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The WORKING GROUP is comprised of representatives of the U. S. Departments of Agriculture; Commerce; Defense; the Interior; Health, Education, and Welfare; State; and Transportation; and the Environmental Protection Agency.

The Pesticide Monitoring Panel consists of representatives of the Agricultural Research Service, Consumer and Marketing Service, Extension Service, Forest Service, Department of Defense, Fish and Wildlife Service, Geological Survey, Food and Drug Administration, Environmental Protection Agency, National Marine Fisheries Service, National Science Foundation, and Tennessee Valley Authority.

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Pesticide monitoring activities of the Federal Government, particularly in those agencies represented on the Pesticide Monitoring Panel which participate in operation of the national pesticides monitoring network, are expected to be the principal sources of data and interpretive articles. However, pertinent data in summarized form, together with interpretive discussions, are invited from both Federal and non-Federal sources, including those associated with State and community monitoring programs, universities, hospitals, and nongovernmental research institutions, both domestic and foreign. Results of studies in which monitoring data play a major or minor role or serve as support for research investigation also are welcome; however, the *Journal* is not intended as a primary medium for the publication of basic research. Manuscripts received for publication are reviewed by an Editorial Advisory Board established by the Monitoring Panel. Authors are given the benefit of review comments prior to publication.

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PESTICIDES IN PEOPLE

DDT and DDE Residues in Blood From Children, South Carolina—1970

Julian E. Keil,¹ William Weston, III,² C. Boyd Loadholt,³
Samuel H. Sandifer,⁴ and James J. Colcolough⁴

ABSTRACT

DDT and DDE residue levels in blood plasma from 192 children in South Carolina, ages 6-9 years, indicated that Negro children had levels two to three times higher than white children. DDT residues averaged 18.4 ppb in Negroes and 6.7 ppb in whites; DDE values for these two races were 55.6 ppb and 24.8 ppb, respectively. White males in this group also had significantly higher levels of both compounds than white females. From the data in this study, baseline levels for a high-risk pediatric group, usually prone to pesticide poisoning, were established.

Introduction

In 1969, the S. C. Community Pesticide Study measured pesticide residues in plasma from 800 persons in Charleston County; the subjects had been selected by sex, race, residence, and 10-year age groupings (6). The outstanding findings of this study were: (1) Negroes had much higher levels of DDT and its metabolites than whites, and (2) children between the ages of 5 and 10 years had significantly higher DDT/DDE levels than the rest of the population studied and, in some instances, as high as DDT formulators.

The present study, partially reported here, was begun with several purposes in mind: (1) to confirm earlier findings of the South Carolina investigators of higher DDT and DDE levels among children and Negroes; (2) to establish new baselines for young children who are prone to poisoning, because analytical methods have changed since the earlier work; and (3) to explore

reasons for the age and racial differences previously reported. This last endeavor is still under study.

Sampling Methods

A total of 192 apparently healthy children, ages 6-9 years from public and private schools in Charleston, S. C., were stratified by race (nonwhites were almost exclusively Negro), sex, residence, and 1-year age spans. After approval by school authorities, written parental permission was obtained for blood sampling of the children. Venous blood samples were collected in heparinized vacutainers. Residue values for this group were compared with those of control subjects of the S. C. Community Pesticide Study.

Analytical Procedures

Plasma extraction was carried out by a modified method of Dale, Curley, and Cueto (1), as recommended by the Perrine Primate Research Branch Laboratory in Perrine, Fla. Two milliliters of plasma were placed in a 16 x 125 mm round-bottom culture tube fitted with screw caps, size 15-415 with Teflon-faced liners; 6 ml of nanograde hexane were added; and the mixture was rotated on a slow speed rotating mixer at 50 rpm for 2 hours. The formation of emulsions was infrequent; when they did occur, centrifugation was used to separate the layers. Following the 2-hour rotation period, a 5-ml aliquot of the hexane layer was quantitatively transferred to a 10-ml graduated concentrator tube. The degree of concentration or dilution that was necessary was determined by a preliminary analysis of the 5-ml aliquot. If concentration was necessary, one 3-mm glass bead was added, and a modified micro-Snyder column was attached. The extract was evaporated on a steam bath to a volume of about 200 μ l. The tubes

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were allowed to cool for about 5 minutes, the micro-Snyder column was removed, and the sides of the tube and column joint were rinsed down with hexane. The final volume was adjusted to 500 μ l for subsequent analysis.

No cleanup procedure was required or utilized. A 5 μ l portion was then injected into a MicroTek 220 gas chromatograph equipped with a tritium foil electron capture detector. All injections were off-column injections. All samples were analyzed on the OV-17/QF-1 column, and the residues were confirmed on the SE-30/QF-1 column. It was necessary in almost all cases to run several sample extract chromatograms of various concentrations to achieve reasonable approximation of peak sizes with those of the standard mixture. The samples were stoppered and mixed on a Vortex mixer for about 30 seconds after each dilution before injection.

GAS-LIQUID CHROMATOGRAPHY

The operating parameters for gas chromatographic analysis were as follows:

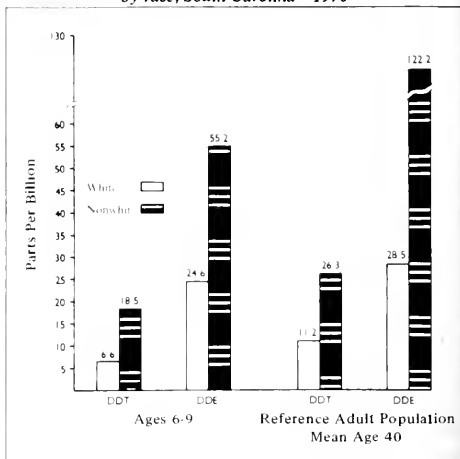
Columns:	1.5% OV-17 and 1.95% QF-1 on Chromosorb W, DMCS, HP, 100/120 mesh; 4% SE-30 and 6% QF-1 on Chromosorb W, DMCS, HP, 100/120 mesh
Temperatures:	Injection chamber 225° C
	Column 200° C
	Transfer block 235° C
	Detector 205° C
Nitrogen flow:	OV-17/QF-1 60 ml/min
	SE-30/QF-1 80 ml/min

All qualitative retention times were based on the retention time of aldrin. Quantitation of pesticide residues was based on active peak areas which were supplied by an Infotronics Chromatograph Integrator Model CRS-101. Recovery, based on the addition of a known quantity of *p,p'*-DDE and *p,p'*-DDT, ranged from 95.0 to 98.6%. Residue results were not corrected for recovery. The minimum reporting limit for *p,p'*-DDE was 1.0 ppb and for *p,p'*-DDT, 2.0 ppb. No polychlorinated biphenyls were detected in any of the samples analyzed. A least squares analysis was used to estimate and test differences between strata means.

Results and Discussion

Results of previous studies indicating unusually high DDT/DDE levels in preadolescents were not confirmed. As shown in Fig. 1, children in the 6-9 age group had lower levels than did their adult counterparts (control subjects of the S. C. Community Pesticide Study). This is at variance with earlier work by Finklea *et al.* (6), but consistent with results reported by Watson *et al.* (7). Improvements in analytical techniques since the

FIGURE 1.—Mean plasma levels of *p,p'*-DDT and *p,p'*-DDE in a pediatric group and adult reference group by race, South Carolina—1970



earlier study (6) may have reduced variances in results and accounted for the lower levels.

DDT and DDE were two to three times higher in the Negro than in the white group. This difference was evident in the juvenile cohorts as well as in the adult reference group (Fig. 1). This study confirms racial differences in DDT/DDE residue levels between whites and nonwhites reported by Davis (2,3), Edmondson (4,5), and Finklea (6).

White males had significantly higher levels of both compounds than white females (Table 1). No significant differences in levels of DDT or DDE were apparent when sex and race were evaluated according to the place of residence of subjects (Table 2); thus, values in Table 1 may be used to estimate "normal" ranges for this high-risk pediatric age group, unusually prone to pesticide poisoning.

Table 3 lists DDT and DDE levels by 1-year age groups and indicates no significant increase or decrease in levels with age.

TABLE 1.—DDT and DDE residue levels in plasma from 192 children, ages 6-9 years, by race and sex, South Carolina—1970

COMPOUND	WHITE*			NONWHITE		
	MALE (N=50)	FEMALE (N=46)	SD [†]	MALE (N=45)	FEMALE (N=51)	SD [†]
DDT (ppb)	8.5	4.8	6.5	16.9	19.7	11.0
DDE (ppb)	29.9	19.2	14.2	55.2	55.9	29.5

[†] Pooled standard deviation for sex within races.

* Differences between levels in white males and females significant at $P < 0.1$.

TABLE 2.—DDT and DDE residue levels in plasma from 192 children, ages 6-9 years, by residence, race, and sex, South Carolina—1970

RACE AND SEX	MEAN RESIDUE LEVELS (PPB)			
	URBAN		RURAL	
	DDT	DDE	DDT	DDE
White Males	7.8 (N=27)	28.2	8.9 (N=23)	31.0
White Females	4.8 (N=24)	16.9	4.7 (N=22)	21.8
Nonwhite Males	16.5 (N=25)	54.5	17.7 (N=20)	54.5
Nonwhite Females	19.1 (N=27)	49.2	20.5 (N=24)	63.5

TABLE 3.—DDT and DDE residue levels in plasma by age of 192 school children, Charleston County, S. C.—1970

AGE (YEARS)	MEAN RESIDUE LEVELS (ppb)	
	DDT	DDE
6 (N=35)	14.5	41.5
7 (N=35)	13.4	43.4
8 (N=42)	11.9	38.1
9 (N=70)	11.7	38.5

See Appendix for chemical names of compounds discussed in this paper.

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Organochlorine Pesticide Residue Levels in Human Milk—Victoria, Australia—1970¹

K. G. Newton and N. C. Greene

ABSTRACT

Samples of human milk were collected in 1970 from 39 rural and 28 urban donors in Victoria, Australia, and were analyzed for organochlorine pesticides using electron capture gas chromatography. All samples contained DDT, DDE, and HCB. Twenty-nine contained dieldrin (mean 0.006 ppm), 12 contained DDD (mean 0.007 ppm), and 3 contained both dieldrin and DDD. Total DDT averaged 0.139 ppm for rural 0.145 ppm for urban donors, and HCB averaged 0.042 ppm and 0.063 ppm, respectively.

Introduction

The Australian State of Victoria has a population of 3.4 million, 2.2 million of whom live in the capital city of Melbourne. The State is self-sufficient in food production; its agriculture, sophisticated and in parts intensive, supplies centralized common markets which distribute food throughout the metropolis.

The use of DDT, aldrin, dieldrin, and other common organochlorine pesticides is restricted to those situations where no suitable alternative is available, and tolerance limits set by State health regulations are low. Over the past 10 years, greater use has been made of the less persistent organophosphate and carbamate pesticides.

In an effort to determine organochlorine pesticide residue levels in human milk, a small survey involving 23 participants (13 rural and 10 urban) was carried out in April 1970. The rural donors lived on or near fruit orchards in Shepparton, the center for a district which produces under irrigation large quantities of tree fruits for the local and export markets. The donors were asked to collect milk during one or more feedings until about

2 ounces was obtained. Simple details on past exposure to pesticides were considered desirable but are not included with the results in Table 2, since efforts to obtain reliable information were unsuccessful.

Differences in the levels of total DDT between the two groups were apparent; however, it was determined that further sampling of other rural areas should be undertaken and that, to maintain parity, more metropolitan samples should be analyzed before valid conclusions could be drawn regarding such differences. A second survey consisting of 26 rural and 18 urban donors was undertaken in December 1970. These donors were selected with on consideration of age, race, weight, medical history, or age of baby.

Resources sufficient to guarantee an accurate history of pesticide exposure and uniform sample collection from each donor were not available. Thus, no exposure information was requested, and the mothers were relied on to collect their own samples according to a designated procedure which was simple enough to ensure cooperation and yet provide a representative sample.

This procedure was established after a preliminary study using one donor to obtain a limited check on variability in fat content of milk during the suckling period. In this study the mother collected a small amount of milk from several feedings at three intervals during each feeding, the beginning, middle, and end. Individual samples taken at each of these intervals were composited by group to provide three samples for analysis, and the results are shown in Table 1. These figures are not included in the general results.

Sampling Procedure

Based on results of the preliminary tests to assess variability in fat content of the milk, the sampling procedure

¹ From the State Health Laboratory, 5 Parliament Place, Melbourne, Victoria, Australia 3002; permission to publish this paper was granted by the Chief Health Officer of Victoria.

TABLE 1.—Organochlorine pesticide residue levels in human milk from one donor obtained at three intervals during each of several feedings

TIME DURING FEEDING	PERCENT FAT	ORGANOCHLORINE PESTICIDE RESIDUE LEVELS (PPM)					
		TOTAL DDT		HCB		DIELDRIN	
		WHOLE MILK	FAT BASIS	WHOLE MILK	FAT BASIS	WHOLE MILK	FAT BASIS
Beginning	1.8	0.014	0.78	0.005	0.25	0.002	0.11
Middle	1.2	0.007	0.58	0.003	0.25	0.001	0.08
End	5.1	0.066	1.29	0.024	0.47	0.006	0.12

described below was followed. Written instructions were given to the volunteers asking them to combine over four or six feedings small quantities of milk taken alternately at the beginning and end of each feeding and to store the samples under refrigeration. A composite milk sample was obtained only once from each donor. Clean glass bottles with aluminum foil-lined caps, preprinted labels, and insulated packing cartons were supplied.

Analytical Procedure

All samples were received in a fresh condition, frozen until ready for analysis, and then thawed at room temperature. The milk was well mixed before aliquots were withdrawn.

All solvents were redistilled in glass before use and tested in conjunction with the other chemical standards for extraneous peaks. The unactivated Florisil used in sample cleanup was "1200 F" Florisil, from supplier's stocks, stored under room conditions. This Florisil was activated by heating at 650° C for 1 hour and stored at 130° C.

Analysis was determined by the Gerber method (8) on a separate 11.0-ml aliquot. Chlorinated hydrocarbon pesticide residues were determined by modified procedures of Giuffrida *et al.* (6) for separating the fat and Mills *et al.* (7) for the cleanup.

Depending on the amount of milk available, 20 to 50 g was weighed into a 350-ml glass-stoppered flask, 100 ml of acetone and 20 g of celite were added, the contents shaken well, and then suction-filtered through a coarse sinter. The retained milk solids and celite were returned to the flask and shaken with 100 ml of hexane and returned. The combined filtrates were transferred to a 1-liter separating funnel and shaken for 30 seconds; 100 ml of water and 10 ml of saturated NaCl were then added and the funnel shaken again for 1 minute. The aqueous layer was discarded, and the dried hexane solution evaporated to 5-10 ml and used to evenly coat, by stirring, 10 g of unactivated Florisil contained in a small beaker. Stirring was maintained to give a dry, free-flowing, fat-coated powder.

This was then poured onto a 1-inch layer of unactivated Florisil in an 18-mm diameter column and eluted with 70 ml of 10% water in acetonitrile; the eluate was collected in a 1-liter separating funnel containing 100 ml of hexane.

From this point the procedures were the same as described in references (6) and (7), except that only one 200-ml elution at 5 ml/minute with 8% ether in hexane was used to remove all organochlorine pesticides, including dieldrin, from the final activated Florisil cleanup column used by Mills *et al.* (7). The removal of dieldrin with a single elution, however, would not have been possible if the Florisil had been made too active by prolonged heating.

Eluates were concentrated to about 0.5 ml and then transferred to glass-stoppered tubes, using hexane, to give a final volume of 2.0 ml. Aliquots from this solution were analyzed on a Varian Aerograph Model 1200 chromatograph equipped with a 250 mc tritium electron capture detector.

Operating conditions were:

Column: Pyrex glass, 5' x 1/8", packed with an equal mixture of 10% DC-200 and 15% QF-1 on individually coated 80/100 mesh Gas Chrom Q. A glass liner was used in the injection port.

Temperatures: Oven 180° C
Detector foil 185° C
Inlet 210° C

Carrier gas: Nitrogen at 35 cc per minute

Injection volumes were held between 2 and 5 µl by sample dilution where necessary. The amounts of pesticides injected were kept within the linear range of the detector.

A hexane solution containing lindane, aldrin, dieldrin, DDD, DDE, and DDT, each at 0.1 ng/µl, was used as a standard. HCB (hexachlorobenzene) and BHC standard solutions were also used at 0.5 ng/µl. Injections of these standards were interspersed with samples during chromatography to provide peak height calibration graphs for quantitation.

All samples were chromatographed on a second column, packed with 12% QF-1 on Gas Chrom Q, to provide qualitative, and a small degree of quantitative, confirmation.

Thin layer chromatography with AgNO_3 -UV visualization, as described by Abbott *et al.* (2), was also used to confirm about one-half of the samples. Additional confirmation of HCB was carried out on four samples by scraping the developed and UV-exposed HCB spot from the plate, extracting the alumina with hexane, and injecting the concentrated extract onto the first column.

Human milk samples spiked with chlorinated hydrocarbon pesticides to levels between 0.2 and 0.4 ppm gave the following recoveries when carried through the full analytical procedure: HCB and lindane—73-77%; aldrin, *p,p'*-DDD, *p,p'*-DDE, and *p,p'*-DDT—80-90%; and dieldrin—83%. The data presented in this report do not include recovery corrections. The lower limit of sensitivity was 0.001 ppm for 50 g of whole milk.

Results

Total DDT shown in the results is the sum, without molecular weight adjustment, of the DDE, DDD, and DDT found.

As stated, the collection of samples occurred in two parts, separated by a period of 7 months. All of the 12 rural samples in the first collection were from one fruit-growing district, whereas the 26 rural samples in the second collection were from 10 other disparate rural areas. The directions given to obtain a representative milk sample from the donors also varied slightly between the two collections.

Table 2 shows the combined results in order to present the information in a unified form. Table 3 presents for total DDT and HCB only, the separate residue levels from the two surveys.

TABLE 2.—Percent fat and organochlorine residues in milk from 67 human donors, Victoria, Australia, 1970

COMPOUND	NUMBER OF SAMPLES WITH DETECTABLE RESIDUES	ARITHMETIC MEAN	RANGE	GEOMETRIC MEAN	SD	SE
PERCENT FAT IN WHOLE MILK						
Rural	36	3.7	1.0-5.6	—	1.24	0.21
Urban	26	4.2	2.5-7.2	—	1.18	0.23
Total	62	3.95	1.0-7.2	—	1.25	0.16
ORGANOCHLORINE RESIDUES IN WHOLE MILK (PPM)						
HCB						
Rural	39	0.042	0.005-0.17	0.031	0.033	0.005
Urban	28	0.063	0.002-0.33	0.040	0.065	0.012
Total	67	0.051	0.002-0.33	0.035	0.050	0.006
DIELDRIN						
Rural	14	0.008	0.002-0.029	—	—	—
Urban	15	0.004	0.001-0.014	—	—	—
Total	29	0.006	0.001-0.029	—	—	—
DDE						
Rural	39	0.100	0.022-0.45	—	0.088	0.012
Urban	28	0.112	0.012-0.29	—	0.070	0.013
Total	67	0.105	0.012-0.45	—	0.081	0.010
DDD						
Rural	4	0.008	0.006-0.014	—	—	—
Urban	8	0.006	0.003-0.010	—	—	—
Total	12	0.007	0.003-0.014	—	—	—
DDT						
Rural	39	0.038	0.007-0.16	—	0.033	0.005
Urban	28	0.034	0.007-0.12	—	0.022	0.004
Total	67	0.036	0.007-0.16	—	0.029	0.004
TOTAL DDT						
Rural	39	0.139	0.033-0.58	0.109	0.119	0.019
Urban	28	0.145	0.015-0.40	0.118	0.089	0.017
Total	67	0.141	0.015-0.58	0.112	0.107	0.013
TOTAL DDT (FAT BASIS)						
Rural	36	4.63	0.81-25.35	3.22	5.21	0.87
Urban	26	3.73	1.11- 6.88	3.29	1.78	0.35
Total	62	4.25	0.81-25.35	3.25	4.16	0.53

¹Insufficient sample in five cases for a fat determination.

TABLE 3.—Total DDT and HCB residues in human milk from rural and urban donors by survey

SAMPLE GROUP	RESIDUES IN PPM					
	TOTAL DDT			HCB		
	ARITHMETIC MEAN	RANGE	GEOMETRIC MEAN	ARITHMETIC MEAN	RANGE	GEOMETRIC MEAN
APRIL 1970						
Rural (13 samples)	0.208	0.045-0.58	0.145	0.039	0.010-0.080	0.031
Urban (10 samples)	0.142	0.015-0.40	0.105	0.040	0.002-0.090	0.025
DECEMBER 1970						
Rural (26 samples)	0.104	0.033-0.20	0.095	0.043	0.005-0.17	0.031
Urban (18 samples)	0.147	0.051-0.31	0.125	0.076	0.016-0.33	0.052

since the distribution of the levels is skew, geometric means are shown, and as the results for some pesticides cover a wide range, standard deviation and standard error of the mean have been included where considered appropriate.

Five samples were too meager to determine the fat content. The total DDT in each was 0.033, 0.015, 0.10, 0.072, and 0.058 ppm, all below the mean. However in the absence of fat determinations, it is difficult to assess the effect of their exclusion on the mean concentration of total DDT calculated on a fat basis.

Forty-three donors supplied the age of their baby at the time of the survey; these ages ranged from 2 to 46 weeks.

Discussion

All samples contained HCB. It is possible that HCB entered the food chain of Victorians from the improper channelling of HCB-treated seed wheat into the local poultry and stock food industries following a series of severe reductions in wheat acreage during the period of worldwide wheat over-production in the past decade.

All samples contained DDT and its first metabolite DDE, but only 12 had the second metabolite DDD. Total DDT ranged from 0.015 to 0.58 ppm, with a mean of 0.141 ppm. These results may be considered in respect to other research findings. Egan *et al.* (5) reported total DDT in the range of 0.075 to 0.170 ppm with a mean of 0.126 ppm for 19 human milks in England. Curley and Kimbrough (4) determined total DDT in the milk of five U.S. women at three stages during a period of from 3 to 96 days postpartum. Although the range was small, 0.05 to 0.15 ppm, two subjects showed an increase in levels, two a decrease, and one a steady state over this period.

In this present study total DDT in human milk, on a fat basis, ranged from 0.81 to 25.35 ppm, with a geometric mean of 3.25 ppm. The ratio of DDE to DDT based on arithmetic means was 2.7 for rural donors, 3.3 for urban, and 2.9 overall.

Abbott *et al.* (1), in a study in the United Kingdom on postmortem human fat (from 91 female subjects over 3 years of age, found total DDT to range from 0.21 to 8.10 ppm, with a geometric mean of 2.2 ppm. For their 248 male and female postmortem samples grouped according to sex, age, and district, they noted a fairly constant ratio for the groupings of 1 part of DDT to 2.6 parts DDE.

Bick (3), in a survey in Victoria using biopsy specimens of human body fat collected from 23 adult males and 30 adult females, found for total DDT a geometric mean of 2.00 ppm for the males, 1.68 ppm for the females, and a combined range of 0.48 to 6.35 ppm. These figures are on a fresh-tissue basis; mention is made that the lipid content of biopsy specimens is variable.

In this study, the mean levels of total DDT in human milk exceeded 0.05 ppm, the limit for total DDT in cow's milk proposed by the FAO WHO and the tolerance set by the U. S. Food and Drug Administration. These limits are based in part on the fact that DDT is now widespread in the biosphere and on the expectation that an individual will consume milk in his diet for a lifetime. They do not provide for residues resulting from the deliberate use of DDT on dairy farms.

The results reported here thus pose a difficult problem for those who are responsible for advising mothers on the feeding of their newborn babies.

Acknowledgment

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See Appendix for chemical names of compounds discussed in this paper.

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Organochlorine Pesticide Levels in Human Serum and Adipose Tissue, Utah—Fiscal Years 1967-71¹

Stephen L. Warnick

ABSTRACT

Organochlorine pesticide residue levels were determined in 1,417 serum and 103 adipose samples collected during fiscal years 1967-71 from residents of Utah; levels agreed closely with values reported for persons from other parts of the United States. The results supported previous evidence of no increase in pesticide storage in the general population since 1951 and a tendency towards decreased storage since 1966. Specific findings from the Utah samples included the following:

- (1) Significantly higher organochlorine levels were found in samples from persons occupationally exposed to pesticides than in those from the general population.
- (2) Significantly higher levels of DDE were found in serum samples from persons 21 years of age and older than in those from persons under 21.
- (3) Although residue levels were higher in males than in females, the difference was not significant.
- (4) Mean values of total DDT in adipose tissue for the years in which these samples were obtained were 9.0 ppm in 1968, 7.2 ppm in 1969, and 5.3 ppm in 1970, indicating a decrease in storage levels.
- (5) Lastly, results from studies of the relation between residue levels in serum and adipose tissue, between levels in serum and food, and between serum and household dust, indicated no significant correlations. However, the latter two correlations closely approximated a significant relationship, and the results suggested that the respiratory route of exposure to pesticides may be as significant as dietary intake in maintaining an individual's body burden of pesticides.

Introduction

A chief concern regarding pesticides is the relationship between pesticide residues in human tissues and human health. Although current evidence indicates that present levels of pesticides in man's environment, food, and body are not adversely affecting human health, it is prudent to ascertain the body burden of pesticides. Numerous studies on pesticide levels in people have been reported (1,3,5-8), including two recently completed studies in Arizona (2) and Idaho (4).

Utah is one of 14 States under contract with the Federal Environmental Protection Agency to investigate the effect of pesticides on human health. One aspect of these studies is to determine levels of pesticides in the environment and human population of each project State. Since the projects are widely scattered throughout the contiguous United States and Hawaii, pooling results from these studies provides up-to-date representative information for the whole country.

This paper reports the results of 5 years' analyses for the presence of organochlorine pesticides in serum and adipose tissue samples from Utah residents. Residue levels are presented according to sex, age, race, and exposure of residents; method of sample analysis; and year of sampling. Results of studies correlating pesticide levels between serum and adipose tissue, between serum and food, and between serum and house dust are also reported. The discussion in this paper is limited to residues of *p,p'*-DDT, *p,p'*-DDE, total DDT (including

¹ From the Utah Community Pesticide Study, State of Utah Department of Social Services, Division of Health, 44 Medical Drive, Salt Lake City, Utah 84113.

DDT, DDE, and DDD), and dieldrin, although several other residues were present in both serum and adipose tissue samples.

Sampling Methods and Analytical Procedures

A total of 1,417 blood samples (fresh serum rather than whole blood) and 103 adipose tissue samples from the population of Utah were collected and analyzed for pesticide residues during the 5-year period, fiscal years 1967-71. The blood samples included 970 from the general population and 447 from persons occupationally exposed to pesticides.

Sample extraction, cleanup, and analyses were performed according to methods prescribed for the Community Pesticide Studies Laboratories. These included the Radomski (21) and the Mills, Onley, and Gaither (22) methods for analyzing adipose tissue; the Dale, Curley, and Cueto (23) method for blood; and modifications of the Mills method for adipose and the Dale method for serum as described in the "Manual of Analytical Methods" (9), prepared by the Primate Research Laboratories, Environmental Protection Agency, Perrine, Fla. The Perrine Laboratory also provided the pesticide standards used in this study and carried out a quality control program in which the Utah Laboratory participated. The MicroTek 220 gas chromatograph equipped with a Ni⁶³ detector was used for identifying pesticides, and a two-column gas chromatographic system was used routinely for confirmation. Recovery studies have shown that, for the pesticides reported, recovery is routinely greater than 90%.

Results

Table 1 presents values from the literature for *p,p'*-DDT, *p,p'*-DDE, DDT + DDE, and dieldrin levels in people from various areas of the United States for comparison with levels in the general population of Utah. Residue levels in Utah residents compared closely with results from other areas of the country despite considerable differences in quantities of pesticides used.

TABLE 1.—Mean organochlorine pesticide concentrations in human blood and adipose tissue from Utah and levels reported for other areas of the United States

COMPOUND	UTAH		CHICAGO ¹		ARIZONA ²		FLORIDA ²		IDAHO ³		UTAH ⁴	
	BLOOD (PPB) N=970	ADIPOSE (PPM) N=103	BLOOD (PPB) N=959	ADIPOSE (PPM) N=959	BLOOD (PPB) N=70	ADIPOSE (PPM) N=70	BLOOD (PPB) N=119	ADIPOSE (PPM) N=159	BLOOD (PPB) N=100	ADIPOSE (PPM) N=100	BLOOD (PPB) N=89	ADIPOSE (PPM) N=89
<i>p,p'</i> -DDT	3.8	1.5		2.4	1.5	7.0	4.3	4.7			4.7	1.3
<i>p,p'</i> -DDE	17.6	5.0		6.4	4.6	11.3	7.0	22.0			22.5	4.5
DDT + DDE	21.4	6.5		8.8	6.1		11.3	26.7			27.2	5.8
DIELDRIN	0.90	0.17		0.14 (N=221)	0.14			0.5				0.04

¹Hoffman *et al.*, 1967.

²Morgan and Roan, 1970.

³Davies *et al.*, 1968.

⁴Watson *et al.*, 1970.

⁵Casarett *et al.*, 1968.

Table 2 is a summary of organochlorine pesticide residue levels in blood of Utah residents according to sex, age, race, and exposure; method of analysis; and year of sampling. Table 3 presents organochlorine residue levels in adipose tissue, collected at autopsy from Utah residents who died in accidents; these data are given according to the residents' sex, age, race, and year of sampling.

TABLE 2.—Average organochlorine pesticide residue levels in human serum, Utah—fiscal years 1967-71

	No. of SAM- PLES	RESIDUES IN PPB			
		<i>p,p'</i> -DDE	<i>p,p'</i> -DDT	TOTAL DDT ¹	DIELDRIN
Total	1,417	19.8	6.2	28.3	1.8
Sex					
Male	1,003	21.9	6.9	31.5	2.1
Female	414	14.6	4.3	20.5	0.9
Year					
FY 1967	72	19.5	9.8	32.8	4.5
FY 1968	237	15.4	7.2	24.7	2.1
FY 1969	267	20.8	7.9	31.2	1.6
FY 1970	439	18.7	4.3	25.1	1.6
FY 1971	402	22.9	5.7	31.2	1.4
Method of Analysis					
Method 1 (Single Extract)	172	15.2	8.4	26.1	3.4
Method 2 (Triple Extract)	843	19.3	5.9	27.4	1.6
Method 3 (2-hour Roto-rack Ext.)	402	22.9	5.7	31.2	1.4
Exposure Group					
Exposed Workers	447	24.7	11.1	38.9	3.7
General Population	970	17.6	3.8	23.4	0.9
Age					
< 21	202	13.4	3.6	18.5	0.6
≥ 21	1,215	20.9	6.6	29.9	2.0
Race					
Caucasian	1,347	19.8	6.0	28.1	1.8
Negro	4	14.3	4.8	20.8	0.8
Oriental	22	35.6	18.6	58.4	1.0
Indian	43	14.0	4.4	20.1	0.5
Mexican	1	16.0	3.0	21.0	1.0

¹Total DDT = DDT + 1.114 (DDE + DDD), formula used to adjust for differences in molecular weight.

TABLE 3.—Average organochlorine pesticide residue levels in human adipose tissue, Utah—fiscal years 1967-71

	No. OF SAMPLES	RESIDUES IN PPB			
		p,p'-DDE	p,p'-DDT	TOTAL DDT ¹	DIELDRIN
Total	103	5.03	1.53	7.31	0.17
Sex					
Male	71	5.36	1.68	7.85	0.19
Female	32	4.31	1.20	6.10	0.14
Year					
FY 1968	48	5.95	2.13	9.01	0.20
FY 1969	15	4.81	1.51	7.15	0.15
FY 1970	40	4.02	0.83	5.33	0.15
FY 1971	No fat samples analyzed				
Age					
< 21	15	4.11	1.25	5.97	0.09
≥ 21	88	5.19	1.58	7.54	0.19
Race					
Caucasian	96	4.75	1.32	6.76	0.16
Negro	4	7.90	3.70	12.70	0.20
Oriental	1	14.40	11.50	29.60	0.80
Indian	1	4.80	1.50	7.10	0.30
Mexican	1	11.40	3.10	16.40	0.30

¹Total DDT = DDT + 1.114 (DDE + DDD), formula used to adjust for differences in molecular weight.

Discussion

There is no evidence of increased storage of organochlorine pesticides in the general population since 1951 (13), and, further, this study and others show a tendency toward decreasing levels since 1966. The fact that there has not been progression in storage can probably be attributed to regulations that have maintained low dietary residues.

The question of changes in pesticide levels with time is interesting but complicated by the need for standardized analytical methodology. Improved methods of analysis the past few years have offset the very subtle changes that may have taken place in tissue stores as related to changing patterns of pesticide usage.

In comparing total DDT in the blood of Utah people as related to sex, the overall average for males was 31.5 ppb and 20.5 ppb for females. Total DDT in adipose was 7.8 ppm for males and 6.1 ppm for females. In both cases, the levels in males were higher, but the difference was not found to be statistically significant.

In comparing total DDT in the blood as related to age, levels in persons 21 years of age and older averaged 29.9 ppb, and levels in those under 21 years averaged 18.5 ppb. In adipose tissue persons 21 years of age and older had average levels of 7.5 ppm, while those under 21 years had 6.0 ppm. In both cases the levels of total DDT in people over 21 years were higher but not significantly so. Of the individual compounds, only for levels of serum DDE was the difference according to age significant.

In comparing total DDT in the blood as related to exposure, results showed that the general population had an average level of 23.4 ppb, while a group of occupationally exposed workers averaged 38.9 ppb; this difference is significant at ($P < 0.05\%$). Adipose tissue was obtained only from the general population; the average level for total DDT was 7.3 ppm.

In comparing total DDT in the blood by method of analysis, residues determined using the single hexane extraction method averaged 26.1 ppb; the triple hexane extraction method, 27.4 ppb; and the 2-hour roto-rack hexane extraction method, 31.2 ppb. The last two methods significantly increased DDE recovery, but not DDT. These more efficient analytical methods may have offset a decline in storage levels of DDE in blood during years 1969, 1970, and 1971. There were some changes in the method for adipose analysis, but the changes did not affect pesticide recovery significantly.

Comparison of total DDT found in the blood by year, is difficult due to changes in analytical methodology, but levels for 1967, 1969, and 1971 were higher than levels in 1968 and 1970. An interesting trend was noted in the adipose samples which averaged 9.0 ppm in 1968, 7.1 ppm in 1969, and 5.3 ppm in 1970. This decrease may have been related to a 75% reduction in the use of DDT in Utah during this same period. A budget cut in fiscal year 1971 prevented collection of adipose tissue this year, so it was not determined if the downward trend continued.

An insufficient number of samples made a comparison of DDT in the blood and adipose as related to race invalid, except perhaps in the case of American Indians. A total of 43 blood samples were obtained from American Indians; these averaged 20.1 ppb compared to 28.1 ppb for Caucasians, perhaps reflecting less exposure for the Indians.

Although many studies have investigated the mechanics of DDT storage and metabolism, even after two decades the process is not completely understood. There does appear to be a direct relationship between the daily intake of DDT and levels in the fat achieved at equilibrium. Hayes (14) reached this conclusion by feeding DDT to human volunteers for 18 months and showing that even when doses were 200 times as much as found in the daily diet, DDT and DDE concentration in the fat leveled off in about a year. Other studies have supported his finding.

When DDT is ingested, carbon-14 studies (15) have shown that a good portion is not absorbed, but excreted directly in the feces. As much as 19% is also excreted

in the feces and urine as DDA. The remainder is in equilibrium between storage in the fat and circulation in the blood where it is gradually metabolized and excreted.

Recent studies have shown that pesticide storage is affected by other pesticides, certain drugs, and diet deficiencies (16-19). A study in Arizona (20) in which human volunteers were given DDT showed that DDT ingestion does not markedly increase DDE storage, indicating that the body burden of DDE is ingested as DDE. Only about 8% of the stored DDE came from the ingested DDT.

There was no attempt in this paper to relate pesticide levels to pathology; but other studies (1,2,10-12) have shown no correlation between levels similar to those found in Utah residents and abnormalities in human health. On this basis, it can be assumed that present levels of organochlorine pesticides in Utah people are not a threat to the health of the community.

CORRELATION STUDIES

A study was done on 40 people supplying both adipose tissue and serum samples to determine if a significant correlation existed between pesticide levels in adipose and serum. The correlation coefficients (r) were 0.22 for p,p' -DDT and 0.06 for p,p' -DDE. To be significant at $P < 5\%$ for 40 samples, the correlation coefficient must be $r = 0.32$, thus no statistically significant relationship was found between levels in the two types of samples.

Another part of the research by the Utah Community Pesticide Study is to determine environmental pesticide levels in food, water, air, house dust, soil, wildlife, etc. To accomplish this, total diet food samples and vacuum cleaner dust samples are collected from selected homes and analyzed for pesticide residues. A study was done to see if DDT levels in the serum correlated with either DDT levels in food or DDT levels in house dust. Ten homes representing the minimally exposed general population were chosen. Blood samples were also collected from the male head-of-household and analyzed for pesticides. The following correlation coefficients were then determined for total DDT residues between serum and food, $r = 0.41$ and between serum and house dust, $r = 0.47$. To be significant at $P < 5\%$ for a sample of 10, the correlation coefficient would have to be $r = 0.52$, thus neither of these relationships was statistically significant. It is interesting to note, however, that the correlation is greater between serum and house dust than between serum and food. This is reasonable based on the fact that the mean level of total DDT in house dust was 1000 times greater than in food (5120 ppb

house dust; 6.13 ppb food). This does indicate that the respiratory route of exposure may be as significant as diet in maintaining a person's body burden of pesticides. Additional samples are being collected and analyzed in an attempt to confirm this theory.

Acknowledgment

Pesticide analyses were performed with the assistance of Lynn Thomas, and data analyses were done by Tom Whitley.

See Appendix for chemical names of compounds discussed in this paper.

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RESIDUES IN FISH, WILDLIFE, AND ESTUARIES

*Mirex and DDT Residues in Wildlife and Miscellaneous Samples in Mississippi—1970*¹

Karl P. Baetcke, Jimmie D. Cain, and William E. Poe

ABSTRACT

Samples of wildlife and a few miscellaneous samples, such as beef, were collected in Mississippi in 1970 and analyzed for the presence of mirex and DDT and its analogs. Levels of mirex residues were found to range from 0 ppm to a high of about 104 ppm; residues of DDT and its metabolites (DDTR = DDT + DDE + DDD) were found to range from <0.001 ppm to 126 ppm. Comparisons of the amounts of the two pesticides found in individual samples showed that mirex residues often exceeded DDT residues. Collections of wildlife were begun in the spring of 1970, 1 year after mirex had been applied by aerial application in the primary study area. The high levels of mirex residues found in some of these samples 1 year after treatment indicate that mirex can be considered a persistent pesticide.

Introduction

The insecticide mirex has been used widely in Mississippi and other parts of the Southeast for the control of the imported fire ant (*Solenopsis saevissima*).

Mirex was first used in Mississippi in 1962. It has since been employed by farmers in many parts of the State for local fire ant control and, in some instances, in county-wide programs involving aerial application. For example, in 1962, 40,000 acres in Washington County in the Delta were aerially treated in an apparently successful attempt to eradicate a fire ant infestation. In 1965, 1966, and 1967, an aerial application program was conducted in portions of eight counties located in north-eastern Mississippi. In 1969, portions of Oktibbeha, Noxubee, and Lowndes Counties were treated with mirex by aerial application; Oktibbeha and Noxubee Counties each received two applications and Lowndes County received three applications. In this instance, 1.25 lb of

bait containing 0.3% mirex was applied per acre. These counties, located in the hill section of east central Mississippi, were thought to be areas in which wildlife might have accumulated mirex as a result of recent exposure.

This report presents data on mirex residues in deer, birds, fish, arthropods, beef, cows' milk, bird eggs, earthworms, silage, and fescue from these counties. The samples were also analyzed for DDT and its analogs in order to make possible some general comparisons between residues representing a pesticide (mirex) which has been employed recently primarily in local situations and one (DDT) which has had long-term widespread usage. A few additional samples were collected in the Delta area of Mississippi where no recent applications of mirex had been made, and results of analysis for mirex residues in these samples are also discussed in this report.

Sampling

The species selected as samples for this investigation were chosen to represent as wide a variety as possible of the larger species in the food chain. The following species were collected for analysis:

Deer	<i>Odocoileus virginianus</i> (Zimmerman)
Birds	
chicken	<i>Gallus gallus</i>
bobwhite quail	<i>Colinus virginianus</i> (Linnaeus)
brown thrasher	<i>Toxostoma rufum</i> (Linnaeus)
blue jay	<i>Cyanocitta cristata</i> (Linnaeus)
meadow lark	<i>Sturnella magna</i> (Linnaeus)
turkey	<i>Meleagris gallopavo</i> (Linnaeus)
eastern kingbird	<i>Tyrannus tyrannus</i> (Linnaeus)
robin	<i>Turdus migratorius</i> (Linnaeus)
barred owl	<i>Strix varia</i> (Barton)

¹ From the Mississippi Community Study on Pesticides, Department of Biochemistry, Mississippi State University, State College, Miss. 39762.

Fish	
channel catfish	<i>Ictalurus punctatus</i> (Rafinesque)
green sunfish (breast)	<i>Lepomis cyanellus</i> (Rafinesque)
Arthropods	
crickets	Orthoptera: Gryllidae
spiders	Arachnida
chinch bugs	Hemiptera: Lygaeidae
pill bugs	Isopoda
beetles	Coleoptera: Carabidae, Elateridae, Coccinellidae, Chrysomelidae
walking sticks	Orthoptera: Phasmidae
praying mantis	Orthoptera: Mantidae
stink bugs	Hemiptera: Pentatomidae
katydid	Orthoptera: Tettigoniidae
grasshoppers	Orthoptera: Locustidae
Miscellaneous	
cattle egret eggs	<i>Bubulcus ibis</i> (Linnaeus)
little blue heron eggs	<i>Florida caerulea</i> (Linnaeus)
cows' milk	
earthworm	<i>Lumbricus terrestris</i> (Linnaeus)
beef	
silage	
fescue	<i>Festuca</i> sp.

Deer samples were obtained from freezer locker plants and from hunters in the Delta area in and around Greenville, Mississippi, and in east central Mississippi near Oktibbeha, Noxubee, and Lowndes Counties (hereafter referred to as the hill area). The bird samples except for two robins collected in the Delta were from the hill area around Starkville and the Noxubee Wildlife Refuge; all arthropods, fish, and miscellaneous samples were from the hill area (Oktibbeha and Lowndes Counties). All samples from the hill area were obtained from locations which had been subjected to mirex applications.

Analytical Procedures

The sampled species, although differing widely in type, did not present any unusual problems in extraction or cleanup for residue analysis. The procedures chosen for extraction and cleanup of mirex, the pesticide of specific interest in this study, as well as DDT were already well established although recovery rates for mirex were not available. Recovery rates for both mirex and DDT and its analogs were determined for this study. Electron capture gas-liquid chromatography was utilized in the determinative step. Thin layer chromatography and infrared spectroscopy were utilized as confirmatory procedures in selected samples. Fish and birds were dissected and the liver, p. ose, and brain tissues were analyzed separately by the procedure described in the Pesticide Analytical Manual, Vol. III (1); however, adipose tissue

was not obtained from all avian species because of an apparent depletion of fat reserves.

Only adipose tissue was selected from beef and deer for analysis, using the same procedures as that for fish and bird tissue analysis. However, because of the variation in the state of the deer samples which included fresh tissues, dehydrated tissues, and, in extreme cases, partially decomposed tissues, meaningful comparisons of residue values based on fresh weights were not possible. Therefore, in order to make the deer fat samples more comparable, the samples were ground in a Duall tissue grinder with petroleum ether (b.p. 30-60° C), the ether evaporated on a 60° C water bath, and sampling for residue analysis made on the resulting petroleum ether soluble lipids according to Enos *et al.* (1).

Residues in cows' milk were determined on a fat basis utilizing the procedure described in the Pesticide Analytical Manual, Vol. I (2) for the extraction of fat. Cleanup of the milk fat prior to GLC analysis was accomplished by the procedure of Langlois, Stemp, and Liska (3) with the following modification: Instead of incorporating fluid milk into the top Florisil layer, 1 g or less of extracted butterfat contained in about 25 ml of 30-60° C petroleum ether was incorporated into Florisil (deactivated with 5% water) to form the top layer. This modification extends the usefulness of this cleanup method making it applicable to a wide variety of plant and animal tissue extracts. Extraction of residues from silage and fescue samples was handled essentially as described by Mills, Onley, and Gaitner (4). This was followed by cleanup as previously described for milk fat.

Extraction and cleanup of cattle egret and little blue heron eggs were accomplished according to Cummings *et al.* (5); no problems were encountered during subsequent gas chromatography. Arthropods and earthworms were composited by maceration in a Duall tissue grinder, and the micromethod of Enos *et al.* (1) was used for the extraction, cleanup, and residue determination on the composite.

Primary identification and quantification of the pesticides were accomplished on a MicroTek NT-220 gas chromatograph. Two columns having different resolution characteristics were utilized on every sample. Instrument parameters were as follows:

- Columns: (A) Borosilicate glass, 6' x 1/4", packed with 1.5% OV-17, 1.95% QF-1 on 80/100 mesh Supelcoport
 (B) Borosilicate glass, 6' x 1/4", packed with 4% SE-30, 6% QF-1 on 80/100 mesh Supelcoport
 Detector: Electron capture, having 130 mc tritium ionizing source

Temperatures: Injector 235° C
 Column 195° C
 Detector 210° C
 Carrier gas: Prepurified nitrogen flowing at 90 ml/min (Column A) and 60 ml/min (Column B)

RECOVERY RATES

To establish recovery rates, adipose, liver, and brain tissues collected from chickens were spiked with known amounts of *o,p'*-DDT, *p,p'*-DDT, *p,p'*-DDE, and mirex, and the tissues were then analyzed for these residues utilizing the procedures described above. Recoveries on each tissue were based on three unspiked samples and two levels of fortification with DDTR (DDT and its analogs) and mirex, each consisting of three replicates. The first level consisted of 0.21 ppm DDTR (0.10 ppm *p,p'*-DDE + 0.10 ppm *p,p'*-DDT + 0.01 ppm *o,p'*-DDT) and 0.10 ppm mirex; the second level consisted of 21.00 ppm DDTR (10.00 ppm *p,p'*-DDE + 10 ppm *p,p'*-DDT + 1.00 ppm *o,p'*-DDT) and 10.00 ppm mirex. Percent recoveries at the first level averaged 110%, 88%, and 98% for DDTR and 71%, 81%, and 110% for mirex in adipose, liver, and brain tissue, respectively. At the second level, recoveries were 93%, 85%, and 102% for DDTR and 82%, 91%, and 102% for mirex in adipose, liver, and brain tissue, respectively.

Efficiency of the analytical technique utilized on arthropods was determined by grinding 10 crickets and dividing the material into four 500 mg samples consisting of three spiked samples and one blank. The spike consisted of 1.0 ppm *p,p'*-DDT, 1.0 ppm *p,p'*-DDE, 0.1 ppm *o,p'*-DDT, and 1.0 ppm mirex. Based on one level of fortification and three replicates, percent recovery averaged 86% for DDTR and 92% for mirex. None of the data presented herein were corrected on the basis of recovery rates.

Confirmation of the presence of mirex was made by thin layer chromatography of six samples (two brown thrashers, two bluejays, one channel catfish, and one robin), on Brinkmann-Silica Gel-G plates and developed with n-heptane solvent. Retention values for the mirex standard and for the mirex extracted from samples both averaged .75, thus confirming the identification of the extracted compound as mirex. Further confirmation of the presence of mirex was obtained from infrared spectra of two samples (the channel catfish and one blue jay); however, additional infrared spectra could not be obtained due to insufficient quantities of mirex in the remaining four samples.

Confirmation of DDT and its analogs was accomplished by extraction p-values in some similar wildlife samplings carried out prior to the experiments relating to this study. Many confirmations by this technique have been made in the past, and to date, polychlorinated biphenyls have not been found to be present in quantities sufficient to

interfere with DDT calculations. It is true that PCB's when present at high enough concentrations, interfere with identification of *p,p'*-DDE, *p,p'*-DDD, *o,p'*-DDT, *p,p'*-DDT, and others. However, PCB's, notably Aroclor 1254 and Aroclor 1248, also exhibit tell-tale electron capture responses in regions of the chromatogram both earlier and later than that occupied by DDT and analogs. None of these patterns were observed in any samples in this study. This fact does not, of course, preclude the presence or absence of PCB's. It does show, however, that they were not present in high enough concentration to interfere with the identification and quantitation of DDT and its analogs under the conditions of these experiments.

Results and Discussion

The results of the analyses for mirex residues are given in Tables 1-4. Each table also shows the amount of DDTR (the sum of *p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDE and *p,p'*-DDD) found in each sample analyzed. Mirex residues were found to range from 0 ppm to a high of about 104 ppm; DDT and its metabolites (DDTR = DDT + DDE + DDD) were found to range from <0.001 ppm to 126 ppm.

By analyzing for DDT residues, in addition to mirex residues, it was possible to make some general comparisons between residues representing a pesticide which has been employed primarily in local situations very recently (mirex) and one which has had long-term widespread usage (DDT). Data are presented on individual samples in the tables in order that more com-

TABLE 1.—Mirex and DDT residues in adipose tissue of deer collected in Noxubee, Oktibbeha, and Monroe Counties, Mississippi—1970

DEER SAMPLE NUMBER	RESIDUES IN PPM					
	<i>p,p'</i> -DDE	<i>p,p'</i> -DDD	<i>o,p'</i> -DDT	<i>p,p'</i> -DDT	DDTR	MIREX
1	0.015	0.010	—	0.096	0.121	—
2	0.015	0.010	—	0.107	0.132	—
3	0.025	0.014	0.011	0.059	0.109	0.09
4	0.035	0.029	—	0.178	0.242	0.17
5	0.023	0.013	0.007	0.102	0.145	—
6	—	0.014	—	0.057	0.070	0.28
7	0.033	0.014	0.007	0.094	0.148	—
8	0.105	0.316	—	1.290	1.711	—
9	0.038	0.015	—	0.083	0.135	—
10	0.059	0.029	—	0.359	0.447	—
11	0.055	0.022	—	0.188	0.265	—
12	0.051	0.034	—	0.128	0.213	0.30
13	0.022	0.016	0.007	0.101	0.146	0.06
14	0.006	0.008	—	0.063	0.077	—
15	0.114	0.035	—	0.405	0.554	—
16	0.098	0.074	—	0.428	0.600	—
17	0.025	0.007	0.009	0.080	0.121	0.04
18	0.032	0.024	0.009	0.112	0.177	0.12
19	0.036	0.025	0.012	0.187	0.270	—

NOTE: — indicates none detected; all results on lipid basis.

plete comparisons can be made between DDTR residues and mirex residues.

The results of the current investigation demonstrate widespread occurrence of mirex in wildlife in the treated area. Of 19 deer adipose samples from the hill area of Mississippi, 7 contained mirex (Table 1). Mirex was not found in any of 51 samples collected from deer in the Delta where mirex use had been sporadic and very localized. Data pertaining to the Delta deer samples are

not presented in the tables since the main purpose of this report is to compare levels of mirex with levels of DDT, as well as to show to what extent mirex is present in samples from the treated area.

Of 42 individual birds representing 9 species analyzed for the presence of mirex, mirex was found present in one or more tissues of 41 birds (Tables 2a-2d). Mirex was not found in any tissue of one turkey (Tables 2a-2c). Of all fish from the hill area analyzed for mirex,

TABLE 2a.—Mirex and DDT residues in liver tissue of fish and birds collected in Oktibbeha, Noxubee, and Lowndes Counties, Mississippi—1970

SPECIMEN	SAMPLE NUMBER ¹	RESIDUES IN PPM					
		p,p'-DDE	p,p'-DDD	o,p'-DDT	p,p'-DDT	DDTR	MIREX
Fish	1	0.046	0.011	T	0.012	0.069	0.281
Channel catfish	2	0.004	—	—	—	0.004	0.023
	3(2)	0.089	0.044	0.046	0.048	0.227	0.337
	4(2)	0.121	—	0.052	0.024	0.197	0.674
	5	0.023	0.028	0.047	0.060	0.158	—
	6	0.054	0.063	0.061	0.068	0.246	—
	7	0.024	0.052	0.056	0.021	0.153	—
Birds							
Chicken	1	0.823	0.047	0.024	0.150	1.044	0.550
Bobwhite quail	1	0.404	—	—	0.006	0.410	0.013
	2	0.389	0.003	0.012	0.007	0.411	0.027
	3	0.101	T	T	T	0.101	—
	4	0.421	T	T	T	0.421	—
	5	0.400	T	T	T	0.400	—
	6	0.156	T	T	T	0.156	0.014
	7	0.281	T	T	0.018	0.299	0.052
	8	0.459	T	0.015	T	0.474	0.016
	9	0.270	0.005	0.012	0.017	0.304	0.077
	10	0.144	0.004	—	0.009	0.157	—
	11	0.487	T	0.016	T	0.503	0.031
	12	0.303	T	T	T	0.303	0.027
	13	0.057	T	T	T	0.057	0.026
14	0.371	T	T	T	0.371	0.147	
15	0.021	T	T	T	0.021	0.034	
16	0.254	—	—	—	0.254	0.034	
17	0.194	T	T	T	0.194	0.209	
18	0.339	T	—	T	0.339	0.216	
19	0.161	—	—	—	0.161	0.050	
20	0.111	—	—	—	0.111	0.295	
Brown thrasher	1	1.369	—	0.029	0.026	1.424	0.456
	2	1.519	—	—	0.118	1.637	1.811
	3	4.228	—	0.046	0.119	4.393	1.838
	4	1.725	—	0.015	0.045	1.785	1.242
Blue jay	1	0.499	—	0.013	0.046	0.558	1.434
	2	0.415	—	—	—	0.415	0.350
	3	0.514	—	—	0.023	0.537	1.506
Meadow lark	1	1.312	0.010	0.014	0.233	1.569	7.564
	2	1.348	0.007	0.011	0.095	1.461	3.744
Turkey	1	0.090	—	—	T	0.090	0.076
	2	—	—	—	—	—	—
	3	—	—	—	—	—	—
	4	—	—	—	—	—	—
	5	1.130	0.010	0.010	0.026	1.176	0.206
	6	0.329	T	T	0.033	0.362	0.475
Eastern kingbird	1	5.462	0.077	0.154	0.308	6.001	0.131
Robin	1	—	—	—	—	—	—
	2	13.879	5.042	0.206	0.158	19.285	0.724
	3	13.332	4.220	0.174	0.255	17.951	0.298
	4	0.583	0.062	0.039	0.089	0.773	1.443
	5	1.683	0.151	0.044	0.086	1.964	0.694
Barred owl	1	4.046	0.012	—	0.052	4.110	4.072

NOTE: Blank = sample not analyzed; — indicates none detected; T = trace = < 0.002 ppm mirex; all results on fresh-weight basis. Numbers in parentheses indicate composite of two or more fish or birds. Samples collected in the Delta.

4 of 5 channel catfish samples were found to contain the pesticide (Tables 2a, 2b, 2d) and 2 of 2 green sunfish (bream) samples were found to contain the pesticide (Table 2b). Of 25 individual and composite samples of arthropods analyzed for pesticides, 10 were found to contain mirex. It should be pointed out that all of the samples containing mirex, with the exception of two robins, were collected from the area that had been treated relatively recently with mirex.

Among the few miscellaneous samples collected, the significant findings were that mirex was present in the two milk samples and the three beef samples (adipose tissue) collected (Table 4). One milk fat sample contained 0.007 ppm of mirex and the other contained 0.016 ppm. The values for mirex in the three beef fat samples were 0.012, 0.113, and 0.042 ppm. The results of the analyses for mirex in beef fat and milk, in addition to results concerning the deer samples, suggest that this

TABLE 2b.—Mirex and DDT residues in adipose tissue of fish and birds collected in Oktibbeha, Noxubee, and Lowndes Counties, Mississippi—1970

SPECIMEN	SAMPLE NUMBER ¹	RESIDUES IN PPM					
		<i>p,p'</i> -DDE	<i>p,p'</i> -DDD	<i>o,p'</i> -DDT	<i>p,p'</i> -DDT	DDTR	MIREX
Fish							
Green sunfish (Bream)	1	0.086	0.010	T	0.016	0.112	0.138
	2	0.068	0.008	0.009	0.024	0.109	0.105
Channel catfish	1	0.579	0.088	0.013	0.087	0.767	11.252
	2	0.211	0.076	0.036	0.058	0.381	5.978
	3(2)	1.170	0.399	0.112	0.559	2.240	3.760
	4(2)	2.382	0.659	0.309	0.783	4.133	2.479
	5	1.953	2.421	1.371	2.189	7.934	—
	6	3.579	2.935	0.753	2.885	10.152	—
	7	1.968	2.021	1.296	2.402	7.687	—
Birds							
Chicken	1	0.067	0.004	—	0.030	0.101	0.087
Bobwhite quail	1	0.073	—	—	—	0.073	0.252
	2	T	—	—	—	T	—
	3	0.014	—	—	—	0.014	0.016
	4	0.141	—	—	T	0.141	0.072
	5	0.074	T	T	0.046	0.120	0.038
	6	0.063	T	T	T	0.063	0.036
	7	0.085	—	—	—	0.085	1.047
	8	0.050	—	—	T	0.050	0.410
	9	0.071	—	—	—	0.071	0.147
	10	0.017	—	—	—	0.017	0.177
	11	0.290	T	T	0.038	0.328	T
	12	0.908	0.015	0.023	0.032	0.987	3.148
	13	0.580	0.015	0.036	0.060	0.515	0.691
	14	0.511	T	T	0.863	1.374	0.843
	15	0.840	0.010	—	0.037	0.887	2.755
	16	0.291	T	T	0.040	0.331	0.292
	17	0.398	0.014	T	0.042	0.454	0.907
	18	0.763	0.024	0.043	0.071	0.901	1.610
	19	0.407	T	T	0.072	0.479	0.717
	20	0.758	T	T	0.092	0.850	1.843
Brown thrasher	1	17.732	0.261	T	1.390	19.122	53.563
	2	28.380	0.176	0.197	2.589	31.342	59.925
	3	60.086	0.337	0.167	4.677	65.267	25.432
	4	33.629	T	T	1.990	35.619	19.978
Blue jay	1	29.779	T	T	2.963	32.742	104.386
	2	8.388	0.077	0.153	0.481	9.029	5.104
	3	50.381	T	T	2.607	52.988	35.720
Turkey	1	0.889	0.036	0.025	0.593	1.543	1.614
	2	2.936	0.110	0.084	2.290	5.420	0.156
	3	0.091	T	T	0.074	0.165	0.014
	4	0.053	0.005	—	0.099	0.157	—
	5	0.471	—	—	0.099	0.570	0.098
	6	0.436	—	—	0.128	0.564	1.207
Eastern kingbird	1	8.068	0.190	T	0.351	8.609	0.436
Robin	1	0.677	0.062	0.030	0.047	0.816	1.002
	2	—	—	—	—	—	—
	3	—	—	—	—	—	—
	4	17.976	0.220	0.559	2.815	21.570	35.138
	5	96.341	1.840	0.924	26.829	125.934	56.536

NOTE: Blank = sample not analyzed; — indicates none detected; T = trace = <0.001 ppm DDT or <0.002 ppm mirex; all results on fresh-weight basis.

¹ Numbers in parentheses indicate composite of two or more fish or birds.

² Samples collected in the Delta.

pesticide may have entered the food chain of human beings. However, additional data involving larger numbers of samples must be collected before any definite statements can be made concerning mirex residues in food products since the possibility of misleading information exists when the number of samples is so small.

When mirex values are compared with DDTR values, it can be seen from Tables 1-4 that in many cases, mirex residue values were found to exceed DDTR values. Among all birds sampled, mirex residues exceeded DDTR residues in 20 of 38 adipose tissues (Table 2b), 10 of 39 liver tissues (Table 2a), and 10 of 37 brain issues (Table 2c); in fish, mirex values exceeded DDTR values in 4 of 9 adipose samples (Table 2b), in 4 of 7

liver samples (Table 2a), and 1 of 5 muscle samples (Table 2d); similarly, in 6 of 25 arthropod samples (Table 3), mirex values exceeded DDTR values. Additional comparisons between DDTR and mirex values can be made from the information presented in the tables, but the above data are sufficient to illustrate the point that mirex has indeed become a prominent pesticide residue in some wildlife, at least in Mississippi. The existence of some mirex levels which exceed DDTR levels is somewhat surprising since the mirex has been applied at rates which represent grams of toxicant per acre, whereas DDT has been applied at a rate of several pounds per acre, although not necessarily in the area studied. DDT has also been used for a much longer period of time than mirex.

TABLE 2c.—Mirex and DDT residues in brain tissue of birds collected in Oktibbeha, Noxubee, and Lowndes Counties, Mississippi—1970

SPECIMEN	SAMPLE NUMBER	RESIDUES IN PPM				DDTR	MIREX
		<i>p,p'</i> -DDF	<i>p,p'</i> -DDD	<i>o,p'</i> -DDT	<i>p,p'</i> -DDT		
Chicken	1	0.049	—	—	—	0.049	0.029
Bobwhite quail	1	—	0.002	—	—	0.002	—
	2	0.014	—	—	—	0.014	T
	3	0.028	0.003	0.007	0.015	0.053	0.012
	4	0.009	—	—	0.013	0.022	—
	5	0.010	—	—	—	0.010	—
	6	0.226	T	T	0.052	0.278	0.045
	7	0.010	—	—	—	0.010	—
	8	0.015	—	—	—	0.015	—
	9	0.008	—	—	—	0.008	—
	10	0.008	—	—	—	0.008	—
	11	—	—	—	—	—	—
	12	—	—	—	—	—	—
	13	0.012	T	T	T	0.012	T
14	0.036	T	T	T	0.036	T	
15	0.029	T	T	T	0.029	0.093	
16	0.008	—	—	—	0.008	T	
17	0.008	T	T	T	0.008	T	
18	0.017	T	T	T	0.017	—	
19	0.019	—	—	—	0.019	—	
20	0.019	—	—	—	0.019	0.067	
Brown thrasher	1	0.595	—	—	0.043	0.638	0.318
	2	0.244	—	—	—	0.244	0.429
	3	1.256	—	—	0.086	1.342	1.057
	4	0.281	—	—	0.022	0.303	0.374
Blue jay	1	0.486	—	—	—	0.486	1.195
	2	0.118	—	—	0.011	0.129	0.099
	3	0.396	—	0.010	0.030	0.436	1.513
Meadow lark	1	0.080	0.002	0.003	0.016	0.101	0.462
	2	0.338	T	T	0.031	0.369	0.524
Turkey	1	0.009	—	—	—	0.009	—
	2	—	—	—	—	—	—
	3	0.015	—	0.002	0.008	0.025	—
	4	—	—	—	—	—	—
	5	—	—	—	—	—	—
	6	0.031	—	0.002	0.008	0.041	0.028
Eastern kingbird	1	1.305	0.016	—	0.071	1.392	0.029
Robin	1	—	—	—	—	—	—
	2	1.381	0.698	0.078	2.197	4.354	0.197
	3	9.819	1.238	0.138	2.521	13.716	0.172
	4	0.171	0.032	—	0.031	0.234	0.404
	5	1.057	0.033	0.039	0.180	1.309	0.572
Barred owl	1	1.651	0.010	—	0.032	1.703	1.820

NOTE: Blank = samples not analyzed; — indicates none detected; T = trace = <0.001 ppm DDT or <0.002 ppm mirex; all results on fresh-weight basis.

TABLE 2d.—Mirex and DDT residues in heart and muscle tissue of fish and birds collected in Oktibbeha, Noxubee, and Lowndes Counties, Mississippi—1970

SPECIMEN	SAMPLE NUMBER ¹	TISSUE	RESIDUES IN PPM					
			p,p'-DDD	p,p'-DDE	o,p'-DDT	p,p'-DDT	DDTR	MIREX
Fish								
Channel catfish	3(2)	Muscle	—	0.016	0.026	0.030	0.072	0.060
	4(2)	Muscle	—	0.024	—	—	0.024	0.079
	5	Muscle	0.118	0.105	0.083	0.142	0.448	—
	≠6	Muscle	0.121	0.219	0.037	0.160	0.537	—
	≠7	Muscle	0.327	0.251	0.289	0.449	1.316	—
Birds								
Meadow lark	1	Heart	0.004	0.113	0.005	0.041	0.163	1.058
	2	Heart	T	0.911	T	0.045	0.956	1.931
Turkey	5	Heart	—	0.018	0.002	0.010	0.030	0.020
	6	Heart	—	0.636	—	0.117	0.753	0.127
	7	Heart	—	0.029	—	T	0.029	0.229
Barred owl	1	Muscle	0.005	1.036	—	0.020	1.061	0.934

NOTE: — indicates none detected; T = trace = <0.001 ppm DDT or <0.002 ppm mirex; all results on fresh-weight basis.

¹ Numbers in parentheses indicate composite of two or more samples.

² Samples collected in the Delta.

TABLE 3.—Mirex and DDT residues in arthropods collected in Oktibbeha County, Mississippi—1970

SPECIMEN	NUMBER OF INDIVIDUALS PER SAMPLE	RESIDUES IN PPM					
		p,p'-DDD	p,p'-DDE	o,p'-DDT	p,p'-DDT	DDTR	MIREX
Praying mantis	3	0.001	0.008	0.003	0.010	0.022	0.008
Walking sticks	4	0.002	0.003	0.004	0.006	0.015	0.021
Stink bugs	19	0.004	0.061	0.018	0.048	0.131	—
Lady bugs	15	0.012	0.692	0.039	0.052	0.793	—
Beetles	17	0.003	0.033	0.020	0.048	0.131	—
Spiders	5	0.040	0.368	0.336	0.308	1.052	1.219
Spider	1	—	0.017	0.019	0.013	0.049	0.065
Spider	1	—	0.010	0.014	0.016	0.040	0.267
Spider	1	—	0.015	0.020	0.029	0.064	0.189
Katydid	2	—	0.011	0.019	0.029	0.059	—
Katydid	2	—	0.007	0.019	0.017	0.043	—
Cricket	2	—	0.013	0.016	0.052	0.081	0.008
Cricket	2	0.049	0.049	0.013	0.240	0.351	0.030
Cricket	2	0.007	0.010	0.016	0.022	0.055	—
Cricket	2	—	0.012	0.011	0.031	0.054	—
Cricket	2	—	0.012	0.042	0.042	0.075	0.026
Cricket	2	—	—	—	—	—	0.075
Cricket	2	—	0.005	0.022	0.017	0.044	—
Cricket	2	—	—	—	—	—	—
Cricket	2	—	0.006	0.009	—	0.015	—
Cricket	2	0.007	0.012	—	—	0.019	—
Grasshopper	1	—	—	0.034	0.033	0.067	—
Grasshopper	2	—	—	0.010	0.019	0.029	—
Grasshopper	2	0.021	—	0.030	0.019	0.049	—
Grasshopper	4	0.077	0.011	0.012	0.015	0.038	—

NOTE: — indicates none detected; all results on fresh-weight basis.

It is difficult at the present time to assess the implications of the results reported in the present paper. Lacking in most reports to date is any information pertaining to the buildup of mirex in animals following treatment with mirex and what toxic effects, if any, can be expected at various tissue levels. A number of LD₅₀ values have been established for mirex and these values indicate that mirex is rather low in its toxicity to animals. Gaines and Kimbrough (6), for example, in a study of the toxicity of mirex to rats found the single dose LD₅₀ to

be 365 mg/kg for adult female rats. The 90-dose LD₅₀, the amount daily given in a single dose for 90 days which will result in 50% mortality, was reported to be 6 mg/kg. Earlier, Gaines (7) found that when corn oil was used as the carrier, the oral LD₅₀ for mirex in male and female rats was 740 and 600 mg/kg, respectively. Mirex administered in a peanut oil solution resulted in a LD₅₀ value greater than 3,000 mg/kg, in males and females alike. The pronounced difference in the LD₅₀ values found, based on which carrier was used, has not been explained.

TABLE 4.—Mirex and DDT residues in miscellaneous samples collected in Lowndes and Oktibeha Counties, Mississippi—1970

SPECIMEN	SAMPLE NUMBER ²	RESIDUES IN PPM					MIREX
		p,p'-DDD	p,p'-DDE	o,p'-DDT	p,p'-DDT	DDTR	
Cattle egret eggs	1	0.056	0.759	0.017	0.089	0.921	1.555
	2	0.011	0.834	0.013	0.044	0.902	0.035
	3	0.012	1.944	—	0.014	2.070	0.285
	4	0.024	1.769	0.014	0.353	2.160	0.055
	5	0.068	2.605	0.042	0.230	2.945	0.277
	6	0.006	0.655	0.011	0.036	0.708	0.618
	7	0.018	0.302	0.022	0.139	0.481	0.073
Blue heron eggs	1	0.084	1.561	0.031	0.605	2.281	0.051
	2	0.049	0.511	0.097	0.239	0.896	0.166
	3	0.053	3.479	0.406	0.517	4.455	0.316
	4	0.012	0.220	0.017	0.062	0.311	T
	5	0.010	0.369	0.017	0.059	0.455	0.083
	6	0.075	1.139	0.098	0.562	1.874	0.694
Cow's milk (lipid basis)	1	0.003	0.013	T	0.007	0.023	0.007
	2	—	0.007	—	—	0.007	0.016
	3	0.056	0.291	—	0.054	0.401	—
Earthworms	1	—	0.005	—	—	0.005	0.030
	2(10)	0.005	0.005	T	0.008	0.018	0.076
Beef adipose	1	0.013	0.134	—	0.067	0.214	0.012
	2	0.023	0.118	—	0.074	0.215	—
	3	0.035	0.868	—	0.068	0.971	0.113
	4	0.021	0.227	—	0.055	0.303	0.042
	5	0.156	1.324	0.019	0.410	1.909	—
Silage (dry weight)	1	0.015	0.011	—	—	0.026	—
	2	0.016	0.011	—	—	0.027	—
Fescue (dry weight)	1	0.003	0.010	0.012	0.037	0.062	—

NOTE: — indicates none detected; T = trace = < 0.001 ppm DDT or < 0.002 ppm mirex, all results on fresh-weight basis unless otherwise noted. Numbers in parentheses indicate composite of two or more samples.

Ware and Good (8) found that a rather low level of mirex in the diet (10 ppm) of mice resulted in 100% mortality by 60 days. This report also showed that 5 ppm of mirex increased parent mortality and decreased litter size.

In addition to mortality data on mammalian species, some evidence has been accumulated which suggests that toxic effects can be produced in mammals following mirex ingestion. Gaines and Kimbrough found that female rats fed 25 ppm of mirex exhibited ultrastructural changes in liver tissue (6). Also, some offspring from females fed 25 ppm for 102 days developed cataracts; the incidence of cataracts at this level, which correspond to an average intake of 2.3 mg/kg day, was 46%.

In relating the findings reported for avian species in the current report to previous work, one must rely on amounts of mirex reported which cause mortality. Toxicity data on a number of different species of birds are given in Circular 199 of the Fish and Wildlife Service (9). Data in that publication show that 12% mortality was produced in quail fed 300 ppm for 111 days; young mallards fed a diet containing 500 ppm of mirex experienced a maximum of 81% mortality in 30 days; 20% mortality was observed in pheasants fed a diet which contained 200 ppm of mirex for 30 days;

and 8% of a group of cowbirds fed 500 ppm of mirex died after 30 days.

In addition to mortality data, it has been shown that when mirex (600 ppm) was fed to laying hens over a 16-week period, the hens lost weight and the hatchability of eggs and survival of chicks was reduced (10). Based on the high amounts of mirex required to reach an LD₅₀ or to produce toxic effects, it would not appear that the levels of mirex observed in animals sampled in the current investigation represent a potential hazard.

The most relevant information reported regarding effects of mirex in fish is that provided by Van Valin *et al.* (11). These workers showed that lesions in gills and kidneys of goldfish occurred following feeding of mirex. Tissue levels of mirex in goldfish at completion of the experiment ranged from 0 to 1,350 ppm in liver tissue and ranged from 20.8 to 232 ppm in muscle tissue. These fish had been exposed to 1.0 ppm mirex in water for 224 days. The levels reported by Van Valin *et al.* which resulted in histological changes in fish are considerably higher than the values for mirex residues found for the same tissues in the limited number of fish samples in the current investigations.

To summarize the above discussion, it appears that for the most part mirex is low in its toxicity to animals based

on LD₅₀ values. However, rather low amounts of this material in the diet of some animals, mice for example, can produce mortality. Because of the lack of information regarding tissue levels of mirex encountered with the feeding of LD₅₀ dosages, it is not possible to interpret the meaning of the levels observed in samples discussed in this report, although some previous data on fish indicate that the values reported in the current study do not represent toxic levels.

Acknowledgment

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See Appendix for chemical names of compounds discussed in this paper.

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Chemical Residues in Lake Erie Fish—1970-71¹

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ABSTRACT

Yellow perch, coho salmon, carp, channel catfish, freshwater drum, and white bass from the Ohio shore of Lake Erie were analyzed during 1970-71 for residues of chlorinated pesticides (DDE, TDE, DDT, and dieldrin), polychlorinated biphenyls (PCB's), and mercury. All but 1 of the 80 samples analyzed contained DDT and/or its metabolites; PCB's were found in all samples. Fifty-three of the 80 samples were analyzed for mercury, and all were found positive.

Average levels of residues for the species sampled ranged from 0.06 to 0.42 ppm for DDE; 0.07 to 0.52 ppm, TDE; 0.03 to 0.25 ppm, DDT; 0.18 to 0.90 ppm, total DDT; 0.01 to 0.07 ppm, dieldrin; 0.08 to 4.4 ppm, PCB's; and 0.12 to 0.64 ppm, mercury. The highest average residue levels of total DDT were in coho salmon and channel catfish. Average levels of PCB's were significantly higher in channel catfish, and levels of mercury were significantly higher in white bass.

Introduction

In the spring of 1970 the Cincinnati District of the U. S. Food and Drug Administration began monitoring yellow perch (*Perca flavescens*) and, in cooperation with the Ohio Department of Natural Resources, Division of Wildlife, coho salmon (*Oncorhynchus kisutch*) to determine the extent of pesticide contamination in these species in Lake Erie. The next spring (1971) monitoring was broadened to include white bass (*Roccus chrysops*), freshwater drum (*Aplodinotus grunniens*), channel catfish (*Ictalurus punctatus*), and carp (*Cyprinus carpio*). These six species were selected because they had the greatest commercial significance and/or greatest potential for residues due to their predatory eating habits and size. The investigation of these species was limited to the Ohio shore of Lake Erie, since this was the legal boundary of Cincinnati District of FDA.

Prior to the expansion of the program (1970), the coho

salmon and yellow perch (15 and 12 samples, respectively) were analyzed for DDT and its isomers and metabolites, dieldrin, and PCB's (calculated as Aroclor[®] 1254); after the expansion (1971), all species were analyzed for mercury as well.

Sampling Procedures

The coho salmon, collected by the Ohio Division of Wildlife, were random samples and represented catches from almost the entire Ohio shore of Lake Erie (Fig. 1); the other species were collected by Food and Drug inspectors at commercial fisheries in Ohio. The coho salmon represent catches on a year-round basis, while the other species were obtained during the fishing season on Lake Erie, a period from latter March to early October. Each coho sample consisted of 1 or 2 fish and the other samples consisted of 5 to 10 fish.

On collection, the samples were frozen and shipped to the laboratory in Cincinnati. At the laboratory, the samples were thawed, and the heads, viscera, and scales removed. The rest of the fish was then thoroughly ground and mixed in a meat grinder.

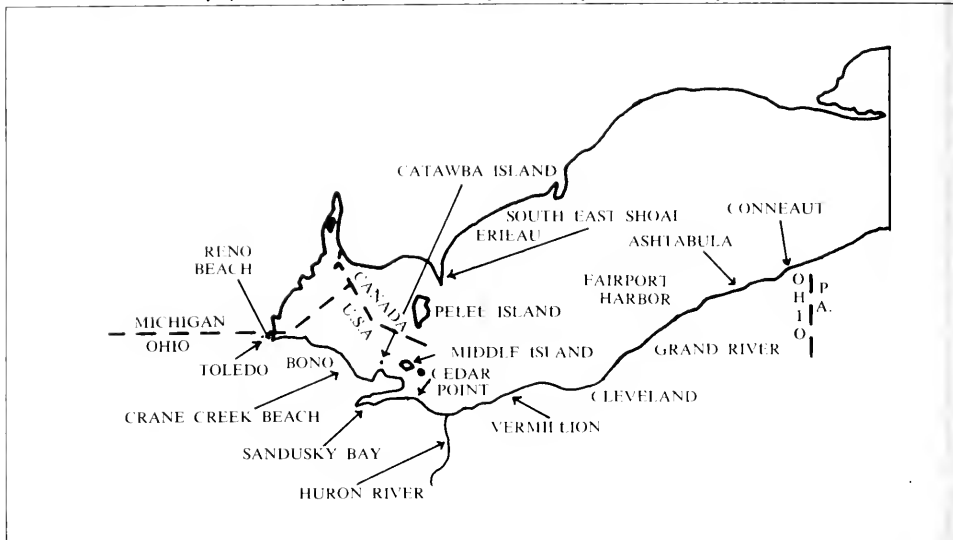
Analytical Procedures

ORGANOCHLORINES AND PCB'S

The method employed for extraction and cleanup of samples to determine DDT residues, dieldrin, and PCB's was that described by Porter, Young, and Burke (6). The procedure involved dehydrating the sample with sodium sulfate and then isolating the fat by blending 50 g of the sample three times with petroleum ether. The fat was partitioned with acetonitrile and the residues isolated by a Florisil column, eluted with 6% and 15% ethyl petroleum ether. The 15% eluate which contained any residues of dieldrin was additionally cleaned up using alkaline saponification (4). PCB residues in the

¹ From the U. S. Food and Drug Administration, 1141 Central Parkway, Cincinnati, Ohio 45202.

FIGURE 1.—Map of Ohio Shore of Lake Erie showing collection points of fish samples analyzed—1970-71



6% eluate were isolated from DDT residues by using the silicic acid column method described by Armour and Burke (1).

A Barber-Coleman Model 10 gas-liquid chromatograph (GLC), equipped with a 150-mc tritium electron capture detector, was used for all quantitative assays (4). Two columns were used: (1) 10% DC-200 on Chromosorb W/HP and (2) a 1 to 1 mixture of 10% DC-200 and 15% QF-1 on Chromosorb W/HP. Parameters were those given in the FDA Pesticide Analytical Manual (3).

Confirmation tests for the chlorinated pesticide residues consisted of thin layer chromatography (3) of 10 randomly selected samples. PCB residues were confirmed by their stability to alkaline saponification and by micro-coulometric GLC using a chlorine specific detector (3). Quantitation of pesticide residues was accomplished using peak height. PCB residues in the fish had a pattern most similar to Aroclor[®] 1254 and were calculated by comparing the entire area of the PCB pattern with the area of Aroclor[®] 1254. Standards were supplied by the FDA Pesticide Reference Standard Section.

Recovery data for the above procedure provided by Carr in a collaborative study (2) were *p,p'*-DDE, 87.5 ± 13.5%; *p,p'*-TDE, 88.6 ± 11.4%; *p,p'*-DDT, 90.6 ± 9.7%; and dieldrin 82.2 ± 25.4%.

MERCURY

Mercury was determined using the method described and collaborated by Munns and Holland (5). The method

employs a perchloric acid digestion and subsequent determination by "cold vapor" atomic absorption. The instrument was a Perkin-Elmer Model 303 AA with a 17-cm cell length in a closed system. The 2533 A absorption line was used. The average recovery for samples fortified with methyl mercury chloride was 85.4%.

Results

Table 1 lists the individual residue levels for each fish sample in the study by species; Table 2 summarizes these results. These data were not corrected for percent recovery, and the minimum level recorded in all determinations was 0.01 ppm; all residues are reported on a wet-weight basis. Average residues of total DDT were highest in coho salmon (0.90 ppm) and catfish (0.89 ppm); however, none of the samples approached the FDA guideline of 5 ppm. Average dieldrin residue: were highest in coho salmon (0.07 ppm), although again the levels were low. Polychlorinated biphenyl residue: were significantly higher in channel catfish (average 4.4 ppm). White bass continued the highest average level of mercury with residues in five of the six white bass samples examined in excess of FDA's 0.5 ppm guideline. In general, the species lowest in the food chain, yellow perch, and the "rougher" fish (fish that are not sport fish or of significant food value), carp and freshwater drum contained the lowest amounts of chemical residues.

See Appendix for chemical names of compounds discussed in this paper.

TABLE 1.—Chemical residue levels in six species of fish, Lake Erie—1970-71

LOCATION OF CATCH	DATE COLLECTED ¹	RESIDUES IN PPM						
		DDE	TDE	DDT	TOTAL DDT	DIELDRIN	PCB'S	MERCURY
COHO SALMON								
Sandusky Bay (I)	4/1/70	0.39	0.15	0.25	0.79	0.06	1.6	
Sandusky Bay (M)	4/1/70	0.38	0.28	—	0.66	0.06	1.7	
Sandusky Bay (I)	4/1/70	0.34	0.10	0.31	0.75	0.05	1.6	
NNW Toledo (M)	5/18/70	0.32	0.28	0.18	0.78	0.07	1.3	
NNW Toledo (M)	5/18/70	0.32	0.15	0.37	0.84	0.06	1.0	
NNW Toledo (M)	5/18/70	0.43	—	0.67	1.10	0.08	2.2	
East Pelee Island (M) ²	6/10/70	0.62	0.45	0.23	1.30	0.08	2.6	
East Pelee Island (M)	6/10/70	0.53	0.32	0.23	1.09	0.06	1.6	
Southeast Shoal	6/30/70	0.72	0.21	0.38	1.31	0.06	3.2	
Southeast Shoal	6/30/70	0.64	0.25	0.50	1.39	0.06	3.2	
Southeast Shoal (I)	6/30/70	0.55	0.42	0.11	1.08	0.05	4.3	
NE Erieau Ontario (M)	7/28/70	0.39	0.25	0.04	0.68	0.05	1.5	
NE Erieau Ontario (M)	7/28/70	0.88	0.50	0.07	1.45	0.15	3.1	
NE Erieau Ontario (M)	7/28/70	0.60	0.42	0.15	1.17	0.10	3.8	
W Erieau Ontario (M)	7/30/70	0.90	0.30	0.49	1.69	0.14	3.2	
NE Fairport Harbor (M)	8/1/70	0.53	0.23	0.50	1.26	0.07	2.2	0.42
NE Fairport Harbor (M)	8/11/70	0.27	0.20	0.24	0.71	0.06	1.6	0.59
NE Fairport Harbor (M)	8/11/70	0.31	0.18	0.06	0.55	0.03	1.3	0.39
N Conneaut (M)	9/2/70	0.04	0.46	0.13	0.63	0.09	3.8	0.58
N Conneaut (M)	9/2/70	0.06	0.25	0.61	0.91	0.06	1.5	0.21
N Ashtabula (I)	9/2/70	0.17	0.17	0.44	0.78	0.05	1.10	0.11
N Conneaut (I)	9/3/70	—	0.10	0.11	0.21	0.04	1.2	0.14
W Huron (M)	10/12/70	0.46	0.20	0.08	0.74	0.03	1.3	0.47
W Huron (M)	10/12/70	0.42	0.24	0.11	0.78	0.04	1.9	0.36
W Huron (M)	10/12/70	0.47	0.16	0.26	0.89	0.03	1.2	0.20
Huron River Dam (M)	11/4/70	0.47	0.22	0.10	0.79	0.04	2.9	0.35
Huron River Dam (M)	12/8/70	0.37	0.21	0.07	0.65	0.03	2.6	0.41
Sandusky Bay (I)	4/1/71	0.28	0.16	0.11	0.53	0.07	1.1	0.24
Bono (M)	4/23/71	0.50	—	0.50	1.00	0.10	1.4	0.12
Middle Island (M)	6/14/71	0.21	0.22	0.07	0.50	0.07	2.6	0.36
YELLOW PERCH								
North of Cleveland	4/13/70	0.04	0.03	—	0.09	0.02	0.5	
North of Fairport	4/19/70	—	—	—	—	—	0.26	
Sandusky Bay	4/30/70	0.05	0.05	0.05	0.15	0.02	0.6	
Catawba Island	5/13/70	0.08	0.06	—	0.14	—	1.1	
ENE of Toledo	5/16/70	0.08	0.06	0.07	0.21	—	0.8	
Crane Creek-Toledo	5/17/70	0.09	0.07	—	0.16	—	1.2	
NW Conneaut	5/18/70	0.17	0.05	0.13	0.35	—	0.8	
NE Cleveland	5/18/70	0.08	0.05	0.11	0.24	—	0.8	
East of Conneaut	6/15/70	0.09	0.03	0.05	0.17	—	0.5	
North of Cleveland	6/16/70	0.10	0.08	0.06	0.24	—	0.8	
East of Reno Beach	6/16/70	0.10	0.11	—	0.21	—	1.3	
East of Reno Beach	7/14/70	0.14	0.15	0.05	0.34	0.02	0.9	
East of Cedar Point	8/18/70	0.02	0.04	—	0.09	—	0.2	0.15
North of Conneaut	8/26/70	0.06	0.08	0.20	0.34	—	0.5	0.22
East of Toledo	9/15/70	—	0.19	0.11	0.30	—	2.4	0.37
NW of Vermillion	10/5/70	0.06	0.05	0.04	0.15	0.02	0.6	0.25
Ashtabula	10/7/70	0.04	0.05	0.07	0.16	0.02	0.2	0.16
East of Toledo	10/8/70	0.08	0.10	0.02	0.20	0.01	0.9	0.40
Grand River	11/18/70	0.03	0.04	—	0.07	—	0.7	0.13
Sandusky Bay	3/29/71	0.05	0.07	0.02	0.14	0.01	0.8	0.25
ENE of Toledo	3/30/71	0.06	0.10	0.01	0.17	0.01	0.8	0.45
Conneaut	5/11/71	0.02	0.04	0.03	0.09	0.01	0.4	0.16
Vermillion	5/13/71	0.05	0.03	0.03	0.11	0.01	0.5	0.19
WHITE BASS								
Cedar Point	8/11/70	0.24	0.27	0.13	0.64	0.06	2.1	0.62
East of Toledo	9/15/70	0.02	0.30	0.05	0.37	0.03	3.5	0.70
Sandusky Bay	9/23/70	0.26	0.30	0.31	0.87	0.03	1.9	0.60
East of Toledo	10/12/70	0.11	0.22	0.10	0.43	0.03	1.4	0.58
Vermillion	3/29/71	0.33	0.34	0.06	0.73	0.06	2.4	0.86
ENE of Toledo	3/30/71	0.11	0.21	0.02	0.34	0.03	1.6	0.45

TABLE 1.—Chemical residue levels in six species of fish, Lake Erie—1970-71—Continued

LOCATION OF CATCH	DATE COLLECTED ¹	RESIDUES IN PPM						
		DDE	TDE	DDT	TOTAL DDT	DIELDRIN	PCB'S	MERCURY
CARP								
East of Cedar Point	8/18/70	0.04	0.08	—	0.12	—	0.4	0.07
East of Toledo	9/15/70	0.02	0.06	0.02	0.10	—	0.3	0.11
Sandusky Bay	9/23/70	—	0.39	0.12	0.51	0.02	0.9	0.16
East of Toledo	10/8/70	0.14	0.24	0.04	0.38	0.05	4.3	0.08
Vermillion	3/13/71	0.61	0.26	0.04	0.91	0.06	5.3	0.13
Vermillion	3/20/71	0.11	0.21	—	0.32	0.04	1.1	0.17
ENE of Toledo	3/30/71	0.13	0.32	—	0.45	0.04	2.3	0.12
CHANNEL CATFISH								
East of Cedar Point	8/18/70	0.53	0.66	0.12	1.41	0.10	3.9	0.31
East of Toledo	9/15/70	0.07	0.85	0.26	1.18	0.05	4.7	0.45
Sandusky Bay	9/23/70	0.04	0.09	0.09	0.22	0.01	1.4	0.14
East of Toledo	10/8/70	0.43	0.65	0.13	1.21	0.07	4.2	0.72
ENE of Toledo	3/30/71	0.28	0.46	0.06	0.80	0.07	6.2	0.48
Sandusky Bay	4/23/71	0.38	0.33	0.06	0.77	0.04	2.4	0.32
Vermillion	5/12/71	—	0.59	0.15	0.74	0.04	7.8	0.49
FRESHWATER DRUM								
Cedar Point	8/18/70	0.08	0.11	—	0.19	—	0.8	0.40
East of Toledo	9/15/70	0.12	0.19	0.09	0.40	—	1.4	0.45
Sandusky Bay	9/23/70	—	0.18	0.02	0.10	0.02	0.6	0.21
East of Toledo	10/8/70	0.17	0.18	0.12	0.47	0.02	1.3	0.37
Vermillion	3/29/71	0.08	0.10	0.04	0.22	0.04	0.8	0.24
ENE of Toledo	3/30/71	0.09	0.15	0.02	0.26	0.03	1.2	0.43
Vermillion	5/12/71	0.17	0.11	0.15	0.43	0.05	1.5	0.29

NOTE: — = not detected; blank = not analyzed

(M) = Mature fish; (I) = Immature fish.

¹ Although the monitoring program was not expanded to include carp, channel catfish, freshwater drum, and white bass until 1971, some of the analyses were done on samples collected in 1970.

² Although sample was found to be a chinook salmon, it was not eliminated from the study.

TABLE 2.—Average and range of chemical residue levels in six species of fish, Lake Erie—1970-71

SPECIES	NUMBER OF SAMPLES	RESIDUES IN PPM													
		DDE		TDE		DDT		TOTAL DDT		DIELDRIN		PCB'S		MERCURY	
		AVG.	RANGE	AVG.	RANGE	AVG.	RANGE	AVG.	RANGE	AVG.	RANGE	AVG.	RANGE	AVG.	RANGE
Coho salmon	30-15 ¹	0.42	0.04-0.90	0.24	0.00-0.50	0.25	0.00-0.67	0.90	0.21-1.69	0.07	0.03-0.15	2.1	1.0-4.3	0.32	0.11-0.5
Yellow perch	23/11 ¹	0.06	0.00-0.17	0.07	0.00-0.19	0.05	0.00-0.20	0.18	0.07-0.35	0.01	0.00-0.02	0.8	0.2-2.4	0.25	0.13-0.4
White bass	6	0.18	0.02-0.33	0.27	0.21-0.34	0.11	0.02-0.31	0.56	0.34-0.87	0.04	0.03-0.06	2.1	1.4-3.5	0.64	0.45-0.8
Carp	7	0.15	0.00-0.61	0.22	0.06-0.39	0.03	0.00-0.12	0.40	0.10-0.91	0.04	0.00-0.06	2.0	0.3-5.3	0.12	0.07-0.1
Channel catfish	7	0.25	0.00-0.53	0.52	0.09-0.85	0.12	0.06-0.26	0.89	0.22-1.31	0.05	0.01-0.10	4.4	1.4-7.8	0.42	0.14-0.7
Freshwater drum	7	0.10	0.00-0.17	0.13	0.08-0.19	0.06	0.00-0.15	0.30	0.10-0.47	0.02	0.00-0.05	1.1	0.6-1.5	0.34	0.21-0.4

¹ Total samples/samples analyzed for mercury

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Mercury and Lead Residues in Starlings—1970¹

William E. Martin

ABSTRACT

Starling (*Sturnus vulgaris*) samples from 125 randomly selected sites were analyzed for mercury residues. Except for three locations, all samples had residues well below 0.5 ppm. Lead residues were identified in all samples from 23 survey sites that had been tentatively selected to evaluate use of starlings as a biological measure of environmental contamination by lead. Residues of lead ranged from 0.4 to 13.3 ppm with a mean of 3.18 ppm and a standard error of 0.62 ppm.

Introduction

The starling (*Sturnus vulgaris*) has been used since 1967 by the Bureau of Sport Fisheries and Wildlife as an indicator species for measuring occurrence and relative amounts of selected persistent environmental contaminants. The overall wildlife monitoring scheme, the randomized nationwide sampling design, and analytical methods used for determining residue levels of persistent organochlorine insecticides, mercury, and lead in starling tissue are described by Dustman *et al.* (1) and Martin (2).

Determination of mercury residues in starlings is conducted to aid in evaluation of the relative distribution of this element in terrestrial fauna other than game birds. Concern over possible widespread environmental contamination by lead provided the impetus for the preliminary survey of lead residues in starlings. The starling was selected because it is present throughout the contiguous States in both urban and rural areas, it is omnivorous in its feeding habits, and normally is not exposed to lead shotgun pellets.

This report presents lead and mercury residue data gathered from starling monitoring collections made during November and December 1970.

Sampling Procedures

MERCURY

A randomized, nationwide sampling design was used to obtain birds for mercury analysis. This standard design allows for sampling at up to four randomly selected sites within each of 40 five-degree blocks drawn from 24 to 49 latitude and 64 to 124 longitude, as shown in Fig. 1. Sampling locations are identified by a row number, a column letter, and a site number; e.g., the site near Tacoma, Wash., is designated 1-A-1. Tissues used for mercury analysis in the study reported here were taken from the same bird samples used for persistent organochlorine insecticide analyses reported separately by Martin and Nickerson (3). Collections were successful at 125 of the planned 139 sites (Table 1).

LEAD

A total of 25 sampling sites were selected to reflect the degree of lead residues expected from man's activities and related pollution sources, such as a high incidence of industrialization, heavy automobile traffic, etc. Samples were not obtained from two critical preselected locations—Los Angeles, Calif., a site which was expected to reflect relatively high lead residue, and Carlsbad, N. Mex., predicted to reflect relatively low lead residues. Selected sampling sites are listed in Table 2 with results of lead analyses.

As described by Martin and Nickerson (3), each sample normally consisted of a "pool" of 10 birds. "Pools" containing fewer than 10 birds are indicated by footnote in the appropriate tables. Birds analyzed for mercury residues had been either trapped or shot. Starlings collected for lead analysis were taken by means other than lead shot or poison. The bird "pools" were wrapped in aluminum foil, placed in polyethylene bags, and frozen immediately for later laboratory analyses.

¹ From the Pesticide Appraisal and Monitoring Branch, Division of Wildlife Services, Bureau of Sport Fisheries and Wildlife, U. S. Department of the Interior, Washington, D. C. 20240.

FIGURE 1.—Starling monitoring sites—1970

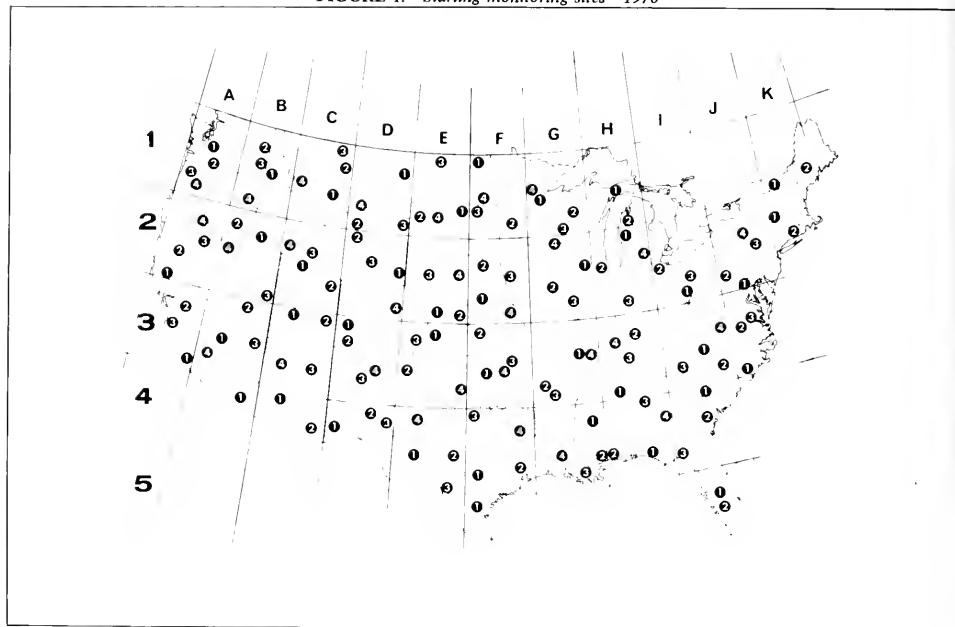


TABLE 1.—Mercury and lead sampling site locations, 1970

STATE	CITY OR COUNTY	SAMPLING SITE NUMBER 1,2	STATE	CITY OR COUNTY	SAMPLING SITE NUMBER 1,2
Alabama	Marion	3-H-1	Georgia	Pike	4-H-4
	Talladega	4-H-3		Wayne	4-I-2
Arizona	Navajo	3-C-3		Atlanta (Lead)	19
	Yavapi	3-C-4	Idaho	Nezperce	1-B-1
	Maricopa	4-C-1		Owyhee	2-B-1
	Graham	4-C-2		Franklin	2-C-3
	Phoenix (Lead)	7		Minidoka	2-C-4
Arkansas	Yell/Pope	3-G-2		Boise (Lead)	5
	Lonoke/Pulaski	3-G-3	Illinois	Stephenson	2-G-1
	Stuttgart (Lead)	16		Sangamon	2-G-3
California	Colusa	2-A-1		Cook	2-H-2
	Shasta	2-A-2		Chicago (Lead)	13
	Modoc	2-A-3	Indiana	Henry	2-H-3
	Ventura	3-A-1	Iowa	Pottawattamie	2-F-3
	Stanislaus	3-A-2		Polk	2-G-2
	Monterey	3-A-3		Butler	2-G-4
	Inyo	3-B-1	Kansas	Rawlins	2-E-1
	Kern	3-B-4		Smith	2-E-2
	Imperial	4-B-1		Hamilton & Kearny	3-E-1
	Los Angeles (Lead)	3		Nemaha	2-F-4
Colorado	Adams	2-D-4		Marion	3-F-2
	Montrose	3-D-1	Kentucky	Ohio	3-H-2
	La Plata & Rio Grande	3-D-2		Hopkins	3-H-4
	Otero	3-E-3	Louisiana	Jefferson	4-G-3
	Greteley (Lead)	8		Rapides	4-G-4
Connecticut	New London	2-K-2		Baton Rouge (Lead)	17
Florida	Bay	4-H-1	Maine	Penobscot	1-K-2
	Madison	4-I-3		Gray (Lead)	25
	Polk	5-I-1	Maryland	Prince Georges	2-J-1
	Hardec	5-I-2		Patuxent (Lead)	22
	Gainesville (Lead)	20			

TABLE 1.—Mercury and lead sampling site locations, 1970—Continued

STATE	CITY OR COUNTY	SAMPLING SITE NUMBER ^{1,2}	STATE	CITY OR COUNTY	SAMPLING SITE NUMBER ^{1,2}
Michigan	Chippewa	1-H-1	Ohio (Cont'd.)	Sandusky (Lead)	14
	Grand Traverse	1-H-2		Columbus (Lead)	15
	Kent	2-H-1	Oklahoma	Greer	3-E-4
	Inaham	2-H-4		Canadian	3-F-1
Minnesota	Swift	1-F-2	Snowata	3-F-3	
	Pine	1-G-1	Oklmulgee	3-F-4	
	Aitkin	1-G-4	Tishomingo (Lead)	10	
Mississippi	Twin Cities (Lead)	12	Oregon	Yamhill	1-A-3
	Leake	4-G-1		Lane	1-A-4
	Harrison	4-G-2		Klamath	2-A-4
Jackson	4-H-2	Baker		1-B-4	
Missouri	Stoddard	3-G-1		Harney	2-B-2
	Bollinger	3-G-4	Corvallis (Lead)	2	
Montana	Meagher	1-C-1	Pennsylvania	Somerset	2-J-2
	Missoula	1-C-4		Cuzerne	2-J-3
	Richland	1-D-1		South Carolina	Aiken
	Yellowstone	1-D-4	South Dakota		Potter
Nebraska	Keith	2-E-3		Hughes	1-E-4
	Lincoln	2-E-4		Brown	1-F-3
	Clay	2-F-1		Mitchell (Lead)	11
	Antelope	2-F-2	Tennessee	Davidson	3-H-3
Nevada	White Pine	2-B-3		Nashville (Lead)	18
	Humboldt	2-B-4	Texas	Clay	4-F-3
	Nye	3-B-2		Morris	4-F-4
	Clark	3-B-3	Utah	Weber	2-C-1
	Reno (Lead)	4		Sevier Millard	3-C-1
New Mexico	Bernalillo	3-D-3		Salt Lake City (Lead)	6
	Santa Fe & Torrance	3-D-4	Vermont	Addison	1-K-1
	Luna	4-D-1		Virginia	Amherst
	Chaves	4-D-3	Prince George		3-J-2
	Quay	3-E-2	Caroline		3-J-3
Carlsbad (Lead)	9	Washington	Pierce	1-A-1	
New Jersey	N. Brunswick (Lead)		23	Yakima	1-A-2
	New York		Oswego	2-J-4	Spokane
Rensselaer			2-K-1	Whitman	1-B-3
Jamestown (Lead)		24	Yakima (Lead)	1	
North Carolina	Wilkes	3-I-1	Wisconsin	Curtiss	1-G-2
	Union	3-I-2		Trempleau	1-G-3
	Macon	3-I-3	Wyoming	Big Horn	1-D-2
	Pender	3-J-1		Brook	1-D-3
Raleigh (Lead)	21	Goshen		2-D-1	
North Dakota	Ward	1-E-3	Washakie	2-D-2	
	Cavelier	1-F-1			
	Dickey	1-F-4			
Ohio	Washington	2-I-1			
	Erie	2-I-2			
	Jefferson	2-I-3			

¹Mercury sampling sites are described by the counties in which collections were taken and are identified by a three place site number (e.g., 1-A-1).

²Lead samples were taken near the cities indicated in the Table. A single number identified the site locations.

Analytical Procedures

Residue analyses were done by the Wisconsin Alumni Research Foundation² under contract with the Bureau of Sport Fisheries and Wildlife. Birds were prepared by skinning and removing the beak and wings at the first joint out from the body; the removed parts were discarded. Each 10-bird "pool" was ground together in a Hobart food chopper, and a subsample taken for analysis

by atomic absorption spectrophotometry. Results are reported on a whole body, wet-weight basis.

MERCURY

A cold vapor atomic absorption technique was suggested by the contracting laboratory as being more rapid with greater selectivity and less interference. Digestion for this technique was a modification of one reported by the Joint Mercury Residue Panel (4). The methodology was demonstrated by the contractor to provide results comparable to the WARF "boat" method for both bird and

² Mention of this commercial laboratory is for identification only and does not constitute endorsement by the U. S. Department of the Interior.

fish tissue and was accepted for use. A 25 ml sulfuric-nitric acid mixture (4:1) was added to a 10-g aliquot of the sample, and the mixture was heated slowly for 30 to 45 minutes to reach full temperature; the sample was then refluxed for 1 hour. After the digest had cooled to room temperature, it was transferred to a 100-ml volumetric flask quantitatively with ice water; the container was stoppered, and the sample again was allowed to return to room temperature. Determination was made with a Perkin-Elmer atomic absorption spectrophotometer Model 303 fitted with a cold vapor device and a Perkin-Elmer Model 304 recorder.

Instrument conditions were:

Wavelength	2537 Å (Setting 254)
Slit	3 mm, 20 Å
Range	UV
Source	Mercury hollow cathode lamp
Air	3 liters per minute
Recorder noise suppression	—1, expansion—3x

The standard curve used for determination had almost a 20-fold range starting with 0.010 μg of mercury. The curve was plotted using peak height versus micrograms of mercury.

An 0.5 gram equivalent aliquot was used to measure for mercury for an indicated sensitivity of 0.05 ppm. This was used since this is the level at which natural occurring and background residues could be expected. In terms of practical instrument sensitivity, 0.01 μg was the lower level of sensitivity for the analytical method. The procedures include methylmercury as part of the total mercury reported.

Recovery studies were conducted on the analytical method using samples not reported in this paper. Recoveries ranged from 86% to 106%. Figures reported in this paper were not corrected for recovery.

LEAD

A 25-g portion of the sample homogenate was dried and charred on a hot plate, then transferred to a 500° C muffle furnace and ashed overnight. The ash was cooled, wet with nitric acid, then taken to dryness, and returned to the muffle for 20 minutes. After the sample was again cooled, 2 ml of concentrated hydrochloric acid and about 15 ml of water were added. The mixture then was brought to a boil, cooled, and made to volume for analysis. Determination was made with a Model 303 Perkin-Elmer atomic absorption spectrophotometer in accordance with standard Perkin-Elmer procedures for lead (5).

Instrument settings were:

Wavelength	283.3 background at 280.0 negative
Recorder noise suppression	—3, expansion—3x

Recovery studies were not done for lead analysis. The lower level of sensitivity for lead was 0.1 ppm.

Results and Discussion

MERCURY

Residue levels of mercury in the 125 bird pools analyzed are shown in Table 2. Values are reported for individual sites grouped by 5-degree blocks, and block means are given. These preliminary findings for mercury residues in starlings appear somewhat encouraging in light of recent concern about relatively high residues in a variety of fish and game birds. With the exceptions of two high levels of 1.5 ppm and 1.9 ppm found in northwestern Oregon (1-A-3 and 1-A-4, respectively) and one sample approaching 0.5 ppm in California (2-A-1), the residue levels for mercury in starlings appear to be uniformly low throughout the contiguous United States. Of the 125 samples analyzed, 102 showed levels equal to or less than 0.09 ppm on a whole body, wet-weight basis; 53 of these 102 samples were below the upper value of the World Health Organization's "practical residue limit" (range, 0.02-0.05 ppm) (8). The practical residue limit describes residues expected to be found in food from background and natural environmental contamination. Of the 23 samples in which mercury was found in excess of 0.09 ppm, only the 3 West Coast samples (1-A-3, 1-A-4, 2-A-1) and 1 sample from Virginia (3-J-2) exceeded 0.2 ppm.

The mercury levels reported in this paper could well reflect environmental conditions that are not associated with contaminated aquatic food webs or with birds feeding on mercury-treated seeds. The wide-ranging, omnivorous, terrestrial ground-feeding habits of the starling support this hypothesis. On the other hand, the residues could be a result of a short physiological retention time or could possibly be directly influenced by the season during which sample collections were made (November and December). Data illustrating the half-life of mercury residues in starlings on a whole-body basis would be helpful in interpreting these monitoring findings.

LEAD

Lead residues were found in all 23 survey samples collected. Whole body, wet-weight residues ranged from a low of 0.4 ppm at Yakima, Wash., to a high of 13.3 ppm at Chicago, Ill. The mean level for the survey was 3.18 ppm with a standard error of 0.62 ppm. When the exceptionally high reading of 13.3 ppm (almost twice as much as the next highest residue) is dropped, the mean becomes 2.70 ppm with a standard error of 0.45 ppm. Lead monitoring findings are presented in Table 3.

Little is known about environmental dispersal of lead or about its assimilation by wildlife other than that reported as a result of ingestion of shotgun pellets by

TABLE 2.—Mercury residues in starlings, 1970

SAMPLING SITE NUMBER	MERCURY RESIDUE ¹ (PPM)	SAMPLING SITE NUMBER	MERCURY RESIDUE ¹ (PPM)	SAMPLING SITE NUMBER	MERCURY RESIDUE ¹ (PPM)	SAMPLING SITE NUMBER	MERCURY RESIDUE ¹ (PPM)
1-A-1	0.05	3-D-1	0.05	4-G-1	<0.05	3-I-1	<0.05
2	0.06	2	<0.05	2	<0.05	2	0.05
3	1.50	3	<0.05	3	<0.05	3	<0.05
4	1.90	4	<0.05	4	0.10	4	0.08
Mean	0.878	Mean	<0.05	Mean	<0.062	Mean	<0.058
SE	0.417	SE	<0.011	SE	<0.011	SE	<0.007
2-A-1	0.05	4-D-1	0.08	1-H-1	0.05	4-I-1	<0.05
2	<0.05	2	—	2	0.06	2	<0.05
3	0.07	3	<0.05	Mean	0.055	3	<0.05
4	0.11	Mean	<0.065	SE	0.067	Mean	<0.050
Mean	<0.175	SE	<0.010	2-H-1	0.05	SE	<0.050
SE	<0.085	1-E-1	<0.05	2	0.07	5-I-1	<0.05
3-A-1	<0.05	2	—	3	0.19	2	<0.05
2	0.07	3	0.05	4	0.05	Mean	<0.050
3	<0.05	4	<0.05	Mean	<0.090	SE	<0.008
Mean	<0.057	SE	<0.050	SE	<0.029	2-J-1	0.10
SE	<0.018	2-E-1	0.10	3-H-1	<0.05	2	0.10
1-B-1	0.05	2	0.07	2	<0.05	3	0.10
2	0.07	3	0.05	3	0.05	4	0.06
3	0.06	4	<0.05	4	0.10	Mean	0.090
4	0.14	Mean	<0.068	Mean	0.062	SE	0.008
Mean	0.080	SE	<0.010	SE	<0.011	3-J-1	<0.05
SE	0.023	3-E-1	0.15	4-H-1	<0.05	2	0.22
2-B-1	0.05	2	<0.05	2	0.08	3	0.18
2	0.10	3	<0.05	3	<0.05	Mean	<0.150
3	0.11	4	0.08	4	<0.05	SE	<0.053
4	0.06	Mean	<0.083	Mean	0.058	1-K-1	0.05
Mean	0.080	SE	<0.020	SE	0.007	2	0.05
SE	0.016	1-F-1	0.11	2-I-1	0.08	Mean	0.050
3-B-1	0.08	2	<0.05	2	0.11	SE	<0.050
2	0.09	3	0.05	3	<0.05	2-K-1	0.10
3	0.06	4	<0.05	4	<0.05	2	0.05
4	0.08	Mean	0.065	Mean	<0.080	Mean	0.075
Mean	0.078	SE	0.013	SE	<0.014	SE	0.019
SE	0.055	2-F-1	<0.05	NOT	—	SE	<0.014
4-B-1	0.09	2	<0.05	NOTE	no sample taken		
Mean	0.090	3	<0.05	1	Parts per million whole body,	7 birds,	
SE	—	4	0.18	2	wet-weight basis,	8 birds,	
1-C-1	0.06	Mean	0.083	3	2 birds	14 birds	
2	—	SE	0.028	4			
3	—	3-E-1	0.13				
4	<0.05	2	0.15				
Mean	<0.060	3	0.13				
SE	<0.007	4	0.09				
2-C-1	0.09	Mean	0.125				
2	—	SE	0.010				
3	<0.05	4-F-1	—				
4	0.06	2	—				
Mean	<0.067	3	0.08				
SE	<0.009	4	<0.05				
1-C-1	0.08	Mean	0.065				
2	—	SE	0.010				
3	<0.05	1-G-1	<0.05				
4	<0.05	2	0.05				
Mean	0.060	3	<0.05				
SE	0.008	4	0.05				
4-C-1	<0.05	Mean	<0.050				
2	<0.05	SE	<0.050				
Mean	<0.050	2-G-1	<0.05				
SE	<0.050	2	<0.05				
1-D-1	0.08	3	<0.05				
2	0.05	4	<0.05				
3	<0.05	Mean	<0.058				
4	<0.05	SE	<0.006				
Mean	<0.058	3-G-1	0.05				
SE	<0.006	2	<0.05				
2-D-1	<0.05	3	<0.05				
2	0.06	4	<0.05				
3	—	Mean	0.053				
4	<0.05	SE	<0.003				
Mean	0.053						
SE	<0.003						

NOTE — no sample taken

1 Parts per million whole body, wet-weight basis.

7 birds,
8 birds,
14 birds

watertowl and certain game birds. Available information indicates that contamination of vegetation may be directly related to the proximity of lead pollution levels in air (6).

Bagley and Locke (7) reported occurrence of lead residues in the liver and tibia of birds that were free of lead shot in the gizzards at necropsy. These birds had been pen raised, shot, or found dead. Sampled species associated with aquatic environments contained average lead residues in the liver of from 0.5 to 2.0 ppm on a wet-weight basis. Residues in the liver of terrestrial feeding birds averaged 3.7 ppm for cowbirds (*Molothrus ater*), 3.3 ppm for mourning doves (*Zenaidura macroura*), 2.1 ppm for rock ptarmigan (*Lagopus mutus*), and 0.5 ppm for ring-necked pheasant (*Phasianus colchicus*). Lead was found in the tibia of selected aquatic birds in the order of 2 ppm to 13 ppm. The rock ptarmigan averaged 7.1 ppm of lead in the tibia, and a single dusky grouse (*Dendragapus obscurus*) was reported to have 3.0 ppm tibia lead residues. These data appear to be consistent with the survey sample findings.

TABLE 3.—Lead residues in starlings

SAMPLING SITE NUMBER	LOCATION	LEAD RESIDUE ¹ (PPM)
1	Yakima, Wash.	0.4
2	Corvallis, Oreg.	0.8
3	Los Angeles, Calif.	
4	Reno, Nev.	1.2
5	Boise, Idaho	1.6
6	Salt Lake City, Utah	1.5
7	Phoenix, Ariz.	2.1
8	Greeley, Colo.	1.1
9	Carlsbad, N. Mex.	
10	Tishomingo, Okla.	1.7
11	Mitchell, S. Dak.	1.5
12	Twin Cities, Minn.	2.9
13	Chicago, Ill.	13.3
14	Sandusky, Ohio	.)
15	Columbus, Ohio	1.2
16	Stuttgart, Ark.	2.0
17	Baton Rouge, La.	0.8
18	Nashville, Tenn.	2.6
19	Atlanta, Ga.	4.9
20	Gainesville, Fla.	1.2
21	Raleigh, N. C.	5.2
22	Patuxent, Md.	3.6
23	N. Brunswick, N. J.	7.3
24	Jamestown, N. Y.	7.0
25	Gray, Maine	2.4

NOTE: Mean = 3.18.

Standard Error = .621.

Standard Deviation = 2.98.

Blank indicates no sample obtained.

¹ Parts per million whole body, wet-weight basis.

Acknowledgment

Collections were made by field personnel of the Wildlife Services Division, Bureau of Sport Fisheries and Wildlife. Regional Pesticide Specialists of the Division of Wildlife Services were responsible for coordinating and reporting collections and assuring that samples were

received by the contracting laboratory in proper condition. The Regional Pesticide Specialists are:

James B. Elder	Minneapolis, Minn.
Robert H. Hillen	Albuquerque, N. Mex.
David J. Lenhart	Portland, Oreg.
John C. Oberheu	Atlanta, Ga.
John W. Peterson	Boston, Mass.

Paul R. Nickerson, Division of Wildlife Services, did the statistical computations and assisted in collating the data presented in this report.

See Appendix for chemical names of compounds discussed in this paper.

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Organochlorine Residues in Starlings—1970¹

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ABSTRACT

As part of the National Pesticide Monitoring Program, starlings were collected in November and December 1970 from 125 sites throughout the contiguous United States and analyzed for certain persistent organochlorine insecticides. DDT and its metabolites, dieldrin, heptachlor epoxide, and benzene hexachloride were found in all samples. Polychlorinated biphenyls, which were estimated using Aroclor 1254 as a standard, were also found in all samples. A comparison of 1970 residue data with baseline information from 1967-68 indicates an apparent decline in levels of DDT and its metabolites and dieldrin; however, the decline is not significant at the 95% confidence level.

Introduction

As outlined in the recent description of the national pesticide monitoring program for wildlife (1), a nationwide sample of starlings (*Sturnus vulgaris*) is collected every other year and analyzed to help measure environmental levels of persistent organochlorine insecticides. Baseline data for residues of these contaminants, as reported by Martin in 1969 (2), were developed through the analysis of three complete random sample collections taken from 1967 through 1968. This paper presents data from the 1970 collections and compares it to the baseline findings.

Sample Design and Collection

The random sampling design used for this study is described by Dustman *et al.* (1). Basically, the design consists of 40 blocks of 5 degrees latitude (24 -49) and longitude (64 -124), with up to four sites randomly selected from each block. Fig. 1 shows the locations of the sampling sites, and Table 1 lists the locations sampled

during the November-December 1970 collection. Sampling locations are identified by a row number, a column letter, and a site number; e.g., the site near Tacoma, Wash., is designated 1-A-1. Collections were made at 125 of the possible 139 sampling locations (90%).

Starlings again proved difficult to collect in some areas. Sampling sites in Texas presented the greatest collection problem, with no samples being taken at seven of the nine preselected locations. Since the area in question is one of high pesticide use, new collection techniques will be developed or an alternate representative species will be selected prior to the 1972 collection.

There is no specific information, other than general observation, concerning starling population variation from block to block. The sampling design takes into account the fact that differences in number of birds do occur between and within the blocks. The study, however, is designed to measure trends of environmental residues rather than effects on starling populations.

The sample from each site normally consisted of a "pool" of birds. Pools containing fewer than 10 birds are indicated by footnote in the appropriate tables. Birds collected for organochlorine residue determination were either trapped or shot. Each 10-bird pool was wrapped in aluminum foil, placed in polyethylene bags, and frozen immediately for later laboratory analyses.

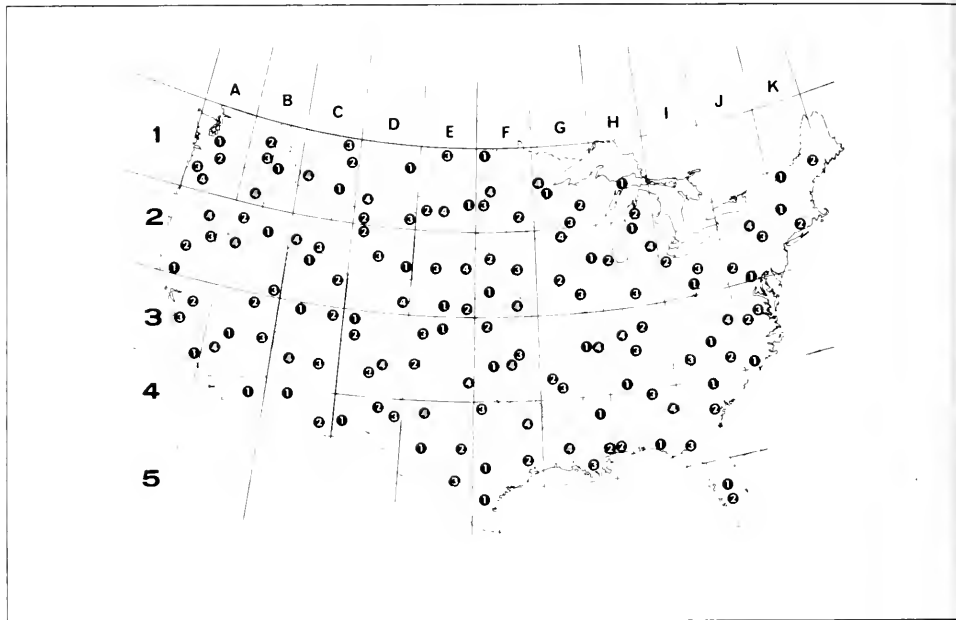
Sample Preparation and Analytical Methods

Residue analyses were done by the Wisconsin Alumni Research Foundation² under contract with the Bureau of Sport Fisheries and Wildlife. Birds were prepared by skinning and removing the beak and wings at the first

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² Mention of this commercial laboratory is for identification only and does not constitute endorsement by the U. S. Department of the Interior.

FIGURE 1.—Starling monitoring sites—1970



joint out from the body: the removed parts were discarded. Each 10-bird pool was ground together in a Hobart food chopper and a subsample taken for analysis.

Every effort was made to use essentially the same procedures for preparation and analyses of samples as in the baseline report (2). Determination was made using two columns with a Barber-Coleman Pesticide Analyzer, Model 5360. Normal instrument conditions were:

Column: Glass, 4 ft x 4 mm, packed with 5% DC-200 on 80/100 Gas Chrom Q
 Temperatures: Injector 230° C
 Column 195° C
 Detector 250° C
 Carrier gas: Nitrogen
 Flow rate: Such that *p,p'*-DDT had a retention time of 6 to 8 minutes

Modifications of techniques were used for a greater degree of accuracy and discrimination in identifying the isomers (alpha, beta, delta, and gamma) of benzene hexachloride. For this determination, the column was packed with 3% OV-17 rather than 5% DC-200, and appropriate instrument changes were made as follows:

Column: Glass, 4 ft x 4 mm, packed with 3% OV-17 on 80/100 Gas Chrom Q

Temperatures: Injector 230° C
 Column 180° C
 Detector 240° C

Flow rate: Such that gamma BHC has a retention time of 2 minutes

Polychlorinated biphenyls (PCB's) were estimated by using the peaks between DDD and DDT on the DC-200 column with Aroclor 1254 as a standard. On the basis of the findings of Risebrough, Reiche, and Olcott (3) regarding the lack of significant PCB interference with analytical findings for *p,p'*-DDE and because of a tight budgetary allowance for starling monitoring efforts, the PCB question was not pursued. However, it is expected that a closer analytical scrutiny would reveal PCB's other than the Aroclor 1254 estimates. The PCB estimates are provided only to help place in context the readings for DDT and DDD and to demonstrate the common occurrence of PCB's in starling tissues.

The residue data were not corrected for recovery. The standardized analytical procedure results in an 88% to 105% recovery rate for the chemicals reported in this paper. Recovery and confirmatory tests were done internally by the commercial laboratory on a quality control basis. Limits of detection ranged from 0.005 ppm to 0.01 ppm.

TABLE 1.—Starling sampling site locations by State and county, 1970

STATE	SAMPLING SITE NUMBER	COUNTY	STATE	SAMPLING SITE NUMBER	COUNTY	
Alabama	3-H-1	Marion	Montana	1-C-1	Meagher	
	4-H-3	Talladega		1-C-4	Missoula	
Arizona	3-C-3	Navajo		1-D-1	Richland	
	3-C-4	Yavapi		1-D-4	Yellowstone	
	4-C-1	Maricopa	Nebraska	2-E-3	Keith	
	4-C-2	Graham		2-E-4	Lincoln	
Arkansas	3-G-2	Yell/Pope		2-F-1	Clay	
	3-G-3	Lonoke/Pulaski		2-F-2	Antelope	
California	2-A-1	Colusa	Nevada	2-B-3	White Pine	
	2-A-2	Shasta		2-B-4	Humboldt	
	2-A-3	Modoc		3-B-2	Nye	
	3-A-1	Ventura	3-B-3	Clark		
	3-A-2	Stanislaus	New Mexico	3-D-3	Bernalillo	
	3-A-3	Monterey		3-D-4	Santa Fe/Torrance	
	3-B-1	Inyo		4-D-1	Luna	
	3-B-4	Kern		4-D-3	Chaves	
Colorado	4-B-1	Imperial	3-E-2	Quay		
	2-D-4	Adams	New York	2-J-4	Oswego	
	3-D-1	Montrose		2-K-1	Rensselaer	
	3-D-2	La Plata/Rio Grande	North Carolina	3-J-1	Wilkes	
3-E-3	Otero	3-J-2		Union		
Connecticut	2-K-2	New London		3-J-3	Macon	
				3-J-1	Pender	
Florida	4-H-1	Bay	North Dakota	1-E-3	Ward	
	4-I-3	Madison		1-F-1	Cavelier	
	5-I-1	Polk		1-F-4	Dickey	
	5-I-2	Hardee	Ohio	2-I-1	Washington	
Georgia	4-H-4	Pike		2-I-2	Errie	
	4-I-2	Wayne		2-I-3	Jefferson	
	Idaho	1-B-1	Nezperce	Oklahoma	3-E-4	Greer
		2-B-1	Owhee		3-F-1	Canadian
2-C-3		Franklin	3-F-3		Nowata	
2-C-4		Muldoka	3-F-4		Okmulgee	
Illinois	2-G-1	Stephenson	Oregon	1-A-3	Yamhill	
	2-G-3	Sangamon		1-A-4	Lane	
	2-H-2	Cook		2-A-4	Klamath	
Indiana	2-H-3	Henry		1-B-4	Baker	
			2-B-2	Harney		
Iowa	2-F-3	Pottawattamie	Pennsylvania	2-J-2	Somerset	
	2-G-2	Polk		2-J-3	Cuzco	
	2-G-4	Butler		South Carolina	4-I-1	Aiken
Kansas	2-E-1	Rawlins	South Dakota		1-E-1	Potter
	2-E-2	Smith			1-E-4	Hughes
	3-E-1	Hamilton/Kearny			1-F-3	Brown
	2-F-4	Nemaha		Tennessee	3-H-3	Davidson
	3-F-2	Marion	Texas		4-F-3	Clay
Kentucky	3-H-2	Ohio		4-F-4	Morris	
	3-H-4	Hopkins	Utah	2-C-1	Weber	
Louisiana	4-G-3	Jefferson		3-C-1	Sevier/Millard	
	4-G-4	Rapides		Vermont	1-K-1	Addison
Maine	1-K-2	Penobscot	Virginia		3-I-4	Amherst
Maryland	2-J-1	Prince Georges		3-I-2	Prince George	
	1-H-1	Chippewa		3-J-3	Caroline	
Michigan	1-H-2	Grand Traverse	Washington	1-A-1	Pierce	
	2-H-1	Kent		1-A-2	Yakima	
	2-H-4	Inaham		1-B-2	Spokane	
Minnesota	1-F-2	Swift	1-B-3	Whitman		
	1-G-1	Pine	Wisconsin	1-G-2	Curtiss	
	1-G-4	Aitkin		1-G-3	Fremont	
Mississippi	4-G-1	Leake	Wyoming	1-D-2	Big Horn	
	4-G-2	Harrison		1-D-3	Brook	
	4-H-2	Jackson		2-D-1	Goshen	
Missouri	3-G-1	Stoddard		2-D-2	Washakie	
	3-G-4	Bollinger				

Total sites = 125

Results and Discussion

Residue levels of organochlorine insecticides and PCB's for the November/December 1970 collection are presented in Table 7. The sampling design used in this study was chosen primarily because it permitted statistical comparison of the distribution of pesticide residues on a nationwide basis. The arbitrarily selected 5-degree blocks are the basic units for evaluating nationwide trends. Comparison of the distribution of average residues by frequency of occurrence in different quantitative ranges for the 1967-68 and 1970 collections (Table 4) offers an additional tool for evaluating changes in residue levels.

DDT AND METABOLITES

As in 1967-68, DDT and its metabolites were found in all samples taken. The 1970 block averages appear to reflect a general nationwide decline in levels, although this is only apparent and is not significant at the 95% confidence level (Table 2).

TABLE 2.—Block averages of residues of DDT and its metabolites

SAMPLING BLOCK	BASELINE DATA 1967-68		FALL 1970	
	AVERAGE RESIDUE LEVEL (PPM)	STANDARD ERROR	AVERAGE RESIDUE LEVEL (PPM)	STANDARD ERROR
1A	1.734	.350	.775	.260
2A	.755	.167	.472	.093
3A	2.281	.318	1.779	.768
1B	.809	1.320	.345	.064
2B	1.070	.331	.975	.400
3B	1.767	.590	1.053	.519
4B	3.450	1.490	2.192	—
1C	.341	.120	.145	.002
2C	3.616	2.190	.373	.088
3C	1.988	.651	.470	.152
4C	12.966	4.511	7.903	4.930
1D	.159	.038	.106	.017
2D	.432	.104	.263	.078
3D	1.162	.237	.502	.069
4D	12.574	8.155	3.130	1.160
1E	.919	.433	.099	.010
2E	.342	.065	.256	.051
3E	1.736	.706	1.460	1.114
1F	.219	.044	.244	.098
2F	.297	.050	.171	.032
3F	.713	.144	.229	.065
4F	.667	—	.172	.007
1G	.305	.064	.178	.023
2G	.443	.080	.266	.023
3G	2.134	.870	1.822	1.008
4G	4.201	.996	2.449	.877
1H	1.012	.312	.231	.042
2H	1.089	.182	1.213	.402
3H	1.463	.553	.937	.446
4H	2.110	.423	2.135	.425
2I	.576	.209	.455	.155
3I	.673	.136	.409	.121
4I	4.335	.812	2.619	.706
5I	1.550	.299	.316	.117
2J	.689	.102	.716	.321
3J	.795	.211	.709	.136
1K	.364	.053	.303	.077
2K	.593	.081	.411	.068

Individual sites having DDT and metabolite residues greater than 3.0 ppm in baseline and/or 1970 data are listed in Table 3, and frequency of occurrence of residues in different quantitative ranges is shown in Table 4. Sites containing the highest DDT and metabolite levels continue to be found in southern Arizona and New Mexico and in areas of the Southeast, including parts of Florida, Georgia, Alabama, Mississippi, Louisiana, and Arkansas. Because starlings were not available, we do not have 1970 data for two sites in Utah that had high residue levels in the baseline collections. Other States yielding relatively high residue levels (greater than 3.0 ppm) at one or more sites include Oklahoma, California, and South Carolina.

DIELDRIN

As with DDT, dieldrin residue levels are apparently declining from the baseline findings, although again the decline is not significant at the 95% confidence level (Table 5). Two exceptionally high residue levels of 3.59 and 1.52 ppm were found, however, along the upper Mississippi River drainage area (Sites 2-G-3 and 2-G-4). States from which samples were obtained with relatively high residue levels (greater than 0.3 ppm) at one or more sites include Georgia, Illinois, Iowa, Kansas, Missouri, and Washington. Individual sites having dieldrin residue levels greater than 0.3 ppm in baseline and/or 1970 collections are listed in Table 6, and frequency of occurrence of residues in different quantitative ranges is shown in Table 4.

BHC

BHC was found in all 125 samples collected in 1970; whereas, in the 1967-68 baseline study of three collections (375 samples), it was found in only 45 samples. The lindane reported in 105 samples of the baseline study is thought to have resulted as a product of technical BHC (Table 7).

BHC figures presented for 1970 cannot be related directly to the 1967-68 baseline figures, and it was decided not to re-analyze the 1967-68 baseline samples with the new techniques at this time because the levels were relatively low compared to established tolerances for edible food and feed.

HEPTACHLOR EPOXIDE

Heptachlor epoxide was found in all samples collected in 1970 (Table 7) and in 168 of 375 samples in the 1967-68 baseline study. Although the frequency of occurrence of heptachlor epoxide was greater for the 1970 collection, there is no statistically significant difference between the residue levels found in the 1970 sample and the baseline data. Any attempt at describing a trend in heptachlor epoxide residue levels in starlings should

be delayed until after residue levels for the 1972 collections are determined. Improvements in analytical methodology may be partly responsible for the apparent increase in frequency of occurrence of heptachlor epoxide residues.

TABLE 3.—Sites with residue levels of DDT and its metabolites greater than 3.0 ppm in baseline and/or 1970 collections

SAMPLING SITE NUMBER	DDT RESIDUES IN PPM	
	BASELINE DATA 1967-68	1970
3-A-1	1.903	3.660
3-B-4	4.376	2.837
2-C-2	9.551	no sample
3-C-2	3.163	no sample
4-C-1	23.902	14.874
4-D-1	1.930	4.780
4-D-3	19.680	1.479
3-E-4	4.948	5.318
3-G-3	5.950	5.313
4-G-1	8.128	3.413
4-G-2	1.580	4.801
4-G-4	4.220	1.210
4-H-3	2.347	3.060
4-H-4	3.510	2.546
4-I-1	5.483	3.026
4-I-3	5.668	3.872

TABLE 4.—Distribution of average residues of DDT and its metabolites and dieldrin by frequency of occurrence in different quantitative ranges—1967-68 and 1970 collections

RESIDUE RANGE (PPM)	FREQUENCY OF OCCURRENCE (SITES)	
	1967-68	1970
DDT AND METABOLITES		
≤1.0	76	103
>1.0 and ≤2.0	25	5
>2.0 and ≤3.0	12	7
>3.0 and ≤4.0	3	5
>4.0 and ≤5.0	3	2
>5.0 and ≤10.0	5	2
>10.0 and ≤15.0	0	1
>15.0 and ≤20.0	1	—
>20.0 and ≤25.0	1	—
Total sites	126	125

RESIDUE RANGE (PPM)	FREQUENCY OF OCCURRENCE (SITES)	
	1967-68	1970
DIELDRIN		
≤0.1	65	98
>0.1 and ≤0.2	40	12
>0.2 and ≤0.3	11	6
>0.3 and ≤0.4	2	2
>0.4 and ≤0.5	3	1
>0.5 and ≤1.0	4	4
>1.0 and ≤1.5	1	1
>1.5 and ≤2.0	—	—
>2.0 and ≤2.5	—	—
>2.5	—	1
Total sites	126	125

TABLE 5.—Block averages of dieldrin residues

SAMPLING BLOCK	BASELINE DATA 1967-68		FALL 1970	
	AVERAGE RESIDUE LEVEL (PPM)	STANDARD ERROR	AVERAGE RESIDUE LEVEL (PPM)	STANDARD ERROR
1A	.339	.094	.129	.019
2A	.066	.031	.020	.005
3A	.074	.016	.041	.008
1B	.339	.120	.237	.116
2B	.073	.019	.023	.005
3B	.128	.029	.030	.005
4B	.167	.061	.026	—
1C	.062	.025	.005	—
2C	.021	.030	.015	.007
3C	.102	.029	.035	.012
4C	.069	.018	.014	.007
1D	.028	.010	.012	.005
2D	.089	.028	.005	—
3D	.107	.037	.021	.005
4D	.041	.009	.088	.058
1E	.093	.040	.009	.001
2E	.143	.049	.095	.034
3E	.095	.019	.124	.086
1F	.169	.067	.052	.024
2F	.123	.033	.093	.060
3F	.079	.035	.038	.018
4F	.034	—	.020	.010
1G	.094	.040	.065	.023
2G	.338	.099	1.458	.665
3G	.755	.060	.215	.095
4G	.297	.119	.061	.017
1H	.080	.037	.032	.012
2H	.127	.043	.162	.068
3H	.104	.031	.067	.016
4H	.142	.035	.120	.047
2I	.118	.048	.124	.052
3I	.360	.233	.020	.005
4I	.045	.005	.305	.183
5I	.073	.023	.018	.007
2J	.133	.060	.063	.020
3J	.120	.078	.072	.036
1K	.023	.009	.014	—
2K	.012	.001	.015	.001

TABLE 6.—Sites with residue levels of dieldrin greater than 0.3 ppm in baseline and/or 1970 collections

SAMPLING SITE NUMBER	DIELDRIN RESIDUES IN PPM	
	BASELINE DATA 1967-68	1970
1-A-3	0.528	0.160
1-A-4	0.492	0.140
1-B-3	0.587	0.600
1-B-4	0.418	0.018
3-E-1	0.102	0.420
2-G-1	0.280	0.590
2-G-3	0.657	3.590
2-G-4	0.032	1.520
3-G-1	0.403	0.230
3-G-3	0.317	0.067
3-G-4	0.207	0.520
4-G-2	0.970	0.067
2-H-2	0.208	0.330
3-I-1	1.385	0.018
4-I-2	0.027	0.750
3-J-1	0.333	0.160

TABLE 7.—Pesticide residue levels in starlings, 1970

SAMPLING SITE NUMBER	WET WEIGHT (GRAMS)	LIPID WEIGHT (GRAMS)	RESIDUES IN PPM ($\mu\text{G/G}$, WET-WEIGHT)							
			DDE	DDD	DDT	DDT AND METABOLITES	ESTIMATED PCB'S	DIELDRIN	HEPTACHLOR EPOXIDE	BHC
1-A-1	19.98	0.115	0.430	0.029	0.022	0.481	0.31	0.150	0.053	0.009
1-A-2	20.02	0.370	0.320	0.016	0.021	0.357	0.15	0.066	0.009	0.390
1-A-3	19.99	0.814	0.520	0.033	0.045	0.598	0.38	0.160	0.016	0.015
1-A-4	20.02	0.869	1.620	0.013	0.029	1.662	0.23	0.140	0.012	0.110
2-A-1	20.00	0.789	0.670	0.013	0.023	0.706	0.26	0.022	0.013	0.013
2-A-2	20.03	0.742	0.180	0.009	0.022	0.211	0.25	0.016	0.009	0.009
2-A-3	19.98	1.210	0.550	0.008	0.014	0.572	0.17	0.034	0.019	0.013
2-A-4	20.00	1.281	0.380	0.006	0.013	0.399	0.14	0.006	0.008	0.021
3-A-1	20.03	0.813	3.590	0.017	0.053	3.660	0.65	0.021	0.034	0.005
3-A-2	20.00	0.803	0.710	0.023	0.090	0.823	0.96	0.057	0.016	0.015
3-A-3	19.99	0.830	0.770	0.031	0.053	0.854	0.39	0.045	0.019	0.015
1-B-1	20.05	1.285	0.150	0.014	0.015	0.179	0.14	0.050	0.014	0.230
1-B-2	19.99	1.063	0.280	0.034	0.038	0.352	0.36	0.280	0.016	0.160
1-B-3	20.00	1.203	0.450	0.040	0.048	0.538	0.39	0.600	0.013	0.590
1-B-4	19.99	1.443	0.230	0.026	0.055	0.311	0.50	0.018	0.027	0.031
2-B-1	20.00	1.184	0.410	0.006	0.011	0.427	0.09	0.018	0.026	0.013
2-B-2	19.96	1.069	0.200	0.039	0.160	0.399	2.19	0.008	0.011	0.240
2-B-3	20.04	1.305	2.250	0.065	0.234	0.78	0.15	0.032	0.032	0.035
2-B-4	20.01	1.422	0.700	0.012	0.017	0.729	0.15	0.034	0.016	0.016
3-B-1	20.01	1.377	0.580	0.021	0.033	0.634	0.36	0.045	0.039	0.010
3-B-2	20.00	1.388	0.240	0.016	0.016	0.272	0.25	0.018	0.170	0.012
3-B-3	19.99	1.411	0.360	0.038	0.069	0.467	0.79	0.035	0.038	0.022
3-B-4	20.00	0.753	2.750	0.026	0.061	2.837	0.51	0.023	0.016	0.018
4-B-1	20.01	1.000	2.110	0.029	0.053	2.192	0.53	0.026	0.120	0.025
1-C-1	20.00	2.454	0.110	0.015	0.024	0.149	0.17	0.005	0.010	0.031
1-C-4	20.01	0.737	0.064	0.028	0.049	0.141	0.66	0.005	0.009	0.015
2-C-1	20.07	0.903	0.140	0.005	0.012	0.157	0.13	0.008	0.026	0.013
2-C-3	19.98	0.924	0.480	0.006	0.013	0.499	0.14	0.031	0.039	0.014
2-C-4	20.06	1.303	0.420	0.019	0.024	0.463	0.25	0.005	0.017	0.036
3-C-1	19.99	1.479	0.750	0.021	0.044	0.815	0.27	0.039	0.022	0.013
3-C-3	20.01	0.969	0.350	0.022	0.047	0.419	0.36	0.008	0.010	0.007
3-C-4	19.98	0.777	0.150	0.009	0.017	0.176	0.15	0.057	0.012	0.008
4-C-1	20.06	0.924	14.800	0.036	0.038	14.874	0.32	0.005	0.007	0.034
4-C-2	20.10	0.638	0.870	0.022	0.040	0.932	0.31	0.022	0.015	0.008
1-D-1	20.00	0.978	0.078	0.005	0.005	0.088	0.05	0.006	0.006	0.016
1-D-2	19.97	0.860	0.050	0.014	0.025	0.089	0.29	0.029	0.015	0.005
1-D-3	20.03	2.397	0.094	0.011	0.017	0.122	0.17	0.007	0.080	0.017
1-D-4	19.99	1.045	0.064	0.025	0.034	0.123	0.30	0.008	0.010	0.014
2-D-1	20.03	1.025	0.310	0.005	0.010	0.325	0.14	0.005	0.006	0.160
2-D-2	19.98	0.704	0.047	0.008	0.022	0.077	0.19	0.005	0.011	0.019
2-D-4	20.04	1.355	0.350	0.016	0.022	0.388	0.14	0.005	0.019	0.022
3-D-1	19.99	1.323	0.790	0.015	0.028	0.733	0.29	0.031	0.019	0.012
3-D-2	20.00	1.554	0.380	0.014	0.029	0.423	0.20	0.018	0.009	0.045
3-D-3	20.02	1.181	0.340	0.007	0.035	0.382	0.72	0.019	0.012	0.009
3-D-4	19.99	0.839	0.430	0.014	0.026	0.470	0.20	0.016	0.008	0.012
4-D-1	19.90	0.735	4.750	0.013	0.017	4.780	0.17	0.170	0.026	0.012
4-D-3	19.99	1.156	1.450	0.009	0.020	1.479	0.25	0.006	0.006	0.016
1-E-1	20.06	1.385	0.062	0.014	0.023	0.099	0.26	0.005	0.006	0.013
1-E-3	20.02	0.976	0.071	0.017	0.032	0.120	0.37	0.008	0.009	0.011
1-E-4	20.00	1.400	0.056	0.010	0.013	0.079	0.11	0.013	0.024	0.014
2-E-1	20.02	1.069	0.130	0.028	0.044	0.202	0.41	0.21	0.015	0.017
2-E-2	20.01	0.943	0.082	0.030	0.034	0.146	0.29	0.041	0.009	0.010
2-E-3	20.00	1.561	0.320	0.043	0.055	0.418	0.47	0.044	0.016	0.014
2-E-4	19.99	1.518	0.200	0.017	0.039	0.256	0.38	0.086	0.120	0.023
3-E-1	20.00	1.176	0.140	0.005	0.006	0.151	0.09	0.420	0.017	0.012
3-E-2	19.93	0.739	0.190	0.019	0.049	0.258	0.36	0.026	0.012	0.010
3-E-3	19.99	1.018	0.099	0.006	0.009	0.114	0.17	0.014	0.008	0.008
3-E-4	20.09	0.794	5.290	0.011	0.117	5.318	0.12	0.034	0.010	0.008
1-F-1	19.99	1.639	0.015	0.016	0.019	0.050	0.14	0.021	0.015	0.018
1-F-2	20.00	1.639	0.110	0.044	0.063	0.217	0.58	0.048	0.100	0.017
1-F-3	20.01	1.799	0.520	0.021	0.028	0.569	0.19	0.130	0.019	0.015
1-F-4	20.01	1.521	0.081	0.017	0.041	0.139	0.26	0.007	0.009	0.016

TABLE 7.—Pesticide residue levels in starlings, 1970—Continued

SAMPLING SITE NUMBER	WET WEIGHT (GRAMS)	LIPID WEIGHT (GRAMS)	RESIDUES IN PPM ($\mu\text{G G. WET-WEIGHT}$)							
			DDE	DDD	DDT	DDT AND METABOLITES	ESTIMATED PCB'S	DIELDRIN	HEPTACHLOR EPOXIDE	BHC
2-F-1	19.98	1.571	0.130	0.009	0.016	0.155	0.13	0.015	0.013	0.011
2-F-2	19.98	1.351	0.180	0.006	0.028	0.214	0.25	0.049	0.056	0.020
2-F-3	20.00	2.269	0.180	0.025	0.034	0.239	0.33	0.300	0.093	0.048
2-F-4	20.04	0.910	0.047	0.009	0.019	0.075	0.14	0.009	0.007	0.016
3-F-1	20.02	0.909	0.091	0.007	0.014	0.112	0.21	0.012	0.015	0.010
3-F-2	19.96	1.195	0.140	0.008	0.015	0.163	0.25	0.034	0.019	0.012
3-F-3	19.96	1.089	0.390	0.019	0.039	0.448	0.45	0.009	0.016	0.012
3-F-4	19.98	0.896	0.170	0.006	0.015	0.191	0.18	0.098	0.031	0.011
4-F-3	20.07	0.755	0.140	0.009	0.015	0.164	0.15	0.035	0.024	0.012
4-F-4	20.06	0.568	0.160	0.005	0.015	0.180	0.17	0.005	0.007	0.013
1-G-1	19.98	1.443	0.096	0.031	0.082	0.209	1.00	0.013	0.011	0.013
1-G-2	19.99	1.196	0.069	0.010	0.024	0.103	0.26	0.026	0.016	0.013
1-G-3	20.06	2.358	0.100	0.028	0.053	0.181	0.18	0.120	0.045	0.016
1-G-4	20.01	2.510	0.160	0.025	0.034	0.219	0.29	0.100	0.028	0.020
2-G-1	19.98	1.763	0.250	0.019	0.038	0.307	0.29	0.59	0.340	0.024
2-G-2	20.02	1.909	0.150	0.019	0.028	0.197	0.29	0.13	0.028	0.120
2-G-3	20.04	1.636	0.200	0.029	0.080	0.309	0.87	3.59	0.970	0.030
2-G-4	20.01	1.870	0.210	0.014	0.028	0.252	0.22	1.52	0.110	0.034
3-G-1	20.02	2.002	0.630	0.032	0.047	0.709	0.48	0.230	0.084	0.021
3-G-2	20.00	0.827	0.680	0.019	0.031	0.730	0.38	0.043	0.100	0.014
3-G-3	19.98	0.916	5.240	0.034	0.039	5.313	0.28	0.067	0.013	0.017
3-G-4	19.97	2.384	0.450	0.040	0.045	0.535	0.43	0.520	0.120	0.041
4-G-1	20.00	0.703	3.300	0.038	0.075	3.413	0.56	0.075	0.036	0.019
4-G-2	20.01	0.733	4.690	0.041	0.070	4.801	0.40	0.067	0.099	0.011
4-G-3	20.00	0.619	0.260	0.014	0.099	0.373	1.260	0.005	0.210	0.017
4-G-4	20.00	0.842	1.030	0.050	0.130	1.210	1.82	0.095	0.130	0.051
1-H-1	20.06	2.594	0.150	0.068	0.073	0.291	0.82	0.016	0.034	0.026
1-H-2	19.98	0.897	0.120	0.015	0.036	0.171	0.46	0.048	0.021	0.039
2-H-1	20.00	2.319	0.410	0.099	0.260	0.769	3.13	0.031	0.110	0.030
2-H-2	20.00	1.703	2.410	0.050	0.120	2.580	1.36	0.330	0.630	0.039
2-H-3	19.97	1.348	0.360	0.069	0.110	0.539	1.35	0.260	0.063	0.140
2-H-4	19.96	1.935	0.830	0.041	0.091	0.962	0.93	0.025	0.050	0.011
3-H-1	20.09	0.665	2.400	0.010	0.022	2.432	0.25	0.085	0.027	0.013
3-H-2	19.98	1.087	0.140	0.007	0.019	0.166	0.41	0.012	0.007	0.011
3-H-3	20.00	1.137	0.320	0.013	0.023	0.356	0.28	0.081	0.099	0.030
3-H-4	19.99	0.874	0.750	0.010	0.034	0.794	0.28	0.091	0.019	0.012
4-H-1	20.08	1.171	0.730	0.012	0.024	0.766	0.36	0.087	0.100	0.012
4-H-2	20.01	0.873	1.720	0.170	0.280	2.170	2.27	0.280	0.150	0.026
4-H-3	20.03	1.074	1.640	0.340	1.080	3.060	24.30	0.062	0.100	0.024
4-H-4	20.02	1.055	2.500	0.015	0.031	2.546	0.25	0.051	0.022	0.016
2-I-1	19.98	2.176	0.410	0.050	0.079	0.539	0.80	0.130	0.056	0.030
2-I-2	20.06	1.192	0.510	0.055	0.082	0.647	0.89	0.230	0.032	0.017
2-I-3	20.05	1.050	0.110	0.025	0.045	0.180	0.62	0.011	0.084	0.013
3-I-1	19.99	0.822	0.150	0.010	0.015	0.175	0.20	0.018	0.028	0.012
3-I-2	19.99	0.984	0.770	0.013	0.025	0.808	0.25	0.019	0.018	0.014
3-I-3	20.00	0.957	0.200	0.021	0.048	0.269	0.36	0.028	0.024	0.013
3-I-4	20.02	0.661	0.330	0.015	0.039	0.384	0.41	0.013	0.037	0.011
4-I-1	20.01	0.763	3.000	0.009	0.017	3.026	0.13	0.120	0.019	0.013
4-I-2	19.99	0.801	0.880	0.016	0.064	0.960	0.73	0.750	0.038	0.026
4-I-3	20.02	0.738	3.820	0.015	0.037	3.872	0.47	0.044	0.032	0.061
5-I-1	19.98	0.561	0.390	0.009	0.034	0.433	0.38	0.025	0.040	0.067
5-I-2	19.99	0.701	0.140	0.016	0.043	0.199	0.43	0.010	0.009	0.005
2-J-1	20.09	0.763	0.230	0.013	0.053	0.296	0.75	0.030	0.093	0.018
2-J-2	20.02	1.601	0.240	0.041	0.087	0.368	0.81	0.097	0.046	0.025
2-J-3	20.00	1.097	1.750	0.033	0.044	1.827	0.56	0.110	0.044	0.015
2-J-4	20.00	1.989	0.260	0.035	0.078	0.373	0.65	0.016	0.025	0.013
3-J-1	20.00	0.867	0.320	0.033	0.120	0.473	1.18	0.160	0.018	0.009
3-J-2	20.06	0.834	0.940	0.014	0.077	1.031	0.79	0.022	0.055	0.011
3-J-3	20.02	0.676	0.520	0.067	0.037	0.624	0.31	0.033	0.160	0.019
1-K-1	20.00	1.095	0.110	0.031	0.053	0.194	0.75	0.013	0.010	0.011
1-K-2	20.01	1.202	0.320	0.032	0.060	0.412	0.69	0.014	0.022	0.016
2-K-1	20.04	0.920	0.450	0.028	0.028	0.506	0.33	0.017	0.021	0.017
2-K-2	19.99	0.654	0.290	0.009	0.016	0.315	0.25	0.013	0.024	0.012

1 2 birds.

2 7 birds.

3 8 birds.

4 14 birds.

Acknowledgment

Collections were made by field personnel of the Bureau of Sport Fisheries and Wildlife. Regional Pesticide Specialists of the Division of Wildlife Services were responsible for coordinating and reporting collections and assuring that samples were received by the contracting laboratory in proper condition. The Regional Pesticide Specialists are:

James B. Elder	Twin Cities, Minn.
Robert H. Hillen	Albuquerque, N. Mex.
David J. Lenhart	Portland, Oreg.
John C. Oberheu	Atlanta, Ga.
John W. Peterson	Boston, Mass.

See Appendix for chemical names of compounds discussed in this paper.

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The Occurrence of Mirex in Starlings Collected in Seven Southeastern States—1970¹

John C. Oberheu

ABSTRACT

Selected samples of starlings collected for the National Pesticides Monitoring Program were analyzed for mirex, the chemical used for eradication of the imported fire ant. Whole bodies (less skin, beak, feet, and outer wings) of 10 birds from each sampling site were pooled for analysis. Residues present in 10 of the 12 sample pools ranged from 0.01 to 1.66 ppm.

Introduction

During the fall of 1970, the U. S. Department of Agriculture planned to treat 120 million acres in the southeastern United States with mirex to eradicate the imported fire ant. Very little information was available on the uptake of mirex in animal food chains, and there was widespread concern among conservationists about the environmental hazards of this chemical.

During November and December 1970, samples of starlings (*Sturnus vulgaris*) were collected by the Bureau of Sport Fisheries and Wildlife from designated sampling sites throughout the country as a part of the national pesticides monitoring network, described by Dustman *et al.* (2). Since the standard analysis of these monitoring samples does not include determination of mirex residues, arrangements were made for special analysis of samples collected in seven of the States which either had been or would be treated in the fire ant eradication program. This paper presents the results of these analyses. Data on organochlorine residues in the nationwide starling monitoring samples are reported separately by Martin and Nickerson (5); mercury and lead data are included in Martin (4).

Sampling and Analytical Procedures

Details on selection of sample sites, methods of collection, and analytical procedures for the starling samples were described by Martin (3,4) and Martin and Nickerson (5). Each sample consisted of a pool of 10 starlings collected by trapping or shooting. Specimens were wrapped in aluminum foil, placed in plastic bags, and frozen immediately. They were kept frozen until processed for analysis by the Wisconsin Alumni Research Foundation.²

The birds were skinned, and beaks, legs, and outer wings were clipped off. The bodies were ground thoroughly in a Hobart food chopper. A 20-g portion of the homogenate was weighed into a 150-ml beaker and placed in a 40 °C oven for 72-96 hours. After dry-weight calculation, the samples were ground with 100 g of Na₂SO₄ and placed in a 33-x 94-mm Whatman extraction thimble. Samples were extracted for 8 hours on a Soxhlet extractor using 70 ml of ethyl ether and 170 ml of petroleum ether. The solvent was concentrated to 10-15 ml on a steam bath and made to 50 ml with petroleum ether.

Cleanup was accomplished by placing an aliquot of the sample on previously standardized Florisil (1). Typical elutions were 150 ml of 5% ethyl ether in petroleum ether, followed by 240 ml of 15% ethyl ether in petroleum ether. The resulting solutions were concentrated on a steam bath (10-15 ml) and made to 25 ml with hexane.

¹ From the Bureau of Sport Fisheries and Wildlife, U. S. Department of the Interior, Atlanta, Ga. 30323.

² Mention of this commercial laboratory is for identification only and does not constitute endorsement by the U. S. Department of the Interior.

A 10- μ l portion of the sample solution was injected into a Barber-Coleman, Model 5360, gas chromatograph, operating with the following instrument conditions:

Column: Glass, 4' x 4 mm, packed with 5% DC-200 on 80/100 Gas Chrom Q
 Temperatures: Column 210° C
 Injector 230° C
 Detector 240° C
 Carrier gas: Nitrogen
 Flow rate: Mirex off column in 12-15 minutes

Results

Table 1 presents the residue levels of mirex in starlings from the 12 collection sites, a history of the nearest mirex treatments, and each site's rated potential for accumulating residues.

All but two of the samples contained detectable levels of mirex. The highest levels occurred at Anniston, Ala., and Thomaston, Ga., both locations of high exposure potential. Although starlings are known to be migratory birds that can range widely for food, the residues occurring in most of the samples correspond generally

with the exposure potential. The significance of these residue levels in birds is not presently known. The occurrence in 10 out of 12 samples does, however, prove that mirex will readily appear in animal food chains despite the very low dosage (1.7 g per acre) at which it is applied (6).

See Appendix for chemical name of mirex.

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TABLE 1.—Levels of mirex in starlings, history of mirex treatment, and exposure potential for 1970 collection sites

SAMPLE SITE LOCATION	MIREX RESIDUES (PPM) ¹	NEAREST MIREX TREATMENT	DATES OF TREATMENT	EXPOSURE POTENTIAL
Penderlee, N. C.	ND	65 miles	Spring and fall of 1966, 1968, and 1969	Low
Monroe, N. C.	ND	100 miles	Spring and summer 1963, summer 1964, spring 1965 and fall 1967	Low
Aiken, S. C.	0.08	Treated (5,000 acres)	Fall of 1966 and 1967, spring 1970	Moderate to high
Carthage, Miss.	0.08	15 miles (855,000 acres)	Spring 1967	Low to moderate
Gulfport, Miss.	0.42	Treated (75,000 acres)	Summer 1969	Moderate to high
Alexandria, La.	0.10	Treated (802,000 acres)	Spring and fall 1965, spring 1966	Low
Anniston, Ala.	1.61	Immediately adjacent (5,000 acres)	Spring 1970. Bait available for landowner treatments	High
Thomaston, Ga.	1.66	3 miles (1,840,000 acres)	Fall 1969	Moderate to high
Jessup, Ga.	0.62	Treated (200,000 acres)	Spring and fall 1970	High
Panama City, Fla.	0.31	Same county (800 - 2,240 acres)	Fall 1964 and 1965, spring 1966 and 1967	Low to moderate
Madison, Fla.	0.01	75 miles (1,000 acres)	Annual treatment since 1965	Low
Winter Haven, Fla.	0.34	Treated (368,000 acres)	Spring and summer 1967. County treatments of 2,500-368,000 acres annually 1964-69	Moderate

NOTE: ND = not detected.
¹ Wet-weight basis.

Organochlorine Pesticide Residues in Commercially Caught Fish in Canada—1970

J. Reinke,¹ J. F. Uthe,¹ and D. Jamieson²

ABSTRACT

Organochlorine pesticide residues were determined for commercially caught fish from a total of 78 locations in 68 central Canadian lakes and rivers. Only a few of these waters yielded fish with appreciable concentrations of DDT and its analogs (>1 ppm), and in only a few cases did the concentrations exceed the maximum permissible level of 5 ppm. Of the other organochlorine pesticides commonly used, namely lindane, aldrin, heptachlor, heptachlor epoxide, endrin, dieldrin, and chlordane, only dieldrin was found at significant levels in a number of samples, but these amounts were still below the maximum permissible level. Trace amounts of lindane were found in some samples. The presence of polychlorinated biphenyls (PCB's) was noted in samples from the Great Lakes and the south end of Lake Winnipeg. All results were confirmed by multiple-column gas chromatography and thin layer chromatography. PCB's were separated from DDE on aluminum oxide G (type E) plates run in a triethylamine-hexane solvent system.

Introduction

In recent years the fishing industry as a whole has suffered adversely from the effects of a wide variety of pollutants. The finding of extremely high concentrations of DDT in coho salmon (*Oncorhynchus kisutch*) from the Great Lakes (6) and northern anchovy (*Engraulis mordax*) off Los Angeles (7) has led to certain species being banned from the market and, undoubtedly, has resulted in public distrust of some fish products. Similarly, the recent fishery disasters due to mercury, although eliminating contaminated products from the market, have caused adverse publicity resulting in reduced sales (3). To meet the need created by these incidents for a detailed survey of organochlorine pesticide residues in fish from commercially fished lakes and

rivers in central Canada, a study was carried out in 1970 on fish collected from 78 locations in 68 Canadian waters (Fig. 1) and is reported here.

All analyses were done in duplicate, with all pesticides being determined by quantitation of gas-liquid chromatograms obtained from two different column packings; the second column packing was chosen to separate pesticides not widely separated by the first column. The identities of the pesticides were confirmed by thin layer chromatography.

Materials and Methods

SAMPLING PROCEDURES

Fish samples were collected from commercial fishermen by officers of the Inspection Service of the Department of Fisheries and Forestry. A sample consisted of 5 lb of headless dressed fish; if the fish were large, as few as three were pooled to make up a sample. The fish were frozen and shipped to Winnipeg where they were ground (Hobart, Model L800); a 2-lb portion of each sample was refrozen in a block and stored at -40°C until analyzed.

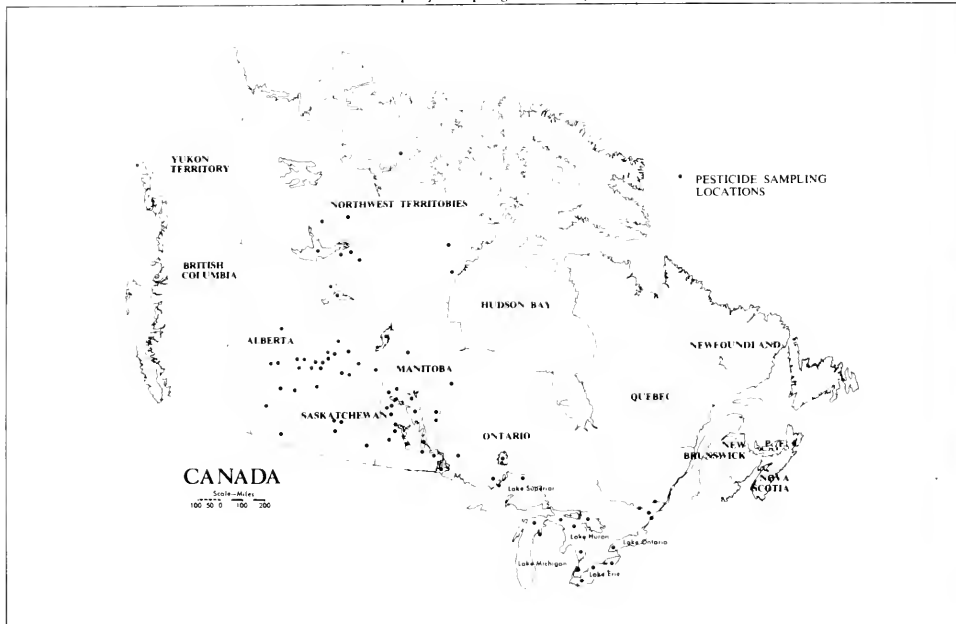
ANALYTICAL PROCEDURES

For sample extraction, the method of Mills *et al.* (4) was used with slight modifications. Hexane, petroleum ether, and acetonitrile were purchased as reagents certified for pesticide analysis. Acetone was redistilled prior to use, and diethyl ether was purified according to the method of Grussendorf *et al.* (2). A 25-g portion of fish was taken from the center of the frozen sample block and blended with 75 ml of acetonitrile for 2 minutes at high speed in a Waring blender (Model B). The homogenate was suction filtered through a sintered glass funnel directly into a 1-liter separatory funnel and re-extracted twice with 25 ml of acetonitrile; then, 350 ml of distilled water, 7 ml of saturated aqueous NaCl.

¹ Fisheries Research Board of Canada, Freshwater Institute, Winnipeg 19, Manitoba.

² Department of Fisheries, Inspection Branch, Winnipeg, Manitoba

FIGURE 1.—Map of sampling locations, Canada—1970



and 57 ml of petroleum ether were added to the separatory funnel, shaken vigorously, and allowed to separate into layers. The aqueous layer was discarded, and the organic layer was washed twice with 100 ml of water and drained into a 100-ml graduated cylinder; the recovered petroleum ether was then measured to determine the correction factor, transferred to a 250-ml round bottom flask, and reduced to approximately 1 ml on a rotary evaporator at 40° C.

The sample was quantitatively transferred to the cleanup column by rinsing the round bottom flask with two 10-ml aliquots of 8% ether-hexane. The column was made up of 30 g of 2% H₂O-deactivated Florisil (60/80 mesh, PR grade, Floridin Co.) with ½ inch of anhydrous Na₂SO₄ (reagent grade, Fisher Scientific) above and below the Florisil. The Florisil had been stored at 130° C prior to deactivation; after deactivation, it had been slowly tumbled for ½ hour, then left to equilibrate further for 24 hours. It was then stored in a glass-stoppered bottle and was stable for 1 week.

The pesticides were eluted with 8% diethyl ether:hexane (8:92; v/v) to give a total eluate of 220 ml. This eluate was evaporated on a rotary evaporator to approximately 1 ml and then made up to the required volume with hexane prior to gas chromatography. All pesticides reported were confirmed by thin layer chromatography.

Gas-Liquid Chromatography

A Hewlett-Packard Model 5750, fitted with a pulsed d.c. Ni⁶³ electron capture detector, was used for gas chromatographic analysis. Operating parameters were as follows:

Columns:	Glass, 6' x ¼", o.d., packed with either 2% SE-30/3% QF-1 or 3% OV-225 on 80/100 mesh HMDS treated Chromosorb W.
Temperatures:	Injection port 230° C Column oven 200° C Detector 260° C
Carrier gas:	Helium
Purge gas:	10% methane in argon

Standard solutions (1 µg/ml) of lindane, aldrin, heptachlor, heptachlor epoxide, endrin, *p,p'*-DDE, *p,p'*-DDT, *p,p'*-DDD, *o,p'*-DDT, and dieldrin were prepared in hexane. Each standard gave a single peak on gas chromatography and a single spot on a thin layer plate. The chlordane standard was reported to be 95% α -chlordane; gas chromatography showed two peaks only.

Quantitation was based on the peak heights obtained on injection of known amounts of pesticides. Care was taken to ensure that all heights were within the linear range of the detector, and, rather than make extrapolations, the samples were diluted. The following calcula-

tions were used to determine parts per million of residues in fish samples:

$$\text{ppm of residue} = \frac{\text{ng}/5 \mu\text{l}}{x/n} \times \text{c.f.}$$

x = weight of samples
 n = number of milliliters in final volume
 $5 \mu\text{l}$ = volume injected

$$\text{ng in volume injected} = \frac{\text{peak height of sample}}{\text{peak height of standard}}$$

$$\text{correction factor (c.f.)} = \frac{\text{ml petroleum ether recovered}}{\text{ml petroleum ether used}}$$

Thin Layer Chromatography

Aluminum oxide G (E. Merck, Darmstadt) plates, 0.5 mm thick, were prepared according to the method of Moats (5). The plates were run in a hexane:triethylamine (Eastman Co.) solution (100:7, v/v).

Retention factors for pesticides confirmed by TLC are as follows:

Compounds	Retention Factors
Methoxychlor	.119
Lindane	.223
<i>p,p'</i> -DDD	.224
Dieldrin	.268
Chlordane	
<i>cis</i>	.283
<i>trans</i>	.328
Heptachlor epoxide	.328
BHC	.335
Endrin	.358
<i>p,p'</i> -DDT	.388
<i>o,p'</i> -DDT	.447
Heptachlor	.597
<i>p,p'</i> -DDE	.611
Aldrin	.642
PCB's (five spots)	.436, .530, .536, .657, .751

This particular solvent system was chosen because it separated the pesticides and at the same time separated the PCB spots in a manner which allowed for DDE identification and isolation. This method is sensitive to 1.0 μg for visible identification. If spots were not visible, the area was scraped and eluted with 1:1 hexane:acetone and injected to the gas chromatograph for confirmation.

RECOVERY STUDIES

During the course of the survey, recovery studies were performed every other month. The studies were carried out by spiking previously analyzed samples with known amounts of the desired pesticides. The difference between the spiked and the unspiked result was used to indicate

the percent recovered. Similar studies were performed with blanks at various steps in the analyses. Recovery values shown below represent the average values of all eight recovery studies carried out:

Heptachlor	72%	<i>p,p'</i> -DDT	84%
Aldrin	72%	<i>p,p'</i> -DDD	90%
Heptachlor epoxide	77%	<i>p,p'</i> -DDE	90%
Lindane	80%	<i>o,p'</i> -DDT	94%
Endrin	81%	Dieldrin	100%

The standard deviation for all pesticides was $\pm 6\%$.

The results of recovery studies showed lower recovery values for the compounds with lower boiling points, i.e., lindane, heptachlor, aldrin, endrin, and heptachlor epoxide. Also, recovery values for these compounds were lower than results of Brown (1) in his comparison of the Mills and Langlois cleanup procedures using fatty and non-fatty samples. Brown's recovery results (1), however, are lower for the remaining pesticides examined except for DDD in fish oil in which he had a recovery of 111%; this was apparently due to both *o,p'*-DDT and *p,p'*-DDD peaks eluting simultaneously under his GLC conditions. Because small amounts of only a few pesticides with low boiling points were found, their lower recovery values, in this instance, are not considered to be critical.

The cause of the low recovery values for the compounds mentioned above could be the fact that the sample volume was reduced two times by rotary vacuum evaporator before it was ready for gas-chromatographic analyses.

Results were expressed without correction for percent recovery.

Results and Discussion

Pesticides were found in fish from most of the 68 lakes and rivers sampled (Table 1). However, the only waters which showed significant amounts of DDT and its analogs (>1 ppm) were in the Provinces of Ontario and Alberta and are as follows:

Province of Ontario	Province of Alberta
The Great Lakes	Lake St. Paul
Huron	Sturgeon River
Erie	Cold River
Ontario	Bow River
Michigan	
Superior	
Lake St. Clair	
Lake Nipigon	
Ottawa River	

Of the pesticides analyzed for, DDT and its analogs appeared most often with dieldrin appearing almost as

frequently. PCB's were quite prominent throughout the Great Lakes and appeared to a lesser degree in southern Lake Winnipeg in Manitoba and in the Bow River area of Alberta. It should be noted that, because of the interference of PCB's with DDT and its analogs, the DDT results of the survey in the areas where PCB's were found may, to some extent, be erroneous, but thin layer studies indicated that the reported values would be higher by 10% at most. The fact that not all regions with DDT present in fish samples had PCB's present would indicate that PCB's are not being used to extend the kill life of organochlorine pesticides and that these compounds do not enter the environment as a breakdown product of pesticides. Trace amounts of other pesticides (especially lindane, heptachlor, and heptachlor epoxide) were found in the Great Lakes, some lakes in northern Manitoba and Saskatchewan, and in many lakes in the Northwest Territories.

Results showed that pesticide concentrations differed for various species in the same lake and that the pesticide concentrations in the same species varied with body size and weight.

See Appendix for chemical names of compounds discussed in this paper.

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- (3) Kroeger, Wally. 1971. Report on a speech by Dave F. Corney (Freshwater Fish Marketing Corporation) to the 13th Annual Meeting of the Manitoba Federation of Fishermen. Winnipeg Tribune. 1 Apr. p. 19.
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- (7) Riechbrough, R. W. 1969. Chlorinated hydrocarbons in marine ecosystems, p. 5 to 23. In Morton W. Miller and George G. Berg [ed.] Chemical fallout-current research on persistent pesticides. Charles C. Thomas, Springfield, Ill.

TABLE 1.—Pesticide residue levels in fish, by provinces and waters, Canada—1970
[T = trace = <0.005 ppm; — = not detected]

LAKE OR RIVER ¹	LATITUDE AND LONGITUDE		FISH SPECIES	PESTICIDE RESIDUE LEVELS (PPM)					TOTAL DDT
				DIELDRIN	p,p'-DDE	o,p'-DDT	p,p'-DDD	p,p'-DDT	
PROVINCE OF ONTARIO									
Lake Huron	44° 30'	82° 15'	Coho salmon	—	2.20	0.70	0.80	1.30	5.00
			do.	—	8.90	1.20	1.50	3.90	15.50
			do.	—	6.70	0.70	1.00	2.60	11.00
(Manitoulin Is.)			do.	0.16	1.80	0.20	0.50	0.80	3.30
(Georgian Bay)			do.	0.40	11.10	0.90	1.40	3.00	16.40
Do.			do.	0.20	5.60	0.90	1.40	2.00	10.60
Do.			do.	0.20	1.00	0.30	0.30	0.70	2.30
(Southern)			do.	0.07	0.49	0.12	0.17	0.51	1.20
Do.			do.	0.08	0.55	0.17	0.16	0.56	1.44
Do.			do.	0.08	0.43	0.18	0.13	0.41	1.15
			Whitefish	0.15	0.20	0.11	0.08	0.38	0.83
			do.	0.15	0.23	0.12	0.07	0.33	0.75
			do.	0.12	0.26	0.13	0.08	0.44	0.91
(Northern)			do.	0.21	0.36	0.20	0.15	0.86	1.57
Do.			do.	0.25	0.47	0.20	0.12	0.58	1.37
			Kokanee	0.20	0.50	0.10	0.10	0.60	1.30
(Northern)			Yellow pickerel	0.01	0.19	0.04	0.55	0.18	0.47
(Georgian Bay)			do.	0.15	1.68	0.28	0.46	1.87	4.29
(Northern)			Sturgeon	—	0.32	—	0.12	0.18	0.62
(Georgian Bay)			Yellow perch	0.03	0.43	0.14	0.14	0.75	1.46
Do.			Mulletts	0.14	0.90	0.26	0.38	1.45	2.99
(Northern)			Chub	0.31	0.72	0.46	0.31	1.59	3.08
(Georgian Bay)			Rainbow trout	0.17	0.60	0.32	0.22	0.88	2.02
Lake Erie	42° 53'	78° 56'	Mulletts	0.04	0.18	0.01	0.18	0.13	0.50
			do.	0.03	0.17	0.02	0.18	0.13	0.50
(Western)			Perch	0.04	0.14	0.01	0.14	0.13	0.49
Do.			do.	0.04	0.16	0.02	0.17	0.14	0.92
			Catfish	0.11	0.49	0.13	0.73	0.33	1.68
			Carp	0.08	0.51	0.02	0.37	0.06	0.96

TABLE 1.—Pesticide residue levels in fish, by provinces and waters, Canada—1970—Continued

LAKE OR RIVER ¹	LATITUDE AND LONGITUDE		FISH SPECIES	PESTICIDE RESIDUE LEVELS (PPM)					TOTAL DDT
				DIELDRIN	p,p'-DDE	o,p'-DDI	p,p'-DDD	p,p'-DDT	
PROVINCE OF ONTARIO—Continued									
Lake Erie (Cont'd)			Alewife Sheepshead	0.56 0.06	0.17 0.27	— 0.03	— 0.35	0.18 0.27	0.34 0.92
Lake Ontario	43° 45'	78° 00'	Pickeral do. do. Rockbass Lake herring Crappies Pike Perch Whitefish	0.06 0.07 0.08 0.01 0.28 0.02 — 0.15 0.37	0.71 0.53 0.55 0.07 2.64 0.17 — 0.67 0.65	0.20 0.21 0.19 — 0.58 0.28 — — —	0.35 0.33 0.40 0.04 0.43 0.12 — 0.42 0.67	0.37 0.41 0.40 0.02 2.84 0.12 — 0.69 0.57	1.63 1.48 1.47 0.13 6.49 0.45 — 1.78 1.89
Lake Michigan	43° 45'	87° 00'	Alewife Chub	0.14 0.03	3.72 2.50	0.37 0.68	0.56 0.44	1.19 3.00	4.28 6.62
Lake Superior	48° 00'	88° 00'	Smelt Lake trout	— 0.06	0.16 0.68	— 0.11	— 0.16	T 0.36	0.16 1.32
St. Clair	47° 14'	84° 00'	Coho Pickeral	0.03 0.10	0.12 1.11	0.05 0.12	0.05 0.58	0.15 1.09	0.37 2.90
Sturgeon River	46° 19'	79° 58'	Mullet	0.01	0.14	0.05	0.20	0.52	0.91
Nipigon	49° 50'	88° 30'	Northern pike Pickeral Whitefish Tullibee	— — 0.05 0.06	0.23 0.20 0.80 0.80	0.04 0.04 0.15 0.28	0.04 0.03 0.05 0.43	0.14 0.09 0.47 0.92	0.45 0.36 1.47 2.45
Lake of the Woods	49° 00'	94° 50'	Whitefish do.	0.01 0.03	0.12 0.16	0.01 0.04	0.05 0.08	0.02 0.05	0.20 0.33
Ottawa River	45° 34'	74° 23'	Crappies Bullheads do. Sturgeon	0.01 0.03 0.03 0.52	0.11 0.08 0.08 0.82	0.02 — — 0.51	0.02 0.95 0.95 0.89	0.05 0.67 0.67 1.14	0.20 0.24 0.24 3.36
St. Lawrence	45° 20'	73° 58'	Yellow perch Bullheads	— 0.03	0.01 0.88	— —	0.80 0.24	0.01 0.05	0.23 0.38
Lake St. Francis	45° 08'	74° 25'	Yellow perch	0.03	0.05	—	0.04	0.06	0.15
Clay	50° 05'	93° 30'	Pike Whitefish	0.01 0.07	0.02 0.02	— —	0.02 0.02	0.07 0.06	0.17 0.10
PROVINCE OF MANITOBA									
Winneposis (Dawson Bay) (N) (S) (Duck Bay) (Dawson) (N) (Duck Bay) (S) (S) (N) (Duck Bay) (N) (S) (Duck) (Dawson)	52° 30'	100° 00'	Goldeye do. Mullet do. do. do. Sauger do. do. Northern pike do. do. do. Pickeral do. do. do.	T 0.01 0.01 T T T T T T 0.01 T T T T T T	0.01 0.02 0.01 0.01 T 0.01 T T T 0.01 0.01 T T 0.02 0.01 0.01	T 0.02 T T — T T T 0.01 T T T T T T T	T 0.01 T T T T T T 0.01 T T T T T T T	— — — — T T T T 0.01 T T T T T T —	0.01 0.05 0.01 0.01 T T T T 0.01 0.04 0.01 T T 0.02 0.02 0.01
God's	50° 40'	94° 15'	Pike Walleye	— —	— —	— —	— —	— —	— —
Reindeer	57° 20'	102° 00'	Lake trout	—	—	—	—	—	—
Family	51° 54'	95° 27'	Walleye Pike	— —	T T	— —	— —	— —	T T
Fishing	52° 08'	95° 24'	Walleye	—	T	—	—	—	T
Winnipeg (Southern) (Northern) (Southern)	52° 00'	97° 00'	Pike Sauger Northern pike do.	0.01 0.02 0.01 0.01	0.15 0.11 0.05 0.19	0.01 0.03 0.01 0.02	0.09 0.09 0.01 0.12	0.05 0.02 0.02 0.08	0.30 0.25 0.09 0.41

TABLE 1.—Pesticide residue levels in fish, by provinces and waters, Canada—1970—Continued

LAKE OR RIVER 1	LATITUDE AND LONGITUDE	FISH SPECIES	PESTICIDE RESIDUE LEVELS (PPM)					TOTAL DDT
			DIELDRIN	p,p'-DDE	o,p'-DDT	p,p'-DDD	p,p'-DDT	
PROVINCE OF MANITOBA—Continued								
Winnipeg (Cont'd)		Pickere1	0.01	0.04	0.01	0.01	0.01	0.07
(Northern)		do.	0.01	0.16	0.02	0.04	0.31	0.53
(Southern)		Whitefish	0.05	0.06	0.01	0.02	0.02	0.11
(Northern)		do.	0.01	0.08	0.03	0.06	0.09	0.26
(Southern)		Perch	T	0.18	0.01	0.10	0.04	0.33
Do.		Sheepshead	0.02	0.18	0.03	0.07	0.06	0.34
		Tullibee	0.03	0.12	0.07	0.11	0.07	0.37
Manitoba	51° 00' 98° 45'	Mullet	T	T	T	T	T	T
		Sauger	T	0.03	—	0.01	T	0.04
		do.	T	0.03	—	0.01	T	0.04
(Northern)		Northern pike	T	0.02	T	0.02	T	0.04
(Western)		do.	—	0.02	T	0.01	T	0.03
(Southern)		Pickere1	—	0.02	0.01	—	—	0.03
(Western)		do.	T	0.03	0.01	0.01	0.01	0.06
St. Martin	51° 37' 98° 29'	Mullet	T	0.02	T	0.01	T	0.03
		Northern pike	T	0.02	T	T	0.01	0.03
		Pickere1	T	0.02	T	T	0.01	0.03
		Perch	T	0.02	T	0.01	T	0.03
Dauphin	51° 17' 99° 48'	Mullet	T	0.02	—	0.01	T	0.03
		do.	T	0.02	—	0.01	—	0.03
		Northern pike	T	0.02	—	0.01	—	0.03
		do.	T	0.01	T	0.01	—	0.02
		Pickere1	T	0.02	T	0.01	—	0.03
		do.	T	0.02	0.01	0.01	T	0.04
Lockport	50° 05' 96° 56'	Maria	T	0.03	T	0.03	0.02	0.08
South Indian	57° 10' 98° 30'	Pickere1	T	0.01	T	T	T	0.01
Dogskin	51° 43' 95° 12'	Pickere1	T	T	—	—	T	T
Cedar	49° 55' 96° 27'	Northern pike	T	0.05	T	0.02	0.03	0.10
Clear	50° 41' 100° 00'	Pickere1	—	0.10	—	0.08	0.17	0.34
Summerberry River	53° 23' 100° 22'	Pike	T	T	—	—	—	T
PROVINCE OF SASKATCHEWAN								
Dillon	55° 45' 109° 30'	Northern pike	—	—	—	—	—	—
Buffalo Narrows	55° 51' 108° 29'	Pickere1	T	T	T	—	T	T
Pelican	50° 32' 106° 00'	Whitefish	T	0.01	0.01	0.01	—	0.03
Montreal	54° 20' 105° 40'	Whitefish	0.01	0.01	0.02	0.01	—	0.04
Jackfish	51° 39' 101° 35'	Whitefish	0.01	0.02	0.03	0.01	—	0.06
Last Mountain	51° 05' 105° 14'	Whitefish	0.01	0.01	—	T	—	0.01
Primrose (Long Bay)	54° 55' 109° 45'	Whitefish	0.01	0.25	0.05	0.06	0.19	0.55
		do.	T	0.05	0.01	0.04	0.05	0.15
Pinehouse	55° 32' 106° 35'	Whitefish	T	0.01	0.01	T	—	0.02
Weyakwin	54° 30' 106° 00'	Whitefish	T	0.01	0.01	T	—	0.02
Lac La Ronge	55° 04' 105° 19'	Whitefish	T	0.01	0.01	T	—	0.02
Ile La Cross	55° 40' 107° 45'	Whitefish	—	0.01	0.02	—	—	0.03
Descharme	57° 05' 109° 13'	Whitefish	0.01	T	—	0.01	—	0.01
Wicken Camp	57° 29' 109° 38'	Whitefish	T	T	T	T	T	T
North Sask. River	53° 15' 105° 05'	Burbot	T	0.20	—	0.01	0.01	0.04
		do.	T	0.10	—	0.01	0.01	0.03
		do.	T	0.10	—	0.01	0.01	0.03
		do.	—	0.02	—	0.19	0.19	0.40
		Goldeye	—	0.22	0.29	0.22	0.22	0.95
		do.	0.08	0.09	0.06	0.10	0.10	0.43
Athabasca	59° 15' 109° 15'	Pickere1	—	T	—	—	—	T
Utikumak	54° 50' 108° 14'	Whitefish	—	0.01	0.01	—	—	0.01

TABLE 1.—Pesticide residue levels in fish, by provinces and waters, Canada—1970—Continued

LAKE OR RIVER ¹	LATITUDE AND LONGITUDE		FISH SPECIES	PESTICIDE RESIDUE LEVELS (PPM)					TOTAL DDT
				DIELDRIN	p,p'-DDE	o,p'-DDT	p,p'-DDD	p,p'-DDT	
PROVINCE OF ALBERTA									
Bellis	54° 09'	112° 09'	Goldeye do.	0.01 0.02	0.12 0.23	0.02 0.02	0.08 0.15	0.06 0.12	0.28 0.52
St. Paul	59° 59'	111° 17'	Goldeye	0.02	0.60	—	0.55	0.79	1.94
			Burbot	T	0.04	—	0.02	0.03	0.09
			do.	T	0.02	—	0.01	0.02	0.05
			do.	T	0.02	—	0.02	0.02	0.06
			Walleye	T	0.40	0.30	0.20	0.20	1.1
			Pike	—	0.40	0.08	0.22	0.25	0.95
Pickereel	0.01	0.04	—	0.03	0.03	0.10			
Sturgeon River	53° 46'	113° 10'	Whitefish	0.03	0.08	0.08	0.08	0.23	0.47
			Goldeye	0.06	0.37	0.48	0.28	1.08	2.21
			Pickereel	0.01	0.04	—	0.03	0.05	0.12
Kinnaird	54° 47'	111° 31'	Northern Pike	T	T	0.01	—	—	0.01
Myrnam	53° 40'	111° 14'	Pickereel	0.01	0.06	—	0.04	0.05	0.15
			do.	0.01	0.05	—	0.04	0.05	0.14
Cold	52° 18'	112° 41'	Tullibee	0.01	0.34	0.02	0.02	0.10	0.48
			Whitefish	0.02	0.89	0.08	0.09	0.70	1.76
Lac La Biche	54 46'	111° 58'	Whitefish	—	0.01	0.01	—	—	0.02
Kackson			Whitefish	0.01	T	—	0.01	—	0.02
Bow River	49° 51'	111° 41'	Trout eggs	0.92	0.57	—	0.39	0.37	1.33
			Rainbow trout	0.05	0.50	—	—	0.32	0.87
			do.	0.01	0.88	0.03	0.04	0.05	1.00
			do.	0.05	0.35	0.29	0.26	0.43	1.33
Whitemud Creek	53° 27'	113 33'	Northern sucker	0.01	0.07	0.02	0.06	0.09	0.24
NORTHWEST TERRITORIES									
Great Slave	61° 23'	115° 38'	Whitefish	0.01	0.01	T	T	T	0.01
Hjalmar	61° 33'	109° 25'	Whitefish	0.01	0.03	—	—	—	0.03
			Lake trout	T	0.03	—	—	—	0.03
Nonacho	61° 42'	109° 40'	Whitefish	T	T	—	—	T	T
Rutledge	61° 33'	110 47'	Whitefish	0.01	0.01	0.01	T	T	0.02
			Lake trout	T	0.02	—	—	—	0.02
			do.	—	T	—	—	—	T
			Muktuk	0.07	0.16	0.05	0.10	0.10	0.41
Merkley	69° 45'	107° 40'	Whitefish	0.01	T	T	T	—	T
Gymer			Whitefish	0.01	0.01	0.01	T	T	0.02
Gordon	63 10'	113° 12'	Trout	0.01	0.05	0.03	0.01	0.02	0.11
			Whitefish	0.03	0.10	—	0.04	0.05	0.19
Mackay	63° 55'	110 25'	Whitefish	0.02	0.08	0.03	0.02	0.03	0.16
Cambridge Bay	68° 03'	105 05'	Arctic char	T	T	—	—	—	T
			do.	—	—	—	—	—	—
			Whale	—	—	—	—	—	—
Baker	64° 00'	96° 00'	Whitefish	—	—	—	—	—	—
Kaminak	62° 10'	95° 00'	Lake trout	—	—	—	—	—	—
Jackson	62° 35'	114° 18'	Whitefish	T	—	—	0.01	—	0.01

¹ Specific location within waters, if known, given in parentheses.

Residues of Organochlorine Pesticides,, Polychlorinated Biphenyls, and Mercury in Bald Eagle Eggs and Changes in Shell Thickness—1969 and 1970

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ABSTRACT

Twenty-three bald eagle eggs collected in Alaska, Maine, Michigan, Minnesota, and Florida during 1969 and 1970 were analyzed for organochlorine pesticides, polychlorinated biphenyls, and mercury. All eggs contained residues of DDE, dieldrin, PCB's, and mercury. Average residue concentrations were lowest in eggs from Alaska. Significant eggshell thinning has occurred among eggs from most major areas sampled. Some eggs contained DDE residues of the same magnitude as those that produced shell thinning in experimental species. High dieldrin residues in some eggs could be having an adverse effect on reproductive success.

Introduction

Bald eagle (*Haliaeetus leucocephalus*) populations and the reproductive success of this species have declined in many areas of the United States within the last 20 years (1,3,32). Several authors have related the decline of eagles and of other species of raptorial and fish-eating birds at the top of food chains to the adverse effects of organochlorine pesticides that are widespread in the environment (6,7,11,26,27). A reduction in eggshell thickness since the introduction and widespread use of organochlorine pesticides has been shown for a number of bird species (7,26,27); the adverse effects of DDT and dieldrin in combination and of DDE on reproductive success of a raptorial species have been shown

experimentally (23,35). Previous studies of field-collected bald eagles and their eggs have reported organochlorine or heavy metal residues (9,22,28,33), causes of mortality (4,22), and mortality due to pesticide poisoning (4,22,29,33).

This paper reports the results of analyses for organochlorine pesticides, polychlorinated biphenyls (PCB's), and mercury residues in bald eagle eggs collected in Alaska, Maine, Michigan, Minnesota, and Florida during 1969 and 1970. Eggshell characteristics and reproductive success are discussed in relation to these residues.

Sampling Procedures

One egg was collected from each of 15 nests during the normal incubation period: 11 of the nests were located in Alaska, 3 in Maine, and 1 in Michigan. Eight additional eggs collected after the normal incubation period included five eggs from four nests in Minnesota, two eggs from nests on the west coast of Florida, and one egg from a nest on Kodiak Island, Alaska.

Following collection in the field, each egg was usually wrapped in aluminum foil, individually packed in a container, and shipped to the Patuxent Wildlife Research Center. The length and breadth of each egg was measured in millimeters, and the volume of some eggs was measured by water displacement. The eggs were opened, and the age of embryos was estimated based on a 35-day incubation period. Egg contents were frozen prior to chemical analysis. Shells were air dried, and shell thickness was measured as previously described (9).

Chemical Analysis

Eggs were analyzed individually for residues of organochlorine pesticides, polychlorinated biphenyls (PCB's), and mercury. Each egg was mixed in an Omnimixer; a

¹Bureau of Sport Fisheries and Wildlife, Patuxent Wildlife Research Center, Laurel, Md. 20810

²National Audubon Society, Corkscrew Swamp Sanctuary, Immokalee, Fla. 33934

³Bureau of Sport Fisheries and Wildlife, Kodiak National Wildlife Refuge, Kodiak, Alaska 99615. Present address: Bureau of Sport Fisheries and Wildlife, Alaska Area Office, 6917 Seward Highway, Anchorage, Alaska 99502

⁴Forest Service, Chippewa National Forest, Cass Lake, Minn. 56633

⁵Bureau of Sport Fisheries and Wildlife, P.O. Box 1287, Juneau, Alaska 99801

⁶Department of Wildlife Ecology, University of Wisconsin, Madison, Wis. 53706.

20-g aliquot was removed for pesticide and PCB analysis; and a 5-g aliquot was removed for mercury analysis. The 20-g aliquots were ground with anhydrous sodium sulfate and extracted for 7 hours with hexane in a Soxhlet apparatus. Extracts were cleaned up and then divided into halves; one half was used for pesticide analysis and the other saved for PCB analysis.

For pesticide analysis, each cleaned extract was spotted on a thin layer plate, developed, and removed in four fractions as described by Mulhern *et al.* (22). All four fractions were analyzed separately by electron capture gas chromatography on a 3.8% UCW-98 on 80/100 Diatoport S column, and DDT residues in Fractions III or IV were confirmed on a 3% XE-60 column. The operating parameters for the UCW-98 and XE-60 columns are given in Table 1. Average recovery by this method was 85-96% with a detection limit of approximately 0.05 ppm. Residues were not corrected for recovery. In addition to zonal separation by thin layer chromatography (TLC) and analysis on two columns, residues in 20% of the samples were confirmed by TLC (AgNO₃ incorporated Al₂O₃ plate—5% benzene in hexane solvent).

TABLE 1.—Chromatographic operating conditions using electron capture detection for pesticide analysis

	COLUMNS, GLASS 14" O.D.	
	A	B
Column length	4 feet	6 feet
Liquid phase	3.8% UCW-98	3% XE-60
Support	Diatoport S	Gas Chrom Q
Mesh size	80/100	60-80
Column flow rate	60 ml/min	100 ml/min
Purge	40 ml/min	—
Gas	5% methane/argon	nitrogen
Temperature	200° C	170° C
Retention time of dielrdrn	8.6 minutes	16.3 minutes

PCB analysis was by TLC as described by Mulhern *et al.* (21).

Mercury analyses were for total mercury by a method developed by R.S. Christensen (*personal communication*). The procedure involves acid digestion of the tissue and extraction of the mercury from the liquid digest with dithizone (20). Mercury determination was made on the dithizone extract by flame atomic absorption spectrophotometry using the "sampling boat" technique (8). The average recovery from tissue samples fortified with both inorganic and organic mercury compounds was 91%. The limit of detection was 0.05 ppm.

The residue concentrations were calculated as micrograms per milliliter on the basis of total egg volume. This is converted to a ppm basis assuming a specific gravity of 1.0 as described by Stieckel *et al.* (33).

Measurements of length and breadth were used to estimate the volume of those eggs whose volumes were not determined by water displacement. The equation for this estimate was volume (ml) = 3.73 X length (cm) X breadth (cm) — 35.3 (L. F. Stieckel, S. N. Wiemeyer, and L. J. Blus, *manuscript in preparation*).

Collection Dates and Embryonic Development of Samples

Six eggs were collected from Kodiak Island between May 8 and 26, 1969; two were fresh with no signs of embryonic development while four contained 7- to 23-day-old embryos. An additional egg from Kodiak Island (Karluk Weir), collected on July 21, 1969, after it failed to hatch during the normal incubation period, was decomposed with no evidence of embryonic development; a small portion of this egg's contents may have been lost before it was opened.

Five eggs from the Admiralty Island area, Alaska, collected between May 13 and 15, 1970, contained embryos estimated to be more than 15 days old. One (Tiedman Island) was decomposed and had lost a small portion of its contents before it was opened.

Three eggs from nests in Maine were collected on April 14 and 15, 1969. One (Dyer Neck) was decomposed with no signs of embryonic development; however, the other two contained embryos 15 to 32 days old.

The single egg from Baraga County, Michigan, collected on April 14, 1969, appeared fresh and exhibited no visible signs of embryonic development.

Two eggs from Minnesota (Star Island, Rabideau Lake), collected on June 4 and 6, 1969, contained embryos 8 to 20 days old. Two additional eggs were collected from one nest in Minnesota (Six Mile Lake) on May 14, 1970; one contained a decomposed embryo 5 to 10 days old, and the added contents of the other contained no evidence of embryonic development. An additional egg from Minnesota, collected on June 5, 1970, exhibited no indication of embryonic development in its added contents.

Two eggs collected in Lee County, Florida, on March 7 and May 2, 1969, were badly dehydrated and contained no signs of embryonic development.

Results

Results of the analyses for organochlorine pesticides, PCB's, and mercury are shown in Table 2. Average residue concentrations of organochlorines in bald eagle eggs from Alaska were the lowest found for any area in the United States to date. DDE residues in the eggs from the Admiralty Island area, excluding the one egg from South Midway Island, averaged only 0.95 ppm.

TABLE 2.—Organochlorine pesticides, polychlorinated biphenyls, and mercury in bald eagle eggs collected in Alaska, Maine, Michigan, Minnesota, and Florida in 1969 and 1970
(ND = not detected; T = <0.05 ppm)

NEST LOCATION	SHELL THICKNESS (MM) ¹	EGG VOLUME (ML) ²	RESIDUE CONCENTRATION IN PPM						
			p,p'-DDE	p,p'-DDD	p,p'-DDT	DIELDRIN	HEPTACHLOR EPOXIDE	PCB'S	MERCURY
ALASKA—KODIAK 1969									
Island Point	0.51	[134]	1.60	0.07	ND	0.09	0.02	1.9	0.2
Bird Island	0.52	[136]	4.93	0.23	ND	0.30	0.06	5.4	0.2
Spiridon Bay	0.52	[144]	1.43	0.07	ND	0.06	0.01	1.7	0.2
Grassey Point	0.53	[129]	1.89	0.21	ND	0.12	0.02	1.8	0.1
Uganik Island	0.60	[128]	1.28	0.06	ND	0.06	0.02	1.7	0.3
Fraser Lake	0.58	[126]	1.16	0.05	ND	0.02	0.01	1.6	0.1
Karluk Weir	0.59	[123]	1.15	0.14	ND	0.05	0.01	1.4	0.1
Average	0.55		1.92	0.12	ND	0.10	0.02	2.2	0.2
ALASKA, ADMIRALTY ISLAND AREA—1970									
Tiedman Island	[0.60]	102	0.83	T	ND	T	ND	0.85	0.1
Fools Inlet	0.60	103	1.81	T	ND	T	ND	0.84	0.3
South Midway Island	[0.55]	125	10.75	T	ND	0.22	ND	2.3	0.2
Windfall Harbor	0.68	151	0.25	T	ND	0.02	ND	0.43	0.2
Swan Island	0.60	133	0.91	T	ND	0.03	T	0.88	0.2
Average ³	0.61		2.91	T	ND	0.06	T	1.1	0.2
MAINE—1969									
Franklin	0.49	142	11.86	0.55	0.24	0.22	0.02	4.9	0.3
Dyer Neck	[0.47]	129	20.55	0.84	0.49	0.29	0.03	15.2	0.3
Boydton Pond	0.59	131	12.44	0.35	0.25	0.15	0.02	9.7	0.4
Average	0.52		14.95	0.58	0.33	0.22	0.02	9.9	0.3
MICHIGAN—1969									
Van Zellen's Camp	0.51	112	39.46	1.12	ND	1.02	0.07	27.7	0.4
MINNESOTA—1969 AND 1970*									
Star Island	0.48	[130]	21.62	2.42	ND	2.29	0.17	12.4	0.3
Rabideau Lake	0.61	[120]	3.68	0.91	ND	0.63	0.11	8.0	0.3
Cass Lake	0.55	123	8.15	0.48	ND	0.62	T	6.2	0.3
Six Mile Lake (1)	0.53	[124]	2.37	0.07	ND	0.16	ND	2.2	0.2
(2)	0.55	[135]	7.27	0.42	ND	0.65	T	5.8	0.3
Average	0.55		9.57	1.02	ND	0.99	0.08	7.7	0.3
FLORIDA—1969									
Lee County—1	0.61	[104]	18.21	1.13	0.19	0.68	0.02	13.3	0.3
Lee County—3	0.52	[98]	18.52	2.49	0.29	1.54	0.07	11.1	0.7
Average	0.57		18.37	1.81	0.24	1.11	0.05	12.2	0.5

¹Shell thicknesses in brackets may not be totally accurate due to disruption and/or loss of the shell membranes.

These data have been excluded from Table 3 and from Discussion in the text.

²Volumes in brackets computed as stated in text.

³Trace considered to equal 0.025 ppm when computing averages.

*Parentheses designate different eggs from the same nest. Average computed on a nest basis.

Residues of dieldrin averaged 0.06 ppm in all of the eggs from the Admiralty Island area, while DDD was present in only trace amounts. Residues in eggs from Kodiak Island were slightly higher, averaging 1.92 ppm DDE, 0.12 ppm DDD, and 0.10 ppm dieldrin.

Residues in eggs from Maine collected in 1969 averaged 0.22 ppm dieldrin, 14.95 ppm DDE, 0.58 DDD, and 0.33 ppm DDT. Residues in Maine eggs collected in 1967 and 1968 averaged somewhat higher and contained 1.63 ppm dieldrin, 21.01 ppm DDE, 0.98 ppm DDD, and 0.50 ppm DDT (9).

The concentration of organochlorine pesticides in the eggs from Chippewa National Forest, Minnesota, in 1969 and 1970 was generally similar to that found in eggs collected in Wisconsin in 1968 (9), with the exception of the egg from Star Island collected in Minnesota in 1969, which contained much higher residue concentrations. Excluding the Star Island egg, the residues in the eggs from Minnesota (on a nest basis) averaged 5.55 ppm DDE, 0.55 ppm DDD, and 0.55 ppm dieldrin. The single egg from the Michigan shore of Lake Superior contained an unusually high concentration of DDE and a moderate concentration of dieldrin.

TABLE 3.—Bald eagle eggshell parameters for eggs collected in 1968 to 1970 and changes from pre-1946 norms

AREA	MEAN EGGSHELL THICKNESS ¹ (MM)	PERCENT CHANGE FROM PRE-1946 NORMS ²	MEAN SHELL WEIGHT ¹ (G)	PERCENT CHANGE FROM PRE-1946 NORMS ²	MEAN THICKNESS INDEX ^{1,2}	PERCENT CHANGE FROM PRE-1946 NORMS ²
Alaska—Kodiak	0.5500 (7)	-10.4**	12.460 (6)	-11.4**	2.770 (6)	-14.1**
Alaska—Admiralty	0.6267 (3)	-2.2	15.715 (2)	—	3.345 (2)	—
Great Lake States	0.5486 (14)	-10.2**	11.617 (11)	-10.8*	2.777 (11)	-12.1**
Florida	0.5183 (6)	-11.3*	9.650 (6)	-19.1**	2.567 (6)	-17.2**
Maine	0.5425 (4)	-11.0*	12.830 (2)	—	2.750 (2)	—

NOTE: Sample size is given in parentheses.

All parameters for all eggs could not be used because of the presence of the following conditions: loss and or disruption of shell membranes, loss of portion of shell, residue of egg contents adhering to shell surfaces.

The current samples of eggs may not be entirely random.

¹ Means are calculated on a nest basis; data are, in part, from Krantz *et al.* (9).

² Pre-1946 data from D. W. Anderson and J. J. Hickey (*unpublished*).

³ Ten times shell weight (g) divided by the product of the length and breadth (cm) as devised by Ratcliffe (26).

⁴ Sample variances unequal; method of determining significance level from Snedecor and Cochran (37).

* Significant change from pre-1946 norm $P < 0.01$; T-test.

** Significant change from pre-1946 norm $P < 0.001$; T-test.

DDE in the eggs from Lee County, Florida, averaged 18.37 ppm, which was considerably higher than the 10.72 ppm average in the eggs from Everglades National Park in 1968 (9). Dieldrin residues in eggs from Lee County were 0.68 and 1.54 ppm, about two and six times the highest level found in the Everglades eggs, which ranged from 0.11 to 0.28 ppm (9).

Residues of PCB's were lower than residues of DDT plus its metabolites for most eggs in all areas except for the majority of eggs taken from Kodiak, Alaska. The highest concentration of PCB's occurred in the egg from Michigan. Average PCB concentrations in descending order were: Florida 12.2 ppm, Maine 9.9 ppm, Minnesota 7.7 ppm, Kodiak, Alaska 2.2 ppm, and Admiralty, Alaska 1.1 ppm.

Mercury concentrations were uniformly low in the eggs sampled and averaged 0.2-0.3 ppm, with the exception that a higher concentration (0.7 ppm) occurred in one of the two eggs from Florida. Borg *et al.* (2) found reduced hatchability of pheasant (*Phasianus colchicus*) eggs that contained 1.3 to 2.0 ppm mercury.

Eggshell thickness and weight of bald eagle eggs collected from 1968 to 1970 are compared in Table 3 with measurements of shells of eggs collected prior to the widespread use of organochlorine pesticides (D. W. Anderson and J. J. Hickey, *unpublished data*). Thickness and weight were found to decline significantly ($p < 0.01$) in all areas where sufficient data were available, except for a nonsignificant ($p > 0.05$) change in eggshell thickness for eggs from the Admiralty Island area. Hickey and Anderson (7) reported declines in shell weight of 18.0 and 19.8% for bald eagle eggs collected from two Florida counties since organochlorine pesticides were initially used.

Shells of eggs from individual nests in the recent sample from Kodiak, Alaska, were from 2 to 17% thinner than the pre-1946 norm. Shells from Admiralty, Alaska, were 2% thinner to 11% thicker than the earlier norm. The shells of some eggs from the Lake States were equal in thickness to the pre-1946 norm, and others varied from that to as much as 21% thinner than the earlier norm. Eggshells from Florida were 25% thinner to 4% thicker than the pre-1946 norm, and those from Maine were 3 to 20% thinner than the earlier norm. Only those eggs that could be measured accurately (those used in Table 3) were included in the above data.

Discussion

Data on reproduction of bald eagles in the areas in which eggs were collected are included in Table 4.

Lockie, Ratcliffe, and Balharry (11), in a study of golden eagles (*Aquila chrysaetos*) in west Scotland, found that the proportion of eagles successfully rearing young doubled (from 31% to 69%) following the ban of dieldrin use in sheep dips, the average dieldrin residues in eagle eggs dropped significantly (from 0.87 ppm to 0.38 ppm) during the same period. The low residues of DDE and other organochlorine pesticides, other than dieldrin, in the eggs of the Scottish eagles are not believed to have been a significant factor in reproductive success. Lockie and Ratcliffe (10) earlier correlated reproductive failure with amounts of dieldrin exceeding 1.0 ppm in the eggs of these golden eagles. Potts (25) found a significant correlation between the residues of dieldrin in the eggs of shag (*Phalacrocorax aristotelis*) and reproductive failure; there was a sizeable increase in the percentage of clutches with no chicks surviving to the 10th day when there was more than 2.0 ppm dieldrin in the egg.

TABLE 4.—Reference data on reproduction of bald eagles in the areas in which eggs were collected

LOCATION	YEAR	AVERAGE NUMBER OF YOUNG PER ACTIVE NEST/YEAR	PERCENT OF ACTIVE NESTS IN WHICH YOUNG WERE RAISED	REFERENCE
Karluk Lake, Kodiak National Wildlife Refuge, Alaska	1959, 1961, 1962	1	—	Hensel and Troyer (5)
Kodiak National Wildlife Refuge, Alaska	1963	1.1	66	Troyer and Hensel (34)
Admiralty Island, Alaska	1966	1.4	88	Robards and King (30)
Chippewa National Forest, Minnesota	1963-70	0.8	52	Mathisen (12-18) Mathisen and Stewart (19)
Maine	1965	—	18	Sprunt and Ligas (32)
Florida (west coast)	1965	—	45	Sprunt and Ligas (32)
Michigan (shores of Great Lakes)	1967	—	6	Postupalsky (24)

One-half of the bald eagle eggs from Maine that have been analyzed, as well as single eggs from Michigan, Minnesota, Wisconsin, and Florida have contained more than 1.0 ppm dieldrin—see also Krantz *et al.* (9). If the results of Lockie *et al.* (11), regarding the adverse effects of dieldrin on golden eagle reproduction, are also applicable to reproductive success in bald eagles, then dieldrin could be a factor in bald eagle reproductive success in these areas.

The DDE residues in a few of the eggs reported here, as well as in a number of eggs reported previously (9), especially many of those from Maine, were similar in magnitude to those that have produced shell thinning in experimental sparrow hawks (*Falco sparverius*) fed a low dietary level of DDE (35).

Average declines in shell thickness, as expressed by shell weight or thickness index, greater than 17% have been accompanied by severe declines in populations and/or reproductive success in several species of raptorial birds (7,26,27); declines in shell thickness are commonly associated with an increased frequency of egg breakage (7,23,26,27). The decline in shell weight and thickness index of bald eagle eggs from Florida equalling that of some declining raptor populations, the presence of eggshell fragments in two Kodiak, Alaska, nests at the time of the egg collection, and the moderate declines in shell thickness for most of the areas sampled induce considerable concern regarding the effects that shell thinning may be having on reproductive success and status of these bald eagle populations.

Conclusion

DDE residues in some eggs were of a magnitude that has produced shell thinning in experimental studies of other species. Significant shell thinning in most areas sampled causes concern for ultimate effects on reproductive success and populations. Dieldrin residues in the eggs from some areas could have an adverse effect on reproductive success.

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See Appendix for chemical names of compounds discussed in this paper.

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PESTICIDES IN WATER

*Organochlorine Pesticide Residues in Water, Sediment, Algae, and Fish, Hawaii—1970-71*¹

Arthur Bevenue, John W. Hylin, Yoshihiko Kawano, and Thomas W. Kelley

ABSTRACT

Rainwater, drinking water, and nonpotable waters in Hawaii were sampled and found to contain chlorinated insecticide residues in the low parts-per-trillion range. Dieldrin, p,p'-DDT, and lindane were the pesticides most prevalent; pentachlorophenol was present in samples from a sewage fallout. The ratio of chlorinated pesticide residues in canal waters to residues in algae, sediment, and fish from the same canals was 1:4,000:9,000:32,000, respectively. According to proposed water quality standards, results of this study indicated that pollution of Hawaii's water by organochlorine pesticides does not occur to any significant degree.

Introduction

The study reported here was conducted to determine the extent of organochlorine pesticide contamination of water, sediment, algae, and fish in the State of Hawaii, which consists of eight major islands: in order of descending size, they are Hawaii, Maui, Oahu, Kauai, Molokai, Lanai, Niihau, and Kahoolawe. These islands comprise an area of 6,425 square miles (1-3).

The Hawaiian Islands have an abundance of potable water due to moisture-laden trade winds which blow southerly from a string of high-pressure areas to the north of the Island chain. As the trade winds strike the mountains of the Islands, rise in altitude, and are cooled, the water vapor is converted to rain. The amount of rainfall is related to the heights of the mountains. For example, the islands of Niihau and Kauai are at approximately equal latitudes; however, Niihau at an elevation of 1,281 feet has 26 inches of rainfall while Kauai at 5,170 feet has up to 482 inches. The unique formation of the Islands trap and store this rainwater:

i.e., marine sediments and alluvial and talus sediments, deposited through the ages, have formed a relatively impermeable caprock around the Islands. At the highest elevations there may also be impermeable faults or dikes which impede the flow of rainwater—generally, however, rain falling at high elevations is rapidly absorbed into the porous volcanic basalt and percolates through the mountains to the water table where it accumulates to heights above sea level because of the caprock. Since fresh water is less dense than sea water, the fresh water in the water table literally floats on the sea water which has permeated the basalt. This relationship between the two waters was first discovered by Ghyben-Herzberg and is known as the Ghyben-Herzberg lens.

Because the major part of the State's population resides on Oahu and also for convenience, this Island was sampled much more extensively than the other Islands of the State. Subterranean Oahu may be classed as a water bank. The Board of Water Supply of the City and County of Honolulu taps this zone of fresh water with skimming tunnels, shafts, and artesian wells (3). It has been conservatively estimated that Oahu's current sources of potable water are sufficient to meet anticipated needs until the year 2000; more optimistic reports predict that these sources will provide sufficient water to meet demands over the next 90 years. About 1.8 billion gallons of water, as rain, falls on Oahu each day, 700 million gallons of which returns to the basin. At this time, Oahu uses about 430 million gallons each day.

Some areas of Oahu are only a few feet above sea level, and canals have been constructed to eliminate swamp-like areas and to receive surface runoff and storm-drain waters which eventually drain into the sea; much of this drainage into the canals originates from either industrial or residential areas.

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Hawaii produces about 75% of the world's pineapple crop and 25% of the United States' domestically grown sugar crop. Other agricultural activities include the commercial production of macadamia nuts, papaya, coffee, guava, passion fruit, avocados, bananas, taro, vegetable crops, beef cattle, dairy products, poultry, and swine. The sugar companies apply no insecticides to the land used for sugarcane production, but use primarily herbicides and rodenticides. The pineapple growers use insecticides, herbicides, and nematocides. Both use small amounts of plant growth regulators. Pasture lands are treated regularly with herbicides to control heavy brush foliage. In the other agricultural areas, the entire spectrum of pesticides is used. Termites are a constant problem in the islands, and commercial pest control operators use aldrin, dieldrin, chlordane, heptachlor, and pentachlorophenol in addition to the fumigants Vikane® (sulfuryl fluoride) and methyl bromide for their control. Numerous commercial formulations of insecticides, fungicides, and herbicides are sold locally for use in the average household.

Waters obtained for this study included 10 samplings of rainwater, 45 drinking waters, and 46 nonpotable waters.

Potable waters were sampled from 45 stations, including 28 wells, 4 shafts, 12 tunnels, and 3 springs on Oahu Island (Fig. 1). Nonpotable waters on Oahu were obtained from the Honolulu area on the leeward side and the Kaneohe-Kailua section on the windward side. Several swimming and surfing areas were also sampled—Waialua Bay and Waimea Bay to the northwest and Kahana Bay in the northeastern area. In addition, two harbors, one industrial site, one sewage outfall, and two canals were sampled; sediment samples were also obtained from both canals (Fig. 2), and algae and fish specimens were obtained from one, the Ala Wai Canal. Nonpotable samples from the Island of Hawaii were from two populated areas (Hilo and Kona areas) and one unpopulated area of Punaluu (Black Sand Beach) (Fig. 3). Samplings from Kauai included five estuaries and one freshwater area (Fig. 3). The eight samples from Maui included water from a harbor, a pond, several streams, a golf trap at a resort, and a sump area which emptied excess drainage from a sugarcane field (Fig. 3).

Materials and Methods

WATER AND SEDIMENT

New, 1-gal glass bottles with metal screw caps and teflon liners were used to collect all water samples. Prior to use, these bottles and separatory funnels used in the analytical procedure were rinsed thoroughly with a solution of concentrated sulfuric acid and sodium dichromate, followed by rinsing with water, redistilled acetone, and redistilled hexane. All other glassware used

in the analytical procedure was heat-treated for 16 hours at 200° C in an air-oven prior to use (4).

One-gallon grab samples of potable and nonpotable waters were obtained from various areas of the State, including representative samples of the 50 outlet stations of the drinking water supply for the City and County of Honolulu on the Island of Oahu. Rain-water samples were collected from different areas on Oahu during the period from December 1970 through February 1971.

FIGURE 1.—Sampling sites for drinking waters, Oahu, Hawaii—1970-71

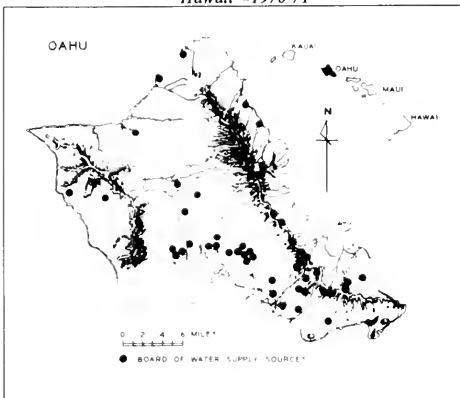


FIGURE 2.—Sampling sites for nonpotable waters, Oahu, Hawaii—1970-71

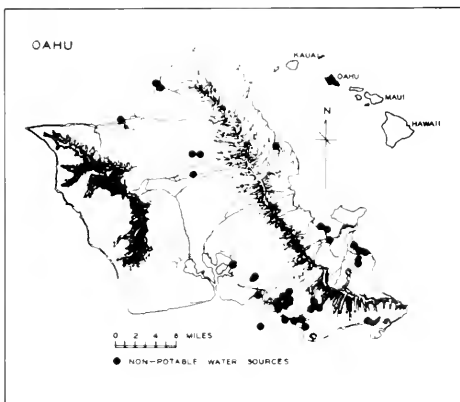


FIGURE 3.—Sampling sites for nonpotable waters from the Islands of Hawaii, Kauai, and Maui—1970-71

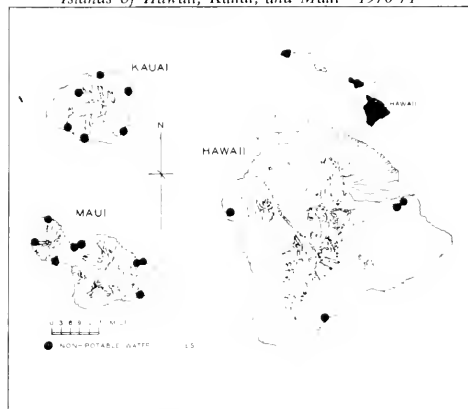


TABLE 1.—pH and chloride ion concentration of water samples, Hawaii—1970-71

WATER SAMPLE	pH (RANGE)	CHLORIDE ION MG LITER (RANGE)
Rainwater	6.6 - 6.8	4 - 7
Potable Water		
Wells and Springs	7.0 - 8.4	15 - 200 (66)
Shafts and Tunnels	6.8 - 9.4	13 - 78 (28)
Nonpotable Waters ¹		
Streams	7.2 - 7.8	350 - 17,150
Ponds	7.3	1,400 - 5,680
Bays	7.4 - 8.1	2,450 - 19,525
Canals	7.1 - 7.8	10,300 - 14,800
Lakes	7.9 - 8.1	12,070 - 14,630
Harbors	6.7 - 8.1	13,000 - 19,460
Basin	7.9	18,970
Lagoon	8.2	19,180

¹One sample of a total of 45 samples had a pH of 9.4 (sample was obtained from a lime-treated station).

²(Average) value.

³Average value for sea water is usually given as 18,980 mg liter.

A portion of each water sample was used to determine pH and chloride ion concentration (Table 1). Total chloride was determined by titration of the acidified water with 0.1N or 0.01N silver nitrate solution and use of a Fisher Ag-AgCl, Model No. 36 titrimer.

Sufficient water was removed from each sample to leave 3,000 ml of water in the bottle. Redistilled hexane (100 ml) was added to each sample, and the bottle was rotated at high speed for 1 hour on a roller-type jar mill. After mixing, each sample was allowed to stand until the hexane and water phases separated. Fifty milliliters of the hexane layer was removed with a volumetric pipette, transferred to a 125-ml round-bottom flask, and concentrated to a small volume using a rotary

flash evaporator. The concentrate was transferred to a calibrated centrifuge tube and the final volume adjusted to 0.5 ml with the aid of a stream of nitrogen. Suitable aliquots, usually 10 μ l, were applied to the gas chromatograph.

The sewage water sample was collected from an outfall discharge over a 24-hour day beginning at 7:00 a.m. December 14, 1970, and ending at 6:00 a.m. December 15, 1970. The sample was obtained at the sluice gate which was the last exposed point in the stream before flowing through the outfall sewer pipe. Samples were obtained hourly and then composited in proportion to hourly flow rates. The extraction procedure for the sewage water was modified in order to isolate any pentachlorophenol (PCP) in the water. Prior to the addition of hexane and the mixing step, the sample was adjusted to pH 13 with sodium hydroxide solution. The alkaline water was extracted three times with 100-ml portions of redistilled hexane in a separatory funnel; the hexane extracts were discarded. The sample was adjusted to pH 2 with concentrated sulfuric acid, 100 ml of redistilled hexane was added, the sample was mixed for 1 hour, and 50 ml of the hexane phase was concentrated as previously described.

The residue was concentrated to near dryness in a centrifuge tube, and excess diazomethane solution was added to convert any PCP in the sample to its ether derivative. Nitrogen was passed over the sample to remove excess diazomethane, the residue was made to a definite volume with hexane, and suitable aliquots were applied to the gas chromatograph.

A second sewage water sample, used for the analysis of chlorinated pesticides other than PCP, was cleaned up prior to analysis using the combined Florisil and silicic acid procedure of Armour and Burke (5). As noted by Armour and Burke, the Aroclors and aldrin are isolated in the first fraction of the silicic acid cleanup; the second fraction contains any remaining chlorinated pesticides except for dieldrin, which would be present in the 15% Florisil fraction. In the present survey, all data were compared with 10 available types of Aroclors. If any polychlorinated biphenyls were present in the examined samples, either they did not relate to the standard Aroclors used for comparison or were not detected; the latter was more often the case.

Sediment samples were obtained from several canals on the Island of Oahu with a dredge sampler or a 1-gal paint can. The solids content of the sediments was determined by air-drying a portion of each sample for 24 hours, followed by drying in an air-oven for 16 hours at 110 C. The wet sediment samples (50-100 g) were mixed thoroughly with equal weights of anhydrous sodium sulfate which had been previously heated 16 hours at 400 C; 100 ml of redistilled hexane was added

to each sample, the mixtures were shaken on a wrist-action shaker for 1 hour, and then allowed to stand until the suspended material settled. A measured amount of the hexane solution was removed and subjected to the Mills' Florisil cleanup procedure (6). The cleaned up extracts were concentrated, and aliquots of the concentrate were applied to the gas chromatograph.

GAS CHROMATOGRAPHY

The following two gas chromatographs were used for analyzing the water and sediment samples: The first was a Varian-Aerograph Model 204, electron capture concentric tritium detector with $1/8'' \times 6'$ borosilicate glass column, with column temperature at 190°C, injector temperature at 270°C, detector temperature at 200°C, carrier gas—nitrogen, and flow rate—25 ml/min. The second was an F&M Model 810, electron capture parallel plate tritium detector with a $1/4'' \times 4'$ borosilicate glass column, with column temperature at 190°C, injector and detector temperatures at 200°C, carrier gas—argon-methane (90:10), and flow rate—75 ml/min. Leeds and Northrup Speedomax H recorders, 1 mv full scale with a chart speed of 0.5" min were used.

Four types of column packing were used, namely:

- (1) 3% SE-30 on Chromosorb W, AW, DMCS, 80/100 mesh;
- (2) 3% QF-1 and 2% DC-200 on Chromosorb W, AW, DMCS, 80/100 mesh;
- (3) 4% SE-30 and 6% QF-1 on Chromosorb W, HP, 80/100 mesh; and
- (4) 1.50% OV-17 and 1.95% QF-1 on Supelcoport, 100/200 mesh.

The retention times of the pesticides on these columns, relative to aldrin, are given in Table 2.

The limits of detectability of the pesticides ranged from about 0.05 ppt (lindane the most sensitive) to 0.5 ppt

(DDT the least sensitive); dieldrin was detected at about 0.2 ppt. This is an approximate range only, because detection limits varied with the degree of purity of the water sample or the degree of success in cleaning up nonpotable water, sediment, and biota samples. The reported residue data are the amounts found by analysis; no attempt was made to correct the data based on recovery values obtained from fortified samples. Repeated attempts to obtain consistent and good recoveries of pesticides from samples spiked at the 1 and 2 ppt levels produced poor results. Efforts on confirmatory analyses by thin layer chromatography (TLC) were confined to samples containing at least 10 ppt of a particular pesticide. Confirmation by the "p" value procedure was difficult, if not impossible, in the residue range studied. Neither TLC nor "p" values were practical when working with residues in the low ppt range. Confirmation procedures were therefore limited to at least two different types of gas chromatograph columns, a practice commonly used by many laboratories especially in preliminary survey work and where the amount of a sample may be limited.

ALGAE AND FISH

The residue data on the algae and fish specimens were provided by Cynthia Schultz of the Department of Oceanography of the University of Hawaii and were obtained from the Ala Wai Canal during the same time period of this investigation. The cleanup procedure for these specimens was a modification of the procedure of Kadoum (7), whereby the extracts, previously partitioned with acetone-hexane, were eluted through a micro-column of silica gel (Woelm, Activity Grade No. 1, activated with 3% water) with a mixture of benzene-hexane (70:30). Analytical data were obtained with a MicroTek Model 220 gas chromatograph utilizing an electron capture detector.

TABLE 2.—Retention times of pesticides in gas chromatographic columns relative to retention of time of aldrin, Hawaii—1970-71

COMPOUND	3% SE-30	3% QF-1 2% DC-200	4% SE-30 6% QF-1	1.50% OV-17 1.95% QF-1
	CHROMOSORB W AW, DMCS 80/100 MESH	CHROMOSORB W AW, DMCS 80/100 MESH	CHROMOSORB W HP 80/100 MESH	SUPELCOPORT 100/120 MESH
Lindane	0.43	0.57	0.54	0.64
Heptachlor	0.77	0.81	0.81	0.81
Aldrin	1.00	1.00	1.00	1.00
Heptachlor epoxide	1.26	1.47	1.43	1.62
<i>p,p'</i> -DDE	1.97	1.92	1.95	2.43
Dieldrin	2.26	2.19	2.19	2.60
<i>p,p'</i> -DDD	2.60	2.79	2.76	3.89
<i>p,p'</i> -DDT	3.46	3.36	3.44	4.69
Pentachlorophenol ethyl ether			0.57	0.55
Pentachlorophenol methyl ether			0.46	0.46

Results and Discussion

The residue data on rainwater samples from Oahu are given in Table 3. Hawaiian rainwater contained *p,p'*-DDT, dieldrin, and lindane in the low ppt range. The only comparative data that could be found in the literature were from 1965 (8,9); the residues in the present study were much lower than the levels noted previously in the cities of Cincinnati, Coshocton, and Ripley, Ohio, (8) but similar to the amounts observed in Central England (9). It is possible that with increased restricted usage of chlorinated pesticides in the United States in the past 2 years, new measurements of rainwater in Ohio might show considerably lower quantities of these pesticides than reported in the earlier study.

TABLE 3.—Organochlorine pesticide residues in 10 rainwater samples from Oahu, Hawaii, and in other studies

COMPOUND	HAWAII 1970-71		REPORTED IN OHIO 1965 (8)		REPORTED IN CENTRAL ENGLAND 1965 (9)	
	RESIDUES IN PPT					
	RANGE	AVERAGE	RANGE	AVERAGE	RANGE	AVERAGE
<i>p,p'</i> -DDT	1-13	3	70-340	187	2.4	3
Dieldrin	1-27	5	(1)		3-16	9
Lindane	1-19	5	6-50	25	12-52	29

¹ Detected, but no values reported

Residues in the drinking water samples were chlordane, DDT, dieldrin, and lindane and were in the low ppt range and about one-fifth or less the amounts noted in rainwater (Table 4). DDT was the predominant pesticide, appearing in 87% of the samples. Although extreme precautions were taken with the sampling procedures, pesticide contamination could have been present at the external source of sampling, because some of the stations are surrounded by sugarcane and pineapple growing areas or subdivision development projects.

The residues for drinking water were insignificant compared to the allowable amounts (Table 4) proposed by the Federal Committee to the Secretary of the Interior on Water Quality Criteria (10). The Committee has defined "permissible criteria" as "those characteristics and concentrations of substances in raw surface waters which will allow the production of a safe, clear, potable, aesthetically pleasing, and acceptable public water supply which meets the limits of drinking water standards after treatment." This Committee has also stated that it would be desirable to have no pesticides in drinking water supplies.

Ettinger and Mount (11) have stated that the use of waters for fish propagation must be considered in any set of acceptable State water quality criteria and that it has a bearing on drinking water standards. They have proposed the maximum stream allowance for certain pesticides (Table 4) which are one-tenth or less the

TABLE 4.—Organochlorine pesticide residues in 45 potable water samples from Oahu, Hawaii—February-May 1971

COMPOUND	RANGE	AVERAGE	PERCENT OF SAMPLES	MAXI- MUM PER- MISSIBLE CRITERIA (10)	MAXIMUM REASONABLE STREAM ALLOW- ANCES (11)
Chlordane	0.5-5.0	1.0	9	3,000	250
<i>p,p'</i> -DDT	0.6-2.2	1.0	87	42,000	500
Dieldrin	0.2-0.7	0.3	15	17,000	250
Lindane	0.06-0.4	0.2	4	56,000	5,000

amounts permissible by the Public Health Service. These allowances are greater than the amounts found in the local drinking water supply in Oahu. It is of interest to note that in 1962 the standards were revised to specify that the carbon chloroform extract (which is a measurement of synthetic chemicals in the environment including chlorinated insecticides) should not exceed 200 ppb (12,13). As late as 1967, an effort was made to include specific pesticides under these standards; however, it was pointed out that this could not be done in a legal sense, because pesticides were not classed in the "communicable disease category" (14).

Organochlorine pesticide residues in nonpotable waters were at the low ppt level, with *p,p'*-DDT representing the highest level (Table 5). In all areas sampled, dieldrin was consistently present and, in some instances, at a higher level than the other observed pesticides.

A crude attempt was made to classify the sampled areas into rural and urban sections of the State. The data show that the only major difference in residues in the waters was for dieldrin, which averaged about seven times higher in the urban areas (Table 6).

A detailed study was made of two canals and a stream, which were located primarily in the urban areas of Oahu, and a sewage outfall which received all sewage from the Honolulu area. One of the canals (Kapalama) and the stream passed through industrial areas. The water samples from the Kalihi stream were obtained near a wood treatment plant where, on the day of sampling, a heavy rain had drenched a large pile of pentachlorophenol-treated lumber. Drainage ditches from this lumber yard migrated to the sampled stream. The sample obtained from the ditch area nearest the lumber yard contained 1,143 ppt pentachlorophenol, whereas the sample obtained some distance from the yard and more a part of the stream contained 168 ppt pentachlorophenol, indicating a dispersion or diluent effect of the larger water area. Other pesticides, DDD, DDT, and dieldrin were also detected in the low ppt range, with dieldrin predominating in about 3-fold the quantities of DDT (Table 7). The sample from the sewage outfall contained 2,600 ppt pentachlorophenol, 198 ppt dieldrin, 107 ppt DDT, and 41 ppt lindane.

TABLE 5.—Average amount and range of organochlorine pesticide residues in nonpotable waters, Hawaii—August 1970-February 1971

ISLAND	No. OF SAMPLES	p,p'-DDE	p,p'-DDD	RESIDUES IN PPT		LINDANE	CHLORDANE ¹
				DIETRIN			
Hawaii	4	—	—	3.5 (3.2-3.8)	5.5 (1.9-8.3)	—	—
Kauai	6	—	—	4.4 (3.4-7.1)	1.1 (0.4-2.1)	—	—
Maui	8	0.7 (0.5-0.8)	—	4.9 (2.6-6.8)	1.6 (0.5-5.1)	0.9 (0.2-3.4)	—
Oahu							
Streams	4	0.5 (0.2-0.8)	0.1 (0.1)	1.4 (1.0-1.7)	1.5 (1.2-1.7)	0.7 (0.4-1.1)	—
Lakes	5	0.3 (0.1-0.5)	7.8 (0.2-18.0)	14.0 (0.2-64.0)	2.0 (0.5-4.0)	0.1 (0.1)	—
Bays	6	0.3 (0.1-0.6)	3.6 (0.1-10.0)	9.0 (0.4-41.0)	1.0 (0.1-3.0)	0.9 (0.3-2.0)	—
Basin	1	—	—	0.5	14.0	—	—
Harbors	2	—	—	0.5 (0.5)	3.5 (0.0-7.0)	—	—
Lagoon	1	—	—	0.5	11.0	—	—
Canals	9	0.1 (0.0-1.0)	2.4 (1.3-3.9)	4.0 (1.0-7.1)	13.0 (0.4-18.6)	0.9 (0.3-2.0)	6.9 (3.0-17.6)

NOTE: — = not detected; figures in parenthesis = range.
¹ Chlordane analysis was not included in the early part of the study.

TABLE 6.—Pesticide residues in waters from 13 rural and 24 urban areas in Hawaii—1970-71

COMPOUND	RURAL		URBAN	
	RANGE	AVERAGE	RANGE	AVERAGE
	RESIDUES IN PPT			
p,p'-DDE	0.1-0.5	0.3	0.2-0.8	0.5
p,p'-DDD	0.1-10.0	3.1	0.8-18.0	3.4
p,p'-DDT	0.2-41.0	6.1	0.8-64.0	6.3
Dieldrin	0.1-4.0	1.2	0.3-19.0	8.4
Lindane	0.1-1.1	0.5	0.5-3.4	1.1
Total chlorinated pesticides		11.2		19.7

Extensive samplings of the waters of the two canals were made; these pesticide residue values are shown in Table 7. Again, the residues were in the low ppt range, with dieldrin predominating in quantity; however, the residues in the algae, fish, and sediments obtained from the same canals, were larger. Converting the total residues in the algae, sediment, and fish to parts per million places the values in the 0.1-1.0 ppm range, which is considerably lower than the 5.0 ppm DDT guideline value allowed in fish by Federal regulations. Converting dieldrin residue values found in the fish to parts per million gives values approximating the maximum permissible amounts. The amounts of chlordane and dieldrin found in the waters and the sediments and the pentachlorophenol residues observed in the sewage suggests that these pesticides originated primarily in the urban areas where many households and home builders use these compounds for the control of termites.

Summarizing the data in Table 7, the total amount of chlorinated insecticide residues in the waters of the two canals was the same at about 0.03 ppb; the residues in sediments (dry-weight basis) were 600 ppb. The total

residues in the biota from the Ala Wai Canal on a fresh-weight basis were algae 130 ppb, small fish (guppies and mollies) 800 ppb, carnivore fish and detrital feeder fish each about 1,000 ppb. All three types of fish had similar amounts of total residue; however, the carnivore species contained a predominant amount of DDD, whereas the guppies, mollies, and detrital feeders contained predominant amounts of dieldrin.

The ratio of residues in the canal waters to those in the biota from the canals, assigning the value of 1 to water, were:

Water	1
Algae	4,300
Sediment (wet-weight basis)	9,000
Small fish	27,000
Carnivore fish	33,000
Detrital feeder fish	36,000

As noted by Gunther *et al.* (15) and others (16,17), published data on the solubility of pesticides in water are inconsistent and, at times, actually misleading and meaningless (Table 8). In waters containing salts, organic substances, and colloidal material, the solubility of a given pesticide may vary greatly, no doubt due to absorption or binding of the pesticide to suspended soil particles, plankton, and other types of matter in the water.

Pesticide residues found in natural water analyzed as received may vary considerably from residues found in the same water after it has been filtered through a glass fiber membrane. As stated by Walker (18), water containing suspended material appears to carry residues while water without this material may not contain measurable amounts of pesticides.

TABLE 7.—Pesticide data on nonpotable waters from selected areas, Oahu, Hawaii—
August 1970-February 1971

	DATE SAMPLED	p,p'-DDE	p,p'-DDD	p,p'-DDT	DIELDRIN	LINDANE	CHLORDANE	PCP ¹
RESIDUES IN PPT								
ALA WAI CANAL								
Water								
Station 1	8-12-70	1.0	2.0	1.0	5.0	1.0	(²)	
	2-18-71	—	1.3	1.6	9.6	—	9.4	
Station 2	8-12-70	—	3.0	3.0	17.0	2.0	(²)	
	2-26-71	—	1.3	1.6	18.6	1.3	13.0	
Station 3	8-12-70	—	3.0	2.0	16.0	2.0	(²)	
	2-18-71	—	2.6	1.6	0.4	0.3	4.8	
	Average	0.2	2.2	1.8	11.1	1.1	9.1	
Sediment (dry-weight basis)								
Station 1	2-18-71	100,000	220,000	150,000	100,000	—	720,000	
Station 2	2-18-71	10,000	100,000	30,000	30,000	—	290,000	
Station 3	2-18-71	10,000	40,000	40,000	100	—	125,000	
	Average	40,000	120,000	73,333	43,366	—	378,000	
Biota (fresh-weight basis)								
Algae		10,000	30,000	45,000	45,000	—	—	
Fish								
Guppy, Molly	June 1970	80,000	210,000	170,000	340,000	—	—	
Carnivore (<i>Elops hawaiiensis</i>) ²	through Feb. 1971	182,000	581,000	101,000	141,000	—	—	
Plankton and detrital feeder (<i>Chanos chanos</i>) ²		298,000	149,000	159,000	486,000	—	—	
KAPALAMA CANAL								
Water								
Station 1	2-9-71	—	—	7.1	16.4	1.0	(²)	
	2-16-71	—	3.9	4.7	10.2	—	3.0	
	3-1-71	—	3.9	6.4	18.0	0.5	4.4	
	Average	—	2.6	6.1	14.9	0.5	3.7	
Sediment (dry-weight basis)								
Station 2	2-19-71	—	—	50,000	370,000	—	255,000	
Station 3	2-19-71	20,000	90,000	170,000	140,000	—	125,000	
	Average	10,000	45,000	110,000	255,000	—	190,000	
KALIHI STREAM								
Water (Industrial area)								
Station 1	12-15-70	—	—	3.7	12.7	—	(²)	1,143
Station 2	12-15-70	—	1.6	2.8	7.1	—	(²)	168
	Average	—	0.8	3.3	9.9	—	—	655
SAND ISLAND OUTFALL								
Sewage (24-hr. composite discharge)	12-14-70	—	—	107	198	41	(²)	2,600

NOTE: — = not detected.

¹ PCP = pentachlorophenol; analysis performed only on samples indicated.

² Chlordane analysis was not included in the early part of the study.

³ Muscle tissue.

Water standards or "water quality criteria" are defined differently by different specialists; in fact, at this time, it is impossible to give an all-inclusive definition. For example, the publication of the Public Health Service on drinking water standards (13) states that "The Drinking Water Standards are regarded as a standard of quality which is generally attainable by good water quality control practices. Poor practice is an inherent health hazard. It has been the policy of the Committee to set limits which are not so low as to be impracticable nor so high as to encourage pollution of water.

"No attempt has been made to prescribe specific limits for every toxic or undesirable contaminant which might enter a public water supply. While the Committee is fully cognizant of the need for continued attention to chemical contamination of water, the standards are limited to recognized need. Standards for innumerable substances would require an impossible burden of analytical examination."

Another ill-defined area is toxicity data on fish. The publication "Water Quality Criteria" of the State of

California (19) states that "... the results by several investigators of the same pollutant may not compare closely. This wide discrepancy arises from variations in the species of fish or other organism used, its prior handling, the temperature, the season, the dissolved oxygen content, synergistic and antagonistic substances, the hardness and other mineral content of the water, and the time of exposure, ... sometimes vague terms such as toxic concentration of lethal dose are used without further definition. ... The effects of long-term exposures of fish populations to very low sublethal concentrations are not clearly understood."

It has been noted (23) that water analyses have limited meaning when evaluating the effects of a pesticide on animal populations. Johnson (20) concluded that water data alone on pesticides are an inadequate criterion for determining the relative danger or safety to the fish population in a given area; but it has also been stated that the very low concentrations observed in water may be important because natural mechanisms can concentrate residues many thousands of times. This is evident in the data obtained from the canal samples (Table 7).

Results given below from a survey of the literature indicate no consistency in the relation between residues found in various parts of a given ecosystem:

Study	Compound	Ratio of Residues in Various Parts of Ecosystem
Macek (21) ¹	DDT	Water to fish-1:8,500
Holden (22)	Dieldrin	Water to algae-1:1,000
Woodwell et al. (23) ²	DDT	Water to fish-1:20,000
Johnson (20)	DDF	Water, sediment, and fish-1:1,500:20,000
Hickey et al. (24)	DDT, DDD, DDE	Sediment to fish-1:100
Hannon et al. (25) ³	DDT, DDD and DDE	Water, sediment, and algae, fish-1:20:40:800 accounted for the greatest percentage of pesticide residues

There is a general consensus that as many factors as possible should be examined in a given ecosystem. Pesticides are leached from soils by water, moved by erosion, and absorbed by mud-seavenging organisms (23); they are soluble in fats and oils; they tend to be concentrated in organic matter, algae, bacterial films, and slimes; and they can be suspended in colloidal forms (22). Most probably the food chain is the major

source of pesticide contamination in fish (21,26), and, as stated by Eichelberger and Lichtenberg (27), there is no predictable safe level for pesticides in waters where food-chain buildup can occur.

TABLE 8.—Reported water solubilities of some organochlorine pesticides at room temperature, 25° C

COMPOUND	GUNTER <i>et al.</i> (15)	ROBEK <i>et al.</i> (16)	BEVENE AND BUKMAN (17)
	RESIDUES IN PPB		
Chlordane	1 Insoluble		
<i>p,p'</i> -DDD	1 Insoluble		
<i>p,p'</i> -DDT	1 Insoluble 3.4 6 and 25	16 and 40	
Dieldrin	150 and 195 250	140 and 180	
Lindane	600 and 6,800 7,300	500 and 6,000	
Pentachlorophenol			14,000 to 19,000
Sodium pentachlorophenate			79,000 (pH 5) 4,000,000 (pH 8)

¹ "Insoluble" = solubility up to about 1% or 10,000 ppm
² Particle size 0.05 and 5.0 μ
³ Particle size 0.04 and 5.0 μ
⁴ At temperatures 20° to 30° C

Several investigators have suggested that bottom muds and sediments should be given more attention in any pesticide study of water (20,24,26) and that these data should be combined with other available ecological information to give a good pesticide-pollution index.

In conclusion, it is emphasized that the present report is based on a preliminary survey of randomly selected samples. Future studies will include silt and strata sampling of the shoreline waters, seasonal sampling, an in-depth study of the sewage outfall, and, perhaps, concentrated sampling of areas in the State suspected of potentially higher pesticide residues. However, based on these preliminary data, on the present usage pattern of the pesticides studied, and comparison with the proposed water quality standards, pollution of Hawaii's waters by organochlorine pesticides does not occur to a significant degree.

See Appendix for chemical names of compounds discussed in this paper.

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PESTICIDES IN SOIL

*DDT Residues in Forest Floor and Soil After Aerial Spraying, Oregon—1965-68*¹

R. F. Tarrant, D. G. Moore, W. B. Bollen, and B. R. Loper

ABSTRACT

One month after aerial application of DDT (12 oz/acre) in eastern Oregon forest, 3 oz/acre of DDT residues (DDT, its isomers and metabolites—DDD, DDE, *p,p'*-DDT, and *o,p'*-DDT) were detected in the forest floor; 3 years later, the DDT content had decreased by more than 50%, and had not leached into the surface mineral soil.

At the time of spraying, water from two streams draining the sprayed area had a total DDT content of about 0.3 ppb. This low concentration decreased rapidly to levels below limits of analytical detection. No effect of the spraying was noted on soil microbial populations, nitrification rate, or amount of nitrate nitrogen in the soil.

Of the 12 oz of DDT applied per acre, about 26% reached the ground surface initially; and over 36 months, about 6% more was brought to the ground in litterfall. Thus, approximately one-third of the sprayed chemical reached the forest floor. The need for more efficient aerial methods of chemical application is evident.

Introduction

Only a few studies have been concerned with residues of DDT, its isomers and metabolites, in the forest environment. Woodwell (41) determined DDT residues in a forest in New Brunswick, Canada, which had been sprayed between 1952 and 1958 with a total of 4 lb/acre of DDT. He concluded that the maximum residue persistence time in forest soil would be 10 years and that *o,p'*-DDT was leached into the subsoil. Woodwell and Martin (42) reported that DDT residues in heavily sprayed forest soils in Maine and New Brunswick increased over a period of 3 years after final spraying. These authors hypothesized that DDT residues persist in the forest canopy and are carried to the soil by rain and litterfall.

Yule (43) stated, however, that Woodwell's hypothesis of differential weathering and preferential retention of *o,p'*-DDT by New Brunswick forest soils is untenable. He concluded from a study in the same locality that about 16% of the DDT originally applied still remained in surface soils after almost 20 years, but mainly in the form of the most toxic isomer, *p,p'*-DDT. He further demonstrated that the acidic, highly organic, forest surface soils held these DDT residues unavailable in toxic amounts to soil insects.

DDT residues were also measured in a northern Pennsylvania forest soil, 380 days after aerial spraying at a rate of 0.5 lb/acre (9). The only environmental change reported was a significant accumulation of DDT in the forest floor and surface soil. One year after spraying, no measurable increase in DDT residues was noted in fish, crayfish, or stream sediments. Belyea (4) studied DDT residues in soils and a related food chain in northern Maine forests. He concluded that DDT would disappear from these soils in 10-12 years. In western Washington, DDT was applied to the surface of a gravelly soil beneath a stand of Douglas-fir [*Pseudotsuga menziesii* (Mirb.) Franco] at 0.5 and 5.0 lb/acre (29). Regardless of application rate, less than 1% of the DDT applied leached through the surface soil.

In 1965, an opportunity to further study DDT residues in forests resulted from a spray project conducted by the U. S. Forest Service between June 10 and July 1 to control a serious outbreak of the Douglas-fir tussock moth (*Heemerocampa pseudotsugata* McD.) in eastern Oregon. Helicopters were used to spray 66,000 acres of forest with an insecticide formulation of 0.75 lb of technical grade DDT dissolved in 0.94 qt of hydrocarbon solvent and sufficient No. 2 fuel oil to make 1 gal of solution at 60° F; the application rate was 12 oz/acre.

¹ From the Pacific Northwest Forest and Range Experiment Station, U. S. Department of Agriculture, Forest Service, Corvallis, Oregon 97330.

A number of public agencies have already conducted surveillance and monitoring activities to determine residues in fish and wildlife, cattle, and forage, and to ascertain public health effects from the spray project (10,35). The present study reports observations of the persistence of the applied DDT in the forest floor and soil and some related effects for the first 3 years after the spray project.

The area studied was along a 1-mile transect in the Malheur National Forest of eastern Oregon. Elevation of the area is 5,700 feet. The subhumid continental climate includes dry, warm summers ($>100^{\circ}$ F maximum temperature) and cold winters (-20° F minimum temperature). The daily range of temperatures in summer is often 40 to 50 degrees and the monthly range may exceed 60 degrees. Annual precipitation averages about 20 inches, one-third to one-half of which is snow. Most moisture available to plants is stored in the soil at the beginning of the growing season, and by midsummer soils become very dry; summer flow in small streams is intermittent.

The soil of the study area, representative of perhaps several million acres of eastern Oregon forest land, is tentatively identified as a member of the Klicker series. This is a well-drained, moderately fine-textured, forested soil developed in residuum from basalt bedrock; small fragments of the basaltic parent material occur throughout the solum, and rock outcrops are common throughout the area. The A horizon, which may contain volcanic ash, ranges in texture from silt loam to silty clay loam, with the percent clay increasing gradually with soil depth. The soils are slightly acidic throughout the solum which ranges in depth from 15 to 35 inches. Permeability is moderate, and surface runoff is medium. Major tree species in the study area which partially determine the amount of litter and duff on the forest floor are ponderosa pine [*Pinus ponderosa* (Laws.)], White fir [*Abies concolor* (Gord. & Glend.) Mill.], and Douglas-fir [*Pseudotsuga menziesii* (Mirb.) Franco].

Sampling Procedures

Five 1-10-acre plots were established along the transect nearest to equidistant points where maximum uniformity of stand composition and density could be found. Within each of the five plots, four sampling points were randomly selected. At each point, the forest floor over a 4-sq-ft area was carefully removed, and 1-qt samples of soil were collected at each of two depths, 0 to 3 inches and 3 to 6 inches, in step-like trenches. Extreme care was taken to avoid contaminating the lower sample with material from above, and all tools were cleaned frequently with acetone. Samples were placed in new 1-qt paper freezer containers, stored during the day in portable cooler chests, and frozen the same day they

were collected. Samples of forest floor and soil were collected prior to the spray project, 1 month after the spraying, and at 1-year intervals. Litterfall, throughfall precipitation, and water samples from streams were collected every 6 months.

Eight trays, each with a surface area of 10.9 sq ft, were randomly placed throughout each sample plot to collect litterfall, and four 1-gal containers fitted with funnels having openings approximately 1 sq ft in size were randomly located in each sample plot to collect precipitation. Water samples from the east and west forks of Rattlesnake Creek within 50 feet of their confluence were taken in 1-gal containers, submerged just deep enough to prevent undue disturbance of bottom sediments.

Analytical Procedures

Soil samples were air-dried, ground, passed through a 10-mesh sieve, mixed, and subsampled. For samples intended for microbial analysis, the sieve was cleaned with 30% ethanol and flamed between collections of each sample. Frozen litter and forest floor samples were chopped with dry ice, mixed, and subsampled. All samples, including water, were stored at 0° F until extracted.

EXTRACTION

Soils: A 100-g subsample was extracted with 41:59 hexane:acetone (azeotropic) in a Soxhlet extractor for 16 hours (27).

Litter and forest floor: A 25-g subsample was extracted with acetone in a Soxhlet extractor for 16 hours.

Water: Volumes up to 4,000 ml were extracted with hexane in a continuous-cycling liquid-liquid extractor for 16 hours.

CLEANUP

The soil and litter extracts were transferred to separatory funnels. Water was added to form a 2:1 water-acetone solution. The pesticides were partitioned into hexane by shaking with three 100-ml aliquots of hexane (17).

The hexane extracts of soil, litter, and water were dried with anhydrous sodium sulfate, evaporated to a small volume (5- to 10-ml), and transferred to a 15-g Florisil column (40). The pesticides were eluted from the column with 100 ml of 1:3 dichloromethane:hexane. Dichloromethane was removed by evaporation, and samples were transferred to volumetric flasks for analysis.

ANALYSIS

The concentration of DDE, *o,p'*-DDT, DDD, and *p,p'*-DDT was quantified in a MicroTek 2000 MF gas chromatograph with a 130-me tritium electron capture detector. This system gave good individual peak resolution at the following retention times: DDE, 5.2 minutes;

o,p'-DDT, 6.8 minutes; DDD, 8.0 minutes; and *p,p'*-DDT, 9.5 minutes. Other operating parameters were:

Column: Pyrex glass, 180 cm x 2 mm i.d., packed with 5% QF-1 (0.7 of length) and 5% DC-11 (0.3 of length) on 60-80 mesh Gas Chrom Q, preconditioned for 48 hours at 220°C.
Temperature: Column 185°C
Detector 190°C
Injector 205°C
Carrier gas: Nitrogen at 30 ml/min

Minimum residue levels for quantitative determinations were 0.001 ppm for soil, 0.01 ppm for forest floor and litter, and 0.01 ppb for water. Average percent recovery and range for DDT isomers and metabolites were as follows:

FORM OF DDT	SOIL (PPM)	FOREST FLOOR AND LITTER (PPM)	WATER (PPB)
DE	99(92-103)	97(93-100)	99(98-100)
<i>p'</i> -DDT	82(71-99)	99(96-100)	98(97-100)
DD	82(78-91)	85(80-94)	94(93-95)
<i>p</i> -DDT	97(92-100)	94(90-97)	99(98-100)

NOTE: () = range

CONFIRMATION

It is most necessary to positively identify any apparent DDT determined by gas-liquid chromatography (GLC), especially in samples of materials to which no known DDT application was made. A number of industrial pollutants are similar to DDT in structure and properties and can interfere with the detection or identification of DDT (19,22,28,30,31); some naturally occurring plant or soil substances may also be potential sources of analytical error in determining the presence of chlorinated hydrocarbon compounds (14,20). To confirm apparent DDT residues in this study, about half the samples were analyzed by GLC with a chloride-specific, microcoulometric detection system (Infotronics Instrument Corp.). This step confirmed that substances with the same retention time as the DDT standards detected by the electron capture detector did contain chlorine, but did not rule out the possible misinterpretation of polychlorinated biphenyls (PCB's) as DDT isomers and metabolites. Therefore, all samples analyzed with the microcoulometric detector were hydrolyzed with alcoholic potassium hydroxide which would chemically alter DDT and DDD, but not PCB's (17). Hydrolyzed samples were then re-analyzed by both electron capture and microcoulometric detection systems. DDD, *o,p'*-DDT, and *p,p'*-DDT peaks disappeared after hydrolysis, indicating that PCB's were not present in detectable quantities and that the quantitative measurement of DDT isomers and metabolites by the electron capture detection system was correct.

The mass spectrophotometer provides the most positive means of identifying pesticides in biological samples; but in this study, only a few forest floor samples contained sufficient DDT to use this instrument. Two composite samples of forest floor from two different plots were extracted and purified for this particular analysis. The DDT isomers and metabolites were separated by chromatography of the final hexane extract on 500- μ silica gel H thin layer plates developed with 4:96 benzene:hexane. DDT standards were co-chromatographed on both edges of the 20- by 20-cm plates. After development, a 15-cm strip in the middle of the plate was covered, and the DDT standards were located by spraying the edge of the plate with 0.5% silver nitrate and exposing to UV light for 15 minutes. The *o,p'*-DDT, *p,p'*-DDT, and DDE were scraped from the appropriate section of the center of the plate, extracted from the silica gel with hexane, and analyzed by gas chromatography (electron capture detector). The pesticides separated by the thin layer method had the same retention times as the standards.

Extracts containing the individual pesticides were introduced into a Model CH 7 (Varian Mat. Bmg. H.) mass spectrometer with a direct inlet probe. The mass spectra for *o,p'*-DDT, *p,p'*-DDT, and DDE isolated from the forest floor samples agreed with spectra of appropriate standards and with published spectra (34). Sample spectra compared with published PCB spectra (3) indicated no PCB's present in the isolated pesticides. All confirmation steps gave positive evidence that the substances isolated and measured were indeed DDT isomers and metabolites and that PCB's were not present in detectable quantities.

MICROBIAL ANALYSIS

The number of bacteria in soil was estimated by plating sterile tap water dilutions of soil on sodium albuminate agar; mold counts were made on plates of peptone-glucose-acid agar (37). In each case, five plates were poured of each dilution: 1:50, 1:500, and 1:5,000 for molds; 1:5,000, 1:50,000, and 1:500,000 for bacteria. Mold colony counts were made on plates showing approximately 30 to 100 colonies after 3 days' incubation; major genera were differentiated as mucors, *Penicillium*, and *Aspergillus* after 3 to 7 days. Total bacteria and *Streptomyces* were counted on plates showing colonies in the range of 50 to 300 after 10 to 14 days' incubation; *Streptomyces* were differentiated and their numbers expressed as a percentage of the total count. Incubation was 28°C.

Nitrite and nitrate were determined colorimetrically by the diazotization (2) and phenoldisulfonic acid (18) methods, respectively. For the nitrification study, 200 ppm N was added as ammonium sulfate to 100 g of soil (oven-dry basis) in 1-pt bottles. Moisture was adjusted

to 50% of water-holding capacity, and the bottles were capped with polyethylene film and incubated for 30 days at 28°C. Distilled water sufficient for a 1:5 dilution was then added, and the samples shaken for 15 minutes. After pH was measured, nitrite and nitrate in the supernatant were determined.

Results and Discussion

FOREST FLOOR SAMPLES

A very small amount (0.13 ppm) of "apparent" total DDT residues (*p,p'*-DDE, *p,p'*-DDD, *o,p'*- and *p,p'*-DDT) were found in prespray samples of the forest floor (Table 1); this was not confirmed as DDT, its isomers and metabolites by means other than GLC because of its insignificant contribution (<0.01 oz/acre) to the totals found after spraying. One month after spraying, concentration of total DDT in the forest floor was slightly more than 7.5 ppm (Table 1), or 3.08 oz/acre (Fig. 1). Thus, an estimated 26% of the applied DDT (12 oz/acre) reached the forest floor shortly after spraying. DDT residues in the forest floor decreased steadily with time, and at the end of 3 years, more than half the DDT originally added had disappeared. Volatilization, chemical or photochemical degradation, and bacterial decomposition are possible removal mechanisms (12).

TABLE 1.—Concentration of DDT isomers and metabolites in the forest floor before and after aerial spraying, Oregon—1965-68

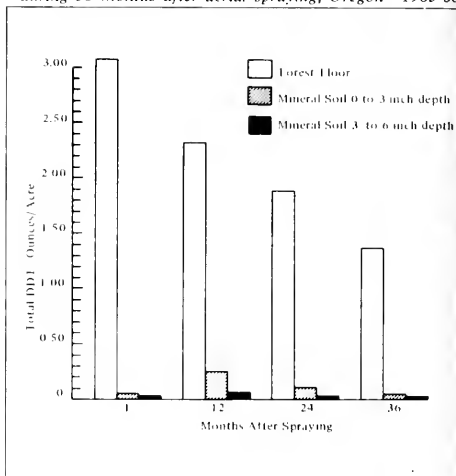
MONTHS AFTER SPRAYING	RESIDUES IN PPM ON A DRY-WEIGHT BASIS ¹				TOTAL DDT
	<i>p,p'</i> -DDE	<i>o,p'</i> -DDT	<i>p,p'</i> -DDD	<i>p,p'</i> -DDT	
0	.008	.025	.008	.089	130
1	.190	1.294	.348	5.708	7.540
12	.214	.957	.352	3.914	5.437
24	.298	.584	.198	3.332	4.412
36	.096	.473	.114	2.641	3.324

¹ Each value for DDE, DDD, *o,p'*- and *p,p'*-DDT represents the average of duplicate determinations on 20 replicate samples. The relationship between concentrations of all isomers and metabolites and months after spraying is negatively linear and significant at the 5% probability level.

SOIL SAMPLES

DDT did not leach from the forest floor to underlying mineral soil. Apparent total DDT in prespray samples was 0.006 ppm (Table 2, or 0.05 oz/acre (Fig. 1) at the 0- to 3-inch depth; at the 3- to 6-inch depth, total DDT concentration was 0.002 ppm, or 0.02 oz/acre. One month after spraying, these levels had not changed, indicating that the forest floor effectively intercepted the spray solution. One year after spraying, the residue level of total DDT in soil at the 0- to 3-inch depth was 0.26 oz/acre; at the 3- to 6-inch depth, it was 0.05 oz/acre. This small difference in residues 1 month after spraying and 1 year later was attributed to the physical action of soil animals and, most probably, to minor, unavoidable contamination during sampling. DDT has a solu-

FIGURE 1.—Total DDT in forest floor and mineral soil during 36 months after aerial spraying, Oregon—1965-68



bility in water of only about 1 ppb (7), and thus, does not leach readily in soil (16,29,33). At the end of the second year, total DDT at the upper and lower soil depth has decreased to 0.11 and 0.03 oz/acre, respectively, and by the end of 3 years, was at prespray level:

LITTERFALL SAMPLES

DDT in litterfall totaled 0.73 oz/acre over the 3-year sampling period, about 6% of the original application (Fig. 2). DDT concentration decreased with time at a greater rate than it did in the forest floor and soil (Table 3), suggesting that photochemical decomposition and volatilization may be effective mechanisms of chemical degradation in tree canopies exposed to sunlight. DDT concentration is also reduced in successive litterfall samples because of the constantly decreasing proportion of needles and twigs originally subjected to the spray. The contribution of DDT from litterfall to the forest floor after spraying did not contribute strongly to total amount observed. Total loss of DDT from the forest floor over 3 years amounted to 2.46 oz/acre, more than three times the amount brought down in litterfall over the same period.

THROUGHFALL PRECIPITATION SAMPLES

Additions of DDT to the forest floor by throughfall precipitation were insignificant—0.02 oz/acre for the 3-year period following application (Fig. 2). Concentrations varied with season (summer-fall vs. winter-spring) and, in general, showed a gradual decrease with time (Table 4). DDT concentrations in samples representing the dry summer and fall months were approximately three times greater than those for the wet winter-spring

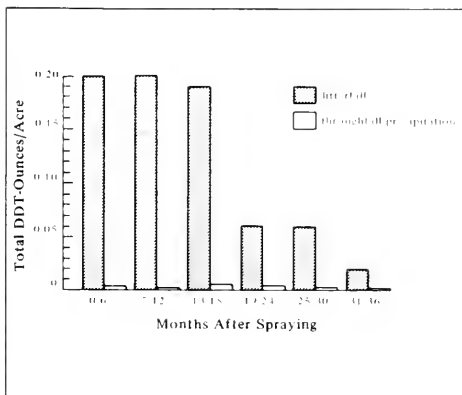
TABLE 2.—Concentration of DDT isomers and metabolites in surface soil before and after aerial spraying, Oregon—1965-68

MONTHS AFTER SPRAYING	RESIDUES IN PPM ON A DRY-WEIGHT BASIS ¹				TOTAL DDT
	<i>p,p'</i> -DDE	<i>o,p'</i> -DDT	<i>p,p'</i> -DDD	<i>p,p'</i> -DDT	
0- to 3-inch depth					
0	.001	.002	—	.003	.006
1	.001	.001	—	.004	.006
12	.003	.005	—	.021	.029
24	.001	.002	—	.009	.012
36	—	.001	—	.005	.006
3- to 6-inch depth					
0	—	.001	—	.001	.002
1	—	.001	—	.001	.002
12	—	.002	—	.004	.006
24	—	.001	—	.002	.003
36	—	.001	—	.001	.002

NOTE: — = not detected.

¹ Each value for DDE, DDD, *o,p'*- and *p,p'*-DDT represents the average of duplicate determinations on 20 replicate samples.

FIGURE 2.—Total DDT brought to forest floor in litterfall and throughfall precipitation during 36 months after aerial spraying, Oregon—1965-68



season. Precipitation samples for the 13- to 18-month period after treatment contained higher DDT concentrations than expected relative to the amount of rainfall for the period and the concentrations found at 6 and 30 months. However, the DDT levels, their seasonal variations, and the total range in concentrations found in this study are consistent with normal climatological variations and similar to those reported for other regions (1,36,39).

TABLE 3.—Concentration of DDT isomers and metabolites added to the forest floor in litterfall after aerial spraying, Oregon—1965-68

MONTHS AFTER SPRAYING	RESIDUES IN PPM ON A DRY-WEIGHT BASIS ¹				TOTAL DDT
	<i>p,p'</i> -DDE	<i>o,p'</i> -DDT	<i>p,p'</i> -DDD	<i>p,p'</i> -DDT	
0-6	66	1.57	.44	8.65	11.32
7-12	.19	1.65	.30	8.18	10.32
13-18	.13	1.06	.22	5.71	7.12
19-24	15	1.07	.26	6.01	7.49
25-30	.09	.62	.13	3.08	3.92
31-36	.07	.47	.10	2.44	3.08

¹ Each value for DDE, DDD, *o,p'*- and *p,p'*-DDT represents the average of duplicate determinations on 20 replicate samples. The relationship between concentrations of DDT isomers and metabolites and months after spraying is negatively linear and significant at the 5% probability level in all cases.

TABLE 4.—Concentration of DDT added to the forest floor in throughfall precipitation after aerial spraying, Oregon—1965-68

MONTHS AFTER SPRAYING	PRECIPITATION ¹ (MM)	TOTAL DDT RESIDUE ² (PPB)
0-6	162	.176
7-12	212	.075
13-18	185	.364
19-24	455	.066
25-30	110	.103
31-36	241	.036

¹ Rainfall level extrapolated from monthly climatological data for Burns, Oregon, using Mean Annual Precipitation Isohyetal Map, U. S. Weather Bureau River Forecast Center, Portland, July 1964.

² Total DDT residue includes *p,p'*-DDE, *o,p'*-DDT, *p,p'*-DDD and *p,p'*-DDT. DDE residue accounted for an average 6.58% of total DDT residue and showed no statistically significant change with time.

At the end of 3 years, DDT concentrations in throughfall precipitation had decreased appreciably, but still were 5 to 10 times greater than levels found in samples from an untreated forested area in western Oregon; however, the total amount of DDT brought down over this period in throughfall precipitation was negligible compared with that part of the intended application that initially reached the forest floor or was deposited in litterfall. Thus, throughfall precipitation was not a significant factor in determining the fate of applied DDT or in maintaining DDT concentrations in the forest floor.

DDT IN STREAMWATER SAMPLES

Streamwater was monitored in Rattlesnake Creek immediately above the confluence of the east and west forks, both of which flow from the sprayed area. The maximum total DDT concentration found over a period of 3½ years after spraying was 0.277 ppb; this was in a sample taken a few hours after spraying (Table 5). This level was similar to those reported from North Carolina (15) and northeastern California (5) and less than those from northern Pennsylvania (9) and New Brunswick (44). Most samples, including those taken 3½ years after the spraying, contained concentrations of DDT near the lower limit of detection (0.01 ppb).

TABLE 5.—Total DDT content of streamwater flowing from sprayed area—before treatment and during 3 years after treatment, Oregon—1965-68

DATE	DAYS FROM TIME OF SPRAYING	TOTAL DDT RESIDUES IN PPB	
		RATTLESNAKE CREEK EAST FORK	WEST FORK
1965			
5/24	-30	—	—
6/19	-4	—	—
6/23	1	.104	.277
7/14	21	.031	.022
8/26	64	.028	.015
11/17	147	.014	—
1966			
6/7	349	—	—
7/19	391	.010	—
11/9	505	—	—
1967			
7/4	742	—	—
11/7	869	.032	.010
1968			
7/16	1,131	—	—
11/12	1,251	.010	—

NOTE: — = not detected.
Blank = levels of DDT isomers and metabolites less than 0.01 ppb but greater than 0.002 ppb.

Surveillance operations by the Bureau of Sport Fisheries and Wildlife (25) indicated that the DDT spraying had little effect on the waters and organisms of Malheur Lake, toward which Rattlesnake Creek flows. Levels of total DDT accumulation in the food chain of Rattlesnake Creek were very low in all components of the sampled community (8).

MICROBIAL SOIL PROPERTIES AND NITROGEN RELATIONS

The small amount of DDT in the top 6 inches of mineral soil had no significant effect on microbial populations, soil nitrification rate, or amount of nitrate nitrogen (Table 6). A number of findings similar to these are found in the literature; these reports, however, are based on laboratory studies in which DDT was added to soil at extremely high rates—50-2,000 lb/acre (13,21,24,26,32). Effects of pesticide residues on microbes in agricultural soils are usually negligible when the chemicals are used at recommended field rates (6, 11,23). Such field rates were usually much greater than those encountered in this study. Thus, it may be concluded that the small amount of total DDT residue in soil found after low-volume aerial spraying to control insects is not hazardous to soil microbes or their role in maintaining soil fertility.

FATE OF AERIALY APPLIED DDT

Of a total aerial application of 12 oz of DDT per acre, 26% reached the forest floor initially, 6% was brought to the forest floor in litterfall over a 3-year period, and a fraction of 1% of the total was washed from the tree canopy over 3 years (Table 7). Thus, about one-third of the total application reached the forest floor.

TABLE 6.—Soil microbial populations, nitrate nitrogen content, and nitrification rate before treatment and during 24 months after aerially spraying with DDT at 12 oz/acre, Oregon—1965-68

VARIABLE AND UNIT OF MEASUREMENT	SOIL DEPTH (INCHES)	MONTHS AFTER AERIAL SPRAYING ¹			
		0	1	12	24
Total bacteria—millions/g of soil	0-3	2.51	2.81	3.46	7.58
	3-6	1.83	2.05	4.04	4.51
Streptomycetes—percent of total bacteria	0-3	15.25	23.63	27.25	63.81
	3-6	13.13	19.50	35.56	34.00
Total molds—thousands/g of soil	0-3	338.19	302.88	464.69	168.31
	3-6	93.88	33.88	323.31	35.06
Penicillia—percent of total molds	0-3	78.23	94.46	95.06	62.72
	3-6	77.10	68.45	82.78	68.45
Nitrate nitrogen in soil—ppm	0-3	58.00	40.94	7.00	17.50
	3-6	5.81	4.31	1.63	2.13
Nitrification of added ammonium sulfate—percent of total added	0-3	9.59	—56	—50	—17
	3-6	.27	.04	—26	—13

¹ Results of regression analysis indicated no significant relation between soil DDT content and any of the variables measured at the 5% probability level.

TABLE 7.—Total DDT deposited on the ground surface (forest floor) during 3 years after aerially spraying of DDT at 12 oz/acre, Oregon—1965-68

SOURCE OF DDT AT GROUND SURFACE	TOTAL DDT	
	OZ/ACRE	PERCENT OF TOTAL APPLIED
Initial deposit from spraying	3.08	25.66
Total deposit during 3 years from:		
Litterfall	.74	6.17
Throughfall precipitation	.02	.01
Total, all sources	3.84	31.84

Because this study did not include direct measurement of the amounts of DDT that reached the forest canopy, the extent of chemical loss from drift during spraying or by volatilization or degradation in the canopy after spraying cannot be assessed. In a study in Arizona, less than 50% of aerially sprayed insecticides were deposited on the agricultural target during summer months (38); in the same study, the distance from the spray aircraft to the target was inversely correlated with amount of on-target chemical application. Aircraft spraying forest lands must fly at far greater heights than those operating over level agricultural fields. Thus, the comparatively low amount of on-target application suggested by the present study is not surprising. This finding reaffirms that efficient methods of aerial spraying must be developed in order to avoid great loss of chemical to nontarget areas.

See Appendix for chemical names of compounds discussed in this paper.

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GENERAL

Decay of Parathion and Endosulfan Residues on Field-Treated Tobacco, South Carolina—1971

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ABSTRACT

Parathion and endosulfan were applied three times at rates of 1.5 and .5 lb/acre, respectively, to field tobacco in South Carolina in an effort to determine the time required for these pesticides to degrade to "zero" residue levels. The maximum times were estimated to be 7 days for parathion and 10 days for endosulfan. During the study period, weather was characterized by high rainfall and temperatures averaging 80° F.

Introduction

The recent increased use of organophosphate pesticides such as parathion as substitutes for the restricted chlorinated hydrocarbons has revived concern about residues of these compounds. Interest was further stimulated by the reported cluster of parathion poisonings among North Carolina tobacco workers in 1970 (J. I. Freeman, personal communication).

Dowart *et al.* (2) in measuring the hydrolysis rate of parathion reported that 85% remained undegraded after 1 week, 75% after 2 weeks, and 17% was still undegraded at 6 weeks. Maier-Bode (3), however, reported that the question of how long parathion residues remained in and on food plants has been answered. He concluded that parathion as a spray or powder is absorbed into plant tissues where metabolism occurs; any remaining surface residues are rapidly broken down by photolysis of the sun or evaporated into the atmosphere. Maier-Bode advised that German law requires a waiting

time of 14 days between parathion application and harvest to reduce residues to levels nonhazardous to human health. He also suggested that rain during this waiting period does not significantly affect the residue level.

The U. S. Department of Agriculture has long suggested that workers entering tobacco fields within 5 days after application of parathion be protected against skin contact by use of protective clothing (1).

The experiment reported here was designed to observe the decline with time of levels of parathion normally applied to field tobacco. The ultimate goal was to estimate and verify the safe interval suggested by USDA for entering fields after parathion treatment.

Endosulfan, a chlorinated hydrocarbon insecticide still registered for use on tobacco, was included as another treatment and also mixed with parathion to assay decay interaction of the two chemicals.

Methods and Procedures

Cokers 319 variety tobacco was planted on April 12, 1971. Plots consisting of three 12-foot rows were randomly selected for treatment with parathion, endosulfan, or parathion in combination with endosulfan or as appropriate controls. Each treatment or control plot was replicated four times in a completely randomized design for a total of 16 plots. Guard rows were used to reduce pesticide drift. Parathion and endosulfan were applied as sprays at rates of 1.5 and .5 lb active ingredient (A.I.) per acre, respectively, on June 8, 21, and July 8, 1971.

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Twelve foliage sample collections were made at times indicated in Table 1 and Fig. 1 and 2. Each time, 25-g. field-weighted samples were secured from the center alley of each plot from the foliage of two plants at locations approximately one-half the plant height. The samples were placed in acetone-washed, oven-dried, 1-pint containers.

Rainfall data, shown in Fig. 1 and 2, were obtained as recorded at the Clemson University Truck Experiment Station, and temperature information was secured from the U. S. Weather Bureau.

Since all plots were observed at 12 different times, the statistical analysis computed (Tables 2 and 3) was for a split plot in time where the whole plots were in a completely randomized design.

Analytical Procedures

To each 25-g sample of tobacco, 50 ml of nanograde hexane and 5 g of anhydrous sodium sulfate were added. The samples were shaken and allowed to stand for 3 hours. The hexane was then decanted into a Kuderna-Danish evaporator. This extraction was repeated twice more with 50 ml of hexane, and extracts were combined with the original. The extract was taken to near dryness on a steam bath using a three-half Snyder column and rediluted to an exact volume. The extraction procedure used in this experiment was a modification of that of Reed and Priester (4). Sensitivity of the method allowed for detection of each compound at 0.01 ppm.

A 5 μ l of column injection was made from dilutions ranging from 1 ml to 50 ml of the residues redissolved in hexane. Determinations of residues were made using a MicroTek 220 gas chromatograph equipped with an electron capture (tritium) detector and a flame photometric detector.

Instrument parameters were as follows:

Column:	Glass, 6' x 1/4", packed with 1.5% OV-17/1.95% QF-1 on Chromosorb W 100/120; DMCS, HP
Carrier gas:	Repurified nitrogen at 60 ml/min
Temperatures:	Inlet 230° C Column 200° C Transfer 235° C
Detectors: (A)	Electron capture (tritium) Temperature 205° C
(B)	Flame photometric
	1. Temperature 225° C
	2. Mode—sulfur and phosphorus
	3. Gas flows: Helium-200 ml/min; oxygen-20 ml/min Air-100 ml/min; nitrogen-60 ml/min

The 1.5% OV-17/1.95% QF-1 column was efficient in resolving the parathion and endosulfan peaks and the was used for both detectors. As the sensitivity of the electron capture detector is greater, it was used for quantitation of the peaks on the basis of relative peak heights. The flame photometric detector was used for confirmation of compounds detected. The two endosulfan isomers were quantitated independently but reported as total endosulfan.

Recoveries averaged 92.6% for parathion and 87.3% for endosulfan. Results were not corrected for recovery.

Results and Discussion

Fig. 1 presents parathion residue levels in relation to application time. Within the framework of this experiment regarding temperature, moisture, and sunlight, the maximum time required for parathion to degrade to "zero" levels was estimated to be 7 days and the mir-

TABLE 1.—Parathion and endosulfan residues on field-treated tobacco, South Carolina—1971

APPLICATION DATE	SAMPLING TIME IN DAYS FROM LAST APPLICATION	MEAN RESIDUAL LEVEL IN PPM OF FOUR REPLICATES PER TREATMENT							
		PARATHION (1.5 A.I. LB/ACRE)		ENDOSULFAN (.5 A.I. LB/ACRE)		PARATHION (1.5 A.I. LB/ACRE)/ENDOSULFAN (.5 A.I. LB/ACRE)		CONTROL	
		PARATHION	ENDOSULFAN	PARATHION	ENDOSULFAN	PARATHION	ENDOSULFAN	PARATHION	ENDOSULFAN
June 8	0	0	0	0	0	0	0	0	0
	1	.619	.161	.108	.905	1.049	1.489	.140	.091
	10	.146	.195	.144	.459	.146	.305	.091	.069
June 21	1	.436	.076	.009	.695	.736	1.189	.041	.100
	3	.091	.224	.130	.234	.159	.231	.091	.221
	5	.069	.106	.071	.152	.070	.174	.089	.072
	9	.017	.056	.012	.156	.014	.148	.020	.067
July 8	15	.004	.037	.003	.077	.007	.106	0	.052
	1	1.123	.679	.380	1.751	1.304	1.741	.584	.529
	4	.632	.418	.169	.969	.694	.954	.493	.389
	8	0	.145	.062	.311	0	.235	0	.147
	18	0	.047	.002	.207	0	.174	0	.037

NOTE: LSD₀₅ = least significant difference at 95% probability level = .166 for parathion and .363 for endosulfan; residues were not corrected for percent recovery (see methods).

num time 2 days. "Zero" levels are considered within the limits of actual zero, i.e., undetectable amounts, plus the calculated least difference considered statistically significant at the 5% level of significance.

Similar information is provided for endosulfan in Figure 2 and indicates reductions of residues to "zero" in 10, 2, and 7 days, respectively, after three successive pesticide applications.

Although the experimental design included guard rows, there were still measurable amounts of drift of the insecticide between all plots. Table 1 lists the actual amounts of each insecticide detected in each of the four treatments. In most instances, the contaminant or "drifted" chemical (e.g., endosulfan detected in the parathion treatment) was less than the LSD_{05} . Thus, it is felt that experiment safeguards such as guard rows and control plots were highly effective in reducing the contamination between plots.

The parathion-endosulfan treatment was included to determine if any interaction existed between the two chemicals. It was found that more parathion residue was present when endosulfan was present; however, the converse was not true as can be seen from Tables 1, 2, and 3.

FIGURE 1.—Average parathion residues on tobacco leaves

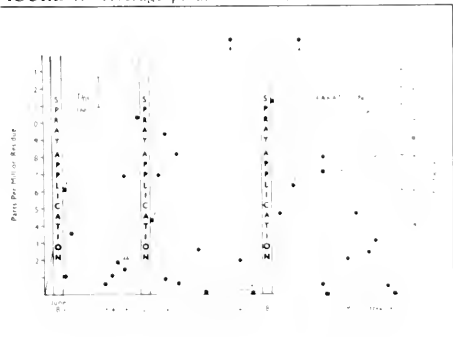


FIGURE 2.—Average residues of endosulfan on tobacco leaves

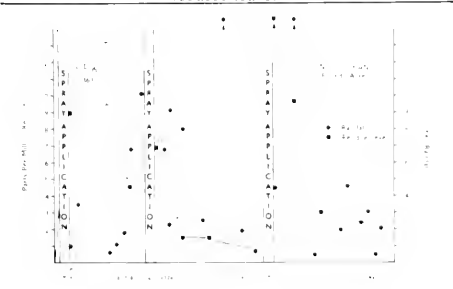


TABLE 2.—Analysis of variance for the variable parathion

SOURCE OF VARIATION	DEGREES OF FREEDOM	SUM OF SQUARES	MEAN SQUARE	F
Treatment				
Parathion	1	1.8217	1.8217	100.765**
Total Endosulfan	1	.0282	.0282	1.559
Para x Endo	1	.1884	.1884	10.421**
Error (a)	12	2169	.0181	
Time	11	12.9523	1.1775	81.665**
Ti x Para	11	4.0406	.3673	25.476**
Ti x Endo	11	.2898	.0263	1.827*
Ti x Para x Endo	11	.4412	.0401	2.782**
Error (b)	132	1.9032	.0144	

* $P < .05$

** $P < .01$

TABLE 3.—Analysis of variance for the variable endosulfan

SOURCE OF VARIATION	DEGREES OF FREEDOM	SUM OF SQUARES	MEAN SQUARE	F
Treatment				
Parathion	1	.1201	.1201	1.034
Total Endosulfan	1	6.3731	6.3731	54.873**
Para x Endo	1	.0178	.0178	.154
Error (a)	12	1.3937	.1161	
Time	11	21.1820	1.9256	27.939**
Ti x Para	11	.5584	.0508	.737
Ti x Endo	11	8.0654	.7332	10.638**
Ti x Para x Endo	11	.6309	.0574	.832
Error (b)	132	9.0979	.0689	

** $P < .01$

In excess of 12 inches of rain fell during the period of the experiment, and it is felt that the unusual rainfall may have physically effected some residue reduction. Daily temperature means averaged 80.9 F with a standard deviation of 2.3 and a range of 76 to 85 F.

This experiment should be repeated for confirmation purposes and to observe results under different environmental conditions including different moisture conditions to determine effects of weather on degradation.

See Appendix for chemical names of compounds discussed in this paper.

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APPENDIX

Chemical Names of Compounds Discussed in This Issue

ALDRIN	Not less than 95% of 1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4- <i>endo-exo</i> -5,8-dimethanonaphthalene
BHC	1,2,3,4,5,6-hexachlorocyclohexane, mixed isomers
CHLORDANE	1,2,4,5,6,7,8,8-octachloro-3a,4,7,7a-tetrahydro-4,7-methanoindane
DDE	1,1-dichloro-2,2-bis(<i>p</i> -chlorophenyl) ethylene
DDT (including its isomers and dehydrochlorination products)	1,1,1-trichloro-2,2-bis(<i>p</i> -chlorophenyl)ethane; technical DDT consists of a mixture of the <i>p,p'</i> -isomer and <i>o,p'</i> -isomer (in a ratio of about 3 or 4 to 1)
DIELDRIN	Not less than 85% of 1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8a-octahydro-1,4- <i>endo-exo</i> -5,8-dimethanonaphthalene
ENDOSULFAN (THIODAN®)	6,7,8,9,10,10-hexachloro-1,5,5a,6,9,9a-hexahydro-6,9-methano-2,4,3-benzodioxathiepin 3-oxide
ENDRIN	1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4- <i>endo-endo</i> -5,8-dimethanonaphthalene
HCB	hexachlorobenzene
HEPTACHLOR	1,4,5,6,7,8,8-heptachloro-3a,4,7,7a-tetrahydro-4,7-methanoindene
HEPTACHLOR EPOXIDE	1,4,5,6,7,8,8-heptachloro-2,3-epoxy-3a,4,7,7a-tetrahydro-4,7-methanoindan
LEAD	Pb
LINDANE	1,2,3,4,5,6-hexachlorocyclohexane, 99% or more gamma isomer
MERCURY	Hg
MIREX	dodecachlorooctahydro-1,3,4-metheno-1 <i>H</i> -cyclobuta[<i>c</i> , <i>d</i>]pentalene
PARATHION	<i>o</i> , <i>o</i> -diethyl <i>o</i> - <i>p</i> -nitrophenyl phosphorothioate
PCP	pentachlorophenol
POLYCHLORINATED BIPHENYLS (PCB's)	Mixtures of chlorinated biphenyl compounds having various percentages of chlorination
TDE (DDD) (Including its isomers and dehydrochlorination products)	1,1-dichloro-2,2-bis(<i>p</i> -chlorophenyl)ethane; technical TDE contains some <i>o,p'</i> -isomer also

Information for Contributors

The PESTICIDES MONITORING JOURNAL welcomes from all sources qualified data and interpretive information which contribute to the understanding and evaluation of pesticides and their residues in relation to man and his environment.

The publication is distributed principally to scientists and technicians associated with pesticide monitoring, research, and other programs concerned with the fate of pesticides following their application. Additional circulation is maintained for persons with related interests, notably those in the agricultural, chemical manufacturing, and food processing industries; medical and public health workers; and conservationists. Authors are responsible for the accuracy and validity of their data and interpretations, including tables, charts, and references. Accuracy, reliability, and limitations of the sampling and analytical methods employed must be clearly demonstrated through the use of appropriate procedures, such as recovery experiments at appropriate levels, confirmatory tests, internal standards, and inter-laboratory checks. The procedure employed should be referenced or outlined in brief form, and crucial points or modifications should be noted. Check or control samples should be employed where possible, and the sensitivity of the method should be given, particularly when very low levels of pesticides are being reported. Specific note should be made regarding correction of data for percent recoveries.

- Preparation of manuscripts should be in conformance to the *STYLE MANUAL FOR BIOLOGICAL JOURNALS*, American Institute of Biological Sciences, Washington, D. C., and/or the *STYLE MANUAL* of the United States Government Printing Office.
- An abstract (not to exceed 200 words) should accompany each manuscript submitted.
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- All copy, including tables and references, should be double spaced, and all pages should be numbered. The first page of the manuscript must contain authors' full names listed under the title, with affiliations, and addresses footnoted below.
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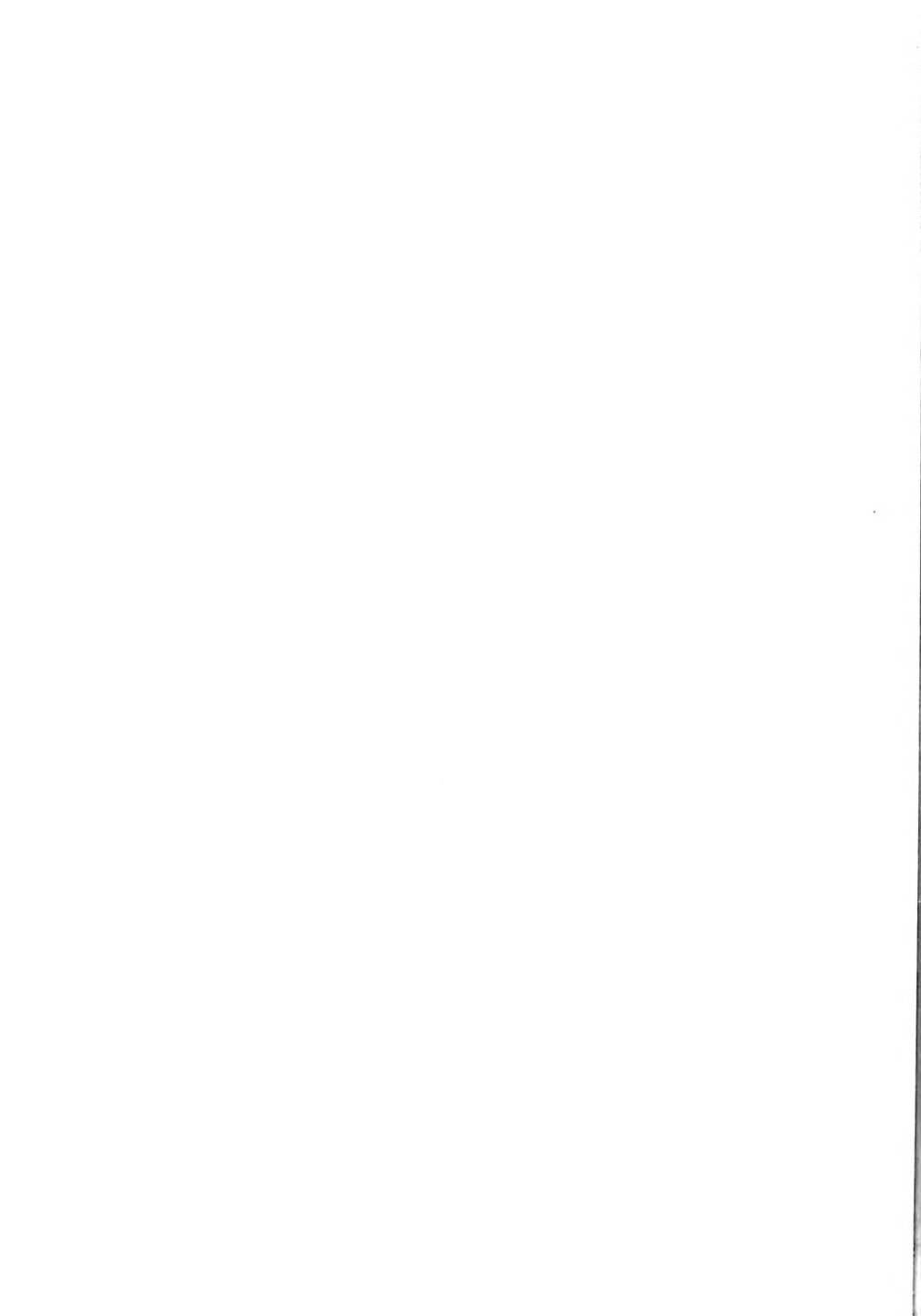
- Charts should be drawn so the numbers and texts will be legible when considerably reduced for publication. All drawings should be done in black ink on plain white paper.
- Photographs should be made on glossy paper. Details should be clear, but size is not important.
- The "number system" should be used for literature citations in the text. List references alphabetically, giving name of author/s, year, full title of article, exact name of periodical, volume, and inclusive pages.

The Journal also welcomes "brief" papers reporting monitoring data of a preliminary nature or studies of limited scope. A section entitled *Briefs* will be included, as necessary, to provide space for papers of this type to present timely and informative data. These papers must be limited in length to two Journal pages (850 words) and should conform to the format for regular papers accepted by the Journal.

Pesticides ordinarily should be identified by common or generic names approved by national scientific societies. The first reference to a particular pesticide should be followed by the chemical or scientific name in parentheses—assigned in accordance with *CHEMICAL ABSTRACTS* nomenclature. Structural chemical formulas should be used when appropriate. Published data and information require prior approval by the Editorial Advisory Board; however, endorsement of published information by any specific Federal agency is not intended or to be implied. Authors of accepted manuscripts will receive edited typescripts for approval before type is set. After publication, senior authors will be provided with 100 reprints.

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The WORKING GROUP is comprised of representatives of the U. S. Departments of Agriculture; Commerce; Defense; the Interior; Health, Education, and Welfare; State; and Transportation; and the Environmental Protection Agency.

The pesticide MONITORING PANEL consists of representatives of the Agricultural Research Service, Consumer and Marketing Service, Extension Service, Forest Service, Department of Defense, Fish and Wildlife Service, Geological Survey, Food and Drug Administration, Environmental Protection Agency, National Marine Fisheries Service, National Science Foundation, and Tennessee Valley Authority.

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Pesticide monitoring activities of the Federal Government, particularly in those agencies represented on the pesticide MONITORING PANEL which participate in operation of the national pesticides monitoring network, are expected to be the principal sources of data and interpretive articles. However, pertinent data in summarized form, together with interpretive discussions, are invited from both Federal and non-Federal sources, including those associated with State and community monitoring programs, universities, hospitals, and nongovernmental research institutions, both domestic and foreign. Results of studies in which monitoring data play a major or minor role or serve as support for research investigation also are welcome; however, the *Journal* is not intended as a primary medium for the publication of basic research. Manuscripts received for publication are reviewed by an Editorial Advisory Board established by the MONITORING PANEL. Authors are given the benefit of review comments prior to publication. Editorial Advisory Board members are:

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EDITORIAL

These Changing Times

The well-worn phrase that "time changes everything," or as some prefer to say, "everything changes with time," has been very much in evidence recently.

First of all, you have probably noticed that there is a new "face" on this issue of the JOURNAL. The editorial staff has arranged to upgrade the quality of the cover to be more attractive and durable, yet retaining those characteristics by which the periodical is quickly recognized. Mr. Reo Duggan, Chairman of the Editorial Advisory Board of the PMJ since publication began in June 1967, has retired from service with the Food and Drug Administration. This issue of the PMJ is a fitting tribute to his untiring efforts which have brought about international recognition of the JOURNAL as one of stature in its field.

Another Monitoring Panel member, Dr. Eugene Dustman, has also retired. "Dusty" as we know him, was Director of the Patuxent Wildlife Research Center, USDI. Both Dr. Dustman and Mr. Duggan were charter members of the Panel dating back to about 1964 when it was known as the Subcommittee on Pesticide Monitoring of the Federal Committee on Pest Control. These gentlemen have provided careful and expert guidance, and their absence is a challenge to us to uphold the respected manner in which they carried out their obligations.

In May, the Working Group approved the revised Charter of the Monitoring Panel, rewritten to be compatible with the new charter for the Federal Working Group on Pest Management. Among the charges to the Panel, is to continue to have a task group serve as the interagency Editorial Advisory Board of the PMJ for publication of monitoring data. New members have been appointed. Mr. John Wessel, FDA, was chosen Chairman, and Dr. Paul Sand, USDA, Vice Chairman. The Panel pledges its full support to the Advisory Board and editorial staff.

What else has time wrought? We'll no doubt see changes in monitoring programs in keeping with monetary and personnel constraints, and changes in methods for pest control or pest management—but that's another story for another time.

Best wishes to our retirees and those who have assumed new responsibilities.

Herman Feltz
Chairman, Monitoring Panel

PESTICIDES IN PEOPLE

*Total Mercury in Hair From 1,000 Idaho Residents—1971*¹

W. W. Benson and Joe Gabica

ABSTRACT

In a study of mercury in hair from 1,000 people throughout Idaho, mercury was found in all samples. The mean concentration was 4.18 ppm, and mean levels for samples from males and females were 2.45 and 5.90 ppm, respectively. Mercury levels ranged from a low of 0.12 ppm in a male to a high of 139.0 ppm in a female. No common source of mercury exposure was found, and there is no explanation at this time for the higher mercury levels in hair samples from women.

Introduction

During the fall of 1970, a study was conducted in Idaho to determine the presence of mercury in the environment, especially in pheasants. Because of the high mercury levels found in pheasants (1) and subsequently in fish (2) and other environmental samples, the study reported here was initiated to determine mercury levels in Idaho residents.

Interest in environmental mercury as a factor in human health has increased greatly during the past several years. Reports have been published based on accidental poisonings (3) and poisonings resulting from occupational hazards (4). Most of these studies, however, have been carried out on human tissues and have not included hair samples.

The current study was designed to survey mercury levels in hair samples from persons throughout Idaho to determine if levels were higher for residents of particular areas and to determine if the consumption of fish or other sources of exposure could be correlated with mercury levels in an individual. The study was also designed to provide baseline data for future studies.

Hair was studied since it was known that mercury, especially methyl mercury which is considered dangerous to human health (5), could be detected in hair and because such samples could be readily obtained.

Sampling Procedures

In order to compare levels in residents for different areas, sampling was carried out on the basis of the seven Idaho health districts which are apportioned by population and are approximately equal. Various organizations and individuals in each district were provided with return envelopes, printed to include name, address, age, sex, and occupation of participants as well as their consumption of fish and frequency of consumption. Initial requests met with an inadequate response; consequently, return envelopes were provided to barbers and beauticians in each district who supplied the required hair samples.

During analysis, if a sample tested above 15 ppm mercury, an additional hair sample was requested from that individual and a blood sample was also collected. Each of these persons was also asked to complete a questionnaire listing known mercury products or activities involving mercury products and was interviewed in an effort to determine, if possible, other sources of mercury exposure. Individuals with high levels of mercury in their hair were specifically asked if they used hair coloring and conditioning preparations, diuretics, skin bleaches, and ointments; however, no queries were made concerning dental fillings.

All samples collected from men were short hair obtained close to the scalp usually from the sides and neck and, thus, represented mercury in the body at the time of sampling. In contrast, the initial samples from women were usually terminal hair ends and, depending on the

¹ From the Juano Community Study on Pesticides, Idaho Department of Health, Statehouse, Boise, Idaho 83707.

length of hair, represented a body burden of mercury experienced previously. For example, the terminal end of hair 18 inches long, would represent that body burden experienced 12 to 18 months previously. Consequently, any repeat samples from women were requested to be hair from next to the scalp which would more accurately represent the present body burden. In all but one instance, the mercury levels were lower in subsequent samples from women. In the one exception, the level increased from 39 to 62 ppm. The current study presents only levels in initial hair samples; the subsequent samples will be correlated with blood levels of mercury in a future report.

Analytical Procedures

Each man's hair sample, since it was received as fine clippings, was mixed and an aliquot taken for analysis; a woman's hair sample, which was usually longer, had to be cut into fines and mixed before an aliquot was taken. In each case, a 1-g subsample was digested by either of two methods: Method A employed nitric and sulfuric acid digestion using the A.O.A.C. method and equipment (6); Method B used nitric and sulfuric acid (in a ratio of 10:1) digestion in an open 50-ml pyrex tube, placed in a boiling water bath for 2 hours. The two methods were compared by determining the final mercury levels for samples digested by both methods; and there was no significant difference (Table 1).

TABLE 1.—Mercury levels determined after A.O.A.C. method and tube method of digestion

RESIDUES IN PPM	
A O.A.C.	TUBE (50-ML)
1.60	1.54
2.40	3.08
0.54	0.80
21.40	21.40
86.68	100.00
2.00	1.92
1.90	2.08
4.90	5.00
1.10	1.40

The digested hair samples were then diluted to volume with distilled water in a 100-ml volumetric flask. The subsequent preparation procedure followed that outlined in the Manual of Analytical Methods (7) with the following modifications: The 100-ml solution was quantitatively transferred to a 300-ml BOD bottle; 2 ml of 5% potassium persulfate was added to each bottle and allowed to stand for 1 minute; 4 ml of 5% potassium permanganate was added to each bottle and allowed to stand for 1 minute; 2 ml of sodium chloride hydroxylamine sulfate was added to reduce the excess permanganate; and 5 ml of well-mixed stannous sulfate (or stannous chloride) suspension was added and the bottle immediately attached to the aeration assembly.

Prior to analyzing the hair samples, two types of equipment—the Coleman 50 mercury analyzer (C-50) and the Perkin-Elmer 303 atomic absorption spectrophotometer (P-E 303)—were compared by analyzing pheasant breast tissue. Results of analysis by each machine are given in Table 2. The C-50 provided accurate and rapid results when the mercury levels were above 0.05 μg , but for lower levels, the P-E 303 with a detection limit of 0.003 μg (8) was required.

Hair samples were analyzed using the C-50; however, to insure that accuracy was being maintained, the work was frequently checked by the A.O.A.C. method using the P-E 303.

If a hair sample contained residues over 15 ppm mercury, the remaining portion of hair taken from the individual was washed with a detergent having no measurable mercury, air-dried, and then reanalyzed. A

TABLE 2.—Mercury residues in pheasant samples determined by two instruments

PHEASANT SAMPLE NUMBER	MERCURY RESIDUES IN μG	
	P-E 303 ¹	C-50 ²
1	.152	.06
2	.175	.10
3	.187	.18
4	.225	.18
5	.601	.81
6	.352	.37
7	.130	.10
8	.150	.10
9	.100	.10
10	.190	.10
11	.120	.07
12	.135	.10
13	.098	.09
14	.225	.19
15	.225	.16
16	.086	.05
17	.088	.07
18	.742	.70
19	.913	.82
20	.691	.59
21	.404	.47
22	.233	.27
23	.231	.21
24	.154	.29
25	.203	.28
26	.264	.32
27	.600	.42
28	.124	.23
29	1.024	1.10
30	1.300	1.22
31	.384	.42
32	.940	.81
33	.790	.77
34	.261	.14
35	.480	.07
36	.100	.07
37	.210	.15
38	.131	.10
39	.065	.07
40	.474	.53
41	.100	.10
42	.090	.07
43	.065	.06
44	.081	.05
45	.070	.07
46	.125	.22
47	.055	.14

NOTE: Coefficient of Correlation: $r = .97$; Regression Line of Y on X: $Y = .970X - .001$; and Standard Error of Estimate of Y on X is $.067$.

¹ Perkin-Elmer 303 atomic absorption spectrophotometer

² Coleman 50 mercury analyzer.

solvent extract of hair was not undertaken because of the solubility of many alkyl mercury compounds (10). Results of analyses of 12 random hair samples are given in Table 3 and indicate that there were no appreciable differences in mercury levels before and after hair washing. Any variance that did occur may have been due to the inability to make two homogeneous subsamples from the same sample. Thus, the level of mercury was not considered attributable to external contamination, but was a part of the highly proteinized hair.

After every 100 samples were analyzed, a test to determine recovery values was done by analyzing an homogenized hair sample twice, first as received and second containing a spike control with the amount of the spike being unknown to the analyst. The average recovery of spike controls was 99.7%, regardless of the digestion method used. In addition, each day a known standard was analyzed to calibrate both analytical instruments.

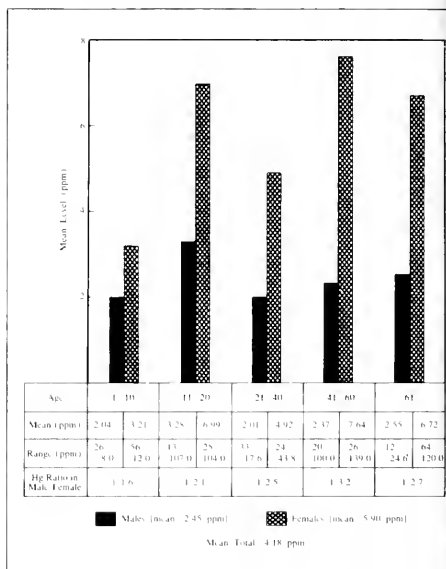
TABLE 3.—Mercury levels in 12 random samples of hair, analyzed as received and after washing

MERCURY RESIDUES IN PPM		
HAIR AS RECEIVED		WASHED HAIR
13.86		25.60
17.40		16.40
107.00		132.00
22.40		17.60
38.00		26.40
36.00		24.00
39.00		31.00
15.20		10.00
18.40		22.00
13.60		12.00
21.20		23.00
16.40		19.60

Results and Discussion

Mercury was found in all 1,000 hair samples, with the average concentration being 4.18 ppm. Mercury levels ranged from a low of 0.12 ppm in a male to a high of 139.0 ppm in a female. These levels in samples were subsequently evaluated according to the sex of the donor and by age groups for each sex (Fig. 1). The average level was higher for females (5.90 ppm) than males (2.45 ppm); similarly, according to the age group, females had mercury levels 1.6- to 3.2-fold higher than those for males. One or more individuals in every age group, however, exceeded the normally expected level of mercury in human hair of 10 ppm (9); however, only 20 men (3.37%) had levels above 10 ppm compared with 61 women (14.99%). In two age groups (41 to 60 years and 60+ years), the maximum range approached the 150 ppm mercury level in hair considered danger-

FIGURE 1.—Mercury residues in hair samples from 100 Idaho residents by age and sex of donors



ous (9). The highest level of mercury for males occurred for the age group 11-20 years; males within this age group are statistically the nation's largest food consumers (11).

In comparing those individuals with high mercury levels, diet did not seem to play any great role in determining these levels. Generally, individuals with high levels ate little or no fish and, in most instances, no wild game. None of these subjects stated that they were taking drugs containing mercury compounds, and no environmental exposure could be traced to individuals with higher levels. Since few persons with high levels used hair coloring or conditioning preparations, this was also eliminated as a source of mercury exposure. There did not appear to be any specific reason for the higher levels of mercury in females than males, as there was no available data to show evidence of environmental exposure differentially affecting males and females (5,12-14).

Sex differences in mercury levels might be influenced by hormonal mechanisms or the biochemical composition of the sexes; however, further studies in this area are needed.

This research was supported under Contract No. 68-02-0552 by the Division of Pesticide Community Studies, Office of Pesticides Programs, Environmental Protection Agency, through the Idaho State Department of Health.

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Comparative Organochlorine Pesticide Residues in Serum and Biopsied Lipoid Tissue: A Survey of 200 Persons in Southern Idaho—1970¹

Joe Wyllie, Joe Gabica, and W. W. Benson

ABSTRACT

Paired samples of serum and adipose tissue from patients undergoing abdominal surgery were studied in order to determine residue levels of organochlorine pesticides in the two tissues. Although the residue levels were found to vary with the type of pesticide and with sex and age of the donors, levels in Idaho residents did not differ greatly from persons elsewhere in terms of body burden. Attempts at establishing the degree of compartmental equilibria for *p,p'*-DDE and *p,p'*-DDT between the two tissues revealed that the distribution of these two residues was not directly proportional.

Introduction

Beginning with the report of Howell (1) in 1948 that DDT residues were present in human fat, the biological storage of persistent insecticides has been studied rather extensively. The literature on human storage was reviewed by Robinson (10). The pesticide content of human adipose tissue using samples obtained primarily from autopsy (2-6) has been determined for various areas of the world. Fat samples taken from living persons, by biopsy or during routine surgery, have understandably been examined less frequently (7-9).

For obvious reasons, blood (and in many cases, serum) has become the tissue of choice for monitoring pesticide residues in humans. However, despite the abundance of available information concerning the respective occurrence of pesticides in blood and fat, relatively little is known of their quantitative relationship to one another. To better understand this relationship, the Idaho Community Study on Pesticides examined 202 paired serum and adipose tissue samples obtained voluntarily from

hospitalized patients who lived in the highly agricultural region of southern Idaho.

Sampling Procedures

Both blood and adipose tissue samples were obtained from 141 female and 61 male Caucasian patients undergoing abdominal surgery at Saint Luke's and Saint Alphonsus Hospitals, Boise, Idaho, and Mercy Hospital, Nampa, Idaho. A 10-ml fasting whole blood sample was obtained from each volunteer during the hospital's routine blood collection for biochemistries and approximately 5 g of panniculus fat was taken during the subsequent surgery. In an effort to maintain a maximum degree of validity, only adequately nourished patients were surveyed.

Analytical Procedures

Whole blood was not extracted and analyzed. Because the Community Studies on Pesticides have evaluated serum analysis and use only serum samples in population studies, serum, rather than whole blood, was analyzed for serum-adipose tissue correlations.

Following whole blood centrifugation, serum samples were extracted for organochlorine insecticides by a revised Dale-Cueto triple hexane extraction method (11,12).

Two ml of serum was combined with 6 ml of nano-grade hexane containing a 20-ng internal standard of aldrin for subsequent recovery determinations and agitated for 3 minutes on a Vortex mixer. The mixture was then centrifuged for 10 minutes at 2,000 rpm and the hexane layer pipetted into a 50-ml concentrator tube. This procedure was repeated three times with unspiked

¹ From the Idaho Community Study on Pesticides, Idaho Department of Health, Statehouse, Boise, Idaho 83707.

hexane, and the combined hexane fractions were then concentrated by means of a modified Snyder column on a steam bath to a final volume of 500 μ l.

Adipose tissues were extracted by a modified Mills (13) procedure, as follows: the 5-g sample was placed in a mortar and pestle containing 10 g of clean, sharp sand and 1 g of anhydrous sodium sulfate. This mixture was then ground vigorously into a uniform dry granular mass. One ml of nongrade hexane containing 1 μ g of methoxychlor as an internal standard was added for subsequent calculation of percent recoveries. The resulting pulverized mixture was transferred to a 150-ml breaker by washing three times with 50-ml petroleum ether. This mixture was then filtered, evaporated to near dryness under nitrogen, cooled to room temperature in a desiccator, and reweighed for percent of fat content. Two grams of this fat weighed on an analytical balance was transferred to a 125-ml separatory funnel where partitioning, extractions with hexane, and subsequent column fractionation were carried out following the procedures previously reported by Mills (13) and Mills, Onley, and Gaither (14).

The concentrated hexane extracts of sera and fat were injected in 5- μ l aliquots into a MicroTek 220 gas chromatograph equipped with two different columns and tritium foil electron capture detectors. The operating analytical parameters were as follows:

Columns:	1/4" x 6' glass columns, packed with 1.5% OV-17 and 1.95% QF-1 on 100/120 mesh Chromosorb W, DMCS, HP or 4% SE-30 and 6% QF-1 on 80/100 mesh Chromosorb W, DMCS, HP
Temperatures:	Column 220° C Injection chamber 220° C Detector 205° C
Carrier gas:	Nitrogen
Flow rate:	OV-17 QF-1—70 ml/min SE-30 QF-1—100 ml/min

The two gas chromatographic columns used have a complete capability of separating for both quantitation and identification of the three isomers alpha-, beta-, and gamma-BHC and many other pesticides not before separable by a one-column determination.

All quantitation of pesticide residues was based on relative peak heights. Each fifth fat sample extract and pooled sera were qualitatively analyzed by thin layer chromatography for confirmation of pesticides reported. Recovery for the aldrin spike in sera and the methoxychlor spike in adipose tissues was 60-90% and 75-95%, respectively; results were corrected to 100%.

Results and Discussion

Seven organochlorine residues (*p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDD, *p,p'*-DDE, dieldrin, β -BHC, and heptachlor epoxide) were present in both fat and serum samples.

The means, ranges, and percent occurrence of these residues in samples are given in Table 1. *P,p'*-DDE was the most prevalent and the most highly concentrated compound in both tissues followed by *p,p'*-DDT. Although dieldrin was the third most prevalent residue in sera, it was the sixth most frequently detected in adipose samples. This finding for dieldrin supports the conclusions of Morgan and Roan (5) that dieldrin might be more effectively stored in nonlipid tissues. The remaining four compounds occurred quite frequently in adipose tissue, but were detected much less often in serum. The current findings in sera differ slightly from a previous study (16) of pesticide levels in sera from 1,000 persons from this region of Idaho in that mean serum *p,p'*-DDE concentrations are somewhat lower (15.5 ppb, this study versus 22.0 ppb) in the previous study and dieldrin in sera was detected much more frequently in this study (88%, this study versus 33%). The present results for pesticide residues in fat generally agree quantitatively with those of recent investigations in other geographic areas, with the possible exceptions of Hawaii (2) which reported lower *p,p'*-DDE mean fat levels and Holland (17) where studies with adipose tissue revealed *p,p'*-DDE at mean concentrations of only 1.7 ppm. Overall, however, residues in persons in Idaho appear to be comparable to those found elsewhere.

TABLE 1.—Residue levels of organochlorine pesticides in serum and adipose tissue—Idaho, 1970

COMPOUND	SERUM			ADIPOSE TISSUE		
	RESIDUES IN PPB		PERCENT OCCURRENCE	RESIDUES IN PPM		PERCENT OCCURRENCE
	MEAN	RANGE		MEAN	RANGE	
<i>p,p'</i> -DDE	15.5	2-70	100	7.2	0.2-30	100
<i>p,p'</i> -DDT	4.0	0-14	98	1.9	0.1-6.6	100
Dieldrin	0.9	0-10	88	0.2	0-0.7	87
β -BHC	0.3	0-15	23	0.3	0-2.5	98
<i>p,p'</i> -DDD	0.1	0-4	12	0.1	0-1.2	75
Heptachlor epoxide	0.1	0-2	18	0.1	0-1.3	97
<i>o,p'</i> -DDT	0.1	0-2	4	0.1	0-1.0	89

Concentrations of the four most commonly occurring pesticide residues in serum and adipose tissue (*p,p'*-DDE, *p,p'*-DDT, dieldrin, and β -BHC), as a function of sex of the persons sampled, are shown in Table 2. In both serum and fat, males were found to have significantly higher average levels of *p,p'*-DDE. Males also had higher average levels of *p,p'*-DDT than females in both serum and adipose tissue, but the difference was narrower in adipose tissue. Both sexes had comparable respective levels of dieldrin in both tissues. Mean β -BHC levels in sera were considerably higher in males than females, but mean levels in fat were the same for both sexes. The detection of higher pesticide levels in men agrees with previous findings in Idaho (16) and other locations (6,18), but is at variance with the results

TABLE 2.—Residue levels of *p,p'*-DDE, *p,p'*-DDT, dieldrin, and β -BHC in serum and adipose tissue by sex of persons sampled—Idaho, 1970

	COMPOUND	MALES (N = 61)			FEMALES (N = 141)		
		MEAN	RANGE	PERCENT OCCURRENCE	MEAN	RANGE	PERCENT OCCURRENCE
Serum (Residues in ppb)	<i>p,p'</i> -DDE	20.5	3-70	100	13.4	2-65	100
	<i>p,p'</i> -DDT	4.9	0-14	99	3.7	0-14	98
	Dieldrin	1.0	0-3	92	0.9	0-10	85
	β -BHC	0.6	0-15	26	0.2	0-7	20
Adipose Tissue (Residues in ppm)	<i>p,p'</i> -DDE	8.5	0.2-23	100	6.6	0.3-30	100
	<i>p,p'</i> -DDT	2.1	0.1-7	100	1.8	0.2-7	100
	Dieldrin	0.2	0-1	84	0.2	0-1	89
	β -BHC	0.3	0-3	97	0.3	0-2	99

TABLE 3.—Residues levels of *p,p'*-DDE, *p,p'*-DDT, dieldrin, and β -BHC in serum and adipose tissue by age of persons sampled—Idaho, 1970

	COMPOUND	AGE 0-20 (N = 16)		AGE 21-40 (N = 47)		AGE 41-60 (N = 80)		AGE 61-90 (N = 59)	
		MEAN	PERCENT OCCURRENCE	MEAN	PERCENT OCCURRENCE	MEAN	PERCENT OCCURRENCE	MEAN	PERCENT OCCURRENCE
Serum (Residues in ppb)	<i>p,p'</i> -DDE	7.57	100	14.27	100	16.11	100	17.87	100
	<i>p,p'</i> -DDT	2.55	100	4.46	98	3.92	98	4.52	98
	Dieldrin	.56	94	.88	83	1.06	89	.85	86
	β -BHC	.09	13	.34	21	.55	25	.36	20
Adipose Tissue (Residues in ppm)	<i>p,p'</i> -DDE	3.47	100	6.73	100	7.06	100	8.72	100
	<i>p,p'</i> -DDT	1.01	100	1.91	100	1.67	100	2.08	100
	Dieldrin	.14	100	.13	83	.17	88	.16	86
	β -BHC	.15	94	.24	100	.36	98	.34	97

of Fiserova-Bergerova *et al.* (19), who found no differences, as a function of sex, with respect to *p,p'*-DDT-derived materials in a group of 71 people in Florida. This tendency for males to store organochlorines at higher levels may be due in part to the relative complexity of female hormonal interrelationships which could conceivably result in increased microsomal enzyme activity and subsequent body burden reduction. The differing female pattern of fat deposition and the likelihood of males having greater environmental exposure to pesticides may also be contributing factors.

Residue levels of *p,p'*-DDE, *p,p'*-DDT, dieldrin, and β -BHC in serum and adipose tissue by age are shown in Table 3. Persons under 20 years of age had the lowest mean concentrations of all four pesticides in both sera and adipose; however, *p,p'*-DDE was the only residue that showed a consistent progression from the lowest level in people below 20 to its highest mean levels in persons ages 61 to 90 years. The tendency for *p,p'*-DDE levels to increase with age is what might be expected, and analysis of variance confirms this— $p = .01$ in serum and $p < .01$ in tissue. Mean levels of *p,p'*-DDT increased significantly in both tissues between age groups 0-20 and 21-40 years; however, there was no significant difference among the age groups 21-40, 41-60, and 61-90 years. This trend also carried a statistical significance in serum of $p = .05$ and in tissue, $p = .05$.

Both dieldrin and β -BHC had similar patterns, i.e. there were no significant differences between the mean values found in the different age groups for either of these compounds. The means of positive findings are low for each compound in the total sampling, but the levels found varied widely. The statistical evaluation of this is negative. In Florida (19) dieldrin levels in fat were reported to increase considerably in persons over 20 years of age; this could be due to differences in patterns of pesticide application and analytical techniques.

The increase of *p,p'*-DDE in man with age may be expected in terms of *in vivo* organochlorine catabolism, according to the generally accepted scheme of Peterson and Robinson (20). DDT is dechlorinated in the body to DDD which then degrades to the excretable DDA or is excreted as DDT. DDE storage is not appreciably derived from ingestion of DDT, however, but by ingestion of DDE previously broken down in the environment from DDT (15,21). Failure of DDE to be effectively eliminated would then result in body burden levels that increase with age. DDT, in contrast, would be broken down and excreted much more readily, and thus, tissue levels would not be expected to increase so dramatically with time.

The considerable difference between serum and adipose tissue in the relative frequencies of occurrence of *p,p'*-DDD, *o,p'*-DDT, β -BHC, and heptachlor epoxide

(Table 1) probably reflects the limitations of present analytical capabilities using serum samples of this size, rather than implying that these residues are sometimes present at high levels in fat but absent from serum. However, since *p,p'*-DDE and *p,p'*-DDT were detectable in both tissues in nearly all cases (Table 1), it would seem reasonable to correlate their respective tissue levels in an attempt to assay the degree of proportionality between serum and adipose tissue residues. This was done by plotting the individual mean serum levels of *p,p'*-DDE and *p,p'*-DDT against their corresponding values for fat. From this information, Pearson correlation coefficients (*r*) were then derived. These point distribution studies are portrayed graphically in Fig. 1 through 4. It appears from these data that *p,p'*-DDE and *p,p'*-DDT levels in serum and adipose tissue from the persons sampled are not directly proportional, although females do seem to show a more positive correlation (Fig. 1 and 3) between levels in the two tissues than do males (Fig. 2 and 4). However, differences could

be due to the occupationally and sexually related factors previously cited, as well as the smaller number of males sampled.

Barquet *et al.* (18) have postulated that serum and whole blood titres reflect both the DDE- and the total DDT-derived contents of adipose tissue; this assumption was based on two studies consisting of 59 and 65 persons each. This relationship has also been proposed by Radomski *et al.* (22) who examined 20 specimens of whole blood and adipose tissue obtained from autopsy sources and reported pesticide levels between the two tissues to be highly proportional. In contrast, we must tentatively conclude that lipid stored organochlorines are not always accurately predictable on the basis of serum levels. It thus appears that although serum residues may provide a convenient and useful estimate of acute exposure, their validity as an index of chronically acquired adipose body burden may be somewhat limited. To be sure, samples obtained from hospital

FIGURE 1.—Distribution of *p,p'*-DDE residue levels in serum and adipose tissue from 141 females

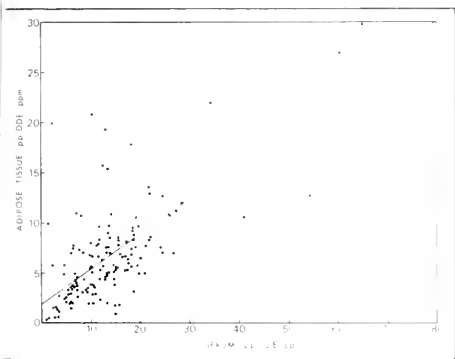


FIGURE 3.—Distribution of *p,p'*-DDT residue levels in serum and adipose tissue from 141 females

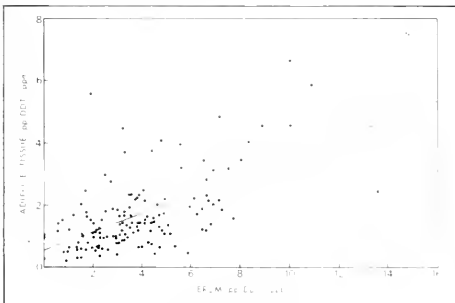


FIGURE 2.—Distribution of *p,p'*-DDE residue levels in serum and adipose tissue from 61 males

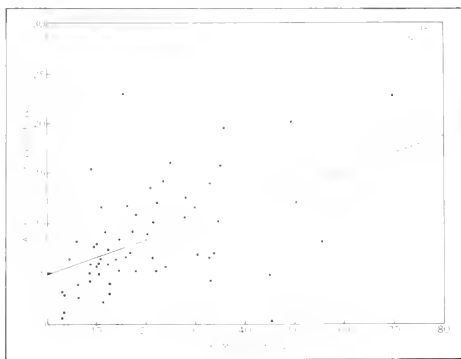
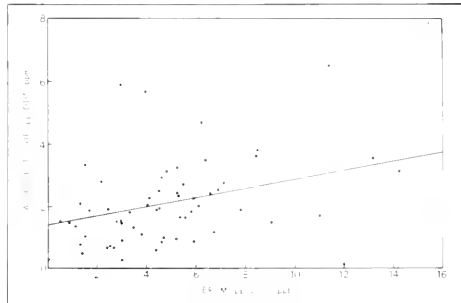


FIGURE 4.—Distribution of *p,p'*-DDT residue levels in serum and adipose tissue from 61 males



patients awaiting surgery are subject to some bias; for example, pathologically and chemotherapeutically induced biochemical changes could conceivably affect pesticide metabolism in such persons. However, the difficulties involved in obtaining adipose biopsies of sufficient size and number from the general population makes the use of hospital specimens a more practical choice at this time.

See Appendix for chemical names of compounds discussed in this paper.

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RESIDUES IN FOOD AND FEED

Arsenic Residues in Soil and Potatoes From Wisconsin Potato Fields—1970¹

D. R. Steevens, I. M. Walsh, and D. R. Keeney

ABSTRACT

Potato fields in Wisconsin known to have been treated with sodium arsenite (NaAsO_2) were surveyed in 1970 to determine residue levels of arsenic (As) in potato tuber peelings and flesh and in the soil. Total soil As residues ranged from 2.2 to 25.7 ppm and were generally related to the amounts applied. Potato tuber peelings contained 0.2 to 2.6 ppm As, but regardless of the amount of NaAsO_2 applied, the tuber flesh did not exceed 0.6 ppm As. It was concluded that As had not accumulated in these Wisconsin potato fields to potentially harmful levels.

Introduction

Accumulation of arsenic (As) in soil as a result of repeated use of pesticides containing As in orchards, cotton, and tobacco fields has been found to adversely affect the quality and quantity of crops grown (1,4). Studies to investigate the possible toxic effects and plant uptake of As applied to a Plainfield sand in Wisconsin (3,6) showed that phytotoxic effects to vegetable crops did not occur until 90 kg of As per hectare (ha) or more had been applied. Residual phytotoxicity remained for four cropping seasons after application of As at rates of 90 to 720 kg/ha. Except for potato peelings, edible portions of vegetable crops were not contaminated with As. In view of this finding and the fact that sodium arsenite had been used extensively in Wisconsin as a potato vine defoliant (applied at about 9 kg As/ha) from 1950 to 1968, this study was undertaken to survey commercial potato fields in Wisconsin to ascertain if excessive levels of As had accumulated in the soil and potato tubers (flesh and peelings) due to use of sodium arsenite.

Materials and Methods

In August 1970, soil and potato tuber samples were collected from a total of 18 potato fields in the three principal potato-growing areas of Wisconsin. At each site, a composite soil sample consisting of 10 randomly selected cores was taken from the plow layer. A composite potato tuber sample was also obtained at each site by taking tubers from five potato hills selected at random. The soil samples were air-dried (60 C) and ground (< 80 mesh). Potato tuber samples were scrubbed using distilled water and a nylon fiber brush. Potato tuber peelings and flesh were analyzed separately; samples were air-dried (60 C) and ground (20 Mesh).

For total As analyses, plant tissue samples were digested by using the $\text{HNO}_3\text{-HClO}_4$ procedure of Blanchard *et al.* (2), and soil samples were digested by the $\text{H}_2\text{SO}_4\text{-HClO}_4$ procedure of Small and McCants (5) with modification made by Jacobs *et al.* (3). This procedure gave quantitative (>95%) recovery of 10-100 ppm As added as As_2O_3 to soils. In addition, the amount of available soil As was determined by extracting with Bray P-1 solution (0.025N HCl and 0.03N NH_4F), an extractant commonly used to determine available soil phosphorus (3). Arsenic in the potato and soil digests and soil extracts was determined by the reduction-distillation method of Small and McCants (5) with modification made by Jacobs *et al.* (3). The sensitivity limit for this method was 0.1 ppm of As.

Results and Discussion

Results of analyses of soil and potato tubers are reported in Table 1; arsenic residues reported represent the average of analyses of duplicate samples.

¹ From the Department of Soil Science, Univ. of Wisconsin, Madison, Wis. 53706.

In general, total soil As levels were related positively with As application as reported by growers, although wide variations were noted (Table 1). Part of this disparity is probably due to the fact that the stated amount of As application was usually only an estimate. The level of naturally occurring As in soils is considered to be from 2 to 10 ppm (1,4). In this survey, As levels in the soil were <10 ppm at 7 sites and 10.0 to 25.7 ppm at 11 sites. The available Bray P-1 extractable As in soil was less than 5 ppm for all sites sampled. In comparison with the data obtained in experimental field plots (3,6), soil As levels were below any concentration detrimental to crops.

The As concentration in potato peelings taken from the survey sites ranged from 0.2 to 2.3 ppm, while the greatest As level in the flesh was 0.6 ppm. The As tolerance limit for several vegetable crops has been established at 2.6 ppm (7); all potato samples obtained in this survey were well below this limit, especially considering that the peelings constitute only a small portion of the total potato tuber.

Sodium arsenite was removed from registration as a potato vine defoliant in 1969. The results of this survey indicate that past usage of NaAsO₂ has not resulted in

levels of As in Wisconsin soils which would be phytotoxic or which would cause harmful levels of As to accumulate in potato tubers.

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TABLE 1.—Amount of arsenic applied as NaAsO₂, total and extractable Bray P-1 arsenic in soil samples, and arsenic concentration in potato tuber peelings and flesh from Wisconsin potato fields—1970

TYPE OF SOIL	SAMPLING SITES	SOIL			POTATO	
		TOTAL ARSENIC APPLIED (KG HA) ¹	TOTAL ARSENIC (PPM) ²	BRAY P-1 EXTRACTABLE ARSENIC (PPM) ²	PEELINGS (PPM) ²	FLESH (PPM) ²
Sandy loams (Northwestern Wisconsin, Spoooner area)	1	56	8.0	2.4	1.1	.4
	2	32	6.0	1.0	.5	.2
	3	32	9.8	2.8	1.6	.2
	4	25	17.4	2.9	1.8	.2
Silt loams (North-central Wisconsin, Antigo area)	5	65	25.7	1.4	.2	F
	6	20	5.4	.6	.2	T
	7	96	25.9	4.6	.5	.1
	8	25	10.4	1.0	.4	.2
	9	8	13.9	1.6	.4	.4
	10	48	16.0	1.1	.4	.2
Loamy sands (Central Wisconsin, Stevens Point area)	11	25	10.0	.5	.5	.4
	12	40	8.6	1.0	1.6	.4
	13	53	14.9	1.9	1.6	.4
	14	28	10.1	1.2	1.3	.6
	15	48	12.3	1.6	2.3	.1
	16	40	10.4	1.7	1.8	T
	17	0	2.2	.3	.4	T
18	16	5.7	1.0	1.3	T	
Plainfield sand [Data from a previous study in Wisconsin (6)]	—	0	3.6	.4	2.4	.1
	—	45	12.5	2.0	5.2	.1
	—	90	24.3	5.2	9.8	.1
	—	180	45.0	10.1	22.2	.5
	—	720	121.0	25.2	53.7	.6

NOTE: T = trace = <.1ppm.

¹ Growers' estimate of total arsenic applied to sites 1-18.

² Average of duplicate samples.

³ Phytotoxicity was noted in studies (3,6) after application of arsenic at rates of 90-720 kg/ha.

RESIDUES IN FISH, WILDLIFE, AND ESTUARIES

*Mercury Concentrations in Game Birds, State of Washington—1970 and 1971*¹

Frank F. Adley and Donald W. Brown

ABSTRACT

In a survey to determine the presence of mercury in the liver of game birds, 246 specimens contributed by local hunters during October 1970 to January 1971 and 4 obtained in May 1971 were analyzed. The species included pheasant, quail, chukar partridge, duck, geese, and grouse. Muscle samples were submitted with some of the liver specimens and, in those instances, these tissues also were analyzed.

The highest average mercury concentrations were in livers from mergansers (11.67 ppm) and teal (0.29 ppm), species with diets including aquatic organisms and thus differing from the other samples. The average mercury levels in liver from the other species were 0.08 ppm in pheasant; 0.03 ppm, quail; 0.06 ppm, chukar partridge; 0.12 ppm, ducks (other than teal and mergansers); 0.16 ppm, geese; and 0.02 ppm, grouse.

Of the species with both muscle and liver tissue analyzed, quail had the highest muscle to liver ratios of mercury and mergansers, the lowest.

In view of the practice of fall seeding with mercury-dressed seeds, the importance of the period of exposure to the treated seeds was investigated. No apparent correlation was evident between liver concentrations and the time of year the birds were killed.

Introduction

Relatively recent findings of elevated mercury levels in ducks from the Lake St. Clair-Detroit River area of Michigan (1) prompted investigations by several States and Canadian Provinces to determine levels of mercury in their wildlife. In some instances, (1) the concentrations of mercury were found to exceed the guideline limit of 0.5 ppm of mercury set by the Food and Drug Administration for edible portions of fish, and, in 1969, the pheasant season in the Province of Alberta was closed due to excessive mercury levels. In the interest of

game management, the State of Washington Department of Game instigated a study to monitor pheasant in certain areas where mercury-treated seeds were being used (Burton J. Lauckhart, *personal communication*).

Mercury concentrations above 0.5 ppm were detected in several pheasant livers and one breast; however, from an evaluation of the overall findings, it was concluded that mercury residues were not present at levels that would constitute a hazard to consumers.

To supplement this study, the Hanford Environmental Health Foundation of Richland, Wash., initiated a program to monitor, during the 1970-71 hunting season, other field-killed game birds native to the Columbia Basin, which is a consistently popular hunting area. Specimens were contributed largely by sportsmen and field personnel of the State Department of Game and were from the area of the Basin from Ephrata to the north down to Paterson, and from Prosser on the west to Dayton, a productive upland and migratory bird hunting area of about 8,000 square miles.

In view of the practice of fall seeding with mercury-dressed seeds in this region, the study was designed to detect any apparent increase in body burden of mercury as the season progressed. Thus, samples were collected from the beginning of the hunting season in October through the close of the hunting season in January.

Sampling Procedures

Uniform instructions for sampling were disseminated to those who would have specimens to contribute. It was requested that samples of whole liver be placed in plastic bags and kept frozen for delivery to the laboratory. Essentially all samples were identified with the following information: location and date of kill, type and sex of bird, statement as to general age, and name of submitter.

¹ From the Hanford Environmental Health Foundation, Inc., P. O. Box 100, Richland, Wash. 99352.

Analytical Procedures

Samples accurately weighed to .5 g were dissolved in 1:1 sulfuric-nitric acids at about 60 °C. On dissolution, the samples were cooled and a 6% w/v solution of potassium permanganate was added until purple color persisted. This was made to 100-ml volume, and an aliquot was analyzed by flameless atomic absorption spectrophotometry. In this procedure (2,3) the mercury compounds are reduced to elemental mercury by the addition of stannous chloride, and the mercury is swept by an air stream through an absorption cell mounted on the burner of an AA unit. The resulting absorption is measured and compared to a calibration curve. This procedure has a sensitivity of 0.01 ng of mercury as mercuric chloride per sample per 1% absorption. Reproducibility was determined to be better than $\pm 10\%$ using mercuric chloride standard.

Results and Discussion

During the season, over 300 specimens were received by the laboratory. Of these, 250 were selected, Table 1, which could provide the best coverage of the parameters selected for investigation. These included four mergansers collected in May 1971 as part of a Federal monitoring program. All others were collected between October 1970 and January 1971.

The results of mercury analysis of liver samples by month of collection are given in Table 2. For the 85 pheasant analyzed, the mercury concentration averaged 0.08 ppm; three of these samples had concentrations exceeding 0.5 ppm, the highest being 0.71 ppm. The average level in the 64 ducks (mostly mallard and excluding teal and mergansers) was 0.12 ppm with none exceeding 0.39 ppm. The mercury level for the 15 geese averaged 0.16 ppm; for the 13 chukar partridge, 0.04 ppm; the 10 quail, 0.03 ppm; and the 1 grouse, 0.02 ppm. The highest average levels were in the 6 mergansers and 10 teal, 11.67 ppm and 0.29 ppm, respectively.

The higher values for mergansers and teal are interesting since they differ in diet from the other birds. The normal diet for mergansers consists of small fish, insects, and crustacea, and the diet of the teal may include certain aquatic life having an elevated methylmercury content.

There appeared to be no marked increase in body burden of mercury through the hunting season, October through January, although ducks (excluding teal and mergansers), chukar, and quail did show higher levels in the latter part of the season (Table 2).

In some instances, muscle tissue was also submitted with liver samples. The analytical findings are shown in Table 3. Quail and geese had the highest muscle to liver ratios, whereas the mergansers had consistently

TABLE 1.—Upland and migratory birds analyzed—Washington, 1970-71

SPECIES	NUMBER ANALYZED
Pheasant	85
Ducks	
Teal ¹	10
Mergansers ¹	6
Other	64
Geese	15
Chukar Partridge	59
Quail	10
Grouse	1
TOTAL	250

¹ Separated due to eating habits.

TABLE 2.—Mercury concentrations in livers of birds by month of collection

SPECIES	DATE OF COLLECTION	NUMBER ANALYZED	AVERAGE Hg CONCENTRATION (PPM)	RANGE (PPM)
Pheasant	October	35	0.05	0.01-0.15
	November	23	0.12	0.01-0.71
	December	27	0.09	0.003-0.45
	Total	85	0.08	0.003-0.71
Ducks Teal ¹	October	6	0.39	0.009-0.47
	November	4	0.13	0.01-0.17
	Total	10	0.29	0.009-0.47
Mergansers ¹	November	2	31.10	4.15-58.00
	May 1971 ²	4	1.97	0.80-3.40
	Total	6	11.67	0.80-58.00
Other	October	16	0.07	0.03-0.08
	November	10	0.07	0.02-0.11
	December	14	0.12	0.06-0.17
	January	24	0.16	0.07-0.39
Total	64	0.12	0.02-0.39	
Geese	November	7	0.226	0.14-0.34
	December	7	0.097	0.05-0.16
	January	1	0.08	—
	Total	15	0.16	0.05-0.34
Chukar Partridge	October	39	0.03	0.02-0.09
	November	7	0.05	0.02-0.21
	December	13	0.06	0.03-0.11
	Total	59	0.04	0.02-0.21
Quail	October	5	0.02	0.01-0.02
	November	5	0.03	0.01-0.07
	Total	10	0.03	0.01-0.07
Grouse	October	1	0.02	—

¹ Separated due to eating habits.

² Federal monitoring sample.

TABLE 3.—Muscle/liver mercury concentration ratios by species

SPECIES	MUSCLE-LIVER RATIOS
Pheasant	0.65, 0.75
Quail	3.0, 2.5, 1.0, 1.0
Geese	0.8, 1.25, 3.8, 0.7
Mergansers	0.20, 0.16, 0.20, 0.21

low muscle to liver ratios. It is possible that mergansers, due to their diet of aquatic life high in the food chain, possess relatively less mercury in their edible muscle tissue than in liver tissue.

The pheasant is one of the most important upland bird species since over 200,000 pheasants are harvested annually by hunters in the Columbia Basin area (4). During the 1969-70 winter season, the State of Washington Department of Game monitored 37 wild pheasants taken as road kills from Eastern Washington (Burton J. Lauckhart, *personal communication*). The average mercury content of liver in this group was 0.24 ppm. Since that time ranchers, seed producers, and the Federal and State governments have taken steps to minimize the use of mercury-bearing seed dressings. It is noteworthy that the findings of this study reflect a reduction in the mercury content of pheasants from an average of 0.24 ppm in 1969-70 to 0.08 ppm in 1970-71, which may be attributed to a general reduction under way in the use of mercury-dressed seeds.

Acknowledgment

This study could not have been performed without the cooperation of the members of the Richland Rod and Gun Club and other interested sportsmen. The advice and cooperation of the Division of Game Management, Washington State Department of Game, and publicity by radio station KONA, were also helpful. The authors gratefully acknowledge all those who made this work possible.

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Effects of Estuarine Dredging of Toxaphene-Contaminated Sediments in Terry Creek, Brunswick, Ga.—1971¹

Charles J. Durant and Robert J. Reimold

ABSTRACT

The possible reintroduction of toxaphene into estuarine biota from dredging and displacement of contaminated sediment in Terry Creek, Brunswick, Ga., was studied. In the estuary, the sediments near a toxaphene plant outfall were found to be contaminated with toxaphene approaching 2,000 ppm, and oysters collected 2 miles from the outfall were found to contain residue levels near 6 ppm. Analyses of oysters and sediment before and after dredging operations revealed no significant increase of toxaphene residues resulting from the dredging and resultant spoil runoff.

Introduction

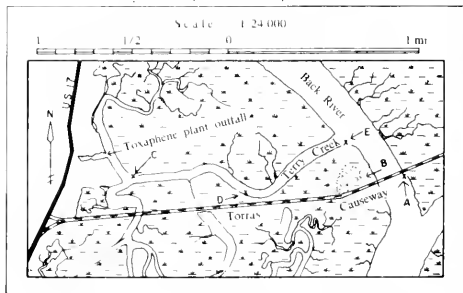
Terry Creek, Brunswick, Ga., is an estuarine watershed through which industrial wastes have been discharged to the ocean for the past 50 years; among the contaminants introduced over the past 20 years have been the manufacturing residues and wastes of toxaphene. Previous surface and subsurface sediment core samples had revealed high concentrations of toxaphene in the sediments. It was reasonable to assume that a proposed dredging operation from June 10-13, 1971, to widen and deepen the existing channel of Terry Creek (Fig. 1) would release concentrations into the water and that the toxaphene would be incorporated into the biota, perhaps causing mortality. The purpose of this study was to determine levels of toxaphene in nearby oysters and sediment, measure any increase in these levels, and assess any ecological damage caused by the dredging operation.

Sampling Procedures

The oyster (*Crassostrea virginica*) was used as an indicator of contamination because it is a sessile filter feeder with the ability to concentrate pesticides up to 70,000 times that found in the surrounding water (1). Oysters were collected at the Torras Causeway toll bridge crossing Back River (Fig. 1, location A) weekly from April 1 to June 23, 1971, and monthly from then until October 12, 1971. Each sample was taken from as near as possible to mean low water and included a minimum of 12 shucked oysters which were placed in a mason jar.

Three sediment core samples were taken from Terry Creek on June 10, 1971, one each from locations C, D, and E (Fig. 1). Location C was a point on the north shore of Terry Creek, 50 yd from the junction with Dupree Creek and .2 mile from the toxaphene plant outfall; location D was on the south shore of Terry Creek, .8 mile from the plant outfall and one-half the distance

FIGURE 1.—Geographic locations of collection sites near Terry Creek, Brunswick, Ga.—1971



¹ Contribution 330 from the Marine Institute, The University of Georgia, Sapelo Island, Ga. 31327.

from the outfall to Back River; and location E was on the south shore of Terry Creek, 1.4 miles from the plant outfall and 50 yd west of Back River. Each sample (70 mm diameter, 80 cm long) was collected with a clear plastic core barrel. Samples were extruded from the liner in 10-cm increments by forcing the sediment out with a tightly fitting rubber plunger. In addition to the core samples, a composite sediment sample consisting of six surface grab samples was collected from a transect across the dredge spoil area (location B, Fig. 1) on June 23, 1971, about 10 days after dredging ceased.

In addition to sampling, field observations of biota were made in the vicinity of the dredging operation on June 10 and 11.

Analytical Procedures

Analytical procedures generally followed those reported by Wilson (2). Each oyster sample (12 specimens) was homogenized in an Osterizer, and a 30-g aliquot of the homogenate was then mixed with a desiccant consisting of 9 parts anhydrous powdered sodium sulfate and 1 part Quiso (a micro fine precipitated silica). The mixture was alternately frozen and blended until a free-flowing powder was obtained. The samples were then Soxhlet extracted for 4 hours with glass-distilled petroleum ether, concentrated and partitioned with acetonitrile, and evaporated to dryness at room temperature. The residue was eluted from a Florisil column with 6% ethyl ether in petroleum ether.

Sediment samples were processed as follows: Each 10-cm increment in the core was placed in a separate beaker and thoroughly mixed. An aliquot of each sample was thinly spread in the bottom half of an open petri dish and dried in darkness at ambient temperature for a minimum of 5 days. A 5- to 15-g aliquot of the dried sediment was mixed with a desiccant in a 1:3 ratio by weight. The extraction and cleanup procedures were identical to those previously described for oyster samples.

The oyster and sediment extracts were then quantified on Varian model 600 D gas chromatographs equipped with tritium electron capture detectors and glass columns (5' by 1/8", o.d.) packed with 3% DC-200. Operating parameters were: oven temperature—193° C; injector and detector temperatures—210° C; and carrier gas—N₂ at 20 ml/min.

For confirmation, extracts were injected on a mixed column of 5% QF-1 and 3% DC-200 (1:1 by weight) with the same operating parameters as above.

Recovery rates of toxaphene in oysters and in sediments were above 85% and 90%, respectively. Toxaphene

concentrations below 0.25 ppm were considered insignificant for the purpose of this study. The data were not corrected for percent recovery.

Results and Discussion

The results of toxaphene analyses of oyster samples are presented in Fig. 2. The mean of the weekly concentrations in May was 3.30 ppm. Oysters collected on June 10, 1971, at about 10 a.m. on the day the dredging operation began, had a higher concentration than subsequent samples but contained 1.24 ppm less than the May average.

The results of toxaphene analysis of the sediment core samples are summarized in Table 1. The concentrations in the first 10-cm increment (surface to 10-cm depth) of the three samples ranged from 35.5 to 1,858.3 ppm. The composite sample of surface sediment obtained after dredging (site B) contained 32.8 ppm toxaphene on a dry-weight basis.

Analyses of oysters and sediment (before and after dredging operations) revealed no significant increase of toxaphene residues resulting from the dredging and resultant spoil runoff.

During the course of field observations, a few anchovies (which are also filter feeders) were observed in distress as a result of the dredging operation. The cause appeared to be related to the heavy suspended sediment

FIGURE 2.—Concentration of toxaphene (in ppm, wet-weight basis) in the American oyster (*Crassostrea virginica*) collected from near the intersection of Terry Creek with Back River, Brunswick, Ga. (as depicted in Fig. 1)

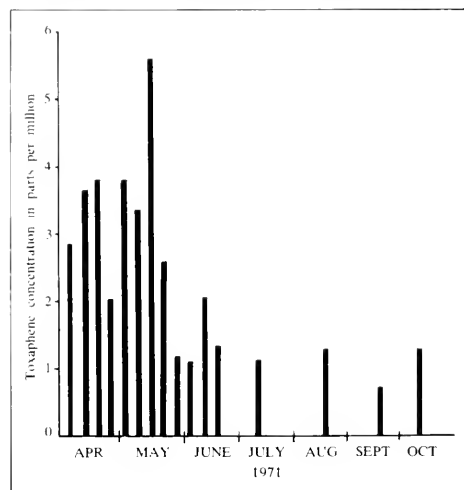


TABLE 1.—Toxaphene concentrations in three sediment cores, by 10-cm increments, collected from Terry Creek, Brunswick, Ga.—June 10, 1971

SAMPLING LOCATION	RESIDUES IN PPM AND CONTENT OF SEDIMENT							
	SURF. TO 10 CM	10-20 CM	20-30 CM	30-40 CM	40-50 CM	50-60 CM	60-70 CM	70-80 CM
Site C ¹	1,858.3 mud & some chips	1,340.5 mud & few chips	1,324.0 mud	1,367.2 mud	1,236.7 mud	433.6 mud	68.5 mud	83.2 mud
Site D ²	111.85 mud & few chips	615.64 mud & many chips	16.04 mud	17.46 mud	5.42 mud	3.4 mud	2.88 mud	—
Site E ³	35.5 mud	35.47 mud	21.9 mud	70.65 mud	79.8 mud	21.0 mud	18.5 mud	5.27 mud

NOTE: — = no sample taken.

¹ North shore of Terry Creek, 50 yd from junction with Dupree Creek and .2 mile from toxaphene plant outfall.

² South shore of Terry Creek, .8 mile from the plant outfall and one half the distance from the outfall to Back River.

³ South shore of Terry Creek, 1.4 miles from the plant outfall and 50 yd west of Back River.

load in the water which clogged the gill cavities of the fish, probably causing them to suffocate, rather than related to pesticide poisoning. Several weeks prior to the Terry Creek operation, menhaden and anchovies were observed in distress with clogged gill covers in the vicinity of another dredging operation in the Intra-coastal Waterway just north of Altamaha Sound. No lethal concentrations of pesticides were suspected in the Altamaha Sound area, since oysters collected there at monthly intervals for several years have contained no more than trace amounts of pesticides and no toxaphene.

The anticipated sudden kill of fish and shellfish from toxaphene poisoning in Terry Creek did not occur, probably for a combination of reasons: (1) There was less runoff of toxaphene contaminated sediments than expected from the dredging operation. (2) The toxaphene was bound to the clay particles and therefore not available to the biota. (3) The toxaphene in the lower sediments (below 10 cm) had undergone degradation and detoxification; chromatograms of sediments showed a marked difference with depth (illustrated by a buildup of peaks in the first portion of the chromatograms and diminishing peaks in the later portion) suggesting *in situ* dechlorination. (4) Lastly, the possibility exists that some fin fish were able to avoid the area in question.

Any future dredging activities should be planned and coordinated with further research. The relation of suspended sediment load to toxaphene concentration should be of primary concern. Additional research is needed to document whether or not sediment toxaphene concentration increases in the direction from the mouth of Terry Creek toward the toxaphene plant outfall (Fig. 1). Detoxification studies in the form of static bioassays using toxaphene extracted from core sediments are currently in progress.

See Appendix for chemical name of toxaphene.

This work was funded in part by a grant from Hercules, Inc. to the University of Georgia.

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Organochlorine Insecticide Residues in Water, Sediment, and Organisms, Aransas Bay, Texas—September 1969 - June 1970

Roger R. Fay¹ and Leo W. Newland²

ABSTRACT

An investigation was conducted to determine the presence and distribution of organochlorine insecticide residues in Aransas Bay and its contributing bays at Rockport, Tex. A total of 80 water samples, 29 sediment samples, and 11 samples of 8 different types of organisms were collected and analyzed from September 1969 through June 1970.

Organochlorine insecticide residues were detected in only 3 water samples and 3 sediment samples and, although residues were detected in 8 of the 11 organism samples, these were all at low levels (<67 ppb). The predominant residues found in the organisms were dieldrin, *p,p'*-DDD, and *p,p'*-DDE. The occurrence or concentration of residues could not be related to salinity or pH of the water or percent organic content of the sediments.

Introduction

The extensive use of organochlorine insecticides has led to an accumulation of these residues in the environment. In estuaries, the persistence of organochlorine residues creates a hazard to fish and other forms of marine life (3). Estuaries, in addition to supporting large shellfish populations, provide spawning and nursery grounds for many species of fish. Shellfish larvae and young fish are extremely susceptible to chemical pollutants and other forms of environmental stress. The problem is compounded by the fact that some fish and shellfish are known to concentrate organochlorine residues in their tissues and, thus, pass these residues along the food chain.

Insecticides entering the estuary in water can be introduced into the food chain (1) when residues are absorbed by plankton or other organisms and (2) when residues are adsorbed to suspended particles which settle and become part of the bottom sediment where benthic fauna feed (5). Insecticides entering on silt and detritus also may become part of the sediment.

The food chain at the same time plays an important role in removing insecticides from estuaries since fish and porpoise immobilize insecticide residues in their body tissues and, thus, these insecticides are removed from estuaries when fish migrate to sea. Similarly, fish-eating birds accumulate large amounts of insecticides and then distribute them along migratory pathways. The major portion of insecticides entering estuaries, however, are lost through dilution, chemical decay, and adsorption by bottom deposits (5).

The effects on aquatic organisms of chronic exposure to organochlorine insecticides were first realized with the reproductive failure of lake trout in New York and high mortalities in hatchery-reared coho salmon related to *p,p'*-DDT residues in the eggs (2). Investigations have shown that, of the commonly used organochlorine insecticides, endrin has the highest acute toxicity to fish. Fish subjected to chronic exposure of low concentrations of endrin were found to be more susceptible than control fish to lethal concentrations of endrin during the first 24 hours of exposure; however, by 72 hours the two groups exhibited similar survival (10).

Extensive experimentation has been conducted to determine the effects on oysters of chronic exposure to sublethal concentrations of *p,p'*-DDT and other organochlorine insecticides. Under controlled laboratory conditions, oysters exposed to *p,p'*-DDT at concentra-

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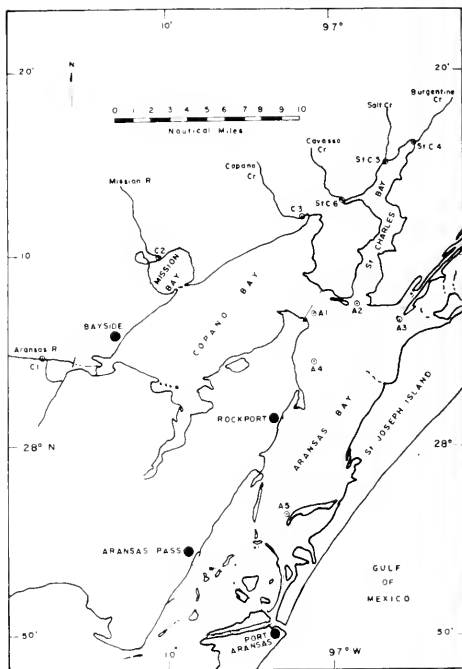
tions of 1 to 2 parts per billion (ppb) at 30 °C appeared to grow and behave in a manner similar to that of the control groups; analyses of the tissues, however, revealed concentrations of *p,p'*-DDT many times that of the environment (4). A concentration of 7 to 10 ppb *p,p'*-DDT in water inhibited 50% of the normal shell deposition in oysters (3). Physiological irritation was shown by spasmodic shell movements when the concentration of *p,p'*-DDT in the water was increased to 0.1 parts per million (ppm), and at a concentration of 1 ppm, the oysters usually remained closed (4). In the presence of continuous low level *p,p'*-DDT pollution, oysters concentrated the insecticide in their tissues to more than 25 ppm within 1 week without showing any harmful effects; however, 50% of the small fish and shrimp fed this oyster meat died within 2 days (4).

Dissection of oysters containing *p,p'*-DDT residues has shown that 67% of the *p,p'*-DDT was stored in the intestinal tract, digestive gland, and gonads. The gonads were found to be a major site of *p,p'*-DDT storage with residues in excess of 25 ppm reported in the gametes. An attempt was made to culture oyster larvae from such highly contaminated gametes in order to determine the effects of inherent *p,p'*-DDT on oyster development. Unfortunately, the experiment failed at an early stage due to technical difficulties in the laboratory (4).

The Gulf Breeze Laboratory of the Environmental Protection Agency (formerly under the Bureau of Commercial Fisheries, USDI) at Gulf Breeze, Fla., is engaged in a monitoring program for organochlorine insecticides. Oyster samples from coastal States are collected at 30-day intervals and analyzed for organochlorine insecticide residues. In 1967, oyster samples were obtained on a monthly basis from 12 stations in Texas. Of the 129 samples analyzed, 90% contained one or more organochlorine insecticide residues (6). A similar investigation of Galveston Bay following extensive mosquito control programs in the fall of 1964 (7) reported no indication of elevated insecticide levels in water and oyster samples. Insecticide levels were low in both water and oysters, usually less than 0.01 ppm if detected.

The study reported here was conducted to determine the organochlorine residues in Aransas Bay, Tex., and its adjacent contributing bays, Copano, Mission, and St. Charles, (Fig. 1) from September 1969 through June 1970. The land area surrounding Aransas, Copano, and Mission Bays is primarily ranch land with secondary agricultural usage. The area is drained by the Aransas and Mission Rivers and the Copano, Cavasso, Salt, and Burgentine Creeks. Samples collected for analysis included water, sediment, and various types of organisms. Locations of sampling stations are shown in Fig. 1.

FIGURE 1.—The area of investigation showing sampling stations, Aransas Bay, Texas—1969-70



Collection of Samples

Water samples were collected monthly from September 1969 through June 1970. Samples were taken at the surface in clean 2-qt glass jars with aluminum foil lined lids and stored in darkness to prevent photochemical alteration of insecticide residues.

Sediment samples were collected in September and November 1969, and January, May, and June 1970 using a grab sampler; samples were then refrigerated to minimize loss of the insecticide due to biochemical degradation.

Dates of collection of organism samples listed below depended on their availability; shrimp, oysters, and crabs were obtained in January and March 1970, and the fish samples were collected in July 1969.

sea trout	<i>Cynoscion arenarius</i>
drum	<i>Paralichthys crotmis</i>
oysters	<i>Crassostrea virginica</i>
blue crab	<i>Callinectes sapidus</i>
silverside minnows	<i>Menidia menidia</i>
yellowtail croaker	<i>Bairdella chrysura</i>
ribbon fish	<i>Trichiurus lepturus</i>
brown shrimp	<i>Penaeus aztecus</i>

Each sample represented an average of at least 12 individuals of a species. All organisms were collected with a trawl, except for oysters which were picked from the reef, and then frozen until analysis.

Analytical Procedures

The procedure for quantitatively extracting organochlorine insecticides from water, sediments, and organisms is described below. The solvents, hexane and hexane:acetone (41:59) used in the extraction techniques, were repurified by glass distillation employing a 3-ball Snyder column.

WATER

One liter of water was separated from suspended material by centrifugation or settling; then 500 ml of the sample was extracted with 100 ml of hexane by shaking for 1 minute in a separatory funnel. After separation of the layers was complete, the water was drawn off; the remaining 500 ml of water added, and the sample extracted by shaking for 1 minute. If emulsification was encountered between the hexane and water, anhydrous Na₂SO₄ was added to break the emulsion. The hexane phase was quantitatively transferred to a Kuderna-Danish concentrating apparatus equipped with a 3-ball Snyder column and concentrated in a water bath of 5 ml. The extract was transferred to a 10-ml volumetric flask and brought to volume. The hexane extract was analyzed directly by gas-liquid chromatography (GLC without further cleanup).

SEDIMENT

On removal from refrigeration, the sediment samples were spread and allowed to dry at room temperature. The dry sediments were ground with a mortar and pestle to pass through a #30 mesh sieve, thus insuring uniform saturation of the sample with solvent. A 20- to 30-g subsample (dry weight) was weighed, placed in a Soxhlet extraction apparatus, and refluxed for 6 hours using hexane:acetone as the extracting solvent. Since acetone is unsuitable as a solvent in GLC employing an electron capture (EC) detector, the insecticides were partitioned into the hexane phase by the addition of water. The hexane phase was then transferred to a Kuderna-Danish evaporating apparatus with a Snyder column and concentrated to approximately 10 ml for cleanup by column chromatography.

ORGANISMS

Immediately after thawing, the individual organisms making up a sample were ground to a fine mixture in a blender. A portion of this homogenate was weighed, and three times the subsample's weight of anhydrous Na₂SO₄ was added. The mixture was blended thoroughly to insure adequate distribution of the Na₂SO₄ and adsorption of the water. The sample was extracted following the same procedure as described for sediments.

Following the extraction and partitioning of the insecticides into the hexane phase, the sample was concentrated in the Kuderna-Danish apparatus to approximately 10 ml for cleanup by column chromatography.

CLEANUP OF SAMPLE EXTRACTS

The presence of relatively large amounts of interfering materials in sediments and biological samples requires that these samples be cleaned up prior to analysis by GLC. Column chromatography was selected as a method of cleanup because of its large carrying capacity and its ability to pre-separate insecticide mixtures, thereby aiding in the analysis of subsequent gas chromatograms. Florisil (activated magnesium silicate) was employed as the adsorbent, and elutions were made with 5% ether in hexane and 15% ether in hexane. The procedure followed was essentially that described in reference (9) with modifications (8).

GAS-LIQUID CHROMATOGRAPHY

A Varian Aerograph model 204B equipped with a H-foil EC detector was employed for the separation and identification of organochlorine insecticide residues. The use of all glass columns and on-column injection was required to avoid sample degradation resulting from contact of the sample with metal surfaces. Gas chromatographic conditions were as follows:

Columns	2 m long x 4 mm i.d. glass packed with 10% DC-200 or 10% QF-1 on 60/80 mesh Gas Chrom Q
Temperatures:	Columns: 195° C (QF-1) 215° C (DC-200) Injector: 230° C
Carrier gas:	N ₂ with a flow rate of 75 ml/min
Sensitivity	1.2 x 10 ⁻¹¹ amps full scale

Identification was based on retention times from the polar (QF-1) and non-polar (DC-200) columns. The use of two columns of differing polarity decreases the possibility of erroneous identifications. An L&N Speedomax W recorder with a disc integrator was used to calculate peak areas.

All of the samples were analyzed for the following organochlorine insecticides: γ -BHC, heptachlor, heptachlor epoxide, aldrin, dieldrin, endrin, *p,p'*-DDE, *p,p'*-DDD, and *p,p'*-DDT. Analysis was not performed for polychlorinated biphenyls, since the investigation was planned and executed prior to the availability of methods and standards for these compounds.

RECOVERY

Organochlorine insecticide standards of greater than 99% purity were obtained from the following sources: heptachlor, heptachlor epoxide, and endrin from Velsicol Corporation; aldrin and dieldrin from Shell Chemical Company; *p,p'*-DDD from Rohm and Haas; γ -BHC from the U. S. Food and Drug Administration; *p,p'*-DDT from City Chemical Corporation; and *p,p'*-DDE

from the Pesticide Repository, Perrine Primate Laboratory, Environmental Protection Agency. Recovery rates were assumed to be in the range of 90-100% according to the methods used (1,8,9).

Results and Discussion

The results of the analysis for organochlorine residues are considered below, followed by an interpretation of these data. An attempt was made to relate the presence or concentration of insecticide residues to the physical and chemical characteristics of the area. Salinity and pH data of the water samples were obtained from the Texas Department of Parks and Wildlife at Rockport, and organic content of the sediments was determined by combustion of the sample at 500° C. A summary of these characteristics of the water and sediment from each sampling station is presented in Table 1.

TABLE 1.—Salinity and pH of water and organic matter content of sediments at the sampling stations, Aransas Bay, Texas—1969-70

SAMPLING STATION	WATER		SEDIMENTS
	SALINITY (PPT)	pH	PERCENT ORGANIC CONTENT (DRY-WEIGHT BASIS)
A-1	15.0	8.3	2.1
A-2	12.2	8.3	1.7
A-3	11.1	8.3	14.3
A-4	16.1	8.3	5.4
A-5	20.5	7.9	5.0
C-1	6.6	8.1	5.7
C-2	1.1	8.2	4.7
C-3	8.9	8.0	1.8
St. C-4	11.1	8.4	6.9
St. C-5	12.2	8.3	8.2
St. C-6	8.3	8.4	10.2

TABLE 2.—Organochlorine residues in water, Aransas Bay, Texas—1969-70

[T = Trace = <0.1 ppb; — = not detected]

COLLECTION DATE	SAMPLING LOCATION	RESIDUES DETECTED IN PPB (NG/G)
1969		
Sept.	A-1	p,p'-DDT (T) endrin (4.4)
	A-2	—
	A-3	—
	A-4	—
	A-5	—
Oct.	A-1	p,p'-DDT (T)
	A-2	—
	A-3	—
	A-4	—
	A-5	—
Nov.	A-1	—
	A-2	—
	A-3	—
	A-4	—
	A-5	—
	C-2	—
	St. C-4	—
	St. C-5	—
	St. C-6	—

TABLE 2.—Organochlorine residues in water, Aransas Bay, Texas—1969-70—Continued

[T = Trace = <0.1 ppb; — = not detected]

COLLECTION DATE	SAMPLING LOCATION	RESIDUES DETECTED IN PPB (NG/G)
Dec.	A-1	—
	A-2	—
	A-3	—
	A-4	—
	A-5	—
	C-1	heptachlor (T)
	C-2	—
	St. C-4	—
	St. C-5	—
	St. C-6	—
1970		
Jan.	A-1	—
	A-2	—
	A-3	—
	A-4	—
	A-5	—
	C-1	—
	C-2	—
	St. C-4	—
	St. C-5	—
	St. C-6	—
Feb.	A-1	—
	A-2	—
	A-3	—
	A-4	—
	A-5	—
	C-1	—
	C-2	—
	C-3	—
	St. C-4	—
	St. C-5	—
	St. C-6	—
Mar.	A-1	—
	A-2	—
	A-3	—
	A-4	—
	A-5	—
	C-1	—
	C-2	—
	C-3	—
	St. C-4	—
	St. C-5	—
	St. C-6	—
Apr.	A-1	—
	A-2	—
	A-3	—
	A-4	—
	A-5	—
	C-1	—
	C-2	—
	St. C-4	—
	St. C-5	—
	St. C-6	—
May	A-1	—
	A-2	—
	A-3	—
	A-4	—
	A-5	—
	C-1	—
	C-2	—
	St. C-4	—
	St. C-5	—
	St. C-6	—
June	A-1	—
	A-2	—
	A-3	—
	A-4	—
	A-5	—
	C-1	—
	C-2	—
	St. C-4	—
	St. C-5	—
	St. C-6	—

WATER

The results from the analyses of water samples for organochlorine insecticides are shown in Table 2. Sensitivities were 0.05 ppb for all insecticides in water except DDT and its isomers; the sensitivity for DDT and its isomers was 0.1 ppm. Concentrations below 0.1 ppb could not be accurately quantified and were reported as trace (<0.1 ppb). Eighty water samples were analyzed, and 3 (3.8%) showed the presence of organo-

chlorine insecticide residues. Heptachlor, endrin, and *p,p'*-DDT were the only organochlorine residues detected in the water samples. No correlation could be made between the insecticide residues detected and the chemical properties of the area.

These amounts represent only the insecticides dissolved in the water and do not include the suspended load of the water, i.e., suspended plankton, detritus, and silt. Therefore, the data are reasonable considering the low solubility of organochlorine insecticides in water, the adsorptive properties of sediments, and the ability of organisms to concentrate insecticide residues in their tissues.

SEDIMENT

The results from the analyses of sediment samples for organochlorine insecticides are shown in Table 3. The limit of detection was 0.1 ppb. Organochlorine insecticides were detected in only 3 (10.3%) of the 29 samples analyzed.

No correlation could be made between insecticide residues detected in the sediment and the chemical properties of the area. There was no relationship between insecticides reported in water samples and insecticides detected in the sediment; in each instance the residues detected in the water samples differed in type from the residues detected in the sediments. Heptachlor, endrin, and *p,p'*-DDT were detected in the water, but only dieldrin and *p,p'*-DDD were found in the sediments.

TABLE 3.—Organochlorine residues in sediment, Aransas Bay, Texas—1969-70
(— = not detected)

COLLECTION DATE	SAMPLING LOCATION	RESIDUES DETECTED IN PPB (NG G)	
1969 Sept.	A-1	dieldrin (0.7)	
	A-2	—	
	A-3	dieldrin (0.5)	
	A-4	—	
	A-5	—	
Nov.	C-2	<i>p,p'</i> -DDD (24.6)	
	St. C-4	—	
	St. C-5	—	
	St. C-6	—	
1970 Jan.	A-1	—	
	A-2	—	
	A-3	—	
	A-4	—	
	A-5	—	
	C-1	—	
	C-2	—	
	St. C-4	—	
	St. C-5	—	
	St. C-6	—	
	May	A-1	—
		A-2	—
A-3		—	
A-4		—	
A-5		—	
June	C-1	—	
	C-3	—	
	St. C-4	—	
	St. C-5	—	
	St. C-6	—	

ORGANISMS

The results from the analyses of organisms for organochlorine insecticides are shown in Table 4. The limit of detection was 0.1 ppb. Dieldrin, *p,p'*-DDE, *p,p'*-DDD, and *p,p'*-DDT were the only insecticides present in any of the organisms. The concentrations ranged from a trace (<0.1 ppb) to 66.5 ppb. The predominant insecticide residues detected in this investigation coincided with those found for this area by the Gulf Breeze Laboratory, and concentrations were similar.

TABLE 4.—Organochlorine residues in organisms, Aransas Bay, Texas—1969-70
(T Trace = < 0.1 ppb; — = not detected)

COLLECTION DATE	TYPE OF SAMPLE	SAMPLING LOCATION	RESIDUES DETECTED IN PPB (NG G)
1969 July	2nd year, sea trout (muscle)	St. Chas. Bay	dieldrin (3.4) <i>p,p'</i> -DDE (25.5) <i>p,p'</i> -DDD (2.1) <i>p,p'</i> -DDT (5.5)
	2nd year, drum (whole)	St. Chas. Bay	dieldrin (9.7) <i>p,p'</i> -DDE (3.6) <i>p,p'</i> -DDT (2.1)
1970 Jan.	oysters	A-1	<i>p,p'</i> -DDE (5.9) <i>p,p'</i> -DDD (T)
Mar	blue crab (muscle)	Aransas Bay	<i>p,p'</i> -DDE (7.8) <i>p,p'</i> -DDD (T)
	blue crab (viscera)	Aransas Bay	—
	blue crab (eggs)	Aransas Bay	—
	silverside minnows	Aransas Bay	<i>p,p'</i> -DDE (33.3) <i>p,p'</i> -DDD (30.0) dieldrin (13.3)
	yellowtail croaker	Aransas Bay	<i>p,p'</i> -DDE (66.5) <i>p,p'</i> -DDD (60.0) <i>p,p'</i> -DDT (T)
	ribbon fish	Aransas Bay	<i>p,p'</i> -DDE (33.6) <i>p,p'</i> -DDD (41.0)
	brown shrimp	Aransas Bay	—
yellowtail croaker	Aransas Bay	<i>p,p'</i> -DDE (25.0) <i>p,p'</i> -DDD (T)	

Of particular interest is the lack of insecticide residues detected in shrimp and the low concentrations in the blue crabs and oysters. Butler (5) has postulated that the absence of insecticide residues in shrimp and other crustaceans is due to their high sensitivity to organochlorine insecticides, and that these animals are killed when the concentration of insecticides reaches a threshold value; this threshold may be low and thus, death may occur before these organisms have had time to metabolize the insecticide.

The low insecticide concentration in the oysters (*p,p'*-DDE, 5.9 ppb; *p,p'*-DDD, <0.1 ppb) is accounted for by the uncontaminated water and sediment at Station A-1. Although oysters can concentrate insecticides in their tissues when placed in contaminated water, they also have the ability, unlike many other organisms, to "flush" insecticide residues from their tissues when placed in uncontaminated water (4).

Butler (6) interpreted the presence of a high percentage of *p,p'*-DDT as indicating direct exposure to the insecticide and the presence of metabolites alone or at disproportionately high levels, as indicating that the residues have been transmitted through the food chain. A high percentage of *p,p'*-DDT was reported in the tissues of the sea trout and drum taken in July 1969. The first water samples analyzed (September 1969 and October 1969) indicated a trace of *p,p'*-DDT at Station A-1. No *p,p'*-DDT was found in any other water samples and only at one other time in any organism; this organism, the yellowtail croaker, (collected March 1970) had a trace of *p,p'*-DDT (<0.1 ppb) which constituted less than 0.1% of the total residues detected in the sample.

The small number of sediment and water samples in which organochlorine insecticides were present indicates that insecticide contamination of Aransas Bay is relatively low, and the presence of low organochlorine insecticide concentrations in the organisms may be attributed to transmission by the food chain.

Acknowledgment

Appreciation is expressed to the Texas Parks and Wildlife Department, Rockport, Tex., for their help in the collection of the samples and especially to Mr. T. I. Heffernan.

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See Appendix for chemical names of compounds discussed in this paper

Total Mercury in Largemouth Bass (*Micropterus salmoides*) in Ross Barnett Reservoir, Mississippi—1970 and 1971

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ABSTRACT

Total mercury in 73 largemouth bass, *Micropterus salmoides*, from Ross Barnett Reservoir, Mississippi, was measured by atomic absorption spectrophotometry. The fish analyzed were collected between November 1970 and October 1971 at intervals representing winter, spring, summer, and fall. Specimens ranged in weight from 0.10 to 3.15 kg. Fish contained from <0.05 to 0.74 ppm total mercury; levels generally increased with weight of the fish.

Introduction

Mercury compounds have been used for many years in agriculture, industry, and medicine, but only recently has there been much concern about the effects of mercurials on the aquatic environment. Previously, the assumption was generally accepted that metallic mercury settled to the bottom of a body of water and remained innocuous in the mud. However, research has shown that metallic mercury is converted through the action of microorganisms to methylmercury, and in this form is readily taken up and retained by living organisms (7). Most of this biological methylation of mercury is assumed to take place in the upper part of the bottom sediment (10). Eventually, by absorption and through the food chain, high levels of mercury may build up in carnivorous fishes such as the largemouth bass.

In 1970, concentrations of total mercury were found in fishes in Pickwick Lake, Mississippi, sufficient to warrant closing the lake to commercial fishing. With virtually little or no data available on the extent of contamination in Mississippi waters, a study was initiated to measure

total mercury in Ross Barnett Reservoir. Preliminary results on total mercury content in largemouth bass (*Micropterus salmoides*) from the reservoir are given in this paper.

Materials and Methods

Fish were collected between November 1970 and October 1971 at intervals representing winter, spring, summer, and fall. All collections were made from the State Highway 43 area of Ross Barnett Reservoir except the September - October 1971 collections which included specimens from several other points in the reservoir as well (Fig. 1).

A flameless atomic absorption method described by Hatch and Ott (8) and Lee, Noble, and Randall (11) was used to measure total mercury. Hatch and Ott (8) stated that this method is accurate for determination of mercury at levels as low as 0.001 ppm. Bache, Gutenmann, and Lisk (3) stated that it is sensitive to at least 0.1 ppm mercury in fish. In the analyses reported here the sensitivity limit was determined to be 0.05 ppm and recovery was 87%. The results reported are corrected for recovery and for values obtained in analyzing control blanks.

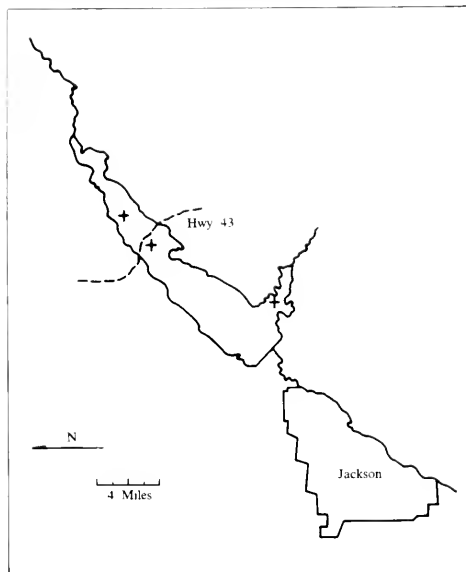
Samples of flesh taken from the trunk region of the fish below the dorsal fin and above the lateral line were mechanically chopped and mixed in a Waring Blendor: 2- to 3-g subsamples were dissolved in 250-ml flasks with 5 ml each of concentrated nitric and sulfuric acids and then diluted with 50 ml of distilled water. After cooling the sample to room temperature, a 5-ml aliquot of 10% stannous sulfate was added as a reducing agent and the flask placed in the absorption system. Once maximum absorbance was obtained, the mercury vapor

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was vented to the hood and the flask removed. Standards were run in the same manner. Foaming caused by aeration of samples was abated with Dow Corning Antifoam A. A Beckman Atomic Absorption Spectrophotometer System equipped with a 10-inch Potentiometric Recorder was used for these analyses. Standards were obtained from Beckman Instruments, Inc., Fullerton, Calif.

FIGURE 1.—Map of Ross Barnett Reservoir, Mississippi, showing locations (+) where fish were collected



Results and Discussion

Residues of mercury in muscle tissues and length and weight of the fish sampled are given in Table 1. Mercury concentrations ranged from less than 0.05 to 0.74 ppm. In Table 2 average levels of total mercury in fish sampled are given according to weight groups. In general, average mercury levels increased with weight; fish weighing 0.5 kg or less contained less than 0.12 ppm mercury, and those weighing over 3 kg averaged 0.45 ppm. A study using largemouth bass from Ross Barnett Reservoir (5) has shown that length and weight are indicative of age. Further, Bache *et al.* (3) using tagged lake trout (*Salvelinus namaycush*), demonstrated positive correlation between age and accumulation of total mercury.

During the 12-month period in which fish were collected, temperatures ranged from about 8.0° to 29.0° C. Table 3 shows mercury residues in fish as a function of weight for each season of collection. All winter samples weighed less than 2 kg and with the exception of one sample containing 0.62 ppm generally had low levels of mercurial residues. All spring samples weighed 2 kg or less with the exception of two fish; residue levels in the smaller fish ranged from 0.10 to 0.48 ppm; the two larger fish contained 0.36 and 0.74 ppm. Samples collected in the summer showed less variation in the amount of mercury (range, 0.10 - 0.21 ppm); the two specimens weighing over 2 kg each contained 0.17 ppm. The fall collection contained a greater proportion of larger fish than collections from other seasons, and residues increased with weight more noticeably.

TABLE 1.—Concentrations of total mercury (Hg) in largemouth bass by sample collection date—Ross Barnett Reservoir, 1970-71

LENGTH (CM)	WEIGHT (KG)	SEX	TOTAL MERCURY RESIDUE (PPM)
NOVEMBER 18—DECEMBER 30, 1970 (WINTER SAMPLES)			
22.23	0.14	—	<0.05
22.23	0.15	M	<0.05
22.23	0.15	—	<0.05
22.86	0.16	F	<0.05
22.86	0.17	—	<0.05
22.86	0.17	—	<0.05
22.86	0.17	F	<0.05
23.50	0.17	F	0.06
21.59	0.20	—	0.06
27.94	0.33	M	0.05
27.94	0.35	M	0.07
32.39	0.45	F	0.05
32.39	0.46	F	0.06
33.02	0.53	M	0.16
35.56	0.61	F	<0.05
37.47	0.82	F	<0.05
40.64	0.96	M	0.19
43.82	1.02	M	0.19
45.09	1.13	F	0.62
43.18	1.13	F	0.25
43.18	1.18	F	0.16
41.28	1.20	M	0.07
46.99	1.23	F	0.21
45.72	1.25	F	0.25
43.82	1.37	F	0.05
50.80	1.77	F	0.30
45.72	1.81	F	0.18
MARCH 29, 1971 (SPRING SAMPLES)			
20.32	0.13	F	0.10
22.23	0.15	M	0.38
23.50	0.24	F	0.36
25.40	0.24	M	0.10
26.04	0.29	F	0.24
30.48	0.36	F	0.10
30.48	0.39	M	0.23
30.48	0.46	M	0.17
32.00	0.48	M	0.18
32.39	0.48	M	0.11
30.48	0.52	M	0.24
33.02	0.59	F	0.27
37.08	0.70	M	0.14
38.10	0.70	M	0.20
37.08	0.74	M	0.48
38.10	0.79	M	0.27
46.99	1.35	F	0.27
57.79	2.61	F	0.36
58.42	3.15	F	0.74

TABLE 1.—Concentrations of total mercury (Hg) in largemouth bass by sample collection date—Ross Barnett Reservoir, 1970-71—Continued

LENGTH (CM)	WEIGHT (KG)	SEX	TOTAL MERCURY RESIDUE (PPM)
JUNE 28—JULY 1, 1971 (SUMMER SAMPLES)			
19.69	0.10	F	0.11
20.32	0.12	F	0.13
21.16	0.14	M	0.12
—	0.14	F	0.10
25.40	0.19	M	0.12
29.85	0.40	F	0.14
31.75	0.42	F	0.15
33.02	0.50	M	0.16
38.10	0.68	F	0.16
39.12	0.72	M	0.14
39.37	0.74	F	0.13
41.91	1.04	F	0.16
46.36	1.37	F	0.21
59.06	2.66	F	0.17
54.61	3.09	F	0.17
SEPTEMBER 30 AND OCTOBER 1, 1971 (FALL SAMPLES)			
31.12	0.40	M	0.05
36.83	0.77	M	0.05
41.91	1.02	F	0.10
45.09	1.29	F	0.14
48.90	1.55	F	0.18
48.26	1.86	F	0.27
54.61	2.34	F	0.36
57.15	2.76	F	0.33
60.69	3.10	F	0.44
45.72	1.40	F	0.08
50.17	2.13	F	0.18
52.07	2.56	F	0.13
Total Number of Samples = 73			

TABLE 2.—Average concentrations of total mercury in Ross Barnett Reservoir largemouth bass according to weight groups

WEIGHT GROUP (KG)	AVERAGE MERCURY CONCENTRATION IN PPM	NUMBER OF FISH
0.0-0.50	0.12	31
0.51-1.00	0.18	15
1.01-1.50	0.20	14
1.51-2.00	0.23	4
2.01-2.50	0.27	2
2.51-3.00	0.25	4
3.01-3.15	0.45	3
Total		73

NOTE: In calculating average concentrations, those less than 0.05 ppm were computed at 0.05 ppm.

The ability of aquatic organisms to concentrate mercury above the level found in their environment is well known (1,9). However, the mechanism by which fish concentrate mercury is not fully understood. Rucker and Amend (12) found that mercury levels in muscle tissues from lake trout treated 1 hour per week for 12 weeks with 6.25% ethylmercury phosphate reached maximum concentrations of 4.4 ppm and returned to normal approximately 17 weeks following cessation of treatment. Studies have shown that fish absorb mercury compounds directly through their gills and through ingestion of a contaminated food (7,12).

TABLE 3.—Total mercury in fish by weight of fish sampled

TOTAL MERCURY (PPM)	NUMBER OF FISH PER WEIGHT GROUP			
	< 1-1 KG	1-2 KG	> 2-3 KG	> 3-4 KG
NOV. 18 & DEC. 30, 1970 (WINTER SAMPLES)				
0.05-0.1	15	2		
< 0.1-0.2	2	3		
< 0.2-0.3		4		
< 0.3-0.4				
< 0.4-0.5				
< 0.5-0.6				
< 0.6-0.7		1		
< 0.7-0.8				
MARCH 29, 1971 (SPRING SAMPLES)				
0.05-0.1	3			
< 0.1-0.2	5			
< 0.2-0.3	8	1		
< 0.3-0.4	2		1	
< 0.4-0.5	1			
< 0.5-0.6				
< 0.6-0.7				
< 0.7-0.8				1
JUNE 28 & JULY 1, 1971 (SUMMER SAMPLES)				
0.05-0.1	1			
< 0.1-0.2	10	1	1	1
< 0.2-0.3		1		
< 0.3-0.4				
< 0.4-0.5				
< 0.5-0.6				
< 0.6-0.7				
< 0.7-0.8				
SEPT. 30 & OCT. 1, 1971 (FALL SAMPLES)				
0.05-0.1	2	2		
< 0.1-0.2		2	2	
< 0.2-0.3		1		
< 0.3-0.4			2	
< 0.4-0.5				
< 0.5-0.6				1
< 0.6-0.7				
< 0.7-0.8				

Feeding activities and levels of mercury in food chain organisms no doubt influence total mercury accumulation in largemouth bass. Dubets (6) and Schneidermeyer and Lewis (13) concluded that gizzard shad, when available, is the principal food of largemouth bass, and Applegate and Mullan (2) found that largemouth bass, after reaching 40 mm length, subsist principally on small shad which feed extensively on plankton and other small aquatic organisms. Barkley (4,5, and personal communication) found that on a percent by weight basis, shad comprised approximately 51.9% of the total fish population of Barnett Reservoir during the period from 1963 to 1970.

The sources of mercury available to the organisms in Ross Barnett Reservoir appear to be naturally occurring mercurials, items containing mercury which have been disposed of by the consumer public, and agricultural products; there are few industrial sources of mercury pollution in the drainage area of the Reservoir.

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A Survey of the Selenium Content of Fish From 49 New York State Waters

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ABSTRACT

A survey was made of the selenium content of 438 fish of various species collected in 1969 from 49 New York State waters and a group of lake trout sampled in 1970 from Cayuga Lake only. Concentrations of selenium on a fresh-weight basis were usually below 1 ppm. There was little apparent correlation between selenium concentrations and species or sampling locations except that sturgeon from the Hudson River, lake trout from Lakes George and West Canada, whitefish from Raquette Lake, and several species from Lake Pleasant had consistently higher levels of selenium than other samples; all fish from Lakes Butterfield and Champlain and the Chenango and Salmon Rivers had consistently lower levels. No correlation was apparent between selenium levels and size or sex of fish. Selenium did not appear to be cumulative in lake trout of known age up to 12 years from Cayuga Lake.

Introduction

The contamination of fish by heavy metals and other toxic elements is presently of major concern. Selenium, although essential to man and animals in trace amounts, is very toxic at slightly higher levels. It is widely distributed in the lithosphere at a level of about 0.09 ppm (5) and is largely concentrated in sulfide materials. A major source of selenium as an air pollutant is the combustion of coal and oil, which have been found to contain concentrations up to 5 ppm (6) and 1.4 ppm (11), respectively. Selenium is also present as an impurity in fertilizers (13).

Although data on the levels of selenium in fish and aquatic organisms are scant, levels in zooplankton in Lake Michigan have been reported up to about .7 ppm (4); and another study has shown relatively higher levels, 1 to 2 ppm, in sea foods (10). The death of stocked game fish in a Colorado reservoir has been associated repeatedly with high levels of selenium in bottom sediments (3).

The present study was undertaken to determine the selenium concentrations in freshwater fish from 49 waters in New York State. A total of 438 fish were collected in 1969 by the New York State Conservation Department. In addition, a series of lake trout of increasing age up to 12 years were collected in an effort to determine if selenium was cumulative in these fish. Most of the samples, including the lake trout, were also analyzed for lead, and results of these analyses are reported in a previous issue of this Journal (7).

Sampling and Analytical Methods

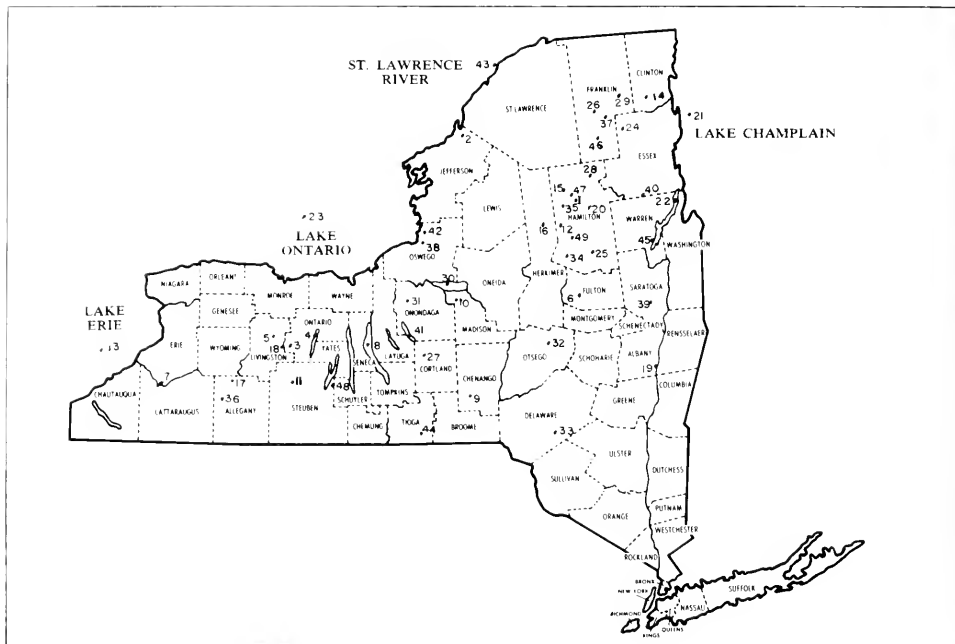
The fish were netted, and species, sex, length, and weight recorded. All fish were decapitated and eviscerated. The remainder was chopped in a food chopper, mixed, and frozen in polyethylene bags prior to analysis. Selenium was determined by an adaptation of the method of Allaway and Cary (1) involving oxygen-flask combustion of 1 g of dried fish (2) and determination of the fluorescence of the selenium-2,3-diaminonaphthalene complex. Isolation of selenium by co-precipitation with arsenic (1) was found unnecessary and was thus omitted. The percent recoveries of selenium added to fish are given in Table I. The method was sensitive to about 0.1 ppm of selenium in fish.

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FIGURE 1.—New York State map showing approximate locations of waters where fish were collected



Results and Discussion

Table 2 lists the fish samples by their common and scientific names, and Fig. 1 designates numerically the approximate locations of the waters where fish were collected. The results of analysis of the fish for selenium, given in ppm on a fresh-weight basis and not corrected for percent recovery, are presented in Table 3. The fish species, sex, length, and weight are included to the extent that recording was complete.

Some general observations can be made concerning the selenium concentrations found (Table 3). All fish contained levels below 1 ppm, with the exception of four samples. The maximum concentration found was 1.50 ppm in a lake trout of unrecorded sex and size from Lake George. Levels were consistently higher in sturgeon from the Hudson River, lake trout from Lakes George and West Canada, whitefish from Raquette Lake, and various species of fish from Lake Pleasant. However, levels of selenium in fish from Lakes Erie and Ontario, generally considered quite polluted, were not notably high. Levels were consistently lower in fish from Lakes Butterfield and Champlain and the Chenango and Salmon Rivers. Selenium concentrations did not appear

to be related to the size, sex, or species of fish, and, with the exception of the observations noted above, levels did not appear to be related to location of waters from which fish were collected. Other observations might have been apparent had more fish from specific waters been analyzed.

Virtually no data are available on the soil and mineral content of selenium in New York and, therefore, attempts to correlate selenium concentrations in fish with natural background levels were unsuccessful.

Table 4 lists the selenium concentrations in lake trout, of increasing age up to 12 years, from Cayuga Lake. The ages were accurately known since fingerling lake trout are stocked there annually and marked with the date of stocking. Based on these limited data, selenium did not seem to be cumulative in this species.

The levels of selenium found in fish in this study are in the same range as those present in many other common food classes (10) and are not considered significant. In this regard, it is interesting to note that selenium has been reported to be markedly protective against the toxic effects of mercury and cadmium (8,9,12).

Acknowledgment

The authors thank W. H. Allaway and E. E. Cary for advice on the analysis of selenium and for supplying analytical reagents. The authors also thank W. D. Youngs of the Conservation Department, Cornell University, for collecting the Cayuga Lake trout of various ages.

TABLE 1.—Percent recovery of selenium from fish

SPECIES	SELENIUM ADDED (PPM)	PERCENT RECOVERY
Brown catfish	0.2	105.85
	0.5	68.98
	1.0	84.94
Lake trout	0.5	100
	1.0	97
Largemouth bass	0.2	75.60
	0.5	86.88
	1.0	85.69
Northern pike	0.2	80.90
	0.5	80.68
	1.0	89.59

NOTE: Sensitivity level = 0.1 ppm

TABLE 2.—Common and scientific names of fish analyzed in this study

COMMON NAME	SCIENTIFIC NAME
Black crappie	<i>Pomoxis macromaculatus</i>
Bowfin	<i>Amia calva</i>
Brook trout (Speckled trout)	<i>Salvelinus fontinalis</i>
Brown trout	<i>Salmo trutta</i>
Brown catfish (bullhead) and Channel catfish	<i>Ictalurus sp.</i>
Burbot	<i>Lota lota</i>
Carp	<i>Cyprinus carpio</i>
Chain pickerel	<i>Esox niger</i>
Cisco	<i>Coregonus artedii</i>
Coho salmon	<i>Oncorhynchus kisutch</i>
Freshwater drum	<i>Aplodinotus grunniens</i>
Gizzard shad	<i>Dorosoma cepedianum</i>
Goldfish	<i>Carassius auratus</i>
Lake trout	<i>Salvelinus namaycush</i>
Lake whitefish	<i>Coregonus clupeaformis</i>
Largemouth bass	<i>Micropterus salmoides</i>
Muskellunge	<i>Esox masquinongy</i>
Northern pike	<i>Esox lucius</i>
Perch	<i>Perca sp.</i>
Rainbow trout	<i>Salmo gairdneri</i>
Rock bass	<i>Ambloplites rupestris</i>
Smallmouth bass	<i>Micropterus dolomieu</i>
Splake (Brook and Lake trout cross)	
Striped bass	<i>Morone saxatilis</i>
Sturgeon, Atlantic sturgeon, and Shortnose sturgeon	<i>Acipenser sp.</i>
Walleye pike	<i>Stizostedion vitreum vitreum</i>
White bass (Silver bass)	<i>Roccus chrysops</i>
White sucker (Sucker)	<i>Catostomus commersoni</i>

TABLE 3.—Residues of total selenium in fish from New York State waters in 1969

SPECIES	TAG NO.	SEX	LENGTH (CM)	WEIGHT (KG.)	SELENIUM RESIDUE LEVEL (PPM)
BLUE MOUNTAIN LAKE—NO. 1					
Bullhead catfish	6R6527	F	28.4	.379	0.26
	6R6528	M	28.9	.377	0.48
	6R6529	F	27.9	.378	0.38
Rainbow trout	6R6522	F	33.0	.393	0.65
	6R6521	M	33.8	.173	0.57
Smallmouth bass	6R6526	M	30.5	.337	0.64
	6R6524	M	28.2	.275	0.50
BUITERFIELD LAKE—NO. 2					
Bowfin	22BL-4	M	55.9	1.702	0.15
Bullhead catfish	24BL-4	F	32.2	.669	0.10
	23BL-4	F	33.0	.685	0.11
	25BL-4	F	32.0	.680	0.10
Largemouth bass	19BL-4	F	34.8	.657	0.21
	21BL-4	M	34.5	.626	0.18
	20BL-4	F	33.0	.573	0.11
Northern pike	9BL-4	—	—	—	0.15
	8BL-4	—	—	—	0.15
	10BL-4	—	—	—	0.21
	7BL-4	—	—	—	0.15
Rock bass	14BL-4	M	22.9	.263	0.22
	15BL-4	M	22.2	.277	0.15
Smallmouth bass	11BL-4	—	—	—	0.22
	12BL-4	—	—	—	0.21
Sucker	25BL-4	M	38.6	.916	0.18
Walleye pike	26BL-4	M	55.9	1.753	0.17
	29BL-4	M	47.3	1.071	0.20
	27BL-4	M	48.7	1.268	0.19
CANADIAN LAKE—NO. 3					
Bullhead catfish	J6220	—	35.0	.574	0.12
Lake trout	J6293	M	47.8	1.246	0.42
	J6295	M	76.8	4.855	0.44
Chain pickerel	J6228	F	46.0	.699	0.32
	J6222	F	40.7	.471	0.24
Rainbow trout	J6225	—	29.2	.284	0.36
	J6296	M	30.5	.370	0.31
	J6297	M	32.5	.430	0.27
Rock bass	J6212	M	21.8	.248	0.39
Smallmouth bass	J6210	M	36.1	.751	0.41
	J6211	M	28.4	.386	0.53
	J6291	M	29.2	.469	0.50
	J6290	F	33.6	.603	0.48
	J6292	F	29.2	.378	0.37
CANANDAIGUA LAKE—NO. 4					
Brown trout	J6288	M	56.7	2.595	0.39
Lake trout	J6287	M	55.2	1.821	0.36
	J6286	M	52.6	1.545	0.41
	J6231	M Imm.	44.4	.722	0.55
Largemouth bass	J6167	M	28.7	.370	0.18
Smallmouth bass	J6162	F	28.7	.359	0.63
CONESUS LAKE—NO. 5					
Black crappie	J6102	—	—	—	0.20
CAROGA LAKE—NO. 6					
Smallmouth bass	6R6192	F	40.9	1.295	0.42

TABLE 3.—Residues of total selenium in fish from New York State waters in 1969—Continued

SPECIES ¹	TAG NO.	SEX	LENGTH (CM)	WEIGHT (KG)	SELENIUM RESIDUE LEVEL (PPM)	
CATARAUGUS CREEK—NO. 7 ²						
Coho salmon	2F1201	M	41.6	.683	0.40	
	2F1202	M	40.2	.588	0.28	
	2F1203	M	37.9	.588	0.45	
	2F1204	F	54.4	1.77	0.26	
	2F1205	F	56.8	2.09	0.32	
	2F1206	F	61.0	2.19	0.24	
	2F1207	F	60.2	2.23	0.24	
	2F1208	F	63.0	3.04	0.27	
	2F1209	F	64.0	3.00	0.25	
CAYUGA LAKE—NO. 8 ²						
Largemouth bass	3-CL-2	—	15.25	—	0.25	
	3-CL-1	M	31.7	.621	0.29	
Chain pickerel	CAY-1	—	—	—	0.23	
	CAY-2	—	—	—	0.22	
	CAY-3	—	—	—	0.34	
CHENANGO RIVER—NO. 9 ²						
Smallmouth bass	3-CHR-1	F	22.2	.168	0.28	
	3-CHR-3	F	27.9	.343	0.24	
	3-CHR-2	Imm.	24.8	.198	0.23	
	3-CHR-4	M	28.6	.295	0.27	
	3-CbR-Ch.8	F	30.5	.436	0.21	
	3-CHR-5	M	34.9	.550	0.23	
CHITTENANGO CREEK—NO. 10 ²						
Brown trout	3-CH-1	F	43.2	1.072	0.26	
	3-CH-2	F	31.7	.447	0.24	
	3-CH-3	F	23.4	.151	0.27	
COHOCTON RIVER—NO. 11 ²						
Brown trout	2CohR1	M	27.6	.256	0.40	
	2CohR2	F	26.7	.251	0.32	
	2CohR3	M	25.4	.194	0.33	
EIGHTH LAKE—NO. 12 ²						
Rainbow trout	6R6493	—	—	—	0.41	
LAKE ERIE—NO. 13 ²						
Burbot	2-LAER-6	—	58.4	1.980	0.33	
Coho salmon	2-LE-Hg-7	F	41.3	.665	0.37	
	2-LE-Hg-6	F	44.5	.764	0.33	
	LE-CS	—	—	—	—	
(Sunset Bay)	1-9-4-69	M	56.2	2.110	0.35	
	2-LE-Hg-8	M	48.3	1.147	0.45	
	1E-CS-9	F	57.5	2.671	0.35	
Do.	1E-CS-8	F	55.0	2.392	0.34	
Do.	1E-CS-6	M-Imm.	35.0	.493	0.36	
Do.	1E-CS-7	F	56.8	2.520	0.31	
Freshwater drum	LE-FWD-1	M	45.0	1.005	0.81	
	2-LE-Hg-3	F	40.0	.733	0.45	
	2-LE-Hg-42	F	36.8	.670	0.42	
	2-LE-Hg-37	M	36.2	.650	0.35	
	2-LE-Hg-47	F	48.3	1.385	0.28	
	2-LE-Hg-38	M	37.5	.715	0.48	
	2-LE-Hg-45	M	34.3	.570	0.62	
	2-LE-Hg-43	M	35.6	.610	0.43	
	2-LE-Hg-41	F	37.5	.750	0.59	
	2-LE-Hg-46	M	36.2	.565	0.42	
	2-LE-Hg-40	F	32.4	.415	0.38	
	2-LE-Hg-44	M	36.8	.610	0.41	
	2-LE-Hg-39	—	—	—	0.42	
	2-LE-Hg-48	M	48.9	1.395	0.24	
	2-LE-1	—	—	43.2	.695	0.29
	2-LE-3	F	30.7	.270	0.43	
	2-LE-Hg-2	F	36.2	.612	0.32	
	2-LE-2	F	50.8	1.750	0.37	
	2-LE-Hg-1	M	38.1	.615	0.36	
LE-FWD-2	M	29.2	.364	0.54		

TABLE 3.—Residues of total selenium in fish from New York State waters in 1969—Continued

SPECIES ¹	TAG NO.	SEX	LENGTH (CM)	WEIGHT (KG)	SELENIUM RESIDUE LEVEL (PPM)	
LAKE ERIE—NO. 13 ² —Continued						
Gizzard shad	2-LE-4	—	Composite of 31 fish		0.42	
Lake trout	2-LEI-2	—	—	—	0.33	
Rock bass	2-LE-Hg-31	M	27.3	.358	0.31	
	2-LE-Hg-51	F	37.5	.680	0.52	
Silver bass	2-LE-Hg-68	F	36.2	.487	0.37	
	2-LE-Hg-71	F	35.6	.487	0.43	
	2-LE-Hg-53	F	36.2	.615	0.32	
	2-LE-Hg-70	M	34.9	.503	0.46	
	2-LE-Hg-52	F	36.2	.625	0.35	
	2-LE-Hg-72	F	40.0	.840	0.41	
	2-LE-Hg-69	M	34.9	.425	0.51	
	Smallmouth bass	LE-SMB-1	M	43.5	.932	0.40
		2-LE-Hg-54	F	40.7	.860	0.30
2-LE-Hg		—	—	—	0.31	
2-LE-Hg-50		F	34.9	.645	0.29	
2-LE-Hg-60		F	39.4	.649	0.57	
2-LE-Hg-62		F	45.7	1.300	0.43	
2-LE-Hg-58		M	33.0	.360	0.37	
Sucker	2-LE-Hg-56	F	35.6	.682	0.38	
	2-LE-Hg-59	F	37.5	.741	0.54	
	2-LE-Hg-57	M	40.0	.695	0.26	
	2-LAER-5	—	50.8	1.474	0.59	
	2-LE-Hg-13	F	51.5	1.530	0.47	
	2-LAER-4	M	30.5	.334	0.34	
	2-LE-Hg-12	F	48.9	1.296	0.37	
Walleye pike	2-LE-Hg-65	F	44.5	1.150	0.51	
	2-LE-Hg-63	M	46.4	1.190	0.71	
	2-LAER-3	M	48.3	1.097	0.21	
	2-LE-Hg-11	M	45.7	1.040	0.36	
	2-LE-Hg-64	F	46.4	1.260	0.44	
	2-LE-Hg-67	F	43.2	1.120	0.40	
	2-LE-Hg-66	F	49.5	1.150	0.33	
	2-LE-Hg-23	F	69.3	4.210	0.30	
	2-LE-Hg-25	F	64.7	2.920	0.20	
	2-LE-Hg-32	F	63.5	3.375	0.25	
Yellow perch	2-LE-Hg-4	F	63.5	2.310	0.37	
	2-LE-Hg-33	M	73.7	5.070	0.40	
	2-LE-Hg-35	F	67.4	3.800	0.23	
	2-LE-Hg-36	F	62.3	3.380	0.33	
	2-LE-Hg-21	F	68.5	4.219	0.25	
	2-LE-Hg-16	F	31.7	.515	0.31	
	2-LE-Hg-29	F	33.0	.609	0.37	
	2-LE-Hg-30	F	27.9	.485	0.27	
	2-LE-Hg-17	F	31.1	.516	0.35	
	2-LE-Hg-28	F	34.3	.697	0.28	
FERN LAKE—NO. 14 ²	2-LE-Hg-26	F	32.4	.612	0.27	
	2-LE-Hg-20	F	33.3	.605	0.32	
	2-LE-Hg-19	F	36.2	.735	0.35	
	2-LE-Hg-18	F	34.9	.694	0.34	
	2-LE-Hg-27	F	30.5	.450	0.36	
	Rock bass	5-FLK-3	M	26.2	.426	0.25
	FORKED LAKE—NO. 15 ²					
	Brook trout	6R6641	—	43.7	.945	0.40
	Lake trout	6R6642	M	53.3	.854	0.33
	Smallmouth bass	6R6648	F	37.1	.950	0.39
Speckled trout	6R6640	F	35.6	.645	0.57	
FOURTH LAKE—NO. 16 ²						
Lake trout	4-4L-12	F	55.9	1.872	0.39	
	4-4L-10	M	37.8	.497	0.37	
	4-4L-11	—	—	—	0.33	
Sucker	4-4L-23	M	29.2	.301	0.27	
	4-4L-22	F	30.2	.336	0.40	
	4-4L-24	M	35.6	.554	0.72	

TABLE 3.—Residues of total selenium in fish from New York State waters in 1969—Continued

SPECIES ¹	TAG NO.	SEX	LENGTH (CM)	WEIGHT (KG)	SELENIUM RESIDUE LEVEL (PPM)
GENESSEE RIVER—NO. 17 ²					
Smallmouth bass	J-6267	—	—	—	0.35
HEMLOCK LAKE—NO. 18 ²					
Bullhead catfish	J6203	M	36.1	.739	0.1
	J6202	—	28.2	.341	0.18
Chain pickerel	J6198	—	31.7	.229	0.44
	J6199	M	45.7	.754	0.56
	J6197	—	50.8	1.245	0.38
Lake trout	J6299	M	67.5	3.987	0.42
	J6298	F	77.8	4.270	0.39
	J6300	M	75.5	4.580	0.47
Largemouth bass	J6229	M	26.2	.344	0.19
	J6201	—	28.4	.468	0.42
Rainbow trout	J6200	M	38.1	.688	0.45
HUDSON RIVER—NO. 19 ²					
Atlantic sturgeon	J2067	♀—Imm.	46.0	.410	0.69
	J2065	♀ F—Imm.	62.0	1.285	0.84
	J2064	M	98.0	5.076	0.80
	J2066	♂ M—Imm.	64.0	1.075	0.26
	J2068	♂—Imm.	64.5	1.348	0.88
Goldfish	6R6667	—	—	—	0.18
	6R6666	—	—	—	0.17
	6R6665	—	—	—	0.14
Shortnose sturgeon	J2076	♂—Imm.	47.3	.636	0.37
Striped bass	J2070	M	57.6	2.256	0.25
	J2071	M	53.4	2.042	0.37
	J2072	F	41.1	.780	0.24
	J2069	M	54.4	2.072	0.65
	8H4UDR2	M	40.6	.800	0.38
	8H4UDR3	M	43.7	.985	0.30
	8H4UDR1	M	40.2	.850	0.45
INDIAN LAKE—NO. 20 ²					
Northern pike	6R6554	M	73.0	2.735	0.27
	6R6142	M	64.3	1.433	0.35
Walleye pike	6R6553	M	70.6	2.562	0.27
	6R6144	M	29.7	.380	0.33
Whitefish	6R6139	F	38.6	.595	0.43
	6R6140	M	43.2	.789	0.32
	6R6141	M	45.3	.641	0.71
	6R6143	M	46.0	.873	0.24
LAKE CHAMPLAIN—NO. 21 ²					
Channel catfish	5-LCCC-1	F	53.4	1.630	0.14
	5-LCCC-2	M	76.1	6.200	0.11
	5-LCCC-3	—	61.0	2.630	0.12
	6R6592	F	52.0	1.700	0.12
(South Bay)	6R6591	M	47.8	1.130	0.15
Do.	6R6593	M	60.5	2.760	0.14
Freshwater drum	6R6588	F	31.7	.306	0.24
(South Bay)	6R6590	F	48.8	1.165	0.11
Do.	6R6589	M	36.8	.423	0.22
Walleye pike	6R6587	M	64.8	3.070	0.24
(South Bay)	6R6585	F	40.2	.566	0.23
Do.	6R6586	M	54.9	1.455	0.26
LAKE GEORGE—NO. 22 ²					
Black crappie	6R6676	F	20.3	.109	0.38
	6R6675	F	31.2	.584	0.35
	6R6452	F	28.7	.352	0.47

TABLE 3.—Residues of total selenium in fish from New York State waters in 1969—Continued

SPECIES ¹	TAG NO.	SEX	LENGTH (CM)	WEIGHT (KG)	SELENIUM RESIDUE LEVEL (PPM)
LAKE GEORGE—NO. 22 ² —Continued					
Brown bullhead	6R6680	F	32.8	.686	0.28
	6R6454	F	32.8	.508	0.29
Chain pickerel	6R6450	M	35.6	.258	0.30
Lake trout	6R6672	F	71.0	3.570	0.46
	6R573	—	—	—	1.50
	6R6674	M	68.8	3.125	0.78
	6R6673	F	70.1	3.241	0.68
Largemouth bass	6R6441	M	41.1	.973	0.51
	6R6453	F	36.3	.743	0.40
	6R6440	M	40.4	.789	0.57
Northern pike	6R6677	F	64.0	1.820	0.38
	6R6671	F	59.5	1.780	0.41
	6R6404	F	75.0	3.180	0.44
	6R6407	F	64.7	2.950	0.35
	6R6406	M	60.5	3.000	0.45
Rainbow trout	6R6681	M	55.3	2.440	0.55
	6R6682	M	50.8	1.690	0.53
	6R639	F	41.9	.692	0.52
	6R6425	F	58.6	2.410	0.63
Smallmouth bass	6R6449	F	42.4	1.235	0.49
White sucker	6R6433	F	53.0	1.139	0.59
	6R6434	M	46.3	.872	0.60
	6R6432	M	47.3	.906	0.48
LAKE ONTARIO—NO. 23 ²					
Black crappie	2-LO-4	—	—	—	0.26
Carp	J6067	—	27.4	.419	0.21
Coho salmon	69-LO-CS1	F	57.7	2.555	0.34
	20-LO-4	M	58.5	2.640	0.30
	21-LO-4	F	48.3	.965	0.29
	22-LO-4	M	50.0	1.400	0.30
	23-LO-4	F	55.9	2.180	0.39
	73-30-69	F	51.3	2.060	0.39
(False Duck Is.)	73-29-69	F	55.8	2.040	0.38
(Outlet Beach)	73-49-69	F	63.0	3.110	0.43
(Amherst Bar)	73-18-69	M	60.1	3.190	0.47
(Pt. Petre)	73-27-69	F	56.1	2.090	0.31
(Perch Cove)	73-23-69	M	60.4	2.090	0.50
(Shelter Valley Mouth)	73-47-69	F	64.0	2.890	0.44
(Pennicott Reef)	73-16-69	F	56.8	2.460	0.37
Do.	73-13-69	M	50.3	1.740	0.29
	73-13-69	M	—	—	0.25
	(testes)	M	—	—	—
Do.	73-17-69	M	64.8	3.780	0.38
(Wellington Beach)	73-51-69	M	60.5	2.120	0.46
Do.	73-44-69	M	64.8	3.010	0.40
Do.	73-53-69	M	59.9	2.100	0.68
Do.	73-52-69	F	66.0	3.290	0.44
Do.	73-48-69	F	64.8	3.010	0.47
Do.	73-50-69	M	54.0	1.620	0.49
Rock bass	J6038	F	22.8	.268	0.31
	J6097	F	24.1	.288	0.35
	J6098	M	25.5	.436	0.31
	J6085	F	22.8	.301	0.31
Smallmouth bass	J-6165	—	—	—	0.28
	9-LO-4	F	37.6	.826	0.43
	16-LO-4	M	40.7	1.014	0.35
(Carleton Is.)	4C1-1	M	28.9	.320	0.37
Do.	4C1-2	M	27.4	.258	0.39
Do.	4C1-3	M	34.0	.573	0.38

TABLE 3.—Residues of total selenium in fish from New York State waters in 1969—Continued

SPECIES ¹	TAG NO.	SEX	LENGTH (CM)	WEIGHT (KG)	SELENIUM RESIDUE LEVEL (PPM)
LAKE ONTARIO—NO. 23 ² —Continued					
Smallmouth bass (Cont'd)					
(Pt. Peninsula)	4RA2637	F	37.4	.855	0.30
Do.	4RA2635	F	33.0	.558	0.37
Do.	4RA2632	—	—	—	0.32
Do.	4RA2638	F	33.6	.644	0.37
Do.	4RA2646	F	30.5	.533	0.38
Do.	4RA2644	M	38.1	.892	0.40
Do.	4RA2650	M	27.9	.320	0.49
Splake	17-LO-4	♂—Imm.	34.8	.523	0.48
Sucker	15-LO-4	M	39.1	.657	0.24
	14-LO-4	F	39.1	.703	0.20
White bass	J6027	F	25.5	.272	0.24
	J6030	—	24.9	.207	0.50
LAKE PLACID—NO. 24 ²					
Brook trout	LP-NE-2	F	30.5	.360	1.10
Lake trout (North End)	5-NELP-2	—	41.5	.726	0.77
	5-NELP-1	M	31.7	.299	0.80
Northern pike	3LP-5	M	76.6	3.870	0.40
	1LP-5	M	57.4	1.408	0.34
	2LP-5	M	64.0	1.642	0.34
Rainbow trout	5-LKP-1	F	25.4	.225	0.29
	5-LKP-3	F	40.9	.715	0.51
	5-LKP-2	F	42.1	.779	0.55
Smallmouth bass	5-LKP-4	M	29.2	.336	0.76
	5-LKP-5	F	46.3	1.420	0.45
White sucker	5-LP-WS-1	—	43.7	1.440	0.37
	5-LP-WS-3	—	30.5	.417	0.65
Yellow perch	5-LP-5	F	31.7	.435	0.77
LAKE PLEASANT—NO. 25 ²					
Brown bullhead	6R6118	M	34.3	.681	0.38
	6R6119	F	34.8	.673	0.31
Bullhead catfish	6R6120	M	25.5	.309	0.24
Chain pickerel	6R6126	F	35.0	.255	0.40
	6R6127	F	38.4	.392	0.29
	6R6128	F	39.2	.405	0.25
Largemouth bass	6R6131	F	35.0	.622	0.24
Rainbow trout	6R6129	F	42.7	1.068	1.05
Rock bass	6R6124	F	26.4	.461	0.98
	6R6123	F	25.4	.325	0.78
Smallmouth bass	6R6125	—	—	—	0.84
	6R6132	F	32.7	.484	0.40
	6R6130	F	49.3	1.775	0.74
Whitefish	6R6117	M	53.8	2.153	0.62
	6R6116	—	61.0	2.035	0.82
Yellow perch	6R6135	M	30.0	.438	0.86
	6R6133	M	27.4	.335	0.92
LITTLE GREEN POND—NO. 26 ²					
Rainbow trout	5-lgp-8	F	34.1	.392	0.67
	5-lgp-7	M	32.0	.370	0.37
	5-lgp-9	F	38.6	.608	0.44
Whitefish	5-lgp-1	F	53.4	1.805	0.97
	5-lgp-2	F	57.1	2.041	0.74
	5-lgp-3	M	50.8	2.050	0.49

TABLE 3.—Residues of total selenium in fish from New York State waters in 1969—Continued

SPECIES ¹	TAG NO.	SEX	LENGTH (CM)	WEIGHT (KG)	SELENIUM RESIDUE LEVEL (PPM)
LITTLE YORK LAKE—NO. 27 ²					
Brown trout	LY-3-1	M	34.3	.456	0.23
Bullhead catfish	LY-3-3	F	31.7	.414	0.10
Largemouth bass	LY-3-2	M	33.2	.720	0.15
LONG LAKE—NO. 28 ²					
Largemouth bass	6R6546	F	43.2	1.128	0.45
Northern pike	6R6541	F	53.8	1.167	0.40
	6R6540	M	71.4	2.305	0.54
	6R6542	M	61.0	1.335	0.55
Smallmouth bass	6R6545	F	40.9	.767	0.54
	6R6544	F	35.6	.672	0.51
LOON LAKE—NO. 29 ²					
Smallmouth bass	6R6414	—	—	.201	0.15
Walleye pike	6R6409	F	—	.190	0.22
	6R6410	F	—	.187	0.19
	6R6408	F	—	.232	0.24
ONEIDA LAKE—NO. 30 ²					
Walleye pike	3-OneL-1	F	59.6	2.060	0.23
	3-OneL-2	F	48.3	1.030	0.17
ONONDAGA CREEK—NO. 31 ²					
Brown trout	OS-P154-4-1	F	32.2	.386	0.43
	OS-P154-4-2	—	29.5	.314	0.43
	OS-P154-4-3	—	24.1	.181	0.44
OTSEGO LAKE—NO. 32 ²					
Cisco	7-OTSL-1	—	25.9	.161	0.30
	7-OTSL-2	M	34.3	.365	0.33
	7-OTSL-3	F	36.1	.488	0.36
Lake trout	7-OTSL-6	—	—	—	0.34
	7-OTSL-5	—	—	—	0.33
	7-OTSL-4	—	—	—	0.33
Whitefish	7-OTSL-9	—	—	—	0.33
	7-OTSL-7	—	—	—	0.56
	7-OTSL-8	—	—	—	0.42
PEPACTON RESERVOIR—NO. 33 ²					
Brown trout	1PR-7	F	64.8	3.115	0.40
	3PR-7	F	51.5	1.880	0.35
	2PR-7	F	41.2	.953	0.37
PISECO LAKE—NO. 34 ²					
Brown bullhead	6R6105	M	31.7	.425	0.14
Smallmouth bass	6R6104	F	35.3	.571	0.43
Whitefish	6R6103	F	38.1	.431	0.52
	6R6102	F	31.7	.295	0.48
	6R6101	M	34.8	.328	0.43
Yellow perch	6R6111	M	29.2	.262	0.65
RAQUETTE LAKE—NO. 35 ²					
Lake trout	6R6523	—	—	—	0.97
Smallmouth bass	6R6207	F	28.9	.287	0.37
	6R6206	F	27.6	.287	0.31
	6R6208	F	27.4	.251	0.35

TABLE 3.—Residues of total selenium in fish from New York State waters in 1969—Continued

SPECIES ¹	TAG No.	SEX	LENGTH (CM)	WEIGHT (KG)	SELENIUM RESIDUE LEVEL (PPM)
RAQUETTE LAKE—NO. 35—Continued					
Whitefish	6R6245	—Imm.	33.0	.337	0.59
	6R6244	F	34.8	.453	0.76
	6R6625	F	31.7	.349	0.93
	6R6243	F	35.6	.497	0.75
	6R6627	M	34.0	.388	0.52
	6R6626	M	35.3	.439	0.84
RUSHFORD LAKE—NO. 36—					
Lake trout	2RL-1	F	75.2	4.195	0.23
SARANAC LAKE—NO. 37—					
Smallmouth bass	5SL-5	F	33.2	.684	0.30
SALMON RIVER—NO. 38—					
Coho salmon (Pulaski, N.Y.)	69-SR-CS-6	M	61.5	1.910	0.29
	69-SR-CS-4	F	56.8	1.983	0.27
	69-SR-CS-2	F	63.0	2.072	0.31
	69-SR-CS-10	M	61.8	1.920	0.34
	69-SR-CS-10 (testes)	M	—	—	0.18
	69-SR-CS-7	M	60.5	2.046	0.31
	69-SR-CS-8	M	57.3	1.984	0.44
	SR-CS-1	F	48.8	1.036	0.27
	69-SR-CS-5	F	53.9	1.661	0.31
	69-SR-CS-11	F	61.8	1.840	0.29
	CSJ-SR-2	M	35.6	.620	0.31
	CSJ-SR-1	M	40.7	.663	0.31
	CSJ-SR-3	M	38.1	.544	0.30
	69-SR-CS-9	F	61.8	1.735	0.35
	SRCJSJ-70-1	—Imm.	33.0	.444	0.35
	SRCJSJ-70-2	do.	30.5	.280	0.34
	SRCJSJ-70-3	do.	34.9	.520	0.33
SRCJSJ-70-4	do.	32.4	.400	0.27	
SRCJSJ-70-5	do.	30.9	.326	0.34	
SRCJSJ-70-6	do.	30.9	.327	0.36	
SARATOGA LAKE—NO. 39—					
Walleye pike	6R6403	F	—	.357	0.22
SCHROON LAKE—NO. 40—					
Lake trout	1	—	—	—	0.27
SKANEATELES LAKE—NO. 41—					
Rainbow trout	3-SKL-1	F	50.8	1.235	0.56
	3-SKL-3	F	38.1	.644	0.71
SPRING BROOK—NO. 42—					
Coho salmon	5M381	M	54.4	1.710	0.36
	4RA2991	—	56.8	2.160	0.37
	4RA3601	—	61.5	2.680	0.38
ST. LAWRENCE RIVER—NO. 43—					
Brown bullhead	9SL-5	—	—	—	0.14
Muskellunge	10ISL-4	M	77.5	3.832	0.27
Smallmouth bass	1STL-4	M	29.5	.398	0.34
	6STL-4	M	26.8	.263	0.41
	2STL-4	M	26.9	.280	0.41
	6SL-5	—	—	—	0.35
	3STL-4	—Imm.	23.9	.242	0.43
	4STL-4	F	30.0	.339	0.34
Sturgeon	5STL-4	M	29.5	.344	0.39
	G429	—	102.5	6.830	0.33
	G435	—	89.6	4.540	0.34
	G430	—	—	3.630	0.23
	G432	—	97.3	5.690	0.33
	G434	—	100.8	6.590	0.25
	G431	—	103.5	8.200	0.29
	G433	—	96.3	5.910	0.33

TABLE 3.—Residues of total selenium in fish from New York State waters in 1969—Continued

SPECIES ¹	TAG No.	SEX	LENGTH (CM)	WEIGHT (KG)	SELENIUM RESIDUE LEVEL (PPM)
ST. LAWRENCE RIVER—NO. 43—Continued					
Sturgeon (Cont'd) (Massena)	1-SL-54	—	—	—	0.38
	3-SL-56	—	—	—	0.35
	2-SL-56	—	—	—	0.46
	4-SLS-6	M	95.5	7.210	0.31
Walleye pike (Massena)	1-STL-Hg-4	—	—	—	0.31
	4-MAN-1	—	—	—	0.30
	4-MAN-2	F	45.3	1.060	0.31
	4-MAN-3	F	65.5	3.527	0.30
SUSQUEHANNA RIVER—NO. 44—					
Walleye pike	3-SQR-1	—	—	—	0.19
	3-Susq R-6	M	22.8	.125	0.20
	3-Susq R-7	M	33.0	.305	0.24
Yellow perch	3-Susq R-5	M	27.9	.268	0.18
	3-Susq R-4	M	24.8	.166	0.21
	3-Susq R-3	—	—	—	0.19
TROUT LAKE—NO. 45—					
Chain pickerel (West Shore)	6R6456	F	62.0	1.570	0.20
	6R6457	M	46.3	.785	0.27
	6R6458	F	49.5	.918	0.24
Smallmouth bass	6R6459	F	43.7	1.197	0.30
UPPER SARANAC LAKE—NO. 46—					
Largemouth bass	5-US-1	F	34.8	.904	0.34
	5-US-2	F	31.2	.432	0.32
UTOWANA LAKE—NO. 47—					
Smallmouth bass	6R6504	F	42.9	.949	0.53
	6R6505	M	42.5	1.030	0.60
Rainbow trout	6R6506	M	43.0	.844	0.40
	6R6508	F	41.5	.775	0.40
WANETA LAKE—NO. 48—					
Muskellunge	1MI-2	—	—	—	0.15
WEST CANADA LAKE—NO. 49—					
Lake trout	6RS69	M	53.3	1.268	0.91
	6RS68	F	38.6	.426	1.14

NOTE: — indicates data unknown

¹ Specific location within waters, if known, given in parentheses.

Numbers refer to location of water in Fig. 1

Imm. = immature.

¹ One fillet.

TABLE 4.—Total selenium in Cayuga Lake trout by age of trout, 1970

AGE (YEARS)	SELENIUM (PPM)
1	0.53
2	0.52
3	0.40
4	0.29
5	0.40
6	0.52
7	0.49
8	0.61
9	0.44
12	0.52

NOTE: Sensitivity level = 0.1 ppm.

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*Effects of Insecticides on Populations of Rodents in Kansas—1965-69*¹

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ABSTRACT

The general objective of this study was to determine the effect of recommended applications of commonly used insecticides on population dynamics of unconfined rodents in two sites in Ellis County, Kans. Six insecticides (diazinon, endrin, heptachlor, parathion, methyl parathion, and aldrin) were applied to one field, while a second field served as a control. Insecticide applications began in summer 1965 and continued through summer 1968. Live trapping of rodents was conducted on the treated field and the control field from 1965 through 1969. Specimens were collected each month for residue analysis.

During 19 separate 10-day trapping periods, 4,661 rodents were captured of which 162 were analyzed for the six insecticides. Insecticidal residues were detected in 36 of these specimens. Dieldrin residues were detected in 33 specimens and heptachlor epoxide in 8 specimens (five of these specimens had residues of both dieldrin and heptachlor epoxide). These residue levels were low with no dieldrin levels in specimens exceeding 0.50 ppm, and no heptachlor epoxide levels exceeding 0.02 ppm. No residues of the other four insecticides were detected in any of the specimens.

Species composition of the trapped rodents was similar for the two study areas as were the population levels, with *Peromyscus maniculatus* comprising about 74% of the rodent population on the two areas. Population fluctuation trends for the two areas were also similar. Average minimal longevity for *P. maniculatus* was 45.7 days on the treated area and 50.9 days on the control area. Monthly survival between June and September was about 45% on both areas. Recaptures of *P. maniculatus* the year following their initial marking were slightly greater on the untreated area than on the treated area. The slight differences in dynamics of the

rodent populations on the two areas could easily have been due to chance, minor differences between the two study areas, or differential emigration of specimens from the areas as well as direct or indirect effects of insecticidal applications.

Introduction

Although much research has been done on various aspects (persistence, toxicity, species susceptibility, biological accumulation, etc.) of insecticides, little effort has been expended to study the long-term effects of insecticides on populations of unconfined animals. This lack of research is even more apparent when one tries to ascertain the effects of recommended insecticidal applications on wildlife populations under natural conditions. Because of this lack of fundamental data, research on the effects of recommended insecticidal applications on the population dynamics of unconfined small rodents was initiated in west central Kansas in 1965, as a subunit of a larger research project involving studies of fish populations, and insecticidal residues in soil, water, and vegetation. The results of the rodent population dynamics investigation are reported in this paper, while the results of other aspects of the comprehensive study were published separately in an earlier issue of the *Pesticides Monitoring Journal* (14).

The Study Area

This study was conducted on two sites in Ellis County, approximately 21 km (13 miles) southwest of Hays, Kans. Because the region was part of a newly created Cedar Bluffs Irrigation District and had not been intensively cultivated prior to 1965, insecticide use on the locale had been minimal. No insecticidal residues were detected in samples of water, soil, plants, or animals collected from the two study sites before the study began in 1965. The history and development of the general region are described in detail by Knutson *et al.* (14).

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Average annual rainfall for the area is 59 cm (23 in), of which 75% normally occurs in localized convective storms during the growing season (April through September). The mean annual temperature is 12 C (54 F), with mean monthly extremes of 27 C (80 F) for July and -2 C (29 F) for January. Silty clay loam is the most common soil type on the study sites. A more detailed discussion of climate and physical characteristics of the study area is presented in Leonard (16) and Knutson *et al.* (14).

The two sites utilized during this study were a "treated area" which had insecticides applied to it at recommended levels and an "untreated area" which had no insecticides applied to it. The treated area was a 7.0-hectare (ha) (19.5 acre) field with three terraces. The untreated area consisted of a similar 8.4-ha (22.7 acre) field with one terrace and situated 1.6 km (1 mile) south of the treated area.

Corn (*Zea mays*) and sorghum (*Sorghum vulgare*) were produced on both areas during the study period. The density and composition of vegetation on terraces and areas bordering both fields were similar and consisted mainly of small kochia (*Kochia scoparia*), dandelion (*Taraxacum officinale*), giant foxtail (*Setaria faberii*), yellow foxtail (*Setaria lutescens*), goldenrod (*Solidago* spp.), ragweed (*Ambrosia* spp.), and sandbur (*Cenchrus pauciflorus*).

Materials and Methods

SAMPLING PROCEDURES

Insecticidal applications on agricultural crops grown on the treated area were carefully monitored and were in accordance with recommended dosages and procedures of the Extension Entomologists of the Kansas Extension Service. Application rates varied from year to year and reflected the general insecticidal usage pattern of the region. A summary of the amounts of diazinon, endrin, heptachlor, parathion, methyl parathion, and aldrin applied to the treated area is presented in Table 1. The dates of insecticidal applications appear in Table 2. A more detailed discussion of time and method of insecticidal application on the treated area appears in Knutson *et al.* (14). No insecticides were intentionally applied to the untreated area during the course of the study, however, some dieldrin-treated sorghum seeds may have been planted inadvertently on the untreated area during one or two of the years.

TABLE 1.—Summary of insecticides applied to the treated area (pounds per acre) from 1965 to 1968

YEAR	DAIZINON	ENDRIN	HEPTACHLOR	PARATHION	METHYL PARATHION	ALDRIN
1965	0.60	0.00	0.00	0.38	0.00	2.45
1966	2.29	0.30	0.67	1.40	0.38	1.20
1967	2.13	0.36	0.43	0.79	0.50	0.84
1968	3.46	0.28	0.41	1.01	0.53	0.30

TABLE 2.—Dates of insecticide application to the treated area

TREATMENT	DATES OF APPLICATION			
	1965	1966	1967	1968
Soil	May 10-11	May 12	May 15-16	May 25
Foliage	Aug. 19	Aug. 3	Aug. 10	Aug. 5

¹ Diazinon, heptachlor, parathion, and aldrin were used as soil treatments while foliage was treated with diazinon, endrin, and methyl parathion.

Data on small rodent populations on the two areas were obtained by the capture-recapture method. Live traps similar to those of Scheffer (19) were placed at 18-m (60-ft) intervals on the terraces and along the edges of each study area; 215 live traps were used throughout the study, 151 on the treated area and 64 on the untreated area. More traps were set on the treated area than on the untreated area because of extra terraces in the treated area.

Initial trapping in August 1964 determined that rodents lived mainly in habitats along field edges and on the terraces of the two areas and moved laterally out into the fields foraging for food. At that time, 50 traps located within 60 feet of a terrace captured 21 rodents (42% success), while 50 traps located 120 feet or more from a terrace captured only 3 rodents (6% success). The soil under the crops was bare and weedless, affording little cover in which rodents could reside. Thus, trapping intensity, i.e., traps per 305 m (1,000 ft) of rodent habitat, was similar for both areas during the course of this study. Trapping was conducted on each area during 19 separate 10-day trapping periods, spring and summer 1965-1969. To reduce heat induced mortality of trapped animals, which could occur during daytime, all traps were baited and set at dusk, then emptied and or sprung the following morning, and left unset until dusk.

Traps were baited with a mixture of rolled oats and peanut butter (4). Bait from the previous day was removed before a new ball of the mixture, approximately 13 cm (0.5 in) in diameter, was placed in each trap. Additional bait was used during cold weather (11).

Trapped animals were identified, marked, and released at the capture site. Toe clipping and ear notching combinations (2) were used to mark captured animals. To facilitate analysis, all data were placed on computer input cards following a format similar to Brotzman and Giles (3). All data were analyzed on an IBM 360 50 computer.

Population estimates were made using capture-recapture data and the procedures of Schnabel (20) and Schumacher and Eschemeyer (21). Compensation was made for differences in available trapping areas on the two study sites by calculating population estimates for each 305-m (1,000 ft) unit of trapline. There were 8.7 such

units on the treated area and 3.8 units on the untreated area. An index to annual mortality was determined by dividing the number of June and July recaptures of animals marked during the previous year by the total number of animals marked in the previous year.

During the last night of each trapping period of 1965 to 1968, five rodent specimens from each study area were collected for insecticidal residue analysis.

ANALYTICAL PROCEDURES

The specimens for insecticide analysis were quick frozen the morning they were captured. Prior to analysis for insecticidal residues, each specimen was allowed to thaw before being homogenized in a Warning high-peed blender. After homogenization, a 10-g sample of each specimen was analyzed separately for insecticidal residues. The 10-g sample was placed in an Omnimixer with 50 ml of redistilled hexane and sufficient anhydrous sodium sulfate to take up the water. The mixture was then blended at high speed for 1-2 minutes, then decanted through No. 43 Whatman filter paper into a 100-ml suction flask. The residue was extracted with two additional portions of hexane as described above, then filtered and combined in the suction flask. The container, filter paper, and contents were then washed with a final portion (10 ml) of hexane. The total hexane extract was transferred to a round-bottomed flask for concentration under vacuum at 35-40°C to 2-3 ml of hexane, then transferred quantitatively to a 15 ml-centrifuge tube using 5 ml of hexane. An aliquot was used for cleanup and gas chromatographic analysis. All solvents were glass-distilled and purified for gas chromatographic analysis.

Cleanup methods for extracts, recoveries of insecticides from fortified samples, and sensitivities by gas chromatographic analysis are reported by Kadoum (12,13). Insecticide residues were not corrected for recovery with the exception of methyl parathion, since recovery from fortified samples was essentially 100%. The insecticides were separated and detected by electron capture gas chromatography using a 6-ft glass column, packed with 3% DC-11 on 60/80 mesh silanized Gas Chrom

P (Applied Science Labs, State College, Pa.). Operating conditions were as follows: carrier gas—nitrogen at a flow rate of 36 ml/min; column temperature—200°C; injection temperature—240°C; detector cell—220°C; volume of extract injected—4 µl. The analytical procedure was capable of detecting residues of as low as 0.01 ppm of diazinon, parathion, methyl parathion, malathion, endrin, aldrin, dieldrin, heptachlor, heptachlor epoxide, *p,p'*-DDE, *o,p'*-DDT, and *p,p'*-DDT.

Results

Nineteen 10-day trapping periods on the two study areas produced data from 40,850 trap-nights (number of trapping periods × number of nights per period × number of traps set), 28,690 on the treated area and 12,160 on the untreated area. Totals of 3,306 and 1,355 individual rodents were captured on the treated and untreated areas, respectively. Fifty-four percent were recaptured later, and 35.8% of the individuals were recaptured more than once. Data from 17 trapping periods during the summers of 1965-68 were used to estimate rodent populations on the two areas. June and July recaptures of mammals marked during the previous summer were used to estimate annual mortality and survival for 1965-68. Two additional 10-day trapping periods were conducted in June and July of 1969 to estimate annual mortality and survival for the 1968-69 period.

P. maniculatus was the predominant species on both areas (Table 3). During the entire study, *P. maniculatus* made up 74.2% of the captures on the treated area and 74.0% of those on the untreated area. *Mus musculus* was the next most abundant small mammal trapped, contributing 7.4% and 8.6% to the total captures on the treated and untreated areas, respectively. *Sigmodon hispidus* was the third most commonly trapped mammal constituting 7.5% and 5.1% of the total captures on the treated and untreated areas, respectively. *Onychomys leucogaster*, *Microtus ochrogaster*, and *Reithrodontomys megalotis* each contributed less than 5.0% to the number of mammals trapped on either of the two areas. *Reithrodontomys montanus*, *Perognathus flavus*, *P. flavesens*, *P. hispidus*, *Spermophilus tridecemlineatus*,

TABLE 3.—Species composition of 3,306 and 1,355 mammals captured on the treated and untreated area, respectively, during this study

SPECIES	SPECIES COMPOSITION OF CAPTURES—(PERCENT)											
	1965		1966		1967		1968		1969		1965-69	
	TA	UA	TA	UA	TA	UA	TA	UA	TA	UA	TA	UA
<i>Peromyscus maniculatus</i>	64.1	68.4	64.8	63.6	83.0	85.5	78.9	73.7	82.3	81.5	74.2	74.0
<i>Mus musculus</i>	16.6	14.8	9.1	10.4	3.0	7.2	4.3	6.1	3.3	0.4	7.4	8.6
<i>Onychomys leucogaster</i>	3.3	0.0	1.0	8.2	0.7	1.2	5.8	5.3	4.0	13.5	2.7	4.6
<i>Sigmodon hispidus</i>	3.5	4.8	11.4	10.8	9.0	0.9	7.2	6.6	3.1	0.4	7.5	5.1
<i>Microtus ochrogaster</i>	4.7	8.9	10.8	3.3	0.0	0.0	0.2	1.8	2.0	0.7	3.7	3.2
<i>Reithrodontomys megalotis</i>	1.5	2.1	0.5	1.3	3.8	1.5	0.9	0.9	4.7	2.2	2.1	1.5
Other species ¹	6.1	0.9	2.4	2.3	0.3	3.5	1.9	5.6	0.7	1.4	2.3	2.9

NOTE: TA = Treated Area; UA = Untreated Area.

¹ Other species included *Reithrodontomys montanus*, *Perognathus flavus*, *P. flavesens*, *P. hispidus*, *Spermophilus tridecemlineatus*, and *Dipodomys ordii*.

and *Dipodomys ordii*, each constituted less than 1.0% of the mammals trapped on either of the two areas.

Early in the analysis of data, attempts were made to estimate populations of each species of rodent on the area, but period-to-period and season-to-season variations were extremely large (Table 3). Therefore, population estimates of individual species were abandoned in favor of crude estimates of the composite rodent population and an estimate of the most abundant rodent species, *P. maniculatus*. The trappable populations estimated by the Schumacher-Eschmeyer method were slightly higher than those calculated by the Schnabel method. The differences in estimates by these two methods averaged 3.4% (range of 0.0% to 18.4%) for the entire study, and this difference was not significant for the entire study nor for any specific trapping period during the study.

Insecticidal residues were detected in 36 of 162 specimens analyzed. Residues of dieldrin were detected in 33 specimens and heptachlor epoxide in 8 specimens (5 specimens contained residues of both dieldrin and heptachlor epoxide). No residues of the other four insecticides were detected in any of the specimens. Residue concentrations were low ranging from 0.01 to 0.50 ppm of dieldrin and 0.01 to 0.02 ppm of heptachlor epoxide. Seven (7.1%) of 98 specimens from the treated area had detectable residues of heptachlor epoxide, while only 1 (1.6%) of 64 specimens from the untreated area

showed heptachlor epoxide residues. Residues of dieldrin were detected in 26 (27.6%) of the 98 specimens from the treated area and in 7 (10.9%) of the 64 specimens analyzed from the untreated area (Table 4). Average dieldrin residue concentrations (0.07 ppm) were higher in specimens from the treated area than the average residue concentrations (0.04 ppm) in specimens from the untreated area.

Of nine species of rodents analyzed for insecticidal residues, five contained detectable amounts of dieldrin (Table 5). Specimens of *Mus musculus*, *Spermophilus tridecemlineatus*, and *P. maniculatus* contained more dieldrin (average concentrations of 0.07 to 0.10 ppm) in their carcasses than the other specimens. *Onychomys leucogaster* and *Reithrodontomys megalotis* also contained dieldrin residues in their carcasses, but in lesser amounts (average concentrations of 0.03 ppm). With the exception of specimens of *Reithrodontomys megalotis*, average residue concentrations were greater in carcasses of species collected from the treated area than those from the untreated area. Heptachlor epoxide residues (0.01 to 0.02 ppm) were found in four specimens of *P. maniculatus* and in one each of *Onychomys leucogaster*, *Signiodon hispidus*, and *Spermophilus tridecemlineatus* from the treated area. One *Onychomys leucogaster* from the untreated area had residues (0.01 ppm) of heptachlor epoxide. Of the 36 specimens in which residues were detected, 20 (56.0%) were males and 16 (44%) were females.

TABLE 4.—Summary of dieldrin residues (≥ 0.01 ppm) in 162 collected during this study

YEAR	TREATED AREA						UNTREATED AREA					
	NO. ANALYZED	DIELDRIN RESIDUES PRESENT			NO. ANALYZED	DIELDRIN RESIDUES PRESENT						
		NO. POSITIVE	PERCENT POSITIVE	CONCENTRATIONS (PPM)			NO. POSITIVE	PERCENT POSITIVE	CONCENTRATIONS (PPM)			
AVERAGE	MEDIAN	RANGE	AVERAGE	MEDIAN	RANGE	AVERAGE	MEDIAN	RANGE				
1965	19	3	15.8	0.09	0.01	0.01-0.24	14	0	0.0	—	—	—
1966	22	3	13.0	0.08	0.09	0.02-0.13	18	3	16.7	0.02	0.02	0.01-0.03
1967	31	14	45.2	0.05	0.02	0.01-0.50	12	2	16.7	0.01	0.01	0.01-0.01
1968	26	6	23.1	0.13	0.03	0.01-0.44	20	2	10.0	0.09	0.09	0.04-0.15
Total	98	26	27.6	0.07	0.02	0.01-0.50	64	7	10.9	0.04	0.02	0.01-0.15

TABLE 5. Dieldrin residues (≥ 0.01 ppm) in nine rodent species analyzed during this study

SPECIES	TREATED AREA						UNTREATED AREA					
	NUMBER ANALYZED	NUMBER CONTAMINATED	PERCENT CONTAMINATED	CONCENTRATION IN THOSE CONTAMINATED (PPM)			NUMBER ANALYZED	NUMBER CONTAMINATED	PERCENT CONTAMINATED	CONCENTRATION IN THOSE CONTAMINATED (PPM)		
				AVERAGE	MEDIAN	RANGE				AVERAGE	MEDIAN	RANGE
<i>Peromyscus maniculatus</i>	31	12	38.7	0.08	0.01	0.01-0.50	21	3	14.3	0.03	0.03	0.01-0.04
<i>Mus musculus</i>	14	6	42.9	0.10	0.01	0.01-0.44	11	2	18.2	0.08	0.08	0.02-0.15
<i>Onychomys leucogaster</i>	9	3	33.3	0.03	0.02	0.02-0.05	7	1	14.3	0.01	—	—
<i>Reithrodontomys megalotis</i>	6	1	16.7	0.03	—	—	5	1	20.0	0.03	—	—
<i>Spermophilus tridecemlineatus</i>	7	4	57.1	0.07	0.02	0.01-0.24	0	0	0	—	—	—
<i>Signiodon hispidus</i>	15	0	0	—	—	—	6	0	0	—	—	—
<i>Microtus ochrogaster</i>	9	0	0	—	—	—	8	0	0	—	—	—
<i>Perognathus flavescens</i>	5	0	0	—	—	—	2	0	0	—	—	—
<i>Perognathus hispidus</i>	2	0	0	—	—	—	4	0	0	—	—	—

Fluctuations of the total rodent populations on the treated and untreated areas followed the same general trend during the study (Fig. 1). The longest time between the date of first capture and date of last capture of a specific specimen was 34 months, a *Sigmodon hispidus* male on the treated area. Two *P. maniculatus* males survived at least 24 months on the treated area. Several individuals were known to have survived at least 14 months on the untreated area. For *P. maniculatus*, the average time between date of initial capture and date of last recapture varied from a high of 53.4 days on the untreated area to a low of 42.8 days on the treated area (Table 6). The average minimal longevity during 1965-68 for *P. maniculatus* on the treated area was less than the average minimal longevity for *P. maniculatus* on the untreated area.

FIGURE 1.—Estimated (Schumacher-Eschmeyer method) trappable population per 305 m (1,000 ft) of trapline on the treated and untreated areas. Vertical lines represent 95% confidence intervals about the mean.

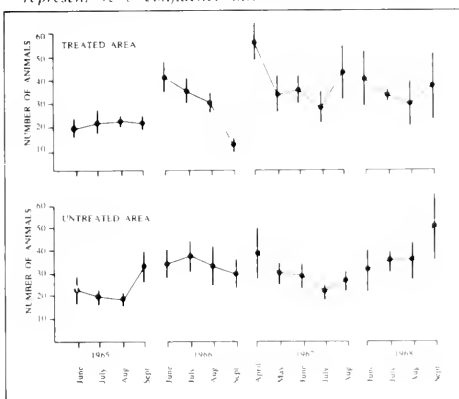


TABLE 6.—Average minimal longevity (\pm S.D.) for *Peromyscus maniculatus* recaptured during this study

YEAR ¹	TREATED AREA (DAYS)	UNTREATED AREA (DAYS)
1965	48.1 \pm 7.8	49.8 \pm 8.7
1966	46.7 \pm 11.4	50.6 \pm 14.2
1967	42.8 \pm 6.2	53.4 \pm 8.9
1968	45.2 \pm 5.8	49.9 \pm 9.1
Mean (\pm S. E.)	45.7 \pm 2.3	50.9 \pm 1.7

¹ Indicates year in which the individuals were first captured.

An index to summer mortality within each rodent population was determined by following a single group of marked individuals through a summer. Rodents marked during the June trapping period of each year on each area were considered separate groups and later recaptures of these rodents were used as evidence of survival. Life tables were constructed to obtain an index

to mortality (10). The index to monthly mortality was strikingly similar for both areas, averaging 54.5% and 54.9% on the treated and untreated areas, respectively (Table 7).

The number of animals marked one year and recaptured during June or July the following year was small, averaging 3.4% and 5.9% for the treated and untreated areas, respectively (Table 8).

TABLE 7.—Summer mortality of *Peromyscus maniculatus* on the two study areas determined from recaptures of *P. maniculatus* marked in the initial trap period of each year

YEAR	NUMBER MARKED IN JUNE PERIOD	NUMBER RECAPTURED IN SUCCEEDING MONTHS			TOTAL
		JULY	AUGUST	SEPTEMBER	
TREATED AREA					
1965	310	194	173	154	521
1966	389	204	176	155	535
1967	493	175	154	—	329
1968	376	188	164	142	494
Totals	1,568	761	667	451	1,879
Marked <i>P. maniculatus</i>					
Available	1,568	1,568	1,075	—	—
Number Recaptured	761	667	451	1,879	—
Number Alive at Start	1,879	1,118	451	3,448	—
Percent Monthly Mortality	40.5	59.7	100.0	54.5	—
UNTREATED AREA					
1965	107	60	54	47	161
1966	112	56	50	44	150
1967	128	57	50	—	107
1968	100	48	42	34	124
Totals	447	221	196	125	542
Marked <i>P. maniculatus</i>					
Available	447	447	319	—	—
Number Recaptured	221	196	125	542	—
Number Alive at Start	542	321	125	988	—
Percent Monthly Mortality	40.8	61.1	100.0	54.9	—

TABLE 8.—*Peromyscus maniculatus* recaptured during June and July trapping periods the year following initial marking on the two areas

YEAR MARKED	TREATED AREA		UNTREATED AREA	
	NUMBER MARKED	RECAPTURES SUCCEEDING YEAR	NUMBER MARKED	RECAPTURES SUCCEEDING YEAR
1965	319	10	160	5
1966	437	21	165	16
1967	733	18	274	14
1968	502	18	192	12
Total	1,991	67	791	47
Percent Recaptured		3.4		5.9

Discussion

This study was meant to be a comparison of two rodent populations; one exposed to insecticides and one not exposed to insecticides. However, the experimental design did not permit replications or a study of two closed populations. As far as could be discerned from field observations, the only fundamental difference between

the two study areas which could have markedly influenced rodent populations was the insecticide treatment. Since the populations were not closed, rodent immigration and emigration were unknowns in the study. Likewise since no large enclosures were used, natural predation on the two areas could not be controlled. It appears though that the rodent populations on the two areas were very similar in composition and dynamics. Without adequate replications, only major changes in the rodent populations could have been detected. Since no attempt was made in this study to accurately determine the total population of rodents on each of the two study areas, the many limitations of the basic capture-recapture method discussed by Manly (18), Cormack (5), Tanton (22), and Leslie (17) did not influence the results. The same methods of trapping and marking were used on both study areas, thus, although the population estimates could not be regarded as entirely unbiased, the estimates are comparable. The population parameters calculated from these capture-recapture data are likewise comparable.

The relative abundance of some mammals changed from year to year, but since these changes occurred on both areas, these changes could not be considered insecticide induced. Year-to-year changes in population density and species composition are not uncommon in prairie regions of Kansas (6-9).

The average longevities calculated from capture-recapture data in this study are minimal values since they represent the time between first and last capture of the animal, not between birth and death. Unless there are differences in emigration rates of rodents from the two areas, these minimal longevity figures are comparable. Longevity of *P. maniculatus* on the treated area was slightly less than that of the population on the untreated area. Because the difference was slight and egress from the two populations not controlled, it is doubtful that the 5.2-day difference in longevity can be specifically attributable to insecticide applications on the treated area.

An index to mortality of *P. maniculatus* during summer months as estimated from recaptures was almost identical on the two areas. Again, unless there was differential emigration and trap vulnerability on the two areas, these indices to mortality are comparable. Carry-over of marked *P. maniculatus* from one trapping season to the next may have been a bit greater on the untreated area, but this could not be confirmed since emigration was not controlled.

The actual pathway of insecticidal contamination was not studied; however, the rodents probably acquired insecticides through their food. *Sigmodon*, *Microtus*, and *Perognathus* feed primarily on plant material and seeds and had no insecticide residues in their carcasses while

rodents (*Peromyscus*, *Mus*, *Onychomys*, *Reithrodontomys*, and *Spermophilus*) which had a diet that included insects in addition to seeds and plant material had insecticide residues in their carcasses. Contaminated insects (dead, dying, or those containing low levels of insecticides) could have been a source of contamination for insect eating rodents in our study as was found for insect eating rodents in Missouri (15).

Probably the most significant findings of this study were the low incidence and levels of contamination observed in the rodent population on the treated area. Only one species, *Spermophilus tridecemlineatus*, exhibited an incidence of contamination exceeding 50%. *P. maniculatus*, the most abundant species on the area, had an incidence of contamination of only 38.7% on the treated area. Levels of contamination never exceeded 0.50 ppm of dieldrin for any specimen analyzed. Although levels of dieldrin contamination were higher in rodents from the treated area early in the study, levels of contamination increased on the untreated area during the last year of our study. Collection of contaminated specimens on the untreated area was a result of immigration onto that area by rodents from surrounding areas; similar movements probably were occurring on the treated area. It appears therefore that unless closed populations of rodents can be assured, their population dynamics are probably a poor indicator of insecticide contamination. It is doubtful that monitoring of rodent populations can detect anything but changes of extremely large magnitude under unconfined conditions.

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See Appendix for chemical names of compounds discussed in this paper

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Mercury Residues in Fish From Saskatchewan Waters With and Without Known Sources of Pollution—1970¹

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ABSTRACT

In 1970, mercury concentrations were determined in muscle tissue of 125 fish from 10 Saskatchewan waters, 6 of which were part of the Saskatchewan River system receiving industrial and/or municipal wastes containing mercury and 4 of which had no known sources of mercury pollution.

Concentrations in fish from the Saskatchewan River system ranged from 0.18 to 8.88 ppm depending on the degree of pollution of the fish collection site. Levels were higher (0.18 to 8.88 ppm) in fish caught downstream from a chlor-alkali plant in Saskatoon than those (0.25 to 1.2 ppm) in fish from sites upstream from this plant whose sources of mercury were municipal or industrial wastes other than the chlor-alkali industry. Levels in fish from the four lakes without any known source of mercury pollution ranged from 0.11 to 1.13 ppm. These results indicate that mercury levels in fish from apparently unpolluted waters may exceed the Canadian actionable level of 0.5 ppm for mercury; such residues may be of natural origin.

Introduction

Both nature and man contribute to the contamination of our environment with mercury (7). It has been estimated that the surface of the earth receives about 100,000 tons of mercury annually from precipitation (14) as compared to the world production of 10,000 tons of mercury per year (16). The earth's crust contains an average of 0.5 ppm mercury (6), and traces of natural mercury are to be expected everywhere; for example, mercury was found in sea water as early as 1777 (11). Subsequent studies by Stock and Cueliel (15)

during the 1930's showed that mercury was truly ubiquitous, being present in air, water, foods, and soils. Man by his activities has increased the concentration of mercury in some foods, particularly in fish, in some localized areas. The consequence of the pollution of water with mercury-containing industrial effluent was evident in Minamata, Japan, where in the 1950's several persons died or became seriously ill after eating fish or shellfish containing very high levels (27-102 ppm, dry-weight basis) of mercury (10).

To understand the contamination of fish with mercury from human activities, the various uses of mercury have to be considered. Of the 3,006 tons of mercury used in the United States in 1969, 1,156 tons (38.4%) were used in the chlor-alkali industry, 710 tons (23.6%) in the electrical industry, 116 tons (3.8%) in dentistry, 102 tons (3.4%) in agriculture, 370 tons (12.3%) in the paint industry, and the rest (18.5%) in laboratories, pulp and paper, pharmaceutical, and other industries (3). In the same year, 150 tons were used in Canada: 100 tons (66.7%) in the chlor-alkali industry and 12.5 tons (8.3%) in agriculture (8). Some mercury used in chlor-alkali and other industries escapes into local waters. Large urban centers may also contribute to water pollution with mercury since a significant amount of mercury is used in paints, hospitals (e.g., thermometers), laboratories, dental preparations, electrical equipment, batteries, light-bulbs, etc., some of which may enter sewage and become part of local waters. Elevated mercury levels are to be expected in fish from waters that receive mercury-containing industrial and/or municipal wastes.

The use of mercury in agriculture, mainly as seed-dressings, accounts for only a small part of total mercury consumption. Furthermore, mercurial seed-dressings are

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used at about 500 mgHg/acre or 0.00055 ppm/6-inch acre (13). Such small quantities of mercury in a vast land area should not appreciably contribute to the pollution of water, as this amount, even after 30 years of continuous use on the same land, adds up to only 0.016 ppm. This amount is insignificant when compared to the natural mercury content of soil (2.15) and is less than the amount of mercury received by soil through precipitation (2). Thus, it is perhaps safe to assume that the mercury in fish from waters in agricultural areas not receiving any industrial and/or municipal wastes can be considered to be of natural origin.

Numerous studies from many countries have reported the mercury content of fish from contaminated waters (12). Mercury levels in fish from many rivers and lakes in Canada and the United States have been reported in excess of 1 ppm (1,4,5,17), and commercial fishing in many waters has been banned. Little, however, is known about the mercury content of fish from apparently unpolluted waters. Studies of "background" levels would be useful in establishing a practical actionable level since such levels of natural mercury have probably been present in fish all the time and may have done man no harm.

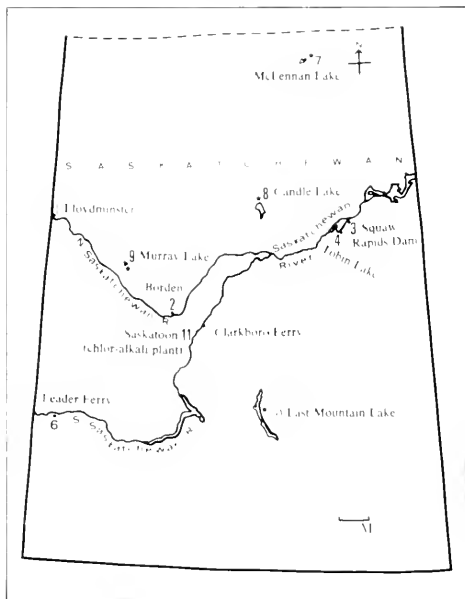
The object of the study reported here was to determine the mercury levels in fish from 10 Saskatchewan waters, 6 with industrial and/or municipal sources of mercury pollution (Group I sites) and 4 with essentially only natural sources of mercury (Group II sites).

Sampling Sites and Procedures

The fish collection sites are shown in Fig. 1. Group I sites were located on the Saskatchewan River system and included Lloydminster (Site 1) on the North Saskatchewan River at the Alberta-Saskatchewan border; Borden (Site 2); Squaw Rapids Dam (Site 3); Tobin Lake (Site 4); Clarkboro Ferry (Site 5); and Leader Ferry (Site 6). The city of Saskatoon (designated Site 11) is a principal source of pollution, since a chlor-alkali plant there discharges effluents containing mercury into the river system; all sampling sites downstream from Saskatoon would be contaminated by this plant as well as by municipal wastes from Saskatoon and several other cities upstream. Sampling sites upstream from Saskatoon and on the South Saskatchewan River would be contaminated only by cities at appreciable distance upstream, such as Calgary in Alberta; Edmonton, also in Alberta, may contribute to pollution of the North Saskatchewan River.

The Group II sampling sites, McLennan Lake (Site 7), Candle Lake (Site 8), Murray Lake (Site 9), and Last Mountain Lake (Site 10), did not receive industrial or municipal wastes. McLennan Lake in Northern Saskatchewan is remote from human activities, while

FIGURE 1.—Map of Saskatchewan showing the collection sites—1970



agricultural land forms a substantial part of the precipitation area of Candle Lake, and Murray and Last Mountain Lakes are within agricultural areas. It has been pointed out that agricultural uses of mercury do not contribute to pollution of water or fish (12). Although water or bottom mud from the lakes were not analyzed, these Group II sites were considered apparently unpolluted by man.

During the summer of 1970, a total of 125 fish were netted at the 10 sampling locations and brought to the laboratory as rapidly as possible. Muscle tissue samples were taken from the longitudinal dorsal muscles on the anterior section of each fish and frozen in plastic bags at -18°C until analyzed.

Analytical Procedures

The total mercury content of each muscle tissue sample was determined by atomic absorption spectrometry according to the method of Saha *et al.* (13). A 4- to 5-g sample of fish tissue was digested under reflux with concentrated nitric acid and perchloric acid to destroy organic matter. The acidity of the digest was adjusted to approximately 1N, and hydroxylamine hydrochloride was added to destroy excess oxidants. The solution was then extracted twice with chloroform to remove any

organic matter. Mercuric ions were then extracted with a chloroform solution of dithizone and determined by atomic absorption spectrometry. About 93 to 98% of the mercury added to muscle tissue as $HgCl_2$ could be recovered by this method, and the minimum detectable amount was 0.005 ppm Hg. All fish specimens were analyzed in duplicate, and the mercury levels were not corrected for recovery.

Results and Discussion

The average mercury concentration with standard deviation, range, and median values for fish species from each sampling site are given in Table 1.

The mercury concentrations in fish from Group I sites ranged from 0.18 to 8.88 ppm and generally averaged above the 0.5 ppm Canadian actionable level. The goldeye fish from Lloydminster (Site 1) averaged 0.74 ppm of mercury; and long-nose suckers from Borden (Site 2), a point downstream from Lloydminster, had a rather high average level for the species (0.57 ppm) since this species is known to accumulate less mercury than other species, for example, pike. The waters at those two sites on the North Saskatchewan River were probably contaminated by industrial and municipal wastes from cities along the river bank. Squaw Rapids Dam (Site 3) and Tobin Lake (Site 4) were only 30 miles apart and both received contaminated waste from the chlor-alkali plant in Saskatoon (about 300 miles upstream) and other industrial and municipal wastes from many cities along the river system. The average mercury content of pike varied considerably for these two sites (Site 3—1.94 ppm and Site 4—0.86 ppm) although the lower ranges were similar (Site 3—0.49 ppm and Site 4—0.63 ppm). These results are not unusual, since other

studies have reported widely differing amounts of mercury in the same species from the same location (9,17). The highest average mercury concentrations were in goldeye (2.05 ppm) and pike (4.80 ppm) from Clarkboro Ferry (Site 5); these levels reflect the effects of direct discharge of individual wastes containing mercury into water, since the sampling site is only a few miles downstream from the chlor-alkali plant in Saskatoon (Site 11).

It has been estimated that about 0.25 to 0.5 lb mercury can be lost to the environment for every ton of chlorine produced, and plants producing 100 tons or more of chlorine per day are rather common (3,4). A plant of this size can then discharge 9 to 18 thousand lb of mercury per annum into the environment, mainly in water.

Fish (Sauger) from Leader Ferry (Site 6), a site upstream from the chlor-alkali plant in Saskatoon and contaminated with municipal or other types of industrial wastes averaged only 0.67 ppm mercury.

The mercury concentrations in fish from Group II sites ranged from 0.11 to 1.13 ppm. Mean levels of mercury were low in walleye from Candle Lake and pike from McLennan Lake, averaging 0.18 ppm and 0.24 ppm of mercury, respectively. The walleye from Last Mountain Lake, however, averaged 0.45 ppm, a concentration barely within the 0.5 ppm Canadian actionable level. Of particular interest was the mercury content of perch from Murray Lake (0.71 ppm), a level about three times higher than in perch from Tobin Lake (0.24 ppm), a site with known industrial and municipal mercurial contamination. These results indicate that mercury levels in fish in excess of 0.5 ppm do not necessarily result

TABLE 1.—Mercury residues in fish from some Saskatchewan waters with and without known sources of pollution—1970

SAMPLING SITE NO. AND LOCATION	FISH SAMPLES	NO. OF FISH	MERCURY CONCENTRATION (PPM, FRESH-WEIGHT BASIS)		
			MEAN \pm S.D.	RANGE	MEDIAN
GROUP I (INDUSTRIAL AND/OR MUNICIPAL POLLUTION)					
1. Lloydminster ¹	Goldeye	10	0.74 \pm 0.25	0.52-1.2	0.63
2. Borden ¹	Longnose sucker	15	0.57 \pm 0.19	0.25-0.90	0.57
3. Squaw Rapids Dam ²	Pike	10	1.94 \pm 2.50	0.49-8.88	1.06
4. Tobin Lake ²	Pike	10	0.86 \pm 0.24	0.63-1.2	0.91
	Sauger	6	0.98 \pm 0.26	0.79-1.50	0.89
	Goldeye	16	0.42 \pm 0.08	0.30-0.55	0.41
	Perch	12	0.24 \pm 0.07	0.18-0.42	0.21
5. Clarkboro Ferry ³	Goldeye	5	2.05 \pm 1.37	0.96-4.25	1.30
	Pike	3	4.80 \pm 2.25	2.60-6.11	6.09
6. Leader Ferry ³	Sauger	4	0.67 \pm 0.20	0.42-0.84	0.71
GROUP II (NO KNOWN SOURCES OF POLLUTION)					
7. McLennan Lake	Pike	4	0.24 \pm 0.07	0.16-0.32	0.23
8. Candle Lake	Walleye	9	0.18 \pm 0.04	0.11-0.24	0.19
9. Murray Lake	Perch	11	0.71 \pm 0.20	0.42-1.13	0.70
10. Last Mountain Lake	Walleye	10	0.45 \pm 0.13	0.23-0.68	0.41
Total		125			

¹ Polluted with industrial and municipal wastes from cities, but no chlor-alkali plant.

² Polluted with industrial and municipal wastes and by effluents from a chlor-alkali plant at considerable distance upstream.

³ Polluted with industrial and municipal wastes and direct discharge of effluents from a chlor-alkali plant.

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PESTICIDES IN SOIL

Pesticide Residues in Soil From Eight Cities—1969

G. B. Wiersma¹, H. Tai¹, and P. F. Sand²

ABSTRACT

Soil samples from eight cities were analyzed for pesticide residues. Besides DDT and its metabolites (DDTR), other pesticides detected were dieldrin, chlordane, heptachlor, heptachlor epoxide, toxaphene, and endrin. No organophosphate residues were detected. Levels of DDTR varied significantly among the eight cities, with the highest average residue level in Miami, Fla. (5.98 ppm) and the lowest in Houston, Tex. (0.35 ppm). When residue levels in lawn or garden areas were compared to those in unkept areas within the cities, DDTR residues were significantly greater for lawn areas.

Introduction

Much information has been developed on pesticide residues in soils; however, little is available on residue levels in soil from urban areas. Fahey, Butcher, and Murphy (1) sampled urban soil in Battle Creek, Mich., for chlorinated hydrocarbons and found residues of DDT ranging from 0.07 to 79.92 ppm. Purves (2) studied the difference between residues of trace elements found in urban garden plots and those in rural areas and found residues of boron, lead, and zinc to be greater in urban garden plots than rural plots.

The present report covers a preliminary survey of pesticide residues in soil initiated in response to the need for more intensive studies of pesticide residues in urban areas of the United States.

Sampling Procedures

Eight U.S. cities at different geographical locations throughout the country were selected for soil sampling during the summer and fall of 1969. Fifty sampling sites were randomly chosen within each city. Each site was a 50- by 50-ft plot, modified at times to meet local conditions, but always containing 2,500 square feet. Within each site, 9 soil cores (2 inches in diameter by 3 inches deep) were collected on a 3- by 3-grid. These cores were sifted through a 1/4-inch mesh, composited, and shipped to the Pesticide Monitoring Laboratory in Gulfport, Miss.

Analytical Procedures

A subsample of soil, weighing 300 g wet weight, was placed in a 2-qt jar with 600 ml of 3:1 hexane-isopropanol solvent. The jars were sealed and rotated for 4 hours. After rotation, the soil was allowed to settle, and 200 ml of the extract solution was filtered into a 500-ml separatory funnel. The isopropanol was removed by two washings with distilled water. The hexane was then filtered through a funnel containing glass wool and anhydrous sodium sulfate (Na₂SO₄). No further cleanup was normally required before analysis.

Gas chromatography was employed for the analysis of organochlorine pesticides using the electron affinity detector and for organophosphorus pesticides using the flame photometric detector. The essential experimental conditions were as follows:

Gas Chromatographs:

1. Hewlett-Packard Model 402 and Model 810—tritium foil electron affinity detector (pulsed-cell potential); carrier gas—5% methane in argon and flow rate—about 80 ml/min.
2. MicroTek Model 220—tritium foil electron affinity detector (DC-cell potential); flame photometric detector, phosphorus filter, hydrogen-oxygen flame (hydrogen flow rate—150-200 ml/min, oxygen flow rate—30-35 ml/min); carrier gas—purified nitrogen and flow rate—about 100 ml/min.

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Columns: Glass, 183 cm long by 6 mm, o.d. and 4 mm, i.d. with the following packings:
 3% or 10% DC-200 on 100 120 mesh Gas Chrom Q
 9% QF-1 on 100 120 mesh Gas Chrom Q
 5% XE-60 on 100 120 mesh Chromosorb W

Temperatures: Detector 200° C
 Injection port 250° C
 Column DC-200 180° C
 Columns QF-1 and XE-60 160° C

Peak height was used to measure the amount of pesticide residues. A dual-column system employing polar and nonpolar columns was utilized to identify and confirm pesticides. Further confirmation, when necessary, was made by thin layer chromatography or partition values. Average recovery values for most of the organochlorine pesticides ranged from 90% to 100%, and with the exception of chlordane and toxaphene, the detection limits were 0.01 to 0.02 ppm. Chlordane was calculated using gamma-chlordane as a standard (gamma-chlordane constitutes an average of 10% of technical chlordane), with a detection limit of 0.04 ppm. Toxaphene was measured by comparing the summation of peak heights of several peaks (usually four selected peaks from its multiple-peak gas chromatogram) with the corresponding peaks of a known standard, and the detection limit was 0.05 ppm. All residues reported were corrected for recovery.

The present analytical methodology should detect common organophosphate pesticides such as DEF², diazinon, EPN, ethion, azinphosmethyl, malathion, parathion, phorate, and carbofenthothion. The recovery values ranged from 80% to 100%, with detection limits ranging from 0.01 ppm for phorate to 0.1 ppm for azinphosmethyl and EPN. It should be noted, however, that the present extraction technique would not recover many of the possibly oxidized metabolic products.

Arsenic was determined by atomic absorption spectrophotometry. The soil sample was first extracted with 9.6N hydrochloric acid and then reduced to the As⁻³ state with stannous chloride. As⁻³ was partitioned from the acid to benzene, then further partitioned from benzene into water for the absorption measurement. A Perkin-Elmer Model 303 instrument was used, and absorbance was measured with an arsenic cathode lamp at 1970 Å with argon as an aspirant to an air-hydrogen flame. This method was rapid and precise, but indicated an average recovery rate of 56% for soil samples. The arsenic levels reported in this study were corrected for the recovery value; the detection limit after correction, was 0.2 ppm.

Results and Discussion

Table 1 shows the average residues detected for all cities. Miami had the highest combined residues of DDTR (DDT and related degradation products). It also

had the highest dieldrin and chlordane residues. The dieldrin residues were particularly outstanding, because the average residue in Miami was over 10 times greater than the next highest average dieldrin residue, which was detected in Bakersfield. The highest arsenic residues were found in Salt Lake City and the lowest, in Houston. No organophosphate residues were detected.

The range of residues and the percent of sites with detectable residues are also given in Table 1. Although Bakersfield, Calif., had an average residue of 0.36 ppm for DDTR, the range was narrow (0.02 - 3.08 ppm) and 94% of the sites had detectable residues. This indicates an equal distribution of DDTR residues over the entire city. In contrast, Waterbury, Conn., has an average DDTR residue of 0.98 ppm and a wide range (0.01 - 10.35 ppm), with only 56% of the sites having detectable residues, indicating an unequal distribution over the city.

With the exception of Houston and Manhattan, cities in this study were located in States where croplands had previously been sampled as part of the National Soils Monitoring Program. Residues of arsenic, DDTR, dieldrin, and chlordane, four commonly occurring pesticides, are compared in Table 2. In most cases, the average residues detected in the cities tended to be greater than the corresponding cropland residues; however, because no tests of significance can be made on the data at this time, caution should be exercised in interpreting the results in Table 2.

Pesticide residue data can usually be described by a log normal distribution. Frequently, however, the data contain a large number of zero values, resulting either from the absence of pesticides or their presence at levels below the analytical sensitivity. To include these zero values when taking logarithms, a value must be substituted for the zero figure. After repeated tests for significant kurtosis and/or skewness, the log (X-0.1) transformation most closely approximated the normal distribution. Using this transformation, logarithmic means were determined for DDTR and arsenic residues. The 95% confidence interval about each of these means was determined. The antilogs of these figures were taken to give estimates of the geometric mean and its 95% confidence interval in the untransformed dimension. The results are given in Table 3. All figures are in parts per million.

For DDTR, the geometric mean for soil from Miami, Fla., (1.60 ppm) far exceeded means from the other cities; the confidence interval ranged from 0.91 to 2.82 ppm. Camden, N. J., had the second highest mean (0.37 ppm) with an upper limit of 0.62 ppm. Houston, Tex., had the lowest mean (0.02 ppm), and its upper limit (0.04 ppm) was the same as the lower limits for Manhattan, Kan., and Waterbury, Conn., which had virtually identical means and confidence intervals. The

TABLE 1.—Pesticide residues in soil from eight cities, 1969

City	RESIDUES IN PPM AND PERCENT POSITIVE SITES														
	ARSENIC	<i>o,p'</i> -DDT	<i>p,p'</i> -DDT	<i>o,p'</i> -TDE	<i>p,p'</i> -TDE	<i>o,p'</i> -DDE	<i>p,p'</i> -DDE	DDTR	DIELDRIN	CHLORDANE	HEPTA-CHLOR EPOXIDE	HEPTA-CHLOR EPOXIDE	TOXAPHENE	ENDRIN	
Bakersfield, Calif. Range Average Percent Positive sites ¹	1.2-52.2 7.1 100.0	0.01-0.72 0.05 66.0	0.01-1.72 0.15 90.0	ND 0.03 70.0	0.01-0.33 0.01 32.0	0.01-0.92 0.12 94.0	0.02-3.08 0.36 94.0	0.01-1.98 0.07 28.0	0.07-20.48 0.78 30.0	0.02-0.10 <0.01 4.0	0.05-0.18 0.01 8.0	ND	0.01-0.03 <0.01 6.0		
	Camden, N.J. Range Average Percent Positive sites ¹	1.0-46.3 11.2 100.0	0.01-2.06 0.19 66.0	0.03-6.74 0.75 88.0	0.01-0.28 0.02 34.0	0.01-2.19 0.16 78.0	± 0.01 <0.01 2.0	0.02-3.11 0.23 84.0	0.09-13.44 1.36 88.0	0.02-0.21 <0.01 4.0	0.39-5.90 0.36 16.0	0.02-0.39 0.01 6.0	ND	ND	
		Houston, Tex. Range Average Percent Positive sites ¹	0.2-15.3 2.1 98.0	0.01-3.06 0.11 32.0	0.01-3.79 0.17 34.0	± 0.07 <0.01 2.0	0.01-0.49 0.02 18.0	0.01-0.06 <0.01 4.0	0.01-0.90 0.05 28.0	0.01-7.68 0.35 40.0	0.01-1.47 0.04 20.0	0.04-12.94 0.66 34.0	0.01-0.02 <0.01 6.0	± 0.11 <0.01 2.0	± 0.01 <0.01 2.0
Manhattan, Kans. Range Average Percent Positive sites ¹			0.7-72.0 11.5 100.0	0.01-1.24 0.11 48.0	0.01-4.65 0.45 56.0	± 0.01 <0.01 2.0	0.01-0.78 0.06 50.0	0.01-0.03 <0.01 4.0	0.01-1.53 0.15 54.0	0.01-8.20 0.78 58.0	0.01-0.72 0.04 20.0	0.03-4.86 0.30 40.0	0.02-0.09 <0.01 10.0	± 12.07 0.24 2.0	ND
	Miami, Fla. Range Average Percent Positive sites ¹		0.3-33.9 2.3 80.0	0.01-5.91 0.79 86.0	0.04-42.56 2.67 94.0	0.01-0.27 0.02 24.0	0.01-5.06 0.41 84.0	0.01-0.15 0.01 8.0	0.01-11.15 2.09 100.0	0.01-53.38 5.98 100.0	0.01-8.58 0.72 64.0	0.04-16.87 1.59 64.0	0.01-0.02 <0.01 6.0	0.01-0.45 0.05 50.0	14.79-52.73 1.34 4.0
		Milwaukee, Wis. ² Range Average Percent Positive sites ¹	1.2-54.4 14.4 100.0	0.01-1.77 0.11 79.6	0.03-15.91 0.60 87.7	0.01-0.49 0.03 40.8	0.01-1.35 0.11 83.7	0.01-0.11 <0.01 10.2	0.01-2.85 0.21 89.8	0.02-22.11 1.07 91.8	0.01-1.42 0.04 20.0	0.05-10.21 0.45 34.0	0.02-0.45 0.02 12.0	0.01-0.52 0.04 32.0	ND
Salt Lake City, Utah Range Average Percent Positive sites ¹			1.8-74.5 15.7 100.0	0.01-1.38 0.09 62.0	0.02-2.64 0.23 66.0	0.03-0.09 <0.01 6.0	0.01-0.59 0.06 64.0	0.02-0.17 <0.01 4.0	0.01-0.85 0.10 66.0	0.01-5.00 0.49 72.0	0.01-1.14 0.03 26.0	0.02-7.50 0.41 38.0	0.01-0.24 0.01 12.0	0.01-0.23 0.02 26.0	± 0.06 <0.01 2.0
	Waterbury, Conn. Range Average Percent Positive sites ¹		0.9-37.9 8.5 100.0	0.01-1.26 0.14 44.0	0.01-6.46 0.43 48.0	0.01-0.67 0.04 22.0	0.01-1.29 0.13 50.0	0.01-0.04 <0.01 4.0	0.01-2.27 0.19 52.0	0.01-10.35 0.98 56.0	0.02-0.22 0.01 8.0	0.02-8.74 0.96 28.0	0.01-0.53 0.01 8.0	0.01-0.53 0.02 30.0	ND

NOTE: ND = not detected.

¹ Percent based on number of sites with residues greater than or equal to the sensitivity limits.² One value.³ Based on results for 49 sampling sites.

confidence intervals show some distinct variation in levels between the cities, but, in general, there is considerable overlap.

The geometric means of the arsenic values tended to separate the cities into two general classes—those with residues greater than 5 ppm and those with residues less than 2 ppm. Houston and Miami had geometric means of <2 ppm, and the other cities had residues >5 ppm. These variations in elemental arsenic are probably attributable to differences in geological conditions or possibly contamination from industrial or combustion sources rather than to differences in use of various arsenical pesticides.

The residue levels of DDT and some of the other pesticides were too high to have resulted from general environmental contamination, suggesting that they may have resulted from application of pesticides by city governments, home owners, or other urban activities.

In an effort to determine the sources of contamination from DDTR and arsenic in the cities, the soil residue data for each compound were pooled and divided into "lawn" and "unkept" areas. These areas are defined as follows:

Lawn Areas

1. Mowed grass in close proximity to a house, factory, or other structure.
2. Mowed grass in municipal parks or other town-owned or -operated land.
3. Garden or cultivated areas.
4. A yard that is in obvious proximity to a home

Unkept Areas

1. Vacant lots where grass is apparently uncared for
2. Small wooded lots, brush, or overgrown fields.
3. Areas such as power lines, gas lines, etc.
4. Bare exposed soil around construction sites, eroded areas, etc

A "t" test, for unequal numbers of observations, was used to test for significance. When arsenic residues were tested, no significant difference could be detected in residues between lawn and unkept areas. This supports the hypothesis that arsenic residues resulted either from background levels or from general pollution sources such as coal or petroleum combustion. However, a significant difference ($t=4.15$) was detected for DDTR residues, with the higher residues found in the lawn areas. This indicates that DDTR residues in the city areas probably occurred as a direct result of pesticides used within the city.

In conclusion, the cities sampled in this study generally had heavy loads of chlorinated hydrocarbon pesticide residues in soil. These tended to be greater than cropland residues from the same State, although tests of significance could not be made on these data. There was some distinct variation in residue levels between cities. Finally, within the cities, DDTR residues in lawn areas were significantly greater than those in unkept areas.

TABLE 2.—Comparison of residue levels of compounds present in both urban and cropland soils in six States

LOCATION	RESIDUES IN PPM			
	ARSENIC	DDTR	DIELDRIN	CHLORDANE
California				
Bakersfield	7.1	0.36	0.07	0.78
Cropland	5.2	1.43	0.02	0.01
New Jersey				
Camden	11.2	1.36	< 0.01	0.36
Cropland ¹	6.8	0.14	0.02	< 0.01
Florida				
Miami	2.3	5.98	0.72	1.59
Cropland	0.8	0.85	0.08	0.36
Wisconsin				
Milwaukee	14.4	1.07	0.04	0.45
Cropland	3.8	0.02	0.01	0.01
Utah				
Salt Lake City	15.7	0.49	0.03	0.41
Cropland ²	4.8	0.20	0.01	0.02
Connecticut				
Waterbury	8.5	0.98	0.01	0.96
Cropland ³	10.0	0.59	0.01	0.01

¹ Average of results for New Jersey, Delaware, and Maryland.

² Average of results for Arizona, New Mexico, Nevada, and Utah.

³ Average of results for Maine, New Hampshire, Vermont, Massachusetts, Rhode Island, and Connecticut.

TABLE 3.—Geometric means and 95% confidence intervals for DDTR and arsenic residues in soil from six cities—1969

CITY	DDTR			ARSENIC		
	UPPER (PPM)	G.M. (PPM)	LOWER (PPM)	UPPER (PPM)	G.M. (PPM)	LOWER (PPM)
Bakersfield, Calif.	0.27	0.20	0.13	6.7	5.5	4.5
Camden, N.J.	0.62	0.37	0.21	10.0	7.6	5.8
Houston, Tex.	0.04	0.02	0.01	1.5	1.1	0.7
Manhattan, Kans.	0.18	0.09	0.04	8.2	6.2	4.7
Miami, Fla.	2.82	1.60	0.91	1.1	0.6	0.3
Milwaukee, Wis.	0.50	0.31	0.19	13.1	8.4	5.4
Salt Lake City, Utah	0.21	0.12	0.06	12.6	9.6	7.4
Waterbury, Conn.	0.18	0.08	0.04	7.8	6.1	4.8

See Appendix for chemical names of compounds discussed in this paper.

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APPENDIX

Chemical Names of Compounds Discussed in This Issue

ALDRIN	Not less than 95% of 1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4-endo-exo-5,8-dimethanonaphthalene
ARSENIC	As ₂ O ₃
AZINPHOSMETHYL	<i>o,o</i> -dimethyl <i>S</i> -(4-oxo-1,2,3-benzotriazin-3[4 <i>H</i>]-yl)methyl phosphorodithioate
BHC	1,2,3,4,5,6-hexachlorocyclohexane, mixed isomers
CARBOPHENTHION (TRITHION®)	<i>S</i> -[(<i>p</i> -chlorophenylthio)methyl] <i>o,o</i> -diethyl phosphorodithioate
CHLORDANE	1,2,4,5,6,7,8,8-octachloro-3a,4,7,7a-tetrahydro-4,7-methanoindane
γ-CHLORDANE	-----, gamma isomer
DDE	1,1-dichloro-2,2-bis(<i>p</i> -chlorophenyl) ethylene
DDT (including its isomers and dehydrochlorination products)	1,1,1-trichloro-2,2-bis(<i>p</i> -chlorophenyl)ethane; technical DDT consists of a mixture of the <i>p,p'</i> -isomer and the <i>o,p'</i> -isomer (in a ratio of about 3 or 4 to 1)
DEF®	<i>S,S,S</i> -tributyl phosphorotrithioate
DIAZINON	<i>o,o</i> -diethyl <i>o</i> -(2-isopropyl-4-methyl-6-pyrimidyl) phosphorothioate
DIELDRIN	Not less than 85% of 1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8a-octahydro-1,4-endo-exo-5,8-dimethano= naphthalene
ENDRIN	1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4-endo-endo-5,8-dimethanonaphthalene
FPN	<i>o</i> -ethyl <i>o</i> -(<i>p</i> -nitrophenyl) phenylphosphonothioate
ETHION	<i>o,o,o',o'</i> -tetraethyl <i>S,S'</i> -methylenebisphosphorodithioate
HEPTACHLOR	1,4,5,6,7,8,8-heptachloro-3a,4,7,7a-tetrahydro-4,7-methanoindene
HEPTACHLOR EPOXIDE	1,4,5,6,7,8,8-heptachloro-2,3-epoxy-3a,4,7,7a-tetrahydro-4,7-methanoindan
MALATHION	diethyl mercaptosuccinate, <i>S</i> -ester with <i>o,o</i> -dimethyl phosphorodithioate
MERCURY	Hg
METHYL PARATHION	<i>o,o</i> -dimethyl <i>o</i> - <i>p</i> -nitrophenyl phosphorothioate
PARATHION	<i>o, o</i> -diethyl <i>o</i> - <i>p</i> -nitrophenyl phosphorothioate
PHORATE	<i>o,o</i> -diethyl <i>S</i> -(ethylthio)methyl phosphorodithioate
SELENIUM	Se
TDE (DDD) (including its isomers and dehydrochlorination products)	1,1-dichloro-2,2-bis(<i>p</i> -chlorophenyl)ethane; technical TDE contains some <i>o,p'</i> -isomer also
FOXAPHENE	chlorinated camphene containing 67% to 69% chlorine

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BRIEFS

Residues of Chlorinated Hydrocarbon Pesticides in the Northern Quahog (Hard-Shell Clam), Mercenaria mercenaria—1968 and 1969

Ronald M. Check¹ and Manuel T. Canario, Jr.²

ABSTRACT

Samples of the northern quahog (hard-shell clam), *Mercenaria mercenaria*, were collected monthly, when possible, from September 1968 to September 1969 at five locations in Narragansett Bay, Rhode Island, and one location in nearby Mount Hope Bay. All 56 composite samples contained dieldrin at an average level of 0.040 ppm; p,p'-DDD was present in 3 samples at an average level of 0.026 ppm. Quahogs from upper reaches of Narragansett Bay contained higher levels of residues than samples from lower Bay areas.

Introduction

Preliminary examinations of the northern quahog (hard-shell clam), *Mercenaria mercenaria*, revealed the presence of chlorinated hydrocarbon pesticides in samples from Narragansett Bay, Rhode Island. As a result, a one-year survey was conducted with sampling at monthly intervals, when possible, from September 1968-September 1969, to determine the amounts of various chlorinated pesticides present in this species.

Sampling and Analytical Procedures

In September 1968, sampling stations were established at five sites in upper Narragansett Bay and one station in the southeastern corner of nearby Mount Hope Bay (Fig. 1). Shellfish samples were obtained by dredging. In the laboratory, clams were shucked and drained; a 300-g composite sample of meat, generally representing 14-18 clams from each location, was blended in a Waring Blender until homogenized. Blended samples were frozen until analyzed.

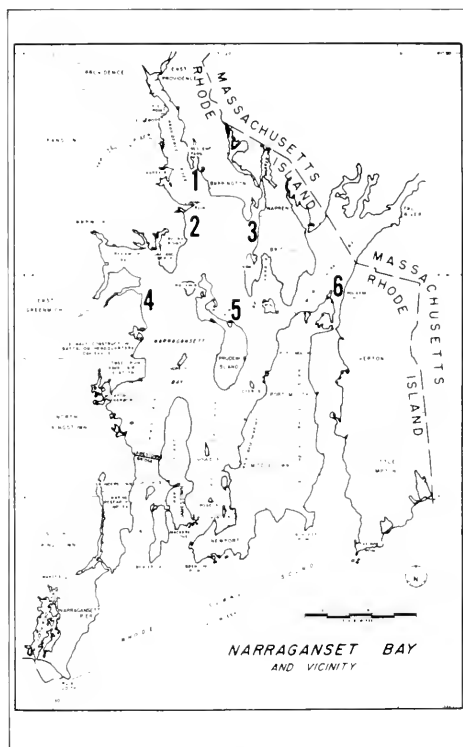


FIGURE 1.—Locations of shellfish sampling sites in Narragansett Bay and Mount Hope Bay, R. I., September 1968-September 1969

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The frozen 300-g samples were thawed, and 50-g subsamples were slurried with 100 ml of acetonitrile and allowed to stand overnight. A 25-g aliquot was filtered from the slurry and partitioned into petroleum ether according to the procedure outlined in the Pesticide Analytical Manual (1).

A 5-g aliquot was evaporated at room temperature to 2-3 ml and subjected to chemical cleanup by the sweep co-distillation method of Storherr *et al.* (2). The Florisil column step was included in the cleanup method. Purified sample extracts were concentrated to the equivalent of 1 g of sample per ml and analyzed by gas chromatography. The chromatograph was equipped with a Ni⁶³ electron capture detector and two 6-ft glass columns, 1/4 inch in diameter. One column contained 10% DC-200 and the other, 5% QF-1. These liquid phases were coated on Gas Chrom Q solid support. Nitrogen was used as the carrier gas. Injections onto the two columns provided tentative peak identification. All pesticide residues found were confirmed by thin layer chromatography using precoated aluminum oxide G plates. Standard solutions of the suspected pesticides were spotted beside samples and developed in *n*-heptane which had been redistilled over molten metallic sodium. Sample areas only were covered with aluminum foil, sprayed with Kovac's chromagenic reagent (3), and the plates were exposed to UV light. The foil was removed, and zones corresponding to standards were scraped from the plate and extracted from the aluminum oxide with 2 ml of 10% ethyl ether in petroleum ether. The 2-ml extracts were concentrated to 0.2 ml and reinjected through the two gas chromatographic columns for pesticide confirmation.

Further tests using chemical derivatization and partition coefficients were employed on initial samples to confirm the presence of dieldrin. The lower limits of detection for pesticides reported were 0.01 ppm for dieldrin and 0.02 ppm for *p,p'*-DDD, *p,p'*-DDE, and *p,p'*-DDT.

Occasional samples were fortified with known amounts of dieldrin, *o,p'*-DDE, *p,p'*-DDE, *p,p'*-DDD, and *p,p'*-DDT and subjected to the methodology described. Recoveries averaged 85%, 80%, 71%, 71%, and 73%, respectively. Ranges of recovery were 65%-100% for dieldrin and 50%-87% for *p,p'*-DDT. The recovery for *p,p'*-DDT and *p,p'*-DDE, while lower than that for dieldrin, was sufficient to show the presence of these compounds above the 0.02 ppm detection level used in this study. Several methods of analysis were evaluated initially including the traditional Florisil column cleanup procedure, but interfering peaks prevented positive identification and accurate quantitation of DDE and DDT if these compounds were present. Many of these peaks may have been residues of various polychlorinated

biphenyls, and it was necessary to eliminate them before an accurate pesticide profile could be obtained. The co-distillation method provided a means of separating unknown peaks from pesticides present in the samples. Future monitoring programs in conjunction with currently available methodology should yield more information about possible PCB residues.

Results and Discussion

Analyses for chlorinated pesticides in quahogs taken from the sampling stations indicated in Fig. 1, revealed the presence of dieldrin in all 56 samples and *p,p'*-DDD in 3 samples (Table 1). The absence of DDE and DDT in amounts greater than 0.02 ppm was unexpected in the samples analyzed. The average levels of pesticides found during the year were 0.040 ppm for dieldrin and 0.026 ppm for *p,p'*-DDD. In general, residues of dieldrin and DDD were higher in those samples from upper reaches of the Narragansett Bay than from the lower Bay areas.

TABLE 1.—Dieldrin and *p,p'*-DDD residues detected in northern quahogs, Narragansett and Mount Hope Bays—September 1968-September 1969

MONTH OF SAMPLING	DIELDRIN AND <i>p,p'</i> -DDD* RESIDUES IN PPM, WET-WEIGHT BASIS, DRAINED MEAT					
	STATION 1	STATION 2	STATION 3	STATION 4	STATION 5	STATION 6
1968						
Sept.	.060	.050	.010	.006	.021	.027
Oct.	.090	.056	.043	.063	.055	.048
1969						
Feb.	.070	.082	.043	.036	.037	.029
Mar.	.086	.066	.045	.062	.015	.026
Apr.	.060	.063	.060	.049	.031	.024
May	.066	.050	.042			
June	.047	.047	.049		.039	.013
July	.044	.039	.053	.039	.029	.016
		*.028	*.030	*.020		
Aug.	.030	.023	.028	.014	.013	.007
Sept	.030	.026	.029	.026	.025	.023

NOTE: Blank = sample not analyzed.

* *p,p'*-DDD residues were detected only in the July samples from Stations 2, 3, and 4.

See Appendix for chemical names of compounds discussed in this paper.

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APPENDIX

Chemical Names of Compounds Discussed in This Issue

ALDRIN	Not less than 95% of 1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4-endo-exo-5,8-dimethanonaphthalene
ARSENIC	As ₂ O ₃
ATRAZINE	2-chloro-4-ethylamino-6-isopropylamino-s-triazine
BHC	1,2,3,4,5,6-hexachlorocyclohexane, mixed isomers
CARPHENOTHION (TRITHION®)	S-[(<i>p</i> -chlorophenylthio)methyl] 0,0-diethyl phosphorodithioate
CHLORDANE	1,2,4,5,6,7,8,8-octachloro-3a,4,7,7a-tetrahydro-4,7-methanoindane
2,4-D	2,4-dichlorophenoxyacetic acid
DCPA (DACTHAL®)	dimethyl ester of tetrachloroterephthalic acid
DDE	1,1-dichloro-2,2-bis(<i>p</i> -chlorophenyl) ethylene
DDT (including its isomers and dehydrochlorination products)	1,1,1-trichloro-2,2-bis(<i>p</i> -chlorophenyl)ethane; technical DDT consists of a mixture of the <i>p,p'</i> -isomer and the <i>o,p'</i> -isomer (in a ratio of about 3 or 4 to 1)
DIAZINON	0,0-diethyl 0-(2-isopropyl-4-methyl-6-pyrimidyl) phosphorothioate
DCBP	4,4'-dichlorobenzophenone
DICOFOL (KELTHANE®)	4,4'-dichloro- <i>a</i> -(trichloromethyl)benzhydrol
DIELDRIN	Not less than 85% of 1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8a-octahydro-1,4-endo-exo-5,8-dimethano-naphthalene
DURSBAN®	0,0-diethyl 0-(3,5,6-trichloro-2-pyridyl) phosphorothioate
ENDOSULFAN (THIODAN®)	6,7,8,9,10,10-hexachloro-1,5,5a,6,9,9a-hexahydro-6,9-methano-2,4,3-benzodioxathepin 3-oxide
ENDRIN	1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4-endo-endo-5,8-dimethanonaphthalene
ETHION	0,0,0'-tetraethyl S,S'-methylenebisphosphorodithioate
HEPTACHLOR	1,4,5,6,7,8,8-heptachloro-3a,4,7,7a-tetrahydro-4,7-methanoindene
HEPTACHLOR EPOXIDE	1,4,5,6,7,8,8-heptachloro-2,3-epoxy-3a,4,7,7a-tetrahydro-4,7-methanoindan
HCB	hexachlorobenzene
ISODRIN	1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4-endo, endo-5,8-dimethanonaphthalene
LINDANE	1,2,3,4,5,6-hexachlorocyclohexane, 99% or more gamma isomer
MALATHION	diethyl mercaptosuccinate, S-ester with 0,0-dimethyl phosphorodithioate
MERCURY	Hg
METHOXYCHLOR	1,1,1-trichloro-2,2-bis(<i>p</i> -methoxyphenyl)ethane
METHYL MERCURY	R ₂ CH ₂ Hg
PARATHION	0, 0-diethyl 0- <i>p</i> -nitrophenyl phosphorothioate
PCNB	pentachloronitrobenzene
POLYCHLORINATED BIPHENYLS (PCB's)	Mixtures of chlorinated biphenyl compounds having various percentages of chlorination
TDE (DDD) (including its isomers and dehydrochlorination products)	1,1-dichloro-2,2-bis(<i>p</i> -chlorophenyl)ethane; technical TDE contains some <i>o,p'</i> -isomer also
TOXAPHENE	chlorinated camphene containing 67% to 69% chlorine
TRIFLURALIN	α,α,α -trifluoro-2,6-dinitro- <i>N,N</i> -dipropyl- <i>p</i> -toluidine

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RESIDUES IN FISH, WILDLIFE, AND ESTUARIES

*Residues of Organochlorine Pesticides, Polychlorinated Biphenyls, and Mercury and Autopsy Data for Bald Eagles, 1969 and 1970*¹

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ABSTRACT

Thirty-nine bald eagles found sick or dead in 13 States during 1969 and 1970 were analyzed for pesticide residues. Residues of DDE, dieldrin, polychlorinated biphenyls (PCB's), and mercury were detected in all bald eagle carcasses; DDD residues were detected in 38; DDT, heptachlor epoxide, and dichlorobenzophenone (DCBP) were detected less frequently. Six eagles contained possible lethal levels of dieldrin in the brain, and one contained a lethal concentration of DDE (385 ppm) in the brain together with 235 ppm of PCB's. Autopsy revealed that 18 bald eagles were illegally shot; other causes of death were impact injuries, electrocution, emaciation, and infectious diseases.

Introduction

Reichel *et al.* (4) presented pesticide residue data from bald eagles (*Haliaeetus leucocephalus*) collected in 1964 and 1965; a more recent report by Mulhern *et al.* (2) presented residue and autopsy data on bald eagles for 1966 through 1968. The purpose of this paper is to report the organochlorine pesticides, polychlorinated biphenyls (PCB's), and mercury residues and autopsy data for eagles collected in 1969 and 1970.

Sampling and Autopsy Procedures

As mentioned in previous report (2, 4), a systematic sampling procedure could not be used because of the relatively low populations of these birds and their protected status. Bald eagles found dead or moribund in the field are collected by Federal, State, and private cooperators, packed in dry ice, shipped air express to this laboratory, and stored intact in plastic bags at -25° C. The 13 States from which the 39 samples included in

this report were collected are shown in Table 1: 28 birds were collected in 1969 and 11 in 1970. Only field specimens that were not decomposed were analyzed.

Autopsies were performed on all specimens to determine possible causes of death. Tissues for histological study were fixed in 10% formalin, embedded in paraffin, sectioned, and stained with either hematoxylin and eosin, Ziehl-Neelsen acid-fast, periodic-acid Schiff (PAS), Giesmsa, Perl's Prussian blue, or the Van Kossa stain. The entire brain was removed and placed in an acetone-rinsed glass jar, and the remaining carcass (except for skin, feet, wings, liver, and gastrointestinal tract) was

TABLE 1.—Distribution of eagles collected, by State and year of death, 1969 and 1970

STATE	NUMBER OF EAGLES COLLECTED	
	1969	1970
Florida		1
Illinois	2	1
Iowa	1	
Maine	1	
Maryland		1
Michigan	4	3
Minnesota	6	1
Missouri	5	
North Dakota	3	
Ohio	2	
South Carolina		1
Wisconsin	4	2
Virginia		1
Total	28	11

¹ From the Bureau of Sport Fisheries and Wildlife, Patuxent Wildlife Research Center, Laurel, Md. 20810.

wrapped in aluminum foil. The samples were usually processed for residue analysis within 1 week after dissection.

Analytical Procedures

The remaining carcass was ground and homogenized in a Hobart food cutter.

ORGANOCHLORINE PESTICIDES

The entire brain and a 20-g aliquot of the carcass were then extracted and cleaned up separately, using the procedure described by Mulhern *et al.* (2). The method consisted of Soxhlet extraction and cleanup by acetonitrile partitioning and Florisil column procedures. The eluate was concentrated and divided in half. One half was analyzed for pesticides, and the other half was set aside for PCB analysis.

The portion to be analyzed for organochlorine pesticides was streaked on a thin layer (TL) plate, and the pesticides were separated into four fractions and extracted with hot benzene (1). The pesticides present would be found in the following fractions: Fraction I—dieldrin, lindane, heptachlor epoxide, endrin, dichlorobenzophenone (DCBP), methoxychlor, and dicofol; Fraction II—*p,p'*- and *o,p'*-DDD; Fraction III—*p,p'*- and *o,p'*-DDT and *o,p'*-DDE; Fraction IV—*p,p'*-DDE heptachlor, aldrin, and mirex. Each fraction was analyzed by gas chromatography (GC) on a 4% SE-30/6% QF-1 column and confirmed on a 3% QF-1 or a 3% XE-60 column; instruments and operating parameters are described in Table 2.

TABLE 2.—Gas chromatographic operating parameters using electron capture detection

COLUMNS, GLASS 6' × 1/4" O.D.			
Instrument	Hewlett-Packard 5750	Packard	Packard
Detector	nickel 63	tritium	tritium
Liquid phase	4% SE-30/6% QF-1	3% QF-1	3% XE-60
Support	Supelcoport	Gas Chrom Q	Gas Chrom Q
Mesh size	100/120	100/120	60/80
Flow rate	argon/methane at 60 ml/min	N ₂ at 40 ml/min	N ₂ at 100 ml/min
Temperature	190° C	175° C	175° C

In addition to the TL zonal separations and dual-column gas chromatographic confirmation, the residues in 10% of the samples were semiquantitatively confirmed by TLC (silica gel plate—2% ethyl ether in hexane developing solvent).

Positive identification of dieldrin residues in brains of certain eagles was accomplished with the use of an LKB gas chromatograph-mass spectrometer (GC-MS)

equipped with a 2% SE-30, 80/100 mesh column. Operating conditions were: flow rate—35 ml/min of helium; oven temperature—200° C; flash heater—220° C; separator—240° C; and ion source—290° C. The ionization potential was 70 ev, and accelerating voltage 3.5 kv. The sensitivity was approximately 50 ng. The mass spectra obtained from the samples were compared with spectra obtained from a dieldrin standard in respect to parent peak, C17 multiplet peaks, base peak, and fragmentation pattern.

Polychlorinated biphenyls (PCB's) with GC patterns most closely resembling Aroclor 1254® were found in all samples in Fraction IV and, in smaller amounts, in Fraction III. A previous study (2) showed that PCB compounds (Aroclor 1254® as reference) did not interfere in the quantitative determination of DDE unless the ratio of PCB's to DDE exceeded 2:1; samples were run on a 3% OV-17 column and quantities were measured by a disk integrator. Thus, a separate study was made to determine whether PCB compounds interfered with DDE when the DDE was quantitated by peak height using the 4% SE-30/6% QF-1 column. Standards of Aroclor 1248®, 1254®, and 1260® were each mixed with *p,p'*-DDE to obtain the following ratios of PCB's to DDE—1:1, 3:1, 5:1, 10:1, 15:1, and 20:1. The GC instrument parameters were the same as those used to measure DDE in the eagle samples. The results are given in Table 3.

TABLE 3.—The interference from PCB compounds in the determination of *p,p'*-DDE

<i>p,p'</i> -DDE ¹	RATIO OF PCB TO DDE					
	1:1	3:1	5:1	10:1	15:1	20:1
AROCLOR 1248®						
% Recovered	100	102	100	105	105	105
% Error	—	+2	—	+5	+5	+5
AROCLOR 1254®						
% Recovered	100	100	100	103	110	122
% Error	—	—	—	+3	+10	+22
AROCLOR 1260®						
% Recovered	100	100	103	95	92	90
% Error	—	—	+3	-5	-8	-10

¹ Quantitation by peak height.

Using peak height, the amount of DDE recovered remained within ± 5% of true value with all three Aroclors up to a 10:1 PCB's to DDE ratio; the error increased to +10% and -8% at a 15:1 ratio of PCB's to DDE using Aroclor 1254® and 1260®, respectively.

To determine the efficiency of the analytical procedure, a pesticide recovery study was run with duck wings; 20-g aliquots of duck wing homogenates, containing trace quantities of pesticides, were spiked to contain the following: 5.0 ppm DDE, 0.2 ppm DDD, 0.1 ppm DDT, 0.2 ppm dieldrin, and 5.0 ppm Aroclor 1254®. The average recoveries from three determinations were 77% DDE, 95% DDD, 79% DDT, 85% dieldrin, and 90% PCB's.

In reporting residues in eagle samples, no corrections were made for recoveries. The lower limit of detection was arbitrarily set at 0.05 ppm; quantities <0.05 ppm were reported as trace.

PCB's

The second half of the eluate was analyzed for PCB's using the semiquantitative determination by thin layer chromatography (TLC) as described by Mulhern *et al.* (3) with the following modification: the sample eluate was evaporated to dryness, followed by the addition of 2 ml of oxidizing agent (1.5 g of CrO₃ in 59 ml of HOAc and 1 ml of H₂O). This agent was prepared by saturating 59 ml HOAc with a portion of the 1.5 g CrO₃; the remaining CrO₃ crystals were dissolved with 1 ml H₂O. The lower detection limit was set at 1.0 ppm; a trace value for PCB's was considered to be <1.0 ppm.

TOTAL MERCURY

Total mercury was determined by cold vapor atomic absorption. A separate 2-g portion of the homogenized carcass was digested by refluxing with sulfuric and nitric acids and then oxidized with hydrogen peroxide. Hydroxylamine hydrochloride and stannous chloride were added to the digest to reduce mercury (II) ions to mercury metal. The sample was aerated, and the mercury in the air stream passing through a gas cell was measured by atomic absorption. The lower limit of detection was approximately 0.02 ppm in a 2-g sample.

Methyl mercuric hydroxide and mercuric chloride were added to eight 2-g portions of an eagle carcass, and from 90% to 107% of the mercury was recovered.

METHYL MERCURY

Methyl mercury in ground bald eagle carcasses was determined in duplicate by the method of Jensen (Naturvardsvérkets Specialanalytiska Laboratorium Lanktbrukshogskolan Uppsala, Sweden. *Unpublished method sent to Organization for Economic Cooperation and Development members, Sept. 1968*). In this procedure the lipids were extracted from a 1-g sample, wet tissue, by shaking with 30 ml methanol:ethyl ether, 3:1 mixture. After centrifuging and decanting, the remaining tissue was dried *in vacuo*, and 1 ml each of 0.5 M HBr and 0.5 M CuBr₂ followed by 10 ml benzene:tol-

uene (4:1) were added. The mixture was shaken thoroughly, centrifuged, and chilled until the water layer was frozen. The organic layer was then decanted into a tube for injection into a MicroTek MT220 gas chromatograph equipped with a Ni⁶³ detector on pulsed voltage operation. When chopped mallard muscle tissues spiked with 2 or 6 µg of mercury as methyl mercury hydroxide were analyzed by this procedure, 81% recoveries were obtained. Two column packings were used: one consisted of 15% Carbowax 20 M on 60/80 mesh Chromosorb G, acid washed, DCMS treated, at 185° C. Under these conditions injection of 1 ng of Hg as methyl mercury bromide produced a peak height of 10% of full-scale deflection. Some fouling of the detector was encountered with this column, and the degree of sensitivity was only fair. Thus, a second column packing, 5% Hi-Eff-10B (phenyl diethanolamine succinate) on 60/80 mesh Gas Chrom Q, was used. Operating temperatures of this column ranged from 145° to 160° C. Injection of approximately 0.4 ng Hg as methyl mercury bromide produced a peak height of 10% of full-scale deflection. This was considered to be the lower limit of reliable detection for the instrument and these operating conditions. This column packing apparently did not foul the detector, although continual clogging of the long exit tube on the detector was observed. This problem was partially alleviated by wrapping the exit tube with heating tape.

Results and Discussion

RESIDUES

The data for chlorinated pesticides, PCB's and total mercury residues found in bald eagles collected in 1969 and 1970 are summarized in Table 4. Results for eagle carcasses and brains are reported on a wet-weight basis. Medians rather than arithmetic means were used because the residue levels were asymmetrically distributed. All 39 carcass samples contained DDE, dieldrin, PCB's, and mercury; 38 contained *p,p'*-DDD.

The median value of DDE in the 1970 carcass samples was 2.6 times larger than in 1969, but both values were close to or within the median range of 4.9-16.6 ppm found for 1964-1968 (2, 4). For 1970, the median concentration of DDE in the brain was over 14 times greater than any median value observed since 1964; however, no trend can be established from these data because of the small number of samples collected, the wide range in DDE levels, and the absence of a systematic sampling procedure. Only two States, Minnesota and Wisconsin, were represented continuously from 1964-1970, often by one or two birds.

Significant correlations between residues of PCB's and DDE in the brain were obtained for both 1969 and 1970 samples; the correlations indicate that in the 28

samples statistically analyzed, there was an association between the levels of PCB's and DDE in the brain (Table 5). A more general inference may become possible with the analysis of future samples. The correlation between residues of PCB's and DDE in the brain could not be attributed to interference of PCB's with GC quantitation of DDE since the study of PCB interference in DDE quantitation showed only a 5% error in the amount of DDE recovered at the 10:1 level of PCB's to DDE and only one sample had a PCB's to DDE ratio greater than 10:1 (*viz* 11:1). The median value of PCB's to DDE in brains was 3.1 in 1969 and 2.2 in 1970.

The data for methyl mercury residues in 29 of the 39 carcasses are presented in Table 6; the 10 carcasses containing <1.0 ppm total mercury were not analyzed.

Seven samples, which were reanalyzed in duplicate, contained less than 65% methyl mercury, presumably due to the presence of significant amounts of inorganic mercury in the soft tissues or in the bone fragments mixed with the ground carcass. The equivalent ppm mercury present as methyl mercury ranged from 0.38 to 44.56 ppm Hg. Lethal levels of methyl mercury for bald eagles have not been established.

One finding of prime interest was that pesticide poisoning possibly accounted for the death of seven bald eagles. Residues in these birds were semiquantitatively confirmed by TLC. In addition, dieldrin residues in four of these birds were analyzed and confirmed by GC-MS. Six of the eagles contained possible lethal levels of dieldrin in the brain (Table 7); two of these birds were collected in 1969 and four, in 1970. The effects of the

TABLE 4.—Pesticide residues in bald eagles, 1969 and 1970

[T = <0.05 ppm]¹

COMPOUND	YEAR	RESIDUES IN PPM ¹					
		CARCASS			BRAIN		
		MEDIAN	RANGE	N ²	MEDIAN	RANGE	N ²
<i>p,p'</i> -DDE	1969	6.9	0.16-30	28	0.68	T-62	28
	1970	18	1.5-78	11	26	0.22-385	11
<i>p,p'</i> -DDD	1969	1.0	0.06-0.27	27	0.20	T-2.7	18
	1970	1.5	0.28-11	11	1.5	0.05-7.2	11
<i>p,p'</i> -DDT	1969	0.22	0.07-0.75	12	0.06	0.06	1
	1970	0.10	0.09-1.1	6	0.34	0.23-0.69	4
Dieldrin	1969	0.41	T-6.5	28	0.27	T-8.0	18
	1970	0.74	<0.10-18	11	2.0	0.23-11	10
Heptachlor epoxide	1969	0.06	T-0.35	22	0.04	0.003-0.06	6
	1970	0.10	T-0.41	9	0.36	0.06-1.0	7
Dichlorobenzophenone	1969	0.42	0.08-0.77	7	0.31	0.07-0.81	4
	1970	T	T	1	0.30	T-1.1	4
PCB Compounds	1969	10	T-150	28	2.5	T-65	28
	1970	20	4-200	11	46	T-230	11
Mercury	1969	1.5	0.52-43	28			
	1970	2.5	0.47-9.4	11			

NOTE: A total of 28 birds were collected in 1969 and 11 birds in 1970.

¹ Calculated on a wet-weight basis.

² Number of specimens that contained residues; the median is based on this number.

TABLE 5.—Association between PCB and DDE residues in bald eagles, 1969 and 1970

YEAR	CARCASS				BRAIN			
	PCB/DDE		N ¹	CORRELATION COEFFICIENT	PCB/DDE		N ¹	CORRELATION COEFFICIENT
	MEDIAN	RANGE			MEDIAN	RANGE		
1969	1.5	0.4-10	26	0.287	3.1	0.9-11	18	² 0.908
1970	1.3	0.5-3.3	11	0.451	2.2	0.6-7.5	10	³ 0.752

¹ Number of specimens that contained more than trace amounts of both PCB compounds and DDE.

² P = <0.01.

³ P = <0.05.

high levels of dieldrin in the brains of the two eagles that drowned may have caused the birds to fall into the water and drown. The dieldrin levels ranged from 4.6 to 11 ppm, wet-weight basis. Experimental and field data have shown that the lowest lethal brain residue for dieldrin is about 4 or 5 ppm (5). The seventh eagle, an adult female from Michigan, contained 385 ppm of DDE and 6 ppm DDD in the brain, together with 235 ppm of PCB's. The level of DDE alone is well within the lethal range (6), but the high level of PCB's suggests that these compounds may have also contributed to death. In this study and previous studies (2, 4) since 1964, with the exception of one eagle of unknown sex, all bald eagles found to be possibly poisoned by dieldrin or DDT metabolites have been females. Of the 153 eagles analyzed during 1964-1970, 15 (9.8%) possibly died of dieldrin poisoning.

AUTOPSY DATA

The autopsy results for the 39 bald eagles are summarized in Table 8. Illegal shooting remained the most frequent single cause of death among the bald eagles

TABLE 6.—Mercury found in 29 bald eagle carcasses, 1969 and 1970

TOTAL PPM Hg ¹	METHYL MERCURY EQUIVALENT PPM Hg	% OF TOTAL Hg FOUND AS METHYL MERCURY
1.01	0.68	67.3
1.02	0.96	94.1
1.12	1.01	90.2
1.13	1.09	96.5
1.17	0.98	83.8
1.26	0.76	60.3
1.31	0.90	68.7
1.42	0.92	64.8
1.49	1.28	85.9
1.53	1.41	92.2
1.53	0.38	24.8
1.70	0.80	47.0
1.72	1.36	79.1
1.77	1.75	98.9
2.07	1.74	84.0
2.18	1.45	66.5
2.20	1.88	85.4
2.50	1.99	79.6
2.74	2.44	89.0
2.96	0.48	16.2
3.01	2.85	94.7
3.58	1.56	43.6
4.42	2.05	46.4
5.65	4.68	82.8
5.83	3.98	68.3
8.32	6.71	80.6
9.40	2.72	28.9
11.00	7.60	69.1
43.00	44.56	103.6
MEDIAN		
2.07	1.45	79.6

¹ Of the 39 samples collected, 10 contained <1.0 ppm total mercury and, thus, were not analyzed for methyl mercury.

examined in this laboratory with 46% of the birds collected during 1969-1970 having been shot.

The four eagles dying from impact injuries, most frequently as the result of hitting a power line or tower, included one eagle from Virginia which had been struck by a private jet aircraft.

Three eagles were extremely emaciated; two of these were later found to have possible lethal levels of dieldrin in the brain (Table 7), and the third, a bird from Wisconsin, had multiple injuries from porcupine quills. Two quills were still embedded in the back of the eagle's oral cavity, suggesting that this eagle died from the effects of starvation and secondary bacterial infection (see below).

TABLE 7.—Data on six suspected cases of possible dieldrin poisoning and one suspected case of DDE poisoning, 1969 and 1970

STATE	YEAR	AGE ¹	SEX	RESIDUE IN BRAIN (PPM)	AUTOPSY FINDINGS
DIELDRIN					
Missouri	1969	Ad	F	≈ 4.6	Drowning, emaciation
Wisconsin	1969	Ad	F	≈ 6.5	Open
Illinois	1970	Im	F	4.6	Open
Michigan	1970	Ad	F	≈ 4.8	Drowning
Minnesota	1970	Im	F	≈ 5.9	Emaciated; shotgun pellets around tail
Maryland	1970	Ad	F	11	Open
DDE PCB					
Michigan	1970	Ad	F	385 235	Parasitic enteritis

¹ Ad = adult, Im = immature.

² Dieldrin analyzed and confirmed by gas chromatography-mass spectrometry.

Open = no diagnosis could be made on the basis of autopsy findings.

TABLE 8.—Probable causes of bald eagle mortality, 1969 and 1970

CAUSE OF DEATH	NUMBER OF EAGLES
Shot	18
Dieldrin ¹	6
DDE ¹	1
Impact	4
Electrocution	2
Emaciation	1
Nephrosis	1
Streptococcal infection	1
Avian cholera	1
Trapping injuries	1
Open	3
Total	39

¹ See Table 7 for details.

One eagle from Michigan had an old trapping injury which had become secondarily infected and resulted in a fibrinous pericarditis. A pure culture of a gram-positive rod, subsequently identified as a *Lactobacillus*, was isolated from the pericardium. This isolate of *Lactobacillus* was noninfectious to laboratory mice and domestic pigeons and probably should be regarded as a post-mortem contaminant.

Although *Pseudomonas aeruginosa* was isolated from the kidney and from an old infected bullet leg wound of an eagle from Wisconsin, the cause of death was a more recent gunshot wound which produced massive hemorrhage into the pericardial sac.

Pasteurella multocida, the causative organism of avian cholera, was isolated from three eagles. The first isolation was from an adult male from Ohio and apparently represents a frank case of avian cholera. The second isolation of *P. multocida* was from the heart of the emaciated eagle which had been injured by porcupine quills; the third isolation was from the liver of an eagle shot in Florida.

Several parasitic conditions were observed in these 39 eagles. Schizonts of *Leucocytozoon* were found in the hearts of an eagle from Maryland and one from Illinois; both eagles had possible lethal levels of dieldrin in the brain (Table 7). Another eagle from Michigan, subsequently found to have high levels of DDE and PCB's in the brain, had a severe enteritis caused by a large number of as yet unidentified flukes; although the enteritis was severe, it was not regarded as the cause of the eagle's death.

Conclusion

Since the bald eagle is located at the top of food chains and is the final recipient of environmental pollutants, the presence of PCB's, DDE, dieldrin, and mercury in all 39 samples from 13 States, demonstrates continued widespread environmental contamination by these compounds.

Acknowledgment

Appreciation is extended to J. O. Knisley and P. Jones who assisted in the autopsies and dissections of eagles, to L. T. Young who prepared sections for histological examination, and to the cooperators from many States and organizations who submitted eagles for analysis. Special acknowledgment is due to Dr. R. Shillinger and his staff (Animal Health Laboratories, Maryland Department of Agriculture, College Park, Md.) for the identification of the bacterial isolates and to the National Animal Disease Laboratory, USDA, Ames, Iowa, for identification of the *Lactobacillus* isolate.

See Appendix for chemical names of compounds discussed in this paper.

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DDT, DDE, and Polychlorinated Biphenyls in Biota From the Gulf of Mexico and Caribbean Sea—1971

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ABSTRACT

Residue levels of DDT, DDE, and PCB's were determined in various species of fish, shrimp, crabs, and other biota from the Gulf of Mexico and Caribbean Sea. Samples were collected from the Gulf during two Gulf-wide cruises in May and October 1971 and from part of the Caribbean Sea during the October cruise. DDT, DDE, and PCB's were found widely distributed in all biota; however, samples from coastal areas generally had higher levels than samples from the open waters.

Introduction

The occurrence of DDT and its metabolites and polychlorinated biphenyls (PCB's) in fish and wildlife is of current interest; however, data concerning the levels of these compounds in organisms from the open ocean are scarce (1-5).

During 1971 and 1972 about 50 scientists are participating in an intensive study program sponsored by the International Decade for Ocean Exploration (National Science Foundation) to identify problems related to oceanic environmental quality. This paper reports some of the results of this study, i.e., the concentrations of DDT, DDE, and PCB's in fish and other marine organisms collected in the Gulf of Mexico and the Caribbean Sea in May and October 1971. These "baseline concentrations" should be useful as comparison data to investigators working in estuaries around the Gulf of Mexico. Fig. 1 shows the locations where samples were collected.

The Gulf of Mexico receives runoff from approximately two-thirds of the United States and one-half of Mexico. This large amount of runoff with its high load of pollutants is swept generally westward and trapped in the western Gulf where waters remain possibly as long as 100 years. The primary flushing mechanism is exchange in the eastern Gulf with the loop current which passes quickly through the Yucatan Strait and out through the Straits of Florida. Because of the characteristics of this unique system, a buildup in concentration of man-made toxic materials is possible in the western Gulf, and baseline concentrations reported here together with future analyses should provide an early indication of this.

Sampling and Analytical Procedures

Samples were collected by the scientific party aboard cruises 71-A-5 (in May 1971) and 71-A-12 (in October 1971) of the R/V Alaminos, the oceanography vessel of Texas A&M University. Most samples were obtained using nets, but a few tuna and shark were caught by hook and line. Smaller samples were transferred immediately to glass mason jars with caps lined with aluminum foil and frozen until analysis. The jars and foil had been pre-washed with absolute ethanol which was free of any chlorinated hydrocarbons. Appropriate organs and muscle samples were taken from the larger fish, placed in mason jars, and frozen until analysis.

Small fish, shrimp, crabs, and other crustaceans were analyzed whole as composites of two or more fish or six crustaceans. Generally 50- to 100-g subsamples were taken from each composite for analysis. Subsamples of muscle tissue and organs analyzed generally weighed 50- to 100-g. The extraction and cleanup procedure used was that described in the "Pesticide Analytical Manual"

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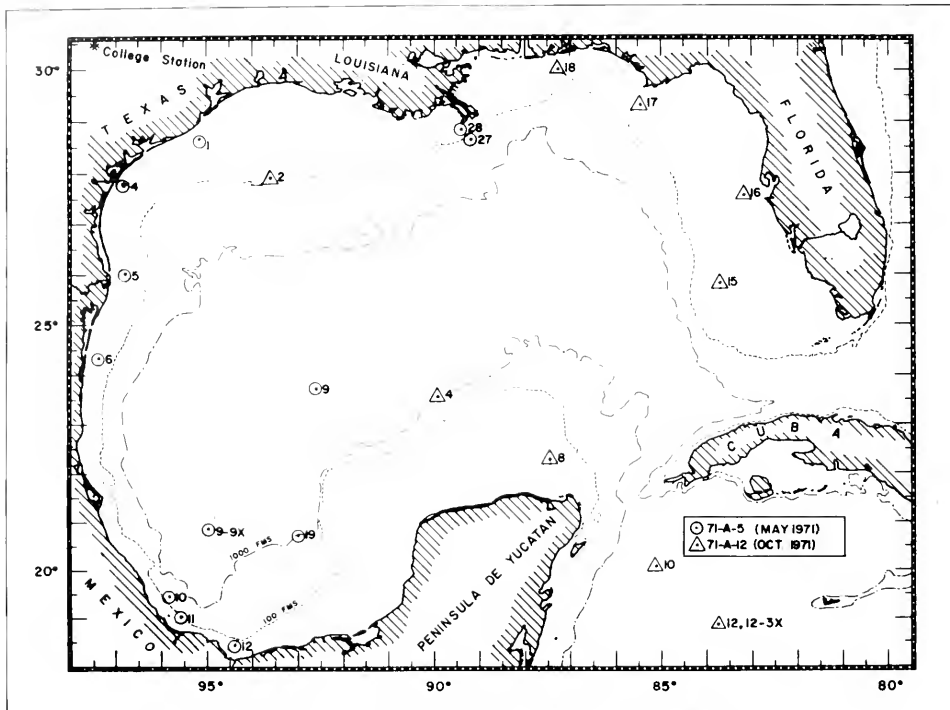


FIGURE 1.—Sampling stations in the Gulf of Mexico and Caribbean Sea—May and October 1971

of the U. S. Food and Drug Administration (6). The final residue extracts were adjusted to a suitable volume (between 2 and 10 ml) for gas chromatographic analysis. No attempt was made to concentrate the eluate to less than 2 ml, and not more than 10 μ l of extract was injected into the chromatographic column.

A Tracor gas chromatograph (Model MT 220) equipped with a ^{63}Ni electron capture detector and U-shaped glass columns, 6 ft x $\frac{1}{4}$ inch, o.d., and packed with 5% DC-200 on 80/100 mesh HP Chromosorb W was used. Columns packed with 5% OV-1 on 80/100 mesh HP Chromosorb W and a 6% mixture of OV-17 and QF-1 (in the ratio of 7:9) on 80/100 mesh HP Chromosorb W were also employed for further characterization. Nitrogen was used as the carrier gas at a flow rate of 60 cc/min. The injector, oven, and detector temperatures were 225° C, 200° C, and 275° C, respectively.

Identification of PCB's as commercial Aroclor® formulations was based on good matching of the sample peaks with those of standard Aroclor® mixtures. Quantification

was carried out when at least 50% of the peaks from a sample chromatogram matched peaks of the Aroclor® formulation; three different columns were used for matching the arrays of Aroclor®. In instances where the concentrations of PCB's were so high that they interfered with the quantification of DDT and DDE, separations of the PCB's from DDT and its metabolites were performed using a silica gel column (7). The presence of DDT and PCB's was further confirmed by the disappearance of the *p,p'*-DDT peak (and a corresponding increase in the *p,p'*-DDE peak) in the chromatograms of sample extract after alkaline alcoholysis treatment (8). The PCB peaks were not affected.

Percent recovery studies were performed using spiked liver and muscle samples. Recovery was 85% or better for all compounds identified in this study. (The group performing these analyses took part in a national and international cooperative analysis of chlorinated hydrocarbon insecticides and PCB's in marine samples, and their results were in excellent agreement with results from other laboratories.) The sensitivity of detection,

depending on the weight of the sample, ranged from 0.1 to 0.3 $\mu\text{g}/\text{kg}$ wet weight for DDT and DDE and 1 to 3 $\mu\text{g}/\text{kg}$ for PCB's. Results were not corrected for percent recovery.

Results and Discussion

Results of analysis of samples from the Gulf of Mexico and the Caribbean Sea are given in Tables 1 and 2. DDT, DDE, and the PCB's were detected in nearly all the samples analyzed, indicating that these compounds were widely distributed in the Gulf and Caribbean Sea; however, the levels were generally low, as may be expected in open ocean biota (5, 9). *P*, *p'*-DDD was not detected, and the analytical procedures excluded other organochlorine insecticides.

The levels of residues in marine organisms varied appreciably, but because the number of samples analyzed from each location was small, no firm conclusions can be made at this time. Certain general trends were evident, however, from the results of these analyses: (1) The samples from coastal areas, regardless of species, generally had higher levels of DDT, DDE and PCB's than samples from open waters. For example, samples obtained in May 1971 from Stations 27 and 28—two sites near the Mississippi Delta, a highly polluted area—showed relatively higher concentrations. (2) The ratios

of DDE to DDT varied widely between samples. A few samples from the coastal areas, i.e., shrimp collected at Stations 4, 6, 10, and 28 and other crustaceans from Station 27, showed rather high levels of DDT but relatively low levels of DDE (Table 1). Residue data from these specific samples may be fortuitous or may indicate that most of the DDT had not yet been metabolized. (3) In the livers obtained from larger fish (Table 2), concentrations of DDE were generally higher than DDT, possibly indicating the capability of this organ to metabolize and degrade DDT. (4) In individual fish (Table 2), the liver generally had the highest levels of DDT, DDE, and PCB's and muscle tissue the lowest; appreciable concentrations were also detected in the gonads and the digestive tract.

Acknowledgments

We gratefully acknowledge the National Science Foundation (International Decade of Ocean Exploration program) for financial support of this work, Mr. W. Dill for identification of organisms analyzed in this study, and Mr. Jerry Burke of the Food and Drug Administration and Dr. George Harvey of the Woods Hole Oceanographic Institute for intercalibration samples.

See Appendix for chemical names of compounds discussed in this paper.

TABLE 1.—DDT, DDE, and PCB residues in biota from the Gulf of Mexico and Caribbean Sea—May and October 1971

MONTH OF COLLECTION (1971)	SAMPLING STATION (SEE FIG. 1)	LOCATION	SAMPLE ¹	RESIDUES IN $\mu\text{G}/\text{KG}$, WET WEIGHT BASIS			
				<i>p,p'</i> -DDT	<i>p,p'</i> -DDE	TOTAL DDT	PCB's ²
May	1	Lat. 28°43.1' Long. 95°08.4'	Flounder (<i>Syacium ouanteri</i>)	84	10	94	32
			Squid	65	4.6	70	(3)
	4	Lat. 27°47.1' Long. 96°49.6'	Crabs (<i>Callinectes sp.</i>)	1.8	7.4	9	4 17
			Fish (Unidentified)	141	18	159	5 53
			Flounder (<i>Syacium papillosum</i>)	21	14	35	36
			Sea pansy (Order Pennatulacea)	128	161	289	4 850
	5	Lat. 26°02.8' Long. 96°48.5'	Shrimp (Family Penaeidae)	26	7.0	33	(3)
			Fish (<i>Paraques acuminatus</i>)	20	6.6	27	27
	6	Lat. 24°23.4' Long. 97°23.9'	Flounder (<i>Syacium papillosum</i>)	(5)	(5)	(5)	59
			Flounder (<i>Syacium ouanteri</i>)	3.6	3.3	7	34
Shrimp (Family Penaeidae)			134	18	152	(3)	

TABLE 1. —DDT, DDE, and PCB residues in biota from the Gulf of Mexico and Caribbean Sea—
May and October 1971—Continued

MONTH OF COLLECTION (1971)	SAMPLING STATION (SEE FIG. 1)	LOCATION	SAMPLE ¹	RESIDUES IN µG/KG, WET/WEIGHT BASIS			
				p,p'-DDT	p,p'-DDE	TOTAL DDT	PCB's ²
May	9-9X	Lat. 20°53.5' Long. 94°59.0'	Flying fish (Family Exocoetidae)	7.2	4.5	12	20
			Colonial tunicate	188	8.7	197	4 139
	10	Lat. 19°28.0' Long. 95°51.0'	Shrimp (Family Penaeidae)	154	11	165	(3)
			Fish (<i>Peristedion oracile</i>)	16	16	32	4 54
			Fish (<i>Serranus atrobranchus</i>)	33	8	41	(3)
			Fish (<i>Saurida brasiliensis</i>)	111	7	118	68
			Fish (<i>Halicutichthys aculeatus</i>)	78	4.8	83	4 64
			Fish (<i>Trichosetta vontralis</i>)	8.5	5.2	14	(3)
			Squirrel fish (<i>Holocentrus sp.</i>)	86	5.6	92	4 150
	27	Lat. 28°40.9' Long. 89°10.0'	Crustacean (<i>Aristacus antillensis</i>)	86	16	102	151
			Crustacean (<i>Nephropsis aculeate</i>)	151	6.0	157	22
			Fish (<i>Benthodesmus atlanticus</i>)	91	4.3	95	36
			Fish (Unidentified)	33	7.3	40	56
			Holothuroids	(5)	(5)	(5)	4 8
	28	Lat. 28°21.7' Long. 90°14.0'	Bat fish	12	65	77	527
Croakers (<i>Micropogon undulatus</i>)			(5)	11	11	50	
Shrimp (<i>Parapenaeus longirostris</i>)			304	34	338	167	
October	10	Lat. 20°05.4' Long. 85°07.7'	Flying fish (Family Exocoetidae)	5.1	4.6	10	26
	12-3X	Lat. 18°54.1' Long. 83°44.4'	Flying fish (Family Exocoetidae)	46	19	65	(3)
	15	Lat. 25°49' Long. 83°43'	Fish (<i>Synodus intermedius</i>)	4.5	10	15	14
	16	Lat. 27°34' Long. 83°10'	Flying fish (Family Exocoetidae)	18	12	30	65
17	Lat. 29°19.5' Long. 85°28.0'	Rock shrimp	1.0	2.1	3	(3)	
18	Lat. 30°00.5' Long. 87°17.5'	Rock shrimp	5.2	2.7	8	6	
		Squid	4.6	4.3	9	40	

¹ Represents 50- to 100-g subsamples from a composite of two or more whole fish or six whole crustaceans/other invertebrates.

² Calculated as Aroclor 1260® unless otherwise indicated.

³ Not estimated because of insufficient number of peaks on the chromatogram to characterize PCB formulations.

⁴ Calculated as Aroclor 1254®.

⁵ Not analyzed due to negligible concentrations of DDT and interference by PCB peaks.

TABLE 2.—DDT, DDE, and PCB residues in organs and muscle tissue of fish from the Gulf of Mexico and Caribbean Sea—May and October 1971

MONTH OF COLLECTION (1971)	SAMPLING STATION (SEE FIG. 1)	LOCATION	FISH OR COLLECTED	TISSUE OR ORGAN SAMPLED ¹	RESIDUES IN µG KG, WET-WEIGHT BASIS			
					p,p'-DDT	p,p'-DDE	TOTAL DDT	PCB'S ²
May	5	Lat. 26°02.8' Long. 96°48.5'	Shark (<i>Carcharinus falciformis</i>)	liver	200	499	699	1300
				9	Lat. 23°45.8' Long. 92°37.4'	White tip shark (<i>Pterolamiops longimanus</i>)	gut	53
	12	Lat. 18°27.5' Long. 94°24.0'	King mackerel (<i>Scomberomus cavalla</i>)	gonads			188	60
				muscle	15	15	30	32
				liver	406	1100	1506	536
				liver	72	51	123	83
	19	Lat. 20°44.0' Long. 92°50.0'	Parrot fish (<i>Holchoeres radiatus</i>)	gut	14	13	27	76
				gonads	44	31	75	58
				muscle	45	39	84	59
				liver	36	(³)	36	(⁴) 284
				liver	3.9	9.5	13	18
				gonads	34	6.1	40	16
October	2	Lat. 27°54.5' Long. 93°36.0'	Jack (<i>Thunnus atlanticus</i>)	liver	83	52	135	(⁵)
				gonads	10	19	29	43
	4	Lat. 23°33.4' Long. 89°54.5'	Trigger fish (<i>Canthidermis sufflamen</i>)	liver	37	7.4	44	22
				gut	56	3.7	60	9
	8	Lat. 22°16.4' Long. 87°27.3'	Tuna (<i>Euthynnus alleteratus</i>)	muscle	36	13	49	43
				gonads	8.2	36	44	36
	12	Lat. 18°54.1' Long. 83°44.4'	Barracuda (<i>Sohvaena barracuda</i>)	gonads	4.4	8.4	13	35
				liver	40	111	151	153
				muscle	4.0	4.2	8	9
				liver	16	26	42	57
				muscle	2.5	1.2	4	<1
				muscle	(³)	1.2	1	8
12	Fish (<i>Haemulon plumieri</i>)	liver	44	116	160	310		
		muscle	1.4	0.6	2	<1		
		muscle	1.7	0.9	3	<1		
		liver	44	116	160	310		

¹ Represents 50 to 100 g of organ or muscle samples.

² Calculated as Aroclor 1260[®] unless otherwise indicated.

³ Not analyzed due to negligible concentrations and interference by PCB peaks.

⁴ Calculated as Aroclor 1254[®].

⁵ Not estimated because of insufficient number of peaks on the chromatogram to characterize PCB formulations.

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Mercury Residues in Fish, 1969-1970—National Pesticide Monitoring Program¹

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ABSTRACT

As part of the fish monitoring program conducted by the Bureau of Sport Fisheries and Wildlife since 1967, composite fish samples collected during the fall of 1969 and 1970 were analyzed for mercury. Three composite samples, each of a different species and consisting of 3-5 adult fish, were collected at each of 50 monitoring stations in 1969; similarly, three composite samples and in most cases a replicate sample of one of the species were collected at each of 100 stations in 1970. Stations were located on major rivers and lakes throughout the United States. Total mercury residues equal to or exceeding the sensitivity level of 0.05 ppm were found in 129 of the 145 samples in 1969 and 373 of the 393 samples in 1970. Values ranged from <0.05 to 1.25 ppm in 1969 samples and from <0.05 to 1.80 ppm in 1970 samples. Analyses by two different laboratories of 40 selected samples from the 1970 collection gave comparable results. Analyses of 24 selected 1970 samples indicated that 90% or more of the mercury in fish was in the form of methyl mercury.

Introduction

A nationwide monitoring program to determine pesticide residue levels in fish has been conducted by the Bureau of Sport Fisheries and Wildlife each year since 1967. Composite fish samples collected from 50 stations during the first 2 years of the program (1967, 1968) were analyzed for whole body residues of 11 organochlorine insecticides; these results were reported in a previous issue of this Journal (3). In 1969, fish were collected from the same 50 stations, and analyses were expanded to also include lipids, polychlorinated biphenyls (PCB's), and mercury; results for lipids, organochlorine insecticides and PCB's were reported by Henderson, Inglis, and Johnson (4). In 1970, the number of sampling stations was increased to 100 and analyses again included organochlorine insecticides, PCB's, lipids, and mercury.

This report presents the data on mercury residues in fish collected from 50 stations during 1969 and from 100 stations during 1970. Common names of fishes as designated by the American Fisheries Society (1) are used throughout this report.

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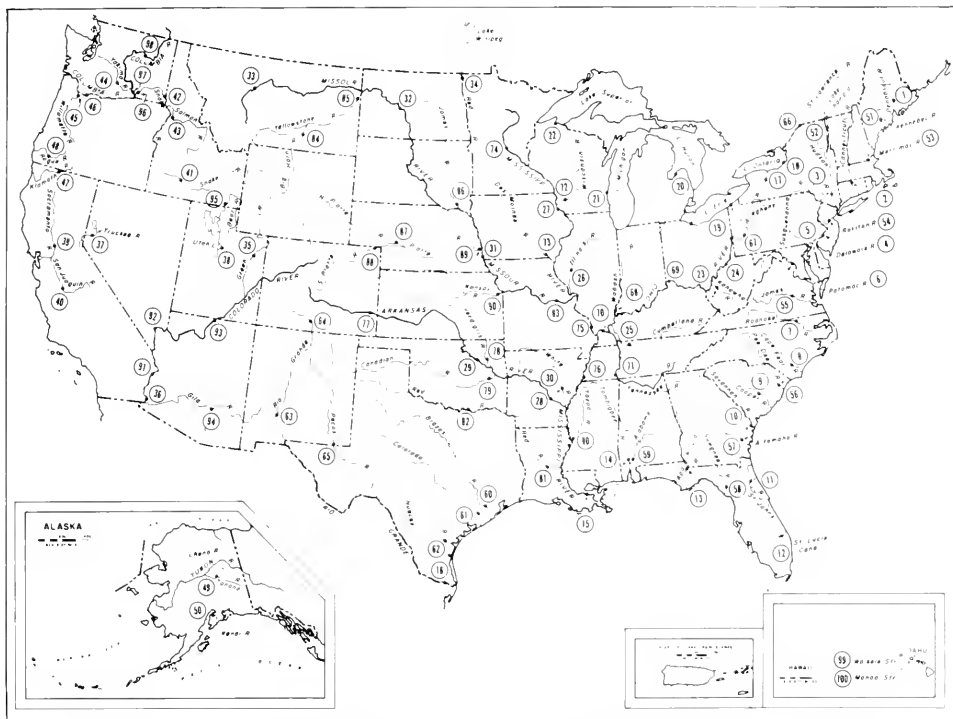


FIGURE 1.—Locations of fish sampling stations, National Pesticide Monitoring Program—1969 and 1970

Methods

FISH COLLECTIONS

The locations of sampling stations are shown in Fig. 1 and listed in Table 1. Fish were collected at Stations 1-50 in both 1969 and 1970, but at Stations 51-100 in 1970 only.

As in previous collections, three composite samples, each of a different species and consisting of 3-5 adult fish of uniform size for each species, were collected at each station. In 1970, a replicate composite sample of one of the three species was also collected in order to determine possible variation in residue levels in similar samples from the same station. A special effort has been made to collect the same species each year.

Fish collections were made by biologists from Fishery Services and other Divisions of the Bureau of Sport Fisheries and Wildlife with considerable assistance

from State conservation agencies. Fish were collected by various means including seines, gill nets, traps, hook and line, electrofishing, etc. The use of fish toxicants was not permitted. Collections were made once each year, usually in September, October, or November.

Each composite sample was wrapped in aluminum foil, frozen, packed in dry ice, and shipped to a laboratory for whole body residue analyses. Accompanying the shipment was a legend showing location, date collected, name of collector, collection method, species of fish, and the length, weight, and estimated age of each fish in each composite sample.

LABORATORY ANALYSES

A commercial laboratory (designated Laboratory C) conducted total mercury analyses on all of the 1969 and 1970 samples. The same laboratory conducted methyl mercury analyses on 24 selected samples from the 1970

fish collection. Subsamples of 40 selected homogenates from the 1970 collection were sent to another laboratory (designated Laboratory H) for total mercury analyses in order to cross-check or further confirm the results from Laboratory C. Analytical methods as furnished by participating laboratories are as follows:

Laboratory C—Analyses for Total Mercury

Each composite sample was thawed, cut into small pieces, and ground in a Hobart food chopper until homogenized. An aliquot sample was removed for mercury analysis. The method used is described in a report by the Joint Mercury Residues Panel (7), modified by atomic absorption spectrometry with the boat technique for 1969 samples and the cold vapor technique for 1970 samples. The procedures for digestion and analysis by the cold vapor technique were as follows:

A 10-g portion of the fish homogenate was transferred to a 1-liter round-bottom flask using little or no water; several glass beads were added. The flask was placed in a mantle and condensers and tap funnel inserted; 25 ml of a sulfuric-nitric acid mixture (4:1) was carefully introduced through the tap funnel (over approximately 10 minutes). The sample was heated slowly, so that the reaction would not become violent, for approximately $\frac{1}{2}$ to $\frac{3}{4}$ of an hour increasing the heat until full heat was reached. When necessary, small amounts of nitric acid were added to prevent carbonization. After refluxing for one hour, the sample was cooled to room temperature. When the digest was cool, the condenser and tap funnel were disconnected and the digest was transferred quantitatively with ice water to a 100-ml volumetric flask. The flask was stoppered and allowed to come to room temperature. The digest was made to volume, mixed, and analyzed on an atomic absorption spectrophotometer.

For atomic absorption analysis, 50 ml of a reducing solution was transferred into a 300-ml Erlenmeyer flask. The reducing solution was made up with 5.0 g of NaCl, 10 g of hydroxylamine hydrochloride, 20 g of stannous chloride in 20% H_2SO_4 , and diluted to 1 liter with 20% H_2SO_4 . Additional 20% sulfuric acid was added to make a total volume of 75 ml with the amount of sample to be added. The sample was added with a pipet by draining down the side of the flask, the flask stoppered rapidly, and then stirred vigorously with a magnetic stirrer (teflon bar) for exactly 30 seconds. The stirrer was shut off and the teflon bar allowed to stop; then the air pump was turned on to force mercury through the cell. To clean out the cell, the air flow was reversed between each sample analysis.

To establish a standard curve, an approximately 20-fold range of standards was used starting with 0.010 μg of mercury. The standard curve was plotted using peak height versus micrograms of mercury.

Analyses were performed on a Perkin-Elmer atomic absorption spectrometer, model 303, and a Perkin-Elmer recorder, model 304, with the following conditions:

Wavelength—2537 Angstroms (254 Setting—303).
Slit—3 mm, 20 Angstroms (5 Setting—303)
Range—UV
Source—Mercury hollow cathode lamp
Air—3 liters per minute
Recorder noise suppression—1, expansion—3 \times

The same digestion procedure as above was used for the boat method. Mercury was extracted from the acid solution into a dithizone-chloroform solution with two 5-ml extractions. Extractions were placed into a small tantalum boat and chloroform vaporized off. The boat was then placed directly into the flame, and the mercury vaporized.

Fourteen samples analyzed by both the boat and cold vapor techniques gave comparable results. However, the cold vapor technique was found to be more rapid and to have greater sensitivity with less interference. Recovery experiments showed an average recovery rate of 97%. The results were not corrected for recovery; sensitivity was reported as 0.05 ppm.

Laboratory C—Analyses for Methyl Mercury

Methyl mercury was determined by the procedure developed by Kamps and McMahon (8).

Laboratory H—Analyses for Total Mercury

The digestion method used was patterned after that reported by the Analytical Methods Committee (2); the flameless atomic absorption method was quite similar to that reported by others (6). A description of the methods follows:

For digestion, up to 10 g (wet weight) of sample was placed in a 250-ml flat-bottom flask with 3 boiling beads. This was placed under a condenser, and 10 ml of HNO_3 added through the condenser. When necessary, the sample was warmed slightly to start the reaction, but heating was discontinued when foaming started. When the initial reaction was finished, the sample had been dissolved. The flasks were allowed to cool slightly, and then 10 ml of 1:1 $H_2SO_4:HNO_3$ was added. The sample was heated slowly, without boiling, until refluxed, then refluxed for 3 hours. The flask was cooled until just warm to touch, and 10 ml of 30% H_2O_2 added in 1-ml increments, waiting between additions for foaming

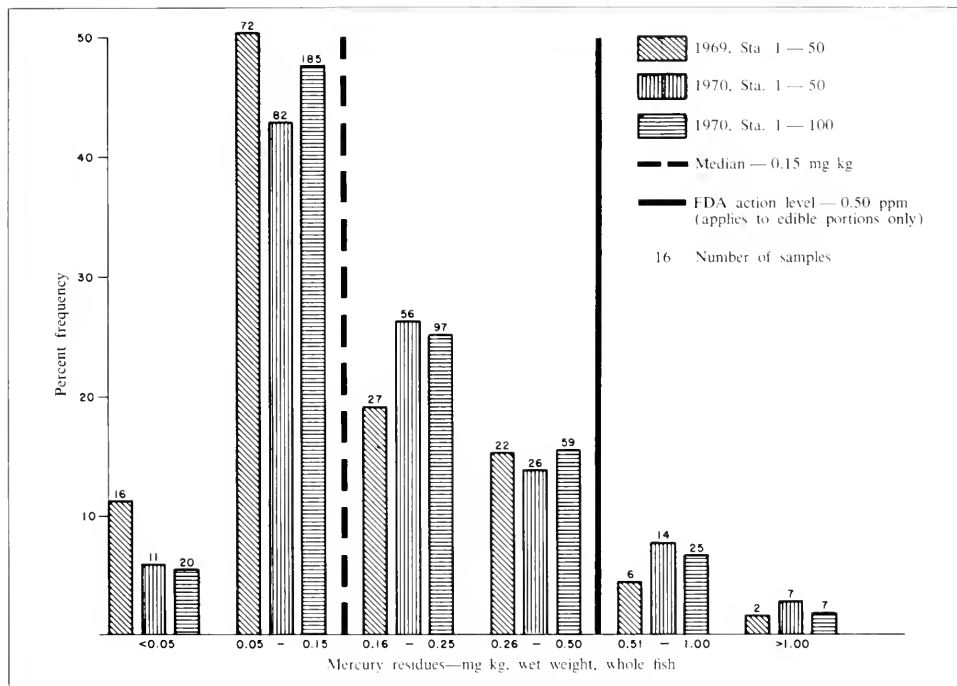


FIGURE 2.—Mercury residues in fish by frequency of occurrence—1969 and 1970

to stop. If no foaming occurred on the first addition, the sample was heated slightly until foaming began. After all 10 ml had been added, the sample was brought to a boil, and boiled for 1 hour (by then most of the brown NO_2 fumes had gone). The sample was chilled in ice water, and the condensers washed down with 50 ml of distilled water; the sample was removed from the condensers after the fat solidified. At this point, the sample was analyzed or covered and left in the refrigerator overnight.

For atomic absorption analysis, the cold digestion sample was filtered through glass wool into a 100-ml graduated cylinder, and the flask and filter were washed with enough distilled water to make the volume up to 100 ml. A 1-ml or some other suitable aliquot was removed and added to a gas bubbling flask containing sufficient 1N H_2SO_4 to make a total volume of 20 ml. Two milliliters of the reductant solution (100 ml of H_2SO_4 , 5 g of NaCl, 5.3 g of $\text{NH}_2\text{OH}\cdot\text{HCl}$, and 20 g of $\text{SnCl}_2\cdot 2\text{H}_2\text{O}$, made to 1 liter) was added, the flask closed, the chart

recorder started, the gas flow turned on, and the maximum pen deflection on chart noted. Then, the gas flow was turned off and a suitable quantity of standard (from 0.1 ppm Hg in 1N H_2SO_4) added. The gas was turned on and measured as before, and the nanograms of Hg were plotted against the peak area on linear graph paper.

Analyses were performed on a Jarrell-Ash Model 82 atomic absorption spectrometer equipped with a 25-cm quartz cell.

Results and Discussion

FISH COLLECTIONS

A total of 145 composite samples were collected from the 50 sampling stations in 1969 and 393 samples from the 100 stations in 1970. Fifty-five species of fish were represented in the collections. Some species such as carp, channel catfish, and largemouth bass were collected at many different stations. For example, samples of carp were obtained from 56, channel catfish from

34, and largemouth bass from 33 of the 100 stations in 1970. Many of the species, however, were collected at only 1 or 2 of the 100 stations. Generally, collectors were successful in obtaining the same species at a station in both 1969 and 1970 collections.

RESIDUE LEVELS IN FISH

Total mercury residue levels for each composite sample and average values at each station for both 1969 and 1970 collections are shown in Table 1. All values are reported as ppm (mg/kg), wet-weight, whole fish. Also shown in Table 1 are station locations and species of fish; the number and the average length and weight of fish in the composite samples collected in both years are included. The analyses for mercury in 1969 samples were conducted on subsamples of fish homogenates that had been prepared primarily for organochlorine insecticide analyses.

In order to make the 1969 and 1970 data more comparable, the 1970 average residue values were computed as follows: the replicate samples of the same species were averaged and this average used with the values for the other species to compute the final average. Values less than the sensitivity level of 0.05 ppm were considered to be zero in computing averages.

Mercury residues equal to or exceeding the sensitivity limit were found in 129 of the 145 samples in 1969 and 373 of the 393 samples in 1970. Values ranged from <0.05 to 1.25 ppm mercury in 1969 samples and from <0.05 to 1.80 ppm Hg in 1970 samples.

While high residue levels were found in fish from some waters in most drainage basins, these high levels appeared to occur more frequently in samples from Atlantic Coastal streams and the Columbia River system (Fig. 1). Lowest levels were found in fish from Alaskan streams, the Colorado River system, and Mississippi River tributaries in the Great Plains region.

Total mercury residues exceeding the Food and Drug Administration action level of 0.5 ppm, set for edible portions of the fish only, were found in 8 samples from 8 of the 50 stations in 1969 and in 32 samples from 21 of 100 stations in 1970. Average values exceeded 0.5 ppm at 3 stations in 1969 and 6 stations in 1970.

The frequency of occurrence of various residue levels is shown in Fig. 2. The range of values was comparable for 1969 and 1970 samples with the median residue level the same (0.15 ppm) for both sampling periods.

High mercury levels occurred much more often in some species of fish than in others. With few exceptions, the highest levels were found in predatory species near the top of the food chain such as bass, perch, and squawfish. Generally, the residue results for the two composite samples of the same species collected at each station in 1970 were in close agreement indicating no great variation in similar samples.

CONFIRMATORY ANALYSIS AND METHYL MERCURY

The results of the initial analyses by Laboratory C of 40 selected samples from the 1970 collections and the analyses of subsamples of the same homogenates by Laboratory H for total mercury are shown in Table 2. Also shown are results of analyses for methyl mercury reported as mg/kg of Hg on 24 selected samples by Laboratory C. The samples selected for methyl mercury analyses were among those having the highest total mercury residues.

The total mercury residue results for both laboratories are in very close agreement, although slightly different methods were used.

From a comparison of the total mercury and methyl mercury results, it appears that at least 90% and possibly more of the mercury in fish is methyl mercury.

The significance of the mercury residue results with respect to possible adverse effects on fish and wildlife is unknown at the present time. Research is underway to determine possible correlation of residue levels in tissues with possible effects such as mortality, reproduction, etc. From the standpoint of the use of fish for human food, it may be significant that residue levels in some samples exceeded the FDA action level of 0.5 ppm. It is also possible that such levels may have an adverse effect on fish-eating birds or other animals.

Monitoring for mercury and other residues in fish is a continuing program (5). Fish samples were collected at the same 100 sampling stations again in the fall of 1971 and are presently being analyzed for residues of mercury, other metals, organochlorine insecticides, and PCB's.

Acknowledgment

We greatly appreciate the help of the Bureau of Sport Fisheries and Wildlife biologists and fishery personnel in many of the States for collecting samples for the monitoring program. Without their assistance, such a program would not have been possible.

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TABLE 1.—Mercury residues in fish, 1969 and 1970

STATION NUMBER AND LOCATION	SPECIES	1970				1969				
		NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	
			LENGTH (INCHES)	WEIGHT (LB.)			LENGTH (INCHES)	WEIGHT (LB.)		
ATLANTIC COASTAL STREAMS										
#1	Stillwater River Old Town, Maine	White sucker	5	15.1	1.3	.43	5	15.0	1.5	.23
		White sucker (R)	5	15.3	1.3	.60				
		Yellow perch	5	7.9	0.2	.29				
		Chain pickerel	4	16.4	1.0	.64				
						Avg.				
#51	Kennebec River Hinckley, Maine	Yellow perch	5	9.3	0.4	1.20	5	7.7	0.2	.48
		Yellow perch (R)	5	9.0	0.4	.71				
		White perch	2	8.9	0.5	.38				
		Smallmouth bass	2	11.3	0.9	.64				
						Avg.				
#52	Lake Champlain Burlington, Vt.	Pumpkinseed	5	6.5	0.3	.38	5	16.2	1.1	.47
		Yellow perch	5	8.6	0.3	.27				
		Yellow perch (R)	5	9.4	0.5	.49				
		Chain pickerel	5	16.2	1.1	.47				
						Avg.				
#53	Merrimac River Lowell, Mass.	White sucker	5	12.0	0.6	.27	5	10.0	0.9	.14
		White sucker (R)	5	10.4	0.5	.28				
		Pumpkinseed	5	5.6	0.1	.15				
		Yellow perch	3	7.9	0.2	.33				
						Avg.				
#2	Connecticut River Windsor Locks, Conn.	White catfish	5	12.6	0.9	.22	5	8.8	0.4	.13
		White catfish (R)	5	12.9	1.0	.18				
		Yellow perch	2	10.5	0.6	.30				
		White perch	5	9.9	0.6	.51				
						Avg.				
#3	Hudson River Poughkeepsie, N. Y.	Goldfish	5	8.4	0.5	.07	5	9.2	0.5	.25
		Goldfish (R)	5	10.5	1.0	.11				
		Largemouth bass	4	12.8	1.3	.19				
		Pumpkinseed	5	6.2	0.2	.10				
						Avg.				
									Avg.	.36
										.19
										.20

TABLE 1.—Mercury residues in fish, 1969 and 1970—Continued

STATION NUMBER AND LOCATION	SPECIES	1970				1969				
		NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	
			LENGTH (INCHES)	WEIGHT (LB)			LENGTH (INCHES)	WEIGHT (LB)		
ATLANTIC COASTAL STREAMS—Continued										
#54	Raritan River Highland Park, N. J.	Golden shiner	5	5.9	0.1	.08				
		White perch	5	9.1	0.5	.28				
		White perch (R)	5	8.3	0.3	.29				
		Rock bass	3	6.9	0.2	.25				
					Avg.	.21				
#4	Delaware River Camden, N. J.	White sucker	5	14.7	1.41	.07	5	14.7	1.4	.08
		Brown bullhead	5	10.6	0.6	.07	5	11.7	0.9	.05
		White perch	5	9.8	0.6	.21	5	9.4	0.5	.22
		White perch (R)	5	9.7	0.6	.17				
					Avg.	.11			Avg.	
									.12	
#5	Susquehanna River Conowingo Dam, Md.	Carp	3	21.6	5.0	.07	4	20.0	3.5	.05
		Channel catfish	4	12.1	0.6	.05	5	13.6	0.9	.06
		Yellow perch	5	8.8	0.3	.09	5	9.1	0.4	.09
		Yellow perch (R)	5	7.9	0.2	.10				
					Avg.	.07			Avg.	
									.07	
#6	Potomac River Little Falls, Md.	Carp	3	13.8	1.3	.08	5	15.1	1.9	.10
		Carp (R)	3	14.1	1.1	.05				
		White sucker	2	11.5	0.7	<.05	5	12.7	0.8	.08
		Black crappie	5	8.7	0.3	.09				
					Avg.	.05	5	10.5	0.5	Avg.
									.09	
#55	James River Richmond, Va.	Redhorse sucker	5	10.8	0.7	.11				
		Channel catfish	4	18.8	2.5	.12				
		Channel catfish (R)	3	17.3	2.5	.20				
		Largemouth bass	5	10.0	0.7	.14				
					Avg.	.14				
#7	Roanoke River Roanoke Rapids, N. C.	Redhorse sucker	3	21.7	3.7	.08	5	19.0	2.8	.10
		Redhorse sucker (R)	3	20.0	3.2	.12				
		Brown bullhead	5	9.6	0.3	.21	5	9.6	0.4	.13
		Largemouth bass	3	12.0	1.0	.12	4	9.7	0.6	.09
					Avg.	.14			Avg.	
									.11	
#8	Cape Fear River Elizabethtown, N. C.	Gizzard shad	5	6.2	0.2	.25	5	12.0	0.6	.18
		Channel catfish	5				2	21.5	3.9	.35
		Brown bullhead	5	8.2	0.3	.25	5	10.8	0.6	.17
		Brown bullhead (R)	5	8.2	0.3	.25				
					Avg.	.60			Avg.	
									.23	
#56	Pee Dee River Dongola, S. C.	White catfish	5	7.4	0.2	.19				
		White catfish (R)	5	6.6	0.1	.20				
		Bluegill	5	5.2	0.1	.20				
		Largemouth bass	5	12.5	1.5	1.00				
					Avg.	.47				
#9	Cooper River Summerton, S. C.	Carp	2	17.5	3.5	.05				
		Spotted sucker					5	12.6	1.2	.13
		Bluegill	5	5.6	0.2	.20	5	6.8	0.2	.09
		Largemouth bass	4	11.3	0.6	.20	5	13.4	1.3	.14
					Avg.	.24			Avg.	
									.12	
#10	Savannah River Savannah, Ga.	Carp	2	26.0	6.9	.17	4	15.8	1.8	.36
		Bluegill	4	8.3	0.5	.56	3	7.3	0.3	.45
		Largemouth bass	3	11.3	1.0	1.80	4	10.5	0.8	1.00
		Largemouth bass (R)	3	9.0	0.5	1.40				
					Avg.	.78			Avg.	
									.60	
#57	Altamaha River Doctortown, Ga.	Spotted sucker	5	15.2	1.4	.25				
		Spotted sucker (R)	5	11.2	0.7	.11				
		Bluegill	5	8.2	0.4	.15				
		Largemouth bass	5	12.2	1.3	.51				
					Avg.	.28				
#11	St. Johns River Welaka, Fla.	Striped mullet	3	16.3	1.7	.07				
		Channel catfish	5	9.0	0.3	.06	5	11.0	0.9	<.05
		Channel catfish (R)	5	8.0	0.2	<.05				
		Redbreast sunfish					4	5.8	0.2	.07
					Avg.	.43	3	17.3	3.2	Avg.
									.05	

TABLE 1.—Mercury residues in fish, 1969 and 1970—Continued

STATION NUMBER AND LOCATION	SPECIES	1970				1969				
		No. of FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	No. of FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	
			LENGTH (INCHES)	WEIGHT (LB.)			LENGTH (INCHES)	WEIGHT (LB.)		
ATLANTIC COASTAL STREAMS—Continued										
#12	St. Lucie Canal Indiantown, Fla.	Channel catfish	2	18.5	2.2	.27	1	27.0	10.0	.23
		Channel catfish (R)	2	18.0	2.3	.15				
		Bluegill	5	5.6	0.2	.06				
		Largemouth bass	5	11.2	0.9	.28				
		Avg.				.18				
GULF COASTAL STREAMS										
#58	Suwanee River Old Town, Fla.	Spotted sucker	4	15.8	1.7	.13				
		Redbreast sunfish	4	7.0	0.5	.13				
		Redbreast sunfish (R)	4	7.0	0.5	.21				
		Largemouth bass	5	9.0	0.4	.37				
		Avg.				.22				
#13	Apalachicola River Jim Woodruff, Fla.	Spotted sucker	3	17.3	2.5	.10	5	17.4	2.3	.23
		Channel catfish	5	11.0	0.7	.11				
		Largemouth bass	5	10.8	0.7	.13				
		Largemouth bass (R)	5	9.4	0.5	.10				
		Avg.				.11				
#59	Alabama River Chrysler, Ala.	Striped mullet	4	15.0	1.3	<.05				
		Striped mullet (R)	4	15.0	1.2	<.05				
		Bluegill	5	6.6	0.3	.48				
		Largemouth bass	3	17.3	2.6	.60				
		Avg.				.36				
#14	Tombigbee River McIntosh, Ala.	Carp	5	17.0	3.7	.15	5	20.8	4.6	.36
		Striped mullet	4	15.8	1.6	.17				
		Striped mullet (R)	4	15.8	1.6	.35				
		Largemouth bass	5	13.0	1.2	.92				
		Avg.				.44				
#15	Mississippi River Luling, La.	Carp	4	15.3	2.1	<.05	5	13.4	1.7	.11
		Carp (R)	4	15.3	2.4	.05				
		Freshwater drum	3	12.0	0.7	.30				
		Striped mullet	5	13.8	1.0	.10				
		Channel catfish				.14				
Avg.				.14						
#60	Brazos River Richmond, Tex.	Smallmouth buffalo	3	17.2	3.2	.06				
		Channel catfish	3	20.9	3.4	.10				
		Channel catfish (R)	3	17.1	1.8	.08				
		Longnose gar	5	25.1	1.5	.24				
		Avg.				.13				
#61	Colorado River Wharton, Tex.	River carpsucker	3	13.3	1.5	.15				
		River carpsucker (R)	3	12.6	1.5	.19				
		Channel catfish	3	13.7	1.2	.18				
		Spotted bass	3	8.4	0.5	.14				
		Avg.				.16				
#62	Nueces River Mathis, Tex.	Gizzard shad	5	11.4	0.5	<.05				
		Gizzard shad (R)	5	10.0	0.4	.06				
		Channel catfish	4	12.9	0.7	.06				
		Largemouth bass	5	10.9	0.9	.40				
		Avg.				.17				
#16	Rio Grande Brownsville, Tex.	Gizzard shad	5	10.7	0.4	.08	5	11.4	0.6	<.05
		Gizzard shad (R)	5	12.1	0.7	.12				
		Channel catfish	2	16.8	1.5	.17				
		Blue catfish	2	16.2	1.3	<.05				
		Avg.				.09				
#63	Rio Grande Elephant Butte Reservoir, N. Mex.	Channel catfish	2	23.9	5.9	.35				
		Bluegill	4	4.6	0.1	.10				
		Bluegill (R)	3	4.7	0.1	.13				
		Largemouth bass	2	18.6	4.5	.52				
		Avg.				.33				

TABLE 1.—Mercury residues in fish, 1969 and 1970—Continued

STATION NUMBER AND LOCATION	SPECIES	1970				1969					
		NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹		
			LENGTH (INCHES)	WEIGHT (LB)			LENGTH (INCHES)	WEIGHT (LB)			
GULF COASTAL STREAMS—Continued											
#64	Rio Grande	Carp	5	17.2	2.4	<.05					
	Alamosa,	White sucker	5	12.7	0.9	.16					
	Colo.	White sucker (R)	5	13.4	1.0	.28					
		Brown trout	3	10.8	0.5	.18					
					Avg.	.13					
#65	Pecos River	Gizzard shad	5	12.3	0.8	.05					
	Red Bluff Lake,	Gizzard shad (R)	5	12.4	0.8	.05					
	Tex.	Channel catfish	5	15.8	1.4	.42					
		Largemouth bass	5	10.6	0.7	.06					
					Avg.	.18					
GREAT LAKES DRAINAGE											
#17	Genessee River	White sucker	4	14.0	1.2	.15	5	15.1	1.5	.13	
	Scottsville,	Redhorse sucker (R)	4	13.2	0.9	.19					
	N. Y.	Rock bass	4	8.1	0.5	.39	5	7.2	0.3	.22	
		Walleye					2	17.2	1.6	.25	
		Northern pike	4	13.7	0.7	.17					
					Avg.	.24				Avg.	.20
#66	St. Lawrence River	White sucker	3	17.2	1.5	.22					
	Massena, N. Y.	Yellow perch	5	7.0	0.2	.20					
		Yellow perch (R)	5	8.3	0.3	.18					
		Northern pike	4	20.6	2.1	.39					
					Avg.	.27					
#18	Lake Ontario	Yellow perch	5	8.5	0.4	.86	4	10.4	0.6	.48	
	Port Ontario,	Yellow perch (R)	5	8.4	0.4	1.00					
	N. Y.	White perch	5	8.3	0.4	1.30	5	9.5	0.5	.43	
		Rock bass	5	6.5	0.2	.30	3	8.6	0.6	.65	
					Avg.	.84				Avg.	.52
#19	Lake Erie	White sucker	5	18.0	2.7	.31	3	14.8	1.5	.10	
	Erie, Pa.	Freshwater drum	5	14.1	1.3	.43	5	13.5	1.1	.15	
		Yellow perch	5	9.7	0.4	.23	5	9.4	0.4	.13	
		Yellow perch (R)	5	8.9	0.3	.15					
					Avg.	.31				Avg.	.13
#20	Lake Huron	Carp	5	19.6	4.0	.07	5	16.3	2.1	<.05	
	Bay Port,	Channel catfish	5	16.2	1.4	.07	5	15.9	1.5	.13	
	Mich.	Yellow perch	5	9.1	0.3	.08	5	9.9	0.5	.09	
		Yellow perch (R)	5	9.1	0.3	.05					
					Avg.	.07				Avg.	.07
#21	Lake Michigan	Bloater	5	11.2	0.6	.09	5	12.0	0.8	.09	
	Sheboygan,	Bloater (R)	5	9.4	1.9	.10					
	Wis.	Yellow perch	5	11.0	0.6	.07	5	10.3	0.6	.27	
					Avg.	.09				Avg.	.18
#22	Lake Superior	Bloater	5	10.3	0.3	.15	5	11.2	0.4	.16	
	Bayfield,	Lake whitefish	5	17.6	1.7	.08	5	16.1	1.2	<.05	
	Wis.	Lake whitefish (R)	5	18.1	1.9	.06					
		Lake trout	5	23.4	4.0	.29	4	22.0	3.0	.14	
					Avg.	.17				Avg.	.10
MISSISSIPPI RIVER SYSTEM											
#67	Allegheny River	Carp	5	14.8	2.0	.11					
	Natrona,	Carp (R)	5	14.8	2.0	.05					
	Pa.	Bluegill	5	5.5	0.2	.11					
		Walleye	5	12.7	0.6	.18					
					Avg.	.12					
#23	Kanawha River	Carp	5	8.9	0.4	.08	4	8.6	0.4	<.05	
	Winfield,	Brown bullhead	4	6.4	0.2	<.05	4	11.9	0.8	<.05	
	W. Va.	Brown bullhead (R)	4	6.5	0.2	<.05					
		White crappie	5	6.8	0.2	.11	5	7.6	0.2	<.05	
					Avg.	.06				Avg.	<.05

TABLE 1.—Mercury residues in fish, 1969 and 1970—Continued

STATION NUMBER AND LOCATION	SPECIES	1970				1969				
		NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	
			LENGTH (INCHES)	WEIGHT (LB)			LENGTH (INCHES)	WEIGHT (LB)		
MISSISSIPPI RIVER SYSTEM—Continued										
#68	Wabash River New Harmony, Ind.	Carp	5	16.8	2.3	.25				
		Channel catfish	3	12.4	0.7	.15				
		White crappie	5	8.6	0.3	.16				
		White crappie (R)	5	7.6	0.2	.15				
		Avg.				.19				
#24	Ohio River Marietta, Ohio	Carp	2	15.1	2.4	.24	4	10.1	1.6	.22
		Redhorse sucker	4	11.7	0.6	.07	10	7.9	0.04	.20
		Channel catfish	4	14.8	0.8	.83	1	15.7	1.3	.39
		Channel catfish (R)	4	14.9	0.9	.68				
		Largemouth bass					5	11.3	0.7	.50
Avg.				.36				.33		
#69	Ohio River Cincinnati, Ohio	Carp	3	18.5	3.1	.20				
		Carp (R)	3	18.6	3.3	.09				
		White crappie	5	8.6	0.3	.15				
		Sauger	2	13.9	0.9	.30				
		Avg.				.20				
#70	Ohio River Metropolis, Ill.	Carp	5	15.0	1.8	.14				
		Channel catfish	5	11.5	0.5	.11				
		White crappie	5	9.9	0.5	.43				
		White crappie (R)	5	10.0	0.4	.37				
		Avg.				.22				
#25	Cumberland River Clarksville, Tenn.	Carp	4	13.5	1.4	.11	5	11.8	0.8	.09
		Bluegill	5	5.8	0.1	.05	5	6.2	0.1	.11
		Bluegill (R)	5	5.8	0.1	.05				
		Largemouth bass	2	11.0	0.8	.15	5	11.8	0.8	.27
		Avg.				.10				.16
#71	Tennessee River Savannah, Tenn.	Carp	5	16.0	1.9	.40				
		Channel catfish	4	12.8	0.6	.28				
		Channel catfish (R)	4	13.0	0.6	.27				
		Largemouth bass	5	15.2	2.2	.67				
		Avg.				.45				
#72	Wisconsin River Woodman, Wis.	Carp	3	18.2	3.3	.37				
		Carp (R)	2	19.3	4.0	.42				
		Channel catfish	3	16.7	2.1	.11				
		Smallmouth bass	3	11.7	1.0	.60				
		Avg.				.37				
#73	Des Moines River Keosauqua, Iowa	Carp	5	12.8	1.3	.07				
		Carp (R)	5	12.1	0.9	.11				
		Channel catfish	2	10.3	0.4	.07				
		Walleye	3	12.6	0.9	.24				
		Avg.				.17				
#26	Illinois River Beardstown, Ill.	Carp	5	14.2	1.4	.10	5	15.3	1.9	.09
		Carp (R)	5	16.3	2.1	.10				
		Bigmouth buffalo	3	18.6	3.9	.08	5	16.7	2.7	.07
		White crappie	5	7.3	0.2	.21	5	8.9	0.4	.13
		Avg.				.13				.10
#74	Mississippi River Little Falls, Minn.	White sucker	4	13.5	1.2	.21				
		White sucker (R)	4	13.8	1.2	.55				
		Yellow bullhead	2	10.2	0.8	.68				
		Northern pike	5	14.3	0.8	.37				
		Avg.				.48				
#27	Mississippi River Gutenberg, Iowa	Carp	4	18.8	2.9	.11	5	13.9	1.4	.10
		Bluegill	4	8.3	0.4	.20	5	7.1	0.4	.13
		Bluegill (R)	4	8.0	0.4	.12				
		Largemouth bass	3	15.3	2.1	.33	5	12.0	1.0	.25
		Avg.				.20				.16
#75	Mississippi River Cape Girardeau, Mo.	Carp	3	18.2	3.0	.20				
		Channel catfish	5	15.4	1.3	.11				
		Channel catfish (R)	5	15.7	1.2	<.05				
		White crappie	3	10.7	0.7	.15				
		Avg.				.14				

TABLE 1.—Mercury residues in fish, 1969 and 1970—Continued

STATION NUMBER AND LOCATION	SPECIES	1970				1969				
		NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	
			LENGTH (INCHES)	WEIGHT (LB.)			LENGTH (INCHES)	WEIGHT (LB.)		
MISSISSIPPI RIVER SYSTEM—Continued										
#76	Mississippi River Memphis, Tenn.	Carp	2	19.0	4.0	.17				
		Carp sucker	2	16.5	2.4	.46				
		Carp sucker (R)	2	17.0	2.9	.14				
		Freshwater drum	2	20.0	3.9	.15				
						Avg.	.21			
#28	Arkansas River Pine Bluff, Ark.	Carp	2	25.0	8.9	.09	3	21.0	3.8	.14
		Smallmouth buffalo	2	16.5	2.9	.11	4	16.3	2.5	.08
		Smallmouth buffalo (R)	2	19.5	4.8	.13				
		Flathead catfish	2	23.5	5.5	.22	2	21.0	4.6	.15
						Avg.	.14			Avg.
#29	Arkansas River Keystone Reser- voir, Okla.	Carp	5	13.4	1.2	.06	5	14.5	1.5	.08
		Carp (R)	5	13.9	1.3	.10				
		Bluegill	4	5.4	0.1	.13	5	6.2	0.2	<.05
		Largemouth bass	3	14.3	1.7	.22	5	15.0	2.5	.14
						Avg.	.14			Avg.
#77	Arkansas River John Martin Reservoir, Colo.	Carp	5	14.7	1.4	.05				
		Carp (R)	5	12.9	1.1	.06				
		Channel catfish	3	9.2	0.3	.05				
		Black bullhead	3	8.5	0.4	.07				
						Avg.	.06			
#78	Verdegris River Oologah, Okla.	Carp	5	14.8	1.6	.13				
		Carp (R)	5	15.0	1.7	.05				
		Bluegill	5	4.7	0.1	.07				
		Largemouth bass	3	14.3	2.3	.44				
						Avg.	.20			
#79	Canadian River Eufaula, Okla.	Carp	5	13.6	1.1	.06				
		Carp (R)	5	13.2	1.2	.21				
		Bluegill	4	5.5	0.1	.07				
		Largemouth bass	2	11.0	0.8	.10				
						Avg.	.10			
#30	White River DeValls Bluff, Ark.	Carp	2	18.0	2.9	.14				
		Bigmouth buffalo	2	17.0	2.6	.35	3	15.7	2.0	.22
		Bigmouth buffalo (R)	2	21.0	4.8	.25				
		Channel catfish	2	15.5	1.4	.12	4	14.5	0.8	.13
						Avg.	.19			Avg.
#80	Yazoo River Redwood, Miss.	Carp	2	20.0	4.5	.19				
		Smallmouth buffalo	2	16.5	3.0	.16				
		Smallmouth buffalo (R)	2	15.5	2.3	.10				
		Gizzard shad	5	5.8	0.1	.15				
						Avg.	.16			
#81	Red River Alexandria, La.	Smallmouth buffalo	2	21.0	5.8	.10				
		Smallmouth buffalo (R)	2	20.0	5.2	.10				
		Freshwater drum	3	13.7	1.1	.24				
		White catfish	3	13.0	0.6	.21				
						Avg.	.18			
#82	Red River Lake Texoma, Okla.	Carp	5	20.5	4.5	.13				
		Carp (R)	5	20.2	4.3	.14				
		Bluegill	4	6.6	0.4	<.05				
		Largemouth bass	4	13.2	0.9	.15				
						Avg.	.10			
#83	Missouri River Hermann, Mo.	Carp	2	19.4	3.7	<.05				
		Carp (R)	2	18.8	2.9	.08				
		Bigmouth buffalo	2	17.9	3.7	.14				
		Channel catfish	3	18.3	2.0	.15				
						Avg.	.11			
#31	Missouri River Nebraska City, Nebr.	Carp	3	17.1	2.4	.06	5	13.6	1.2	<.05
		Carp (R)	3	16.9	2.2	.07				
		Goldeye	4	13.0	0.9	.20	5	12.5	0.8	.07
		Channel catfish					5	13.0	0.8	<.05
						Avg.	.14			Avg.

TABLE 1.—Mercury residues in fish, 1969 and 1970—Continued

STATION NUMBER AND LOCATION	SPECIES	1970				1969				
		NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	
			LENGTH (INCHES)	WEIGHT (LB)			LENGTH (INCHES)	WEIGHT (LB)		
MISSISSIPPI RIVER SYSTEM—Continued										
#32	Missouri River Garrison Dam, N. Dak.	Carp	3	17.2	2.2	.11	2	15.2	1.6	.14
		Goldeye	5	12.4	0.5	.24				
		Goldeye (R)	5	11.9	0.5	.17				
		Walleye	3	15.8	1.3	.20				
		Avg.				.17				
									Avg. .19	
#33	Missouri River Great Falls, Mont.	Goldeye	5	11.2	0.5	.20	5	12.9	0.5	.14
		Goldeye (R)	5	11.2	0.5	.13				
		Redhorse sucker	5	15.9	1.7	.16				
		Sauger	5	12.5	0.7	.24				
		Avg.				.19				
									Avg. .14	
#84	Big Horn River Hardin, Mont.	Carp	5	15.5	2.4	.35				
		Goldeye	5	10.6	0.4	.23				
		Goldeye (R)	5	10.9	0.5	.25				
		Channel catfish	5	19.1	3.7	.18				
		Avg.				.26				
#85	Yellowstone River Sidney, Mont.	Carp	5	11.9	1.0	.11				
		Goldeye	5	11.3	0.5	.15				
		Goldeye (R)	5	10.3	0.4	.15				
		Channel catfish	5	9.1	0.4	.08				
		Avg.				.11				
#86	James River Olivet, S. Dak.	Carp	3	15.8	1.0	.13				
		Carp (R)	3	15.3	0.9	.13				
		Goldeye	5	11.0	0.8	.22				
		Freshwater drum	4	9.0	0.3	.08				
		Avg.				.14				
#87	North Platte River Lake McConaughy, Nebr.	Carp	3	15.8	1.2	.14				
		Carp (R)	3	15.9	1.2	.10				
		Channel catfish	3	16.5	1.0	.12				
		Walleye	5	16.2	1.8	.09				
		Rainbow trout	3	19.6	3.2	.11				
		Avg.				.11				
#88	South Platte River Brule, Nebr.	Carp	5	5.6	0.1	.08				
		White sucker	5	12.0	0.7	.11				
		White sucker (R)	5	10.2	0.5	.15				
		Green sunfish	3	4.8	0.1	.10				
		Avg.				.10				
#89	Platte River Louisville, Nebr.	Carp	3	19.7	3.7	.24				
		Carp (R)	3	12.2	0.8	.11				
		Channel catfish	5	12.6	0.6	.12				
		White crappie	3	7.6	0.3	.13				
		Avg.				.14				
#90	Kansas River Bonner Springs, Kans.	Carp	5	14.8	1.9	.10				
		Carp (R)	5	15.8	2.8	.13				
		Gizzard shad	5	7.2	0.2	.08				
		Freshwater drum	5	9.9	0.7	.33				
		Avg.				.18				
HUDSON BAY DRAINAGE										
#34	Red River Noyes, Minn.	Goldeye	5	12.3	0.6	.19				
		Goldeye (R)	5	12.4	0.7	.19				
		White sucker								
		Channel catfish	3	15.6	0.7	.13				
		Sauger	2	12.2	0.5	.60				
Avg.				.31	2	16.4	2.1	.36		
									Avg. .41	
									Avg. .39	

TABLE 1.—Mercury residues in fish, 1969 and 1970—Continued

STATION NUMBER AND LOCATION	SPECIES	1970				1969				
		NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	
			LENGTH (INCHES)	WEIGHT (LB)			LENGTH (INCHES)	WEIGHT (LB)		
COLORADO RIVER SYSTEM										
#35	Green River Vernal, Utah	Carp	5	10.8	0.6	.14	5	11.0	0.9	<.05
		Flannelmouth sucker	5	16.7	1.4	.17		3	19.2	2.6
		Flannelmouth sucker (R)	5	16.6	1.5	.24				
		Channel catfish	5	9.2	0.3	.20				
		Black bullhead					3	5.4	0.2	<.05
					Avg. .18				Avg. <.05	
#36	Colorado River Imperial Reser- voir, Ariz.	Carp	5	16.4	3.9	<.05	5	16.9	2.5	<.05
		Carp (R)	4	14.8	2.5	<.05				
		Channel catfish	5	12.3	0.9	<.05	3	9.2	0.2	<.05
		Largemouth bass	5	14.5	0.9	.05	5	10.3	0.5	<.05
							Avg. <.05			
#91	Colorado River Havasu Lake, Ariz.	Carp	5	14.1	1.4	<.05				
		Carp (R)	5	14.1	1.4	.08				
		Channel catfish	5	10.6	0.7	<.05				
		Largemouth bass	5	13.1	0.7	.08				
							Avg. .06			
#92	Colorado River Lake Mead, Nev.	Carp	4	15.8	2.8	.05				
		Carp (R)	3	16.1	3.0	.05				
		Largemouth bass	4	14.7	2.0	.09				
							Avg. .07			
#93	Colorado River Lake Powell, Ariz.	Carp	3	12.7	0.5	.27				
		Largemouth bass	4	13.9	1.2	.23				
		Largemouth bass (R)	4	13.0	0.9	.18				
		Rainbow trout	5	13.4	1.1	.10				
							Avg. .19			
#94	Gila River San Carlos Reservoir, Ariz.	Carp	6	14.5	1.0	.11				
		Bluegill	6	4.7	0.1	.13				
		Largemouth bass	3	13.1	1.2	.22				
							Avg. .15			
INTERIOR BASINS										
#37	Truckee River Fernley, Nev.	Carp	5	13.2	1.1	.21	5	14.6	1.5	.37
		Carp (R)	5	12.7	1.1	.23				
		Brown bullhead	5	10.1	0.6	.35	5	10.2	0.7	.22
		Largemouth bass	5	11.5	1.1	.65	5	12.3	1.1	.53
							Avg. .41			
#38	Utah Lake Provo, Utah	Carp	5	16.7	2.4	<.05	5	17.0	2.1	.06
		Black bullhead	5	10.2	0.6	.07		5	9.8	0.5
		Black bullhead (R)	5	9.9	0.6	.05				
		White bass	5	10.4	0.5	.06	5	10.2	0.5	.06
							Avg. .04			
#95	Bear River Preston, Idaho	Carp	3	18.6	3.8	.21				
		Largescale sucker	5	17.1	2.7	.34				
		Largescale sucker (R)	5	16.1	1.9	.37				
		Yellow perch	5	7.9	0.3	.13				
							Avg. .23			
CALIFORNIA STREAMS										
#39	Sacramento River Sacramento, Calif.	Carp	5	10.9	0.8	.18	5	12.9	0.9	.11
		Carp (R)	5	12.3	1.2	.18				
		White catfish	5	8.3	0.3	.20	5	14.1	1.3	.19
		Largemouth bass	5	11.9	1.0	.22	3	10.8	0.6	.13
							Avg. .20			
#40	San Joaquin River Los Banos, Calif.	Carp	5	15.1	1.7	.23	5	14.7	1.3	.20
		Channel catfish	5	16.6	2.3	.16		5	16.1	1.2
		Black crappie	5	11.6	1.2	.09	5	10.4	0.6	.14
		Black crappie (R)	5	10.8	1.1	.11				
							Avg. .16			

TABLE 1.—Mercury residues in fish, 1969 and 1970—Continued

STATION NUMBER AND LOCATION	SPECIES	1970				1969								
		NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹					
			LENGTH (INCHES)	WEIGHT (LB)			LENGTH (INCHES)	WEIGHT (LB)						
COLUMBIA RIVER SYSTEM														
#41	Snake River Hagerman, Idaho	Largescale sucker	5	13.0	1.0	.08	5	15.0	1.4	.08				
		Largescale sucker (R)	5	12.6	0.9	.10								
		Rainbow trout	5	14.2	1.3	.18								
		Northern squawfish	3	15.4	1.7	.43								
		Avg.				.23								Avg.
#42	Snake River Lewiston, Idaho	Carp	4	15.3	2.3	.25	5	15.9	2.0	.10				
		Largescale sucker	5	15.8	1.8	.23								
		Largescale sucker (R)	5	15.3	1.6	.18								
		Smallmouth bass	5	11.0	0.8	.21								
		Northern squawfish									3	7.0	0.3	.15
Avg.				.22				2	13.0	0.8	1.25	Avg.	.50	
#43	Salmon River Riggins, Idaho	Carp	4	13.1	1.5	.29	5	15.2	1.6	.23				
		Largescale sucker	4	17.4	2.3	.42								
		Largescale sucker (R)	3	17.7	2.3	.39								
		Northern squawfish	2	13.1	1.0	1.70								
		Avg.				.80								
#96	Snake River Ice Harbor Dam, Wash.	Bridgelip sucker	5	12.1	0.8	.12								
		Channel catfish	5	13.1	1.2	.90								
		Channel catfish (R)	5	14.3	1.3	.10								
		Northern squawfish	4	12.6	0.7	1.20								
		Avg.				.61								
#44	Yakima River Granger, Wash.	Carp	5	11.2	1.1	.23								
		Carp (R)	5	11.3	1.0	.23								
		Black crappie									5	7.4	0.3	.13
		Largescale sucker	5	14.0	1.2	.07					5	13.3	0.9	.30
		Smallmouth bass	3	12.3	1.4	.27					2	10.0	0.9	.14
Avg.				.19					Avg.	.19				
#45	Willamette River Oregon City, Oreg.	Carp	3	19.0	5.0	.17								
		Largescale sucker	5	15.3	1.8	.37								
		Largescale sucker (R)	5	15.9	1.7	.33								
		Chiselmouth									4	10.2	0.5	.13
		White crappie									5	6.9	0.2	.23
Avg.				.26					Avg.	.18				
#46	Columbia River Bonneville Dam, Oreg.	Largescale sucker	5	16.9	2.0	.21	5	16.5	2.0	.27				
		Chiselmouth									4	7.9	0.3	.15
		Northern squawfish	5	12.7	0.8	.67					5	13.1	1.0	1.25
		Northern squawfish (R)	5	13.8	1.1	1.10								
		Avg.				.55								
#97	Columbia River Pasco, Wash.	Carp	5	12.6	1.3	.07								
		Largescale sucker	5	17.2	2.6	.16								
		Largescale sucker (R)	5	16.3	2.1	.14								
		Mountain whitefish	3	12.0	0.5	.19								
		Avg.				.14								
#98	Columbia River Grand Coulee Dam, Wash.	Bridgelip sucker	5	14.1	1.2	.07								
		Walleye	5	14.3	1.1	.11								
		Walleye (R)	5	14.8	1.3	.12								
		Northern squawfish	5	12.6	0.9	.25								
		Avg.				.15								
PACIFIC COASTAL STREAMS														
#47	Klamath River Hornbrook, Calif.	Klamath sucker	5	15.7	2.1	.27	5	13.5	1.2	.12				
		Yellow perch	5	7.9	0.3	.20								
		Yellow perch (R)	5	8.2	0.3	.19								
		Largemouth bass	5	8.4	0.3	.12								
		Rainbow trout												
Avg.				.20					Avg.	.16				
#48	Rogue River Gold Ray Dam, Oreg.	Carp	5	15.2	2.6	.18	5	14.0	1.5	.16				
		Bridgelip sucker	5	10.8	0.7	.47								
		Brown bullhead	5	10.8	0.6	.51								
		Brown bullhead (R)									5	9.5	0.5	.90
		Black crappie												
Largemouth bass	2	9.5	0.6	.14										
Avg.				.27						Avg.	.44			

TABLE 1.—Mercury residues in fish, 1969 and 1970—Continued

STATION NUMBER AND LOCATION	SPECIES	1970				1969				
		NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	NO. OF FISH	AVERAGE SIZE		TOTAL MERCURY (PPM) ¹	
			LENGTH (INCHES)	WEIGHT (LB)			LENGTH (INCHES)	WEIGHT (LB)		
ALASKAN STREAMS										
#49	Chena River Fairbanks, Alaska	Longnose sucker	2	13.5	1.7	.05	5	15.0	1.2	.09
		Round whitefish	3	11.7	1.0	.06		10.0	0.2	.06
		Arctic grayling	3	10.2	0.6	.06		11.7	0.5	.07
					Avg.	.06				Avg.
#50	Kenai River Soldatna, Alaska	Longnose sucker	5	12.1	0.7	.10	5	15.5	1.5	.08
		Round whitefish	5	16.3	1.5	.06		13.2	0.9	.06
		Rainbow trout	5	13.7	0.9	.26		14.6	0.9	.12
		Lake trout	5			.14				Avg.
				Avg.	.14			Avg.	.09	
HAWAIIAN STREAMS										
#99	Waieke Stream Waipahu, Hawaii	Tilapia ²	2	6.1	0.1	.11				
		Cuban limia ³	13	2.6	0.1	.06				
		Chinese catfish ⁴	3	8.1	0.1	.43				
		Chinese catfish (R)	2	9.4	0.2	.22				
					Avg.	.17				
#100	Manoa Stream Honolulu, Hawaii	Tilapia ²	3	8.0	0.3	.09				
		Cuban limia ³	6	3.5	0.1	.13				
		Chinese catfish ⁴	3	10.3	0.3	.18				
		Chinese catfish (R)	3	8.8	0.2	.15				
					Avg.	.13				

NOTE: (R) designates replicate field sample. To compute residue average, the replicate samples at a sampling station were averaged and this average used with the values for the other species at a station to compute the average; values less than the sensitivity level of 0.05 ppm were considered to be zero in computing averages.

¹ mg/kg—wet-weight basis—whole fish.

² *Tilapia mossambica*.

³ *Limia vittata*.

⁴ *Clarias fuscus* (Identification uncertain).

TABLE 2.—Results of analyses for total mercury and methyl mercury by Laboratory C and confirmatory analyses for total mercury by Laboratory H—1970

STATION NUMBER AND LOCATION	SPECIES	TOTAL MERCURY (MG/KG) ¹		METHYL MERCURY ² (MG/KG OF Hg) LABORATORY C	
		LABORATORY C	LABORATORY H		
#1	Stillwater River	Chain pickerel	.64		.69
#2	Connecticut River	White perch	.51	.54	.43
		Yellow perch	.30	.38	
#3	Hudson River	Goldfish	.07	.13	
#4	Delaware River	White perch	.21	.24	
		White perch (R)	.17	.22	
#8	Cape Fear River	Largemouth bass	.60		.61
		Gizzard shad	.25	.17	
#10	Savannah River	Largemouth bass	1.80	1.73	1.70
#14	Tombigbee River	Largemouth bass	.92	.80	1.10
#15	Mississippi River	Carp (R)	<.05	.04	
#16	Rio Grande	Channel catfish	.17	.15	
#18	Lake Ontario	White perch	1.30	.97	1.00
#20	Lake Huron	Carp	.07	.04	
#21	Lake Michigan	Bloater (R)	.07	.09	

TABLE 2.—Results of analyses for total mercury and methyl mercury by Laboratory C and confirmatory analyses for total mercury by Laboratory H—1970—Continued

STATION NUMBER AND LOCATION	SPECIES	TOTAL MERCURY (MG/KG) ¹		METHYL MERCURY ² (MG/KG OF HG) LABORATORY C
		LABORATORY C	LABORATORY H	
#23 Kanawha River	Brown bullhead (R)	<.05	.03	
#24 Ohio River	Channel catfish	.83	.63	.65
	Channel catfish (R)	.68	.57	.56
#26 Illinois River	White crappie	.21	.12	
#28 Arkansas River	Smallmouth buffalo (R)	.13	.16	
#30 White River	Bigmouth buffalo (R)	.25	.24	
#31 Mississippi River	Goldeye	.20	.18	
#34 Red River (North)	Sauger	.60		.48
#37 Truckee River	Largemouth bass	.65		.65
#39 Sacramento River	Carp (R)	.18	.20	
#40 San Joaquin River	Black crappie	.09	.08	
#43 Salmon River	Northern squawfish	1.70		1.40
#45 Willamette River	Largescale sucker (R)	.37	.45	.36
#46 Columbia River	Northern squawfish (R)	1.10	.97	.88
#48 Rogue River	Brown bullhead	.47		.39
#51 Kennebec River	Yellow perch	1.20		1.10
#52 Lake Champlain	Yellow perch (R)	.49		.48
#53 Merrimac River	Yellow perch	.33	.34	
#54 Raritan River	Golden shiner	.08	.06	
#55 James River	Channel catfish	.12	.07	
#56 Pee Dee River	Largemouth bass	1.00	1.00	1.00
#57 Altamaha River	Spotted sucker	.25	.23	
	Largemouth bass	.51		.52
#59 Alabama River	Largemouth bass	.60	.74	.60
#63 Rio Grande	Largemouth bass	.52		.52
#67 Allegheny River	Walleye	.28	.19	
#69 Ohio River	Carp (R)	.20	.10	
	White crappie	.15	.19	
#71 Tennessee River	Largemouth bass	.67		.69
#72 Wisconsin River	Smallmouth bass	.60		.56
#74 Mississippi River	Yellow bullhead	.68		.62
#75 Mississippi River	Carp	.20	.16	
#76 Mississippi River	Freshwater drum	.15	.23	
#80 Yazoo River	Carp	.19	.21	
	Smallmouth buffalo	.16	.15	
#81 Red River	Smallmouth buffalo (R)	.10	.06	
#83 Missouri River	Channel catfish	.15	.19	
#96 Snake River	Northern squawfish	1.20	1.13	.94

NOTE: (R) designates replicate sample.

¹ Confirmatory analyses at Laboratory H on 40 samples also analyzed at Laboratory C.

² Analyses for methyl mercury at Laboratory C based on 24 samples.

Dursban® and Diazinon Residues in Biota Following Treatment of Intertidal Plots on Cape Cod—1967-69

Vahe M. Marganian¹ and William J. Wall, Jr.²

ABSTRACT

The effects of the organophosphorous pesticides Dursban® and diazinon on intertidal biota were studied over a period of 3 years, 1967-69. One percent granular Dursban® applied manually at an optimum concentration of 0.05 lb/acre controlled *Culicoides* larvae effectively with no noticeable harm to fiddler crabs or other organisms. Residues recovered ranged from trace amounts to 2.30 ppm in white oligochaete, 2.58 ppm in ribbed mussel, 4.62 ppm in fiddler crab, 14.0 ppm in horsefly, and 15.7 ppm in marsh snail. Two percent granular diazinon applied manually at 0.20 lb/acre controlled *Culicoides* effectively, but killed small sand organisms. In general, concentrations of diazinon residues recovered were higher than those for Dursban® in the same organisms reported above. This report describes the sampling and gas chromatographic analytical procedures employed in this work, discusses data collected on residues in organisms at various periods after treatment, and gives persistence periods for these pesticides in substrates of intertidal sand, salt marsh sod, salt marsh mud, and sea water.

Introduction

In recent years, the deleterious effects of organochlorine pesticides, particularly DDT, on intertidal biota have received much criticism from environmentalists and the public. In an effort to find alternate means to organochlorine pesticide control, the effects of a number of organophosphorous pesticides on the breeding patterns of bloodsucking insects in intertidal areas have been studied (1).

This paper reports the results of a 3-year study (1967-69) conducted on Cape Cod, Mass., to determine if the use of Dursban® and diazinon for control of larvae of *Culicoides melleus* breeding in intertidal sand, *C. hollensis* and *C. furens* breeding in salt marsh mud, and *Tabanus nigrovittatus* and *T. lineola* breeding in salt marsh sod, would result in harmful effects to nontarget organisms and to determine residue levels in the intertidal biota. Reports on the field tests and on the classification of organisms are being published separately.

Because techniques for the chemical analysis of Dursban® and diazinon were not readily available, it was necessary to develop analytical methodology in the early

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phases of the study for the detection and quantitative measurement of both compounds.

Treatment and Sampling Procedures

The majority of the tests were conducted in small intertidal sand and salt marsh plots ranging from 0.08 to 0.25 acres in size. For each test, two plots were treated with a single concentration of Dursban® or diazinon and a nearby untreated plot was used as a control. These pesticides, in granular form, were applied by hand at low tide, and in some instances uncoated granules were added to the pesticide-coated granules to provide additional materials for better distribution. During the latter phases of the program, two extended areas of intertidal sand or beach areas were treated using a Kiekens Whirlwind gasoline-operated duster and a salt marsh was treated using a helicopter.

Samples of intertidal biota, sand, and water were collected at regular intervals from treated and untreated areas before and after treatment as described by Wall and Marganian (1). When it was necessary to separate small organisms from the underlying sand, mud, or sod, the methods used for separation and counting of the larvae were based on that of Jamnback and Wall (2), Wall and Jamnback (3), and Wall and Doane (4).

The procedure for sample storage was based on methodology recommended by Van Middeltem (5), Beckman (6), and Lykken (7). All specimens were stored at low temperatures (about 0° C) to prevent possible decomposition of the insecticides, and, when possible, samples were analyzed within 24 hours of their arrival at the laboratory.

Analytical Procedures

Extraction and cleanup procedures were related to techniques outlined by Thornburg (8) and Morley and Chiba (9). The wet weight of analyzed samples, representing composites of several specimens of the same species, as a rule, was in the following ranges: 1-10 g for clams, fiddlers, ribbed mussels, oysters, and shrimp; 0.1-1 g for mud snails, marsh snails, mud whelks, and periwinkles; 0.01-0.1 g for amphipods, crabs, tanai-*daceans*, horseflies, white oligochaetes, and red oligochaetes; 15-20 g for sand; 50-55 g for sod; and 50 ml for water. Each sample of biota, sand, mud, and water was treated in a blender with 25 ml of reagent grade acetonitrile (density = 0.78 g/ml) for 5 minutes, and a 10- μ l solution of internal standard dissolved in petroleum ether was added to the blender to insure uniform losses of insecticides during subsequent extractions. For the quantitative analysis of Dursban®, diazinon was used as an internal standard, and vice versa.

This was followed by a second extraction of the sample with 15 ml of acetonitrile, and the two extracts were then combined. The combined extracts were next treated with an equal volume of petroleum ether (b.p. = 30°-60° C, density = 0.62 g/ml) by shaking the mixture vigorously in a separatory funnel and removing the acetonitrile phase. This step was repeated twice using 25 ml of petroleum ether in each treatment of the acetonitrile phase, and all three petroleum ether extracts were combined. Five milliliters of a saturated NaCl solution and 25 ml of distilled water were added to the combined petroleum ether phase, shaken again, and the ether phase was transferred to a second separatory funnel, where it was washed again with distilled water. The two aqueous layers were treated together with small amounts of additional petroleum ether, and all ether phases were combined and dried over anhydrous Na₂SO₄. The petroleum ether extracts were next concentrated to about 0.3 ml with a gentle stream of nitrogen, and 2- μ l aliquots were subjected to gas chromatographic analysis.

An F&M series Model 400 Biomedical gas chromatograph with a Honeywell strip chart recorder and a modified flame detector was used. This modification was achieved by coating a 27-gauge platinum wire with Na₂SO₄ beads and mounting the coiled wire over the tip of the flame. The operating parameters were as follows:

Columns:	Glass, 6' x 1/4", o.d., packed with 5% UC-W98 on 60/80 mesh, acid-washed Diatoport S.
Temperatures:	Detector 245° C Injector 250° C Oven 185° C
Flow rates:	Carrier gas—He at 45 ml/min Oxygen (air) at 340 ml/min Hydrogen at 22 ml/min
Retention times:	Diazinon 4.2 min Dursban® 7.8 min

The lower limit of sensitivity was 0.01 ppm (mg/kg, net fresh weight). Quantitative calculations were carried out on a computer. A 6% precision of results was obtained on several samples determined at random. Recovery rates for both insecticides ranged from 80-85%. Data in this report do not include a correction factor for percent recovery. A few samples containing high concentrations of Dursban® and diazinon were analyzed by ultraviolet spectroscopy (at 231 m μ and 246 m μ , respectively) and were found to give comparable results on gas chromatographic analysis.

Results and Discussion

The effects of applications of Dursban® and diazinon on nontarget intertidal biota are shown in Table 1. The applications listed in Table 1 effectively controlled *Culicoides* larvae. In addition, Dursban® and diazinon

TABLE 1.—Effects of Dursban® and diazinon on intertidal biota after effective control of Culicoidae larvae

APPLICATION RATE LB/ACRE	MEANS OF APPLICATION	AREA TREATED	ORGANISM CONTROLLED	EFFECTS ON NONTARGET ORGANISMS
DURSBAN®				
0.05	duster	0.5 mile of intertidal shoreline	<i>C. melleus</i> larvae (up to 2 weeks following application)	Killed numerous fiddler crabs
0.05	manually	Sand plots	<i>C. melleus</i> larvae	No visible effects
0.1 and 0.2	manually	Sand plots	<i>C. melleus</i> larvae	Killed numerous fiddler crabs
0.20	manually	0.08—acre salt marsh plots	<i>Culicoides</i> larvae, specifically <i>C. jurens</i> and <i>C. hollensis</i> (for 18 days)	Killed numerous larvae (75%)
DIAZINON				
0.20	helicopter	17 acres of salt marsh	<i>C. hollensis</i> larvae (up to 29 days after treatment)	Numerous fish were found dead in the tidal creek up to 3 days following treatment
0.2	duster	1.1 mile of intertidal sand	<i>C. melleus</i> larvae	Killed numerous marine fish including <i>Fundulus</i> spp.
0.2	manually	Sand plots	<i>C. melleus</i> larvae	Killed small sand organisms such as naidids and dolichopods
0.20	manually	0.12—acre salt marsh plots	<i>Culicoides</i> larvae, specifically <i>C. jurens</i> and <i>C. hollensis</i> (for 7 days)	Killed numerous larvae (80%)

applied by hand at low tide to 0.25-acre salt marsh plots at rates of 0.05 and 0.3 lb/acre, respectively, failed to effectively control larvae of *Tabanus* sp.; numerous confined *Fundulus* spp. appeared to have died on the second day following this Dursban® treatment, and others placed daily in potholes were killed up to 4 days following the diazinon treatment.

An optimum concentration of 0.05 lb/acre of 1% granular Dursban®, applied manually, effectively controlled *Culicoides* larvae in small plots without visibly harming fiddler crabs or other organisms. However, Dursban® applied at the same rate by duster to 0.5 mile of intertidal sand shoreline controlled *C. melleus* larvae up to 2 weeks following treatment but also killed numerous fiddler crabs.

Two percent granular diazinon applied manually at 0.20 lb/acre controlled all *Culicoides* larvae effectively, but killed small sand organisms, such as naidids and dolichopods.

Table 2 gives the results of analyses of various species from sand and salt marsh plots for residues of Dursban® following treatment at 0.05 lb/acre. Results of analyses of biota following diazinon treatments at 0.2 and 0.25 lb/acre are given in Table 3.

Some specimens from most of the species or groups (where species could not be determined) analyzed during the first 4 days after treatment were found to contain measurable amounts of Dursban® (Table 2) or diazinon (Table 3). Organisms taken from sand plots treated with diazinon applied at the rate of 0.2 pound of technical material per acre generally contained more residues than similar organisms taken from comparable plots treated with Dursban® applied at the rate of 0.05 pound of technical material per acre. Table 4 shows the periods of persistence of these pesticides in intertidal sand, salt marsh sod, salt marsh mud, and sea water. Each figure in Tables 2-4 represents one analysis per sample.

With a few exceptions, no measurable residues of Dursban® or diazinon were found in the organisms from or the substrate of the treated or control plots prior to treatment, and none were found in organisms or substrate from control plots following treatment.

In most of the species or groups analyzed, no obvious pattern of accumulation or disappearance of residues could be established for the two pesticides studied. In some specimens, residues up to 50 ppm were found during the first few days following treatment, gradually declining to immeasurable amounts at the end of 30 days.

TABLE 2.—Dursban® residues in marine organisms at various intervals after treatment at a concentration of 0.05 lb/acre

ORGANISM	SUBSTRATE	DURSBAN® RESIDUES IN PPM (FRESH-WEIGHT BASIS) ¹								
		DAYS								
		1	2	3	4	7	9	10	12	22
Oligochaeta										
Enchytraeidae (white oligochaete)	Salt marsh mud	1.0	—	2.30	—					
<i>Mys arenaria</i> (clam)	Intertidal sand		T	—	—	0.01				
Mollusca										
<i>Modiolus demissus</i> (ribbed mussel)	Salt marsh mud	—	2.58	0.12		—				
<i>Nassarius obsoletus</i> (mud snail)	Intertidal sand	T	T	1.10	—	0.62				
<i>Melampus bidentatus</i> (marsh snail)	Salt marsh mud	—	T	2.24	—	—		15.70		
Arthropoda										
<i>Leptochelia</i> sp. (tanaidacean)	Salt marsh mud	T	—	16.70		—				T
<i>Palaemonetes</i> sp. (prawn)	Intertidal sand	—	—							
<i>Uca</i> spp. (fiddler crab)	Intertidal sand	T	T	—		4.62				
<i>Culicoides</i> spp. (gnat)	Salt marsh mud	0.43	—							
<i>Tabanus</i> spp. (horsefly)	Salt marsh mud	T	—	3.80		0.43		14.00	—	

NOTE: Dursban® applied as 1% technical grade Dursban® in granular form; blank = no sample taken, — = no residue detected; T = trace = <0.01 ppm.

¹ Represents one analysis per sample.

In other organisms of the same species or group, collected during the second or third week following treatment, residues over 20 ppm were found.

From 1 to 7 days following treatment, many annelids, particularly Oligochaeta, did not contain measurable amounts of residues, but others were found to contain large quantities (over 10 ppm) of the particular pesticide employed. From 1 to 21 days following treatment, samples of the *Fundulus* spp. (killifish) as well as many samples of Pelecypoda and Arthropoda analyzed contained small measurable amounts of residues (usually <1 ppm) of the pesticide distributed. In general, Mollusca (Gastropoda and Pelecypoda) were found to consistently contain more residues than the other organisms analyzed.

Organisms taken from salt marsh habitats generally contained more residue of the pesticide employed than the same species or group taken from intertidal sand habitats.

Dursban® applied at the rate of 0.05 pound of technical material per acre persisted in measurable amounts in the top one-half inch of intertidal sand for an average of 2

days with a maximum of 4 days; in the top inch of salt marsh sod for an average of 5 days with a maximum of 12 days; and in the top inch of salt marsh mud for an average of 15 days with a maximum of 22 days (Table 4). Diazinon applied at the rate of 0.2 pound of technical material per acre persisted in measurable amounts in the top one-half inch of intertidal sand for an average of 4 days with a maximum of 12 days; in the top inch of marsh sod for an average of 6 days with a maximum of 10 days; and in the top inch of marsh mud for an average of 10 days with a maximum of 22 days (Table 4). For both pesticides, residues on the day following treatment were greatest, gradually declining thereafter.

With one minor exception, analysis of sea water taken from the bay or creek immediately adjacent to each treated plot showed no residues on the day following treatment.

During the summer following the original application of Dursban® in small intertidal sand and marsh plots, organisms and substrates from these plots were analyzed, and no measurable amounts of residues of either pesticide were found.

TABLE 3.—Diazinon residues in marine organisms at various intervals following treatment

ORGANISM	SUBSTRATE	APPLI- CATION RATE LB./ACRE	DIAZINON RESIDUES IN PPM (FRESH-WEIGHT BASIS) ¹																
			DAYS																
			1	2	3	4	5	6	7	8	9	11	14	15	22	29	36	38	45
Oligochaeta Enchytraeidae (white oligochaete)	Intertidal sand	0.20	14.82	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	
Naididae and Tubificidae (red oligochaete)	Salt marsh	0.25	—	—	—	—	—	—	—	—	—	—	—	36.80	—	—	—	—	
Polychaeta (sandworm)	Salt marsh	0.25	—	—	0.73	—	—	—	—	—	—	23.51	—	—	—	—	—	—	
Mollusca <i>Mercenaria mercenaria</i> (quahog)	Salt marsh	0.25	4.36	T	0.07	1.80	—	—	—	—	—	—	—	—	—	—	—	—	
<i>Crassostrea virginica</i> (oyster)	Salt marsh	0.25	—	—	0.56	—	1.69	0.81	—	—	—	0.11	0.03	3.16	T	0.03	—	—	
<i>Modiolus demissus</i> (ribbed mussel)	Salt marsh	0.25	50.00	2.06	2.93	3.93	0.99	—	—	—	—	0.23	—	—	—	—	—	—	
<i>Nassarius obsoletus</i> (mud snail)	Intertidal sand	0.20	—	—	0.16	—	2.45	—	—	—	—	0.30	0.23	—	—	—	—	0.17	
<i>Melampus bidentatus</i> (marsh snail)	Salt marsh mud	0.20	2.22	1.31	2.22	—	—	—	—	—	—	1.66	—	—	—	—	—	—	
<i>Littorina littorea</i> (periwinkle)	Intertidal sand	0.20	0.62	—	0.11	2.16	14.26	T	—	—	—	0.16	—	0.40	—	—	—	—	
<i>Uca</i> spp. (fiddler crab)	Salt marsh mud	0.20	0.05	—	0.33	T	—	0.11	—	—	—	0.04	T	0.01	—	—	—	0.09	
<i>Carcinus maenas</i> (green crab)	Intertidal sand	0.20	0.82	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	
<i>Parasita longicarpus</i> (hermit crab)	Salt marsh	0.25	3.61	—	0.46	1.21	—	—	—	—	—	—	0.12	—	—	—	—	—	
<i>Palaeomonetes</i> and <i>Crangon</i> (prawn)	Salt marsh mud	0.20	—	—	0.86	—	—	—	—	—	—	—	—	—	—	—	—	—	
Pisces <i>Fundulus</i> spp. (killifish)	Intertidal sand	0.20	1.42	0.95	0.95	0.10	—	—	—	—	—	—	—	—	—	—	—	0.49	

NOTE: Diazinon applied as 2% technical grade in granular form; blank = no sample taken; — = no residue detected; T = trace = <0.01 ppm.
¹ Representative analysis per sample.

TABLE 4.—Persistence of Dursban® and diazinon in various marine substrates

SUBSTRATE	DAYS			
	DURSBAN		DIAZINON	
	AVERAGE	MAXIMUM ¹	AVERAGE	MAXIMUM ¹
Intertidal sand	2	4	4	12
Salt marsh sod	5	12	6	10
Salt marsh mud	15	22	10	22
Sea water		0		0

¹ Time of last detectable trace

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See Appendix for chemical names of compounds discussed in this paper.

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PESTICIDES IN WATER

Seasonal Variations in Residues of Chlorinated Hydrocarbon Pesticides in the Water of the Utah Lake Drainage System—1970 and 1971¹

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ABSTRACT

Definite surges of pesticides (1 ppb or more) enter Utah Lake three times per year—early spring, late spring, and fall, generally corresponding to the application times of pesticides by farmers in the area. The pesticides involved were mainly aldrin and BHC in the early spring; heptachlor (plus heptachlor epoxide) and methoxychlor in the late spring; and aldrin, heptachlor, and methoxychlor in the late fall. The fish samples collected from Utah Lake contained only small amounts of pesticides, the highest level being 956 ppb DDE.

Introduction

Utah Lake is a large freshwater lake in central Utah (Fig. 1), 25 miles long and 11 miles wide with an average depth of 9 feet. The major inlets to Utah Lake are on the east side, and more than half of the estimated 600,000 acre-feet of water entering Utah Lake in an average year enters through the Provo, Spanish Fork, and American Fork Rivers. The rest of the water enters through agricultural drains, small creeks, and underground springs. The only outlet from Utah Lake is the Jordan River to the north. Utah Lake serves as a reservoir for the water users of the Utah Lake and Jordan River Irrigation System.

Previous studies concerning the overall chemistry of Utah Lake were summarized by Bradshaw *et al.* (1, 2). The only previous pesticide residue studies of Utah

Lake showed that at least two surges of pesticides enter Utah Lake every year (1, 3). Fish from Utah Lake were reported to contain minute amounts (0.1-0.8 ppm) of chlorinated hydrocarbon residues (4, 5). This study was undertaken to catalog the sources and types of pesticides entering the Lake in a given year and to determine the extent of pesticide residues in Utah Lake fish.

Sampling and Analytical Procedures

WATER SAMPLES

Water from 15 tributaries and 1 outlet point (16 Stations) was sampled biweekly from March 1 to July 1, 1970, and weekly or semiweekly through February 1971 (Fig. 1). Station 2 was soon abandoned because it dried up. One gallon of water was collected at each station in large mouth glass jars. The water (3 liters) was extracted as soon as possible (within 3 days) with nano-grade petroleum ether (Mallinkrodt Chemical Co.) in a continuous liquid-liquid extractor for 24 hours. Care was taken to insure that a representative mixture of water and suspended material was extracted. The petroleum ether extract was dried over anhydrous sodium sulfate, filtered, and evaporated to 10 ml. The sample was then analyzed on a dual glass column, Varian Aerograph 202 gas-liquid chromatograph (GLC) using electron capture detectors. Each sample was analyzed simultaneously on the two columns. One column (1/8" x 5') was packed with a mixture of 4% SE-30 and 6% QF-1 on Chromosorb W and the other, with a mixture of 1.5% OV-17 and 1.95% QF-1 on Chromosorb W. The columns were heated to 200° C. The GLC peak areas were determined using a Disc Integrator. Quantitation was accomplished by comparing the peaks with

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those of known standards. Only pesticides verified by the second column were evaluated (6). The final pesticide levels were calculated using a Hewlett-Packard 9100 B "programmable" calculator.

FISH SAMPLES

Fish were caught from the east side of Utah Lake between the Orem Pier and Provo Airport on July 21, 1970, by Utah State Wildlife Resources personnel. The fish were treated in a manner similar to that reported by Kleinert, Degurse, and Wirth (7). Each frozen fish sample was ground and homogenized in a heavy duty meat grinder. A 10-g portion of the homogenized fish was added to a blender and mixed with 60 g of anhydrous sodium sulfate and 200 ml of hexane (distilled through a glass column in the laboratory). The solvent from the blended material was decanted through a filter paper into a 500-ml round-bottom flask. The fish sample was washed four times with 50-ml portions of hexane. The combined hexane extracts were evaporated at about 50-mm pressure to 5 ml using a rotary evaporator. This material was then placed on a column containing 200 g of Florisil and eluted from the column with a mixture of 180 ml of hexane and 12 ml of ethyl ether which contained 2% ethyl alcohol. The solution was eluted at a rate of 5 ml/min. The resulting solution was evaporated at about 50-mm pressure to 10 ml and subjected to GLC analysis as reported above.

RECOVERY STUDIES

Spiked water samples treated as above gave 75-85% recovery of the various pesticide residues. The results were not corrected to reflect this recovery.

Results and Discussion

WATER SAMPLES

The concentrations of pesticides detected in water samples (aldrin, total alpha- and gamma-BHC, heptachlor and heptachlor epoxide, methoxychlor, and DDT-type compounds) by collection stations on Utah Lake are shown in Fig. 2. The results showed that definite surges of pesticides entered the Lake at specific times during 1970. Except for these surges, extremely low levels of pesticides were detected in the water entering the Lake. Aldrin and BHC were the main residues entering Utah Lake in the early spring (March-April); heptachlor or its epoxide and methoxychlor in the late spring (May-June); and aldrin, heptachlor or its epoxide, and methoxychlor in the late fall (October-November). These surges corresponded to the times of application of the respective pesticides in Utah County.

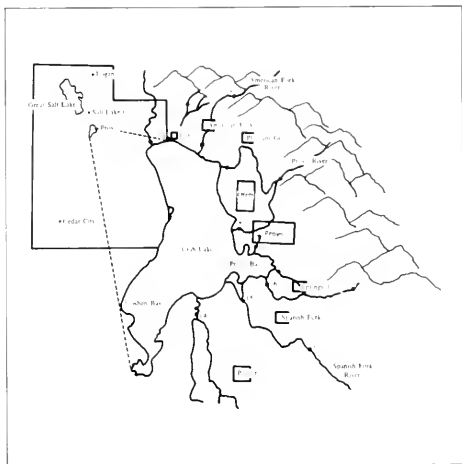


FIGURE 1.—Sampling stations on the Utah Lake tributaries, 1970 and 1971

Most of the pesticide residues were detected in tributaries in valley locations, with the exception of residues detected in the early spring (probably resulting from forest runoff) and at one location in the fall (high aldrin at Station 4). Station 13 (Fig. 1) was also considered a valley location since there are many farms above this point.

Aldrin (Fig. 2) was observed mainly in the northern part of the Utah Valley where it is used for grasshopper control. The spring and fall surges of this pesticide entering Utah Lake were at levels over 1 ppb.

The greatest surge of BHC, which is used extensively on livestock in Utah County, was observed in the early spring at a level of 1.3 ppb (Fig. 2) in the south central area where there is much livestock.

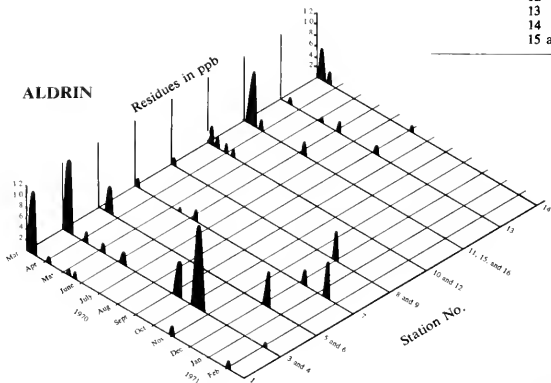
The high incidence of heptachlor or its epoxide in the alfalfa-producing area (Fig. 2—Stations 5, 6, and 13) was somewhat surprising, however, since heptachlor is no longer registered for use on alfalfa (8) and has not been sold in Utah County for 3-4 years. It is likely that the heptachlor used was that left over from previous purchases.

Methoxychlor, due to its relatively rapid degradation (9), is finding wide use as a replacement for DDT and was used extensively throughout Utah County. It was never detected in samples at the Jordan River outlet from Utah Lake probably because of its degradation (Fig. 2).

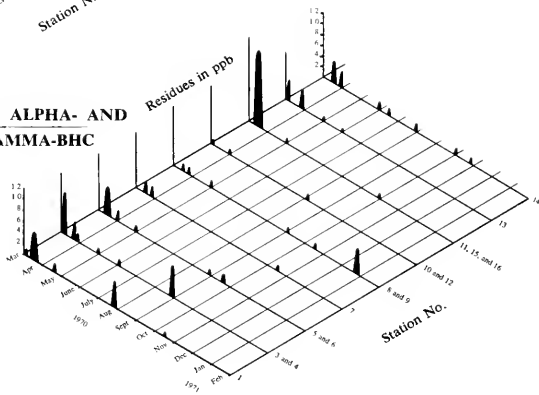
FIGURE 2.—Pesticide residues in water samples from the Utah Lake drainage system, 1970 and 1971

SAMPLING STATION NUMBER	LOCATION
1	Jordan River
3 and 4	North Upper
5 and 6	North Lower
7	North Central Upper
8 and 9	North Central Lower
10	South Central Upper
11	South Central Lower
12	South Central Upper
13	South Upper
14	South Lower
15 and 16	South Central Lower

ALDRIN



TOTAL ALPHA- AND GAMMA-BHC



HEPTACHLOR AND HEPTACHLOR EPOXIDE

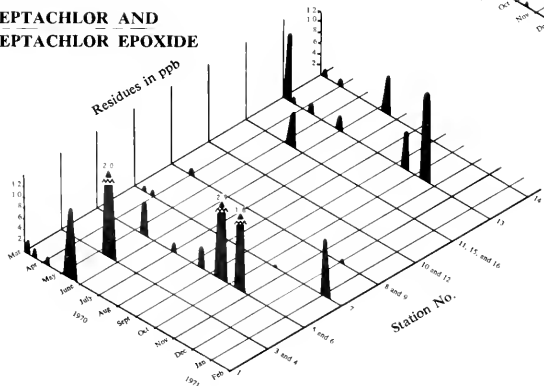
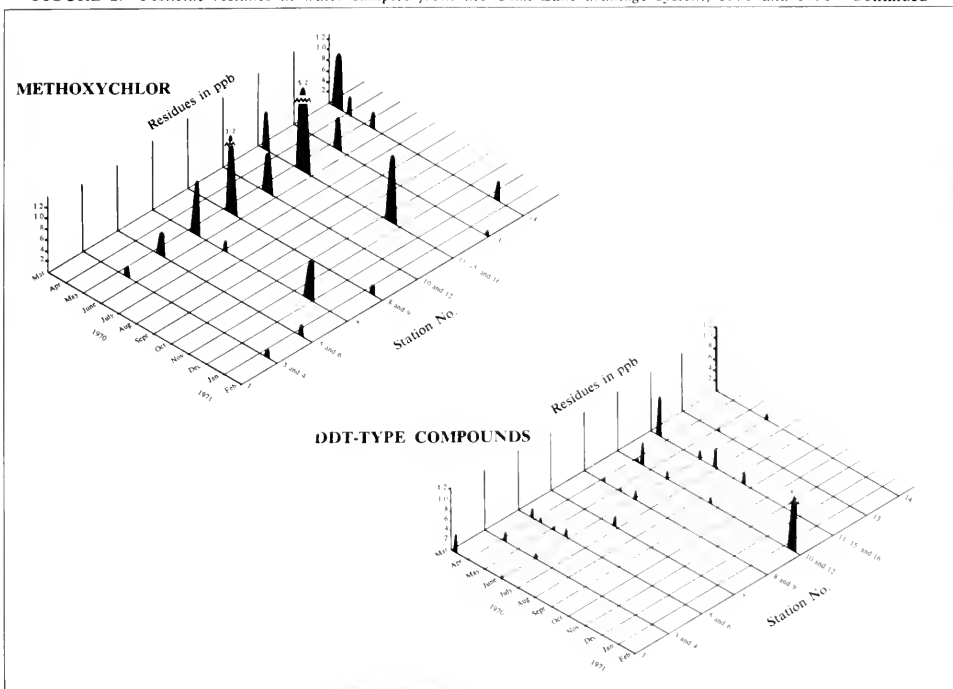


FIGURE 2.—Pesticide residues in water samples from the Utah Lake drainage system, 1970 and 1971—Continued



Except for isolated instances, DDE, the principal degradation product of DDT, was observed only at low concentrations in water samples (Fig. 2). The observed DDE could have been leached from the soil or washed into the waterway on soil particles. Indeed, every occurrence of DDE was preceded by a rain storm in the area (11). DDT was observed only once, at Station 12 in January 1971 at a high concentration (4.1 ppb). Very little DDT is presently being used in Utah Valley, and this high level may have been due to supplies of DDT having been dumped in Spanish Fork Canyon.

There are no known studies of the pesticide variations in the water of a small isolated basin such as the Utah Lake drainage system. There have been a number of national studies of pesticide residue levels in surface waters (10) in which one water sample from a large number of rivers was analyzed per year. The pesticides were found to be widespread throughout the country, but were below permissible limits as they relate to human intake directly from a domestic water supply (10).

FISH SAMPLES

Pesticide residue levels in the fish of Utah Lake have been reported previously (4, 5). In the study reported here, the most common pesticide observed in fish was DDE, probably due to its great persistence (Table 1). The smaller and younger fish contained lesser amounts of DDE.

It is evident that pesticide levels in fish were much higher than those found in water. Reinert (12) and Crosby and Tucker (13) have shown that accumulation of pesticides in fish results primarily from absorption from the water. Reinert (12) has shown that uptake of pesticides from water can be very rapid and that fish can attain up to 1 ppm of pesticide from water with concentrations as low as 1 ppt.

Although a limited number of fish samples were collected in this study, it is evident that the levels of pesticide residues in the fish in Utah Lake are very low when compared to those of fish in many other areas. Fish studied in the Midwest had from 2-10 ppm total pesti-

TABLE 1.—Pesticide concentrations in Utah Lake fish, 1970

FISH ¹	LENGTH (CM)	WEIGHT (G)	AGE ² (YEARS)	PESTICIDES ³ (PPB)
Carp (<i>Cyprinus carpio</i>)	39.5	844	3	302 DDE, 230 Dieldrin
	43.5	1,044	4	288 DDE, 215 Dieldrin
	45.5	1,036	4	123 DDE, 30 BHC, 62 Methoxychlor
	44.5	1,135	5	369 DDE, 56 Methoxychlor
	49	1,667	7	956 DDE
Channel catfish (<i>Ictalurus punctatus</i>)	51	1,264	6	652 DDE, 79 DDT
Bullhead catfish (<i>Ictalurus melas</i>)	25.5	274	2	126 DDE
White bass ⁴ (<i>Morone chrysops</i>)	19	85	2	53 Dieldrin

¹ Fish were caught by Utah State Wildlife Resources personnel on the east side of Utah Lake between the Orem Pier and Provo Airport on July 21, 1970.

² Determined from scales and spine cross sections.

³ Reported in parts per billion (ppb) of a whole fish.

⁴ Four fish were ground together to provide a large enough sample.

cides (4, 5) as compared to 0.05-0.96 ppm in Utah Lake. The major reason for the low pesticide residues in Utah Lake fish might be the fact that relatively little farming is carried out in the area as compared to the Midwest. Another possible reason is that the pesticides are being adsorbed by the suspended silt particles in Utah Lake; however, this was not confirmed in this study:

Acknowledgment

The authors wish to thank those who participated in the project by collecting and extracting the water samples, in particular, E. M. Michenor, B. Hague, J. F. Brown, E. Loveless, and M. Morrison, and to thank the Utah State Agriculture Agents for their assistance in evaluating the data. We wish also to thank Joe White, Utah State Wildlife Resources, who provided the fish samples.

See Appendix for the chemical names of compounds discussed in this paper.

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Sampling Procedures and Problems in Determining Pesticide Residues in the Hydrologic Environment¹

Herman R. Feltz² and James K. Culbertson³

ABSTRACT

Diligent use of standardized sampling and analytical techniques is essential to meaningful assessments of the occurrence, distribution, and fate of pesticide residues in the hydrologic environment. The validity of analytical data and subsequent interpretations are interdependent and limited to the confidence level of adequate, representative sampling of various components. Equally important are appropriate sample-preservation practices and procedures for sample preparation and cleanup and identification, measurement, and confirmation of residues. Analytical schedules should include pesticides listed in the Revised Chemicals Monitoring Guide for the National Pesticide Monitoring Program and should be responsive to special interests. Samplers are available for collecting acceptable water, fluvial material, and bottom-material samples in about 75% of the river miles and in lakes and estuaries throughout the United States; however, more experience is needed in sampling hydrosols and low density deposits at the active water-sediment interface.

Introduction

Scientific literature contains many reports pointing out the "pesticide problem" and presenting data used to assess the occurrence and distribution of pesticides in various components of the hydrologic environment. Most assessments have been thorough but limited to an individual component. Concentration levels in each aquatic phase must be related to the whole aquatic environment because each component exerts a control, creating an interdependence. To maintain a current

assessment, data from all investigators have become increasingly important because these data are used to derive trends in residue concentrations and to identify possible problem areas.

Because measuring pesticide residues in the Nation's water resources is far too great for any single group, contributors are numerous and represent Federal, State, private, and academic organizations. Therefore, standardized sampling and analytical techniques must be used to acquire the valid and interrelated data needed for meaningful assessments of the occurrence, distribution, and fate of pesticide residues. Standardization should not be rigid or deter technical advancement, but there should be some minimum criteria established for sampling and analysis which investigators should fulfill.

Pesticides are distributed throughout the hydrologic environment—the atmosphere, precipitation, surface water, ground water, suspended and bed sediments, flora, and fauna. Measurements in each component are equally important, and validity begins immediately upon sampling. Irrespective of scrutiny and quality control applied in performing laboratory analyses, reported data are no better than the confidence that can be placed in the representativeness of the sampling. Webster's dictionary states that "a sample is a representative part from a whole whose properties are studied to gain information about the whole." Because the whole cannot be analyzed, the sample thus becomes paramount.

Although a high degree of analytical capability has generally been attained, the use of adequate samplers and sampling techniques has not been emphasized. Perhaps, the greatest weakness in assessing the "pesticide situation" is the lack of representative samples for analysis.

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This paper directs attention to the state of the art, the problems, and some of the pitfalls in collecting representative samples from streams, lakes, and estuaries.

Occurrence of Pesticide Residues

Pesticide residues have been detected in all components of the dynamic hydrosystem, attesting to the ubiquitous nature of these residues (1, 10-13, 15). The occurrence and distribution of residues are as divergent as the components. Concentrations range from the generally accepted lower detection limit of 0.01 $\mu\text{g/liter}$ in filtrates to 100,000 times this value in particulate matter separated from the same aqueous samples. The environment ranges from the most humid to the driest, and residues may originate from local applications or as fallout transported from remote areas.

In assessing the quality of water resources, the significance of measuring residues throughout the aquatic system is shown by the wide range in concentrations found within a single hydrologic area as illustrated in Table 1. This table, based on results of recent work in south Florida, indicates the order of magnitude of total DDT (DDT+DDE+DDD) residue values which ranged from 0.02 $\mu\text{g/liter}$ in water up to 16,500 $\mu\text{g/kg}$ in birds at the top of the food chain. The biological accumulation of DDT family residues in the ecosystem is several hundred thousand times the average concentration found in surface and ground water.

The least valid assessment of the distribution of pesticide residues is one based on data from the aqueous phase alone. Although water is primarily considered when studying the aquatic environment, evaluating pesticide residues is not truly significant without relating all components as an interdependent system.

Sampling Techniques

The data presented in Table 1 represent a cross section of an entire ecosystem in which each component presents sampling problems. The following discussion, however, emphasizes the identification of problems in sampling water, fluvial sediment, and bottom materials.

WATER AND FLUVIAL SEDIMENT

Preliminary results of an investigation of pesticide residues in runoff to streams draining different land-use areas in central Pennsylvania indicated that the concentration of the DDT family sometimes is directly correlated with suspended-sediment concentration (L. A. Reed and J. F. Truhlar, *personal communication*, 1970). Samples were collected using a depth-integrating sampler and the equal-transit-rate sampling procedure. Correlation curves for two of the streams in the study area are shown in Fig. 1.

TABLE 1.—Scheme of biological accumulation of total DDT (DDT+DDD+DDE) in south Florida¹

ENVIRONMENTAL COMPONENT	CONCENTRATION OF TOTAL DDT (DDT+DDD+DDE) IN $\mu\text{g/LITER}$ FOR WATER AND $\mu\text{g/KG}$ FOR PLANTS AND ANIMALS
Water:	
Surface	.02
Ground (in Biscayne Aquifer)	.02
Rain	.30
Everglades algal mats or periphyton (producers)	7
Everglades vascular plants (producers)	8
Everglades submerged soils	16
Everglades crustaceans (omnivores)	23
Everglades marsh fishes (omnivores and primary carnivores)	196
Everglades alligators (higher carnivores)	352
Everglades kite eggs and eagle (higher carnivores)	16,500

¹ Data from U.S. Geological Survey except eagle and kite data from National Park Service (Department of the Interior).

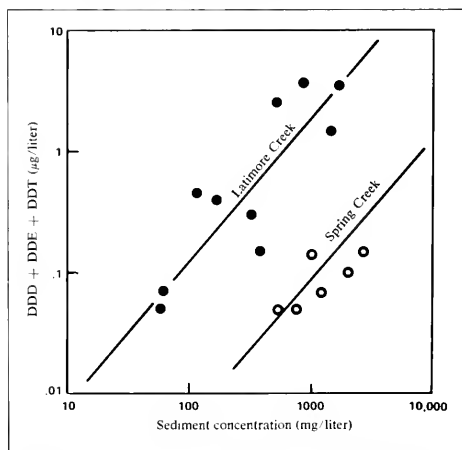


FIGURE 1.—Correlation between suspended-sediment concentration and total concentration of DDD+DDE+DDT in unfiltered water samples

More representative sampling of pesticide residues is based on a better understanding of the role of fluvial-sediment transport. Numerous reports reveal that residues associated with sediment are dependent upon both particle-size distribution and organic matter content (12). Therefore, the variability of pesticide concentrations may be correlated with the variability of suspended-sediment concentration in the cross section of a stream.

Water is usually sampled by filling a container held just beneath the surface of a body of water. Published data and discussions by investigators reveal that a high percentage of all samples have been obtained in this manner. This sample is commonly referred to as a dip sample. Using a weighted bottle holder which would allow the bottle to be lowered to any desired depth and returned to the surface would improve this method. If the person taking a sample could be assured that the bottle was lowered to the bottom and raised to the surface at a uniform rate, he would have roughly approached collection of what is known as a depth-integrated sample.

A true depth-integrated sample is collected by means of a depth-integrating sampler which integrates discharge as a function of depth (8, 18). Sediment is maintained in suspension because of velocity and turbulence. Velocity varies from the water surface to the stream bed, being generally highest near the surface and lowest at the bed. Sediment concentration varies also from the surface to the bed, being lowest at the surface and greatest at the bed. Fine sediment (finer than about 0.062 mm) is easily kept in suspension and is distributed relatively uniformly throughout the depth of flow. However, as particle size increases, more energy is required to maintain suspension in a given flow, and the average size of sediment in suspension increases from the water surface to the bed. Depth integration is used to collect a water-sediment sample that is weighted according to velocity at each increment of depth. This means that the water-sediment mixture must enter the sample container at the same velocity as the flow passing the intake. If the depth-integrating sampler is lowered from the water surface to the bed and back at the same transit rate, each increment of flow in that vertical is sampled proportionately to the velocity.

The open-mouth weighted-bottle sampler, therefore, does not collect a truly representative sample in a flowing stream if there are many particles coarser than about 0.062 mm carried in suspension (17). Another disadvantage to using an open-mouth bottle sampler in flowing streams is that there is no assurance when the bottle was filled, compounding the uncertainty that the sample collected truly represented the distribution of both dissolved and suspended material in the sampled water column. This method of sampling may be extremely poor for flowing streams but used effectively for slow-moving bodies of water, lakes, and estuaries.

Recent research studies of flow and sediment transport in the Rio Grande conveyance channel near Bernardo, N. Mex., a typical sand-bed channel (4), illustrates variability in the vertical and lateral distribution of sediment in a cross section. Velocity was measured (2, 3), and

suspended-sediment samples were collected (8, 18) at five points in each of six verticals in a cross section. Sediment samples were collected using a standard depth-integrating, suspended-sediment sampler modified to collect point integrated samples. Particle-size distributions were determined using standard laboratory, fall-velocity techniques. Concentration of sediment in the following size classes was determined for each sample: silt and clay (finer than 0.062 mm) and sand (between 0.062 and 2.0 mm).

Fig. 2 shows the velocity distribution in the 80-ft wide cross section. There were four cells of high velocity, resulting largely from the turbulent characteristics of the dune-bed form. The highest velocity was near mid-channel and the lowest velocity was along the right bank.

The distribution of concentration of the silts and clays is shown in Fig. 3. The open circles indicate the location of the six sampled verticals. Concentration values (mg/liter) are shown at the top of the figure. The upper values are the concentrations at the surface, and the lower values are the mean concentration in the vertical. The differences are evident; the surface samples generally were lower than the mean in the verticals, and in this case, averaged about 4% less than the mean in the cross section. The concentration at the surface of the right bank was the lowest in the cross section, coinciding with the low-velocity zone, and was about 92% of the mean in the cross section. A dip sample taken from the right bank would reflect an 8% minimum error with respect to fine suspended material.

Distribution of the concentration of sand is shown in Fig. 4. The distribution of sand is quite different from the distribution of silt and clay. Concentrations at the surface generally were about one-half the mean concentration in the verticals, and the concentration at the surface at the right bank was only about one-half the concentration at the surface at the left bank. The solid circles represent depths at which samples have to be collected to sample the mean concentration of sand in that vertical. In this case, which is rather typical in sand-channel streams, a surface sample collected along the right bank would represent only 26% of the mean in the cross section and is not representative of all sizes of particles in transport. A sample collected along the left bank would be about 47% of the mean. Thus, a real sampling problem related to sand in transport is apparent.

One sampling technique currently accepted by hydrologists for use in such situations is the equal-transit-rate method (ETR) (8). In this method, standard suspended-sediment samplers are used to collect a velocity-weighted

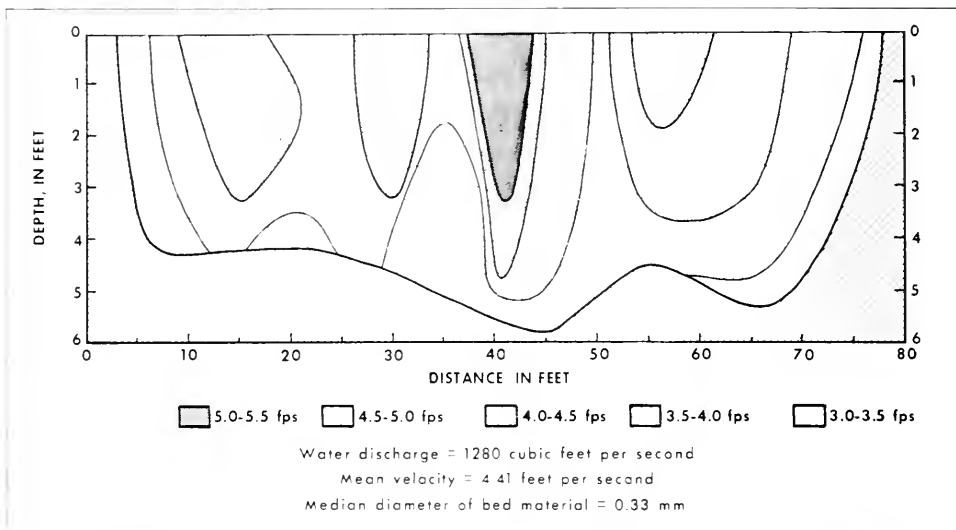


FIGURE 2.—Velocity distribution in cross section of Rio Grande conveyance channel near Bernardo, N. Mex.

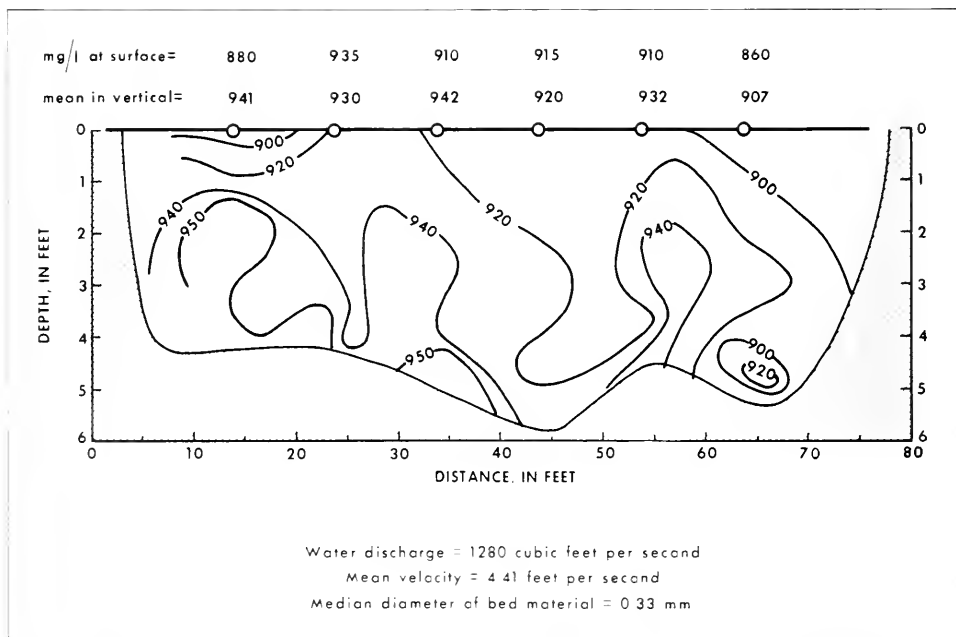


FIGURE 3.—Distribution of silt and clay (finer than 0.062 mm) in cross section of Rio Grande conveyance channel near Bernardo, N. Mex.

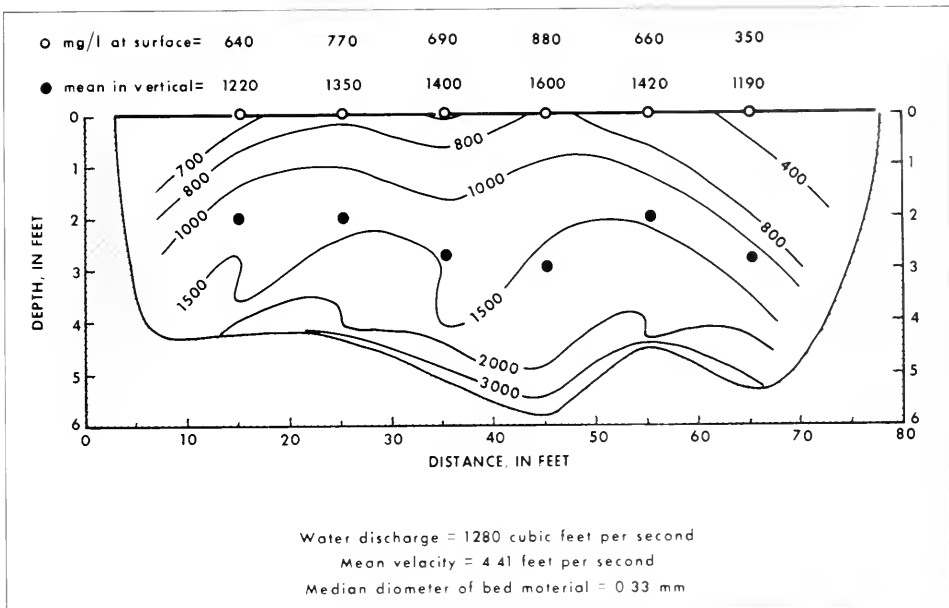


FIGURE 4.—Distribution of sand (between 0.062 mm and 2.0 mm) in cross section of Rio Grande conveyance channel near Bernardo, N. Mex.

sample. Samples are taken at a number of equally spaced verticals in the cross section depending on stream width. The transit rate of the sampler, which is the rate of movement of the sampler from the water surface to the stream bed and back to the surface, is the same at all verticals. Samples collected in each vertical are all composited into a single sample representative of the entire flow in the cross section. In this manner, the composite sample of the water-sediment mixture flowing in the cross section is velocity- and discharge-weighted.

Traditional dip samples, collected at the six points in the cross section and composited, would have contained concentrations of suspended sediment representative of about 96% of the silts and clays but only about 49% of the sands being transported. The discharge of silts and clays was at the rate of 3,200 tons per day, and sand at 4,700 tons per day; thus, the dip samples would have been representative of only 3,070 and 2,300 tons per day for fine and coarse material, respectively. Pesticides associated with about 32% of the suspended sediment in transport would not have been accounted for.

The significance of the sediment-distribution data presented is that without prior knowledge of the hydraulic

and sediment transport conditions at a given site, gross sampling errors may accrue. There is no justification in striving for 5% allowable error in the laboratory analysis of poorly collected samples that may represent only $\pm 30\text{-}40\%$ of the true water-sediment mixture. It is incumbent upon all investigators to recognize that pesticide concentrations currently reported for streams can reflect gross errors, and that these values may be extremely misleading in any assessment of residue distribution. Although a western sand-bed stream has been used for illustration, the authors recommend that the ETR sampling technique described herein should be used in all flowing streams.

BOTTOM MATERIALS

Pesticides associated with bottom material, irrespective of the ratio of inorganic to organic composition, may reflect an integration of chemical and biological processes. They serve the indispensable historical role by reflecting input to non-scouring streams, lakes, and estuaries with respect to time, application of pesticides, and land use. Recalling that fluvial materials tend to settle out during periods of low streamflow or of calm conditions in lakes and are additive to solids that have accumulated on stream beds or in lake bottoms, periodic

sampling of water overlying these deposits might not reveal the presence of pesticides, even though they may be present in the solid material which acts as a sink and reservoir.

The loss of low-density deposits must be kept minimal during any sampling process, requiring a bottom-material sampler that is capable of collecting and retaining the "fines" which sometimes contain the highest concentration of pesticide residues.

Few data have appeared in the literature regarding the presence of pesticide residues at the liquid-solid interface or in the hydrosol area. Depending upon ambient physical, biological, and chemical controls, residues may be transformed into metabolites, degraded, or taken into solution. Further study of these processes is an open and challenging field of endeavor. An evaluation of this portion of the hydrosystem is essential to comprehensive assessments of the occurrence and distribution of pesticides.

Sample Collection and Preservation

To overcome the problems associated with collecting representative samples, equipment that has been specifically designed and thoroughly tested is favored. There are several depth-integrating samplers available that are suitable. In shallow streams and wetlands that can be waded, the US DH-59 suspended-sediment sampler (Fig. 5) can be used successfully. The sampler is simple and of clean design. A sample container is easily inserted into position and held firmly by spring action.

The US D-49 suspended-sediment sampler (Fig. 6) has been used for many years in collecting depth-integrated suspended-sediment samples in the larger streams and rivers. It is provided with a choice of nozzles (shown beneath the tail fins), $\frac{1}{8}$ -, $\frac{3}{16}$ -, and $\frac{1}{4}$ -inch in diameter, with which inflow of the water-sediment mixture can be controlled. The sampler, which weighs about 60 pounds, is suspended on a cable and operated with a reel attached to a boom. The open-mouth, weighted-bottle sampling technique used in the National Pesticide Monitoring Program is acceptable for sluggish streams, lakes, and reservoirs (5, 7).

Bottom deposits present a more difficult set of sampling conditions than those in flowing streams or in lakes, primarily because of the varying firmness of bed materials. The US BMH-60 bed-material sampler (Fig. 7), is a 30-pound impact type sampler designed primarily for use in sand-bed streams. However, it works equally well for firm and partly consolidated bottom materials. One man can collect samples with this sampler under most conditions. The bucket takes a bed-material sample

that is approximately 2.2 x 5 x 1.7 inches. This sampler can be used in about 75% of the river miles across the United States. It also can be used in many lakes and reservoirs to collect excellent samples (Fig. 8). The equipment is described in detail in references 8 and 18.

Numerous investigators prefer to collect core samples when interest is primarily in historical data and when the bed is sufficiently compacted to be sampled with a coring device. The value of this type of sample should not be minimized; however, fresh deposition is of prime importance in a continuous evaluation of the occurrence, distribution, and movement of pesticide residues.

Most core samplers lack positive seals to hold a core or moist sample in place as the sampler is withdrawn from the water. The Controlled-Depth Volumetric Bottom Sampler described by Jackson (9), and the Core Sampler for *in situ* Freezing of Benthic Sediments designed by Gleason and Ohlmacher (6) are examples of efforts to overcome the drawbacks of the common core sampler and to provide the investigator with reliable equipment.

Little progress has been made in developing equipment to collect representative samples at the liquid-solid interface because it is difficult to sample without disturbance. The Gleason core sampler offers an advantage in sampling this interface because it can be used as a point sampler. The Hydrosol Sampler designed by Lawrence (14) may be used in many sampling situations. Reporting of experiences in using these and other hydrosol samplers will help to determine the adequacy of the samplers.

Deteriorated samples negate all the effort and cost expended in obtaining good samples. Samples of water plus suspended material should be forwarded to the laboratory in cleansed glass containers by the fastest transportation available. Bottom materials can be placed in the same type of container as suspended-sediment samples when they have a high percentage of water; solid bed-material samples may be sealed in foil. All samples should be kept near freezing, and extractions should be carried out in minimum elapsed time, preferably within 2 days from time of sample collection. Fresh plant material should be wrapped in foil and kept chilled or frozen; fauna should be wrapped in foil and frozen.

Sampling Frequency

In conducting reconnaissance studies, defined here as short-term, one-time evaluations, both bottom deposits and the overlying water should be sampled at each site. Monitoring, which consists of repetitive, continuing measurements to define variations and trends at a given



FIGURE 5.—US DH-59 suspended-sediment sampler



FIGURE 6.—US D-49 suspended-sediment sampler

location, should include collection of water samples during each of the four seasons, with particular emphasis on the fall and spring collections. Most flood events should be sampled. Bed-material samples should be collected for analysis of fresh deposits at least once per year at monitoring sites, and preferably during both the spring and fall seasons (5).

Evaluation of the variability in available pesticide data must precede any decision as to the number of samples and collection frequency required to maintain an effective monitoring program.

Analytical Schedule

To use fully all reported data, analytical and quality-control procedures must be agreed upon and standardized. Data contributions by all investigators constitute what may be regarded as a National Data Bank. Each piece of information should be virtually equivalent in terms of accuracy and reliability. To insure this condition, analytical schedules should be derived from consideration of the persistence, toxicity, and total usage of pesticides, the number of reported positive identifications, and the listings in the Revised Chemicals Monitoring Guide for the National Pesticide Monitoring Program (16). Schedules must be flexible and responsive to local problems. All significant peaks on chromatograms should be identified and quantified, if possible, and a report made of products that are pesticidal or toxic. Identification and quantification by dual-column electron-capture gas chromatography should be considered minimal. Back-up tools are essential, and confirmations can be made by use of a third column with diverse retention times, thin layer chromatography, column chromatography, microcoulometry, flame-photometric detection, infrared, and mass spectrometry (5).



FIGURE 7.—US BMH-60 bed-material sampler in use on South River below Atlanta, Ga.



FIGURE 8.—Bottom material being released from BMH-60 sampler

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Organochlorine Insecticides in Surface Waters in Germany—1970 and 1971¹

Fritz Herzel

ABSTRACT

As part of a series of studies initiated in 1969 to determine the organochlorine insecticide content of major waters in the Federal Republic of Germany, unfiltered water and suspended solids were analyzed from approximately 25 sites sampled in May 1971, and unfiltered water was analyzed from 7 sites sampled monthly from April 1970—June 1971. As in former studies (June and October 1969, April and September 1970), the insecticide concentrations found in waters and suspended solids were almost exclusively in the ppt range (ng/liter).

The compounds found most frequently were γ -BHC (lindane) and α -BHC; α - and β -endosulfan were detected in the Main, Regnitz, and Rhine Rivers. DDT and particularly its metabolites DDD and DDE were found infrequently except in samples from the Berlin Teltowkanal. Findings of heptachlor, heptachlor epoxide, dieldrin, and parathion (the only organophosphorus insecticide included in the study) were rare.

Introduction

Extensive surveys have been carried out to determine the presence of chlorinated pesticides in surface waters (1-14, 18-36); however, until recent years studies of this type had not been conducted in the Federal Republic of Germany. To obtain data on the organochlorine insecticide content of surface waters in the Federal Republic, a series of studies was initiated in 1969. The overall program consisted of the analysis of water and suspended solids or sediment samples obtained during five sampling excursions and water samples collected each month over the period April 1970-June 1971. The five sampling excursions which extended, as a rule, over a period of 1 week involved sampling the major bodies of water in the Federal Republic at approximately 25 pre-

selected sites. Because some organochlorine insecticides are almost insoluble in water but will adsorb to suspended matter, in addition to the analysis of water samples, suspended solids were filtered from other water samples collected at most of these same sites and analyzed separately. The sampling excursions were as follows:

- (1) June 1969—water only, with additional sampling at some sites which had been designated as potentially endangered, e.g., waters in fruit- and hop-growing areas, agricultural drainage ditches, etc.;
- (2) October 1969—water and suspended solids;
- (3) April 1970—water and sediment;
- (4) September 1970—water and suspended solids; and
- (5) May 1971—water and suspended solids.

Because of unfavorable weather conditions, a sixth sampling excursion scheduled for the winter months did not take place.

Results from the first four sampling excursions have been published elsewhere (15, 16). This paper presents the results of the May 1971 excursion during which approximately 25 sites were sampled for water and suspended sediments; data are also reported on the analysis of water samples collected monthly from 7 sites during the period April 1970-June 1971. Specific locations of sampling sites are shown on Fig. 1.

Sampling Procedures

One of the most difficult problems in sampling surface waters is obtaining a representative sample, particularly in flowing waters, since concentrations vary considerably

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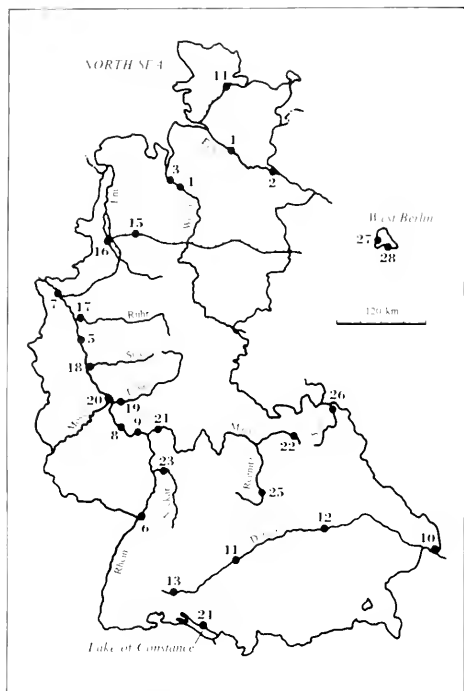


FIGURE 1.—Location of sites for sampling surface waters in the Federal Republic of Germany

with time in a downstream direction as well as horizontally and vertically across the stream. Theoretically, these variations can be overcome with an ideal device for continuous sampling; however, the general experience of this investigator with continuous sampling devices has been unfavorable, because they did not collect suspended matter in a uniform proportion to water collected.

WATER SAMPLES

Water samples were collected in 1-liter round-bottom flasks attached with a clamp to the end of a 2-m steel rod. The flasks were filled by moving them evenly up and down at an approximate depth of 50 cm. Before this, 0.5 ml toluene and 30 mg mercuric chloride had been added to the flasks as preservatives for the water samples; the flasks were stored in heat-insulation fitted transport boxes of foamed polystyrene, and sent to the laboratory with as little delay as possible.

SUSPENDED SOLIDS

Water from which suspended solids were obtained was collected in a steel bucket that was cleaned between

samplings with alcohol. Suspended solids were filtered from the water using a Buchner funnel (32 cm in diameter) fitted with filter paper (Selecta No. 1450) that had been carefully pre-extracted with acetone. The water was drawn through the filter into a 10-liter suction flask and a vacuum pump operated by a portable small-size generator until the filter was filled to capacity. The paper filters holding the filtration residues were folded and put into 100-ml flasks each containing 20 ml of a 1:1 mixture of hexane and acetone to reduce biological degradation.

An effort was made to select sampling sites with sufficient depth to avoid stirring of sediment at the bottom, particularly where samples were taken with a bucket. In several cases, however, the water was too shallow and samples for analysis of suspended solids could not be collected. Sampling of water for water analysis (using flasks) and water for obtaining suspended solids (using buckets) took place at the same site whenever possible; however, bank conditions did not permit this in all instances.

Analytical Procedures

PREPARATION OF SAMPLES

Water Samples

Water samples were extracted in a separatory funnel with subsequent quantities of 15, 10, and 10 ml of hexane, (b.p. 68-70° C, column distilled). The extracts were then combined and without cleanup (17) were analyzed by electron capture gas-liquid chromatography (EC-GC).

Suspended Solids (Filtration Residues)

Solvents used in extraction and cleanup of suspended solids were hexane, b.p. 68-70° C; acetone, reagent grade; and benzene, reagent grade—each column distilled.

Suspended solids, which had been collected on filters and placed in a mixture of acetone and hexane, were extracted with an additional 50 ml of a 1:1 mixture of acetone-hexane in a Soxhlet apparatus, and poured into 5 times their volume of water. The nonpolar phase was separated, reduced, and then cleaned up on a Florisil column, 20 cm by 10 mm, i.d., filled with 7 cm of Florisil, 60/100 mesh, and 1 cm of anhydrous sodium sulphate on top; the Florisil had been prepared by heating at 130° for 16 hours and 3% w/w of water added, and the anhydrous sodium sulphate by heating at 400° C for 16 hours. The column was pretreated with hexane, and the samples were eluted with 30 ml of a 1:1 mixture of hexane and benzene. First, these eluates were analyzed by EC-GC. Then, they were evaporated to dryness in a rotary vacuum evaporator

and taken up in 0.1 ml amylacetate for subsequent microcoulometric gas chromatography and thin layer chromatography. The extracts from the water samples were treated likewise after their EC-GC detection.

Previous experience has shown that in using a rotary vacuum evaporator, there is no risk of volatilization of insecticide from extracts even if complete reduction to dryness takes place. Even in pure solutions, as contrasted with air drying, there were no notable losses when the procedure took place at room temperature and when the plunger was removed from the vacuum as soon as the solvent had evaporated. To insure that there was no loss of insecticides, a few milliliters of decyl acetate was added.

GAS CHROMATOGRAPHY

Gas chromatographic determination was carried out on Aerograph gas chromatographs, models 205-B and 1740, equipped with two electron capture detectors (tritium) and, additionally, a microcoulometric detector for confirmation. The operating parameters were as follows:

Columns:	Glass, 1/8" by 5' packed with 2-3% Dow-11, OV-101, XE-60, and Dexsil 300 on 80/100 mesh Chromosorb W, AW, DMCS treated
Temperatures:	Injector—210° C Column—185° C Detector—195° C
Recorder:	1 mv full scale deflection

The detection limits of organochlorine insecticides in 30 ml of extract were between 5 and 10 ng for hexachlorobenzene (HCB) and the hexachlorocyclohexane isomers α - and γ -BHC (lindane), 100-200 ng for DDT and its derivatives, and 10-30 ng for the remaining organochlorines. In a number of cases, particularly DDT, the sample was concentrated to one-tenth of the original volume, thus lowering the detection limit by one power of ten.

The detection limit for parathion was 1 ng; quantitative determination was based on the quantity of 0.1-ml amylacetate which had been used, as described above, to take up the residue from the 30-ml hexane extract. An alkali FID was employed for quantitative gas chromatographic determination of parathion because of its comparatively low susceptibility in respect to high concentrations of interfering substances in a concentrated extract of this type. Since the concentration per liter of parathion in suspended solids was based on the amount of residue detected divided by the number of liters of water filtered, very low concentrations could be detected.

RECOVERY

The recovery rates from water samples were checked repeatedly; reproducibility was quite good, at rates exceeding 90% in all cases. The recovery values of residues

in suspended solids varied between 80% and 95%, depending on the insecticide residue. Results were not corrected for percent recovery.

CONFIRMATION STUDIES

In doubtful cases, prepreparation by thin layer chromatography (TLC) was used for further confirmation of results. After the volume of the extract had been reduced to dryness and then taken up by 0.1 ml of amyl acetate, approximately one-half the amount (0.05 ml) was placed on 20- by 20-cm TLC plates coated with 0.3-mm silica gel (Kieselgel HF Merck) which had been dried 1 hour at 105° C. Hexane was used as the solvent, and the suspected substances served as references. After removal of the corresponding zone and elution by 1 ml of benzene, gas chromatographic analysis using an electron capture detector was repeated.

As a rule, a particular insecticide was considered present only if corresponding peaks were obtained from all GC columns; therefore, a major number of peaks were considered unidentifiable. Prepreparation by TLC was performed in addition when the comparatively low microcoulometric reading of less sensitive substances like DDT was obtained. The polychlorinated biphenyls were not considered present, however, without confirmation by mass spectrometry even where typical fingerprints were observed. These compounds had not been detected in any samples taken during this series of studies.

Results and Discussion

Results of analyses of unfiltered water and suspended solids are given in Tables 1 and 2. Residue levels in water are expressed as ng liter (ppt) and values obtained from suspended solids as nanograms of insecticides in solids suspended in 1 liter of water, i.e., the quantities obtained from the filter residues were divided by the number of liters of water that had been filtered.

Generally, insecticide concentrations were low in both types of samples. In water samples, heptachlor, heptachlor epoxide, and dieldrin were found seven times, three times, and once, respectively. In the suspended solids heptachlor was detected only once, and aldrin, dieldrin, and heptachlor epoxide were not found in any of the samples. The results were similar to those from earlier studies. Concentrations of endosulfan, however, were clearly lower as compared with earlier studies; residues of this compound, originating from industrial effluents, were found in samples from the Rhine, the lower Main (downstream from its junction with the Rhine), and the Regnitz.

The insecticides found most frequently were the hexachlorocyclohexane isomers, α -BHC and lindane (γ -BHC); however, the proportion of these compounds

found in suspended solids was low because of their greater solubility in water. Because of technical difficulties, only a few samples were analyzed for α -BHC and hexachlorobenzene (HCB), and concentrations were determined with certainty only in some samples of suspended solids. The results obtained, however, showed a notable increase of lindane and α -BHC concentrations in samples from the upper Rhine and at the Berlin Teltowkanal over values obtained in previous studies.

DDT and its main metabolites, DDD and DDE, occupied a subordinate rank with regard to the frequency of positive findings and concentrations in waters. Because of extremely low solubility in water, DDT was found mainly adsorbed to suspended solids.

Parathion, the only organophosphate analyzed in this study, was found in only a few samples and in very small amounts.

In evaluating the degree of pollution of major German surface waters by organochlorine insecticides, results obtained in this study as well as former studies show that the Rhine River is the most contaminated.

Acknowledgment

A great number of samples were furnished regularly from various sources. The author wishes to express his gratitude, in particular, to the Hygienisches Institut, Hamburg; Wasserwerke Bremen AG; the Düsseldorf Field Station of this Institute; the Institut für Gastechnik, Feuerungstechnik und Wasserchemie, Technical University Karlsruhe, and Regierung von Niederbayern, Abt. Gewässergüteaufsicht, Landshut. Special thanks for technical assistance are expressed to Mrs. H. Junge and Mrs. F. Reichert.

See Appendix for chemical names of compounds discussed in this paper.

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TABLE 1.—Concentrations of insecticides in unfiltered surface waters in the Federal Republic of Germany—May 1971 sampling excursion and monthly collections from April 1970-June 1971

[blank = not analyzed, — = not detected]

SAMPLING SITE NO. (SEE FIG. 1)	BODY OF WATER	LOCATION	DATE OF SAMPLING	INSECTICIDE CONCENTRATIONS IN NG LITER (PPT)														
				α-BHC	LINDAN	HEPTACHLOR	HEPTACHLOR EPOXID	DIELDRIN	α-CYHUTHIOLAN	β-CYHUTHIOLAN	DDD	DDD	DDT	PARATHION				
1	Elbe	Hamburg	5 21 70	—	215	—	—	—	—	—	—	—	—	—	—	—		
			6 11 70	—	250	—	—	—	—	—	—	—	—	—	—	—	—	
			7 23 70	—	395	—	—	—	—	—	—	—	—	—	—	—	—	
			8 70	—	310	—	—	—	—	—	—	—	—	—	—	—	—	
			9 01 70	—	175	—	—	—	—	—	—	—	—	—	—	—	—	
			10 28 70	—	125	—	—	—	—	—	—	—	—	—	—	—	—	—
			11 05 70	—	160	—	—	—	—	—	—	—	—	—	—	—	—	—
			12 14 70	—	185	20	—	—	—	—	—	—	—	—	—	—	—	—
			1 27 71	—	810	255	—	—	—	—	—	—	—	—	—	—	—	—
			2 25 71	—	1500	430	—	—	—	—	—	—	—	—	—	—	—	—
			5 03 71	—	1050	275	—	—	—	—	—	—	—	—	—	—	—	—
6 15 71	—	690	250	—	—	—	—	—	—	—	—	—	—	—	—			
2	Elbe	Lauenburg	5 10 71	—	145	—	25	—	—	—	—	—	—	—	165	—		
			3	Weser	Bremen	4 13 70	—	10	—	—	—	—	—	—	—	—	—	—
5 11 70	—	5	—			—	—	—	—	—	—	—	—	—	—			
6 08 70	—	30	—			—	—	—	—	—	—	—	—	—	—	—		
7 13 70	—	25	—			—	—	—	—	—	—	—	—	—	—	—		
8 10 70	—	30	—			—	—	—	—	—	—	—	—	—	—	—		
9 07 70	—	20	—			—	—	—	—	—	—	—	—	—	—	—		
10 12 70	—	25	10			—	—	—	—	—	—	—	—	—	—	—		
11 09 70	—	30	—			—	—	45	—	—	—	—	—	—	—	—		
12 07 70	—	5	—			—	—	—	—	—	—	—	—	—	—	—		
1 11 71	—	5	—			—	—	—	—	—	—	—	—	—	—	—		
2 08 71	—	10	—			—	—	—	—	—	—	—	—	—	—	—		
3 08 71	—	5	—	—	—	—	—	—	—	—	—	—	—	—				

TABLE 1.—Concentrations of insecticides in unfiltered surface waters in the Federal Republic of Germany—
May 1971 sampling excursion and monthly collections from April 1970-June 1971—Continued

SAMPLING SITE NO. (SEE FIG. 1)	BODY OF WATER	LOCATION	DATE OF SAMPLING	INSECTICIDE CONCENTRATIONS IN NG/LITER (PPT)														
				α -BHC	LINDANE	HEPTACHLOR	HEPTACHLOR EPOXIDE	DIELDRIN	α -ENDOSULFAN	β -ENDOSULFAN	DDE	DDD	DDT	PARATHION				
3	Weser	Bremen	4/13/71	—	30	—	—	—	—	—	—	—	—	—	—	—		
			5/10/71	—	50	—	—	—	—	—	—	—	—	—	—	—	—	
			6/14/71	—	60	—	—	—	—	—	—	—	—	—	—	—	—	
4	Weser	Achim	5/10/71	25	45	—	—	—	—	—	—	—	—	—	—	—		
			5	Rhine	Düsseldorf	4/70	720	245	—	—	—	70	—	—	—	—	—	—
6/70	215	—	—			—	—	—	—	—	—	—	—	—	—			
7/70	105	—	—			—	—	—	—	—	—	—	—	—	—	—		
8/70	165	—	—			—	—	55	95	—	—	—	—	—	—	—		
9/70	105	—	—			—	—	35	—	—	—	—	—	—	—	—		
10/70	140	—	—			—	—	15	20	—	—	—	—	—	170	—		
11/70	100	—	—			—	—	15	—	—	—	—	—	—	—	—		
12/70	220	—	—			—	—	—	—	—	—	—	—	—	—	—		
1/71	905	115	205			—	—	—	—	—	—	—	—	—	—	—	55	
2/71	555	175	—			—	—	—	—	—	—	—	—	—	—	—	5	
3/71	125	110	—			—	—	—	—	—	—	—	—	—	—	—	—	
6	Rhine	Karlsruhe	4/15/70			1900	380	—	—	—	—	—	—	—	—	—	—	—
			5/14/70	155	130	—	—	—	—	—	—	—	—	—	—	—	—	
			6/70	50	—	—	—	—	—	—	—	—	—	—	—	—	—	
			7/15/70	60	—	—	—	—	—	—	—	—	—	—	—	—	—	
			8/10/70	225	—	—	—	—	—	—	—	—	—	—	—	—	—	
			9/70	455	—	—	—	—	—	—	—	—	—	—	—	—	—	
			10/13/70	125	—	—	—	—	—	—	—	—	—	—	—	—	55	
			11/09/70	265	—	—	—	—	—	—	—	—	—	—	—	—	—	
			12/10/70	175	—	—	—	—	—	—	—	—	—	—	—	—	—	
			1/13/71	1000	325	—	—	—	—	—	—	—	—	—	—	—	—	
			2/71	945	330	—	—	—	—	—	—	—	—	—	—	—	—	5
			3/16/71	1100	330	—	—	—	—	—	—	—	—	—	—	—	—	
			4/71	1300	260	—	—	—	—	—	—	—	—	—	—	—	—	
5/71	2400	535	—	—	—	—	—	—	—	—	—	—	—	—				
5/12/71	890	240	—	—	—	—	—	—	—	—	—	—	—	—				
7	Rhine	Wesel	5/11/71	115	155	—	—	—	—	—	—	—	—	—	—			
8	Rhine	St. Goar	5/11/71	110	260	—	—	—	—	—	—	—	—	—	—			
9	Rhine	Oestrich	5/12/71	550	130	—	—	—	—	55	—	—	—	—	—			
10	Danube	Jochenstein	4/15/70	—	5	—	—	—	—	—	—	—	—	—	—	—		
			5/13/70	5	20	—	—	—	—	—	—	—	—	—	—	—		
			6/10/70	30	—	—	—	—	—	—	—	—	—	—	—	—		
			7/08/70	25	—	—	—	—	—	—	—	—	—	—	—	—		
			8/19/70	20	—	—	—	—	—	—	—	—	—	—	—	—		
			9/16/70	25	—	—	—	—	—	—	—	—	—	—	—	—		
10/14/70	15	—	—	—	—	—	—	—	—	—	—	—	—					

TABLE 1.—Concentrations of insecticides in unfiltered surface waters in the Federal Republic of Germany—
May 1971 sampling excursion and monthly collections from April 1970-June 1971—Continued

SAMPLING SITE No. (SEE FIG. 1)	BODY OF WATER	LOCATION	DATE OF SAMPLING	INSECTICIDE CONCENTRATIONS IN NG LITER (PPT)													
				α -BHC	LINDANE	HEPTACHLOR	HEPTACHLOR EPOXIDE	DILUOREN	α -ENDOSULFAN	β -ENDOSULFAN	DDE	DDD	DDT	PARATHION			
10	Danube	Jochenstein	11/11/70		45	—	—	—	—	—	—	—	—	—	—	—	
			12/09/70		35	—	—	—	—	—	—	—	—	—	—	—	—
			1/20/71		—	5	—	—	—	—	—	—	—	—	—	—	—
			2/17/71		—	30	—	—	—	—	—	—	—	—	—	—	—
			3/17/71		—	15	—	—	—	—	—	—	—	—	—	—	—
			4/14/71		—	30	—	—	—	—	—	—	—	—	—	—	—
			5/12/71		—	5	—	—	—	—	—	—	—	—	—	—	—
			6/09/71		50	40	—	—	—	—	—	—	—	—	—	—	—
11	Danube	Ulm	5/13/71	—	25	—	—	—	—	—	—	—	—	—	—		
12	Danube	Ingolstadt	5/14/71	40	30	—	—	—	—	—	—	—	—	—	—		
13	Danube	Geisingen	5/13/71	20	40	—	—	—	—	—	—	—	—	—	—		
14	Nord-Ostsee-Kanal	Rendsburg	5/10/71	—	—	—	—	—	—	—	—	—	—	—	—		
15	Mittelland-Kanal	Bramsche	5/11/71	—	15	—	35	—	—	—	—	—	—	—	—		
16	Ems	Rheine	5/11/71	—	—	—	—	—	—	—	—	—	—	—	—		
17	Ruhr	Duisburg	5/11/71	—	—	—	—	—	—	—	—	—	—	—	—		
18	Sieg	Siegburg	5/11/71	295	615	—	—	—	—	—	—	—	—	—	—		
19	Lahn	Fachbach	5/11/71	—	—	—	—	—	—	—	—	—	—	—	—		
20	Moselle	Koblenz	5/11/71	25	20	—	—	—	—	—	—	—	—	—	—		
21	Main	Raunheim	5/12/71	170	64	—	—	—	100	45	—	—	—	—	—		
22	Main	Bad Berneck	5/14/71	5	30	—	—	—	—	—	—	—	—	—	—		
23	Neckar	Heidelberg	5/12/71	—	—	—	—	—	—	—	—	—	—	—	—		
24	Lake Constance	Langenargen	5/13/71	40	25	—	—	—	—	—	—	—	—	—	—		
25	Regnitz	Erlangen	5/14/71	170	140	20	40	—	10	—	—	—	—	100	—		
26	Saale	Hof	5/15/71	95	55	—	—	—	—	—	—	—	—	—	—		
			4/29/70	10	10	—	—	—	—	—	—	—	—	—	—		
27	Havel	Berlin-Gatow	5/22/70	—	5	—	—	—	—	—	—	—	—	—	—		
			6/24/70		50	—	—	—	—	—	—	—	—	—	—	—	
			7/08/70		80	—	—	—	—	—	—	—	—	—	—	—	
			8/12/70		40	—	—	—	—	—	—	—	—	—	—	—	
			9/17/70		55	—	—	—	—	—	—	—	—	—	—	—	
			10/27/70		20	—	—	—	—	—	—	—	—	—	—	45	
			11/02/70		45	—	—	—	—	—	—	—	—	—	—	—	
			12/01/70		80	15	—	—	—	—	—	—	—	—	—	—	
			1/21/71		5	70	—	—	—	—	—	—	—	—	—	—	
			2/10/71		5	5	—	—	—	—	—	—	—	—	—	—	
3/25/71		95	25	25	—	—	—	—	—	—	—	—	—				
4/21/71		165	50	30	—	—	—	—	—	—	—	—	—				
5/12/71		190	70	—	—	—	—	—	—	—	—	—	—				
6/21/71		115	55	—	—	—	—	—	—	—	—	—	—				

TABLE 1.—Concentrations of insecticides in unfiltered surface waters in the Federal Republic of Germany—
May 1971 sampling excursion and monthly collections from April 1970-June 1971—Continued

SAMPLING SITE NO. (SEE FIG. 1)	BODY OF WATER	LOCATION	DATE OF SAMPLING	INSECTICIDE CONCENTRATIONS IN NG/LITER (PPT)													
				α -BHC	LINDANE	HEPTACHLOR	HEPTACHLOR EPOXIDE	DIELDRIN	α -ENDOSULFAN	β -ENDOSULFAN	DDE	DDD	DDT	PARATHION			
28	Teltow-Kanal	Berlin-Lichterfelde	4/29/70	135	170	—	—	—	—	—	—	—	—	—	—	—	
			5/11/70	515	950	—	—	—	—	—	—	—	—	—	—	—	—
			6/24/70		370	—	—	—	—	—	—	—	—	—	—	65	—
			7/08/70		420	—	—	—	—	—	—	—	—	250	—	—	—
			8/12/70		190	—	—	—	—	—	—	—	—	95	—	—	—
			9/17/70		450	—	—	—	—	—	—	—	—	190	—	—	—
			10/27/70		60	—	—	—	—	—	—	—	—	80	60	—	—
			11/02/70		1350	—	—	—	—	—	—	—	35	180	—	—	—
			12/01/70		7100	—	—	—	—	—	—	85	—	135	—	—	—
			1/21/71		415	1650	—	—	—	—	—	—	—	85	—	—	—
			2/10/71		1700	800	—	—	—	—	—	—	—	130	—	65	—
			3/25/71		400	1750	—	—	—	—	—	—	—	200	—	—	—
			4/21/71		530	715	—	—	—	—	—	—	—	830	—	—	—
			5/12/71		545	540	—	—	—	—	—	—	70	775	—	—	—
6/21/71		1245	80	—	—	—	—	—	—	—	—	—	—	—			

TABLE 2.—Concentrations of insecticides in suspended solids from surface waters in the Federal Republic of Germany—
May 1971

[— = not detected]

SAMPLING SITE NO. (SEE FIG. 1)	BODY OF WATER	LOCATION	DATE OF SAMPLING	LITERS OF WATER FILTERED	INSECTICIDE CONCENTRATIONS IN NG/SOLIDS SUSPENDED IN 1 LITER OF WATER (PPT) ¹								
					HCB	α -BHC	LINDANE	HEPTACHLOR	α -ENDOSULFAN	β -ENDOSULFAN	DDT ²	PARATHION	
2	Elbe	Lauenburg	5/10/71	18	57	—	37	—	—	—	—	36	—
4	Weser	Achim	5/10/71	14	—	—	53	—	—	—	—	—	—
6	Rhine	Karlsruhe	5/12/71	18	—	2.5	21	—	—	—	—	—	—
7	Rhine	Wesel	5/11/71	12	—	—	53	—	—	—	—	—	0.4
8	Rhine	St. Goar	5/11/71	21	—	37	22	—	—	—	—	—	—
9	Rhine	Oestrich	5/12/71	12	36	42	39	—	—	—	—	—	0.15
11	Danube	Ulm	5/13/71	34	—	—	0.4	—	—	—	—	—	—
12	Danube	Ingolstadt	5/14/71	24	0.6	—	—	2	—	—	—	4.4	—
14	Nord-Ostsee-Kanal	Rendsburg	5 10 71	32	—	—	—	—	—	—	—	—	—
15	Mittelland-Kanal	Bransche	5/11/71	13	—	—	3.1	—	—	—	—	—	—
16	Ems	Rheine	5/11/71	12	—	—	0.8	—	—	—	—	—	—

TABLE 2.—Concentrations of insecticides in suspended solids from surface waters in the Federal Republic of Germany—
May 1971—Continued

SAMPLING SITE No. (SEE FIG. 1)	BODY OF WATER	LOCATION	DATE OF SAMPLING	LITERS OF WATER FILTERED	INSECTICIDE CONCENTRATIONS IN NG SOLIDS SUSPENDED IN 1 LITER OF WATER (PPT) ¹								
					HCB	α-BHC	LINDANE	HEPTACHLOR	α-ENDOSULFAN	β-ENDOSULFAN	DDT ²	PARATHION	
17	Ruhr	Duisburg	5/11/71	20	—	—	12	—	—	—	—	—	—
18	Sieg	Siegburg	5/11/71	20	—	—	—	—	—	—	—	—	—
19	Lahn	Fachbach	5/11/71	12	1	—	2.1	—	—	—	—	8.7	—
20	Moselle	Koblenz	5/11/71	25	0.6	—	1.2	—	—	—	—	4.2	—
21	Main	Raunheim	5/12/71	13	19	25	18	—	22	9.6	264	0.15	—
23	Neckar	Heidelberg	5/12/71	32	—	—	0.15	—	—	—	—	—	—
24	Lake Constance	Langenargen	5/13/71	25	—	—	0.4	—	—	—	—	—	—
25	Regnitz	Erlangen	5/14/71	11	21	—	159	—	24	—	119	—	—
26	Saale	Hof	5/15/71	13	6.5	—	6.9	—	—	—	31	—	—

NOTE: Aldrin, dieldrin, and heptachlor epoxide were not detected in samples of suspended matter.

¹ Quantities obtained from the filter residues were divided by the number of liters of water that had been filtered.

² Only DDT was found in suspended solids; the presence of DDD and DDE could not be confirmed in any case.

Insecticide Residues in Water and Sediment From Cisterns on the U.S. and British Virgin Islands—1970¹

Herbert Lenon², LaVerne Curry², Andrew Miller², and Daniel Patulski²

ABSTRACT

In the Virgin Islands the potential exists for pesticide contamination of water cisterns which supply and store all water used by local populations; cistern water and sediment samples on four islands were analyzed for pesticide residues in 1970 by gas-liquid chromatography. In the past, chlorinated hydrocarbon pesticides were used quite extensively on the Islands, however, malathion is more commonly used today.

Evidence of an unknown malathion metabolite was found in all 49 water samples analyzed, whereas malathion was found in only two (0.01 and 0.14 ppb). DDT, its metabolites, and dieldrin were not commonly found in the water samples except those from St. John where dieldrin was detected in approximately 50% of the samples (average concentration—0.19 ppb).

Sediment samples from cisterns, in general, contained much higher concentrations of pesticides than water, with DDT and its metabolites occurring most frequently. In many of these sediment samples, the residue levels were high enough to be concern. As a result it is strongly recommended that cisterns be cleaned frequently to remove sediment.

Introduction

Although the Virgin Islands receive an average annual rainfall of about 41 inches, much of this water is lost immediately by runoff and evaporation (1). Thus, the local populations depend on rain water for drinking and domestic use which is collected from roofs and stored in cisterns beneath their homes. With this type of rain catchment system, the nearby use of insecticides in mosquito eradication programs has been of concern, since insecticides could be carried by wind

to roofs and then flushed into the cisterns during any of the Islands' frequent showers.

Cistern water supplies from four of the Virgin Islands were surveyed for insecticide accumulations. The cisterns sampled were on the three major U.S. Virgin Islands of St. Croix, St. Thomas, and St. John and one British Virgin Island, Anegada (Fig. 1). The Virgin Islands are at the end of a 1,300-mile chain of islands known as the Greater Antilles—including Cuba, Jamaica, and Puerto Rico—which begins off the southern tip of Florida and extends south and west toward Central America.

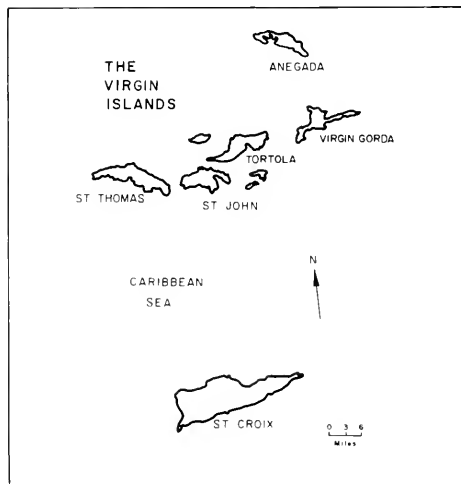


FIGURE 1.—The U.S. and British Virgin Islands

¹ Contribution 103 from the Virgin Islands Ecological Station, Great Lameshur Bay, St. John's, U.S. Virgin Islands.

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The U.S. Virgin Islands are of continental formation and were created during the Eocene Epoch by extrusion and folding from the ocean floor (6). St. John and St. Thomas are about 2 miles apart and are quite similar geographically. Both are rugged and hilly with numerous outcrops of metamorphic rocks. St. John is a small island, with 20 square miles of land, about two-thirds of which is national park. The 1960 population was 925 comprised mostly of natives who are supported largely by government funds. There is no commercial agriculture or significant tourism. Most of the population is centered around Cruz Bay and, to some extent, Coral Bay (Fig. 2). Small scattered native communities are also found on Bordeaux Mountain located in the interior south of Coral Bay. St. Thomas is larger and more densely populated than St. John, to the extent of being crowded, with 30 square miles of land and a population of 16,201 in 1960 (Fig. 3). For its economy, this island relies heavily on tourist spending which is approximately one million dollars a day. There is little industry and no major agriculture, although there is an Agricultural Experiment Station and some dairy farming.

St. Croix is 40 miles south of St. John. It has 85 square miles of surface area and had a population of 14,973 in 1960. Most of the population is centered around the two major cities of Frederiksted and Christiansted (Fig. 4). The economy is primarily industrial with rum as the main product, although there are a few remaining pineapple plantations. This island is also an important tourist center.

Anegada, one of the British Virgin Islands, is a flat coral island, almost entirely surrounded by reefs, 35 miles northeast of St. John and about 15 miles north of Virgin Gorda, the closest major island. The island is nearly 10 miles long and is almost divided by three large estuaries (Fig. 5). Nowhere is its elevation higher than 60 feet. It has the appearance of a large paved area and is exceedingly desolate. The population is very sparse, estimated at about 225 with only one small native community called "The Settlement." It is very poor with only shacks and junk accumulated for over 100 years in the front yards. The natives once fished for a

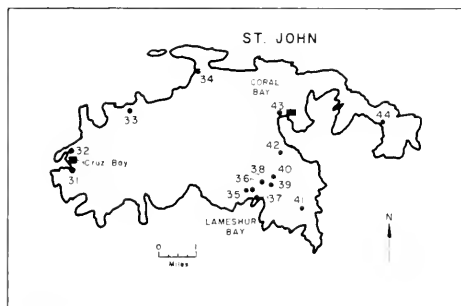


FIGURE 2.—Sampling stations on the U.S. Virgin Island of St. John

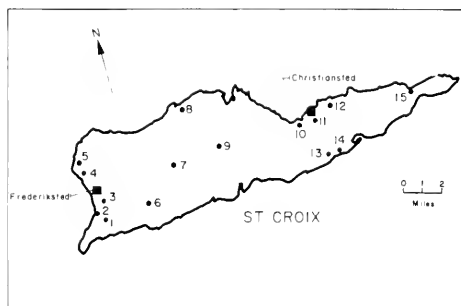


FIGURE 4.—Sampling stations on the U.S. Virgin Island of St. Croix

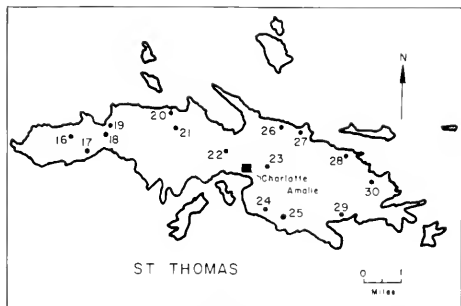


FIGURE 3.—Sampling stations on the U.S. Virgin Island of St. Thomas

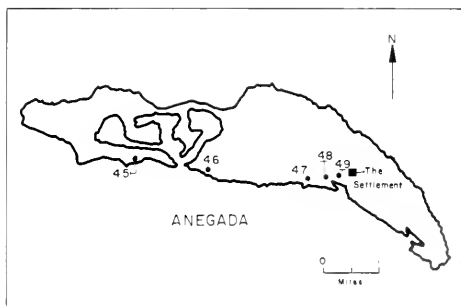


FIGURE 5.—Sampling stations on the British Virgin Island of Anegada

living but now work for a British corporation preparing for a jet airport. There is no tourism, agriculture, or industry.

Cisterns for catching rainwater are especially important for the basic supply of water since these islands have no other fresh water (1). The approximate average annual rainfalls for these four islands are: St. John, 47 inches; St. Thomas, 42 inches; St. Croix, 40 inches; and Anegada, 30 inches. Yet, these islands appear very dry, and as explained by Bowden *et al.* (1) "In essence rain falling on most rain-days is evaporated and transpired almost immediately, and is obviously of little or no consequence for growth of crops and vegetation."

Although no large-scale agricultural spraying is conducted on these islands, insecticides are used locally by individuals and by the U.S. Virgin Islands Health Department to control certain pest insects, especially mosquitoes. On St. Thomas and St. Croix, hotel owners spray areas to facilitate the tourist trade. The health department indicated that on St. Thomas DDT was used from 1960 to 1962 and dieldrin, from 1962 to 1964. Since then, malathion has been used, primarily because DDT and dieldrin were no longer effective against mosquitoes. According to Director Francois of Environmental Health (*personal communication, 1970*), it is applied to the ground, shrubbery, exposed water containers, and around the outside of water barrels and cisterns. Stored drinking water is often a source of mosquitoes, and private cisterns may be treated directly with Abate or DDVP resin strips. A similar program has been followed on St. Croix. On St. John, all insecticide use appears to be by individuals who apply much malathion. All insecticides are prohibited inside the national park.

Sampling Methods and Analytical Procedures

Samples of water and sediment from cisterns were taken on the following dates: St. Croix Island—March 18 and 19, 1970; St. Thomas Island—March 10-12, 1970; St. John Island—February 23 and March 9, 1970; and Anegada Island—March 26, 1970.

Each drinking water sample was collected in two 1-gal glass bottles. These were immediately extracted by liquid-liquid partitioning with purified petroleum ether using 70 ml per liter of water. This solvent was purified by twice distilling reagent grade petroleum ether with 10 g of dri-sodium per 3 liters at between 30° and 60° C. Each sample was extracted twice. Combined extracts were partially evaporated and sealed in screw-capped vials, packed, and sent by airmail to the biology laboratory at Central Michigan University, Mt. Pleasant, Mich. They were then immediately dried with anhydrous

sodium sulfate, further evaporated, and adjusted to 25 ml for analysis. It is believed that little if any breakdown of the extracted pesticides occurred in the short period of transport as was also noted by Guerrant, Fetzer, and Miles (2) using hexane.

Sediment samples were collected with a plankton net attached to a metal pole from the bottoms of cisterns containing significant amounts of sediment. They were then sent in screw-capped bottles directly to Central Michigan University for extraction and analysis. Water in the samples was removed by filtering on Whatman No. 1 paper with a Buchner funnel. Then the paper containing the sediment was oven-dried overnight at 50° C. The sample (3-7 g) was weighed on the pre-weighed filter paper and then shaken thoroughly several times with purified petroleum ether. The sediment was washed three times in petroleum ether, then the entire extract was dried with anhydrous sodium sulfate, evaporated, and adjusted to 25 ml.

Sample extracts were identified and quantified by gas chromatography (Beckman GC-4) using electron capture detection. Column packing and operating parameters were based on those of Mills, Onley, and Gaither (5) and were very similar to those used later by Guerrant *et al.* (2) with malathion:

Column: 6' x 1/8" stainless steel, packed with 3% SE-30 on 60/80 mesh Chromosorb W
Temperatures: Detector 300° C
Column 180° C
Inlet 210° C
Carrier gas: Helium at a flow rate of 40 ml/min

Samples were verified by analysis using 3% OV-17 on Chromosorb W as described by Menzie and Prouty (4) and also used by Guerrant *et al.* (2) with good agreement of retention times between the two systems.

Laboratory tests gave the following average recoveries: DDE, 76.1%; TDE (DDD), 72.8%; DDT, 82.2%; dieldrin, 83.5%; and malathion, 85%. Data in this report have been corrected using these recovery rates.

The standards used were from:

- A. Beckman Poly-Science Quant-Kits, 1% by weight in benzene, and included:
 - Technical *p,p'*-DDT, 99.5%
 - Technical *p,p'*-TDE, 99.5%
 - Technical dieldrin, 99.5%
 - Technical malathion, 99.5%
- B. Pesticide Repository, Perrine Primate Laboratory, Environmental Protection Agency, prepared 1% by weight in petroleum ether:
 - Analytical Standard—*p,p'*-DDE, 99%
 - Analytical Standard—*o,p'*-TDE, 99%
 - Analytical Standard—*o,p'*-DDT, 99%

A given quantity of malathion standard was allowed to hydrolyze naturally at room temperature for 6 months in order to characterize its metabolite in gas chromatographic analysis. This common metabolite was not identified but was used as a "qualitative standard."

Results and Discussion

DDT, its metabolites, and dieldrin were not commonly found in the water samples (Tables 1-4). DDT was not present in any of the water samples and its metabolites (DDE and TDE) were found in only one (Table 3). Dieldrin, which presents the greatest hazard in the water supply at levels found because of its toxicity and persistence, occurred in 48% (6 of 14) of the samples from St. John and 13% (2 of 15) of those from St. Thomas; however, no residues of dieldrin were found in samples from St. Croix and Anegada.

Malathion was present in only two water samples (Stations 19 and 34) and at low concentrations (0.14 and 0.01 ppb, respectively) (Tables 1-4). However, evidence of some metabolite of malathion was found in all water samples taken. Because this metabolite was not identified, values are not reported. Detection of this compound is reported only to suggest the previous presence of its precursor, malathion. Since malathion is a short-term insecticide, it was expected that it would be found only as one of its hydrolyzed products (2). Widespread use of this insecticide is, nevertheless, suggested through the common appearance of its metabolite in every water

sample. The potential hazard of the insecticide would not appear to be great since hydrolysis occurs rather rapidly, especially under conditions of neutral-to-alkaline pH. Because the metabolite was not identified, however, the importance of its presence cannot be interpreted.

The highest indicated malathion occurrence, as evidenced by the greatest relative quantities of its metabolite, was on St. Thomas and St. John. It is reasonable to expect St. Thomas Island to have one of the highest levels of pesticide residues, since the local health department as well as individuals spray the area to reduce the annoyance of mosquitoes and the risk of malaria to both tourists and its large local population. It may be possible to explain the high occurrence of malathion in water samples from the cisterns of St. John on the basis of the large amounts of water for this island brought in from areas like Puerto Rico where insecticide use is probably more common. This would also perhaps, explain the more frequent occurrence of the chlorinated hydrocarbon pesticides, especially dieldrin (Table 3). Water samples from St. Croix were next highest in relative malathion metabolite, as might be expected, since it is also quite heavily populated and supports tourists interest.

TABLE 1.—Stations from which cistern water was sampled on St. Croix Island

CISTERN SOURCE	APPROXIMATE AGE OF CISTERN (YEARS)	SAMPLE STATION ¹	RESIDUES IN PPB	
			MALATHION	DIELDRI
Residence	75-100	1	—	—
Business	9	2	—	—
Business	3-4	3	—	—
Hotel	>100	4	—	—
Residence	12	5	—	—
Museum	<10	6	—	—
Residence	13	7	—	—
School	10-15 (not in use)	8	—	—
Agric. Exp. Stn.	100 (plastic liner)	9	—	—
Hotel	5	10	—	—
Business	Unknown	11	—	—
Hotel	>100	12	—	—
Residence	Unknown	13	—	—
Residence	1	14	—	—
Hotel	Unknown (cleaned often)	15	—	—

NOTE: No residues of DDT, its metabolites, dieldrin, or malathion were detected. Evidence of some metabolite of malathion was found in all water samples taken.

¹ Sample station numbers correspond with those shown on map (Fig. 4).

TABLE 2.—Stations from which cistern water was sampled on St. Thomas Island showing pesticide residue levels detected

CISTERN SOURCE	APPROXIMATE AGE OF CISTERN (YEARS)	SAMPLE STATION ¹	RESIDUES IN PPB	
			MALATHION	DIELDRI
Estate	>100	16	—	—
Estate	5	17	—	—
Estate	30	18	—	—
Residence	>30	19	0.14	0.04
Dept. of Agric.	>30 (artesian)	20	—	—
Nursery	39	21	—	—
Public	4 (open)	22	—	—
Hospital	40 (open)	23	—	0.10
Residence	3	24	—	—
Residence	200	25	—	—
Camp (Peace Corp)	>30	27	—	—
Residence	5	28	—	—
Race track	1	29a	—	—
Race track	21	29b	—	—
School	>30	30	—	—

NOTE: — = no residue detected; no residues of DDT or its metabolites were detected. Evidence of some metabolite of malathion was found in all water samples taken.

¹ Sample station numbers correspond with those shown on map (Fig. 3).

Sediment was found in cisterns and consequently sampled at 35 of the 46 stations where water samples were obtained. Pesticide residues were detected in sediment samples from only 15 of the Stations (Table 5). Generally sediment samples had much higher residues with more variation in levels than water samples (Table 5—values for sediment were reported in ppm rather than in ppb). This suggests that when sediment occurs, the insecticides tend to be bound and accumulate over longer periods of time. Of the pesticides detected, DDT, its metabolites, or both occurred most frequently and were present in 4 of 8 sediment samples from St. John and 9 of 14 from St. Thomas. The relative absence of these compounds in both sediment and water samples from St. Croix and Anegada indicates little usage of the pesticides on these islands. Many of the residue levels of DDT and its metabolites in sediment samples from St. John and St. Thomas were high enough to be of considerable concern, particularly two from Stations 19 and 41 (Table 5). It appears that DDT was used in these two situations in place of malathion because malathion was not available. Sediment from Station 41 contained a high amount of DDT and smaller amounts of

its degradation products indicating a short-term occurrence of the pesticide in the sediment, while sediment from Station 19 contained very high amounts of metabolites and little DDT, thus, indicating a longer presence of the pesticide. The fact that unusually high amounts were found only at these two locations suggests individual sprayings and, perhaps, careless application.

Dieldrin was not found in sediment as frequently (only one sample) as in water, a finding which remains unexplained. Malathion was found in only one sediment sample, while its metabolite was found in all but five (Stations 19, 22, 32, 41, and 43). It is interesting to note that even though evidence of this product was found in all water samples, it was absent in five of the sediment samples.

Most sediment samples were composed of silty loam or organic matter and sometimes coral sand. DDT and its degradation products can accumulate in sediment, since they are bound to solid particles. Lichtenstein and Schulz (3) demonstrated that the persistence of residues in soils is dependent on various factors such as the insecticide itself, soil types, as well as climatic conditions. Soils of higher organic content tend to bind the greatest amount of chlorinated hydrocarbon pesticide.

The survey and evaluation of pesticide levels in private and public cisterns on these islands were complicated for several reasons. The exact sources of the water are never known. Although rainwater is collected in all cisterns, these supplies are frequently insufficient and additional water must be purchased from various places, often as far away as Puerto Rico, but including other islands as well. This is known to be true, at least, for St. John where water arrives by barge and is then

TABLE 3.—Stations from which cistern water was sampled on St. John Island showing pesticide residue levels detected

CISTERN SOURCE ¹	SAMPLE STATION ²	RESIDUES IN PPB				
		MALATHION	DDT	DDE	DDDE	DIELDRIN
Public	31	—	—	—	—	—
National park	32	—	—	—	—	—
Camp ground	33	—	—	—	—	—
Residence	34	0.01	—	—	—	0.02
Old commissary	36	—	—	—	—	0.01
Laboratory	37	—	—	—	—	—
Residence (Park Ranger)	38	—	—	—	—	0.04
Camp	39a	—	—	—	—	—
Camp	39b	—	—	—	—	—
Laboratory	40	—	—	—	—	1.03
Residence	41	—	—	0.15	0.02	—
Clinic	42	—	—	—	—	0.04
Church	43	—	—	—	—	0.01
School	44	—	—	—	—	—
Average						0.19

NOTE: — = no residue detected. Evidence of some metabolite of malathion was found in all water samples taken.

¹ Approximate ages of these cisterns are unknown.

² Sample station numbers correspond with those shown on map (Fig. 2).

TABLE 4.—Stations from which cistern water was sampled on Anegada Island

CISTERN SOURCE	APPROXIMATE AGE OF CISTERN (YEARS)	SAMPLE STATION ¹
Business	<1 (open)	45
Residence	<1	46
Public well	(plastic liner) very old	47
Public well	Unknown	48
School	>25	49

NOTE: No residues of DDT, its metabolites, dieldrin, or malathion were detected. Evidence of some metabolite of malathion was found in all water samples taken.

¹ Sample station numbers correspond with those shown on map (Fig. 5).

trucked around the Island. Consequently, this may account for the occurrence in water of pesticides not used locally. Incomplete information was also a problem. Data regarding the age and characteristics of the individual water supplies were often sparse. Some cisterns were cleaned regularly and contained no sediment, while others were never cleaned and contained several inches of bottom material.

No apparent correlations are evident between the age of cisterns and residue levels, or between commercial or residential establishments and residue levels. It appears to be more a matter of the extent of individual use of pesticides, care in application, as well as, perhaps, the location where water was obtained.

Results do not suggest a potential danger to the people who use water from the cisterns examined with the possible exception of some on St. John containing significant levels of dieldrin. Sediment, contained in many of the cisterns, represents a much greater potential hazard with accumulated levels of residues, representing the past presence of insecticides at various stages of degradation. It is suggested that these sediment accumulations be removed by frequent cleaning of the cisterns, thus keeping total residue levels in the cisterns to a minimum.

Acknowledgment

We wish to thank the U.S. National Park Service, Mr. Bromberg, and Mr. Tony Cook for personnel and equipment; Drs. Edward Towle and Arthur Dammon for facilities at St. John; and Mr. John Yntema for help with field work.

See Appendix for the chemical names of compounds discussed in this paper.

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TABLE 5.—Stations where sediment of cisterns was sampled and pesticide residue concentrations detected

SAMPLE STATION ¹	RESIDUES IN PPM				
	MALATHION	DDT	DDE	DDE	DIELDRIN
ST. CROIX					
3	—	—	—	—	—
4	—	—	—	—	—
5	—	—	—	—	—
6	—	—	—	—	—
8	—	—	—	—	—
9	—	—	—	—	—
10	—	—	—	—	—
12	—	—	—	—	—
15	—	—	—	—	—
ST. THOMAS					
16	—	0.61	0.37	0.09	—
18	0.19	37.49	11.50	0.96	—
19	—	16.36	401.37	1,250.26	—
20	—	13.49	4.12	1.58	—
22	—	—	—	—	—
23	—	—	—	—	—
24	—	—	—	—	—
25	—	—	—	—	—
26	—	—	—	—	—
27	—	0.96	0.43	0.17	—
28	—	—	—	0.07	—
29a	—	—	0.66	0.89	—
29b	—	0.28	0.22	0.32	—
30	—	0.09	0.10	0.12	—
ST. JOHN					
32	—	—	—	—	—
33	—	—	—	—	—
34	—	—	—	—	—
35	—	1.82	0.21	0.05	—
36	—	—	—	—	0.08
41	—	271.29	34.11	6.27	—
43	—	—	—	0.14	—
44	—	2.77	0.44	0.07	—
ANEGADA					
45	—	—	—	—	—
47	—	—	—	—	—
48	—	—	—	—	—
49	—	7.18	1.79	3.68	—

NOTE: — = no residue detected; evidence of some metabolite of malathion was found in all sediment samples with the exception of the five samples from the following stations: Stations 19, 22, 32, 41, and 43.

¹ Sample station numbers correspond to those for water samples (Tables 1-4; Figs. 2-5).

PESTICIDES IN SOIL

Pesticide Residue Levels in Soils, FY 1969—National Soils Monitoring Program

G. B. Wiersma¹, H. Tai², and P. F. Sand³

ABSTRACT

This report is a summary of the FY 1969 results of the National Soils Monitoring Program, an integral part of the National Pesticide Monitoring Program (NPMP). Pesticide residues in cropland soil for 43 States and noncropland soil for 11 States are reported. Tables for each State give the number of samples collected, arithmetic means and ranges of residue levels detected, and the percent of sites with detectable residues. In addition, for selected pesticides and various States and State groupings, a frequency distribution of pesticide residues was determined. Use records for FY 1969 are given by the pesticides used, the percent of sites treated, the average application rates, and the average amounts applied per site. Comparisons are made between residue levels in different land-use areas.

Introduction

The National Soils Monitoring Program is an integral part of the National Pesticide Monitoring Program (NPMP), which was initiated as a result of a recommendation made by the President's Science Advisory Committee in its report of 1964 entitled "Use of Pesticides" that the appropriate Federal agencies "develop a continuing network to monitor residue levels in air, water, soil, man, wildlife, and fish." The NPMP as originally designed was described in the first issue of the *Pesticides Monitoring Journal* (1), and a revised description to reflect certain program realignments and

other changes was published in the June 1971 issue of this *Journal* (2).

The objectives of the NPMP are to determine levels and trends of pesticides in the various components of the environment (2). The establishment of baseline or background levels of pesticide residues through the NPMP will provide a basis for comparison of subsequently identified pesticide residue levels in an environmental component.

The Panel on Pesticides Monitoring of the Working Group on Pesticides (2) listed five bases for concern to be used in evaluating pesticide residue levels in the various environmental components. They are:

- (1) any concentration of a pesticide known to be potentially harmful;
- (2) increasing trends;
- (3) exceeding standards;
- (4) recognition of adverse effects on humans; and
- (5) erratic variability (a statistically oriented observation that is potentially common to each stratum sampled).

The results of this study serve to establish a baseline of pesticide residues in cropland and noncropland soils at a particular point in time (FY 1969). The present data and all future data will be evaluated using applicable criteria included in the five bases of concern outlined above.

Sampling Procedures and Methods

In general, sampling techniques involved in this study were the same as those described by Wiersma, Sand, and Cox (3).

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In FY 1969, cropland soil was sampled in every State except Alaska, Hawaii, Kansas, Minnesota, Montana, Oregon, and Texas. Noncropland was sampled in 11 States—Arizona, Georgia, Idaho, Iowa, Maine, Maryland, Nebraska, Virginia, Washington, West Virginia, and Wyoming. Samples collected in FY 1969 included both soil and mature crops and/or those ready for harvest; however, results of crop analyses are not published in this report.

Analytical Procedures

ORGANOCHLORINE AND ORGANOPHOSPHOROUS COMPOUNDS

A subsample of soil weighing 300 g. wet weight, was placed in a 2-qt fruit jar with 600 ml of 3:1 hexane-isopropanol solvent. The jars were sealed and rotated for 4 hours. After rotation, the soil was allowed to settle, and 200 ml of the extract solution was filtered into a 500-ml separatory funnel. Isopropanol was removed with two washings of distilled water, and the remaining solution was then filtered through a funnel containing glass wool and anhydrous sodium sulfate (Na_2SO_4). Further cleanup was normally not required before analysis.

Gas-Liquid Chromatography

Analyses were performed on gas chromatographs equipped with tritium foil electron affinity detectors for organochlorine compounds and thermionic or flame photometric detectors for organophosphorous compounds. A dual-column system employing polar and nonpolar columns was utilized to identify and confirm pesticides. Instrument parameters were as follows:

Columns:	Glass, 183 cm long by 6 mm, o.d., and 4 mm, i.d., with one of the following packings: 3% DC-200 on 100-120 mesh Gas Chrom Q or 9% QF-1 on 100-120 mesh Gas Chrom Q
Carrier gas:	5% methane-argon at a flow rate of 80 ml/min
Temperatures:	Detector 200° C Injection port 250° C Column QF-1 166° C Column DC-200 170°-175° C

When necessary, confirmation of residues was made by thin layer chromatography or p-values. The lower limit of detection was 0.01 ppm. The average recovery rate for all pesticides was 100% (with a $\pm 10\%$ error); the data were corrected for recovery and also adjusted to a dry-weight basis by determining the moisture content on a separate portion of each sample using the oven drying method.

ATRAZINE

After a 4-hour Soxhlet extraction of a 50-g subsample of soil with 25 ml of water and 300 ml of methanol, the sample extract was transferred to a 1-liter separatory funnel and 200 ml of water added. The sample extract

was partitioned three times with a portion of 150 ml of freon 113 for each partitioning. The freon 113 fractions were combined and concentrated to incipient dryness. The sample was then dissolved in hexane, adjusted to a 5-ml volume, and injected into a gas-liquid chromatograph.

Gas-Liquid Chromatography

A thermionic flame detector with rubidium sulfate coating on a helix coil was used. Instrument parameters were as follows:

Column:	Glass, 183 cm long by 6 mm, o.d., and 4 mm, i.d., packed with 3% Versamid 900 on 100-120 mesh Gas Chrom Q
Carrier gas:	Helium
Detector fuel gas:	Oxygen (200-300 ml/min); Hydrogen (20-30 ml/min)
Temperatures:	Detector 240° C Injection port 240° C Column 240° C

Confirmation was made using a DC-200 column at 180° C and a Coulson detector (reductive mode) at the following temperature settings: pyrolysis tube—850° C, transfer line—220° C, and block—220° C.

The minimum detection limit was 0.01 ppm, and recovery was about 100%.

2,4-D

Analyses were made following the procedure developed by Woodham *et al.* (4). The analytical method involved a diethyl ether extraction of acidified soil, an alkali wash to remove interfering substances, and an esterification procedure using 10% boron trichloride in 2-chloroethanol reagent. The 2-chloroethyl ester of 2,4-D was then analyzed by gas chromatography. The minimum detection limit was 0.01 ppm, and the average recovery was 85%. Results were corrected for percent recovery.

ARSENIC

Arsenic was determined by atomic absorption spectrophotometry. The soil sample was first extracted with 9.6N hydrochloric acid (HCL) and reduced to trivalent arsenic with stannous chloride. The trivalent arsenic was partitioned from HCL solution to benzene, then further partitioned into water for the absorption measurement. A Perkin-Elmer Model 303 instrument was used, and absorbance was measured with an arsenic lamp at 1972 Å with argon as an aspirant to an air-hydrogen flame. The minimum detection limit was 0.1 ppm, and the recovery value for arsenic averaged 70%. Results were corrected for percent recovery.

Results

The data in this report are for soils only (both cropland and noncropland) and include results for all States

sampled in the study. Caution should be exercised when interpreting the arithmetic means presented in the tables, because pesticide residue data are not normally distributed, and the arithmetic means for pesticide residues tend to be greater than the corresponding median. Therefore, they cannot be considered an indication of the central tendency of the data. Information accompanying

the arithmetic means in this report such as the percent occurrence, range of detected residues, and number of observations can aid in evaluating the arithmetic mean.

RESIDUES—ALL STATES

Table 1 presents a summary of pesticide residues in cropland soils for all 43 States sampled. Percent occur-

TABLE 1.—Summary of pesticide residues in cropland soil from 43 States—FY 1969

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
Aldrin	1,729	189	10.9	0.02	0.01-3.06
Arsenic	1,726	1,713	99.3	6.43	0.25-107.45
Atrazine	199	28	14.1	0.01	0.01-1.55
Carbophenothion	66	1	1.5	<0.01	0.23
Chlordane	1,729	151	8.7	0.04	0.01-6.30
2,4-D	188	3	1.6	<0.01	0.01-0.03
DCPA (Dacthal®)	1,729	1	0.1	<0.01	0.54
<i>o,p'</i> -DDE	1,729	79	4.6	<0.01	0.01-0.20
<i>p,p'</i> -DDE	1,729	429	24.8	0.06	0.01-6.99
<i>o,p'</i> -DDT	1,729	243	14.1	0.03	0.01-6.29
<i>p,p'</i> -DDT	1,729	384	22.2	0.17	0.01-35.92
DDTR	1,729	451	26.1	0.31	0.01-78.36
DEF	1,729	1	0.1	<0.01	0.12
Diazinon	66	2	3.0	<0.01	0.02-0.15
Dicofol	1,729	9	0.5	<0.01	0.03-1.07
Dieldrin	1,729	480	27.8	0.03	0.01-1.60
Endosulfan (I)	1,729	5	0.3	<0.01	0.01-0.24
Endosulfan (II)	1,729	9	0.5	<0.01	0.01-0.53
Endosulfan sulfate	1,729	11	0.6	<0.01	0.01-0.94
Endrin	1,729	39	2.3	<0.01	0.01-0.56
Endrin aldehyde	1,729	1	0.1	<0.01	0.03
Endrin ketone	1,729	9	0.5	<0.01	0.01-0.13
Ethion	66	1	1.5	<0.01	0.03
Heptachlor	1,729	68	3.9	<0.01	0.01-0.97
Heptachlor epoxide	1,729	139	8.0	<0.01	0.01-1.08
Isodrin	1,729	11	0.6	<0.01	0.01-0.03
Lindane	1,729	15	0.9	<0.01	0.01-0.35
Malathion	66	2	3.0	0.01	0.04-0.36
Methoxychlor	1,729	1	0.1	<0.01	0.28
Ethyl parathion	66	7	10.6	0.06	0.01-3.01
PCNB	1,729	1	0.1	<0.01	0.69
<i>o,p'</i> -TDE	1,729	49	2.8	0.01	0.01-4.52
<i>p,p'</i> -TDE	1,729	265	15.3	0.05	0.01-31.43
Toxaphene	1,729	73	4.2	0.07	0.10-11.72
Trifluralin	1,729	60	3.5	<0.01	0.01-0.25

¹ One sample per site.

² Percent based on number of sites with residues greater than or equal to the sensitivity limits.

rence of residues is based on the number of sites with residues greater than or equal to the sensitivity limit.

The data for atrazine, 2,4-D, and the organophosphates are not truly comparable with those determined for the organochlorines or arsenic, because analyses for atrazine and 2,4-D were made only when use records indicated that they had been applied—199 and 188 times, respectively, and analyses for organophosphates were performed on only 66 of the 1,729 samples.

Elemental arsenic residues were found most frequently, with 99.3% of the sites having detectable residues and a mean level of 6.4 ppm. It is probable that most of this arsenic was from natural sources, although agricultural sources cannot be ruled out at this time.

The most widely distributed organochlorine pesticide was dieldrin, with 27.8% of the sites having detectable residues, followed by DDTR residues (a compilation of all members of the DDT group) found at 26.1% of the sites; aldrin, found at 10.9%; and chlordane, found at 8.7%. DDTR had the highest mean residue level, with 0.31 ppm found in cropland soils. With the exception of individual members of the DDT group, the other organochlorines had average residues ranging from <0.01 to 0.07 ppm.

Based on the 66 samples analyzed for organophosphates, ethyl parathion was detected 10.6% of the time, with a mean residue level of 0.06 ppm. Malathion and diazinon were each detected 3.0% of the time, with mean residue levels of 0.01 and <0.01 ppm, respectively.

In the 188 samples analyzed for 2,4-D and other chlorophenoxy herbicides, 2,4-D was the only one detected; 2,4-D was found in 1.6% of 188 samples analyzed, with a mean residue level of <0.01 ppm. Atrazine was detected in 14.1% of the 199 samples analyzed, with a mean residue level of 0.01 ppm—the highest mean residue of the herbicides detected. Trifluralin was detected in 3.5% of the 1,729 samples, with a mean residue level of <0.01 ppm.

The residues found in noncropland soils for the 11 States sampled are presented in Table 2. The mean arsenic residue level was 5.0 ppm, occurring in 98.5% of the samples. DDTR was detected in 16.1% of the noncropland soils at levels ranging from 0.01 to 0.62 ppm, with a mean level of 0.01 ppm. With the exception of members of the DDT group, dieldrin was the most widely distributed pesticide, occurring in 4.0% of the samples, with residues ranging between 0.01 to 0.09 ppm and a mean residue level of <0.01 ppm.

RESIDUES—INDIVIDUAL STATES

The pesticide residue summaries for cropland by individual States are given in Table 3, and similar results are shown for noncropland in Table 4. It would be impractical to attempt to comment on the results for each State; therefore, in order to facilitate summarizing the data, Figs. 1, 2, and 3 are presented. These are for three of the most commonly occurring residues—arsenic, DDTR, and dieldrin. Means for each pesticide in each State were calculated, and distribution of these averages are indicated on the corresponding Figures.

TABLE 2.—Summary of pesticide residues in noncropland soil from 11 States—FY 1969

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
Aldrin	199	1	0.5	<0.01	0.02
Arsenic	198	195	98.5	5.01	0.33-54.17
Chlordane	199	3	1.5	<0.01	0.04-0.50
<i>o,p'</i> -DDE	199	1	0.5	<0.01	0.02
<i>p,p'</i> -DDE	199	27	13.6	0.01	0.01-0.31
<i>o,p'</i> -DDT	199	7	3.5	<0.01	0.01-0.05
<i>p,p'</i> -DDT	199	18	9.1	0.01	0.01-0.23
DDTR	199	32	16.1	0.01	0.01-0.62
Dicofol	199	2	1.0	<0.01	0.10-0.29
Dieldrin	199	8	4.0	<0.01	0.01-0.09
Heptachlor epoxide	199	2	1.0	<0.01	0.01
<i>p,p'</i> -TDE	199	6	3.0	<0.01	0.01-0.18
Toxaphene	199	1	0.5	<0.01	0.52

¹ One sample per site.

² Percent based on number of sites with residues greater than or equal to the sensitivity limits.

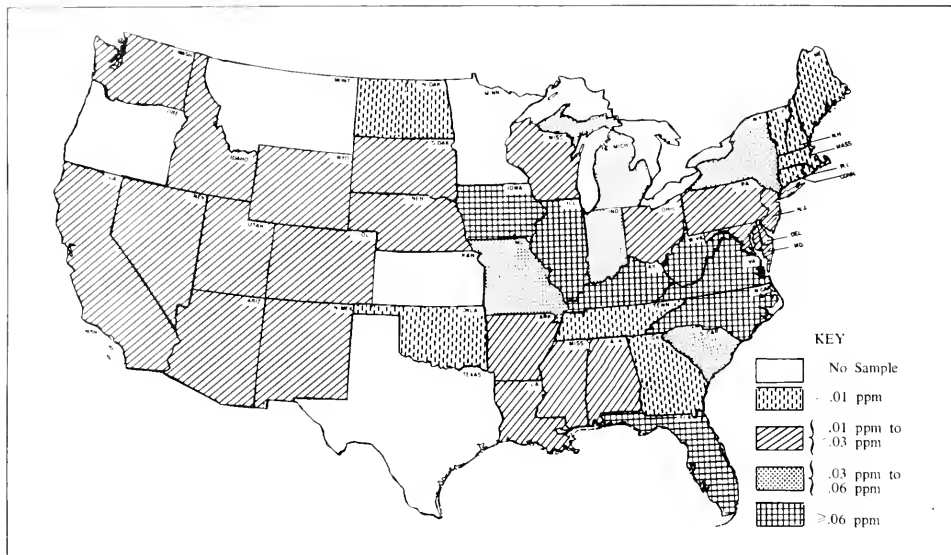


FIGURE 1.—Arsenic residues in cropland soil

The class intervals for the keys accompanying each Figure were obtained in the following manner: The range of residues for the Nation was obtained, and the highest value was converted to a logarithm. This value was then divided by the number of desired classes. The resulting intervals were added to obtain the class boundaries which, in turn, were converted to the untransformed dimensions. Essentially, this took advantage of the fact that most residue data are logarithmically distributed.

Distribution of arsenic residues across the United States is presented in Fig. 1. The highest residue levels were found in the New England States (Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, and Vermont), Arkansas, Kentucky, New York, North Dakota, Ohio, and Pennsylvania; these individual States and the New England States had mean residues of arsenic >8.4 ppm. The remaining residues were distributed primarily in the 2.0 to 8.4 ppm range, with Wyoming and Florida having less than 2.0 ppm. Those States left blank were not sampled.

The distribution of DDT residues (DDTR) is shown in Fig. 2. Once again, the key indicates the range of residues for each of the class intervals. A similar map for dieldrin residues is presented in Fig. 3.

The mean residue levels, the percent positive sites, and the range of residue levels for the 12 States with the highest arsenic residues are shown in Table 5.

Residue data for the five States with the highest DDTR residues are presented in Table 6. Although Michigan had a mean residue of 2.09 ppm and a range of 0.01 to 78.36 ppm, only 23.5% of the samples had detectable residues, indicating that the residues were not widely distributed. By contrast, Mississippi had a mean residue of 2.06 ppm with 89.7% of its sites having detectable residues and a narrower range (0.03 to 13.14 ppm). Although the range was narrower, pesticide residues were more widely distributed in Mississippi than in Michigan.

The seven States with the highest dieldrin residues are listed in Table 7. The highest mean residue level, 0.11 ppm, was found in Illinois, with 61.3% of the sites having detectable residues. In general, the other six States tended to have mean residues approximating one another, 0.06, 0.07, or 0.08 ppm.

PESTICIDE USE RECORDS

When soil samples were collected, an attempt was made to determine what pesticides had been used on the sites for the year of sampling. The summary tables for the use records show the percent of times a pesticide was

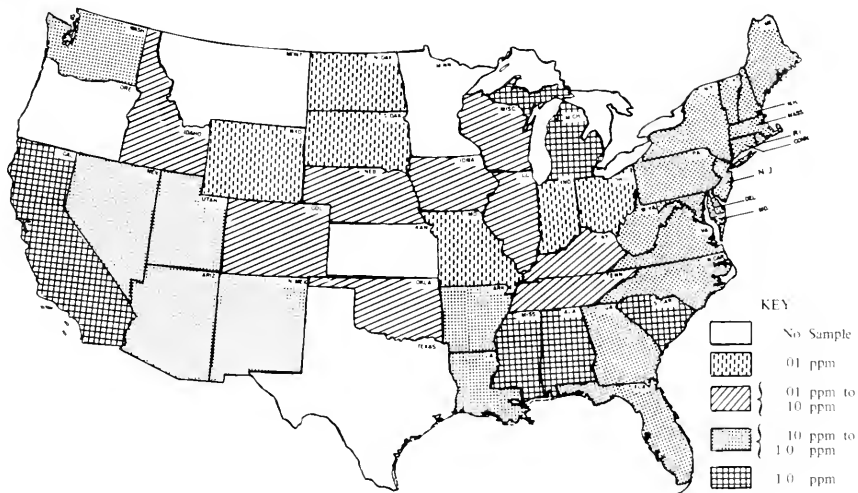


FIGURE 2.—DDTR residues in cropland soil

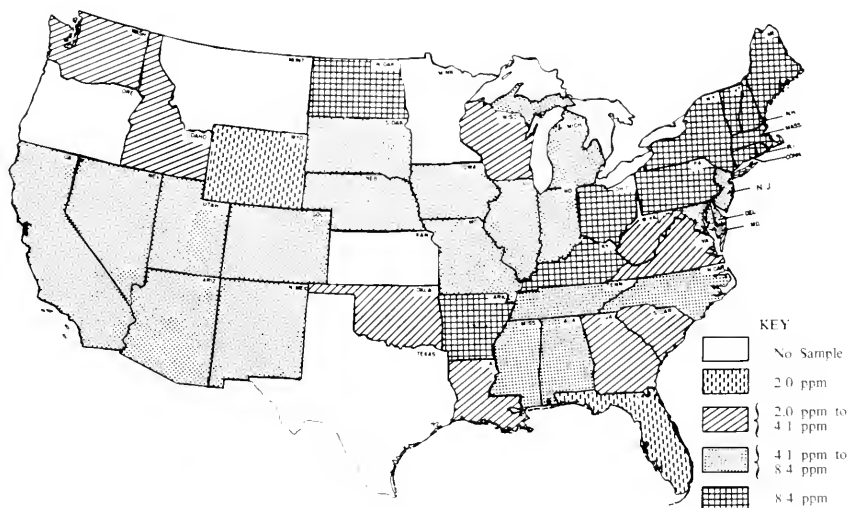


FIGURE 3.—Dieldrin residues in cropland soil

used, the average application rate expressed in pounds per acre of the active ingredients, and the average amount applied per site. The average amount per site was determined by dividing the total amount of active ingredient of a pesticide used by the total number of sites surveyed.

Table 8 shows 130 different pesticides reported to have been used on cropland in the year of sampling. Those most commonly used were atrazine, captan, 2,4-D, malathion, and methylmercury dicyandiamide. Technical DDT was used on 3.44% of the sites, aldrin on 4.16% of the sites, and dieldrin on 1.19% of the sites.

On noncropland sites 2,4-D, malathion, and mirex were reported to have been used (Table 9). However, these should not be considered the only pesticides used on noncropland sites. In general, records of treatment of noncropland sites are less accurate than those kept for cropland. The breakdown of pesticide usage by individual States for cropland and noncropland soils, respectively, are shown in Tables 10 and 11. Of the 43 States with cropland soil analyzed, use records for 4 showed no pesticides used on the sampling sites: Nevada (2 sites); New Hampshire (2 sites); Vermont (5 sites); and Wyoming (17 sites). Of the 11 States with noncropland soil analyzed, 8 reported no pesticides used on the sampling sites: Arizona (43 sites); Iowa (7 sites); Maine (11 sites); Maryland (3 sites); Virginia (14 sites); Washington (11 sites); West Virginia (9 sites); and Wyoming (37 sites).

Because of the number of States and pesticides presented in Tables 10 and 11, it is difficult to make all possible comparisons between the use patterns indicated and the detected residues shown in Tables 3 and 4. Therefore, comparisons have been restricted to those States having the highest residues as shown in Figs. 1, 2, and 3 (arsenic, DDTR, and dieldrin, respectively).

Table 12 compares those States having the highest arsenic residues with the average amount applied per site and the percent of sites which reported using an arsenic compound. The amount of arsenic applied did not seem to be directly related to the amount detected in the soil. Arkansas, Kentucky, North Dakota, and Ohio reportedly used no arsenic compounds, whereas New England, New York, and Pennsylvania reported using sodium arsenite and lead arsenate. The application rates were below the detected residue levels, and the percent of times used was below the percent of times residues were detected. It also must be considered that the application rates were for the active ingredients of sodium arsenite and lead arsenate, and not for elemental arsenic alone. A fair assumption would be that most arsenic residues detected in cropland soils probably resulted from natural levels of arsenic.

A similar comparison for the five States with the highest DDTR residues is found in Table 13. It is interesting to note that use records for four of the States listed (California, Michigan, Mississippi, and South Carolina) indicate that the amount applied was less than the mean level detected in the soil. Also, in all five States, the percent of sites positive for DDTR was approximately three or four times greater than the percent of sites reportedly treated with DDT. Unlike arsenic, the residues of DDTR could only result from the use of DDT either in the year of sampling or in previous years.

Table 14 lists the seven States with the highest dieldrin residues. In most cases, the average amount of aldrin/dieldrin applied approximated the mean residue of dieldrin detected in the soil, but the percent of sites reportedly treated with dieldrin or aldrin was always considerably less than the percent of sites with dieldrin residues. This wider distribution of dieldrin residues, when compared to use records for the year of sampling, probably indicates residues from previous years.

PESTICIDE FREQUENCY DISTRIBUTION

The statistics discussed thus far, namely the mean, the range, and the percent of sites at which residues were detected, do not describe their distribution. To describe this distribution, probit analysis was used. The residue levels were ranked from lowest to highest, accumulated, and the percentages computed. The residues were transformed to logarithms, the percentages to probits, and the relationship between the logarithms of the residues and the probits of the accumulated percentages was calculated by regression analysis. The computer program used was that of Daum, (5); the theory and techniques as applied in the cited reference were modified slightly.

The residue levels at the fiftieth percentile point (median) for the individual pesticides in soil for each State along with the upper and lower 95% fiducial limits are presented in Table 15. For example, in the State of Alabama, the fiftieth percentile point (median) for arsenic was 4.09 ppm. Thus, 50% of the sites had residues less than 4.09 ppm. The upper and the lower fiducial limits of the residues establish the 95% confidence interval about the residue value for the fiftieth percentile. It should be noted that the mean for a particular State is not the same as the fiftieth percentile point (median) from the frequency distribution. For example, the mean level of arsenic for Alabama was 6.1 ppm, while the frequency distribution indicated 4.09 ppm for the fiftieth percentile point. This is an example of the fact that residue data are not normally distributed and the mean and median are not identical.

Not all pesticides are shown for all States. A cutoff point of six or more pairs of observations was used to eliminate

situations where there were too few observations to calculate a reliable distribution. Space did not permit printing tables showing distribution of pesticide residues for percentiles other than the fiftieth.

CROPPING REGIONS ANALYSIS

The data were grouped by counties into various cropping regions, and these are shown in Tables 16 and 17. The boundaries for the various cropping areas were based on a major land-use map of the United States compiled by F. J. Marschner of the U.S. Department of Agriculture, Bureau of Agricultural Economics, 1950. No effort was made to make a land-use division within counties. This resulted in a good definition of the larger land-use areas such as the corn belt and cotton-growing areas. The land in the United States was grouped into several major land-use areas—corn, cotton, general farming, hay, small grain, vegetables, and fruit. In some cases, two areas overlapped. Irrigated land was determined from information obtained at the time of sample collection in this study.

It is of interest to make a few individual comparisons between the cropping regions and the national means. For example, note that in the corn region, aldrin occurred 23.5% of the time (Table 17) with a mean residue level of 0.05 ppm (Table 16). However, nationally, aldrin only occurred 10.9% of the time with a mean level of 0.02 ppm (Table 1), an indication of the heavier use of aldrin in the corn region. But, in the corn region, the mean residue level of DDTR was 0.14 ppm which is well below the national mean of 0.31 ppm.

The vegetable and fruit cropping region had the highest level of DDTR, over two times higher than the next highest cropping region and over six times higher than the national mean for DDTR. This might result from a high use of DDT in various orchard operations. The next highest residue was found in the cotton and vegetable region, with approximately equal amounts detected between them. The rest of the amounts of DDT in the cotton and general farming, general farming, hay and general farming, and irrigated land were similar to one another. The two areas with the least amount

of DDTR in the soil were the corn and small grains cropping regions.

The corn, vegetable, and vegetable and fruit cropping regions had the heaviest residues of dieldrin. Residues of dieldrin in the other cropping regions were either equal to or below the mean residues detected for all States (Table 1).

The cotton cropping region had the highest toxaphene residues. The cotton and general farming and general farming cropping regions had residue levels of about half those detected in the cotton cropping region.

Acknowledgment

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See Appendix for chemical names and compounds discussed in this paper.

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TABLE 3.—Pesticide residues in cropland soil from 43 States—FY 1969

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
ALABAMA					
Arsenic	23	23	100.0	6.11	0.70-28.60
Chlordane	22	3	13.6	0.04	0.07-0.62
<i>o,p'</i> -DDE	22	1	4.6	<0.01	0.01
<i>p,p'</i> -DDE	22	19	86.4	0.17	0.01-0.72
<i>o,p'</i> -DDT	22	16	72.7	0.09	0.01-0.65
<i>p,p'</i> -DDT	22	20	90.9	0.78	0.02-6.60
DDTR	22	20	90.9	1.13	0.05-8.08
Dieldrin	22	5	22.7	0.01	0.01-0.14
Endrin	22	2	9.1	<0.01	0.03-0.05
Heptachlor	22	2	9.1	<0.01	0.01
Heptachlor epoxide	22	3	13.6	<0.01	0.01-0.04
Lindane	22	2	9.1	<0.01	0.01
<i>o,p'</i> -TDE	22	1	4.6	<0.01	0.08
<i>p,p'</i> -TDE	22	13	59.1	0.07	0.01-0.73
Toxaphene	22	6	27.3	0.69	0.68-4.95
Trifluralin	22	7	31.8	0.01	0.01-0.08
ARIZONA					
Arsenic	8	8	100.0	6.58	2.82-9.97
<i>o,p'</i> -DDE	8	4	50.0	0.02	0.01-0.07
<i>p,p'</i> -DDE	8	8	100.0	0.46	0.06-0.84
<i>o,p'</i> -DDT	8	5	62.5	0.07	0.08-0.17
<i>p,p'</i> -DDT	8	7	87.5	0.20	0.03-0.57
DDTR	8	8	100.0	0.76	0.06-1.56
Endosulfan (I)	8	1	12.5	0.03	0.24
Endosulfan (II)	8	1	12.5	0.07	0.53
Endosulfan sulfate	8	1	12.5	0.04	0.29
Endrin	8	3	37.5	0.07	0.10-0.22
Endrin ketone	8	2	25.0	0.01	0.01-0.07
<i>p,p'</i> -TDE	8	2	25.0	0.01	0.03-0.06
Toxaphene	8	6	75.0	1.09	0.57-4.27
Trifluralin	8	1	12.5	0.02	0.13
ARKANSAS					
Aldrin	47	7	14.9	<0.01	0.01-0.06
Arsenic	47	47	100.0	8.98	1.70-28.25
<i>o,p'</i> -DDE	47	5	10.6	0.01	0.01-0.07
<i>p,p'</i> -DDE	47	32	68.1	0.24	0.01-2.81
<i>o,p'</i> -DDT	47	22	46.8	0.07	0.01-1.11
<i>p,p'</i> -DDT	47	32	68.1	0.29	0.01-3.28
DDTR	47	34	72.3	0.67	0.03-7.20
Dieldrin	47	12	25.5	0.02	0.01-0.24
Endrin	47	5	10.6	0.01	0.01-0.29
Endrin ketone	47	2	4.3	<0.01	0.10-0.13
<i>p,p'</i> -TDE	47	27	57.5	0.07	0.01-1.19
Toxaphene	47	8	17.0	0.27	0.32-3.40
Trifluralin	47	4	8.5	0.01	0.01-0.20
CALIFORNIA					
Aldrin	65	1	1.5	<0.01	0.03
Arsenic	65	65	100.0	5.15	0.74-23.67
Carbophenothion	17	1	5.9	0.01	0.23
Chlordane	65	2	3.1	0.01	0.10-0.32
DCPA	65	1	1.5	0.01	0.54
<i>o,p'</i> -DDE	65	24	36.9	0.01	0.01-0.14

TABLE 3.—Pesticide residues in cropland soil from 43 States—FY 1969—Continued

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
CALIFORNIA—Continued					
<i>p,p'</i> -DDE	65	55	84.6	0.37	0.01-5.93
<i>o,p'</i> -DDT	65	32	49.2	0.08	0.01-1.33
<i>p,p'</i> -DDT	65	48	73.9	0.54	0.01-11.09
DDTR	65	55	84.6	1.47	0.01-41.81
Diazinon	17	1	5.9	<0.01	0.02
Dicofol	65	6	9.2	0.02	0.03-1.07
Dieldrin	65	20	30.8	0.02	0.01-0.31
Endosulfan (I)	65	1	1.5	<0.01	0.01
Endosulfan (II)	65	5	7.7	<0.01	0.01-0.09
Endosulfan sulfate	65	5	7.7	0.01	0.02-0.15
Endrin	65	9	13.9	0.01	0.01-0.16
Heptachlor epoxide	65	8	12.3	<0.01	0.01-0.03
Lindane	65	2	3.1	<0.01	0.02
Ethyl parathion	17	1	5.9	<0.01	0.02
<i>o,p'</i> -TDE	65	13	20.0	0.09	0.01-4.52
<i>p,p'</i> -TDE	65	40	61.5	0.38	0.01-20.13
Toxaphene	65	10	15.4	0.16	0.16-2.07
Trifluralin	65	7	10.8	<0.01	0.01-0.10
COLORADO					
Aldrin	60	1	1.7	<0.01	0.02
Arsenic	58	58	100.0	4.60	1.78-9.46
<i>p,p'</i> -DDE	60	7	11.7	0.01	0.01-0.17
<i>o,p'</i> -DDT	60	2	3.3	<0.01	0.01-0.03
<i>p,p'</i> -DDT	60	5	8.3	0.01	0.01-0.22
DDTR	60	8	13.3	0.01	0.01-0.42
Dieldrin	60	5	8.3	0.01	0.01-0.61
Endrin	60	3	5.0	<0.01	0.01-0.02
Endrin ketone	60	1	1.7	<0.01	0.05
<i>p,p'</i> -TDE	60	1	1.7	<0.01	0.01
CONNECTICUT					
Arsenic	2	2	100.0	3.96	2.33-5.59
<i>p,p'</i> -DDE	2	1	50.0	0.01	0.01
<i>p,p'</i> -DDT	2	1	50.0	0.03	0.05
DDTR	2	1	50.0	0.03	0.06
Dieldrin	2	1	50.0	0.01	0.01
DELAWARE					
Arsenic	3	3	100.0	2.97	0.95-5.88
<i>p,p'</i> -DDE	3	1	33.3	<0.01	0.01
DDTR	3	1	33.3	<0.01	0.01
Dieldrin	3	1	33.3	<0.01	0.01
FLORIDA					
Aldrin	18	1	5.6	0.03	0.47
Arsenic	18	16	88.9	0.77	0.25-3.08
Chlordane	18	9	50.0	0.36	0.04-3.32
<i>o,p'</i> -DDE	18	2	11.1	0.01	0.03-0.06
<i>p,p'</i> -DDE	18	13	72.2	0.25	0.01-2.40
<i>o,p'</i> -DDT	18	9	50.0	0.10	0.01-0.98
<i>p,p'</i> -DDT	18	14	77.8	0.37	0.01-2.08
DDTR	18	14	77.8	0.85	0.01-5.03

TABLE 3.—Pesticide residues in cropland soil from 43 States—FY 1969—Continued

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
FLORIDA—Continued					
Diazinon	5	1	20.0	0.03	0.15
Dieldrin	18	7	38.9	0.08	0.01-0.52
Endrin	18	2	11.1	0.03	0.13-0.38
Endrin aldehyde	18	1	5.6	<0.01	0.03
Endrin ketone	18	1	5.6	<0.01	0.03
Ethion	5	1	20.0	0.01	0.03
Heptachlor	18	1	5.6	<0.01	0.05
Heptachlor epoxide	18	3	16.7	0.01	0.01-0.07
Ethyl parathion	5	2	40.0	0.60	0.01-3.01
<i>o,p'</i> -TDE	18	1	5.6	0.02	0.34
<i>p,p'</i> -TDE	18	11	61.1	0.11	0.01-0.64
Toxaphene	18	2	11.1	0.08	0.62-0.77
Trifluralin	18	1	5.6	<0.01	0.03
GEORGIA					
Arsenic	29	29	100.0	2.61	0.37-10.72
Chlordane	22	1	4.6	0.01	0.19
2,4-D	3	1	33.3	<0.01	0.01
<i>o,p'</i> -DDE	22	5	22.7	0.01	0.01-0.08
<i>p,p'</i> -DDE	22	20	90.9	0.18	0.01-1.04
<i>o,p'</i> -DDT	22	13	59.1	0.09	0.01-0.73
<i>p,p'</i> -DDT	22	18	81.8	0.56	0.01-4.64
DDTR	22	21	95.5	0.96	0.01-6.31
DEF	22	1	4.6	0.01	0.12
Dieldrin	22	4	18.2	<0.01	0.01-0.03
Endrin	22	1	4.6	0.02	0.42
Heptachlor epoxide	22	1	4.6	<0.01	0.02
PCNB	22	1	4.6	0.03	0.69
<i>o,p'</i> -TDE	22	1	4.6	0.02	0.34
<i>p,p'</i> -TDE	22	15	68.2	0.10	0.01-1.23
Toxaphene	22	8	36.4	0.60	0.43-5.63
Trifluralin	22	3	13.6	<0.01	0.02-0.04
IDAHO					
Arsenic	33	32	97.0	3.22	0.47-8.58
Chlordane	33	2	6.1	<0.01	0.03-0.07
<i>p,p'</i> -DDE	33	8	24.2	0.01	0.01-0.09
<i>o,p'</i> -DDT	33	6	18.2	0.01	0.01-0.13
<i>p,p'</i> -DDT	33	8	24.2	0.04	0.01-0.67
DDTR	33	8	24.2	0.07	0.02-1.03
Dieldrin	33	3	9.1	0.01	0.03-0.11
Heptachlor epoxide	33	1	3.0	<0.01	0.01
<i>o,p'</i> -TDE	33	3	9.1	<0.01	0.01
<i>p,p'</i> -TDE	33	6	18.2	0.01	0.01-0.15
Trifluralin	33	2	6.1	0.01	0.01-0.24
ILLINOIS					
Aldrin	142	60	42.3	0.13	0.01-2.24
Arsenic	142	142	100.0	8.05	1.54-33.40
Atrazine	43	2	4.7	<0.01	0.02-0.10
Chlordane	142	36	25.4	0.23	0.02-5.20
<i>p,p'</i> -DDE	142	16	11.3	<0.01	0.01-0.05
<i>o,p'</i> -DDT	142	4	2.8	<0.01	0.01-0.02
<i>p,p'</i> -DDT	142	12	8.5	<0.01	0.01-0.06
DDTR	142	16	11.3	0.01	0.01-0.29

TABLE 3.—Pesticide residues in cropland soil from 43 States—FY 1969—Continued

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
ILLINOIS—Continued					
Dieldrin	142	87	61.3	0.11	0.01-1.42
Heptachlor	142	31	21.8	0.03	0.01-0.59
Heptachlor epoxide	142	36	25.4	0.02	0.01-1.08
Isodrin	142	2	1.4	< 0.01	0.02
<i>o,p'</i> -TDE	142	1	0.7	< 0.01	0.06
<i>p,p'</i> -TDE	142	5	3.5	< 0.01	0.01-0.16
Trifluralin	142	2	1.4	< 0.01	0.05-0.16
INDIANA					
Aldrin	78	13	16.7	0.07	0.01-3.06
Arsenic	78	78	100.0	7.88	1.28-19.65
Chlordane	78	4	5.1	0.02	0.07-0.53
<i>p,p'</i> -DDE	78	1	1.3	< 0.01	0.03
<i>o,p'</i> -DDT	78	2	2.6	< 0.01	0.01-0.03
<i>p,p'</i> -DDT	78	2	2.6	< 0.01	0.02-0.09
DDTR	78	2	2.6	< 0.01	0.06-0.14
Dieldrin	78	21	26.9	0.03	0.01-0.58
Heptachlor	78	2	2.6	< 0.01	0.02-0.08
Heptachlor epoxide	78	1	1.3	< 0.01	0.02
Isodrin	78	1	1.3	< 0.01	0.03
<i>p,p'</i> -TDE	78	2	2.6	< 0.01	0.01
Trifluralin	78	1	1.3	< 0.01	0.03
IOWA					
Aldrin	151	48	31.8	0.04	0.01-1.37
Arsenic	152	152	100.0	7.51	0.86-107.45
Atrazine	48	13	27.1	0.05	0.01-1.55
Chlordane	151	32	21.2	0.13	0.04-6.30
<i>p,p'</i> -DDE	151	21	13.9	0.01	0.01-0.18
<i>o,p'</i> -DDT	151	6	4.0	< 0.01	0.01-0.05
<i>p,p'</i> -DDT	151	23	15.2	0.01	0.01-0.34
DDTR	151	25	16.6	0.03	0.01-0.60
Dieldrin	151	81	53.6	0.06	0.01-0.42
Heptachlor	151	14	9.3	0.02	0.01-0.97
Heptachlor epoxide	151	31	20.5	0.01	0.01-0.33
Isodrin	151	2	1.3	< 0.01	0.01-0.02
<i>o,p'</i> -TDE	151	1	0.7	< 0.01	0.10
<i>p,p'</i> -TDE	151	8	5.3	< 0.01	0.01-0.50
Trifluralin	151	5	3.3	< 0.01	0.02-0.08
KENTUCKY					
Aldrin	31	8	25.8	0.03	0.01-0.42
Arsenic	31	31	100.0	8.41	2.60-12.80
Chlordane	31	4	12.9	0.02	0.06-0.27
<i>o,p'</i> -DDE	31	1	3.2	< 0.01	0.03
<i>p,p'</i> -DDE	31	5	16.1	0.01	0.01-0.17
<i>o,p'</i> -DDT	31	3	9.7	0.02	0.01-0.34
<i>p,p'</i> -DDT	31	6	19.4	0.04	0.02-1.00
DDTR	31	6	19.4	0.08	0.03-1.84
Dieldrin	31	17	54.8	0.06	0.01-0.65
Heptachlor	31	2	6.5	< 0.01	0.01
Heptachlor epoxide	31	1	3.2	< 0.01	0.02
Isodrin	31	2	6.5	< 0.01	0.01
<i>o,p'</i> -TDE	31	1	3.2	< 0.01	0.02
<i>p,p'</i> -TDE	31	4	12.9	0.01	0.02-0.28

TABLE 3.—Pesticide residues in cropland soil from 43 States—FY 1969—Continued

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
LOUISIANA					
Aldrin	27	5	18.5	<0.01	0.01-0.02
Arsenic	27	26	96.3	2.15	0.26-6.34
Chlordane	27	1	3.7	<0.01	0.11
<i>o,p'</i> -DDE	27	2	7.4	<0.01	0.04-0.05
<i>p,p'</i> -DDE	27	12	44.4	0.19	0.01-2.55
<i>o,p'</i> -DDT	27	9	33.3	0.10	0.02-1.45
<i>p,p'</i> -DDT	27	13	48.2	0.61	0.02-7.17
DDTR	27	13	48.2	0.99	0.03-10.99
Dieldrin	27	10	37.0	0.02	0.01-0.13
Endrin	27	1	3.7	<0.01	0.06
Endrin ketone	27	1	3.7	<0.01	0.02
<i>p,p'</i> -TDE	27	9	33.3	0.08	0.02-1.63
Toxaphene	27	4	14.8	0.57	0.59-11.72
Trifluralin	27	1	3.7	<0.01	0.07
MAINE					
Arsenic	8	8	100.0	16.01	5.06-44.06
Chlordane	8	1	12.5	0.02	0.12
<i>p,p'</i> -DDE	8	6	75.0	0.12	0.02-0.36
<i>o,p'</i> -DDT	8	5	62.5	0.13	0.02-0.46
<i>p,p'</i> -DDT	8	6	75.0	0.54	0.04-1.87
DDTR	8	6	75.0	0.85	0.08-2.86
Endrin	8	1	12.5	0.02	0.15
Heptachlor	8	1	12.5	<0.01	0.01
Heptachlor epoxide	8	1	12.5	<0.01	0.01
<i>p,p'</i> -TDE	8	6	75.0	0.06	0.01-0.19
MARYLAND					
Arsenic	12	12	100.0	5.69	3.40-11.90
Chlordane	12	1	8.3	0.01	0.09
<i>p,p'</i> -DDE	12	2	16.7	<0.01	0.02
<i>p,p'</i> -DDT	12	1	8.3	<0.01	0.03
DDTR	12	2	16.7	0.01	0.02-0.05
Dieldrin	12	1	7.3	0.01	0.06
Heptachlor epoxide	12	1	8.3	<0.01	0.02
MASSACHUSETTS					
Arsenic	2	2	100.0	9.75	7.35-12.15
<i>p,p'</i> -DDE	2	1	50.0	0.17	0.34
<i>o,p'</i> -DDT	2	1	50.0	0.10	0.20
<i>p,p'</i> -DDT	2	1	50.0	0.49	0.97
DDTR	2	1	50.0	0.78	1.55
<i>p,p'</i> -TDE	2	1	50.0	0.02	0.04
MICHIGAN					
Aldrin	51	2	3.9	<0.01	0.01-0.10
Arsenic	52	52	100.0	4.85	0.13-11.94
Atrazine	13	1	7.7	0.01	0.18
<i>o,p'</i> -DDE	51	4	7.8	<0.01	0.01-0.14
<i>p,p'</i> -DDE	51	12	23.5	0.16	0.01-4.58
<i>o,p'</i> -DDT	51	3	5.9	0.18	0.15-6.29
<i>p,p'</i> -DDT	51	5	9.8	1.10	0.01-35.92
DDTR	51	12	23.5	2.09	0.01-78.36

TABLE 3.—Pesticide residues in cropland soil from 43 States—FY 1969—Continued

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
MICHIGAN—Continued					
Dieldrin	51	11	21.6	0.05	0.01-1.01
Endosulfan (1)	51	2	3.9	0.01	0.03-0.24
Endosulfan sulfate	51	2	3.9	0.02	0.25-0.94
Endrin	51	1	2.0	<0.01	0.01
<i>p,p'</i> -TDE	51	5	9.8	0.65	0.02-31.43
MISSISSIPPI					
Arsenic	30	30	100.0	5.70	1.10-16.90
<i>o,p'</i> -DDE	29	9	31.0	0.01	0.01-0.08
<i>p,p'</i> -DDE	29	26	89.7	0.31	0.01-1.43
<i>o,p'</i> -DDT	29	22	75.9	0.20	0.02-1.35
<i>p,p'</i> -DDT	29	26	89.7	1.36	0.01-9.28
DDTR	29	26	89.7	2.06	0.03-13.14
Dieldrin	29	10	34.5	0.01	0.02-0.10
Endrin	29	1	3.5	0.01	0.19
Endrin ketone	29	1	3.5	<0.01	0.11
Lindane	29	2	6.9	<0.01	0.01-0.04
<i>o,p'</i> -TDE	29	2	6.9	0.03	0.33-0.49
<i>p,p'</i> -TDE	29	20	69.0	0.15	0.01-0.81
Toxaphene	29	14	48.3	0.78	0.10-8.80
Trifluralin	29	6	20.7	0.02	0.02-0.25
MISSOURI					
Aldrin	82	18	22.0	0.05	0.01-1.59
Arsenic	81	80	98.8	5.99	0.49-24.51
Chlordane	82	6	7.3	0.03	0.17-0.60
<i>p,p'</i> -DDE	82	3	3.7	<0.01	0.01
<i>o,p'</i> -DDT	82	2	2.4	<0.01	0.01-0.02
<i>p,p'</i> -DDT	82	3	3.7	<0.01	0.02-0.09
DDTR	82	3	3.7	<0.01	0.03-0.12
Dieldrin	82	26	31.7	0.04	0.01-0.55
Endrin	82	1	1.2	<0.01	0.01
Heptachlor	82	5	6.1	<0.01	0.01-0.04
Heptachlor epoxide	82	5	6.1	<0.01	0.01-0.06
Isodrin	82	1	1.2	<0.01	0.03
Toxaphene	82	1	1.2	0.04	3.15
Trifluralin	82	5	6.1	<0.01	0.02-0.10
NEBRASKA					
Aldrin	106	2	1.9	<0.01	0.01
Arsenic	106	106	100.0	5.81	0.33-15.80
Atrazine	72	12	16.7	<0.01	0.01-0.12
Chlordane	106	11	10.4	0.01	0.03-0.18
<i>p,p'</i> -DDE	106	14	13.2	0.01	0.01-0.10
<i>o,p'</i> -DDT	106	6	5.7	<0.01	0.01-0.08
<i>p,p'</i> -DDT	106	12	11.3	0.01	0.01-0.19
DDTR	106	16	15.1	0.01	0.02-0.31
Dicofol	106	2	1.9	<0.01	0.10
Dieldrin	106	37	34.9	0.02	0.01-0.19
Endrin	106	1	0.9	<0.01	0.02
Heptachlor	106	1	0.9	<0.01	0.01
Heptachlor epoxide	106	12	11.3	<0.01	0.01-0.03
Malathion	2	1	50.0	0.18	0.36
<i>p,p'</i> -TDE	106	4	3.8	<0.01	0.01-0.05

TABLE 3.—Pesticide residues in cropland soil from 43 States—FY 1969—Continued

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
NEVADA					
Arsenic	2	2	100.0	2.32	1.77-2.86
NEW HAMPSHIRE					
Arsenic	2	2	100.0	5.35	1.31-9.38
<i>p,p'</i> -DDE	2	1	50.0	0.02	0.03
DDTR	2	1	50.0	0.02	0.03
NEW JERSEY					
Arsenic	5	5	100.0	11.72	4.55-17.21
<i>o,p'</i> -DDE	5	1	20.0	<0.01	0.02
<i>p,p'</i> -DDE	5	2	40.0	0.17	0.18-0.66
<i>o,p'</i> -DDT	5	1	20.0	0.06	0.28
<i>p,p'</i> -DDT	5	2	40.0	0.24	0.05-1.17
DDTR	5	2	40.0	0.55	0.26-2.48
Dieldrin	5	2	40.0	0.05	0.05-0.21
Endosulfan (II)	5	1	20.0	<0.01	0.02
Endosulfan sulfate	5	1	20.0	0.02	0.11
Heptachlor epoxide	5	1	20.0	<0.01	0.01
Lindane	5	1	20.0	0.01	0.03
Ethyl parathion	1	1	100.0	0.02	0.02
<i>o,p'</i> -TDE	5	1	20.0	0.02	0.09
<i>p,p'</i> -TDE	5	2	40.0	0.06	0.03-0.26
NEW MEXICO					
Arsenic	10	10	100.0	4.64	0.66-15.82
<i>p,p'</i> -DDE	10	4	40.0	0.02	0.01-0.11
<i>o,p'</i> -DDT	10	1	10.0	<0.01	0.01
<i>p,p'</i> -DDT	10	4	40.0	0.01	0.01-0.03
DDTR	10	4	40.0	0.02	0.02-0.15
Dieldrin	10	1	10.0	<0.01	0.01
NEW YORK					
Arsenic	37	35	94.6	9.38	1.24-43.90
Chlordane	38	1	2.6	0.08	3.19
<i>o,p'</i> -DDE	38	3	7.9	<0.01	0.01-0.06
<i>p,p'</i> -DDE	38	15	39.5	0.23	0.01-3.70
<i>o,p'</i> -DDT	38	11	29.0	0.07	0.01-1.45
<i>p,p'</i> -DDT	38	13	34.2	0.53	0.02-7.67
DDTR	38	15	39.5	0.91	0.01-13.29
Dieldrin	38	13	34.2	0.05	0.01-0.96
Endrin	38	1	2.6	0.01	0.56
Endrin ketone	38	1	2.6	<0.01	0.05
Lindane	38	3	7.9	0.01	0.01-0.23
Methoxychlor	38	1	2.6	0.01	0.28
<i>o,p'</i> -TDE	38	2	5.3	0.01	0.06-0.37
<i>p,p'</i> -TDE	38	10	26.3	0.07	0.01-1.49
NORTH CAROLINA					
Aldrin	31	3	9.7	0.05	0.01-1.12
Arsenic	27	27	100.0	6.18	0.73-22.00
Chlordane	31	1	3.2	<0.01	0.11

TABLE 3.—Pesticide residues in cropland soil from 43 States—FY 1969—Continued

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
NORTH CAROLINA—Continued					
<i>o,p'</i> -DDE	31	6	19.4	<0.01	0.01-0.03
<i>p,p'</i> -DDE	31	22	71.0	0.08	0.01-0.44
<i>o,p'</i> -DDT	31	14	45.2	0.07	0.03-0.83
<i>p,p'</i> -DDT	31	19	61.3	0.28	0.01-1.75
DDTR	31	22	71.0	0.53	0.02-2.88
Dieldrin	31	10	32.3	0.08	0.01-1.53
Endrin	31	2	6.5	<0.01	0.01-0.08
Heptachlor	31	2	6.5	<0.01	0.01-0.02
Heptachlor epoxide	31	4	12.9	<0.01	0.01-0.03
Isodrin	31	1	3.2	<0.01	0.01
Ethyl parathion	6	1	16.7	<0.01	0.02
<i>o,p'</i> -TDE	31	11	35.5	0.03	0.03-0.17
<i>p,p'</i> -TDE	31	19	61.3	0.07	0.01-0.27
Toxaphene	31	7	22.6	0.28	0.34-3.20
Trifluralin	31	2	6.5	<0.01	0.03-0.11
NORTH DAKOTA					
Aldrin	157	1	0.6	<0.01	0.03
Arsenic	158	158	100.0	8.50	0.98-37.53
Chlordane	157	3	1.9	<0.01	0.08-0.15
<i>p,p'</i> -DDE	157	10	6.4	<0.01	0.01-0.14
<i>o,p'</i> -DDT	157	5	3.2	<0.01	0.01-0.19
<i>p,p'</i> -DDT	157	8	5.1	0.01	0.01-0.56
DDTR	157	10	6.4	0.01	0.01-0.95
Dieldrin	157	12	7.6	<0.01	0.01-0.20
Endrin	157	1	0.6	<0.01	0.01
Heptachlor epoxide	157	3	1.9	<0.01	0.02-0.07
<i>p,p'</i> -TDE	157	7	4.5	<0.01	0.01-0.06
OHIO					
Aldrin	68	10	14.7	0.03	0.01-0.74
Arsenic	69	69	100.0	11.23	1.15-41.49
Chlordane	68	3	4.4	0.01	0.01-0.71
<i>o,p'</i> -DDE	68	1	1.5	<0.01	0.20
<i>p,p'</i> -DDE	68	11	16.2	0.03	0.01-1.77
<i>o,p'</i> -DDT	68	2	2.9	0.01	0.19-0.22
<i>p,p'</i> -DDT	68	6	8.8	0.04	0.01-1.27
DDTR	68	11	16.2	0.08	0.01-3.38
Dieldrin	68	19	27.9	0.02	0.01-0.30
Endosulfan (I)	68	1	1.5	<0.01	0.07
Endosulfan (II)	68	1	1.5	<0.01	0.29
Endosulfan sulfate	68	1	1.5	0.01	0.40
Heptachlor	68	2	2.9	<0.01	0.01
Heptachlor epoxide	68	1	1.5	<0.01	0.01
Isodrin	68	2	2.9	<0.01	0.01-0.03
Lindane	68	1	1.5	0.01	0.35
<i>p,p'</i> -TDE	68	3	4.4	<0.01	0.04-0.12
Trifluralin	68	1	1.5	<0.01	0.06
OKLAHOMA					
Arsenic	62	60	96.8	3.30	0.24-14.58
Chlordane	64	1	1.6	<0.01	0.07
<i>p,p'</i> -DDE	64	10	15.6	<0.01	0.01-0.09
<i>o,p'</i> -DDT	64	1	1.6	<0.01	0.01
<i>p,p'</i> -DDT	64	9	14.1	<0.01	0.01-0.09
DDTR	64	10	15.6	0.01	0.02-0.17

TABLE 3.—Pesticide residues in cropland soil from 43 States—FY 1969—Continued

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
OKLAHOMA—Continued					
Dieldrin	64	2	3.1	<0.01	0.01
Heptachlor epoxide	64	1	1.6	<0.01	0.01
<i>p,p'</i> -TDE	64	2	3.1	<0.01	0.01-0.02
Trifluralin	64	1	1.6	<0.01	0.03
PENNSYLVANIA					
Arsenic	29	29	100.0	10.80	2.96-64.94
Chlordane	29	6	20.7	0.07	0.02-0.92
<i>o,p'</i> -DDE	29	1	3.5	<0.01	0.08
<i>p,p'</i> -DDE	29	9	31.0	0.07	0.01-1.52
<i>o,p'</i> -DDT	29	5	17.2	0.03	0.01-0.67
<i>p,p'</i> -DDT	29	8	27.6	0.12	0.01-2.99
DDTR	29	11	37.9	0.27	0.01-5.50
Dicofol	29	1	3.5	0.02	0.53
Dieldrin	29	10	34.5	0.02	0.01-0.14
Endosulfan (II)	29	1	3.5	<0.01	0.02
Endosulfan sulfate	29	1	3.5	<0.01	0.01
Heptachlor epoxide	29	4	13.8	<0.01	0.01-0.03
Ethyl parathion	3	1	33.3	<0.01	0.01
<i>o,p'</i> -TDE	29	4	13.8	0.01	0.03-0.20
<i>p,p'</i> -TDE	29	7	24.1	0.04	0.01-0.55
Trifluralin	29	2	6.9	<0.01	0.01-0.07
RHODE ISLAND					
Arsenic	1	1	100.0	21.30	21.30
<i>p,p'</i> -DDE	1	1	100.0	0.23	0.23
<i>o,p'</i> -DDT	1	1	100.0	0.25	0.25
<i>p,p'</i> -DDT	1	1	100.0	2.46	2.46
DDTR	1	1	100.0	3.00	3.00
Dieldrin	1	1	100.0	0.11	0.11
<i>p,p'</i> -TDE	1	1	100.0	0.06	0.06
SOUTH CAROLINA					
Aldrin	17	1	5.9	0.01	0.14
Arsenic	17	17	100.0	3.28	0.53-19.54
2,4-D	2	1	50.0	0.02	0.03
<i>o,p'</i> -DDE	17	7	41.2	0.01	0.01-0.05
<i>p,p'</i> -DDE	17	14	82.4	0.24	0.01-0.93
<i>o,p'</i> -DDT	17	12	70.6	0.15	0.01-0.95
<i>p,p'</i> -DDT	17	11	64.7	0.64	0.12-3.15
DDTR	17	15	88.2	1.17	0.01-4.78
Dieldrin	17	3	17.7	0.04	0.02-0.56
Endrin	17	3	17.7	<0.01	0.01-0.05
Heptachlor epoxide	17	3	17.7	<0.01	0.01
Lindane	17	1	5.9	<0.01	0.01
<i>o,p'</i> -TDE	17	5	29.4	0.03	0.03-0.19
<i>p,p'</i> -TDE	17	14	82.4	0.10	0.01-0.34
Toxaphene	17	1	5.9	0.10	1.74
Trifluralin	17	5	29.4	0.01	0.01-0.08
SOUTH DAKOTA					
Arsenic	101	101	100.0	5.80	0.47-34.54
Chlordane	106	3	2.8	0.01	0.10-0.66
<i>p,p'</i> -DDE	106	2	1.9	<0.01	0.01-0.03

TABLE 3.—Pesticide residues in cropland soil from 43 States—FY 1969—Continued

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
SOUTH DAKOTA—Continued					
<i>o,p'</i> -DDT	106	2	1.9	<0.01	0.01-0.03
<i>p,p'</i> -DDT	106	2	1.9	<0.01	0.02-0.04
DDTR	106	4	3.8	<0.01	0.01-0.10
Dieldrin	106	9	8.5	0.01	0.01-0.25
Heptachlor	106	1	0.9	<0.01	0.01
Heptachlor epoxide	106	3	2.8	<0.01	0.01-0.03
Lindane	106	3	2.8	<0.01	0.01-0.02
<i>p,p'</i> -TDE	106	1	0.9	<0.01	0.02
TENNESSEE					
Arsenic	27	27	100.0	8.05	2.31-15.63
Chlordane	27	1	3.7	0.01	0.20
<i>p,p'</i> -DDE	27	10	37.0	0.02	0.01-0.26
<i>o,p'</i> -DDT	27	7	25.9	0.01	0.01-0.08
<i>p,p'</i> -DDT	27	10	37.0	0.05	0.01-0.38
DDTR	27	11	40.7	0.11	0.01-0.70
Dieldrin	27	6	22.2	<0.01	0.01-0.03
Endrin	27	1	3.7	<0.01	0.02
<i>p,p'</i> -TDE	27	6	22.2	0.03	0.02-0.36
Toxaphene	27	4	14.8	0.14	0.13-2.19
Trifluralin	27	2	7.4	<0.01	0.04-0.05
UTAH					
Arsenic	12	11	91.7	4.16	0.62-12.66
Chlordane	12	4	33.3	0.04	0.02-0.25
<i>p,p'</i> -DDE	12	2	16.7	<0.01	0.01-0.02
<i>p,p'</i> -DDT	12	1	8.3	<0.01	0.03
DDTR	12	2	16.7	0.01	0.01-0.05
Dieldrin	12	2	16.7	0.01	0.02-0.15
Heptachlor	12	2	16.7	0.02	0.02-0.26
Heptachlor epoxide	12	3	25.0	0.01	0.02-0.05
VERMONT					
Arsenic	4	4	100.0	1.79	0.99-2.30
<i>p,p'</i> -DDE	5	1	20.0	<0.01	0.01
DDTR	5	1	20.0	<0.01	0.01
Dieldrin	5	1	20.0	<0.01	0.01
VIRGINIA					
Aldrin	21	1	4.8	<0.01	0.01
Arsenic	20	20	100.0	3.34	0.69-12.34
Chlordane	21	5	23.8	0.01	0.01-0.11
<i>p,p'</i> -DDE	21	11	52.4	0.02	0.01-0.22
<i>o,p'</i> -DDT	21	4	19.1	0.01	0.01-0.17
<i>p,p'</i> -DDT	21	8	38.1	0.11	0.01-1.31
DDTR	21	11	52.4	0.17	0.01-1.75
Dieldrin	21	6	28.6	0.08	0.01-1.60
Heptachlor epoxide	21	4	19.1	0.01	0.01-0.05
Malathion	1	1	100.0	0.04	0.04
Ethyl parathion	1	1	100.0	0.90	0.90
<i>o,p'</i> -TDE	21	1	4.8	<0.01	0.07
<i>p,p'</i> -TDE	21	7	33.3	0.02	0.01-0.19
Toxaphene	21	1	4.7	0.01	0.28

TABLE 3.—Pesticide residues in cropland soil from 43 States—FY 1969—Continued

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
WASHINGTON					
Aldrin	45	2	4.4	<0.01	0.09-0.10
Arsenic	45	45	100.0	2.61	0.71-7.02
2,4-D	6	1	16.7	<0.01	0.01
<i>o,p'</i> -DDE	45	2	4.4	<0.01	0.01-0.09
<i>p,p'</i> -DDE	45	10	22.2	0.17	0.01-6.99
<i>o,p'</i> -DDT	45	6	13.3	0.06	0.01-2.58
<i>p,p'</i> -DDT	45	10	22.2	0.46	0.01-19.75
DDTR	45	11	24.4	0.72	0.01-30.69
Dieldrin	45	8	17.8	0.02	0.01-0.30
<i>o,p'</i> -TDE	45	1	2.2	<0.01	0.17
<i>p,p'</i> -TDE	45	3	6.7	0.03	0.01-1.11
Toxaphene	45	1	2.2	0.02	0.73
Trifluralin	45	1	2.2	<0.01	0.08
WEST VIRGINIA					
Arsenic	6	6	100.0	6.33	4.36-8.17
Chlordane	6	3	50.0	0.21	0.09-0.78
<i>p,p'</i> -DDE	6	2	33.3	0.02	0.04-0.10
<i>p,p'</i> -DDT	6	2	33.3	0.01	0.01-0.07
DDTR	6	2	33.3	0.04	0.05-0.17
Dieldrin	6	1	16.7	0.04	0.23
Heptachlor epoxide	6	3	50.0	0.06	0.08-0.18
WISCONSIN					
Aldrin	67	5	7.5	<0.01	0.01-0.04
Arsenic	68	68	100.0	3.78	0.34-10.01
Chlordane	67	3	4.5	0.01	0.04-0.32
<i>o,p'</i> -DDE	67	1	1.5	<0.01	0.02
<i>p,p'</i> -DDE	67	9	13.4	0.01	0.01-0.27
<i>o,p'</i> -DDT	67	3	4.5	0.01	0.05-0.20
<i>p,p'</i> -DDT	67	7	10.5	0.01	0.01-0.30
DDTR	67	9	13.4	0.02	0.01-0.71
Dieldrin	67	9	13.4	0.01	0.01-0.17
Heptachlor	67	2	3.0	<0.01	0.01
Heptachlor epoxide	67	2	3.0	<0.01	0.01
<i>p,p'</i> -TDE	67	4	6.0	<0.01	0.01-0.12
Trifluralin	67	1	1.5	<0.01	0.01
WYOMING					
Arsenic	17	14	82.4	1.71	0.40-10.88
Chlordane	17	4	23.5	0.05	0.03-0.48
Dieldrin	17	6	35.3	0.02	0.02-0.19
Heptachlor epoxide	17	3	17.7	0.01	0.02-0.05

¹ One sample per site.² Percent based on number of sites with residues greater than or equal to the sensitivity limits.

TABLE 4.—Pesticide residues in noncropland soil from 11 States—FY 1969

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
ARIZONA					
Arsenic	44	44	100.0	6.63	1.35-30.64
Chlordane	44	1	2.3	<0.01	0.08
<i>p,p'</i> -DDE	44	8	18.2	<0.01	0.01-0.06
<i>p,p'</i> -DDT	44	1	2.3	<0.01	0.03
DDTR	44	8	18.2	<0.01	0.01-0.09
Dieldrin	44	1	2.3	<0.01	0.03
GEORGIA					
Arsenic	19	18	94.7	1.47	0.53-4.29
<i>p,p'</i> -DDE	10	6	60.0	0.02	0.01-0.07
<i>o,p'</i> -DDT	10	2	20.0	<0.01	0.01-0.02
<i>p,p'</i> -DDT	10	5	50.0	0.02	0.01-0.10
DDTR	10	7	70.0	0.05	0.01-0.12
Dieldrin	10	1	10.0	<0.01	0.01
<i>p,p'</i> -TDE	10	1	10.0	<0.01	0.01
IDAHO					
Arsenic	26	26	100.0	7.73	1.01-39.07
<i>p,p'</i> -DDE	26	3	11.5	<0.01	0.01
<i>o,p'</i> -DDT	26	1	3.9	<0.01	0.02
<i>p,p'</i> -DDT	26	1	3.9	<0.01	0.06
DDTR	26	3	11.5	0.01	0.01-0.11
<i>p,p'</i> -TDE	26	1	3.9	<0.01	0.02
IOWA					
Aldrin	7	1	14.3	<0.01	0.02
Arsenic	7	7	100.0	7.08	1.71-17.39
MAINE					
Arsenic	8	8	100.0	5.14	1.40-13.00
<i>p,p'</i> -DDE	11	1	9.1	0.02	0.18
<i>o,p'</i> -DDT	11	1	9.1	<0.01	0.03
<i>p,p'</i> -DDT	11	1	9.1	0.02	0.23
DDTR	11	1	9.1	0.06	0.62
<i>p,p'</i> -TDE	11	1	9.1	0.02	0.18
MARYLAND					
Arsenic	3	3	100.0	8.43	5.20-11.97
<i>p,p'</i> -DDE	3	1	33.3	0.02	0.05
<i>o,p'</i> -DDT	3	1	33.3	0.01	0.03
<i>p,p'</i> -DDT	3	2	66.7	0.05	0.03-0.11
DDTR	3	2	66.7	0.09	0.03-0.23
<i>p,p'</i> -TDE	3	1	33.3	0.01	0.04
NEBRASKA					
Arsenic	17	16	94.1	2.18	0.33-8.42
Chlordane	19	1	5.3	<0.01	0.04
<i>p,p'</i> -DDE	19	3	15.8	<0.01	0.01-0.04
<i>o,p'</i> -DDT	19	1	5.3	<0.01	0.01
<i>p,p'</i> -DDT	19	1	5.3	<0.01	0.02

TABLE 4.—Pesticide residues in noncropland soil from 11 States—FY 1969—Continued

COMPOUND	NUMBER OF SAMPLES ANALYZED ¹	NUMBER OF POSITIVE SAMPLES	PERCENT POSITIVE SITES ²	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
NEBRASKA—Continued					
DDTR	19	3	15.8	<0.01	0.01-0.07
Dicofol	19	2	10.5	0.02	0.10-0.29
Dieldrin	19	2	10.5	<0.01	0.01
Heptachlor epoxide	19	1	5.3	<0.01	0.01
VIRGINIA					
Arsenic	10	10	100.0	4.07	0.50-12.42
<i>p,p'</i> -DDE	13	3	23.1	0.01	0.03-0.07
DDTR	13	3	23.1	0.01	0.03-0.09
Dieldrin	13	2	15.4	0.01	0.03-0.09
<i>p,p'</i> -TDE	13	1	7.7	<0.01	0.02
WASHINGTON					
Arsenic	21	21	100.0	6.94	1.58-54.17
<i>p,p'</i> -DDE	21	3	14.3	<0.01	0.01-0.02
<i>p,p'</i> -DDT	21	2	9.5	<0.01	0.01
DDTR	21	3	14.3	<0.01	0.01-0.03
WEST VIRGINIA					
Arsenic	6	6	100.0	5.16	2.67-13.26
<i>p,p'</i> -DDE	8	1	12.5	<0.01	0.02
<i>p,p'</i> -DDT	8	1	12.5	0.01	0.05
DDTR	8	1	12.5	0.01	0.08
Dieldrin	8	1	12.5	0.01	0.04
<i>p,p'</i> -TDE	8	1	12.5	<0.01	0.01
WYOMING					
Arsenic	37	36	97.3	2.73	0.35-19.33
Chlordane	37	1	2.7	0.01	0.50
<i>o,p'</i> -DDE	37	1	2.7	<0.01	0.02
<i>p,p'</i> -DDE	37	1	2.7	0.01	0.31
<i>o,p'</i> -DDT	37	1	2.7	<0.01	0.05
<i>p,p'</i> -DDT	37	1	2.7	<0.01	0.18
DDTR	37	1	2.7	0.02	0.56
Dieldrin	37	1	2.7	<0.01	0.02
Heptachlor epoxide	37	1	2.7	<0.01	0.01
Toxaphene	37	1	2.7	0.01	0.52

¹ One sample per site.² Percent based on number of sites with residues greater than or equal to the sensitivity limits.

TABLE 5.—Arsenic residue data for the 12 States having the highest residue levels—FY 1969

STATE	NUMBER OF SAMPLES ANALYZED	PERCENT POSITIVE SITES ¹	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
Arkansas	47	100.0	9.0	1.7-28.2
Kentucky	31	100.0	8.4	2.6-12.8
New England ²	19	100.0	10.2	1.0-14.1
New York	37	94.6	9.4	1.2-43.9
North Dakota	158	100.0	8.5	1.0-37.5
Ohio	69	100.0	11.2	1.2-41.5
Pennsylvania	29	100.0	10.8	3.0-64.9

¹ Percent based on number of sites with residues greater than or equal to the sensitivity limits.

² Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, and Vermont

TABLE 6.—Pesticide residue data for 5 States having the highest DDTR residue levels—FY 1969

STATE	NUMBER OF SAMPLES ANALYZED	PERCENT POSITIVE SITES ¹	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
Alabama	22	90.9	1.13	0.05-8.08
California	65	84.6	1.47	0.01-41.81
Michigan	51	23.5	2.09	0.01-78.36
Mississippi	29	89.7	2.06	0.03-13.14
South Carolina	17	88.2	1.17	0.01-4.78

¹ Percent based on number of sites with residues greater than or equal to the sensitivity limits.

TABLE 7.—Residue data for the 7 States with the highest dieldrin residue levels—FY 1969

STATE	NUMBER OF SAMPLES ANALYZED	PERCENT POSITIVE SITES ¹	MEAN RESIDUE LEVEL (PPM)	RANGE OF DETECTED RESIDUES (PPM)
Florida	18	38.9	0.08	0.01-0.52
Illinois	142	61.3	0.11	0.01-1.42
Iowa	151	53.6	0.06	0.01-0.42
Kentucky	31	54.8	0.06	0.01-0.65
North Carolina	31	32.3	0.08	0.01-1.53
Virginia/West Virginia	27	25.9	0.07	0.01-1.60

¹ Percent based on number of sites with residues greater than or equal to the sensitivity limits.

TABLE 8.—Summary of pesticides used in FY 1969 on cropland for all 43 States

ALL STATES—1,684 SITES

COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB/ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB/ACRE)	COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB/ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB/ACRE)
Aldrin	4.16	1.25	0.0522	Dithane M-45	0.30	5.82	0.0173
Amiben	2.14	1.07	0.0229	Diuron	1.13	0.93	0.0105
Aramite	0.12	2.35	0.0028	DSMA	0.36	1.52	0.0054
Atrazine	7.66	1.88	0.1442	Endosulfan (I)	0.48	1.11	0.0053
Azinphosmethyl	0.59	1.70	0.0101	Endrin	0.48	2.21	0.0105
Azodrin	0.42	2.07	0.0086	EPN	0.06	1.50	0.0009
Bacillus thuringiensis	0.12	9.50	0.0113	EPTC	0.36	2.65	0.0094
Barban	0.12	0.17	0.0002	Ethion	0.24	2.06	0.0049
Benefin	0.18	1.36	0.0024	Ethylene dibromide	0.12	14.62	0.0174
Benzene hexachloride	0.06	3.00	0.0018	Falone	0.06	2.00	0.0012
Bidrin	0.24	0.18	0.0004	Ferbam	0.06	9.12	0.0054
Binapacryl	0.06	2.12	0.0013	Folex	0.06	1.50	0.0009
Bordeaux mixtures	0.06	0.50	0.0003	Heptachlor	1.96	0.33	0.0065
Cacodylic acid	0.06	0.01	0.0000	Herbasan	0.06	10.00	0.0060
Captan	11.16	0.12	0.0133	Hexachlorobenzene	0.06	0.01	0.0000
Carbaryl	1.72	3.64	0.0627	Lead arsenate	0.06	3.80	0.0023
Carbophenothion	0.18	1.83	0.0033	Lindane	0.65	0.03	0.0002
CDA	0.89	1.78	0.0158	Linuron	0.77	0.73	0.0056
Ceresan L	1.25	0.01	0.0002	Malathion	7.54	0.17	0.0127
Ceresan M	1.48	0.01	0.0001	Maleic hydrazide	0.36	1.43	0.0051
Ceresan red	1.84	0.01	0.0003	Maneb	0.30	2.14	0.0064
Chevron RE-5353	0.30	1.72	0.0051	MCPA	1.07	0.33	0.0035
Chlordane	0.12	3.10	0.0037	Methoxychlor	2.20	0.04	0.0008
Chlorobenzilate	0.12	1.31	0.0016	Methyl demeton	0.06	1.50	0.0009
Chloroneb	0.36	0.05	0.0002	Methylmercury dicyandiamide	5.46	0.01	0.0006
Chloroxirone	0.30	1.65	0.0049	Methylmercury nitrite	0.06	0.01	0.0000
Chromophon	0.06	0.15	0.0001	Mevinphos	0.36	1.48	0.0053
CIPC	0.12	1.50	0.0018	Mirex	0.24	0.01	0.0000
Copper carbonate	0.06	0.60	0.0004	Monuron	0.06	1.60	0.0010
Copper oxide	0.18	4.23	0.0075	MSMA	0.48	1.21	0.0058
Copper oxychloride sulfate	0.12	4.68	0.0056	Nabam	0.24	1.78	0.0042
Copper-8-quinolinate	0.06	0.01	0.0000	Naled	0.30	1.62	0.0048
Copper sulfate	0.36	13.53	0.0482	Nitralin	0.36	0.76	0.0027
Cotoran	0.48	0.74	0.0035	Nitrate	1.13	64.58	0.7286
2,4-D	15.14	0.54	0.0825	Norea	0.12	0.46	0.0006
2,4-DB	0.89	0.48	0.0042	NPA	0.36	1.01	0.0036
Dalapon	0.42	2.12	0.0088	Oxydemetonmethyl	0.18	0.40	0.0007
DDT technical	3.44	5.56	0.1915	Ethyl parathion	1.84	1.48	0.0272
DEF	0.59	1.66	0.0099	Methyl parathion	3.03	3.07	0.0929
Demeton	0.18	0.59	0.0011	PCNB	0.42	1.59	0.0066
Diazinon	1.96	1.22	0.0240	PCP	0.06	1.50	0.0009
Dicamba	0.30	0.39	0.0012	Phenylmercury urea	0.06	0.01	0.0000
Dichlone	0.12	2.00	0.0024	Phorate	0.65	2.17	0.0142
Dichloropropene	0.06	54.43	0.0323	Phosphamidon	0.12	0.13	0.0002
Dichloropropene	0.36	70.07	0.2496	Picloram	0.12	0.63	0.0007
Dichloroprop	0.06	2.00	0.0012	PMA	0.18	0.06	0.0001
Dicofol	0.42	2.12	0.0088	Polyram	0.06	10.40	0.0062
Dieldrin	1.19	0.17	0.0021	Prometryne	0.06	2.00	0.0012
Difolatan	0.06	0.01	0.0000	Propanil	0.42	3.96	0.0165
Dimetan	0.06	0.01	0.0000	Propazine	0.06	2.00	0.0012
Dimethoate	0.12	0.75	0.0009	Ramrod	1.37	1.45	0.0198
Dinitrobutylphenol	0.95	3.78	0.0359	Ro-Neet	0.12	1.88	0.0022
Dinitrocresol	0.06	3.00	0.0018	Roundup	0.12	0.78	0.0009
Dinocap	0.12	0.22	0.0003	Randox T	0.12	0.90	0.0011
Dioxathion	0.12	2.60	0.0031	Silvex	0.12	0.63	0.0007
Diphenamid	0.24	2.19	0.0052	Simazine	0.12	2.07	0.0025
Diquat	0.06	0.83	0.0005	Simetryne	0.06	2.00	0.0012
Disulfoton	1.72	1.77	0.0305	Sodium arsenite	0.24	5.25	0.0125
				Sodium chlorate	0.06	6.00	0.0036

TABLE 8.—Summary of pesticides used in FY 1969 on cropland for all 43 States—Continued

COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB./ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB./ACRE)
Strobane	0.12	16.50	0.0196
Sulfur	0.71	34.00	0.2423
2,4,5-T	0.18	0.83	0.0015
TCA	0.06	2.00	0.0012
TDE technical	0.36	2.31	0.0082
Tetradifon	0.12	0.50	0.0006
Thiram	1.07	0.03	0.0003
Toxaphene	1.90	9.87	0.1876
Trichlorofon	0.06	0.80	0.0005
Trifluralin	4.33	0.76	0.0327
Vernolate	0.53	1.29	0.0069
Zineb	0.18	4.90	0.0087
Ziram	0.06	0.80	0.0005

TABLE 9.—Summary of pesticides used in FY 1969 on noncropland for all 11 States

ALL STATES—195 SITES

COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB./ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB./ACRE)
2,4-D	0.51	2.00	0.0103
Malathion	0.51	0.61	0.0031
Mirex	0.51	0.01	0.0001

TABLE 10.—Summary of pesticides used in FY 1969 on cropland by State

COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB./ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB./ACRE)
ALABAMA—23 SITES			
Azodrin	4.35	0.84	0.0365
Benzene hexachloride	4.35	3.00	0.1304
Captan	21.74	0.04	0.0083
Carbaryl	4.35	0.40	0.0174
Ceresan M	4.35	0.01	0.0004
Copper sulfate	8.70	36.08	3.1374
Cotoran	4.35	1.50	0.0652
DDT technical	39.13	10.73	4.2000
DEF	4.35	1.50	0.0652
Disulfoton	8.70	0.35	0.0304
Diuron	8.70	0.95	0.0826
DSMA	4.35	1.00	0.0435
Endrin	8.70	1.20	0.1043
EPN	4.35	1.50	0.0652

TABLE 10.—Summary of pesticides used in FY 1969 on cropland by State—Continued

COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB./ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB./ACRE)
ALABAMA—23 SITES—Continued			
Malathion	8.70	2.50	0.2174
Ethyl parathion	4.35	1.00	0.0435
Methyl parathion	52.17	3.42	1.7848
MSMA	8.70	1.50	0.1304
Phorate	4.35	1.00	0.0435
Prometryne	4.35	2.00	0.0870
Thiram	4.35	0.02	0.0009
Toxaphene	17.39	3.45	0.6000
Trifluralin	47.83	0.61	0.2913
Vernolate	8.70	1.05	0.0913
ARIZONA—9 SITES			
Azodrin	11.11	6.25	0.6944
Captan	11.11	0.01	0.0011
Ceresan I	11.11	0.01	0.0011
Demeton	11.11	0.13	0.0144
Dieldrin	11.11	0.01	0.0011
Diuron	11.11	1.00	0.1111
Endosulfan (I)	11.11	2.00	0.2222
Naled	11.11	0.50	0.0556
Ethyl parathion	22.22	5.50	1.2222
Methyl parathion	44.44	2.75	1.2244
PCNB	11.11	0.75	0.0833
Phorate	11.11	1.50	0.1667
Strobane	11.11	15.00	1.6667
Toxaphene	11.11	2.00	0.2222
Trifluralin	22.22	1.12	0.2500
ARKANSAS—45 SITES			
Aldrin	2.22	0.25	0.0056
Captan	13.33	0.03	0.0036
Ceresan M	2.22	0.01	0.0002
Chloroxuron	6.67	1.00	0.0667
2,4-D	2.22	0.05	0.0011
2,4-DB	2.22	1.75	0.0389
DEF	2.22	1.00	0.0222
Dinitrobutylphenol	6.67	1.58	0.1056
Disulfoton	2.22	0.01	0.0002
Diuron	2.22	0.75	0.0167
DSMA	4.44	3.00	0.1333
Endrin	2.22	12.00	0.2667
Linuron	8.89	0.94	0.0833
NPA	6.67	0.54	0.0362
Nitralin	2.22	0.44	0.0098
Methyl parathion	2.22	12.00	0.2667
Propanil	4.44	5.50	0.2444
2,4,5-T	4.44	0.88	0.0389
Thiram	4.44	0.03	0.0016
Trifluralin	15.56	0.79	0.1222
CALIFORNIA—66 SITES			
Aramite	3.03	2.35	0.0712
Atrazine	1.52	2.50	0.0379

TABLE 10.—Summary of pesticides used in FY 1969 on cropland by State—Continued

COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLI-CATION RATE (LB/ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB/ACRE)	COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLI-CATION RATE (LB/ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB/ACRE)
CALIFORNIA—66 SITES—Continued				DELAWARE—3 SITES			
Azinphosmethyl	3.03	0.48	0.0145	Captan	33.33	0.04	0.0133
Bacillus thuringiensis	3.03	9.50	0.2879	Lindane	33.33	0.08	0.0267
Benefin	1.52	1.83	0.0277	FLORIDA—15 SITES			
Binapacryl	1.52	2.12	0.0321	Atrazine	6.67	0.80	0.0533
Bordeaux mixtures	1.52	0.50	0.0076	Azinphosmethyl	6.67	2.50	0.1667
Captan	1.52	2.30	0.0348	Captan	6.67	7.50	0.5000
Carbaryl	6.06	10.76	0.6521	Carbophenothion	6.67	2.00	0.1333
Carbophenothion	3.03	1.75	0.0530	Chlorobenzilate	13.33	1.31	0.1753
Ceresan red	3.03	0.01	0.0003	Copper oxide	6.67	7.50	0.5000
Chlordane	1.52	5.00	0.0758	Copper oxychloride sulfate	6.67	8.00	0.5333
2,4-D	3.03	0.63	0.0189	2,4-D	6.67	1.50	0.1000
DDT technical	13.64	2.82	0.3844	Dalapon	6.67	1.70	0.1133
Diazinon	6.06	0.99	0.0603	DDT technical	6.67	7.00	0.4667
Dichloropropene	4.55	8.67	0.3939	Diazinon	6.67	2.90	0.1933
Dichloroprop	1.52	2.00	0.0303	Dichloropropene	6.67	194.40	12.9600
Dicofol	7.58	2.59	0.1962	Dicofol	6.67	1.50	0.1000
Dimethoate	1.52	1.00	0.0152	Ethion	13.33	2.75	0.3667
Dioxathion	3.03	2.60	0.0788	Ferbam	6.67	9.12	0.6080
Diphenamid	1.52	2.62	0.0397	Mirex	6.67	0.01	0.0007
Disulfoton	3.03	4.00	0.1212	Ethyl parathion	13.33	2.85	0.3800
Diuron	1.52	0.75	0.0114	Methyl parathion	6.67	10.00	0.6667
Dithane M-45	3.03	5.00	0.1515	Sulfur	6.67	46.50	3.1000
Endosulfan (1)	7.58	0.99	0.0750	2,4,5-T	6.67	0.75	0.0500
Ethion	3.03	1.38	0.0417	TDE technical	6.67	10.00	0.6667
Malathion	4.55	1.65	0.0750	Toxaphene	6.67	2.00	0.1333
MCPA	3.03	0.76	0.0230	Zineb	13.33	6.55	0.8733
Mevinphos	9.09	1.48	0.1345	GEORGIA—28 SITES			
Nabam	1.52	3.50	0.0530	Atrazine	7.14	3.00	0.2143
Naled	6.06	1.90	0.1152	Azodrin	3.57	4.00	0.1429
Ethyl parathion	6.06	2.03	0.1230	Benefin	7.14	1.12	0.0800
Methyl parathion	4.55	6.10	0.2773	Captan	39.29	0.08	0.0307
Propanil	3.03	3.63	0.1098	Ceresan red	10.71	0.01	0.0011
Simazine	1.52	3.75	0.0568	Copper oxychloride sulfate	3.57	1.36	0.0486
Simetryne	1.52	2.00	0.0303	Copper sulfate	3.57	2.72	0.0971
Sulfur	7.58	35.79	2.7115	2,4-D	10.71	0.50	0.0536
Tetradifon	3.03	0.50	0.0152	DDT technical	21.43	14.18	3.0396
Toxaphene	6.06	9.75	0.5908	Disulfoton	3.57	2.00	0.0714
Trichlorofon	1.52	0.80	0.0121	Folex	3.57	1.50	0.0536
Trifluralin	6.06	0.88	0.0530	Malathion	21.43	0.34	0.0732
COLORADO—60 SITES				Maleic hydrazide	7.14	2.41	0.1725
Aldrin	3.33	0.08	0.0027	Methoxychlor	28.57	0.02	0.0046
Carbaryl	1.67	1.00	0.0167	Mirex	3.57	0.01	0.0004
Ceresan M	1.67	0.01	0.0002	Ethyl parathion	3.57	1.00	0.0357
2,4-D	10.00	0.51	0.0508	Methyl parathion	14.29	1.84	0.2632
2,4-DB	1.67	0.70	0.0117	PCNB	3.57	10.00	0.3571
Endrin	5.00	0.33	0.0167	Sulfur	7.14	40.20	2.8714
Malathion	1.67	0.60	0.0100	Thiram	10.71	0.04	0.0043
Ethyl parathion	1.67	0.25	0.0042	Toxaphene	17.86	14.45	2.5804
Picloram	1.67	1.00	0.0167	Trifluralin	7.14	1.25	0.0893
PMA	1.67	0.15	0.0025	CONNECTICUT—2 SITES			
CONNECTICUT—2 SITES				Atrazine	50.00	2.50	1.2500

TABLE 10.—Summary of pesticides used in FY 1969 on cropland by State—Continued

COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB/ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB ACRE)	COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB/ACRE)
IDAHO—33 SITES				INDIANA—75 SITES—Continued			
Captan	12.12	0.01	0.0015	Methylmercury dicyandiamide	2.67	0.01	0.0003
Ceresan M	6.06	0.01	0.0006	Ramrod	2.67	1.40	0.0373
Ceresan L	15.15	0.01	0.0015	Roundup	1.33	1.50	0.0200
CIPC	3.03	2.00	0.0606	Trifluralin	2.67	0.75	0.0200
2,4-D	12.12	2.12	0.2576	Zineb	1.33	1.60	0.0213
2,4-DB	3.03	0.50	0.0152				
DDT technical	6.06	0.50	0.0303	IOWA—151 SITES			
Dieldrin	3.03	0.01	0.0003	Aldrin	8.61	0.68	0.0587
Diquat	3.03	0.83	0.0252	Amiben	8.61	1.12	0.0966
EPTC	3.03	0.38	0.0115	Atrazine	10.60	2.00	0.2123
Hexachlorobenzene	3.03	0.01	0.0003	Captan	2.65	0.03	0.0008
Ro-Neet	6.06	1.87	0.1136	Carbaryl	0.66	1.00	0.0066
Trifluralin	6.06	0.56	0.0342	CDA A	1.32	1.50	0.0199
				Chevron RE-5353	0.66	1.00	0.0066
ILLINOIS—141 SITES				2,4-D	20.53	0.62	0.1278
Aldrin	19.15	1.52	0.2914	Diazinon	6.62	1.19	0.0791
Amiben	7.80	0.91	0.0707	Dicamba	0.66	0.50	0.0033
Atrazine	9.22	2.19	0.2023	Dieldrin	0.66	0.15	0.0010
Captan	49.65	0.06	0.0317	Heptachlor	4.64	0.35	0.0164
Carbaryl	0.71	4.80	0.0340	Lindane	1.32	0.06	0.0009
CDA A	7.80	1.51	0.1176	Ethyl parathion	0.66	0.32	0.0021
Ceresan red	0.71	0.01	0.0001	Phorate	1.32	0.95	0.0126
Ceresan L	0.71	0.06	0.0004	Ramrod	3.97	2.02	0.0801
Chevron RE-5353	1.42	2.56	0.0363	Randox T	0.66	0.40	0.0026
2,4-D	20.57	0.42	0.0864	Thiram	0.66	0.06	0.0004
2,4-DB	2.13	0.35	0.0075	Trifluralin	2.65	0.47	0.0125
Diazinon	3.55	1.86	0.0659				
Dieldrin	2.13	0.23	0.0050	KENTUCKY—31 SITES			
Heptachlor	9.93	0.46	0.0455	Aldrin	9.68	2.00	0.1935
Linuron	1.42	0.66	0.0094	Atrazine	19.35	1.33	0.2581
Malathion	39.72	0.03	0.0104	2,4-D	3.23	0.50	0.0161
Methoxychlor	10.64	0.01	0.0011	Dalapon	3.23	1.50	0.0484
Ramrod	7.80	1.22	0.0955	DDT technical	3.23	3.00	0.0968
Roundup	0.71	0.07	0.0005	EPTC	3.23	1.50	0.0484
Silvex	0.71	0.25	0.0018				
Thiram	0.71	0.01	0.0001	LOUISIANA—27 SITES			
Trifluralin	2.84	0.97	0.0277	Aldrin	22.22	0.08	0.0178
Vernolate	0.71	0.37	0.0026	Captan	3.70	0.25	0.0093
				Carbaryl	3.70	12.00	0.4444
				Ceresan L	3.70	0.01	0.0004
				Cotoran	3.70	1.00	0.0370
				2,4-D	11.11	1.58	0.1759
				Dalapon	3.70	2.00	0.0741
				DDT technical	7.41	23.25	1.7222
				DEF	3.70	9.00	0.3333
				Dimetan	3.70	0.01	0.0004
				Malathion	3.70	1.00	0.0370
				Methylmercury dicyandiamide	3.70	0.01	0.0004
				Methylmercury nitrile	3.70	0.01	0.0004
				MSMA	3.70	1.50	0.0556
				Nitrate	22.22	72.00	16.0000
INDIANA—75 SITES							
Aldrin	10.67	1.11	0.1187				
Amiben	5.33	0.85	0.0453				
Atrazine	13.33	1.79	0.2393				
Captan	26.67	0.01	0.0027				
Carbaryl	1.33	1.60	0.0213				
CDA A	1.33	1.07	0.0143				
Ceresan L	2.67	0.01	0.0003				
2,4-D	10.67	0.35	0.0373				
DDT technical	1.33	0.01	0.0001				
Dieldrin	1.33	0.01	0.0001				
Difolatan	1.33	0.01	0.0001				
Heptachlor	5.33	0.32	0.0172				
Malathion	17.33	0.01	0.0017				
Methoxychlor	4.00	0.01	0.0004				

TABLE 10.—Summary of pesticides used in FY 1969 on cropland by State—Continued

COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB/ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB/ACRE)	COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB/ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB/ACRE)
LOUISIANA—27 SITES—Continued				MISSISSIPPI—29 SITES			
Methyl parathion	7.41	7.20	0.5333	Azinphosmethyl	3.45	0.25	0.0086
Propanil	11.11	3.17	0.3519	Azodrin	6.90	0.76	0.0524
Silvex	3.70	1.00	0.0370	Bidrin	6.90	0.03	0.0024
Strobane	3.70	18.00	0.6667	Captan	24.14	0.09	0.0210
TCA	3.70	2.00	0.0741	Ceresan M	3.45	0.01	0.0003
Toxaphene	3.70	75.00	2.7778	Ceresan red	27.59	0.02	0.0045
Trifluralin	3.70	1.00	0.0370	Ceresan L	3.45	0.01	0.0003
MAINE—8 SITES				Chloroneb	17.24	0.06	0.0100
Dalapon	12.50	4.90	0.6125	Cotoran	13.79	0.47	0.0652
Dinitrobutylphenol	37.50	1.37	0.5125	DDT technical	31.03	3.47	1.0759
Disulfoton	25.00	8.50	2.1250	DEF	20.69	0.82	0.1690
Malathion	12.50	1.00	0.1250	Disulfoton	31.03	0.05	0.0152
Maneb	12.50	0.70	0.0875	Diuron	17.24	0.63	0.1079
Sodium arsenite	25.00	8.80	2.2000	DSMA	10.34	0.70	0.0728
MARYLAND—13 SITES				Endrin	3.45	2.00	0.0690
Atrazine	30.77	1.26	0.3885	Linuron	3.45	0.42	0.0145
Captan	30.77	0.03	0.0100	Malathion	6.90	1.40	0.0969
2,4-D	15.38	0.54	0.0838	Methoxychlor	3.45	0.01	0.0003
Dieldrin	7.69	0.01	0.0008	Mirex	6.90	0.01	0.0007
Lindane	15.38	0.01	0.0015	MSMA	10.34	1.44	0.1486
Malathion	23.08	0.01	0.0023	Norea	3.45	0.33	0.0114
Methoxychlor	7.69	0.01	0.0008	Nitralin	13.79	0.99	0.1372
Thiram	7.69	0.01	0.0008	Methyl parathion	41.38	2.14	0.8841
MASSACHUSETTS—2 SITES				PCNB	10.34	0.13	0.0131
Carbaryl	50.00	0.83	0.4150	Sodium chlorate	3.45	6.00	0.2069
Dinitrobutylphenol	50.00	3.06	1.5300	Toxaphene	34.48	7.50	2.5862
Disulfoton	50.00	1.50	0.7500	Trifluralin	37.93	0.85	0.3241
Dithane M-45	50.00	12.40	6.2000	Vernolate	3.45	0.30	0.0103
Maleic hydrazide	50.00	2.32	1.1600	MISSOURI—81 SITES			
Oxydemetonmethyl	50.00	0.25	0.1250	Aldrin	4.94	1.65	0.0815
Ethyl parathion	50.00	0.53	0.2650	Amiben	4.94	1.15	0.0568
MICHIGAN—51 SITES				Atrazine	12.35	1.87	0.2315
Atrazine	11.76	1.49	0.1753	Bidrin	1.23	0.10	0.0012
Azinphosmethyl	1.96	8.00	0.1569	Captan	1.23	0.01	0.0001
Captan	1.96	0.01	0.0002	Ceresan M	1.23	0.01	0.0001
CDA	1.96	6.00	0.1176	2,4-D	11.11	0.77	0.0856
Ceresan red	1.96	0.01	0.0002	2,4-DB	2.47	0.25	0.0062
CIPC	1.96	1.00	0.0196	Diazinon	1.23	0.93	0.0115
Chloroxuron	3.92	2.63	0.1029	Dinitrobutylphenol	2.47	0.53	0.0131
2,4-D	9.80	0.53	0.0524	Heptachlor	1.23	0.19	0.0023
DDT technical	1.96	1.50	0.0294	Linuron	2.47	0.37	0.0091
Dinitrobutylphenol	1.96	11.25	0.2206	NPA	2.47	1.07	0.0264
Diuron	1.96	2.00	0.0392	Methyl parathion	1.23	0.50	0.0062
EPTC	1.96	2.00	0.0392	Propazine	1.23	2.00	0.0247
Herbisan	1.96	10.00	0.1961	Ramrod	1.23	1.10	0.0136
Malathion	3.92	0.50	0.0198	Trifluralin	7.41	0.91	0.0673
Methoxychlor	1.96	0.01	0.0002	Vernolate	2.47	1.38	0.0340
NEBRASKA—103 SITES				Amiben	0.97	2.50	0.0243
				Atrazine	4.85	0.82	0.0400
				Captan	17.48	0.04	0.0069
				Ceresan red	0.97	0.01	0.0001

TABLE 10.—Summary of pesticides used in FY 1969 on cropland by State—Continued

COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB/ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB 'ACRE)	COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB ACRE)
NEBRASKA—103 SITES—Continued				NEW YORK—38 SITES—Continued			
Ceresan L	0.97	0.01	0.0001	Nabam	2.63	2.40	0.0632
Chevron RE-5353	1.94	1.25	0.0243	Nitrate	7.89	26.17	2.0658
2,4-D	14.56	0.44	0.0644	Oxydemetonmethyl	2.63	0.15	0.0039
Diazinon	4.85	0.98	0.0476	Ethyl parathion	5.26	0.45	0.0239
Dieldrin	3.88	0.01	0.0004	Phosphamidon	2.63	0.15	0.0039
Disulfoton	0.97	0.22	0.0021	Sodium arsenite	2.63	0.90	0.0237
EPTC	0.97	3.00	0.0291				
Malathion	17.48	0.01	0.0017	NORTH CAROLINA—29 SITES			
Methoxychlor	1.94	0.01	0.0002	Aldrin	6.90	1.75	0.1207
Methylmercury dicyandiamide	4.85	0.01	0.0005	Atrazine	6.90	2.75	0.1897
Nabam	0.97	0.01	0.0001	Carbaryl	20.69	1.43	0.2966
Norea	0.97	0.60	0.0058	Ceresan red	3.45	0.10	0.0034
Ethyl parathion	3.88	0.50	0.0194	Chromophon	3.45	0.15	0.0052
Phorate	0.97	0.90	0.0087	Copper carbonate	3.45	0.60	0.0207
Ramrod	0.97	0.83	0.0081	Copper-8-quinolinolate	3.45	0.01	0.0003
Thiram	4.85	0.03	0.0014	2,4-D	20.69	1.00	0.2079
				2,4-DB	3.45	0.07	0.0024
NEW JERSEY—5 SITES				DDT technical	13.79	0.70	0.0962
2,4-D	40.00	0.31	0.1240	Diazinon	13.79	1.12	0.1545
Monuron	20.00	1.60	0.3200	Dicamba	3.45	1.20	0.0414
Ethyl parathion	20.00	0.54	0.1080	Dichloropropene	3.45	20.00	0.6897
Sulfur	20.00	9.00	1.8000	Dieldrin	6.90	1.25	0.0866
				Dinitrobutylphenol	3.45	1.50	0.0517
NEW MEXICO—10 SITES				Diphenamid	6.90	1.07	0.0741
Azodrin	10.00	1.50	0.1500	EPTC	3.45	4.00	0.1379
Carbaryl	10.00	2.50	0.2500	Ethylene dibromide	3.45	6.00	0.2069
DDT technical	10.00	1.00	0.1000	Lindane	3.45	0.01	0.0003
Diuron	20.00	1.12	0.2250	Maleic hydrazide	10.34	0.47	0.0486
Ethyl parathion	10.00	2.50	0.2500	Ethyl parathion	6.90	0.50	0.0345
Toxaphene	10.00	1.00	0.1000	Methyl parathion	3.45	0.83	0.0286
				Phorate	6.90	1.13	0.0783
NEW YORK—38 SITES				Sulfur	3.45	11.10	0.3828
Atrazine	23.68	1.36	0.3211	TDE technical	10.34	0.26	0.0272
Azinphosmethyl	5.26	1.15	0.0605	Thiram	6.90	0.01	0.0007
Captan	13.16	0.87	0.1150	Toxaphene	6.90	8.65	0.5966
Carbaryl	5.26	1.55	0.0816	Trifluralin	3.45	0.57	0.0197
Copper sulfate	2.63	3.00	0.0789	Vernolate	6.90	1.85	0.1276
2,4-D	7.89	0.21	0.0166				
Dalapon	2.63	2.50	0.0658	NORTH DAKOTA—159 SITES			
DDT technical	5.26	1.35	0.0711	Barban	1.26	0.17	0.0021
Demeton	2.63	0.04	0.0011	Captan	0.63	0.01	0.0001
Diazinon	2.63	1.00	0.0263	Ceresan M	0.63	0.01	0.0001
Dichlone	2.63	2.20	0.0579	Ceresan red	2.52	0.01	0.0003
Dinitrobutylphenol	5.26	15.22	0.8013	Ceresan L	5.03	0.01	0.0005
Diuron	5.26	2.40	0.1263	2,4-D	42.14	0.40	0.1673
Endosulfan (1)	5.26	0.95	0.0500	Dicamba	1.89	0.08	0.0016
Lead arsenate	2.63	3.80	0.1000	Disulfoton	0.63	3.00	0.0189
Malathion	5.26	0.01	0.0005	Endrin	0.63	0.25	0.0016
MCPA	5.26	0.33	0.0176	Heptachlor	1.26	0.04	0.0005
Methoxychlor	2.63	0.01	0.0003	Lindane	1.89	0.02	0.0004
Methylmercury dicyandiamide	10.53	0.01	0.0011	Malathion	0.63	0.01	0.0001
				Maneb	0.63	1.50	0.0094
				MCPA	5.66	0.30	0.0171
				Methylmercury dicyandiamide	41.51	0.01	0.0042

TABLE 10.—Summary of pesticides used in FY 1969 on cropland by State—Continued

COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLI- CATION RATE (LB/ ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB/ ACRE)	COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLI- CATION RATE (LB/ ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB/ ACRE)
NORTH DAKOTA—159 SITES—Continued				PENNSYLVANIA—31 SITES			
Phenylmercury urea	0.63	0.01	0.0001	Atrazine	19.35	1.57	0.3032
PMA	1.26	0.01	0.0001	Azinphosmethyl	3.23	0.50	0.0161
Polyram	0.63	10.40	0.0654	Captan	3.23	0.01	0.0003
OHIO—66 SITES				Carbaryl	3.23	0.32	0.0103
Aldrin	6.06	3.00	0.1818	Chlordane	3.23	1.20	0.0387
Amiben	3.03	1.75	0.0530	Copper sulfate	3.23	1.70	0.0548
Atrazine	12.12	1.20	0.1455	2,4-D	16.13	0.92	0.1484
Captan	12.12	0.02	0.0021	DDT technical	9.68	0.83	0.0806
Ceresan M	1.52	0.01	0.0002	Dicofol	3.23	0.42	0.0135
Copper sulfate	1.52	1.60	0.0242	Dinitrobutylphenol	3.23	0.82	0.0265
2,4-D	19.70	0.44	0.0859	Dinitroresol	3.23	3.00	0.0968
Dalapon	1.52	1.50	0.0227	Dinocap	3.23	0.44	0.0142
Diazinon	1.52	0.50	0.0076	Diuron	3.23	0.32	0.0103
Dichlone	1.52	1.80	0.0273	Lindane	3.23	0.02	0.0006
Dieldrin	1.52	0.01	0.0002	Linuron	3.23	1.00	0.0323
Dinocap	1.52	0.01	0.0002	Maneb	3.23	7.00	0.2258
Dithane M-45	1.52	0.30	0.0045	Methyl demeton	3.23	1.50	0.0484
Linuron	1.52	0.75	0.0114	Nitrate	3.23	100.00	3.2258
Malathion	10.61	0.01	0.0011	Ethyl parathion	6.45	0.45	0.0290
Maneb	3.03	0.75	0.0227	Phorate	3.23	12.50	0.4032
Methylmercury dicyandiamide	1.52	0.05	0.0008	Simazine	3.23	0.40	0.0129
NPA	1.52	2.27	0.0344	Sodium arsenite	3.23	2.50	0.0806
PCP	1.52	1.50	0.0227	Trifluralin	3.23	0.75	0.0242
Picloram	1.52	0.25	0.0038	RHODE ISLAND—1 SITE			
Randox T	1.52	1.40	0.0212	Carbaryl	100.00	0.80	0.8000
Sulfur	1.52	25.00	0.3788	DDT technical	100.00	2.00	2.0000
TDE technical	1.52	0.80	0.0121	Disulfoton	100.00	2.00	2.0000
Trifluralin	1.52	1.00	0.0152	Dithane M-45	100.00	6.40	6.4000
Ziram	1.52	0.80	0.0121	EPTC	100.00	5.00	5.0000
OKLAHOMA—65 SITES				Oxydemetonmethyl	100.00	0.80	0.8000
Cacodylic acid	1.54	0.01	0.0002	SOUTH CAROLINA—17 SITES			
Captan	4.62	0.01	0.0005	Azodrin	5.88	0.40	0.0235
Carbaryl	1.54	0.30	0.0046	Carbaryl	17.65	7.19	1.2682
Ceresan M	20.00	0.01	0.0020	2,4-D	11.76	0.40	0.0471
Ceresan red	12.31	0.01	0.0012	DDT technical	29.41	2.46	0.7229
Chloroneb	1.54	0.01	0.0002	DEF	5.88	0.20	0.0118
2,4-D	6.15	0.86	0.0531	Demeton	5.88	1.60	0.0941
2,4-DB	4.62	0.50	0.0231	Diuron	5.88	0.72	0.0424
Dieldrin	4.62	0.01	0.0005	MSMA	5.88	0.45	0.0265
Dimethoate	1.54	0.50	0.0077	Nabam	5.88	1.20	0.0706
Dinitrobutylphenol	1.54	2.00	0.0308	Ethyl parathion	11.76	0.51	0.0600
Disulfoton	7.69	0.58	0.0445	Methyl parathion	11.76	5.10	0.6000
Falone	1.54	2.00	0.0308	Phorate	5.88	0.20	0.0118
Methylmercury dicyandiamide	1.54	0.01	0.0002	TDE technical	5.88	2.25	0.1324
Nitrate	10.77	16.64	1.7923	Toxaphene	17.65	6.17	1.0894
Ethyl parathion	3.08	0.75	0.0231	Trifluralin	35.29	0.21	0.0753
Methyl parathion	12.31	0.65	0.0800	SOUTH DAKOTA—106 SITES			
PCNB	1.54	0.01	0.0002	Atrazine	1.89	1.40	0.0264
Phosphamidon	1.54	0.12	0.0018	Captan	10.38	0.01	0.0010
Thiram	1.54	0.01	0.0002	Carbaryl	0.94	1.05	0.0099
Trifluralin	4.62	1.10	0.0508				

TABLE 10.—Summary of pesticides used in FY 1969 on cropland by State—Continued

COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB/ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB/ACRE)
SOUTH DAKOTA—106 SITES—Continued			
Ceresan M	0.94	0.01	0.0001
2,4-D	26.42	0.47	0.1230
Dalapon	0.94	0.74	0.0070
Dieldrin	1.89	0.01	0.0002
Heptachlor	3.77	0.02	0.0007
Lindane	0.94	0.01	0.0001
Malathion	6.60	0.01	0.0007
MCPA	4.72	0.20	0.0095
Methoxychlor	3.77	0.01	0.0004
Methylmercury dicyandiamide	10.38	0.01	0.0010
Phorate	0.94	0.60	0.0057
Ramrod	0.94	1.00	0.0094
Thiram	0.94	0.01	0.0001
TENNESSEE—28 SITES			
Atrazine	21.43	1.98	0.4250
Bidrin	3.57	0.54	0.0193
Captan	10.71	0.01	0.0011
Ceresan M	7.14	0.01	0.0007
Ceresan red	3.57	0.01	0.0004
Cotoran	7.14	0.78	0.0557
2,4-DB	7.14	0.43	0.0307
Disulfoton	3.57	0.01	0.0004
Diuron	7.14	0.06	0.0046
Linuron	7.14	0.75	0.0536
Malathion	3.57	0.01	0.0004
Methylmercury dicyandiamide	3.57	0.01	0.0004
MSMA	3.57	0.46	0.0164
Nitrate	7.14	250.00	17.8571
Nitralin	3.57	0.15	0.0054
PCNB	3.57	0.01	0.0004
Trifluralin	14.29	0.38	0.0543
UTAH—12 SITES			
Dichloropropene	8.33	180.00	15.0000
Heptachlor	8.33	0.34	0.0283
VIRGINIA—20 SITES			
Atrazine	5.00	4.00	0.2000
Azinphosmethyl	5.00	2.00	0.1000
Carbaryl	5.00	2.75	0.1375
Copper oxide	10.00	2.60	0.2600
2,4-D	10.00	1.12	0.1125
2,4-DB	5.00	0.20	0.0100
DDT technical	5.00	2.00	0.1000
Diazinon	5.00	0.50	0.0250
Dinitrobutylphenol	5.00	1.50	0.0750
Diphenamid	5.00	4.00	0.2000
Disulfoton	10.00	6.80	0.6800
Ethylene dibromide	5.00	23.24	1.1620
Dichloropropane	5.00	54.43	2.7215

COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE (LB/ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB/ACRE)
VIRGINIA—20 SITES—Continued			
Malathion	5.00	0.95	0.0475
Methoxychlor	5.00	1.00	0.0500
Ethyl parathion	5.00	6.00	0.3000
Phorate	5.00	3.00	0.1500
Sulfur	5.00	57.00	2.800
Vernolate	5.00	2.40	0.1200
WASHINGTON—2 SITES			
Ceresan L	50.00	0.01	0.0050
2,4-D	50.00	1.00	0.5000
WEST VIRGINIA—5 SITES			
Azinphosmethyl	20.00	0.50	0.1000
Ethyl parathion	20.00	1.50	0.3000
WISCONSIN—68 SITES			
Atrazine	29.41	2.61	0.7684
Ceresan red	1.47	0.01	0.0001
2,4-D	2.94	0.75	0.0221
Ramrod	1.47	2.00	0.0294
Trifluralin	1.47	2.00	0.0294

NOTE: Of the 43 States with cropland soil analyzed, use records for 4 showed no pesticides used on the sampling sites: Nevada (2 sites); New Hampshire (2 sites); Vermont (5 sites); and Wyoming (17 sites).

TABLE 11.—Summary of pesticides used in FY 1969 on noncropland, by State

COMPOUND	PERCENT OF SITES TREATED	AVERAGE APPLICATION RATE PER SITE TREATED (LB/ACRE)	AVERAGE AMOUNT APPLIED PER SITE (LB/ACRE)
GEORGIA—15 SITES			
Mirex	6.67	0.01	0.0007
IDAHO—26 SITES			
Malathion	3.85	0.61	0.0235
NEBRASKA—19 SITES			
2,4-D	5.26	2.00	0.1053

NOTE: Of the 11 States with noncropland soil analyzed, 8 reported no pesticides used on the sampling sites: Arizona (43 sites); Iowa (7 sites); Maine (11 sites); Maryland (3 sites); Virginia (14 sites); Washington (11 sites); West Virginia (9 sites); and Wyoming (37 sites).

TABLE 12.—Comparison of residues detected with use records for 12 States with highest arsenic residues, FY 1969

STATE	AVERAGE AMOUNT APPLIED (LB/ACRE)	PERCENT OF SITES TREATED	MEAN RESIDUE LEVEL (PPM)	PERCENT POSITIVE SITES ¹
Arkansas	³ 0.13	4.4	9.0	100.0
Kentucky	No Arsenic Compounds Used		8.4	100.0
New England ²	⁴ 0.88	10.0	10.2	100.0
New York	⁵ 0.12	5.3	9.4	94.6
North Dakota	No Arsenic Compounds Used		8.5	100.0
Ohio	No Arsenic Compounds Used		11.2	100.0
Pennsylvania	⁴ 0.08	3.2	10.8	100.0

¹ Percent based on number of sites with residues greater than or equal to the sensitivity limits.

² Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, and Vermont.

³ Calculated for DSMA.

⁴ Calculated for sodium arsenite.

⁵ Calculated for sodium arsenite and lead arsenate.

TABLE 13.—Comparison of residues detected with use records for 5 States with highest DDTR residues, FY 1969

STATE	AVERAGE AMOUNT APPLIED (LB/ACRE)	PERCENT OF SITES TREATED	MEAN RESIDUE LEVEL (PPM)	PERCENT POSITIVE SITES ¹
Alabama	4.20	39.1	1.13	90.9
California	0.38	13.6	1.47	84.6
Michigan	0.03	2.0	2.09	23.5
Mississippi	1.07	31.0	2.06	89.7
South Carolina	0.72	29.4	1.17	88.2

¹ Percent based on number of sites with residues greater than or equal to the sensitivity limits.

TABLE 14.—Comparison of residues detected with use records for 7 States with highest dieldrin residues, FY 1969

STATE	AVERAGE AMOUNT APPLIED (LB/ACRE)	PERCENT OF SITES TREATED	MEAN RESIDUE LEVEL (PPM)	PERCENT POSITIVE SITES ¹
Florida	0.00	0.0	0.08	38.9
Illinois	0.29 aldrin	19.2		
	0.01 dieldrin	2.1	0.11	61.3
Iowa	0.06 aldrin	8.6		
	<0.01 dieldrin	0.7	0.06	53.6
Kentucky	0.19 aldrin	9.7	0.06	54.8
North Carolina	0.12 aldrin	6.9		
	0.09 dieldrin	6.9	0.08	32.3
Virginia/West Virginia	0.00	0.0	0.07	25.9

¹ Percent based on number of sites with residues greater than or equal to the sensitivity limits.

TABLE 15.—Fiftieth percentile value for pesticide residues in cropland soil including the 95% confidence interval by State

PESTICIDE	UPPER LIMIT (PPM)	FIFTIETH PERCENTILE RESIDUE LEVEL (PPM)	LOWER LIMIT (PPM)	PESTICIDE	UPPER LIMIT (PPM)	FIFTIETH PERCENTILE RESIDUE LEVEL (PPM)	LOWER LIMIT (PPM)
ALABAMA				IDAHO			
Arsenic	4.42	4.09	3.76	Arsenic	2.85	2.53	2.21
<i>p,p'</i> -DDE	0.10	0.07	0.04	<i>p,p'</i> -DDT	0.00	0.00	0.00
<i>o,p'</i> -DDT	0.03	0.02	0.01	DDTR	0.01	0.00	0.00
<i>p,p'</i> -DDT	0.27	0.21	0.15	ILLINOIS			
DDTR	0.48	0.38	0.29	Aldrin	0.00	0.00	0.00
<i>p,p'</i> -TDE	0.02	0.01	0.01	Arsenic	6.28	6.20	6.13
ARKANSAS				Chlordane	0.01	0.01	0.00
Arsenic	7.42	7.26	7.10	DDTR	0.00	0.00	0.00
<i>p,p'</i> -DDE	0.02	0.02	0.02	Dieldrin	0.03	0.02	0.02
<i>o,p'</i> -DDT	0.01	0.01	0.00	Heptachlor	0.00	0.00	0.00
<i>p,p'</i> -DDT	0.04	0.03	0.03	Heptachlor epoxide	0.00	0.00	0.00
DDTR	0.10	0.09	0.08	INDIANA			
Dieldrin	0.00	0.00	0.00	Aldrin	0.00	0.00	0.00
<i>p,p'</i> -TDE	0.01	0.01	0.01	Arsenic	7.24	7.03	6.82
Toxaphene	0.15	0.04	0.00	Dieldrin	0.00	0.00	0.00
CALIFORNIA				IOWA			
Arsenic	4.02	3.92	3.83	Aldrin	0.00	0.00	0.00
<i>o,p'</i> -DDE	0.00	0.00	0.00	Arsenic	5.86	5.78	5.71
<i>p,p'</i> -DDE	0.05	0.04	0.04	Atrazine	0.01	0.00	0.00
<i>o,p'</i> -DDT	0.01	0.01	0.00	Chlordane	0.01	0.01	0.00
<i>p,p'</i> -DDT	0.04	0.03	0.03	<i>p,p'</i> -DDE	0.00	0.00	0.00
DDTR	0.14	0.13	0.12	<i>p,p'</i> -DDT	0.00	0.00	0.00
Dieldrin	0.00	0.00	0.00	DDTR	0.00	0.00	0.00
<i>o,p'</i> -TDE	0.00	0.00	0.00	Dieldrin	0.02	0.01	0.01
<i>p,p'</i> -TDE	0.02	0.01	0.01	Heptachlor	0.00	0.00	0.00
Toxaphene	0.02	0.01	0.00	Heptachlor epoxide	0.00	0.00	0.00
COLORADO				KENTUCKY			
Arsenic	4.26	4.20	4.15	Aldrin	0.00	0.00	0.00
FLORIDA				Arsenic	8.45	7.89	7.30
Arsenic	0.64	0.58	0.53	Dieldrin	0.01	0.01	0.00
Chlordane	0.05	0.03	0.02	LOUISIANA			
<i>p,p'</i> -DDE	0.03	0.02	0.01	Arsenic	1.80	1.65	1.51
<i>o,p'</i> -DDT	0.01	0.00	0.00	<i>p,p'</i> -DDE	0.01	0.00	0.00
<i>p,p'</i> -DDT	0.07	0.05	0.03	<i>o,p'</i> -DDT	0.01	0.00	0.00
DDTR	0.10	0.08	0.06	<i>p,p'</i> -DDT	0.02	0.01	0.00
<i>p,p'</i> -TDE	0.02	0.01	0.00	DDTR	0.02	0.01	0.01
GEORGIA				Dieldrin	0.01	0.01	0.00
Arsenic	1.96	1.88	1.80	MICHIGAN			
<i>p,p'</i> -DDE	0.06	0.05	0.04	Arsenic	4.92	3.83	2.93
<i>o,p'</i> -DDT	0.01	0.01	0.01	<i>p,p'</i> -DDE	0.00	0.00	0.00
<i>p,p'</i> -DDT	0.17	0.01	0.09	DDTR	0.00	0.00	0.00
DDTR	0.30	0.23	0.17	Dieldrin	0.00	0.00	0.00
<i>p,p'</i> -TDE	0.01	0.01	0.01	MICHIGAN			
Toxaphene	0.36	0.28	0.18	Arsenic	4.92	3.83	2.93

TABLE 15.—Fiftieth percentile value for pesticide residues in eropland soil including the 95% confidence interval by State—Continued

PESTICIDE	UPPER LIMIT (PPM)	FIFTIETH PERCENTILE RESIDUE LEVEL (PPM)	LOWER LIMIT (PPM)	PESTICIDE	UPPER LIMIT (PPM)	FIFTIETH PERCENTILE RESIDUE LEVEL (PPM)	LOWER LIMIT (PPM)
MID-ATLANTIC STATES GROUP ¹				NORTH CAROLINA—Continued			
Arsenic	5.87	5.34	4.83	<i>p,p'</i> -TDE	0.03	0.02	0.01
MISSISSIPPI				Toxaphene	0.16	0.07	0.01
Arsenic	4.86	4.68	4.51	NORTH DAKOTA			
<i>p,p'</i> -DDE	0.11	0.09	0.08	Arsenic	6.82	6.79	6.75
<i>o,p'</i> -DDT	0.06	0.06	0.05	<i>p,p'</i> -DDT	0.00	0.00	0.00
<i>p,p'</i> -DDT	0.36	0.29	0.24	DDTR	0.00	0.00	0.00
DDTR	0.67	0.55	0.45	OHIO			
<i>p,p'</i> -TDE	0.03	0.02	0.01	Aldrin	0.00	0.00	0.00
Toxaphene	0.24	0.16	0.09	Arsenic	7.85	7.58	7.32
MISSOURI				DDTR	0.00	0.00	0.00
Aldrin	0.00	0.00	0.00	Dieldrin	0.00	0.00	0.00
Arsenic	5.43	5.05	4.69	OKLAHOMA			
Dieldrin	0.00	0.00	0.00	Arsenic	2.19	2.11	2.02
NEBRASKA				PENNSYLVANIA			
Arsenic	4.73	4.56	4.40	Arsenic	7.22	6.65	6.15
Chlordane	0.01	0.00	0.00	<i>p,p'</i> -DDE	0.00	0.00	0.00
<i>p,p'</i> -DDE	0.00	0.00	0.00	<i>p,p'</i> -DDT	0.00	0.00	0.00
DDTR	0.00	0.00	0.00	DDTR	0.00	0.00	0.00
Dieldrin	0.00	0.00	0.00	Dieldrin	0.01	0.00	0.00
NEW ENGLAND STATES GROUP ²				<i>p,p'</i> -TDE	0.00	0.00	0.00
Arsenic	6.39	5.71	5.09	SOUTH CAROLINA			
<i>p,p'</i> -DDE	0.02	0.01	0.00	Arsenic	1.98	1.82	1.68
<i>p,p'</i> -DDT	0.05	0.02	0.00	<i>p,p'</i> -DDE	0.08	0.06	0.03
DDTR	0.04	0.02	0.01	<i>o,p'</i> -DDT	0.04	0.03	0.02
<i>p,p'</i> -TDE	0.01	0.01	0.00	<i>p,p'</i> -DDT	0.18	0.13	0.08
NEW YORK				DDTR	0.31	0.15	0.06
Arsenic	6.34	6.12	5.90	<i>p,p'</i> -TDE	0.06	0.04	0.02
<i>p,p'</i> -DDE	0.00	0.00	0.00	SOUTH DAKOTA			
<i>o,p'</i> -DDT	0.00	0.00	0.00	Arsenic	3.86	3.76	3.68
<i>p,p'</i> -DDT	0.01	0.00	0.00	Dieldrin	0.00	0.00	0.00
DDTR	0.00	0.00	0.00	TENNESSEE			
Dieldrin	0.00	0.00	0.00	Arsenic	7.18	7.00	6.81
<i>p,p'</i> -TDE	0.00	0.00	0.00	<i>p,p'</i> -DDE	0.00	0.00	0.00
NORTH CAROLINA				<i>p,p'</i> -DDT	0.01	0.00	0.00
Arsenic	3.42	3.07	2.77	DDTR	0.01	0.01	0.00
<i>p,p'</i> -DDE	0.03	0.03	0.02	NEW ENGLAND STATES GROUP ²			
<i>o,p'</i> -DDT	0.02	0.01	0.01	Arsenic	6.39	5.71	5.09
<i>p,p'</i> -DDT	0.05	0.03	0.02	<i>p,p'</i> -DDE	0.02	0.01	0.00
DDTR	0.18	0.13	0.08	<i>p,p'</i> -DDT	0.05	0.02	0.00
Dieldrin	0.01	0.00	0.00	DDTR	0.04	0.02	0.01
<i>o,p'</i> -TDE	0.03	0.02	0.01	<i>p,p'</i> -TDE	0.01	0.01	0.00

TABLE 15.—Fiftieth percentile value for pesticide residues in cropland soil including the 95% confidence interval by State—Continued

PESTICIDE	UPPER LIMIT (PPM)	FIFTIETH PERCENTILE RESIDUE LEVEL (PPM)	LOWER LIMIT (PPM)	PESTICIDE	UPPER LIMIT (PPM)	FIFTIETH PERCENTILE RESIDUE LEVEL (PPM)	LOWER LIMIT (PPM)
VIRGINIA AND WEST VIRGINIA				WESTERN STATES GROUP—Continued			
Arsenic	2.90	2.72	2.56	<i>p,p'</i> -DDT	0.00	0.00	0.00
<i>p,p'</i> -DDT	0.01	0.00	0.00	DDTR	0.01	0.01	0.00
DDTR	0.02	0.01	0.01	WISCONSIN			
Heptachlor epoxide	0.01	0.00	0.00	Arsenic	3.33	3.14	2.97
WASHINGTON				DDTR	0.00	0.00	0.00
Arsenic	2.30	2.22	2.14	Dieldrin	0.00	0.00	0.00
<i>p,p'</i> -DDT	0.00	0.00	0.00	WYOMING			
DDTR	0.00	0.00	0.00	Arsenic	0.92	0.85	0.78
WESTERN STATES GROUP ³							
Arsenic	3.57	3.40	3.23				
<i>p,p'</i> -DDE	0.01	0.00	0.00				

¹ Includes Delaware, Maryland, and New Jersey.
² Includes Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, and Vermont.
³ Includes Arizona, Nevada, New Mexico, and Utah.

TABLE 16.—Mean pesticide residues in ppm in soil for various cropping regions, FY 1969

COMPOUND	CORN	COTTON	COTTON AND GENERAL FARMING	GENERAL FARMING	HAY AND GENERAL FARMING	IRRIGATED LAND	SMALL GRAINS	VEGETABLE	VEGETABLE AND FRUIT
Aldrin	0.05	<0.01	<0.01	0.02	<0.01	<0.01	<0.01	<0.01	0.01
Arsenic	7.44	6.72	4.88	5.35	6.42	4.77	5.70	8.75	3.27
Atrazine	0.02	—	—	—	—	—	<0.01	—	—
Carbophenothion	—	—	—	—	—	—	—	—	—
Chlordane	0.09	<0.01	0.01	0.01	0.03	0.03	<0.01	<0.01	0.14
2,4-D	—	—	—	<0.01	—	—	<0.01	—	—
DCPA	—	—	—	—	—	<0.01	—	—	—
<i>o,p'</i> -DDE	<0.01	0.01	<0.01	<0.01	<0.01	0.01	—	<0.01	0.01
<i>p,p'</i> -DDE	0.01	0.16	0.13	0.07	0.05	0.18	<0.01	0.18	0.37
<i>o,p'</i> -DDT	0.01	0.09	0.04	0.05	0.03	0.05	<0.01	0.07	0.06
<i>p,p'</i> -DDT	0.06	0.54	0.22	0.25	0.20	0.19	<0.01	0.50	0.64
DDTR	0.14	0.87	0.44	0.43	0.30	0.48	<0.01	0.81	1.92
DEF	—	—	—	<0.01	—	—	—	—	—
Diazinon	—	—	—	—	—	0.01	—	—	—
Dicofol	<0.01	—	—	—	<0.01	0.01	—	—	—
Dieldrin	0.05	0.01	<0.01	0.03	0.02	0.02	<0.01	0.05	0.04
Endosulfan (I)	<0.01	—	—	—	<0.01	<0.01	—	—	—
Endosulfan (II)	<0.01	—	—	—	<0.01	0.01	—	<0.01	—
Endosulfan sulfate	<0.01	—	—	—	<0.01	0.01	—	<0.01	—
Endrin	<0.01	0.01	<0.01	<0.01	—	0.01	<0.01	0.01	0.01
Endrin aldehyde	—	—	—	—	—	—	—	—	<0.01
Endrin ketone	—	<0.01	<0.01	—	—	<0.01	—	<0.01	<0.01
Ethion	—	—	—	—	—	<0.01	—	—	—
Ethyl parathion	—	—	—	<0.01	—	<0.01	—	—	—
Heptachlor	0.01	—	<0.01	<0.01	<0.01	<0.01	—	—	<0.01
Heptachlor epoxide	0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Isodrin	<0.01	—	—	<0.01	—	—	—	—	—
Lindane	<0.01	—	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	—
Malathion	—	—	—	—	—	—	—	—	—
Methoxychlor	—	—	—	—	—	—	—	<0.01	—
PCNB	—	—	—	<0.01	—	—	—	—	—
<i>o,p'</i> -TDE	<0.01	<0.01	<0.01	0.01	<0.01	0.01	—	0.01	0.15
<i>p,p'</i> -TDE	0.05	0.07	0.04	0.04	0.01	0.04	<0.01	0.05	0.70
Toxaphene	<0.01	0.42	0.20	0.16	—	0.14	—	0.01	0.08
Trifluralin	<0.01	0.01	<0.01	<0.01	—	0.01	<0.01	<0.01	<0.01

NOTE: Blank = not analyzed; — = not detected.

TABLE 17.—Percent of sites with detectable pesticide residues in ppm in soil for various cropping regions, FY 1969

COMPOUND	CORN	COTTON	COTTON AND GENERAL FARMING	GENERAL FARMING	HAY AND GENERAL FARMING	IRRIGATED LAND	SMALL GRAINS	VEGETABLE	VEGETABLE AND FRUIT
Aldrin	23.5	6.4	0.9	6.6	2.1	6.5	0.6	1.1	3.0
Arsenic	100.0	100.0	98.4	99.4	99.3	99.1	99.4	98.9	93.9
Atrazine	14.5	—	—	—	—	—	8.3	—	—
Carbophenothion	—	—	—	—	—	—	—	—	—
Chlordane	14.5	1.8	2.6	7.2	5.5	11.1	0.9	4.3	21.2
2,4-D	—	—	—	14.3	—	—	1.7	—	—
DCPA	—	—	—	—	—	0.9	—	—	—
<i>o,p'</i> -DDE	0.5	15.6	5.2	10.8	2.8	19.4	—	5.3	15.1
<i>p,p'</i> -DDE	9.5	69.7	44.8	46.4	21.4	58.3	5.8	38.3	60.6
<i>o,p'</i> -DDT	3.0	51.4	25.0	27.1	10.3	33.3	3.0	23.4	36.4
<i>p,p'</i> -DDT	7.7	66.1	43.1	42.2	16.5	53.7	5.8	31.9	57.6
DDTR	10.9	72.5	47.4	49.4	22.8	60.2	6.1	39.4	63.6
DEF	—	—	—	0.6	—	—	—	—	—
Diazinon	—	—	—	—	—	12.5	—	—	—
Dicofol	0.6	—	—	—	0.7	5.6	—	—	—
Dieldrin	41.8	24.8	14.7	25.3	15.2	39.8	7.0	23.4	21.2
Endosulfan (I)	0.3	—	—	—	0.7	1.8	—	—	—
Endosulfan (II)	0.2	—	—	—	0.7	5.6	—	1.1	—
Endosulfan sulfate	0.3	—	—	—	1.4	5.6	—	1.1	—
Endrin	0.3	7.3	2.6	3.6	—	11.1	0.9	3.2	6.1
Endrin aldehyde	—	—	—	—	—	—	—	—	3.0
Endrin ketone	—	2.8	0.9	—	—	2.8	—	1.1	3.0
Ethion	—	—	—	—	—	6.3	—	—	—
Ethyl parathion	—	—	—	10.0	—	12.5	—	—	—
Heptachlor	8.6	—	1.7	1.8	1.4	0.9	—	—	3.0
Heptachlor epoxide	13.3	0.9	3.4	7.8	4.1	13.0	0.9	4.3	12.1
Isodrin	1.2	—	—	1.8	—	—	—	—	—
Lindane	0.3	—	2.6	1.2	0.7	1.8	0.6	3.2	—
Malathion	—	—	—	—	—	—	—	—	—
Methoxychlor	—	—	—	—	—	—	—	1.1	—
PCNB	—	—	—	0.6	—	—	—	—	—
<i>o,p'</i> -TDE	0.3	0.9	2.6	10.8	2.1	13.0	—	5.3	6.1
<i>p,p'</i> -TDE	3.3	47.7	25.9	35.5	11.7	38.0	1.8	27.7	45.4
Toxaphene	0.2	22.9	12.1	10.2	—	12.0	—	1.1	6.1
Trifluralin	2.0	12.8	7.8	6.0	—	9.3	0.3	2.1	3.0

NOTE: Blank = not analyzed; — = not detected.

RESIDUES IN FOOD AND FEED

A Study of the Sources of Insecticide Residues in Milk on Dairy Farms in Illinois—1971¹

Steve Moore III, W. N. Bruce, D. E. Kuhlman, and
Roscoe Randell

ABSTRACT

In 1971, a study was conducted to determine the sources of chlorinated hydrocarbon insecticide contamination to dairy cows in order to help dairy farms avoid, or recover from, a residue problem in their milk supply. Twelve dairy farms were selected from 40 herds surveyed initially in February 1971; the 12 farms had dieldrin milk residues ranging from low to medium to high. Samples of milk were collected from the 12 farms in February, March, and September; in March, samples of feed, well water, and soil were also collected. Cropping history, insecticide usage history, and cattle management practices were obtained for each farm for the preceding 10-year period, 1961-70. The following conclusions may be drawn from this study: (1) the chances of dieldrin residues in milk exceeding the Food and Drug Administration's current administrative guideline of 0.3 ppm (fat basis) are greatest on dairy farms having a history of aldrin soil treatment within the last 6 or 7 years; (2) hay and oat straw supply significant amounts of dieldrin to dairy cattle; in addition, roasted soybeans could be an important source of dieldrin contamination in milk; (3) corn silage, commercial feed concentrate, and well water are usually not important sources of dieldrin contamination to dairy cattle.

Introduction

This report is not based on controlled detailed research, but rather constitutes a gross field investigation of a problem situation having many variables. The purpose of this study, initiated in February 1971, was to determine the sources of chlorinated hydrocarbon insecticide contamination to dairy cows to help dairy farms avoid, or recover from, a residue problem in their milk supply.

During 1969-70 in Illinois, a total of 27 dairy farms were found to be producing milk with illegal amounts of chlorinated hydrocarbon insecticide residues and, in January 1971, the Illinois Department of Public Health found dieldrin residues approaching an illegal level in a sample of cheese taken from a manufacturer in northern Illinois.

As early as 1951, the Illinois Cooperative Extension Service and Illinois Natural History Survey had advised against the use of DDT on dairy farms. In 1965, they advised against the use also of aldrin, chlordane, dieldrin, endrin, heptachlor, and lindane on dairy farms and, in 1969, they recommended against the use of these compounds on all farms. The vast majority (at least 80%) of Illinois dairymen followed these recommendations and had not used these insecticides in recent years, although alternative controls and insecticides were often more expensive, less effective in controlling the insects, and more hazardous to handle.

Continued use of certain of these insecticides by a few dairymen caused the residue problem that prompted the investigation reported here, but this represented only a fraction of 1% of the total dairy farms in Illinois. Of the 27 dairy farms found to be producing milk with illegal amounts of chlorinated hydrocarbon insecticide residues, on 4 the insecticide had been accidentally fed to the cattle; on the remaining 23 the insecticide had been used intentionally but in accordance with label directions.

In September 1971, State regulations were prescribed prohibiting dairymen from applying or storing on their farms, for agricultural purposes, the chlorinated hydrocarbon insecticides mentioned above.

¹ From the Cooperative Extension Service, College of Agriculture, University of Illinois and the Illinois Natural History Survey, 280 Natural Resources Building, Urbana, Ill. 61801.

Sampling Procedures

Twelve dairy farms with dieldrin milk residues ranging from low to medium to high were selected from a total of 40 herds surveyed on February 2, 1971. The herds surveyed were Grade B dairy herds located in an intensive grain-producing area in the northwest section of Illinois; they did not represent a cross section of the State's dairy herds. The dieldrin residue in a composite milk sample from all 40 farms was 0.22 ppm (butterfat basis). In 1967, Duggan (1) reported that the average level of dieldrin in dairy products produced in the United States was 0.042 ppm (butterfat basis); heptachlor epoxide, 0.036 ppm; and DDT, 0.042 ppm.

Cropping history, insecticide usage history, and cattle management practices were obtained for each of the 12 farms for the preceding 10 years, 1961-70. On March 16 and 17, 1971, samples of all the feed (including concentrates and roughages), a sample of well water, a composite milk sample, and a soil sample from every field on each of the farms were obtained for insecticide analysis. The samples were collected in 1-quart screw-cap glass jars, and a piece of aluminum foil was used to cover the jar mouth before the cap was fastened. Although sampling procedures varied depending on the material, approximately 1 quart by volume of each material was obtained. Hay and oat straw samples were taken from several representative bales (from each cutting still available) with a core sampler attached to an electric drill. Corn silage was collected by running the silage unloader for about 2-3 minutes and holding the sample jar in the stream of falling silage until full. Concentrate feeds were sampled by obtaining small portions from bins or sacks at several locations. The milk sample from each farm was taken from the bulk tank after the milk was well mixed. The water sample was obtained from the well water consumed by the cattle. The soil samples were taken with a standard soil sampling probe to a depth of 6 inches using the standard method of taking samples for soil nutrient analyses.

Analytical Methods

EQUIPMENT AND REAGENTS

1. Chromatographic columns, Kontes (K-420600) Chromaflex 22 x 330 mm
2. 1-liter separatory funnels
3. Three-ball Snyder columns with 24 40 ground glass fittings
4. 250-ml Erlenmeyer flasks with 24 40 ground glass fittings
5. Aerograph Model 204 GLC with a nickel detector
6. Nanograde ether and hexane
7. 95% ethyl alcohol redistilled in glass
8. Reagent grade anhydrous sodium sulphate heated overnight at 400° C to eliminate interference
9. 50% reagent grade potassium hydroxide in water
10. Florisil heated 450° C overnight, partially deactivated by adding 5% water by weight, mixed, and stored in a glass-stoppered flask for at least 24 hours before use

EXTRACTION

Butterfat for dieldrin analysis was extracted by placing 100 ml of milk and 100 ml of ethyl alcohol in a 1-liter separatory funnel; 100 ml of ether was added to this

mixture and shaken briefly, then 100 ml of hexane was added. The mixture was shaken about 200 times and allowed to stand until the phases separated. The lower aqueous phase was drained from the separatory funnel and discarded. The ether-hexane layer was then washed 3 times with 100 ml of water to remove ethyl alcohol from the nonaqueous phase. If emulsions were formed, 50% ethyl alcohol was added to break the emulsions. The ether-hexane layer was drained into a 250-ml Erlenmeyer flask, and about 10 g of sodium sulphate was added to remove water. The ether-hexane was transferred to a clean, dry 250-ml Erlenmeyer flask. A few grains of sodium sulphate were added to act as boiling chips, and the extract was placed on a steam bath, a Snyder column attached, and the solvent was removed by distillation. After cooling and solidification, 1 g of butterfat was weighed for analysis.

Ten-gram aliquots of thoroughly mixed soil samples were extracted by adding 5 ml of water, 10 ml of alcohol, and 100 ml of 50% ether in hexane. These were shaken and allowed to stand overnight before filtering into a separatory funnel. The soil residues were rinsed several times with hexane to quantitatively transfer the extract. The extracted soil was dried in an oven overnight to determine the percent moisture; 100 ml of water was added to the separatory funnel, and the mixture was shaken briefly to remove the ethyl alcohol. The ether-hexane layer was dried over 10 g of sodium sulphate in a 250-ml Erlenmeyer flask. The dried extract was transferred to a 250-ml Erlenmeyer flask. A few grains of sodium sulphate were added, a Snyder column was attached, and all but 10 ml of the solvent was removed by distillation. This extract was then ready for cleanup through a chromatographic column containing 30 g of Florisil.

Chopped hay, oat straw, corn silage, and soybean and other feeds, were extracted by the same method; 10 ml of ethyl alcohol and 100 ml of 50% ether in hexane were added to these materials in a 250-ml flask. These were gently warmed on the steam bath and allowed to stand overnight. The solvents were filtered into a separatory funnel, and the feed residues were rinsed several times with hexane to quantitatively transfer the extract; 100 ml of water was added to the organic solvents to remove ethyl alcohol. The separatory funnel was shaken briefly and the phases allowed to separate before draining off the lower aqueous phase. The extract was dried over sodium sulphate in a 250-ml flask and transferred to a 250-ml flask. A Snyder column was attached, and the solvents were removed by distillation through the column.

Five hundred milliliters of each water sample were extracted three times with 250 ml of 15% ethyl ether in hexane. Extracts were then dried over anhydrous sodium sulphate, combined in a 1000-ml Erlenmeyer flask, a Snyder column attached, and all but 2 ml of the solution removed by distillation.

The 1-g butterfat sample and the extracts from the hay, straw, corn silage, and other feed were saponified by adding 10 ml of 50% potassium hydroxide in 100 ml of ethyl alcohol. These mixtures were heated on the steam bath for 1 hour. Each saponified mixture was transferred to a 1-liter separatory funnel with 100 ml of water. The Erlenmeyer flask was then rinsed several times with hexane into the separatory funnel, and 100 ml of hexane was transferred to the separatory funnel. The mixture was shaken 200 times, and after the phases separated, the lower aqueous phase was drained and discarded. The hexane layer was washed several times with water to remove soaps and alcohol. The hexane was dried over sodium sulphate, transferred to a clean flask, and most of the solvent removed by distillation through a Snyder column. These extracts were then cleaned up through a chromatographic column containing 30 g of Florisil topped with one-half inch of sodium sulphate. The Florisil was prewet with 30 ml of hexane and a receiving glass was placed beneath the column. The hexane extract was then transferred to a column with several rinses of hexane. The aldrin and dieldrin residues were eluted from the column with 200 ml of 10% ether in hexane. After all of the solvent had passed through the column, a few grains of sodium sulphate were added, a Snyder column was attached, and most of the solvent removed by distillation on the steam bath. The butterfat eluate was made up to 50 ml with hexane for injection into the gas chromatograph. The extracts from the soil, hay, oat straw, and other feed were made up to 10 ml, and 2 to 10 μ l of the extract was injected into the gas chromatograph. Recovery of dieldrin and aldrin put through this system of extraction and cleanup was found to be between 90% and 98%.

GAS-LIQUID CHROMATOGRAPHY

The operating conditions of the Aerograph Model 204 gas chromatograph were as follows:

Column:	3 mm by 2m packed with Supelcoport 100/120 mesh containing 2% QF-1 and 1.25% OV-17.
Temperatures:	Injection port 225° C Column 190° C Detector 225° C
Carrier gas:	Nitrogen at 40 ml/min

The retention time of dieldrin was approximately 9 minutes with a 25% full-scale deflection for 50 picograms. The noise level at an attenuation of 1 was less than 0.3%.

Results and Discussion

Residues of dieldrin were significant in the milk and in many of the other materials sampled on the 12 farms studied (Tables 1 and 2); trace levels of aldrin were present in some soil samples; and residues of heptachlor, heptachlor epoxide, DDT, and DDE were found only at trace levels.

Aldrin, which converts to the more stable dieldrin in soil, water, plants, and animals, has been the major soil insecticide used for corn in Illinois. During the 10-year period 1961-70, approximately 4 million acres were treated each year up to 1967 (2); use of aldrin has been on the decline since then, with only about 2½ million acres being treated in 1971. Six of the 12 dairymen reported using aldrin as a corn soil treatment within the 10-year period; these use records are presented below:

FARM NO.	NUMBER OF YEARS ALDRIN WAS USED	LAST YEAR ALDRIN WAS APPLIED
1	8	1968
2	5	1967
3	2	1966
4	5	1966
5	5	1968
7	4	1964

Farm No. 6 had use records only from 1966 to date but did not report using aldrin during that period.

Five of the 12 dairymen reported using a heptachlor seed treatment (an insignificant amount), and none reported using DDT during the last 10 years.

A definite correlation was found to exist between the overall dieldrin soil residue on each farm and the level of dieldrin present in the milk (Tables 1 and 2). *To convert the amount of dieldrin soil residue in parts per million to pounds per acre, the dieldrin soil residue figure given in parts per million for the top six inches of soil should be multiplied by two. The weight of one acre of typical Illinois soil 6 inches deep is approximately 2 million pounds; also, 1 lb of insecticide incorporated into this soil leaves an initial residue of 0.5 ppm.*

The several months that cattle were grazing on pasture and cropland containing dieldrin residues did not result in an increase in dieldrin residues in September milk

TABLE 1.—Dieldrin residues in milk from 12 dairy farms in Illinois, 1971

DAIRY FARM NUMBER	RESIDUES IN PPM FOR THREE SAMPLE COLLECTIONS (BUTTERFAT BASIS)		
	FEBRUARY 2	MARCH 17	SEPTEMBER 24
1	2.5560	2.3360	2.3561
2 ¹	2.5294	2.740	2.773
3	2.4343	2.940	1.894
4 ¹	2.4037	2.300	2.561
5	.2786	2.800	2.757
6	2.3333	2.800	2.348
7	1.864	1.900	0.833
8	1.823	2.071	0.833
9	1.784	1.571	0.788
10	1.608	1.243	0.492
11	1.512	0.929	0.492
12	1.314	1.000	0.492

¹ Dry lot operation.

² Residues exceed the Food and Drug Administration's current administrative guideline of 0.3 ppm dieldrin in milk (fat basis).

TABLE 2.—Dieldrin residues in feed and soils on 12 dairy farms in Illinois, 1971

(Blank = no sample taken)

MATERIAL SAMPLED	RESIDUES IN PPM FOR 12 DAIRY FARMS SAMPLED IN MARCH 1971											
	FARM 1	FARM 2 ¹	FARM 3	FARM 4 ¹	FARM 5	FARM 6	FARM 7	FARM 8	FARM 9	FARM 10	FARM 11	FARM 12
Hay	.0373	.0401	.0213	.0313	.0206	.0393	.0256	.0203	.0129	.0159	.0155	.0160
Hay soil ²	.3531	.2271	.6551	.4223	.2115	.0864	.0287	.0151	<.0005	.0006	.0013	.0044
Oat straw	.0208	.0303	.0221	.0332	.0223	.0510	.0230	.0184	.0150	.0170	.0197	.0117
Oat soil ²	.2730	.1700	.3003	.3762	.0422	.3144	.0314	.0142	<.0005	<.0005	³	<.0005
Corn silage	.0098	.0047	.0061				.0028		.0032	.0038	.0014	.0021
Corn soil ²	.4507	.3590	.2583				.0095		.0008	.0006	.0027	.0013
Soybean feed ³	.0062		.0133			.0444	.0458	.0081				.0064
Concentrate feed ⁴	.0024	.0011	.0007	.0009	.0008	.0038	.0051	.0009	.0017	.0010	<.0005	.0009
Commercial protein supplement	.0030	.0037		<.0005	.0037			.0016		.0038	<.0005	.0052
Permanent pastures												
6-inch composite core			.0251		.0173		.0009	.0046		<.0005		<.0005
Surface sample ⁵			.0397		.0277		.0081	.0104		.0065		.0109
Soil average for all fields ⁶	.3859	.2183	.3558	.3153	.3515	.1344	.0219	.0566	.0047	.0009	.0020	.0010

NOTE: Residues of heptachlor, heptachlor epoxide, DDT, and DDE found only at trace levels in some samples. Residues of aldrin found at trace levels in some soil samples. Each of the 12 water samples, one from each farm, had a dieldrin level of <.0005 ppm.

¹ Dry lot operation.

² Soil samples taken with a standard soil sampling probe to a depth of 6 inches.

³ Purchased, no field history.

⁴ Soybean meal.

⁵ Roasted soybeans.

⁶ Made from home-grown corn and oats plus commercial protein supplement and minerals.

⁷ Commercial concentrate feed.

⁸ Sampled May 25, 1971.

⁹ Represents an average of residue levels in 6-inch soil samples from every field on each of the 12 farms.

samples as expected. There was a drop in the dieldrin milk residue level on most of the farms between the February 2 and March 17 samplings. Indications are that this general downward trend continued on at least 8 of the 12 farms through the spring and summer months as shown by the September 24 samples. It can be speculated that these findings are attributable to an overall decrease in dieldrin residues in crops and soils, but with a peak being reached during the winter months due to the following factors:

1. November and December are the months of heavy freshening of cattle on these farms. Dry stock and young stock could release higher than normal amounts of dieldrin in their milk for a time after freshening.

2. Cattle subjected to extremely cold temperatures could draw on body fat reserves (dieldrin storage site) at a faster than normal rate and release greater amounts of dieldrin into their milk.

Significant amounts of dieldrin milk residues occurred even on dairy farms (Farms 8 through 12) where no aldrin was used in the 10-year period, 1961-70. Only trace levels of dieldrin existed in the soil on these farms.

The dairyman on Farm 7 had used aldrin from 1961 to 1964. Soil residues after 7 years showed just trace levels

of dieldrin, and dieldrin milk residues were also low. There is some indication from the data that dairy cattle on a farm having a history of aldrin soil treatments within the last 6 to 7 years will produce milk with dieldrin residues near or above the Food and Drug Administration's current administrative guideline of 0.3 ppm dieldrin in milk (fat basis).

Decker, Bruce, and Bigger (3) showed that 75% of the aldrin applied to soil dissipates by the end of the first year. The remaining residue in the form of dieldrin in the soil dissipates more slowly (about 12% per year) with a half-life of 4 years.

All the samples of hay and oat straw contained significant levels of dieldrin residues (Table 2). Farms 1 through 6 had slightly higher levels of dieldrin in these materials than Farms 7 through 12. However, the dieldrin in the hay and oat straw on these latter farms could still account for a significant part of the dieldrin residue occurring in the milk.

Gannon, Link, and Decker (4) demonstrated that dairy cattle fed 0.05 ppm of dieldrin in their total diet produced milk with a dieldrin residue (butterfat basis) of 0.25 ppm.

Roasted soybeans being fed to cattle on Farms 6 and 7 had high dieldrin residues. Significant dieldrin residues were also present in the roasted soybeans from Farm 3, while only trace levels of dieldrin occurred in the roasted soybeans from Farm 8. The two samples of soybean meal obtained from Farms 1 and 12 showed only trace levels of dieldrin. This is to be expected since the oil (main storage site for dieldrin in soybeans) is extracted in the preparation of meal. All soybean feed had been purchased.

Corn silage, commercial feed concentrates and protein supplements, and the water on all 12 farms showed only trace amounts of dieldrin (Table 2). There was no apparent correlation between the dieldrin soil residue and the dieldrin residue in the silage. The silage was cut at heights varying from 4 to 7 inches above ground. Farms 1, 2, and 3 had the highest dieldrin soil residues, and the silage on these farms was cut at 4, 7, and 6 inches above ground, respectively. The overall dieldrin milk residue is still increased slightly by feeds with trace residues of dieldrin. In addition, cattle can absorb dieldrin directly through their skin. The amount of dieldrin absorbed by cattle lying on contaminated soil and from consuming dieldrin-contaminated soil is currently not predictable. The soil type, soil moisture level, and other factors would vary greatly the amount of dieldrin cattle obtain directly from the soil. As already pointed out, there is a fivefold increase from the level of dieldrin in the diet to the level of dieldrin occurring in the milk fat.

There was no good correlation between the dieldrin soil residues and the dieldrin residues in hay and oat straw (Table 2). Levels of dieldrin in hay and oat straw exceeded 0.01 ppm on all farms including those where no aldrin had been used for at least 10 years. The waxy coating on hay and straw can readily absorb dieldrin; therefore, it is suggested that direct contact with contaminated ground soil or blowing soil particles could account for the dieldrin residues in the hay and straw.

A higher dieldrin soil residue was present at the soil surface than in a 6-inch deep composite core sample in permanent pastures on six farms (Table 2). Aldrin had never been applied to this land, and no soil erosion from adjacent fields had occurred at the sampling sites. This would indicate that soil particles contaminated with dieldrin were transported by the wind to these fields.

Cohen and Pinkerton (5) in a study conducted at Cincinnati, Ohio, reported the average monthly dust fall to be 15 tons per square mile. The dust source was from the southern high plains of Texas and contained 0.003 ppm dieldrin. They further stated that the movement of soil particles by air within a localized area is a certainty.

Conclusions

1. The chances of dieldrin residues in milk exceeding the FDA's current administrative guideline of 0.3 ppm (fat basis) are greatest on dairy farms having a history of aldrin soil treatment within the last 6 or 7 years. Therefore, it can be expected that additional herds, but in lessening numbers, will be found producing milk with illegal residues for about 4 to 6 more years.
2. Hay and oat straw supply significant amounts of dieldrin to dairy cattle. In addition, roasted soybeans, a relatively new feed for dairy cattle, could be an important source of dieldrin contamination in milk.
3. Corn silage, commercial feed concentrate, and well water are usually not important sources of dieldrin contamination to dairy cattle.

See Appendix for chemical names of compounds discussed in this paper.

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RESIDUES IN FISH, WILDLIFE, AND ESTUARIES

*Organochlorine Residues in Estuarine Mollusks,
1965-72—National Pesticide Monitoring Program*¹

Philip A. Butler²

Part I. General Summary and Conclusions

Part II. Residue Data—Individual States

SECTION A: ALABAMA
SECTION B: CALIFORNIA
SECTION C: DELAWARE
SECTION D: FLORIDA
SECTION E: GEORGIA
SECTION F: MAINE
SECTION G: MARYLAND
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SECTION L: SOUTH CAROLINA
SECTION M: TEXAS
SECTION N: VIRGINIA
SECTION O: WASHINGTON

ABSTRACT

This paper describes the development of the national program for monitoring estuarine mollusks in 15 coastal States and reports the findings for the period 1965-72. The report is presented in two parts: Part I. General Summary and Conclusions, and Part II. Residue Data—Individual States.

Analyses of the 8,095 samples for 15 persistent organochlorine compounds showed that DDT residues were ubiquitous; the maximum DDT residue detected was 5.39 ppm. Dieldrin was the second most commonly detected compound with a maximum residue of 0.23 ppm. Endrin, mirex, toxaphene, and polychlorinated biphenyls were found only occasionally. Results indicate a clearly defined trend towards decreased levels of DDT residues, beginning in 1969-70. At no time were residues observed of such a magnitude as to imply damage to mollusks; however, residues were large enough to pose a threat to other elements of the biota through the processes of recycling and magnification.

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Part I. General Summary and Conclusions

Introduction

Initial investigations of the effects of pesticides on estuarine fauna were undertaken at the Gulf Breeze Laboratory in 1958 to determine if the pesticide lindane might be safely used directly in estuarine waters to control crabs preying on shellfish populations. The unexpected acute toxicity of this chemical, not only to crabs but also to nontarget organisms, revealed by these early experiments prompted a broad investigation of both the direct and indirect effects of persistent synthetic pesticides. The extent of the problem was not known, and the investigators were concerned about the potential harm to estuarine fauna exposed to drainage waters from large river basins where significant quantities of pesticides were used. Marine commercial fisheries were recognized as being especially vulnerable since a major portion of their catch, both in tonnage and dollar value, is made up of estuarine-dependent species.

The acute toxicity of a broad spectrum of pesticides was determined under laboratory conditions (14-17). These data, however, could be most useful only if there were information on the actual levels of organochlorines reaching the estuarine environment. Accordingly, a search was undertaken for meaningful tools to measure this type of pollution (6).

The decision to use mollusks as bioassay tools was based on the findings of laboratory experiments designed to measure the uptake and flushing rates of an array of organochlorine pesticides under controlled conditions. Various species of mollusks, but primarily the eastern oyster, *Crassostrea virginica*, were exposed to appropriate concentrations of pesticides added continuously to a flowing seawater system. Results indicated that oysters detect DDT in the ambient water supply at levels as low as 10 parts per trillion (10⁻¹¹). By the process of biomagnification, residues of DDT as high as 25 ppm accumulate in oyster tissues within 96 hours at a level of environmental contamination of only 1.0 ppb (7). Oysters tolerate tissue residues of DDT at least as high as 150 ppm without apparent ill effect provided residues are accumulated slowly. However, as little as 0.1 ppm of DDT in the oyster's water supply terminates feeding activities and at summer water temperatures (31°C) will cause death.

Organochlorine residues are flushed rapidly from molluscan tissues when the water supply is no longer contaminated. In one experimental series, for example, DDT residues of about 25 ppb in oysters and soft

clams, *Mya arenaria*, diminished by 50-90% after a week of flushing in clean water. Consequently, it is possible to learn much about the periodicity of organochlorine pollution in estuaries from samples of sedentary species collected at appropriately brief intervals.

As a result of these studies, it was possible for the Bureau of Commercial Fisheries to undertake a program for monitoring pesticide residues in estuarine mollusks to determine the extent of organochlorine pollution. The collection of samples was not begun immediately in some areas, while in others, sample collection was terminated at an early date. The program was continuously operative, however, from July 1965 through June 1972. In 1971, the Gulf Breeze Laboratory and the monitoring program became a part of the U.S. Environmental Protection Agency.

The following report describes the 7-year (1965-72) data collection and discusses, specifically, the well-defined trends in the magnitude of DDT residues in estuarine mollusks. Except where noted, the term DDT includes the metabolites TDE and DDE. All residue analyses are presented, by State, in Part II of this report. A report summarizing the first 3 years of this program was published in 1969 (3).

Data Interpretation

Although the eastern oyster has a wide distribution, it was obvious that some other species might be more available for monitoring in different geographical areas or salinity regimes; thus, different species of mollusks were tested in the laboratory to determine their relative capabilities in the uptake and retention of organochlorine pollutants (2). Such information is all important for the understanding of these monitoring data.

In the tests, all species were exposed to the same hydrographic conditions with low turbidity and a salinity level about 80% that of seawater. It is probable that species accustomed to different ecological conditions would react more efficiently in nature than in the Laboratory. Caution must be exercised in the extrapolation of laboratory results to field conditions, and, at best, such data serve only as guidelines for the interpretation of residue levels in monitored samples.

In general, any of three species of oysters, four species of mussels and two species of clams were found to be reliable indicators of the magnitude of organochlorine pollution (Table 1). In some areas it was necessary to use the hard clam, *M. mercenaria*, although it is the least satisfactory of the species evaluated. Under similar

laboratory conditions, for example, hard clams accumulated pesticide residues less than half as large as those in oysters. Moreover, the residues were flushed from the clam much more quickly than from the oyster when clean water was restored.

Sample Collection and Preparation

The management of estuarine molluscan resources is the responsibility of the individual States; therefore, in each coastal area there is a cadre of specialists who are not only interested in estuarine pollution but who also have the knowledge and equipment necessary to collect shellfish samples. Without the continuing cooperation

of these agencies (see Acknowledgment), this program could not have achieved its objectives.

Estuaries with well defined drainage basins and bays that could be considered "nursery areas" for estuarine fauna were selected for monitoring.

Some sites were monitored because of suspected pollution problems. To insure continuity of data, permitting detection of not only seasonal but yearly trends in pesticide pollution levels, it was essential, too, that the stations selected have shellfish populations large enough for monthly collections over a number of years. Duplicate samples of 15 or more mature mollusks were collected and prepared for shipment at about 30-day intervals. About 10% of all samples were analyzed in replicate; the remaining duplicates were discarded after satisfactory analysis of the sample. Sample collections were interrupted by the loss of shellfish populations to vandals, floods, and hurricanes, but the overall continuity of the data was good.

Coverage of coastal estuaries was incomplete in this program because other agencies were monitoring shellfish in some states, notably Alabama, Louisiana, and Massachusetts. The number of sample collections by State and year is tabulated in Table 2. The original plan was to monitor each area for 5 years so that trends in pesticide residue levels could be detected. The general absence of residues in Washington estuaries, however, prompted an earlier termination of monitoring in that State. In addition to the samples tabulated, about 2,000 miscellaneous samples of other species of vertebrates and invertebrates were collected and analyzed. These frequently had more varied pesticide residues and at higher levels than mollusks but are omitted from this report because of difficulty in determining their source.

The analysis of all samples by a single laboratory to insure uniformity seemed important in planning the program. Various potential preservatives were examined to find a method for shipping samples without resorting to refrigeration. Eventually, it was discovered that by dehydrating the homogenized tissues of mollusks or other marine animals with a desiccant mixture, the dry samples could be wrapped in aluminum foil and held without refrigeration for 15 or more days without degradation or loss of organochlorine residues (2). This made it possible to ship the samples by regular mail.

In practice, samples of 15 or more mature oysters or other mollusks were collected and taken to the cooperating agency's laboratory. Samples not to be processed immediately could be refrigerated for 2 or 3 days in the shell. If longer storage was necessary, animals were shucked and the undrained meats frozen in mason jars. The shucked meats were homogenized in an electric blender, and approximately 25-g aliquots were blended

TABLE 1.—*Pelecypod mollusks used to monitor organochlorine residues in 15 States—1965-72*

SCIENTIFIC AND COMMON NAMES OF MOLLUSKS	
<i>Crassostrea gigas</i>	Pacific oyster
<i>Crassostrea virginica</i>	eastern oyster
<i>Ostrea lurida</i>	Olympia oyster
<i>Modiolus demissus</i>	ribbed mussel
<i>Modiolus modiolus</i>	northern horse mussel
<i>Mytilus californianus</i>	Californian mussel
<i>Mytilus edulis</i>	blue mussel
<i>Mercenaria mercenaria</i>	hard clam
<i>Mya arenaria</i>	soft clam
<i>Corbicula fluminea</i>	Asiatic clam, fresh water

STATE	SPECIES COLLECTED
Alabama	<i>C. virginica</i>
California	<i>C. gigas</i> <i>O. lurida</i> <i>M. demissus</i> <i>M. californianus</i> <i>M. edulis</i> <i>C. fluminea</i>
Delaware	<i>C. virginica</i> <i>M. demissus</i> <i>M. mercenaria</i>
Florida	<i>C. virginica</i>
Georgia	<i>C. virginica</i>
Maine	<i>M. modiolus</i> <i>M. edulis</i> <i>M. arenaria</i>
Maryland	<i>C. virginica</i>
Mississippi	<i>C. virginica</i>
New Jersey	<i>C. virginica</i>
New York	<i>C. virginica</i> <i>M. demissus</i> <i>M. edulis</i> <i>M. mercenaria</i> <i>M. arenaria</i>
North Carolina	<i>C. virginica</i>
South Carolina	<i>C. virginica</i>
Texas	<i>C. virginica</i>
Virginia	<i>C. virginica</i>
Washington	<i>C. gigas</i>

with precisely three times their weight of desiccant to yield a total sample weight of about 100 g. Alternate blending and chilling (not freezing) of sample is required to achieve a dry, free-flowing product. The amount of desiccant used depends on the moisture content of the sample. Less desiccant is required for fish (two times body weight), while up to nine times as much desiccant may be used with small samples, plankton for example, to achieve a 100-g final weight of the sample to be processed. The desiccant is made up of about 90% sodium sulfate and 10% Quso (Quso G30, manufactured by Philadelphia Quartz Co., Philadelphia, Pa.), a micro fine precipitated silica.

Analytical Procedures

Throughout the monitoring program samples were routinely screened for the following substances: aldrin, chlordane, *p,p'*-DDT, *p,p'*-TDE (DDD), *p,p'*-DDE, dieldrin, endrin, heptachlor, heptachlor epoxide, lindane, methoxychlor, mirex, and toxaphene. On the few occasions when the *o,p'* isomers of DDT were detected in quantifiable amounts, they were included with the *p,p'* residues.

On receipt in the laboratory, samples were extracted for 4 hours with petroleum ether in a Soxhlet apparatus. Extracts were concentrated and transferred to 250-ml separatory funnels. The extracts were diluted to 25 ml with petroleum ether and partitioned with two, 50-ml

portions of acetonitrile previously saturated with petroleum ether. The acetonitrile was evaporated just to dryness, and the residue eluted from a Florisil column (12). The sample was then identified and quantitated by electron capture gas chromatography. Three columns of different polarity (DC-200, QF-1, and mixed DC-200/QF-1) were used to confirm identification. Operating parameters on Varian Aerograph gas chromatographs were as follows:

Columns: Pyrex glass 6' x 1/4" (o.d.) packed with 3% DC-200, 5% QF-1, and a 1:1 ratio of 3% DC-200 and 5% QF-1, all on 80/100 mesh Gas Chrom Q
 Temperatures: Detector—210° C
 Injector—210° C
 Oven—190° C
 Carrier gas: Prepurified nitrogen at a flow rate of 40 ml/min

A few samples were analyzed by thin layer chromatography. All residues are reported on a wet-weight basis. The lower limit of quantification was 10 ppb. Laboratory tests conducted during the sampling period gave the following recovery rates: *p,p'*-DDE, 80-85%; *p,p'*-TDE, 92-95%; *p,p'*-DDT, 91-95%. Data in this report do not include a correction factor for percent recovery.

Toxaphene sometimes interfered with the quantification of DDT residues which, in these cases, are reported as approximate values. The lower limit of quantification of toxaphene was 250 ppb. The presence of polychlorinated biphenyls (PCB's) also interfered with the quantification of DDT residues. In most instances, DDT was calculated as though PCB's were not present. Acquisition

TABLE 2.—Summary of sample collections by State and year—1965-72

STATE	PRINCIPAL SPECIES MONITORED	NUMBER OF SAMPLE COLLECTIONS								
		1965	1966	1967	1968	1969	1970	1971	1972	TOTALS
Alabama	<i>C. virginica</i>				13	20				33
California	<i>C. gigas</i>		136	180	167	139	45	75	30	772
Delaware	<i>M. mercenaria</i>		16	101	99	71				287
Florida	<i>C. virginica</i>	6	80	102	82	44	35	19	6	374
Georgia	<i>C. virginica</i>			112	127	124	121	120	60	664
Maine	<i>M. arenaria</i>	6	95	89	79	83	44			396
Maryland	<i>C. virginica</i>		18	20	26	9	15			88
Mississippi	<i>C. virginica</i>	30	71	72	72	63	66	60	36	470
New Jersey	<i>C. virginica</i>		23	44	45	39	33	27	8	219
New York	<i>M. mercenaria</i>		148	183	175	174	148	143	88	1,059
North Carolina	<i>C. virginica</i>		96	201	204	204	124	136	66	1,031
South Carolina	<i>C. virginica</i>	72	142	143	145	108				610
Texas	<i>C. virginica</i>	53	133	125	93	97	103	95	29	728
Virginia	<i>C. virginica</i>	56	117	123	120	112	105	27	9	669
Washington	<i>C. gigas</i>	40	218	223	214					695
Total		263	1,293	1,718	1,661	1,287	839	702	332	8,095

of the appropriate standards permitted the identification of Aroclor 1254³ in samples from California, Florida, Georgia, Texas, and Virginia, and Aroclor 1242³ in samples from Virginia. Since 1970, PCB residues have been approximately quantified in samples from Florida and, more recently, from Virginia.

There is some question as to how much interference by PCB's exists in the sample analyses reported in the early years of the monitoring program. At this time there is no way of knowing with certainty. It is considered significant that in the period 1965-70 there was a 3-8% annual increase in the domestic sale of these chemicals, and total domestic sales in 1970 were more than double sales in 1960; however, PCB residues were identified in samples from relatively few estuaries in 1971.

During the course of the program, several States extended the monitoring of their estuaries and collected more samples than the Gulf Breeze Laboratory was equipped to process. Analytical equipment similar to that used at Gulf Breeze was provided to these agencies as well as a manual of operations (Prepared by A. J. Wilson, Jr., Research Chemist, Gulf Breeze Laboratory), to insure similar methodology in analytical techniques. For the first few collections under the new arrangement, samples were split and analyses made by both the State and Federal laboratories. Excellent comparability in data was obtained (13) and thereafter, the State agency submitted only the monthly data reports to the Gulf Breeze

Laboratory. Such arrangements were in effect during portions of the monitoring program in California, Georgia, Maine, New York, and Virginia.

Data Summaries and Discussion

DDT with its analogs was the most commonly identified pesticide and occurred in 63% of all samples analyzed (Table 3). Dieldrin was the second most commonly detected residue with an incidence of 15%. DDT and dieldrin were detected in some samples from all States monitored (Tables 4 and 5). Other organochlorine residues were encountered infrequently and generally at low levels, with the exception of toxaphene. The large number of Georgia samples containing toxaphene reflects the direct contamination of the marine environment by the effluent from a single manufacturing plant.

The incidence of DDT residues varied markedly from one drainage basin to another and was not correlated with the magnitude of the residues. Only in New Jersey and Alabama, for example, did all samples contain detectable residues of DDT, but the size of DDT residues was greater in several other States (Table 4). It is true that in both Alabama and New Jersey, monitored oyster populations were exposed primarily to the runoff from a single, although complex, drainage basin. In other States, samples were collected from several distinct drainage basins.

TABLE 3.—Summary of organochlorine residues detected in estuarine mollusks by State—1965-72

STATE	TOTAL NUMBER OF SAMPLES	NUMBER OF SAMPLES WITH RESIDUES > 5 PPB ($\mu\text{g}/\text{kg}$) AND MAXIMUM RESIDUE () DETECTED IN PPB ($\mu\text{g}/\text{kg}$)					
		DDT	DIELDRIN	ENDRIN	MIREX	TOXAPHENE	PCB'S
Alabama	33	33 (616)	6 (21)				
California	772	712 (3,970)	194 (57)	14 (19)		4 (11,000)	¹ 21
Delaware	287	216 (205)	37 (25)				
Florida	374	230 (5,390)	27 (28)				25 (390)
Georgia	664	96 (96)	141 (230)			128 (54,000)	¹ 16
Maine	396	72 (359)	14 (38)				
Maryland	88	71 (70)	11 (22)				
Mississippi	470	285 (135)	19 (20)				
New Jersey	219	219 (278)	52 (26)				¹ 6
New York	1,059	858 (596)	456 (132)				
North Carolina	1,031	768 (566)	12 (19)				
South Carolina	610	332 (154)	24 (154)			12 (540)	
Texas	728	530 (1,249)	134 (87)	22 (32)		¹ 1	¹ 5
Virginia	669	585 (678)	112 (40)				19 (2,800)
Washington	695	78 (176)	1 (120)				

¹ Present but not quantified.

TABLE 4.—Listing of States in order of frequency and maximum value of DDT residues in mollusks

STATE	FREQUENCY OF RESIDUES (%)	STATE	MAXIMUM VALUE IN PPB
Alabama	100	Florida	5,390
New Jersey	100	California	3,970
California	92	Texas	1,249
Virginia	87	Virginia	678
New York	81	Alabama	616
Maryland	81	New York	596
Delaware	75	North Carolina	566
North Carolina	75	Maine	359
Texas	73	New Jersey	278
Florida	62	Delaware	205
Mississippi	61	Washington	176
South Carolina	54	South Carolina	154
Maine	18	Mississippi	135
Georgia	15	Georgia	96
Washington	11	Maryland	70

NOTE: These comparisons are limited in that the number of samples, number of sampling stations, periods (years) of sampling, and species of mollusks differ for each State.

TABLE 5.—Listing of States in order of frequency and maximum value of dieldrin residues in mollusks

STATE	FREQUENCY OF RESIDUES (%)	STATE	MAXIMUM VALUE IN PPB
New York	43	Georgia	230
California	25	South Carolina	154
New Jersey	24	New York	132
Georgia	21	Washington	120
Alabama	18	Texas	87
Texas	18	California	57
Virginia	17	Virginia	40
Delaware	13	Maine	38
Maryland	13	Florida	28
Florida	7	New Jersey	26
Mississippi	4	Delaware	25
South Carolina	4	Maryland	22
Maine	4	Alabama	21
North Carolina	1	Mississippi	20
Washington	<1	North Carolina	19

NOTE: These comparisons are limited in that the number of samples, number of sampling stations, periods (years) of sampling, and species of mollusks differ for each State.

The magnitude of all DDT residues was low compared to residues reported in carnivores such as fish-eating birds. By extrapolation from laboratory experiments, the monitoring data indicate that, in most cases, estuarine pollution with DDT was intermittent and at levels in the low parts-per-trillion range. In only 38 samples (0.5%) did the residue exceed 1.0 ppm. These samples were collected in California, Florida, and Texas in drainage basins having intensive agricultural development. The single highest residue of 5.39 ppm (DDT-3.70 ppm, TDE-0.76 ppm, DDE-0.93 ppm) was observed in oysters from the Caloosahatchee River drainage basin in Florida where the seasonal pattern of residue fluctuations indicated an agricultural or at least a scheduled use of the pesticide (Fig. 1). It is significant that extensive acreage in this drainage basin was devoted to sugarcane and sweetcorn that would be maturing and receiving fairly heavy applications of pesticides during the peak residue periods indicated in Fig. 1 (R. G. Curtis, 1972, Florida Cooperative Extension Service, *personal communication*). In controlled feeding experiments in the laboratory, from 50 to 100% mortality was observed in small populations of commercial species of shrimp, crabs, and fish fed exclusively diets containing less than 3.0 ppm of *p,p'*-DDT (4).

In a survey of 7,000 analyses of mollusk samples completed in the period 1965-71, the mean residue composition was 24% DDT, 39% TDE, and 37% DDE. Exceptions to this average picture were Station 2 in New Jersey where DDT comprised only 4% (mean of 47 samples in 5 years) and Station 18 in Washington where DDT made up 75% of the residues (mean of

36 samples in 3 years). Biotic recycling of persistent residues is usually associated with the high percentages of DDT metabolites found in dominant carnivores. It is of interest that the metabolites were the only residues detected in many of these analyses of filter-feeding mollusks. Results of a study by Johnson *et al.* (10) indicated that there are some animals, however, such as aquatic insects, in which direct exposures to DDT result in tissue residues that are more than 80% DDE. The large percentage of the parent compound DDT in residues from Washington mollusks does imply a direct contamination of the estuarine environment, perhaps, for insect control. But in general, the percentage distribution of DDT metabolites in these samples revealed little about the kinetics of DDT in the estuary.

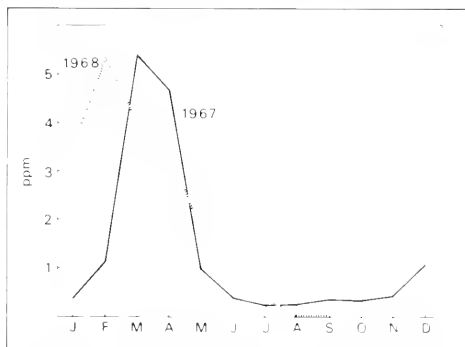


FIGURE 1.—DDT residues in the eastern oyster from the Caloosahatchee River Basin, Lee County, Fla., by month of collection—1967 and 1968

Trends in DDT Residues

Many of the estuaries were monitored over a sufficient period of time to permit detection of clearly defined trends in DDT residue patterns. Average DDT residue levels detected in the first 2 to 5 years and average levels in the final year of monitoring in each State are presented in Table 6. The overall picture is that of a pronounced decline, about 55%, in the number of samples containing DDT residues in excess of 100 ppb. There was a 20% decrease in the 10-100 ppb range, and a concomitant 44% increase in the number of samples having negligible or undetectable DDT residues.

There are important exceptions to this average picture. In California, New York, and Virginia, for example, more samples had residues in excess of 10 ppb in 1971 than in earlier years. On the other hand, the percent of samples having residues higher than 100 ppb declined in these States. It would appear that in some areas, DDT pollution has become more widespread, but has resulted in residues of lower magnitude in the estuarine food web.

Since organochlorine residues in mollusks showed a continuing decline in most areas during the years that

domestic sales and presumably usage of PCB compounds were increasing, PCB's were not considered to be a significant factor in the early monitoring data.

Too few samples from Alabama were analyzed in this program to indicate any trend in residue magnitude. The mean value of 88 ppb of DDT in 33 samples collected in 1969-70 may be compared, however, with a mean residue level of 330 ppb in a series of 82 samples of oysters collected in 1965-66 (7).

Exact comparisons by States of the data in Table 6 are not valid since in succeeding years there were different numbers of samples and occasionally different species of mollusks collected at the same station. A more critical review of data on DDT residues is possible for 10 stations in North Carolina. These stations were selected for the continuity of sampling of the eastern oyster at monthly intervals for more than 5 years. The number of samples containing less than 11 ppb of DDT increased steadily until, in 1971, 76% of all residues were in this category as compared to only 8% in 1966 and 1967. The corresponding decrease in the number of samples containing larger residues is shown in Fig. 2 and Table 7.

TABLE 6.—Percent distribution of DDT residues of different magnitude in estuarine mollusks by State—1965-71 (7,000 samples)

STATE	PERCENT DISTRIBUTION OF SAMPLES							
	<11 PPB		11-100 PPB		101-1,000 PPB		>1,000 PPB	
	FIRST 2 TO 5 YEARS	1971	FIRST 2 TO 5 YEARS	1971	FIRST 2 TO 5 YEARS	1971	FIRST 2 TO 5 YEARS	1971
Alabama			69		31			
California	14	7	30	64	51	28	5	1
Delaware	23	30	62	67	15	3		
Florida (1 station)	43	100	57					
Georgia	85	96	15	4				
Maine	82	98	17	2	1			
Maryland	19	50	81	50				
Mississippi	42	72	56	27	2	1		
New Jersey		7	69	74	31	19		
New York	26	22	60	74	14	4		
North Carolina	22	76	68	24	10			
South Carolina	52	82	47	18	1			
Texas	34	52	53	45	13	3	<1	
Virginia	18		67	95	15	5		
Washington (1 station)	92	94	8	6				
Mean	39	56	49	39	11	5	<0.5	

TABLE 7.—Trends in magnitude of DDT residues in oysters, 10 stations, North Carolina

YEAR	TOTAL NUMBER OF SAMPLES	<11 PPB		11-100 PPB		101-1,000 PPB	
		NUMBER OF SAMPLES	PERCENT DISTRIBUTION	NUMBER OF SAMPLES	PERCENT DISTRIBUTION	NUMBER OF SAMPLES	PERCENT DISTRIBUTION
1966	60	5	8	45	75	10	17
1967	119	9	8	90	76	20	16
1968	120	26	22	70	58	24	20
1969	120	29	24	77	64	14	12
1970	109	61	56	46	42	2	2
Subtotal	528	130	25	328	62	70	13
1971	115	87	76	26	22	2	2
Percent change in 1971 from average for 1966-70		+204%		-65%		-85%	

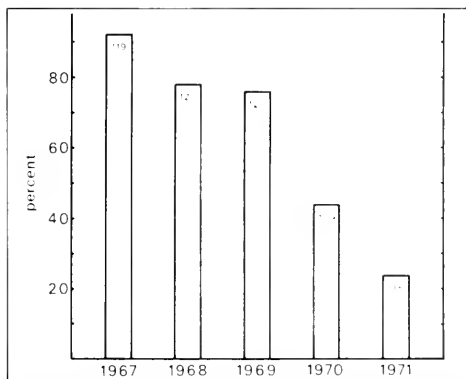


FIGURE 2.—Percent of eastern oyster samples containing more than 10 ppb of DDT, average of monthly samples collected at 10 stations in North Carolina (Numbers in bars indicate total number of samples)

Conclusions

The data demonstrate that in most estuaries monitored, detectable DDT residues have declined in both number and magnitude in several species of estuarine mollusks in recent years. DDT pollution in many estuaries, as judged by the magnitude of the residues in mollusks, peaked in 1968 and has been declining markedly since 1970.

The sensitivity of mollusks to organochlorine pollutants plus the fact that they are filter-feeders warrant the assumption that the contribution of particulate DDT to estuaries from one or more primary sources such as drainage basin runoff waters, atmospheric fallout, and persistent reservoirs in bottom sediments, has declined significantly.

In view of the efficiency of mollusks in detecting and storing residues of the persistent organochlorines, it is clear that relatively low levels of this type of pollution were present in the monitored areas during the period 1965 to 1972.

Appropriate correlations of the residue data reported here with available records of drainage-basin discharge rates, precipitation, and hydrographic factors in the various types of estuaries should provide a useful model for predicting the effects of future introductions of unspecified synthetic substances chemically similar to DDT.

See Appendix for chemical names of compounds discussed in this paper.

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We are greatly indebted to the graduate students and technicians whose diligence in the collection and processing of samples made the program a reality. I trust that the results will make them pleased with their participation.

The program could not have been developed without the interest and skills of Alfred J. Wilson, Jr., Research Chemist at the Gulf Breeze Laboratory.

Lastly, I thank the administrators and professional staffs of the cooperating agencies who kindly let me think that the monitoring program had the number one priority on their busy schedules.

In view of the volume of data in this report, it is inevitable that there are sins of both omission and commission. The writer would be most grateful for have these called to his attention so that the record can be appropriately emended.

COOPERATING AGENCIES—This alphabetical listing by States includes the names of investigators and, where appropriate, chemists and their titles at the time they were participating in the program. Where chemists are not listed, the samples were analyzed at the Gulf Breeze Laboratory under the supervision of Alfred J. Wilson, Jr., with the assistance of Jerrold Forrester and Johnny Knight. The listing of more than one principal investigator or agency in any one State reflects changes taking place during the monitoring period 1965-72. Operational funds were provided by the U.S.F.W.S., Bureau of Commercial Fisheries (BCF) for the collection of samples and for analytical equipment where contracts are indicated. In States participating by agreement, the BCF provided equipment and chemicals. In 1971-72, the program was jointly funded by the National Marine Fisheries Service (NMFS) and the Environmental Protection Agency.

ALABAMA	Alabama Marine Resources Laboratory Johnie H. Crance, Director; E. B. May, Principal Investigator. Agreement.
CALIFORNIA	California Dept. of Fish and Game, Marine Resources Operations Dr. H. C. Orcutt, Laboratory Supervisor; John Modin, Chemist. Contracts, BCF: 14-17-0007-332; 14-17-0002-211; -265; -337; -532. California Department of Fish and Game, Resources Agency W. H. Griffith, Principal Investigator. Contract, NMFS: N-042-10-72(N).
DELAWARE	University of Delaware Dr. F. C. Daiber, Principal Investigator. Contracts, BCF: 14-17-0002-117; -261; -326.
FLORIDA	State Board of Conservation Marine Laboratory R. M. Ingle, Director of Research. Agreement. Bureau of Commercial Fisheries—Environmental Protection Agency, Gulf Breeze Laboratory. Dr. T. W. Duke, Director. Agreement.
GEORGIA	The University of Georgia Dr. T. L. Linton, Principal Investigator. Contracts, BCF: 14-17-0002-220; -267. C. J. Durant, Principal Investigator and Chemist. Contracts, BCF: 14-17-0002-344; -454. Dr. R. J. Reimold, Principal Investigator. Contract, NMFS: N-042-12-71(N).
MAINE	Department of Sea and Shore Fisheries L. Varney, Principal Investigator; John Hurst, Laboratory Director and Chemist. Contracts, BCF: 14-17-0007-333; 14-17-0002-206; -263; -332; -434.
MARYLAND	BCF Biological Laboratory Dr. A. Rosenfield, Principal Investigator. Agreement.
MISSISSIPPI	Gulf Coast Research Laboratory Dr. W. P. Abbott, Principal Investigator. Contracts, BCF: 14-17-0002-133; -172; -235; -341. Dr. G. Gunter, Laboratory Director. Contract, NMFS: N-042-11-71(N).
NEW JERSEY	Rutgers—The State University, Oyster Research Laboratory Dr. H. H. Haskin and D. E. Kunkle, Principal Investigators. Agreement.
NEW YORK	New York State Department of Environmental Conservation D. H. Wallace, Director of Marine Fisheries; J. Fochrenbach, Chemist. Contracts, BCF: 14-17-0002-163; -219; -268; -345; -455; NMFS: N-042-14-71(N).
NORTH CAROLINA	University of North Carolina, Institute for Marine Sciences Dr. A. F. Chestnut, Principal Investigator. Contracts, BCF: 14-17-0002-182; -239; -343; NMFS: N-042-15-71(N).
SOUTH CAROLINA	Bears Bluff Laboratories, Inc. Dr. G. R. Lunz, Director (deceased). Contracts, BCF: 14-17-0002-130; -171; -234; -340; -426.
TEXAS	State of Texas, Parks and Wildlife Department T. R. Leary, Coastal Fisheries Coordinator; R. Childress, Principal Investigator. Agreement.
VIRGINIA	Virginia Institute of Marine Science Dr. M. L. Brehmer, Principal Investigator; Dr. R. J. Hargett, Principal Investigator and Chemist. Contracts, BCF: 14-17-0002-138; -174; -237; -342; -452; NMFS: N-042-13-71(N).
WASHINGTON	State of Washington, Department of Fisheries C. Lindsay, R. E. Westley, Principal Investigators. Contracts, BCF: 14-17-0002-134; -173; -236.

Part II. Residue Data—Individual States

The following sections present residue data for the 15 coastal States where estuarine mollusks were monitored for organochlorine residues. A map showing sampling sites in the respective States together with a discussion of the findings are included in each section.

SECTION A.—ALABAMA

Samples of the eastern oyster, *Crassostrea virginica*, were collected in Alabama at 3-month intervals during 1968-69 from four commercial reefs in or near Mobile Bay. Samples were processed at the Alabama Marine Resources Laboratory and mailed to the Gulf Breeze Laboratory for chemical analysis.

Approximate station locations are shown in Fig. A-1. Stations 1 and 2 on the eastern shore of Mobile Bay are influenced more by the presumably cleaner Gulf of Mexico waters than Stations 3 and 4 which are more exposed to drainage waters from the Alabama-Tombigbee River Basin. Both Stations 1 and 4 are influenced to an unknown extent by small drainage basins in the coastal areas of Alabama. A summary of data on organochlorine residues in the monitored species, *C. virginica*, is presented in Table A-1, and the distribution of residues in this species for each sampling station by date of collection in Table A-2. Many of these data have already been published by the cooperating agency (10).

All 33 samples contained detectable amounts of DDT, but the sampling series was conducted in Alabama for too few years to indicate annual trends in pollution levels. An earlier study of pesticide residues in Mobile Bay oysters (7) also reported a 100% incidence of DDT in 82 samples analyzed; however, maximum DDT residues at Shell Bank and Cedar Point reefs were 13 and 25 times higher in 1965 than those observed in

this study in 1969. Because of differences in sample preparation in the two studies, 1965 residues could be expected to be only about 10% higher than the 1969 data had there been no change in DDT pollution levels in the bay. Alabama and New Jersey were the only States of the 15 monitored in which 100% of the samples contained detectable residues of DDT. The maximum level of DDT in Alabama oysters (616 ppb) was lower than residues found in four other States.

Dieldrin residues were small, but the 18% incidence was significantly higher than the average incidence for all States of 15%. The incidence and magnitude of dieldrin residues in the 1965 study (7) were significantly higher.

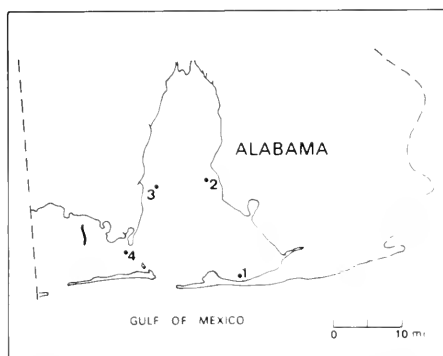


FIGURE A-1.—Diagram of coastal Alabama showing approximate location of monitoring stations

1. Shellbank—Bon Secour Bay
2. Klondike—Mobile Bay
3. Whitehouse—Mobile Bay
4. Cedar Point Reef—Mississippi Sound

TABLE A-1.—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1968-69—Alabama

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (μg/KG)	
				DDT	DIELDRIN
1	Shellbank	1968-69	8	8 (214)	1 (14)
2	Klondike	1968-69	8	8 (445)	1 (14)
3	White House	1968-69	7	7 (616)	2 (21)
4	Cedar Point	1968-69	8	8 (372)	2 (13)
	Occasional stations (2)	1968-69	2	2 (237)	
Total number of samples			33		
Percent of samples positive for indicated compound				100	18

¹ Each sample represents 15 or more mature mollusks.

TABLE A-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Alabama

[Blank = no sample collected; — = no residue detected above 5 ppb; T = >5 but <10 ppb]

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)												
		JAN.	FEB	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.	
STATION 1.—SHELL BANK—8 SAMPLES ¹														
1968	DDE				110			110				33		
	TDE				88			52				15		
	DDT				16			12				—		
1969	DDE	48			94			48			26	T		
	TDE	31			70			25			17	T		
	DDT	—			—			—			13	—		
	Dieldrin	—			14			—			—	—		
STATION 2.—KLONDIKE—8 SAMPLES ¹														
1968	DDE				230			210				45		
	TDE				180			130				37		
	DDT				35			34				12		
1969	DDE	120			18			170			73	22		
	TDE	100			94			110			53	18		
	DDT	—			44			23			62	—		
	Dieldrin	—			14			—			—	—		
STATION 3.—WHITE HOUSE—7 SAMPLES ¹														
1968	DDE				320			120				32		
	TDE				240			57				20		
	DDT				56			11				—		
	Dieldrin				20			—				—		
1969	DDE	110			15			56			46			
	TDE	83			98			37			40			
	DDT	—			36			—			36			
	Dieldrin	—			21			—			—			
STATION 4.—CEDAR POINT—8 SAMPLES ¹														
1968	DDE				180			86				41		
	TDE				160			51				23		
	DDT				32			17				23		
1969	DDE	84			77			110			26	30		
	TDE	55			71			78			22	23		
	DDT	—			30			—			26	T		
	Dieldrin	—			13			—			—	T		

¹ Each sample represents 15 or more mature mollusks.

SECTION B.—CALIFORNIA

The monthly collection of mollusks to monitor pesticide pollution in 12 estuaries in California was initiated in January 1966. Some of these stations were terminated and other estuaries were added during the course of the program. Samples were analyzed at the Gulf Breeze Laboratory until May 1968; from then until May 1970 they were analyzed at the Marine Resources Operations Laboratory of the Department of Fish and Game, Menlo Park, Calif. During the period July 1970 - June 1972, samples were collected and analyzed at approximately 3-month intervals by the Department of Fish and Game, Pesticides Investigations at Sacramento, Calif.

Six different mollusks (*Crassostrea gigas*, *Corbicula fluminea*, *Modiolus denissus*, *Mytilus californianus*, *Mytilus edulis*, and *Ostrea lurida*) were utilized for monitoring; for the most part, a single species was collected at each station. The relative ability of these different mollusks to store organochlorine residues appears to be reasonably similar and, thus, comparisons of the magnitude of residues in different estuaries can be made with some confidence. In general, residue levels at different stations followed patterns of suspected pollution loading in the associated drainage basin, regardless of the species monitored.

The approximate station locations are shown in Fig. B-1. A summary of data on organochlorine residues in the monitored species is presented in Table B-1, and the distribution of residues in these species for each sampling station by date of collection in Table B-2. Results of some of the analyses conducted by the Gulf Breeze Laboratory during the period January 1966 - December 1967 have been published by the cooperating agency (13).

DDT residues in mollusks were consistently larger in California than in any other area monitored with the exception of a single station in south Florida. There is a clear pattern of maximum pesticide residues being correlated with proximity of the monitoring station to runoff from agricultural lands. In southern California, where most samples contained typically large residues, residues were consistently higher at Hedionda and Mugu Lagoons, the recipients of agricultural runoff waters, than at Anaheim Slough which receives intermittent runoff from the urban and industrialized sections of Los Angeles. Residues in samples from estuaries draining the intensely cultivated central and southern parts of the State were larger, by one order of magnitude usually, than those in samples collected from watersheds north of San Francisco Bay where dairy land predominates.

The incidence of dieldrin residues (25%) was second only to New York samples although residues were lower in magnitude than in five other States. California and Texas were the only States where endrin and toxaphene

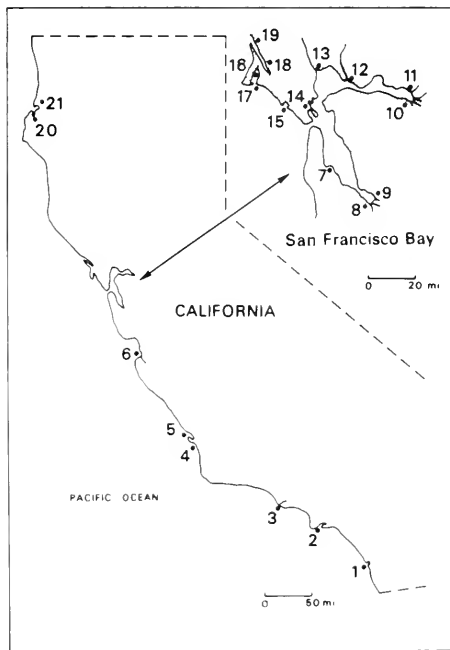


FIGURE B-1.—Diagram of coastal California and the San Francisco Bay area showing approximate location of monitoring stations

1. Hedionda Lagoon
2. Anaheim Slough
3. Point Mugu
4. Baywood Park—Morro Bay
5. Los Osos Creek—Morro Bay
6. Elkhorn Slough
7. Coyote Point—San Francisco Bay, South
8. Guadalupe Slough—San Francisco Bay, South
9. Alviso Slough—San Francisco Bay, South
10. West Island—Sacramento-San Joaquin River Basin
11. False River—Sacramento-San Joaquin River Basin
12. Napa River—San Pablo Bay
13. Petaluma River—San Pablo Bay
14. Point San Quentin—San Francisco Bay, North
15. Bolinas Lagoon
16. Schooner Bay—Drakes Estero
17. Berries Bay—Drakes Estero
18. Tomales Bay—Tomales Bay
19. Nicks Cove—Tomales Bay
20. Gunther Island—Humboldt Bay
21. Bird Island—Humboldt Bay

from presumably agricultural sources were detected. Polychlorinated biphenyl compounds were detected in samples beginning in 1971, but were not quantified. They occurred in a few samples from nearly all drainage basins monitored.

Late in 1970 or early 1971, there was a sharp decline in DDT residues in samples collected in estuaries draining predominantly agricultural areas, i.e., San Francisco Bay and the southern parts of the State. Decreased frequency

of sample collection in 1970-71 makes it impossible to pinpoint when this decline in DDT pollution occurred. The typically small DDT residues in samples

from drainage basins north of San Francisco Bay remained about the same throughout the monitoring period.

TABLE B-1.—Summary of data on organochlorine residues in the monitored species, 1966-72—California

STATION NUMBER	LOCATION	MONITORING PERIOD	PRINCIPAL MONITORED SPECIES	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (μG/KG)				
					DDT	DIELDRIN	ENDRIN	TOXAPHENE	PCB'S ²
1	Hedionda Lagoon	1967-72	<i>M. edulis</i>	31	31 (3,970)	4 (T)		2 (11,000)	2
2	Anaheim Slough	1967-72	<i>M. edulis</i>	33	33 (833)	10 (31)	1 (T)		2
3	Point Mugu	1967-72	<i>M. edulis</i>	29	29 (1,758)	9 (16)	1 (T)		2
4	Baywood Park	1966-72	<i>C. gigas</i>	52	52 (601)	3 (24)			
5	Los Osos Creek	1966-72	<i>C. gigas</i>	52	52 (412)	4 (27)			1
6	Elkhorn Slough	1966-72	<i>C. gigas</i>	57	57 (2,305)	24 (57)	2 (19)		2
7	Coyote Point	1966-72	<i>O. lurida</i>	55	54 (362)	26 (43)	2 (19)		1
8	Guadalupe Slough	1968-72	<i>M. demissus</i>	27	25 (407)	9 (37)			1
9	Alviso Slough	1968-72	<i>M. demissus</i>	28	28 (328)	6 (25)			1
10	West Island	1967-72	<i>C. fluminea</i>	28	28 (2,280)	23 (22)	3 (T)		1
11	False River	1967-71	<i>C. fluminea</i>	26	26 (1,850)	12 (24)	1 (18)		
12	Napa River	1968-72	<i>M. demissus</i>	28	26 (210)	5 (T)	2 (T)		1
13	Petaluma River	1968-72	<i>M. demissus</i>	28	25 (268)	4 (10)			1
14	Point San Quentin	1966-70	<i>C. gigas</i>	50	49 (440)	22 (23)			
15	Bolinas Lagoon	1966-68	<i>C. gigas</i>	17	14 (45)				
16	Schooner Bay	1966-72	<i>C. gigas</i>	33	25 (43)	2 (T)			1
17	Berries Bar	1966-68	<i>C. gigas</i>	27	25 (44)		1 (19)		
18	Tomales Bay	1966-72	<i>C. gigas</i>	34	28 (45)	2 (T)	1 (T)		
19	Nicks Cove	1966-68	<i>C. gigas</i>	25	20 (37)				
20	Gunther Island	1966-72	<i>C. gigas</i>	33	31 (78)	5 (T)			1
21	Bird Island	1966-68	<i>C. gigas</i>	25	3 (T)				
	Occasional stations (15)	1966-72	Mixed	54	51 (1,144)	25 (26)		2 (1,000)	4
Total number of samples				772					
Percent of samples positive for indicated compound					92	25	2	<1	3

NOTE: T = >5 but <10 ppb.

¹ Each sample represents 15 or more mature mollusks.

² Present but not quantified.

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California

[Blank = no sample collected; -- = no residue detected above 5 ppb or no residue detected (PCB's); T = >5 but <10 ppb]

YEAR	COMPOUND	RESIDUES IN PPB (μG/KG)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—HEDIONDA LAGOON— <i>M. EDULIS</i> UNLESS OTHERWISE INDICATED—31 SAMPLES ¹													
1967	DDE										100	2 130	2 90
	TDE										72	240	84
	DDT										130	3,600	740
	Toxaphene										--	11,000	970

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—HEDIONDA LAGOON— <i>M. EDULIS</i> , UNLESS OTHERWISE INDICATED—31 SAMPLES ¹ —Continued													
1968	DDE	130	52	200	210	91	130	120	168	^a 105	120	136	
	TDE	73	31	88	220	103	154	80	171	74	73	58	
	DDT	920	200	440	300	42	86	59	129	63	120	164	
1969	DDE	52	211	242	118	227	95		139	347	466	76	
	TDE	—	207	101	172	124	53		35	99	115	64	
	DDT	123	291	486	214	99	91		34	61	68	108	
1970	DDE												114
	TDE												102
	DDT												54
	Dieldrin												T
1971	DDE		19				36		54				18
	TDE		11				58		56				13
	DDT		16				T		285				10
	Dieldrin		T				—		—				—
	PCB's		—				—		—				—
1972	DDE	14				31							
	TDE	T				31							
	DDT	10				10							
	Dieldrin	T				T							
	PCB's	—				—							
STATION 2.—ANAHEIM SLOUGH— <i>M. EDULIS</i> , UNLESS OTHERWISE INDICATED—33 SAMPLES ¹													
1967	DDE										360	330	200
	TDE										100	150	87
	DDT										85	120	120
1968	DDE	270	^a 110	^b 170	310	464	203	265	432	^c 464	440	354	
	TDE	91	45	62	110	186	102	68	109	127	170	118	
	DDT	160	43	110	77	108	52	33	51	65	110	70	
	Dieldrin	—	31	—	—	—	T	T	—	—	12	—	
	Endrin	—	—	—	—	—	—	T	—	—	—	—	
1969	DDE	157	273	127	51	388	547	323	466	451	168	494	
	TDE	37	55	—	136	172	189	107	115	107	129	130	
	DDT	123	217	94	222	131	97	37	60	64	282	88	
1970	DDE		157										305
	TDE		49										126
	DDT		38										10

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G/KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 2.—ANAHEIM SLOUGH— <i>M. EDULIS</i> . UNLESS OTHERWISE INDICATED—33 SAMPLES ¹ —Continued													
1971	DDE		75			103			185			92	
	TDE		53			164			101			41	
	DDT		23			T			22			10	
	Dieldrin		T			T			T			T	
	PCB's		—			—			—			(⁶)	
1972	DDE	64				80							
	TDE	24				53							
	DDT	18				10							
	Dieldrin	T				T							
	PCB's	—				(⁶)							
STATION 3.—POINT MUGU— <i>M. EDULIS</i> . UNLESS OTHERWISE INDICATED—29 SAMPLES ¹													
1967	DDE										130	160	220
	TDE										150	230	280
	DDT										270	440	650
	Dieldrin										—	—	T
1968	DDE	250	200	370	170	366	207	255	465	269		360	
	TDE	230	210	350	180	494	168	65	388	278		443	
	DDT	460	340	790	430	749	363	32	432	566		955	
	Dieldrin	—	—	16	—	—	—	T	—	—		T	
	Endrin	—	—	—	—	—	—	—	—	—		T	
1969	DDE	226		560	334	365	273	298		918	349	112	
	TDE	121		—	301	63	31	116		117	34	146	
	DDT	161		391	245	120	92	40		580	176	185	
1970	DDE											238	
	TDE											141	
	DDT											56	
1971	DDE		49				65		22			112	
	TDE		24				73		T			50	
	DDT		45				11		—			20	
	Dieldrin		T				T		—			T	
	PCB's		—				—		—			(⁶)	
1972	DDE	11				24							
	TDE	12				10							
	DDT	T				10							
	Dieldrin	T				T							
	PCB's	—				(⁶)							

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California—Continued

YEAR	COMPOUND	RESIDUES IN PPB (μ G KG)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 4.—BAYWOOD PARK—C. GIGAS—52 SAMPLES ¹													
1966	DDE	54	74	82	75	76	52	55	62	59	69	69	100
	TDE	26	34	35	25	32	19	22	25	35	34	33	37
	DDT	25	25	26	20	24	14	—	18	26	24	25	46
	Dieldrin	—	—	—	24	16	—	—	—	—	—	—	—
1967	DDE	110	110	62	130	80	120	51	82	55	48	35	46
	TDE	29	42	50	47	34	49	29	40	23	21	10	13
	DDT	58	73	96	130	67	70	37	49	46	26	15	10
1968	DDE	96	43	40	160	48	49	48	48	44	—	74	—
	TDE	24	13	T	40	17	19	—	13	—	—	—	—
	DDT	25	13	30	61	—	T	T	T	—	—	—	—
1969	DDE	123	111	139	180	148	119	110	—	97	165	184	162
	TDE	23	38	—	70	57	40	57	—	31	53	70	75
	DDT	24	164	131	351	189	150	43	—	31	58	64	69
1970	DDE	—	220	226	215	56	—	—	—	—	—	—	—
	TDE	—	74	87	58	21	—	—	—	—	—	—	—
	DDT	—	69	64	46	—	—	—	—	—	—	—	—
1971	DDE	—	—	—	—	—	—	—	22	—	—	21	—
	TDE	—	—	—	—	—	—	—	11	—	—	16	—
	DDT	—	—	—	—	—	—	—	—	—	—	10	—
1972	DDE	33	—	—	—	—	—	—	—	—	—	—	—
	TDE	12	—	—	—	—	—	—	—	—	—	—	—
	DDT	T	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	T	—	—	—	—	—	—	—	—	—	—	—
STATION 5.—LOS OSOS CREEK—C. GIGAS, UNLESS OTHERWISE INDICATED—52 SAMPLES ¹													
1966	DDE	83	58	43	88	65	40	43	53	73	10	71	72
	TDE	33	27	17	39	25	16	16	22	34	27	33	31
	DDT	23	21	14	30	20	—	—	14	23	23	25	37
	Dieldrin	—	—	—	27	10	—	—	—	—	—	—	—
1967	DDE	62	120	63	110	93	130	64	81	56	43	37	29
	TDE	29	47	43	42	57	56	46	44	33	20	14	10
	DDT	41	96	130	120	92	80	52	49	72	25	20	12
1968	DDE	100	61	42	70	65	42	31	25	55	—	69	—
	TDE	32	21	13	24	T	T	—	11	T	—	—	—
	DDT	37	21	T	36	T	—	T	—	—	—	—	—
1969	DDE	66	70	126	104	155	144	201	—	115	223	—	137
	TDE	T	37	—	56	61	80	83	—	43	72	—	51
	DDT	T	72	131	239	183	188	128	—	34	93	—	35

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California—Continued

YEAR	COMPOUND	RESIDUES IN PPB (µG/KG)											
		JAN	FEB	MAR	APR	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 5—LOS OSOS CREEK— <i>C. GIGAS</i> . UNLESS OTHERWISE INDICATED—52 SAMPLES ¹ —Continued													
1970	DDE		186	182	221	63							
	TDE		60	54	70	13							
	DDT		53	50	41	—							
1971	DDE								29				25
	TDE								15				21
	DDT								T				10
1972	DDE	30				12							
	TDE	12				T							
	DDT	T				10							
	Dieldrin	T				T							
	PCB's	—				6							
STATION 6—ELKHORN SLOUGH— <i>C. GIGAS</i> . UNLESS OTHERWISE INDICATED—57 SAMPLES ¹													
1966	DDE	160	220	96	96	89	88	86	72	79	84	130	190
	TDE	160	220	120	110	95	82	79	77	66	65	77	160
	DDT	250	290	110	96	85	65	64	55	41	56	76	210
	Dieldrin	—	19	11	20	18	—	—	—	—	10	—	30
1967	DDE	200	220	200	230	210	300	160	200	190	62	190	250
	TDE	160	230	200	260	340	390	200	260	210	55	150	230
	DDT	260	440	390	690	860	920	390	500	390	110	340	370
	Dieldrin	26	25	29	30	39	33	10	10	15	—	14	17
1968	DDE	260	130	120	170	214		173	122	168		95	65
	TDE	160	85	92	160	212		129	70	100		71	63
	DDT	250	97	61	230	411		237	159	200		113	110
	Dieldrin	13	—	—	—	—		27	—	—		—	—
	Endrin	—	—	—	—	—		19	—	—		—	—
1969	DDE	178	191	126	280	215	324	424			1,373	237	191
	TDE	102	338	156	393	223	253	358			502	171	117
	DDT	120	441	346	808	304	96	704			630	284	189
1970	DDE	208	230	445	270	353	325						31
	TDE	173	300	582	285	276	236						19
	DDT	204	444	808	491	411	375						26
	Dieldrin	—	—	—	—	—	—						T
	Endrin	—	—	—	—	—	—						T
1971	DDE		76			29			43				28
	TDE		37			24			71				37
	DDT		72			10			—				17
	Dieldrin		—			T			—				11
	PCB's		—			—			—				(4)

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California—Continued

YEAR	COMPOUND	RESIDUES IN PPB (20 KG)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 6.—ELKHORN SLOUGH— <i>C. GIGAS</i> . UNLESS OTHERWISE INDICATED—5" SAMPLES—Continued													
1972	DDE		42			T							
	TDE		25			T							
	DDT		36			10							
	Dieldrin		T			5"							
	PCB's		—			4							
STATION 7.—COYOTE POINT— <i>O. LURIDA</i> —55 SAMPLES													
1966	DDE	54	93	42	71	71	47	30	35	50	42	33	53
	TDE	74	120	54	88	91	66	33	46	65	71	51	79
	DDT	63	100	43	70	74	46	39	42	55	62	40	75
	Dieldrin	—	27	20	29	23	—	—	—	—	21	15	21
1967	DDE	41	49	51	61	65	52	51	47	39	33	46	46
	TDE	60	68	74	82	76	78	84	54	58	37	86	84
	DDT	53	72	79	89	69	70	80	50	50	43	110	51
	Dieldrin	26	26	28	23	21	25	17	43	13	—	16	16
	Endrin	—	—	—	—	—	19	—	—	—	—	—	—
1968	DDE	44	59	47	47	46	27	42	—	30		33	
	TDE	57	95	69	77	103	41	57	—	57		69	
	DDT	53	100	84	89	103	45	60	—	56		58	
	Dieldrin	13	—	18	20	—	—	10	—	—		—	
	Endrin	—	—	—	10	—	—	—	—	—		—	
1969	DDE	51	52	172	25	—	65	34	153	36		52	24
	TDE	46	—	—	—	89	57	33	96	T		78	50
	DDT	48	158	171	—	176	88	33	100	39		59	26
1970	DDE	18	33	102	93	37	33						
	TDE	33	55	102	100	—	50						
	DDT	33	42	87	76	42	24						
1971	DDE		17									14	
	TDE		67									24	
	DDT		47									24	
	Dieldrin		T									T	
1972	DDE		15			T							
	TDE		32			T							
	DDT		53			T							
	Dieldrin		T			T							
	PCB's		—			4							

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 8.—GUADALUPE SLOUGH— <i>M. DEMISSUS</i> —27 SAMPLES ¹													
1968	DDE		36	77	48	74	67	34	24	19	24	34	T
	TDE		90	180	100	185	140	68	53	57	58	T	24
	DDT		110	150	60	91	130	34	T	40	26	—	T
	Dieldrin		18	23	14	—	14	—	—	—	—	—	—
1969	DDE	—	—	34	70	26	11	10	24	—	29		
	TDE	42	—	—	—	27	108	22	34	48	50		
	DDT	—	—	—	—	—	204	T	T	T	T		
1970	No Samples Collected												
1971	DDE		T			T			—				T
	TDE		40			26			—				T
	DDT		28			10			—				10
	Dieldrin		T			T			—				37
1972	DDE		11			T							
	TDE		12			T							
	DDT		10			10							
	Dieldrin		T			T							
	PCB's		—			(⁶)							
STATION 9.—ALVISO SLOUGH— <i>M. DEMISSUS</i> —28 SAMPLES ¹													
1968	DDE		43	46	69	74	47	26	28	18	11	—	13
	TDE		140	59	170	169	95	72	80	45	79	T	30
	DDT		93	78	77	85	66	35	34	35	11	T	27
	Dieldrin		18	12	25	—	—	—	—	—	—	—	—
1969	DDE	38	—	55	98	52	12	17	30	—	39		
	TDE	59	61	55	—	73	161	42	45	56	27		
	DDT	—	—	88	—	108	111	T	20	T	T		
1970	DDE		42										
	TDE		76										
	DDT		33										
1971	DDE		13			T			—				T
	TDE		38			15			T				T
	DDT		17			T			—				10
	Dieldrin		—			T			—				—
1972	DDE	T				T							
	TDE	T				T							
	DDT	10				10							
	Dieldrin	T				T							
	PCB's	—				(⁶)							

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 10.—WEST ISLAND— <i>C. FLUMINEA</i> —28 SAMPLES ¹													
1967	DDE	280	330	320	270	320	230	170	140	170	180	690	390
	TDE	250	370	350	250	250	210	130	93	150	150	490	310
	DDT	210	300	310	250	260	270	150	130	230	270	1,100	770
	Dieldrin	20	20	22	17	12	18	T	15	20	10	20	18
	Endrin	T	T	T	—	—	—	—	—	—	—	—	—
1968	DDE	390	500	280	370	251	196	134	104	41		71	
	TDE	290	400	200	220	224	183	160	182	97		138	
	DDT	240	290	190	210	320	223	150	235	150		184	
	Dieldrin	16	22	15	16	—	21	19	13	—		—	
1969	DDE			177									
	TDE			—									
	DDT			168									
1970						No Samples Collected							
1971	DDE					198			91			15	
	TDE					126			71			T	
	DDT					173			111			—	
	Dieldrin					T			T			T	
1972	DDE		11			T							
	TDE		10			T							
	DDT		10			10							
	Dieldrin		—			T							
	PCB's		—			(^a)							
STATION 11.—FALSE RIVER— <i>C. FLUMINEA</i> —26 SAMPLES ¹													
1967	DDE				470	460	320	420	270				400
	TDE				410	320	200	260	180				350
	DDT				970	910	640	780	500				210
	Dieldrin				24	19	20	16	16				17
1968	DDE	470	500	340	330	315	199	250	122	53		96	
	TDE	400	590	230	230	281	167	190	212	109		144	
	DDT	220	420	200	190	312	225	290	296	92		103	
	Dieldrin	17	22	19	23	—	—	—	16	—		—	
	Endrin	18	—	—	—	—	—	—	—	—		—	
1969	DDE		151	54		93	41	88	139	57	42	—	
	TDE		152	66		75	47	88	165	91	76	91	
	DDT		378	167		214	46	135	136	61	43	44	
1970						No Samples Collected							

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 11.—FALSE RIVER— <i>C. FLUMINEA</i> —26 SAMPLES ¹ —Continued													
1971	DDE		21										
	TDE		41										
	DDT		20										
	Dieldrin		T										
STATION 12.—NAPA RIVER— <i>M. DEMISSUS</i> —28 SAMPLES ¹													
1968	DDE		T	25	21	24	23	15	21	18		T	
	TDE		22	46	72	100	83	38	42	62		24	
	DDT		T	26	39	45	47	—	23	18		T	
1969	DDE	10	—	100	62	—	10	13	T	—	T	12	
	TDE	30	—	48	—	—	24	45	30	58	24	37	
	DDT	T	—	—	—	—	T	T	T	—	T	T	
1970	DDE		11									16	
	TDE		33									93	
	DDT		T									46	
	Dieldrin		—									T	
	Endrin		—									T	
1971	DDE		27			13			T				T
	TDE		143			68			T				T
	DDT		41			10			T				T
	Dieldrin		T			T			—				—
	Endrin		T			—			—				—
1972	DDE		T			T							
	TDE		21			T							
	DDT		23			10							
	Dieldrin		T			T							
	PCB's		—			10							
STATION 13.—PETALUMA RIVER— <i>M. DEMISSUS</i> —28 SAMPLES ¹													
1968	DDE		—	27	27	26	47	19	—	—		92	
	TDE		—	58	63	72	104	35	T	13		68	
	DDT		—	15	19	31	41	T	—	—		108	
1969	DDE	T	—	124	49	12	T	17	—	—	22	10	
	TDE	T	—	37	—	28	T	38	—	57	37	22	
	DDT	—	—	—	—	—	T	T	—	—	T	T	
1970	DDE		T									28	
	TDE		T									71	
	DDT		T									26	
	Dieldrin		—									10	

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 13—PETALUMA RIVER—M. DEMISSUS ¹ —28 SAMPLES ¹ —Continued													
1971	DDE		T			T			—				T
	TDE		18			24			T				T
	DDT		10			T			—				—
	Dieldrin		T			—			—				—
1972	DDE		T			T							
	TDE		T			T							
	DDT		10			10							
	Dieldrin		T			T							
	PCB's		—			(4)							
STATION 14.—POINT SAN QUENTIN—C. GIGAS—50 SAMPLES ¹													
1966	DDE	12	30	47	52	69	59	37	52	57	51	55	55
	TDE	20	37	60	83	120	92	47	82	90	88	84	110
	DDT	14	12	19	23	45	38	24	43	49	33	40	98
	Dieldrin	—	—	—	14	20	—	—	—	—	11	15	20
1967	DDE	52	34	30	42	23	39	45	53	30	31	100	45
	TDE	130	65	59	75	55	85	120	130	74	68	50	84
	DDT	88	49	49	70	34	64	89	63	38	36	85	45
	Dieldrin	23	11	15	19	13	21	19	17	11	—	10	11
1968	DDE	43	44	43	43	36	59	59	25	38			40
	TDE	79	96	78	97	95	110	110	60	86			120
	DDT	44	89	67	63	69	100	100	T	52			82
	Dieldrin	12	17	17	12	—	—	12	—	—			18
1969	DDE	74	53	62	47	143	25	13	30		—	—	31
	TDE	80	149	134	—	143	54	23	41		—	55	51
	DDT	130	193	76	182	154	24	T	T		—	32	26
1970	DDE	18	19	54	66	51							
	TDE	18	T	31	64	39							
	DDT	T	T	37	45	23							
STATION 15.—BOLINAS LAGOON—C. GIGAS—17 SAMPLES ¹													
1966	DDE									10	T	T	—
	TDE									—	T	10	—
	DDT									—	—	—	—
1967	DDE	T	T	10	10	11	T	11	13	—	T	T	T
	TDE	11	13	16	17	16	14	21	20	—	15	11	T
	DDT	T	T	T	11	14	12	13	11	—	11	—	—

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 15.—BOLINAS LAGOON—C. GIGAS—17 SAMPLES ¹ —Continued													
1968	DDE	—											
	TDE	—											
	DDT	—											
STATION 16.—SCHOONER BAY—C. GIGAS—33 SAMPLES ¹													
1966	DDE	—	T	T	—	T	T	T	T	—	T	T	—
	TDE	—	—	T	—	T	—	—	—	—	T	T	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	T	T	10	11	T	11	T	15	—	—	T	—
	TDE	—	—	T	13	13	13	T	18	—	—	—	—
	DDT	—	—	—	10	—	T	—	10	—	—	—	—
1968	DDE	—											
	TDE	—											
	DDT	—											
1969						No Samples Collected							
1970	DDE		11									10	
	TDE		T									11	
	DDT		T									10	
1971	DDE		14			T			T				T
	TDE		16			T			T				T
	DDT		10			T			T				10
	Dieldrin		T			—			—				—
1972	DDE		T			T							
	TDE		T			T							
	DDT		10			10							
	Dieldrin		—			T							
	PCB's		—			(4)							
STATION 17.—BERRIES BAR—C. GIGAS—27 SAMPLES ¹													
1966	DDE	—	13	T	10	13	10	17	11	12	T	T	11
	TDE	—	15	T	17	16	10	13	11	T	T	T	10
	DDT	—	T	—	—	T	—	—	—	—	—	—	—
1967	DDE	T	11	17	14	16	12	13	T	13	15	14	T
	TDE	T	14	20	18	17	14	15	12	16	18	17	T
	DDT	—	—	T	10	11	10	T	—	10	T	T	—

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 17.—BERRIES BAR—C. GIGAS—27 SAMPLES ¹ —Continued													
1968	DDE	—			12				17				
	TDE	—			13				14				
	DDT	—			T				—				
	Endrin	19			—				—				
STATION 18.—TOMALES BAY—C. GIGAS—34 SAMPLES ¹													
1966	DDE	T	T	—	—	T	—	14	11	14	14	T	T
	TDE	—	T	—	—	11	—	T	T	11	12	—	—
	DDT	—	—	—	—	T	—	—	—	—	—	—	—
1967	DDE	T	12	11	11	T	11	T	T	T	—	11	—
	TDE	—	T	T	12	T	14	T	—	—	—	T	—
	DDT	T	10	T	11	T	13	T	—	—	—	T	—
1968	DDE	—			11				T				
	TDE	—			—				T				
	DDT	—			10				—				
1969	DDE									22		T	
	TDE									T		T	
	DDT									18		T	
1970	DDE												T
	TDE												T
	DDT												10
	Dieldrin												T
	Endrin												T
1971	DDE		12			T							T
	TDE		T			T							T
	DDT		10			10							10
1972	DDE		T										
	TDE		T										
	DDT		10										
	Dieldrin		T										
STATION 19.—NICKS COVE—C. GIGAS—25 SAMPLES ¹													
1966	DDE	—	12	11	—	T	T	11	11	T	T	T	T
	TDE	—	T	—	—	—	—	—	T	—	—	T	—
	DDT	—	T	—	—	—	—	—	—	—	—	—	—
1967	DDE	12	13	13	14	10	T	11	T	T	—	14	—
	TDE	T	10	12	12	T	T	T	—	T	—	—	—
	DDT	T	11	T	11	T	T	T	—	—	—	—	—

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	Nov.	DEC.
STATION 19.—NICKS COVE—C. GIGAS—25 SAMPLES ¹ —Continued													
1968	DDE	—											
	TDE	—											
	DDT	—											
STATION 20.—GUNTHER ISLAND—C. GIGAS—33 SAMPLES ¹													
1966	DDE	—	—	—	—	—	T	11	T	T	T	T	T
	TDE	—	—	—	—	—	T	17	—	21	T	T	T
	DDT	T	10	—	47	11	14	—	11	—	18	14	20
1967	DDE	10	T	T	T	T	T	T	T	—	T	T	T
	TDE	T	—	—	T	—	T	T	—	T	T	T	T
	DDT	30	28	12	19	19	19	24	12	16	15	21	22
1968	DDE	—			13								
	TDE	—			11								
	DDT	—			54								
1969		No Samples Collected											
1970	DDE												11
	TDE												11
	DDT												10
	Dieldrin												T
1971	DDE		T			T			T				T
	TDE		13			T			—				T
	DDT		17			10			—				10
	Dieldrin		T			T			—				T
1972	DDE			T			T						
	TDE			T			T						
	DDT			10			10						
	Dieldrin			—			T						
	PCB's			—			(4)						
STATION 21.—BIRD ISLAND—C. GIGAS—25 SAMPLES ¹													
1966	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	T	—	—	T	—	T

TABLE B-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—California—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 21—BIRD ISLAND— <i>C. GIGAS</i> —25 SAMPLES ¹ —Continued													
1968	DDE	—											
	TDE	—											
	DDT	—											

¹ Each sample represents 15 or more mature mollusks

² DDE, TDE, and DDT values approximated because of presence of toxaphene.

³ *C. gigas*.

⁴ Present but not quantified.

⁵ *M. demissus*.

⁶ *M. californianus*.

⁷ *M. edulis*.

SECTION C.—DELAWARE

Samples were collected at nine stations at monthly intervals during the period October 1966 - August 1969. The eastern oyster (*Crassostrea virginica*), ribbed mussel (*Modiolus demissus*), and hard clam (*Mercenaria mercenaria*) were each collected at three stations. All samples were analyzed at the Gulf Breeze Laboratory. The approximate locations of the stations are shown in Fig. C-1. The Cape Henlopen station was in Delaware Bay; the other stations were adjacent to the Bay but exposed primarily to the runoff from large agricultural areas in separate drainage basins. A summary of data on organochlorine residues in the monitored species is presented in Table C-1, and the distribution of residues in these species for each sampling station by date of collection in Table C-2.

The use of three different species for monitoring obscured pollution patterns in Delaware estuaries to some extent. The relative inefficiency of hard clams in storing organochlorine residues makes Rehoboth Bay (Stations 7 and 8) appear to be generally free from this type of pollution. The first samples of clams collected in adjacent Indian River Bay (Station 9) also were free of detectable residues; however, subsequent monitoring, using the ribbed mussel, showed Indian River Bay to be moderately but continuously polluted. It is probable that Rehoboth Bay was similarly polluted during the monitoring period. This same reasoning suggests that the waters at Cape Henlopen were continually more polluted with DDT than the small residues in the hard clams would imply.

The magnitude of DDT residues in clams and oysters showed no trend towards increased or decreased levels during the 3-year monitoring period. In ribbed mussels, however, there was a marked decline in the average level

of residues in the final year at Stations 1 and 2 as well as Station 9. Delaware monitoring samples ranked 6th in frequency and 10th in magnitude of DDT residues in comparison with the other 14 States. The 13% incidence of dieldrin residues was about the average for all States.

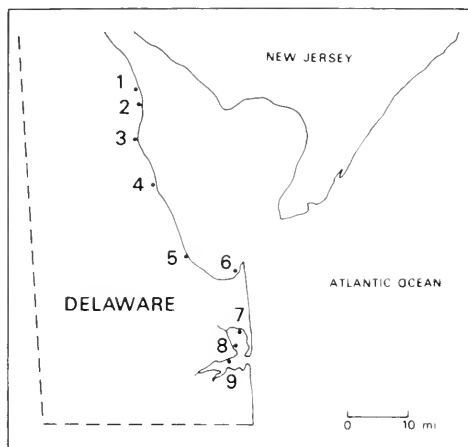


FIGURE C-1.—Diagram of coastal Delaware showing approximate location of monitoring stations

1. Leipsic River
2. Simons River
3. Bowers Beach—Murderkill River
4. Mispillion River
5. Broadkill River
6. Cape Henlopen—Delaware Bay
7. Thompson Island—Rehoboth Bay
8. Arrowhead Point—Rehoboth Bay
9. West Gables—Indian River Bay

TABLE C-1.—Summary of data on organochlorine residues in the monitored species, 1966-69—Delaware

STATION NUMBER	LOCATION	MONITORING PERIOD	PRINCIPAL MONITORED SPECIES	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (µg/kg)	
					DDT	Dieldrin
1	Leipsic River	1967-69	M. demissus	27	23 (156)	4 (13)
2	Simons River	1967-69	M. demissus	25	23 (205)	6 (19)
3	Bowers Beach	1966-69	C. virginica	34	34 (172)	25 (25)
4	Mispillion River	1966-69	C. virginica	35	33 (90)	2 (10)
5	Broadkill River	1966-69	C. virginica	34	34 (90)	
6	Cape Henlopen	1966-69	M. mercenaria	32	30 (65)	
7	Thompson Island	1966-69	M. mercenaria	33	5 (16)	
8	Arrowhead Point	1966-69	M. mercenaria	34	4 (35)	
9	West Gables	1966-69	M. demissus	33	30 (96)	
Total number of samples				287		
Percent positive for indicated compound					75	13

¹ Each sample represents 15 or more mature mollusks.

TABLE C-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—Delaware

[Blank = no sample collected; — = no residue detected above 5 ppb; T = >5 but <10 ppb]

YEAR	COMPOUND	RESIDUES IN PPB (µg/kg)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—LEIPSIC RIVER—M. DEMISSUS—27 SAMPLES ¹													
1967	DDE			12	25	26	47	33	21	17	22	18	32
	TDE			11	53	46	91	77	51	47	29	47	77
	DDT			—	T	12	18	23	51	47	—	T	17
	Dieldrin			—	10	13	—	10	—	10	—	—	—
1968	DDE			27	30	32	26	33	T	12	23	19	
	TDE			68	69	91	41	45	20	18	29	37	
	DDT			17	17	14	—	22	—	—	T	18	
1969	DDE	—		T	15	T	—	17	—				
	TDE	—		14	18	T	—	33	—				
	DDT			—	—	—	—	19	—				
STATION 2.—SIMONS RIVER—M. DEMISSUS—25 SAMPLES ¹													
1967	DDE	13		17	31	31	43	37	25		18	29	29
	TDE	43		37	65	150	89	79	47		88	75	66
	DDT			—	15	24	19	28	38		29	18	16
	Dieldrin			—	15	19	12	T	—		12	—	—
1968	DDE			28	23	39			23	—	20	11	
	TDE			65	78	100			30	—	26	16	
	DDT			18	39	16			—	—	22	—	
	Dieldrin			—	—	10			—	—	—	—	

TABLE C-2—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—Delaware—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G KG}$)												
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.	
STATION 2.—SIMONS RIVER— <i>M. DEMISSUS</i> —25 SAMPLES ¹ —Continued														
1969	DDE	—	13	T	21	14	21	13	21					
	TDE	—	22	T	23	19	29	26	35					
	DDT	—	—	—	—	—	24	26	31					
STATION 3.—BOWERS BEACH— <i>C. VIRGINICA</i> —34 SAMPLES ¹														
1966	DDE											25	19	
	TDE											29	17	
	DDT											—	—	
	Dieldrin											16	—	
1967	DDE	27	25	⁽²⁾	⁽²⁾	42	43	41	40	40	42	41	32	
	TDE	35	26	⁽²⁾	⁽²⁾	65	70	66	64	56	98	51	38	
	DDT	—	—	⁽²⁾	⁽²⁾	T	10	24	68	17	25	14	10	
	Dieldrin	18	14	20	25	24	18	11	13	16	16	13	15	
1968	DDE	50	46	48	52	66	75	82	49	41	52	57	41	
	TDE	56	42	47	48	73	78	53	34	32	44	57	35	
	DDT	T	—	T	—	10	T	25	T	T	13	18	T	
	Dieldrin	12	13	14	15	12	16	—	—	—	—	16	15	
1969	DDE	52	48	42	49	41	79	60	39					
	TDE	37	47	36	39	39	70	57	29					
	DDT	—	—	—	—	—	20	29	11					
	Dieldrin	14	15	11	—	11	—	—	—					
STATION 4.—MISPILLION RIVER— <i>C. VIRGINICA</i> —35 SAMPLES ¹														
1966	DDE											31	21	22
	TDE											27	24	24
	DDT											15	—	—
1967	DDE	17	24	⁽²⁾	25	35	23	22	16	20	T	32	27	
	TDE	18	22	⁽²⁾	31	41	26	30	20	23	44	36	32	
	DDT	—	—	⁽²⁾	—	—	—	T	11	T	T	—	T	
	Dieldrin	—	—	10	—	10	—	—	—	—	—	—	—	
1968	DDE	23	28	Lost	41	39	39	47	34	32	25	39	38	
	TDE	40	31	—	40	36	34	33	23	25	18	32	27	
	DDT	T	—	—	—	15	—	—	—	—	—	—	—	
1969	DDE	40	50	36	29	36	26	35	28					
	TDE	29	31	28	25	32	26	22	20					
	DDT	—	—	—	—	—	—	—	—					

TABLE C-2—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—*Delaware*—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	Nov.	DEC.
STATION 5.—BROADKILL RIVER— <i>C. VIRGINICA</i> —34 SAMPLES ¹													
1966	DDE										18	22	
	TDE										20	17	
	DDT										T	—	
1967	DDE	28	18	23	17	23	24	16	30	27	35	35	37
	TDE	23	11	17	13	18	21	19	30	27	27	31	33
	DDT	—	—	—	—	—	T	T	16	11	T	T	T
1968	DDE	25	29	23	32	39	43	36	48	37	31	44	51
	TDE	21	22	16	21	45	32	20	32	25	22	33	39
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1969	DDE	40	38	43	41	39	42	43	35				
	TDE	24	28	26	20	28	28	24	15				
	DDT	—	—	—	—	—	—	—	—				
STATION 6.—CAPE HENLOPEN— <i>M. MERCENARIA</i> —32 SAMPLES ¹													
1966	DDE												12
	TDE												11
	DDT												—
1967	DDE	12	13	—	(2)	14	12	20	12	T	14	16	16
	TDE	16	12	—	(2)	14	14	24	14	T	13	15	15
	DDT	—	—	—	(2)	—	—	—	—	—	—	—	—
1968	DDE	13	15	18	18	28	39	25	25	19	13	T	21
	TDE	12	14	14	15	24	26	16	15	12	T	T	11
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1969	DDE	12	14	22	Lost	16	19	22	35				
	TDE	—	—	T		11	T	10	20				
	DDT	—	—	—		—	—	—	—				
STATION 7.—THOMPSON ISLAND— <i>M. MERCENARIA</i> —33 SAMPLES ¹													
1966	DDE												T
	TDE											T	11
	DDT											T	—
1967	DDE	T	—	—	T	T	—	—	—	—	—	—	—
	TDE	T	—	—	—	T	—	—	—	—	—	—	—
	DDT	—	—	—	—	T	—	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

TABLE C-2—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—Delaware—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G. KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 7.—THOMPSON ISLAND— <i>M. MERCENARIA</i> —33 SAMPLES ¹ —Continued													
1969	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 8.—ARROWHEAD POINT— <i>M. MERCENARIA</i> —34 SAMPLES ¹													
1966	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	—	(2)	1	—	—	—	—	—	—	—	—
	TDE	—	—	(2)	T	—	—	—	—	—	—	—	—
	DDT	—	—	(2)	—	—	—	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1969	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 9.—WEST GABLES— <i>M. DEMISSUS</i> UNLESS OTHERWISE INDICATED—33 SAMPLES ¹													
1966	DDE	—	—	—	—	—	—	—	—	—	—	(2)	(3)
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	11	—	18	17	22	19	15	18	13	19	19	21
	TDE	25	—	37	32	33	24	21	29	13	29	30	33
	DDT	32	—	19	14	24	13	14	41	21	21	22	26
1968	DDE	19	24	18	20	18	23	T	T	—	T	12	13
	TDE	29	35	28	32	30	37	11	T	—	T	16	18
	DDT	21	30	22	26	23	36	T	—	—	—	T	—
1969	DDE	10	16	13	16	17	21	13	11	—	—	—	—
	TDE	14	16	17	20	22	27	18	T	—	—	—	—
	DDT	—	—	—	T	10	T	11	—	—	—	—	—

¹ Each sample represents 15 or more mature mollusks.

² Present but not quantified.

³ *M. mercenaria*

SECTION D.—FLORIDA

Investigation of the effects of pesticide pollution on estuarine fauna in Florida was initiated at the Gulf Breeze Laboratory, Gulf Breeze, Fla., in 1959. During the next 5 years, sufficient headway in the understanding of uptake and flushing rates of persistent synthetic compounds as well as the technology for handling samples made a continuing monitoring program feasible. Local oysters (Station 9, East Bay) were analyzed monthly during 1964, and the concept of a national monitoring program was developed and implemented in 1965. The eastern oyster, *C. virginica*, was the only species monitored in Florida; all samples were analyzed at the Gulf Breeze Laboratory. The approximate locations of monitoring stations are shown in Fig. D-1. A summary of data on organochlorine residues in the monitored species, *C. virginica*, is presented in Table D-1, and the distribution of residues in this species for each sampling station by date of collection in Table D-2.

Oyster samples from Florida contained the highest levels of DDT residues and the most persistent contamination with PCB's observed in the entire monitoring program.

The polychlorinated biphenyl, Aroclor 1254[®], was identified in studies of estuarine fauna following a 1969 fish kill in Escambia Bay, Fla., (8). Station 9 is about 25 miles from the presumed source of this PCB pollution and is in a contiguous but distinct drainage basin. Monitoring samples from this station contained PCB residues about one-third the magnitude of residues in Escambia Bay oysters and continued to have residues of similar magnitude for at least 3 years after the presumed primary source of PCB's had been eliminated.

The trend in DDT residues is most clearly shown in the Station 9 data. Some DDT had been used in this geographic area for agricultural purposes. However, its primary use had been for the control of stable-fly larvae, *Stomoxys calcitrans*, that develop in seaweed windrows on estuarine beaches. In 1969, methoxychlor

was substituted for this purpose, and DDT residues virtually disappeared from all succeeding monitoring samples. Methoxychlor residues were not detected in the monitored samples. There are not enough recent data to determine DDT pollution trends in other estuaries along the Florida Gulf coast.

The incidence of DDT in Florida samples (62%) is about the average for all States monitored. The incidence of dieldrin (7%) may be compared with the average incidence of 15% for all States.

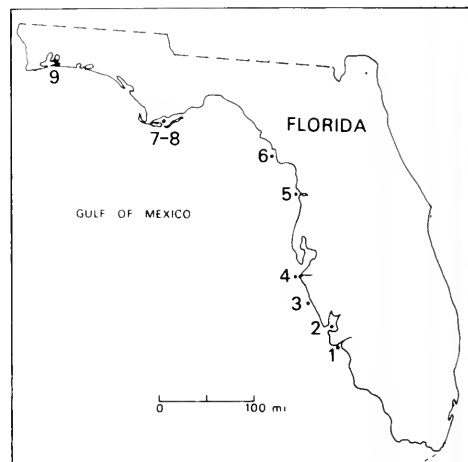


FIGURE D-1.—Diagram of coastal Florida showing approximate location of monitoring stations

1. Iona Point—Caloosahatchee River
2. Charlotte Harbor—Peace River
3. Coral Cove—Little Sarasota Bay
4. Manatee River
5. Crystal River
6. Suwanee River
7. St. Vincents Bar (North)—Apalachicola Bay
8. St. Vincents Bar (South)—Apalachicola Bay
9. East Bay—Blackwater River

TABLE D-1.—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1965-72—Florida

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (μg/kg)		
				DDT	DIELDRIN	PCB'S ²
1	Iona Point	1967-69	31	31 (5,390)	1 (11)	
2	Charlotte Harbor	1966-69	31	28 (338)	13 (27)	
3	Coral Cove	1966-69	32	32 (129)		
4	Manatee River	1966-69	32	32 (159)		
5	Crystal River	1966-71	43	7 (27)		
6	Suwanee River	1966-69	32	6 (22)		

TABLE D-1.—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1965-72—Florida—Continued

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (µG/KG)		
				DDT	Dieldrin	PCB's ²
7	St. Vincents Bar (North)	1966-67	17	12 (50)	3 (28)	
8	St. Vincents Bar (South)	1966-67	16	10 (70)	3 (22)	
9	East Bay	1965-72	84	46 (65)		25 (390)
	Occasional stations (21)	1966-71	56	26 (101)	7 (12)	
Total number of samples			374			
Percent of samples positive for indicated compound				62	7	7

¹ Each sample represents 15 or more mature mollusks

² Calculated as Aroclor 1254®.

TABLE D-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Florida (Blank = no sample collected; -- = no residue detected above 5 ppb or no residue detected (PCB's); T = >5 but <10 ppb)

YEAR	COMPOUND	RESIDUES IN PPB (µG/KG)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—IONA POINT—31 SAMPLES ¹													
1966	DDE								30	13	24	35	T
	TDE								39	20	48	79	T
	DDT								--	--	--	28	--
1967	DDE	91	320	930	1,450	290	110	53	60	87	72	140	240
	TDE	94	170	760	705	310	200	110	160	160	150	220	310
	DDT	190	630	3,700	2,550	350	68	57	32	97	110	68	520
1968	DDE	760	1,200	1,100	1,500	780	340	180	--	T	77	84	82
	TDE	44	560	580	560	390	310	190	T	16	160	120	120
	DDT	2,800	3,600	2,300	1,200	650	220	33	--	--	69	140	60
	Dieldrin	--	--	--	--	--	--	--	--	11	--	--	--
1969	DDE		710	940									
	TDE		1,400	400									
	DDT		1,700	1,100									
STATION 2.—CHARLOTTE HARBOR—31 SAMPLES ¹													
1966	DDE								T	T	17	--	52
	TDE								10	16	23	--	91
	DDT								--	--	15	--	41
1967	DDE	83	14	15	39	18	30	13	T	T	--	T	18
	TDE	85	20	24	33	27	43	20	13	T	--	11	28
	DDT	170	T	13	--	T	13	T	--	--	--	--	21
	Dieldrin	--	--	--	--	--	--	14	11	19	--	--	15

TABLE D-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Florida—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 2.—CHARLOTTE HARBOR—31 SAMPLES ¹ —Continued													
1968	DDE	19	23	18	34	27	22	20	—	T	—	T	T
	TDE	22	28	18	36	29	18	26	T	17	—	11	T
	DDT	14	13	—	20	16	10	T	—	—	—	—	—
	Dieldrin	—	11	—	18	11	—	27	—	16	—	13	19
1969	DDE		14	17									
	TDE		19	22									
	DDT		—	12									
STATION 3.—CORAL COVE—32 SAMPLES ¹													
1966	DDE								24	T	10	25	17
	TDE								21	—	T	33	16
	DDT								—	—	—	T	T
1967	DDE	34	26	24	25	20	24	25	23	12	16	13	10
	TDE	28	23	22	26	21	21	24	20	16	16	10	10
	DDT	12	12	T	13	T	10	17	T	T	T	T	T
1968	DDE	29	27	21	35	49	39	31	19	20	28	21	23
	TDE	30	30	14	36	43	40	26	16	23	28	23	29
	DDT	10	14	—	13	37	49	32	22	14	28	11	T
1969	DDT	30	41	36									
	TDE	38	40	40									
	DDT	T	20	15									
STATION 4.—MANATEE RIVER—32 SAMPLES ¹													
1966	DDE								23	37	25	T	30
	TDE								39	47	33	T	33
	DDT								—	13	11	—	12
1967	DDE	37	39	22	31	18	T	19	21	23	33	34	13
	TDE	30	45	24	41	23	T	20	42	46	59	45	14
	DDT	19	19	10	13	T	—	T	20	17	13	14	14
1968	DDE	26	24	18	42	16	18	31	16	18	18	17	24
	TDE	24	29	30	65	19	61	88	37	38	16	19	27
	DDT	26	13	10	22	T	25	40	T	13	—	—	14
1969	DDE	22	32	24									
	TDE	33	46	26									
	DDT	—	17	T									

TABLE D-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Florida—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 5.—CRYSTAL RIVER—43 SAMPLES ¹													
1966	DDE								12			T	
	TDE											T	
	DDT												
1967	DDE	—	T	—	—	—	—	T	—	—	—	—	—
	TDE	—	—	—	—	—	—	T	—	—	—	—	—
	DDT	—	—	—	—	13	—	T	—	—	—	—	—
1968	DDE	—	—	—	T	—	11	—	—	—	—	—	—
	TDE	—	—	—	T	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	16	—	—	—	—	—	—
1969	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1970	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1971	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 6.—SUWANEE RIVER—32 SAMPLES ¹													
1966	DDE								T			T	
	TDE											T	
	DDT												
1967	DDE	—	12	T	—	—	—	—	—	—	—	—	—
	TDE	—	T	T	—	—	—	—	—	—	—	—	—
	DDT	—	T	T	—	11	11	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1969	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 7.—ST. VINCENTS BAR (NORTH)—17 SAMPLES ¹													
1966	DDE			T	16	T			14	—	—	11	T
	TDE			T	19	T			15	—	—	10	T
	DDT			—	T	—			—	—	—	—	—
	Dieldrin			—	10	—			—	—	—	—	—

TABLE D-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Florida—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 7.—ST. VINCENTS BAR (NORTH)—17 SAMPLES ¹ —Continued													
1967	DDE	T	—	T		22	T	13	—	—		T	
	TDE	—	—	T		23	T	10	—	—		—	
	DDT	—	—	—		T	—	T	—	—		—	
	Dieldrin	11	—	—		28	—	—	—	—		—	
STATION 8.—ST. VINCENTS BAR (SOUTH)—16 SAMPLES ¹													
1966	DDE			18	25			17	—	—		—	
	TDE			21	30			T	—	—		—	
	DDT			—	15			—	—	—		—	
	Dieldrin			—	13			—	—	—		—	
1967	DDE	T	—	14		21	T	13	T	T		—	
	TDE	—	—	13		22	T	12	T	—		—	
	DDT	—	—	—		T	38	T	—	—		—	
	Dieldrin	—	—	15		22	—	—	—	—		—	
STATION 9.—EAST BAY—84 SAMPLES ¹													
1965	DDE							19	T	T	—	T	T
	TDE							18	I	T	—	T	—
	DDT							13	T	T	—	T	—
1966	DDE	12	13	13	17	26	24	15	14	T	—	T	T
	TDE	—	14	—	15	24	19	11	—	—	—	—	T
	DDT	—	—	—	15	15	14	—	—	—	—	—	—
1967	DDE	T	18	21	18	18	12	20	T	T	T	T	T
	TDE	—	18	17	22	24	13	23	—	—	—	—	—
	DDT	—	14	16	15	15	—	18	—	10	—	—	—
1968	DDE	16	12	17	22	15	15	—	T	20	—	T	—
	TDE	15	20	—	—	—	—	—	T	—	—	—	—
	DDT	10	12	—	—	—	—	—	—	—	—	—	—
1969	DDE	T	16	11	—	13	14	—	—	10	T	—	—
	TDE	T	14	10	—	13	—	—	—	T	—	—	—
	DDT	—	T	—	—	—	—	—	—	14	—	—	—
1970	DDE	—	T	—	T	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	PCB's ²	—	—	—	—	380	180	170	73	92	50	55	140
1971	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	PCB's ²	160	160	200	220	230	390	190	230	100	55	120	—

TABLE D-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Florida—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 9.—EAST BAY—84 SAMPLES ¹ —Continued													
1972	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	PCB's ²	50	82	140	160	190	300						

¹ Each sample represents 15 or more mature mollusks.

² Calculated as Aroclor 1254 Φ .

SECTION E.—GEORGIA

Monthly collections of the eastern oyster (*C. virginica*) were made at 11 estuarine areas in Georgia during the period February 1967 - June 1972. Analyses were done at the Gulf Breeze Laboratory until September 1969, and thereafter at the Marine Institute of the University of Georgia. The approximate locations of monitoring stations are shown in Fig. E-1. A summary of data on organochlorine residues in the monitored species, *C. virginica*, is presented in Table E-1, and the distribution of residues in this species for each sampling station by date of collection in Table E-2. The 15% incidence of DDT residues in Georgia samples was next to the lowest of all States monitored (Washington, lowest at 11%). The maximum level of DDT observed was also next to the lowest of any of the other States monitored. By contrast, the largest dieldrin residue detected in the nationwide program was in Georgia, (230 ppb) and the incidence of dieldrin residues (21%) was well above the average incidence (15%) for all States.

The occurrence of substantial toxaphene residues in the samples collected in St. Simons Sound was unexpected. A special sampling program was initiated in the area that included the placement of trays of oysters in creek beds where oysters did not normally occur. Analyses of these samples pinpointed the industrial source of the toxaphene and precipitated a schedule for control of the effluent discharge by the manufacturer. The magnitude of toxaphene residues at Stations 8 - 11 illustrates well the importance of dilution (distance) in the abatement of pollution.

Polychlorinated biphenyl residues were analyzed for beginning in 1969. A few samples collected in the Ogeechee and Satilla River basins contained residues of Aroclor 1254 Φ , but the amounts were not quantified.

DDT residue levels were generally low and there was an approximate increase of 13% in the number of samples

with negligible residues in 1971 as compared to earlier years. Stations 1 and 2 in the Savannah River basin, however, showed a reversal of this trend in 1972 when oysters contained substantially higher residue levels than in 1971.

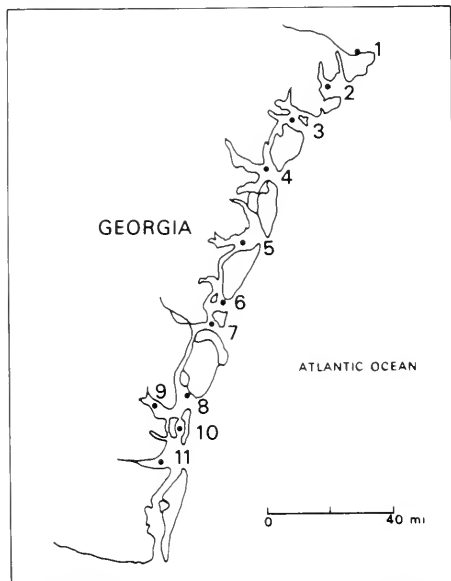


FIGURE E-1.—Diagram of coastal Georgia showing approximate location of monitoring stations

1. Lazeretta Creek—Savannah River Basin
2. Wilmington River—Savannah River Basin
3. Ogeechee River—Ogeechee River Basin
4. St. Catherine Sound—Ogeechee River Basin
5. Sapelo Sound—Ogeechee River Basin
6. Doboy Sound—Ogeechee River Basin
7. Egg Island—Altamaha River Basin
8. St. Simons Sound—Satilla River Basin
9. Terry Creek—Satilla River Basin
10. Jekyll Island—Satilla River Basin
11. Satilla River—Satilla River Basin

TABLE E-1—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1967-72—Georgia

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB ($\mu\text{g}/\text{kg}$)			
				DDT	DIELDRIN	TOXAPHENE	PCB'S ²
1	Lazeretta Creek	1967-72	64	30 (96)	58 (230)		
2	Wilmington River	1967-72	65	21 (86)	27 (90)		
3	Ogeechee River	1967-72	65	13 (50)	15 (26)		1
4	St. Catherine Sound	1967-72	65	7 (15)	2 (T)		1
5	Sapelo Sound	1967-72	65	12 (50)	6 (12)		2
6	Doboy Sound	1967-72	64	7 (27)	8 (14)		1
7	Egg Island	1967-72	65	3 (52)	22 (23)		
8	St. Simons Sound	1967-72	65	⁽³⁾	3 (T)	64 (7,500)	2
9	Terry Creek	1967-70	16	⁽³⁾		16 (54,000)	
10	Jekyll Island	1967-72	62	⁽³⁾		37 (3,500)	8
11	Satilla River	1967-72	64	3 (15)		8 (1,000)	1
	Occasional stations (2)	1968-69	4	⁽³⁾		3 (13,000)	
Total number of samples			664				
Percent of samples positive for indicated compound				15	21	19	2

NOTE: T = >5 but <10 ppb.

¹ Each sample represents 15 or more mature mollusks.² Present but not quantified.³ Presence of toxaphene prevented quantification of DDT and its metabolites.TABLE E-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Georgia

YEAR	COMPOUND	JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
RESIDUES IN PPB ($\mu\text{g}/\text{kg}$)													
[Blank = no sample collected; — = no residue detected above 5 ppb or no residue detected (PCB's); T = >5 but <10 ppb]													
STATION 1.—LAZERETTA CREEK—64 SAMPLES ¹													
1967	DDE		14	13	21	T	T	53	—	—	—	—	12
	TDE		17	14	29	13	11	25	14	—	—	—	16
	DDT		—	—	T	—	T	18	11	—	—	—	T
	Dieldrin		98	65	56	32	30	30	33	18	42	33	46
1968	DDE	—	13	12	17	—	—	15	—	—	—	—	T
	TDE	—	16	12	23	—	—	23	—	—	—	—	T
	DDT	—	—	—	T	—	—	28	—	—	—	—	—
	Dieldrin	22	42	37	46	—	20	39	22	18	—	42	56
1969	DDE	—	—	—	—	—	T	—	—	—	—	—	T
	TDE	—	—	—	—	—	13	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	39	23	47	51	16	35	28	23	20	180	230	20
1970	DDE	T	13	—	T	T	—	—	T	—	23	—	—
	TDE	—	—	—	T	T	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	30	31	40	32	17	23	80	T	—	—	T	T

TABLE E-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—
Georgia—Continued

YEAR	COMPOUND	JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
RESIDUES IN PPB ($\mu\text{g}/\text{kg}$)													
[Blank = no sample collected; — = no residue detected above 5 ppb or no residue detected (PCB's); T = >5 but <10 ppb]													
STATION 1.—LAZERETTA CREEK—64 SAMPLES ¹ —Continued													
1971	DDE	—	—	20	—	—	—	—	—	—	T	T	—
	TDE	—	—	—	—	—	—	—	—	—	T	T	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	19	19	T	17	13	10	—	T	T	—	—	13
1972	DDE	T	T	23	18	T	T	—	—	—	—	—	—
	TDE	—	T	14	12	T	T	—	—	—	—	—	—
	DDT	—	—	T	T	—	T	—	—	—	—	—	—
	Dieldrin	15	13	T	T	22	T	—	—	—	—	—	—
STATION 2.—WILMINGTON RIVER—65 SAMPLES ¹													
1967	DDE	—	T	T	T	—	—	T	12	—	—	—	—
	TDE	—	T	T	T	—	—	T	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	17	19	22	—	—	—	—	—	—	—	—
1968	DDE	—	—	T	T	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	10	21	12	—	—	—	—	—	—	—	—	—
1969	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	—	—	10	—	—	—	—	—	—	90	T
1970	DDE	—	11	—	T	—	—	—	T	86	—	—	—
	TDE	—	—	—	T	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	T	T	10	25	—	T	—	—	—	—	—	T
1971	DDE	—	—	10	—	—	—	—	—	—	T	T	T
	TDE	—	—	—	—	—	—	—	—	—	T	T	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	T	12	12	T	T	—	—	—	—	—	T	T
1972	DDE	T	12	15	12	T	T	—	—	—	—	—	—
	TDE	—	13	T	11	T	T	—	—	—	—	—	—
	DDT	—	—	—	—	T	T	—	—	—	—	—	—
	Dieldrin	10	17	15	—	T	T	—	—	—	—	—	—

TABLE E-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—
Georgia—Continued

YEAR	COMPOUND	JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
RESIDUES IN PPB ($\mu\text{g}/\text{kg}$)													
[Blank = no sample collected; — = no residue detected above 5 ppb or no residue detected (PCB's); T = >5 but <10 ppb]													
STATION 3.—OGEECHEE RIVER—65 SAMPLES ¹													
1967	DDE		T	T	T	—	—	—	T	—	—	—	T
	TDE		T	10	—	—	—	—	—	—	—	—	T
	DDT		—	—	—	—	—	—	—	—	—	—	T
	Dieldrin		13	26	10	—	—	—	—	—	—	—	—
1968	DDE		—	13	T	—	—	—	—	—	—	—	—
	TDE		—	13	11	—	—	—	—	—	—	—	—
	DDT		—	24	—	—	—	—	—	—	—	—	—
	Dieldrin		18	16	—	—	—	—	—	—	—	—	—
1969	DDE		—	—	—	—	—	—	—	—	—	—	—
	TDE		—	—	—	—	—	—	—	—	—	—	—
	DDT		—	—	—	—	—	—	—	—	—	—	—
	Dieldrin		—	—	—	T	—	—	16	—	—	—	T
	PCB's		—	—	—	—	—	—	—	—	—	—	(2)
1970	DDE		10	—	—	—	—	—	T	12	—	—	—
	TDE		—	—	—	—	—	—	—	—	—	—	—
	DDT		—	—	—	—	—	—	—	—	—	—	—
	Dieldrin		T	T	T	—	—	—	—	—	—	—	—
1971	DDE		—	—	—	—	—	—	—	—	—	—	—
	TDE		—	—	—	—	—	—	—	—	—	—	—
	DDT		—	—	—	—	—	—	—	—	—	—	—
	Dieldrin		—	T	T	T	—	—	11	—	—	—	—
1972	DDE		—	T	—	T	T	—	—	—	—	—	—
	TDE		—	—	—	—	T	—	—	—	—	—	—
	DDT		—	—	—	—	—	—	—	—	—	—	—
STATION 4.—ST. CATHERINE SOUND—65 SAMPLES ¹													
1967	DDE		T	T	T	—	—	—	—	—	—	—	—
	TDE		T	—	—	—	—	—	—	—	—	—	—
	DDT		T	—	—	—	—	—	—	—	—	—	—
1968	DDE		—	—	T	—	—	—	—	—	—	—	—
	TDE		—	—	T	—	—	—	—	—	—	—	—
	DDT		—	—	—	—	—	—	—	—	—	—	—
1969	DDE		—	—	—	—	—	—	—	—	—	—	—
	TDE		—	—	—	—	—	—	—	—	—	—	—
	DDT		—	—	—	—	—	—	—	—	—	—	—
	PCB's		—	—	—	—	—	—	—	—	—	—	(2)

TABLE E-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—
Georgia—Continued

YEAR	COMPOUND	JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
RESIDUES IN PPB ($\mu\text{g/kg}$)													
[Blank = no sample collected; — = no residue detected above 5 ppb or no residue detected (PCB's); T = >5 but <10 ppb]													
STATION 4.—ST. CATHERINE SOUND—65 SAMPLES ¹ —Continued													
1970	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1971	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	T	—	T	—	—	—	—	—	—	—	—
1972	DDE	—	—	T	—	T	T	—	—	—	—	—	—
	TDE	—	—	—	—	—	T	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 5.—SAPELO SOUND—65 SAMPLES ¹													
1967	DDE	—	T	T	T	—	T	T	—	—	—	—	T
	TDE	—	T	—	T	—	T	22	—	—	—	—	T
	DDT	—	T	—	—	—	T	23	—	—	—	—	—
	Dieldrin	—	12	—	—	—	—	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1969	DDE	—	—	T	—	—	—	—	—	—	—	—	—
	TDE	—	—	13	—	—	—	—	—	—	—	—	—
	DDT	—	—	19	—	—	—	—	—	—	—	—	—
	Dieldrin	—	—	—	T	—	—	—	—	—	—	—	—
	PCB's	—	—	—	—	—	—	—	—	—	—	12 ¹	12 ¹
1970	DDE	—	11	—	T	—	—	—	—	—	—	—	—
	TDE	—	—	—	T	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	—	—	T	—	—	—	—	—	—	—	—
1971	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	T	T	T	—	—	—	—	—	—	—	—
1972	DDE	—	—	T	—	T	T	—	—	—	—	—	—
	TDE	—	—	—	—	—	T	—	—	—	—	—	—
	DDT	—	—	—	—	T	—	—	—	—	—	—	—

TABLE E-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—
Georgia—Continued

YEAR	COMPOUND	JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
RESIDUES IN PPB ($\mu\text{g}/\text{kg}$)													
[Blank = no sample collected; — = no residue detected above 5 ppb or no residue detected (PCB's); T = >5 but <10 ppb]													
STATION 6.—DOBOY SOUND—64 SAMPLES ¹													
1967	DDE		T	T	—	—	—	T	—	—	—	—	—
	TDE		—	T	—	—	—	11	—	—	—	—	—
	DDT		—	—	—	—	—	11	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1969	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	14	14	13	—	—	—	—	—	—	—	—
1970	PCB's	—	—	—	—	—	—	—	—	—	—	—	(2)
	DDE	—	10	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1971	Dieldrin	T	—	—	—	—	—	—	—	—	—	—	—
	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1972	Dieldrin	T	T	T	—	T	—	—	—	—	—	—	—
	DDE	—	—	T	T	T	—	—	—	—	—	—	—
	TDE	—	—	T	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 7.—EGG ISLAND—65 SAMPLES ¹													
1967	DDE	—	—	—	—	—	—	15	—	—	—	—	—
	TDE	—	—	—	—	—	—	19	—	—	—	—	—
	DDT	—	—	—	—	—	—	18	—	—	—	—	—
	Dieldrin	—	—	—	—	—	—	—	—	—	—	—	20
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	13	15	23	14	—	—	—	—	—	—	—	21
1969	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	16	—	15	15	—	—	—	—	—	—	—	T

TABLE E-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—
Georgia—Continued

YEAR	COMPOUND	JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
RESIDUES IN PPB ($\mu\text{g}/\text{kg}$)													
[Blank = no sample collected; — = no residue detected above 5 ppb or no residue detected (PCB's); T = >5 but <10 ppb]													
STATION 7.—EGG ISLAND—65 SAMPLES ¹ —Continued													
1970	DDE	—	16	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	T	T	T	—	T	—	—	—	—	T	T	T
1971	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	11	T	T	T	—	—	—	—	—	—	—	—
1972	DDE	—	—	T	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	T	—	—	—	—	—	—	—	—	—	—
RESIDUES IN PPM (mg/kg)													
[Blank = no sample collected; — = no residue detected above 0.1 ppm or no residue detected (PCB's); T = >0.1 but <0.25 ppm]													
STATION 8.—ST. SIMONS ISLAND—65 SAMPLES ^{1,2,5}													
1967	Toxaphene	—	—	2.5	1.5	1.5	1.0	1.0	1.1	0.8	0.7	2.0	2.0
1968	Toxaphene	0.8	5.0	6.0	4.3	1.6	2.0	2.0	0.6	T	—	5.4	2.8
1969	Toxaphene	2.0	1.2	2.5	7.5	5.0	1.5	1.0	1.0	1.5	1.6	1.6	1.8
1970	Toxaphene	3.8	3.8	7.2	3.3	1.8	1.1	1.0	0.8	T	0.6	0.7	1.6
	PCB's	—	—	—	—	—	—	—	—	—	—	—	—
1971	Toxaphene	1.3	0.7	1.1	1.6	0.7	0.1	0.6	T	T	0.6	T	0.6
	PCB's	—	—	—	—	—	—	—	—	—	—	—	—
1972	Toxaphene	0.6	1.0	1.1	1.0	0.8	0.6	—	—	—	—	—	—
RESIDUES IN PPM (mg/kg)													
[Blank = no sample collected; — = no residues detected above 0.1 ppm; T = >0.1 but <0.25 ppm]													
STATION 9.—TERRY CREEK—16 SAMPLES ¹													
1967	Toxaphene	—	—	—	12.0	—	—	4.7	—	—	—	18.0	13.0
1968	Toxaphene	—	23.0	6.0	54.0	—	5.0	—	—	—	6.3	12.0	—
1969	Toxaphene	9.0	—	—	—	12.0	17.0	—	8.0	—	—	—	—
1970	Toxaphene	—	—	—	6.2	8.2	—	—	—	—	—	—	—

TABLE E-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Georgia—Continued

YEAR	COMPOUND	JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
RESIDUES IN PPM (mg/kg)													
[Blank = no sample collected; — = no residue detected above 0.1 ppm; T = >0.1 but <0.26 ppm]													
STATION 10.—JEKYLL ISLAND—62 SAMPLES ^{1,6}													
1967	Toxaphene	—	—	T	0.5	0.4	0.4	—	—	—	—	1.0	—
1968	Toxaphene	0.7	2.1	0.7	0.7	0.5	T	0.4	—	—	—	—	—
	PCB's	—	—	—	—	—	—	—	—	—	—	—	—
1969	Toxaphene	1.0	1.0	1.0	1.0	—	—	—	—	3.5	T	—	0.7
	PCB's	—	—	—	—	—	—	—	—	—	(2)	—	—
1970	Toxaphene	0.8	—	T	T	—	—	—	—	—	—	—	0.6
	PCB's	—	(2)	(2)	(2)	(2)	—	—	—	—	—	—	—
1971	Toxaphene	0.5	T	0.6	0.8	T	T	—	—	—	0.5	T	T
	PCB's	—	(2)	—	—	(2)	—	—	—	—	—	—	—
1972	Toxaphene	0.3	T	1.0	0.6	T	—	—	—	—	—	—	—
RESIDUES IN PPM (mg/kg)													
[Blank = no sample collected; — = no residue of DDT detected above 0.005 ppm or no residue detected above 0.1 ppm (toxaphene and PCB's); t = >0.1 but <0.25 ppm; T = >0.005 but <0.010 ppm]													
STATION 11.—SATILLA RIVER—64 SAMPLES ¹													
1967	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Toxaphene	—	—	t	t	t	t	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Toxaphene	1.0	0.5	0.7	t	—	—	—	—	—	—	—	—
1969	DDF	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1970	DDE	—	15	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	PCB's	—	—	—	—	—	—	—	—	—	—	—	—
1971	DDE	—	—	T	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	PCB's	—	—	—	—	—	—	—	—	—	—	—	—
1972	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	T	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

¹ Each sample represents 15 or more mature mollusks.

² Aroclor 1254B present but not quantified.

³ Presence of toxaphene prevented quantification of DDT and its metabolites in these samples.

⁴ Toxaphene present but not quantified.

⁵ One sample each in April 1969, April 1970, and February 1972 contained a trace of dieldrin.

⁶ DDT and its metabolites not detected in any samples.

SECTION F.—MAINE

The monthly monitoring of Maine estuaries for persistent synthetic residues was initiated in December 1965 and continued until November 1970. There were 10 principal stations; about 40 other sites were sampled occasionally. Samples were analyzed at the Gulf Breeze Laboratory until June 1969 and, thereafter, at the Fisheries Research Station, Maine Department of Sea and Shore Fisheries.

The soft clam (*Mya arenaria*) and the blue mussel (*Mytilus edulis*) were the principal mollusks monitored and, on occasion, both eastern oysters (*Crassostrea virginica*) and horse mussels (*Modiolus modiolus*) were collected at the same sites. In the laboratory, the uptake of DDT was greater in the soft clam than in other species tested as was the flushing rate, and 90% of DDT residues was lost within 7 days after the toxicant was removed. This may explain why in simultaneous collections of two or more species of mollusks, DDT residues in soft clams examined at 30-day intervals were usually lower than those in the oyster or horse mussel. A summary of data on organochlorine residues in the monitored species, is presented in Table F-1, and the distribution of residues in these species for each sampling station by date of collection in Table F-2.

The Maine samples are characterized by the low incidence (18%) of detectable DDT residues as compared to most other monitored areas, despite the fact that substantial amounts of DDT are reported to have been used agriculturally in some watersheds in Maine. The maximum magnitude of DDT residues detected was, however, larger than that found in seven other States. Analysis of occasional collections of fish and invertebrates other than mollusks revealed DDT residues larger than those in mollusks. Presumably organochlorine pollution in Maine estuaries was usually too low and too

transitory to be detected except in animals that retain residues for a long period of time.

Despite the generally low incidence of DDT residues at most stations, there was sufficient continuity in detectable DDT residues at Station 10 on the Piscataqua River to show a gradual decline from an average of about 28 ppb in 1966 to an undetectable level in 1970. A similar trend is clearly shown in samples collected at Station 7, Small Point.

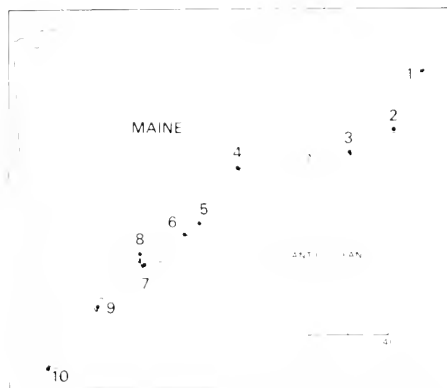


FIGURE F-1.—Diagram of coastal Maine showing approximate location of monitoring stations

- 1 Mill Cove—St. Croix River
- 2 Machiasport—Machias River
- 3 Millbridge—Naraguagus River
- 4 Fort Point—Penobscot River
- 5 Thomaston—St. George River
- 6 Medomak—Medomak River
- 7 Small Point—Kennebec-Androscoogin River
- 8 Phippsburg—Kennebec-Androscoogin River
- 9 Biddeford Pool—Saco River
- 10 Eliot—Piscataqua River

TABLE F-1.—Summary of data on organochlorine residues in the monitored species, 1965-70—Maine

STATION NUMBER	LOCATION	MONITORING PERIOD	PRINCIPAL MONITORED SPECIES	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (µG/KG)	
					DDT	DIELDRIN
1	Mill Cove	1965-66	<i>M. arenaria</i>	12		
2	Machiasport	1965-70	<i>M. arenaria</i>	52	2 (15)	
3	Millbridge	1966-70	<i>M. arenaria</i>	37	1 (12)	
4	Fort Point	1965-70	<i>M. arenaria</i>	42	1 (15)	
5	Thomaston	1965-70	<i>M. arenaria</i>	42	1 (80)	1 (11)
6	Medomak	1967-70	<i>M. arenaria</i>	23	2 (11)	
7	Small Point	1968-70	<i>M. edulis</i>	18	12 (359)	

TABLE F-1.—Summary of data on organochlorine residues in the monitored species, 1965-70—Maine—Continued

STATION NUMBER	LOCATION	MONITORING PERIOD	PRINCIPAL MONITORED SPECIES	NUMBER OF SAMPLES :	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB ($\mu\text{g/kg}$)	
					DDT	DIELDRIN
4	Phlipsburg	1965-69	<i>M. arenaria</i>	39	7 (24)	
9	Biddeford Pool	1968-70	<i>M. edulis</i>	24	7 (64)	
10	Eliot	1966-70	<i>M. arenaria</i>	45	22 (67)	9 (38)
11	Occasional stations 40	1965-69	Mixed	62	16 (93)	4 (18)
Total number of samples				396		
Percent of samples positive for indicated compound					18	4

Each sample represents 15 or more mature mollusks.

TABLE F-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—Maine

Blank = no sample collected; — = no residue detected above 5 ppb; T = >5 but <10 ppb.]

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{g/kg}$)											
		JAN	FEB	MAR	APR	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV	DEC.
STATION 1.—MILL COVE— <i>M. ARENARIA</i> —12 SAMPLES :													
1965	DDE												
	TDE												
	DDT												
1966	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 2.—MACHIASPORT— <i>M. ARENARIA</i> —52 SAMPLES :													
1965	DDE												—
	TDE												—
	DDT												—
1966	DDE	—	—	—	—	T	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	T	—	—
	TDE	—	—	—	—	—	—	—	—	—	T	—	—
	DDT	—	—	—	—	—	—	—	—	—	T	—	—
1969	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

TABLE F-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—Maine—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 2.—MACHIASPORT—M <i>ARENARIA</i> —52 SAMPLES ¹ —Continued													
1970	DDE						—	—	—	—	—	—	—
	TDE						—	—	—	—	—	—	—
	DDT						—	—	—	—	—	—	—
STATION 3.—MILLBRIDGE—M <i>ARENARIA</i> —3 ¹ SAMPLES ¹													
1966	DDE												—
	TDE												—
	DDT												—
1967	DDE					—	—	—	—	—	—	—	—
	TDE					—	—	—	—	—	—	—	—
	DDT					—	—	—	—	—	—	—	—
1968	DDE			—	—	—	—	—	—	—	—	—	—
	TDE			—	—	—	—	—	—	—	—	—	—
	DDT			—	—	—	—	—	—	—	—	—	—
1969	DDE		—	—	—	—	—	—	—	—	—	—	—
	TDE		—	—	—	—	—	—	—	—	—	—	—
	DDT		—	—	—	—	—	—	—	—	—	—	—
1970	DDE						—	—	—	12	—	—	—
	TDE						—	—	—	—	—	—	—
	DDT						—	—	—	—	—	—	—
STATION 4.—FORT POINT—M <i>ARENARIA</i> —42 SAMPLES ¹													
1965	DDE												—
	TDE												—
	DDT												—
1966	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE			—	—	—	—	—	—	—	—	—	—
	TDE			—	—	—	—	—	—	—	—	—	—
	DDT			—	—	—	—	—	—	—	—	—	—
1968	DDE			—	—	—	—	—	—	—	—	—	—
	TDE			—	—	—	—	—	—	—	—	—	—
	DDT			—	—	—	—	—	—	—	—	—	5
1969	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

TABLE F-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—Maine—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 4.—FORT POINT— <i>M. ARENARIA</i> —42 SAMPLES ¹ —Continued													
1970	DDE	—	—										
	TDE	—	—										
	DDT	—	—										
STATION 5.—THOMASTON— <i>M. ARENARIA</i> , UNLESS OTHERWISE INDICATED—42 SAMPLES ¹													
1965	DDE												—
	TDE												—
	DDT												—
1966	DDE										21	—	
	TDE		—	—	—	—	—	—	—	—	35	—	
	DDT		—	—	—	—	—	—	—	—	24	—	
	Dieldrin		—	—	—	11	—	—	—	—	—	—	
1967	DDE	—		—	—	—	—	—	—	—	—	—	—
	TDE	—		—	—	—	—	—	—	—	—	—	—
	DDT	—		—	—	—	—	—	—	—	—	—	—
1968	DDF												—
	TDE			—	—	—	—	—	—	—	—	—	—
	DDT			—	—	—	—	—	—	—	—	—	—
1969	DDE				—	—	—	—	—	—	—	—	—
	TDE	—			—	—	—	—	—	—	—	—	—
	DDT	—			—	—	—	—	—	—	—	—	—
1970	DDE				—	—	—	—	—	—	—	—	—
	TDE	—			—	—	—	—	—	—	—	—	—
	DDT	—			—	—	—	—	—	—	—	—	—
STATION 6.—MEDOMAK— <i>M. ARENARIA</i> —23 SAMPLES ¹													
1967	DDE				—	—	—	—	—	—	—	—	—
	TDE				—	—	—	—	—	—	—	—	—
	DDT				11	—	1	—	—	—	—	—	—
1968	DDF					—	—	—	—	—	—	—	—
	TDE				—	—	—	—	—	—	—	—	—
	DDT				—	—	—	—	—	—	—	—	—
1969	DDF				—	—	—	—	—	—	—	—	—
	TDE				—	—	—	—	—	—	—	—	—
	DDT				—	—	—	—	—	—	—	—	—
1970	DDF				—	—	—	—	—	—	—	—	—
	TDE				—	—	—	—	—	—	—	—	—
	DDT				—	—	—	—	—	—	—	—	—

TABLE F-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—Maine—Continued

YEAR	COMPOUND	RESIDUES IN PPB (µG/KG)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 7.—SMALL POINT— <i>M. EDULIS</i> —18 SAMPLES ¹													
1968	DDE							35	21	17	12	25	19
	TDE							44	25	20	14	27	18
	DDT							280	77	68	18	26	26
1969	DDE		11		T	T	T	12	—	—	—	—	—
	TDE		14		T	T	T	21	—	—	—	—	—
	DDT		18		15	13	T	13	49	—	—	—	—
1970	DDE	—	—		—								
	TDE	—	—		—								
	DDT	—	—		—								
STATION 8.—PHIPPSBURG— <i>M. ARENARIA</i> —39 SAMPLES ¹													
1965	DDE												—
	TDE												—
	DDT												—
1966	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	—	—	T	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	11	11	T	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	T	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	T	19	—	—	—	—	T	11
1969	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 9.—BIDDEFORD POOL— <i>M. EDULIS</i> —24 SAMPLES ¹													
1968	DDE			—		—	T	—	T	1	T	T	
	TDE			—		—	T	—	T	T	T	T	
	DDT			—		—	21	—	54	22	15	13	
1969	DDE			T	—	T	—	—	—	—	—	—	—
	TDE			T	—	—	—	—	—	—	—	—	—
	DDT			21	—	12	—	—	—	—	—	—	—
1970	DDE	—			—	—	—	—	—	—	—	—	—
	TDE	—			—	—	—	—	—	—	—	—	—
	DDT	—			—	—	—	—	—	—	—	—	—

TABLE F-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—Maine—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 10.—ELIOT— <i>M. ARENARIA</i> . UNLESS OTHERWISE INDICATED—45 SAMPLES ¹													
1966	DDE	T		12	14	13	13	T	—	T	T	T	—
	TDE	T		16	21	19	21	11	—	T	11	T	—
	DDT	21	T	32	32	23	16	T	—	T	T	T	—
	Dieldrin	32	—	38	—	27	23	—	—	—	—	—	T
1967	DDE	—	—	—	T	T	—	—	—	T	T	—	T
	TDE	—	—	—	11	14	T	—	—	15	T	—	T
	DDT	—	—	—	20	18	30	—	—	T	22	—	15
	Dieldrin	—	—	16	Lost	10	T	—	—	—	—	—	—
1968	DDE	—	—	—	T	—	—	—	T	—	—	—	—
	TDE	—	—	—	T	12	—	—	12	—	—	—	—
	DDT	—	—	—	18	15	—	—	T	—	—	—	—
	Dieldrin	—	—	—	10	—	—	—	—	—	—	—	—
1969	DDE	—	—	—	11	² 12	T	—	—	—	—	—	—
	TDE	—	—	—	23	22	T	—	—	—	—	—	—
	DDT	—	—	—	18	14	T	—	—	—	—	—	—
1970	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

¹ Each sample represents 15 or more mature mollusks.

² *M. edulis*.

³ *M. demissus*.

SECTION G.—MARYLAND

Eastern oysters, *Crassostrea virginica*, were collected in upper Chesapeake Bay and its tributaries at irregular intervals (usually twice yearly) from August 1966 to November 1970. The sampling was made possible because of oyster surveys being conducted for other programs. All samples from the 10 locations in Maryland were analyzed at the Gulf Breeze Laboratory. The approximate station locations are shown in Fig. G-1. A summary of data on organochlorine residues in the monitored species, *C. virginica*, is presented in Table G-1, and the distribution of residues in this species for each sampling station by date of collection in Table G-2.

Maryland was fifth among all States, in the incidence of DDT residues (81%), but the magnitude of residues in oysters was surprisingly low in view of the size of the Susquehanna River watershed and the extent of its agricultural development. More selective monitoring might show that the major pesticide burden of the river is precipitated with silt in the headwaters of the Bay and does not enter the trophic web of the estuarine system extensively. DDT residues detected at monitoring stations probably reflected pollution primarily in the adjacent and usually small drainage basins.

Despite the small number of samples, the decline in average DDT residues from 26 ppb in 1966 to 10 ppb in 1970 together with a more than 150% increase in samples containing less than 11 ppb suggests a real change in average pollution levels.

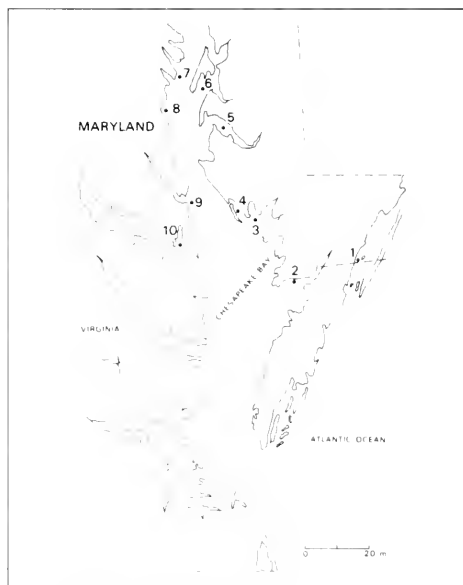


FIGURE G-1.—Diagram of coastal Maryland showing approximate location of monitoring stations

1. Franklin City—Chincoteague Bay
2. Pocomoke Sound
3. Tangier Sound
4. Honga River
5. Choptank River
6. Eastern Bay
7. Tollys Bar—Chesapeake Bay
8. Herring Bay—Chesapeake Bay
9. Cedar Point—Chesapeake Bay
10. St. Marys River

TABLE G-1.—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1966-70—Maryland

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (µg/kg)	
				DDT	DIELDRIN
1	Franklin City	1966-70	8	8 (43)	
2	Pocomoke Sound	1966-69	6	5 (47)	
3	Tangier Sound	1966-70	10	5 (48)	
4	Honga River	1966-70	10	8 (43)	
5	Choptank River	1966-70	8	4 (30)	
6	Eastern Bay	1966-70	10	8 (70)	
7	Tollys Bar	1967-70	8	8 (44)	7 (22)
8	Herring Bay	1966-70	10	9 (46)	4 (18)
9	Cedar Point	1966-70	10	9 (70)	
10	St. Marys River	1966-70	8	7 (33)	
Total number of samples			88		
Percent of samples positive for indicated compound				81	13

¹ Each sample represents 15 or more mature mollusks.

TABLE G-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Maryland

[Blank = no sample collected. — = no residue detected above 5 ppb; T = >5 but <10 ppb]

YEAR	COMPOUND	RESIDUES IN PPB (µG/KG)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—FRANKLIN CITY—8 SAMPLES:													
1966	DDE								10				T
	TDE								14				—
	DDT								T				—
1967	DDE			11				10					
	TDE			—				—					
	DDT			—				13					
1968	DDE				26			T					T
	TDE				17			T					—
	DDT				—			16					T
1969								No Samples Collected					
1970	DDE				14								
	TDE				—								
	DDT				—								
STATION 2.—POCOMOKE SOUND—6 SAMPLES:													
1966	DDE								—				T
	TDE								17				T
	DDT								T				—
1967	DDE			T					T				
	TDE			T					T				
	DDT			—					T				
1968	DDE							11					
	TDE							12					
	DDT							24					
1969	DDE												
	TDE												
	DDT												
STATION 3.—TANGIER SOUND—10 SAMPLES:													
1966	DDE								13				T
	TDE								24				T
	DDT								11				—
1967	DDE			—				T					
	TDE			—				—					
	DDT			—				10					

TABLE G-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Maryland—Continued

YEAR	COMPOUND	RESIDUES IN PPB (µG/KG)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 3.—TANGIER SOUND—10 SAMPLES—Continued													
1968	DDE				T			17				—	
	TDE				T			10				—	
	DDT				—			—				—	
1969	DDE			—									
	TDE			—									
	DDT			—									
1970	DDE				—								—
	TDE				—								—
	DDT				—								—
STATION 4.—HONGA RIVER—10 SAMPLES													
1966	DDE									T			12
	TDE									12			20
	DDT									11			11
1967	DDE			T						T			
	TDE			12						T			
	DDT			—						T			
1968	DDE					T				T		T	
	TDE					—				T		—	
	DDT					—				28		10	
1969	DDE			—									
	TDE												
	DDT			—									
1970	DDE					—							T
	TDE					—							10
	DDT					—							—
STATION 5.—CHOPTANK RIVER—8 SAMPLES													
1966	DDE									T			T
	TDE									11			13
	DDT									—			12
1967	DDE			T						T			
	TDE			11						T			
	DDT			—						—			
1968	DDE												
	TDE												
	DDT												

TABLE G-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Maryland—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 5.—CHOPTANK RIVER—8 SAMPLES ¹ —Continued													
1969	DDE			—									
	TDE			—									
	DDT			—									
1970	DDE					—							—
	TDE					—							—
	DDT					—							—
STATION 6.—EASTERN BAY—10 SAMPLES ¹													
1966	DDE								—				14
	TDE								—				17
	DDT								—				—
1967	DDE		11					T					
	TDE		15					11					
	DDT		—					T					
1968	DDE					—		11					11
	TDE					—		11					—
	DDT					—		48					16
1969	DDE			10									
	TDE			T									
	DDT			—									
1970	DDE					11							10
	TDE					T							10
	DDT					—							—
STATION 7.—TOLLYS BAR—8 SAMPLES ¹													
1967	DDE		18					13					
	TDE		19					17					
	DDT		T					11					
	Dieldrin		13					13					
1968	DDE					14		12				15	
	TDE					11		14				13	
	DDT					—		T				16	
	Dieldrin					15		—				11	
1969	DDE			15									
	TDE			14									
	DDT			—									
	Dieldrin			16									

TABLE G-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Maryland—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 7.—TOLLYS BAR—8 SAMPLES ¹ —Continued													
1970	DDE			16									T
	TDE			17									10
	DDT			—									—
	Dieldrin			22									15
STATION 8.—HERRING BAY—10 SAMPLES ¹													
1966	DDE								10				T
	TDE								15				—
	DDT								T				—
1967	DDE		12					10					
	TDE		11					17					
	DDT		—					T					
	Dieldrin		—					13					
1968	DDE					T		12			12		
	TDE					T		14			11		
	DDT					—		20			11		
1969	DDE			10									
	TDE			11									
	DDT			—									
	Dieldrin			13									
1970	DDE					14							—
	TDE					16							—
	DDT					—							—
	Dieldrin					18							12
STATION 9.—CEDAR POINT—10 SAMPLES ¹													
1966	DDE							18					20
	TDE							27					24
	DDT							25					15
1967	DDE		22					15					
	TDE		20					16					
	DDT		T					13					
1968	DDE					T		21				T	
	TDE					—		13				T	
	DDT					—		16				24	
1969	DDE			T	11								
	TDE			T	12								
	DDT			—	—								

TABLE G-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Maryland—Continued

YEAR	COMPOUND	RESIDUES IN PPB (μ G KG)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 9.—CEDAR POINT—10 SAMPLES ¹ —Continued													
1970	DDE												—
	TDE												—
	DDT												—
STATION 10.—ST. MARYS RIVER—8 SAMPLES ¹													
1966	DDE							—					12
	TDE							—					16
	DDT							—					T
1967	DDE		15					T					
	TDE		17					T					
	DDT		—					T					
1968	DDE					T		11					T
	TDE					—		11					—
	DDT					—		11					12
1969													No Samples Collected
1970	DDE			15									
	TDE			11									
	DDT			—									

Each sample represents 15 or more mature mollusks.

SECTION H.—MISSISSIPPI

Mississippi Sound and tributaries were monitored for organochlorine residues in eastern oysters, *C. virginica*, during the period August 1965 - June 1972. All samples from the eight sampling stations were analyzed at the Gulf Breeze Laboratory. Approximate station locations are shown in Fig. H-1. A summary of data on organochlorine residues in the monitored species, *C. virginica* is presented in Table H-1, and the distribution of residues in this species for each sampling station by date of collection in Table H-2.

Only four States had a lower incidence of DDT residues in oysters, and the maximum residue detected in Mississippi (125 ppb) was lower than that in 12 of the other 14 States. Maximum DDT residues appeared to be more directly related to runoff from urban and industrialized centers rather than from agricultural areas.

In 1971, there was a more than 70% increase in the number of DDT residues of less than 10 ppb as com-

pared to earlier years. This trend was reversed in the first 6 months of 1972 when 44% of the residues were more than 10 ppb as compared to only 25% in 1971.

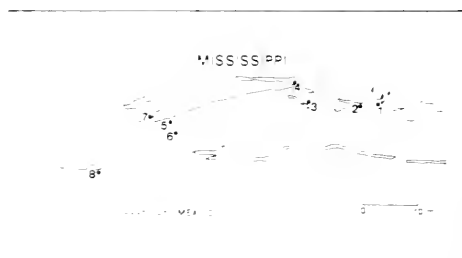


FIGURE H-1.—Diagram of coastal Mississippi showing approximate location of monitoring stations

1. Pascagoula—Pascagoula River
2. Graveline—Graveline Bay
3. Deer Island—Biloxi Bay
4. Biloxi Bay—Biloxi Bay
5. Pass Christian (Inshore)—Mississippi Sound
6. Pass Christian (Offshore)—Mississippi Sound
7. Bay St. Louis—St. Louis Bay
8. St. Joseph Point—Mississippi Sound

TABLE H-1.—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1965-72—Mississippi

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE (% DETECTED IN PPB (µg/kg))	
				DDT	DIELDRIN
1	Pascagoula	1965-72	78	47	74
2	Graveline	1965-72	79	56	99
3	Deer Island	1965-69	49	33	105
4	Biloxi Bay	1965-72	78	71	135
5	Pass Christian (Inshore)	1965-66	13	7	(53)
6	Pass Christian (Offshore)	1965-72	74	29	42
7	Bay St. Louis	1966-72	66	31	124
8	St. Joseph Point	1969-72	29	11	(69)
Total number of samples			470		
Percent of samples positive for indicated compound				61	4

¹ Each sample represents 15 or more mature mollusks

TABLE H-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Mississippi

[Blank = no sample collected; — = no residue detected above 5 ppb; T = >5 but <10 ppb]

YEAR	COMPOUND	RESIDUES IN PPB (µg/kg)											
		JAN.	FEB.	MAR.	APR.	MAY.	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—PASCAGOULA—78 SAMPLES ¹													
1965	DDE								T	T	T	14	—
	TDE								T	—	T	55	—
	DDT								T	—	—	T	—
1966	DDE	T	17		13	19	—	T	T	—	—	T	11
	TDE	T	41		10	19	—	T	—	—	—	—	12
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	13	12	11	14	T	—	—	—	T	T	T	—
	TDE	14	47	T	13	T	—	—	—	T	—	—	—
	DDT	T	10	—	—	—	—	—	—	—	—	—	—
1968	DDE	T	T	T	T	10	—	—	—	—	—	—	—
	TDE	—	T	T	T	11	—	—	—	—	—	—	—
	DDT	—	T	—	—	T	—	—	—	—	—	—	—
1969	DDE	T	—	11	T	15	T	T	—	—	11	12	—
	TDE	13	—	12	T	19	12	T	—	—	14	T	—
	DDT	—	—	—	—	T	—	—	—	—	T	—	—
1970	DDE	13	12	15	16	10	T	—	—	—	—	T	—
	TDE	40	12	18	14	13	64	—	—	—	—	T	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

TABLE H-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Mississippi—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—PASCAGOULA—78 SAMPLES ¹ —Continued													
1971	DDE	—	—	—	—	T	—	—	—	—	—	—	T
	TDE	—	—	—	—	12	55	—	—	—	—	—	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1972	DDE	T	T	—	10	11	—	—	—	—	—	—	—
	TDE	11	T	—	10	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 2.—GRAVELINE—79 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	—	T	T	T	T	T
	TDE	—	—	—	—	—	—	—	T	T	T	—	20
	DDT	—	—	—	—	—	—	—	T	—	—	—	—
1966	DDE	10	T	12	29	21	22	16	12	18	T	13	13
	TDE	27	T	10	10	60	68	19	31	69	13	17	36
	DDT	T	—	—	T	T	—	—	—	—	—	—	—
1967	DDE	14	12	23	24	T	T	T	—	T	12	14	15
	TDE	11	10	66	36	T	18	T	—	12	23	18	23
	DDT	13	—	10	T	—	—	—	—	21	29	10	T
1968	DDE	15	16	13	11	16	22	10	—	—	—	—	T
	TDE	22	23	19	18	23	25	14	—	T	—	—	—
	DDT	T	12	—	T	T	T	—	—	—	—	—	—
1969	DDE	—	—	11	T	15	T	—	—	—	—	11	17
	TDE	—	—	14	T	15	12	—	—	—	—	13	20
	DDT	—	—	—	—	T	—	—	—	—	—	—	10
1970	DDE	—	—	14	14	15	10	T	—	—	—	—	—
	TDE	—	—	16	17	14	11	10	—	—	—	—	—
	DDT	—	—	10	T	—	—	T	—	—	—	—	—
1971	DDE	15	—	—	—	12	—	—	—	—	—	—	T
	TDE	17	—	—	—	23	—	—	—	—	—	—	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1972	DDE	T	T	—	15	T	16	—	—	—	—	—	—
	TDE	T	T	—	—	T	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 3.—DEER ISLAND—49 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	—	10	T	—	T	T
	TDE	—	—	—	—	—	—	—	21	T	—	17	17
	DDT	—	—	—	—	—	—	—	T	—	—	—	—

TABLE H-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Mississippi—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 3.—DEER ISLAND—49 SAMPLES ¹ —Continued													
1966	DDE	14	20	15	22	27	23	17	T	—	T	—	T
	TDE	27	43	25	45	62	65	38	T	—	T	—	11
	DDT	T	16	T	12	16	—	—	—	—	—	—	—
1967	DDE	10	15	14	15	11	—	—	—	—	—	—	T
	TDE	12	T	14	11	17	—	—	—	—	—	—	T
	DDT	T	—	—	—	—	—	—	—	12	—	—	—
1968	DDE	T	11	T	12	12	—	—	—	—	—	—	—
	TDE	13	13	T	12	12	—	—	—	—	—	—	—
	DDT	—	T	—	—	—	—	—	—	—	—	—	—
1969	DDE	T	T	11	T	T	—	T	T	—	—	—	—
	TDE	T	—	13	13	T	—	27	34	—	—	—	—
	DDT	—	—	—	—	—	—	—	22	—	—	—	—
STATION 4.—BILOXI BAY—78 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	—	14	T	—	T	T
	TDE	—	—	—	—	—	—	—	23	11	—	23	18
	DDT	—	—	—	—	—	—	—	T	—	—	—	—
1966	DDE	14	16	16	29	32	—	20	19	—	T	15	T
	TDE	43	30	33	73	87	—	47	48	—	T	27	15
	DDT	14	T	11	15	16	—	—	—	—	—	T	—
1967	DDE	16	19	23	28	15	13	T	T	—	—	T	T
	TDE	31	40	43	50	33	21	43	25	17	17	19	22
	DDT	15	23	15	20	—	T	—	—	T	T	—	—
	Dieldrin	—	—	—	13	—	—	—	—	—	—	—	—
1968	DDE	12	14	16	20	30	17	T	—	—	—	T	16
	TDE	32	30	39	34	94	61	52	25	24	T	43	49
	DDT	—	T	—	T	T	—	—	—	—	—	—	—
1969	DDE	T	18	17	14	20	17	T	—	—	14	14	20
	TDE	28	46	47	46	67	54	22	25	—	42	28	53
	DDT	—	T	T	11	T	—	—	—	—	18	—	12
	Dieldrin	—	—	—	—	—	—	—	—	—	—	16	18
1970	DDE	—	—	18	19	28	—	—	—	—	—	—	—
	TDE	—	—	60	50	78	—	77	40	—	12	12	17
	DDT	—	—	15	T	14	—	—	—	—	—	—	—
	Dieldrin	—	—	19	16	15	—	11	—	16	—	—	—
1971	DDE	T	24	16	24	T	—	—	22	—	—	—	T
	TDE	26	65	43	69	85	—	—	81	—	—	19	27
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

TABLE H-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Mississippi—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 4.—BILOXI BAY—78 SAMPLES ¹ —Continued													
1972	DDE	T	10	12	18	17	T						
	TDE	20	28	35	58	63	30						
	DDT	—	—	—	—	—	—						
STATION 5.—PASS CHRISTIAN (INSHORE)—13 SAMPLES ¹													
1965	DDE								T	T	—	T	—
	TDE								T	—	—	—	—
	DDT								—	—	—	—	—
1966	DDE	T	—	—	T	11	19	—	—				
	TDE	T	—	—	T	12	34	—	—				
	DDT	—	—	—	—	11	—	—	—				
STATION 6.—PASS CHRISTIAN (OFFSHORE)—78 SAMPLES ¹													
1965	DDE								T	—	—	T	—
	TDE								T	—	—	T	—
	DDT								—	—	—	T	—
1966	DDE	T	—	—	11	14	—	—	—	—	—	—	—
	TDE	T	—	—	12	15	—	—	—	—	—	—	—
	DDT	—	—	—	T	13	—	—	—	—	—	—	—
1967	DDE	—	—	T	—	—	—	—	—	—	—	—	T
	TDE	—	—	T	—	—	—	—	—	—	—	—	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1968	DDE	T	T	T	T	T	—	—	—	—	—	—	—
	TDE	—	T	T	11	11	—	—	—	—	—	—	—
	DDT	—	T	—	—	—	—	—	—	—	—	—	—
1969	DDE		—	—	—	T	T	—	—	—	—	T	T
	TDE		—	—	—	T	13	—	—	—	—	T	T
	DDT		—	—	—	T	—	—	—	—	—	—	—
1970	DDE		12	13	15	11	T	—	—	—	—	—	—
	TDE		T	22	22	16	T	—	—	—	—	—	—
	DDT		—	T	—	T	—	—	—	—	—	—	—
	Dieldrin		11	16	—	—	—	—	—	15	—	—	—
1971	DDE	—	—	—	—	T	—	—	—	—	—	—	T
	TDE	—	—	—	—	14	—	—	—	—	—	—	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1972	DDE	T	T	12	12	T	11						
	TDE	T	T	10	16	T	—						
	DDT	—	—	—	T	—	—						

TABLE H-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Mississippi—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)												
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	Nov.	DEC.	
STATION 7.—BAY ST. LOUIS—66 SAMPLES ¹														
1966	DDE										—	—	—	—
	TDE										—	—	—	—
	DDT										—	—	—	—
1967	DDE	T	T	T	T	T	—	—	—	—	—	11	T	—
	TDE	T	T	14	T	T	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	10	—	—	—	—
1968	DDE	T	T	T	T	10	—	—	—	—	—	—	—	—
	TDE	T	T	T	T	11	—	—	—	—	—	—	—	—
	DDT	—	T	—	—	T	—	—	—	—	—	—	—	—
1969	DDE	—	T	T	—	T	T	—	—	—	—	T	10	—
	TDE	T	—	12	—	T	11	—	—	—	—	11	12	—
	DDT	—	—	—	—	T	—	—	—	—	—	T	11	—
	Dieldrin	T	14	11	T	—	—	—	—	—	—	—	—	—
1970	DDE		T	12	17	34	32	—	—	—	—	—	—	—
	TDE		13	15	21	76	12	—	—	—	—	—	—	—
	DDT		—	—	T	14	—	—	—	—	—	—	—	—
	Dieldrin		17	20	—	14	—	—	—	—	—	—	—	—
1971	DDE	—	—	—	—	—	—	—	—	—	—	—	T	—
	TDE	—	—	—	—	13	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	—
1972	DDE	T	T	—	11	T	—	—	—	—	—	—	—	—
	TDE	T	T	—	11	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	T	—	—	—	—	—	—	—	—	—
STATION 8.—ST. JOSEPH POINT—29 SAMPLES ¹														
1969	DDE												—	—
	TDE												—	—
	DDT												—	—
1970	DDE		T	T	T	14	15	—	—	—	—	—	—	—
	TDE		—	—	—	22	54	—	—	—	—	—	—	—
	DDT		—	—	—	—	—	—	—	—	—	—	24	—
	Dieldrin		18	—	—	—	—	—	—	—	—	—	—	—
1971	DDE	—	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	24	—
1972	DDE	T	T	—	—	T	T	—	—	—	—	—	—	—
	TDE	—	15	—	—	T	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	—

¹ Each sample represents 15 or more mature mollusks.

SECTION I.—NEW JERSEY

Samples of eastern oysters, *Crassostrea virginica*, were collected at five principal stations in the New Jersey waters of Delaware Bay during the period June 1966 - June 1972. All samples were analyzed at the Gulf Breeze Laboratory. The approximate station locations are shown in Fig. I-1. A summary of data on organochlorine residues in the monitored species, *C. virginica*, is presented in Table I-1, and the distribution of residues in this species for each sampling station by date of collection in Table I-2.

Oyster samples collected in Delaware Bay were characterized by a 100% incidence of DDT residues and a relatively high incidence (24%) of dieldrin residues as compared to other areas monitored.

The maximum DDT residue observed, 272 ppb, is low compared to that in many other estuaries; most residues, from New Jersey were less than half this amount. The fact that DDE was the principal component of these residues suggests that the pesticide had been metabolized in other links of the trophic web before its acquisition by the oyster.

DDT residues appear to have been somewhat higher in the 1968-69 period than earlier, but the 1971 data show a clear-cut trend towards decreased residue levels.

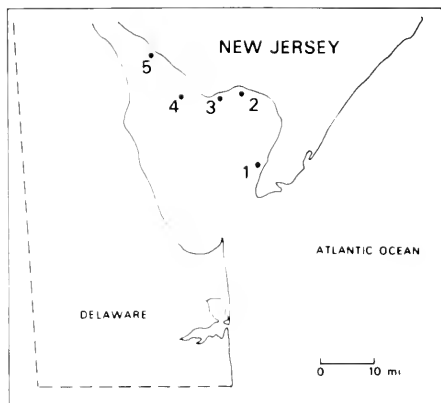


FIGURE I-1.—Diagram of coastal New Jersey showing approximate location of monitoring stations

1. Drum Beds—Delaware River
2. Maurice River—Delaware River
3. Dividing Creek—Delaware River
4. Lease 564/496D—Delaware River
5. Cohansey—Delaware River

TABLE I-1.—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1966-72—New Jersey

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (µg/kg)		
				DDT	DIELDRIN	PCB'S ²
1	Drum Beds	1966-72	49	49 (213)	3 (12)	2
2	Maurice River	1966-72	50	50 (143)	1 (T)	1
3	Dividing Creek	1966-71	7	7 (125)	1 (12)	
4	Lease 564/496D	1966-72	52	52 (278)	28 (26)	2
5	Cohansey	1966-72	49	49 (245)	16 (23)	1
	Occasional Stations (7)	1966-71	12	12 (166)	3 (29)	
Total number of samples			219			
Percent of samples positive for indicated compound				100	24	3

NOTE: T = >5 but <10 ppb.

¹ Each sample represents 15 or more mature mollusks.

² Present but not quantified.

TABLE I-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—New Jersey

[Blank = no sample collected; — = no residue detected above 5 ppb or no residue detected (PCB's); T = >5 but <10 ppb]

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{g}/\text{kg}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—DRUM BEDS—49 SAMPLES ¹													
1966	DDE						19		33	24		28	17
	TDE						18		38	24		26	14
	DDT						—		15	11		10	—
1967	DDE			43	39	30	31	27	18	27	37	42	
	TDE			34	28	27	30	40	25	33	46	50	
	DDT			11	T	—	—	21	16	13	22	14	
1968	DDE	43	50	55		50	110	110	55	44	75	52	
	TDE	43	43	54		57	98	83	38	35	48	44	
	DDT	10	T	T		—	T	13	13	T	11	—	
1969	DDE			49	19	48	51	73	52	63	67	56	58
	TDE			35	18	15	45	44	30	34	32	41	28
	DDT			—	—	—	—	—	T	—	T	T	T
1970	DDE			99			100	110	34	37		46	38
	TDE			42			52	47	12	15		21	16
	DDT			11			—	—	—	—		—	—
	Dieldrin			—			—	12	—	—		—	—
1971	DDE			35	59		70	38		27			53
	TDE			10	35		30	26		14			28
	DDT			—	—		—	—		—			—
1972	DDE				^a 10		^a 52						
	TDE				20		29						
	DDT				52		—						
	Dieldrin				T		T						
	PCB's				(b)		(b)						
STATION 2.—MAURICE RIVER—50 SAMPLES ¹													
1966	DDE						11		T	14		13	T
	TDE						15		12	15		16	T
	DDT						—		—	—		—	—
1967	DDE			12	12	13	15	12	T	18	26	19	
	TDE			T	T	19	28	23	10	30	40	31	
	DDT			17	—	—	—	T	—	13	13	—	
1968	DDE	19	24	22	21		45	37	17	20	16	24	
	TDE	27	31	32	43		68	38	21	24	17	19	
	DDT	T	—	—	—		—	16	T	10	T	T	

TABLE 1-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—New Jersey—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 2.—MAURICE RIVER—50 SAMPLES ¹ —Continued													
1969	DDE		21		16	77	75	13	22	12	T	19	13
	TDE		21		15	26	68	14	23	18	11	14	T
	DDT		—		—	—	—	—	T	—	—	—	—
1970	DDE			16		24	25	32	17	13	18		13
	TDE			T		22	36	34	16	15	19		10
	DDT			—		—	—	—	—	—	—		—
1971	DDE		12		T		26	23		14	T		
	TDE		T		—		29	29		13	—		
	DDT		—		—		—	—		—	—		
1972	DDE			14			^a 17						
	TDE			—			17						
	DDT			—			—						
	Dieldrin			—			T						
	PCB's			—			(^b)						
STATION 3.—DIVIDING CREEK—7 SAMPLES ¹													
1966	DDE										T		
	TDE										13		
	DDT										—		
1967	DDE				26			41					22
	TDE				28			66					33
	DDT				—			18					11
	Dieldrin				—			12					—
1968	DDE						49						
	TDE						64						
	DDT						T						
1969	DDE				60								
	TDE				35								
	DDT				—								
1970							No Samples Collected						
1971	DDE										13		
	TDE										13		
	DDT										—		
STATION 4.—LEASE 564/496D—52 SAMPLES ¹													
1966	DDE						34		29	41		51	58
	TDE						42		39	53		53	110
	DDT						—		T	12		67	15

TABLE I-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—New Jersey—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{g}/\text{kg}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	Nov.	DEC.
STATION 4.—LEASE 564/496D—52 SAMPLES ¹ —Continued													
1967	DDE			47	41	48	41	44	29	38	26	39	39
	TDE			48	39	86	81	72	47	56	41	56	56
	DDT			T	—	—	—	18	17	19	10	—	—
	Dieldrin			10	—	18	20	18	—	12	—	—	—
1968	DDE	35	38	37	23		110	77	51	69	67	57	
	TDE	50	54	60	45		140	82	46	57	57	56	
	DDT	T	T	T	—		18	23	16	T	T	T	
	Dieldrin	11	14	15	—		—	14	—	—	—	11	
1969	DDE		100		66	95	27	82	47	72	84	72	
	TDE		87		51	74	34	68	39	45	48	56	
	DDT		13		—	—	—	17	T	13	14	14	
	Dieldrin		13		12	18	21	—	—	19	13	11	
1970	DDE	130		140		180	75	35	62	95	120	42	12
	TDE	61		67		98	92	18	33	36	33	36	43
	DDT	15		16		—	—	—	—	—	—	—	—
	Dieldrin	14		16		20	26	—	—	—	—	—	12
1971	DDE		150		180		180	190		49			56
	TDE		45		87		62	78		25			18
	DDT		—		T		—	—		—			—
	Dieldrin		13		19		19	T		—			—
1972	DDE			* 67			* 46						
	TDE			42			35						
	DDT			11			—						
	Dieldrin			T			14						
	PCB's			(8)			(5)						
STATION 5.—COHANSEY—49 SAMPLES ¹													
1966	DDE						12		17		20	22	11
	TDE						24		37		35	33	23
	DDT						—		—		—	—	—
1967	DDE			29	35	41	54	24	29	17	17	32	
	TDE			45	53	19	150	59	66	39	38	59	
	DDT			T	T	22	25	10	13	—	T	—	
	Dieldrin			—	—	23	18	12	T	—	—	11	
1968	DDE	20	30	22		49	81	68	32	33	52	28	
	TDE	41	62	66		12	150	110	60	50	70	32	
	DDT	—	—	—		—	14	11	15	T	T	—	
	Dieldrin	—	12	14		13	19	12	—	—	—	—	

TABLE 1-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—New Jersey—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 5—COHANSEY—49 SAMPLES ¹ —Continued													
1969	DDE		33		59	57	36	37	29	24	42	38	
	TDE		46		43	76	49	46	45	44	48	42	
	DDT		—		—	—	—	T	T	—	—	T	
	Dieldrin		—		—	16	—	—	—	—	—	—	
1970	DDE			45	41	55		30	27	53	36	42	
	TDE			46	51	98		30	34	54	42	36	
	DDT			—	—	—		T	—	—	—	—	
	Dieldrin			—	11	21		—	—	—	—	—	
1971	DDE		52		28	38		27		21			22
	TDE		42		24	61		30		20			17
	DDT		—		—	T		—		—			—
	Dieldrin		13		—	16		—		—			—
1972	DDE			23		* 40							
	TDE			114		49							
	DDT			—		—							
	Dieldrin			—		T							
	PCB's			—		(^b)							

¹ Each sample represents 15 or more mature mollusks.

² DDT values are approximate because of presence of unidentified PCB's.

³ Present but not quantified.

SECTION J.—NEW YORK

Several different species of mollusks (*Crassostrea virginica*, *Modiolus domissus*, *Mytilus edulis*, *Mercenaria mercenaria*, and *Mya arenaria*) were collected at 16 principal sites in New York's coastal waters to monitor organochlorine pollution during the period March 1966 - June 1972. Samples were analyzed at the Gulf Breeze Laboratory until February 1969 and thereafter by the New York Conservation Department. Analyses of aliquots of some of the samples collected during the period October 1968 - July 1970 have been reported by the cooperating agency (9) and do not differ significantly from the data reported here.

Approximate station locations are shown in Fig. J-1. A summary of data on organochlorine residues in the monitored species is presented in Table J-1, and the distribution of residues in these species for each sampling station by date of collection in Table J-2.

The hard clam, *M. mercenaria*, was the principal species collected because of its ubiquity and despite its recognized inefficiency in storing organochlorine residues. This lack of sensitivity to low levels of DDT pollution is especially well documented in the analytical record of samples collected in Conscience Bay, Station 6. Hard clams were the only mollusk of four species collected there in which DDT residues were undetected. DDT pollution apparently disappeared at this station during the period July 1968 - March 1969, but this was because of the substitution of hard clams for the blue mussel as monitors.

These data emphasize the fact that in areas where hard clams did show DDT residues, there were probably significant levels of DDT in the water or food supply. This parallels the situation in Delaware, where hard clams collected in Delaware Bay (Cape Henlopen) consistently had DDT residues while residues were



FIGURE J-1.—Diagram of coastal New York showing approximate location of monitoring stations

- | | |
|---------------------------|---------------------------------|
| 1. Mamaroneck | 9. Mecox Bay |
| 2. Hempstead Harbor | 10. Shinnecock Bay |
| 3. Oyster Bay Harbor | 11. Moriches Bay |
| 4. Huntington Bay | 12. Bellport—Great South Bay |
| 5. Nissequogue River | 13. Sayville—Great South Bay |
| 6. Conscience Bay | 14. Amityville—South Oyster Bay |
| 7. Southold—Gardiners Bay | 15. East Bay |
| 8. Flanders Bay | 16. West Bay |

usually not detected in hard clams collected in inner bays. There was generally good agreement in the magnitude of residues in two or more species, other than the hard clam, collected at the same station on the same day.

The New York samples ranked fifth among the States in incidence and sixth in magnitude of DDT residues. More samples (43%) contained dieldrin residues than in any other area monitored. PCB's were present in some samples in 1972, but they were not identified or quantified.

Despite the large number of samples collected over a period of 7 years, no clearly defined trends in pollution levels can be identified. This may be the result of having used a variety of species. The overall impression is one of no significant change in DDT residue levels in mollusks.

TABLE J-1.—Summary of data on organochlorine residues in the monitored species, 1966-72—New York

STATION NUMBER	LOCATION	MONITORING PERIOD	PRINCIPAL MONITORED SPECIES	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB ($\mu\text{g}/\text{kg}$)	
					DDT	DIELDRIN
1	Mamaroneck	1966-69	<i>M. mercenaria</i>	36	34 (96)	27 (29)
2	Hempstead Harbor	1966-72	<i>M. mercenaria</i>	74	70 (201)	61 (132)
3	Oyster Bay	1966-72	<i>M. mercenaria</i>	73	54 (99)	48 (86)
4	Huntington Bay	1966-72	<i>M. edulis</i>	74	72 (588)	52 (104)
5	Nissequogue River	1966-72	<i>M. edulis</i>	74	70 (138)	58 (117)
6	Conscience Bay	1966-72	<i>M. edulis</i>	73	61 (112)	52 (75)
7	Southold	1969-72	<i>C. virginica</i>	34	32 (149)	26 (78)
8	Flanders Bay	1966-72	<i>M. mercenaria</i>	69	63 (199)	15 (107)
9	Mecox Bay	1966-72	<i>C. virginica</i>	67	65 (596)	14 (22)

TABLE J-1.—Summary of data on organochlorine residues in the monitored species, 1966-72—New York—Continued

STATION NUMBER	LOCATION	MONITORING PERIOD	PRINCIPAL MONITORED SPECIES	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (µG/KG)	
					DDT	Dieldrin
10	Shinnecock Bay	1966-72	<i>M. mercenaria</i>	73	43 (188)	19 (46)
11	Moriches Bay	1966-72	<i>M. mercenaria</i>	71	49 (83)	13 (49)
12	Bellport Bay	1966-72	<i>M. mercenaria</i>	71	51 (132)	10 (53)
13	Sayville	1966-72	<i>M. mercenaria</i>	74	41 (107)	16 (59)
14	Amityville	1966-72	<i>M. mercenaria</i>	73	49 (64)	13 (42)
15	East Bay	1966-72	<i>M. mercenaria</i>	57	43 (98)	13 (38)
16	West Bay	1966-72	<i>M. mercenaria</i>	57	51 (111)	19 (20)
	Occasional stations (8)	1967-72	Mixed	9	9 (159)	3 (31)
Total number of samples				1,059		
Percent of samples positive for indicated compound					81	43

¹ Each sample represents 15 or more mature mollusks.

TABLE J-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—New York

[Blank = no sample collected; — = no residue detected above 5 ppb; T = >5 but <10 ppb]

YEAR	COMPOUND	RESIDUES IN PPB (µG/KG)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—MAMARONECK— <i>M. MERCENARIA</i> —36 SAMPLES ¹													
1966	DDE				—	T	—	—	—	—	T	16	T
	TDE				—	T	—	13	T	12	18	49	29
	DDT				—	—	—	—	—	—	—	14	13
	Dieldrin				—	—	—	—	—	—	—	29	12
1967	DDE	T	19	12	11	11	11	T	T	T	12	T	T
	TDE	35	50	31	27	27	28	20	15	26	28	32	30
	DDT	24	27	11	11	11	15	T	T	—	T	14	T
	Dieldrin	16	21	16	16	15	15	15	14	14	13	14	15
1968	DDE	T	T	T	11	10	T	—	—	—	—	—	T
	TDE	27	25	27	30	27	26	22	20	20	18	23	19
	DDT	T	—	—	T	12	15	T	—	—	—	—	—
	Dieldrin	11	12	14	11	12	—	10	12	12	11	15	14
1969	DDE	—	10	—									
	TDE	18	24	13									
	DDT	—	—	—									
	Dieldrin	11	11	—									

TABLE J-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—New York—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 2.—HEMPSTEAD HARBOR— <i>M. MERCENARIA</i> . UNLESS OTHERWISE INDICATED—74 SAMPLES ¹													
1966	DDE			—	—	—	T	—	² 24	T	11	15	13
	TDE			—	—	—	29	—	48	17	29	39	35
	DDT			—	—	—	13	—	26	—	—	17	14
	Dieldrin			—	—	—	—	—	—	—	—	17	15
1967	DDE	T	14	13	12	12	14	T	T	12	13	12	10
	TDE	17	36	35	30	33	39	23	25	29	30	32	30
	DDT	—	13	13	11	15	28	15	16	—	T	11	11
	Dieldrin	—	15	16	17	15	19	15	17	15	16	14	16
1968	DDE	11	13	13	13	10	T	—	—	—	T	—	T
	TDE	29	31	31	34	26	27	17	18	17	24	22	23
	DDT	11	—	10	10	10	—	T	—	—	—	—	—
	Dieldrin	12	16	14	15	20	16	—	—	11	50	93	66
1969	DDE	—	10	² 15	² 16	18	15	T	—	² 34	² 30	² 23	² 26
	TDE	12	29	33	34	18	19	22	10	93	62	57	57
	DDT	—	T	34	38	12	T	T	—	74	49	47	46
	Dieldrin	47	70	22	86	85	13	33	38	132	28	40	31
1970	DDE	² 41	11	² 24	² 13	² 12	² 18	10	10	² 21	² 18	² 18	² 20
	TDE	71	33	51	28	33	48	22	35	71	40	48	50
	DDT	76	15	62	29	54	70	17	18	28	35	43	51
	Dieldrin	29	30	30	25	30	33	20	18	26	22	19	25
1971	DDE	² 10	² 13	² 13	² 15	² 20	—	² 16	—	² 13	² 22	² 23	
	TDE	22	26	33	38	27	—	16	37	—	31	46	58
	DDT	18	31	34	44	23	—	10	30	—	17	32	51
	Dieldrin	10	14	16	31	—	—	13	21	—	19	23	20
1972	DDE	² 19	² 17	² 23	² 34	² 9	—	—	—	—	—	—	—
	TDE	41	—	48	67	15	T	—	—	—	—	—	—
	DDT	22	24	44	53	9	—	—	—	—	—	—	—
	Dieldrin	19	14	19	21	—	T	—	—	—	—	—	—
STATION 3.—OYSTER BAY HARBOR— <i>M. MERCENARIA</i> . UNLESS OTHERWISE INDICATED—73 SAMPLES ¹													
1966	DDE			—	—	—	—	—	—	—	—	T	T
	TDE			—	—	—	—	—	T	—	—	22	11
	DDT			—	—	13	—	—	—	—	—	T	—
	Dieldrin			—	—	—	—	—	—	—	—	13	11
1967	DDE	13	T	T	T	T	T	—	T	T	T	T	T
	TDE	50	15	19	14	20	22	26	17	13	T	13	15
	DDT	18	—	—	—	—	13	T	12	—	—	—	—
	Dieldrin	10	—	14	13	14	14	11	14	—	12	11	—

TABLE J-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—New York—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 3.—OYSTER BAY HARBOR— <i>M. MERCENARIA</i> . UNLESS OTHERWISE INDICATED—73 SAMPLES ¹ —Continued													
1968	DDE	T	T	—	T	T	—	—	—	—	—	—	—
	TDE	16	T	—	T	11	—	T	—	—	—	—	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	—	11	—	—	—	—	—	—	—	—	10
1969	DDE	—	T	—	^a 11	^a 24	—	—	—	—	^a 27	T	—
	TDE	—	T	—	25	28	11	15	—	—	55	13	T
	DDT	—	—	—	16	32	—	T	—	—	17	—	—
	Dieldrin	T	T	12	17	24	86	16	—	17	—	15	18
1970	DDE	—	^a 27	^a 10	—	^a 21	T	^a 16	T	—	^a 22	^a 27	^a 24
	TDE	—	52	19	—	44	16	37	18	—	48	55	50
	DDT	—	20	11	—	34	T	24	T	—	20	15	16
	Dieldrin	—	31	12	37	23	11	19	16	—	29	30	26
1971	DDE	^a 19	^a 19	^a 22	—	^a 13	—	—	—	^a 19	^a 23	^a 19	^a 23
	TDE	38	38	45	T	32	—	T	—	30	41	34	44
	DDT	16	T	15	—	18	—	—	—	13	10	17	18
	Dieldrin	18	25	27	16	18	—	10	—	21	20	14	17
1972	DDE	^a 18	^a 19	—	—	^a 10	^a 13	—	—	—	—	—	—
	TDE	36	31	T	—	24	22	—	—	—	—	—	—
	DDT	24	18	—	—	10	11	—	—	—	—	—	—
	Dieldrin	12	15	—	10	11	19	—	—	—	—	—	—
STATION 4.—HUNTINGTON BAY— <i>M. EDULIS</i> . UNLESS OTHERWISE INDICATED—74 SAMPLES ¹													
1966	DDE	—	—	^b T	^b —	^b 32	98	53	40	^b —	^b —	^b 21	^b 29
	TDE	—	—	16	—	75	280	190	110	—	T	64	71
	DDT	—	—	—	—	81	210	60	25	—	—	17	16
	Dieldrin	—	—	—	—	—	—	18	13	—	—	12	T
1967 ^a	DDE	20	27	21	19	19	16	T	13	12	T	T	17
	TDE	88	65	54	57	54	63	50	40	39	27	22	56
	DDT	44	20	17	12	24	26	13	11	—	—	—	12
	Dieldrin	21	—	11	11	—	12	12	12	11	—	—	12
1968 ^a	DDE	T	—	12	16	15	12	—	—	—	T	T	10
	TDE	27	40	49	48	46	35	35	49	43	47	60	48
	DDT	—	T	—	T	14	—	T	—	—	—	T	11
	Dieldrin	11	11	—	12	—	—	—	—	—	—	11	T
1969	DDE	^b —	^b 11	^b T	18	^b T	^b 30	29	^b 15	34	24	28	30
	TDE	26	28	32	35	24	104	100	23	127	73	84	80
	DDT	—	—	—	21	21	46	50	—	47	28	55	50
	Dieldrin	T	T	T	11	14	104	26	T	18	—	23	34

TABLE J-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—New York—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{g}/\text{kg}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 4.—HUNTINGTON BAY— <i>M. EDULIS</i> , UNLESS OTHERWISE INDICATED—74 SAMPLES ¹ —Continued													
1970	DDE	38	15	23	—	20	13	21	20	19	20	24	21
	TDE	90	58	60	22	50	59	146	70	40	66	89	73
	DDT	61	40	49	12	40	40	55	29	44	28	28	29
	Dieldrin	18	16	17	38	22	21	26	21	26	22	17	17
1971	DDE	21	18	15	11	21	—	15	26	—	33	21	19
	TDE	70	57	50	37	51	—	63	74	—	14	56	60
	DDT	35	30	24	22	22	—	22	27	—	T	23	28
	Dieldrin	17	18	22	14	—	—	21	14	—	13	—	14
1972	DDE	—	14	17	15	10	14	—	—	—	—	—	—
	TDE	T	33	40	36	20	41	—	—	—	—	—	—
	DDT	—	20	15	18	—	15	—	—	—	—	—	—
	Dieldrin	—	13	15	12	—	60	—	—	—	—	—	—
STATION 5.—NISSEQUOGUE RIVER— <i>M. EDULIS</i> , UNLESS OTHERWISE INDICATED—74 SAMPLES ¹													
1966	DDE	—	—	21	21	24	18	T	19	17	22	42	40
	TDE	—	—	33	47	45	49	17	43	38	48	76	59
	DDT	—	—	23	37	50	34	13	30	29	22	20	30
	Dieldrin	—	—	16	20	23	—	—	—	—	—	31	17
1967	DDE	—	23	24	21	23	21	18	14	20	19	17	18
	TDE	33	53	59	50	18	51	42	38	49	42	35	44
	DDT	13	27	29	33	45	42	33	32	33	27	18	32
	Dieldrin	13	17	27	27	27	22	18	19	15	15	14	17
1968	DDE	14	16	13	17	17	13	11	—	5	5	6	6
	TDE	32	33	36	40	44	49	44	31	—	—	—	T
	DDT	20	18	21	23	42	51	47	25	—	—	—	—
	Dieldrin	14	18	16	18	—	—	—	—	—	—	—	12
1969	DDE	6	T	T	12	T	5	15	5	10	14	18	20
	TDE	—	T	14	30	12	T	22	T	34	35	49	42
	DDT	—	—	T	22	11	T	14	—	17	30	36	25
	Dieldrin	T	T	T	16	12	11	—	T	24	21	20	20
1970	DDE	16	17	T	—	21	14	18	15	T	14	11	T
	TDE	37	44	18	16	46	37	48	46	17	38	29	14
	DDT	25	33	T	T	38	26	34	30	—	22	17	—
	Dieldrin	27	17	10	16	23	20	24	25	T	24	12	T
1971	DDE	14	T	T	5	—	—	10	17	—	14	11	5
	TDE	30	16	17	15	12	—	38	T	—	10	34	T
	DDT	24	12	T	T	—	—	20	—	—	—	13	—
	Dieldrin	14	T	17	20	T	—	27	—	—	13	10	—

TABLE J-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—New York—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 5.—NISSEQUOGUE RIVER— <i>M. EDULIS</i> , UNLESS OTHERWISE INDICATED—74 SAMPLES ¹ —Continued													
1972	DDE	¹ —	15	T	² —	T	T						
	TDE	12	33	12	T	20	14						
	DDT	—	18	T	—	10	T						
	Dieldrin	10	13	T	—	T	12						
STATION 6.—CONSCIENCE BAY— <i>M. EDULIS</i> , UNLESS OTHERWISE INDICATED—73 SAMPLES ¹													
1966	DDE			³ T	³ —	³ —	⁵ 18	⁵ —	⁵ —	20	21	24	⁶ 17
	TDE			18	—	—	26	—	—	35	34	46	40
	DDT			15	12	—	15	—	—	22	22	30	17
	Dieldrin			—	12	—	—	—	—	—	—	25	—
1967	DDE	18	23	18	18	20	22	21	21	20	18	17	16
	TDE	48	43	33	34	42	44	48	44	36	31	29	31
	DDT	24	23	18	21	29	41	36	35	24	21	18	20
	Dieldrin	17	20	20	30	24	22	21	16	13	15	14	13
1968	DDE	14	16	14	15	T	12	⁶ —	⁶ —	⁶ —	⁶ —	⁶ —	⁶ —
	TDE	23	24	22	27	24	24	—	—	—	—	—	—
	DDT	14	13	17	19	21	21	—	—	—	—	—	—
	Dieldrin	15	14	14	14	—	—	—	—	—	—	—	T
1969	DDE	⁷ —	⁷ —	⁷ —	16	15	⁶ 16	⁸ 18	⁸ 14	⁸ 13	17	23	21
	TDE	—	—	—	23	24	36	22	20	14	29	39	36
	DDT	—	—	—	14	32	48	20	—	T	15	26	17
	Dieldrin	—	T	T	14	15	—	T	—	15	26	18	13
1970	DDE		17	22	¹ —	25	21	24	26	⁹ —	24	23	18
	TDE		37	37	11	49	45	59	59	17	46	37	26
	DDT		25	36	—	36	29	28	27	—	24	15	14
	Dieldrin		16	19	20	26	20	23	23	T	16	T	10
1971	DDE	⁹ T	⁹ T	¹ —	16	⁵ 10		⁶ T	⁸ —	⁸ —	T	12	⁸ T
	TDE	12	15	13	29	15		24	13	13	16	23	11
	DDT	T	10	—	13	—		10	—	T	T	11	—
	Dieldrin	T	14	22	18	—		10	10	17	T	T	11
1972	DDE	⁴ —		—	⁴ —	—	13						
	TDE	10		18	T	11	21						
	DDT	—		14	—	—	—						
	Dieldrin	—		T	—	75	49						

TABLE J-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—New York—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 7.—SOUTHOLD—C. VIRGINICA, UNLESS OTHERWISE INDICATED—34 SAMPLES ¹													
1969	DDE				^a 22	^b T	^c —	^d 22		^e T	^f T		^g 20
	TDE				48	11	T	38		14	T		41
	DDT				28	—	—	89		—	—		63
	Dieldrin				—	78	T	—		—	—		T
1970	DDE		^a 16	29	27	27	^b 10	21	19		17	22	20
	TDE		32	30	32	35	13	18	19		21	21	21
	DDT		21	26	23	22	—	22	23		18	15	12
	Dieldrin		—	21	20	26	T	11	27		14	15	14
1971	DDE	^a T	17	17	18	^b —		10	^c —	^d —	^e —	^f 15	^g —
	TDE	16	19	19	17	—		10	T	T	—	18	16
	DDT	—	14	11	12	—		13	—	—	—	13	—
	Dieldrin	21	12	14	16	14		—	—	12	—	T	T
1972	DDE	16	17	18	16	^b 10	21						
	TDE	27	17	22	21	18	28						
	DDT	16	T	T	11	—	31						
	Dieldrin	14	11	T	10	T	39						
STATION 8.—FLANDERS BAY—M. MERCENARIA, UNLESS OTHERWISE INDICATED—69 SAMPLES ¹													
1966	DDE			56	28	22	24	—	11	T	T	10	17
	TDE			54	25	21	44	—	23	14	—	15	23
	DDT			89	29	15	—	—	—	—	—	—	—
1967	DDE	15	18	25	23	^b 23	^b 20	17	17	16	17	16	15
	TDE	20	27	42	32	58	53	46	47	36	42	37	33
	DDT	—	—	—	—	12	12	—	T	—	—	—	—
1968	DDE	13	10	12	17	10	19	10	T	10	—	T	10
	TDE	25	17	25	41	24	59	44	38	38	18	39	34
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1969	DDE	—	T	12	^a 33	15	14	10	12	^a 20	—	—	13
	TDE	12	32	25	84	33	28	19	23	49	T	—	28
	DDT	—	—	—	27	—	—	—	—	75	—	—	—
	Dieldrin	—	—	—	—	17	107	11	—	14	93	—	—
1970	DDE			14	^a T	13	^b 14	^b 14			^c T	T	^d T
	TDE			26	22	30	29	29			19	T	14
	DDT			—	12	—	—	T			—	—	—
	Dieldrin			11	—	—	—	T			T	—	—

TABLE J-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—New York—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{g}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 8.—FLANDERS BAY— <i>M. MERCENARIA</i> , UNLESS OTHERWISE INDICATED—69 SAMPLES ¹ —Continued													
1971	DDE	—	T	—	T	^a 16	—	^a —	T	—	13	^a —	—
	TDE	10	16	—	16	50	—	16	12	—	24	—	11
	DDT	—	—	—	—	15	—	—	—	—	—	—	—
	Dieldrin	10	T	12	13	T	—	—	—	—	T	—	—
1972	DDE	T	—	—	—	—	—	—	—	—	—	—	—
	TDE	12	—	—	10	T	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	T	—	—	—	—	—	—	—	—	—	—	—
STATION 9.—MECOX BAY— <i>C. VIRGINICA</i> , UNLESS OTHERWISE INDICATED—67 SAMPLES ¹													
1966	DDE	—	—	300	120	^a 22	^a T	^a 27	^a 21	^a —	46	42	83
	TDE	—	—	240	120	41	13	45	29	—	46	37	83
	DDT	—	—	56	22	20	—	19	—	—	12	—	—
1967	DDE	63	67	77	T	^a 37	190	^a 15	49	64	69	93	110
	TDE	66	60	62	11	67	180	22	52	74	74	100	73
	DDT	—	—	—	—	29	27	T	18	17	11	15	T
1968	DDE	—	100	53	130	150	130	68	32	34	37	75	30
	TDE	—	81	48	87	120	85	48	29	18	32	50	18
	DDT	—	—	—	20	38	48	T	—	—	—	—	—
1969	DDE	44	62	26	60	^a 20	^a 21	^a 19	^a T	^a 10	^a —	^a 12	—
	TDE	31	45	20	39	31	29	21	10	10	—	23	—
	DDT	—	—	—	T	24	10	—	—	—	—	—	—
	Dieldrin	—	—	—	—	12	—	—	22	—	—	—	—
1970	DDE	^a 19	^a 21	16	32	^a 16	21	—	—	22	36	^a T	^a T
	TDE	27	27	14	45	36	25	—	—	38	48	10	12
	DDT	52	49	10	—	32	T	—	—	13	13	—	—
	Dieldrin	—	—	—	—	T	10	—	—	—	16	—	10
1971	DDE	—	—	30	41	34	—	T	14	37	—	—	35
	TDE	—	—	24	32	34	—	10	15	40	—	—	46
	DDT	—	—	—	T	T	—	—	—	15	—	—	T
	Dieldrin	—	—	—	12	T	—	—	10	19	—	—	—
1972	DDE	45	37	26	34	26	35	—	—	—	—	—	—
	TDE	52	39	26	40	27	38	—	—	—	—	—	—
	DDT	13	T	—	T	—	12	—	—	—	—	—	—
	Dieldrin	10	T	10	—	T	—	—	—	—	—	—	—

TABLE J-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—New York—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)												
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.	
STATION 10.—SHINNECOCK BAY— <i>M. MERCENARIA</i> . UNLESS OTHERWISE INDICATED—73 SAMPLES														
1966	DDE			—	—	—	^a T	—	T	—	—	—	T	
	TDE			—	—	—	T	—	14	—	—	—	13	
	DDT			—	—	—	—	—	—	—	—	—	—	
1967	DDE	T	T	T	59	T	T	—	T	—	T	—	—	
	TDE	12	12	12	50	T	T	—	T	—	T	—	—	
	DDT	—	—	—	—	—	T	—	—	—	—	—	—	
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—	
	TDE	—	—	—	—	—	—	—	—	—	10	—	T	
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	
1969	DDE	—	—	—	T	^a 20	—	—	^a 12	^a 14	^a 10	^a —	^a 13	
	TDE	—	T	—	T	38	T	T	21	20	16	—	24	
	DDT	—	—	—	—	21	—	—	12	T	T	—	22	
	Dieldrin	—	—	—	—	14	T	—	—	T	46	—	—	
1970	DDE	^a 11	^a —	^a —	^a —	^a 10	^a 18	^a 10	^a 12	—	^a —	^a —	^a T	
	TDE	18	13	T	—	19	34	18	22	—	—	T	11	
	DDT	—	—	—	—	T	21	24	20	T	—	—	—	
	Dieldrin	14	—	12	—	—	12	T	T	—	12	—	—	
1971	DDE	^a 10	^a T	^a —	^a —	^a —	^a —	^a —	^a —	^a —	^a —	^a —	^a —	
	TDE	17	18	10	10	—	—	—	T	—	—	—	T	
	DDT	12	14	T	—	—	—	—	—	—	—	—	—	
	Dieldrin	—	10	10	—	—	—	—	44	18	T	T	—	
1972	DDE	^a —	^a —	^a —	^a 11	^a —	^a 11	—	—	—	—	—	—	
	TDE	T	T	—	22	11	165	—	—	—	—	—	—	
	DDT	—	—	—	12	T	12	—	—	—	—	—	—	
STATION 11.—MORICHES BAY— <i>M. MERCENARIA</i> . UNLESS OTHERWISE INDICATED—71 SAMPLES ¹														
1966	DDE			—	—	—	T	—	—	—	—	—	^a 15	T
	TDE			—	—	—	10	—	—	—	—	—	33	14
	DDT			—	—	—	—	—	—	—	—	—	—	—
1967	DDE	T	T	T	T	T	T	T	T	T	T	T	—	
	TDE	13	14	18	13	18	17	10	T	17	T	18	—	
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	
1968	DDE	T	—	T	T	—	—	—	—	—	—	—	—	
	TDE	10	—	14	T	—	—	—	T	—	12	T	T	
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	
1969	DDE	—	—	—	—	—	—	—	—	—	—	—	—	
	TDE	—	11	—	—	13	—	T	T	—	—	—	T	
	DDT	—	—	—	—	—	—	—	—	—	24	—	—	

TABLE J-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—New York—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 11.—MORICHES BAY— <i>M. MERCENARIA</i> . UNLESS OTHERWISE INDICATED—71 SAMPLES ¹ —Continued													
1970	DDE		—	T	—	T	T	^a T	^a 20		^a 25	^a 25	^a 13
	TDE		T	13	—	14	20	21	40		29	19	13
	DDT		—	T	—	—	—	11	23		17	17	T
	Dieldrin		—	—	—	—	T	—	—		14	T	—
1971	DDE		T	^a 20	^a 18	—	—	—	—		^a 22	^a 10	^a 21
	TDE		10	27	25	11	—	—	T		15	26	34
	DDT		—	15	14	—	—	—	—		17	T	25
	Dieldrin		T	T	16	—	—	—	17	18	—	T	T
1972	DDE	^a 17	^a 16	^a 17	^a 20	^a 17	^a 25						
	TDE	27	20	27	26	13	33						
	DDT	19	T	—	15	—	22						
	Dieldrin	T	—	—	T	—	49						
STATION 12.—BELLPORT BAY— <i>M. MERCENARIA</i> . UNLESS OTHERWISE INDICATED—71 SAMPLES ¹													
1966	DDE			57	—	—	12	T	T	T	14	16	10
	TDE			44	—	10	27	18	13	16	28	30	20
	DDT			31	—	—	T	—	—	—	—	—	—
1967	DDE	20	21	24	19	12	12	11	10	15	T	13	14
	TDE	42	40	45	36	30	31	25	19	26	20	22	27
	DDT	—	—	—	—	T	—	—	T	—	—	—	—
1968	DDE	15	13	15	30	10	T	T	—	T	—	—	—
	TDE	28	24	27	50	23	14	12	—	T	—	—	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1969	DDE	—	T	—	^a 25	T	—	T	—	—	—	—	—
	TDE	—	10	—	50	10	15	18	—	—	—	—	—
	DDT	—	—	—	33	—	—	—	—	—	—	—	—
1970	DDE			T	—	11	10	T	T	—	—	—	—
	TDE			13	—	26	17	21	17	—	11	—	—
	DDT			—	—	—	T	—	—	—	—	—	—
	Dieldrin			—	—	—	T	—	—	37	T	—	—
1971	DDE	—	T	T	—	—	—	—	—	—	—	—	—
	TDE	10	12	11	13	10	—	—	—	—	—	—	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	—	12	T	T	—	T	—	10	—	—	53
1972	DDE	—	—	—	—	—	T	—	—	—	—	—	—
	TDE	—	T	T	T	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	16	—	—	—	—	—	—	—	—	—	—

TABLE 1-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—New York—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G KG}$)												
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	Nov.	DEC.	
STATION 13.—SAYVILLE— <i>M. MERCENARIA</i> . UNLESS OTHERWISE INDICATED—74 SAMPLES ¹														
1966	DDE			—	—	—	T	—	—	—	—	—	T	
	TDE			—	—	T	10	—	—	—	—	—	10	
	DDT			—	—	T	T	—	—	—	—	—	—	
1967	DDE	T	T	11	T	T	T	T	T	T	—	T	T	
	TDE	14	19	24	16	24	14	T	10	T	—	16	19	
	DDT	T	—	—	—	T	—	—	T	—	—	—	—	
1968	DDE	—	—	—	—	—	—	T	—	—	—	—	—	
	TDE	—	—	—	—	—	—	T	—	—	—	—	—	
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	
1969	DDE	—	—	—	—	—	—	—	—	—	—	4 11	T	
	TDE	—	—	—	—	—	—	—	—	T	T	24	13	
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	
	Dieldrin	—	—	—	—	—	—	—	—	59	—	19	12	
1970	DDE	—	—	—	—	—	T	—	T	1	10	—	T	
	TDE	T	13	11	—	T	16	T	14	16	12	T	20	
	DDT	—	—	—	—	—	T	10	—	T	T	—	10	
	Dieldrin	—	T	—	—	—	T	—	—	—	T	10	T	
1971	DDE	T	—	—	—	—	—	—	—	—	—	—	—	
	TDE	12	T	11	11	10	—	—	—	—	—	—	16	
	DDT	—	—	—	—	—	—	—	—	—	—	—	12	
	Dieldrin	T	T	—	—	—	—	13	—	—	41	T	11	
1972	DDE	4 22	4 22	—	17	—	—	—	—	—	—	—	—	
	TDE	56	38	—	16	—	—	—	—	—	—	—	—	
	DDT	29	T	T	10	—	T	—	—	—	—	—	—	
	Dieldrin	15	—	—	11	—	—	—	—	—	—	—	—	
STATION 14.—AMITYVILLE— <i>M. MERCENARIA</i> —73 SAMPLES														
1966	DDE			—	—	—	—	—	—	—	—	—	T	T
	TDE			—	—	—	18	—	—	—	—	—	10	17
	DDT			—	—	—	—	—	—	—	—	—	—	—
1967	DDE	T	T	T	T	T	T	—	—	—	—	—	T	—
	TDE	20	15	16	16	18	14	17	13	T	11	14	—	
	DDT	T	—	—	—	T	—	—	T	—	—	—	—	
1968	DDE	—	—	—	T	—	—	T	—	T	—	—	—	
	TDE	—	—	—	13	—	—	11	T	T	—	13	T	
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	

TABLE J-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—New York—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 14.—AMITYVILLE—M. MERCENARIA—73 SAMPLES ¹ —Continued													
1969	DDE	—	—	—	—	—	—	—	—	—	—	20	—
	TDE	—	T	—	T	14	—	T	—	11	T	11	—
	DDT	—	—	—	—	—	—	—	—	—	T	33	—
	Dieldrin	—	—	—	—	T	—	—	—	T	—	T	T
1970	DDE	—	—	—	—	10	T	T	T	—	—	—	—
	TDE	—	11	—	T	20	19	15	15	10	T	T	10
	DDT	—	—	—	—	11	—	—	—	—	—	—	—
	Dieldrin	—	—	T	—	T	—	—	—	—	18	—	T
1971	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	11	12	T	16	—	—	T	T	10	—	10
	DDT	—	—	—	—	—	—	—	—	T	—	—	—
	Dieldrin	—	—	—	10	10	—	—	—	T	—	42	—
1972	DDE	T	—	—	—	—	—	—	—	—	—	—	—
	TDE	16	T	T	T	—	—	—	—	—	—	—	—
	DDT	T	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	11	—	—	—	—	—	—	—	—	—	—	—
STATION 15.—EAST BAY—M. MERCENARIA. UNLESS OTHERWISE INDICATED—57 SAMPLES ¹													
1966	DDE	—	—	—	—	—	—	—	—	—	—	T	T
	TDE	—	—	—	—	T	—	—	—	—	—	18	19
	DDT	—	—	—	—	T	—	—	—	—	—	T	—
1967	DDE	T	—	T	T	—	T	—	—	—	T	T	T
	TDE	21	12	13	15	19	19	15	T	T	13	T	14
	DDT	T	—	—	—	T	T	T	T	—	—	—	T
1968	DDE	—	—	T	T	T	—	—	—	—	—	—	—
	TDE	—	—	16	10	12	—	—	T	—	12	—	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1969	DDE	—	—	—	—	—	—	—	—	—	—	—	T
	TDE	16	10	—	—	16	14	14	T	T	T	—	16
	DDT	—	—	—	—	—	—	—	—	—	T	—	—
	Dieldrin	—	—	—	—	T	10	16	17	T	14	—	14
1970	DDE	—	—	—	—	—	—	—	—	—	—	2	10
	TDE	T	—	12	T	—	—	—	—	—	—	—	21
	DDT	—	—	—	—	—	—	—	—	—	—	—	18
	Dieldrin	—	—	T	—	—	—	—	—	—	—	—	16

TABLE J-2.—Distribution of organochlorine residues in the monitored species for each sampling station by date of collection—New York—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)												
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.	
STATION 15.—EAST BAY— <i>M. MERCENARIA</i> , UNLESS OTHERWISE INDICATED—57 SAMPLES ¹ —Continued														
1971	DDE		² 12		³ T	² 18		² —					—	T
	TDE		29		21	57		19					15	26
	DDT		29		11	23		12					10	23
	Dieldrin		15		11	38		11					—	—
1972	DDE	—			⁴ T	⁵ —	⁵ —							
	TDE	T			14	T	—							
	DDT	—			T	—	—							
STATION 16.—WEST BAY— <i>M. MERCENARIA</i> , UNLESS OTHERWISE INDICATED—57 SAMPLES ¹														
1966	DDE				—	T	—	—	—	—	—	—	T	T
	TDE				—	15	—	—	13	—	—	—	26	22
	DDT				—	T	—	—	—	—	—	—	14	13
1967	DDE	T	T	T	—	T	10	T	T	—	T	T	T	T
	TDE	20	17	24	24	27	32	—	17	T	20	14	19	19
	DDT	13	T	12	18	17	14	—	13	—	—	—	13	13
	Dieldrin	—	—	11	12	10	—	—	—	—	—	—	—	—
1968	DDE				1	T	—	—	—	—	—	—	—	—
	TDE			20	18	13	17	T	15	14	16	15	14	14
	DDT			—	1	—	—	—	—	—	—	—	—	—
1969	DDE	—	T			11	—	—	—	—	—	—	—	10
	TDE	19	20	T	—	28	13	18	19	14	21	—	21	21
	DDT	—	—	—	—	33	T	—	—	—	12	—	43	43
	Dieldrin	—	—	—	—	T	T	18	17	20	12	—	17	17
1970	DDE	—		T	—								² T	
	TDE	13		20	19								15	
	DDT	—		11	40								15	
	Dieldrin	—		14	1								T	
1971	DDE		¹ 11		¹ 1	² 15							—	10
	TDE		23		22	47			15				14	25
	DDT		28		12	44			—				T	20
	Dieldrin		T		10	16			—				T	10
1972	DDE			—	—	⁴ 30	⁴ 13							
	TDE	10		18	T	59	28							
	DDT	—		14	—	22	T							
	Dieldrin	—		T	—	—	—							

¹ Each sample represents 15 or more mature mollusks.

² *M. edulis*.

³ *M. arenaria*.

⁴ *C. virginica*.

⁵ *M. mercenaria*.

⁶ *M. demissus*.

SECTION K.—NORTH CAROLINA

The monthly collection of eastern oysters, *Crassostrea virginica*, to monitor pollution was initiated in July 1966 and continued until July 1972. During the program, 17 stations were sampled routinely for periods ranging from 3 to 6 years. All samples were analyzed by the Gulf Breeze Laboratory.

Approximate station locations are shown in Fig. K-1. A summary of data on organochlorine residues in the monitored species, *C. virginica*, is presented in Table K-1, and the distribution of residues in this species for each sampling station by date of collection in Table K-2.

North Carolina samples are noteworthy for the continuity of collections of a single species of mollusk at short intervals over a relatively long period of time. For this reason the data present a good picture of annual and seasonal trends of a persistent synthetic pollutant in this estuarine environment.

The incidence of DDT residues (75%) and maximum magnitude (566 ppb) are about the median of the 15 States monitored. The 1% incidence of dieldrin residues was somewhat lower than most other states. PCB compounds were not detected.

Although there are exceptions from one estuary to another, the magnitude of DDT residues in oysters showed little seasonal variation during the period 1967-69 when maximum levels of DDT pollution were detected. The overall decline in DDT residues (Part I, Table 7 and Fig. 2) is notable and undoubtedly associated with the decreased agricultural use of this chemical in North Carolina.

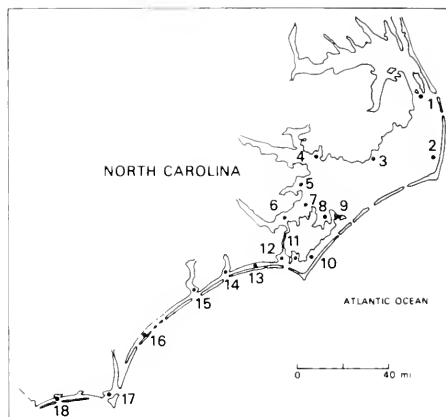


FIGURE K-1.—Diagram of coastal North Carolina showing approximate location of monitoring stations

1. Wanchese—Croatan Sound
2. Salvo—Pamlico Sound
3. Wysocking Bay
4. Rose Bay
5. Bay River
6. Neuse River
7. Point of Marsh—Neuse River
8. West Bay
9. Back Bay—Core Sound
10. Jarrett Bay—Core Sound
11. North River
12. Newport River
13. Bogue Sound
14. White Oak River
15. New River
16. Wrightsville Beach—Wrightsville Sound
17. Southport—Cape Fear River
18. Shallotte River

TABLE K-1.—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1966-72—North Carolina

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (μg/KG)	
				DDT	DIELDRIN
1 *	Wanchese	1966-72	72	49 (264)	
2 *	Salvo	1966-72	71	58 (566)	
3	Wysocking Bay	1966-70	43	35 (64)	
4 *	Rose Bay	1966-72	71	46 (121)	3 (14)
5 *	Bay River	1966-72	71	69 (310)	2 (12)
6	Neuse River	1966-70	43	43 (176)	
7 *	Point of Marsh	1966-72	71	53 (139)	2 (19)
8	West Bay	1967-72	58	34 (74)	
9	Back Bay	1966-67	9	8 (103)	
10 *	Jarrett Bay	1966-72	66	42 (106)	
11 *	North River	1966-72	64	48 (172)	2 (10)
12 *	Newport River	1966-72	68	54 (121)	3 (13)

TABLE K-1.—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1966-72—
North Carolina—Continued

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (μ G/KG)	
				DDT	DIELDRIN
13	Bogue Sound	1967-72	51	33 (71)	
14	White Oak River	1966-70	43	30 (60)	
15 *	New River	1966-72	72	61 (118)	
16	Wrightsville Beach	1966-70	43	35 (57)	
17 *	Southport	1966-72	72	32 (116)	
18	Shallotte River	1966-70	43	38 (51)	
Total number of samples			1,031		
Percent of samples positive for indicated compound				75	1

* Data from these stations summarized in Part I Table 7, and Fig. 2.

¹ Each sample represents 15 or more mature mollusks.

TABLE K-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—North Carolina

[Blank = no sample collected; — = no residue detected above 5 ppb; T = >5 but <10 ppb]

YEAR	COMPOUND	RESIDUES IN PPB (μ G/KG)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—WANCHESE—72 SAMPLES ¹													
1966	DDE							19	20	12	22	20	20
	TDE							12	17	T	17	18	32
	DDT							13	—	T	22	—	11
1967	DDE	14	25	28	16	26	21	21	19	20	24	43	28
	TDE	16	20	32	17	32	31	35	15	15	11	57	10
	DDT	15	10	17	—	29	21	64	17	13	17	64	53
1968	DDE	140	140	85	78	62	T	30	—	43	35	29	21
	TDE	56	51	59	16	32	1	13	—	22	27	16	15
	DDT	68	29	37	42	86	—	12	—	60	49	57	—
1969	DDE	40	17	13	10	37	11	T	10	—	T	T	12
	TDE	15	16	18	10	44	21	21	10	—	1	13	15
	DDT	T	T	11	—	43	T	19	13	—	—	—	T
1970	DDE	T	T	13	T	17	—	—	—	—	—	—	—
	TDE	11	T	15	—	19	—	—	—	—	11	—	—
	DDT	—	—	T	—	—	—	—	—	—	—	—	—
1971	DDE	—	—	—	—	—	T	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1972	DDE	—	—	—	T	—	1	—	—	—	—	—	—
	TDE	—	—	—	—	—	10	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

TABLE K-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—North Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB	MAR	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 2.—SALVO—71 SAMPLES ¹													
1966	DDE							12	14	14	14	32	23
	TDE							—	17	—	17	28	17
	DDT							—	—	—	—	28	15
1967	DDE	26	24	31	35	24	21	17	24		120	74	87
	TDE	11	13	21	25	21	30	21	19		56	45	29
	DDT	T	T	T	11	19	21	29	44		390	190	66
1968	DDE	35	85	66	85	65	34	58	28	95	45	58	66
	TDE	16	26	20	24	37	29	38	21	40	33	37	35
	DDT	20	50	36	21	28	19	87	36	240	76	120	80
1969	DDE	100	73	100	59	120	39	27	—	15	13	10	15
	TDE	31	31	38	27	51	38	37	—	21	15	19	13
	DDT	87	35	50	22	67	16	28	—	T	T	—	11
1970	DDE	23	34	24	19	27	14	—	12	11	—	10	19
	TDE	25	27	20	16	33	20	—	17	—	—	—	16
	DDT	14	20	13	10	19	—	—	14	—	—	—	11
1971	DDE	16	15	—	17	21	—	12	—	—	—	—	—
	TDE	17	16	—	18	21	—	14	—	—	—	—	—
	DDT	T	—	—	—	11	—	—	—	—	—	—	—
1972	DDE	—	—	—	T	13	13						
	TDE	—	—	—	—	T	22						
	DDT	—	—	—	—	—	—						
STATION 3.—WYSOCKING BAY—43 SAMPLES ¹													
1966	DDE							17	—	17	T	T	T
	TDE							17	—	18	13	12	11
	DDT							—	—	20	—	—	—
1967	DDE	T	14	15	17	16	12	15	T	—	—	T	10
	TDE	12	17	17	22	29	22	35	T	—	—	T	15
	DDT	T	10	—	—	14	13	14	T	—	—	T	T
1968	DDE	T	T	T	18	13	15	—	T	T	—	T	—
	TDE	T	T	—	20	20	13	—	T	T	—	13	—
	DDT	—	—	—	—	T	13	—	—	21	—	—	—
1969	DDE	T	—	T	12	T	T	T	T	—	T	T	14
	TDE	14	—	—	16	T	T	T	—	—	T	12	19
	DDT	—	—	—	T	—	—	T	12	—	—	—	12
1970	DDE	T											
	TDE	T											
	DDT	T											

TABLE K-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—North Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 4.—ROSE BAY—71 SAMPLES ¹													
1966	DDE							28	16	19	18	16	20
	TDE							34	13	22	23	23	36
	DDT							21	—	32	16	T	14
1967	DDE	12	T	23	15	16	15	35	T	T	—	14	12
	TDE	18	14	32	26	32	30	63	15	10	—	30	15
	DDT	T	T	T	11	16	40	23	13	T	—	19	T
1968	DDE	T	T	13	15	21	14	T	—	T	T	T	16
	TDE	T	T	12	15	27	25	T	—	T	17	17	17
	DDT	—	—	—	22	13	T	—	—	T	T	T	29
1969	DDE	11	16	11	T	12	T	T	T	12	T	—	10
	TDE	15	18	12	—	17	T	T	13	17	T	—	14
	DDT	20	—	—	—	T	—	T	69	—	—	—	T
1970	DDE	17	13	14	T	T	11	—	—	—	—	—	—
	TDE	19	19	19	—	15	19	—	—	—	—	—	—
	DDT	15	10	10	—	—	—	12	—	—	—	—	—
	Diieldrin	—	—	14	—	—	—	—	—	—	—	—	—
1971	DDE	—	—	—	—	T	—	—	—	—	—	—	—
	TDE	—	—	—	—	13	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Diieldrin	—	—	10	—	12	—	—	—	—	—	—	—
1972	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 5.—BAY RIVER—71 SAMPLES ¹													
1966	DDE							36	55	52	23	30	26
	TDE							16	78	73	46	61	60
	DDT							29	25	34	19	11	20
1967	DDE	36	30	81	39	32	29	22	15	T	28	25	24
	TDE	69	49	100	56	65	56	46	48	T	43	34	46
	DDT	23	16	35	20	27	19	27	27	—	51	—	16
1968	DDE	35	44	37	52	75	22	18	23	13	T	24	16
	TDE	35	43	24	49	71	33	24	28	15	—	36	21
	DDT	26	32	28	47	55	T	13	18	12	—	29	T
1969	DDE	19	29	36	43	54	45	37	18	T	13	T	20
	TDE	25	39	36	56	59	80	71	16	T	T	20	35
	DDT	T	20	37	37	32	17	26	18	14	—	—	21

TABLE K-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—North Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 5.—BAY RIVER—71 SAMPLES ¹ —Continued													
1970	DDE	24	40	36	42	55	10	15	11	13	—		23
	TDE	33	43	29	31	94	27	23	12	22	16		39
	DDT	17	33	18	14	22	—	18	—	14	—		13
1971	DDE	12	16	—	22	110	16	52	49	18	17	13	T
	TDE	15	13	—	21	170	19	97	96	34	26	11	—
	DDT	—	10	—	—	30	—	27	22	—	18	12	—
	Dieldrin	—	—	—	—	10	—	—	—	—	—	—	—
1972	DDE	16	41	—	71	85	43						
	TDE	11	37	—	48	130	91						
	DDT	—	21	—	37	87	43						
	Dieldrin	—	—	—	—	12	—						
STATION 6.—NEUSE RIVER—43 SAMPLES ¹													
1966	DDE							29	18	32	36	24	29
	TDE							46	24	48	57	55	60
	DDT							13	15	49	30	13	18
1967	DDE	17	16	24	29	21	16	19	14	20	25	24	24
	TDE	29	28	41	50	42	30	37	33	32	47	49	46
	DDT	—	T	T	T	T	16	25	14	14	13	T	T
1968	DDE	19	20	15	30	49	32	26	29	13	T	19	25
	TDE	24	25	15	44	110	68	42	56	22	T	32	25
	DDT	11	T	—	27	17	15	19	39	T	—	T	20
1969	DDE	20	17	23	16	37	29	30	13	T	10	T	16
	TDE	17	13	26	23	56	75	40	23	T	17	19	35
	DDT	10	T	11	—	20	21	20	11	—	—	—	16
1970	DDE	27											
	TDE	45											
	DDT	17											
STATION 7.—POINT OF MARSH—71 SAMPLES ¹													
1966	DDE							33	26	15	29	16	20
	TDE							37	26	20	31	33	40
	DDT							38	28	25	19	16	16
	Dieldrin							T	—	—	—	—	—
1967	DDE	15	17	22	20	20	33	13	22	11	12	11	18
	TDE	29	24	28	32	39	82	26	24	19	19	18	25
	DDT	15	10	—	11	45	24	27	33	10	27	15	10

TABLE K-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—North Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 7.—POINT OF MARSH—71 SAMPLES ¹ —Continued													
1968	DDE	11	—	12	18	11	15	10	15	—	T	14	11
	TDE	15	—	18	23	18	20	13	19	—	T	21	16
	DDT	T	—	—	21	10	16	11	21	—	—	—	—
1969	DDE	13	T	12	T	T	T	10	37	—	T	T	T
	TDE	14	14	17	13	T	11	16	27	—	T	11	10
	DDT	—	T	T	—	—	—	18	48	—	T	12	—
	Dieldrin	—	—	—	—	—	—	—	19	—	—	—	—
1970	DDE	11	22	20	T	T	—	—	—	—	—	T	—
	TDE	21	29	21	12	17	—	—	—	—	—	T	—
	DDT	T	15	—	—	—	—	—	—	—	—	—	—
1971	DDE	—	T	T	T	T	—	—	—	—	—	—	T
	TDE	—	T	T	32	—	—	—	—	—	—	—	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	T
1972	DDE	11	—	—	T	T	—	—	—	—	—	—	—
	TDE	20	—	—	—	T	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 8.—WEST BAY—58 SAMPLES ¹													
1967	DDE	—	18	25	—	—	22	14	T	—	16	T	15
	TDE	—	25	39	—	—	22	19	T	—	22	10	24
	DDT	—	T	11	—	—	13	11	T	—	17	12	15
1968	DDE	T	11	25	T	16	11	—	T	—	T	—	10
	TDE	—	11	30	T	13	T	—	T	—	—	—	16
	DDT	—	—	19	—	T	—	—	15	—	—	—	—
1969	DDE	16	T	20	16	11	T	12	—	—	—	T	T
	TDE	10	T	17	19	T	12	16	—	—	—	16	17
	DDT	T	—	12	12	10	T	T	—	—	—	—	T
1970	DDE	—	—	—	—	—	—	—	—	—	—	T	T
	TDE	—	—	—	—	—	—	—	—	—	—	T	16
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1971	DDE	—	T	—	—	T	—	—	—	—	T	—	—
	TDE	—	T	—	—	—	—	—	—	—	T	—	—
	DDT	—	—	—	—	—	—	—	—	—	T	—	—
1972	DDE	—	—	13	14	11	—	—	—	—	—	—	—
	TDE	—	—	10	T	T	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

TABLE K-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—North Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 9.—BACK BAY—9 SAMPLES ¹													
1966	DDE							10	—	15	17	24	10
	TDE							12	—	11	32	74	14
	DDT							14	—	12	15	T	—
1967	DDE	10			26	10							
	TDE	10			23	13							
	DDT	—			17	T							
STATION 10.—JARRETT BAY—66 SAMPLES ¹													
1966	DDE							22	—	13	14	T	12
	TDE							22	—	11	16	T	18
	DDT							15	—	19	16	—	14
1967	DDE	11	17	14	29	19	17	10	12	12	14	10	21
	TDE	T	12	19	24	22	18	12	T	T	18	17	17
	DDT	—	T	—	17	14	10	T	12	T	39	13	12
1968	DDE	18	13	47	T	10	T	—	13	T	18	—	T
	TDE	14	10	44	T	—	—	—	T	T	13	—	15
	DDT	T	T	15	—	—	—	—	T	T	66	—	T
1969	DDE	12	T	12	21	13	T	10	—	—	—	10	12
	TDE	T	11	16	22	12	10	15	—	—	—	18	22
	DDT	—	T	T	—	T	—	—	—	—	—	—	11
1970	DDE	T				—	—	—	—	—	—	—	—
	TDE	T				—	—	—	—	—	—	—	—
	DDT	T				—	—	—	—	—	—	—	—
1971	DDE	—	T	—	—	11	—	—	—	—	T	—	—
	TDE	—	T	—	—	10	—	—	—	—	T	—	—
	DDT	—	—	—	—	—	—	—	—	—	T	—	—
1972	DDE			—	12	T	—						
	TDE			—	—	—	—						
	DDT			—	—	—	—						
STATION 11.—NORTH RIVER—64 SAMPLES ¹													
1966	DDT							64	43	43	47	36	35
	TDE							50	33	41	47	33	29
	DDT							58	36	31	30	10	—
	Dieldrin							10	—	—	—	—	—
1967	DDE	27	26	45	11	52	53	34	21	16	17	10	20
	TDE	15	18	40	12	46	49	28	20	12	12	10	16
	DDT	10	16	27	—	29	31	15	26	T	10	—	T
	Dieldrin	—	—	10	—	—	—	—	—	—	—	—	—

TABLE K-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—North Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 11.—NORTH RIVER—64 SAMPLES ¹ —Continued													
1968	DDE	18	14	T	35	34	T	32	21	T	22	38	27
	TDE	13	—	—	25	26	—	27	19	T	17	20	—
	DDT	T	—	—	43	36	—	57	26	T	36	27	—
1969	DDE	18	13	16	32	16	T	—	—	12	53	—	T
	TDE	T	—	T	15	12	—	—	—	T	73	—	T
	DDT	—	—	12	14	—	—	—	—	11	34	—	T
1970	DDE	T	—	—	—	11	—	—	—	—	—	—	T
	TDE	T	—	—	—	11	—	—	—	—	—	—	—
	DDT	T	—	—	—	—	—	—	—	—	—	—	—
1971	DDE	—	—	—	12	T	T	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1972	DDE	—	—	20	10	T	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	11	—	—	—	—	—	—	—	—	—
STATION 12.—NEWPORT RIVER—68 SAMPLES ¹													
1966	DDE	—	—	—	—	—	—	20	17	14	16	24	14
	TDE	—	—	—	—	—	—	21	13	19	26	44	22
	DDT	—	—	—	—	—	—	—	T	T	—	11	—
	Dieldrin	—	—	—	—	—	—	—	T	—	—	—	—
1967	DDE	14	18	25	20	21	19	T	1	14	16	12	16
	TDE	21	24	85	27	29	30	12	15	25	18	17	26
	DDT	T	T	11	T	—	T	10	T	15	23	—	T
1968	DDE	11	16	17	25	—	T	T	1	11	18	19	15
	TDE	14	16	23	31	—	—	T	T	13	24	29	18
	DDT	—	—	T	—	—	—	—	—	T	23	11	T
1969	DDE	21	21	20	27	17	12	T	—	T	13	15	18
	TDE	21	22	21	29	23	19	T	—	T	16	15	17
	DDT	—	10	T	13	T	T	—	—	—	25	17	17
	Dieldrin	—	—	—	—	—	—	—	—	—	T	—	—
1970	DDE	16	12	12	18	T	—	T	—	—	—	T	—
	TDE	14	T	10	23	T	—	—	—	—	—	T	—
	DDT	13	—	T	10	—	—	—	—	—	—	—	—
1971	DDE	—	11	—	—	13	—	—	17	—	—	—	—
	TDE	—	11	—	—	18	—	—	16	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

TABLE K-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—North Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{g}/\text{kg}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 12.—NEWPORT RIVER—68 SAMPLES ¹ —Continued													
1972	DDE	—		30	T	T	T						
	TDE	—		24	—	—	—						
	DDT	—		36	—	—	—						
	Dieldrin	—		13	—	—	—						
STATION 13.—BOGUE SOUND—51 SAMPLES ¹													
1967	DDE			31	20	15	T	10	10	13	15	14	15
	TDE			26	23	17	T	12	12	14	—	13	T
	DDT			12	T	T	—	11	16	24	T	12	T
1968	DDE	12	T	16	15	T	—	T	T	13	13	21	18
	TDE	T	—	—	T	—	—	—	T	13	13	17	11
	DDT	—	—	—	—	—	—	—	T	21	T	13	11
1969	DDE	20	29	17	25	19	T	15	—	—	12	13	12
	TDE	15	23	11	20	19	11	16	—	—	14	17	—
	DDT	T	19	T	12	11	11	37	—	—	T	15	—
1970	DDE					—							
	TDE					—							
	DDT					—							
1971	DDE			—	—	T	—	—	—	—	—	—	—
	TDE			—	—	—	—	—	—	—	—	—	—
	DDT			—	—	—	—	—	—	—	—	—	—
1972	DDE	—	—	T	—	—	—						
	TDE	—	—	—	—	—	—						
	DDT	—	—	—	—	—	—						
STATION 14.—WHITE OAK RIVER—43 SAMPLES ¹													
1966	DDE							T	20	T	29	T	T
	TDE							—	25	T	31	T	T
	DDT							—	T	—	—	—	—
1967	DDE	—	—	I	—	I	—	T	—	T	T	T	T
	TDE	—	—	—	—	T	—	13	—	T	—	T	T
	DDT	—	—	—	—	—	—	10	—	T	—	—	—
1968	DDE	T	T	13	T	—	—	—	T	11	T	T	14
	TDE	T	T	13	T	—	—	—	T	11	—	T	T
	DDT	T	—	—	—	—	—	—	T	T	—	—	T
1969	DDE	I	12	14	T	T	—	—	—	—	T	T	—
	TDE	—	—	T	—	T	—	—	—	—	T	T	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

TABLE K-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—North Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 14.—WHITE OAK RIVER—43 SAMPLES ¹ —Continued													
1970	DDE	T											
	TDE	—											
	DDT	—											
STATION 15.—NEW RIVER—72 SAMPLES ¹													
1966	DDE							25	T	23	21	28	36
	TDE							21	T	27	26	34	44
	DDT							28	—	T	—	—	14
1967	DDE	39	16	30	45	20	16	11	19	16	28	21	28
	TDE	49	18	30	59	23	13	13	28	20	34	26	37
	DDT	16	T	T	14	—	—	—	11	—	—	—	21
1968	DDE	42	16	35	39	25	15	T	10	14	15	15	14
	TDE	38	14	31	29	25	15	—	T	14	12	12	T
	DDT	31	T	T	—	—	—	—	T	T	—	—	—
1969	DDE	25	18	21	28	13	15	11	13	14	19	23	27
	TDE	19	19	27	35	13	15	15	13	12	22	26	27
	DDT	—	T	T	T	—	—	—	T	—	11	—	11
1970	DDE	21	28	29	26	26	T	—	—	—	—	—	17
	TDE	19	24	22	31	20	—	—	—	—	—	—	—
	DDT	—	T	T	12	—	—	—	—	—	—	—	—
1971	DDE	11	T	15	T	T	—	—	—	—	12	—	20
	TDE	—	T	12	—	—	—	—	—	—	11	—	22
	DDT	—	—	—	—	—	—	—	—	—	T	—	T
1972	DDE	19	17	24	11	T	—	—	—	—	—	—	—
	TDE	18	21	21	—	—	—	—	—	—	—	12	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 16.—WRIGHTSVILLE BEACH—43 SAMPLES ¹													
1966	DDE							11	14	15	15	19	12
	TDE							12	18	12	16	24	10
	DDT							—	—	—	13	14	—
1967	DDE	11	16	19	14	T	T	T	10	—	T	—	11
	TDE	14	18	15	14	T	T	14	18	—	12	—	13
	DDT	—	T	T	—	T	—	T	14	—	22	—	T
1968	DDE	T	T	10	13	T	—	—	10	T	T	T	T
	TDE	T	—	—	10	T	—	—	—	—	—	12	—
	DDT	T	—	—	—	—	—	—	T	—	—	—	—

TABLE K-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—North Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 16.—WRIGHTSVILLE BEACH—43 SAMPLES ¹ —Continued													
1969	DDE	T	T	T	T	T	—	—	—	—	T	T	12
	TDE	—	—	—	T	T	—	—	—	—	T	11	16
	DDT	—	—	—	—	—	—	—	—	—	—	—	12
1970	DDE	T	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 17.—SOUTHPORT—72 SAMPLES ¹													
1966	DDE	—	—	—	—	—	—	11	21	11	29	T	T
	TDE	—	—	—	—	—	—	T	16	10	25	T	T
	DDT	—	—	—	—	—	—	12	—	—	—	—	—
1967	DDE	T	13	11	T	—	T	10	14	11	T	—	T
	TDE	—	12	T	T	—	—	10	T	—	—	—	T
	DDT	—	T	—	—	—	—	17	30	15	—	—	—
1968	DDE	18	13	T	T	11	—	—	10	11	T	17	17
	TDE	13	T	T	T	14	—	—	—	T	11	—	T
	DDT	T	—	—	—	—	—	—	13	T	—	21	10
1969	DDE	T	—	12	T	11	—	—	—	—	—	—	—
	TDE	—	—	17	—	12	—	—	—	—	—	—	—
	DDT	—	—	87	—	T	—	—	—	—	—	—	—
1970	DDE	—	T	—	—	—	—	—	T	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1971	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1972	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 18.—SHALLOTTE RIVER—43 SAMPLES ¹													
1966	DDE	—	—	—	—	—	—	—	T	T	T	19	T
	TDE	—	—	—	—	—	—	—	—	T	—	18	T
	DDT	—	—	—	—	—	—	—	—	—	—	11	—
1967	DDE	T	16	11	14	19	T	T	T	10	T	12	T
	TDE	—	13	T	15	22	T	11	T	T	—	11	T
	DDT	—	T	—	—	10	—	10	T	T	—	T	—

TABLE K-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—North Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 18.—SHALLOTTE RIVER—43 SAMPLES ¹ —Continued													
1968	DDE	13	14	16	15	—	—	—	T	T	13	—	17
	TDE	11	10	11	12	—	—	—	—	—	T	—	10
	DDT	T	T	—	—	—	—	—	—	—	11	—	T
1969	DDE	T	T	12	11	15	T	14	T	T	11	T	T
	TDE	—	—	10	T	19	—	14	T	—	T	13	—
	DDT	—	—	19	—	T	—	16	16	12	T	—	—
1970	DDE	T	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

¹ Each sample represents 15 or more mature mollusks.

SECTION L.—SOUTH CAROLINA

Monthly collections of eastern oysters, *Crassostrea virginica*, to identify estuarine pollution were made from August 1965 through November 1969. The 17 stations (Fig. L-1) were monitored for periods ranging from 1 to 5 years. All samples were analyzed at the Gulf Breeze Laboratory. A summary of data on organochlorine residues in the monitored species, *C. virginica*, is presented in Table L-1, and the distribution of residues in this species for each sampling station by date of collection in Table L-2.

South Carolina samples are characterized by the uniformly low level of DDT residues and moderately low incidence of positive samples. Samples from only three other States indicated generally lower levels of DDT contamination.

In those areas with adequate numbers of samples for annual comparison, there was an obvious decline at most stations in the magnitude and incidence of DDT residues in 1968-69 as compared to earlier years (Part I. Table 6).

South Carolina was the only State in which mirex residues were detected in mollusks. These residues were observed only in the period March - May 1969. They were found at nine stations widely distributed along the South Carolina coast. Largest residues were found in samples collected in the Charleston area, i.e., Stations 8 and 9.

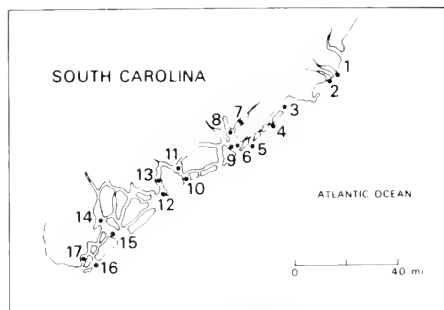


FIGURE L-1.—Diagram of coastal South Carolina showing approximate location of monitoring stations

1. North Santee Bay—Santee River
2. South Santee Bay—Santee River
3. Bull Creek
4. Price Creek
5. Inlet Creek
6. Hog Island Channel—Ashley, Cooper, and Wando Rivers
7. Wando River—Ashley, Cooper, and Wando Rivers
8. Ashley River—Ashley, Cooper, and Wando Rivers
9. Fort Johnson—Ashley, Cooper, and Wando Rivers
10. Steamboat Creek—North Edisto River
11. Toogoodoo Creek—North Edisto River
12. Big Bay Creek—South Edisto River
13. St. Pierre Creek—South Edisto River
14. Whale Branch—Broad River
15. Skull Creek—Broad River
16. May Creek
17. New River

TABLE L-1.—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1965-69—South Carolina

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (µG/KG)		
				DDT	Dieldrin	MIREX
1	North Santee Bay	1965-68	41	10 (19)	4 (19)	
2	South Santee Bay	1965-68	40	14 (80)	3 (19)	
3	Bull Creek	1969	12	2 (10)	2 (13)	2 (35)
4	Price Creek	1965-68	42	25 (81)	2 (12)	
5	Inlet Creek	1965-68	42	21 (52)		
6	Hog Island Channel	1965-68	41	33 (73)		
7	Wando River	1965-68	42	31 (44)		
8	Ashley River	1965-69	54	45 (154)	8 (90)	1 (190)
9	Fort Johnson	1969	12	4 (10)		1 (540)
10	Steamboat Creek	1965-69	54	26 (32)		1 (38)
11	Toogoodoo Creek	1965-69	53	40 (98)		1 (38)
12	Big Bay Creek	1965-69	54	32 (91)		1 (T)
13	St. Pierre Creek	1969	12	7 (88)		1 (38)
14	Whale Branch	1965-68	41	21 (79)		
15	Skull Creek	1965-68	39	12 (30)	1 (35)	
16	May Creek	1969	12	3 (15)	1 (11)	3 (37)
17	New River	1969	12	1 (16)	1 (21)	1 (27)
	Occasional stations (6)	1965-68	7	5 (201)	2 (15)	
Total number of samples			610			
Percent positive for indicated compound				54	4	2

NOTE: T = >5 but <10 ppb.

¹ Each sample represents 15 or more mature mollusks.

TABLE L-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—South Carolina

[Blank = no sample collected; — = no residue detected above 5 ppb; T = >5 but <10 ppb]

YEAR	COMPOUND	RESIDUES IN PPB (µG/KG)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—NORTH SANTEE BAY—41 SAMPLES ¹													
1965	DDE							T	T	T	—	—	—
	TDE							T	T	T	—	—	—
	DDT							—	—	—	—	T	—
1966	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	—	—	—	—	T	T	T	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	T	T	—	—	—	—
	Dieldrin	—	—	15	—	—	—	—	—	—	—	—	—

TABLE L-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—South Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—NORTH SANTEE BAY—41 SAMPLES ¹ —Continued													
1968	DDE	T	—	—	T	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	19	—	—
	Dieldrin	15	—	12	19	—	—	—	—	—	—	—	—
STATION 2.—SOUTH SANTEE BAY—40 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	T	T	T	—	—	—
	TDE	—	—	—	—	—	—	T	T	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	T	—	—
1966	DDE	—	—	—	—	T	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	—	T	—	—	—	T	T	T	—	—	—
	TDE	—	—	—	—	—	—	T	—	—	—	—	—
	DDT	—	—	—	—	—	—	13	T	T	—	—	—
1968	DDE	T	46	—	10	T	—	—	T	—	—	—	—
	TDE	T	—	—	T	—	—	—	—	—	—	—	—
	DDT	T	34	—	—	—	—	—	12	—	—	—	—
	Dieldrin	—	13	10	19	—	—	—	—	—	—	—	—
STATION 3.—BULL CREEK—12 SAMPLES ¹													
1969	DDE	—	—	—	T	T	—	—	—	—	—	—	—
	TDE	—	—	—	T	T	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	—	—	13	12	—	—	—	—	—	—	—
	Mirex	—	—	22	—	35	—	—	—	—	—	—	—
STATION 4.—PRICE CREEK—42 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	T	—	T	—	—	—
	TDE	—	—	—	—	—	—	T	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1966	DDE	T	—	T	—	—	—	—	—	19	T	—	T
	TDE	—	—	—	—	—	—	—	—	36	—	—	T
	DDT	—	—	—	—	—	—	—	—	26	—	—	—
	Dieldrin	—	—	12	—	—	—	—	—	—	—	—	—
1967	DDE	T	11	12	T	T	T	T	13	T	—	10	T
	TDE	—	T	—	—	—	10	T	12	—	—	T	T
	DDT	—	T	—	—	T	11	10	11	T	—	T	—

TABLE 1-1—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—South Carolina—Continued

Year	Compound	Residues in PPB $\mu\text{g/g}$											
		Jan	Feb	Mar	Apr	May	June	July	Aug	Sept	Oct	Nov	Dec
STATION 4—FRICE CREEK—41 SAMPLES—Continued													
84	DDC	T	T	T	11	T	—	T	—	—	T	—	—
	TDE	11	T	—	T	—	—	—	—	—	—	—	—
	DDT	T	—	—	—	—	—	—	—	—	—	—	—
	Diastz	—	11	—	—	—	—	—	—	—	—	—	—
STATION 5—INLET CREEK—41 SAMPLES:													
85	DDC	—	—	—	—	—	—	T	T	—	—	T	T
	TDE	—	—	—	—	—	—	T	T	—	—	—	T
	DDT	—	—	—	—	—	—	—	T	—	—	—	—
86	DDC	T	—	—	—	14	—	—	T	—	T	T	T
	TDE	T	—	—	—	14	—	—	—	—	—	—	T
	DDT	T	—	—	—	—	—	—	—	—	—	—	—
87	DDC	T	T	T	4	T	T	T	16	—	—	—	T
	TDE	—	—	T	13	—	—	15	21	—	—	—	—
	DDT	—	—	—	1	T	—	17	15	—	—	—	—
88	DDC	T	—	11	—	—	—	—	—	—	—	—	—
	TDE	T	—	T	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 6—HOG ISLAND CHANNEL—41 SAMPLES:													
89	DDC	—	—	—	—	—	—	T	T	T	T	T	—
	TDE	—	—	—	—	—	—	T	T	—	T	—	—
	DDT	—	—	—	—	—	—	—	—	—	T	—	—
90	DDC	T	3	T	14	13	—	20	—	14	10	—	T
	TDE	T	T	—	—	5	T	15	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
91	DDC	1	20	20	16	14	T	11	19	T	—	T	T
	TDE	4	13	—	—	11	T	13	24	T	—	T	T
	DDT	—	T	—	—	1	27	14	24	17	—	16	—
92	DDC	T	3	1	20	—	—	T	—	—	T	T	—
	TDE	—	T	—	16	—	—	11	—	—	—	—	—
	DDT	—	—	—	4	—	—	T	—	—	—	—	—
STATION 7—WANDO RIVER—41 SAMPLES:													
93	DDC	—	—	—	—	—	—	T	T	T	T	—	T
	TDE	—	—	—	—	—	—	T	T	T	T	—	T
	DDT	—	—	—	—	—	—	T	T	—	T	—	—

TABLE 1-C.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—South Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPE (ug/g)											
		JAN	FEB	MAR	APR	MAY	JUNE	JULY	AUG.	SEPT	OCT	NOV	DEC
STATION 7—WANDO RIVER—42 SAMPLES—Continued													
1966	DDE	10	—	7	11	10	—	—	—	10	11	7	7
	TDE	10	—	—	—	11	—	—	—	10	11	11	14
	DDT	7	—	—	—	—	—	—	—	7	—	—	7
1967	DDE	10	11	15	7	7	7	—	10	7	7	7	11
	TDE	17	15	17	—	7	7	11	20	10	7	—	16
	DDT	—	—	—	—	—	7	10	14	10	—	—	7
1968	DDE	14	13	11	14	13	—	7	—	—	—	—	—
	TDE	19	7	7	7	14	—	10	—	—	—	—	—
	DDT	11	—	—	—	—	—	7	—	—	—	—	—
STATION 8—ASHLEY RIVER—54 SAMPLES													
1965	DDE	—	—	—	—	—	—	15	7	7	7	15	11
	TDE	—	—	—	—	—	—	7	7	7	15	17	11
	DDT	—	—	—	—	—	—	4	7	—	10	15	11
1966	DDE	13	14	13	15	15	7	3	—	—	7	15	15
	TDE	15	24	15	13	15	24	7	—	—	7	15	24
	DDT	15	30	15	11	14	—	—	—	—	7	15	11
	Dieldrin	—	13	11	20	30	—	—	—	—	—	—	—
1967	DDE	16	16	11	15	11	—	3	3	7	4	15	15
	TDE	11	15	15	11	15	—	20	15	11	15	15	17
	DDT	14	15	15	15	15	14	3	11	—	3	11	14
	Dieldrin	—	—	7	—	—	—	—	—	—	—	—	—
1968	DDE	11	15	20	15	11	7	3	15	7	7	7	8
	TDE	10	15	15	11	15	—	15	3	11	—	—	11
	DDT	15	—	15	15	15	—	14	—	3	—	7	15
	Dieldrin	7	—	15	—	—	—	—	—	—	—	—	—
1969	DDE	11	13	7	—	15	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	15	—	—	—	—	—	—	—
	Dieldrin	—	—	—	—	—	—	—	—	—	—	—	15
	Mirex	—	—	30	—	—	—	—	—	—	—	—	—
STATION 9—FORT JOHNSON—12 SAMPLES													
1968	DDE	—	—	7	7	—	—	—	—	—	7	—	7
	TDE	—	—	—	—	—	—	—	—	—	7	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Mirex	—	—	30	—	—	—	—	—	—	—	—	—

TABLE L-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—South Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 10.—STEAMBOAT CREEK—54 SAMPLES ¹													
1965	DDE							T	T	T	—	T	—
	TDE							T	T	—	—	—	—
	DDT							—	T	—	—	—	—
1966	DDE	T	T	—	14	—	11	—	T	—	T	—	—
	TDE	—	—	—	11	—	11	—	—	T	T	—	—
	DDT	—	—	—	—	—	—	—	—	T	—	—	—
1967	DDE	T	11	10	—	13	T	—	—	T	—	—	—
	TDE	T	T	10	—	14	T	—	—	T	—	—	T
	DDT	—	—	T	—	T	16	—	—	T	—	—	T
1968	DDE	T	T	—	T	T	—	T	—	—	—	—	—
	TDE	T	—	—	—	—	—	—	—	—	—	—	—
	DDT	T	—	—	—	—	—	—	—	—	—	—	—
1969	DDE	—	T	—	—	—	—	—	—	—	—	T	T
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Mirex	—	—	38	—	—	—	—	—	—	—	—	—
STATION 11.—TOOGODOO CREEK—53 SAMPLES ¹													
1965	DDE							32	14	18	16	20	16
	TDE							43	20	19	20	33	16
	DDT							T	T	T	—	T	—
1966	DDE	27	18	15	38	24	19	—	15	13	18	11	22
	TDE	25	20	13	36	33	20	—	16	13	18	12	26
	DDT	—	T	T	24	16	—	—	—	11	10	—	T
1967	DDE	10	20	21	25	21	14	T	—	—	12	T	30
	TDE	T	17	18	23	22	15	—	—	—	—	—	26
	DDT	—	—	T	—	14	T	—	—	—	—	—	12
1968	DDE	17	20	16	18	25	21	T	18	11	T	T	13
	TDE	14	16	—	16	20	16	T	19	10	—	—	T
	DDT	T	—	—	—	—	—	—	17	T	—	—	—
1969	DDE	—	—	—	—	—	—	—	—	—	—	T	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Mirex	—	—	38	—	—	—	—	—	—	—	—	—
STATION 12.—BIG BAY CREEK—54 SAMPLES ¹													
1965	DDE							T	T	T	T	—	T
	TDE							T	T	—	T	—	T
	DDT							—	—	—	—	—	—

TABLE L-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—South Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 12—BIG BAY CREEK—54 SAMPLES ¹ —Continued													
1966	DDE	11	—	—	—	T	T	12	T	—	—	—	T
	TDE	T	—	—	—	T	T	T	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	11	14	14	12	12	T	13	—	T	—	—	T
	TDE	T	12	14	11	15	T	14	—	—	—	—	—
	DDT	T	T	11	12	13	21	31	—	—	—	—	—
1968	DDE	T	T	T	13	12	—	—	T	—	—	—	—
	TDE	11	—	T	T	—	—	—	T	—	—	—	—
	DDT	T	—	—	—	—	—	—	T	—	—	—	—
1969	DDE	—	—	—	T	T	24	—	—	—	12	14	17
	TDE	—	—	—	—	—	23	—	—	—	13	T	13
	DDT	—	—	—	—	33	44	—	—	—	27	13	13
	Mirex	—	—	T	—	—	—	—	—	—	—	—	—
STATION 13.—ST. PIERRE CREEK—12 SAMPLES ¹													
1969	DDE	—	T	—	T	T	22	—	—	—	15	13	T
	TDE	—	—	—	—	—	21	—	—	—	15	T	—
	DDT	—	—	—	—	30	45	—	—	—	29	13	—
	Mirex	—	—	38	—	—	—	—	—	—	—	—	—
STATION 14.—WHALE BRANCH—41 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	T	T	—	T	T	T
	TDE	—	—	—	—	—	—	T	T	—	T	—	T
	DDT	—	—	—	—	—	—	T	T	—	—	—	T
1966	DDE	—	—	—	T	T	—	—	—	—	11	33	T
	TDE	—	—	—	T	—	—	—	—	—	11	20	—
	DDT	—	—	—	—	—	—	—	—	—	—	26	—
1967	DDE	13	T	—	11	T	—	T	—	11	—	—	—
	TDE	T	—	—	T	T	—	T	—	—	—	—	—
	DDT	T	—	—	T	—	—	T	—	T	—	—	—
1968	DDE	11	T	—	14	—	—	—	T	T	—	—	—
	TDE	T	T	—	14	—	—	—	12	—	—	—	—
	DDT	—	—	—	—	—	—	—	15	—	—	—	—
STATION 15.—SKULL CREEK—39 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	T	T	—	T	T	—
	TDE	—	—	—	—	—	—	T	T	—	T	—	—
	DDT	—	—	—	—	—	—	—	T	—	—	—	—

TABLE L-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—South Carolina—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 15.—SKULL CREEK—39 SAMPLES ¹ —Continued													
1966	DDE	—	—	—	—	12	T	—	—	—	—	T	—
	TDE	—	—	—	—	12	T	—	—	—	—	T	—
	DDT	—	—	—	—	T	—	—	—	—	—	—	—
1967	DDE	—	—	—	11	—	T	—	—	—	—	—	—
	TDE	—	—	—	14	—	T	—	—	—	—	—	T
	DDT	—	—	—	T	—	16	—	—	—	—	—	T
1968	DDE	T	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	17	—	—
	Dieldrin	—	—	—	—	—	—	—	—	—	—	—	35
STATION 16.—MAY CREEK—12 SAMPLES ¹													
1969	DDE	—	—	—	15	T	—	—	T	—	—	—	—
	TDE	—	—	—	—	T	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	—	—	11	—	—	—	—	—	—	—	—
	Mirex	—	—	23	37	27	—	—	—	—	—	—	—
STATION 17.—NEW RIVER—12 SAMPLES ¹													
1969	DDE	—	—	—	T	—	—	—	—	—	—	—	—
	TDE	—	—	—	11	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	—	—	21	—	—	—	—	—	—	—	—
	Mirex	—	—	—	—	27	—	—	—	—	—	—	—

¹ Each sample represents 15 or more mature mollusks.

SECTION M.—TEXAS

The eastern oyster, *Crassostrea virginica*, was used to monitor pollution in Texas estuarine waters during the period July 1965 - June 1972. All samples were analyzed at the Gulf Breeze Laboratory. Approximate locations of the 13 sampling stations are shown in Fig. M-1. A summary of data on organochlorine residues in the monitored species, *C. virginica*, is presented in Table M-1, and the distribution of residues in this species for each sampling station by date of collection in Table M-2. In some instances, more than one reef was sampled at different times in a particular bay. In these instances, the data have been integrated to reflect bay conditions as a whole. At some times, floods resulting from tropical storms decimated oyster reefs and interrupted routine monitoring. On at least one occasion, sample preparation reagents were contaminated with chlordane leading to spurious analytical results. Consequently, all findings of chlordane have been omitted from the data tabulations.

In conjunction with oyster monitoring in Texas, many samples of fish and other vertebrates were analyzed throughout the monitoring program. These analyses indicated, as might be expected, more kinds of pollutants and of greater magnitude than those found in oysters. PCB's, for example, were commonly found in fish samples but were detected in only five collections of oysters. In the Arroyo Colorado, Station 12, findings of consistently large DDT residues in oysters were paralleled by DDT residues about 10 times larger in fish. A causal relationship between DDT residues in the eggs and reproductive failure of the spotted sea trout, *Cynoscion nebulosus*, there in 1969, has been postulated (5).

Although the incidence of DDT residues was higher in eight other States, samples from monitoring stations in Texas bays that receive runoff from the agricultural areas were consistently contaminated with DDT. The maximum DDT residue detected, 1,249 ppb, was in an isolated sample; more typically the residues in contaminated areas were in the range of 100 - 500 ppb of DDT.

Toxaphene of presumably agricultural origin was detected in only one sample.

There is a clearly defined trend of declining DDT residues in oysters. In 1971, there was a more than 50% increase in the number of samples containing negligible DDT residues (i.e., <11 ppb) over previous years and a 75% decrease in the number of samples in the 100 - 1,000 ppb range.

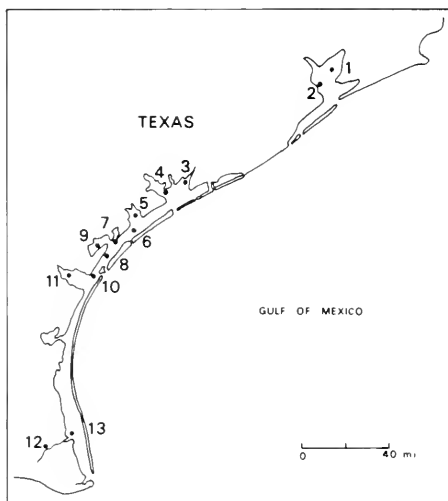


FIGURE M-1.—Diagram of coastal Texas showing approximate location of monitoring stations

1. Trinity Bay—Trinity-San Jacinto River basins
2. Galveston Bay—Trinity-San Jacinto River basins
3. Tres Palacios Bay—Lavaca River Basin
4. Lavaca Bay—Lavaca River Basin
5. San Antonio Bay, North—Guadalupe-San Antonio River Basin
6. San Antonio Bay, South—Guadalupe-San Antonio River Basin
7. St. Charles Bay—San Antonio-Nueces Coastal Area
8. Aransas Bay—San Antonio-Nueces Coastal Area
9. Copano Bay—San Antonio-Nueces Coastal Area
10. Red Fish Bay—San Antonio-Nueces Coastal Area
11. Nueces Bay—Nueces River Basin
12. Arroyo Colorado—Rio Grande Coastal Area
13. Lower Laguna Madre—Rio Grande Coastal Area

TABLE M-1.—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1965-72—Texas

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (µg/kg)				
				DDT	DIELDRIN	ENDRIN	TOXAPHENE ²	PCB'S ³
1	Trinity Bay	1965-69	47	28 (51)	1 (20)			
2	Galveston Bay	1965-72	71	60 (88)	31 (87)			
3	Tres Palacios Bay	1965-72	74	71 (974)	6 (18)			
4	Lavaca Bay	1965-72	66	59 (400)	4 (24)			2
5	San Antonio Bay, North	1965-72	59	38 (78)	8 (27)			
6	San Antonio Bay, South	1965-72	75	40 (488)	3 (56)	1 (10)		

TABLE M-1.—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1965-72—Texas—Continued

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (µG KG)				
				DDT	DIELDRIN	ENDRIN	TOXAPHENE ²	PCB'S ³
7	St. Charles Bay	1966-72	66	33 (93)	11 (80)			
8	Aransas Bay	1965-67	19	18 (83)	2 (48)			
9	Copano Bay	1967-71	51	24 (96)				
10	Red Fish Bay	1966-72	67	52 (82)				2
11	Nueces Bay	1965-68	20	20 (450)	4 (33)	3 (18)		
12	Arroyo Colorado	1965-71	48	48 (710)	45 (46)	18 (32)	1	
13	Lower Laguna Madre	1965-67	24	15 (57)	1 (46)			
	Occasional stations (16)	1965-72	41	24 (1,249)	16 (64)			1
Total number of samples			728					
Percent of samples positive for indicated compound				73	18	3		<1

¹ Each sample represents 15 or more mature mollusks

² Present but not quantified.

TABLE M-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Texas

[Blank = no sample collected; — = no residue detected above 5 ppb or no residue detected (PCB s); T = >5 but <10 ppb.]

YEAR	COMPOUND	RESIDUES IN PPB (µG KG)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—TRINITY BAY—47 SAMPLES ¹													
1965	DDE							T	—	T	T	T	11
	TDE							T	—	—	T	10	16
	DDT							—	—	—	—	—	—
1966	DDE	12	18	15		18	—	—	—	—	—	T	T
	TDE	17	27	28		33	—	—	—	—	—	T	12
	DDT	—	—	—		—	—	—	—	—	—	—	—
1967	DDE	T	T	12	—	—	T	—	—	—	T	T	T
	TDE	T	12	18	—	—	T	—	—	—	T	T	—
	DDT	—	—	—	—	—	T	—	—	—	T	—	—
1968	DDE	T	T	T	T	T	—	—	T	—	10	T	
	TDE	11	T	T	11	T	—	—	—	—	T	—	
	DDT	—	—	—	—	—	—	—	—	—	—	—	
1969	DDE		—			T	—	—	—	—			T
	TDE		—			13	—	—	—	—			—
	DDT		—			—	—	—	—	—			—
	Dieldrin		—			20	—	—	—	—			—

TABLE M-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Texas—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 2.—GALVESTON BAY—71 SAMPLES ¹													
1965	DDE							—	T	T		T	T
	TDE							—	—	—		11	17
	DDT							—	—	—		—	—
1966	DDE	15	18	13		11	—	—	—	—	T	T	T
	TDE	36	43	32		33	—	—	—	—	T	24	24
	DDT	T	—	—		—	—	—	—	—	—	—	—
	Dieldrin	—	—	—		—	—	—	—	—	—	12	—
1967	DDE	T	11	15	13	—	T	—	T	T	11	11	13
	TDE	26	29	44	34	15	18	—	10	T	21	23	32
	DDT	T	—	—	—	—	—	—	—	—	11	—	—
	Dieldrin	12	14	15	—	—	—	—	—	—	—	14	19
1968	DDE	10	16	T	14	20	T	T	T	T	14	T	—
	TDE	24	43	36	37	46	41	30	13	15	46	34	49
	DDT	—	T	—	—	—	—	—	—	—	13	—	—
	Dieldrin	13	25	20	—	—	—	—	—	—	—	19	14
1969	DDE		T			24	—	T	10	—			19
	TDE		44			64	23	13	11	T			19
	DDT		—			—	—	—	—	—			—
	Dieldrin		30			19	—	—	—	—			—
1970	DDE		13	15	T	12	T	—	—	—	—	11	—
	TDE		35	34	34	32	18	20	—	—	17	31	32
	DDT		—	10	T	—	—	—	—	—	—	—	—
	Dieldrin		18	19	11	14	—	—	—	13	—	24	18
1971	DDE	T	11			—			—	—	—	T	T
	TDE	39	38		48				—	—	—	17	17
	DDT	—	—		—				—	—	—	T	11
	Dieldrin	16	23		30				65	46	—	—	26
1972	DDE	T	T	T	T	—							
	TDE	18	T	26	15	T							
	DDT	—	—	—	—	—							
	Dieldrin	42	26	87	24	36							
STATION 3.—TRES PALACIOS BAY—74 SAMPLES ¹													
1965	DDE							11	11	T	T	21	93
	TDE							T	1	—	—	T	29
	DDT							—	—	—	—	T	65

TABLE M-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Texas—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 3.—TRES PALACIOS BAY—74 SAMPLES ¹ —Continued													
1966	DDE	78	78	250	190	300	47	210	97	21	11	23	26
	TDE	36	36	44	53	89	22	97	29	T	—	T	T
	DDT	43	53	80	59	130	12	23	—	—	—	—	—
1967	DDE	42	42	67	51	34	58	18	12	18	72	150	240
	TDE	14	17	31	19	14	24	—	—	—	41	52	57
	DDT	—	T	11	T	15	11	—	—	—	T	71	38
1968	DDE	270	220	230	320	300	91	62	43	20	19	22	24
	TDE	66	57	23	590	77	62	95	10	42	13	19	15
	DDT	83	21	81	64	22	T	17	—	T	—	—	—
	Dieldrin	—	18	—	—	—	—	—	—	—	—	—	—
1969	DDE	25	24	83	—	91	55	95	58	43	—	—	25
	TDE	19	18	35	—	44	62	15	56	T	—	—	40
	DDT	—	T	27	—	21	15	—	—	—	—	—	12
	Dieldrin	—	—	—	—	—	—	—	—	10	—	—	—
1970	DDE	70	36	110	230	—	44	33	41	—	50	—	16
	TDE	48	29	51	44	—	38	55	72	—	—	—	12
	DDT	31	—	19	56	—	31	—	—	—	—	—	—
	Dieldrin	—	13	17	13	—	—	—	—	—	—	—	—
1971	DDE	—	43	14	13	—	10	13	15	T	—	14	T
	TDE	—	30	12	12	—	23	27	44	—	—	15	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1972	DDE	100	59	—	—	—	—	—	—	—	—	—	—
	TDE	25	20	—	—	—	—	—	—	—	—	—	—
	DDT	15	T	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	T	—	—	—	—	—	—	—	—	—	—
STATION 4.—LAVACA BAY—66 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	—	T	—	T	13	22
	TDE	—	—	—	—	—	—	—	—	—	—	T	T
	DDT	—	—	—	—	—	—	—	—	—	—	T	10
1966	DDE	33	40	43	56	51	140	39	26	17	T	T	11
	TDE	12	17	T	27	25	30	16	11	—	—	—	—
	DDT	11	18	T	16	21	23	—	—	—	—	—	—
1967	DDE	22	16	20	25	14	T	25	14	14	T	19	26
	TDE	13	T	12	14	T	—	13	—	—	—	—	12
	DDT	—	—	—	T	14	—	34	—	—	—	—	T

TABLE M-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Texas—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 4.—LAVACA BAY—66 SAMPLES ¹ —Continued													
1968	DDE	39	38	39	69	46	140						24
	TDE	16	17	22	62	39	120						13
	DDT	26	40	18	49	34	140						—
	Dieldrin	—	—	—	—	—	24						—
1969	DDE	20	19	33		41	33	—	12	—	26		40
	TDE	T	—	11		19	22	—	—	—	18		33
	DDT	—	—	—		14	16	—	—	—	15		48
1970	DDE	—	120		30	(2)		37	(2)	—	—		16
	TDE	—	42		16	(2)		53	(2)	—	—		T
	DDT	—	26		11	(2)		—	(2)	—	—		—
	Dieldrin	—	—		10	—		—	—	—	—		—
	PCB's	—	—		—	(3)		—	(3)	—	—		—
1971	DDE	48		43	43	22	T	T	—		12	T	13
	TDE	29		15	—	—	—	—	—		25	—	—
	DDT	T		—	—	—	—	—	—		—	—	—
	Dieldrin	—		—	—	—	—	—	—		—	—	14
1972	DDE	18											
	TDE	T											
	DDT	T											
	Dieldrin	21											
STATION 5.—SAN ANTONIO BAY (NORTH)—59 SAMPLES ¹													
1965	DDE							T	T	T	T	11	30
	TDE							T	T	T	T	T	25
	DDT							—	T	—	T	T	16
	Dieldrin							—	—	—	—	—	11
1966	DDE	31	30	32	29	33	29	16	—	T	13	15	19
	TDE	24	27	30	23	27	22	10	—	—	—	T	14
	DDT	14	16	15	—	18	13	—	—	—	—	—	—
	Dieldrin	—	—	17	—	—	—	—	—	—	—	—	—
1967	DDE	17	20	22	29	13	12	T	—	11	14		
	TDE	13	18	18	30	T	—	—	—	T	13		
	DDT	—	—	—	T	—	—	—	—	14	—		
	Dieldrin	—	10	—	—	—	—	—	—	—	—		
1968								No Samples Collected					
1969	DDE							—	—	—	T		18
	TDE							—	—	—	T		12
	DDT							—	—	—	—		T

TABLE M-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Texas—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 5.—SAN ANTONIO BAY (NORTH)—59 SAMPLES ¹ —Continued													
1970	DDE	—	—	18	14	17	—	—	—	—	—	—	—
	TDE	—	—	—	—	33	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	—	—	—	—	—	27	—	—	—	17	—
1971	DDE	T	—	—	—	—	—	—	10	—	T	13	—
	TDE	12	—	—	—	—	—	—	—	—	11	—	—
	DDT	—	—	—	—	—	—	—	19	—	—	—	—
	Dieldrin	—	—	—	—	—	—	—	—	—	—	12	17
1972	DDE	12	12	—	—	T	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	T	—	—	—	—	—	—	—	—	—	—	—
STATION 6.—SAN ANTONIO BAY (SOUTH)—75 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	T	T	T	12	17	—
	TDE	—	—	—	—	—	—	—	T	T	10	T	—
	DDT	—	—	—	—	—	—	—	T	T	T	T	—
1966	DDE	13	13	14	19	14	T	—	—	—	T	15	10
	TDE	T	—	—	14	10	—	—	—	—	—	T	T
	DDT	T	—	—	—	10	—	—	—	—	—	T	—
1967	DDE	17	20	20	14	T	—	—	—	T	—	—	—
	TDE	11	10	11	—	—	—	—	—	T	—	—	—
	DDT	10	—	11	—	—	—	—	—	T	—	—	—
1968	DDE	—	—	—	21	T	—	—	110	—	—	—	T
	TDE	—	—	—	19	—	—	—	310	—	—	—	T
	DDT	—	—	—	13	—	—	—	68	—	—	—	—
	Dieldrin	—	—	—	56	—	—	—	14	—	—	—	—
	Endrin	—	—	—	10	—	—	—	—	—	—	—	—
1969	DDE	T	16	20	16	T	—	—	—	—	T	T	12
	TDE	—	—	—	14	T	—	—	—	—	T	T	15
	DDT	—	—	—	T	—	—	—	—	—	—	—	T
	Dieldrin	14	—	—	—	—	—	—	—	—	—	—	—
1970	DDE	—	—	—	—	11	—	—	—	—	—	13	—
	TDE	—	—	—	—	25	—	—	—	—	—	41	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1971	DDE	—	—	—	—	T	—	—	—	—	—	T	13
	TDE	—	—	—	—	—	—	—	—	—	—	—	10
	DDT	—	—	—	—	—	—	—	—	—	—	—	T

TABLE M-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Texas—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 6.—SAN ANTONIO BAY (SOUTH)—75 SAMPLES ¹ —Continued													
1972	DDE	13	16			T							
	TDE	—	—			—							
	DDT	—	—			—							
STATION 7.—ST. CHARLES BAY—66 SAMPLES ¹													
1966	DDE	11	T	11		11	—	—	—	—	T	T	15
	TDE	—	—	—		—	—	—	—	—	—	—	T
	DDT	—	—	—		T	—	—	—	—	—	—	T
	Dieldrin	—	—	—		—	—	—	—	—	—	—	11
1967	DDE	15	20	17	23	16	—	—	—	12	15	16	17
	TDE	T	T	T	52	47	—	—	—	—	—	T	12
	DDT	T	T	—	—	23	—	—	—	—	—	—	10
	Dieldrin	13	12	—	28	39	78	80	—	—	—	—	15
1968 ⁴	DDE	22	30	—	24	19	—	T	—	—	—	—	—
	TDE	—	20	—	19	16	—	—	—	—	—	—	—
	DDT	—	43	—	11	23	—	—	—	—	—	—	—
1969	DDE	—	—	14	12	T	T	—	—	—	—	—	T
	TDE	—	—	T	21	17	15	—	—	—	—	—	—
	DDT	—	—	—	—	T	—	—	—	—	—	—	—
	Dieldrin	49	—	—	—	—	—	27	—	T	—	—	—
1970	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1971 ⁴	DDE	—	—	—	—	—	—	—	—	T	T	—	13
	TDE	—	—	—	—	—	—	—	—	T	T	—	T
	DDT	—	—	—	—	—	—	—	—	21	10	—	15
1972 ⁴	DDE	14	23	27	16	—	—	—	—	—	—	—	—
	TDE	T	T	10	—	—	—	—	—	—	—	—	—
	DDT	14	15	—	—	—	—	—	—	—	—	—	—
STATION 8.—ARANSAS BAY—19 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	—	T	21	—	—	—
	TDE	—	—	—	—	—	—	—	T	T	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1966	DDE	12	16	20	16	16	10	T	T	T	27	15	15
	TDE	T	45	57	—	43	35	30	33	20	26	42	43
	DDT	T	—	T	—	14	—	—	—	—	—	—	T

TABLE M-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Texas—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 8.—ARANSAS BAY—19 SAMPLES ^{1, 4} —Continued													
1967	DDE	23	24	27	—	T							
	TDE	54	49	56	—	—							
	DDT	T	T	—	—	—							
	Dieldrin	—	—	—	28	48							
STATION 9.—COPANO BAY—51 SAMPLES ^{1, 4}													
1967	DDE						—	—	—	—	T	—	21
	TDE						—	—	96	—	20	—	30
	DDT						—	—	—	—	—	—	T
1968	DDE	14	18	—	21	T	12	T	—	—	—	—	—
	TDE	23	28	—	24	—	—	—	—	—	—	—	—
	DDT	—	—	—	T	—	—	—	—	—	—	—	—
1969	DDE	—	T	—	50	15	17	—	—	—	—	—	15
	TDE	T	—	—	21	27	23	—	—	—	—	—	T
	DDT	—	—	—	T	18	T	—	—	—	—	—	T
1970	DDE	17	15	25	14	—	—	—	—	—	—	—	—
	TDE	11	T	T	14	—	—	—	—	—	—	—	—
	DDT	T	T	—	T	—	—	—	—	—	—	—	—
1971	DDE	10	13	15	17	10	—	—	—	—	—	—	—
	TDE	10	11	10	39	10	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 10.—RED FISH BAY—67 SAMPLES ^{1, 4}													
1966	DDE					29		24	12	T	T	T	T
	TDE					25		21	18	T	11	T	T
	DDT					12		12	—	—	—	—	—
1967	DDE	T	15	17	—	T	14	T	—	—	—	—	—
	TDE	T	21	32	—	14	39	19	—	—	—	—	—
	DDT	—	T	T	—	—	13	14	—	—	—	—	—
1968	DDE		23	25	18		12	10	10	11	—	—	10
	TDE		14	26	43		18	21	18	15	21	—	17
	DDT		—	12	21		—	—	T	23	T	—	T
1969	DDE	15	T	10	15	—	T	—	—	12	—	11	17
	TDE	27	T	19	27	18	T	—	—	—	—	15	22
	DDT	13	—	T	19	—	—	—	—	—	—	T	13

TABLE M-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Texas—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 10.—RED FISH BAY—67 SAMPLES ¹ —Continued													
1970	DDE	16	14	18	17	10	12	14	12	—	—	—	T
	TDE	20	19	29	25	23	38	25	18	—	—	—	29
	DDT	T	10	13	12	—	—	—	10	—	—	—	15
	PCB's	—	—	—	—	—	—	—	—	—	—	(^b)	—
1971	DDE	—	—	17	—	T	—	—	13	T	T	T	12
	TDE	—	—	14	—	—	—	—	13	—	T	T	11
	DDE	—	—	—	—	—	—	—	17	26	17	22	26
1972	DDE	14	20	16	18	—	T	—	—	—	—	—	—
	TDE	11	15	16	12	—	14	—	—	—	—	—	—
	DDT	22	30	45	42	—	34	—	—	—	—	—	—
	PCB's	—	—	—	—	—	—	—	—	—	—	—	(^b)
STATION 11.—NUECES BAY—20 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	—	—	—	—	T	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1966	DDE	—	—	34	22	32	—	120	18	18	T	T	—
	TDE	—	—	30	17	26	—	200	20	16	—	12	—
	DDT	—	—	12	—	14	—	130	—	—	—	—	—
	Dieldrin	—	—	—	—	—	—	33	—	—	—	—	—
1967	DDE	31	30	43	46	34	29	20	32	57	51	—	—
	TDE	36	61	110	110	48	52	20	20	25	22	—	—
	DDT	T	22	20	26	22	49	17	37	35	15	—	—
	Dieldrin	—	11	13	19	—	—	—	—	—	—	—	—
	Endrin	—	18	12	11	—	—	—	—	—	—	—	—
1968	DDE	—	—	45	—	—	—	—	—	—	—	—	—
	TDE	—	—	28	—	—	—	—	—	—	—	—	—
	DDT	—	—	15	—	—	—	—	—	—	—	—	—
STATION 12.—ARROYO COLORADO—48 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	—	—	170	24	55	64
	TDE	—	—	—	—	—	—	—	—	520	33	80	80
	DDT	—	—	—	—	—	—	—	—	20	T	17	16
	Dieldrin	—	—	—	—	—	—	—	—	19	T	29	34
	Endrin	—	—	—	—	—	—	—	—	—	—	32	19
1966	DDE	80	120	120	74	96	230	300	270	98	12	180	63
	TDE	110	140	130	70	69	140	230	93	57	—	50	58
	DDT	21	19	17	—	26	31	53	24	—	—	19	12
	Dieldrin	32	23	24	18	16	30	45	27	14	—	18	20
	Endrin	18	17	14	—	22	23	28	13	—	—	14	12

TABLE M-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Texas—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 12.—ARROYO COLORADO—48 SAMPLES ¹ —Continued													
1967	DDE	120	140	210	170	110	160	160	79				
	TDE	73	110	180	75	49	63	92	49				
	DDT	27	—	28	19	26	24	23	16				
	Dieldrin	23	16	42	46	19	33	30	16				
	Endrin	11	29	19	—	—	—	12	—				
1968	DDE			48									160
	TDE			150									68
	DDT			—									49
	Dieldrin			—									33
1969	DDE	260	330	220	320	180	260	280	86	100	110	210	54
	TDE	110	100	48	100	35	63	55	28	33	30	T	21
	DDT	15	57	35	110	77	48	22	—	—	T	—	24
	Dieldrin	14	16	17	18	14	25	18	17	12	T	16	—
1970	DDE	23	120	140	110	130	96						
	TDE	35	20	25	29	25	54						
	DDT	32	19	22	T	T	60						
	Dieldrin	21	18	23	25	13	25						
	Endrin	T	T	—	—	—	12						
1971	DDE				65	280	380	220					
	TDE				14	61	46	78					
	DDT				—	—	—	—					
	Dieldrin				11	27	24	38					
	Toxaphene				—	—	—	(a)					
STATION 13.—LOWER LAGUNA MADRE—24 SAMPLES ¹													
1965	DDE									T	T	—	—
	TDE									—	—	—	—
	DDT									—	—	—	—
1966	DDE	—	—	—	—	27	T	T	13	—	12	—	T
	TDE	—	—	—	—	19	—	—	—	—	—	—	—
	DDT	—	—	—	—	11	—	—	—	—	—	—	—
	Dieldrin	—	—	—	—	—	46	—	—	—	—	—	—
1967	DDE	11	15	13	13	10	12	T	—				
	TDE	—	—	—	—	—	T	—	—				
	DDT	—	—	—	—	14	T	—	—				

¹ Each sample represents 15 or more mature mollusks.

² DDT present but not quantified due to presence of PCB's in sample.

³ Present but not quantified.

⁴ Dieldrin data omitted because of possible sample contamination

SECTION N.—VIRGINIA

The eastern oyster, *Crassostrea virginica*, was monitored at 10 principal stations in estuarine areas of Virginia during the period July 1965 - February 1972. Samples were analyzed at the Gulf Breeze Laboratory until June 1968, and thereafter at the Virginia Institute of Marine Science. The approximate station locations are shown in Fig. N-1. A summary of data on organochlorine residues in the monitored species, *C. virginica*, is presented in Table N-1, and the distribution of residues in this species for each sampling station by data of collection in Table N-2.

The 87% incidence of DDT residues in Virginia samples and the maximum residue of 678 ppb were fourth highest of the States monitored. The higher residues were clearly associated with intensive truck farming (Station 2) and a combination of urban and industrial development (Station 9).

The presence of PCB's was noted in 1970 samples, but not until 1971 was equipment acquired to identify and quantify these compounds. The residue of 2,800 ppb of Aroclor 1254[®] detected in oysters in the Elizabeth River, a highly industrialized area, has prompted a special study to pinpoint the source of this pollution.

Trends in DDT residues in Virginia oysters differ somewhat from other areas in that, while the larger residues (those above 100 ppb) decreased by 66% in 1971, 100% of the 1971 samples contained residues in excess of 11 ppb as compared to 82% in earlier years. It appears that DDT residues are more widely dispersed but at relatively lower levels, presumably through the processes of recycling.

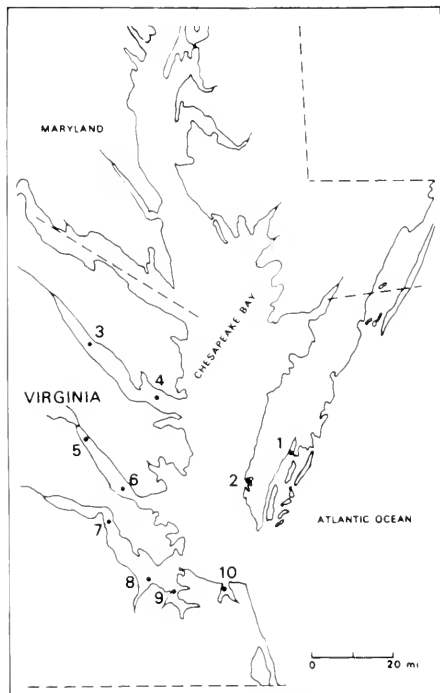


FIGURE N-1.—Diagram of coastal Virginia showing approximate location of monitoring stations

1. Machipongo River
2. Cherrystone Inlet—Chesapeake Bay
3. Bowers Rock—Rappahannock River
4. Urbana—Rappahannock River
5. Bell Rock—York River
6. Pages Rock—York River
7. Deep Water Shoals—James River
8. Nansemond Ridge—James River
9. Hospital Point—Elizabeth River
10. Lynnhaven Bay

TABLE N-1.—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1965-72—Virginia

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (µG KG)		
				DDT	DIELDRIN	PCB'S ²
1	Machipongo River	1965-72	67	56 (127)	2 (11)	1 (390)
2	Cherrystone Inlet	1965-72	68	67 (678)		2 (510)
3	Bowers Rock	1965-72	70	62 (60)		2 (400)
4	Urbana	1965-72	69	59 (45)		2 (270)
5	Bell Rock	1965-72	69	35 (54)		2 (450)
6	Pages Rock	1965-72	68	50 (100)	3 (T)	2 (400)
7	Deep Water Shoals	1965-72	69	69 (144)	38 (40)	3 (1,000)

TABLE N-1.—Summary of data on organochlorine residues in the monitored species (*C. virginica*), 1965-72—
Virginia—Continued

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (μg/kg)		
				DDT	DIELDRIN	PCB'S ²
8	Nansemond Ridge	1965-72	64	63 (128)	29 (22)	2 (1,000)
9	Hospital Point	1966-72	58	58 (300)	38 (24)	3 (2,800)
10	Lynnhaven Bay	1965-70	62	61 (113)	4 (16)	
	Occasional stations (4)	1965-67	5	5 (241)		
Total number of samples			669			
Percent positive for indicated compound				87	17	3

NOTE: T = >5 but < 10 ppb.

¹ Each sample represents 15 or more mature mollusks

TABLE N-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Virginia
(Blank = no sample collected; -- = no residue detected above 5 ppb or no residue detected (PCB'S); T = >5 but <10 ppb)

YEAR	COMPOUND	RESIDUES IN PPB (μg/kg)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 1.—MACHIPONGO RIVER—67 SAMPLES ¹													
1965	DDE							34	15	18	T	13	15
	TDE							20	13	28	T	T	T
	DDT							73	T	24	--	--	--
	Dieldrin							11	--	--	--	--	--
1966	DDE	12	T	11	14	19	17	19	--	T	T	--	T
	TDE	--	--	--	T	14	18	17	--	--	--	--	--
	DDT	--	--	--	--	--	--	--	--	--	--	--	--
1967	DDE	--	34	T	T	17	14	T	T	--	--	T	11
	TDE	--	59	--	--	12	13	--	T	--	--	--	--
	DDT	--	10	--	--	--	--	T	--	T	--	--	--
1968	DDE	T	--	T	--	T	18	15	T	--	T	11	T
	TDE	--	--	--	--	--	10	20	T	--	T	T	--
	DDT	--	--	--	--	--	--	10	--	--	--	--	--
1969	DDE	T	T	T	T	T	13	--	11		T	20	T
	TDE	--	T	--	--	T	13	--	16		T	27	T
	DDT	--	T	--	--	--	T	--	--		--	16	--
	Dieldrin	--	--	--	--	--	--	--	--		T	--	--
1970	DDE	15	10	--	--	11	24		T	T	T	11	
	TDE	14	T	--	--	T	35		13	T	T	T	
	DDT	--	11	--	--	--	17		T	--	--	--	
1971	DDE						10		18		17		
	TDE						--		T		T		
	DDT						--		--		--		
	PCB'S						--		--			2 390	

TABLE N-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Virginia—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)												
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.	
STATION 1.—MACHIPONGO RIVER—67 SAMPLES ¹ —Continued														
1972	DDE		12											
	TDE		—											
	DDT		—											
STATION 2.—CHERRYSTONE INLET—68 SAMPLES ¹														
1965	DDE							60	35	16	24	25	45	
	TDE							89	60	14	42	32	55	
	DDT							230	71	T	35	20	35	
1966	DDE	45	42	33	43	36	32	90	41	14	25	19	36	
	TDE	49	46	45	40	36	37	110	86	35	66	62	73	
	DDT	23	26	25	15	11	16	130	59	T	20	11	14	
1967	DDE	49	31	32	37	45	37	34	44	55	26	24	20	
	TDE	74	52	36	53	75	68	61	63	81	42	29	18	
	DDT	17	14	—	12	10	21	110	110	120	22	13	T	
1968	DDE	19	27	35	59	42	40	146	63	20	33	33	T	
	TDE	18	20	46	58	55	52	210	172	76	31	31	15	
	DDT	T	—	T	21	12	17	322	42	12	T	T	T	
1969	DDE	24	11	21	17	12	T	15	35	17	24	16	35	
	TDE	16	14	22	16	13	14	10	31	22	34	10	39	
	DDT	—	—	—	—	—	—	—	20	11	13	—	17	
1970	DDE	32	33	—	20	28			24	T	22	18	19	
	TDE	30	42	—	21	29			26	T	34	19	22	
	DDT	T	16	—	—	—			23	T	T	—	T	
1971	DDE							30				29		
	TDE							30		14		23		
	DDT							—		14		T		
	PCB's							—		—		350		
1972	DDE		43											
	TDE		18											
	DDT		—											
	PCB's		510											
STATION 3.—BOWLERS ROCK—70 SAMPLES ¹														
1965	DDE							16	T	T	T	T	11	
	TDE							23	T	T	T	T	12	
	DDT							21	T	T	T	—	—	
1966	DDE	11	10	—	13	11	13	13	13	—	—	T	—	
	TDE	T	T	—	T	11	15	15	T	—	—	T	—	
	DDT	—	—	—	—	—	T	—	—	—	—	—	—	

TABLE N-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Virginia—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 3.—BOWLERS ROCK—70 SAMPLES ¹ —Continued													
1967	DDE	—	T	T	10	T	T	10	—	—	T	T	T
	TDE	—	—	T	11	T	11	14	15	—	T	T	T
	DDT	—	—	—	—	—	—	11	18	—	T	T	—
1968	DDE	—	T	T	T	T	13	10	10	—	17	T	12
	TDE	—	—	T	—	—	14	12	15	—	17	T	T
	DDT	—	—	—	—	—	—	—	T	—	T	—	—
1969	DDE	T	T	11	12	10	T	T	T	T	11	T	11
	TDE	10	T	10	14	10	T	T	T	10	18	14	13
	DDT	—	—	—	—	T	—	—	T	T	14	T	T
1970	DDE	16	10	T	T	10	10	T	T	—	T	—	T
	TDE	14	T	—	11	10	17	12	T	T	12	T	T
	DDT	T	32	—	—	—	T	—	—	T	T	—	—
1971	DDE					11				T			14
	TDE					T				—			12
	DDT					—				—			—
	^a PCB's					T				—			400
1972	DDE		17										
	TDE		12										
	DDT		—										
STATION 4.—URBANA—69 SAMPLES ¹													
1965	DDE							T	10	T	T	T	13
	TDE							13	16	T	—	—	10
	DDT							14	19	T	—	—	—
1966	DDE	10	T	—	—	10	T	15	—	T	—	—	T
	TDE	T	—	—	—	—	—	—	—	—	—	—	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	T
1967	DDE	12	10	11	T	T	T	T	T	—	T	11	11
	TDE	—	T	T	T	T	T	T	T	—	11	T	12
	DDT	—	—	—	—	—	—	—	T	—	12	—	T
1968	DDE	T	T	T	11	T	11	14	T	—	T	T	T
	TDE	T	—	T	T	—	—	13	T	—	T	12	T
	DDT	—	—	—	—	—	—	T	—	—	—	T	—
1969	DDE	T	T	—	T	T	T	T	T	T	11	T	T
	TDE	T	11	—	—	T	T	T	13	T	18	10	10
	DDT	T	T	—	—	—	—	—	—	—	16	—	—
1970	DDE		T	—	T	10	15	T	—	10	—	T	T
	TDE		10	—	T	T	17	T	T	17	—	T	T
	DDT		16	—	—	—	—	T	T	—	14	—	T

TABLE N-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Virginia—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 4.—URBANA—69 SAMPLES ¹ —Continued													
1971	DDE					10				T		10	
	TDE					T				—		T	
	DDT					—				—		—	
	* PCB'S					T				—		270	
1972	DDE		10										
	TDE		T										
	DDT		—										
STATION 5.—BELL ROCK—69 SAMPLES ¹													
1965	DDE							11	T	T	T	11	16
	TDE							24	T	13	10	13	21
	DDT							19	T	10	T	—	13
1966	DDE	—	—	—	—	—	—	—	—	—	—	—	T
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	T	—	T	—	—	—	—	—	—	—	T
	TDE	—	T	—	—	—	—	—	—	—	12	—	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	T	T	—	T	T	11
	TDE	—	—	—	—	—	—	10	T	—	T	T	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	11
1969	DDE	—	—	—	—	—	T	—	T	T	11	—	17
	TDE	—	—	—	—	—	T	—	T	14	20	—	T
	DDT	—	—	—	—	—	—	—	—	—	10	—	—
1970	DDE	T	T	—	T	T	10	10	—	—	—	—	T
	TDE	T	12	11	11	T	18	13	T	T	—	—	T
	DDT	—	—	—	—	—	—	T	—	—	—	—	—
1971	DDE					T				—		T	
	TDE					—				T		T	
	DDT					—				—		—	
	PCB'S					—				—		³ 390	
1972	DDE		T										
	TDE		T										
	DDT		—										
	PCB'S		² 450										
STATION 6.—PAGES ROCK—68 SAMPLES ¹													
1965	DDE							10	T	11	11	17	19
	TDE							15	12	17	14	20	16
	DDT							17	14	14	12	24	13

TABLE N-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Virginia—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{g}/\text{kg}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 6.—PAGES ROCK—68 SAMPLES ¹ —Continued													
1966	DDE	14	—	—	T	10	11	—	T	—	T	—	T
	TDE	13	—	—	—	T	11	—	T	—	T	—	T
	DDT	T	—	—	—	—	—	—	11	—	—	—	—
1967	DDE	—	—	T	10	—	T	T	—	T	T	T	13
	TDE	—	—	—	T	—	T	T	—	16	14	11	14
	DDT	—	—	—	—	—	—	—	T	14	T	—	—
1968	DDE	T	T	—	T	—	12	13	T	—	T	T	10
	TDE	—	T	—	—	—	—	14	10	—	10	T	11
	DDT	—	—	—	—	—	—	—	—	—	—	—	T
1969	DDE	—	—	T	—	T	13	—	—	14	18	—	T
	TDE	—	—	T	—	T	T	—	—	12	16	—	T
	DDT	—	—	T	—	—	—	—	—	T	T	—	—
	Dieldrin	—	—	—	—	—	—	—	—	—	T	—	—
1970	DDE	—	T	—	T	T	T	T	T	—	10	90	T
	TDE	T	20	—	T	T	29	12	T	—	T	10	T
	DDT	—	—	—	—	—	11	T	—	—	—	—	—
1971	DDE	—	—	—	—	T	—	—	—	—	—	—	T
	TDE	—	—	—	—	—	—	—	—	—	—	—	T
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
	PCB's	—	—	—	—	—	² T	—	—	—	—	—	³ 400
1972	DDE	—	T	—	—	—	—	—	—	—	—	—	—
	TDE	—	T	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 7.—DEEP WATER SHOALS—69 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	21	18	10	13	30	40
	TDE	—	—	—	—	—	—	52	31	17	22	41	56
	DDT	—	—	—	—	—	—	63	35	T	13	17	23
	Dieldrin	—	—	—	—	—	—	14	T	—	—	11	13
1966	DDE	37	11	24	26	30	40	37	23	11	18	19	17
	TDE	43	15	24	32	45	63	57	41	21	29	28	32
	DDT	15	—	—	—	14	20	30	22	T	16	15	10
	Dieldrin	—	—	—	23	34	38	16	17	—	12	—	—
1967	DDE	21	26	19	31	19	20	20	T	17	13	21	19
	TDE	29	30	21	41	25	32	37	25	33	20	23	20
	DDT	12	13	—	19	13	12	18	24	28	10	11	24
	Dieldrin	—	14	—	40	28	21	22	11	12	—	—	—
1968	DDE	15	14	17	15	15	23	18	14	T	15	12	19
	TDE	15	12	15	15	19	29	26	25	T	20	T	18
	DDT	—	—	—	—	11	14	15	18	—	T	T	T
	Dieldrin	—	—	12	12	16	16	T	T	—	—	T	—

TABLE N-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Virginia—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 7.—DEEP WATER SHOALS—69 SAMPLES ¹ —Continued													
1969	DDE	T	T	10	11	T	10	18	14	17	20	28	
	TDE	10	T	12	T	T	16	12	16	23	28	40	
	DDT	—	—	—	—	T	T	—	16	21	20	29	
	Dieldrin	—	Lost	—	—	T	11	13	T	—	T	14	
1970	DDE	41	19	10	27	10	12	T	40	20	25	12	17
	TDE	43	22	11	12	T	21	22	71	40	43	17	28
	DDT	60	—	T	—	—	17	T	10	14	25	T	12
	Dieldrin	T	—	14	—	—	12	16	—	—	—	—	—
1971	DDE					40				T		16	
	TDE					35				13		21	
	DDT					T				—		—	
	Dieldrin					31				T		17	
	^a PCB's					1,000				—		560	
1972	DDE		15										
	TDE		15										
	DDT		—										
	Dieldrin		10										
	PCB's		^a 760										
STATION 8.—NANSEMOND RIDGE—64 SAMPLES ¹													
1965	DDE							16	17	17	T	27	36
	TDE							59	43	29	14	37	49
	DDT							53	39	25	T	24	31
	Dieldrin							17	T	—	—	T	11
1966	DDE	28	14	16	30	30	36		34	13	10	11	11
	TDE	35	14	17	34	29	52		55	18	15	16	19
	DDT	15	—	—	13	13	29		29	—	10	16	10
	Dieldrin	—	—	—	17	14	22		14	—	—	—	—
1967	DDE	14	17	22	18	15	14	14	18	T	13	17	20
	TDE	17	16	20	18	17	20	24	35	16	24	24	25
	DDT	T	T	11	T	T	12	11	20	T	10	13	10
	Dieldrin	—	—	—	15	12	T	12	14	—	—	10	11
1968	DDE	12	15	14	12	32	24	23	14	—	T	16	16
	TDE	14	16	15	12	47	32	38	29	—	12	13	22
	DDT	—	T	T	—	45	27	23	15	—	—	T	T
	Dieldrin	—	11	—	10	21	15	T	—	—	—	—	—
1969	DDE	—	T	10	T	T	11	T	50	11	16		16
	TDE	T	T	11	T	T	23	T	37	14	28		26
	DDT	—	—	—	—	—	10	—	23	T	13		12
	Dieldrin	—	—	—	—	—	10	—	—	—	11		T

TABLE N-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Virginia—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 8.—NANSEMOND RIDGE—64 SAMPLES ¹ —Continued													
1970	DDE	12	15	—				10	16	10	T		T
	TDE	18	40	13				27	23	14	12		14
	DDT	22	11	35				—	13	11	10		T
	Dieldrin	T	—	T				T	—	—	—		—
1971	DDE					22				11			16
	TDE					18				13			15
	DDT					T				—			—
	Dieldrin					15				T			20
	PCB's					^a 1,000				—			^a 440
1972	DDE		16										
	TDE		14										
	DDT		—										
	Dieldrin		—										
STATION 9.—HOSPITAL POINT—58 SAMPLES ¹													
1966	DDE			140	82	63	83	66	37	20	24	26	27
	TDE			120	73	60	130	96	63	36	53	42	40
	DDT			40	32	31	89	39	43	22	35	27	24
	Dieldrin			13	18	15	20	—	—	—	—	—	T
1967	DDE	42	52	26	34	33	37	29	11	20	39	43	54
	TDE	48	53	17	25	42	76	67	55	59	83	78	64
	DDT	31	29	T	—	20	58	36	31	62	63	100	37
	Dieldrin	T	16	—	—	11	16	—	—	10	15	16	19
1968	DDE	68	92	60	57	48	50	30	20	14	13	11	26
	TDE	79	67	48	49	48	93	67	62	30	24	17	21
	DDT	52	55	18	20	34	83	35	19	12	10	T	T
	Dieldrin	13	19	15	12	12	13	10	T	—	10	11	—
1969	DDE	12	17	31	22	15	15	15	28	33	32		29
	TDE	T	11	32	21	28	28	33	70	84	92		56
	DDT	—	—	11	19	11	27	15	41	46	71		36
	Dieldrin	—	T	10	10	T	T	10	T	19	10		T
1970	DDE	15	32	15				T	18	24	13	T	15
	TDE	21	54	35				22	37	57	30	13	23
	DDT	T	29	—				10	19	28	14	—	11
	Dieldrin	T	14	—				—	—	—	—	—	—
1971	DDE					40				27			27
	TDE					40				49			32
	DDT					20				—			—
	Dieldrin					T				12			24
	^a PCB's					2,800				—			960

TABLE N-2.—Distribution of organochlorine residues in *C. virginica* for each sampling station by date of collection—Virginia—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 9.—HOSPITAL POINT—58 SAMPLES ¹ —Continued													
1972	DDE		34										
	TDE		32										
	DDT		—										
	Dieldrin		Lost										
	^a PCB's		1,440										
STATION 10.—LYNNHAVEN BAY—62 SAMPLES ¹													
1965	DDE							26	18	13	14	13	31
	TDE							49	33	10	24	14	40
	DDT							17	12	T	—	T	T
	Dieldrin							10	—	—	—	—	—
1966	DDE	19	20	17	32	16	25	36	16	16	14	17	14
	TDE	25	29	20	39	20	41	59	21	24	22	27	26
	DDT	—	—	—	T	—	11	15	—	—	T	—	T
1967	DDE	16	22	19	29	15	24	34	17	21	20	20	18
	TDE	19	27	21	36	18	35	57	33	33	32	27	25
	DDT	T	T	—	T	T	20	22	17	16	17	11	10
	Dieldrin	—	—	—	16	—	—	—	—	—	—	—	—
1968	DDE	19	T	29	18	27	30	15	14	11	16	15	19
	TDE	22	—	43	18	28	45	T	21	13	20	12	20
	DDT	T	—	12	—	10	27	12	T	—	T	—	T
	Dieldrin	—	—	13	—	—	—	—	—	—	—	—	—
1969	DDE	18	16	12	11	16	12		27	14	11	20	18
	TDE	16	22	21	11	21	16		28	18	20	28	23
	DDT	—	T	T	—	T	—		17	T	10	—	—
	Dieldrin	—	—	—	—	—	—		—	—	—	10	—
1970	DDE	—	20	—	T	11	28			18		18	14
	TDE	—	26	—	—	—	29			20		12	23
	DDT	T	—	—	—	—	T			10		T	T

¹ Each sample represents 15 or more mature mollusks.

² Calculated as Aroclor 1242®.

³ Calculated as Aroclor 1254®.

SECTION O.—WASHINGTON

The Pacific oyster, *Crassostrea gigas*, was used to monitor 19 estuarine sites at monthly intervals in the period October 1965 - December 1968. All samples were analyzed at the Gulf Breeze Laboratory. The approximate station locations are shown in Fig. O-1. A summary of data on organochlorine residues in the monitored species, *C. gigas*, is presented in Table O-1, and the distribution of residues in this species for each sampling station by date of collection in Table O-2.

The monitoring program was terminated in Washington after 3 years because of the absence of detectable DDT residues in most samples. This was due to the absence of DDT pollution and not because of any lack of sensitivity on the part of the monitored species. Analyses of samples of the Pacific oyster in California waters had demonstrated its ability to store organochlorine residues at levels comparable to other molluscan species in the same estuary.

The overall incidence of DDT residues in Washington samples was only 11%. The maximum residue detected, 176 ppb, was the obvious result of a single pollution incident. Station 18 was the only one demonstrating a continuing, but low-level pollution problem. The fact that residues at this station were primarily DDT rather than one of its metabolites suggests a direct application of the pesticide to coastal waters. Analytical data are too few, even at Station 18, to indicate any trend in DDT pollution. The overall picture is that of an estuarine area of the United States that was remarkably free from DDT pollution in the period 1965-68.

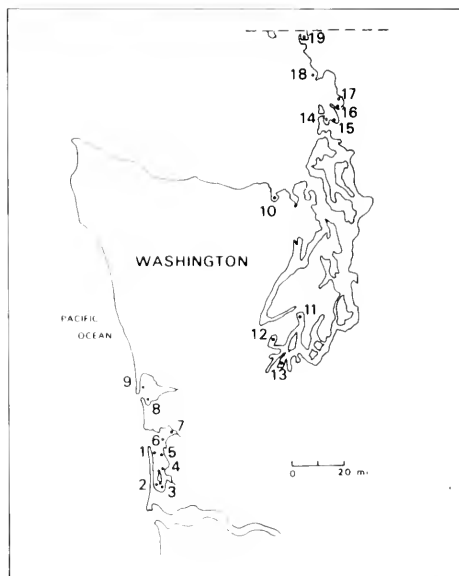


FIGURE O-1.—Diagram of coastal Washington showing approximate location of monitoring stations

- | | |
|----------------------------------|-------------------------------------|
| 1. Stackpole Harbor—Willapa Bay | 11. North Bay Reserve—Willapa Bay |
| 2. Olson Slough—Willapa Bay | 12. Oakland Bay Reserve—Puget Sound |
| 3. Bear River—Willapa Bay | 13. Mud Bay—Puget Sound |
| 4. Naselle River—Willapa Bay | 14. Padilla Bay—Padilla Bay |
| 5. Nemah River—Willapa Bay | 15. Swinomish—Padilla Bay |
| 6. Stony Point—Willapa Bay | 16. Scott Point—Samish Bay |
| 7. South Bend—Willapa Bay | 17. Rock Point—Samish Bay |
| 8. Beardslee Slough—Grays Harbor | 18. Lummi—Lummi Bay |
| 9. Oyehut—Grays Harbor | 19. Blaine—Drayton Harbor |
| 10. Sequim Bay | |

TABLE O-1.—Summary of data on organochlorine residues in the monitored species (*C. gigas*), 1965-68—Washington

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (µg/kg)	
				DDT	DIELDRIN
1	Stackpole Harbor	1965-68	38	9 (25)	
2	Olson Slough	1966-68	30	7 (55)	
3	Bear River	1965-68	38	3 (17)	
4	Naselle River	1965-68	38	1 (11)	1 (120)
5	Nemah River	1965-68	39	4 (21)	
6	Stony Point	1965-68	39	10 (176)	
7	South Bend	1965-68	39	6 (23)	
8	Beardslee Slough	1965-68	37	2 (27)	
9	Oyehut	1966-68	36		
10	Sequim Bay	1966-68	31		
11	North Bay Reserve	1965-68	33		
12	Oakland Bay Reserve	1965-68	33		

TABLE O-1.—Summary of data on organochlorine residues in the monitored species (*C. gigas*), 1965-68—
Washington—Continued

STATION NUMBER	LOCATION	MONITORING PERIOD	NUMBER OF SAMPLES ¹	NUMBER OF POSITIVE SAMPLES AND MAXIMUM RESIDUE () DETECTED IN PPB (µg/kg)	
				DDT	DIELDRIN
13	Mud Bay	1965-68	32		
14	Padilla Bay	1965-68	39	8 (17)	
15	Swinomish	1965-68	38	1 (T)	
16	Scott Point	1965-68	39	4 (10)	
17	Rock Point	1965-68	37		
18	Lummi	1965-68	38	23 (99)	
19	Blaine	1965-68	38		
	Occasional stations (2)	1966	3		
Total number of samples			695		
Percent of samples positive for indicated compound				11	<1

NOTE: T = >.5 but <10 ppb.

¹ Each sample represents 15 or more mature mollusks.

TABLE O-2.—Distribution of organochlorine residues in *C. gigas* for each sampling station by date of collection—Washington
[Blank = no sample collected; — = no residue detected above 5 ppb; T = .5 but <10 ppb]

YEAR	COMPOUND	RESIDUES IN PPB (µg/kg)													
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.		
STATION 1.—STACKPOLE HARBOR—38 SAMPLES ¹															
1965	DDE											T	T	—	
	TDE												T	—	—
	DDT												T	—	—
1966	DDE	—	—	T	T	T	13	11	—	T	—	—	—	—	
	TDE	—	—	T	—	—	12	—	—	—	—	—	—	—	
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	—	
1967	DDE	—	—	—	—	T	—	—	—	—	—	—	—	—	
	TDE	—	—	—	—	T	—	—	—	—	—	—	—	—	
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	—	
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—	—	
	TDE	—	—	—	—	—	—	—	—	—	—	—	—	—	
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	—	
STATION 2.—OLSON SLOUGH—30 SAMPLES ¹															
1966	DDE							20	T	—	—	—	—	—	
	TDE							19	10	—	—	—	—	—	
	DDT							16	—	—	—	—	—	—	
1967	DDE	—	T	—	T	—	T	—	—	—	—	—	—	—	
	TDE	—	11	—	T	—	12	—	—	—	—	—	—	—	
	DDT	—	14	—	14	—	14	—	—	—	—	—	—	—	

TABLE O-2.—Distribution of organochlorine residues in *C. gigas* for each sampling station by date of collection—Washington—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{g}/\text{kg}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 2.—OLSON SLOUGH—30 SAMPLES ¹ —Continued													
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	T	—	—	—	—	—	—	—	—	—	—	—
	DDT	15	—	14	—	—	—	—	—	—	—	—	—
STATION 3.—BEAR RIVER—38 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	—	—	—	—	—	T
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1966	DDE	—	—	—	—	—	T	—	—	—	—	—	—
	TDE	—	—	—	—	—	12	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	—	—	—	—	T	—	—	—	—	—	—
	TDE	—	—	—	—	—	T	—	—	—	—	—	—
	DDT	—	—	—	—	—	T	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 4.—NASELLE RIVER—38 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1966	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	11	—	—	—	—	—	—	—	—	—	—	—
	Dieldrin	—	—	—	—	—	—	120	—	—	—	—	—
STATION 5.—NEMAH RIVER—39 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	—	—	—	—	—	T
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

TABLE O-2.—Distribution of organochlorine residues in *C. gigas* for each sampling station by date of collection—Washington—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 5.—NEMAH RIVER—39 SAMPLES ¹ —Continued													
1966	DDE	—	—	—	—	—	11	T	—	—	—	—	—
	TDE	—	—	—	—	—	10	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	T	—	—	—	—	—	—	—	—	—	—	—
STATION 6.—STONY POINT—39 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	—	—	—	T	T	—
	TDE	—	—	—	—	—	—	—	—	—	14	T	—
	DDT	—	—	—	—	—	—	—	—	—	T	—	—
1966	DDE	T	—	—	T	T	T	13	—	—	—	—	—
	TDE	T	—	—	T	—	11	11	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	26	—	—	—	—
	DDT	—	—	—	—	—	—	—	150	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	T	—	T	—	—	—	—	—	—	—	—	—
STATION 7.—SOUTH BEND—39 SAMPLES ¹													
1965	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1966	DDE	T	—	T	T	T	—	10	—	—	—	—	—
	TDE	T	—	16	T	—	—	13	—	—	—	—	—
	DDT	—	—	—	T	—	—	—	—	—	—	—	—
1967	DDE	—	T	—	—	—	—	—	—	—	—	—	—
	TDE	—	T	—	—	—	—	—	—	—	—	—	—
	DDT	—	T	—	—	—	—	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—

TABLE 2-1.—Continued.—Organochlorine residues in *C. fluminea* in each sampling station in each of collection months, 1966-67.

STATION	COMPOUND	RESIDUES IN P.P.M. IN FAT											
		JUN	JUL	AUG	SEP	OCT	NOV	DEC	JAN	FEB	MAR	APR	
STATION 1—BEAVERCREEK SLUDGE—17 SAMPLES													
1	DDE												
	DDT												
	DDE												
2	DDE												
	DDT												
	DDE												
3	DDE												
	DDT												
	DDE												
4	DDE												
	DDT												
	DDE												
5	DDE												
	DDT												
	DDE												
6	DDE												
	DDT												
	DDE												
STATION 2—CREEK—19 SAMPLES													
1	DDE												
	DDT												
	DDE												
2	DDE												
	DDT												
	DDE												
3	DDE												
	DDT												
	DDE												
4	DDE												
	DDT												
	DDE												
5	DDE												
	DDT												
	DDE												
6	DDE												
	DDT												
	DDE												

TABLE C-1—Distribution of reported vehicle fatalities in 1966 by each reporting county in each of counties—A through G—Continued

Year	County	Percent of total for											
		Jan	Feb	Mar	Apr	May	June	July	Aug	Sept	Oct	Nov	Dec
STATION 1—NORTH 3-1-1 FIFTEEN 3-1-1 SAMPLES													
1965	DOE												
	TDE												
	DOT												
1966	DOE	-	-	-	-	-	-	-	-	-	-	-	-
	TDE	-	-	-	-	-	-	-	-	-	-	-	-
	DOT	-	-	-	-	-	-	-	-	-	-	-	-
1967	DOE	-	-	-	-	-	-	-	-	-	-	-	-
	TDE	-	-	-	-	-	-	-	-	-	-	-	-
	DOT	-	-	-	-	-	-	-	-	-	-	-	-
1968	DOE	-	-	-	-	-	-	-	-	-	-	-	-
	TDE	-	-	-	-	-	-	-	-	-	-	-	-
	DOT	-	-	-	-	-	-	-	-	-	-	-	-
STATION 2—NORTH 3-1-1 FIFTEEN 3-1-1 SAMPLES													
1965	DOE												
	TDE												
	DOT												
1966	DOE	-	-	-	-	-	-	-	-	-	-	-	-
	TDE	-	-	-	-	-	-	-	-	-	-	-	-
	DOT	-	-	-	-	-	-	-	-	-	-	-	-
1967	DOE	-	-	-	-	-	-	-	-	-	-	-	-
	TDE	-	-	-	-	-	-	-	-	-	-	-	-
	DOT	-	-	-	-	-	-	-	-	-	-	-	-
1968	DOE	-	-	-	-	-	-	-	-	-	-	-	-
	TDE	-	-	-	-	-	-	-	-	-	-	-	-
	DOT	-	-	-	-	-	-	-	-	-	-	-	-
STATION 3—NORTH 3-1-1 FIFTEEN 3-1-1 SAMPLES													
1965	DOE												
	TDE												
	DOT												
1966	DOE	-	-	-	-	-	-	-	-	-	-	-	-
	TDE	-	-	-	-	-	-	-	-	-	-	-	-
	DOT	-	-	-	-	-	-	-	-	-	-	-	-
1967	DOE	-	-	-	-	-	-	-	-	-	-	-	-
	TDE	-	-	-	-	-	-	-	-	-	-	-	-
	DOT	-	-	-	-	-	-	-	-	-	-	-	-
1968	DOE	-	-	-	-	-	-	-	-	-	-	-	-
	TDE	-	-	-	-	-	-	-	-	-	-	-	-
	DOT	-	-	-	-	-	-	-	-	-	-	-	-

TABLE O-2.—Distribution of organochlorine residues in *C. gigas* for each sampling station by date of collection—Washington—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)												
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.	
STATION 13.—MUD BAY—32 SAMPLES ¹ —Continued														
1968	DDE		—	—	—	—	—	—	—	—				
	TDE		—	—	—	—	—	—	—	—				
	DDT		—	—	—	—	—	—	—	—				
STATION 14.—PADILLA BAY—39 SAMPLES ¹														
1965	DDE											T	T	—
	TDE											—	—	—
	DDT											—	—	—
1966	DDE	—	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	T	—	—	—	T	—	—	—	—	—	—	—
1967	DDE	—	—	—	—	—	—	—	—	T	T	T	—	
	TDE	—	—	—	—	—	—	—	—	—	—	T	—	
	DDT	—	—	—	—	—	—	—	—	12	T	T	—	
1968	DDE	T	—	—	—	—	—	—	—	—	—	—	—	
	TDE	—	—	—	—	—	—	—	—	—	—	—	—	
	DDT	T	—	—	—	—	—	—	—	—	—	—	—	
STATION 15.—SWINOMISH—38 SAMPLES ¹														
1965	DDE											—	—	—
	TDE											—	—	—
	DDT											—	—	—
1966	DDE	—	—	—	—	—	—	—	—	—	—	—	—	
	TDE	—	—	—	—	—	—	—	—	—	—	—	—	
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	
1967	DDE	—	—	—	—	—	—	—	—	—	—	—	—	
	TDE	—	—	—	—	—	—	—	—	—	—	—	—	
	DDT	—	T	—	—	—	—	—	—	—	—	—	—	
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—	
	TDE	—	—	—	—	—	—	—	—	—	—	—	—	
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	
STATION 16.—SCOTT POINT—39 SAMPLES ¹														
1965	DDE											—	—	—
	TDE											—	—	—
	DDT											—	—	—

TABLE O-2.—Distribution of organochlorine residues in *C. gigas* for each sampling station by date of collection—Washington—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{G}/\text{KG}$)											
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
STATION 16.—SCOTT POINT—39 SAMPLES ¹ —Continued													
1966	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	T	T	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	T	T	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	T	—	—	—	—
STATION 17.—ROCK POINT—37 SAMPLES ¹													
1965	DDE										—	—	—
	TDE										—	—	—
	DDT										—	—	—
1966	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—
STATION 18.—LUMMI—38 SAMPLES ¹													
1965	DDE										—	—	—
	TDE										—	—	—
	DDT										—	—	—
1966	DDE	—	—	—	—	—	—	—	—	—	—	T	T
	TDE	—	—	—	—	—	—	—	—	—	—	15	—
	DDT	—	—	—	—	—	T	—	—	—	—	—	18
1967	DDE	T	—	T	T	T	11	T	T	—	T	—	T
	TDE	10	—	T	11	T	14	T	T	T	T	—	T
	DDT	25	19	29	44	42	74	34	14	15	21	21	19
1968	DDE	T	—	—	—	10	T	T	T	—	—	—	—
	TDE	—	—	—	—	T	T	—	—	—	—	—	—
	DDT	17	19	22	33	25	24	14	17	—	—	—	—

TABLE O-2.—Distribution of organochlorine residues in *C. gigas* for each sampling station by date of collection—Washington—Continued

YEAR	COMPOUND	RESIDUES IN PPB ($\mu\text{g}/\text{kg}$)												
		JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.	
STATION 19.—BLAINE—38 SAMPLES ¹														
1965	DDE											—	—	—
	TDE											—	—	—
	DDT											—	—	—
1966	DDE	—	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	—
1967	DDE	—	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	—
1968	DDE	—	—	—	—	—	—	—	—	—	—	—	—	—
	TDE	—	—	—	—	—	—	—	—	—	—	—	—	—
	DDT	—	—	—	—	—	—	—	—	—	—	—	—	—

¹ Each sample represents 15 or more mature mollusks.

PESTICIDES IN WATER

Organochlorine Insecticide Residues in Streams Draining Agricultural, Urban-Agricultural, and Resort Areas of Ontario, Canada—1971¹

J. R. W. Miles and C. R. Harris

ABSTRACT

Organochlorine insecticide residues in water systems draining agricultural, urban-agricultural, and resort areas of Ontario, Canada, were compared by analysis of water, bottom mud, and fish, collected during the period from mid-April to mid-October 1971. Insecticides detected were p,p'-DDT, o,p'-DDT, p,p'-DDE, o,p'-TDE, p,p'-DDE, γ -chlordane, dieldrin, endrin, endosulfan, heptachlor, heptachlor epoxide, lindane, and aldrin. Insecticide concentrations in water from all three areas were less than the "Maximum Reasonable Stream Allowances" for growing fish that are safe for human consumption.

The concentrations of total DDT in the water were combined with water flow data to calculate the weekly rate of transport of total DDT at each sampling time. The greatest transport of total DDT was by the Muskoka River which drains the Muskoka Lakes resort area where DDT was used until 1966 for control of biting flies: a peak of 11.8 lb total DDT per week was recorded in May, but this transport quickly lessened, resulting in a May to October average of 1.9 lb total DDT per week. Corresponding figures for the Thames River (urban-agricultural) were peak 2.5 lb and average 0.4 lb total DDT per week and for Big Creek (agricultural), peak 0.5 lb and average 0.2 lb per week.

The ratio of concentration of total DDT in mud to total DDT in water was as great as 800; total DDT in fish to total DDT in water was as great as 1 million. The ratio of p,p'-TDE to p,p'-DDT was <1 in water but >1 in bottom mud, indicating possible dechlorination of p,p'-DDT to p,p'-TDE in the bottom mud. Polychlorinated biphenyls (PCB's) were present in the urban-agricultural area samples of bottom mud and fish at levels up to 217 ppm and about 0.4 ppm, respectively.

Introduction

In a previous issue of this Journal (4), this laboratory reported on the transport of insecticides into Lake Erie by two water systems draining agricultural areas in southwestern Ontario (Big Creek, which flows into Lake Erie, and a controlled drainage system near Erieau, Ontario, the water of which is pumped into Lake Erie). In 1971, in order to compare residue contributions from differing areas of insecticide usage, the present study was made of insecticides transported by water systems draining agricultural, urban-agricultural, and resort areas. The agricultural area studied (Fig. 1, Area 1) was the same 280 square miles of predominantly tobacco farms in Norfolk County, Ontario, reported in the earlier study, whose drainage enters Lake Erie by Big Creek. DDT has been used extensively in this area for many years primarily for cutworm and hornworm control. Since January 1970, use of DDT has been restricted to cutworm control on tobacco and requires a provincial government permit prior to its use. The urban-agricultural area (Fig. 1, Area 2) was the City of London, Ontario, (population 200,000) on the Thames River which drains 1,200 square miles of mixed agricultural land (chiefly dairy cattle) before flowing through London; the Thames River flows into Lake St. Clair which empties into Lake Erie via the Detroit River. DDT usage in this area has also been subject to the provincial ban of 1970. The resort area (Fig. 1, Area 3) was the Muskoka Lakes region of Ontario, 1,700 square miles which drains into Georgian Bay and Lake Huron by the Muskoka River. DDT had been used extensively in this area for biting fly control until 1966 when this use was discontinued. Average water flow for the three streams during 1971 was Big Creek (agricultural) 190 cubic feet per second (CFS), Thames River (urban-agricultural) 1,030 CFS, and Muskoka River (resort) 2,421 CFS.

¹ Contribution No. 528 from the Research Institute, Canada Department of Agriculture, University Sub Post Office, London 72, Ontario.

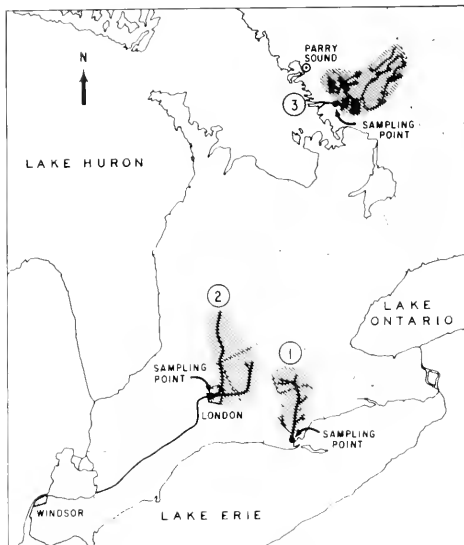


FIGURE 1.—Partial map of Ontario, Canada, showing drainage areas of 1—Big Creek (agricultural), 2—Thames River (urban-agricultural), and 3—Muskoka Lakes (resort)

In Big Creek drainage area, the overburden consists mainly of glacial till of Pleistocene age with dominantly coarse-textured soil formed on sand and gravel. The adjacent Thames River drainage area is also of glacial till but is dominantly fine-textured soil formed on very fine sands and silts. The Muskoka Lakes region is a Precambrian area with little or no true soil and consists of lakes and rock outcrops with conifers growing on shallow soils and detritus trapped between rocks.

Materials and Methods

SAMPLE COLLECTION

Water, bottom mud, and fish samples were collected between mid-April and mid-October 1971. Water and bottom mud samples in Big Creek and the Thames River were taken every 2 weeks. Depth-integrated water samples were collected in 32-fluid ounce, narrow-necked glass bottles clamped to a 24-ft long aluminum pole. The contents of two bottles were combined for one water sample. Bottom mud samples were collected using a sampler (authors' design) consisting of a steel container, 3 $\frac{1}{4}$ inches in diameter and 1 $\frac{3}{4}$ inches deep, attached to the end of a 24-ft aluminum pole. Five samples of mud were taken, ranging from near the bank to mid-stream, and combined into one composite sample. Water and bottom mud samples from the Muskoka River were taken at monthly intervals by personnel of the Ontario Water Resources Commission and shipped

to London, Ontario, for analysis. The difference in frequency from the other two rivers was necessitated by the greater distance of the Muskoka Lakes from the laboratory. Water samples were collected from 10 feet below the surface using Kemmerer bottles. Bottom mud samples were collected with an Eckmann Dredge. Water depth at sampling points was about 30 feet.

Stream flow data were measured and compiled by personnel of the Water Survey of Canada using the "General Procedure for Gaging Streams" (6).

Fish were collected in the fall of 1971 by personnel of the Ontario Department of Lands and Forests, using electric shockers and gill nets.

EXTRACTION AND FRACTIONATION

Water plus any suspended matter was analyzed "as is" without filtration. The contents of the bottles were measured by weighing bottles before and after emptying the contents into a 2-liter separatory funnel. The empty bottles were rinsed with 10 ml acetone to remove any insecticide adsorbed on the walls, and the acetone rinse was added to the separatory funnel. The bottles were then rinsed with 100 ml of hexane which was also added to the separatory funnel. The funnel was shaken for 2 minutes, and the layers allowed to separate. The aqueous layer was withdrawn into a second 2-liter separatory funnel and extracted twice more with 50-ml portions of hexane, the first separatory funnel being used twice. The three hexane extracts were combined in a 500-ml round-bottom flask and concentrated for fractionation on a Florisil column.

Extraction of bottom mud and fish and fractionation of extracts on Florisil have been previously described (4).

GAS-LIQUID CHROMATOGRAPHY

Two Model 1400 and one Model 1200 Varian Aerograph gas chromatographs were used. The column of one Model 1400 was packed with 5% XE-60, the other with mixed 3% DC-200/4.5% QF-1. The column of the Model 1200 was packed with 5% DC-200. Solid support in all columns was 80/100 mesh Chromosorb W, A.W. DMCS treated; all columns, 6 ft long x 2 mm (i.d.), were operated at 180° C. Nitrogen was the carrier gas at a flow rate of 40 ml/min.. Tritium electron capture detectors were used on all three gas chromatographs. All samples were run on all three columns.

In the samples of bottom mud and fish from the urban-agricultural area (Thames River), interference from polychlorinated biphenyls (PCB's) measured as Aroclor 1254[®] (up to 217 ppm in bottom mud, about 0.4 ppm in fish) was too great to allow direct GLC determination of *p,p'*-DDT, *o,p'*-DDT, and *p,p'*-DDE. In these samples the DDT compounds were converted to dichlorobenzophenones, separated from the PCB's on the Florisil column, and then assayed by GLC (5).

RECOVERY

Recoveries of reported insecticides from water fortified at 100 parts per 10¹² (American trillion) ranged from 75% to 102%. Recoveries of the insecticides added to bottom mud at 0.2 ppm were 91% to 103%. Recoveries of the reported insecticides added directly to fish muscle at 0.1 ppm before extraction ranged from 91% to 95%.

Results and Discussion

The concentrations of insecticides found in water samples are given in Table 1. In all three water systems, residues of *p,p'*-DDT were the highest, followed by dieldrin and *o,p'*-DDT. Peak concentrations for all insecticides occurred during spring runoff in May, with subsequent leveling off at lower concentrations during the rest of the sampling period. For these uniform

periods, data have been condensed by reporting average concentrations and ranges (Table 1). For the sampling period, the averages for total DDT in water were similar for the three areas—28, 23, and 18 parts per 10¹² (American trillion) for agricultural, urban-agricultural, and resort areas, respectively. All insecticide concentrations in water were below the "Maximum Reasonable Stream Allowances" (2) in which it is presumably safe to grow fish for human consumption.

The concentrations of total DDT in the water were combined with water flow data measured in cubic feet per second (CFS) to calculate the weekly rate of transport of total DDT at each sampling time (Fig. 2). Although the peak transport (in May) for the Muskoka River (resort area) was 11.8 lb total DDT per week, the average for the period sampled was 1.9 lb per week.

TABLE 1.—*Insecticide concentrations in water of three streams in Ontario, Canada—1971*

DATE (1971)	RESIDUES IN PARTS PER 10 ¹² (AMERICAN TRILLION)											
	TOTAL DDT	<i>p,p'</i> - DDE	<i>o,p'</i> - DDT	<i>p,p'</i> - DDT	<i>p,p'</i> - TDE	DIEL- DRIN	<i>γ</i> -CHLOR- DANE	ENDO- SULFAN	ENDRIN	HEPTA- CHLOR	ALDRIN	LINDANE
BIG CREEK (AGRICULTURAL)												
Apr. 13	30	5	13	10	2	3	<1	2	<1	<1	—	—
26	31	5	13	10	3	41	<1	2	<1	<1	—	—
May 4	75	15	13	40	11	23	<1	11	<1	2	—	—
11	79	5	12	58	4	11	<1	<1	<1	<1	—	—
25	37	4	4	27	2	6	<1	<1	1	<1	—	—
June 8	37	4	5	24	4	5	<1	<1	3	<1	—	—
22	19	1	2	13	3	4	<1	<1	7	<1	—	—
July 26 to Oct. 12 ¹												
Average	16	2	2	9	2	4	<1	1	1	<1	—	—
Range	11-20	1-3	1-3	5-12	1-3	3-6	<1-2	<1-3	<1-3	<1-1	—	—
THAMES RIVER (URBAN-AGRICULTURAL)												
Apr. 15	22	3	10	7	2	4	<1	1	—	<1	<1	1
26	18	3	3	9	3	32	<1	2	—	<1	<1	1
May 5	73	3	13	53	4	7	<1	3	—	1	<1	1
12	50	3	4	40	3	7	<1	<1	—	<1	<1	<1
26	24	2	3	16	3	7	1	<1	—	2	2	<1
June 9 to Sept. 15 ¹												
Average	13	1	2	8	2	4	2	<1	—	<1	<1	1
Range	<10-16	<1-2	1-4	5-9	<1-5	3-5	<1-6	<1	—	<1-1	<1-2	<1-4
Sept. 29	30	2	4	23	1	9	8	<1	—	1	7	<1
Oct. 13	24	1	3	18	2	18	21	<1	—	<1	1	<1
28	19	3	2	11	3	8	3	<1	—	<1	<1	1
MUSKOKA RIVER (RESORT)												
May 5	57	9	6	28	14	11	—	—	2	<1	<1	<1
26	20	3	4	12	1	2	—	—	<1	<1	<1	<1
June 10	17	1	5	11	<1	4	—	—	<1	<1	<1	<1
June 23 to Nov. 4 ¹												
Average	10	1	2	7	1	4	—	—	<1	<1	<1	<1
Range	8-13	<1-2	1-2	4-9	<1-2	3-5	—	—	<1-1	<1	<1	<1

NOTE: — = not detected; <1 indicates that qualitative identification of the compound was made on all three GLC liquid phases, but the amount was less than the lowest level of reporting, i.e., <1 parts per 10¹² (American trillion). Heptachlor epoxide and *o,p'*-TDE were not detected in water samples.

¹ Because of the leveling off of insecticide residues at lower concentrations for these uniform periods, data have been condensed by reporting average concentrations and ranges.

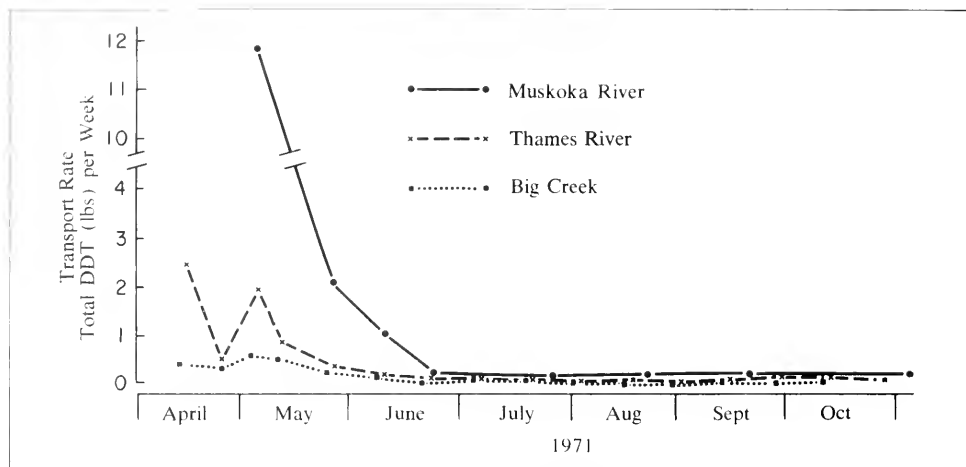


FIGURE 2.—Transport of total DDT per week by water of Big Creek (agricultural), Thames River (urban-agricultural), and Muskoka River (resort)

Similarly, the Thames River (urban-agricultural) averaged 0.4 lb total DDT per week and Big Creek (agricultural) averaged 0.2 lb total DDT per week. To properly relate these data, the size of the three drainage areas must be considered, i.e. resort, 1,700 square miles; urban-agricultural, 1,200 square miles; and agricultural, 280 square miles. Average amounts of total DDT transported per week per 100 square miles were: Muskoka River (resort)-0.11 lb; Big Creek (agricultural)-0.05 lb; and Thames River (urban-agricultural)-0.03 lb. It is surprising that based on area the resort district is still the largest contributor of DDT, since the last official use of DDT for biting-fly control was in 1966. In part, this difference may be explained by the different techniques used in these areas for insect control. In the agricultural area (Big Creek), DDT, applied primarily for cutworm control on tobacco, was incorporated into the soil. By contrast, in the resort area (Muskoka River), DDT was applied by ground or air application over land and water. As noted in the introduction, the Muskoka district has little or no true soil, thus the insecticide would tend to accumulate in the surface detritus with the possibility of greater surface erosion.

The concentrations of insecticides found in the bottom mud are listed in Table 2. These concentrations were uniform for each compound, and results for each month have been averaged for presentation in this table. Concentrations of total DDT in the bottom mud of Big Creek (agricultural) were 604 times those in the water, while the ratios of total DDT in bottom mud/water for the Thames River (urban-agricultural) and the Muskoka River (resort) were 109:1 and 863:1, respectively. The

reason for the low bottom mud/water ratio for total DDT in the Thames River is that the Thames had the lowest insecticide concentration in the bottom mud, average total DDT = 2.5 parts per 10^9 (American billion). These data may be interpreted to mean that the DDT contamination in the Thames River did not result from erosion of treated soil into the stream, but rather from domestic DDT usage with direct contamination of the water.

Ratios of *p,p'*-DDE:*p,p'*-TDE:*p,p'*-DDT in bottom mud may indicate the metabolic history of residues in specific areas. These ratios were as follows: Big Creek (agricultural) 0.7:1.0:1.0; Thames River (urban-agricultural) 1.9:5.7:1.0; and Muskoka River (resort) 2.5:1.8:1.0. The higher DDE/DDT ratios in the Thames and Muskoka Rivers compared to Big Creek could indicate that these residues are older (the last official use of DDT in the resort area was in 1966). The high TDE/DDT ratio in the Thames River bottom mud suggests greater anaerobic dechlorination of DDT (1,3) and may indicate the presence of sewage.

Residues found in fish from the three areas are listed in Table 3. Magnification of total DDT concentration from water to fish was greatest (1 million times) in the lake trout from the resort area. No direct comparisons can be made because the same species of fish were not obtained from all three areas, but it is interesting to note the differences in the ratios of *p,p'*-DDE:*p,p'*-TDE:*p,p'*-DDT in fish from Big Creek (agricultural)-3.6:0.7:1.0 and from Muskoka River (resort)-0.6:1.0:1.0, indicating more metabolism to DDE in the smaller fish from the agricultural area than in the larger fish from

the resort area. In the fish from the urban-agricultural area (Thames River), levels of total DDT were very low (0.02-0.04 ppm), and the major part of this residue was *p,p'*-DDE and *p,p'*-TDE; levels of *p,p'*-DDT were <0.01 ppm.

Summary

Insecticide residues in water, bottom mud, and fish were compared in streams draining agricultural, urban-agricultural, and resort areas during spring runoff and summer of 1971. Transport of total DDT was calculated by combining insecticide concentrations in water with stream flow measurements. Average transports (April-October) of total DDT were Muskoka River (resort) 0.11 lb per week per 100 square miles, Big Creek (agricultural) 0.05 lb per week per 100 square miles, and Thames River (urban-agricultural) 0.03 lb per week per 100 square miles. The heaviest contributor of DDT among these three water systems was the resort area in spite of the fact that official DDT usage in that area ceased in 1966. Fish from the resort area contained up to 19 ppm total DDT compared to a high of 1.3 ppm

from the agricultural area stream. Bottom mud and fish from the urban-agricultural river contained PCB's (up to 217 ppm in the mud and about 0.04 ppm in the fish). The very small concentration of DDT in the bottom mud of the urban river indicated that the DDT in this water system did not come from erosion of treated soil.

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- (4) stream flow data, the Water Survey of Canada, Division of Environment, Canada.

See Appendix for chemical names of compounds discussed in this paper

TABLE 2.—Insecticide concentrations in bottom mud from three streams in Ontario, Canada—1971

DATE (1971)	RESIDUES IN PARTS PER 10 ⁶ (AMERICAN BILLION), DRY-WEIGHT BASIS ¹								
	TOTAL DDT	<i>p,p'</i> -DDE	<i>o,p'</i> -DDT	<i>p,p'</i> -DDT	<i>o,p'</i> -TDE	<i>p,p'</i> -TDE	DIELDRIN	γ -CHLORDANE	ENDRIN
BIG CREEK (AGRICULTURAL)									
Apr.	21.0	4.0	1.5	10.2	0.4	4.9	0.8	0.2	<0.2
May	15.6	3.2	0.7	6.3	0.7	4.7	0.7	0.1	0.2
June	19.2	4.8	0.7	6.1	1.1	6.5	0.8	<0.1	<0.2
July	17.9	4.8	0.6	5.2	1.0	6.4	0.7	<0.1	<0.2
Aug.	15.9	4.2	0.3	3.9	1.1	6.4	0.7	<0.1	<0.2
Sept.	14.2	4.1	<0.3	3.9	0.9	5.3	0.7	<0.1	0.2
Oct.	22.2	5.0	0.9	7.8	1.0	7.5	4.5	3.1	0.3
THAMES RIVER (URBAN-AGRICULTURAL)									
May	4.3	1.1	—	—	—	3.2	0.4	—	—
June	2.4	0.5	—	0.2	—	1.6	0.5	—	—
July	2.1	0.5	—	0.4	—	1.3	0.4	—	—
Aug.	2.5	0.5	—	<0.1	—	2.0	0.6	—	—
Sept.	2.3	0.4	—	0.3	—	1.7	0.6	—	—
Oct.	2.0	0.4	—	0.7	—	0.9	0.3	—	—
MUSKOKA RIVER (RESORT)									
May	19.1	7.5	<0.4	4.1	1.1	6.5	1.4	1.9	0.3
June	21.7	9.3	<0.4	3.9	1.1	7.4	0.9	<0.1	<0.3
July	12.0	7.0	<0.4	1.6	0.4	3.0	0.6	<0.1	<0.3
Aug.	8.7	3.9	<0.4	1.3	0.4	3.1	0.6	<0.1	<0.3
Sept.	12.7	7.0	<0.4	0.8	0.6	4.3	1.0	0.5	<0.3

NOTE: — = not detected; <0.1 indicates qualitative identification of the compound on all three GLC liquid phases, but the amount was less than the lowest level of reporting. Aldrin, endosulfan, heptachlor, heptachlor epoxide, and lindane were not detected in bottom mud samples.

¹ Because concentrations were very uniform, results for each month have been averaged for presentation in this table.

TABLE 3.—Insecticide concentrations in fish muscle from three streams in Ontario, Canada—1971

SPECIES	NUMBER OF FISH IN SAMPLES	RESIDUES IN PPM										PERCENT FAT
		TOTAL DDT	<i>p,p'</i> -DDE	<i>o,p'</i> -DDI	<i>p,p'</i> -DDT	<i>o,p'</i> -TDE	<i>p,p'</i> -TDE	DIELDRIN	γ -CHLORDANE	ENDRIN	HEPTACHLOR EPOXIDE	
BIG CREEK (AGRICULTURAL)												
Brown trout	6	1.27	0.79	0.04	0.31	0.01	0.13	0.03	—	<0.01	<0.01	1.8
Rainbow trout												
Small (12-15 cm)	7	0.42	0.37	0.01	—	<0.01	0.04	0.01	—	0.01	<0.01	1.1
Medium (17-22 cm)	8	0.23	0.17	0.01	0.03	<0.01	0.03	0.01	—	0.01	<0.01	0.9
Rock bass	8	0.49	0.30	0.02	0.10	<0.01	0.06	0.03	—	0.03	<0.01	4.0
Bluegill	8	0.45	0.27	0.01	0.10	<0.01	0.07	0.03	—	0.03	<0.01	4.0
THAMES RIVER (URBAN-AGRICULTURAL)												
Carp	3	0.03	0.02	—	<0.01	—	0.01	<0.01	<0.01	<0.01	—	0.8
Bass												
Small (9-14 cm)	3	0.02	0.01	—	<0.01	—	0.01	<0.01	<0.01	<0.01	—	1.8
Medium (16-17 cm)	5	0.04	0.02	—	<0.01	—	0.01	<0.01	<0.01	<0.01	—	1.6
MUSKOKA LAKES (RESORT)												
White sucker	1	1.98	0.96	0.04	0.76	0.01	0.21	0.01	<0.01	<0.01	<0.01	2.8
Lake trout	1	2.47	1.25	0.10	0.90	0.01	0.21	0.01	<0.01	<0.01	<0.01	1.6
Lake trout	1	16.57	5.58	0.71	8.92	0.07	1.29	0.04	0.02	0.01	0.02	9.5
Lake trout	1	5.71	2.32	0.15	2.76	0.02	0.46	0.01	0.01	<0.01	0.01	2.6
Cisco	1	5.11	0.95	0.21	3.41	0.01	0.54	0.02	0.02	<0.01	0.01	8.2
Cisco	1	19.75	3.04	1.00	14.82	0.01	0.88	0.03	0.03	0.01	0.01	9.8
Cisco	1	1.41	0.41	0.05	0.84	<0.01	0.11	0.01	0.01	<0.01	<0.01	4.6
Cisco	1	4.05	0.98	0.10	2.78	<0.01	0.19	0.01	0.01	<0.01	<0.01	5.0
Cisco	1	2.87	0.75	0.06	1.87	0.01	0.18	0.01	0.01	<0.01	<0.01	4.2

NOTE: — = not detected; <0.01 indicates qualitative confirmation on all three GLC columns, but less than the reporting level of 0.01 ppm. Aldrin, endosulfan, heptachlor, and lindane were not detected in fish muscle samples.

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PESTICIDES IN SOIL

Organochlorine Pesticide Residues in Soils and Crops of the Corn Belt Region, United States—1970

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ABSTRACT

In order to determine the levels of organochlorine pesticides in the Corn Belt region of the United States, a study was initiated in 1970 to sample 400 sites in 12 States. The sampling areas followed the historical boundaries of the Corn Belt and were selected from sites designated for the National Soils Monitoring Program. At each site a 2-qt soil sample (composite of 50, 2- by 3-inch cores, taken in a grid pattern over each 10-acre site) was collected as well as a composite sample of any available standing crop. In addition, use records were obtained at each site for the kinds and amounts of pesticides used during the 1970 cropping season as well as the names of other pesticides known to have been used in the previous 5 years. These data indicated that pesticides had been applied to most of the agricultural acreages in the study area (up to 85%). Forty compounds were identified in use records: 20 herbicides, 17 insecticides, and 3 fungicides. Atrazine was most widely used, followed by captan, malathion, 2,4-D, propachlor (Ramrod®), amiben, and aldrin. Forty-five percent of the soil samples analyzed contained residues; 11 pesticides or metabolites were detected. Arsenic, which can occur naturally in soil, was detected in nearly all soil samples. The most commonly detected residues were those of aldrin, chlordane, and dieldrin. Seven compounds, including four DDT metabolites, were detected in cornstalks, soybeans, sorghum grain, sorghum fodder, and hay.

Introduction

Historically, the Corn Belt of the United States has been defined as that area stretching from central Ohio to Nebraska and Kansas where agricultural acreage has been devoted almost exclusively to the growing of corn and silage. Although corn is still the main crop, modern agricultural trends have diversified the production in this particular region to include a variety of other food and feed grains. In 1969 the 12 states outlined in Fig. 1 produced 79% of the total national corn crop, and at the same time, produced 73% of the total soybean crop and 44% of the total sorghum grain (1).

Between 1954 and 1959 there was a 22% decrease in the acreage planted to corn; however, the decrease was offset by a 115% increase in yield per harvested acre during the same time period, amounting to a 75% increase in total production. These increases were partly due to more extensive use of fertilizers and pesticides.

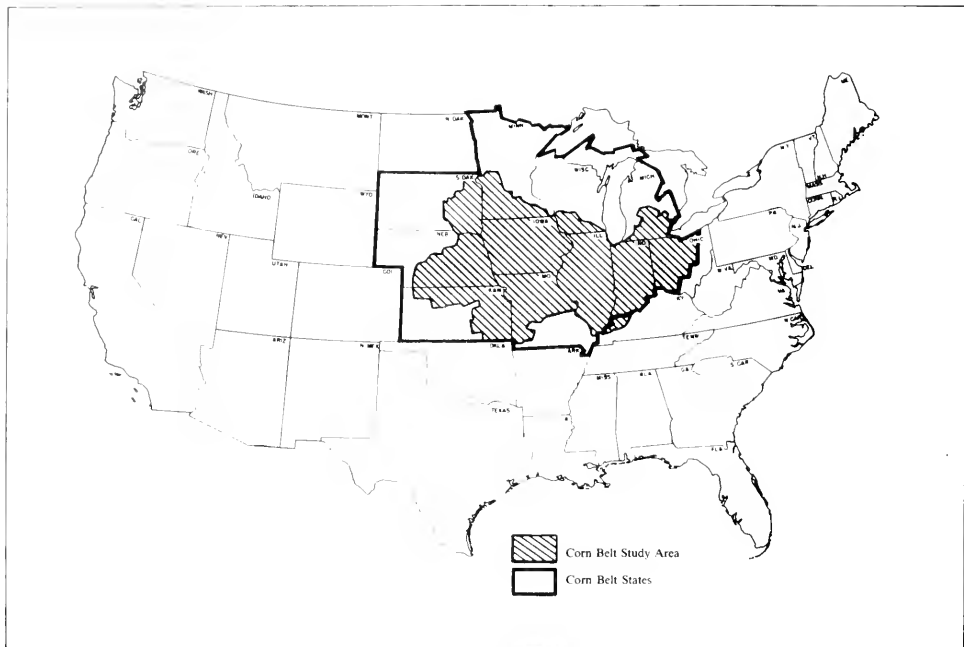
The production and sales of synthetic organic pesticides have followed a generally upward trend which began in the 1940's when many of the chemicals were first introduced; however, in 1969, both production and sales declined for the first time since 1957 (2).

The main objective of this study was to determine the levels in 1970 of organochlorine pesticide residues in soils and crops of the Corn Belt region as shown in Fig. 1. In addition, pesticide use records were obtained for sampling sites, indicating the kinds and amounts of pesticides used for the 1970 cropping season as well as the names of any other pesticides known to have been used on the sites in the previous 5 years.

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FIGURE 1.—Corn Belt region of the United States with area studied—1970



Sampling Procedures

During the late summer of 1970, a total of 400 sites were sampled in the Corn Belt region. These were selected from the 13,300 cropland and noncropland sample sites already designated for the National Soils Monitoring Program, one-fourth of which are sampled each year. The complete sampling scheme for the National Soils Monitoring Program has been reported by Wiersma, Sand, and Cox (3).

Fifty soil cores, each 2 inches in diameter by 3 inches in depth, were collected in a grid pattern over each 10-acre sampling site, composited, and passed through a $\frac{1}{4}$ -inch sieve. A 2-qt metal container was filled from each composite sample, sealed, and shipped to the monitoring laboratory at Gulfport, Miss., for residue analyses.

Crops growing on sites at the time of sampling were collected simultaneously with the soil, i.e., as each soil core was taken, a nearby sample of the standing crop was also taken. The crop samples from each site were composited, air-dried, and packed for shipment to the laboratory. Hay and forage were packed in plastic bags, while corn grain, sorghum grain, and soybeans were packed in 2-qt metal containers.

Analytical Procedures

PREPARATION OF SAMPLES

Soil

A 300-g sample of soil plus 80 ml of water used to wet the soil was extracted with 600 ml of 3:1 hexane isopropanol by concentric rotation for 4 hours. The alcohol was removed by three water washes, and the hexane extract was dried through anhydrous sodium sulfate. The sample extract was then stored at low temperature for subsequent gas-liquid chromatographic analysis.

Crops

For crop samples containing less than 2% fat (corn-stalks, sorghum fodder, and mixed hay), a 100-g sample plus 25 ml of distilled water was blended for 3 minutes in 800 ml of acetonitrile. One-half the sample extract, representing 50 g of original sample, was decanted into a 500-ml graduated cylinder and then transferred to a 500-ml Erlenmeyer flask. After concentration under a three-ball Snyder column to approximately 10 ml, 100 ml of hexane was added, and the hexane-acetonitrile azeotrope was again concentrated to 10 ml. Addition of hexane and concentration to 10 ml were carried out three times to remove essentially all the

acetonitrile. The hexane extract was dried through anhydrous sodium sulfate, the volume adjusted to 100 ml, and the extract stored at low temperature until ready for partitioning.

For crop samples containing more than 2% fat (corn, sorghum grain, and soybeans), a 100-g sample was prewashed with 100 ml of isopropanol and 100 ml of hexane, in that order, and the prewashes discarded. The sample was dried and then dry-blended; 100 ml of isopropanol was added, and the sample was blended again. After the addition of 300 ml of hexane, the isopropanol was removed by two washes with aqueous NaCl solution and one wash with distilled water. The water-alcohol layers were discarded, and the hexane layer was concentrated, adjusted to 100 ml, and held at low temperature for partitioning.

After the above extraction procedure, all crop samples were partitioned with hexane-acetonitrile as follows: a 50-ml portion of the hexane sample extract was shaken with 100 ml of acetonitrile in a 500-ml separatory funnel. The bottom acetonitrile layer was saved. Nanograde acetonitrile (100 ml) was added to the hexane extract, and the separation step above was repeated twice more; then, the hexane was discarded and the three acetonitrile layers combined. The 300-ml acetonitrile extract, which contained essentially all the pesticides in the original hexane extract, was backwashed with 25 ml of acetonitrile-saturated hexane and the hexane layer discarded. The acetonitrile sample extract was concentrated to approximately 10 ml under a three-ball Snyder column, and 100 ml of hexane was added. The addition of hexane and concentration to approximately 10 ml were carried out three times after which the sample was essentially in hexane. The hexane extract was diluted to appropriate volume and held at low temperature for subsequent Florisil column cleanup and fractionation.

GAS-LIQUID CHROMATOGRAPHY

Analyses were performed on gas chromatographs equipped with tritium foil electron affinity detectors for organochlorine compounds and thermionic or flame photometric detectors for organophosphorous compounds. A multiple-column system employing polar and nonpolar columns was utilized to identify and confirm pesticides. Instrument parameters were as follows:

Columns:	Glass, 6 mm o.d. x 4 mm i.d., 183 cm long, packed with one of the following: 9% QF-1 on 100/120 mesh Gas-Chrom Q; 3% DC-200 on 100/120 mesh Gas-Chrom Q; or 1.5% OV-17/1.95% QF-1 on 100/120 mesh Supelcoport
Carrier gases:	5% methane-argon at a flow rate of 80 ml min; prepurified nitrogen at a flow rate of 80 ml min
Temperatures:	Detector 200° C Injection port 250° C Column QF-1 166° C Column DC-200 170°-175° C Mixed column 185°-190° C

Sensitivity (minimum detectable levels) for organochlorine compounds ranged from 0.002 ppm to 0.03 ppm except for mixtures of polychlorinated biphenyls (PCB's), chlordane, toxaphene, etc. whose minimum detectable levels were 0.05 to 0.1 ppm. Minimum detectable levels for organophosphorous compounds were approximately 0.01 to 0.03 ppm. When necessary, confirmation of residues was made by thin layer chromatography or p-values.

Arsenic

Arsenic was determined by atomic absorption spectrophotometry. The soil sample was first extracted with 9.6N hydrochloric acid (HCl) and reduced to trivalent arsenic with stannous chloride. The trivalent arsenic was partitioned from HCl solution to benzene, then further partitioned into water for the absorption measurement. A Perkin-Elmer Model 303 instrument was used and absorbance measured with an arsenic lamp at 1972 Å with argon as an aspirant to an air-hydrogen flame. The minimum detection limit was 0.10 ppm.

RECOVERY STUDIES

For organochlorine pesticides, the average recovery rate in soil was 90% to 110%. Recovery values for stalks and hay ranged from 80% to 95% with an average of 89%; corresponding values for grains were 90% to 100% with a 95% average. For organophosphate pesticides, the average recovery values were 67.1% for soybeans, 86% for sorghum grain, and 60% for corn stalks. Recovery values for arsenic ranged from 70% to 80%. All residue levels were corrected for percent recovery.

Results

PESTICIDE USE RECORDS

The pesticide-use information, as reported by the farmers, was divided into five categories:

	PERCENT OF SITES
(1) No pesticides used in 1970, or in the 5 years prior to 1970	15.5
(2) No pesticides used in 1970, but used in the 5 years prior to 1970	15.5
(3) Pesticides used in 1970 and in the 5 years prior to 1970	53.2
(4) Pesticides used in 1970 only, none in the 5 years prior to 1970	4.8
(5) No use records available	11.0

These data indicate that pesticides had been applied to most of the agricultural acreages sampled in the Corn Belt (up to 85%). Although pesticides reportedly had not been applied in 1970 or the past 5 years on 15.5% of the sites, many of the samples taken from these sites contained pesticide residues; however, this is not unusual. Probable explanations include inaccurate record keeping, spray drift, and the persistent nature of many of the compounds.

Table 1 lists the pesticides used in 1970 on the sample sites, the average application rate, and the number of sites where applied. Table 1 should be considered a conservative estimate of the amounts of pesticides applied to the study areas. In most cases, farmers were most certain of the kinds and amounts of pesticides they had used for the current cropping season. However, in a few cases, farmers knew neither the names nor the amounts of pesticides used on their crops.

The use of unknown seed dressings was common, particularly in Nebraska. Since captan and malathion were indicated as being used almost exclusively as seed dressings, it is likely that they were the "unknown" seed dressings in many cases. Hence their estimates in Table 1 may be particularly low.

TABLE 1.—Pesticides used on sampling sites in the Corn Belt region of the United States during 1970 growing season, average application rate, and number of sites where applied

COMPOUNDS APPLIED	AVERAGE APPLICATION RATE (LB/ACRE)	NUMBER OF SITES WHERE APPLIED
Alachlor	1.22	15
*Aldrin	1.30	27
Amiben	1.08	29
Atrazine	1.51	78
Borax	1.50	1
Butylate (Sutan)	1.10	3
Buxten	1.88	15
*Captan	.03	68
Carbaryl	1.93	6
CDA	.80	1
Ceresan	.01	1
Chloroprotham (CIPC)	.25	2
2,4-D	.55	55
2,4-DB	.50	1
*DDT	2.00	1
*Diazinon	1.34	5
*Dicamba	.94	5
*Dieldrin	.01	2
*Disulfoton	.74	5
*Dyfonate	1.00	1
*Fensulfothion	.80	1
*Fenthion	2.50	2
Furadan	.85	2
*Heptachlor	.19	8
Knoxweed	.35	2
*Lindane	.10	4
Linuron	2.22	5
*Malathion	.02	58
MCPB	2.00	1
*Methoxychlor	.01	5
*Nitralin	.50	1
Norea	.67	3
NPA	.40	2
Panogen	.01	1
*Parathion	.40	4
*Phorate	2.04	13
*Propachlor (Ramrod®)	2.10	31
Propazine	1.60	1
2,4,5-T	.50	1
*Trifluralin	.64	15

* Compounds detectable by described methodology.

In all, 40 different compounds were used: 20 herbicides, 17 insecticides, and 3 fungicides. Atrazine was most widely used, followed by captan, malathion, 2,4-D, propachlor (Ramrod®), amiben, and aldrin. Fenthion was applied most heavily, with an average application rate of 2.5 lb/acre. Although the list of compounds in Table 1 is lengthy, approximately 68% of all the compounds listed were used on five or less sites; 30% of the compounds on the list were used on only one site.

RESIDUES DETECTED IN SOIL

Pesticide residues were detected in 45% of the soil samples analyzed, and 11 pesticides or metabolites were identified. Arsenic, which can occur naturally in soil, was detected in nearly all soil samples. The analytical methods employed were able to detect only 45% of the compounds listed in Table 1 and these are indicated in the table; however, most of the widely used pesticides were included in this group.

Table 2 shows the arithmetic means and ranges of residue levels in soil from each State as well as the percent of positive samples for each compound. The most commonly detected residues were those of dieldrin, aldrin, and chlordane. Since very little dieldrin was used in the study area and the use records indicated that aldrin had been used frequently in the past, most of the dieldrin detected has undoubtedly metabolized from aldrin.

Although chlordane residues were found in soil samples from 7 of the 12 States sampled (approximately 17% of the samples analyzed), no chlordane applications were indicated on any of the use records for 1970 or for the 5 years prior to 1970. Four DDT metabolites (*p,p'*-DDE, *o,p'*-DDT, *p,p'*-DDT, and *p,p'*-TDE) were identified and are listed together as DDTR in Table 2.

Both endrin and isodrin were detected in about 1% of the samples analyzed, but the use records do not indicate that these compounds were applied in 1970 or in previous years. Endrin is currently registered for use on corn and sorghum grain. Since endrin and isodrin are chemically similar to aldrin and dieldrin, it is likely that the small amounts detected have come from the parent compounds. Also, since endrin, isodrin, and chlordane are persistent compounds (4), the amounts detected could have been present from application prior to 1965 or may represent unreported usage.

RESIDUES DETECTED IN CROPS

The crops sampled in the study and the residues detected are listed in Table 3. Seven compounds, including the four DDT metabolites listed above, were detected. The greatest number of residues were detected in cornstalk samples. Approximately 20% of all the cornstalk samples contained pesticide residues; however, no residues were detected in corn grain samples from the same sites.

TABLE 2.—Organochlorine and arsenic residues in soil from the 12 States in the Corn Belt region of the United States—1970
(ND = not detected)

STATE	NO. OF SAMPLING SITES	ALDRIN	CHLORDANE	DIELDRIN	ENDRIN	HEPTACHLOR EPOXIDE	ISOBRIN	TRIFLURALIN	DDTR	ARSENIC
Illinois	69									
	Range of detected residues (ppm)	0.01-0.83	0.05-1.32	0.01-1.08	ND	0.01-0.18	0.01	0.01-0.08	0.06-0.12	0.10-3.60
	Average (ppm)	0.07	0.09	0.14		0.01	<0.01	<0.01	<0.01	1.15
	No. of positive samples ¹ Percent positive sites ²	24 34.8	16 23.2	46 66.7		10 14.5	1 1.4	5 7.2	2 2.9	63 91.3
Indiana	36									
	Range of detected residues (ppm)	0.02-2.41	0.29-4.30	0.01-2.04	0.02	0.27	0.03-0.15	0.08	ND	0.20-14.00
	Average (ppm)	0.15	0.20	0.13	<0.01	0.02	0.01	<0.01	<0.01	4.15
	No. of positive samples ¹ Percent positive sites ²	10 27.8	3 8.3	14 38.9	1 2.8	1 2.8	2 5.6	1 2.8	1 2.8	36 100.0
Iowa	76									
	Range of detected residues (ppm)	0.01-2.98	0.17-3.35	0.01-1.03	0.02	0.01-0.84	0.02-0.31	0.02-0.06	0.01-3.07	0.10-6.10
	Average (ppm)	0.08	0.13	0.10	<0.01	0.02	0.01	<0.01	0.07	1.36
	No. of positive samples ¹ Percent positive sites ²	21 27.6	11 14.5	42 55.3	1 1.3	6 7.9	11 14.5	2 2.6	3 3.9	8 10.5
Kansas	28									
	Range of detected residues (ppm)	0.11	0.32-0.54	0.09-0.21	ND	0.02	0.09	ND	ND	0.20-6.40
	Average (ppm)	0.01	0.03	0.02		<0.01	<0.01			1.55
	No. of positive samples ¹ Percent positive sites ²	1 3.6	2 7.1	3 10.7		1 3.6	1 3.6			28 100.0
Kentucky	1									
	Range of detected residues (ppm)	ND	ND	ND	ND	ND	ND	ND	ND	5.50
	Average (ppm)									5.50
	No. of positive samples ¹ Percent positive sites ²									1 100.0
Michigan	14									
	Range of detected residues (ppm)	ND	ND	0.01	ND	ND	ND	ND	ND	0.20-11.40
	Average (ppm)			<0.01						4.42
	No. of positive samples ¹ Percent positive sites ²			1 7.1					1 7.1	14 100.0

TABLE 2.—Organochlorine and arsenic residues in soil from the 12 States in the Corn Belt region of the United States—1970—Continued
 [ND = not detected]

STATE	NO. OF SAMPLING SITES	ALDRIN	CHLORDANE	DIELDRIN	ENDRIN	HEPTACHLOR	HEPTACHLOR EPOXIDE	ISODRIN	TRIFLURALIN	DDTR	ARSENIC
Minnesota	37										
	Range of detected residues (ppm)	0.05-0.21	0.87	0.08-0.60	ND	ND	0.17	ND	0.09-0.33	ND	0.90-10.80
	Average (ppm)	0.01	0.02	0.05			<0.01		0.02		3.58
	No. of positive samples ¹ Percent positive sites ²	3 8.1	1 2.7	8 21.6			1 2.7		3 8.1		36 97.3
Missouri	31										
	Range of detected residues (ppm)	0.04-0.45	0.14-0.53	0.01-0.66	ND	0.75	0.02-0.05	ND	0.03	ND	0.50-11.50
	Average (ppm)	0.04	0.02	0.05			<0.01		<0.01		3.84
	No. of positive samples ¹ Percent positive sites ²	5 16.1	2 6.4	10 32.3			2 6.4		1 3.2		31 100.0
Nebraska	47										
	Range of detected residues (ppm)	ND	ND	0.01-0.22	0.02	0.02	0.01-0.05	ND	ND	0.10-2.30	0.20-11.90
	Average (ppm)			0.02	<0.01	<0.01	<0.01			0.05	1.73
	No. of positive samples ¹ Percent positive sites ²			20 42.6	1 2.1	1 2.1	5 10.6			2 4.3	33 70.2
Ohio	29										
	Range of detected residues (ppm)	0.02-0.45	ND	0.01-0.45	ND	ND	ND	ND	0.08	ND	0.70-22.75
	Average (ppm)	0.03		0.04					<0.01		6.41
	No. of positive samples ¹ Percent positive sites ²	4 13.8		7 24.1					1 3.4		29 100.0
South Dakota	26										
	Range of detected residues (ppm)	ND	ND	0.09-0.16	0.04	ND	ND	ND	ND	0.02	0.70-7.70
	Average (ppm)			0.01	<0.01					<0.01	2.54
	No. of positive samples ¹ Percent positive sites ²			2 7.7	1 3.8					2 7.7	26 100.0
Wisconsin	5										
	Range of detected residues (ppm)	0.01	0.05	0.21	ND	ND	ND	ND	ND	ND	1.40-4.80
	Average (ppm)	<0.01	0.01	0.04							2.00
	No. of positive samples ¹ Percent positive sites ²	1 20.0	1 20.0	1 20.0							4 80.0

NOTE: Total number of sampling sites used for statistical treatment was 399. One sampling site from Nebraska was omitted.

¹ One sample per site.

² Percent based on number of samples with residues greater than or equal to the sensitivity limits.

TABLE 3.—Residues in crops from the 12 States in the Corn Belt region of the United States—1970
[ND = not detected]

CROP	NO. OF SAMPLES ¹	DIELDRIN	HEPTACHLOR EPOXIDE	p,p'-DDE	p,p'-DDT	o,p'-DDT	p,p'-TDE	DDTR	ETHYL PARATHION	PCB's
Corn	147									
Cornstalks	145									
Range of detected residues (ppm)		0.01-0.04	ND	0.01-0.13	0.01-7.04	0.01-0.51	0.01-0.16	0.01-7.84	0.25	0.53-6.25
Average (ppm)		<0.01		<0.01	0.05	<0.01	<0.01	0.06	<0.01	2.80
No. of positive samples		14		2	3	2	2	3	1	10
Percent positive samples ²		9.7		1.4	3.4	1.4	1.4	3.4	0.7	6.9
Soybeans	75									
Range of detected residues (ppm)		0.01-0.08	0.02-0.03	ND	ND	ND	ND	ND	ND	ND
Average (ppm)		0.01	<0.01							
No. of positive samples		42	2							
Percent positive samples ²		56.0	2.7							
Sorghum grain	24									
Sorghum fodder	21									
Range of detected residues (ppm)		0.01	ND	ND	ND	ND	ND	ND	ND	ND
Average (ppm)		<0.01								
No. of positive samples		1								
Percent positive samples ²		4.8								
Mixed hay	11									
Range of detected residues (ppm)		ND	ND	ND	ND	ND	ND	ND	ND	0.80-2.94
Average (ppm)										0.57
No. of positive samples										3
Percent positive samples ²										27.3

¹ Represents composite samples of available standing crops collected simultaneously with soil samples.

² Percent based on number of samples with residues greater than or equal to the sensitivity limits.

This selective translocation of residues has been observed in other studies (5). The DDTR residues were found in stalk samples from Indiana, Iowa, and Missouri.

Only residues of dieldrin and heptachlor epoxide were found in soybean samples, with 56.0% and 2.7%, respectively, of the samples containing residues. At this time, the tolerance for dieldrin residues in soybeans is zero (6), and the tolerance for combined heptachlor and heptachlor epoxide residues in soybeans has not been established. The sampling may have occurred shortly after an application, and the residues might have been zero if the proper time interval had been observed. Low levels (<0.01 ppm) of dieldrin residues also were detected in sorghum fodder. Crop samples from sites for which records showed usage of organophosphate pesticides in 1970 were analyzed for these compounds; however, only one sample was positive (0.25 ppm ethyl parathion).

In addition to pesticide residues in the crops, two PCB compounds, identified as Aroclor 1232® and 1242®, were also detected. The PCB's were identified by com-

paring retention times of multiple peaks from sample chromatograms to the corresponding multiple peaks from standard chromatograms of the various PCB's. This type of comparison was made on at least two columns of different polarities. Quantitation was based on comparison of the summation of peak height units from the sample chromatogram. At least three peaks were used for quantitation. Standard PCB's were run at least once daily and usually twice or more. The PCB's were found in cornstalk and hay samples from 6 of the 12 States (Ohio, Indiana, Michigan, Illinois, Iowa, and Missouri) but were not detected in corn grain, soybeans, or sorghum. The source of the PCB's has not yet been positively identified but is still under investigation.

Discussion

The presence of residues in crop samples is probably a result of their absorption and translocation to various plant parts. Evidence of translocation of residues has been found in corn (5), soybeans (8-10), alfalfa (10), carrots (14), potatoes (14,15), turnips (8), and pea-

nuts (8). Corn plants have been shown to accumulate dieldrin residues largely in the leaves (5). Crops grown in aldrin-treated soils have been found to contain both aldrin and dieldrin (7).

No trends concerning the pesticide residue levels in agricultural soils of the Corn Belt region can be clearly identified from these data. Pesticides have been widely used throughout the region (on 75% to 85% of the sites sampled), but organochlorine residues in soil were found in less than half of the areas. The ability of a particular soil to retain and/or release pesticide residues is greatly influenced by the interaction between the physical and chemical properties of both the soil and the pesticides applied as well as the microfloral and microfaunal components of the soil system. In general, soils high in organic matter or clay content tend to retain pesticide residues longer than sandy soils or soils low in organic matter (7,10-13,16); this retention ability appears to be most positively correlated with organic matter content. Initial concentration and the formulation of the pesticide are also important. The degradation rate does not remain constant but decreases logarithmically as dosage increases. Sprays tend to disappear sooner than dusts because of a faster initial volatilization, although loss from drift is significant for dusts.

In addition, different crops absorb pesticide residues from the soil at varying rates (5,7,8,14). In this study almost 20% of all crop samples contained detectable residue levels. Future studies should aim for a more comprehensive correlation among soil properties, soil residue levels, and translocation potentials of particular crops.

Acknowledgment

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See Appendix for chemical names of compounds discussed in this paper.

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GENERAL

Decay of Parathion Residues on Field-Treated Tobacco, South Carolina—1972 (II)

Julian E. Keil¹, C. Boyd Loadholt², Samuel H. Sandifer¹, Wayne R. Sitterly³, and Bob L. Brown⁴

ABSTRACT

In an effort to confirm the results of a study in 1971 to determine the length of time required for parathion to degrade to "zero" levels, parathion was applied twice at a rate of .375 lb/acre to field tobacco in South Carolina. After each application, parathion degraded to "zero" levels in 5 days. These results tended to confirm the findings of the original study in which the maximum time required for parathion to degrade to zero levels was estimated to be 7 days and the minimum time 2 days. Weather was characterized by scanty rainfall and temperatures averaging 76° F. During the original study, rainfall was heavy and daily temperatures averaged 80.9° F.

Introduction

A study reported by Keil *et al.* (1) was carried out in June-July 1971 to observe the decline with time of levels of parathion normally applied to field tobacco. This paper reports the results of a second study conducted in June-July 1972 to confirm the findings of the 1971 study.

Methods and Procedures

All growing, application, sampling, and analytical techniques were identical to those of the earlier study (1). Cokers 319 variety tobacco was planted on April 17, 1972. Plots consisting of three 12-foot rows were randomly selected for treatment with parathion, endosulfan, parathion in combination with endosulfan, or as

appropriate controls. Each treatment or control plot was replicated four times in a completely randomized design for a total of 16 plots. Guard rows were used to reduce pesticide drift. In the earlier study, parathion and endosulfan were applied as sprays at rates of .375 and .125 lb active ingredient (A.I.) per acre, respectively, on June 8, 21, and July 8, 1971. In the present study, parathion and endosulfan were sprayed at these same rates on both June 12 and 28, 1972.

Results and Discussion

Results of analyses for parathion residues are given in Table 1 and indicate that parathion residues decayed to "zero" levels in 5 days after each application. Practical "zero" was assumed when statistical differences between actual zero (none detected) and observed values did not exceed the calculated LSD (least significant difference at the .05 level). The results tended to confirm the findings of the original study in 1971 in which the maximum time required for parathion to degrade to zero levels was estimated to be 7 days and the minimum time 2 days.

Endosulfan residues were not measured, but tobacco from the endosulfan-treated plots was analyzed for parathion to determine if there was a measurable amount of drift between plots. The parathion-endosulfan treatment was included to determine if any decay interaction existed between the two chemicals. This was noted at one sampling period, 1 day after application, i.e., more parathion residue was present when parathion-endosulfan was applied. This effect was noted in the previous study up to and including the third day after application.

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Weather during the 1972 study was characterized by scanty rainfall and temperatures averaging 76° F (Table 2). In the 1971 study, rain had been in excess of 12 inches and daily temperatures averaged 80.9° F. Results of both studies support the findings of Maier-Bode (2) who concluded that rainfall does not significantly affect residue levels of parathion.

See Appendix for the chemical names of compounds discussed in this paper.

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TABLE 1.—Parathion residues on field-treated tobacco by treatment plots, South Carolina—1972

APPLICATION DATE	SAMPLING TIME IN DAYS FROM LAST APPLICATION	MEAN PARATHION RESIDUE LEVEL IN PPM ON FOUR REPLICATE TREATMENT PLOTS			
		PARATHION— TREATED PLOT (.375 A.I. LB/ACRE)	ENDOSULFAN— TREATED PLOT (.125 A.I. LB/ACRE)	PARATHION (.375 A.I. LB/ACRE)/ ENDOSULFAN (.125 A.I. LB/ACRE)— TREATED PLOT	CONTROL PLOT
June 12	0	0	0	0	0
	1	1.30	.21	3.37	.05
	3	.46	.05	.26	.03
	5	.03	.01	.09	0
	10	0	0	0	0
	15	0	0	0	0
June 28	1	1.98	.29	1.54	.05
	3	.48	.02	.13	.02
	5	.07	0	0	0
	9	0	0	0	0
	15	0	0	0	0

NOTE: LSD₀₅ = least significant difference at 95% probability level = .50 and may be applied across time or residues.

TABLE 2.—Temperature and rainfall during parathion residue degradation

PERIOD	TIME TO "ZERO" RESIDUE ¹	TEMPERATURE (°F)			RAINFALL (INCHES)
		LOWS	HIGHS	AVG. ²	
June 12-17	5 days	50-67	80-87	72	0
June 28-July 3	5 days	68-70	87-93	80	(July 1) 1.08

¹ "Zero" residue level was assumed when statistical differences between actual zero (none detected) and observed values did not exceed the calculated LSD (least significant difference at the .05 level).

² Overall average temperature, 76° F.

Pesticide Sales and Usage in Kentucky—1968¹

E. Edsel Moore

ABSTRACT

In Kentucky during 1968, 135 pesticide compounds were applied for agricultural and nonagricultural use in a volume of approximately 3.9 million pounds technical material (excluding most pesticides formulated for home, lawn, and garden use). This amounted to about 0.5% of the Nation's total consumption of pesticides. The pesticide poundage used in Kentucky, of which 67% was herbicides, was applied to an estimated 1 million acres, 4% of the State's land. This included over 900,000 lb of herbicides applied to rights-of-way throughout the State by various utility companies. Fourteen of the 135 base compounds sold constituted 73.3% of the total poundage used. These included, in order of volume, methyl bromide, maleic hydrazide, atrazine, DDT, 2,4-D, chlordane, sulfur, copper sulfate, aldrin, sodium chlorate, TDE, carbaryl, malathion, and methoxychlor.

Introduction

In the United States, the manufacture of pesticides is now a billion dollar industry which began its expansion with the introduction of synthetic organic pesticides during the mid-1940's. Modern public health programs and farm production practices are dependent on pesticides. Countless lives have been saved in this country and abroad by the use of pesticides for control of vector-borne diseases. In the United States, the use of pesticides has been an important factor in improving the quality and yield of farm products and has contributed significantly to the annual increase in farm income.

At present, some 60,000 pesticide formulations, containing 1 or more of 900 active pesticide chemicals (1), are available for use as insecticides, herbicides, fungicides, rodenticides, and plant growth regulators. These are packaged in numerous forms and may be purchased at a variety of retail outlets.

The usage patterns of pesticides—past, present, and future—have the potential for profoundly influencing the environment and, consequently existing as well as future pesticide monitoring programs. The purpose of the survey reported in this paper was to outline the patterns of pesticide sales and usage in Kentucky during 1968. To compile the principal pesticide sales and usage data for Kentucky, the following sources were contacted: (1) in-State sales outlets for the pounds of technical materials sold in the State and (2) aerial applicators, utility companies, and State agencies for types and amounts of pesticides used, number of acres treated, and sources of pesticides purchased.

Background Information

Although Kentucky is developing industrially, it is still a rural State. In 1960, Kentucky had a population of 3,038,156, of whom 55% resided in rural areas (2). The boundaries of the State encompass an area of 40,395 square miles or 25,852,800 acres. Nationally, Kentucky ranked 22d and 37th in population and land area, respectively. In 1964, acreage classified as farmland totalled 16,265,180 acres, of which 9,364,980 acres were considered cropland (3); there were approximately 133,000 farms averaging 123 acres in size.

Kentucky's economy depends heavily on agriculture. Economically, tobacco is Kentucky's most important

¹ From the Pesticides Program, Division of Environmental Services, State Department of Health, Frankfort, Ky. 40601.



FIGURE 1.—Agricultural activity by county, Kentucky—1964

farm commodity, providing one-third of the agricultural income, followed by the sale of livestock and the dairy industry. Most of this farm income is derived from central, northern, and western areas. The Eastern Kentucky Coal Field area includes 38 counties, with 16 counties comprising the active coal mining area (Fig. 1) (Div. of Strip Mining and Reclamation, Kentucky State Dep. of Natural Resources, Frankfort 1969, *personal communication*). Due to its topography, this entire area, with the exception of two counties, is a nonproductive agricultural region, with subsistence farming predominating.

In Kentucky, since 1961, approximately 3.7 million acres of crops have been harvested each year; principal crops have included corn, hay, soybeans, small grains, and tobacco (Table 1) (3). Since 1960, the total crop acreage has not fluctuated more than 1%; there have been, however, changes in the acreages planted to specific crops.

During an 8-year period from 1960-1968, the overall agricultural gross product increased from 560 million dollars to well over 910 million dollars (U.S. Dep. of Agric. Statistical Reporting Service, Louisville Branch Off., 1969, *personal communication*), an amount approaching one-third of the State's total gross product. This can be attributed, in part, to a greater shift towards beef cattle, swine, and grain production, particularly

TABLE 1.—Crops harvested in Kentucky, 1967

CROPS	APPROXIMATE ACREAGE HARVESTED
Corn	1,218,000
Popcorn	19,000
Hay	
Clover and Timothy	616,000
Lespedeza	415,000
Alfalfa	383,000
Other	275,000
Soybeans	310,000
Small grains (wheat, barley, oats, and rye)	247,000
Tobacco	177,000
Fruits and Vegetables	11,000
Miscellaneous	30,000
Total	3,701,000

west of a line extending north and south from Jefferson County to Monroe County and west to the Mississippi River. This area encompasses some 45 to 50 counties. Secondly, Kentucky is a major producer of milk and dairy products, and this industry is increasing. In 1968, nationally, Kentucky ranked 13th in milk production and 2d and 7th in production of evaporated milk and cheese, respectively. In addition, pesticide usage is responsible for a significant portion of this increase.

PESTICIDE LEGISLATION

In 1968, Kentucky was one of 47 States that had a pesticide registration law; currently all 50 States have registration laws. Kentucky does not, however, have a law that specifies the reporting of pesticide sales or usage. The State laws and regulations that apply to pesticides are presented in Table 2. In 1968, results of a questionnaire survey (conducted by mail by the Kentucky State Department of Health) showed that the 40 largest cities in Kentucky (4,000 population and above, based on 1960 census figures) had no laws, codes, or ordinances applicable to pesticides on the local level.

METHODS OF APPLICATION

In Kentucky, ground application is the most common method of applying pesticides. Several different types of equipment are used, the most common being the self-propelled "highboy" (a tri-wheeled vehicle with 5-6 ft clearance, the principal application equipment used in the tobacco-growing areas) and the tractor-drawn trailer rig or tractor-mounted spray boom. An estimated 1,500 highboys were in use in 1968 (Kentucky Distributors of Highboys, *personal communication*). In the principal fruit- and vegetable-growing areas, powered speed and row-crop sprayers are the most widely used methods of application. Dust application is diminishing and constitutes only a small portion of the total pesticide usage for all commercial agricultural crops.

Aerial application of pesticides is not a thriving enterprise in Kentucky as it is in the Great Plains States because of the small size and inaccessibility of fields to be treated; this is evidenced by the small number of operators licensed in 1968, 8 compared with 12 in 1960 (Kentucky Dep. of Aeronautics, *personal communication*). In 1968, application by two operators was confined to weed and brush control on rights-of-way of various utility companies, principally in 12 southeastern Kentucky counties (Fig. 2). The total acreage in Kentucky treated for all purposes by aerial application was approximately 14,000 acres.

Pesticide Use Patterns

AGRICULTURAL USAGE

In Kentucky, pesticide usage generally begins in March with application to peaches, apples, and alfalfa and continues until late August or early September, depending on the growing season within the State. The peak periods are usually in April, May, and June (Table 3), with an estimated 85%-90% of the pesticide applications occurring from April through August.

Data abstracted from 1964 U.S. Census of Agriculture (4) indicated that 1.21% of the farmland in Kentucky or 196,808 acres were treated for insect and disease control, and 2.69% or 437,533 acres were treated for

TABLE 2.—State laws and regulations applicable to pesticides, Kentucky—1968

STATE LAWS AND REGULATIONS	AGENCY RESPONSIBLE FOR ADMINISTRATION AND ENFORCEMENT	EXPLANATION OF LAW OR REGULATION
REGISTRATION AND LABELING LAWS		
Economic Poison Law—1956 Kentucky Revised Statutes Chapters 217.540 to 217.640	Division of Regulatory Services University of Kentucky	Requires the registration of all substances or mixtures of substances intended for preventing, destroying, repelling, or mitigating any insects, rodents, fungi, bacteria, weeds, or other forms of plant or animal life or virus (except viruses on or in living man or other animals) which are declared to be a pest.
Food, Drug and Cosmetic Act—Revised 1960 Kentucky Revised Statutes Chapters 217.005 to 217.215, 217.992	Division of Environmental Services State Department of Health	A section of this act requires that pesticide residues on or in raw agricultural products must comply with tolerances established by the U.S. Food and Drug Administration provided a tolerance has been established.
USE AND APPLICATION LAW		
Termite and Pest Control Industry Law—1954, Revised 1960 as the Kentucky Structural Pest Control Act Kentucky Revised Statutes Chapters 249.250 to 249.340, 249.990	Division of Noxious Weed and Pest Control State Department of Agriculture	Basically, requires the licensing of all pest control operators by the State Department of Agriculture. Applicants must pass an examination to secure a license.
REGULATIONS		
Aerial Applicators Regulation—1952 KAV-5-2 to KAV-5-10 Kentucky Revised Statutes Chapter 183	State Department of Aeronautics	Requires the licensing of all aircraft engaged in commercial aerial application.
Occupational Health Regulations—1963, Revised 1966 Kentucky Revised Statutes Sections 211.080, 211.180	Division of Occupational Health State Department of Health	Requires compliance with threshold limit values established for pesticides in the atmosphere in pesticide and chemical manufacturing and formulating plants.

NOTE: In 1972, Kentucky enacted a pesticide use and application act—"Kentucky Pesticide Use and Application Act of 1972" (Ky. Rev. St. 1972 s217B.010 to s217B.099).



□ Commercially treated by licensed aerial applicators (10,000+ acres)
 ▨ Urban company rights-of-way treated for spruce growth by the Kentucky Department of Aeronautics (14 counties)
 □ 26 western Kentucky counties in which the State Department of Aeronautics maintained mosquito control programs are not included

FIGURE 2.—Aerial application of pesticides by county, Kentucky—1968

TABLE 3.—General agricultural and nonagricultural pesticide usage by months, Kentucky

PESTICIDE USAGE	APPLICATION PERIOD											
	JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
AGRICULTURAL CROPS AND FIELDS												
Fruits (excluding dormant)			X	X	X	X	X	X	X			
Vegetables (excluding greenhouse-grown)					X	X	X	X	X			
Corn												
Soil				X	X							
Crop						X	X	X				
Alfalfa			X	X	X	X						
Tobacco												
Soil (excluding tobacco beds)				X	X							
Crop					X	X	X	X				
Small grains				X	X							
Soybeans				X	X	X						
Pasture (Thistle control)			X	X	X							
NONAGRICULTURAL USAGE												
Mosquitoes				X	X	X	X	X	X			
Rights-of-way					X	X	X	X				
Johnson grass				X	X	X						

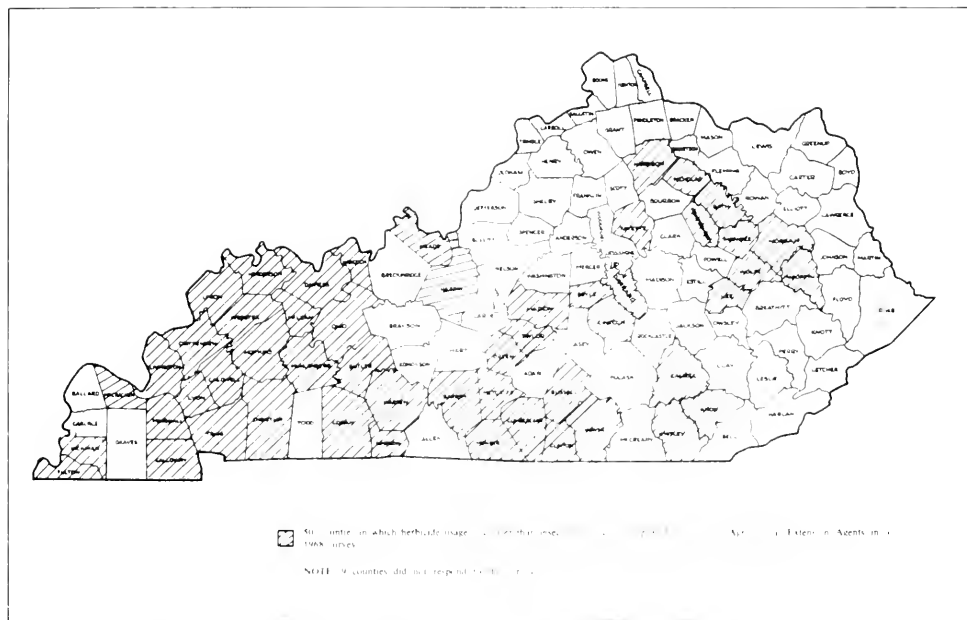


FIGURE 3.—Counties in which herbicide usage is greater than insecticide usage, Kentucky—1968

weed control. Nationally, Kentucky ranked 40th and 35th, respectively, in each category of treatment. By comparison, although the total acreage is small, New Jersey ranked first in the Nation with 13.9% of its farmlands being treated for insect and disease control, and Delaware ranked first in weed control, treating 21.42% of its farmland.

Responses from 111 of Kentucky's 120 County Agents to a questionnaire survey indicated that an estimated 89% of the farmers in Kentucky used some type of pesticide material in 1968. Fifty agents estimated that farmers in their counties used more herbicides than insecticides; these were primarily in the western and central areas of the State (Fig. 3). A decrease in pesticide usage was not reported by any responding agent.

TYPES AND POUNDAGE OF PESTICIDES SOLD BY IN-STATE OUTLETS

In 1968, approximately 3,800 pesticide formulations, involving an estimated 150 base chemicals, were registered for sale in Kentucky (Economic Poison Registration Section, Div. of Regulatory Services, Univ. of Kentucky, Lexington, Ky., *personal communication*). Registrations represented some 480 manufacturers and formulators. Some firms register all their products, although they may not necessarily be sold in the State (there is no additional charge after the first 10 registrations). Registration of pesticides incorporated with

commercial fertilizers for agricultural use and special lawn preparations totaled approximately 200.

Data from in-State sales outlets (distribution outlets, manufacturers or their representatives, dealers buying direct, etc.) reflected the sale of 2,850,734 lb of pesticides in Kentucky during 1968 (Table 4). Some 135 base pesticide chemicals were named; in all probability, additional base chemicals were sold but were not reported by these sources because they were overlooked or the poundage was considered unworthy of mention. These sources estimated that the bulk of the poundage was used to treat farmland, although about 5% of this total may have been formulated and packaged in small containers for the home and garden use market. The total poundage of pesticides sold by in-State outlets is listed in Table 4 as herbicides (growth regulators, soil sterilants, defoliants, and fumigants are grouped with herbicides), insecticides, fungicides, and rodenticides, in order of pounds sold.

It was reported that herbicide usage is increasing more rapidly than any of the other groups of pesticides. Three of the 62 herbicides—methyl bromide, maleic hydrazide (MH-30), and atrazine—constituted 67.5% of the pounds of herbicides sold. The first two were used primarily for tobacco, although an undetermined quantity of methyl bromide was used to fumigate hay and

TABLE 4.—Pesticides sold by in-State outlets, Kentucky—1968 (agricultural usage)

COMPOUNDS	POUNDS OF TECHNICAL MATERIAL SOLD
HERBICIDES—56.1% OF SALES	
Methyl bromide	431,788
MH-30 (maleic hydrazide)	352,956
Atrazine	200,679
2,4-D (all forms)	142,248
Sodium chlorate	99,047
Diphenamid (Enide®)	49,359
2,4,5-T (all forms)	38,356
Dalapon	30,079
NPA (Alanap®)	22,794
Trifluralin (Treflan®)	22,001
EPTC (Eptam®)	19,373
Amiben	18,224
Calcium hydrogen methanearsonate	16,670
Vernolate (Vernam)	15,845
Sodium arsenite	14,580
Linuron (Lorox®)	14,376
DCPA (Dacthal®)	13,513
Simazine	11,498
CIPC (Chloro-IPC)	10,420
Solan	9,090
DSMA (disodium methanearsonate)	8,294
Paraquat	7,184
Sutan	6,387
Picloram (Tordon®)	4,030
DNOC, sodium salt	3,532
Chloroxuron (Tenoran®)	3,369
Planavin	2,500
Dichlobenil (Casoron®)	2,454
Vorlex®	2,440
Isocil (Hyvar®)	2,400
Metham (Vapam®)	2,118
Benfen (Balan®)	1,614
Dicamba (Banvel-D®)	1,600
Silvex	1,240
Diuron (Karmex®)	1,200
Monuron (Telvar®)	1,200
Amitrole	1,006
CDAА (Randox®)	890
Endothall	600
Misc. herbicides (23)	13,456
Total	1,600,410
INSECTICIDES—31.4% OF SALES	
Chlorinated hydrocarbons (21.8%)	
DDT	151,015
Chlordane	127,778
Aldrin	101,079
TDE (Rhothane®)	94,449
Methoxychlor	46,967
Toxaphene	29,881
Dieldrin	26,979
Endosulfan (Thiodan®)	21,393
Dicofol (Keithane®)	8,898
Lindane	4,025
BHC	3,456
Heptachlor	2,347
Tetradifon (Tedio)	1,407
Endrin	500
Subtotal	620,174

TABLE 4.—Pesticides sold by in-State outlets, Kentucky—1968 (agricultural usage)—Continued

COMPOUNDS	POUNDS OF TECHNICAL MATERIAL SOLD
INSECTICIDES—31.4% OF SALES—Continued	
Organophosphates (5.3%)	
Malathion	51,467
Diazinon	20,728
Parathion	19,799
Disulfoton (Di-Syston®)	18,858
Naled (Dibrom®)	14,800
Azinphosmethyl (Guthion®)	11,899
Demeton (Systox®)	6,000
Ethion	1,975
Ciodrin®	1,347
Ronnel (Korlan®)	984
Dimethoate (Cygon®)	871
Mevinphos (Phosdrin®)	720
DDVP (Dichlorvos)	536
Misc. organophosphates (5)	394
Subtotal	150,378
Carbamates (2.8%)	
Carbaryl (Sevin®)	80,704
Subtotal	80,704
Miscellaneous (1.5%)	
Lead arsenate	31,467
Pyrethrins	3,076
Piperonyl butoxide	3,025
Rotenone	2,254
Other misc. (9)	1,952
Subtotal	41,774
Total	893,030
FUNGICIDES—12.0% OF SALES	
Sulfur	126,180
Copper sulfate	107,772
Captan	38,590
Zineb	20,620
Maneb	20,527
Ferbam	8,010
Lime sulfur	6,062
Phalant®	5,905
Cyprax®	3,210
Nabam (Dithane®)	1,168
Polyram® (Metiram)	1,168
Botran®	1,200
Thiram	760
Misc. fungicides (4)	1,523
Total	342,695
RODENTICIDES—0.5% OF SALES	
Warfarin	9,482
Arsenic trioxide	2,916
Zinc phosphide	1,270
Prolan®	699
Misc. rodenticides (4)	232
Total	14,599
GRAND TOTAL	2,850,734

NOTE: For simplicity in reporting, products containing a combination of chemicals, different brand or trade names with the same pesticide(s) were converted to basic chemicals. Salts, acids, etc., of the same basic component are reflected as one compound, such as the salts and acids of 2,4-D. Pesticides of less than 500 pounds are grouped under miscellaneous.

straw for the control of cereal leaf beetle transported into Kentucky from a contaminated out-of-State zone. The remaining 59 herbicides were used to control broad-leaf weeds and grasses.

The persistent chlorinated hydrocarbon insecticides were used more extensively in 1968 than any other insecticide group; their sales constituted 69.4% of the total pounds of insecticides sold. Approximately 4¼ lb were sold for every pound of organophosphate compounds.

The chlorinated hydrocarbon compounds were applied as soil insecticides for field crops; foliar sprays on selective food and feed crops and nonfood crops, such as tobacco and seed crops; for selective control of livestock insects; and for household and industrial pest control. As a soil insecticide for field crops, aldrin was the most widely used. An undetermined amount was incorporated into various commercial fertilizer preparations (168 different preparations containing aldrin were registered in 1968). If the amount used in this form were known, aldrin rather than DDT, would probably be the most widely applied persistent pesticide in 1968. Most of the DDT used in Kentucky in 1968 was applied to tobacco.

The 18 organophosphate compounds listed (Table 4) accounted for only 5.3% of the total pounds of pesticides, almost twice the poundage for the carbamates (exclusively carbaryl). Malathion was the most widely used followed by diazinon, parathion, and disulfoton. The organophosphates were applied as soil insecticides and as foliar sprays on grain, tobacco, fruit, vegetable, and forage crops. A few of these compounds, including co-ral, malathion, and ciodrin, were used to control livestock insects.

Carbaryl was used primarily as a foliar spray on grain, forage, fruit, and vegetable crops.

The most widely used fungicides were sulfur, captan, zineb, and maneb, constituting 60% of the total in this group. Warfarin was the most prevalent rodenticide.

It is estimated that the total poundage in Table 4 reflects 85% of the actual total pounds of pesticides sold in Kentucky in 1968. The ratio for category of pesticides would probably be the same if the total quantity sold by State outlets were known.

Fourteen of the 135 base compounds sold constituted 73.3% of the total pesticide pounds. These included, in order of volume, methyl bromide, MH-30, atrazine, DDT, 2,4-D, chlordane, sulfur, copper sulfate, aldrin, sodium chlorate, TDE, carbaryl, malathion, and methoxychlor.

From a questionnaire survey of aerial applicators licensed in Kentucky, it was determined that commercial

aerial application of pesticides to agricultural crops in 1968 involved less than 10,000 acres, primarily tobacco, small grains, and alfalfa in some 25 counties (Fig. 2). Tobacco accounted for 40% of the total. Approximately 20,500 lb of pesticides (technical material), half of which was MH-30, followed by DDT, toxaphene, and TDE (Rhothane®) were applied by winged aircraft. These four materials constituted 87% of the total. Most of the aerial application to agricultural crops involved application of MH-30 to small plots of less than 10 acres. Pesticides applied by aerial applicators for agricultural purposes were purchased from in-State outlets and are reflected in the data in Table 4.

State agencies, such as the institutional farm system, State universities, the Department of Fish and Wildlife, etc., purchased 38,513 lb of technical materials which included all four groups of pesticides, principally insecticides and herbicides (Division of Purchasing, Kentucky State Department of Finance, Frankfort, Ky., *personal communication*). This poundage was included in 1968 sales data; it was purchased on competitive bids from in-State vendors.

NONAGRICULTURAL USAGE, PURCHASED DIRECTLY FROM OUT-OF-STATE SOURCES

A total of 54 utility companies were surveyed by mail questionnaire concerning the pesticides applied to rights-of-way in Kentucky in 1968. The companies surveyed included railroads; gas transmission, communications, and power companies; and selected rural water districts. All are licensed by the Kentucky Public Service Commission. The rural water districts contacted did not apply any pesticides. The utility companies applied 920,820 lb of materials (exclusively herbicides for weed and brush control) to company-owned property and rights-of-way in numerous areas of the State (Table 5). The principal basic materials used included sodium chlorate-calcium chloride and diuron (Karmex®) mixture; 2,4-D, 2,4,5-T, and various combinations; isocil (Hyvar-X®); and fenuron (Dyhar®). These constituted 95% of the total materials used.

The State Department of Agriculture maintains a mosquito control program in approximately 28 western Kentucky counties and reported spraying 161,500 acres in 1968 (Division of Noxious Weed and Pest Control, Kentucky State Department of Agriculture, Frankfort, Ky., *personal communication*). Officials estimate that about one-half of the acreage was sprayed by winged aircraft either owned by or under contract to the Department. The four materials used in the program included DDT (which is no longer used), Lethane®, malathion, and naled. The 16,800 lb of technical material applied in 1968 were purchased in 1967 directly from the manufacturer and are not part of the sales data.

TABLE 5.—Pesticides purchased directly from out-of-State sources, 1968 (nonagricultural usage)

USERS	POUNDS OF TECHNICAL MATERIALS USED
Utility Companies	
Railroads (8,647 acres treated)	680,010
Power Companies (7,782 acres treated)	191,070
Gas Transmission Companies (1,511 acres treated)	47,400
Rural Water Districts	—
Communcation (75 acres treated)	2,340
Total (18,015 acres treated)	920,820
Department of Agriculture Mosquito Control Program (161,500 acres treated)	16,800
State Highway Department (39,400 acres treated)	122,955
TOTAL	1,060,575

The State Highway Department applied 122,955 lb of technical material (herbicides only) in 1968 on an estimated 39,400 acres of State rights-of-way (Division of Roadside Development, Kentucky State Department of Highways, Frankfort, Ky., *personal communication*). Application was primarily by truck, and principal materials (about 90% of the total), included 2,4-D, 2,4,5-T, MH-30, picloram, and dalapon. These materials were purchased from out-of-State firms.

Railroads used 73.8% of the total poundage, followed by power companies (20.7%), gas transmission companies (5.1%), and communications companies (0.4%). The utility firms reported treating approximately 18,000 acres of which 4,000 were treated by out-of-State based helicopters under contract: The remaining 14,000 acres were treated by surface spray rigs particularly designed for trucks and railcars. A small amount of herbicides in pellet and granular form was applied by cyclone distributors. Small quantities of liquid mix were also applied by knapsack and hand sprayers. Approximately 95% of the herbicides applied by contractors or company employees were purchased from out-of-State firms. These were located generally in Pennsylvania, Indiana, and Georgia. Hence, the volume used in utility operations is not reflected in the pounds of pesticides sold by in-State outlets.

Discussion

In Kentucky in 1968, pesticide usage for all purposes totaled more than 3.9 million pounds of technical material purchased from in-State and out-of-State outlets. This represents about 0.5% of the Nation's estimated total of 800 million pounds used domestically (5). The poundage for herbicides constituted approximately 67%

of the total. Approximately 1,060,575 lb (27% of the total poundage) were purchased from out-of-State sources. Herbicides applied to utility company and State Highway Department rights-of-way constituted all but 16,800 lb (used for mosquito control) of this total.

The total dollar value of pesticides purchased in 1968 for use in Kentucky was estimated at 18 to 20 million dollars. Exact figures cannot be determined because retailers are not required to report sales to any State regulatory agency. The estimate includes small package sales (dusts, liquids, etc.) and specialty pesticides such as aerosols, plunger dispenser devices, pastes, and tablets (all for home or garden use). It is hypothesized that the value would be significantly greater had there been a practical method of estimating dollar sales for special lawn fertilizer preparations containing various pesticides and for pesticides used on Federal Government installations such as military bases or depots, by municipalities, by pest control operators, or by others that may have been excluded in the study who purchase directly from out-of-State sources.

It is likely that millions of dollars are spent on specialty pesticides in various forms for use in and about the home. Most formulations for this market such as aerosols, dusts, and granules are purchased in a form suitable for application without the addition of diluents. Consequently, in terms of pounds of technical material, they constitute only a small segment of the total quantities of pesticides sold and used in 1968, but are much more important in terms of dollar sales.

Although pesticide usage is increasing annually (about 13%-15%), Kentucky ranked very low nationally in 1964 in the percentage of farmland treated, with less than 4% of Kentucky's farmland receiving some type of pesticide treatment (634,341 acres). This may be attributable, in part, to the types of crops grown in Kentucky. In the middle southern and southeastern States few pesticides are applied to soybeans and corn, major crops in Kentucky (Table 1). In addition, tobacco, another major crop in the State, is normally grown on the same soil only once every 2 or 3 years; therefore, the total acreage that receives pesticides in tobacco production is greater than the acreage for a given year (6). Hypothetically, if the total poundage had been confined entirely to agricultural use and evenly distributed over all of the State's farmland (16,265,180 acres), it would total less than 0.25 lb (4 oz) of technical material per acre, or use confined to cropland (9,364,980 acres), would amount to less than 0.5 lb/acre. The bulk of pesticides for agricultural purposes are used on grain, tobacco, and hay crops principally grown in 29 central (north, south) and 16 western Kentucky counties (Hardin, Hart, Barren, and those west) (Fig. 1).

From a monetary standpoint (retail), MH-30 was the most important pesticide in 1968, with sales of about 1.5 million dollars. Furthermore, it appeared that more dollars were expended for pesticide use on tobacco than any other crop. Perhaps the two most significant pesticides to be introduced in recent years in Kentucky are methyl bromide and MH-30. Both are used extensively in the production of tobacco and have been responsible for revolutionizing this particular industry. MH-30, a growth regulator, has all but eliminated the hand control of sucker growth. Methyl bromide, a fumigant, is very effective against soil insects in preparation of tobacco plant beds.

Conclusions

In Kentucky the continued increase in pesticide usage will be an important factor in the growth of the agricultural gross product. It is anticipated that pesticide usage, especially herbicides, will continue to accelerate.

Data on the principal uses, types, and volume of pesticides are important in this era of national concern for the environment, particularly with respect to the persistent pesticides. Such information will provide a basis for projecting future usage trends.

The work reported in this paper was supported under Contract No. PH 21-68-2039 by the Division of Pesticide Community Studies, Office of Pesticide Programs, U.S. Environmental Protection Agency, Chamblee, Ga.

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- (3) *Kentucky Department of Agriculture and Statistical Reporting Service. U.S. Department of Agriculture. 1967. Kentucky agriculture statistics, 1966 and 1967. Kentucky State Dep. Agric., Frankfort, Ky. 178 p.*
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- (5) *U.S. Department of Agriculture, Agricultural Stabilization and Conservation Service. 1969. The pesticide review 1969. Washington, D.C. p. 8, 14.*
- (6) *Sheets, T. J. 1966. The extent and seriousness of pesticide buildup in soils. p. 311-330 in N. C. Brady [ed.] Agriculture and the quality of our environment. The Plimpton Press, Norwood, Mass.*

APPENDIX

Chemical Names of Compounds Discussed in This Issue*

ALDRIN	Not less than 95% of 1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4-endo-exo-5,8-dimethanonaphthalene
ARSENIC	As ₂ O ₃
BHC	1,2,3,4,5,6-hexachlorocyclohexane, mixed isomers
CHLORDANE	1,2,4,5,6,7,8,8-octachloro-3a,4,7,7a-tetrahydro-4,7-methanoindane
DDE	1,1-dichloro-2,2-bis(<i>p</i> -chlorophenyl) ethylene
DDT (including its isomers and dehydrochlorination products)	1,1,1-trichloro-2,2-bis(<i>p</i> -chlorophenyl)ethane; technical DDT consists of a mixture of the <i>p,p'</i> -isomer and the <i>o,p'</i> -isomer (in a ratio of about 3 or 4 to 1)
DIELDRIN	Not less than 85% of 1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8a-octahydro-1,4-endo-exo-5,8-dimethano- \equiv naphthalene
ENDOSULFAN (THIODAN®)	6,7,8,9,10,10-hexachloro-1,5,5a,6,9,9a-hexahydro-6,9-methano-2,4,3-benzodioxatbipin 3-oxide
ENDRIN	1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4-endo-endo-5,8-dimethanonaphthalene
HEPTACHLOR	1,4,5,6,7,8,8-heptachloro-3a,4,7,7a-tetrahydro-4,7-methanoindene
HEPTACHLOR EPOXIDE	1,4,5,6,7,8,8-heptachloro-2,3-epoxy-3a,4,7,7a-tetrahydro-4,7-methanoindan
ISODRIN	1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4-endo, endo-5,8-dimethanonaphthalene
LINDANE	1,2,3,4,5,6-hexachlorocyclohexane, 99% or more gamma isomer
MIREX	dodecachlorooctahydro-1,3,4-metheno-1 <i>H</i> -cyclobuta[<i>cd</i>]pentalene
PARATHION	<i>o</i> , <i>o</i> -diethyl <i>o</i> - <i>p</i> -nitrophenyl phosphorothioate
POLYCHLORINATED BIPHENYLS (PCB's)	Mixtures of chlorinated biphenyl compounds having various percentages of chlorination
TDE (DDD) (including its isomers and dehydrochlorination products)	1,1-dichloro-2,2-bis(<i>p</i> -chlorophenyl)ethane; technical TDE contains some <i>o,p'</i> -isomer also
TOXAPHENE	chlorinated camphene containing 67% to 69% chlorine
TRIFLURALIN	α,α,α -trifluoro-2,6-dinitro- <i>N,N</i> -dipropyl- <i>p</i> -toluidine

*Chemical names of compounds discussed in the paper "Pesticide Sales and Usage in Kentucky—1968" are not included in this Appendix.

ERRATA

PESTICIDES MONITORING JOURNAL, Volume 6,
Number 1, p. 31. In the paper "Mercury and lead residues in starlings—1970," a number of values included in TABLE 2—*Mercury residues in starlings, 1970* were in error. The corrected table is shown below:

TABLE 2.—*Mercury residues in starlings, 1970*

SAMPLING SITE NUMBER	MERCURY RESIDUE ¹ (PPM)	SAMPLING SITE NUMBER	MERCURY RESIDUE ¹ (PPM)	SAMPLING SITE NUMBER	MERCURY RESIDUE ¹ (PPM)	SAMPLING SITE NUMBER	MERCURY RESIDUE ¹ (PPM)
1-A-1	0.05	4-C-1	<0.05	2-F-1	<0.05	3-H-1	<0.05
2	0.06	2	<0.05	2	<0.05	2	<0.05
3	1.50	Mean		3	<0.05	3	0.05
4	1.90	SE		4	0.18	4	0.10
Mean	0.878	1-D-1 ⁵	0.08	Mean	<0.083	Mean	<0.062
SE	0.417	2	0.05	SE		SE	
2-A-1	0.47	3	<0.05	3-F-1	0.13	4-H-1	<0.05
2	<0.05	4	<0.05	2	0.15	2	0.08
3	0.07	Mean	<0.058	3	0.13	3	<0.05
4	0.11	SE		4	0.09	4	<0.05
Mean	<0.175	2-D-1	<0.05	Mean	0.125	Mean	<0.058
SE		2	0.06	SE	0.010	SE	
3-A-1	<0.05	3	—	4-F-1	—	2-I-1 ³	0.08
2	0.07	4	<0.05	2	—	2	0.11
3	<0.05	Mean	<0.060	3	0.08	3	<0.05
Mean	<0.057	SE		4	<0.05	Mean	<0.080
SE		3-D-1	0.05	Mean	<0.065	SE	
1-B-1	0.05	2	<0.05	SE		3-I-1	<0.05
2	0.07	3	<0.05	1-G-1	<0.05	2	0.05
3	0.06	4	<0.05	2	0.05	3	<0.05
4	0.14	Mean	<0.050	3	<0.05	4	0.08
Mean	0.080	SE		4	0.05	Mean	<0.058
SE	0.023	4-D-1	0.08	Mean	<0.050	SE	
2-B-1	<0.05	2	—	SE		4-I-1	<0.05
2	0.10	3	<0.05	2-G-1	<0.05	2	<0.05
3	0.11	Mean	<0.065	2	<0.05	3	<0.05
4	0.06	SE		3	<0.05	Mean	
Mean	<0.080	1-E-1	<0.05	4	<0.05	SE	
SE		2	—	Mean	<0.050	5-I-1	<0.05
3-B-1	0.08	3	0.05	SE		2	<0.05
2	0.09	4	<0.05	3-G-1	0.05	Mean	
3	0.06	Mean	<0.050	2	<0.05	SE	
4	0.08	SE		3	<0.05	2-J-1	0.10
Mean	0.078	2-E-1	0.10	4	<0.05	2	0.10
SE	0.055	2	0.07	Mean	<0.050	3	0.10
4-B-1	0.09	3	0.05	SE		4	0.06
Mean		4	<0.05	4-G-1	<0.05	Mean	0.090
SE		Mean	<0.068	2	<0.05	SE	0.008
1-C-1 ²	0.06	SE		3	<0.05	3-J-1	<0.05
2	—	3-E-1	0.15	4	0.10	2	0.22
3	—	2	<0.05	Mean	<0.062	3	0.18
4 ⁵	<0.05	3	<0.05	SE		Mean	<0.150
Mean	<0.055	4	0.08	1-H-1	0.05	SE	
SE		Mean	<0.083	2	0.06	1-K-1	<0.05
2-C-1 ⁴	0.09	SE		Mean	0.055	2	0.05
2	—	1-F-1	0.11	SE	0.067	Mean	<0.050
3	<0.05	2	<0.05	2-H-1	0.05	SE	
4	0.06	3	<0.05	2	0.07	2-K-1	0.10
Mean	<0.067	4	<0.05	3	0.19	2	<0.05
SE		Mean	<0.065	4	<0.05	Mean	<0.075
3-C-1	0.08	SE		Mean	<0.090	SE	
2	—	2	<0.05	SE			
3 ³	<0.05	3	0.05				
4	<0.05	4	<0.05				
Mean	<0.060						
SE							

NOTE: — = no sample taken.

¹ Parts per million whole body, wet-weight basis.

² 2 birds.

³ 7 birds.

⁴ 8 birds.

⁵ 14 birds.

PESTICIDES MONITORING JOURNAL, Volume 6, Number 1, p. 73-75. In the paper "Decay of parathion and endosulfan residues on field-treated tobacco, South Carolina—1971," the values reported for parathion and endosulfan as pounds of active ingredient applied per acre were in error. Parathion was applied at a rate of .375 lb A.I. per acre rather than the 1.5 lb A.I. per acre as stated, and endosulfan was applied at a rate of .125 lb A.I. per acre rather than .5 lb A.I. per acre. These corrected values should be inserted in the Abstract, in the first paragraph under the Methods and Procedures section, and in Table 1 and Figs. 1 and 2.

PESTICIDES MONITORING JOURNAL, Volume 6, Number 3. In the paper "Residues of organochlorine

pesticides, polychlorinated biphenyls, and mercury and autopsy data for bald eagles, 1969, 1970," on page 138, the word *cholera* was misspelled as "chlorea" in the first and second sentences of the third paragraph.

PESTICIDES MONITORING JOURNAL, Volume 6, Number 3, p. 198 and 199. In the paper "Pesticide residue levels in soils, FY 1969—National Soils Monitoring Program," the maps in Figs. 1 and 3 were reversed. The map shown in Fig. 3 belongs with the caption "FIGURE 1.—Arsenic residues in cropland soil"; the map shown in Fig. 1 belongs with the caption "FIGURE 3.—Dieldrin residues in cropland soil."

Acknowledgment

The Editorial Advisory Board wishes to acknowledge with sincere appreciation the efforts of the following persons who assisted in reviewing papers submitted for publication in Volume 6, Nos. 1-4, of the *Pesticides Monitoring Journal*:

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Mildred L. Porter	Food and Drug Administration
Susan J. Young	Food and Drug Administration

SUBJECT AND AUTHOR INDEXES

Volume 6, June 1972-March 1973

Primary headings in the subject index consist of pesticide compounds, the media in which residues are monitored, and several concept headings, as follows:

Pesticide Compounds (listed alphabetically by common name or trade name where there is no common name)

Media and Concept Headings

Degradation

Experimental Design

Factors Influencing Residues

Food and Feed

Humans

Pesticide Sales and Usage

Plants (other than those used for food and feed)

Sediment

Soil

Water

Wildlife

Compound headings are also used as secondary headings under the primary media and concept headings and vice versa. When a particular paper discusses five or more organochlorines or three or more organophosphates or herbicides, the compounds are grouped by class under the media or concept headings; in the primary headings, however, all compounds are listed individually. The specific compounds or elements which

have been grouped in various combinations by class for certain papers are as follows:

Organochlorines

aldrin

BHC lindane

chlordane

DCBP

DDE

DDT

dicofol

dieldrin

endosulfan

endrin

heptachlor/heptachlor

epoxide

isodrin

methoxychlor

mirex

TDE

toxaphene

Organophosphates

carbophenothion

DEF

diazinon

ethion

malathion

methyl parathion

parathion

Herbicides

atrazine

2,4-D

DCPA

In the author index, the names of both senior and junior authors appear alphabetically. Full citation is given, however, only under the senior author, with a reference to the senior author appearing under junior authors.

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