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PESTICIDES IN PEOPLE

DDT and DDE in the Blood and Diet of Eskimo Children from Hooper Bay, Alaska[1]

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ABSTRACT

An analysis of the levels of DDT and DDE in the blood of some Alaskan Eskimo children and in the fat of some local marine mammals taken for food suggests that the children's pesticide burden is only modestly lower than that of other American children. Authors suggest that some other food source, perhaps packaged food, supplies a portion of the dietary chlorohydrocarbon pesticide.

Introduction

Marine mammals from virtually all waters contain chlorinated hydrocarbon contaminants in their tissues, reflecting the ubiquitous distribution of agricultural and industrial chemicals (*2,3,7,8,15*). Highest levels of DDT have been reported in migratory seals from Canadian waters of the North Atlantic, from the North Sea, and from the Baltic Sea (*8*). Except for nonmigratory harbor seals (*1*), the pesticide and its metabolites are generally less prominent in the tissues of mammals from Antarctic and Pacific waters.

Although perhaps less dependent on aquatic food sources now than in the past, native populations of western coastal Alaska still derive a substantial portion of their diet from the sea. Thus in the absence of any other substantial contact with DDT, levels of the pesticide and its metabolites in tissues of Alaskan Natives likely reflect the marine component of their diet.

Children and adolescents in these Eskimo populations have lived in the era of worldwide contamination by chlorohydrocarbons. Although a portion of any body burden of DDT-type materials may well have been received through the placenta or from breast milk (*4,5,10*), such sources would be difficult to evaluate in the presence of a contaminated marine diet for all but the very young. This paper reports results of a study undertaken to determine whether blood levels of DDT and DDE in Eskimo children of western Alaska are near those of children from other segments of the American population. Alaskan seals and waterfowl which are used as food were also examined for chlorohydrocarbons.

Methods

BLOOD SAMPLES

In May 1972, serum and heparinized whole blood were collected from 40 Eskimo children in Hooper Bay. Thirty-eight sera with 25 matching whole blood samples, and two single whole blood samples were available. They were selected from a listing of 204 specimens which had been collected for other purposes. This list representing nearly every school child in the village, kindergarten through ninth grade, was stratified by grade level and sex. A technique of substitution was used so that no specimens finally selected were from children in the same household. The donors' ages ranged from 6 to 17 years; there were 20 males and 20 females.

Aliquots of 2.0 ml whole blood or sera were extracted with 6.0 ml hexane on a slow rotary mixer. Nanograde (Mallinkrodt) hexane was used in the extraction and as a rinse for all glassware.

Pesticide residues in the extracts were quantitated by gas chromatography using electron-capture detectors. Two 6-ft-by-1/4-in. pyrex glass columns allowed separation of residues. One contained 1.5 percent OV-17/1.95 percent QF-1 on 100/120 mesh Chromosorb WHP, and the other

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contained 5 percent OV-210 on 80/100 mesh Supelcoport. Columns were maintained at 190° C, inlets at 215° C, and detectors at 210° C. Detectors were operated in the pulsed mode, with 10 percent methane in argon carrier gas at 80 ml/min.

Pesticide residue standards were more than 99 percent pure. Recoveries of residues undergoing the analytical regimen were greater than 90 percent and the reliable sensitivity of detection was 0.001 ppm for *p,p'*-DDE and 0.002 ppm for *p,p'*-DDT. Measured residue levels in blood and food source samples were not corrected for recovery values.

FOOD SOURCE SAMPLES

In May 1974, animals which residents had hunted and killed for food near Hooper Bay were tested. Single samples of seal meat, seal fat, and sea duck meat, all components of the native diet, were cleaned by a modification of the procedure of Stanley and Le Favoure (*12*). Following digestion in a mixture of perchloric-acetic acids and extraction with hexane, fats in the extracts were destroyed in large part by treatment with 0.5 ml concentrated H_2SO_4 in a graduated centrifuge tube. After centrifugation, the DDT-DDE residues were quantified by procedures described above. The limit of sensitivity was 0.001 ppm DDE and 0.002 ppm DDT. Neither the digestion mixture nor the H_2SO_4 contained extractable interfering material.

In April 1975, five additional samples of seal oil from hunted species were obtained from food caches in villages 150 miles south of Hooper Bay. Following three extractions with 20 volumes of acetonitrile the extracts were chromatographed. Interfering peaks appeared, so the acetonitrile extracts were mixed with six volumes of water and then extracted with hexane. Acceptable quantitation of chlorohydrocarbons could be made from the hexane solution with minimal interference after reacting with concentrated H_2SO_4. Chromatographic columns were similar to those used to quantitate residues extracted from blood. One column was prepared with 5 percent OV-210 on 100/120 mesh Gas-Chrom Q and the other with 1.5 percent OV-17/1.95 percent QF-1 on 80/100 mesh Gas-Chrom Q. The former column operated at 183° C with the carrier gas at 95 ml/min and the latter operated at 200° C under a gas flow of 80 ml/min. Reliable sensitivities were 0.001 ppm for DDE and 0.002 ppm for DDT.

Polychlorinated biphenyl compounds (PCB's) are reported to be as ubiquitous as DDT-type materials. For this reason [illegible] gas chromatographic peaks in the extract of seal fat were compared with peaks obtained in chromatographing the PCB compound, Aroclor 1242. No correlation could be made between unidentified peaks from the seal fat extract and six prominent peaks from a chromatogram of the PCB. Therefore, authors cannot report the presence of [illegible] contaminant in the fat sample at the sensitivity level of 0.2 ppm for nonmetabolized material by the methods used.

Aroclor 1254 chromatographed on 1.5 percent OV-17/1.95 percent QF-1 on Gas-Chrom Q presented one major peak, from a total of ten, which had a retention time of 6.4 minutes in contrast to 5.0 minutes for *p,p'*-DDE. Thus there is no discernible nonmetabolized PCB (Aroclor 1254) in lipids from the seals indigenous to the coast of western Alaska, determined at a sensitivity of 0.2 ppm.

Results

DDT-DDE

Table 1 summarizes results of analyses of DDE in serum. Pesticide levels in the whole blood samples were, in every case where matching serum levels were available for comparison, lower than serum levels by a factor which would relate to the dilution of serum by red blood cells. DDT levels in serum were beneath the limits of reliable detection (0.002 ppm) in 29 of the 38 samples and ranged from 0.002 to 0.003 ppm in the remaining nine sera.

TABLE 1. *Serum levels of* p,p'*-DDE in children of Hooper Bay, Alaska—1972*

Donor	Number	*p,p'*-DDE Levels, ppm	
		Mean	Range
Total	38	0.011	0.005-0.022
Male	19	0.011	0.005-0.022
Female	19	0.011	0.007-0.016
Ages 6-11 yr	18	0.011	0.005-0.018
Male	9	0.010	0.005-0.018
Female	9	0.012	0.009-0.016
Ages 12-17 yr	20	0.011	0.007-0.022
Male	10	0.012	0.008-0.022
Female	10	0.010	0.007-0.014

DDT-DDE IN FOOD-SOURCE SAMPLES

Pesticide levels in the single samples of seal fat, seal meat, and sea duck meat ranged from undetectable levels in seal meat to 0.110 ppm DDE and 0.020 ppm DDT in seal fat (Table 2). The highest residue in samples of seal oil was 0.80±0.01 ppm DDE (Table 3).

Discussion

A study reported in 1961 (*6*), preceding the present study by at least 11 years, indicated that DDT-related compounds were virtually absent in the natural dietary com-

TABLE 2. *Levels of* p,p'*-DDE and* p,p'*-DDT in three food source samples, Hooper Bay, Alaska—1974*

Sample	Pesticide Level, ppm				Percent Fat
	Weight Basis		Fat Basis		
	p,p' DDT	*p,p'*-DDT	*p,p'*-DDE	*p,p'*-DDT	
Seal fat	0.105	0.019	0.110	0.020	95.5
Seal meat	ND	ND	ND	ND	0.4
Sea duck meat	0.004	ND	0.17	ND	2.4

NOTE: ND = no residues could be detected within limits of reliable sensitivity

TABLE 3. *Levels of chlorohydrocarbon residues in seal oil, western Alaska—1975*

Collection Location	Residues, ppm		
	p,p'-DDE	*o,p'*-DDT	*p,p'*-DDT
Kuskokwim Bay near Kwigillingok			
(Spotted seal)	0.80±0.01	0.02±0.02	0.29±0.02
(Bearded seal, baby mukluk)	0.32±0.01	0.02±0.02	0.23±0.02
Kuskokwim Bay near Kongiganok	0.19±0.01	0.02±0.02	0.02±0.02
Kipnuk	0.36±0.01	0.02±0.02	0.13±0.02
Newtok	0.51±0.01	0.02±0.02	0.28±0.02

ponents of Alaska Natives. In addition the body fat levels of chlorohydrocarbons described for Natives were substantially lower than those of the general population. It might be assumed, on the basis of average values from a number of measurements, that *p,p'*-DDT levels in fat were some 450 times higher than in serum and that *p,p'*-DDE levels were some 400 times higher. Therefore, with reported mean levels of 0.8±0.10 ppm DDT and 2.0±0.41 ppm DDE in fat tissue, an approximate serum level of 0.002 ppm DDT and 0.005 ppm DDE might have been expected. Such estimated serum concentrations of the compounds for the study of 1961 (*6*) are similar to concentrations found now for the children and adolescents from Hooper Bay.

This comparison suggests that the body burden of these materials has remained relatively stable regardless of the route of exposure, and authors have no indication of recent local usage of any pesticide. Table 4 shows that chlorohydrocarbon levels in the serum of children of Hooper Bay are similar or only modestly lower than in children from most other areas (*9,11,13,14*). Children in South Carolina (*9*), especially black children, have demonstrated relatively high mean serum DDE and DDT levels, and reference adult populations had even higher levels.

The absence of chlorohydrocarbons in dietary samples reported in the previous Alaskan study (*6*) is in contrast to findings here, although differences in analytical techniques and corresponding sensitivities in measurements may well account for this.

TABLE 4. *Mean serum levels of chlorohydrocarbons in different populations of five States*

Reported Studies[1]	Age, yr	Chlorohydrocarbon, ppm	
		DDE	DDT
Hooper Bay, Alaska	6-17	0.011	<0.002
South Carolina (9)			
Whites	6-9	0.0246	0.0066
Blacks	6-9	0.0552	0.0185
Whites	Adults	0.0285	0.0112
Blacks	Adults	0.1222	0.0263
Florida (*11*)	—	0.0157	0.0042
Idaho (*14*)	3-10	0.0079	0.0021
	11-15	0.0130	0.0030
	16-20	0.0149	0.0030
Utah (*13*)	<21	0.0134	0.0036
	≥21	0.0209	0.0066

[1]Numbers in parentheses represent literature references cited in present study

The levels of residues in seal fat, in seal oil, and in sea duck meat found in the present study are notably lower than those in fat of seals taken off eastern Scotland (5.5 ppm DDE and 7.8 ppm DDT), northern and western Scotland (3.4 and 3.8 ppm), and Cabot Strait (5.9 and 5.5 ppm) and Magdalene Island (1.2 ppm and 0.36 ppm) in the Gulf of Saint Lawrence, Canada (*8*). Furthermore, the levels found in this study do not approach those reported in fat samples of nonmigratory harbor seals from some regions of the eastern North Pacific Ocean (*1*). Geometric means of ΣDDT and PCB's were 611 ppm in seals from Puget Sound and 11 ppm in seals from Pribilof Islands. Data on the levels of DDE in various tissues from immature males and nursing pups of the northern fur seal from the Pribilof Islands or the coast of Washington (*2,3*) indicate that fat levels of the compound are seven times as high as those in liver. The immature males would thus be expected to contain some 5 ppm DDE in their fat, a value in keeping with those found in seals from North Atlantic waters.

If generally representative, the relatively low levels of chlorohydrocarbons found in the fat and oil samples reported here suggest that lower dietary exposures, at least from an indigenous meat supply, should prevail for Eskimos of western Alaska. This assumption is not borne out by levels of DDE and DDT in serum from the children of Hooper Bay. Environmental levels of the chemically stable compound, DDT, in that locale should not be affected by a recent moratorium on its use, since its introduction into the region would have been largely windborne and in notably smaller quantity than if it had been used in local agriculture. It is possible that prepackaged food available to many Alaska Natives, especially in school lunch programs, is a source of the chlorohydrocarbons in the children's blood. Modest dietary exposure and body burdens of the chlorohydrocarbons appear to have been maintained during the past decade or longer.

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RESIDUES IN FISH, WILDLIFE, AND ESTUARIES

Mercury, Arsenic, Lead, Cadmium, and Selenium Residues in Fish, 1971-73—National Pesticide Monitoring Program

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ABSTRACT

As part of the National Pesticide Monitoring Program, the Fish and Wildlife Service, U.S. Department of Interior, analyzed selected fish samples from 100 monitoring stations for residues of mercury, arsenic, lead, cadmium, or selenium in 1971-73. At most stations, detectable residues of all metals were present in more than 95 percent of the composite samples. Fishes with mercury residues exceeding 0.5 mg/kg wet weight in the whole fish were mainly predators. Fishes with residues of arsenic, lead, cadmium, and selenium exceeding 0.5 mg/kg included predatory and nonpredatory species. The number of composite samples in which residues of these elements exceeded 0.5 mg/kg decreased from 1971 to 1973, whereas the percentage of samples with detectable residues increased slightly. Only selected samples were analyzed in 1973; therefore, these figures should be used only cautiously as trend data. Species of fish collected varied considerably between geographic regions but were similar from year to year within each region.

Introduction

The Fish and Wildlife Service (FWS) has contributed to the National Pesticide Monitoring Program by determining residues of various pollutants in fish. Authors have analyzed for organochlorines since 1967, mercury since 1969, and lead, cadmium, selenium, and the metaloid arsenic since 1971. Results of analyses were published for organochlorines through 1969 (*4,6*) and for mercury through 1970 (*5*). The present report presents results of analyses of heavy metals conducted on fishes collected 1971-73 at 100 stations throughout the United States (Fig. 1). On the basis of 1971 and 1972 results from 100 stations only, selected samples were analyzed for metals in 1973; caution should be exercised in interpreting these data. Except for Redhorse and fishes collected in Hawaii, common names of fishes used throughout this report are those designated by the American Fisheries Society (*1*). Redhorse is used to designate unidentified members of the genus *Moxostoma*. Fishes from the Hawaiian streams were Tilapia (*Tilapia mossambica*), Cuban limia (*Limia vittata*), and Chinese catfish (*Clarias fuscus*).

Methods

FISH COLLECTIONS

Fish were collected by FWS biologists, personnel of State fish and game agencies, and local commercial fishermen. Collection gear included a variety of nets and traps, hook and line, and electrofishing equipment. The use of chemical collecting agents was not permitted. As in previous reports (*1,7*) three composites of three species, each consisting of two to five adult fish, were to be collected at each of the stations from September to November. In the Hawaiian stations up to 26 fish were analyzed. Sample collections included a replicate for each species in 1971, but for only one of the three species from each station in 1972 and 1973. After length and weight of the fish had been determined, each composite was wrapped in foil, frozen, and shipped to the analytical laboratory for preparation and residue analyses. Localities of collection, species, size, and number of fish appear in Tables 1-3.

LABORATORY METHODS

1971—Two subsamples were taken from ground whole body composites: a 1-g sample for mercury and a 15-g sample for arsenic, cadmium, and lead. Mercury determinations followed the procedures of Okuno et al. (*13*).

Arsenic was measured by the Jarrell-Ash procedure (*7*) with the following modifications: a 1:1 (v/v) mixture of concentrated sulfuric acid (H_2SO_4) and nitric acid (HNO_3) was used to digest the 15-g samples, no perchloric acid was added during digestion, and the final volume was adjusted to 60 ml with distilled water. A subsample of the

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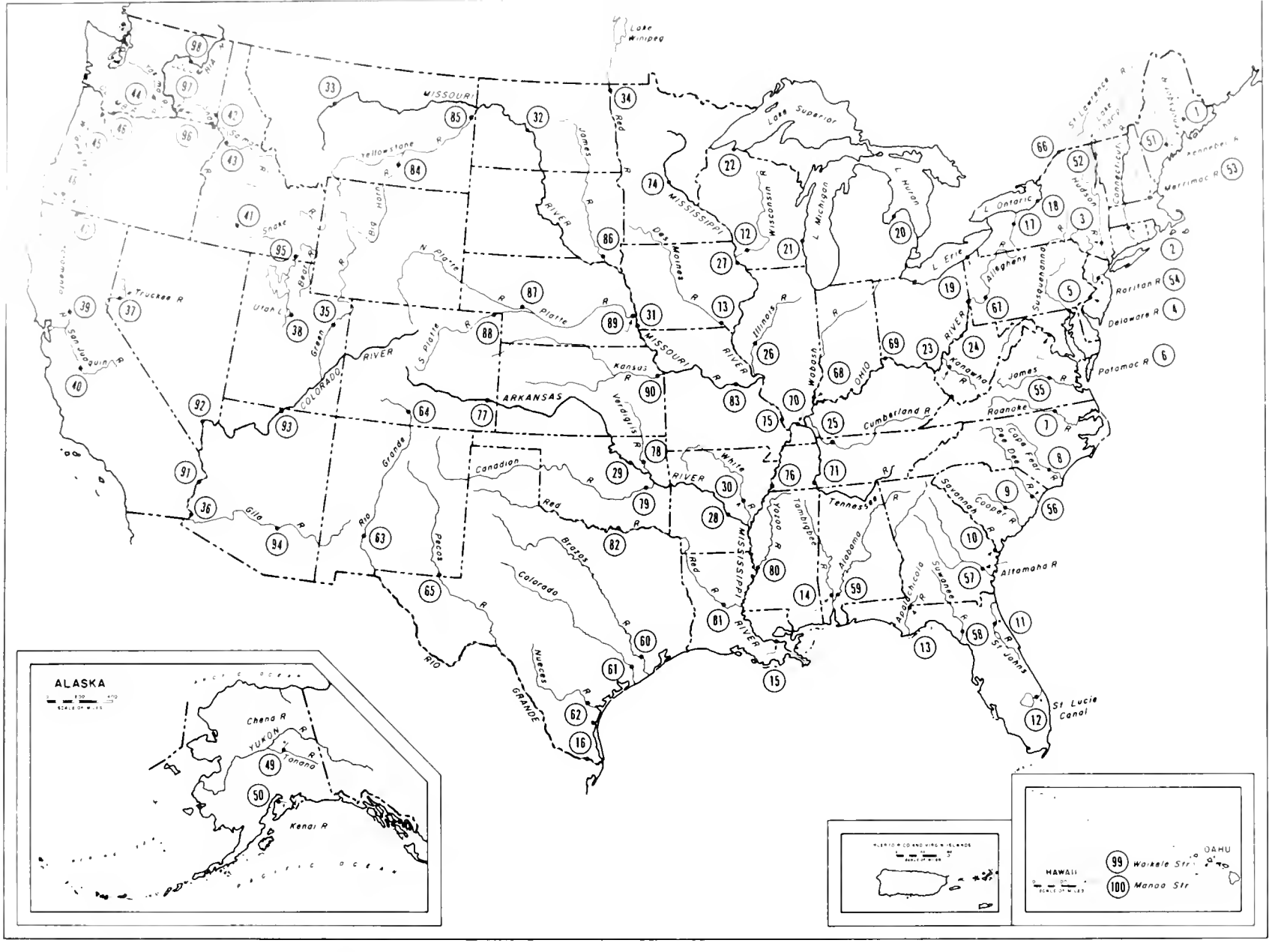

FIGURE 1. *Fish sampling stations, National Pesticide Monitoring Program, 1971-73*

digest representing 2.5 g of sample was analyzed by atomic absorption.

The digest from the arsenic procedure was also used for determining the presence of lead and cadmium. A subsample representing 10 g of sample was placed in a beaker and adjusted to 60 ml with distilled water, three drops of 1 percent (v/v) thymol blue were added, and the pH was adjusted to 5-6 with aqueous solutions of NaOH and H_2SO_4. This solution was transferred to a 250-ml separatory funnel, 10 ml of a 1 percent aqueous solution of a chelator (diethyldithiocarbamic acid, sodium salt) was added, the funnel was shaken for 2 minutes, and the extract was allowed to stand for 10 minutes. A 10-ml portion of water-saturated methyl isobutyl ketone (MIBK) was added, and the funnel was shaken for 2 minutes to extract the metals into the MIBK. The aqueous solution was drawn off, discarded, and the MIBK was collected in a 16-by-125-mm culture tube. This solution was aspirated into the flame of an atomic absorption spectrophotometer.

Recoveries of the metals were determined from the analyses of fish samples fortified at different levels with each metal. The overall mean from triplicate analyses for arsenic at three levels (0.1, 0.25, 0.5 ppm) was 82 percent with a standard deviation of 9.3 percent. The mean and standard deviations for lead (0.1, 1, 5 ppm) and cadmium (0.05, 0.25, 0.5 ppm) were 109 percent ± 21.0 percent and 99 percent ± 17.7 percent, respectively.

1972—Homogenized samples for mercury determinations were dried in a microwave oven for 15 minutes before combustion, but were otherwise analyzed as described for 1971. For lead and cadmium, 1-g subsamples of ground whole-body composites were dried in a beaker on a hotplate, charred with infrared lamps, and ashed in a muffle furnace at 500°C for 4 hours. After cooling, the residue was dissolved in 1 ml concentrated HNO_3, then heated until dry and white. To the cooled residue, 1 ml of concentrated HNO_3 was added, then diluted to 20 ml with water. The solution was warmed on a hotplate, cooled, and adjusted to a pH of 3 ± 0.2, then quantitatively transferred to a 125-ml separatory funnel with 2 ml water. A 1-ml portion of a 1 percent (w/v) aqueous solution of ammonium pyrrolidine-dithiodi-carbamate was added to the funnel and mixed. After 2 minutes, 10 ml of MIBK was added, the funnel was shaken for 1 minute, solvents were allowed to separate, and the lower aqueous layer was drawn off and discarded. A 10-ml solution of 5 percent HNO_3 was added to the funnel and shaken for 30 seconds. Solvents were allowed to separate, and the lower aqueous layer was collected. This sample solution was analyzed for both lead and cadmium, using a carbon rod atomizer on an absorption spectrophotometer.

Arsenic and selenium residues were determined in separate 1-g subsamples of the ground whole-body composites. Analyses for both followed the Jarrell-Ash (*7*) procedure with the following modifications: samples were placed in a Chromel wire sample holder and the holder was hung on the hook of a ground glass stopper placed in a 2-liter combustion flask containing 20 ml of 25 percent hydrochloric acid (HCl) solution for arsenic determinations, or 20 ml of a 1:1 (v/v) mixture of 50 percent HCl and 25 percent H_2SO_4 for selenium determinations. Flasks were flushed with oxygen, stoppered, and the contents were ignited with an infrared igniter. After combustion, flasks were allowed to stand 1 hour in order for the acid solution to entrain combustion products.

For the arsenic determination, the sample solution was transferred to a 100-ml pear-shaped flask and 20 ml of 40 percent (w/v) hydroxylamine hydrochloride was added; after the solution had been allowed to equilibrate for 15 minutes, 1 ml of a 6 percent aqueous solution of potassium iodide was added. After another 15 minutes, 2 ml of a 40 percent $SnCl_2$ solution was added and allowed to stand another 15 minutes; a Teflon-covered magnetic stirring bar was dropped into the flask and the flask was connected to an arsine generator. The solution was stirred briefly, 1 g of 20 mesh zinc was added and mixed for 2 minutes, and the generated arsine was swept with helium into the burner of an atomic absorption spectrophotometer.

Selenium sample solutions were decanted from the combustion flasks and rinsed with 20 ml acid (50 percent HCl and 25 percent H_2SO_4). A 10-ml portion of this solution (0.25 g sample equivalent) was placed in a pear-shaped flask with 30 ml of the HCl-H_2SO_4 acid mixture and 1 ml of stannous chloride ($SnCl_2$). A stirring bar was placed in this flask and the generator was connected as in the procedure for arsenic analysis. A magnetic stirrer was placed under the flask and 2 g of 20 mesh zinc was added; after 15 seconds of stirring, the hydrogen selenide generated was swept into the burner of the atomic absorption spectrophotometer.

Mercury recovery studies were made to compare microwave drying with the former P_2O_5 procedure for drying samples. Analysis of each of three samples using both drying procedures showed no significant differences ($P = 0.55$). Recoveries of lead and cadmium were determined by fortifying samples at varying levels ranging from 0.1 to 1 ppm for lead, and 0.01 to 0.1 ppm for cadmium. The overall mean recovery and standard deviation for lead was 90 percent ± 11.6 percent and 100 percent ± 13.6 percent for cadmium. For arsenic, the overall recovery from samples fortified with levels ranging from 0.05 to 0.3 ppm was 91 percent ± 18 percent. Selenium was determined by the procedure of Church and Robison (*3*). Recoveries of selenium by the procedure averaged 100 percent with a standard deviation of 15 percent.

1973—Mercury, lead, and cadmium were determined as in 1972. Arsenic was determined as in 1972, except that the

zinc used in generating arsine was 200-400 mesh in a slurry of distilled water (10 g zinc to 20 ml distilled water), and an electrodeless discharge lamp (EDL) and power supply were used in place of the hollow cathode lamp. Selenium was measured as described for 1972, but the light source was an EDL.

DETECTION LIMITS AND STANDARDS

The limits of detection of the metals in the composite fish samples were the same in all 3 years. Expressed as mg/kg wet weight, detection levels of each metal were mercury, 0.01; arsenic, 0.05; lead, 0.10; cadmium, 0.05; and selenium, 0.05. Analyses for metal residues were conducted on subsamples of composites prepared as described by Henderson et al. for laboratory C (6).

During all analyses, standard solutions of each metal were used for quantification and reagent blanks were used to detect possible contamination.

CROSS-CHECK ANALYSES

In cross-check analyses, total mercury was determined by the techniques described by the Joint Mercury Residues Panel (*10*) and modified as described by Henderson et al. (5). Arsenic and selenium residues were determined as described in the eleventh (*8*) and twelfth (9) editions, respectively, of the methods book of the Association of Official Analytical Chemists. Lead and cadmium residues were determined as follows: to a 12.5-g sample portion, 5 ml of 10 percent magnesium nitrate was added, the samples were then dried and charred on a hotplate and ashed overnight at 500 C. The samples were wetted with nitric acid, dried on a hotplate, and ashed again at 500°C for 20 minutes. After the sample had cooled, 2 ml concentrated HCL and 15 ml H_2O were added. Samples were boiled and stored in 50-ml volumetric flasks. Final determinations were made with a Perkin-Elmer model 303 spectrophotometer.

PRESENTATION OF RESULTS

The Denver Wildlife Research Center (DWRC) was contracted to conduct the initial analyses and the Wisconsin Alumni Research Foundation (WARF) was contracted to conduct cross-check analyses on selected samples. Also, DWRC conducted a methods check by repeating the analyses on several samples for all 3 years. Samples for cross-checking were selected according to results of initial analysis or the history of high residues at a particular station. A level of 0.5 mg/kg or greater was the criterion generally applied for selection. Mercury, arsenic, lead, and cadmium were analyzed in the samples from 1971, and selenium was added in 1972. In 1973, the rising costs of analytical work precluded the measurement of all metals. Therefore, only selected samples were analyzed for mercury, arsenic, lead, and cadmium, but all samples were analyzed for selenium residues to provide data for 2 consecutive years (1972 and 1973). The initial and the cross-check data for 1973 are presented in Tables 1-3. Results of the initial run and of the in-house rerun by DWRC varied by less than one order of magnitude. For this reason and for a degree of brevity, rerun results are not presented.

The National Academy of Sciences recommends that to protect fish and predatory aquatic organisms, total mercury burdens in these organisms should not exceed 0.5 mg/kg net weight (*12*). For the present purposes, authors have considered that any level exceeding 0.5 mg/kg in whole body components is a high level at which mercury, arsenic, lead, cadmium, or selenium would harm fish. To show annual trends for these elements in all fish, each residue value from the initial analyses was placed into at least one of the following categories: composites analyzed, composites with residues, composites with residues at or below detectable levels, composites with residues between detectable levels and 0.5 mg/kg, or composites with residues above 0.5 mg/kg (Table 4).

Results

Mercury residues were present in all samples of fish collected (Tables 1-3). Of the 100 stations sampled, 25 in 1971, 12 in 1972, and 11 in 1973 yielded composites in which mercury concentrations exceeded 0.5 mg/kg. Henderson et al. (5) reported composites exceeding 0.5 mg/kg from 9 stations in 1969 and 20 in 1970. Only stations 1-50 were sampled in 1969. These data indicate a general increase of mercury contamination from 1969 to 1971 and a decrease from 1971 to 1973.

Henderson et al. (5) also pointed out that certain predator fishes such as bass, perch, and squawfish had the highest mercury residues. Of 12 species in the present study in which mean residues exceeded 0.5 mg/kg during any of the 3 years, 7 were predators: chain pickerel, white perch, smallmouth bass, largemouth bass, whitebass, sauger, and Northern squawfish; and 5 could be considered nonpredator, i.e., nonpiscivorous: bowfin, carp, yellow and brown bullhead, and channel catfish.

Residues of mercury, arsenic, and selenium were generally present in more than 90 percent of samples in the present study (Table 4); lead was detected in 56 percent and cadmium in 76 percent of the 584 composites analyzed in 1971.

Arsenic residues (Tables 1-3) were generally lower than those of mercury; concentrations in mg/kg ranged up to 3.40 in 1971, 1.70 in 1972, and 1.24 in 1973. Residues in excess of 0.5 mg/kg were detected in composites from eight stations during the 3 years. Unlike mercury residues, arsenic residues above 0.5 mg/kg were not confined to the predatory fishes.

Lead residues above 0.5 mg/kg were present in fish from 16 stations in 1971, 34 stations in 1972, and 10 stations in

1973 (Tables 1-3). The highest concentrations in mg/kg were detected in fish from the Hawaiian streams: 1.4 in 1971, 5.2 in 1972, and 1.4 in 1973. Like arsenic residues, lead residues above 0.5 mg/kg were not confined to the predatory fishes.

Composites with cadmium residues above 0.5 mg/kg were few in 1971 (less than 1 percent) and 1972 (4 percent), and, of the 75 selected samples analyzed in 1973, none exceeded 0.5 mg/kg (Table 4). This suggests a decrease in the level of detectable residues of this metal, particularly since authors biased these results by selecting the samples to be analyzed during 1973 from stations at which residues in some samples exceed 0.5 mg/kg during 1972. Like lead and arsenic, cadmium residues above 0.5 mg/kg are not restricted to the predatory fishes. Cadmium is an extremely dangerous metal that accumulates readily in fish, has chronic effects, and is considered a threat to fishery resources (*12*).

The selenium analyses conducted only in 1972 and 1973 showed residues in essentially all samples (Table 4). Residues exceeding 0.5 mg/kg were distributed equally between predatory and nonpredatory fishes. Naturally occurring selenium has been detected in various environmental segments, but the biological significance of selenium residues is unknown (*12*).

Excessive levels of metals, greater than 0.5 mg/kg, were found in some fish from most river systems, but such high levels apparently occurred more frequently in certain stations than in others. Of the 11 stations with excessive mercury levels in 1973, 6 were from the Atlantic coastal streams (one each on the Stillwater and Kennebec Rivers, Maine; Merrimac River, Mass.; Pee Dee River, S. C.; Savannah and Altamaha Rivers, Ga.); 1 on the Gulf coastal streams (Tombigbee River, Ala.); 1 on the Mississippi River system (Little River, Minn.); 3 on the Columbia River system (1 on the Willamette and 2 on the Columbia River). Of those 11 stations, 6 exceeded 0.5 mg/kg during 1972 and 1973 and 3 (Kennebec River, Maine; Savannah River, Ga.; Tombigbee River, Ala.) had residues that exceeded 0.5 mg/kg during all the years reported. McKim, who was quoted in a paper by Olson et al. (*14*), exposed brook trout to methyl-mercuric chloride and determined residues in muscle tissue to be within 90-100 percent of those in the whole body. This suggests that not only should the fish and their predators be protected, but that fish from some rivers should not be consumed by humans.

Olson et al. (*14*) exposed fathead minnows (*Pimephales promelas*) to concentrations of methylmercury ranging from 0.018 to 0.247 mg/liter. After 48 weeks, analysis of whole body samples showed mean residues ranging from 1.47 to 10.9 mg/kg total mercury. For the most part, these residues exceed levels of the present study. However, fish from the control water of Olson's experiment, in which no methylmercury was added and residues averaged <0.01 ppm, had mean residues of 0.21 mg/kg (95 percent confidence interval, 0.17 to 0.25 mg/kg); water used in the experiment was unfiltered water from Lake Superior. In 1973, residues in samples from Lake Superior, Bayfield, Wis., ranged from 0.09 to 0.40 mg/kg. Olson et al. (*14*) point out the potential significance of low concentrations of mercury in natural waters. In a biochemical evaluation of methylmercuric chloride, Christensen (*2*) showed only a decrease in glutamic oxaloacetic transaminase (GOT) activity (L-aspartate: 2-oxoglutarate aminotranslerase, EC 2.6.1.1.) in brook trout embryos, and a decrease in weight and an increase in GOT activity in alevins. He further concluded that the concentrations used (1.03 μg/liter) in the study would be unsafe for the species if exposure were extended from egg through adult. These laboratory studies combined with residue data in the present study suggest that the health of many species collected is in danger.

Arsenic levels exceeding 0.5 mg/kg were found in fish from five stations (Tombigbee River, Ala.; Lake Michigan; Lake Superior; Red River, Okla.); those from the Great Lakes had high residues more frequently than the others. A study by the National Academy of Sciences and National Academy of Engineering showed residues up to 100 mg/kg in shellfish; sea water normally contains 2 to 3 μg /liter (*12*). Authors point out that acute effects of arsenic have been investigated, but little is known about sublethal chronic effects except that arsenic is readily accumulated by marine organisms (*12*).

Geographic distribution of high levels of lead appears to have decreased between 1972 and 1973. For instance, of the 14 stations located from the Stillwater River, Maine, south to the Pee Dee River (Northern Atlantic coastal streams), 9 exceeded 0.5 mg/kg in 1972 and 4 exceeded 1.0 mg/kg in 1973. Only 5 of the 14 stations had concentrations exceeding 0.5 mg/kg and none had composites with residues above 1.0 mg/kg. On the Mississippi River system in 1972, 13 of 35 stations had composites with residues above 0.5 mg/kg; 6 of those exceeded 1.0 mg/kg and 1 exceeded 5.0 mg/kg. In 1973, only 1 station, Des Moines River, Iowa, exceeded 0.5 mg/kg. This trend generally prevailed where excessive lead residues were found in 1972. There are, however, two stations that do not follow this encouraging trend. In 1973, fish from the Columbia River at Grand Coulee, Wash., and Manoa Stream, Hawaii, had composites with residues of 1.0 mg/kg and 1.4 mg/kg, respectively. The former represents an increase and the latter represents only a slight decrease. The source of these residues should be investigated.

As indicated earlier, results of the in-house methods check by DWRC corresponded closely with the study each year; data are not included in this report. The in-house methods check represents the quality control of the laboratory and shows that the data presented are accurate

and can be interpreted within the limits of the methods used. Validity of the present findings is further enhanced by the fact that the cross-check data are from another laboratory which used slightly different techniques, yet agree closely with data here. Furthermore, examination of the results between replicated samples at one station also indicates that the residues are representative of environmental levels.

NAS and NAE have stated, ". . . at present, it is not possible to predict accurately the amount of total metal in any environment that may be lethal, biologically active or contributory to toxicity . . ." (*12*). Authors submit that the data presented in this program are indicative of environmental levels of arsenic, lead, cadmium, and water with a wide variation in such characteristics as hardness and pH, and that criteria should be established for each metal and water. For an update on the effects of pollution on fish, authors recommend an excellent review by McKim et al. (*11*) which includes chemical and biological methodology used, the effects of water quality, pesticides, industrial pollutants, including metals, and domestic and radioactive wastes.

Residues in river fish analyzed in the present study are based on wet weight, whole fish. The concentrations of residues in the edible portions would probably be lower.

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TABLE 1. *Concentrations of mercury, arsenic, lead, and cadmium in whole fish, 1971—National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size		Residues, mg/kg wet weight			
			Length, in	Weight, lb	Mercury	Arsenic	Lead	Cadmium
Atlantic Coast Streams								
1. Stillwater River Old Town, Maine	White sucker	5	12.5	0.8	0.20	<0.05	0.26	<0.05
	White sucker (R)	5	12.5	0.8	0.28	<0.05	ND	<0.05
	Yellow perch	5	7.5	0.2	0.32	<0.05	ND	<0.05
	Yellow perch (R)	5	7.9	0.2	0.32	<0.05	0.26	0.05
	Chain pickerel	5	15.1	0.7	0.35	<0.05	0.16	<0.5
					(0.45)	(<0.05)	(<0.2)	(<0.05)
	Chain pickerel (R)	5	15.0	0.7	0.39	<0.05	ND	ND
51. Kennebec River Hinckley, Maine	White sucker	5	12.7	0.8	0.18	<0.05	ND	<0.05
	White sucker (R)	5	13.8	0.9	0.18	0.05	ND	ND
	Yellow perch	5	10.3	0.5	0.50	<0.05	ND	<0.05
					(0.83)	(<0.05)	(0.4)	(<0.05)
	Yellow perch (R)	5	9.1	0.3	0.50	<0.05	ND	ND
	Smallmouth bass	5	13.0	1.1	0.46	<0.05	0.21	ND
	Smallmouth bass (R)	2	14.4	1.4	1.20	<0.05	ND	<0.05
					(1.88)	(<0.05)	(0.5)	(<0.05)
52. Lake Champlain Burlington, Vt.	Pumpkinseed	5	7.3	0.4	0.12	0.10	ND	<0.05
					(0.23)	(0.08)	(0.4)	(<0.05)
	Pumpkinseed (R)	5	6.9	0.3	0.18	<0.05	0.78	ND
	Yellow perch	5	8.9	0.3	0.37	<0.05	0.57	<0.05
					(0.31)	(<0.05)	(0.7)	(0.06)
	Yellow perch (R)	5	10.0	0.4	0.31	<0.05	ND	ND
	Chain pickerel	3	16.2	1.0	0.39	0.05	0.13	<0.05
	Chain pickerel (R)	2	13.8	0.7	0.34	<0.05	0.11	<0.05
53. Merrimac River Lowell, Mass.	White sucker	5	12.0	0.7	0.15	<0.05	0.36	<0.05
	White sucker (R)	5	11.1	0.5	0.46	<0.05	0.36	<0.05
	Pumpkinseed	5	6.2	0.2	0.38	<0.05	0.24	ND
	Pumpkinseed (R)	5	4.4	0.1	0.42	<0.05	0.16	ND
	Yellow perch	5	10.7	0.6	0.14	<0.05	ND	ND
	Yellow perch (R)	5	10.7	0.6	0.42	<0.05	ND	ND
2. Connecticut River Windsor Locks, Conn.	White catfish	5	12.1	0.8	0.17	0.04	0.41	0.14
	White catfish (R)	5	12.9	1.1	0.21	0.06	0.51	0.17
	Yellow perch	5	8.8	0.3	0.25	<0.05	0.20	<0.05
	Yellow perch (R)	5	10.0	0.6	0.20	<0.05	0.12	<0.05
	White perch	4	8.6	0.4	0.38	<0.05	0.23	0.39
	White perch (R)	4	10.0	0.6	0.44	0.05	0.15	<0.05
3. Hudson River Poughkeepsie, N.Y.	Goldfish	5	9.4	0.8	0.06	0.11	1.30	0.12
	Goldfish (R)	5	9.9	0.8	0.19	0.08	0.52	0.15
	Pumpkinseed	5	6.1	0.1	0.13	0.07	ND	<0.05
	Pumpkinseed (R)	5	6.0	0.2	0.07	0.05	0.12	<0.05
	Largemouth bass	5	10.9	0.9	0.19	<0.05	ND	<0.05
	Largemouth bass (R)	5	10.8	0.9	0.10	0.05	ND	<0.05
54. Raritan River Highland Park, N.J.	Golden shiner	5	7.6	0.2	0.12	0.10	0.13	<0.05
	Golden shiner (R)	6	6.3	0.1	0.26	0.10	ND	<0.05
	White sucker	5	13.4	1.2	0.11	0.08	0.32	0.10
	White sucker (R)	5	12.3	1.0	0.18	<0.05	0.24	0.05
	White perch	5	9.7	0.7	0.34	0.08	0.16	<0.05
	White perch (R)	5	9.6	0.6	0.32	0.07	ND	<0.05
4. Delaware River Camden, N.J.	White sucker	5	14.6	1.3	0.06	0.06	0.51	0.06
	White sucker (R)	5	13.9	1.2	0.06	0.05	0.54	<0.05
	Brown bullhead	5	11.7	0.8	0.12	0.06	0.38	<0.05
	Brown bullhead (R)	5	10.9	0.7	0.04	0.06	0.36	<0.05
					(0.05)	(0.06)	(0.45)	(<0.05)
	White perch	5	9.5	0.5	0.25	0.06	0.78	<0.05
	White perch (R)	5	9.8	0.6	0.20	<0.05	0.47	<0.05
5. Susquehanna River Conowingo Dam, Md.	Carp	4	18.0	3.0	0.12	0.09	0.12	ND
	Carp (R)	4	18.9	3.3	0.05	0.20	ND	0.10
	Channel catfish	5	15.2	1.1	0.04	0.07	0.14	<0.05
	Channel catfish (R)	5	15.4	1.3	0.02	0.06	0.17	<0.05
	Yellow perch	5	8.6	0.3	0.08	0.06	ND	<0.05
	Yellow perch (R)	5	8.4	0.3	0.10	0.08	ND	<0.05
6. Potomac River Little Falls, Md.	Carp	5	17.2	2.6	0.26	0.13	0.11	0.08
	Carp (R)	5	15.7	2.0	0.19	0.11	0.21	0.11
	Redhorse[1]	5	13.5	1.0	0.18	<0.05	ND	0.06
	Redhorse (R)	5	14.7	1.2	0.15	0.06	ND	0.09
	Smallmouth bass	5	14.1	1.4	0.32	<0.05	ND	ND
	Smallmouth bass (R)	4	10.2	0.5	0.14	<0.05	ND	0.05
55. James River Richmond, Va.	Redhorse sucker	2	16.5	1.8	0.15	0.05	ND	ND
	Redhorse sucker (R)	2	16.0	1.6	0.10	<0.05	ND	0.23
	Channel catfish	2	21.5	3.6	0.23	<0.05	ND	0.05
	Channel catfish (R)	2	17.0	1.4	0.11	<0.05	ND	ND
	Largemouth bass	5	11.6	0.9	0.24	<0.05	ND	ND
	Largemouth bass (R)	5	8.6	0.3	0.23	<0.05	ND	ND

(*Continued next page*)

TABLE 1 (cont'd.) *Concentrations of mercury, arsenic, lead, and cadmium in whole fish, 1971 —National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size: Length, in	Average Size: Weight, lb	Residues, mg/kg wet weight: Mercury	Arsenic	Lead	Cadmium
Roanoke River Roanoke Rapids, N.C.	Redhorse	4	18.7	2.5	0.13	<0.05	ND	ND
	Redhorse (R)	4	19.8	2.9	0.12	<0.05	ND	0.17
	Brown bullhead	3	9.7	0.3	0.06	<0.05	ND	ND
	Largemouth bass	5	10.5	0.7	0.14	0.10	ND	0.06
	Largemouth bass (R)	5	9.3	0.5	0.11	0.06	ND	ND
Cape Fear River Elizabethtown, N.C.	Gizzard shad	5	8.7	0.2	0.10	0.09	0.20	<0.05
	Gizzard shad	5	10.0	0.4	0.10	0.05	0.35	<0.05
	Brown bullhead	5	9.5	0.3	0.14	<0.05	0.10	ND
	Brown bullhead (R)	5	9.6	0.3	0.22	<0.05	0.13	<0.05
	Largemouth bass	3	11.1	0.6	0.46	0.09	0.15	ND
Pee Dee River Dongola, S.C.	White catfish	5	14.5	1.6	0.33	<0.05	ND	0.06
	White catfish (R)	5	14.9	2.0	0.36	0.05	ND	<0.05
	Bluegill	4	5.5	0.2	0.04	0.09	0.21	<0.05
	Bluegill (R)	4	5.0	0.1	ND	ND	ND	ND
	Bowfin	4	18.5	2.5	1.03	0.20	ND	ND
	Bowfin (R)	4	17.6	2.1	0.43	0.22	ND	ND
Cooper River Summerton, S.C.	Carp	2	23.2	6.8	0.04	0.08	ND	<0.05
	Carp (R)	2	23.5	7.3	0.29	0.05	0.11	ND
	Bluegill	5	6.5	0.2	0.12	0.05	ND	ND
	Bluegill (R)	5	6.2	0.2	0.08	0.12	ND	ND
	Largemouth bass	5	11.0	0.6	0.05	0.05	0.23	<0.05
	Largemouth bass (R)	5	11.2	0.8	0.12	0.05	0.21	ND
Savannah River Savannah Ga.	Carp	2	20.3	4.1	ND (<0.05)	<0.05 (<0.05)	0.14 (0.3)	<0.05 (<0.05)
	Carp (R)	2	20.9	4.4	0.36 (0.41) <0.05 (<0.05)	ND (<0.2)	ND (<0.05)	
	Bluegill	2	11.2	1.0	0.44 (0.54)	<0.05 (0.12)	ND (0.8)	0.50 (<0.05)
	Bluegill (R)	3	8.8	0.6	0.10 (0.28)	0.06 (0.10)	0.21 (0.3)	<0.05 (<0.05)
	Largemouth bass	3	10.9	1.0	0.60 (0.72)	0.06 (<0.05)	ND (<0.2)	ND (<0.05)
	Largemouth bass (R)	4	10.9	0.7	0.34	<0.05	ND	ND
Altamaha River Doctortown, Ga.	Spotted sucker	4	17.2	2.2	0.23	0.06	0.16	0.12
	Spotted sucker (R)	4	18.2	2.7	0.22	0.06	ND	0.10
	Bluegill	5	7.6	0.4	0.12	0.09	ND	ND
	Bluegill (R)	5	7.4	0.4	0.12	0.09	ND	ND
	Largemouth bass	4	13.4	1.3	0.36	<0.05	ND	ND
	Largemouth bass (R)	4	12.2	1.1	0.43	<0.05	ND	<0.05
St. Johns River Welaka Fla.	Striped mullet	2	21.3	5.1	ND	0.34	0.15	<0.05
	Striped mullet (R)	3	19.1	3.1	ND	0.26	ND	0.05
	Channel catfish	5	10.7	0.4	0.02	<0.05	ND	ND
	Channel catfish	4	11.1	0.4	ND	<0.05	ND	ND
	Largemouth bass	3	17.1	2.9	0.64	0.06	ND	ND
	Largemouth bass (R)	3	15.4	2.3	0.10	0.05	ND	<0.05
St. Lucie Canal Indiantown Fla.	Channel catfish	3	16.6	1.8	0.09	<0.05	ND	ND
	Channel catfish (R)	2	21.8	3.8	0.13	<0.05	ND	0.06
	Bluegill	4	6.5	0.2	0.04	0.06	ND	ND
	Bluegill (R)	4	6.4	0.2	0.07	0.09	ND	ND
	Largemouth bass	4	11.5	1.2	0.26	0.08	ND	ND
	Largemouth bass (R)	4	12.4	1.2	0.32 0.35	0.10 0.09	0.13 ND	<0.05 0.05
Gulf Coast Streams								
[illegible]	Spotted sucker	4	12.5	1.0	0.15	<0.05	ND	<0.05
	Spotted sucker (R)	4	13.8	1.4	0.22	<0.05	ND	<0.05
	Redbreast sunfish	5	7.6	0.2	0.17	<0.05	ND	<0.05
	Redbreast sunfish (R)	5	7.9	0.3	0.12	<0.05	ND	<0.05
	Largemouth bass	4	13.4	1.6	0.42	0.22	ND	ND
	Largemouth bass (R)	4	12.1	1.4	0.76	0.05	ND	<0.05
	Spotted sucker	3	15.6	2.1	0.15	0.10	ND	<0.05
	Channel catfish	3	10.8	0.7	0.06	<0.05	ND	ND
[illegible]	Channel catfish (R)	3	9.7	0.5	0.06	0.05	ND	ND
	[illegible]mouth bass	4	10.0	0.8	0.09	0.07	ND	ND
	[illegible]mouth bass (R)	4	9.4	0.4	0.09	0.09	ND	ND
[illegible]	[illegible]	4	16.3	2.0	0.02	0.24	ND	<0.05
[illegible]	[illegible] (R)	4	15.0	1.7	0.19	0.16	0.12	<0.05
[illegible]		5	8.0	0.4	0.08	<0.05	0.14	<0.05
		5	7.6	0.4	0.16	0.05	ND	ND
		4	17.1	3.2	0.59	0.07	ND	0.10
	[illegible] (R)	5	16.6	2.6	0.09	0.07	ND	<0.05

(Continued next)

TABLE 1 (cont'd.). *Concentrations of mercury, arsenic, lead, and cadmium in whole fish, 1971 —National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size		Residues, mg/kg wet weight			
			Length, in	Weight, lb	Mercury	Arsenic	Lead	Cadmium
14. Tombigbee River	Carp	3	19.7	3.9	0.10	0.07	ND	<0.05
McIntosh,	Carp (R)	3	19.6	3.8	0.42	0.11	ND	ND
Ala.	Striped mullet	3	15.8	1.9	0.07	0.50	ND	ND
	Striped mullet (R)	3	15.6	1.8	0.30	0.70	0.13	<0.05
	Largemouth bass	4	15.7	2.0	1.10	0.13	ND	<0.05
	Largemouth bass (R)	5	14.7	1.6	1.10	0.09	ND	ND
15. Mississippi River	Carp	3	14.2	2.1	0.04	0.08	0.15	<0.05
Luling,	Carp (R)	3	14.4	1.9	0.02	0.07	ND	<0.05
La.	Bigmouth buffalo	3	18.6	4.8	0.11	0.07	0.22	0.06
	Bigmouth buffalo (R)	3	19.2	4.9	0.15	0.12	0.23	<0.05
	Channel catfish	3	14.2	1.3	0.07	0.05	0.30	<0.05
	Channel catfish (R)	3	15.7	1.8	0.12	0.05	0.37	<0.05
60. Brazos River	Smallmouth buffalo	2	21.4	7.0	0.17	0.07	0.13	0.05
Richmond,	Smallmouth buffalo (R)	3	19.1	4.0	0.09	0.06	0.19	<0.05
Tex.	Blue catfish	3	14.3	1.6	0.09	<0.05	0.26	<0.05
	Longnose gar	3	25.1	1.6	0.34	<0.05	ND	ND
	Spotted gar	3	22.4	2.1	0.44	0.05	0.10	<0.05
61. Colorado River	River carpsucker	5	11.8	1.1	0.16	0.06	0.21	<0.05
Wharton,	River carpsucker (R)	5	14.3	1.4	0.11	0.07	0.17	ND
Tex.	Channel catfish	3	15.5	1.0	0.05	<0.05	0.13	<0.05
	Flathead catfish	3	22.5	4.7	0.30	ND	0.10	<0.05
	Longnose gar	3	26.1	1.7	0.24	ND	0.14	<0.05
	Longnose gar (R)	3	24.2	1.6	0.31	<0.05	0.12	<0.05
62. Nueces River	Gizzard shad	6	9.7	0.4	0.06	0.10	0.10	<0.05
Mathis,	Gizzard shad (R)	6	7.3	0.2	0.02	0.16	0.15	<0.05
Tex.	Blue catfish	6	12.8	0.6	0.11	<0.05	0.13	<0.05
	Blue catfish (R)	6	11.3	0.4	0.10	<0.05	0.16	<0.05
	Black crappie	6	7.0	0.3	0.06	0.16	<0.10	<0.05
	White crappie	6	8.7	0.3	0.13	0.14	0.11	<0.05
16. Rio Grande	Gizzard shad	3	9.6	0.3	0.02	0.32	0.20	<0.05
Brownsville,					(0.06)	(0.43)	(0.5)	(0.06)
Tex.	Gizzard shad (R)	3	10.6	0.4	0.15	0.23	0.10	ND
	Channel catfish	3	13.4	1.1	0.10	<0.05	0.24	ND
	Channel catfish (R)	3	11.9	0.6	0.11	<0.05	0.26	ND
	Blue catfish	3	13.5	1.4	0.11	0.12	0.26	ND
	Blue catfish (R)	3	13.0	1.1	0.11	<0.05	0.44	ND
					(0.08)	(0.06)	(0.5)	(<0.05)
63. Rio Grande	Channel catfish	6	13.2	1.3	0.48	<0.05	ND	<0.05
Elephant Butte,	White bass	2	15.8	2.0	0.58	0.24	ND	<0.05
N. Mex.	White bass (R)	3	16.2	2.0	0.68	0.14	ND	ND
	Longear sunfish	5	4.9	0.1	0.03	0.22	ND	<0.05
	Largemouth bass	2	16.9	2.8	0.68	0.15	ND	<0.05
	Largemouth bass (R)	2	16.6	2.7	0.53	0.11	ND	ND
64. Rio Grande	Carp	3	10.1	0.5	0.02	<0.05	0.27	<0.05
Alamosa,	Carp (R)	3	10.4	0.5	0.30	0.06	0.14	ND
Colo.	White sucker	5	8.0	0.2	0.04	0.06	0.22	<0.05
	White sucker (R)	3	10.7	0.5	0.04	0.07	0.35	<0.05
	Brown trout	3	13.6	0.9	0.07	<0.05	0.20	ND
	Brown trout (R)	3	12.7	0.6	0.21	<0.05	0.19	<0.05
65. Pecos River	Gizzard shad	5	7.4	0.2	<0.01	0.08	0.11	<0.05
Red Bluff Lake,	Gizzard shad (R)	4	9.2	0.3	0.04	0.28	ND	ND
Tex.	Smallmouth buffalo	3	14.7	1.2	0.16	0.16	0.12	<0.05
	Smallmouth buffalo (R)	3	14.8	1.5	0.18	0.06	0.12	<0.05
	Channel catfish	3	15.8	1.3	0.06	<0.05	ND	<0.05
	Channel catfish (R)	3	17.3	1.8	0.17	<0.05	ND	<0.05
17. Genessee River	White sucker	5	14.4	1.1	0.14	0.06	<0.10	<0.05
Scottsville, N.Y.	White sucker (R)	4	14.1	1.0	0.16	0.08	0.10	<0.05
	Rock bass	5	8.0	0.4	0.32	<0.05	0.30	<0.05
	Rock bass (R)	5	7.5	0.4	0.28	0.05	<0.10	ND
	Walleye	5	19.3	3.0	0.64	0.08	0.10	<0.05
	Walleye (R)	5	13.4	0.9	0.28	0.11	0.12	ND
66. St. Lawrence River	White sucker	5	13.9	1.1	0.12	0.05	0.14	ND
Massena, N.Y.	White sucker (R)	5	14.1	1.2	0.11	0.05	0.21	ND
	Yellow perch	5	9.2	0.3	0.36	0.06	0.10	ND
	Yellow perch (R)	5	8.4	0.2	0.23	<0.05	0.19	ND
	Smallmouth bass	5	12.2	0.9	0.34	0.28	ND	ND
	Smallmouth bass (R)	5	12.1	0.9	0.38	0.34	ND	ND
18. Lake Ontario	Yellow perch	5	11.4	0.8	0.65	0.15	0.20	<0.05
Port Ontario, N.Y.					(0.58)	(0.18)	(0.4)	(<0.05)
N.Y.	Yellow perch (R)	5	11.3	0.8	0.50	0.10	0.16	ND
	White perch	5	10.0	0.6	0.36	0.14	0.85	<0.05
	White perch (R)	5	9.6	0.6	0.46	0.14	1.40	<0.05
					(0.56)	(0.31)	(0.4)	(<0.05)
	Rock bass	5	8.3	0.5	0.45	0.10	ND	ND
					(0.45)	(0.12)	(0.4)	(<0.05)
	Rock bass (R)	5	8.4	0.6	0.52	0.10	0.51	<0.05
					(0.48)	(0.14)	(0.4)	(<0.05)

(*Continued next page*)

TABLE 1 (cont'd.). *Concentrations of mercury, arsenic, lead, and cadmium in whole fish, 1971 —National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size		Residues, mg/kg wet weight			
			Length, in.	Weight, lb.	Mercury	Arsenic	Lead	Cadmium
19 Lake Erie	White sucker	5	14.6	1.3	0.13	0.11	ND	ND
Erie, Pa.	White sucker (R)	5	14.4	1.4	0.14	0.16	0.18	0.06
	Freshwater drum	5	13.3	1.1	0.29	0.11	0.20	<0.05
	Freshwater drum (R)	5	13.3	1.3	0.13	0.09	0.20	<0.05
	Yellow perch	5	9.5	0.5	0.09	<0.05	ND	<0.05
	Yellow perch (R)	5	9.0	0.4	0.13	<0.05	0.12	<0.05
20 Lake Huron	Carp	4	16.7	2.4	0.02	<0.05	ND	<0.05
Bay Port, Mich.	Carp (R)	4	17.7	2.7	0.04	0.08	0.15	<0.05
	Channel catfish	5	17.0	1.6	0.11	0.12	ND	<0.05
	Channel catfish (R)	5	17.3	1.7	0.12	0.16	0.11	<0.05
	Yellow perch	5	9.4	0.4	0.04	<0.05	0.11	ND
	Yellow perch (R)	5	9.1	0.3	0.02	<0.05	ND	ND
21 Lake Michigan	Bloater	3	9.5	0.5	0.07	2.80	0.54	<0.05
Sheboygan, Wis.	Bloater (R)	2	9.3	0.4	0.13	3.40	ND	<0.05
	Lake trout	5	26.0	6.4	0.52	1.00	ND	<0.05
	Lake trout (R)	5	25.1	6.6	0.49	1.30	0.10	0.05
	Yellow perch	5	9.8	0.3	0.16	0.07	0.15	ND
	Yellow perch (R)	3	9.6	0.3	0.15	0.07	0.13	ND
22 Lake Superior	Bloater	5	8.5	0.5	0.20	0.80	1.00	<0.05
Bayfield, Wis.	Bloater (R)	5	8.4	0.4	0.13	0.90	0.31	0.09
	Lake whitefish	5	19.7	2.4	0.01	0.60	0.13	<0.05
	Lake whitefish (R)	5	20.0	2.8	0.09	0.60	ND	0.20
	Lake trout	5	19.0	3.2	0.42	0.27	ND	<0.05
	Lake trout (R)	4	22.0	4.4	0.46	0.27	ND	<0.05
67 Allegheny River	Carp	4	16.4	1.9	0.04	<0.05	0.30	0.07
Natrona, Pa.	Carp (R)	4	18.8	3.8	0.12	0.06	0.13	0.07
	Yellow perch	4	9.6	0.5	0.27	<0.05	0.14	<0.05
					(0.10)	(<0.05)	(0.4)	(<0.05)
	Yellow perch (R)	5	7.9	0.2	0.08	ND	ND	<0.05
					(0.08)	(<0.05)	(0.7)	(<0.05)
	Walleye	4	12.4	0.7	0.19	<0.05	0.13	<0.05
	Walleye (R)	5	16.4	1.5	0.19	0.08	0.12	0.06
23 Kanawha River	Carp	5	11.3	0.9	0.01	<0.05	ND	ND
Winfield, W. Va.	Carp (R)	5	12.0	1.0	ND	<0.05	ND	ND
	Brown bullhead	5	10.9	0.8	0.08	<0.05	0.18	0.13
	Brown bullhead (R)	5	11.5	0.8	0.04	0.07	0.14	ND
	White crappie	5	9.2	0.5	0.09	<0.05	ND	<0.05
	White crappie (R)	5	9.2	0.05	0.14	<0.05	ND	ND
68 Wabash River	Carp	3	19.8	4.3	0.28	0.07	ND	0.12
New Harmony, Ind.	Carp (R)	3	19.2	3.5	0.25	0.07	ND	0.05
	Channel catfish	3	16.0	1.6	4.50	<0.05	0.16	<0.05
	Channel catfish (R)	3	17.7	1.9	0.29	<0.05	0.11	0.05
	White crappie	5	9.2	0.4	0.18	0.11	ND	<0.05
	White crappie (R)	5	9.5	0.4	0.16	0.08	ND	<0.05
24 Ohio River	Carp	2	19.1	3.9	0.14	0.52	0.34	0.08
Marietta, Ohio					(0.11)	(0.40)	(0.3)	(0.07)
	Carp (R)	2	14.0	1.4	0.15	0.28	0.40	0.09
	Channel catfish	4	14.8	1.1	0.19	<0.05	0.10	ND
	Channel catfish (R)	4	13.1	0.7	0.30	<0.05	0.28	<0.05
	Largemouth bass	4	12.6	1.5	0.25	0.11	0.12	<0.05
					(0.26)	(0.07)	(0.3)	(<0.05)
	Largemouth bass (R)	4	13.3	1.4	0.22	0.07	ND	ND
69 Ohio River	Carp	5	15.3	1.9	0.09	<0.05	0.28	0.08
Cincinnati, Ohio	Carp (R)	5	14.4	1.7	0.09	0.08	0.15	<0.05
	Channel catfish	4	14.5	1.3	0.17	ND	0.20	<0.05
	Channel catfish (R)	4	14.3	0.9	0.18	<0.05	0.16	<0.05
	Sauger	5	12.9	0.8	0.14	0.06	<0.10	<0.05
	Sauger (R)	5	12.7	0.7	0.13	0.07	<0.10	<0.05
[illegible]	Carp	4	18.4	3.6	0.23	0.25	ND	0.10
[illegible]	Carp (R)	4	17.3	2.8	0.12	0.25	0.13	0.12
	Channel catfish	5	14.1	0.9	0.18	0.22	0.29	<0.05
	Channel catfish (R)	5	14.6	1.0	0.14	0.17	0.14	<0.05
	White crappie	5	10.4	0.7	0.18	0.13	ND	0.30
	White crappie (R)	5	10.1	0.6	0.42	0.16	ND	<0.05
[illegible]	Carp	4	10.8	0.5	0.11	0.06	ND	<0.05
[illegible]	Carp (R)	4	9.5	0.4	0.05	<0.05	ND	<0.05
	Bluegill	4	5.5	0.1	0.09	ND	0.23	<0.05
	Bluegill (R)	4	6.0	0.1	0.04	<0.05	ND	<0.05
	Largemouth bass	2	9.6	0.6	0.10	0.10	ND	0.05

(*Continued*)

TABLE 1 (cont'd.). *Concentrations of mercury, arsenic, lead, and cadmium in whole fish, 1971 —National Pesticide Monitoring Program*

Station Number and Location	Species	No Fish	Average Size		Residues, mg/kg wet weight			
			Length, in.	Weight, lb	Mercury	Arsenic	Lead	Cadmium
71. Tennessee River	Carp	3	17.3	2.4	0.23	0.13	ND	<0.05
Savannah, Tenn.	Carp (R)	3	17.3	2.7	0.20	0.07	ND	<0.05
Tenn	Channel catfish	3	13.4	0.8	0.28	0.10	ND	<0.05
	Channel catfish (R)	4	12.5	0.7	0.34	0.08	0.12	<0.05
	Largemouth bass	3	12.9	1.2	0.24	0.21	ND	<0.05
	Largemouth bass (R)	3	12.9	1.1	0.14	0.18	ND	0.07
72. Wisconsin River	Carp	3	18.7	3.4	0.25	0.05	<0.10	<0.05
Woodman, Wis.	Carp (R)	4	20.5	4.0	0.19	0.06	ND	<0.05
	Channel catfish	4	16.3	1.4	0.20	ND	0.10	<0.05
	Channel catfish (R)	4	17.4	1.6	0.66	<0.05	<0.10	<0.05
					(0.19)	(<0.05)	(0.2)	(<0.05)
	Sauger	4	11.3	0.4	0.55	<0.05	<0.10	<0.05
	Sauger (R)	4	13.9	1.0	1.10	<0.05	ND	ND
					(0.77)	(<0.05)	(0.3)	(<0.05)
	Smallmouth bass	4	10.8	0.7	0.06	<0.05	ND	ND
					(0.46)	(<0.05)	(<0.2)	(<0.05)
	Smallmouth bass (R)	4	14.6	1.7	0.99	<0.05	<0.10	<0.05
					(0.96)	(<0.05)	(<0.2)	(<0.05)
73. Des Moines River	Carp	5	12.5	0.9	0.01	<0.05	0.83	<0.05
Keosauqua, Iowa					(<0.05)	(<0.05)	(0.2)	(<0.05)
	Carp (R)	5	13.0	1.2	0.05	ND	0.32	<0.05
	Channel catfish	5	12.6	0.7	0.06	ND	0.11	ND
	Channel catfish (R)	5	13.0	0.7	0.05	ND	0.17	<0.05
	Walleye	5	13.6	0.8	0.16	<0.05	ND	ND
	Sauger	3	13.9	0.9	0.08	ND	0.11	<0.05
26. Illinois River	Carp	5	15.1	1.6	0.04	<0.05	ND	0.05
Beardstown, Ill.	Carp (R)	5	13.9	1.4	0.08	0.08	0.18	<0.05
	Bigmouth buffalo	3	16.4	3.0	0.06	0.13	ND	<0.05
	Bigmouth buffalo (R)	3	18.7	4.2	0.05	0.13	0.12	<0.05
	White crappie	4	8.9	0.4	0.08	0.16	ND	0.07
	White crappie (R)	4	9.4	0.5	0.05	0.18	ND	0.07
74 Mississippi River	White sucker	4	18.8	2.9	0.96	<0.05	ND	<0.05
Little Falls, Minn.	White sucker (R)	4	18.6	2.8	0.43	<0.05	ND	0.05
	Black bullhead	4	7.1	0.3	0.22	<0.05	0.10	<0.05
	Black bullhead (R)	4	7.0	0.3	0.26	<0.05	ND	ND
	Northern pike	3	18.2	1.4	0.42	<0.05	0.10	<0.05
	Northern pike	3	13.7	0.4	0.18	<0.05	ND	ND
27. Mississippi River	Carp	5	17.9	3.5	0.15	<0.05	0.53	<0.05
Guttenburg, Iowa	Carp (R)	5	19.5	4.4	0.18	<0.05	0.27	<0.05
	Bluegill	5	6.3	0.2	0.12	<0.05	0.30	ND
					(0.12)	(0.11)	(0.7)	(<0.05)
	Bluegill (R)	5	6.7	0.3	0.04	<0.05	0.15	<0.05
	Largemouth bass	5	13.8	2.0	0.26	<0.05	0.14	ND
	Largemouth bass (R)	5	13.2	1.9	0.33	<0.05	0.21	<0.05
75. Mississippi River	Carp	5	18.5	3.0	0.06	0.05	ND	<0.05
Cape Girardeau, Mo.	Carp (R)	5	18.6	3.2	0.15	0.07	0.22	0.08
	Channel catfish	5	16.0	1.2	0.12	<0.05	<0.10	<0.05
	White crappie	3	10.4	0.6	0.12	0.10	<0.10	<0.05
	White crappie (R)	2	11.8	1.2	0.10	0.16	ND	ND
76. Mississippi River	Carp	2	21.3	5.2	0.06	0.26	0.17	0.07
Memphis, Tenn.	Carp (R)	2	20.8	5.0	0.06	0.12	0.12	0.06
	Carpsucker	2	18.5	4.0	0.10	0.11	ND	<0.05
	Carpsucker (R)	2	17.0	2.8	0.08	0.37	0.13	<0.05
28. Arkansas River	Carp	2	16.9	2.1	0.09	0.05	ND	0.05
Pine Bluff, Ark.	Carp (R)	2	17.7	2.4	0.08	<0.05	ND	0.07
	Smallmouth buffalo	2	17.4	3.3	0.03	0.08	ND	<0.05
	Smallmouth buffalo (R)	2	18.3	3.5	0.14	0.33	ND	0.06
	Flathead catfish	2	19.4	2.8	0.16	<0.05	ND	0.06
	Flathead catfish (R)	2	21.6	4.8	0.48	<0.05	ND	0.05
29. Arkansas River	Carp	5	12.8	0.8	0.12	<0.05	<0.10	<0.05
Keystone Reservoir, Okla	Carp (R)	5	14.6	1.4	0.08	0.13	0.13	<0.05
	Channel catfish	5	16.1	1.1	0.14	0.26	0.10	<0.05
	Channel catfish (R)	5	16.7	1.2	0.15	0.07	0.11	<0.05
	Bluegill	5	5.7	0.1	0.07	0.07	<0.10	ND
	Bluegill (R)	4	5.6	0.1	0.03	<0.05	<0.15	<0.05
77. Arkansas River	Carp	5	13.7	1.2	2.70	<0.05	0.13	<0.05
John Martin Reservoir,					(<0.05)	(<0.05)	(<0.2)	(<0.05)
Colo.	Carp (R)	5	13.5	1.2	0.04	<0.05	<0.10	ND
					(<0.05)	(<0.05)	(<0.2)	(<0.05)
	Channel catfish	4	15.0	1.3	0.05	<0.05	0.15	<0.05
	Channel catfish (R)	3	13.2	0.8	0.06	ND	0.26	<0.05
	Black bullhead	5	9.9	0.6	0.16	ND	0.16	<0.05
	Black bullhead (R)	3	9.2	0.4	0.11	ND	0.10	ND

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TABLE 1 (cont'd.). *Concentrations of mercury, arsenic, lead, and cadmium in whole fish, 1971 —National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size		Residues, mg/kg wet weight			
			Length, in	Weight, lb	Mercury	Arsenic	Lead	Cadmium
78 Verdigris River Oologah, Okla	Carp	5	10.6	0.5	0.03	0.09	0.80	<0.05
	Carp (R)	5	11.1	0.7	0.04	<0.05	0.10	<0.05
	Bluegill	5	6.8	0.1	0.04	0.07	ND	ND
	Bluegill (R)	5	6.2	0.1	0.07	<0.05	ND	ND
	Largemouth bass	5	12.1	0.7	0.07	0.19	0.20	ND
	Largemouth bass (R)	5	10.3	0.4	0.06	0.21	ND	ND
79 Canadian River Eufaula, Okla	Carp	5	13.9	1.3	0.12	0.09	ND	<0.05
	Carp (R)	5	14.9	1.4	0.08	0.11	ND	<0.05
	Bluegill	5	5.8	0.2	0.02	0.06	ND	ND
	Bluegill (R)	5	5.9	0.2	0.03	<0.05	<0.10	ND
	Channel catfish	4	15.8	1.4	0.15	<0.05	ND	<0.05
	Channel catfish (R)	4	15.4	1.4	0.16	ND	<0.05	ND
80 White River De Valls Bluff, Ark	Carp	2	19.1	3.5	0.13	<0.05	ND	0.05
	Carp (R)	2	22.4	6.5	0.24	0.11	0.11	0.17
	Bigmouth buffalo	2	18.6	3.9	0.11	0.15	ND	<0.05
	Bigmouth buffalo (R)	2	16.6	2.4	0.03	0.19	ND	<0.05
	Channel catfish	2	22.9	5.0	0.38	0.11	ND	<0.05
	Channel catfish (R)	2	19.6	3.1	0.12	0.14	ND	<0.05
80 Yazoo River Redwood, Miss	Carp	2	18.8	4.0	0.14	0.05	ND	0.05
	Carp (R)	1	21.4	5.5	0.12	<0.05	ND	0.07
	Smallmouth buffalo	2	16.0	2.5	0.19	0.08	0.22	<0.05
	Smallmouth buffalo (R)	2	16.1	2.5	0.19	<0.05	0.20	<0.05
	Channel catfish	2	20.1	2.3	0.16	ND	ND	<0.05
81 Red River Alexandria, La	Smallmouth buffalo	2	16.1	2.3	0.09	0.07	0.15	<0.05
	Smallmouth buffalo (R)	2	15.5	1.9	0.23	<0.05	ND	ND
	Freshwater drum	3	13.5	1.2	0.20	0.09	ND	<0.05
	Freshwater drum (R)	2	14.2	1.3	0.16	0.09	ND	<0.05
	White catfish	3	14.9	1.2	0.10	<0.05	0.15	0.06
	White catfish (R)	2	14.7	1.2	0.03	<0.05	0.10	<0.05
82 Red River Lake Texoma, Okla	Carp	3	20.6	3.9	0.11	<0.05	0.13	<0.05
	Carp (R)	3	19.5	3.4	0.01	<0.05	0.18	0.05
	Bluegill	4	7.1	0.3	0.02	0.10	0.15	<0.05
	Bluegill (R)	4	6.8	0.3	0.02	0.07	<0.10	ND
	Largemouth bass	3	12.5	0.8	0.08	0.22	0.16	<0.05
	Largemouth bass (R)	3	14.5	1.4	0.06	<0.05	0.14	<0.05
83 Missouri River Hermann, Mo	Carp	5	17.5	2.4	0.05	<0.05	0.11	0.08
	Carp (R)	5	17.2	2.4	0.03	<0.05	0.14	0.13
	Smallmouth buffalo	3	15.7	2.2	0.08	0.06	0.35	<0.05
	Smallmouth buffalo (R)	3	17.0	2.9	0.18	<0.05	0.19	<0.05
31 Missouri River Nebraska City, Nebr	Carp	5	17.4	2.8	0.06	<0.05	0.10	0.07
	Carp (R)	5	16.3	2.2	0.06	0.06	0.11	0.06
	Goldeye	5	14.1	1.0	0.14	ND	0.23	<0.05
	Goldeye (R)	5	13.7	0.9	0.10	ND	0.20	<0.05
	White crappie	5	9.5	0.5	0.08	0.14	ND	<0.05
	White crappie (R)	5	8.4	0.3	0.04	0.08	ND	ND
32 Missouri River Garrison Dam, N. Dak	White sucker	3	13.2	0.8	0.12	0.08	ND	<0.05
	White sucker (R)	3	13.5	0.8	0.10	0.17	ND	ND
	Goldeye	5	12.5	0.5	0.17	0.22	ND	<0.05
	Goldeye (R)	5	12.2	0.4	0.74	0.24	ND	ND
	Walleye	3	18.3	2.1	0.17	0.20	0.13	<0.05
	Walleye (R)	3	18.3	2.2	0.20	0.14	0.13	<0.05
33 Missouri River Great Falls, Mont	Goldeye	5	11.8	0.6	0.18	0.08	ND	<0.05
	Goldeye (R)	5	11.8	0.6	0.13	0.24	0.10	0.05
	Redhorse	5	17.2	2.2	0.18	0.08	0.10	0.23
	Redhorse (R)	5	17.5	2.3	0.18	0.06	0.11	0.20
	Sauger	5	10.8	0.4	0.19	<0.05	ND	<0.05
	Sauger (R)	4	9.9	0.3	0.12	<0.05	ND	<0.05
84 Big Horn River Hardin, Mont	Carp	5	15.3	2.4	0.28	<0.05	ND	0.09
	Carp (R)	5	15.8	2.7	0.12	0.09	ND	0.13
	Goldeye	5	11.0	0.6	0.23	0.07	ND	<0.05
	Goldeye (R)	5	10.8	0.5	0.26	0.05	ND	<0.05
	Carpsucker	5	12.0	1.1	0.10	0.30	ND	ND
	Carpsucker (R)	5	11.5	0.9	0.13	0.29	ND	<0.05
85 Yellowstone R. Sidney, Mont	Goldeye	5	10.8	0.6	0.08	0.28	ND	<0.05
	Goldeye (R)	5	10.9	0.6	0.09	0.18	0.10	<0.05
	Carpsucker	4	13.2	1.7	0.13	0.17	0.10	<0.05
	Carpsucker (R)	4	11.8	1.2	0.02	0.14	<0.10	<0.05
	Walleye	3	13.4	1.2	0.20	0.06	0.10	<0.05
	Walleye (R)	3	12.5	0.6	0.13	0.05	<0.10	<0.05
86 James R. Oriska, N. Dak	[illegible]	3	18.3	2.7	0.05 (0.09)	0.05 (<0.05)	ND (0.21)	0.05 (<0.05)
	[illegible] (R)	3	18.0	2.6	0.07	<0.05	0.11	<0.05

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TABLE 1 (cont'd.). *Concentrations of mercury, arsenic, lead, and cadmium in whole fish, 1971 —National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size		Residues, mg/kg wet weight			
			Length, in	Weight, lb	Mercury	Arsenic	Lead	Cadmium
	Goldeye	5	13.2	0.9	0.06 (0.09)	<0.05 (<0.05)	0.24 (0.21)	ND (<0.05)
	Goldeye (R)	5	13.2	0.8	0.32 (0.12)	<0.05 (<0.05)	ND (<0.21)	ND (<0.05)
	Channel catfish	3	13.5	1.0	0.11	0.10	0.14	ND
	Channel catfish (R)	3	14.5	1.2	0.06	<0.05	ND	ND
87. North Platte River Lake McConaughy, Nebr	Carp	5	14.9	2.1	0.21	0.24	ND	<0.05
	Carp (R)	5	16.4	2.1	0.10	0.24	0.13	<0.05
	Channel catfish	5	17.4	1.8	0.25	0.14	ND	<0.05
	Channel catfish (R)	5	19.8	2.4	0.16	0.17	ND	<0.05
	Walleye	5	16.5	1.9	0.14	0.41	ND	0.54
	Walleye (R)	5	15.9	1.6	0.18	1.50	ND	<0.05
88. South Platte River Brule, Nebr	Carp	5	18.1	2.5	0.14	ND	0.38	0.05
	Carp (R)	5	17.3	2.4	0.12	0.06	ND	<0.05
	White sucker	5	12.8	0.8	0.10	0.05	0.15	ND
	White sucker (R)	5	12.3	0.8	0.10	0.08	ND	ND
	Black bullhead	5	8.4	0.4	0.34	<0.05	ND	ND
	Black bullhead (R)	5	8.3	0.3	0.14	<0.05	ND	ND
89. Platte River Louisville, Nebr	Carp	5	16.4	2.0	0.02	<0.05	0.16	0.22
	Carp (R)	5	16.6	2.0	0.12	<0.05	<0.10	0.07
	Channel catfish	5	12.1	0.5	0.08	ND	0.13	<0.05
	Channel catfish (R)	5	10.2	0.4	0.16	ND	0.22	<0.05
	White crappie	3	9.8	0.5	0.11	0.08	<0.10	<0.05
	White crappie (R)	3	8.1	0.3	0.15	0.08	<0.10	ND
90. Kansas River Bonner Springs, Kans	Carp	4	18.7	3.1	0.10	<0.05	0.11	<0.05
	Carp (R)	5	13.6	1.4	0.05	0.09	ND	<0.05
	Gizzard shad	5	7.8	0.3	0.04	0.14	0.10	<0.05
	Gizzard shad (R)	5	7.2	0.3	0.09	0.13	ND	<0.05
	Channel catfish	5	8.1	0.1	0.05	<0.05	0.27	<0.05
	Channel catfish (R)	5	7.8	0.1	0.08	<0.05	0.28	<0.05
Hudson Bay Drainage								
34. Red River Noyes, Minn.	Goldeye	5	13.6	0.8	0.19	<0.05	0.20	<0.05
	Goldeye (R)	5	13.0	0.8	0.18	<0.05	ND	<0.05
	Channel catfish	1	18.0	1.9	0.14	<0.05	0.11	<0.05
	Sauger	5	11.6	0.4	0.12	0.05	0.11	<0.05
	Sauger (R)	5	11.0	0.3	0.62	<0.05	0.10	0.15
Colorado River System								
35. Green River Vernal, Utah	Carp	4	13.7	1.5	0.06	<0.05	ND	ND
	Carp (R)	4	15.2	2.0	0.10	<0.05	ND	<0.05
	Flannelmouth sucker	4	16.4	1.4	0.10	0.13	0.18	<0.05
	Flannelmouth sucker (R)	4	16.1	1.4	0.19	0.09	0.24	<0.05
	Channel catfish	5	8.3	0.2	0.10	<0.05	0.11	0.12
	Channel catfish (R)	5	8.6	0.2	0.12	<0.05	ND	<0.05
36. Colorado River Imperial Reservoir, Ariz	Carp	3	15.9	2.3	ND	0.08	0.14	<0.05
	Carp (R)	3	18.1	2.9	ND	ND	ND	ND
	Redear sunfish	5	7.6	0.3	ND	0.14	ND	<0.05
	Redear sunfish (R)	5	7.7	0.3	0.02	0.08	ND	ND
	Largemouth bass	4	8.7	0.4	0.79	0.13	ND	<0.05
	Largemouth bass (R)	3	10.2	0.5	0.02	<0.05	ND	ND
91. Colorado River Havasu Lake, Ariz	Carp	3	13.7	1.5	0.01	<0.05	0.18	ND
	Carp (R)	4	14.1	1.3	0.02	<0.05	0.12	ND
	Channel catfish	3	16.5	1.4	0.01	0.29	0.15	<0.05
	Channel catfish (R)	4	15.8	1.2	0.02	0.13	ND	<0.05
	Black crappie	3	7.8	0.3	0.04	0.11	ND	ND
	Black crappie (R)	4	8.6	0.4	0.01	0.16	ND	ND
92. Colorado River Lake Mead, Nev.	Carp	3	18.0	2.8	0.06	0.10	1.10	0.14
	Channel catfish	3	14.9	1.1	0.03	0.06	ND	ND
	Channel catfish (R)	2	13.9	0.8	0.08	<0.05	0.10	<0.05
	Largemouth bass	3	13.1	1.5	0.02	0.27	ND	<0.05
	Largemouth bass (R)	3	11.3	0.4	0.52	0.18	ND	<0.05
93. Colorado River Lake Powell, Ariz	Carp	5	13.2	0.9	0.70	0.11	0.13	0.27
	Carp (R)	5	13.0	0.8	0.10	0.08	ND	0.28
	Largemouth bass	4	13.3	1.4	0.20	0.50	0.16	<0.05
	Largemouth bass (R)	3	15.2	2.3	0.33	0.30	ND	<0.05
	Rainbow trout	3	21.1	4.9	0.15	0.26	0.65	<0.05
	Rainbow trout (R)	4	19.4	3.7	0.05	0.13	0.14	<0.05
94. Gila River San Carlos Reservoir, Ariz	Carp	3	12.8	0.9	0.20	0.18	0.16	<0.05
	Carp (R)	5	11.7	0.7	0.06	0.17	0.22	<0.05
	Channel catfish	3	9.2	0.2	0.10	<0.05	0.16	<0.05
	Channel catfish (R)	5	12.6	0.5	0.12	<0.05	0.20	<0.05
	Largemouth bass	3	14.4	1.4	0.25	0.14	0.13	<0.05

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TABLE 1 (cont'd.) *Concentrations of mercury, arsenic, lead, and cadmium in whole fish, 1971 —National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size		Residues, mg/kg wet weight			
			Length, in.	Weight, lb.	Mercury	Arsenic	Lead	Cadmium
Interior Basins								
37 Truckee River Fernley, Nev.	Carp	5	15.5	1.7	0.64	0.09	ND	<0.05
	Carp (R)	5	14.5	1.5	0.46	0.08	ND	<0.05
	Brown bullhead	3	8.9	0.4	0.20	ND	ND	<0.05
	Brown bullhead (R)	3	9.0	0.4	0.19	0.11	ND	<0.05
	Largemouth bass	2	10.7	0.8	0.42	0.05	0.15	0.23
	Largemouth bass (R)	2	11.4	1.0	0.52	0.05	ND	<0.05
38 Utah Lake Provo, Utah	Carp	5	15.2	1.8	0.01	0.26	0.10	<0.05
	Carp (R)	5	13.5	1.3	ND	0.12	0.11	<0.05
	Black bullhead	5	10.1	0.6	0.02	0.06	0.20	ND
	Black bullhead (R)	5	10.9	0.7	0.02	0.06	0.22	ND
	White bass	5	8.9	0.3	0.04	0.15	0.30	ND
	White bass (R)	5	8.4	0.3	0.08	0.20	0.41	ND
95 Bear River Preston, Idaho	Carp	5	14.3	2.1	0.10	0.08	ND	0.08
	Carp (R)	5	14.4	2.3	0.12	0.11	ND	<0.05
	Largescale sucker	5	16.6	2.2	0.20	0.08	0.12	<0.05
	Largescale sucker (R)	5	17.5	2.4	0.24	0.09	ND	<0.05
	Yellow perch	5	8.0	0.3	0.03	<0.05	ND	<0.05
	Yellow perch (R)	5	8.1	0.3	0.15	ND	0.13	<0.05
California Streams								
39 Sacramento River Sacramento, Calif.	Carp	5	13.0	1.4	0.16	0.19	0.19	0.08
	Carp (R)	5	12.7	1.3	0.10	0.29	0.14	<0.05
	White catfish	5	9.1	0.4	0.13	0.08	<0.10	<0.05
	White catfish (R)	5	9.4	0.5	0.13	0.12	0.50	<0.05
					(0.16)	(0.12)	(0.3)	(<0.05)
	Largemouth bass	5	12.5	1.3	0.18	0.07	<0.10	<0.05
	Largemouth bass (R)	5	11.3	1.0	0.12	<0.05	<0.10	<0.05
40 San Joaquin River Los Banos, Calif.	Carp	5	12.9	1.4	0.07	0.06	0.14	<0.05
	Carp (R)	5	11.9	1.4	0.02	<0.05	<0.10	<0.05
	Channel catfish	3	15.6	2.4	0.09	0.06	<0.10	ND
	Channel catfish (R)	3	14.2	2.1	0.12	0.08	0.11	<0.05
	Black crappie	5	10.2	0.8	0.04	0.42	<0.10	ND
	Black crappie (R)	5	10.0	0.8	0.01	0.29	<0.10	<0.05
Columbia River System								
43 Salmon River Riggins, Idaho	Carp	4	15.4	2.4	0.23	0.11	ND	<0.05
	Largescale sucker	5	15.3	1.7	0.36	0.12	ND	ND
	Largescale sucker (R)	5	15.8	1.8	0.19	0.15	0.14	0.21
	Northern squawfish	3	11.2	0.6	1.20	<0.05	ND	<0.05
	Northern squawfish (R)	3	13.9	1.2	0.42	0.20	ND	ND
	Smallmouth bass	3	11.3	1.0	0.36	<0.05	ND	<0.05
41 Snake River Hagerman, Idaho	Largescale sucker	5	12.6	1.0	0.08	0.06	<0.10	<0.05
	Largescale sucker (R)	5	13.0	1.0	0.05	0.08	<0.10	<0.05
	Peamouth chub	5	9.4	0.3	0.08	<0.05	<0.10	<0.05
	Peamouth chub (R)	5	9.5	0.4	0.10	<0.05	<0.10	<0.05
	Northern squawfish	5	14.4	1.3	0.28	0.06	ND	<0.05
	Northern squawfish (R)	5	14.3	1.3	0.35	ND	<0.10	<0.05
42 Snake River Lewiston, Idaho	Carp	5	14.9	2.0	0.36	<0.05	0.11	0.09
	Carp (R)	5	14.5	2.4	0.25	0.22	0.11	<0.05
	Northern squawfish	3	14.0	1.2	1.30	<0.05	ND	ND
	Northern squawfish (R)	3	14.0	1.2	0.89	<0.05	ND	0.05
	Smallmouth bass	5	11.2	1.1	0.28	0.06	ND	ND
	Smallmouth bass (R)	5	10.4	0.7	0.28	0.06	ND	ND
96 Snake River Ice Harbor, Wash.	Carp	5	13.1	1.6	0.19	0.08	0.11	0.11
	Carp (R)	5	12.4	1.6	0.24	0.14	<0.10	0.05
	Largescale sucker	5	14.1	1.1	0.14	0.24	<0.10	<0.05
	Largescale sucker (R)	5	13.5	1.0	0.14	0.22	0.12	<0.05
	Channel catfish	5	14.3	1.6	0.26	0.10	<0.10	<0.05
	Channel catfish (R)	5	14.7	1.9	0.60	0.10	<0.10	0.13
44 Yakima River Granger, Wash.	Carp	5	11.9	1.1	0.14	0.06	ND	ND
	Carp (R)	5	11.9	1.2	0.21	<0.05	ND	<0.05
	Largescale sucker	5	14.0	1.2	0.07	0.09	0.12	<0.05
	Largescale sucker (R)	5	14.1	1.2	0.09	0.10	ND	ND
	Black crappie	5	7.4	0.3	0.06	<0.05	ND	<0.05
	Black crappie (R)	5	7.4	0.3	0.12	<0.05	ND	ND
45 Walla Walla R. Oreg. Oreg.	Largescale sucker	5	15.9	1.8	0.28	0.05	ND	<0.05
	Largescale sucker (R)	5	15.8	1.8	0.32	0.05	<0.10	ND
	Northern squawfish	3	13.7	1.2	1.10	<0.05	ND	ND
	Northern squawfish (R)	3	13.5	1.1	0.99	<0.05	ND	<0.05

(*Continued next page.*)

TABLE 1 (cont'd.). *Concentrations of mercury, arsenic, lead, and cadmium in whole fish, 1971 —National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size		Residues, mg/kg wet weight			
			Length, in	Weight, lb	Mercury	Arsenic	Lead	Cadmium
46. Columbia River Bonneville Dam, Oreg	Carp	5	11 7	1.0	0.05	0 15	ND	0 11
	Carp (R)	5	11 3	0.9	0 05	0.13	0.12	0.05
	Largescale sucker	5	16.2	1.8	0 21	0 12	0 11	<0 05
	Largescale sucker (R)	4	15.8	1 8	0.19	0 28	0.19	<0.05
	Northern squawfish	5	13 6	1.3	0.84	<0 05	ND	<0.05
	Northern squawfish (R)	5	14 6	1 4	0 76	0 05	ND	<0 05
97. Columbia River Pasco, Wash.	Carp	5	12 3	1.3	0 08	0 06	0 34	0.08
	Carp (R)	5	12.1	1 3	0.07	<0 05	0.40	0.12
	Largescale sucker	5	17 2	2 0	0 06	0 15	0.52	0.13
	Largescale sucker (R)	5	17.1	2.1	0.06	0 16	0.38	0 07
	Northern squawfish	5	12.2	0.8	0 15	<0 05	0 18	0.06
	Northern squawfish (R)	5	11.6	0.7	0.16	<0 05	0 11	<0.05
98 Columbia River Grand Coulee, Wash	Largescale sucker	5	13 1	1 0	0 08	0 15	0 58	0 10
	Largescale sucker (R)	5	13 5	1 1	0.03 (<0 05)	0 18 (0 18)	0 90 (0 7)	0 13 (0 08)
	Northern squawfish	5	14 2	1 4	0.22	ND	0 26	0.23
	Walleye	4	16 1	2 0	0.15	0 07	0 12	0.07
	Walleye (R)	5	16 9	2.1	0 15	0.06	<0.10	<0 05
Pacific Coast Streams								
47 Klamath River Hornbrook, Calif	Klamath sucker	4	14 3	1.6	0 24	0 12	ND	<0.05
	Klamath sucker (R)	3	15 5	2 2	0.28	0.10	0.11	0 17
	Brown bullhead	5	8 6	0 3	0 09	<0.05	ND	<0 05
	Brown bullhead (R)	5	8 6	0 4	0.06	<0.05	ND	ND
	Yellow perch	5	7 7	0 2	0 20	<0 05	ND	0 07
	Yellow perch (R)	5	7 6	0 2	0 18	<0.05	0.15	0 12
48. Rogue River Gold Ray Dam, Oreg	Bridgelip sucker	5	12 7	1 2	0 08	0.06	0.11	<0.05
	Bridgelip sucker (R)	4	13 8	1 4	0 15	0 06	ND	0.06
	Brown bullhead	5	10 4	0.6	0.25	<0.05	ND	ND
	Brown bullhead (R)	5	10 2	0.5	0.30	<0.05	ND	<0.05
	Black crappie	4	9 0	0.5	0.14	<0 05	ND	<0.05
	Black crappie (R)	3	9 0	0.6	0 14	<0 05	ND	<0.05
Alaskan Streams								
49 Chena River Fairbanks, Alaska	Longnose sucker	3	14 1	1 6	0 05	0 06	0 29	<0.05
	Longnose sucker (R)	3	14 4	1 6	0 06	0 11	0.22	<0.05
	Round whitefish	3	10 9	0.5	0 07	<0 05	0 24	<0 05
	Round whitefish (R)	3	12 5	0.7	0 04	<0 05	ND	ND
	Arctic grayling	3	11.5	0 7	0.06	<0 05	0 19	ND
	Arctic grayling (R)	3	12 4	0 9	0 05	<0 05	0 23	<0.05
50 Kenai River Soldatna, Alaska	Round whitefish	5	13 8	0.8	0 04	0 14	0 11	<0.05
	Round whitefish (R)	5	11.7	0.5	0 06	0 18	ND	<0 05
	Lake trout	5	16 4	1 2	0 18	0 11	ND	<0 05
	Lake trout (R)	5	16 7	1 2	0.23	0 06	ND	<0.05
	Rainbow trout	5	11 1	0 6	0.06	0 16	ND	0 07
	Rainbow trout (R)	2	10 4	0 5	0.05	0 10	ND	<0.05
Hawaiian Streams								
99 Waikele Stream Waipahu, Hawaii	Tilapia	5	5 5	0 1	0.04 (0 07)	0.07 (0.09)	1.40 (1 7)	0.10 (0 09)
	Tilapia (R)	5	5 2	0 1	0.05	<0.05	0 90	0.05
	Cuban limia	10	3 3	ND	0.07 (0 07)	0.06 (0.06)	ND (0.4)	ND (<0 05)
	Cuban limia (R)	10	3 2	ND	0 07	<0.05	0.19	<0.05
	Chinese catfish	4	8.0	0.2	0 14	0.14	0.35	0.12
	Chinese catfish (R)	4	7 9	0.2	0 19	0 28	0.14	<0 05
100 Manoa Stream Honolulu, Hawaii	Tilapia	2	9 2	0 6	0 02	0.30	0.30	<0.05
	Tilapia (R)	2	9.5	0.5	0 05	0 11	0.12	ND
	Cuban limia	8	3.3	ND	ND	ND	ND	ND
	Cuban limia (R)	10	3 1	ND	0.06	0 11	3 24	<0.05
	Chinese catfish	2	10.3	0.4	0.11	<0 05	0.28	0.10
	Chinese catfish (R)	4	9 8	0.3	0 10	ND	0 12	<0 05

Note: R = replicate sample
ND = not detected
Numbers in parentheses are results of cross-check analyses

[1] Redhorse = *Moxostoma* sp

TABLE 2. *Concentrations of mercury, arsenic, lead, cadmium, and selenium in whole fish, 1972 —National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size: Length, in	Average Size: Weight, lb	Residues, mg/kg wet weight: Mercury	Arsenic	Lead	Cadmium	Selenium
Atlantic Coast Streams									
1 Stillwater River, Old Town, Maine	White sucker	5	13.5	1.0	0.49	0.32	0.22	<0.05	0.13
	Chain pickerel	5	14.1	0.56	0.62	<0.05	0.14	<0.05	0.05
	Yellow perch	5	8.9	0.31	0.50	<0.05	0.60	<0.05	0.25
	Yellow perch (R)	5	8.9	0.32	0.62	<0.05	0.40	<0.05	0.07
51 Kennebec River, Hinckley, Maine	Smallmouth bass	5	11.2	0.6	0.99	<0.05	0.20	<0.05	0.08
					(0.78)	(0.05)	(<0.20)	(0.05)	(0.35
	White perch	5	8.5	0.3	0.23	<0.05	0.30	<0.05	0.76
	Yellow perch	5	9.7	0.4	0.64	0.11	0.40	<0.05	0.04
	Yellow perch (R)	5	9.7	0.4	0.64	0.15	0.30	<0.05	0.05
52 Lake Champlain, Burlington, Vt	Pumpkinseed	5	7.8	0.5	0.32	0.15	0.31	<0.05	0.35
	Chain pickerel	5	17.1	1.1	0.50	0.05	0.50	<0.05	0.92
	Yellow perch	5	10.6	0.6	0.39	0.07	0.40	<0.05	0.29
	Yellow perch (R)	5	11.0	0.7	0.24	0.09	0.40	<0.05	0.38
53 Merrimac River, Lowell, Mass	White sucker	5	12.5	0.5	0.44	0.18	0.50	0.30	0.34
	Carp	5	10.2	0.5	0.09	0.14	0.80	<0.05	0.40
	Yellow perch	5	10.1	0.6	0.56	0.23	0.14	<0.05	0.30
	Yellow perch (R)	5	9.3	0.5	0.49	<0.05	<0.10	0.26	0.34
2 Connecticut River, Windsor Locks, Conn	White perch	5	10.0	0.5	0.32	0.10	0.34	0.17	1.40
					(0.50)	(0.12)	(<0.20)	(0.08)	(1.17)
	Yellow perch	5	9.0	0.4	0.24	0.10	0.30	0.64	0.25
	White catfish	5	13.5	1.0	0.12	0.11	0.40	1.50	0.17
					(0.20)	(0.10)	(0.07)	(0.16)	(0.20)
	White catfish (R)	5	13.3	1.0	0.15	0.20	0.40	0.56	0.17
3 Hudson River, Poughkeepsie, N.Y.	Pumpkinseed	5	5.8	0.1	0.12	0.12	0.90	<0.05	0.37
	Largemouth bass	5	10.5	0.56	0.13	0.12	1.60	<0.05	0.37
	Goldfish	5	11.9	1.1	0.15	0.21	2.60	0.10	0.30
	Goldfish (R)	5	10.1	0.56	0.14	0.09	1.70	0.10	0.28
54 Raritan River, Highland Park, N.J.	Golden shiner	5	7.3	0.2	0.14	0.09	1.10	<0.05	0.72
	Largemouth bass	5	9.7	0.5	0.28	0.12	<0.10	<0.05	0.58
	White perch	5	7.2	0.3	0.10	0.09	<0.10	<0.05	1.30
	White perch (R)	5	6.7	0.3	0.18	<0.05	1.40	<0.05	1.10
4 Delaware River, Camden, N.J.	White sucker	5	15.0	1.5	0.18	0.18	0.72	<0.05	0.41
	Brown bullhead	5	11.5	0.9	0.02	0.10	1.30	<0.05	0.24
	White perch	5	11.1	0.5	0.10	0.09	0.56	<0.05	1.20
	White perch (R)	5	9.9	0.5	0.14	<0.05	0.46	<0.05	0.84
5 Susquehanna River, Conowingo Dam, Md.	Carp	5	14.8	1.7	0.16	0.08	0.28	<0.05	0.55
	Channel catfish	5	13.1	0.8	0.10	0.18	0.28	<0.05	1.51
	Yellow perch	5	7.5	0.2	0.08	0.21	0.26	<0.05	0.46
	Yellow perch (R)	5	7.5	0.3	0.03	0.21	0.36	<0.05	0.80
6 Potomac River, Little Falls, Md.	Carp	4	14.2	1.35	0.22	0.25	1.8	<0.05	0.20
	Redbreast sunfish	4	6.6	0.2	0.06	0.11	0.56	<0.05	0.52
	Smallmouth bass	1	16.2	1.9	0.36	<0.05	0.20	<0.05	0.40
	Redhorse[1]	5	13.3	2.2	0.16	0.07	<0.10	<0.05	0.24
	Redhorse (R)	5	13.2	1.0	0.24	<0.05	1.5	<0.05	0.24
11 James River, Richmond, Va.	White sucker	3	16.2	2.0	0.08	0.12	<0.10	<0.05	0.92
	Redhorse	1	16.2	1.9	0.12	<0.05	<0.10	0.08	0.88
	River chub	5	11.5	0.7	0.05	0.17	0.14	<0.05	0.68
	River chub (R)	4	12.5	0.9	<0.05	0.2	<0.10	<0.05	0.48
[illegible] Rapids, Va.	Largemouth bass	2	11.3	0.9	0.23	0.05	<0.10	<0.05	0.34
	White catfish	5	9.3	0.3	0.04	<0.05	0.30	<0.05	0.17
	Carp	4	19.4	3.5	0.11	0.16	0.56	0.06	0.60
					(0.08)	(<0.05)	(<0.20)	(0.06)	(0.45)
	Bluegill	5	7.2	0.3	0.04	<0.05	<0.11	0.05	0.22
	Bluegill (R)	5	6.4	0.2	0.10	0.11	0.22	<0.05	0.19
[illegible] R.	Gizzard shad	3	9.6	0.5	0.10	0.21	0.22	<0.05	0.24
	Largemouth bass	4	11.0	0.8	0.40	0.14	<0.10	<0.05	0.12
	Brown bullhead	5	10.6	0.5	0.10	0.07	0.12	0.12	0.24
	Brown bullhead (R)	5	10.3	0.5	0.13	0.07	0.64	<0.05	0.32
56 Pee Dee [illegible]	Brown bullhead	5	12.3	0.7	0.36	<0.05	<0.10	<0.05	0.26
Dong	Largemouth bass	5	14.3	1.5	0.10	<0.05	1.2	0.55	0.23
	Bluegill	5	6.8	0.2	0.25	0.09	<0.10	<0.05	0.34
	Bluegill (R)	5	6.6	0.2	0.24	<0.05	<0.10	<0.05	0.54
9 Cooper Ri[illegible]		3	22.1	5.4	0.28	0.24	0.13	<0.05	0.52
Summerton,	ith bass	5	13.2	1.2	0.24	0.09	<0.10	<0.05	0.54
		5	6.9	0.2	0.01	<0.05	0.10	<0.05	0.23
		5	6.3	0.2	0.12	<0.05	<0.10	<0.05	0.39

(Continued next)

TABLE 2 (cont'd.). *Concentrations of mercury, arsenic, lead, cadmium, and selenium in whole fish, 1972 —National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size: Length, in.	Average Size: Weight, lb	Residues, mg/kg wet weight: Mercury	Arsenic	Lead	Cadmium	Selenium
10. Savannah River Savannah, Ga.	Bluegill	5	7.1	0.3	0.14	0.15	0.10	<0.05	0.34
	Channel catfish	5	17.4	2.0	0.56	0.07	0.20	<0.05	0.09
	Largemouth bass	3	13.8	1.4	ND (1.25)	ND (0.10)	ND (<0.20)	ND (<0.05)	ND (0.45)
	Largemouth bass (R)	3	10.3	0.53	0.67	0.05	0.10	<0.05	0.16
57. Altamaha River Doctortown, Ga.	Bluegill	4	5.9	0.2	0.11	0.09	0.30	<0.05	0.16
	Largemouth bass	3	15.8	2.5	0.54	0.22	0.10	0.05	0.12
	Spotted sucker	4	17.5	2.1	0.26	<0.05	0.20	<0.05	0.43
	Spotted sucker (R)	4	15.5	1.5	0.32	0.05	0.20	<0.05	0.40
11. St. Johns River Welaka, Fla	Striped mullet	5	17.1	2.1	0.03	0.12	0.32	<0.05	0.58
	Channel catfish	5	11.8	0.4	0.03	0.10	0.09	<0.05	0.26
	Largemouth bass	5	13.5	1.2	0.10	<0.05	0.12	<0.05	0.42
	Largemouth bass (R)	5	10.3	0.5	0.07	0.06	0.12	<0.05	0.36
12. St Lucie Canal Indiantown, Fla.	Bluegill	5	6.1	0.2	0.09	0.09	<0.10	<0.05	0.60
	Largemouth bass	3	14.3	1.5	0.16	0.10	<0.10	<0.05	0.62
	White catfish	5	13.5	1.4	0.06	<0.05	0.17	<0.05	0.42
	White catfish (R)	5	11.6	0.8	0.06	0.17	<0.10	<0.05	0.45
Gulf Coast Streams									
58 Suwanee River Old Town, Fla	Redbreast sunfish	5	5.9	0.1	0.09	0.08	<0.10	<0.05	0.44
	Largemouth bass	5	11.5	0.7	0.49	0.09	<0.10	<0.05	0.72
	Spotted sucker	5	16.8	1.8	0.11	0.15	<0.10	<0.05	0.76
	Spotted sucker (R)	5	14.1	1.2	0.07	0.06	<0.10	<0.05	0.44
13. Apalachicola River Jim Woodruff Dam, Ala.	Spotted sucker	3	19.0	2.9	0.09	0.10	<0.10	<0.05	0.56
	Largemouth bass	3	14.1	1.4	0.19	0.06	<0.10	<0.05	0.40
	Carp	3	22.6	6.9	0.07	0.10	<0.10	<0.05	0.52
	Carp (R)	3	25.7	9.0	0.05	0.23	<0.10	<0.05	0.40
59. Alabama River Chrysler, Ala.	Striped mullet	1	12.3	0.7	0.04	0.29	<0.10	<0.05	0.28
	Freshwater drum	3	9.1	0.2	0.08	0.05	<0.10	<0.05	0.44
	Channel catfish	3	11.8	0.4	0.04	0.05	<0.10	0.16	0.41
	Largemouth bass	3	14.1	1.3	0.16	0.05	<0.10	<0.05	0.64
	Largemouth bass (R)	3	13.1	1.3	0.36	0.09	<0.10	<0.05	0.10
14. Tombigbee River McIntosh, Ala.	Striped mullet	5	15.6	1.4	0.10	0.28	0.19	<0.05	0.72
	Largemouth bass	5	10.6	0.7	0.96	0.05	<0.10	<0.05	0.40
	Channel catfish	5	13.7	0.7	0.12 (0.08)	0.21 (0.08)	<0.10 (<0.20)	<0.05 (<0.05)	0.40 (0.29)
	Channel catfish (R)	5	12.7	0.5	0.13	0.14	0.10	<0.05	0.56
15. Mississippi River Luling, La.	Carp	3	23.0	6.1	0.08	0.14	0.18	<0.05	0.52
	Channel catfish	5	13.8	1.1	0.06	0.20	0.30	<0.05	1.30
	Freshwater drum	5	14.2	1.4	0.14	0.13	0.16	0.12	1.44
	Freshwater drum (R)	5	9.8	0.7	0.16	0.08	0.16	0.28	0.64
60. Brazos River Richmond, Tex.	Longnose gar	3	25.4	0.87	0.17	0.08	0.40	<0.05	0.40
	River carpsucker	3	10.4	0.58	0.06	0.25	1.1	0.11	0.32
	Channel catfish	3	12.4	0.56	0.10	0.09	1.20	0.07	0.40
	Blue catfish	3	13.9	1.1	0.12	0.06	0.60	<0.05	0.20
61. Colorado River Wharton, Tex	Spotted gar	3	20.5	1.3	0.46	<0.05	0.22	<0.05	0.28
	Channel catfish	3	16.7	1.5	0.12	0.06	0.70	0.17	0.36
	River carpsucker	3	13.2	1.0	0.14	0.28	3.6	<0.05	0.40
	River carpsucker (R)	3	13.2	1.21	0.11	0.13	2.9	<0.05	0.30
62. Nueces River Mathis, Tex.	Channel catfish	5	10.4	0.3	0.02	<0.05	<0.10	<0.05	0.20
	Blue catfish	5	12.8	0.8	0.06	0.05	<0.10	<0.05	0.18
	Gizzard shad	5	12.1	0.7	0.02	<0.05	<0.10	<0.05	0.49
	Gizzard shad (R)	5	11.6	0.6	0.02	0.08	0.26	<0.05	1.10
16. Rio Grande Brownsville, Tex.	Channel catfish	3	15.2	1.1	0.11	<0.05	0.10	<0.05	0.41
	Blue catfish	4	14.3	1.2	0.08	<0.05	0.22	<0.05	0.70
	Gizzard shad	5	8.0	0.2	0.02	0.18	<0.10	<0.05	0.71
	Gizzard shad (R)	5	6.9	0.1	0.01	<0.05	<0.10	<0.05	0.51
63. Rio Grande Elephant Butte, N. Mex.	Largemouth bass	5	15.0	2.0	0.28 (0.35)	0.27 (0.28)	<0.10 (<0.20)	<0.05 (<0.05)	0.66 (0.49
	Carp	5	12.3	1.0	0.16	0.13	0.17	<0.05	0.64
	Channel catfish	5	10.2	0.56	0.24	0.08	<0.10	<0.05	0.20
	Channel catfish (R)	5	8.9	0.3	0.16 (0.21)	0.08 (<0.05)	0.18 (<0.20)	<0.05 (<0.05)	0.23 (0.34)
64. Rio Grande Alamosa, Colo.	Brown trout	5	13.3	0.9	0.10	0.23	0.14	<0.05	0.23
	Carp	5	15.4	2.1	0.05	0.08	4.65	<0.05	0.38
	White sucker	5	11.3	0.6	0.06	0.14	1.4	<0.05	0.23
	White sucker (R)	5	15.1	1.3	0.14	0.05	0.60	<0.05	0.27

(*Continued next page*)

TABLE 2 (cont'd.) *Concentrations of mercury, arsenic, lead, cadmium, and selenium in whole fish, 1972 —National Pesticide Monitoring Program*

	Station Number and Location	Species	No Fish	Average Size		Residues, mg/kg wet weight				
				Length, in	Weight, lb	Mercury	Arsenic	Lead	Cadmium	Selenium
65	Pecos River Red Bluff Lake, Tex	Gizzard shad	5	10 4	0 4	0 01	0.05	0.10	< 0.10	0.48
		Channel catfish	5	14 4	0 9	0 09	<0 05	< 0 10	0.05	0.24
		Smallmouth buffalo	5	13 6	1 1	0 08	0 05	0.10	0.09	1 00
		Smallmouth buffalo (R)	5	13 7	1 1	0 10	0 12	0 10	0.05	0 28
		Great Lakes Drainage								
17	Genessee River Scottsville, N Y	Rock bass	5	7 4	0 4	0 07	0 10	0.16	<0 05	0.70
		Redhorse	5	15 4	1 5	0 07	0 09	0.34	<0.05	0.34
		Northern pike	5	20 7	2.1	0.11	0 13	<0 10	<0 05	0.31
66	St Lawrence River Massena, N Y	White sucker	4	13.6	1 2	0.08	0 09	<0 10	<0.05	0 46
		Northern pike	5	24 7	3 5	0 24	0 08	0 22	<0.05	0.58
		Yellow perch	5	8.8	0 3	0.12	0 09	0.90	<0.05	0 48
		Yellow perch (R)	5	8.7	0.3	0 12	0 09	1.30	<0.05	0.38
		Northern pike	5	19 6	1.8	0.07	<0 05	<0 10	<0.05	0.34
18	Lake Ontario Port Ontario, N Y	White perch	5	8.2	0 4	0.31 (0.15)	0 08 (0 25)	1 1 (<0.2)	<0 05 (<0 05)	1 40 (1.00)
		Rock bass	5	7.7	0 4	0.25	0 21	0 60	<0.05	0.80
		Yellow perch	5	9 1	0 4	0.47	0 12	2 2	<0 05	0 52
		Yellow perch (R)	5	9 2	0 4	0.24	0.20	1 0	<0.05	0.60
19	Lake Erie Erie, Pa	White sucker	4	12.6	1 2	0.16	0 12	0.16	<0.05	0.58
		Freshwater drum	5	13 5	1.1	0 16	0 07	0.18	<0 05	0.84
		Yellow perch	5	8 6	0.3	0 16	<0 05	0.20	<0 05	0.80
		Yellow perch (R)	5	9 0	0.4	0 04 (0 08)	0 09 (0 08)	0 16 (<0.20)	<0 05 (<0.05)	0.64 (0.62)
20	Lake Huron Bay Port, Mich	Yellow perch	5	8.6	0.26	0 07	0 06	<0.10	0.01	0.23
		Yellow perch (R)	5	8.5	0.28	<0 05	0.09	<0.10	0 01	0.19
21	Lake Michigan Sheboygan, Wis	Bloater	5	10.7	0 50	0 16	0 65	0 12	<0 05	0 34
		Bloater (R)	5	10 9	0.62	0 12 (0 11)	1 70 (2 40)	0 12 (<0 20)	<0.05 (<0.05)	0 48 (0.31)
		Yellow perch	5	11.0	0.85	0 26	0 07	<0.10	<0.05	1 00
		Yellow perch (R)	5	10 7	0.82	0 26	0 07	0 12	<0.05	0 82
		Lake trout	5	18 3	2.5	0 20 (0 18)	0 36 (0 69)	<0 10 (<0 20)	<0 05 (<0 05)	0.48 (0 51)
		Lake trout (R)	5	19 2	2 5	0 20	0 15	<0 10	<0 05	0 74
22	Lake Superior Bayfield, Wis	Bloater	5	10 1	0.27	0 20 (0 17)	0 17 (0.50)	0 18 (0 50)	0.08 (0.08)	0 49 (0 40)
		Bloater (R)	5	10.0	0 28	0 10	0 42	0.10	0 08	0 32
		Lake whitefish	5	21 2	3 2	0.08	0 40	0 20	<0.05	0 38
		Lake trout	5	22 9	3.7	0 26	0 13	<0 10	<0 05	0 24
		Lake trout (R)	5	20 9	3 2	0 28	0 21	<0 10	<0 05	0 52
		Mississippi River System								
67	Allegheny River Natrona, Pa	Carp	5	17 8	2.9	0 09	0.17	0 66	0.11	0.52
		Walleye	5	15 6	1 0	0 04	0 07	0 20	< 0 05	0 49
		Smallmouth bass	5	9 9	0 4	0 08	<0 05	0 24	<0 05	0.70
		Smallmouth bass (R)	5	11 0	0 7	0 08	<0 05	0 22	<0 05	0 60
23	Kanawha River Winfield, W Va	Carp	3	10 1	1 4	0 01	0 13	2 0	<0 05	0 80
		White crappie	5	7.5	0 2	0 06	0 07	0 28	< 0 05	0.72
		Brown bullhead	5	10 4	0 5	0 01	< 0 05	2 4	<0 05	0 38
		Brown bullhead (R)	5	10 9	0 5	0 03	0 07	2 9	< 0 05	0.22
68	Wabash River New Harmony, Ind	Carp	5	17 4	2 8	0 16	0 15	< 0 10	1 40	0 40
		Channel catfish	5	11 3	0.3	0 07	< 0 05	0 15	<0 05	0 38
		White sucker	5	9 3	0 4	0 14	< 0 05	6 40	<0 05	0.38
		White sucker (R)	5	8 0	0 4	0 12	0 07	0 31	<0 05	0 56
73	Ohio River Marietta, Ohio	Carp	5	13 1	1 2	0 18	0 25	1 3	0 09	0 72
		Redhorse sucker	5	10 4	0 8	0 04	0 11	0 90	< 0 05	0 14
		Channel catfish	5	14 6	0 9	0 18	0 06	0 60	<0 05	0 34
		Channel catfish (R)	5	14 3	0 8	0 46	< 0 05	0 70	< 0 05	0 29
69	Ohio River Cincinnati, Ohio	Carp	5	14 8	1 6	0 06	0 20	0 20	0 48	0 68
		Channel catfish	5	14 9	1 1	0 12	0 10	0 30	0 76	0 68
		Sauger	5	11 7	0 54	0 10 (0 13)	0 19 (0 11)	< 0 10 (< 0 20)	< 0 05 (< 0 05)	0 44 (0 43)
		Sauger (R)	5	12 8	0 9	0 24	0 17	< 0 10	0 05	0 40
70	Ohio River Metropolis, Ill	Carp	5	17 7	3 0	0 21	0 15	0 18	0 08	0 64
		Channel catfish	5	ND	ND	0 09	0 13	0 16	< 0 05	0 34
		White crappie	5	11 4	0 9	0 09	0 19	< 0 10	< 0 05	0 34
		White crappie (R)	5	11 2	0 9	0 16	0 15	< 0 10	< 0 05	0 30

(Continued next page)

TABLE 2 (cont'd.). *Concentrations of mercury, arsenic, lead, cadmium, and selenium in whole fish, 1972 —National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size: Length, in	Average Size: Weight, lb	Residues, mg/kg wet weight: Mercury	Arsenic	Lead	Cadmium	Selenium
25. Cumberland River Clarksville, Tenn.	Carp	5	16.3	1.9	0.04	<0.05	0.34	0.05	0.55
					(0.37)	(0.16)	(<0.20)	(<0.05)	(0.54)
	Largemouth bass	5	13.4	1.6	0.34	0.10	<0.10	<0.05	0.36
	Bluegill	5	6.5	0.1	0.12	0.06	0.13	<0.05	0.38
	Bluegill (R)	5	6.2	0.1	0.19	0.05	<0.10	<0.05	1.10
71. Tennessee River Savannah, Tenn.	Largemouth bass	3	9.5	0.3	0.22	<0.05	<0.10	<0.05	0.44
	White sucker	3	15.0	1.4	0.78	0.06	0.10	<0.05	0.70
	Carp	3	19.3	3.7	0.28	0.17	0.50	0.12	0.68
	Carp (R)	3	18.4	2.7	0.28	0.24	0.18	0.09	0.56
72. Wisconsin River Woodman, Wis.	Smallmouth buffalo	4	15.6	2.3	0.11	0.14	0.22	<0.05	0.31
	Mooneye	3	10.3	0.4	0.01	0.10	<0.10	<0.05	0.52
	White sucker	5	15.2	1.5	0.15	0.17	0.18	<0.05	0.64
					(0.13)	(0.06)	(<0.20)	(<0.05)	(0.58)
	White sucker (R)	4	13.0	1.2	0.02	0.14	<0.10	<0.05	0.24
73. Des Moines River Keosauqua, Iowa	Largemouth bass	5	13.1	1.2	0.16	0.07	0.80	<0.05	0.76
	Walleye	1	14.7	1.3	0.09	0.24	0.60	0.05	0.96
	Channel catfish	4	14.3	0.7	0.09	<0.05	0.16	<0.05	0.54
	Carp	5	15.2	1.6	0.03	0.09	0.60	0.07	1.10
	Carp (R)	5	13.2	1.0	0.03	0.09	0.16	<0.05	1.00
26. Illinois River Beardstown, Ill.	Carp	5	14.2	1.3	0.06	<0.05	0.34	0.07	1.20
	Smallmouth buffalo	4	13.2	1.2	0.04	0.13	0.14	<0.05	0.76
	White crappie	3	10.1	0.6	0.04	0.16	<0.10	<0.05	0.72
	White crappie (R)	4	9.6	0.5	0.04	0.23	<0.10	0.20	0.38
74. Mississippi River Little Falls, Minn.	Northern pike	3	25.9	3.9	0.20	0.18	0.22	<0.05	0.52
	Yellow bullhead	3	11.5	2.1	0.46	0.20	<0.10	<0.05	0.68
	White sucker	5	18.7	3.1	0.09	0.18	<0.10	<0.05	0.20
	White sucker (R)	5	18.5	2.9	0.09	0.17	<0.10	<0.05	ND
27. Mississippi River Guttenburg, Iowa	Carp	5	16.5	2.2	0.12	<0.05	<0.10	<0.05	0.18
	Bluegill	5	7.3	0.3	0.18	<0.05	0.14	<0.05	0.69
	Largemouth bass	5	11.6	0.9	0.08	0.07	0.36	<0.05	0.23
	Largemouth bass (R)	5	11.2	0.8	0.08	0.05	<0.10	<0.05	0.39
75. Mississippi River Cape Girardeau, Mo.	Blue catfish	4	14.1	1.4	0.07	<0.05	<0.10	<0.05	0.38
	White crappie	5	10.7	0.6	0.10	0.34	<0.10	0.26	0.44
	Carp	5	14.4	1.5	0.06	<0.05	0.10	0.20	0.38
	Carp (R)	5	14.3	1.3	0.06	<0.05	0.10	0.09	0.87
76. Mississippi River Memphis, Tenn.	Freshwater drum	4	12.2	0.7	0.20	0.11	0.80	0.80	0.48
	Channel catfish	4	15.2	1.1	0.06	<0.05	<0.70	<0.05	0.50
	Carp	3	70.2	0.4	0.05	<0.05	0.40	0.36	0.80
	Carp (R)	3	21.1	4.1	0.11	0.16	0.30	0.09	0.70
28. Arkansas River Pine Bluff, Ark.	Carp	3	20.8	5.6	0.16	0.33	0.32	0.06	0.24
	Smallmouth buffalo	3	19.4	5.1	0.14	0.31	0.31	0.06	0.44
	Channel catfish	4	18.9	2.7	0.14	0.31	0.31	<0.05	ND
	Channel catfish (R)	4	10.5	0.2	0.06	0.06	2.0	<0.05	0.32
29. Arkansas River Keystone Reservoir, Okla.	Bluegill	5	6.3	0.3	<0.05	0.10	<0.10	<0.05	1.10
	Largemouth bass	4	15.5	2.3	0.08	0.25	0.38	<0.05	1.10
	Carp	5	11.7	0.8	0.05	0.40	0.10	0.09	0.68
					(0.09)	(0.12)	(0.20)	(<0.05)	(0.51)
	Carp (R)	5	13.3	1.1	0.08	<0.05	0.20	0.48	0.44
77. Arkansas River John Martin Reservoir, Colo.	Carp	5	15.0	1.8	0.04	<0.05	0.13	<0.05	6.60
	Carp (R)	5	12.5	1.0	<0.05	<0.05	<0.10	<0.05	4.60
78. Verdigris River Oologah, Okla.	Carp	3	17.8	3.0	0.06	0.08	0.13	<0.05	0.92
					(0.09)	(0.10	(<2.20)	(<0.05)	(0.65)
	Largemouth bass	4	13.6	1.5	0.15	0.27	<0.10	<0.05	0.60
	Bluegill	5	6.1	0.2	0.06	0.06	0.38	<0.05	1.00
	Bluegill (R)	5	5.1	0.1	0.10	<0.05	0.18	<0.05	1.20
79. Canadian River Eufaula, Okla.	No samples collected								
30. White River De Valls Bluff, Ark.	Carp	3	18.5	3.4	0.14	0.08	0.80	0.05	0.28
	Bigmouth buffalo	2	19.5	4.7	0.27	0.26	0.22	0.06	0.36
	Smallmouth buffalo	3	18.1	2.8	0.16	0.28	0.80	<0.05	0.40
	Smallmouth buffalo (R)	3	16.6	2.3	0.11	0.19	0.80	<0.05	0.28
80. Yazoo River Redwood, Miss.	Freshwater drum	1	16.8	2.6	0.11	0.16	0.12	<0.05	0.80
	Carp	4	22.1	5.1	0.16	<0.05	0.10	1.30	1.00
	Smallmouth buffalo	3	19.5	4.7	0.27	0.09	0.10	0.46	0.64
	Smallmouth buffalo (R)	3	16.7	2.9	0.15	0.11	0.10	0.05	0.44

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TABLE 2 (cont'd). *Concentrations of mercury, arsenic, lead, cadmium, and selenium in whole fish, 1972 —National Pesticide Monitoring Program*

Station Number and Location	Species	No Fish	Average Size: Length, in	Average Size: Weight, lb	Residues, mg/kg wet weight: Mercury	Arsenic	Lead	Cadmium	Selenium
81 Red River Alexandria, La	Smallmouth buffalo	3	18 2	3.6	0 18	<0.05	0 28	<0 05	0.52
	Channel catfish	4	13 7	0.9	0.14	0.16	0 40	0 05	0 36
	Freshwater drum	4	13 8	1.3	0.08	0.11	0 14	<0 05	0.64
	Freshwater drum (R)	3	13 7	1 2	0 18	0.13	0 14	<0 05	0.84
82 Red River Lake Texoma, Okla	Bluegill	5	7 4	0.3	0 05	0.12	0 06	<0.05	0.13
	Largemouth bass	5	16.2	2 2	0 12	0.22	0.18	<0.05	0.62
	Carp	5	15 1	1.3	<0 01	0.25	0.10	<0.05	0.54
					(<0.05)	(0.12)	(0 20)	(<0 05)	(0 44)
	Carp (R)	5	15.9	1 7	<0 05	0.20	0.10	<0.05	0.58
83 Missouri River Hermann, Mo.	Smallmouth buffalo	4	16 8	2.7	0.06	0 11	0.60	<0 05	1 20
	Channel catfish	5	18.4	2.1	0 04	0.07	<0.10	<0.05	0.18
	Blue catfish	4	14 7	1.0	0 13	0 11	<0.10	<0.05	0.14
	Carp	4	20.3	4 0	0 07	0 08	0 5	<0.05	0.90
	Carp (R)	4	19 2	3.4	0 09	<0 05	0.5	<0.05	1.00
31 Missouri River Nebraska City, Nebr	Channel catfish	3	15 0	1.1	<0.05	0 13	0.32	<0.05	0.76
	Goldeye	5	13.6	0.9	0 14	0 13	0.78	<0.05	1.20
	Carp	5	17 9	2 9	<0 05	<0 05	0 28	<0.05	0 76
	Carp (R)	5	16 6	2.2	0 20	<0.05	0.30	0 07	1.30
32 Missouri River Garrison Dam, N Dak	Carp	3	18 1	2 6	0 02	0.20	1.3	0.36	0.38
	Walleye	5	16 9	1 6	0.13	0 15	1.1	0 12	0.80
	Goldeye	5	12 5	0.8	0.18	0 28	0.50	0 12	0.57
	Goldeye (R)	5	13 0	0 8	0.12	0.21	4 2	0.85	0.54
33 Missouri River Great Falls, Mont	Sauger	3	17 1	2.0	0.50	<0 05	<0 10	<0.05	0.54
	Redhorse	4	16.5	1 9	0.24	0 07	0.30	0.32	0 38
	Goldeye	5	12.7	0.6	0.11	0 09	0 11	<0 05	1 10
	Goldeye (R)	5	12.3	0.6	0.10	0 16	<0 10	<0.05	1 20
84 Big Horn River Hardin, Mont.	Carp	5	17 2	2.7	0.04	<0 05	<0.10	0.07	3.20
	Goldeye	2	12.5	0 6	0.16	<0 05	<0.10	<0.05	2.30
	Goldeye (R)	5	12.5	0.7	0.13	<0.05	<0.10	<0.05	3.50
85 Yellowstone River Sidney, Mont	Sauger	1	10.5	0 4	0 13	0.11	0.20	<0.05	0.61
	Carp	3	15.6	2 1	0.05	<0 05	0 20	0 30	0 68
	Goldeye	5	12 3	0.7	0 17	0 10	0.32	<0 05	0 80
	Goldeye (R)	5	11 6	0 6	0.13	0 09	0 26	<0 05	1 20
86 James River Olivet, S Dak	Carp	4	17 4	2.5	0.12	0 10	0 10	<0 05	0 35
	Channel catfish	5	16 4	1.7	0 10	<0 05	0 20	<0 05	0.35
	Goldeye	5	15 7	1 0	0.22	0 23	0 50	<0 05	0 88
	Goldeye (R)	5	13 7	1.2	0 19	<0 05	0.10	<0 05	0 64
87 North Platte River Lake McConaughy, Nebr	Walleye	5	15 6	1 5	0.21	0 09	<0 10	<0 05	1 10
	Channel catfish	5	16 6	1 6	0 23	<0.05	0 13	<0 05	0 76
	Carp	5	18 1	2 9	0 16	0 10	0 24	0 05	1 20
	Carp (R)	5	18 8	3 1	0.23	<0 05	0 22	0 05	1 40
88 South Platte River Brule, Nebr	Green sunfish	5	5 2	0 1	0 21	<0 05	0 10	<0 05	1 10
	White sucker	5	10 6	0 5	0.11	<0 05	0 20	<0 05	1 20
	Carp	5	17 0	2 4	0 34	<0 05	0 28	0 06	1 20
	Carp (R)	5	17 3	2 4	0 26	0 05	0 16	0 07	1 40
89 Platte River Louisville, Nebr	White crappie	5	5.5	0 4	0 12	0 06	<0 10	<0 05	1 20
	Channel catfish	5	15 4	1 2	0.20	0 06	0 20	<0 05	0 72
	Carp	5	15 4	1 7	0 12	0 09	0 20	<0 05	1 10
	Carp (R)	5	16 3	2 0	0.11	0 12	0 30	<0 05	1 20
90 Kansas River Bonner Springs, Kans	Carp	5	20 5	4 1	0 02	0.12	0 60	<0 05	0.96
	Gizzard shad	5	11 3	0 5	0 07	0 07	1 4	<0 05	1 30
	Gizzard shad (R)	5	9 7	0 3	0 05	0 09	0 70	<0 05	0 27
Hudson Bay Drainage									
34 Red River Noyes, Minn	Channel catfish	3	19 3	2 9	0 32	0 11	0 14	0 05	0 31
	Redhorse	4	17 6	1 9	0 49	0 11	<0 10	0 06	0 66
	Sauger	5	14 8	0 9	0 56	<0 05	<0 10	<0 05	0 36
	Sauger (R)	5	12 2	0 36	0 75	<0 05	0 40	<0 05	0 46
Colorado River System									
35 Green River Vernal, Utah	Carp	3	15 3	2 1	0 08	0 05	0 26	<0 05	1 20
	Channel catfish	5	10 8	0 3	0 20	<0 05	<0 10	<0 05	0 92
	Flannelmouth sucker	4	12 7	1 5	0 03	0 27	0 26	<0 05	0 76
	Flannelmouth sucker (R)	3	17 5	1 9	0 20	0 25	<0 10	<0 05	0 60
36 Colorado River Imperial Reservoir	Largemouth bass	4	9 6	0 5	<0 01	0 09	<0 10	<0 05	3 60
	Bluegill	5	6 1	0 2	0 01	<0 10	0 10	<0 05	2 60
	[illegible]	3	17 9	3 0	0 01	0 06	0 26	<0 05	3 20
	[illegible] (R)	3	17 7	2 9	0 01	0 10	0 16	<0 05	3 00

(*Continued next page*)

TABLE 2 (cont'd.). *Concentrations of mercury, arsenic, lead, cadmium, and selenium in whole fish, 1972 —National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size: Length (in)	Average Size: Weight (lb)	Residues, mg/kg wet weight: Mercury	Arsenic	Lead	Cadmium	Selenium
91. Colorado River Lake Havasu, Ariz.	Largemouth bass	4	11.4	1.0	0.08	0.27	0.10	<0.05	1.40
	Yellow bullhead	5	8.9	0.4	0.05	<0.05	0.10	<0.05	3.10
	Carp	5	12.5	1.0	0.05	0.10	0.46	<0.05	3.20
	Carp (R)	4	13.9	1.5	0.05	0.15	0.22	<0.05	3.60
92. Colorado River Lake Mead, Nev.	Carp	5	15.4	1.6	0.03	0.14	2.1	<0.05	2.00
	Largemouth bass	5	12.4	1.0	0.04	0.17	0.17	0.05	3.00
	Channel catfish	5	14.6	0.7	0.01	0.16	1.7	0.05	2.50
	Channel catfish (R)	5	14.5	0.6	0.05	ND	ND	ND	ND
93. Colorado River Lake Powell, Ariz.	Rainbow trout	5	17.4	2.2	0.27	0.13	0.10	0.05	3.00
	Largemouth bass	5	15.5	2.3	0.49	<0.05	<0.10	<0.05	2.20
	Carp	5	14.2	1.1	0.14	0.05	0.20	0.10	1.70
					(0.15)	(0.13)	(0.50)	(0.15)	(1.22)
	Carp (R)	5	13.4	0.9	0.14	0.05	0.20	0.22	2.20
94. Gila River San Carlos Reservoir, Ariz.	Largemouth bass	3	14.8	2.1	0.41	0.19	<0.10	0.05	0.48
	Channel catfish	3	13.5	0.7	0.10	0.08	0.10	<0.05	0.52
	Carp	3	13.0	0.9	0.30	0.09	1.0	0.14	0.36
	Carp (R)	4	11.9	0.7	0.18	0.10	0.50	0.09	0.36
					(0.14)	(0.16)	(<0.20)	0.09	0.47
Interior Basins									
37. Truckee River Fernley, Nev.	Brown bullhead	2	8.9	0.82	0.39	0.09	0.24	<0.05	0.10
	Largemouth bass	5	12.7	1.2	<0.05	0.26	<0.10	<0.05	0.20
	Carp	5	17.3	2.4	0.50	0.15	0.24	<0.05	0.31
	Carp (R)	5	13.3	1.2	0.21	<0.10	0.10	<0.05	0.29
38. Utah Lake Provo, Utah	Carp	5	17.1	1.9	0.02	ND	0.02	<0.05	0.39
	White bass	5	10.3	0.4	0.01	0.20	0.18	<0.05	0.84
	Black bullhead	5	9.7	0.5	0.01	<0.05	<0.10	<0.05	0.29
	Black bullhead (R)	5	10.7	0.8	0.01	0.08	<0.10	<0.05	0.29
95. Bear River Preston, Idaho	Carp	3	17.6	4.1	0.14	<0.05	<0.10	<0.05	0.24
	Largescale sucker	5	15.1	1.6	0.08	0.16	<0.10	<0.05	0.16
	Yellow perch	5	5.2	0.1	0.06	<0.05	<0.10	<0.05	0.40
	Yellow perch (R)	5	6.0	0.1	0.14	<0.05	<0.10	<0.05	0.30
California Streams									
39. Sacramento River Sacramento, Calif.	White catfish	5	10.2	0.5	0.02	0.17	0.10	0.60	0.28
	Largemouth bass	5	11.3	0.9	0.32	0.17	<0.10	0.88	0.24
	Carp	5	11.6	1.0	0.03	<0.05	<0.10	0.80	0.36
	Carp (R)	5	11.5	1.0	0.24	0.06	0.10	1.00	0.28
40. San Joaquin Los Banos, Calif.	Black crappie	5	11.0	0.9	0.04	<0.05	<0.10	0.13	0.31
	Sacramento blackfish	5	11.4	0.8	0.05	0.10	<0.10	0.16	0.30
	Carp	5	11.6	1.0	0.04	<0.05	<0.10	<0.05	0.37
					(0.05)	(<0.05)	(<0.20)	(<0.05)	(0.59)
	Carp (R)	5	12.0	1.2	0.03	<0.05	<0.10	<0.05	0.35
Columbia River System									
43. Salmon River Riggins, Idaho	Northern squawfish	5	9.3	0.3	0.18	0.07	<0.10	<0.05	0.27
	Smallmouth bass	5	9.6	0.6	0.21	<0.05	<0.10	<0.05	0.68
					(0.46)	(0.07)	(<0.20)	(<0.05)	(0.84)
	Largescale sucker	4	14.5	1.3	0.08	0.08	0.10	0.09	0.20
	Largescale sucker (R)	4	13.7	1.2	0.16	0.20	0.20	0.36	0.25
41. Snake River Hagerman, Idaho	Northern squawfish	4	10.6	0.6	0.02	0.14	0.22	<0.05	0.19
	Peamouth chub	5	9.2	0.3	0.26	0.80	0.10	<0.05	0.84
	Largescale sucker	5	11.3	0.7	0.21	<0.05	0.10	0.13	0.25
					(0.11)	(0.09)	(<0.20)	(<0.05)	(1.11)
	Largescale sucker (R)	5	12.1	0.9	0.06	0.05	<0.10	<0.05	0.17
42. Snake River Lewiston, Idaho	Carp	3	17.1	3.1	0.17	0.14	<0.10	0.88	0.17
	Northern squawfish	5	15.4	1.6	0.08	0.12	<0.10	<0.05	0.14
	Largescale sucker	5	14.1	1.2	0.01	0.15	0.10	0.09	0.29
	Largescale sucker (R)	5	15.0	1.4	0.03	0.18	0.10	0.26	0.20
96. Snake River Ice Harbor, Wash.	Largescale sucker	5	13.5	1.0	0.18	0.17	0.20	0.05	0.17
	Channel catfish	5	12.5	0.9	0.14	0.25	<0.10	0.05	0.07
	Northern squawfish	5	8.7	0.3	0.03	0.31	<0.10	<0.05	0.27
	Northern squawfish (R)	4	8.5	0.3	0.04	0.14	<0.10	<0.05	0.15
44. Yakima River Granger, Wash.	Carp	5	10.5	0.8	0.24	0.20	<0.10	0.42	0.40
	Northern squawfish	5	10.8	0.5	0.64	0.10	<0.10	0.05	0.40
	Largescale sucker	5	14.6	1.1	0.03	0.20	<0.10	<0.05	0.24
	Largescale sucker (R)	5	14.0	1.0	0.23	0.16	<0.10	<0.05	0.16

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TABLE 2 (cont'd.). *Concentrations of mercury, arsenic, lead, cadmium, and selenium in whole fish, 1972 —National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size: Length, in.	Average Size: Weight, lb.	Residues, mg/kg wet weight: Mercury	Arsenic	Lead	Cadmium	Selenium
45 Willamette River, Oregon City, Oreg.	Channel catfish	5	13.7	1.0	0.29	<0.05	0.10	<0.05	0.06
	Northern squawfish	5	14.3	1.2	0.04	<0.05	0.20	0.13	0.04
	Largescale sucker	5	14.6	1.4	0.24	0.14	0.10	<0.05	0.12
	Largescale sucker (R)	5	14.8	1.3	0.04	<0.05	0.10	0.02	0.09
46 Columbia River, Bonneville Dam, Oreg.	Carp	5	11.9	1.0	0.08	0.15	0.58	0.16	0.31
	Largescale sucker	3	16.0	1.7	0.23	<0.05	0.10	0.16	0.14
	Northern squawfish	5	13.5	1.0	0.06	0.11	0.30	0.42	0.11
	Carp	5	12.2	1.0	0.12	0.12	0.20	1.80	0.40
97 Columbia River, Pasco, Wash.	Northern squawfish	5	13.5	1.0	0.06	0.11	0.30	0.42	0.11
	Carp	5	12.2	1.0	0.12	0.12	0.20	1.80	0.40
	Largescale sucker	5	14.7	1.2	0.05	0.29	0.30	0.60	0.28
	Largescale sucker (R)	5	14.7	1.2	0.05	0.16	0.20	0.54	0.28
98 Columbia River, Grand Coulee, Wash.	Bridgelip sucker	5	15.3	1.4	0.02	0.18	0.90	0.28	0.24
	Northern squawfish	5	15.0	1.1	0.47	0.13	0.30	1.70	0.23
	Walleye	5	14.9	1.0	0.11	0.22	<0.10	0.16	0.34
	Walleye (R)	5	14.2	0.8	0.12	0.05	<0.10	0.16	0.30
Pacific Coast Streams									
47 Klamath River, Hornbrook, Calif.	No samples collected								
48 Rogue River, Gold Ray Dam, Oreg.	Carp	5	12.8	1.4	0.12	0.13	0.10	<0.05	0.15
	Brown bullhead	5	10.6	0.6	0.04	0.06	0.20	<0.05	0.10
	Brown bullhead (R)	5	10.4	0.6	0.18	<0.05	0.16	<0.05	0.15
Alaskan Streams									
49 Chena River, Fairbanks, Alaska	Round whitefish	3	9.7	0.5	0.06	0.08	0.10	0.05	1.20
	Arctic grayling	4	10.9	0.7	0.08	<0.05	0.20	0.20	1.30
	Longnose sucker	3	16.2	2.0	0.03	0.07	0.10	0.26	0.33
	Longnose sucker (R)	3	15.5	1.8	0.14	0.17	0.10	0.09	0.34
50 Kenai River, Soldatna, Alaska	Rainbow trout	3	9.8	0.4	0.01	<0.05	<0.10	<0.05	0.88
	Round whitefish	5	12.0	0.5	0.03	0.13	1.2	0.07	ND
	Lake trout	5	14.0	0.9	0.01	0.15	0.31	<0.05	0.60
	Lake trout (R)	5	14.6	1.0	0.18	0.16	0.40	<0.05	ND
Hawaiian Streams									
99 Waikele Stream, Waipahu, Hawaii	Tilapia	3	5.0	0.1	0.02	<0.05	4.8	0.08	1.40
	Chinese catfish	3	8.5	0.16	0.22	<0.05	1.70	0.07	1.00
	Chinese catfish (R)	3	8.3	0.15	0.46	0.11	4.60	0.07	1.30
	Cuban limia	8	3.4	ND	0.05	0.12	2.0	<0.05	0.38
100 Manoa Stream, Honolulu, Hawaii	Tilapia	5	6.3	0.26	0.04	0.10	4.2	0.14	0.51
	Chinese catfish	4	8.6	0.20	0.08	0.12	4.2	<0.05	0.35
	Chinese catfish (R)	3	9.2	0.25	0.10	<0.05	5.2	0.07	0.28
	Cuban limia	18	3.2	ND	0.03	0.20	1.4	<0.05	0.42

NOTE: R = replicate sample
ND = not detected
Numbers in parentheses are results of cross-check analyses
Redhorse = *Moxostoma* sp.

TABLE 3. *Concentrations of mercury, arsenic, lead, and cadmium in selected fish and selenium residues in all fish, 1973 —National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size: Length, in.	Average Size: Weight, lb.	Residues, mg/kg wet weight: Mercury	Arsenic	Lead	Cadmium	Selenium
Atlantic Coast Streams									
1 Stillwater River, Old Town, Maine	Yellow perch	5	8.8	0.3	0.44 (0.30)	—	—	—	0.19
	[illegible] sucker	5	14.8	1.3	0.39	—	—	—	0.14
	[illegible] sucker (R)	5	13.6	1.0	0.34	—	—	—	0.15
	[illegible] sucker	5	16.6	1.0	0.56	—	—	—	0.06
51 Kennebec River, Hinckley, Maine	[illegible] bass	5	11.4	0.8	0.80 (0.70)	—	<0.1	—	0.08
	[illegible]	5	9.6	0.4	0.93	—	<0.1	—	0.08
	[illegible]	5	10.2	0.5	1.0	—	0.16	—	0.14
	White [illegible]	5	10.8	0.5	0.21	—	<0.1	—	0.30

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TABLE 3 (cont'd.). *Concentrations of mercury, arsenic, lead, and cadmium in selected fish and selenium residues in all fish, 1973—National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size		Residues, mg/kg wet weight				
			Length, in	Weight, lb	Mercury	Arsenic	Lead	Cadmium	Selenium
52. Lake Champlain	Chain pickerel	5	11.9	1.5	0.40	—	—	—	0.12
Burlington, Vt	Pumpkinseed	5	7.6	0.4	0.18	—	—	—	0.22
	Yellow perch	5	9.5	0.4	0.17	—	—	—	0.17
53. Merrimac River	Carp	5	11.6	0.8	0.16	0.07	0.49	0.10	0.20
Lowell, Mass.	Carp (R)	5	11.5	0.8	0.09	<0.05	0.56	0.06	0.14
	Yellow perch	5	11.5	0.9	0.53	0.05	0.24	<0.05	0.12
	White sucker	5	12.2	0.7	0.23	0.05	0.60	0.06	0.17
					(0.25)	(0.05)	(0.7)	(<0.05)	
2. Connecticut River	Yellow perch	5	9.3	0.4	0.08	—	0.44	<0.05	0.31
Windsor Locks, Conn	White perch	5	10.1	0.6	0.39	—	0.43	0.08	0.83
	White perch (R)	5	9.5	0.5	0.45	—	0.53	0.05	1.13
					(0.37)	(0.08)	(0.5)	(<0.05)	
	White catfish	5	12.6	1.1	0.17	—	0.54	0.13	0.25
3. Hudson River	Largemouth bass	5	11.9	0.9	—	—	<0.1	—	0.20
Poughkeepsie, N.Y.							(0.1)		
	Goldfish	5	10.6	1.0	—	—	0.58	—	0.26
	Pumpkinseed	5	5.8	0.1	—	—	0.28	—	0.30
	Pumpkinseed (R)	5	5.6	0.1	—	—	0.10	—	0.31
54. Raritan River	Carp	5	11.5	0.9	0.02	—	0.46	—	0.44
Highland Park, N.J.	White perch	2	8.0	0.3	0.11	—	ND	—	1.3
									(1.34)
	Golden shiner	5	6.7	0.1	0.10	—	ND	—	0.23
	Golden shiner (R)	5	6.6	0.1	0.08	—	ND	—	0.26
4. Delaware River	White perch	5	9.0	0.4	—	—	0.28	—	1.5
Camden, N.J.							(1.1)		(1.23)
	White perch (R)	5	9.1	0.4	—	—	0.26	—	1.1
	White sucker	5	14.5	1.3	—	—	0.33	—	0.80
	Brown bullhead	5	9.7	0.8	—	—	0.42	—	0.43
5. Susquehanna River	White perch	5	7.6	0.2	—	—	0.73	—	2.1
Conowingo, Md.	White perch (R)	5	7.5	0.2	—	—	0.36	—	0.18
	Carp	5	14.9	1.6	—	—	0.26	—	0.64
	Channel catfish	5	16.1	1.4	—	—	0.15	—	0.30
									(0.35)
6. Potomac River	Channel catfish	4	14.4	0.9	0.14	—	0.13	—	0.21
Little Falls, Md.	Channel catfish (R)	4	11.2	0.4	<0.01	—	0.13	—	0.24
	Largemouth bass	3	7.9	0.2	0.07	—	0.25	—	0.21
	Largemouth bass (R)	3	8.2	0.2	0.13	—	0.17	—	0.13
	Redhorse	4	14.0	1.2	0.03	—	0.20	—	0.17
55. James River	Channel catfish	3	17.4	1.7	0.06	—	—	—	0.68
Richmond, Va.	Channel catfish (R)	2	19.7	2.3	0.07	—	—	—	0.44
	Largemouth bass	4	11.7	0.9	0.24	—	—	—	0.38
	Redhorse	3	15.8	1.9	0.02	—	—	—	0.44
7. Roanoke River	Carp	4	19.0	3.2	—	—	0.56	—	0.48
Roanoke Rapids, N.C.	Carp (R)	3	17.8	2.4	—	—	0.32	—	0.56
	Largemouth bass	3	9.6	0.4	—	—	0.25	—	0.26
	Channel catfish	5	12.9	0.6	—	—	0.26	—	0.16
8. Cape Fear River	Gizzard shad	3	12.7	1.0	0.12	—	—	—	0.26
Elizabethtown, N.C.	Yellow bullhead	5	8.4	0.4	0.14	—	—	—	0.13
	White catfish	5	15.6	1.6	0.24	—	—	—	0.22
	Largemouth bass	2	10.3	0.5	0.07	—	—	—	0.20
56. Pee Dee River	Largemouth bass	2	13.4	1.6	0.59	—	—	—	0.30
Dongola, S.C.	Carp	3	20.9	5.2	0.40	—	—	—	0.44
	Bluegill	5	6.7	0.2	0.27	—	—	—	0.43
	Bluegill (R)	4	6.6	0.2	0.27	—	—	—	0.33
9. Cooper River	Largemouth bass	4	13.7	1.3	—	—	—	—	0.14
Summerton, S.C.	Largemouth bass (R)	4	13.6	1.3	—	—	—	—	0.28
	Carp	2	21.3	3.1	—	—	—	—	0.16
	Bluegill	3	6.5	0.1	—	—	—	—	0.17
10. Savannah River	Carp	3	23.2	6.4	0.28	—	—	—	0.30
Savannah, Ga.	Redbreast sunfish	3	6.8	0.2	0.18	—	—	—	0.50
	Largemouth bass	3	11.6	1.0	0.73	—	—	—	0.12
	Largemouth bass (R)	3	12.7	1.1	0.73	—	—	—	0.05
					(0.08)				
57. Altamaha River	Bluegill	5	8.4	0.5	0.25	—	<0.1	—	0.14
Doctortown, Ga.	Bluegill (R)	5	8.2	0.5	0.60	—	<0.1	—	0.10
	Largemouth bass	5	12.7	1.1	0.53	—	<0.1	—	0.20
					(0.54)				
	Spotted sucker	4	16.9	2.0	0.25	—	<0.1	—	0.26

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TABLE 3 (cont'd.) *Concentrations of mercury, arsenic, lead, and cadmium in selected fish and selenium residues in all fish, 1973—National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size		Residues, mg/kg wet weight				
			Length, in	Weight, lb	Mercury	Arsenic	Lead	Cadmium	Selenium
11 St. Johns River Wallace, Fla.	Striped mullet	3	17.6	2.5	—	0.28	—	—	0.42
	Striped mullet (R)	3	16.2	1.7	—	0.27	—	—	0.39
	Largemouth bass	3	9.2	0.4	—	0.16	—	—	0.24
	Channel catfish	4	10.7	0.4	—	0.13	—	—	0.19
12 St. Lucie Canal Indiantown, Fla.	Channel catfish	2	16.2	1.6	—	—	—	—	0.24
	Largemouth bass	3	10.6	0.5	—	—	—	—	0.26
	Largemouth bass (R)	3	13.2	1.0	—	—	—	—	0.34
	Bluegill	4	8.5	0.4	—	—	—	—	0.37
			Gulf Coast Streams						
58 Suwanee River Old Town, Fla.	Largemouth bass	5	10.7	0.6	0.35 (0.67)	—	—	—	0.36 (0.22)
	Redbreast sunfish	4	7.2	0.3	0.10	—	—	—	0.42
	Spotted sucker	3	14.5	1.6	0.10	—	—	—	0.60
	Spotted sucker (R)	3	14.6	1.6	0.20	—	—	—	2.12
13 Apalachicola River Jim Woodruff Dam, Fla.	Spotted sucker	3	19.3	2.7	—	—	—	—	0.38
	Channel catfish	3	11.0	0.4	—	—	—	—	0.42
	Largemouth bass	3	13.5	1.3	—	—	—	—	0.24
	Largemouth bass (R)	3	15.0	1.9	—	—	—	—	0.12
59 Alabama River Chrysler, Ala.	Largemouth bass	5	14.7	1.5	0.11	—	—	—	0.14
	Striped mullet	5	15.9	1.4	0.13	—	—	—	0.28
	Bluegill	5	7.5	0.2	0.09	—	—	—	0.28
	Bluegill (R)	5	7.1	0.2	0.10	—	—	—	0.14
14 Tombigbee River McIntosh, Ala.	Striped mullet	5	15.5	1.6	0.09	0.75	—	—	0.40
	Carp	3	21.0	4.6	0.07	0.29	—	—	0.44
	Largemouth bass	5	12.6	1.0	0.25	0.15	—	—	0.24
	Largemouth bass (R)	5	12.0	0.7	0.65 (0.66)	0.15	—	—	0.16
15 Mississippi River Luling, La.	Freshwater drum	4	13.4	1.3	—	—	0.19	0.12	0.42
	Blue catfish	5	14.5	1.1	—	—	< 0.1	< 0.05	0.21
	Blue catfish (R)	5	14.5	1.1	—	—	0.12	0.21	0.12 (0.23)
60 Brazos River Richmond, Tex.	Longnose gar	5	23.8	0.9	0.03	—	0.85	—	0.33
	Longnose gar (R)	5	22.2	0.8	0.02	—	< 0.1	—	0.56
	Blue catfish	4	16.1	1.4	0.11	—	0.32	—	0.74
	River carpsucker	4	10.0	0.4	0.13	—	0.11	—	0.42
61 Colorado River Wharton, Tex.	Channel catfish	5	12.1	0.6	0.05	< 0.05	0.40	—	0.65
	Channel catfish (R)	5	13.0	0.7	0.05	0.08	0.36	—	1.0
	Spotted bass	1	12.0	1.0	0.03	0.07	< 0.1	—	0.05
	River carpsucker	2	15.8	1.6	0.03	0.05 (0.09)	0.32 (0.6)	—	0.28
	Carp	2	20.1	4.1	0.05	< 0.05	0.19	—	< 0.05
62 Nueces River Mathis, Tex.	Smallmouth buffalo	5	15.7	2.4	—	—	—	—	0.18
	Smallmouth buffalo (R)	5	14.1	2.0	—	—	—	—	0.18
	Blue catfish (R)	5	17.7	1.9	—	—	—	—	0.14
	White crappie	5	9.4	0.5	—	—	—	—	0.20
	Gizzard shad	5	10.5	0.4	—	—	—	—	0.40 (0.56)
	Largemouth bass	5	13.2	1.3	—	—	—	—	0.16
63 Rio Grande Rio Grande City, Tex.	Channel catfish	4	13.4	0.7	—	—	0.13	—	0.20
	Channel catfish (R)	4	12.1	0.4	—	—	0.13	—	0.16
	Gizzard shad	2	10.0	0.3	—	—	0.23	—	0.25
	Channel catfish	5	12.0	0.5	—	—	0.18	—	0.16
64 Rio Grande Ballez, N. Mex.	Channel catfish	5	11.9	0.4	0.10	0.08	—	—	0.08
	Channel catfish (R)	5	12.8	0.6	0.06	< 0.05	—	—	0.13
	Carp	5	14.3	1.2	0.11	0.05	—	—	0.23
	Largemouth bass	5	12.5	1.3	0.05	0.17	—	—	0.20
65 [illegible]	White sucker	5	12.9	0.9	—	—	0.25	—	0.07
	White sucker (R)	5	12.3	0.7	—	—	0.60	—	0.08
	Brown bullhead	3	13.0	1.0	—	—	0.10	—	0.08
67 Pe[illegible] Ro[illegible]	Gizzard shad	5	8.2	0.1	—	—	—	—	0.95
	Gizzard shad (R)	5	9.3	0.3	—	—	—	—	0.85
	Channel catfish	5	13.5	0.8	—	—	—	—	1.3
	Smallmouth buffalo	5	14.4	1.4	—	—	—	—	1.0 (0.80)
[illegible]		5	16.1	1.4	0.23	—	—	—	0.29
	R	5	17.0	1.6	0.08				0.39
		5	15.4	1.5	0.21				0.49
		5	7.6	0.4	0.21				0.27

(Continued on next page)

TABLE 3 (cont'd.). *Concentrations of mercury, arsenic, lead, and cadmium in selected fish and selenium residues in all fish, 1973—National Pesticide Monitoring Program*

Station Number and Location	Species	No Fish	Average Size		Residues, mg/kg wet weight				
			Length, in	Weight, lb	Mercury	Arsenic	Lead	Cadmium	Selenium
66. St Lawrence River Massena, N Y	Northern pike	3	21 4	2 3	0 25	—	—	—	0 34 (0.36)
	Yellow perch	5	7 0	0 2	0 09	—	—	—	0 33
	Yellow perch (R)	5	7 2	0 2	0.10	—	—	—	0 30
	White sucker	5	14 8	1 4	0.11	—	—	—	0 33
18. Lake Ontario Port Ontario, N Y	White perch	5	9 0	0 5	0 18 (0 16)	0 24	0.34	—	0 56 (0.92)
	Rock bass	5	8 3	0 5	0 39	0 20	0 46	—	0.23
	Yellow perch	5	9 9	0 5	0 24	0 18	0 23	—	0 17
	Yellow perch (R)	5	9 6	0.5	0 21	0 11	<0 1	—	0 29
19 Lake Erie Erie, Pa	Yellow perch	5	8 7	0 4	—	—	—	—	0 64
	Yellow perch (R)	5	9 2	0 4	—	—	—	—	0 44
20. Lake Huron Bay Port, Mich	Carp	5	15 2	1 8	—	0 18	—	—	0 64
	Yellow perch	5	9 5	0 4	—	0 16	—	—	0 64
	Yellow perch (R,	5	9 1	0 4	—	0 09	—	—	0 86
	Channel catfish	3	15 6	1 3	—	0 10	—	—	0.80
21. Lake Michigan Sheboygan, Wis	Bloater	5	10 8	0 4	0 04	0 42 (0 81)	—	—	0 43
	Bloater	5	11 0	0 4	0 07	1 24	—	—	0.44
	Yellow perch	5	9 2	0 3	0 03	0 16	—	—	0 64
	Yellow perch (R)	5	8 8	0 3	0 11	< 0 05	—	—	0.21
	Lake trout	5	21 4	3 2	0.11	0 65	—	—	0 80
	Lake trout (R)	5	21 7	3 3	0.10	0 68	—	—	0 57
22. Lake Superior Bayfield, Wis	Bloater	5	10 3	0 3	0 09	0 60	<0 1	—	0.54
	Bloater (R)	5	10 6	0 3	0 09	0 68	<0 1	—	0 20
	Lake whitefish	4	21 2	3 3	0 08	0 56	0 18	—	2.4
	Lake trout	5	26 1	5 9	0 26	0 27	<0 1	—	0.26
	Lake trout (R)	5	27 0	6 5	0 40	0 43	< 0 1	—	0 16
Mississippi River System									
67. Allegheny River Natrona, Pa	Smallmouth bass	5	10 8	0 8	0 19	—	0 32	—	0 33
	Walleye	5	15 3	1 2	0 30	—	0 11	—	0.24
	Carp	5	16 8	2 3	0 08	—	0 30	—	0 23
	Carp (R)	5	17 2	2 5	0 09	—	0 26	—	0 44
23 Kanawha River Winfield, W Va	Brown bullhead	3	11 9	0 8	—	—	—	—	0 13
	Carp	5	11 4	0 7	—	—	—	—	0 36
	Pumpkinseed	5	7 6	0 2	—	—	—	—	0 30
	Pumpkinseed (R)	5	9 0	0 4	—	—	—	—	0.20
68 Wabash River New Harmony, Ind	Channel catfish	5	14 6	0 9	0 11	—	0 10	<0 05	0 36
	Carp	5	17 5	2 9	0 22	—	0 16 (0 10)	<0 05	0 36
	Carp (R)	5	18 9	3 6	0 11	—	0 23	0 13	0 38
	White crappie	5	9 2	0 4	0 28	—	0 22 (0 2)	<0 05	0 21
24 Ohio River Marietta, Ohio	Spotted sucker	5	12 4	0 7	ND	—	<0 1	—	0 50
	Channel catfish	5	15 0	1 0	0 16	—	<0 1	—	0 30
	Channel catfish (R)	5	14 3	0 9	0 22	—	<0 1	—	0.43
	Carp	5	14 9	1 8	0 10	—	<0 1	—	0 41
69 Ohio River Cincinnati, Ohio	Carp	5	16 4	2 2	—	0 10	—	0 17	0.14
	Channel catfish	5	16 9	1 5	—	0 05	—	0 09	0 12
	Channel catfish (R)	5	16 9	1 5	—	0 05	—	0 05	0 23
	Sauger	5	10 4	0 4	—	0 10	—	<0 05	0.21
70 Ohio River Metropolis, Ill	Carp	5	17 6	2 6	—	0 15	—	—	0 36
	White crappie	5	10 9	0 6	—	0 07	—	—	0 23
	White crappie (R)	5	10 0	0 5	—	0 20	—	—	0.14
	Channel catfish	5	14 6	1 1	—	0 05	—	—	0 23
25 Cumberland River Clarksville, Tenn	Largemouth bass	5	15 3	2 3	0 40	—	0 28	—	0 50
	Bluegill	5	6 0	0 1	0 08	—	< 0 1	—	0.41
	Bluegill (R)	5	5 8	0 1	0 10	—	<0 1	—	0 34
	Carp	5	15 1	1 7	0 09	—	0 20	—	0 41
71. Tennessee River Savannah, Tenn	Carp	3	17 1	2 6	0 26	< 0 05	<0 1	—	0 54
	Carp (R)	3	17 9	3 0	0 38	<0 05	0 20	—	0 46
	Bluegill	4	5 5	0 1	0 29	<0 05	<0 1	—	0 38
	Channel catfish	1	16 3	2 1	0 45	0 07	0 12	—	0 36
	Largemouth bass	2	11 1	0 8	0 16 (0 48)	0 10	< 0 1	—	0 25
72 Wisconsin River Woodman, Wis	Smallmouth bass	5	12 1	1 0	0 13	0 07	—	—	0 46
	Carp	5	21 3	4 6	0 25	0 06	—	—	0 64
	Carp (R)	5	20 0	4 0	0 22	0 08	—	—	0 47
	Sauger	5	14 3	0.7	0 30	< 0 05	—	—	0.18

(*Continued next page*)

TABLE 3 (cont'd.) *Concentrations of mercury, arsenic, lead, and cadmium in selected fish and selenium residues in all fish, 1973—National Pesticide Monitoring Program*

Station Number and Locations	Species	No Fish	Average Size		Residues, mg/kg wet weight				
			Length, in	Weight, lb	Mercury	Arsenic	Lead	Cadmium	Selenium
73 Des Moines River Keosauqua, Iowa	Carp	5	11.8	0.8	—	—	0.19	—	0.36
	Carp (R)	5	11.8	0.8	—	—	0.44	—	0.30
	Channel catfish	4	10.7	0.4	—	—	0.70	—	0.24
	Walleye	7	7.4	0.1	—	—	0.10	—	0.20
26 Illinois River Beardstown, Ill	Black crappie	4	5.6	0.1	—	0.11	—	—	0.70
	Freshwater drum	5	9.4	0.4	—	0.15	—	—	0.33
	Carp	5	12.2	1.0	—	0.15	—	—	0.64
							(0.1)		(0.44)
	Carp (R)	5	12.6	1.1	—	0.17	—	—	0.39
74 Mississippi River Little Falls, Minn	White sucker	1	17.0	2.1	0.22	0.10	—	—	0.11
	Yellow bullhead	2	9.9	0.6	0.50	<0.05	—	—	0.16
	Walleye	2	10.0	0.4	0.15	<0.05	—	—	0.16
27 Mississippi River Guttenburg, Iowa	Carp	5	17.8	2.7	—	—	—	—	0.18
	Largemouth bass	5	12.2	1.2	—	—	—	—	0.30
	Bluegill	5	7.3	0.3	—	—	—	—	0.54
	Bluegill (R)	5	4.7	0.1	—	—	—	—	0.85
75 Mississippi River Cape Girardeau, Mo	Carp	5	14.7	1.7	0.04	—	—	—	0.62
	Carp (R)	5	12.4	1.1	0.08	—	—	—	0.60
	White crappie	5	9.0	0.4	0.08	—	—	—	0.35
	Channel catfish	5	12.3	0.7	0.10	—	—	—	0.73
76 Mississippi River Memphis, Tenn	Flathead catfish	3	15.1	1.6	—	0.07	0.10	<0.05	0.36
	Carp	3	19.7	4.1	—	0.07	0.30	<0.05	0.60
	Freshwater drum	ND	14.9	1.8	—	0.27	0.36	<0.05	0.77
	Freshwater drum (R)	ND	10.5	0.8	—	0.20	0.40	<0.05	0.04
							(1.0)	(<0.05)	(0.60)
28 Arkansas River Pine Bluff, Ark	Smallmouth buffalo	3	17.6	3.2	—	0.08	—	—	0.29
	Smallmouth buffalo (R)	3	19.2	4.1	—	0.15	—	—	0.18
	Flathead catfish	3	22.9	5.6	—	0.12	—	—	0.17
	Carp	4	22.9	6.2	—	<0.05	—	—	0.52
29 Arkansas River Keystone Reservoir, Okla	Carp	5	12.4	0.9	0.03	<0.05	—	<0.05	0.19
	Largemouth bass	5	9.5	0.5	0.05	ND	—	<0.05	0.32
	Bluegill	5	5.6	0.1	0.04	<0.05	—	<0.05	0.29
	Bluegill (R)	5	5.6	0.1	0.04	<0.05	—	<0.05	0.24
77 Arkansas River John Martin Reservoir, Colo	Carp	5	14.2	1.4	—	—	—	—	1.3
									(3.28)
	Carp (R)	5	14.4	1.5	—	—	—	—	1.4
	Channel catfish	5	14.9	1.0	—	—	—	—	0.85
	Black bullhead	2	9.8	0.5	—	—	—	—	0.24
78 Verdigris River Oologah, Okla	Carp	5	16.7	2.4	—	—	0.26	—	0.68
	Largemouth bass	5	10.2	0.5	—	—	0.26	—	0.32
	Bluegill	5	6.0	0.1	—	—	0.24	—	0.33
	Bluegill (R)	5	5.9	0.1	—	—	0.19	—	0.31
79 Canadian River Eufaula, Okla	Bluegill	5	5.4	0.1	—	—	—	—	0.12
	Bluegill (R)	5	5.0	0.1	—	—	—	—	0.16
	Largemouth bass	5	11.0	0.7	—	—	—	—	0.14
	Carp	5	13.7	1.1	—	—	—	—	0.15
30 White River DeValls Bluff, Ark	Channel catfish	3	15.2	1.2	—	<0.05	—	—	0.44
	Channel catfish (R)	3	15.3	1.2	—	0.06	—	—	0.40
	Smallmouth buffalo	4	15.8	2.8	—	<0.05	—	—	0.40
	Carp	4	20.2	4.0	—	<0.05	—	—	0.53
81 Yazoo River Redwood, Miss	Channel catfish	3	18.7	1.9	—	—	—	<0.05	0.22
	Smallmouth buffalo	3	17.3	2.6	—	—	—	ND	ND
	Smallmouth buffalo (R)	3	17.8	2.9	—	—	—	<0.05	0.36
	Carp	3	22.7	6.1	—	—	—	0.09	0.48
								(<0.05)	(0.67)
81 Red River Alexandria, La	Smallmouth buffalo	1	17.0	3.9	—	—	—	—	0.20
	Blue catfish	5	12.3	0.5	—	—	—	—	0.14
	Freshwater drum	5	13.1	1.0	—	—	—	—	0.22
	Freshwater drum (R)	5	13.0	1.0	—	—	—	—	0.26
	Carp	1	21.7	3.8	—	—	—	—	0.24
82 Red River Lake [illegible], Ark	Bluegill	5	5.3	0.1	—	0.10	—	—	0.13
	Bluegill (R)	5	5.3	0.1	—	0.08	—	—	0.15
	Carp	4	19.2	3.2	—	0.28	—	—	0.16
	Largemouth bass	5	11.2	0.7	—	0.65	—	—	0.17
83 Missouri River Hermann, Mo	Carp	3	19.4	4.0	—	—	—	—	0.72
	Carp (R)	3	21.6	4.7	—	—	—	—	0.60
	[illegible] drum	5	16.8	3.0	—	—	—	—	0.68
	[illegible] buffalo	4	18.5	3.6	—	—	—	—	0.38

(Continued next page)

TABLE 3 (cont'd.). *Concentrations of mercury, arsenic, lead, and cadmium in selected fish and selenium residues in all fish, 1973—National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size		Residues, mg/kg wet weight				
			Length, in	Weight, lb	Mercury	Arsenic	Lead	Cadmium	Selenium
31. Missouri River	Carp	5	13.3	1.4	—	—	—	—	0.64
Nebraska City, Nebr.	Carp (R)	5	13.6	1.5	—	—	—	—	1.0
	Goldeye	4	14.3	1.1	—	—	—	—	0.67
							(0.4)		(1.16)
	White crappie	4	9.7	0.6	—	—	—	—	0.71
32. Missouri River	Walleye	5	16.3	1.4	—	0.05	—	<0.05	0.32
Garrison Dam, N. Dak.	Goldeye	5	12.8	0.8	—	0.05	—	<0.05	0.22
	Goldeye (R)	5	12.6	0.8	—	<0.05	—	<0.05	0.18
	Carp	5	15.2	1.7	—	0.07	—	<0.05	0.18
33. Missouri River	Longnose sucker	3	15.6	1.7	0.03	—	0.38	—	0.34
Great Falls, Mont.	Goldeye	5	12.5	0.6	0.08	—	0.29	—	1.4
	Goldeye (R)	3	12.8	0.7	0.02	—	0.26	—	1.5
84. Big Horn River	Carp	4	16.9	2.7	—	—	—	—	1.0
Hardin, Mont.	Channel catfish	3	22.5	4.5	—	—	—	—	1.0
	Goldeye	5	12.6	0.7	—	—	—	—	1.6
	Goldeye (R)	5	12.8	0.7	—	—	—	—	1.6
									(2.59)
85. Yellowstone River	Carp	2	12.7	0.9	—	<0.05	0.17	—	0.48
Sidney, Mont.	Goldeye	5	11.3	0.5	—	0.20	0.10	—	0.72
	Goldeye (R)	5	10.7	0.4	—	0.20	0.28	—	0.72
	Channel catfish	5	16.7	1.4	—	0.05	<0.1	—	0.64
86. James River	Channel catfish	5	14.8	0.8	—	—	<0.10	—	0.24
Olivet, S. Dak.	Goldeye	5	13.6	1.0	—	—	<0.10	—	0.60
	Goldeye (R)	5	13.9	0.9	—	—	<0.10	—	0.60
	Carp	1	10.6	0.7	—	—	<0.10	—	0.36
87. North Platte River	Carp	5	18.1	2.8	—	<0.05	—	—	0.72
Lake McConaughy, Nebr.	Carp (R)	5	17.2	2.3	—	0.26	—	—	0.72
	Walleye	5	16.3	1.2	—	0.24	—	—	0.67
	Channel catfish	5	15.0	1.3	—	0.12	—	—	0.67
88. South Platte River	White sucker	8	7.5	0.3	0.07	—	—	—	0.16
Brule, Nebr.	Green sunfish	13	3.3	<0.1	0.05	—	—	—	0.33
	Carp	5	14.0	1.5	0.06	—	—	—	0.71
	Carp (R)	5	13.3	1.4	0.05	—	—	—	1.4
89. Platte River	Carp	5	13.5	1.5	—	—	—	—	0.88
Louisville, Nebr.	Carp (R)	5	12.9	1.2	—	—	—	—	0.84
	Channel catfish	4	12.5	0.7	—	—	—	—	0.92
	White crappie	4	8.6	0.4	—	—	—	—	0.80
90. Kansas River	Carp	5	16.1	2.3	—	—	—	—	0.80
Bonner Springs, Kans.	Carp (R)	5	16.5	2.3	—	—	—	—	0.72
	Freshwater drum	5	11.4	0.6	—	—	—	—	0.92
	Gizzard shad	5	8.6	0.2	—	—	—	—	0.60
Hudson Bay Drainage									
34. Red River	Goldeye	3	12.6	0.7	0.09	—	<0.10	—	1.7
Noyes, Minn.	Channel catfish	2	16.0	1.5	0.07	—	<0.10	—	0.56
	Sauger	5	12.0	0.5	0.24	—	<0.10	—	0.28
	Sauger (R)	5	11.8	0.5	0.30	—	<0.10	—	0.23
					(0.38)		(0.31		
Colorado River System									
35. Green River	Channel catfish	5	11.6	0.5	—	<0.05	—	—	0.64
Vernal, Utah	Channel catfish (R)	5	11.9	0.4	—	<0.05	—	—	0.73
	Carp	5	15.9	2.2	—	<0.05	—	—	0.56
	Flannelmouth sucker	4	17.8	1.9	—	0.08	—	—	0.49
36. Colorado River	Largemouth bass	4	11.3	0.9	—	—	—	—	2.1
Imperial Reservoir, Ariz.									(2.56)
	Carp	3	18.1	2.8	—	—	—	—	3.2
									(2.32)
	Bluegill	3	5.8	0.2	—	—	—	—	2.5
	Bluegill (R)	3	5.9	0.2	—	—	—	—	0.44
91. Colorado River	Carp	3	17.4	3.1	—	0.05	—	—	3.0
Lake Havasu, Ariz.							(0.3)		(2.91)
	Black crappie	1	12.9	1.3	—	0.19	—	—	2.1
	Largemouth bass	4	9.8	0.5	—	0.10	—	—	1.4

(*Continued next page*)

TABLE 3 (cont'd.) *Concentrations of mercury, arsenic, lead, and cadmium in selected fish and selenium residues in all fish, 1973—National Pesticide Monitoring Program*

Station Number and Location	Species	No. Fish	Average Size: Length (in)	Average Size: Weight (lb)	Residues, mg/kg wet weight: Mercury	Arsenic	Lead	Cadmium	Selenium
92 Colorado River, Lake Mead, Nev.	Largemouth bass	5	12.6	1.1	—	—	0.22	—	1.4
	Largemouth bass (R)	5	12.0	1.0	—	—	0.12	—	1.8
	Channel catfish	5	15.9	1.2	—	—	0.18	—	1.4 (0.31)
	Carp	5	16.8	2.4	—	—	0.20	—	1.6 (1.26)
93 Colorado River, Lake Powell, Ariz.	Largemouth bass	4	14.7	2.3	0.16 (0.20)	0.39	—	—	1.0 (1.39)
	Carp	4	13.6	1.1	0.05	0.09	—	—	0.36
	Carp (R)	4	13.8	1.1	0.13	0.09	—	—	1.3
	Rainbow trout	5	16.7	2.3	0.09	0.26	—	—	2.3
94 Gila River, San Carlos Reservoir, Ariz.	Largemouth bass	5	12.9	1.3	—	< 0.05	—	—	0.56
	Largemouth bass (R)	5	12.6	1.1	—	0.21	—	—	0.23
	Carp	3	12.8	1.0	—	< 0.05	—	—	0.56
	Bluegill	5	6.2	0.2	—	0.15	—	—	0.64
Interior Basins									
37 Truckee River, Fernley, Nev.	Carp	4	15.2	1.7	0.22 (< 0.05)	—	0.42 (< 0.10)	—	0.26 (1.17)
	Carp (R)	4	17.6	2.6	0.24	—	0.33	—	0.28
	Largemouth bass	3	12.6	1.3	0.49	—	0.32	—	0.13
	Brown bullhead	5	10.3	0.7	0.17	—	0.40	—	0.13
38 Utah Lake, Provo, Utah	Carp	5	17.9	2.6	—	0.13	—	—	0.19
	White bass	5	10.4	0.4	—	0.16	—	—	0.32
	Black bullhead	5	11.1	0.7	—	< 0.05	—	—	0.22
	Black bullhead (R)	5	11.3	0.7	—	< 0.05	—	—	0.18
95 Beat River, Preston, Idaho	Largescale sucker	5	18.6	2.6	—	—	—	—	0.34
	Carp	4	14.1	1.6	—	—	—	—	0.26
	Yellow perch	5	8.9	0.3	—	—	—	—	0.34
	Yellow perch (R)	5	7.9	0.2	—	—	—	—	0.34
California Streams									
39 Sacramento River, Sacramento, Calif.	White catfish	1	11.8	0.6	0.08	< 0.05	0.42	< 0.05	0.18
	Carp	5	18.8	2.9	0.03 (0.21)	0.50	0.46	< 0.05 (0.09)	0.33
	Carp (R)	2	20.3	4.2	0.08	0.40	0.32	< 0.05	0.76
40 San Joaquin River, Los Banos, Calif.	Channel catfish	5	10.4	0.3	—	0.05	—	< 0.05	0.18
	Channel catfish (R)	5	12.7	0.6	—	0.07	—	< 0.05	0.27
	Carp	5	13.7	1.2	—	< 0.05	—	< 0.05	0.28
	White catfish	5	12.0	0.7	—	0.05	—	< 0.05	0.20
Columbia River System									
43 Salmon River, Riggins, Idaho	Smallmouth bass	5	13.2	1.4	0.22	0.05	—	—	0.83
	Largescale sucker	5	17.5	1.8	0.15	0.20	—	—	0.32
	Brown bullhead	5	7.5	0.2	0.13	0.08	—	—	0.56
	Brown bullhead (R)	5	8.3	0.3	0.10	0.05	—	—	0.53
41 Snake River, Hagerman, Idaho	Largescale sucker	5	15.9	1.7	0.03	—	—	—	0.27
	Largescale sucker (R)	5	16.1	1.5	0.07	—	—	—	0.20
	Northern squawfish	5	15.4	1.4	0.03	—	—	—	0.30
	Rainbow trout	5	14.8	1.5	0.03	—	—	—	0.22
42 Snake River, Lewistown, Idaho	Black crappie	5	9.6	0.7	0.13	0.06	—	< 0.05	0.20
	Black crappie (R)	5	9.8	0.6	0.10	0.05	—	< 0.05	0.24
	Largescale sucker	5	17.2	1.9	0.04	0.20	—	< 0.05	0.16
	Carp	5	17.3	2.7	0.06	0.18	—	0.06 (0.05)	0.30
186 Snake River, Ice Harbor, Wash.	Largescale sucker	5	14.0	1.0	0.05	0.12	—	—	0.37
	Largescale sucker (R)	5	14.6	1.0	0.07	0.13	—	—	0.38
	Channel catfish	5	16.3	1.4	0.08	0.05	—	—	0.18
	Northern squawfish	5	16.2	1.6	0.65	0.05	—	—	0.18
44 Yakima River, Granger, Wash.	Largescale sucker	5	15.3	1.3	0.04	—	—	—	0.20
	Largescale sucker (R)	5	14.4	1.2	0.03	—	—	—	0.13
	Largemouth bass	3	13.8	1.7	0.18	—	—	—	0.10
	Carp	5	13.4	1.2	0.07	—	—	—	0.22
45 Willamette River, Oregon City, Oreg.	Largescale sucker	5	16.1	1.5	0.08			—	0.09
	Largescale sucker (R)	5	16.7	1.6	0.20			—	0.05
	Northern squawfish	5	16.0	1.6	0.65			—	< 0.05
	Carp	5	14.3	1.3	0.15	—	—	—	0.18

(Continued next page)

TABLE 3 (cont'd.). *Concentrations of mercury, arsenic, lead, and cadmium in selected fish and selenium residues in all fish, 1973—National Pesticide Monitoring Program*

Station Number and Location		Species	No. Fish	Average Size		Residues, mg/kg wet weight				
				Length, in	Weight, lb	Mercury	Arsenic	Lead	Cadmium	Selenium
46	Columbia River Bonneville Dam, Oreg	Largescale sucker	5	17.7	1.9	0.32	<0.05	0.32	0.13	0.13
		Largescale sucker (R)	5	17.6	1.8	0.32	0.08	0.39	0.05	0.06
		Northern squawfish	5	11.9	0.6	0.85	<0.05	0.24	<0.05	0.20
		Carp	5	13.7	1.1	0.06	0.08	0.24	<0.05	0.20
97.	Columbia River Pasco, Wash	Carp	5	11.7	0.8	—	0.18	0.15	<0.05 (<0.05)	0.93
		Carp (R)	5	12.2	0.8	—	0.18	<0.10	<0.05	0.48
		Largescale sucker	5	14.2	1.1	—	0.13	<0.10	<0.05	0.06
		Northern squawfish	5	13.6	0.9	—	0.05	<0.10	<0.05	0.24
98	Columbia River Grand Coulee, Wash	Northern squawfish	5	13.0	0.9	0.16 (0.21)	<0.05	0.30	0.14 (0.11)	0.14
		Bridgelip sucker	5	14.3	1.1	0.03	0.27	0.53	0.08	0.20
		Bridgelip sucker (R)	5	13.7	0.9	0.08	0.18	1.0	0.07	0.26
		Walleye	5	14.4	1.1	0.15	0.05	0.22	<0.05	0.26
		Pacific Coast Streams								
47	Klamath River Hornbrook, Calif	Klamath sucker	5	14.9	1.6	0.04	—	—	0.06	0.32
		Yellow perch	5	8.1	0.3	0.05	—	—	<0.05	0.10
		Yellow perch (R)	5	8.0	0.2	0.04	—	—	<0.05	0.13
		Largemouth bass	5	7.7	0.3	0.10	—	—	<0.05	0.10
48	Rogue River Gold Ray Dam, Oreg	Carp	4	11.9	1.4	0.02	—	—	—	0.20
		Brown bullhead	5	9.4	0.4	0.12	—	—	—	0.13
		Brown bullhead (R)	5	9.0	0.4	0.02	—	—	—	0.13
		Black crappie	5	10.3	0.7	0.15	—	—	—	0.12
49	Chena River Fairbanks, Alaska	Longnose sucker	4	14.3	1.3	—	—	—	<0.05	0.58
		Round whitefish	3	11.1	0.4	—	—	—	<0.05	1.4
		Round whitefish (R)	3	11.3	0.4	—	—	—	<0.05	1.2
		Arctic grayling	5	11.6	0.6	—	—	—	<0.05	1.2 (0.32)
50.	Kenai River Soldatna, Alaska	Round whitefish	5	10.8	0.4	—	<0.05	—	—	1.3
		Rainbow trout	5	13.4	1.1	—	0.11	—	—	0.64
		Lake trout	5	15.2	1.0	—	0.08	—	—	0.52
		Lake trout (R)	5	14.9	0.9	—	0.07	—	—	0.56
		Hawaiian Streams								
99	Waikele Stream Waipahu, Hawaii	Tilapia	2	5.8	0.1	—	0.11	<0.1 (0.4)	—	0.92 (1.35)
		Cuban limia	21	3.0	<0.1	—	<0.05	<0.1	—	1.0
		Chinese catfish	2	8.9	0.2	—	<0.05	0.34	—	1.0
		Chinese catfish (R)	2	8.4	0.2	—	<0.05	0.32 (0.8)	—	0.93 (1.22)
100	Manoa Stream Honolulu, Hawaii	Chinese catfish	2	9.3	0.3	—	0.06	0.70	—	0.58
		Chinese catfish (R)	2	10.0	0.3	—	0.09	0.93 (0.8)	—	0.54
		Tilapia	2	9.8	0.5	—	0.19	0.63	—	0.25
		Cuban limia	26	2.7	0.01	—	0.15	1.4	—	0.29

NOTE: — = residues not determined
R = replicate sample
ND = not detected
Numbers in parentheses are results of cross-check analyses

[1]Redhorse = *Moxostoma* sp

TABLE 4. *Summary of mercury, arsenic, lead, cadmium, and selenium residues in fish, 1971-73—National Pesticide Monitoring Program*

Year	No. Stations Represented	Total Composites Analyzed	Number of Composites							
			With Residues		Less than Detectable Levels[1]		From Detectable Levels to 0.5 mg/kg		>0.5 mg/kg	
			No.	%	No.	%	No.	%	No.	%
Mercury										
1971	100	584	576	99	1	<1	534	93	41	7
1972	98	391	391	100	2	<1	373	95	16	4
1973	47	196	195	100	1	<1	179	92	15	8
Arsenic										
1971	100	584	559	96	210	38	337	60	12	2
1972	98	391	388	99	98	25	291	74	2	1
1973	39	171	171	100	47	27	116	68	8	5
Lead										
1971	100	584	327	56	44	13	261	80	22	7
1972	98	391	390	100	118	30	198	51	73	19
1973	36	161	161	100	42	26	102	63	17	11
Cadmium										
1971	100	584	443	76	329	74	113	26	1	0
1972	98	391	390	99	273	70	100	26	17	4
1973	17	75	75	100	53	71	22	29	0	0
Selenium										
1972	98	391	386	99	0	0	217	56	169	44
1973	100	444	444	100	3	1	308	69	133	30

[1]Detectable levels of mercury, arsenic, lead, cadmium, and selenium were 0.01, 0.05, 0.10, 0.05, and 0.05 mg/kg, respectively.

Nationwide Residues of Mercury, Lead, Cadmium, Arsenic, and Selenium in Starlings, 1973

Donald H. White,[1] Jerry R. Bean,[2] and Jerry R. Longcore [3]

ABSTRACT

Starlings (Sturnus vulgaris) *collected in 1973 at 51 sites throughout the continental United States were analyzed for mercury, lead, cadmium, arsenic, and selenium. All samples contained detectable levels of these elements. In general, residues were low: mercury residues ranged from <0.01 to 0.20 ppm; lead, from <0.10 to 3.20 ppm; cadmium, from <0.05 to 0.20 ppm; arsenic, from <0.05 to 1.40 ppm; and selenium, from 0.10 to 1.10 ppm. There was a significant overall decline in mercury and lead residues in starlings since 1971, and a significant increase in arsenic residues. Lead residues were significantly higher in starlings from urban areas than from rural areas.*

Introduction

The Fish and Wildlife Service, U.S. Department of Interior, began nationwide monitoring of organochlorines in starlings (*Sturnus vulgaris*) in 1967-68 as part of the National Pesticide Monitoring Program. The starling monitoring program was modified to include analyses for mercury and lead in 1970, and cadmium and arsenic in 1971 (*2,3*). Residue data from these collections were to serve as a baseline against which future residue levels might be compared. Analysis for selenium was added in 1973. This paper presents results of analyses of the 1973 starling collection including residue levels of mercury, lead, cadmium, arsenic, and selenium from each collecting site; a comparison of averages of mercury, lead, cadmium, and arsenic residues from the 1971 and 1973 collections; and a comparison of residues from urban and rural collecting sites in 1973.

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Collection Methods

Earlier papers discuss the sampling design and collecting procedures in detail (*2,3*). Twenty-five preselected sites were established in 1970 and 28 new sites were added in 1971. The sites were chosen to reflect varying environmental conditions representing broad geographic areas and including differing degrees of human activity and related pollution sources (*3*). Ten-bird pools of starlings were obtained in November and December 1973 from 51 of the 53 preselected sites (Figure 1) by trapping or shooting with shot other than lead. Six samples contained less than 10 birds; these sites are identified in Table 1. Each pool of birds was wrapped in aluminum foil, placed in a polyethylene bag, frozen as soon as possible, and shipped to the Denver Wildlife Research Center for analysis.

Analytical Procedures

Prior to analysis the feet, beaks, wing tips, and skins were removed from birds in each composite sample; the sample was weighed and then ground in a Hobart food chopper. Four 1-g aliquots were taken from each sample homogenate: 1 g each for mercury, arsenic, and selenium analyses, and 1 g for lead and cadmium analyses.

MERCURY ANALYSIS

The presence of mercury was determined by the method of Okuno et al. (*4*) except that the samples were dried in a microwave oven for 5 minutes and then ignited with oxygen in a combustion flask. The combustion products were collected in dilute HCl and the acid solution was reduced with a 20 percent hydroxylamine hydrochloride solution. The mercury was extracted from the solution by amalgamating it onto a silver wire. The silver wire was placed in an absorption cell of the spectrophotometer and heated, by its own electrical resistance, to volatilize the mercury. Spectrophotometric responses were recorded and micrograms of mercury in the samples were read from a standard curve. A fresh standard curve for 0.001 to

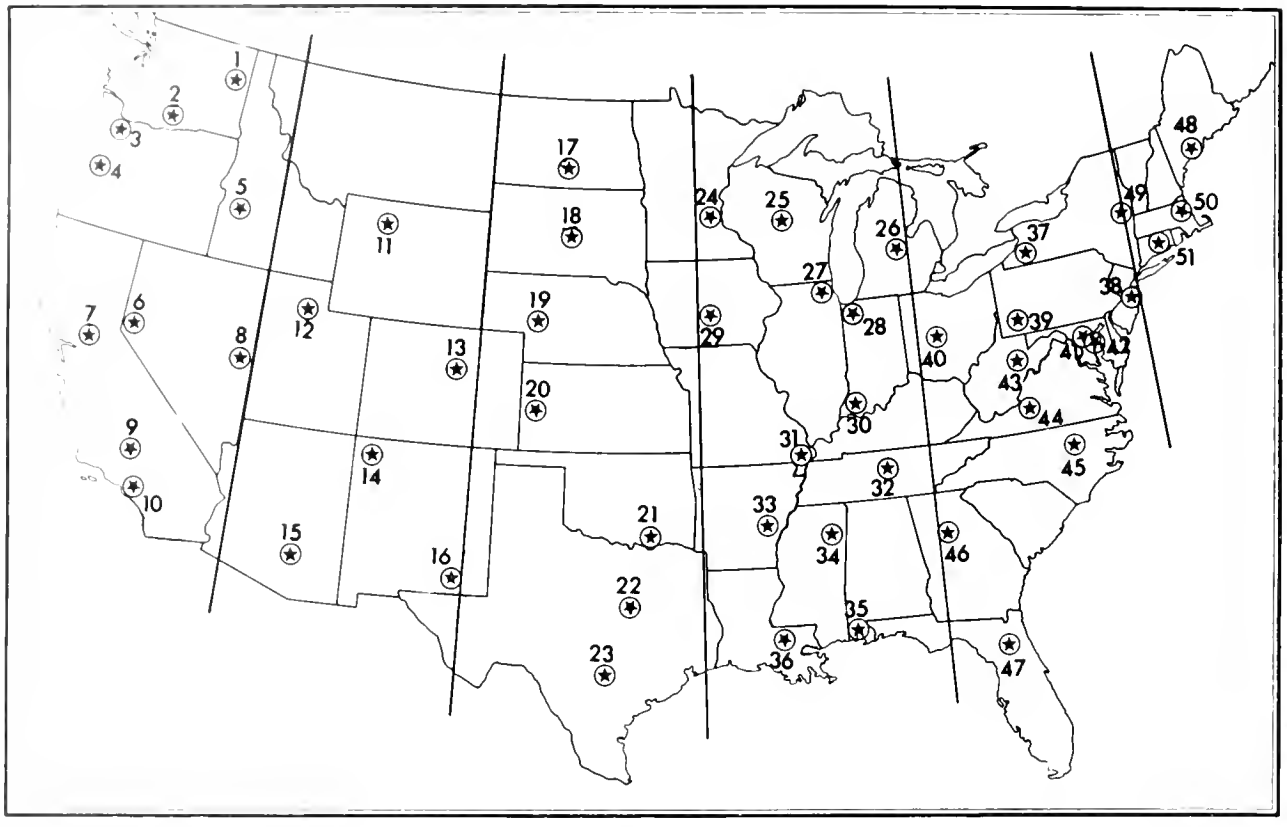

FIGURE 1. *Starling collecting sites, 1973*

0.2 μg mercury was prepared daily. Determinations were made with a Jarrell-Ash Model 82-360 spectrophotometer. The limit of quantification for mercury was 0.01 ppm on a wet-weight basis.

LEAD AND CADMIUM ANALYSIS

The method for detecting lead and cadmium was developed by the Denver Wildlife Research Center (Okuno, 1975: personal communication). A 1-g aliquot was placed in a 50-ml beaker and ashed with a mixture of nitric and perchloric acids; the beaker then was transferred to a 500 C muffle furnace for 4 hours. After cooling, 1 ml concentrated HNO_3 was added and the beaker was heated until the sample was dry and white. After cooling, 1 ml concentrated HNO_3 and 2 ml distilled water were added and the beaker was warmed for 5 minutes on a hotplate. The solution was adjusted to a pH of about 3 and transferred to a 125-ml separatory funnel. One ml of a 1 percent aqueous solution of ammonium pyrrolidine-dithiodicarbamate was added to the funnel; the funnel was swirled and allowed to stand for 2 minutes. Ten ml of methyl isobutyl ketone was added, the funnel was shaken for 1 minute and the aqueous solution was drawn off and discarded. Ten ml 1 N HNO_3 then was added to the funnel and the organic layer was drained off. This solution was analyzed for lead and cadmium using a Varian model 63 carbon rod atomizer on a Jarrell-Ash model 82-360 spectrophotometer. The limits of quantification were 0.1 ppm for lead and 0.05 ppm for cadmium on a wet-weight basis.

ARSENIC AND SELENIUM ANALYSES

A 1-g aliquot for arsenic and selenium analysis was dried in a microwave oven for 5 minutes and ignited in a combustion flask with 20 ml of 25 percent HCl for arsenic analysis or a 1:1 mixture of 50 percent HCl and 25 percent H_2SO_4 for selenium analysis.

For arsenic, the solution was transferred to a 100-ml flask and 2 ml of 40 percent hydroxylamine hydrochloride was added. After 15 minutes, 1 ml 6 percent potassium iodide was added to the flask, the flask was allowed to stand for another 15 minutes, and 2 ml 40 percent $SnCl_2$ was added. After stirring, 1 g of 200-400-mesh zinc in a slurry of water was added. The resulting arsine was swept with helium into the burner of a Perkin-Elmer model 303 spectrophotometer. The limit of quantification for arsenic was 0.05 ppm on a wet-weight basis.

For selenium, 10 ml of the combustion flask solution was transferred to a flask and 30 ml of the 1:1 aqueous acid solution with 1 ml $SnCl_2$ was added. Two g 20-mesh zinc was added to the flask and stirred for 15 seconds. The resulting hydrogen selenide was swept into the burner of a Perkin-Elmer model 303 spectrophotometer. The limit of

TABLE 1. *Residues of mercury, lead, cadmium, arsenic, and selenium in starlings, continental United States—1973*

State	City or County	Site Number	Type of Site[1]	Residues, ppm wet weight				
				Mercury	Lead	Cadmium	Arsenic	Selenium
Alabama	Mobile	35	U	0.07	0.75	<0.05	0.18	0.40
Arizona	Phoenix	15	R	<0.01	0.10	0.05	0.05	0.44
Arkansas	Stuttgart	33	R	0.03	2.40	<0.05	0.12	0.34
California	Bakersfield	9	U	<0.01	0.50	0.05	0.09	0.38
	Los Angeles	10	U	<0.01	0.80	<0.05	0.05	0.38
	Sacramento	7	U	0.06	<0.10	<0.05	<0.05	0.32
Colorado	Brighton	13	R	<0.01	<0.10	0.12	<0.05	0.36
Connecticut	Conn. River Valley	51	R	<0.01	1.80	<0.05	0.12	0.56
Florida	Gainesville	47	R	<0.01	0.65	0.05	0.12	0.10
Georgia	Atlanta	46	U	0.01	1.70	0.09	0.06	0.38
Idaho	Boise	5	R	<0.01	<0.10	0.08	0.09	0.24
Illinois	Chicago	27	U	<0.01	1.00	<0.05	0.07	0.36
Indiana	Evansville	30	R	<0.01	1.20	0.07	0.12	0.72
	Gary	28	U	0.01	1.10	0.08	0.07	0.80
Iowa	Des Moines	29	U	<0.01	0.55	0.05	0.08	0.20
Kansas	Garden City	20[2]	R	0.01	0.55	0.07	0.20	0.60
Louisiana	Baton Rouge	36	R	0.02	0.65	0.09	0.22	0.60
Maine	Gray	48	R	<0.01	0.35	0.10	0.07	0.34
Maryland	Patuxent	41	U	<0.01	1.10	0.08	0.06	0.36
	Annapolis	42	U	<0.01	1.30	0.05	0.13	0.17
Massachusetts	Quincy	50	U	<0.01	0.45	<0.05	0.19	0.28
Michigan	Lansing	26	U	<0.01	0.80	0.06	0.17	0.32
Minnesota	Twin Cities	24[3]	U	<0.01	1.20	0.05	0.17	0.44
Mississippi	Starkville	34	R	<0.01	0.60	<0.05	0.10	0.48
Missouri	Malden	31	R	0.01	0.60	0.13	0.50	0.48
Nebraska	North Platte	19	R	0.01	0.50	0.05	<0.05	0.36
Nevada	McGill	8	R	<0.01	<0.10	<0.05	0.14	0.38
	Reno	6	R	0.20	<0.10	<0.05	0.18	0.80
New Jersey	New Brunswick	38	U	<0.01	3.20	0.06	0.15	0.34
New Mexico	Carlsbad	16	R	<0.01	0.25	<0.05	0.19	0.48
	Farmington	14	R	<0.01	1.30	0.05	<0.05	0.60
New York	Albany	49	U	<0.01	0.65	0.08	0.21	0.20
	Jamestown	37	U	<0.01	1.90	<0.05	0.08	0.32
North Carolina	Raleigh	45	U	<0.01	1.20	0.05	0.05	0.24
North Dakota	Bismarck	17[4]	R	0.01	0.25	0.09	0.28	0.48
Ohio	Columbus	40	R	0.06	1.80	0.05	0.05	0.68
Oklahoma	Tishomingo	21	R	<0.01	1.30	<0.05	0.05	0.60
Oregon	Corvallis	4	R	0.05	<0.10	0.06	0.27	0.38
	Wilsonville	3	R	0.05	0.55	0.07	0.10	0.31
Pennsylvania	Pittsburgh	39	U	<0.01	1.30	0.11	1.40	0.36
South Dakota	Pierre	18[5]	R	0.13	0.14	<0.05	0.52	0.84
Tennessee	Nashville	32	U	<0.01	1.10	0.05	0.24	0.48
Texas	Hillsboro	22	R	0.01	0.80	<0.05	<0.05	0.44
	San Antonio	23	U	<0.01	1.00	<0.05	0.11	0.84
Utah	Salt Lake City	12[3]	R	0.04	0.85	0.20	0.26	0.36
Virginia	Blacksburg	44	R	0.03	0.95	0.05	0.07	0.44
Washington	Spokane	1	R	<0.01	1.30	<0.05	0.25	0.27
	Yakima	2	R	0.03	<0.10	0.05	0.07	1.10
West Virginia	Elkins	43	R	0.01	0.75	0.07	0.11	0.27
Wisconsin	Portage	25	U	0.03	1.20	0.06	0.07	0.72
Wyoming	Worland	11[5]	R	<0.01	0.50	0.05	<0.05	0.68

[1] R = rural areas, U = urban, suburban areas
[2] Sample size = 7
[3] Sample size = 8
[4] Sample size = 3
[5] Sample size = 9

quantification for selenium was 0.05 ppm on a wet-weight basis.

Results and Discussion

Table 1 lists the residues of mercury, lead, cadmium, arsenic, and selenium in starlings by collection site for 1973. Sites are designated either urban or rural. Since some starlings are migratory, caution is advised in making interpretations on a statewide basis. In addition, residue levels do not necessarily reflect year-round levels, because birds were collected in the fall.

MERCURY

As in earlier surveys (*2,3*) mercury levels were uniformly low in starling pools, ranging from <0.01 to 0.20 ppm. Thirty-eight samples, 75 percent, had mercury residues ≤0.01 ppm, and only five samples exceeded 0.05 ppm. Analysis of variance between residues in the 1971 and 1973 collections indicated an overall decline ($P < 0.01$) in mercury residues (Table 2). The 1973 residues were down by 68 percent. The banning of mercuric fungicides for treatment of seeds in 1970-71 may be a major reason for this decline; however, as Baskett (*1*) points out, many of the industrial sources of mercury pollution have been eliminated or greatly reduced in recent years. There was no difference between levels of mercury in starlings from urban and rural areas in 1973 (Table 3).

LEAD

Lead residues were present in all samples, ranging from <0.10 to 3.20 ppm (Table 1). Twenty (39 percent) of the samples had residues of 1.00 ppm or greater but there

TABLE 2. *Arithmetic means, ranges, and geometric means of metals and selenium in starlings, 1971 and 1973*

Element	Year	No. Pools	$\bar{X} \pm SE$	Range	Geom. $\bar{X}$
Mercury	1971	50	0.063[a] ± 0.015	0.01-0.62	0.032
	1973	51	0.020 ± 0.005	<0.01-0.20	0.010
Lead	1971	50	1.310[b] ± 0.190	0.12-6.60	0.840
	1973	51	0.850 ± 0.091	<0.10-3.20	0.550
Cadmium	1971	50	0.048 ± 0.006	0.05-0.24	0.038
	1973	51	0.056 ± 0.005	<0.05-0.20	0.048
Arsenic	1971	50	0.019[c] ± 0.005	<0.01-0.21	0.014
	1973	51	0.156 ± 0.029	<0.05-1.40	0.102
Selenium	1971		No analysis		
	1973	51	0.451 ± 0.028	0.10-1.10	0.410

[a] Means for the two years significantly different $P < 0.01$
[b] Means for the two years significantly different $P < 0.05$
[c] Means for the two years significantly different $P < 0.001$

TABLE 3. *Comparison of metal and selenium residues in starlings from urban and rural sites, 1973*

Element	Site	No. Pools	$\bar{X} \pm SE$	Range	Geom. $\bar{X}$
Mercury	Urban	21	0.012 ± 0.004	<0.01-0.07	0.007
	Rural	30	0.025 ± 0.007	<0.01-0.20	0.012
Lead	Urban	21	1.088[a] ± 0.140	<0.10-3.20	0.885
	Rural	30	0.681 ± 0.110	<0.10-2.40	0.390
Cadmium	Urban	21	0.042 ± 0.007	<0.05-0.11	0.019
	Rural	30	0.053 ± 0.008	<0.05-0.20	0.023
Arsenic	Urban	21	0.171 ± 0.063	<0.05-1.40	0.085
	Rural	30	0.139 ± 0.024	<0.05-0.52	0.057
Selenium	Urban	21	0.395 ± 0.039	0.17-0.84	0.362
	Rural	30	0.491 ± 0.038	0.10-1.10	0.448

[a] Means for the two areas significantly different $P < 0.05$

appeared to be no geographic pattern associated with these higher levels. Those samples with ≥1.00 ppm lead were from widely scattered sites, both urban and rural, throughout the country. There was a significant overall decline ($P < 0.05$) of 35 percent in lead residues since 1971 (Table 2). The reason for this apparent decline is not clear. The increased use of lead-free gasoline in automobiles could be a factor, as well as the reduction of lead contamination in industrial effluents. Analytical methods for lead were essentially the same in 1971 and 1973, as were all the residue determinations; therefore, the data for the two periods are comparable. Lead residues in starlings from urban areas were significantly higher than in those from rural areas in 1973 (Table 3). This is expected because of the higher exposure to automotive and industrial contaminants in urban areas (*3*).

CADMIUM

Cadmium residues were detected in all starling samples, ranging from <0.05 to 0.20 ppm (Table 1). Thirty-one (61 percent) of the samples had residues ≤0.05 ppm and only five samples had levels of 0.10 ppm or greater. As reported earlier (*3*) cadmium residues in whole-body samples of starlings were low. Residues would be higher in major accumulator organs, such as kidneys or livers, than in whole bodies. White and Kaiser (*9*) report levels of cadmium in ruddy duck livers up to 1.60 ppm, and White and Stendell found levels up to 10.0 ppm in kidneys of canvasbacks (Residues of Environmental Pollutants in Canvasback Tissues, Eggs, and Food Materials, 1976. D.H. White and R.C. Stendell [*unpublished*], Patuxent Wildlife Research Center, Laurel, Md. 20811). There was no statistical difference between mean levels of cadmium in starlings in 1971 and those in 1973 (Table 2). Also, there was no difference in cadmium residues from urban and rural sites (Table 3).

ARSENIC

Unlike 1971, arsenic residues were present in all starling samples in 1973, ranging from <0.05 to 1.40 ppm (Table 1). Twenty-eight (55 percent) of the samples contained residues of 0.10 ppm or greater. Arsenic residues in 1973 were 88 percent higher ($P < 0.001$) than in 1971 (Table 2). This overall increase of residues in starlings may reflect an increase in the use of arsenic containing compounds as insecticides or herbicides on lawns. A comparison of urban and rural sites, however, showed no significant difference in residue accumulations from the two areas (Table 3). Wiersma et al. (*10*) found no difference in arsenic residues in soil from lawn and unkept areas in eight different cities; they attribute arsenic residues in the environment mainly to industrial or combustion sources rather than to pesticidal contamination. These data support that hypothesis.

SELENIUM

Selenium was included in the starling analyses for the first time in 1973. Selenium is a necessary dietary element for many animals, including mammals and birds (*5*), but is very toxic in excessive amounts. Sources of environmental contamination by selenium include copper and lead smelting, combustion of fossil fuels, wastes from paper mills, and many other industrial processes (*6*). Selenium residues were present in all starling samples and ranged from 0.10 to 1.10 ppm (Table 1). There was no difference between residues in urban and rural areas (Table 3). These data indicate that selenium is widespread in the environment and is readily available to birds.

Conclusions

Nationwide, residues of mercury and lead have declined significantly in starlings since 1971. On the other hand, arsenic residues have shown a pronounced increase. Selenium, analyzed for the first time, was found in all starling samples, indicating that this element is widespread throughout the country. These data suggest that starlings can serve as indicators of environmental levels of metals, as do starlings and waterfowl for organochlorines (*7,8*), and thus provide information on residue trends over an extended period of time.

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conducted all analyses. Debora Hutchison compiled the tables and typed the manuscript.

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Residues of Organochlorines and Heavy Metals in Tissues and Eggs of Brown Pelicans, 1969-73

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ABSTRACT

Shells of brown pelican (Pelecanus occidentalis) *eggs collected in South Carolina from 1969 through 1973 were significantly thinner than shells of those collected before 1947. Residues of 10 organochlorine pollutants and 10 heavy metals were found in these eggs. Total organochlorine residues were apparently magnified 23 times from fish to pelican eggs, but interpretation of biomagnification was complicated by the migratory habits of both the pelicans and their chief prey fish.*

Residues of organochlorine pollutants and heavy metals were also found in tissues of brown pelicans. Dieldrin was probably involved in the death of a pelican that exhibited myocardial necrosis. Other pelicans died from gunshot wounds, various diseases, or unknown causes.

From 1969 through 1973, there was a significant decline in residues of p,p'-*DDE,* p,p'-*TDE,* p,p'-*DDT, and dieldrin in eggs of the brown pelican in South Carolina, but the rate of decline was different for each pollutant. PCB's peaked in 1972 and then declined in 1973 to the lowest level in 5 years. In 1973, the first time in many years, South Carolina brown pelicans reproduced very well. The excellent reproductivity seemed related to lowered organochlorine residues and favorable tides, weather, and food supply.*

Introduction

The brown pelican (*Pelecanus occidentalis*) provides a classic example of adverse biological effects caused by certain environmental pollutants. Shells of brown pelican eggs have thinned in all parts of their range in the United States (*1,4,9,24,31*) and in parts of Mexico (*20,24*); reproduction has been adversely affected (*2,10,20*); and a population in South Carolina has been substantially reduced (*3,5*). There is speculation that pollution was involved in the extirpation of a population of brown pelicans in Louisiana (*2,21*). In previous studies, authors demonstrated that DDE was the agent principally responsible for eggshell thinning (*5-8*); and that DDE, and to a lesser extent dieldrin, were responsible for limited reproduction and probably the population decline in South Carolina (*10*). In the last several years, however, the status of the brown pelican has improved in several areas of its geographic range (*2,9*). This paper describes the biological significance of certain pollutants in eggs and tissues of brown pelicans in South Carolina and discusses the impact of declining residues on these birds. Pertinent residue data from tissues and eggs of Florida pelicans are also included.

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PROCEDURES FOR SAMPLING, NECROPSY, AND FIELD STUDY

The authors made brief visits to brown pelican colonies in South Carolina in 1969 and 1970; most of the data were published previously (*5*) as were the data from a study of brown pelicans in Louisiana (*9*). The intensive study in South Carolina was conducted in the spring and summer each year from 1971 through 1973. The nesting colony on Marsh Island, Cape Romain National Wildlife Refuge, received most attention, but authors also studied the only other colony in the State, Deveaux Bank. An annual census was conducted of total nests and fledged young in each colony. Eggs in all stages of incubation, both viable and addled, were collected throughout the nesting season. Collectors attempted to gather eggs in all portions of each colony. Some unbroken, rolled eggs were also collected. It was sometimes possible to salvage contents of cracked or crushed eggs. Eggshells of depredated eggs, hatched eggs, and crushed or cracked eggs were collected. One egg was usually taken from each nest selected for sampling.

On Marsh Island two types of nests were marked to determine nest success; one type included nests with full clutches and the other included nests from which one egg was collected. Marked nests were checked for eggs or young on each visit to the colony; collectors usually made two visits to each colony per week with a maximal stay of

1 hour per visit. Several dead pelicans and samples of fish, regurgitated by pelicans, were also collected. Colleagues from Florida provided several pelicans that were found dead on the Atlantic Coast near Stuart in 1972 and in Florida Bay in 1973.

Eggs were weighed and measured. The contents of some eggs were removed before freezing and placed in glass bottles which had been rinsed with dilute nitric acid solution, deionized water, hexane, and acetone. Caps were lined with aluminum foil and the bottles were placed in a freezer. If eggs were not opened immediately, they were wrapped in foil, frozen, and later opened while still frozen. A record was made of the stage of development of each egg. Shells were washed thoroughly with tap water and allowed to dry at room temperature. Shell thickness, which includes shell and shell membranes, was measured at three sites on the waist of the egg with a micrometer graduated in units of 0.01 mm; the average of the three measurements was used to represent the thickness of the shell.

Fish samples were taken from pools of several regurgitated boluses found in the South Carolina colonies. Nesting pelicans and their young regurgitate food when they are disturbed; hence authors collected fish that had undergone little digestion.

The pelicans found dead were frozen; the specimens were removed from the freezer several months later, allowed to thaw, and subsequently necropsied to determine cause of death. Tissues for histological study were fixed in 10 percent formalin, embedded in paraffin, sectioned, and stained. The entire brain was removed and placed in a chemically clean glass bottle, and the remaining carcass, except for skin, feet, wings, liver, kidney, and gastrointestinal tract, was wrapped in foil and refrozen. The brains and carcasses were later analyzed for residues of organochlorine pollutants, and some livers were analyzed for heavy metals. The livers of four pelicans shot in 1970 were analyzed for residues of heavy metals.

Analytical Procedures

Analyses for heavy metals in eggs and tissues were conducted by the Environmental Trace Substances Center, Columbia, Mo. (J. R. Vogt, ed., University of Missouri Nuclear Science Group, 1970-71: unpublished reports). Nickel, selenium, arsenic, and chromium were analyzed by neutron activation techniques. Samples were freeze-dried and placed in vials for irradiation. Irradiation conditions and times were: a neutron flux of 4×10^{13} n /cm^2 /sec—5 to 15 minutes for arsenic and 200 hours for chromium; 5×10^{13} n/cm^{-2}/sec^{-1}—30 to 60 minutes for selenium and approximately 20 minutes for nickel. The samples were allowed to undergo radioactive decay for given periods of time following irradiation. Chemical separation of the metal was then conducted, followed by counts of gamma-ray radiation from each sample. The counter was a Packard Model 9012, 1024 channel Pulse Height analyzer with a Na-I detector. Residues of metals were measured with the 0.32 MeV gamma-ray of ^{51}Cr, the 556 KeV gamma-ray of ^{76}As, the 1.49 MeV gamma-ray of ^{65}Ni, and the 104 KeV gamma-ray of ^{81m}Se.

Samples to be analyzed for copper, zinc, cadmium, lead, and magnesium were wet-ashed with a mixture of nitric and perchloric acids to destroy the organic material and then solubilized in an acidic solution. Lead and cadmium were chelated with ammonium pyrrolidine carbodithionate, and extracted from the aqueous phase into methyl isobutyl ketone (MIBK). The MIBK containing the lead and cadmium complexes was analyzed using a flame atomic absorption (AA) technique. The solubilized samples for copper, magnesium, and zinc analyses were directly aspirated into the AA flame. Absorbance was recorded at the following wave lengths (nm): lead, 283.3; cadmium, 228.8; copper, 324.7; zinc, 213.9; and magnesium, 285.2, using an Instrumentation Laboratories Model 453 AA spectrometer. Samples for mercury analysis were digested with nitric acid. Stannous chloride was added to reduce ionic mercury to elemental mercury, which was then measured photometrically in the vapor phase by AA using a Perkin-Elmer Model 290.

Pelican tissues and the contents of the eggs were analyzed individually for residues of organochlorine pesticides, their metabolites, and polychlorinated biphenyls (PCB's); these analyses were completed at the Patuxent Wildlife Research Center, Laurel, Maryland.

For eggs and pelican tissues collected in 1971, residue analysis was performed as follows: a 10-g sample of the ground carcass, the entire brain, and a 20-g sample of the homogenized egg were mixed separately with anhydrous sodium sulfate in a blendor and extracted for 7 hours with hexane in a Soxhlet apparatus. Extracts were cleaned by acetonitrile partitioning and elution on partly deactivated florisil; half was saved for PCB analysis. For insecticide analysis, residues in the cleaned extract were separated and removed in four fractions from a silica gel thin-layer plate (*30*). Each thin-layer fraction was analyzed by electron-capture gas chromatography (GC) on a column of 3 percent OV-1 or 3.8 percent UCW-98 on Chromosorb WHP. DDT and its metabolites in fractions III or IV were confirmed on a column of 3 percent XE-60 or 3 percent QF-1 on Gas-Chrom Q. PCB's were identified and measured semiquantitatively by thin-layer chromatography (*29*). The average percent recoveries of organochlorine pesticides and their metabolites from spiked bald eagle (*Haliaeetus leucocephalus*) carcasses were: DDE, 95; TDE, 102; DDT, 110; dieldrin, 106; heptachlor epoxide, 112; and *o,p'*-DDT, 107. Residues were not corrected for recovery values.

For eggs and pelican tissues collected in 1972 and 1973, the methodology was modified as described by Cromartie

et al. (14). Lipids were removed from the extract by a column chromatography procedure using washed, recalcined, and partly deactivated florisil. An aliquot of the florisil eluate was concentrated and placed on a silicic acid column to separate the pesticides from PCB's. The organochlorine residues were collected in three fractions and analyzed with an electron-capture gas chromatograph equipped with a 4 percent SE-30/6 percent QF-1 column. Using this method, authors were able to detect additional pesticides: toxaphene, *cis*-chlordane and/or *trans*-nonachlor, and *cis*-nonachlor. In 1972 the laboratory did not have a *cis*-nonachlor standard for quantitation, and authors first developed a procedure to estimate toxaphene levels in 1973; a manuscript is being prepared which describes the procedure. DDE was measured by peak height to avoid errors from PCB interference; the other organochlorines were quantitated with a computing integrator and PCB's were estimated by comparing total area of PCB peaks with that of Aroclor 1260. A combined gas chromatograph – mass spectrometer (GC-MS) was used to confirm residues in about 10 percent of the samples. Average percent recoveries from spiked mallard carcass tissues were: DDE, 96; TDE, 103; DDT, 110; dieldrin, 101; heptachlor epoxide, 104; mirex, 106; oxychlordane, 98; *cis*-chlordane, 100; *cis*-nonachlor, 98; HCB, 69; and Aroclor 1254, 101. Residues in the pelican eggs and tissues were not corrected for recovery values. The lower limit of detection for pesticides or their metabolites was 0.01 μg/g in regurgitated fish and 0.10 μg/g in other samples; the lower limit for PCB's was 0.05 μg/g in regurgitated fish and 0.50 μg/g in other samples.

Results and Discussion

EGGSHELL THICKNESS

The eggshells of South Carolina pelicans were 14 to 17 percent thinner ($P < 0.05$) each year from 1969 through 1973 than they had been before 1947 (Table 1). The average annual thicknesses are similar for the 5-year period 1969-73, although the mean thickness in 1971 was significantly greater ($P < 0.05$) than that recorded in several other years. Mean thicknesses listed in Table 1 are derived from entire eggs found in the nest.

Authors compared eggshell thickness to stage of embryonic development and to fate of the eggs. Eggshell thickness seemed to increase during incubation, although

TABLE 1. *Eggshell thickness of brown pelican eggs, South Carolina*

Eggshell Thickness, mm[1]					
Pre-1947[2]	1969	1970	1971	1972	1973
0.557+ A	0.463+ C	0.461+ C	0.480+ B	0.470+ BC	0.463+ C
(0.012)(23)	(0.006)(49)	(0.007)(38)	(0.005)(65)	(0.005)(67)	(0.003)(104)

Mean + standard error, sample size in parentheses.
A significant difference among thickness means ($P < 0.05$) is indicated for those means not showing a common letter. Means were separated using the new multiple range test (26) with extension for unequal replication (25).

TABLE 2. *Eggshell thickness of brown pelican eggs by stage of incubation, South Carolina—1971-73*

Stage of Incubation	Eggshell Thickness, mm[1]	
	Mean ± Standard Error (Number)	Range
Freshly laid	0.466 ± 0.004 (113) B	0.36-0.61
Early development (1-2.3)	0.467 ± 0.003 (169) B	0.37-0.57
Late development (>2.3)	0.475 ± 0.007 (37) B	0.40-0.56
Hatched	0.506 ± 0.006 (19) A	0.46-0.56

[1]See footnotes, Table 1.

TABLE 3. *Eggshell thickness of brown pelican eggs by condition, South Carolina—1971-73*

Condition	Eggshell Thickness, mm[1]	
	Mean ± Standard Error (Number)	Range
Hatched	0.506 ± 0.006 (19) A	0.46-0.56
Stepped on	0.480 ± 0.018 (6) AB	0.41-0.53
Entire	0.469 ± 0.002 (236) B	0.36-0.61
Rolled	0.468 ± 0.015 (11) B	0.38-0.58
Depredated	0.465 ± 0.009 (17) B	0.42-0.54
Crushed	0.377 ± 0.015 (40) C	0.13-0.47

[1] See footnotes, Table 1.

Kreitzer (26) reported the contrary to be true of Japanese quail (*Coturnix coturnix japonica*). Eggshell thickness is an important factor in the nest life of the egg, and thin-shelled pelican eggs have a shorter nest life than have those with thicker shells. The minimum thickness increased with each stage of incubation; for example, the minimum shell thickness of a hatched egg was 0.46 mm, compared to 0.36 mm in an intact freshly laid egg (Table 2).

Thickness of eggshells also varied with condition of the egg (Table 3). Brown pelican eggs were examined and classified according to six conditions: entire, rolled, depredated, stepped on, crushed, and hatched. Entire eggs were those found intact in the nest; rolled eggs were entire eggs which had been removed from the nest, apparently by the parent. Most rolled eggs were freshly laid and were noted early in the nesting season; on April 5, 1973, over 100 rolled eggs were observed in a colony containing 400 nests. It is probable that pelicans deliberately rolled their eggs out of the nest, especially during periods of unseasonably cold weather. Pelicans desert their nests in the Florida Keys in apparent response to unseasonably cold weather (Alexander Sprunt IV, National Audobon Society, 1969; personal communication), and some deserted their nests at Pelican Island, Fla., in response to a food shortage (19).

Most of the depredated eggs had probably been rolled from nests by pelicans before they were depredated by laughing gulls (*Larus atricilla*). No laughing gulls were observed depredating eggs in active pelican nests, and it appears that these gulls take few viable eggs. The herring gull (*Larus argentatus*) and the ring-billed gull (*Larus delawarensis*) may be more efficient egg predators, but

TABLE 4. *Residues of organochlorine pollutants in eggs of brown pelicans, South Carolina—1971*

Residues, µg/g fresh wet weight						
DDE	TDE	DDT	Dieldrin	Heptachlor Epoxide	Mirex	PCB's
Marsh Island						
1.93	0.20	0.18	0.34	—	—	4.1
2.15	0.23	0.14	0.57	—	—	4.9
6.05	1.04	0.79	0.92	—	—	9.9
1.71	0.37	0.34	0.31	—	—	3.8
3.97	0.77	0.47	0.84	—	—	5.4
2.56	0.74	0.13	0.71	—	—	3.6
3.87	0.80	0.42	1.11	—	0.31	4.7
3.44	0.70	0.30	0.34	—	—	7.4
1.93	0.57	0.37	0.58	—	—	6.5
2.87	0.44	0.41	0.21	—	—	5.4
1.82	0.26	0.18	0.39	—	—	4.5
2.46	0.47	0.52	0.43	0.17	—	36.5
1.39	0.27	0.19	0.19	—	—	3.0
1.90	0.71	0.18	0.54	—	—	17.3
3.99	1.04	—	0.83	—	—	9.2
1.30	0.48	—	0.33	—	—	5.3
3.47	0.80	0.17	0.81	—	—	20.1
2.26	0.21	0.22	—	—	—	19.7
2.49	0.44	0.11	0.47	—	—	3.9
1.12	0.20	0.16	0.31	—	—	1.5
1.20	0.48	—	0.24	—	—	2.4
1.96	0.54	—	0.52	—	—	1.9
1.94	0.39	—	0.32	—	—	2.6
2.25	0.37	0.13	0.42	—	—	18.6
2.23	0.41	0.23	0.56	—	—	5.8
2.46	0.52	0.28	0.49	—	—	3.9
4.55	0.53	0.89	1.02	—	—	19.8
2.77	0.47	0.30	0.62	—	—	6.2
2.64	0.64	—	0.37	—	—	5.7
1.77	0.25	—	0.36	—	—	3.4
3.59	0.47	0.30	0.72	—	—	9.7
2.66	0.37	0.19	0.26	—	—	3.8
1.96	0.43	—	0.42	—	—	5.9
2.85	0.55	0.21	0.32	—	—	3.6
2.37	0.83	—	0.28	—	—	3.6
2.26	0.61	0.42	0.29	—	—	3.9
3.09	0.31	—	0.70	—	—	8.0
3.60	0.96	—	0.66	—	—	7.3
2.95	0.46	—	0.57	—	—	17.9
2.92	0.68	0.31	0.77	—	—	3.8
2.15	0.35	0.20	0.61	—	—	5.9
3.57	0.76	0.23	0.43	—	—	7.1
2.97	0.49	0.39	0.45	—	—	5.9
2.68	0.51	—	0.92	—	—	9.2
GM 2.48	0.48	0.16	0.45			6.02
CL 2.23-2.76	0.42-0.55	0.12-0.21	0.38-0.54			4.89-7.42
Range 1.12-6.05	0.20-1.04	ND-0.89	0.20-1.02	ND-0.17	ND-0.31	1.5-36.5
Deveaux Bank						
1.25	0.18	—	0.21	—	—	5.8
1.60	0.22	0.14	0.21	—	—	3.3
2.29	0.39	0.17	0.38	—	—	9.6
3.02	0.34	0.21	0.61	—	—	5.8
1.42	0.40	0.35	0.25	—	—	2.8
2.11	0.50	—	0.49	—	—	4.5
2.34	0.48	—	0.53	—	—	1.8
3.70	0.75	—	0.55	—	—	7.2
3.30	0.86	—	0.61	—	—	9.2
1.27	0.35	—	0.20	—	—	3.6
2.37	0.40	0.21	0.46	—	—	5.7
2.10	0.57	—	0.80	0.14	—	5.3
1.69	0.38	—	0.34	—	—	9.2
3.19	0.44	—	0.64	—	—	9.1
4.40	0.56	0.37	0.87	0.10	—	26.9
2.19	0.70	0.23	0.31	—	—	5.5
3.71	0.77	—	0.79	—	—	9.4
2.98	0.82	0.16	0.86	—	—	29.8
3.06	0.58	0.13	0.56	—	—	9.5
3.95	1.01	—	0.59	—	—	18.4
3.21	0.27	—	0.54	—	—	26.8
GM 2.46	0.48	0.09	0.47			7.56
CL 2.07-2.93	0.39-0.59	0.06-0.13	0.38-0.58			5.38-10.63
Range 1.25-4.40	0.18-1.01	ND-0.37	0.20-0.87			1.8-29.8
Marsh Island and Deveaux Bank						
GM 2.48	0.48	0.13	0.46			6.48
CL 2.27-2.71	0.43-0.53	0.11-0.17	0.40-0.52			5.44-7.73
Range 1.12-6.05	0.18-1.04	ND-0.89	0.20-1.02	ND-0.17	ND-0.31	1.5-36.5

NOTES: ND or — = no residue detected.
GM = geometric mean.
CL = 95% confidence limits.

TABLE 5. *Residues of organochlorine pollutants in eggs of brown pelicans, South Carolina—1972*

Residues, μg/g fresh wet weight							
DDE	TDE	DDT	Dieldrin	Heptachlor Epoxide	Mirex	*Cis*-Chlordane[1]	PCB's
Marsh Island							
2.33	0.26	—	—	—	—	—	6.0
2.05	0.18	0.24	0.17	—	—	—	3.9
2.71	0.19	0.25	0.17	—	—	0.10	4.4
1.83	0.12	0.10	0.32	—	0.11	0.10	3.2
2.47	0.11	0.10	0.15	—	—	—	4.4
2.30	0.29	0.15	0.37	—	0.15	0.10	3.5
2.42	0.42	0.15	0.40	—	0.11	0.10	6.3
3.75	0.63	0.25	0.63	—	0.16	0.16	7.6
4.48	0.10	0.25	—	—	0.26	—	6.4
1.20	0.16	—	0.28	—	0.11	—	3.9
1.90	0.15	0.12	0.52	—	0.20	0.10	6.0
2.33	0.49	0.27	0.59	—	0.14	—	6.0
2.34	0.22	0.17	0.39	—	0.12	0.10	5
3.97	0.43	0.18	0.61	—	0.18	0.15	9.2
3.00	0.22	0.16	0.46	—	0.11	0.13	5.2
3.08	0.34	0.22	0.68	—	—	0.17	5.4
3.50	0.50	0.28	0.74	—	0.19	0.19	6.8
2.85	0.42	0.21	0.45	—	—	0.15	5.3
3.46	0.38	0.19	0.54	—	0.17	0.16	8.4
2.57	0.23	0.12	0.41	—	—	0.11	5.8
2.15	0.32	0.32	0.56	—	—	—	2.6
2.66	0.30	0.18	0.64	—	0.17	0.10	6.5
5.24	0.61	0.45	0.77	—	0.26	0.25	16.7
2.18	0.49	—	0.29	—	—	—	6.0
2.21	0.18	0.18	0.45	—	—	—	5.8
3.95	0.67	0.39	0.64	—	0.23	0.20	8.4
3.99	0.55	0.30	0.52	—	—	0.17	9.2
3.61	0.34	0.34	0.32	—	0.11	—	6.3
7.04	1.23	0.42	0.38	—	0.13	—	23.6
2.37	0.61	0.13	0.35	—	—	—	5.1
1.46	0.22	0.14	0.28	—	0.14	—	4.9
1.58	0.19	—	0.19	—	0.19	—	3.9
2.25	0.38	—	0.32	—	—	0.11	9.0
2.88	0.60	—	0.50	—	—	0.22	8.6
1.76	0.17	—	0.29	—	—	—	3.8
3.49	0.27	0.27	0.75	—	—	0.13	7.8
1.56	0.29	0.32	0.19	—	—	0.13	5.7
3.99	0.39	0.33	0.99	—	—	0.13	8.3
4.37	0.83	—	0.64	—	—	0.22	10.4
3.32	0.70	0.18	0.58	—	—	0.18	9.0
3.52	0.29	0.15	0.50	—	—	—	7.7
5.35	0.46	0.38	1.11	—	0.16	0.19	16.1
4.14	0.36	0.25	0.71	—	—	0.11	9.8
6.61	0.66	0.37	0.75	—	0.11	0.16	14.5
8.48	0.95	0.39	0.40	—	0.19	0.34	32.3
6.49	1.21	—	0.89	—	0.11	0.24	15.0
3.08	0.20	0.13	0.48	—	0.10	0.10	6.9
3.92	0.33	0.15	0.44	—	0.14	—	9.4
4.12	0.38	0.22	0.75	—	0.15	0.14	6.2
GM 3.02	0.34	0.17	0.41				6.98
CI 2.68-3.42	0.29-0.41	0.14-0.20	0.34-0.50				6.05-8.05
Range 1.20-8.48	0.10-1.23	ND-0.45	ND-1.11	ND	ND-0.26	ND-0.34	2.6-32.3
Deveaux Bank							
4.17	1.05	0.57	1.32	0.13	—	0.17	11.6
8.38	1.92	0.77	1.76	0.53	0.31	0.49	14.6
1.95	0.46	0.22	0.43	—	0.10	—	3.8
1.51	0.32	0.20	0.29	—	0.11	0.11	4.6
3.06	0.26	0.13	0.26	—	0.15	—	7.5
4.35	0.36	0.22	0.41	—	0.21	—	8.1
2.54	0.21	0.12	0.31	—	0.16	—	7.6
4.12	0.35	0.10	0.39	—	0.13	0.10	13.3
1.48	0.14	—	0.23	—	—	—	5.2
3.78	0.45	0.28	0.60	—	—	0.15	8.1
1.47	0.13	—	0.26	—	—	—	5.8
4.55	0.47	—	0.80	—	0.16	0.17	11.7
8.06	0.43	0.17	0.39	—	0.46	0.25	24.5
0.74	0.28	0.31	0.68	—	0.12	0.12	10.3
2.27	0.27	0.13	0.73	—	—	0.25	6.2
3.47	0.84	—	0.42	—	—	0.13	7.3
2.64	0.23	0.23	0.54	—	—	—	7.3
9.39	1.44		0.93	—	0.16	0.42	26.5
6.85	0.70	0.38	1.15	—	0.41	0.24	14.3
2.86	0.40		0.61	0.11	0.15	0.14	7.4
1.28	0.17		0.20	—		—	6.1
3.35	0.29	0.11	0.64	—	0.12	0.11	7.0
4.55	0.70		1.21	—	0.15	0.26	10.0

(Continued next page)

TABLE 5. (cont'd.). *Residues of organochlorine pollutants in eggs of brown pelicans, South Carolina—1972*

Residues, µg/g fresh wet weight							
DDE	TDE	DDT	Dieldrin	Heptachlor Epoxide	Mirex	*Cis*-Chlordane[1]	PCB's
GM 3.05	0.40	0.13	0.53				8.79
CL 2.34-3.98	0.30-0.54	0.09-0.19	0.41-0.69				7.81-9.90
Range 0.74-8.38	0.13-1.92	ND-0.77	0.20-1.76	ND-0.53	ND-0.46	ND-0.49	3.8-26.5
Marsh Island and Deveaux Bank							
GM 3.03	0.36	0.15	0.45				7.51
CL 2.70-3.40	0.31-0.42	0.13-0.18	0.39-0.52				6.68-8.46
Range 0.74-8.48	0.10-1.92	ND-0.77	ND-1.76	ND-0.53	ND-0.46	ND-0.49	2.6-32.3

NOTE: ND or — = no residue detected
GM = geometric mean
CL = 95 percent confidence limits
[1]*Cis*-chlordane and/or *trans*-nonachlor

the small numbers of immature gulls present during the pelican nesting season take few eggs.

Eggs classified as stepped on are those that authors observed the parent stepping on and crushing when taking flight. Some such eggs were well advanced in the incubation period, but all shells were thinner than the pre-1947 mean listed by Anderson and Hickey (1).

Crushed eggs included those that were apparently destroyed by weight stress of the parent during normal incubation; nearly all were freshly laid. Crushed eggs were discovered throughout the nesting season, but most were found early in the season. Some extremely thin eggs were essentially little more than shell membranes; the minimum thickness of a crushed egg was 0.13 mm. Shells of extremely thin-shelled eggs showed interesting variations: the thin shells of eggs in one clutch were light brown instead of the usual white color; in another clutch, shells had lumps of soft calcium carbonate loosely adhering to the shell membrane; and fragile shells of eggs in a third clutch had prominent pimples. It was impossible to measure shell thickness of some eggs with abnormal shells. These extremely thin-shelled eggs were crushed immediately after they were laid.

In 1971, few thin eggshells were noted on the initial visit to Marsh Island in mid-May. The greatest incidence of eggs with extremely thin shells was noted on March 31, 1972. The birds had begun laying only a short time before, and at least 10 of the 300 to 400 nests contained thin-shelled, crushed eggs; at least 3 percent of the nests were unproductive due to thin eggshells. Few thin-shelled eggs were found on the first visit to the island colony in early April 1973. Observations of a few marked nests containing one or more crushed or cracked eggs revealed that all of these nests were unsuccessful. Second clutches in some of these nests had eggs with thicker shells, but authors could not verify that the same female had laid both clutches.

Thickness of eggshells was directly related to the fate of the egg (Table 3). Hatched eggs had significantly thicker shells ($P<0.05$) than had the other five types of eggs except for those that were stepped on. Eggshells of crushed eggs were significantly thinner ($P<0.05$) than were shells of other types of eggs. Shells of entire eggs were considerably thinner ($P<0.05$) than those of hatched eggs and considerably thicker ($P<0.05$) than those of crushed eggs.

RESIDUES

Residues of 10 organochlorines were identified in eggs of brown pelicans (Tables 4-6). Total organochlorine residues in pelican eggs averaged 8.59 µg/g; PCB's and DDE comprised the bulk of these residues. Residues of organochlorines in eggs followed the same pattern in the two colonies; that is, mean residues in a given year were almost the same in each colony; there was much variation in residues of each of the organochlorines in each colony; and there was a general decline from 1969 to 1973 in residues in each colony.

Eleven of the rolled eggs collected in 1973 were analyzed for residues of organochlorines. Mean residues in rolled eggs were essentially the same as those in entire eggs that were collected from nests (Tables 6,7).

Table 8 itemizes residues detected in 13 pelicans which were found dead. Residues in tissues and eggs of brown pelicans provide evidence that the Atlantic Coast of Florida is more heavily contaminated with the organochlorines considered in this study than either the Gulf Coast or Florida Bay. Residues in tissues of pelicans found dead on the Atlantic Coast of Florida in 1972 were several times higher than those in tissues of pelicans found dead in Florida Bay in 1973. The same relationship seemed true of residues in tissues of pelicans shot in the two areas in 1970 (5). Pelican eggs collected in 1969 from the Florida Atlantic Coast colonies contained higher organochlorine residues than did those collected either in Florida Bay or on the Gulf Coast (5). Residues in pelican tissues were generally low, especially in young found dead. This substantiates earlier findings that prefledgling birds have lower residues than have older birds (5).

TABLE 6. *Residues of organochlorine pollutants in eggs of brown pelicans, South Carolina—1973*

Residues, µg/g fresh wet weight									
DDE	TDE	DDT	Dieldrin	Heptachlor Epoxide	Mirex	Cis-chlordane[1]	Cis-nonachlor	Toxaphene	PCB's
Marsh Island									
2.20	0.12	0.17	0.44	—	0.13	0.17	0.13	0.54	4.2
2.04	0.37	0.20	0.32	—	—	0.24	0.10	0.29	3.9
1.83	0.21	0.16	0.44	—	—	0.23	—	0.29	4.1
1.33	—	0.14	0.27	—	—	0.16	—	0.39	3.2
1.31	—	0.10	0.22	—	—	0.11	—	0.13	3.5
1.57	0.22	0.18	0.44	—	0.11	0.23	0.11	0.52	3.7
3.23	0.60	0.35	0.37	—	—	0.32	0.13	0.42	5.5
1.34	0.21	0.23	0.40	—	0.16	—	0.35	2.5	
2.37	0.19	0.24	0.38	—	—	0.25	0.12	0.45	4.0
2.28	0.13	0.15	0.45	—	0.12	0.19	0.12	0.40	3.5
2.75	0.28	0.25	0.40	—	—	0.28	0.12	0.40	5.8
2.66	0.24	0.27	0.53	—	0.17	0.10	0.41	3.9	
1.71	0.61	—	0.27	—	—	0.21	—	0.16	1.7
2.50	0.20	0.18	0.39	—	0.12	0.25	0.12	0.43	5.2
1.92	0.12	0.18	0.35	—	0.13	0.24	0.11	0.43	4.8
4.17	0.57	0.34	0.60	—	—	0.36	0.13	0.42	11.2
1.45	0.16	0.20	0.45	—	—	0.19	—	0.34	3.3
2.72	0.27	0.17	0.48	—	—	0.23	0.10	0.27	4.4
2.23	0.14	0.22	0.53	—	0.17	—	0.38	4.3	
2.45	0.48	0.30	0.55	—	—	0.33	0.10	0.33	4.5
1.59	0.19	0.14	0.41	—	—	0.18	—	0.31	4.1
3.70	0.32	0.42	0.94	—	—	0.44	0.22	0.94	9.4
0.65	—	—	0.20	—	—	—	—	0.17	0.91
3.96	0.74	0.58	0.81	—	—	0.40	0.15	0.53	5.8
2.58	0.59	0.42	0.47	—	—	0.33	0.13	0.49	4.2
4.22	0.30	0.54	1.12	—	0.11	0.37	0.21	0.96	12.4
1.85	0.12	0.22	0.33	—	—	0.14	—	0.40	3.1
1.91	0.27	—	0.58	—	—	0.14	0.14	0.40	2.9
1.81	0.14	0.18	0.60	—	—	0.25	0.10	0.42	4.7
1.52	0.14	0.21	0.21	—	—	0.16	0.12	0.43	1.9
1.19	0.13	0.11	0.23	—	—	0.20	—	0.28	1.9
1.02	0.10	0.12	0.16	—	—	—	—	0.24	1.7
2.73	0.14	0.25	0.35	—	0.12	0.27	0.15	0.57	6.4
1.49	0.18	0.14	0.25	—	—	0.28	0.11	0.35	3.5
3.27	0.58	0.28	0.50	0.16	—	0.50	0.21	0.38	4.0
2.00	0.22	0.22	0.35	—	—	0.23	0.12	0.58	4.5
2.11	0.30	0.28	0.24	—	—	0.33	0.13	0.48	5.8
1.97	0.24	0.14	0.27	—	—	0.23	0.12	0.39	4.4
1.98	0.26	0.11	0.41	—	—	0.26	—	0.25	6.5
1.88	0.38	0.13	0.40	0.10	—	0.30	0.13	0.64	3.8
1.34	0.55	0.35	0.43	—	—	0.35	0.11	0.35	3.3
2.30	—	—	0.38	—	—	0.25	0.14	0.40	5.3
1.50	0.18	—	0.20	—	—	0.25	—	0.25	4.3
2.20	0.30	0.24	0.28	—	—	0.28	0.13	0.30	4.8
3.02	0.55	0.30	0.50	0.13	—	0.65	0.22	0.78	8.8
3.25	0.30	0.25	0.43	0.12	—	0.43	0.16	0.45	7.3
3.02	0.60	0.33	0.55	—	—	0.38	0.13	0.25	6.5
2.77	0.22	0.24	0.38	—	—	0.21	0.12	0.30	4.5
2.38	0.20	0.31	0.26	—	—	0.31	0.14	0.48	5.2
2.96	0.39	0.32	0.67	—	—	0.39	0.14	0.49	8.6
3.12	0.20	0.43	0.94	0.13	—	0.36	0.20	0.88	11.0
1.52	0.14	0.17	0.16	—	—	0.16	—	0.34	3.6
3.25	0.25	0.38	0.85	—	—	0.33	0.17	0.72	7.5
1.66	0.12	—	0.49	—	—	0.11	0.15	0.20	7.2
4.08	0.46	0.61	1.15	0.12	—	0.43	0.20	0.97	7.6
3.44	0.23	0.32	1.06	0.11	—	0.44	0.17	0.71	11.3
5.15	0.49	0.47	0.98	0.13	—	0.56	0.20	0.66	13.7
8.60	0.69	0.76	1.65	0.19	—	0.84	0.24	0.91	18.5
4.97	0.42	0.50	1.30	0.10	—	0.40	0.17	0.85	11.0
1.63	—	—	0.49	—	—	0.14	—	0.37	9.3
1.67	—	—	0.47	—	—	0.14	—	0.29	5.9
0.74	—	—	0.23	—	—	0.10	—	0.25	2.0
0.99	—	—	0.23	—	—	—	—	0.17	2.1
2.21	0.15	0.15	0.63	—	—	0.22	0.11	0.48	8.3
0.90	—	—	0.25	—	—	—	—	0.23	3.0
2.40	0.23	0.17	0.35	—	—	0.18	0.10	0.33	7.0
2.95	0.39	—	0.66	—	—	0.25	0.17	0.34	8.6
GM 2.16	0.21	0.18	0.43			0.23	0.10	0.40	4.80
CL 1.93-2.42	0.17-0.25	0.15-0.22	0.38-0.49			0.20-0.27	0.09-0.12	0.36-0.44	4.21-5.53
Range 0.65-8.60	ND-0.74	ND-0.76	0.16-1.65	ND-0.19	ND-0.13	ND-0.84	ND-0.22	0.13-0.97	0.91-18.5
Deveaux Bank									
2.39	0.32	0.29	0.83	0.13	—	0.54	0.20	0.66	5.8
1.72	0.12	0.21	0.59	—	—	0.14	0.10	0.56	4.0
1.43	—	0.14	0.47	—	—	0.16	0.11	0.49	3.2
0.82			0.25	—	—	—	—	0.23	5.0
3.26	0.22	0.48	0.54	—	—	0.19	0.15	0.54	4.6
2.03	0.14	0.25	0.57	—	—	0.16	0.11	0.55	3.9
3.44	0.29	0.34	0.69	—	—	0.27	0.23	0.79	2.9

(Continued next page)

TABLE 6 (cont'd.). *Residues of organochlorine pollutants in eggs of brown pelicans, South Carolina*—1973

Residues, μg/g fresh wet weight									
DDE	TDE	DDT	Dieldrin	Heptachlor Epoxide	Mirex	*Cis*-chlordane[1]	*Cis*-nonachlor	Toxaphene	PCB's
Deveaux Bank									
0.88	—	—	0 24	—	—	—	—	0 28	1 1
1 43	0 19	0 25	0 63	—	—	0 20	—	0 38	3.8
1 10	—	—	0 33	—	—	—	—	0 22	1 4
1 01	—	—	0 38	—	—	0 14	—	0 28	2.2
2.18	0 19	0 29	0 62	—	—	0 29	0 13	0 60	4 5
2.46	0.23	0 42	0 89	—	—	0 25	0 14	0 76	6 2
1.58	0 13	0 13	0 37	—	—	0 22	0 10	0 30	3 5
2.32	0 33	0.28	0 42	—	—	0 15	—	0 44	2 3
2.20	0 25	0.28	0 39	—	—	0 23	0 10	0 48	4 4
1.27	—	—	0.76	—	—	0 13	—	0 33	2 4
1.54	0.23	0.23	0 37	—	—	0 27	0 13	0 42	3 7
2.32	0.34	0.30	0 40	—	—	0 14	0 11	0 53	3 6
1.82	0.23	0.17	0.41	—	—	0 21	0 10	0 29	4 1
3.68	0.20	0.10	0.50	—	—	0 24	0 14	0 43	12 1
2.69	0.27	0.12	0 61	0 25	—	0.54	0 20	0 25	13 7
3.87	0.21	0.23	0.80	0 21	—	0 44	0 22	0 60	15 2
2.97	0.19	0.27	0.67	—	—	0 19	0 11	0 52	5 9
3.01	0.18	0 28	0 68	—	—	0 19	0 11	0 50	5 5
3 37	0.20	0.31	0 63	—	—	0 19	0 12	0 58	8 4
0.96	—	—	0 29	—	—	—	—	0 23	3 1
1 40	0 13	0 13	0 40	—	—	0 20	—	0 27	4 0
1.31	0.31	0 21	0 69	0.12	—	0 33	0 17	0 58	7 9
2.14	0.33	—	0.51	—	—	0 17	0 14	0 24	5 5
1.61	0.34	—	0 41	—	—	0 24	0 14	0 15	6 2
4 12	0.61	0 36	0 73	—	—	0 36	0 12	0 36	7 3
1.55	0 12	—	0 41	0.13	—	0 20	0 11	0 54	6 1
1 29	—	—	0 26	—	—	—	—	0 26	3 1
3.01	0.30	0 22	0 53	—	—	0 18	0 12	0 48	5 5
4 98	0.30	0 28	0 93	—	—	0 53	0 25	0 53	19 0
1.59	0 15	0 12	0 50	—	—	0 14	—	0 31	3 6
GM 1.97	0 16	0 15	0 50			0.23	0 10	0 40	4 62
CL 1.69-2.30	0.13-0.21	0 12-0 20	0.44-0 57			0 20-0.27	0 09-0.11	0.35-0 46	3 78-5.64
Range 0.82-4.98	ND-0 61	ND-0.48	0.24-0 93	ND-0.25	ND	ND-0 54	ND-0 25	0 15-0 79	1 1-19 0
Marsh Island and Deveaux Bank									
GM 2.09	0 19	0 17	0 45			0.21	0 10	0 40	4 75
CL 1.91-2 29	0.17-0 22	0 15-0 20	0 41-0 50			0 19-0 24	0 09-0 11	0.37-0 43	4 25-5 31
Range 0.65-8.6	ND-0.74	ND-0.76	0 16-1 65	ND-0 25	ND-0 13	ND-0 84	ND-0 25	0 13-0 97	0.91-19 0

NOTE: ND or — = no residue detected
GM = geometric mean
CL = 95 percent confidence limits
[1]*Cis*-chlordane and/or *trans*-nonachlor

Probable causes of deaths of pelicans are listed in Table 9. The female adult found dead in Georgia had myocardial necrosis, but she was also carrying a level of dieldrin in the brain that corresponds to a diagnostic lethal level (*33*). Residues of lead may produce myocardial necrosis if present at high levels, but the Georgia pelican with this disease carried very low levels of lead in its tissues. A pelican found dead in Florida Bay in 1973 apparently died of a gunshot wound. Most of those found dead in Florida in 1972 probably died of bacterial enteritis. *Clostridium perfringens*, a causative agent of hemorrhagic enteritis, was isolated from the gut of three of the Florida pelicans, but it was not determined whether the isolates were toxigenic. Recently, hemorrhagic enteritis in fish-eating birds has been connected with the presence of *Edwardsiella tarda* in the intestinal tract (*3,4*). Unfortunately, the pelican with enteritis was not tested for *E. tarda*.

Each egg and tissue of the brown pelican that was analyzed contained residues of 10 heavy metals (Tables 10 and 11). However, the significance of these metals to the pelicans is undetermined. Generally, there is a highly significant positive correlation between each organochlorine residue: DDE, TDE, DDT, dieldrin, and PCB's. In contrast, residues of metals are usually negatively correlated with residues of organochlorines (Table 12). Of the 10 metals, 5 demonstrated at least one significant negative correlation ($P<0.05$) with an organochlorine. Significant positive correlations ($P<0.05$) were found in comparing metals to each other: chromium and nickel, nickel and mercury, nickel and lead, and mercury and lead. Zinc, copper, cadmium, arsenic, and chromium had no definite tendencies for significant relationships with the organochlorines or with other metals. In contrast, there was a definite tendency for residues of nickel, selenium, mercury, lead, and magnesium to be negatively correlated with organochlorines and positively correlated with other metals. The significance or validity of these relationships of metals in eggs is difficult to determine because different metals may follow different pathways in the body of the bird. For example, females fed cadmium (*32*) or lead (*17*) laid eggs that contained small amounts of these metals.

TABLE 7. *Residues of organochlorine pollutants in rolled eggs of brown pelicans, South Carolina—1973*

Residues, μg/g fresh wet weight							
DDE	TDE	DDT	Dieldrin	*Cis*-chlordane[1]	*Cis*-nonachlor	Toxaphene	PCB's
2 92	—	—	0 18	—	—	0.14	6.8
1 31	0 16	0 16	0.37	0 16	—	0.25	4 0
1 93	0.18	0.23	0.33	0 14	—	0 30	3.8
2 89	0.55	0 36	0 63	0 29	0 11	0 46	6.0
1 47	0.12	—	0 40	0 14	—	0.30	4.3
2 10	0.25	0.19	0.50	0 19	—	0.25	5.1
2 20	0.22	0 19	0 46	0 13	—	0.20	4.1
2 76	0.16	0 33	0 45	0.18	0.11	0 55	9.6
2 07	0 32	0 20	0 44	0 16	—	0.23	4 9
1 89	0.15	0 11	0 47	0 11	0 11	0.39	2.7
2 46	0.25	0 24	0 75	0 30	0 13	0.58	7.5
GM 2.12	0 19	0 16	0 43	0 15		0.30	5.04
CL 1.78-2 52	0 12-0 28	0 10-0.25	0.33-0 55	0.11-0.21		0 23-0 41	3.96-6.41
Range 1 31-2 92	ND-0 55	ND-0.36	0.18-0 75	ND-0 30	ND-0.13	0.14-0.58	2.79-9.6

NOTE: ND or — = no residue detected
GM = geometric mean
CL = 95 percent confidence limits
[1]*Cis*-chlordane and/or *trans*-nonachlor

TABLE 8. *Residues of organochlorine pollutants in tissues of brown pelicans found dead*

Residues, μg/g fresh wet weight												
Sex	Age	Tissue	DDE	TDE	DDT	Dieldrin	Heptachlor Epoxide	Mirex	Oxy-chlordane	*Cis*-chlordane[1]	*Cis*-nonachlor	Toxaphene	PCB
South Carolina													
						1971							
M	12 wk	Carcass	0 47	0.19	0.10	0.10	0.10	—	—				4.0
		Brain	0.10	0.10	—	0.10	—	—					0.6
F	6 wk	Carcass	0 10	0.10	0.10	0.10	0 10	0.10	—				
		Brain	0 10	—	—	—	—	—					—
						1973							
F	8 wk	Carcass	0.48	0.14	0.11	0 13	—	—	—	—	—	—	0.7
		Brain	—	—	—	—	—	—	—	—	—	—	—
Georgia													
						1972							
F	AD	Carcass	5 50	0.78	—	5.30	0 16	0.73	0 15	0.43	0 45		10.0
		Brain	2.90	0.17	0.23	4 40	0.16	0.25	—	0 32	0 23		4.7
Florida													
						1972 (Atlantic Coast near Stuart)							
M	IM	Carcass	8 10	2.00	0 80	1 20	—	—	—	—	—		5.0
		Brain	2 70	0.39	0 33	0.82	—	—	—	—	—		3.2
M	IM	Carcass	5 80	1.30	0.46	0 90	—	0.23	—	0 10			5.0
		Brain	2 36	0.32	0.22	0 59	0 13	—	—	—			6.5
F	AD	Carcass	10 00	2.00	0.94	1 10	0 12	0.40	—	—			25.0
		Brain	0 36	—	—	—	—	—	—	—			0.5
M	IM	Carcass	6 70	1.64	0 55	0 97	0.20	0 28	—	—			15.0
		Brain	5 30	0 46	0 49	0 53	0 12	—	—	—			8.5
M	IM	Carcass	9 65	1 44	0 70	1 17	0 13	—	—	—			10.0
		Brain	4 00	0 75	0 38	0 96	—	0 12	—	—			9.4
						1973 (Florida Bay)							
M	IM	Carcass	1 38	—	—	0.21	—	0.12	—	0.26	—	—	0.9
		Brain	0 36	—	—	0 12	—	—	—	—	—	—	0.3
M	IM	Carcass	1 65	0.57	0 16	0 38	0 16	—	—	1 30	0.18	0.17	1.8
		Brain	0 10	—	—	—	—	—	—	—	—	—	0.3
M	AD	Carcass	0 91	—	—	0 15	—	—	—	—	—	—	1.5
		Brain	0 91	—	—	0 31	—	—	—	0.15	—	0.27	0.9
F	AD	Carcass	1 32	0 16	—	0.23	—	—	—	0 11	0.10	0.26	1.5
		Brain	0 32	—	—	—	—	—	—	—	—	—	0.5

NOTE: — = no residue detected
A blank space indicates that either the sample was not analyzed for that chemical or the chemical was not quantified
IM = fledged pelican that has not attained adult (AD) plumage
[1] *Cis*-chlordane and/or *trans*-nonachlor

Unlike the organochlorine residues, metal residues in pelican eggs from various parts of their geographic range showed little variation, but there were a few exceptions. Eggs from Louisiana contained 0.08 μg/g mercury (9), whereas eggs of Florida and South Carolina pelicans contained four to five times more (Table 10). Residues of arsenic were three times as high in eggs from South Carolina than in those from Florida. It is possible that differences in metal residues reflect local differences in contamination of pelican foods. There was no apparent difference in residues of 10 metals in livers of birds that were found dead compared with those that were shot. Indeed, differences in residues seemed more closely related to place of collection or age than to cause of death. Mercury residues varied more than residues of other metals; the highest residue was 56 times the lowest.

TABLE 9. *Probable causes of brown pelican mortality*

Sex	Age	State	Year	Probable Cause of Mortality
M	12 wk	SC	71	Overwhelming, acute bacteremia
F	5 wk	SC	71	Pneumonia
F	8 wk	SC	73	Open[1]
F	AD	GA	72	Myocardial necrosis and dieldrin poisoning
M	IM	FL	72[2]	Enteritis
M	IM	FL	72	Enteritis
F	AD	FL	72	Enteritis
M	IM	FL	72	Enteritis
M	IM	FL	72	Enteritis
M	IM	FL	73	Gunshot
F	IM	FL	73	Open
M	AD	FL	73	Combination of diseases including early air sacultis, early aspergillosis, and valvular endocarditis
F	AD	FL	73	Fractured humerus, possible gunshot

[1] No diagnosis could be made on the basis of necropsy.
[2] See text for further details.

Connors et al. (*13*) reported that mercury residues (wet weight) in livers of brown pelicans found dead ranged from 1.20 to 1.88 μg/g in California and from 5.14 to 17.36 μg/g in Florida. Residues in both eggs and livers of brown pelicans suggest that pelicans are exposed to less mercury in California or Louisiana than in either South Carolina or Florida.

DECLINE OF RESIDUES

Residues of *p,p'*-DDE, *p,p'*-TDE, *p,p'*-DDT, and dieldrin in brown pelican eggs significantly declined from 1969 through 1973 (Table 13). The rate of change in residues was different for each of the five organochlorines. There was a steep drop in TDE from year to year; the trend for DDT was somewhat erratic. Dieldrin residues dropped steeply until 1971 and then leveled off, whereas there was a significant decline of DDE from one year to the next except for a slight reversal in 1972. The PCB's reached a peak in 1972, then declined in 1973 to the lowest level in 5 years. There was a nine-fold decrease in TDE and 1.6- to 2.6-fold decreases in the other pollutants. Residue trends for certain other organochlorines are unavailable because either the residues of these compounds appear in pelican eggs infrequently, as with mirex and heptachlor epoxide, or the residue methodology needed to detect them was unavailable in 1971 or 1972, as with toxaphene, *cis*-chlordane and/or *trans*-nonachlor and *cis*-nonachlor.

TABLE 10. *Residues of heavy metals in brown pelican eggs*

Residues, μg/g fresh wet weight									
Cr	Ni	Se	As	Hg	Pb	Cd	Cu	Zn	Mg
South Carolina, 1971-72									
0.039	0.053	0.26	0.64	0.66	0.110	0.005	0.92	6.4	85
0.008	0.019	0.30	0.85	0.56	0.068	0.002	1.05	7.2	95
0.013	0.058	0.27	0.65	0.39	0.022	0.003	0.81	5.8	80
0.016	0.010	0.33	0.71	0.17	0.019	0.003	0.70	5.5	79
0.012	0.014	0.29	0.73	0.21	0.010	0.002	1.10	6.7	75
0.005	0.006	0.28	0.48	0.22	0.018	0.003	0.82	6.9	80
0.002	0.011	0.22	0.22	0.55	0.025	0.008	1.07	6.3	67
0.003	0.021	0.31	0.14	0.50	0.025	0.003	1.10	7.4	90
0.001	0.010	0.27	0.18	0.36	0.025	0.003	1.29	6.3	73
0.093	0.072	0.27	0.13	0.58	0.035	0.008	1.06	4.6	68
0.009	0.066	0.23	0.076	0.26	0.025	0.004	1.20	6.2	55
0.150	0.031	0.24	0.18	0.24	0.045	0.003	1.00	8.0	89
GM 0.011	0.022	0.27	0.31	0.36	0.029	0.004	1.00	6.4	77
CL 0.004-0.028	0.013-0.039	0.251-0.292	0.183-0.533	0.263-0.480	0.019-0.043	0.003-0.005	0.890-1.115	5.81-7.00	70.1-85.0
Range 0.001-0.15	0.010-0.072	0.22-0.33	0.076-0.85	0.17-0.66	0.010-0.11	0.002-0.008	0.70-1.29	5.5-8.0	55-95
Florida, 1969-70									
0.067	0.042	0.34	0.09	0.84	0.038	0.003	1.04	8.3	80
0.005	0.009	0.19	0.07	0.22	0.013	0.003	1.02	7.7	86
0.015	0.037	0.38	0.18	0.48	0.045	0.011	1.11	6.8	70
0.006	0.039	0.33	0.08	0.45	0.032	0.003	0.98	5.2	107
0.023	0.027	0.31	0.11	0.45	0.028	0.003	0.78	4.3	78
0.009	0.010	0.21	0.10	0.20	0.013	0.003	0.94	7.4	42
GM 0.014	0.023	0.28	0.10	0.39	0.025	0.004	0.97	6.4	74
CL 0.005-0.037	0.011-0.046	0.211-0.382	0.071-0.140	0.223-0.670	0.014-0.043	0.002-0.006	0.86-1.11	4.93-8.43	53.5-103.3
Range 0.005-0.067	0.009-0.042	0.19-0.38	0.07-0.18	0.20-0.84	0.013-0.045	0.003-0.011	0.78-1.11	4.3-8.3	42-107

[1] GM = geometric mean.
CL = confidence limits.

The decline in organochlorine residues in South Carolina can be attributed partly to local curtailment of these chemicals either for pest control or industrial uses. However, residue trends reflected by the migratory pelican and the migratory Atlantic menhaden (*Brevoortia tyrannus*), their chief prey fish, are influenced by conditions existing over a wide area. Most pelicans nesting in South Carolina winter farther south; about 10 percent winter in South Carolina, about 20 percent in Cuba or more southerly areas, and the remainder in Florida, almost exclusively on the Atlantic Coast (28).

Seven pools of regurgitated menhaden were collected in 1973; organochlorine residues were low (Table 14). Most DDT in menhaden is metabolized to DDE in the body of the pelican. The biomagnification of DDE from fish to

TABLE 11. *Residues of heavy metals in livers of brown pelicans*

Sex	Age[1]	State[2]	Year	Residues, µg/g fresh wet weight									
				Cr	Ni	Se	As	Hg	Pb	Cd	Cu	Zn	Mg
				Found Dead									
F	8 wk	SC	73	0.026	0.078	1.01	1.02	0.12	0.27	0.01	4.7	26	137
M	1M	FL	73	0.049	0.053	4.04	0.54	6.20	0.21	0.23	9.0	50	183
F	AD	GA	72	0.020	0.016	1.71	0.23	0.33	0.10	0.03	8.3	33	165
F	AD	FL	72	0.070	0.019	2.80	0.47	6.30	0.10	0.73	5.4	41	205
				Shot									
F	AD	FL	70	0.110	0.058	3.38	0.63	4.10	0.10	1.06	6.7	55	222
M	1M	FL	70	0.065	0.048	4.42	0.89	1.70	0.10	0.21	7.5	32	226
F	6 wk	SC	70	0.030	0.045	1.16	0.36	0.21	0.10	0.01	4.3	31	198
M	AD	SC	70	0.056	0.039	4.28	0.29	1.10	0.10	0.02	6.5	38	199

[1] 1M = fledged birds that have not attained full adult (AD) plumage
[2] SC = South Carolina; FL = Florida; GA = Georgia

TABLE 12. *Correlation matrix for residues of five organochlorines and ten heavy metals identified in 18 eggs of the brown pelican*

	Simple Correlation Coefficient (R)												
	TDE	DDE	Dieldrin	PCB's	Cr	Ni	Se	As	Hg	Pb	Cd	Cu	Zn	Mg
DDD	0.464	0.468*	0.744**	0.699**	0.288	0.522*	0.417	0.351	0.453	0.492*	0.269	0.055	0.379	−0.255
TDE		0.564*	0.612**	0.439	0.103	0.374	0.650**	0.320	0.466	0.689**	0.146	0.095	0.199	0.484*
DDT			0.444	0.185	0.39	0.627**	0.276	0.52	0.179	0.461	0.089	0.095	0.175	−0.283
Dieldrin				0.741**	0.238	0.486*	0.636**	0.282	0.617**	0.501*	0.111	0.107	0.016	0.482*
PCB's					0.029	0.262	0.585*	0.303	0.271	0.197	0.123	0.121	0.193	0.343
Cr						0.590**	0.165	0.008	0.160	0.397	0.125	0.268	0.065	0.060
Ni							0.247	0.173	0.551*	0.563*	0.332	0.134	0.298	0.089
Se								0.221	0.373	0.322	0.047	0.153	0.225	0.414
As									0.100	0.146	0.069	0.363	0.001	0.241
Hg										0.701**	0.351	0.257	0.117	0.292
Pb											0.318	0.094	0.031	0.376
Cd												0.160	0.254	0.281
Cu													0.345	0.147
Zn														0.013

NOTE: **P = 0.05, *P = 0.01

TABLE 13. *General decline in residues of organochlorine pollutants in eggs of the brown pelican*

Year	Sample Size	Residues, µg/g fresh wet weight				
		DDE	TDE	DDT	Dieldrin	PCB's
1969	15	5.45 A[1] (4.44-6.70)	1.65 A (1.30-2.10)	0.22 A (0.09-0.54)	1.16 A (1.03-1.52)	6.11 ABC (5.00-7.45)
1970	13	3.58 B (2.23-5.72)	0.79 B (0.53-1.20)	0.29 B (0.38-0.69)	0.82 A (0.52-1.32)	5.25 ABC (3.91-7.04)
1971	65	2.48 C (2.27-2.71)	0.48 C (0.43-0.53)	0.13 A (0.11-0.17)	0.46 B (0.40-0.52)	6.49 AB (5.44-7.73)
1972	72	3.03 B (2.70-3.40)	0.36 D (0.31-0.42)	0.15 A (0.13-0.18)	0.45 B (0.39-0.52)	7.51 A (6.68-8.46)
1973	104	2.09 D (1.91-2.29)	0.19 E (0.17-0.22)	0.17 A (0.15-0.20)	0.45 B (0.41-0.50)	4.75 C (4.26-5.31)

[1] Geometric mean with 95 percent confidence limits in parentheses.
[2] A significant difference ($P = 0.05$) among means for each chemical is indicated for those means not sharing a common letter. Means were separated using Duncan's New Multiple Range Test (11) with Kramer's Extension for Unequal Replication (Vogt 1971-73).

pelican eggs is 31 times, only 5 times for both DDT and TDE, and 18 times for DDT and metabolites (DDTR). The biomagnification from fish to pelican eggs is 29 times for PCB's and 23 times for total organochlorines (Figure 1). The migratory habits of the Atlantic menhaden (*15,27*) and the brown pelican confound the significance of biomagnification noted in this study. Residues of DDTR in South Carolina menhaden averaged 0.295 μg/g in the late 1960's (Philip A. Butler, U.S. Environmental Protection Agency, Gulf Breeze, Fla., 1974: personal communication); residues in menhaden have apparently declined over 50 percent from the late 1960's to 1973.

Declining residues of certain organochlorine pesticides and their metabolites were noted in the United States and several other countries over the last few years; the authors have a list of more than 50 literature references concerning residue declines in human food, wild birds, fish, invertebrates, water, soils, and agricultural products.

Johnston (*22,23*) documented the decline of DDTR in passerine birds in Florida and cited several references concerning the decline of organochlorine pesticides and their metabolites in other areas. Butler (*12*) had previously shown a drastic decline in DDTR in oysters (*Crassostrea virginica*) in South Carolina and other States in the 1960's. Investigations have shown both increases and decreases in residues of PCB's in birds in the early 1970's. The only major change involved a significant increase of PCB's in eggs of the osprey (*Pandion haliaetus*) that were collected in Maryland from 1969 through 1973 (*11*).

REPRODUCTIVE SUCCESS AND POPULATION LEVEL

The reproduction record of pelicans in South Carolina was excellent in 1973; 1.66 young were fledged per nest and a total of 2,726 young were fledged in the State (Table 15). From 1969 through 1972, the average number of young fledged per nest ranged from 0.69 to 0.92. For a number of years before 1969, Beckett (*3*) estimated that reproductivity was subnormal. According to Henny (*18*), an average of 1.2 to 1.5 young must be fledged by each breeding female each year in order to maintain a stable population. Thus reproductivity in South Carolina brown pelicans has been subnormal for a number of years, and the population has been declining (*3*). The improvement in

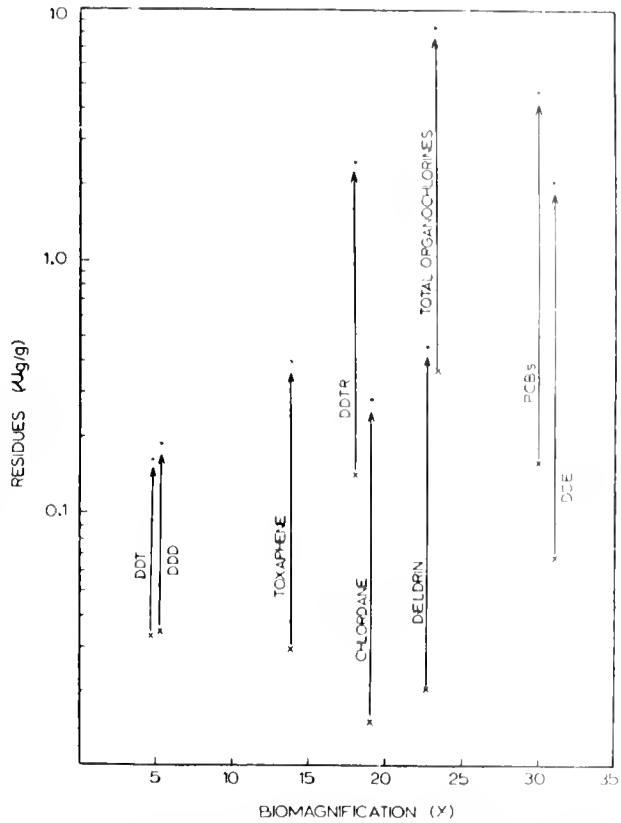

FIGURE 1. *Biomagnification of organochlorine residues from fish (X) to brown pelican eggs (O)*

TABLE 14. *Residues of organochlorine pollutants in Atlantic menhaden regurgitated by brown pelicans, South Carolina—1973*

	Residues, μg/g fresh wet weight						
	DDE	TDE	DDT	Dieldrin	Oxychlordane	*Cis*-chlordane[1]	Toxaphene	PCBs
	0.04	0.04	0.04	0.03	0.01	—	0.03	0.08
	0.06	0.04	0.05	0.03	0.01	0.01	0.04	0.17
	0.07	0.02	0.03	0.02	—	—	0.02	0.25
	0.06	0.03	0.03		—	0.01	0.04	0.14
	0.05	0.03	0.02	0.02	—	0.01	0.02	0.10
	0.08	0.03	0.02	0.02	—	0.01	0.02	0.25
	0.15	0.07	0.06	0.04	—	0.02	0.04	0.24
GM	0.067	0.035	0.033	0.020			0.029	0.161
CL	0.045-0.099	0.024-0.050	0.022-0.049	0.011-0.038			0.021-0.039	0.105-0.248
Range	0.04-0.15	0.02-0.07	0.02-0.06	ND-0.04	ND-0.01	ND-0.02	0.02-0.04	0.08-0.25

NOTE: ND or — no residue detected
GM geometric mean
CL 95 percent confidence limits
[1] *Cis*-chlordane and/or *trans*-nonachlor

TABLE 15. *Reproductive success of brown pelicans in South Carolina*

Year	Colony	Number of Nests	Number of Young Fledged	Young Fledged per Nest
1969	Cape Romain	1016	900	0.82
	Deveaux Bank	*250*	*80*	*0.32*
	Both colonies	1266	980	0.78
1970	Cape Romain	637	500	0.78
	Deveaux Bank	*479*	*445*	*0.93*
	Both colonies	1116	945	0.85
1971	Cape Romain	1094	949	0.87
	Deveaux Bank	*375*	*400*	*1.07*
	Both colonies	1469	1349	0.92
1972	Cape Romain	763	514	0.67
	Deveaux Bank	*652*	*456*	*0.70*
	Both colonies	1415	970	0.69
1973	Cape Romain	836	1082	1.29
	Deveaux Bank	*810*	*1644*	*2.03*
	Both colonies	1646	2726	1.66

reproduction in 1973 was probably related to a decline in residues inasmuch as authors previously found strong correlative evidence that DDE inhibited reproductivity in the brown pelican (*10*). In addition, excellent conditions of tides, weather, and food supply favored the successful reproductive effort.

Conclusion

Residues of organochlorines in brown pelican eggs de creased from 1969 through 1973. In 1973, the first time i many years, pelicans reproduced well. The improved re productivity seemed related to the decline in or ganochlorine residues and favorable conditions of tide weather, and food supply.

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BRIEF

Blood Levels of Chlorinated Hydrocarbon Residues in the Population of a Continental Town in Croatia (Yugoslavia)[1]

Elsa Reiner, Blanka Krauthacker, Marinko Stipcevic, and Zlata Stefanac

ABSTRACT

Plasma or serum samples of 147 individuals from the general population of a continental town in Croatia (Yugoslavia) were analyzed for chlorinated hydrocarbons in 1975. The compounds were determined by gas chromatography and identified by comparing their retention times with those of known standards. All samples contained p,p'*-DDE; mean concentration was 35.3 ppb. Only 20 samples contained* p,p'*-DDT; mean concentration was 22.7 ppb. Concentrations of α-BHC, lindane, and* p,p'*-TDE were 3.25, 4.09, and 11.6 ppb, respectively.*

Introduction

No published data seem to be available on the blood content of chlorinated hydrocarbon residues in the population of Yugoslavia. The use of chlorinated hydrocarbons has recently been restricted in this nation, and it seemed reasonable for the authors to initiate a survey which should establish the present body burden of chlorinated hydrocarbon pesticides and provide the base for a followup study. A total of 147 individuals in a continental town in Croatia were selected, and their plasma or serum was analyzed for compounds listed in Table 1. Lindane and *p,p'*-DDT, which appear in Table 1, are recommended for use as active ingredients in formulations used for plant protection in Yugoslavia (*4*).

TABLE 1 *Concentrations of chlorinated hydrocarbons in 147 samples of human plasma or serum, Croatia (Yugoslavia)—1975*

Compound	Concentration, ppb	
	Mean ± SEM (N)[1,2]	Range
α-BHC	3.25 ± 0.50 (57)	0.11 - 15
Lindane	4.09 ± 0.62 (23)	0.45 - 15
p,p'-DDE	35.3 ± 1.5 (147)	8.4 - 118
p,p'-TDE	11.6 ± 2.7 (7)	3.0 - 23
p,p'-DDT	22.7 ± 3.3 (20)	2.2 - 81

[1] N = no. samples which contain compound
[2] SEM = standard error of mean

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Sampling and Analysis

The presence of chlorinated hydrocarbons was determined b gas chromatography. Compounds were extracted within 2 hours after sampling according to the procedure of Dale et a (*1*). From each individual, two 1.0-ml samples acidified wi formic acid were extracted four times with 3 ml hexane. Th combined extracts were purified on a florisil column containin 1 g florisil and approximately 1 g anhydrous Na_2SO_4. Th hexane was evaporated from the eluates and the compound were redissolved in 1.0 ml hexane.

Conditions and operating parameters for GC analysis were:

Instrument Varian 2800 gas chromatograph

Detector H^3-Sc electron-capture

Columns Glass, packed with

A. 1.95 percent QF-1
1.5 percent OV-17 on Gas-Chrom Q

B. 4 percent SE-30 and 6 percent DC-210 on Gas-Chrom Q 80/100 mesh

C. 3 percent QF-1 and 6 percent DC-200 on Varaport 30

The best separation was obtained on column B. On all column compounds were identified by comparing their retention tim with those of known standards. Solutions of standards hexane were treated in the same way as the hexane extract Although purification of extracts on a florisil column is n included in the procedure of Dale et al. (*1*), it was necessary this study because fatty acids were found in the extract. It w shown, however, that no chlorinated hydrocarbons were lost l that procedure.

Results and Discussion

Samples of plasma (92) or serum (55) of 147 individuals we analyzed for the presence of chlorinated hydrocarbons. The te group included 65 males and 82 females; their mean age was

ears, ranging from 8 to 92 years. Blood samples were obtained om clinical laboratories during 1975. To the authors' knowldge, none of the individuals had had previous occupational or ccidental exposure to chlorinated hydrocarbons or pesticides i general. All samples contained *p,p'*-DDE, but *p,p'*-DDT was ound in only 20 samples (Table 1). In five of the 20 samples the oncentration of *p,p'*-DDT was higher than *p,p'*-DDE; in the ther 15 samples the reverse was true. The mean ratio of *p,p'*-DE to *p,p'*-DDT in these 20 samples was 2.3. Like *p,p'*-DDT, e other compounds were not found in all samples analyzed. he concentration range for a given compound is large and the oncentrations correspond to data published for other popula-ons (*2,3*).

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APPENDIX

Chemical Names of Compounds Discussed in This Issue

AROCLOR	A mixture of chlorinated terphenyls
AROCLOR 1260	PCB, approximately 60% chlorine
BHC (Benzene Hexachloride)	1,2,3,4,5,6-Hexachlorocyclohexane (mixture of isomers)
CHLORDANE	1,2,3,5,6,7,8,8-Octachloro-2,3,3a,4,7,7a-hexahydro-4,7-methanoindene. The technical product is a mixture of several compounds including heptachlor, chlordene, and two isomeric forms of chlordane
DDE	Dichlorodiphenyl dichloro-ethylene (degradation product of DDT) *p,p'*-DDE: 1,1-Dichloro-2,2-bis(*p*-chlorophenyl) ethylene *o,p'*-DDE: 1,1-Dichloro-2-(*o*-chlorophenyl)-2-(*p*-chlorophenyl)ethylene
DDT	Main component (*p,p'*-DDT): α-Bis(*p*-chlorophenyl)-β,β,β-trichloroethane Other isomers are possible and some are present in the commercial product *o,p'*-DDT: [1,1,1-Trichloro-2-(*o*-chlorophenyl)-2-(*p*-chlorophenyl) ethane]
DIELDRIN	Not less than 85% of 1,2,3,4,10,10-Hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4-*endo*-*exo*-5,8-dimethanonaphthalene
HCB	Hexachlorobenezene
HEPTACHLOR EPOXIDE	1,4,5,6,7,8,8-Heptachloro-2,3-epoxy-3a,4,7,7a-tetrahydro-4,7-methanoindan
LINDANE	*Gamma* isomer of 1,2,3,4,5,6-hexachlorocyclohexane
MIREX	Dodecachlorooctahydro-1,3,4-metheno-1H-cyclobuta[cd]pentalene
NONACHLOR	1,2,3,4,5,6,7,8-Nonachlor-3a,4,7,7a-tetrahydro-4,7-methanoindan
PCBS (Polychlorinated Biphenyls)	Mixtures of chlorinated biphenyl compounds having various percentages of chlorine
SELENIUM	Sodium selenate
EDE	2,2-Bis (p-chlorophenyl)-1,1 dichloroethane (including isomers and dehydrochlorination products)
TOXAPHENE	Chlorinated camphene (67-69% chlorine), product is a mixture of polychlor bicyclic terpens with chlorinated camphenes predominating

ERRATA

PESTICIDES MONITORING JOURNAL, Volume 10, Number 2, pages 41-43. Authors of the article "Pesticide Residues in Urban Soils from 14 United States Cities, 1970" are H. S. C. Yang, G. B. Wiersma, H. Tai, and W. G. Mitchell.

PESTICIDES MONITORING JOURNAL, Volume 10, Number 4, page 168. The following table was omitted from the brief, "DDT Residues in Air in the Mississippi Delta, 1975."

TABLE 1. *Monthly atmospheric levels of ΣDDT in the Mississippi Delta, 1972-75*

	Residues, ng/m³			
	1972	1973	1974	1975
January	10.8	3.9	3.0	2.2
February	12.6	4.8	3.6	3.8
March	32.6	11.1	7.6	2.7
April	34.1	11.4	7.7	3.5
May	17.2	18.6	15.6	9.2
June	16.2	49.5	12.8	13.8
July	117.2	9.6	24.3	11.9
August	515.3	25.6	37.9	19.0
September	378.8	24.6	19.4	18.4
October	37.6	18.9	5.1	2.6
November	14.8	11.9	3.3	2.3
December	6.3	2.4	2.1	1.3
Arithmetic mean	99.5	16.0	11.9	7.6
Geometric mean	35.0	11.8	8.1	5.1

The *Pesticides Monitoring Journal* welcomes from all sources qualified data and interpretative information on pesticide monitoring. The publication is distributed principally to scientists, technicians, and administrators associated with pesticide monitoring, research, and other programs concerned with pesticides in the environment. Other subscribers work in agriculture, chemical manufacturing, food processing, medicine, public health, and conservation.

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Monitoring is defined here as the repeated sampling and analysis of environmental components to obtain reliable estimates of levels of pesticide residues and related compounds in these components and the changes in these levels with time. It can include the recording of residues at a given time and place, or the comparison of residues in different geographic areas. The Journal will publish results of such investigations and data on levels of pesticide residues in all portions of the environment in sufficient detail to permit interpretations and conclusions by author and reader alike. Such investigations should be specifically designed and planned for monitoring purposes. The Journal does not generally publish original research investigations on subjects such as pesticide analytical methods, pesticide metabolism, or field trials (studies in which pesticides are experimentally applied to a plot or field and pesticide residue depletion rates and movement within the treated plot or field are observed).

Authors are responsible for the accuracy and validity of their data and interpretations including tables, charts, and references. Pesticides ordinarily should be identified by common or generic names approved by national or international scientific societies. Trade names are acceptable for compounds which have no common names. Structural chemical formulas should be used when appropriate. Accuracy, reliability, and limitations of sampling and analytical methods employed must be described thoroughly, indicating procedures and controls used, such as recovery experiments at appropriate levels, confirmatory tests, and application of internal standards and interlaboratory checks. The procedure employed should be described in detail. If reference is made to procedures in another paper, crucial points or modifications should be noted. Sensitivity of the method and limits of detection should be given, particularly when very low levels of pesticide residues are being reported. Specific note should be made regarding correction of data for percent recoveries. Numerical data, plot dimensions, and instrument measurements should be reported in metric units.

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The Monitoring Panel consists of representatives of the Agricultural Research Service, Animal and Plant Health Inspection 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 Monitoring Panel which participate in operation of the national pesticides monitoring network, are expected to be the principal sources of data and articles. However, pertinent data in summarized form, together with 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. Publication of scientific data, general information, trade names, and commercial sources in the *Pesticides Monitoring Journal* does not represent endorsement by any Federal agency.

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CONTENTS

Volume 11 September 1977 Number 2

PESTICIDES IN PEOPLE

Effects of Reducing DDT Usage on Total DDT Storage in Humans

F. W. Kutz,[1] A. R. Yobs,[2] S.C. Strassman,[1]
and J.F Viar, Jr.[3]

ABSTRACT

Agricultural uses of the insecticide DDT were cancelled by the U.S. Environmental Protection Agency December 31, 1972. However, the domestic use of DDT had begun to decline before this action. Beginning July 1969, residues of DDT and its metabolites were measured in human adipose tissue collected through an annual national survey. Levels of total DDT equivalent residues in human adipose have decreased slightly, but the frequencies of finding DDT or its metabolites have remained high. The most marked decline in residue concentration has been found in the youngest age group (0–14 years). Approximately 80 percent of the total DDT equivalent found in this survey was DDE. These data show that the reduction of the agricultural uses of DDT has decreased human exposure to and storage of this chemical.

Introduction

Agricultural uses of the insecticide DDT were cancelled by the U.S. Environmental Protection Agency (EPA) December 31, 1972. This cancellation order culminated approximately three years of intensive administrative inquiries into all uses of DDT. Most evidence indicated that continued general use of DDT represented an unacceptable risk to humans and their environment. The order did not, however, prohibit the continued use of DDT in controlled situations such as public health programs, quarantine programs, and other official uses in isolated instances (37 FR 13369).

Even prior to this action, DDT use in the United States had declined greatly (Figure 1). Peaking at approximately 80 million pounds in 1959, DDT use decreased to less than 12 million pounds by 1972. Since the 1972 ban, DDT has been used only in emergencies.

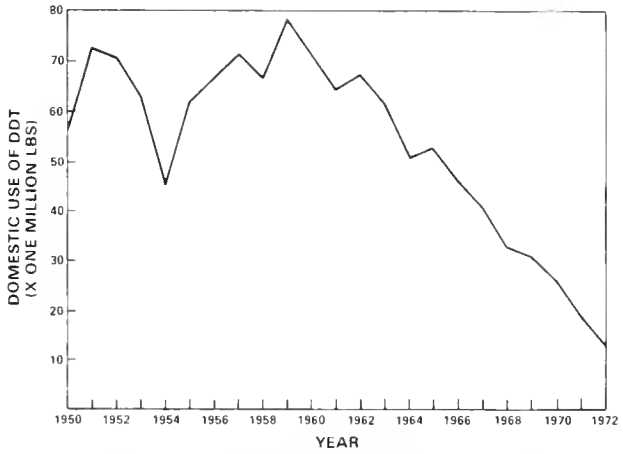

FIGURE 1 *Domestic use of DDT by year, 1950–72*
(D. L. Fowler, Control Stabilization and Conservation Service, U.S. Department of Agriculture, 1975: personal communication.)

Proof that DDT residues were present in human tissues collected over an extensive geographical area contributed to the cancellation decision. Considerable data on residue levels in humans in the United States were provided by the National Human Monitoring Program for Pesticides. This EPA program monitors residue storage, excretion, and metabolic effects in humans on a national scale in order to estimate the incidence and level of exposure to pesticides experienced by the general population and to identify changes and trends in this exposure.

This paper reports survey findings of total DDT equivalent during fiscal years 1970–74. The term, DDT equivalent, reflects the total burden of DDT and its analogs and compensates for differences in molecular weights of these compounds.

Methods and Materials

The Human Monitoring Survey during these five years collected samples of adipose tissue for chemical analysis. Tis-

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sites were obtained through cooperating medical pathologists at sites selected on the basis of an experimental design covering the conterminous 48 States.

SURVEY DESIGN

A proportionate, stratified-random sampling design was followed in selecting cities for sample collection. The population strata for the surveys conducted in fiscal years 1970–72 were the four census regions provided by the 1960 census. 39 collecting sites were used. The population strata for the surveys conducted in fiscal years 1973 and 1974 were the nine census divisions provided by the 1970 census. 75 collecting sites were used, an increase of 66 percent over the earlier survey. A fiscal year begins July 1 and ends June 30 of the year used for identification; i.e., FY 1970 extends from July 1, 1969, to June 30, 1970. The sampling units in each annual survey were cities with populations above 25,000 people. The number of sites needed in each area was based on the proportion of the national population in that area. Cities which served as collecting sites were selected at random.

The design provided samples which statistically represented the general population. For each collection site, an annual sample quota was established which reflected the demographic distribution in that census region or division. This quota in each group was proportioned according to age, sex, and race distribution of the population.

Samples were collected by cooperating pathologists and medical examiners from postmortem examinations and from specimens which had been removed during therapeutic and elective surgery. Thus tissues were received from patients with pathological conditions of varying severity and duration and from victims of sudden death from trauma or acute illness. Age, sex, race, and pathological diagnosis of the donor were recorded. Geographic residence was assumed to be the general location of the hospital. Since the objective of the program was to reflect pesticide levels in the general population, samples were not collected from patients with a suspected or established diagnosis of pesticide poisoning, from patients exhibiting cachexia, or from patients who had been institutionalized for extended periods. Further details of the program have been presented by Yobs (6) and Kutz (7).

CHEMICAL ANALYSIS

Samples were analyzed chemically by contract laboratories using methods specified by the program. Laboratories were required to maintain acceptable performance levels of an interlaboratory quality assurance program established and moderated by the EPA Pesticides and Toxic Substances Effects Laboratory, Research Triangle Park, N.C. The laboratory also provided technical assistance for the analytical portion of the program.

Samples were analyzed for selected chlorinated hydrocarbon insecticides using a modified Mills-Olney-Gaither procedure (5). A 5-g sample of human adipose tissue was dry macerated with sand and disodium sulfate and the lipid material was isolated by repetitive extractions with petroleum ether. Pesticide residues were extracted from the lipid material with acetonitrile and then partitioned back into petroleum ether by aqueous dilution of the acetonitrile extract. The petroleum ether extract was concentrated to 5 ml by Kuderna-Danish evaporation and transferred to a florisil column for successive elutions with 6 percent and 15 percent ethyl ether–petroleum ether. The eluates were concentrated in Kuderna-Danish evaporators and the final extracts were examined by electron-capture gas chromatography. Recovery studies indicated that all DDT isomers and analogs were recovered completely, above 80 percent. Thus the data were not corrected for recovery. Confirmatory techniques included thin-layer chromatography, Coulson detectors, microcoulometry and, in some cases, combined gas chromatography–mass spectrometry.

COMPUTATION AND DESCRIPTIVE STATISTICS

Residue values presented in this paper were calculated on a percent-lipid basis. Conversion was made for each specimen by dividing the whole tissue (wet-weight) value by the proportion of lipid extractable material. This method of reporting reduced the inherent variation in residue data which resulted from differences in the lipid content of individual adipose tissue specimens.

Data were characterized by several statistical parameters: sample size or total number of samples analyzed, frequency as percent positive, and geometric mean.

In calculating the descriptive statistics, only quantifiable amounts of pesticides were considered; trace amounts were converted to zero prior to data processing. Since these residue data were not normally distributed statistically, the geometric mean was considered a reliable measure of central tendency.

Results and Discussion

DDT is stored in greater amounts than are other organochlorine pesticides and is found in almost every adipose tissue sample analyzed (7).

DDT is either dechlorinated in the human body to TDE and then metabolized to the water soluble and excretable DDA, or it is excreted directly as DDT. DDE storage is not appreciably derived from ingestion of DDT, but rather by intake of DDE previously degraded in the environment from DDT. Since DDE is not effectively eliminated from the body, the result is a gradual increase in the body burden of this chemical. On the other hand, DDT is broken down and excreted more rapidly than DDE and thus is more responsive to changes in exposure level than are the other analogs (3, 4).

The annual geometric mean levels of the national survey and the data stratified by age group are presented in Table 1. Sample size and frequency of detecting total DDT equivalent residues are shown in Table 2. The data indicated a nationwide decrease in total DDT equivalent residue concentration. A more marked decrease in concentration was apparent in the youngest age group (0–14 years). Residues decrease noticeably in all age groups when data are stratified by age group. The frequency of detecting DDT and its transformation products in human adipose tissue has remained high, practically 100 percent in all groups and years.

Studies by Kutz et al. (2) showed that residues of DDT and its analogs are racially stratified. Samples from Negroes contained almost twice as much total DDT equivalent residues as did samples from Caucasians. In light of this fact, data in the present study were further stratified by race and age group (Table 3). The five-year trend toward decreasing levels of total DDT equivalent was still apparent for each racial group. Samples from Negroes collected in FY 1971 showed higher levels than did those in FY 1970, but the trend toward decrease compensated for this slight rise over the longer time period.

Conclusions

The concentration of residues of total DDT equivalent in human adipose tissue has decreased slightly; however, the frequency of finding DDT and its analogs has remained high. The slight decline in concentration may be due to reduced human exposure to DDT as a result of decreased use.

TABLE 1. *Total DDT equivalent residues in human adipose tissue from general population, United States*

Age, Yr	FY 1970	FY 1971	FY 1972	FY 1973	FY 1974
	Residues, ppm Lipid Weight				
0–14	4.47	3.74	3.03	2.63	2.32
15–44	7.53	8.29	7.23	6.23	5.46
45 and above	8.88	8.64	7.96	7.13	6.97
National Summary	7.88	7.95	6.88	5.89	5.02

NOTE: Total DDT equivalent = (o,p'-DDT + p,p'-DDT) + 1.114 (o,p'-TDE + p,p'-TDE + p,p'-DDE + o,p'-DDE)

Residues expressed are geometric means

TABLE 3. *Total DDT equivalent residues in human adipose tissue from general population by race, United States*

Age, Yr	FY 1970	FY 1971	FY 1972	FY 1973	FY 1974
	Residues, ppm Lipid Weight				
	Caucasians				
0–14	4.16	3.32	2.79	2.59	2.15
15–44	6.89	6.56	6.01	5.71	4.91
45 and above	8.01	7.50	7.00	6.63	6.55
	Negroes				
0–14	5.54	7.30	4.68	3.16	4.02
15–44	10.88	13.92	11.32	9.97	9.18
45 and above	16.56	19.57	15.91	14.11	11.91

NOTE: See Table 1 for definition of total DDT equivalent
Residues expressed are geometric means

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TABLE 2. *Sample size and frequency of detecting total DDT equivalent residues in human adipose tissue, United States*

	FY 1970		FY 1971		FY 1972		FY 1973		FY 1974	
	Sample Size	Percent Positive	Sample Size	Percent Positive	Sample Size	Percent Positive	Sample Size	Percent Positive	Sample Size	Percent Positive
Age, Yr [1]										
0–14	134	100	133	100	228	100	155	100	194	100
15–49	476	99.78	560	99.64	602	100	405	100	333	100
50 and above	802	100	921	99.78	1090	99.90	532	100	371	100
National Summary	1412	99.83	1616 [2]	99.75	1920	99.95	1092	100	898	100

NOTE: See Table 1 for definition of total DDT equivalent
[1] Age breakdown for FY 1973 and FY 1974 was 0–14, 15–44, and 45 and above
[2] Summary of sample size in FY 1973 and FY 1974 reflects addition of two individuals whose ages were not known

RESIDUES IN FISH, WILDLIFE, AND ESTUARIES

Mirex Residues in Bobwhite Quail After Aerial Application of Bait for Fire Ant Control, South Carolina—1975–76 [1]

Ronald J. Kendall,[2] R. Noblet,[2] J. D. Hair,[2] and H. B. Jackson [3]

ABSTRACT

*Mirex, the organochlorine compound used for control of the imported fire ant (*Solenopsis invicta *Buren), was applied aerially under supervision of the South Carolina Plant Pest Regulatory Service in October 1975 to a game management area in Hampton County, S.C. Influenced by recent reports indicating that low levels of mirex were toxic to certain nontarget organisms in laboratory studies, authors initiated a program for monitoring mirex residues in bobwhite quail (*Colinus virginianus*). Pretreatment residues were recorded on a dry-weight basis in bobwhite quail breast and adipose tissue; conversion factors for determining wet-weight concentrations are approximately as follows: fat, 0.77, and breast, 0.29. Residues ranged from 0.000–0.178 ppm and 0.247–2.763 ppm, respectively. Mirex residues in quail adipose tissue showed up to five-fold increase within the first month after treatment and declined thereafter.*

A residue peak was noticed the spring following mirex treatment, corresponding with insect emergence. Mirex residues in quail collected in summer 1976 following a fall bait application showed slightly higher residue levels than had birds taken in summer 1975; however, little, if any, human food chain concentration would result in the consumption of birds with residue [illegible] and in this study.

Introduction

Mirex, a polycyclic chlorocarbon, is the toxic ingredient in the bait used to control the red imported fire ant (*Solenopsis invicta* Buren) in the southeastern United States. The bait is generally applied aerially in a dosage prescribed by the U. S. Department of Agriculture (USDA) (*12*) to infested areas and becomes available to foraging fire ants. Nontarget species contact mirex by direct consumption of the bait, by scavenging on contaminated carcasses of poisoned ants, and/or by consuming organisms which have concentrated mirex from the environment (*13*).

Mirex has an affinity for lipids and, like certain chlorinated hydrocarbons, is stored and accumulated in the adipose portion of animal tissues (*10*). The persistent nature of mirex is indicated by Kaiser, who detected mirex in fish from Lake Ontario (*9*); however, the contamination source was probably a chemical manufacturer which produced mirex in that area. The movement and accumulation characteristics of mirex are responsible for the widespread presence of this chemical in animal life, especially in avian predators (*3*).

In monitoring mirex residues in nontarget organisms after treatment, Naqvi and de la Cruz (*13*) noted that concentrations of the pesticide in various trophic levels indicated biological magnification. Baeteke et al. (*1*) found significant levels of mirex in wildlife collections one year after treatment; bobwhite quail (*Colinus virginianus*) had up to 3.148 ppm mirex in adipose tissue.

The proposed mirex treatment area had a severe fire ant infestation while serving as game management land for bobwhite quail hunting. The authors wanted to measure residues in quail available for human consumption because the birds' food habits (*14*) allow frequent contact with mirex. Therefore, this study was initiated to monitor mirex residues in bobwhite quail before and after treatment.

[illegible]

[2] [illegible] South Carolina Agricultural Experiment Station [illegible] [3] [illegible] from the South Carolina Plant Pest Regulatory Service.

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Methods and Procedures

SAMPLE AREA AND APPLICATION OF MIREX

Mirex bait (1.7 g mirex/acre in 1.25 lb. corncob grit/soybean oil bait) was applied aerially to the James W.

Webb Wildlife Center, Hampton County, S.C., under supervision of the South Carolina Plant Pest Regulatory Service (SCPPRS). This area is managed by the South Carolina Wildlife and Marine Resources Department (SCWMRD). A 1400-acre tract, excluding the ponds, of the 6000-acre quail plantation was treated. Various habitats were represented in the study area, including pine stands, hardwood forest, broom sedge fields, and farms. Application and collection dates are listed in Table 1.

SAMPLE COLLECTION

The SCWMRD permitted as many as eleven pretreatment quail and five posttreatment quail to be taken from the sprayed area each month. The bobwhites were shot, labeled, and transported to the laboratory on ice and frozen at 0°C until they were thawed and various tissues were removed.

EXTRACTION

Two tissues, fat (50–200 mg) and breast muscle (200–500 mg), were sampled from each bird, weighed, freeze-dried, weighed again, and ground with mortar and pestle using granular sodium sulfate as an abrasive. Mirex was extracted from breast muscles with hexane and from fat samples with petroleum ether. After grinding, the samples were collected in 125-ml flasks and fat samples were lightly boiled on a steam bath for 1 minute. All samples were then shaken on a wrist-action shaker for 1 hour.

CLEANUP

Extracts were cleaned by using standardized florisil column chromatography. Ten cm of 60/100-mesh PR grade florisil topped with 2 cm of anhydrous sodium sulfate was placed in a 1-cm-ID glass column fitted with a fritted glass disc. The florisil had been activated by oven heating at 150°C for 3 hours.

Columns were prewashed with 40 ml petroleum ether, and the washings were discarded. The extract was filtered through Whatman No. 40 paper to remove tissue debris, placed on the column, and allowed to percolate into the florisil; then the mirex was eluted with 40 ml of 6 percent ethyl ether in petroleum ether. The columns were stripped with 40 ml of a 15 percent ethyl ether in petroleum ether solution before introduction of the next sample. Three samples were cleaned on each column. The eluants were collected in 80-ml glass beakers, evaporated to dryness on a steam bath, and transferred into aluminum-lined cap sample vials with 5 ml of nanograde hexane. Stock solutions were adjusted from 0.5 to 5 ml depending on the concentration of mirex.

TABLE 1. *Dates of mirex application and bobwhite quail collection in South Carolina game area, 1975–76*

PRETREATMENT SAMPLES: AUGUST 22–SEPTEMBER 30, 1975	
BAIT APPLIED: OCTOBER 7, 1975	
14 day posttreatment sample	October 21, 1975
28 day " "	November 4, 1975
42 day " "	November 18, 1975
74 day " "	December 20, 1975
100 day " "	January 17, 1976
145 day " "	March 1, 1976
235 day " "	May 30–June 6, 1976
280 day " "	July 15, 1976

QUANTIFICATION AND CONFIRMATION

Mirex residues were detected with a Micro-Tek 220 gas chromatograph using the following instrument parameters and operating conditions:

Detector: Ni 63 electron capture
Columns: 6 ft by 1/4 in., U-shaped glass packed with mixture of 10 percent DC-200 on 80–100 mesh Gas-Chrom Q and 1.5 percent OV-17, 1.95 percent QF-1 on 80–100 mesh Gas-Chrom Q
Temperatures: injection port 220°C
column 210°C
detector 300°C
Carrier gas: nitrogen, flowing at 100 cc/min, purge flow, 10 cc/min

Five-ml aliquots were injected into the gas chromatograph using a Hamilton 10 μl syringe. The detection limit of mirex in tissue samples was set at 0.01 ppm. If mirex was detected in quantity <0.01 ppm it was recorded as a trace. Recovery tests were performed but values given here were not corrected for recovery. The recovery rate for mirex in tissue samples was approximately 70 percent. All concentrations of mirex were reported on a dry weight basis. Mirex was confirmed in several samples by mass spectroscopy.

Results and Discussion

Table 2 presents mirex residues in fat and breast tissue of 48 bobwhite quail according to sex, age, and time of collection.

Mirex residues were detected in pretreatment samples of bobwhite quail. Although the Webb Center had not been recently treated with mirex, the area bordering the plantation was treated by SCPPRS the spring before pretreatment collections of quail. Therefore, some mirex residues were anticipated.

Adult male bobwhites were selected for pretreatment collection because authors believed that they were more likely to contain high residues than females (Kendall et al., 1976: unpublished data). The pretreatment data ranged from 0.000–0.178 and 0.247–2.763 ppm mirex in breast muscle and adipose tissue, respectively. Baeteke et al. reported similar mirex residues in adipose tissue (0.000–3.148 ppm) of bobwhite quail collected 1 year after a mirex treatment (*1*).

Random posttreatment samples of young and adult bobwhite quail were collected (Table 2). The mean residue level of mirex in adipose tissue had up to a five-fold increase 2 weeks after treatment; however, this must be interpreted carefully, due to age variation in the birds. Juvenile quail apparently accumulated mirex quickly after bait application, probably because young quail consume a

TABLE 2. *Mirex residues in fat and breast samples from bobwhite quail collected on South Carolina game area, 1975–76*

Sample	Sex	Age [2]	Residues, ppm dry weight		Residues, ppm dry weight X	
			Breast	Fat	Breast	Fat
Pretreatment	M	Adult	0.178	2.763		
"	"	"	0.045	0.521		
"	"	"	0.034	0.255		
"	"	"	0.173	0.816		
"	"	"	0.046	0.247		
"	F	"	0.000	0.453		
"	M	"	0.025	0.544		
"	—	"	0.000	0.695		
"	M	Adult	0.081	0.886		
"	"	"	0.000	1.212		
"	"	"	0.000	2.733	0.053	1.011
Posttreatment 1	M	Subadult	—	0.211		
"	53		0.499	7.705		
"	F	Adult	0.370	5.602		
"	—	45	0.018	13.631		
"	F	95	0.179	1.501		
"	—	65	0.141	2.231	0.231	5.117
Posttreatment 2	M	69	0.239	1.479		
"	F	Adult	0.035	1.188	0.137	1.330
Posttreatment 3	"	"	0.039	2.735		
"	"	110	0.087	1.484		
"	M	110	0.057	1.839		
"	F	Subadult	0.089	2.878		
"	M	89	0.170	4.730	0.088	2.733
Posttreatment 4	"	Subadult	0.057	1.655		
"	"	"	0.026	1.516		
"	F	"	0.056	1.629		
"	"	"	0.093	6.627		
"	M	"	0.085	5.485	0.063	3.382
Posttreatment 5	"	" [3]	0.000	0.000 [3]		
"	F	"	0.050	2.902		
"	M	"	0.063	2.383		
"	F	Adult	0.287	2.309		
"	F	Adult	0.034	0.477	0.109	2.018
Posttreatment 6	"	Subadult	0.089	0.980		
"	M	"	0.363	11.819		
"	"	"	0.123	4.975		
"	"	"	0.355	8.097	0.233	6.468
Posttreatment 7	F	"	0.023	0.284		
"	M	"	0.156	0.867		
"	"	"	0.125	3.631		
"	"	Adult	0.053	0.502		
"	"	Subadult	0.037	1.566	0.079	1.370
Posttreatment 8	M	Subadult	0.015	0.400		
"	F	"	0.028	3.843		
"	M	Adult	0.024	1.447		
"	"	"	0.042	0.886		
"	"	"	0.117	10.012	0.045	3.318

[1] See Table 1 for schedule of mirex applications and quail collections.
[2] Aged by method of Rosene, 1969 (literature reference 14). Juveniles ≤ 150 days, Subadults > 150 days, Adults > 270 days.
[3] This sample was a banded released pen reared bird which had been in the study area approximately 2 weeks before collection.

much greater proportion of animal matter than do adult birds (6, 14).

Furthermore, Naqvi and de la Cruz (13) reported mirex residues up to 0.7 ppm in grasshoppers and crickets collected from areas receiving a mirex bait application. Examination of crop contents of Posttreatment 1 birds revealed grasshoppers to be a major component of their diets.

The mirex residues showed a slight increase up to the November sample (Posttreatments 2 through 4) and [illegible] the January sample (Posttreatment 5). Insect [illegible] in the area through November and [illegible] afterward, corresponding to the trend [illegible] tissues. Collins et al. (5) reported a similar [illegible] in bobwhite quail in whole-body tissue; a peak occurred approximately 3 months after treatment.

A sharp rise occurred in residues in the Posttreatment 6 sample, which could be attributed to several factors. New plants and insects emerged in February 1976 due to unusually warm conditions and the residue peak may have reflected the quails' selection of these items for food. Barrier et al. (2) studying whitetail deer noticed a spring peak in DDT residues which they speculated to have been translocated from new spring growth consumed by the deer. Uptake and translocation of mirex by bean and pea seedlings have been reported by Mehendale et al. (11). Crop content examination of quail collected in Posttreatment 6 revealed new spring growth, seeds, and insects. Furthermore, fat reserves were low after the winter, possibly causing residue

concentration. Mirex probably entered quail through insect consumption; however, the variables listed above may be magnifying the situation. Residues were high in quail of Posttreatment 8 the summer after the 1975 pretreatment. This likely reflects mirex spraying of the study area rather than the spring 1975 treatment by SCPPRS of border areas. Here quail were exposed directly to mirex bait, rather than indirectly contacting mirex through the food chain.

In pen studies designed to test the toxicological effects of mirex in birds, Hyde et al. (*8*) found no reproductive inhibition in mallard ducks (*Anas platyrhynchos*) fed up to 100 ppm mirex, although some of their ducklings died. Analysis of selected duck tissues from the 100 ppm treatment indicated 2,964 ppm mirex in fat and 37 ppm in breast muscle. Heath et al. (*7*) determined the LC_{50} in 5-day diets of 2-week-old bobwhite quail to be 2,511 ppm mirex in feed. Pen studies testing the effects of 40 ppm mirex in the diet of bobwhite quail revealed no reproductive effects as measured by rates of egg production, shell cracking, embryonation rates, embryo survival, hatching, and hatchling success (*15*). Carcass residues average 113 ppm mirex (wet weight) in males and 172 ppm in females (*15*).

In a similar laboratory study of bobwhite quail receiving mirex treatments of 1, 20, and 40 ppm in feed, Kendall et al. found no harmful reproductive effects (1976: unpublished data). Birds analyzed after the 40 ppm treatment indicated 343 ppm and 197 ppm mirex (dry weight) in adipose tissue of males and females, respectively. Birds exposed to the 1 ppm treatment (F_0 generation) had 17 ppm mirex in fat deposits in males and 6 ppm in females. Furthermore, no reproductive inhibition was noted when a second generation of birds was exposed to 1 ppm mirex. The residue levels in tissue on the F_0 generation quail exposed to 1 ppm mirex closely correlates with residues presented in Table 2.

Action guidelines of the USDA set by the U.S. Environmental Protection Agency (EPA) for mirex residues in meat (lipid weight) of domestic animals intended for human consumption is 0.1 ppm (*4*).

Table 2 reveals that numerous breast muscle residues appear to be above EPA limits. In evaluating these EPA prescribed tolerance levels, Clark and McLane (*4*) reported that in woodcock (*Philohela minor*), breast muscle contains only 1.9 ± 0.1 percent fat, compared to hamburger which may contain up to 28 percent fat, or 15 times the mean percent fat of woodcock. Therefore, in considering the safety of chlorinated hydrocarbon residues in woodcock, it would be appropriate to multiply guideline levels by 15. Tolerance levels have not been established for mirex residues in bobwhite quail. However, in light of lipid weight ppm comparisons, the authors conclude that residue levels reported here would contaminate little if any of the human food chain. The mirex residue data in Table 2 would be further lowered if converted from the reported dry-weight to wet-weight concentrations; approximate conversion factors are fat, 0.77, and breast, 0.29.

The authors conclude that the current rate of mirex bait application (*12*) does not inhibit reproduction of the bobwhite quail. Furthermore, no significant human food chain contamination with mirex would be indicated in the consumption of quail harvested from a mirex-treated area.

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PCB's in Fish from Selected Waters of New York State [1]

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ABSTRACT

PCB residues in fish from 41 stations throughout New York State were monitored in 1975. Nearly all fish contained PCB's in detectable amounts although the levels of contamination and specific Aroclor varied. The Hudson River contained the highest known PCB concentrations within the United States; levels often exceeded 100 ppm. Other waters and fish which were significantly contaminated include Lake Ontario salmonids and Cayuga Lake lake trout. Onondaga Lake, previously closed to fishing because of mercury contamination, also appears to have abnormally high levels of PCB's approaching in some instances the action level of the Food and Drug Administration, U.S. Department of Health, Education, and Welfare. Samples from marine waters generally have contaminant levels substantially below 5.0 ppm.

A summary of PCB data collected during 1970–75 is presented

Introduction

The Division of Fish and Wildlife, New York State Department of Environmental Conservation, began analyzing fish flesh for DDT in the early 1960's. This work was conducted in response to data suggesting that chlorinated hydrocarbons may interfere with reproduction of fish and wildlife resources. Results of the investigation led to State restrictions on use of DDT in 1965. During this time, research indicated that DDT analyses could be biased through sample contamination with similar compounds, polychlorinated biphenyls (PCB's) (*8*). To minimize errors in future work, both DDT and PCB's were extracted and analyzed to obtain separate values. From 1971 through 1975 PCB's were evaluated in all fish flesh analyses as a part of DDT monitoring.

The recent concern over the impact of PCB's on human health and their possible effect on New York's natural resources as well as reports of their ubiquitous nature (*6, 13, 20*) precipitated a shift from low-level monitoring to the statewide monitoring described in this report. The new program promulgated by the Department of Environmental Conservation in August 1975 revised all existing directions. Specific fish species and desired sizes were defined (Table 1). Stations were located in most large waterways throughout the State where PCB contamination could occur (Figure 1, Table 2) and, when possible, where earlier samples had been taken. Several Adirondack streams were included to ascertain statewide ambient conditions. Where contamination was known to exist sampling was intensified. This was the case of the Hudson River where inordinately high concentrations of the contaminant in fish were reported by the U.S. Environmental Protection Agency (EPA (*16*)).

The objectives of this program were to determine the level and extent of PCB contamination in fish inhabiting or migrating through major waterways in New York State and to isolate contaminated portions or reaches of waterways, thereby identifying specific areas to be addressed by agencies which control water pollution.

TABLE 1. *Desired species and size distribution of fish from New York waters analyzed for PCB content, 1975*

Species	Small [1]	Medium [2]	Large [3]
	Length, in		
Smallmouth bass	5–7	10–12	14–16
Largemouth bass	5–7	10–12	14–16
Walleye	7–9	11–13	19–21
Bullheads	4–6	8–10	11–13
Carp	5–7	13–15	23–25
Goldfish	4–6	7–9	10–12
Suckers	6–8	10–12	17–19
Striped bass	11–13	16–18	28–32
	Weight, lb [4]		
Salmon (coho)	1–2	4–6	8–12
(chinook)	3–5	8–12	15–20
Rainbow or steelhead trout	1–2	3–5	5–8
Lake trout	1–2	4–6	8–12

[1] Minimum number of fish per sample was 6
[2] Average number of fish per sample was 5
[3] Maximum number of fish per sample was 5
[4] Minimum number of fish per sample was 5

[1] New York State Department of Environmental Conservation, Bureau of Environmental Protection, 50 Wolf Road, Albany, N.Y. 12233

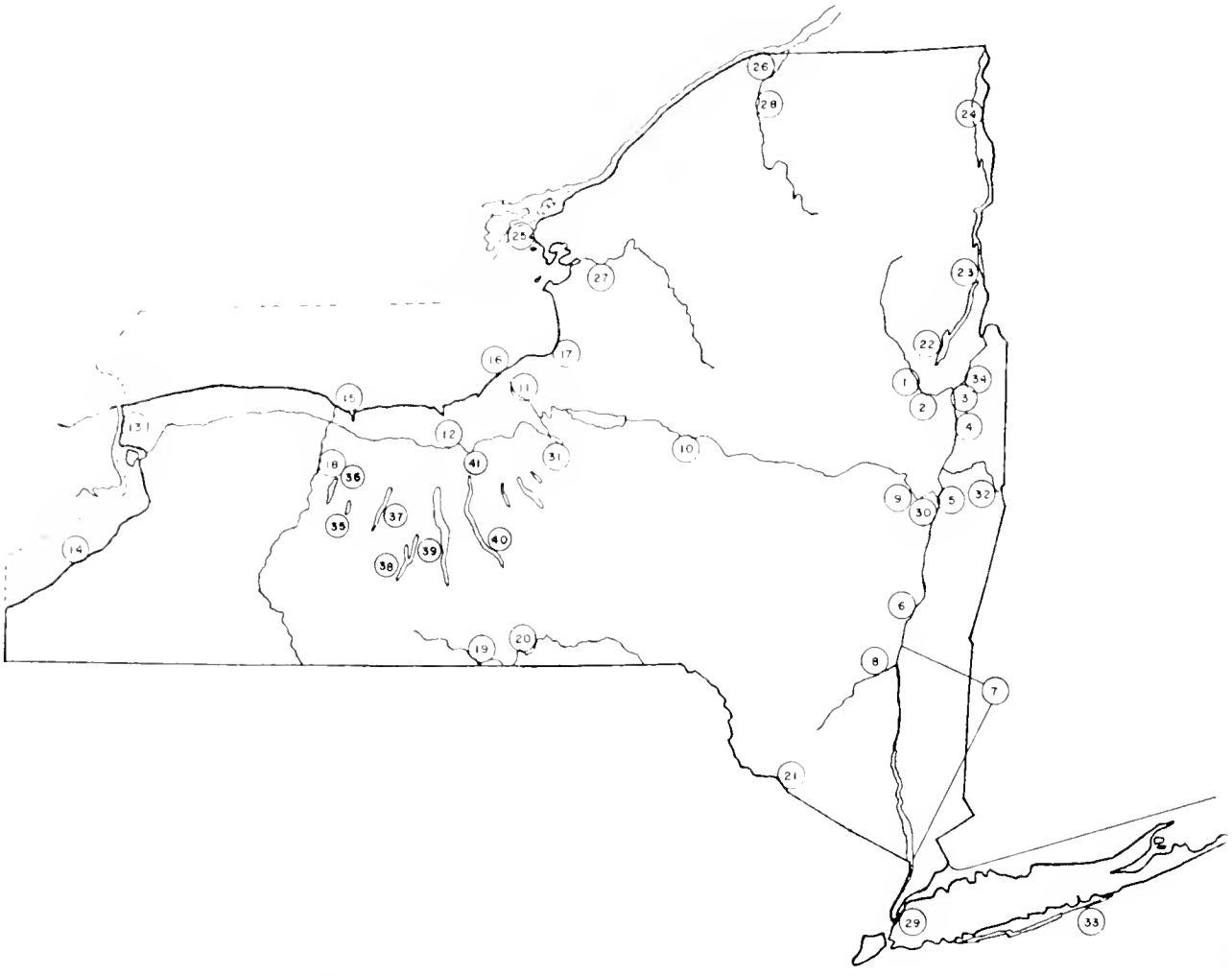

FIGURE 1. *Fish collection stations for PCB studies, New York State–1975*

Stations selected for fish sampling were included in a major water column and sediment monitoring program conducted by the State Division of Pure Waters. Data from all sources are being used to track down sources of PCB contamination and to recommend remedial action.

Fish Collections

Regional Fish and Wildlife personnel were instructed on specific species and sizes of fish desired, general location of the stations, and timetable for collections. Sampling methods were primarily gill nets and electrofishing; angling and rotenone were used when other techniques were inadequate. On occasion, fish were donated by or purchased from commercial and recreational anglers.

The composition of the fish samples directed to the laboratories varied slightly, depending on the success of the collection teams. Specifications in Table 1 were flexible in order to accommodate a sampling program involved with diverse waters. Specific instructions requested that at each location members of two trophic levels, predators and forage species, be sampled in three size categories: small or sublegal, medium or minimally legal, and large.

The collection team chose the representative species and followed the length and number specifications in Table 1. In those areas where a commercial species is harvested, representative specimens were included.

All specimens were individually tagged, measured and weighed, placed by groups in polyethylene bags, and deposited in a cooler or freezer for shipment to a laboratory

TABLE 2. *Statewide sampling stations for PCB analysis, New York—1975*

Station No.[1]	Watershed	Waterway	Location
1	Upper Hudson River	Hudson River	Above Corinth
2			Above Glens Falls
3			At Fort Edward
			below General Electric Co
4			5 miles below Fort Edward
5			Waterford area
6	Lower Hudson River		Coxackie-Catskill area
7			Lower Hudson
			Mouth of Rondout Creek
			Mouth of Wappingers Creek
			Mouth of Esopus Circle
			Kingston Rhinecliff Bridge
			Tappan Zee Bridge area
			West Point
			George Washington Bridge area
			Highland Poughkeepsie area
8		Rondout River	Rondout River above first dam
9	Mohawk River	Mohawk River	Below Schenectady
10			Below Otiskany
11	Seneca Oneida Oswego Rivers	Seneca Oneida Oswego Rivers	Below Fulton
12		Erie Canal	Below Clyde
13	Lake Erie Niagara River	Niagara River	Below Lewiston
14	Lake Erie Niagara River	Lake Erie	Dunkirk
15	Lake Ontario	Lake Ontario	Off Rochester
16			Oswego
17			Pulaski (salmonids)
18	Genesee River	Genesee	Below Industry
19	Chemung River	Chemung	Chemung
20	Susquehanna River	Susquehanna	Smithboro
21	Delaware River	Delaware	Below Pt. Jervis
			(State line)
22	Lake Champlain	Lake George	Bolton Landing
23		Lake Champlain	Ticonderoga
24			Plattsburg
25	St. Lawrence River	St. Lawrence	Cape Vincent
26			Massena
27	Black River	Black River	Watertown
28	St. Lawrence River	Raquette River	Norfolk
29		Fulton Fish Market	
30		Albany Fish Market	
31	Seneca Oneida Oswego Rivers	Onondaga Lake	
32	Upper Hudson River	Walloomsac Hoosick Rivers	
33	Long Island	Marine Waters	
34	Upper Hudson River	Champlain Canal	Lock 8
35	Genesee River	Hemlock Lake	
36		Canadice Lake	
37	Seneca Oneida Oswego Rivers	Canandaigua Lake	
38		Keuka Lake	
39		Seneca Lake	
40		Cayuga Lake	
41		Seneca River	Montezuma National Wildlife Refuge

[1] See map, figure 1

or storage center. Common names of fishes used throughout this report are those adopted by the American Fisheries Society (*1*) and are presented with their scientific nomenclature in Table 3.

TABLE 3. *Common and scientific names of fish and shellfish species analyzed for PCB's –1975*

Common Name	Scientific Name
Spiny dogfish	*Squalus acanthias*
Atlantic sturgeon	*Acipenser oxyrhynchus*
American eel	*Anguilla rostrata*
Blueback herring	*Alosa aestivalis*
Alewife	*Alosa pseudoharengas*
American shad	*Alosa sapidissima*
Atlantic menhaden	*Brevoortia tyrannus*
Cisco or lake herring	*Coregonus artedi*
Coho salmon	*Oncorhynchus kisutch*
Chinook salmon	*Oncorhynchus tshawytscha*
Rainbow trout	*Salmo gairdneri*
Atlantic salmon	*Salmo salar*
Brown trout	*Salmo trutta*
Lake trout	*Salvelinus namaycush*
Rainbow smelt	*Osmerus mordax*
Chain pickerel	*Esox niger*
Northern pike	*Esox lucius*
Goldfish	*Carassius auratus*
Carp	*Cyprinus carpio*
White sucker	*Catostomus commersoni*
Silver redhorse	*Moxostoma anisurum*
Shorthead redhorse	*Moxostoma macrolepidotum*
Brown bullhead	*Ictalurus nebulosus*
Channel catfish	*Ictalurus punctatus*
Goosefish	*Lophius americanus*
Atlantic cod	*Gadus morhua*
Atlantic tomcod	*Microgadus tomcod*
Mummichog	*Fundulus heteroclitus*
Atlantic silverside	*Menidia menidia*
White perch	*Morone americana*
White bass	*Morone chrysops*
Striped bass	*Morone saxatilis*
Rock bass	*Ambloplites rupestris*
Redbreast sunfish	*Lepomis auritus*
Pumpkinseed	*Lepomis gibbosus*
Bluegill	*Lepomis macrochirus*
Smallmouth bass	*Micropterus dolomieui*
Largemouth bass	*Micropterus salmoides*
Black crappie	*Pomoxys nigromaculatus*
Yellow perch	*Perca flavescens*
Walleye	*Stizostedion vitreum*
Bluefish	*Pomatomus saltatrix*
Weakfish	*Cynoscion regalis*
Scup	*Stenotomus chrysops*
Tautog	*Tautoga onitis*
Freshwater drum	*Aplodinotus grunniens*
Atlantic mackerel	*Scomber scombrus*
Bluefin tuna	*Thunnus thynnus*
Grubby	*Myoxocephalus aenaeus*
Summer flounder	*Paralichthys dentatus*
Winter flounder	*Pseudopleuronectes americanus*
Northern puffer	*Sphoeroides maculatus*
Shellfish	
Blue claw crab	*Callinectes sapidus*
Northern lobster	*Hommarus americanus*
Hard clam	*Mercenaria mercenaria*
Surf clam	*Spisula solidissima*
American oyster	*Crassostrea virginica*
Common mussel	*Mytilus edulis*

Laboratory Analyses

Four State laboratories collaborated in the analysis of fish flesh. Department of Environmental Conservation Laboratories at Rome and Stony Brook, Department of Agriculture and Market Food Laboratory in Albany, and the Department of Health Griffin Laboratory in Guilderland. Limited analyses were conducted in the Buffalo, N.Y., laboratory of the Food and Drug Administration, U.S. Department of Health, Education, and Welfare. Specific analytical techniques have been described (5, 9, 10, 14, 15, 17, 21).

Frozen fish samples were defrosted, generally prepared as described below, ground three times, and blended.

FISH SIZE	PREPARATION
Very small less than 100 g	Whole fish composites, see Table 4
Small 100–150 g	Head and viscera removed Individuals composited
Medium 25–61 cm	Head and viscera removed Composite if needed
Large 61–76 cm	Left fillet or anterior half of left fillet with skin intact Composite if needed
Jumbo over 76 cm	Anterior third of left fillet with skin intact

Brown bullhead, American eel, and Atlantic Ocean fish were the only species skinned before grinding; this is the common method of preparing them for consumption. Composites of medium through jumbo fish were made of ground and blended flesh; total weight of each individual was proportional to the total weight of the composite.

Laboratories performing analyses are noted in Tables 4 and 5. Recoveries by the analytical techniques cited above are 85 percent or more. Data were not adjusted to reflect recovery rate. Each procedure has a sensitivity of 0.01 ppm although two laboratories reported results only as low as 0.1 ppm. Very limited numbers of fish from Lake Ontario were subjected to mass spectrometry for confirmation of analytical results.

A quality assurance program was implemented to assure that results from the various laboratories were comparable. Table 6 shows that none of the participating laboratories had contamination problems; all reported zero for sample A. At the level of approximately 1 μg/ml of extract, which corresponds to the action level of 5 μg/g in fish, precision among the laboratories was remarkably good. No gross bias existed, although the laboratories had standard Aroclor mixtures obtained at different times and one laboratory used Aroclor 1242 for quantitation instead of Aroclor 1016. Results at higher concentrations indicate similar agreement.

In Table 6, the two sets of results from Griffin Laboratory illustrate the effects of using separate standards to quantitate the two Aroclor types and of using a mixed standard. Although all laboratories used individual standards, Table 6 results indicate basic similarity to results obtained for a mixed standard, Aroclors 1242 and 1254. This is probably due to differing gas chromatograph column conditions, and differing techniques of measuring the various Aroclors. To measure check samples such as these the use of individual Aroclors is the most desirable, but for environmental samples which are nearly always mixtures, use of mixed standards is analytically faster and probably more accurate. Table 7 supports this contention; some laboratories used mixed standards and some used indi-

TABLE 4. *Concentrations of PCB's in fish and shellfish in New York State, 1975*

Water	Station	Laboratory [1]	Species	Aroclor	No. Fish Analyzed	Number of Analyses	Concentration of PCB's, ppm wet weight [2]		
							Weighted Average	High	Low
Hudson River	1, 2	DEC–R	Smallmouth bass	1242 1016	26	12	0	0	
				1254			TR	TR	0
			Walleye	1242 1016	2	2	0	0	
				1254			TR	TR	0
			Yellow perch	1242 1016	15	11	0	0	
				1254			TR	TR	0
			White sucker	1242 1016	4	4	0	0	
				1254			TR	TR	0
Hudson River (Fort Edward below General Electric Company)	3	DEC–R	Walleye						
			Whole fish	1242 1016	14	2	91 01	104 21	81 12
				1254			36 01	41 80	28 29
			Edible flesh	1242 1016	4	1	9 88	9 88	
				1254			8 31	8 31	
			Yellow perch						
			Whole fish	1242 1016	30	2	164 36	236 40	128 33
				1254			37 14	62 88	24 27
			Edible flesh	1242 1016	10	1	61 10	61 10	
				1254			18 24	18 24	
			Rock bass	1242 1016	2	1	27 35	27 35	
				1254			ND		
			Alewife	1242 1016	1	1	20 35	20 35	
				1254			ND		
			White sucker	1242 1016	9	2	41 74	82 88	8 83
				1254			23 10	47 57	2 72
Hudson River (5 miles below Fort Edward)	4	DEC–R	Smallmouth bass	1242 1016	1	1	100 27	100 27	-
				1254			22 64	22 64	-
			Walleye	1242 1016	3	3	56 73	157 27	5 19
				1254			10 67	20 74	5 56
			Yellow perch						
			Whole fish	1242 1016	26	2	87 29	88 18	34 31
				1254			30 69	34 21	18 95
			Edible flesh	1242 1016	3	1	35 54	35 54	
				1254			15 01	15 01	-
			White sucker [3]	1242 1016	10	5	47 30	78 00	28 03
				1254			19 82	41 10	8 13
			Brown bullhead	1242 1016	5	2	77 04	85 30	73 24
				1254			37 23	38 52	36 38
Hudson River (Stillwater)	4A	DEC–R	Largemouth bass						
			Whole fish	1242 1016	8	2	11 65	14 31	8 99
				1254			8 42	9 84	6 99
			Edible flesh	1242 1016	4	1	19 25	24 85	12 40
				1254			10 59	15 97	4 44
			American eel [4]	1242 1016	2	2	207 33	403 38	11 28
				1254			83 47	155 87	11 07
			Brown bullhead	1242 1016	1	1	4 66	4 66	-
				1254			7 99	7 99	
			White sucker	1242 1016	6	2	58 32	154 28	10 34
				1254			15 93	35 92	5 94
Hudson River (Waterford)	5	DAM	Smallmouth bass	1242 1016	10	2	30 60	36 0	27 0
				1254			15 70	17 5	14 5
			Walleye	1242 1016	2	1	23 30	23 3	-
				1254			9 10	9 1	-
			White sucker	1242 1016	11	2	27 44	36 0	20 3
				1254			10 15	12 9	7 9
Hudson River (Below Troy Dam)	6, 7a-h	DEC–R	Striped bass						
			Whole fish	1242 1016 [5]	21	2	0 10	2 19	0
				1254			1 75	1 96	1 66
				1242 1016	42	31	12 05	56 42	0
			Edible flesh	1254	48	37	11 41	50 06	1 66

(Continued next page)

TABLE 4 (cont'd.) *Concentrations of PCB's in fish and shellfish in New York State, 1975*

Water	Station	Laboratory[1]	Species	Aroclor	No. Fish Analyzed	Number of Analyses	Concentration of PCB's, ppm wet weight[2] Weighted Average	High	Low
			Smallmouth bass	1242 1016	2	2	8.05	13.76	2.34
				1254	1	1	2.18	2.18	–
			Largemouth bass	1242 1016	10	10	5.43	19.73	0
				1254			4.62	7.65	1.73
			Yellow perch	1242 1016	7	2	1.19	4.18	0
				1254	5	1	5.28	5.28	–
			White perch	1242 1016	30	9	9.00	30.67	0
				1254	18	4	6.98	9.65	4.14
			Chain pickerel	1242 1016	1	1	8.68	8.68	–
				1254			ND		
Hudson River (Below Troy Dam)	6–7a,b	DEC, R	Atlantic tomcod Whole fish	1242 1016	47	2	6.49	6.68	6.27
				1254			7.75	9.85	5.37
			Edible flesh	1242 1016	6	2	1.50	2.07	0.93
				1254			1.32	1.63	1.00
			American shad	1242 1016	15	6	1.35	9.00	0
				1254			1.14	1.88	0
			Blueback herring Whole fish	1242 1016	4	4	0.72	1.85	0
				1254			1.41	4.69	0
			Edible flesh	1242 1016	2	1	0	0	–
				1254			5.13	5.13	–
			American eel	1242 1016	11	7	54.34(25.61)[3]	186.22	0
				1254			27.20(19.15)	57.20	3.00
			Atlantic sturgeon	1242 1016	20	5	7.69	13.48	0
				1254			8.15	9.84	6.25
			Brown bullhead	1242 1016	1	1	0	0	–
				1254			3.58	3.58	–
			Atlantic menhaden	1242 1016	8	2	0	0	–
				1254			1.67	1.83	1.41
Rondout Creek	8	DAM	Largemouth bass	1242 1016	7	2	0	0	–
				1254			1.39	1.97	1.16
			White sucker	1242 1016	5	1	0	0	–
				1254			1.60	1.60	–
			Carp	1242 1016	3	1	0	0	–
				1254			1.50	1.50	–
Mohawk River (Schenectady)	9	DAM	Smallmouth bass	1242 1016	3	1	0.70	0.7	–
				1254			1.50	1.5	–
			White sucker	1242 1016	5	1	0.80	0.8	–
				1254			1.90	1.9	
			Black crappie	1242 1016	9	2	1.11	1.6	0.5
				1254			1.13	1.4	0.8
			Largemouth bass	1242 1016	8	2	0.95	1.2	0.7
				1254			3.40	4.7	2.1
			Blueback herring	1242 1016	6	1	2.10	2.1	–
				1254			1.60	1.6	
Mohawk River (Little Falls)	10	DAM	Walleye	1242 1016	10	2	4.45	4.9	4.0
				1254			9.25	9.5	9.0
			Smallmouth bass	1242 1016	2	1	3.90	3.9	
							9.50	9.5	
Mohawk River (Little Falls)		DAM	White sucker	1242 1016	11	2	0	0	–
				1254			2.79	4.2	1.1
			Brown bullhead	1242 1016	5	1	2.70	2.7	
				1254			11.30	11.3	–
[illegible] Canal		DEC	Smallmouth bass	1242 1016	14	3	2.29	4.4	0
				1254			1.13	2.5	0.7

(Continued on next page)

TABLE 4 (cont'd.). *Concentrations of PCB's in fish and shellfish in New York State, 1975*

Water	Station	Laboratory [1]	Species	Aroclor	No. Fish Analyzed	Number of Analyses	Concentration of PCB's, ppm wet weight [2]		
							Weighted Average	High	Low
			Carp	1242 1016 1254	10	2	2 00 5 60	4 0 7 2	0 4 0
Erie Canal	12	DH	Largemouth bass	1242 1016 1254	1	1	0 0 80	0 0 8	- -
			Northern pike	1242 1016 1254	1	1	2 00 1 30	2 0 1 3	-
			Carp	1242 1016 1254	8	2	0 60 0 30	1 6 0 8	0 0
Niagara River	13	DEC-R	Goldfish	1242 1016 1254	5	1	0 TR	0 TR	-
			Largemouth bass	1242 1016 1254	1	1	0 3 10	0 3 10	-
			Smallmouth bass	1242 1016 1254	1	1	0 3 30	0 3 30	-
			White bass	1242 1016 1254	1	1	0 11 31	0 11 31	-
Lake Erie	14	DEC-R DAM	Smallmouth bass	1242 1016 1254	19	4	0 19 0 72	0 4 1 0	0 0 6
			Walleye	1242 1016 1254	19	9	0 19 0 70	0 6 1 45	0 TR
			Yellow perch	1242 1016 1254	10	2	0 40 0 60	0 4 0 6	0 4 0 6
			Goldfish	1242 1016 1254	4	1	1 00 1 40	1 0 1 4	-
			Brown trout	1242 1016 1254	20	9	0 23 1 60	2 4 3 23	0 0 57
Lake Erie	14	DEC R	Rainbow trout	1242 1016 1254	10	9	0 2 03	0 3 77	- 0 76
			Chinook salmon	1242 1016 1254	8	8	0 1 71	0 2 71	- 0 98
			Coho salmon	1242 1016 1254	15	4	0 2 36	0 5 06	- 0 68
Lake Ontario	15-16	DEC R	Coho salmon	1242 1016 1254	23	15	0 8 05	0 18 65	- 2 09
			Chinook salmon	1242 1016 1254	29	17	0 21 14 48	6 07 24 57	0 4 62
			Rainbow trout	1242 1016 1254	9	9	0 5 76	0 12 35	- 1 47
			Brown trout	1242 1016 1254	12	12	0 5 25	0 16 76	- 0 96
			Lake trout	1242 1016 1254	19	14	0 9 38	0 14 62	- 1 35
			White perch	1242 1016 1248 1254	27	5	0 0 05 4 12	0 0 48 12 06	- 2 14
			Yellow perch	1242 1016 1254	25	4	0 38 0 38	0 0 69	
			Northern pike	1242 1016 1254	17	10	0 1 79	0 4 02	0 96
			Largemouth bass	1242 1016 1254	5	5	0 1 79	0 4 25	- 0 42
			Smallmouth bass	1242 1016 1254	7	2	0 1 81	0 2 69	- 1 15
			Rock bass	1242 1016 1254	11	2	0 0 83	0 3 58	0 0 55
			Brown bullhead	1242 1016 1254	10	2	0 0 57	0 0 75	- 0 39

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TABLE 4 (cont'd.) *Concentrations of PCB's in fish and shellfish in New York State, 1975*

Area	Station	Laboratory[1]	Species	Aroclor	No. Fish Analyzed	Number of Analyses	Concentration of PCB's, ppm wet weight[2]		
							Weighted Average	High	Low
			Pumpkinseed	1242 1016 1254	19	4	0 0 01	0 0 26	– 0
			Walleye	1242 1016 1254	8	4	0 2 87	0 7 39	– 0 33
			Rainbow smelt	1242 1016 1254	10	4	0 2 10	0 4 55	– 1 16
			American eel	1242 1016 1254	2	2	0 6 28	0 10 74	– 1 82
			Black crappie	1242 1016 1254	8	6	0 1 35	0 2 07	– 0 20
			White sucker	1242 1016 1254	5	5	0 3 39	0 7 90	– 1 38
Lake Ontario (Salmon River Pulaski Weir)	17	DEC R	Coho salmon	1242 1016 1254	9	20	0 9 31	0 12 67	– 4 48
			Chinook salmon	1242 1016 1254	21	15	0 11 6 98	2 28 17 01	0 2 84
Genesee River	18	DH	Smallmouth bass	1242 1016 1254	13	3	1 77 2 23	4 0 5 6	0 5 0 5
			Silver redhorse	1242 1016 1254	14	3	1 55 1 45	3 0 2 4	1 0 0 5
Chemung River	19	DAM	Smallmouth bass	1242 1016 1254	12	3	0 0 88	0 1 5	– 0 6
			Rock bass	1242 1016 1254	2	1	0 40 0 40	0 4 0 4	– –
			White sucker	1242 1016 1254	13	3	0 29 0 55	0 5 0 9	0 0 2
Susquehanna River	20	DH	Smallmouth bass	1242 1016 1254	12	2	0 20 0 45	0 2 0 5	0 2 0 4
			Walleye	1242 1016 1254	6	2	0 13 0 75	0 3 0 7	0 1 1 0
			White sucker	1242 1016 1254	18	3	0 20 0 80	0 2 0 8	0 2 0 8
			Carp	1242 1016 1254	11	2	0 41 1 25	0 5 1 3	0 3 1 2
Delaware River	21	DH	Smallmouth bass	1242 1016 1254	3	2	1 33 0 67	1 6 0 8	0 8 0 6
			White sucker	1242 1016 1254	5	1	0 40 0 50	0 4 0 5	– –
			American eel	1242 1016 1254	9	3	1 24 0 36	2 8 0 8	0 0
[illegible]	22	DH & DEC R	Lake trout	1242 1016 1254	10	5	0 71 3 94	2 0 15 0	0 0
			Rainbow trout	1242 1016 1254	6	6	0 <1 00	0 <1 00	– <1 00
			Smallmouth bass	1242 1016 1254	14	5	0 29 0 27	1 0 1 0	0 0
			Yellow perch	1242 1016 1254	15	3	0 57 0 31	1 2 0 8	0 0
			Cisco	1242 1016 1254	10	2	0 50 3 95	0 6 4 7	0 4 3 2
Lake Ontario	[illegible]	DH	Smallmouth bass	1242 1016 1254	29	6	0 75 1 21	3 0 3 5	0 0
			Walleye	1242 1016 1254	5	1	0 30 0 20	0 3 0 2	
			Northern Pike	1242 1016 1254	4	2	0 2 0 4	0 3 0 6	0 1 0 2

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TABLE 4 (cont'd.). *Concentrations of PCB's in fish and shellfish in New York State, 1975*

Water	Station	Laboratory[1]	Species	Aroclor	No. Fish Analyzed	Number of Analyses	Concentration of PCB's, ppm wet weight[2]		
							Weighted Average	High	Low
			Yellow perch	1242 1016 1254	17	3	0 40 0 59	0 4 0 8	0 0 2
			Brown bullhead	1242 1016 1254	14	3	0 34 0 45	0 7 1 2	0 0
St Lawrence River	25, 26	DEC R	Smallmouth bass	1242 1016 1254	16	9	0 4 58	0 6 85	1 62
			Walleye	1242 1016 1254	11	3	0 1 83	0 2 05	1 58
Black River	27	DH	Walleye	1242 1016 1254	8	2	1 25 3 11	2 0 4 5	0 0 8
			Brown bullhead	1242 1016 1254	10	2	0 95 2 50	1 5 4 0	0 4 1 0
Raquette River	28	DH	Smallmouth bass	1242 1016 1254	15	4	0 30 0 90	0 4 1 7	0 2 0 3
			Shorthead redhorse	1242 1016 1254	13	3	0 32 0 44	0 4 0 6	0 2 0 3
Onondaga Lake	31	DEC R	Smallmouth bass	1242 1016 1254	9	3	1 82 3 13	1 9 4 0	1 2 1 2
			Walleye	1242 1016 1254	1	1	3 10 2 50	3 1 2 5	
			White perch	1242 1016 1254	11	2	2 28 2 33	4 3 4 9	0 6 0 2
			White sucker	1242 1016 1254	3	1	0 70 0 20	0 7 0 2	-
			Carp	1242 1016 1254	7	2	2 20 6 04	3 0 7 5	0 2 2 4
			Brown bullhead	1242 1016 1254	5	1	3 10 5 30	3 1 5 3	
Wallloomsack Hoosic River	32	DH	Largemouth bass	1242 1016 1254	5	2	2 62 2 98	2 8 3 5	2 5 2 2
			Yellow perch	1242 1016 1254	4	1	2 00 3 40	2 0 3 4	0 0
			White sucker	1242 1016 1254	7	3	1 73 1 58	2 9 2 9	0 7 0 6
Atlantic Ocean	33	DEC SB	Bluefish	1242 1016 1248 1254	26	15	0 0 1 43	0 0 8 39	- - 0 21
			Striped bass	1242 1016 1248 1254	29	17	0 0 1 05	0 0 3 59	- TR
			Porgy	1242 1016 1248 1254	12	2	0 0 0 29	0 0 0 32	- 0 25
			Atlantic menhaden	1242 1016 1248 1254	18	7	0 0 1 07	0 0 1 53	- - 0 44
			Spiny dogfish	1242 1016 1248 1254	60	4	0 0 0 25	0 0 0 27	- - 0 22
			Bluefin tuna	1242 1016 1248 1254	7	7	0 0 1 42	0 0 6 68	- - 0 19
			Atlantic mackerel	1242 1016+ 1254	5	5	0 46	0 52	0 31
			Weakfish	1242 1016 1248 1254	1	1	0 0 0 21	0 0 0 21	- -

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TABLE 4 (cont'd.). *Concentrations of PCB's in fish and shellfish in New York State, 1975*

Water	Station	Laboratory[1]	Species	Aroclor	No. Fish Analyzed	Number of Analyses	Concentration of PCB's, ppm wet weight[2]		
							Weighted Average	High	Low
			Summer flounder	1242 1016	1	1	0	0	–
				1248			0	0	–
				1254			0 56	0 56	–
			Winter flounder	1242/1016	16	10	0	0	–
				1248			0	0	–
				1254			0 09	0 36	TR
			Atlantic cod	1242 1016	9	9	0	0	–
				1248			0	0	–
				1254			0 18	0 34	TR
			Tautog	1242 1016	3	3	0	0	–
				1248			0	0	
				1254			TR	TR	TR
			Scup	1242/1016	1	1	0	0	–
				1248			0	0	
				1254			0 33	0 33	–
			White perch	1242 1016	2	2	0	0	–
				1248			0	0	–
				1254			3 11	5 19	1 02
			Atlantic silversides Whole fish	1242 1016	50	2	0	0	
				1248			0	0	–
				1254			0 98	1 51	0 45
			American eel	1242 1016	1	1	0	0	–
				1248			0	0	–
				1254			3 75	3 75	–
			Goosefish	1242/1016	1	1	0	0	–
				1248			0	0	–
				1254			TR	TR	–
			Northern puffer	1242 1016	1	1	0	0	–
				1248			0	0	–
				1254			TR	TR	–
			Hard clam	1242 1016	36	3	0	0	–
				1248			0 23	0 36	0 01
				1254			0	0	–
			Surf clam	1242 1016	6	1	0	0	–
				1248			0 36	0 36	–
				1254			0	0	–
			Blue claw crab	1242 1016	14	2	0	0	–
				1248			0 32	0 44	TR
				1254			0	0	–
			Northern lobster	1242 1016	1	1	0	0	
				1248			0 13	0 13	–
				1254			0	0	–
			American oyster	1242 1016	48	4	0	0	–
				1248			0	0	–
				1254			TR	0 15	TR
			Common mussel	1242 1016	36	3	0	0	–
				1248			0	0	
				1254			TR	TR	TR
[illegible]	[illegible]	DEC R	Rock bass Whole fish	1242 1016	1	1	4 40	4 40	–
				1254			7 78	7 78	–
			Freshwater drum Whole fish	1242 1016	2	1	0	0	
				1254			1 36	2 33	0 38
			Edible fish	1242 1016	2	2	0	0	
				1254			TR	TR	
			Yellow perch Whole fish	1242 1016	1	1	0	0	
				1254			0 92	0 92	
[illegible]	[illegible]	[illegible]	Mewife	1242 1016	100	1	0	0	
				1254			1 60	1 60	

TABLE 4 (cont'd.). *Concentrations of PCB's in fish and shellfish in New York State, 1975*

Water	Station	Laboratory [1]	Species	Aroclor	No. Fish Analyzed	Number of Analyses	Concentration of PCB's, ppm wet weight [2] Weighted Average	High	Low
			White sucker Whole fish	1242 1016 1254	5	3	0 0.59	0 0.92	TR
			Pumpkinseed Whole fish	1242 1016 1254	5	1	0 2.00	0 2.00	
Hemlock Lake	35	DEC-R	Lake trout	1242 1016 1254	10	10	TR TR	TR TR	0 0
			Rainbow trout	1242 1016 1254	2	2	0 0	0 0	
			Chain pickerel	1242 1016 1254	5	5	0 0	0 0	
			White sucker	1242 1016 1254	3	1	TR 0	TR 0	
			Carp	1242 1016 1254	4	1	0 0	0 0	
			Cisco	1242 1016 1254	3	1	TR 0	TR 0	
			Alewife	1242 1016 1254	5	1	0 0	0 0	
			Rainbow smelt	1242 1016 1254	5	1	TR 0	TR 0	
			Yellow perch	1242 1016 1254	7	2	TR 0	TR 0	
Canadice Lake	36	DEC-R	Lake trout	1242 1016 1254	7	7	0	0	
			Rainbow trout	1242 1016 1254	5	5	0	0	
Canandaigua Lake	37	DEC-R	Lake Trout	1242 1016 1254	17	12	3.11 5.38	25.78 21.69	0 1.12
			Brown trout	1242 1016 1254	1	1	0 2.58	0 2.58	
Keuka Lake	38	DEC-R	Lake trout	1242 1016 1254	18	8	TR 0.09	TR 1.77	0 0
			Rainbow smelt	1242 1016 1254	6	1	0 0	0 0	
			Alewife	1242 1016 1254	9	2	0 0	0 0	
Seneca Lake	39	DEC-R	Lake trout	1242 1016 1254	10	10	0 6.42	0 15.99	2.62
Cayuga Lake	40	DEC-R	Lake trout	1242 1016 1254	7	7	0 6.15	0 8.23	1.76
Montezuma National Wildlife Refuge	41	DEC-R	Carp	1242 1016	3	3	0.66 1.67	1.59 3.53	TR 0.29

[1] Laboratory code: DEC-R = Department of Environmental Conservation, Rome Laboratory, New York State
DAM = Department of Agriculture and Markets, Food Laboratory, New York State
DH = Department of Health, Griffin Laboratory, New York State
DEC-SB = Department of Environmental Conservation, Stony Brook Laboratory, New York State

[2] — indicates absense of data, or no value less than 0 as reported in column titled "High"
TR = trace
ND = not determined

[3] Includes only fish analyzed for both aroclors, 4 fish not included
[4] Sample includes one highly contaminated specimen
[5] Sample does not include 6 fish for which Aroclor 1242 1016 was not determined
[6] Sample does not include 1 fish for which Aroclor 1254 was not determined
[7] Sample does not include 2 fish for which Aroclor 1254 was not determined
[8] Sample does not include 12 fish for which Aroclor 1254 was not determined
[9] Data in parentheses are weighted average PCB concentrations excluding two fish with exceptionally high concentrations

TABLE 5. *Concentrations of PCB's in fish, 1970–74*

Water	Species	Year collected	Aroclor	No. fish analyzed[1]	Number of analyses	Concentration of PCB's, ppm wet weight: Weighted average	High	Low
Hudson River (Wattensburg)	Smallmouth bass	1970	1254	1	1	0.18	0.18	–
Hudson River (Below Troy Dam)	Largemouth bass	1970–72	1254	20(19)	19(18)	6.58(4.09)	53.81	0.66
	Smallmouth bass	1970–72	1254	4(3)	4(3)	6.42(2.70)	17.58	2.56
	Striped bass	1970–72	1254	3	3	4.75	10.48	1.67
		1973	1254	22	22	14.75	49.63	3.70
	Atlantic sturgeon	1970–72	1254	4	4	6.71	7.03	5.73
	Northern pike	1972	1254	1	1	17.78	17.78	–
	Yellow perch	1972	1254	13	2	0.79	0.92	0.79
	White perch	1972	1254	39	31	4.79	15.81	0.38
	Black crappie	1970–72	1254	3	2	0.78	1.42	0.46
	Pumpkinseed	1972	1254	21	3	0.47	0.64	0.46
	Bluegill	1972	1254	4	1	0.52	0.52	–
	Redbreast sunfish	1972	1254	12	1	0.75	0.75	–
Rondout Creek	Smallmouth bass	1970	1254	2	2	14.23	17.58	10.88
	Largemouth bass	1970	1254	7	7	5.05	9.52	2.60
Rampo River	Largemouth bass	1971	1254	1	1	0.57	0.57	–
Lake Champlain	Smallmouth bass	1971	1242 1016	1	1	0.80	0.80	–
	Largemouth bass	1971	1242 1016	1	1	4.21	4.21	–
	Walleye	1971	1242 1016	2	2	28.15	55.46	0.84
	Northern pike	1971	1242 1016	1	1	0.80	0.80	–
	Chain pickerel	1971	1242 1016	1	1	0.15	0.15	–
	Atlantic salmon	1971	1242 1016	1	1	0.98	0.98	–
	Black crappie	1971	1242 1016	1	1	1.45	1.45	–
	Channel catfish	1971	1242 1016	2	2	7.00	7.85	6.15
	Brown bullhead	1971	1242 1016	1	1	0.68	0.68	–
	Freshwater drum	1971	1242 1016	1	1	0.70	0.70	–
	Carp	1971	1242 1016	1	1	1.94	1.94	–
	White sucker	1971	1242 1016	1	1	0.50	0.50	–
	Yellow perch	1971	1254	1	1	TR	TR	
	White fish (species not known)	1971	1254	1	1	0.87	0.87	–
Lake Erie	Coho salmon	1973	1254	13	13	1.30	2.91	0.60
		1974	1254	26	26	1.74	5.88	TR
	Chinook salmon	1973	1254	4	4	1.48	1.89	1.20
		1974	1254	4	4	2.98	4.92	2.24
	Smallmouth bass	1971	1254	1	1	0.42	0.42	
	Freshwater drum	1971	1242 1016	1	1	1.72	1.72	–
[illegible]	Smallmouth bass	1971	1242 1016	2	2	11.30	20.27	2.32
	Walleye	1971	1242 1016	1	1	1.02	1.02	
	Freshwater drum	1971	1242 1016	2	2	16.54	23.57	9.51

[1] [illegible]

TABLE 5 (cont'd.). *Concentrations of PCB's in fish, 1970–74*

Water	Species	Year Collected	Aroclor	No. Fish Analyzed	Number of Analyses	Concentration of PCB's, ppm wet weight: Weighted Average	High	Low
Lake Ontario	Coho salmon	1971	1254	9	9	6.67	21.97	0.66
		1974	1254	29	29	6.26	10.72	2.00
	Chinook salmon	1971	1254	12	12	21.45	34.48	9.24
		1974	1254	9	9	7.15	14.77	1.54
Lake Ontario	Rainbow trout	1974	1254	3	3	1.97	2.81	1.45
	Brown trout	1974	1254	4	4	7.69	14.50	2.70
	Northern pike	1970	1254	4	4	5.32	12.86	1.38
	Smallmouth bass	1970	1242 1016	3	3	7.11	8.81	4.38
			1254	2	2	3.88	4.52	3.24
	Largemouth bass	1970	1254	1	1	2.84	2.84	–
	Rock bass	1970	1254	2	2	0.49	0.59	0.40
	White bass	1970	1254	3	3	10.26	15.46	6.37
	White perch	1970	1254	3	3	3.42	5.15	2.12
	Carp	1970	1254	1	1	0.71	0.71	
	Rainbow smelt							
	Whole	1972	1254	10	10	4.02	7.12	2.26
	Edible	1972	1254	11	11	2.16	3.42	1.15
Lake Ontario (Salmon River Pulaski Weir)	Coho salmon	1970	1254	17	17	7.92	12.10	5.01
		1971	1254	20	20	10.35	21.43	0.83
		1972	1254	20	20	4.65	13.29	0.82
		1974	1254	5	5	7.05	11.29	3.48
Lake Ontario (Salmon River Pulaski Weir)	Chinook salmon	1971	1242 1016	2	2	19.38	28.11	10.65
			1254	4	4	9.37	12.10	5.98
		1973	1254	51	51	5.81	13.14	1.45
	Rainbow trout	1971	1254	3	3	7.24	8.38	5.76
Lake Ontario (Little Salmon River)	Coho salmon	1971	1254	3	3	2.60	6.10	0.76
	Chinook salmon	1971	1254	4	4	15.75	29.36	4.68
Lake Ontario (Spring Brook)	Coho salmon	1972	1254	1	1	6.81	6.81	
St. Lawrence River	Smallmouth bass	1971	1254	7	7	5.35	9.76	1.31
	Walleye	1971	1242 1016	1	1	3.42	3.42	
	Sturgeon (species not known)	1971	1242 1016	1	1	11.89	11.89	
	Channel catfish	1971	1242 1016	2	2	65.85	81.79	49.91
Allegheny River	Smallmouth bass	1971	1242 1016	2	2	2.21	2.36	2.06
	Muskellunge	1970	1242 1016	1	1	2.21	2.21	–
Genesee River Lower portion	Smallmouth bass	1971	1254	2	2	7.36	10.59	4.12
	White bass	1971	1254	1	1	7.76	7.76	
Chemung River	Smallmouth bass	1971	1254	3	3	0.46	0.62	0.23
	Walleye	1971	1254	2	2	1.02	1.09	0.94
	Yellow perch	1971	1254	2	2	0.14	0.28	TR
Cohocton River (above Bath)	Smallmouth bass	1971	1254	1	1	0.90	0.90	
	Brown trout	1971	1254	1	1	2.80	2.80	

(Continued next page)

TABLE 5 (cont'd). *Concentrations of PCB's in fish, 1970–74*

Water	Species	Year Collected	Aroclor	No. Fish Analyzed[1]	Number of Analyses	Concentration of PCB's, ppm wet weight: Weighted Average	High	Low
Mohawk River	Smallmouth bass	1971	1254	3	3	3 57	4 03	2 73
Barge Canal (Spencerport Holley)	Smallmouth bass	1971	1254	1	1	2 41	2 41	–
	Largemouth bass	1971	1254	1	1	1 48	1 48	–
Hooosic River	Smallmouth bass	1971	1254	2	2	2 49	2 62	2 35
	Largemouth bass	1971	1254	3	3	2 70	4 39	0 57
Raquette River	Smallmouth bass	1970	1254	1	1	0 32	0 32	–
	Yellow perch	1970	1254	1	1	0 27	0 27	–
	Pumpkinseed	1970	1254	1	1	0 81	0 81	–
Oswegatchie River	Smallmouth bass	1970	1254	7	7	1 51	4 85	0
	Largemouth bass	1970	1254	1	1	4 17	4 17	–
	Northern pike	1970	1254	5	5	0 9	0 46	0
	Yellow perch	1970	1254	9	9	0 12	1 07	0
	Pumpkinseed	1970	1254	1	1	TR	TR	–
	White sucker	1970	1254	1	1	20 92	20 92	–
Black Lake	Smallmouth bass	1971	1254	1	1	0 13	0 13	–
Brant Lake	Largemouth bass	1971	1254	1	1	TR	TR	–
Cannonsville Reservoir	Brown trout	1971	1254	6	6	0 25	1 65	0
Chautauqua Lake	Smallmouth bass	1971	1254	1	1	0 16	0 16	–
	White sucker	1971	1254	1	1	TR	TR	–
Conesus Lake	Largemouth bass	1971	1254	1	1	0 34	0 34	–
Keuka Lake	Largemouth bass	1971	1254	1	1	0 54	0 54	–
	Rainbow trout	1971	1254	1	1	1 33	1 33	–
Lake George	Lake trout	1971	1254	6	6	4 74	6 07	2 77
Onondaga Lake	Smallmouth bass	1972	1254	14	14	0 71	2 08	TR
	White perch	1972	1254	7	7	1 58	2 62	0 70
	Yellow perch	1972	1254	4	4	0 31	0 68	0
	Northern pike	1972	1254	1	1	1 75	1 75	–
	Black crappie	1972	1254	4	4	0 98	1 54	0 49
	Freshwater drum	1972	1254	4	4	1 17	1 78	0 37
	Brown bullhead	1972	1254	4	4	0 44	0 89	0 12
	Channel catfish	1972	1254	3	3	8 39	16 53	1 40
	American eel	1972	1254	2	2	5 44	6 97	3 91
[illegible] Lake	Smallmouth bass	1971	1254	1	1	0 80	0 80	–
[illegible] River	Brown trout	1971	1254	7	7	0 74	2 62	TR
Saratoga Lake	Smallmouth bass	1971	1254	1	1	1 16	1 16	–
Schoharie Reservoir	Walleye	1971	1254	3	3	0 05	0 16	0
	Brown trout	1971	1254	3	3	0	0	–
[illegible] Lake	Chain pickerel	1971	1254	3	3	TR	TR	TR
[illegible] Point [illegible]	Largemouth bass	1971	1254	1	1	0	0	–
	Eel	1971	1254	5	5	TR	TR	0

[illegible]

TABLE 5 (cont'd.). *Concentrations of PCB's in fish, 1970–74*

Water	Species	Year Collected	Aroclor	No. Fish Analyzed[1]	Number of Analyses	Concentration of PCB's, ppm wet weight: Weighted Average	High	Low
	Black crappie	1971	1254	3	3	TR	TR	0
Atlantic Ocean	Striped bass	1972	1254	5	2	0.57	0.59	0.57
	Bluefish	1972	1254	5	5	1.82	8.49	0.07
	Winter flounder	1973–74	1254	180	9	0.13	0.26	0
	Atlantic silverside	1973–74	1254	900	9	0.16	0.30	0
	Mummichog	1973	1254	11	1	0.30	0.30	
	Grubby	1973	1254	20	1	0.19	0.19	
	Spiny dogfish	1972	1254	4	4	0.25	0.27	0.21
	Northern lobster	1974	1254	1	1	0.15	0.15	-
	Oyster	1974	1254	36	3	0.34	0.75	0.12

NOTE: Results tabulated only for those fish for which the specific PCB was reported. All analyses conducted by Department of Environmental Conservation Laboratory, Rome, N.Y., except Atlantic Ocean analyses which were conducted by the Stony Brook Laboratory.
– indicates absense of data
TR = trace
[1] Parenthetic numbers exclude one individual fish with high PCB concentration

TABLE 6. *Laboratory comparisons of single aroclors in hexane and iso-octane solutions*

Sample Solution	Aroclor	Known Standard	Residues, μg ml⁻¹: DAM	DEC-SB	DEC-R	DH	EPA
Hexane	1016	0	0	0	0	0	NS
	1242	0	0	0	0	0	NS
	1254	0	0	0	0	0	NS
Hexane	1016	0.80	0.89	1.09[3]	1.15	0.71	NS
	1254	0	NV	NV	NV	NV	NS
Hexane	1016	0	NV	NV	NV	NV	NS
	1254	0.95	0.92	1.32	1.01	0.97	NS
Iso-octane (combined standards)	1016	55.5	73.3[3]	NV	39.8[3]	54.2(43.8)	26
	1254	72.0	92.8	NV	90.4	72.2(90.0)	91
	1016+1254	127.5	166.1	177[3]	130.2	126.4(133.8)	117
Iso-octane (combined standards)	1016	61.0	70.0	NV	NV	NV (103.4)	38
	1242	64.5	50.0	NV	70.3	125 NV	60
	1016+1242	125.5	120.0	145[3]			98

[1] Laboratory code: DAM = Department of Agriculture and Markets, Food Laboratory, New York State
DEC-SB = Department of Environmental Conservation, Stony Brook Laboratory, New York State
DEC-RL = Department of Environmental Conservation, Rome Laboratory, New York State
DH = Department of Health, Griffin Laboratory, New York State
EPA = U.S. Environmental Protection Agency Laboratory, Region II, New York City
[2] NV = no value reported for this Aroclor
NS = no sample sent to this laboratory
[3] Residue reported as Aroclors 1242 and 1254
[4] Residue reported as Aroclor 1242

TABLE 7 *Laboratory comparison of PCB's in fish*

Sample ID	Aroclor	Residues (ppm)[1,2] DAM	DEC SB	DEC R	DH	FDA	SD, %[3]
14830	1016	NS	NS	NV	NS	2.32	
	1254			3.69		4.37	
1483	1016	NV	NV	NV	4.3	NS	
	1254	3.3		2.58	6.0		45
	1242 1254		2.03 [4]				
14842	1016	NV	NS	NV	1.0	4.34	
	1254	8.4		7.16	9.5	8.49	11
14843	1016	NV	NV	NV	NS	NA	
	1254	6.6		5.55			
	1242 1254		2.63 [4]				
14845	1016	NV	NV	NV	NS	NA	
	1254	4.2		6.34			
	1242 1254		2.23 [4]				

Laboratory code: DAM — Department of Agriculture and Markets, Food Laboratory, New York State
DEC SB — Department of Environmental Conservation, Stony Brook Laboratory, New York State
DEC R — Department of Environmental Conservation, Rome Laboratory, New York State
DH — Department of Health, Griffin Laboratory, New York State
FDA — Food and Drug Administration Laboratory, New York, N.Y.

NV — no value reported for this Aroclor
NS — no sample sent to this laboratory
NA — sample not analyzed
SD — standard deviation, determined when there were sufficient analyses for comparison

[1] Reported as Aroclors 1242 and 1254

vidual standards, but the agreement for Aroclor 1254 is reasonable. Unfortunately, no split samples contained much Aroclor 1016 but from the synthetic check samples there is no reason to believe that agreement would be different for this mixture.

Results

Tables 4 and 5 summarize results of the 1975 PCB analyses and compilation of earlier results. For the purpose of the discussion concentrations as a function of fish length are not discussed. Data are being compiled for use as a resource management tool wherein harvest of specific lengths may be regulated to protect the consumer while using the resource. Furthermore, concentration length species correlations may shift preferred species management in contaminated waters to favor that group of fish with a low propensity for PCB accumulation. Whole fish analysis results have been separated from flesh analysis results. The data are offered as a weighted average of each individual analyzed from a composite sample and are derived using the following formula:

$$\frac{({}^{x}n)(y_n) + ({}^{x}n+1)(y_{n+1}) + ({}^{x}n+2)(y_{n+2}) + \ldots)}{({}^{x}n + {}^{x}n+1 + {}^{x}n+2 + \ldots)}$$ Weighted average (concentration of PCB's)

Where, for each species of fish:

$n, n+1, n+2$, etc. = individual or group of fish analyzed.

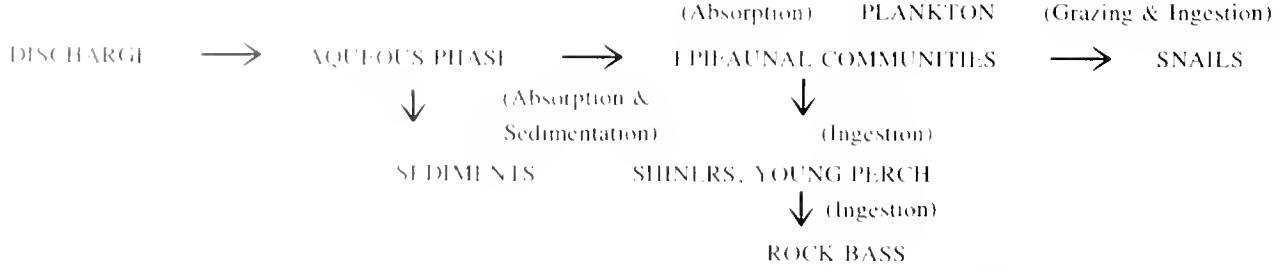
${}^{x}n, {}^{x}n+1, {}^{x}n+2$, etc. = number of fish in group $n, n+1, n+2$, etc.

y_n, y_{n+1}, y_{n+2}, etc. = concentration of PCB of group $n, n+1, n+2$, etc.

Zero concentrations were included in the weighted average. High and low individual readings in each sample are included to indicate end points of the data.

Discussion

PCB's are a ubiquitous contaminant of most environments. The source of this manufactured compound varies but appears to be primarily industries, open burning or incineration of solid wastes and waste oils, or migration from disposal sites (*13*). Once in the environment, PCB's, like organochlorine insecticides, are readily available for biomagnification since they are fat soluble compounds absorbed and stored in animal lipids. Nadeau (*16*) suggests a hypothetical transfer model for the biomagnification of PCB's in the Hudson River:



This model can be modified to show direct uptake by each biotic community from the aqueous phase (*11, 18, 19*). Through this avenue, a steady state may be achieved within a short period of time (±30 days) in highly contaminated waters (*23, 24*). Therefore, monitoring fish populations and analyzing properly composited samples for PCB contamination can indicate whether it is safe to consume fish flesh based on the Food and Drug Administration action level of 5 ppm, and to what degree a waterway is contaminated by PCB's. Indirectly, monitoring studies can show the need for specific studies to address the impact of PCB's on natural communities and/or populations, and the need to re-evaluate existing discharge permit limits.

The 1975 analyses confirmed that PCB's expressed as Aroclor 1016/1242 and/or 1254 were present in detectable amounts in most fish sampled. The level of contamination and specific Aroclor vary according to waterway size, location, and peripheral land use. Table 4 summarizes the data for edible flesh except in the cases indicated. Following is a general discussion of the findings in each waterway.

HUDSON RIVER; Stations 1–8, 32, 34. This major river system contains fish with the highest average concentration of total PCB's of any waterway sampled. Sampling nearly the entire extent of the river shows that contamination is widespread from Hudson Falls to the mouth of the river. The average level of contamination decreases in the lower reaches of the Hudson, perhaps through dilution from tributary waters, deposition in the sediments and aquatic organisms, and the effects of marine waters. Similar results occurred in fish collected from 1970 and 1973 (Table 5).

MOHAWK RIVER; Stations 9 and 10. Concentrations in 5 of 13 analyses in 1975 exceed the Food and Drug Administration action level of 5 ppm. Highest average concentrations appear upstream at the Little Falls station rather than at Schenectady, indicating a source of contamination above the town of Little Falls. The problem appears localized and does not affect the entire waterway as indicated by the Schenectady samples.

Earlier samples (Table 5) were either absent or too small to be compared.

SENECA/ONEIDA/OWSEGO RIVERS; Stations 11 and 12. PCB's are present, averaging 4 ppm. Concentrations are lowest at the Clyde station.

NIAGARA RIVER; Station 13. Largemouth and smallmouth bass have concentrations below 5 ppm; one white bass had levels above 10 ppm. The station was located below Lewiston, thus indicating no major source of PCB contamination from the Buffalo/Niagara Falls industrial complex. Earlier data are too sparse for direct comparison. However, additional study is warranted based on the 1971 analytical results and the single white bass noted above.

LAKE ERIE; Station 14. Samples from Lake Erie were taken at two locations, Dunkirk and the mouth of Cattaraugus Creek at Irving. The several species of fish contained levels of total PCB's well below 5 ppm. This is consistent with the 1973–74 data which indicated the presence of the contaminant at low concentrations.

LAKE ONTARIO; Stations 15–17. Generally, low levels of PCB's were found in warm water species, in contrast to the elevated concentrations commonly found in three species of salmonids taken from the lake proper. However, reduced levels were noted in the salmon that had migrated upstream in one major tributary, the Salmon River (Station 17). A reduction of the body fat through metabolic activities such as spawning and migration apparently caused the loss or displacement of some of the PCB burden in the edible flesh.

Comparison of the 1975 data with earlier findings is difficult. General trends indicate that concentrations similar to the present levels existed as early as 1970.

GENESEE RIVER; Station 18. Analysis of fish taken from the lower portion of the river below the village of Industry indicates a potential PCB problem in large fish. In two samples, one each of smallmouth bass and silver redhorse, levels were noted at or near 5 ppm. However, the 1971 samples, which were analyzed only for Aroclor 1254, indicate levels higher than those found in 1975. Due to the size of the earlier samples no trends can be identified except that PCB's (Aroclor 1254) are still present in substantial concentrations.

CHEMUNG/SUSQUEHANNA/DELAWARE RIVERS; Stations 19–21. These three independent river systems have been grouped for discussion since they reflect low levels of contamination, averaging 1 ppm or less. These figures are comparable to the samples collected in 1971 in the Chemung River.

LAKE GEORGE; Station 21. The larger, fatty lake trout of Lake George contain levels of PCB's (primarily Aroclor 1254) above 5.0 ppm. Lake George lake trout are known to have elevated levels of DDT (*4*). The analytical methodology used converted only DDT metabolites to dichlorobenzophenone rather than DDT itself. Therefore, PCB levels in lake trout and other species in Lake George, particularly ciscos, may be overstated. Further study of the lake is warranted.

LAKE CHAMPLAIN; Stations 23 and 24. These stations were located at Ticonderoga and Plattsburgh, respectively.

Reflected in the sampling are species in both the summer and winter fisheries as well as the limited commercial fishery. In all species sampled, total PCB's did not exceed 5 ppm although smallmouth bass from Plattsburgh approached that limit. These data are consistent with those obtained in 1971.

ST. LAWRENCE RIVER; Stations 25 and 26. Fish were collected from two stations, Massena (26) and Cape Vincent (25). Aroclor 1254 occurred but Aroclor 1242 did not. Walleye averaged about 1.8 ppm; smallmouth bass ranged from 1.62 to 6.85 ppm. Earlier data are consistent for the bass but suggest higher levels in lake sturgeon and catfish, species not sampled in 1975.

BLACK RIVER AND RAQUETTE RIVER; Stations 27 and 28. Fish from both the Black and Raquette Rivers had generally low levels of PCB contamination. These results were expected since the rivers are located in remote areas of the State away from industrial waters.

ONONDAGA LAKE; Station 31. Several species of fish, notably brown bullhead, white perch, smallmouth bass, and walleye, have concentrations of PCB's near or exceeding the FDA action level of 5 ppm. The lake has been closed to fishing since 1970 because of mercury pollution.

MARINE WATERS; Station 33. All species of fish and shellfish tested generally have total concentrations of PCB's less than 1 ppm. On rare occasions large oily fish such as bluefish have elevated levels; in one case the level exceeded 5 ppm.

CHAMPLAIN CANAL; Station 34. Fish from this waterway indicate the presence of PCB's, primarily Aroclor 1254, at low levels. A single individual rock bass, suspected to be a migrant from the Hudson River in the vicinity of Fort Edward, had levels above 5 ppm.

HEMLOCK, CANADICE, AND KEUKA LAKES, AND MONTEZUMA NATIONAL WILDLIFE REFUGE; Stations 35, 36, 38, and 41. These four waters have been grouped because they reflect low levels of PCB contamination.

CANANDAIGUA LAKE; Station 37. Chromatograms obtained for fish from this water were atypical; however, results are initially reported as PCB's. No use or spills of PCB's is known in the watershed.

SENECA AND CAYUGA LAKES; Stations 39 and 40. Previous use of small amounts of PCB's by local industry may have contaminated the fisheries. Results for Cayuga Lake (Station 40) generally confirm the findings of Bache et al. (2). Chromatograms for Seneca Lake display an atypical PCB pattern which is being investigated.

Conclusions

Nonindustrialized or lightly industrialized watersheds in New York State (Chemung, Susquehanna, Delaware, Black, and Raquette) contain fish with low levels of PCB's. Detectable amounts may result from atmospheric fallout, accidental spills, or land fill leachate. Several waterways have PCB's in amounts exceeding the action level of 5 ppm established by the Food and Drug Administration.

Rivers or waterways receiving contributions from industrialized watersheds contain organisms with high levels of PCB's. Hesse (*12*), Bremer et al. (*3*), and Veith (*22*) have indicated that levels in fish from the Great Lakes exceeded 5 ppm and that this level has not declined during the past five years. Information derived during the New York State 1975 study (Table 4) confirms these high concentrations in Lake Ontario. Large, long-lived, or fatty fish such as coho and chinook salmon, brown trout, and lake trout appear to bioaccumulate PCB's, generally above 5 ppm. A recent study (*2*) indicates that in salmonids the PCB level is directly and proportionally related to the age and oil of the fish. Findings in the present study generally support this premise.

The Hudson River below Hudson Falls contains fish with the highest level of total PCB's of any waterway sampled. Results above 50 ppm are not uncommon in the larger oilier fish; the highest individual concentration recorded was 559.25 ppm in a large eel. Comparison of the stations above Fort Edward with those immediately below indicates that a substantial PCB load is added to the river. This total loading probably does not emanate from atmospheric fallout or from other nondescript sources but rather from a major source of contamination or storage. Studies of water and sediment (*7*) have isolated industrial sources that are the subject of legal proceedings.

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RESIDUES IN SOIL

Organochlorine Insecticide Residues in Field Soils of the Kitakyushu District—Japan 1970-74[1]

M. Suzuki,[2] Y. Yamato,[2] and T. Watanabe[3]

ABSTRACT

Residues of organochlorine insecticides were monitored in field soils of the Kitakyushu District, Japan, from 1970 to 1974, before and after the insecticides were banned from agricultural fields. The arithmetic mean (AM) and geometric mean (GM) of BHC reached maximum levels in 1970 and 1971, then decreased rapidly during 1972 and 1974. Among BHC isomers, β-BHC had highest mean levels and was therefore thought to be the most persistent of the isomers in field soils. Residues of aldrin, dieldrin, and endrin peaked in 1970 and or 1971, and then decreased yearly. The percentage of sites which had aldrin contamination have decreased, those with dieldrin contamination have increased. Mean values of p,p'-DDE, p,p'-DDT, and ΣDDT declined more slowly than did BHC and cyclodienes. Values of DDT and its related compounds were higher than were those of BHC, aldrin, dieldrin, and endrin. The residues of p,p'-DDT were most frequently detected of the DDT compounds.

Introduction

Insecticides are introduced into the soil environments by three major routes: direct incorporation of insecticides into field soils to kill soil-inhabiting pests, dislodging of insecticides from treated vegetable foliage, and wind drift during insecticide application.

Edwards (*5*) reported that the years required for 95 percent disappearance of applied insecticides (1.3 lb active ingredient/acre) in field soils was 1–6 (average 3 years) for aldrin, 3–5 (average 4 years) for chlordane, 4–30 (average 10 years) for DDT, 5–25 (average 8 years) for dieldrin, 3–5 (average 3.5 years) for heptachlor, and 3–10 (average 6.5 years) for lindane, respectively. Half-lives of applied insecticides have been reported by Nash and Woolson (*15*).

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Insecticide residues have been detected in field soils 15 years after a single application of aldrin, DDT, and lindane (*10*), 15 years after BHC, DDT, and chlordane (*19*), and 20 years after an organochlorine pesticide application (*17*). Although many monitoring studies have been published (*3, 6, 8, 9, 14*), only limited information has been obtained since the ban of the organochlorine insecticides from agricultural fields.

In Japan, organochlorine insecticides such as BHC, DDT, aldrin, dieldrin, and endrin were applied extensively from the 1950's to the 1960's to control pests in rice paddies, vegetable fields, orchards, and forests. In late 1970, use of these insecticides was banned because of public concern with polluted foods. Among these insecticides, BHC had been applied the most extensively; the amounts of production in 1967 and 1968 were 41,742 tons and 45,695 tons, respectively. The ratio of BHC production to total arable land was approximately 0.81 lb/acre in 1968. That year, 767 tons of cyclodiene insecticides were imported, and 4,936 tons of DDT were produced. Since 1971, however, the production of these insecticides has almost ceased.

The present paper reports organochlorine insecticide residues and related compounds in field soils of the Kitakyushu District, Japan (Fig. 1), before and after a ban of these insecticides.

Sampling Procedures

Soil samples were taken from fields where the primary crops for many years had been vegetables such as radishes, cabbages, spinach, carrots, chinese cabbages, cucumbers, turnips, tomatoes, eggplants, and lettuce. Considerable amounts of organochlorine insecticides had been applied, although no detailed histories of insecticide applications could be obtained. The textures of the soil samples varied from loamy to sandy. Samples were collected in cores 15 cm deep and 10 cm in diameter at diagonal points of the field. Sampling schedules appear in Table 1.

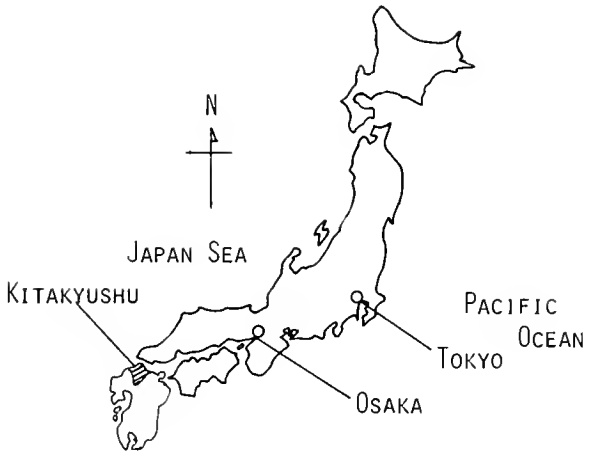


FIGURE 1. *Map of Japan showing Kitakyushu District*

Analytical Methods

EQUIPMENT AND REAGENTS

Kuderna-Danish evaporative concentrator with three-ball Snyder column with ground glass fittings
Chromatographic column, 20 by 300 mm
Five-ml graduated concentrator with a ground glass fitting
Squibb-type 1-liter separatory funnel
Buchner glass funnel, 25G4 (150-200 mesh)
Sanyo mixer, SM–1000
Shimadzu GC–5AIEE gas chromatograph with dual tritium-foil electron-capture detectors (300 mCi)
Acetonitrile redistilled in accord with AOAC methods (*1*)
n-Hexane redistilled twice in an all-glass distillation system
Reagent grade anhydrous sodium sulfate heated 2 hours at 625°C to eliminate contaminants
Redistilled water washed thoroughly with n-hexane
Insecticide standards with 99+percent purity

PRETREATMENT

The soil sample was dried at room temperature, pulverized with a mortar, and screened through a 20-mesh sieve. The screened soil was put into a ball mill jar, rotated without balls for 5 hours, and mixed well.

TABLE 1. *Schedule for soil sampling for organochlorine insecticide analysis, Kitakyushu District, Japan—1970—74*

Collection Dates	No. samples collected
November 26, 1970	24
July 13, 1971	19
September 14, 1971	4
November 5, 1971	45
July 16, 1972	17
November 17, 1972	20
July 9, 1973	16
December 13, 1973	22
November 21, 1974	33

EXTRACTION

All analyses were carried out in duplicate and the results represent an average of duplicate analyses. A 100–g sample was put into a mixer and 0.7 volume of distilled water per 100 g soil sample was added to deactivate. After 30 minutes, 200 ml of acetonitrile was added and the mix was blended 5 minutes at a high speed. An extract and the rinsings were filtered through a Buchner glass funnel, and the filtrate was poured into a 1–liter separatory funnel and shaken with 100 ml of n-hexane for 5 minutes. Then 600 ml of distilled water was added and the funnel was shaken for 1 minute to achieve partitioning. The resulting aqueous phase was drained and the n-hexane phase was washed twice with distilled water. The n-hexane phase was dried by passing through a column of 5 cm anhydrous sodium sulfate.

The dried n-hexane phase was concentrated by a Kuderna-Danish evaporative concentrator. Heptachlor epoxide was added as an internal standard and subjected into a gas chromatograph equipped with a dual channel electron-capture detector. Recovery was well above 90 percent; results were not corrected.

GAS CHROMATOGRAPHY

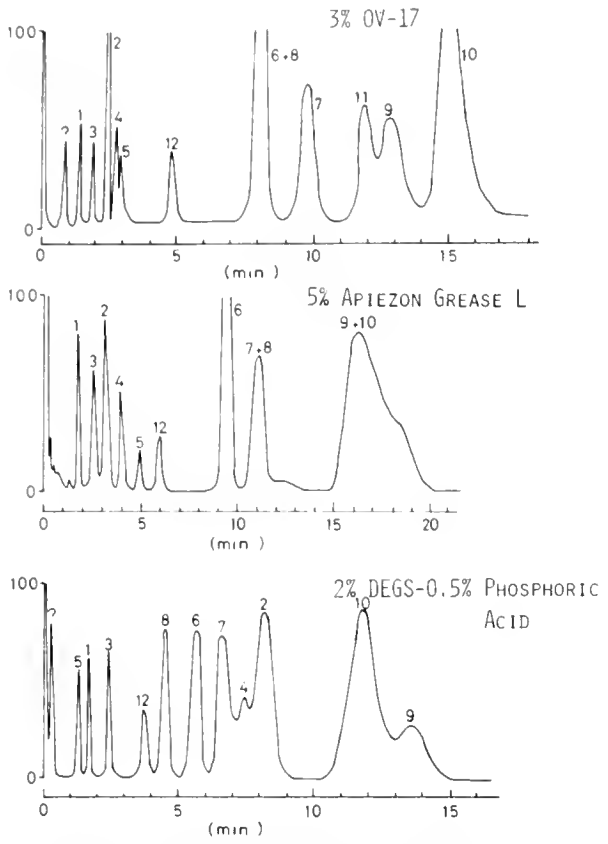
Organochlorine insecticides and related compounds in soil samples were identified and measured as described in the previous report (*22*).

Results and Discussion

The typical gas chromatograms of residual organochlorine insecticides and their related compounds in soil samples are shown in Figure 2. Results of the present monitoring study are presented in Table 2. For each year, the total sampling sites, the number and percent of sites which had insecticides and/or related compounds, the range of residue detected, and the arithmetic mean (AM) and geometric mean (GM) are given.

Residues of α–, β–, and δ–BHC were detected beside insecticidally active γ–BHC (lindane) in the soil samples analyzed, since crude BHC had been formulated and marketed as BHC insecticides in Japan. AM and GM values of BHC were highest during 1970 and 1971, then decreased yearly. The high residues in 1971, one year after the ban, reflect the application of stocked BHC purchased before the ban.

In 1970, the high value of 3.960 ppm reported for α–BHC occurred because the field had been sampled shortly after BHC application. Levels of BHC decreased considerably from 1971 to 1974. Soil in 1974 contained only 10.7 and 22.4 percent of the 1971 AM and GM mean levels of α–BHC, 21.6 and 18.4 percent of the levels for β–BHC, 16.3 and 25.4 percent for γ–BHC, and 17.3 and 16.0 percent for δ–BHC. These results indicate rapid dissipation of



1: α-BHC; 2: β-BHC; 3: γ-BHC; 4: δ-BHC; 5: aldrin; 6: dieldrin; 7: endrin; 8: p,p'-DDE; 9: p,p'-TDE; 10: p,p'-DDT; 11: o,p'-DDT; 12: heptachlor epoxide (internal standard).

FIGURE 2. *Typical gas chromatograms of insecticides and their related compounds in soils*

BHC isomers from field soil. The average residue in 1970 was composed of 28 percent α-BHC, 44 percent β-BHC, 14 percent γ-BHC, and 14 percent δ-BHC. In 1974, ratios were 8 percent α-BHC, 77 percent β-BHC, 7 percent γ-BHC, and 8 percent δ-BHC. The marked increase in percentage of β-BHC in total BHC corroborates the previous report that β-BHC was the most persistent isomer of BHC in the soil (*21*); the residue levels of total BHC could be expected from the level of β-BHC. In contrast to β-BHC, α-BHC and γ-BHC might degrade considerably in field soil. All the BHC isomers were detected in all soil samples analyzed in 1970–73. In 1974, however, one sample had no γ-BHC and four samples had no δ-BHC, which illustrates the different degrees of persistence among BHC isomers in soil.

AM and GM values of aldrin and dieldrin peaked in 1971, then decreased yearly with minor exceptions. The detection ratios of aldrin were 8.3 percent in 1970, 26.8 percent in 1971, 48.6 percent in 1972, 26.3 percent in 1973, and 21.2 percent in 1974. Ratios were highest in 1972 and decreased gradually due to oxidative conversion and microbial degradation of aldrin to dieldrin in soil (*11, 12*). Detection ratios of dieldrin were 50 percent in 1970, 48.5 percent in 1971, 62.2 percent in 1972, 68.4 percent in 1973, and 75.8 percent in 1974. Unlike aldrin, dieldrin ratios increased yearly, due to the conversion of aldrin to dieldrin in soil. Although AM and GM values of dieldrin decreased yearly from 1970 to 1973, no marked decrease of these values was observed in 1974. The maximum AM and GM values of endrin were recorded in 1971, but considerable decreases occurred afterward. Values obtained in 1974 were about a tenth and a twelfth of those in 1971. For dieldrin, values were about a third and two-fifteenths of the 1971 values. Hence, of these stereo-isomers, the endo-type, endrin, was more degradable in soil than the exo-type, dieldrin. Nash et al. (*16*) reported that endrin was readily converted to its delta-ketone, alcohol, and aldehyde in soil with a pH of 6.4. This suggests that endrin is considerably degradable in the acidic soil common in arable fields of Japan.

AM and GM levels of DDT and ΣDDT were considerably higher than those of BHC and cyclodiene insecticides. Among DDT and related compounds, *p,p'*-DDT and *p,p'*-DDE were detected most frequently. Residues of *p,p'*-TDE and *o,p'*-DDT were detected in smaller quantities. Detection ratios of *p,p'*-DDE were 75.0 percent in 1970, 73.5 percent in 1971, 78.4 percent in 1972, and 100 percent in 1973 and 1974. Ratios increased yearly with minor exceptions due to the conversion of *p,p'*-DDT to *p,p'*-DDE in soil by dehydrochlorination. But detection ratios of *p,p'*-DDT have not decreased, suggesting widespread contamination of the soil by DDT as well as BHC. Cliath and Spencer (*4*) reported that the disappearance of DDT from well-aerated soil was due to the conversion to DDE and subsequent dissipation of DDE into the atmosphere. This suggests that DDT residing in field soil might degrade slowly to DDE; however, its dissipation might be accelerated later.

Insecticides in the environment can be degraded by physico-chemical action or by microbial action. Generally, the physico-chemical action occurs shortly after insecticide application, and microbial action occurs more slowly, among the persistent residues. In the soil, microorganisms are the most active factor in insecticide degradation. For instance, BHC is degradable in reductive conditions and in anaerobic microbe-rich soil (*13, 21*).

In the degradation process, γ-BHC is initially converted to γ-pentachlorocyclohexene, and secondly to γ-tetrachlorocyclohexene (*23*). Isomerization of γ-BHC to the other isomers was reported when γ-BHC was incubated with sediments from Pearl Harbor, Hawaii (*2*), and Lake Tomahawk, Wis. (*18*). DDT was easily converted to TDE when the soil was flooded and alfalfa was applied to the field as an organic matter (*7*). In this manner, insecticides might be dissipated from the soils; however, organochlorine insecticides studied here were generally far

TABLE 2. *Insecticide residues in soil from Kitakyushu District, Japan—1970–74*

Pesticide	No Positive Sites	Percent Positive Sites	Residues, ppm dry weight: Range	Arith Mean	Geom Mean
1970 24 Sites					
α-BHC	24	100 0	0 006–3 960	0 233	0 0439
β-BHC	24	100 0	0 062–2 420	0 352	0 2158
γ-BHC	24	100 0	0 008–1 084	0 108	0 0456
δ-BHC	24	100 0	Tr –0 789	0 108	0 0551
∑BHC	24	100 0	0 076–8 253	0 791	0 4325
Aldrin	2	8 3	0 085–0 263	0 174	0 1496
Dieldrin	12	50 0	0 002–1 726	0 308	0 0769
Endrin	5	20 8	0 016–0 125	0 054	0 0275
p,p'-DDE	18	75 0	Tr –0 650	0 131	0 0543
p,p'-DDT	18	75 0	Tr –6 450	0 577	0 1820
∑DDT	18	75 0	Tr –8 254	0 810	0 2999
1971 68 Sites					
α-BHC	68	100 0	0 002–1 948	0 131	0 0425
β-BHC	68	100 0	0 021–2 612	0 631	0 3716
γ-BHC	68	100 0	0 003–0 965	0 080	0 0334
δ-BHC	68	100 0	Tr –0 748	0 081	0 0455
∑BHC	68	100 0	0 026–6 273	0 931	0 5210
Aldrin	25	36 8	0 002–1 012	0 074	0 0189
Dieldrin	33	48 5	0 024–1 014	0 294	0 1950
Endrin	21	30 9	0 036–0 654	0 191	0 1330
p,p'-DDE	50	73 5	0 005–2 048	0 167	0 0656
p,p'-DDT	40	58 8	0 015–9 801	1 259	0 5129
∑DDT	50	73 5	0 049–11 882	1 830	0 7481
1972 37 Sites					
α-BHC	37	100 0	0 002–0 800	0 044	0 0174
β-BHC	37	100 0	0 009–1 250	0 226	0 1492
γ-BHC	37	100 0	0 002–0 073	0 021	0 0178
δ-BHC	37	100 0	0 002–0 500	0 037	0 0189
∑BHC	37	100 0	0 026–6 273	0 328	0 2133
Aldrin	18	48 6	0 002–0 280	0 045	0 0220
Dieldrin	23	62 2	0 003–0 745	0 263	0 1089
Endrin	15	40 5	0 015–0 629	0 145	0 0847
p,p'-DDE	29	78 4	0 009–1 044	0 165	0 0867
p,p'-DDT	28	75 7	0 022–6 661	1 014	0 3467
∑DDT	29	78 4	0 031–10 994	1 487	0 5508

(Continued next page)

TABLE 2 (cont'd.). *Insecticide residues in soil from Kitakyushu District, Japan—1970–74*

Pesticide	No. Positive Sites	Percent Positive Sites	Residues, ppm dry weight		
			Range	Arith. Mean	Geom. Mean
1973—38 Sites					
α-BHC	38	100.0	0.001–0.864	0.034	0.0071
β-BHC	38	100.0	0.005–0.686	0.121	0.0708
γ-BHC	38	100.0	Tr.–0.399	0.025	0.0181
δ-BHC	38	100.0	Tr.–0.125	0.018	0.0079
ΣBHC	38	100.0	0.006–1.573	0.196	0.1069
Aldrin	10	26.3	Tr.–0.007	0.002	0.0033
Dieldrin	26	68.4	0.002–0.250	0.063	0.0268
Endrin	19	50.0	0.006–0.109	0.038	0.0252
o,p'-DDE	38	100.0	0.001–0.831	0.117	0.0455
p,p'-DDT	38	100.0	0.006–4.174	0.686	0.2925
ΣDDT	38	100.0	0.007–4.989	0.863	0.1808
1974—33 Sites					
α-BHC	33	100.0	0.001–0.067	0.014	0.0095
β-BHC	33	100.0	0.002–1.172	0.136	0.0682
γ-BHC	32	97.0	0.001–0.100	0.013	0.0085
δ-BHC	29	87.9	0.001–0.194	0.014	0.0073
ΣBHC	33	100.0	0.003–1.533	0.177	0.1023
Aldrin	7	21.2	0.002–0.009	0.005	0.0039
Dieldrin	25	75.8	0.001–0.506	0.087	0.0263
Endrin	23	69.7	0.001–0.102	0.022	0.0109
p,p'-DDE	33	100.0	0.006–0.762	0.104	0.0388
o,p'-DDT	33	100.0	0.008–4.605	0.518	0.1049
ΣDDT	33	100.0	0.018–5.504	0.671	0.2009

NOTE: Tr. = trace, ≤ 0.001 ppm.
ΣBHC = α-BHC + β-BHC + γ-BHC + δ-BHC.
DDT = *o,p'*-DDE + *p,p'*-TDE + *p,p'*-DDT + *o,p'*-DDT.

more persistent than are organophosphorus and/or carbamate insecticides.

Translocation of insecticides from field soil into vegetables has been reported (*12, 20*). In Japan, vegetables have been highly contaminated with organochlorine insecticides (*22*). Despite a recent ban on their use, residue levels often still exceed the pesticide residue tolerance set by the Japanese government. Therefore, organochlorine insecticides in field soil should be monitored during the next several years, in conjunction with the study of insecticide residues in vegetables.

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GENERAL

Dieldrin Residues in Soybeans in Illinois, 1965, 1966, 1967, 1971, and 1974 [1]

Steve Moore III, H. B. Petty, W. N. Bruce, Roscoe Randell, and D. E. Kuhlman

ABSTRACT

The Illinois soybean crop was monitored in 1965, 1966, 1967, 1971, and 1974 for dieldrin residues resulting from aldrin applied to corn in the years before soybean cultivation. Residue levels of dieldrin in soybeans increased between 1965 and 1974. The percent of fields which had soybeans with a dieldrin level above 0.03 ppm increased between 1965 and 1974. Dieldrin residue levels in soybeans grown in Illinois are expected to decline now that the use of aldrin has diminished and will soon cease. No significant correlation was evident between the dieldrin levels in soybeans and the area of the State where they had grown, the date of planting, or the variety.

Introduction

Aldrin has been the principal chlorinated hydrocarbon insecticide used on corn soil in the United States for the control of underground insects since the early 1950's. It was applied and incorporated into the soil at an average rate of 1.1 pounds of active ingredient per acre. According to Wedberg et al. (*13*), aldrin use reached its peak in Illinois in 1967 when 4,481,258 acres were treated (Table 1). Aldrin use has declined since that time due to the development of insect resistance and concern by farmers about residue problems. Decker et al. (*5*) showed that after an aldrin application small amounts of its epoxide, dieldrin, remained in the soil for at least 12 years and their disappearance could be predetermined with some certainty (Table 2). There is a rapid loss of adrin dieldrin residues the first two years after the aldrin application due to the volatility of the aldrin and the desorption chracteristics of the soil. The determination was based on a 2-year half-life (continuous row cropping) and 4-year half-life (a rotation involving no-till cropping) after the first 2 years. Thus soil residues in a field can be calculated from the aldrin treatment history. The amount and persistence of residues vary with such factors as soil type, cropping history, weather, and cultivation.

Soybeans are exposed to dieldrin residues when growing in fields where aldrin has been used on corn in previous years. Rotation of corn and soybeans is a common practice in Illinois. From 1964 to 1974, 9 to 10 1/2 million acres of corn and 5 to 8 1/2 million acres of soybeans were grown annually. In 1959 and 1960, aldrin was tested for control-

TABLE 1. *Use of aldrin on corn soil in Illinois, 1956–1974* [1]

Year	Acres Treated with Aldrin	Year	Acres Treated with Aldrin
1956	314,679	1966	4,093,284
1957	558,677	1967	4,481,258
1958	663,326	1968	4,136,580
1959	1,126,417	1969	3,569,165
1960	1,607,689	1970	2,883,555
1961	2,187,740	1971	2,042,339
1962	2,979,354	1972	1,449,817
1963	3,441,920	1973	1,216,257
1964	3,407,908	1974	1,320,229
1965	3,635,546		

[1] Adapted from Wedberg et al., 1975, literature reference (*13*).

TABLE 2. *Theoretical residues (aldrin + dieldrin) to be expected in soil at yearly intervals after aldrin applications* [1]

Years after application	Two-year Half-Life	Four-year Half-Life
0	0.5	0.5
1	0.13	0.13
2	0.10	0.10
3	0.07	0.085
4	0.05	0.07
5	0.035	0.06
6	0.025	0.05
7	0.018	0.043
8	0.013	0.035
9	0.009	0.030
10	0.006	0.025

NOTE: Residues measured in ppm each spring after application of 1 lb. a. i. in top 6 inches of soil.

[1] Adapted from Decker, Bruce, and Bigger, 1965, literature reference (*5*).

[1] Contribution from the Illinois Agricultural Experiment Station. [2] Entomologist, Illinois Natural History Survey, 169 Natural Resources Building, Champaign.

ling soil-infesting insects in soybeans. An analysis of the soybeans showed the presence of aldrin/dieldrin residues. These chemicals are translocated from the soil to grain or fruit, as demonstrated by Lichtenstein (*7–10*), Bruce et al. (*2*), Bruce and Decker (*1*), and Eden and Arthur (*6*).

According to Bruce and Decker (*1*), the magnitude of the actual dieldrin residue in soybeans is about 6 to 11 percent (average 8 percent) of the actual dieldrin residue in the soil (Table 3). Turner et al. (*12*) showed that more dieldrin is present in the bean seeds from the lower half of the plant than in those from the upper half. Petty (*11*) reported the magnitude of dieldrin residues in soybeans in Illinois, and suggested that farmers allow at least 2 years to elapse between the last application of aldrin and the planting of soybeans in a field in order to produce soybeans free of significant dieldrin residues. The purpose of the present study was to determine the residue levels of dieldrin in soybean seeds produced in Illinois and to determine trends in dieldrin residue levels through the years.

Sampling Procedures

A random survey of Illinois soybeans was first conducted in 1965 to determine the levels of dieldrin residues (*3*). Similar surveys followed in 1966, 1967, 1971, and 1974. Using a system of random selection for objective yields, the Illinois Crop Reporting Service Enumerators obtained raw soybeans, and field histories of insecticide use and cropping practices. Between 52 and 106 fields of soybeans were randomly selected and sampled during each study year. Soybeans were collected in 1-quart screw-cap glass jars which were covered with aluminum foil before the cap was fastened. The samples were obtained by holding the jar at the edge of the stream of soybeans as they were augered or elevated into the bin or wagon. The samples were taken at harvest and submitted to the Illinois Natural History Survey for analysis. The analyses, which were based on the weight of the whole soybean, were accurate within ±0.001 ppm as shown in trial recoveries (Table 4).

TABLE 3. *Aldrin and dieldrin residues in soil, and corresponding dieldrin residue in soybeans grown in same soil after aldrin application* [1]

Year	Residues, ppm		Percent Recovery
	Soil	Beans	
1961	0.390	0.043	11.0
1962	0.253	0.026	10.3
1963	0.174	0.017	9.8
1964	0.092	0.006	6.5

NOTE: Aldrin applied at 2 lb/a on April 14, 1961

[1] Adapted from Bruce and Decker, 1966, literature reference (*1*)

TABLE 4. *Recovery of dieldrin added to soybean flour*

Dieldrin Added, ppm	Dieldrin Recovered, ppm		
	Replicate 1	Replicate 2	Replicate 3
0.0000	0.0000	0.0040	0.0005
0.0010	0.0004	0.0008	0.0000
0.0020	0.0015	0.0017	0.0018
0.0050	0.0036	0.0051	0.0048
0.0100	0.0095	0.0110	0.0092
0.0200	0.0195	0.0172	0.0190

Analytical Technique

SAMPLES

Fifty-gram lots of soybeans were hexane-rinsed to remove harvest dust which may have contained trace amounts of dieldrin. Soybeans were dried and blended at high speed until reduced to a fine flour. This facilitated the complete extraction of oil from the 10-g flour aliquots used for each replicate. The soybeans contained about 21± percent oil by weight, but the analysis was based upon the weight of the whole soybean, and no special effort was made to survey oil contents of beans. Analyses were made in triplicate.

REAGENTS AND MATERIALS

All solvents used were nanograde or redistilled in glass. Florisil and Na_2SO_4 were baked at 450°C for 18 hours to remove moisture and impurities, then stored in glass-stoppered 1000-ml Erlenmeyer flasks.

EXTRACTION

Ten grams of soybean flour was added to a 250-ml Erlenmeyer flask with 150 ml 10 percent ether in hexane and heated overnight at 50°C. The mixture was filtered through No. 1 Whatman filter paper into another Erlenmeyer flask. The flour residue in the filter paper was rinsed several times with hexane to ensure complete transfer of the extracted oil. A few grains of Na_2SO_4 was added to the filtrate, a Snyder column was attached, and all but about 10 ml of the solvents were removed by distillation through the Snyder column.

PARTITIONING WITH ACETONITRILE

The extracted oil, approximately 2 g, was transferred to a 125-ml separatory funnel with four portions of 5 ml hexane. This hexane/oil mixture was extracted 4 times with 25-ml portions of acetonitrile which had been saturated with hexane. Each successive partition was shaken 100 times, the phases were allowed to separate, and the lower acetonitrile portion was drained into a 1000-ml separatory funnel.

Five hundred ml water and 100 ml hexane were added to the combined acetonitrile in the 1000-ml separatory funnel. The mixture was shaken 200 times and the phases were allowed to separate. The upper hexane was retained and washed twice with water to remove traces of acetonitrile. The hexane layer was drained into a 125-ml Erlen-

meyer flask containing a small quantity (2–3 g) of Na_2SO_4 which effectively removed the water. After standing for about an hour the hexane was transferred to a 250-ml Erlenmeyer flask with several rinses of hexane. A few crystals of Na_2SO_4 were added as boiling chips, a Snyder column was attached, and the solvent was removed by distillation on the steam bath.

FLORISIL COLUMN CHROMATOGRAPHY

Thirty g florisil containing 3 percent water was prepared for chromatographic separation by pre-elution with 50 ml acetone, 50 ml anhydrous diethyl ether, and finally with 100 ml dry nanograde hexane.

The pesticide extract in 2 ml hexane was carefully transferred to the florisil column with 3 to 4 1-ml portions of hexane; 250 ml 10 percent ether in hexane elutes aldrin, dieldrin, heptachlor, and heptachlor epoxide from the column. Eluates were collected in 500-ml Erlenmeyer flasks, Snyder columns were attached, and volume was reduced on a steam bath to approximately 5 ml. This was transferred to a 10 ml volumetric flask for gas chromatographic (GC) determination.

A model 204 Varian Aerograph gas chromatograph equipped with an electron-capture detector was used for dieldrin quantitation. Operating conditions were:

Column	high resolution, 3.3 mm by 3 m, packed with 100/120-mesh Supelcoport containing 2.0 QF-1 and 1.25 percent OV-17
Detector	electron capture, ^{63}Ni
Temperatures	injection port 225 C column 200 C detector 255 C
Carrier gas	nitrogen, at a flow rate of 25 ml/min
Sensitivity	10 picograms dieldrin caused a 10 percent recorder response with a noise level of approximately 1 percent

Results and Discussion

Residue levels of dieldrin in raw soybeans were higher in 1974 than in previous years (Table 5). This is in spite of the declining use of aldrin on Illinois cropland since 1967 (Table 1). Apparently the dieldrin level in Illinois is beginning a gradual decline and residues of dieldrin in raw soybeans probably will also decline; further monitoring of soybeans is needed to prove this theory. Soybeans from fields previously treated with aldrin had a higher residue of dieldrin than had soybeans from untreated fields. It is possible that dieldrin residues occur in raw soybeans from untreated fields due to the movement of dieldrin-contaminated soil particles by air and/or water from previously treated fields. In a study in Cincinnati, Ohio, Cohen and Pinkerton (4) reported the average monthly dust fall to be 15 tons/square mile. The dust, which originated in the southern high plains of Texas, contained 0.003 ppm dieldrin. Authors further stated that soil particles are certain to move by air within a local area.

TABLE 5 *Dieldrin residues in soybeans by years in Illinois*

	All Fields		From Treated Fields[1]		From Untreated Fields[2]	
Year	No.	ppm	No.	ppm	No.	ppm
1965	88	0.0089	57	0.0101	31	0.0068
1966	95	0.0077	43	0.0119	52	0.0043
1967	52	0.0102	17	0.0187	35	0.0061
1971	106	0.0101	43	0.0165	63	0.0058
1974	72	0.0136	23	0.0190	49	0.0110

NOTE: Residues reported in average ppm.

[1] Soil treated with aldrin when field was in corn during previous years.
[2] Soil not treated with aldrin during previous years.

The percentage of fields with high dieldrin residues in soybeans has increased since the initial survey in 1965 (Table 6). In 1965 and 1966, no fields were found with residues above 0.03 ppm in soybeans. In 1967, 1.5 percent of the fields sampled had soybeans with a dieldrin residue above 0.03 ppm. In 1971, this increased to 7.1 percent of the fields and in 1974, to 8.4 percent. There is no official tolerance for dieldrin in soybeans so these residue levels are of concern.

Dieldrin residues in soybeans grown after corn varied greatly even where aldrin use was similar. This can be expected because of variation in soil, slope, and farmers' cultivation practices. Greatest variation occurred where aldrin had been applied as a row or furrow treatment. Authors assume that in some fields the soybean rows were spaced between the corn rows of the previous year, resulting in lower residues than would occur where the soybean rows coincided with the corn rows of the previous year. In one instance in 1971, soybeans from one field contained 0.07–0.08 ppm dieldrin, an extraordinarily high residue. The farmer had used only 1 pound of actual aldrin per acre for the 5 consecutive seasons from 1966 to 1970. The dieldrin residues in the soybeans were greater than expected. Detailed discussions revealed that the farmer had been meticulous in his application rates and had started planting each year precisely the same distance from the fence. A

TABLE 6 *Percent of fields with dieldrin residues in soybeans at various ranges in 1965, 1966, 1967, 1971, and 1974 in Illinois*

		Percent of Fields per Range						
Residue Range	ppm	1965	1966	1967	1971	1974	Avg.	No. Fields
0.0–0.01		67.8	72.5	58.5	58.8	52.8	64.0	264
0.01–0.02		31.1	20.6	29.2	24.7	26.3	26.3	109
0.02–0.03		1.1	6.9	10.8	9.4	12.5	7.3	30
0.03–0.04		0	0	1.5	5.9	4.2	1.2	5
0.04–0.05		0	0	0	0	4.2	0.9	4
0.05–0.06		0	0	0	0	0	0	0
0.06–0.07		0	0	0	0	0	0	0
0.07–0.08		0	0	0	1.2	0	0.3	1

narrow band of aldrin/dieldrin concentration was formed, resulting in an abnormally high pickup by the soybeans.

The area of Illinois in which the soybeans were grown showed no significant variations in the level of dieldrin residues present (Table 7).

Planting dates had no effect on the dieldrin residues in soybeans (Table 8).

Dieldrin residues in soybeans from fields with a history of aldrin application ranged from 0.0140 to 0.0201 ppm for different planting dates; no pattern was indicated.

Soybean varieties showed no apparent difference in dieldrin residue (Table 9).

Table 10 lists dieldrin levels in soybeans according to oil content. Data were insufficient to correlate dieldrin residues with oil content of the soybeans.

Conclusions

Residue levels of dieldrin in Illinois soybeans increased between 1965 and 1974.

The percent of fields which had soybeans with a dieldrin level above 0.03 ppm increased between 1965 and 1974.

Dieldrin residues in Illinois soybeans are expected to decline gradually now that use of aldrin has diminished and will soon cease.

No significant correlation was found between dieldrin levels in soybeans and the area of the State where they were grown, date of planting, or variety.

Acknowledgments

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TABLE 7. *Dieldrin residues in soybeans by districts in Illinois*

District	All Fields		From Treated Fields[1]		From Untreated Fields[2]	
	No	ppm	No	ppm	No	ppm
Northwest	22	0.0093	12	0.0110	10	0.0072
Northeast	22	0.0123	15	0.0148	7	0.0069
West	36	0.0110	27	0.0122	9	0.0073
Central	36	0.0112	20	0.0137	16	0.0081
East	33	0.0101	17	0.0130	16	0.0069
West-southwest	33	0.0115	18	0.0148	15	0.0076
East-southeast	53	0.0095	23	0.0153	30	0.0050
Southwest	24	0.0054	7	0.0092	17	0.0038
Southeast	19	0.0080	10	0.0123	9	0.0032

NOTE. Residues reported in average ppm
[1] Soil treated with aldrin when field was in corn during previous years
[2] Soil not treated with aldrin during previous years

TABLE 8. *Actual dieldrin residues in soybeans by planting date in Illinois*

Planting Date	All Fields		From Treated Fields[1]		From Untreated Fields[2]	
	No	ppm	No	ppm	No	ppm
Up to May 10	30	0.0109	13	0.0165	17	0.0066
May 11–15	27	0.0114	10	0.0201	17	0.0063
May 16–20	23	0.0103	12	0.0146	11	0.0055
May 21–25	21	0.0098	8	0.0140	13	0.0073
May 26–30	21	0.0115	9	0.0194	12	0.0057
May 31–June 4	18	0.0094	5	0.0177	13	0.0062
June 5 and later	18	0.0062	3	0.0199	15	0.0035

NOTE. Residues reported in average ppm
[1] Soil treated with aldrin when field was in corn during previous years
[2] Soil not treated with aldrin during previous years

TABLE 9. *Dieldrin residues in soybeans by variety in Illinois*

Variety	All Fields		From Treated Fields[1]		From Untreated Fields[2]	
	No	ppm	No	ppm	No	ppm
Wayne	51	0.0124	21	0.0209	30	0.0065
Clark	24	0.0043	4	0.0087	20	0.0034
Mixtures	21	0.0080	7	0.0114	14	0.0063
Amsoy	13	0.0137	9	0.0171	4	0.0060
Harosoy	10	0.0127	4	0.0224	6	0.0061
Beeson	10	0.0101	6	0.0115	4	0.0080
Harosoy 63	6	0.0141	3	0.0204	3	0.0078
Clark 5	5	0.0045	0	–	5	0.0045
Shelby	5	0.0122	3	0.0128	2	0.0112
Hawkeye	3	0.0063	0	–	3	0.0063
0102	2	0.179	1	0.0197	1	0.0162
Adams	2	0.0127	1	0.0175	1	0.0078
Dare	2	0.0038	0	–	2	0.0038
0207	1	0.0066	0	–	1	0.0066
Hawkeye 63	1	0.0052	0	–	1	0.0052
Scott	1	0.0019	1	0.0019	0	–
Chippewa	1	0.0241	1	0.0241	0	–
Unknown	183	0.0083	100	0.0109	83	0.0052

NOTE. Residues reported in average ppm
– = samples lost during shipment or analysis
[1] Soil treated with aldrin when field was in corn during previous years
[2] Soil not treated with aldrin during previous years

TABLE 10. *Dieldrin residues and oil content of soybeans in Illinois*

Oil Content, %	Dieldrin Residues					
	All Fields		From Treated Fields[1]		From Untreated Fields[2]	
	No	ppm	No	ppm	No	ppm
0.0–20.0[3]	195	0.0083	102	0.0109	93	0.0053
20.0–21.0	23	0.0106	8	0.0194	15	0.0058
21.0–22.0	46	0.0121	19	0.0209	27	0.0059
22.0–23.0	41	0.0105	16	0.0167	25	0.0064
23.0–24.0	25	0.0091	12	0.0134	13	0.0051
24.0 and over	10	0.0052	3	0.0053	7	0.0052

[1] Soil treated with aldrin when field was in corn during previous years
[2] Soil not treated with aldrin during previous years
[3] Less than 20 percent or no data available. In most instances, oil content was unknown. This figure should be disregarded when comparing oil content and dieldrin residues

recting the random selection of soybeans and for obtaining field histories of insecticide use and cropping practices.

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Preliminary Monitoring of Agricultural Pesticides in a Cooperative Tobacco Pest Management Project in North Carolina, 1971—First-Year Study [1]

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ABSTRACT

A preliminary tobacco pest management pesticide monitoring project was initiated in two counties of North Carolina in 1971 to develop an ecologically, economically, and socially acceptable system for protecting tobacco from damaging insect and disease pests. Results from the first year's study including sampling procedures, sample preparation, and analysis and residue data are presented in this paper.

Introduction

A cooperative tobacco pest management project was initiated in North Carolina in 1971 by the United States Department of Agriculture (USDA), North Carolina Department of Agriculture, and the North Carolina Agricultural Extension Service.

The pest management area was 145 square miles which contained 685 tobacco farms, 1369 tobacco fields, and 4,244 acres. Average tobacco acreage was 6.2 per farm and 3.1 per field.

This pilot study was an attempt to establish an ecologically, economically, and socially acceptable system for protecting tobacco crops from insect pests, as described by Ganyard et al. (*2*).

Another purpose of the preliminary study was to monitor pesticide residues in biotic and abiotic environmental components prior to application of the chemicals and following harvest of the crops, both within and outside program areas. Twenty sampling sites were selected in Wayne and Wilson Counties, ten of which were located within the pest management project area and ten in a geographically adjacent area. Each site consisted of a tobacco field with a farm pond located within 300 feet of the subject field and within the drainage area of the field. Ponds were reasonably uniform in size; duplicate samples were collected at each site according to standard sampling procedures.

Samples were collected in the spring before pesticide applications and again in the fall after pesticide treatments.

Biotic groups sampled in 1971 include bluegill (*Lepomis macrochirus*); four species of turtles, (a) snapping turtles (*Chelydra serpentina*), (b) musk turtles (*Sternotherus odoratus*), (c) painted turtles (*Chrysemys picta*), and (d) yellow-bellied turtles (*Chrysemys scripta*); large frogs (*Rana* sp); tiger beetles (*Megacephala carolina*); and cured tobacco leaves. Abiotic groups sampled were pond sediment, pond water, and tobacco field soil. Not all biotic groups were available from all sites, which accounts for occasional omissions of results from the tables.

SAMPLE COLLECTION

Turtles and fish were captured with a common fish trap constructed of chicken wire. Tiger beetles were captured in pitfall traps on the soil surface within each tobacco field. Traps were serviced daily and trappings for a given species were conducted simultaneously at all 20 sites. Bullfrogs were collected by gigging at night from a boat.

Water samples were collected from a boat by attaching a smallmouth, one-gallon bottle to a pole and slowly lowering it to the bottom of the pond. Ten samples were collected from dispersed positions within each pond, mixed thoroughly, and a one-gallon composite sample was taken for analysis.

Ten random sediment samples collected from each pond with an Eckman dredge were composited and a representative half-gallon sample was taken for analysis. A core sampling device was used to collect 50 soil cores, 2 in. by 5 in., from each tobacco field. Cores from each field were mixed thoroughly and a representative half-gallon sample was taken for analysis.

In a separate aspect of the study, cured tobacco leaves were randomly collected from field research plots. These samples were analyzed primarily for residue determinations following pesticide treatments with certain known chemicals.

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All biotic samples except tiger beetles were weighed in the field and placed in portable freezers containing dry ice. Tiger beetles were immobilized by cooling and returned to the laboratory where accurate weights were obtained on a Mettler balance. All samples except water were stored at 25°F until packed in styrofoam biomailers containing dry ice for shipment to the USDA Methods Development Environmental Quality Laboratory in Brownsville, Tex., for residue analysis. To avoid spoilage, samples were shipped airmail special delivery and normally arrived in Brownsville, still frozen, within 24 hours. Water samples were stored in one-gallon glass bottles and shipped to Brownsville approximately four days after collection. All samples except water were frozen until analysis. Water was stored in a walk-in cooler at approximately 40°F until analyzed. All samples were analyzed as soon as possible after receipt.

ANALYTICAL METHODS

Organochlorine and organophosphate pesticides were extracted from soil essentially as described by Stevens et al. (7); a brief description is given below.

Representative 300-g subsamples of wet soil were transferred to half-gallon Mason jars with 600 ml 3:1 mixture of hexane-isopropyl alcohol solvent and 80 ml distilled water, then rotated 4 hours on a concentric rotator. After the soil had settled, extracts were filtered through glass wool into 1-liter separatory funnels, the isopropanol and water were removed by washing three times with equal volumes of distilled water, and the hexane layer was filtered through a layer of glass wool and anhydrous sodium sulfate into 500-ml graduated cylinders, collecting 300-ml aliquots. The hexane extracts were then stored in capped, amber sample bottles at approximately 40°F pending gas chromatographic (GC) analysis.

Sediment was extracted in an identical manner with the following exceptions: excess water was drained from samples before subsamples were weighed and 250 g anhydrous sodium sulfate was added to the jars containing the solvent and samples.

After mixing water samples thoroughly, 500-g subsamples were extracted three times with 100-ml portions of nanograde dichloromethane in 1-liter separatory funnels. Extracts were drained through a glass wool sodium sulfate filter into 500 ml Erlenmeyer flasks with glass beads and 1 ml 0.01 percent Nujol in hexane holding solution. Solvents were evaporated to approximately 5 ml through Snyder columns on a 60 C water bath, then 100 ml hexane was decanted through the Snyder columns slowly and the evaporation step was repeated. The concentrated extracts were transferred to 15 ml centrifuge tubes with nanograde hexane, solvent volumes were adjusted to 12.5 ml and stored in a 40 F refrigerator pending GC analysis.

Tobacco leaves were ground in a Hobart food chopper and 100-g subsamples were extracted in a half-gallon Mason jar with 800 ml acetonitrile by rotating concentrically at 30 rpm for 4 hours. Three-hundred-ml aliquots were collected through glass wool filters and transferred to 500-ml Erlenmeyer flasks. Acetonitrile was evaporated on hotplates at 95°F to approximately 10 ml and hexane was added and evaporated three times as described in the water extraction procedure. Finally, 100 ml hexane was added to the flask and transferred to 500-ml separatory funnels with 10 ml nanograde isopropyl alcohol and 10 ml hexane. The hexane layer was freed from water and alcohol by shaking with 200 ml distilled water and dried by filtering through anhydrous sodium sulfate into 250-ml Erlenmeyer flasks. Hexane was evaporated on a hot water bath; the final volume was adjusted to 100 ml and the extracts were stored in capped amber bottles pending cleanup.

Fish and frogs were processed whole; turtle shells were removed; composite samples were ground in a Hobart food chopper and 25-g subsamples were blended in a 1-liter blendor jar with 150 ml of a 3:1 mixture of nanograde hexane-isopropanol for 2 minutes. Blended samples were transferred to half-gallon Mason jars with 250 ml additional solvent and extracted as described previously. Extracts were filtered through glass wool into separatory funnels and washed three times with distilled water to remove alcohol. The hexane layer was filtered through sodium sulfate; aliquots were volumetrically measured and stored in amber bottles and refrigerated pending cleanup.

Tiger beetles (0.14–6.58 g) were macerated in 100-ml blendor jars with isopropanol for 2 minutes. Sample transfer to half-gallon jars was aided by two 75-ml rinsings of hexane. The extraction, filtering, drying, and storage procedures were identical to the previously described biological extracts.

Methomyl (Lannate) was extracted from soil, sediment, water and tobacco as described in a previous paper by Reeves and Woodham (6). Dichloromethane was used to extract the methomyl from soil, water, and sediment samples; a mixture of 97.5 percent dichloromethane and 2.5 percent benzene was employed for extracting the insecticide from tobacco samples. Extracts were extracted and stored as described previously for biological samples.

Carbaryl (Sevin) and carbofuran (Furadan) were extracted from soil, sediment, water, and tobacco as described by Reeves and Woodham (USDA Environmental Quality Laboratory, Brownsville, Tex., 1972: unpublished data). Briefly, the insecticides were extracted from sediment and tobacco with dichloromethane in half-gallon Mason jars on a concentric rotator for 4 hours, and water samples were extracted in separatory funnels with dichloromethane. Sample extracts were dried; the solvent was evaporated to 50 ml and refrigerated pending cleanup, hydrolysis, and derivation.

SAMPLE CLEANUP

Extracts from biological samples for organochlorine and organophosphate residue analyses were purified by partitioning 5–g aliquots between two immiscible solvents, hexane and acetonitrile, as described by Wiersma et al. (*8*). After concentrating the samples into hexane, a modification of the florisil column cleanup procedures of Mills et al. (*5*), Johnson (*4*), and Wiersma et al. (*9*) was used to further purfiy the extracts. The following changes were made: a mixture of 15 g florisil and 2 g anhydrous sodium sulfate was used in the columns; a third elution with dichloromethane removed more polar pesticides. Glass beads and 1 ml of a 0.01 percent Nujol in hexane solution were added and each fraction was concentrated through Snyder columns to approximately 5 ml on a hot water bath. The concentrated extracts were quantitatively transferred to stoppered 15–ml centrifuge tubes with hexane and the final solvent volume was adjusted to 12.5 ml. Care was taken to ensure that all diethyl ether or dichloromethane had been removed; then samples were refrigerated pending GC analysis.

Fraction 1 from the florisil columns contained pesticides of low polarity, such as aldrin, heptachlor, DDT, DDE, *o,p'*– and *p,p'*–TDE, toxaphene, strobane, chlordane, mirex, γ–BHC, and PCB's. Fraction 2 contained moderately polar pesticides such as dieldrin, endrin, heptachlor epoxide, methyl and ethyl parathion, endosulfan, and ethion. Fraction 3 contained the highly polar pesticides: malathion, methyl trithion, and endosulfan isomers.

Soil, sediment, water, and tobacco extracts were purified for methomyl analysis as described by Reeves and Woodham (*6*). Briefly, the method involved concentrating solvents to 10 ml, transferring the extracts into florisil chromatographic columns, and eluting from the columns with 90 percent dichloromethane : 10 percent acetone (v/v) for soil, sediment, and water. Water-saturated chloroform eluted methomyl from the florisil columns for the tobacco extracts. Following column cleanup, the eluates were evaporated and transferred to 15–ml centrifuge tubes for refrigeration pending GC analysis.

Soil, sediment, water, and tobacco extracts for carbaryl and carbofuran analysis were purified using a florisil chromatographic column cleanup procedure described by Reeves and Woodham, Brownsville, Tex., as cited earlier. The insecticides were eluted from columns with dichloromethane for soil, water, and sediment; for tobacco, 20 percent diethyl ether in dichloromethane was used. Eluates were evaporated, transferred to centrifuge tubes, and refrigerated pending hydrolysis, derivation, and coagulation.

The purified extracts were hydrolyzed with aqueous sodium hydroxide, then chloracetylated as described by Argauer et al. (*1*) to form derivatives detectable by electron-capture GC. The derivatized soil, sediment, and water samples were stored in benzene in centrifuge tubes with anhydrous sodium sulfate and analyzed by GC as soon as possible. The derivatized tobacco extracts were further purified by coagulation as described by Johnson (*3*). The derivatives were extracted from the aqueous phase with dichloromethane; the solvent was filtered through anhydrous sodium sulfate and evaporated to approximately 5 ml on a hot water bath. Extracts were transferred to centrifuge tubes with benzene and refrigerated pending GC analysis.

GAS CHROMATOGRAPHIC ANALYSIS

Organochlorine pesticides were analyzed by GC using a Tracor MT–200 gas chromatograph with the following instrument parameters and operating conditions:

Detectors	dual Ni–63 electron-capture
Columns	6–ft glass, packed with
(1)	3 percent DC–200 mesh Gas-Chrom Q
(2)	3 percent OV–1 on 80–100 mesh Chromosorb W
(3)	5 percent QF–1 on 100–120 mesh Gas-Chrom Q
(4)	mixture of 1.95 percent QF-1.1 and 5 percent OV–17 on 80–100 mesh Gas-Chrom Q
Temperatures	column 200°C
	injector 225°C
	detector 300°C
Carrier gas	nitrogen flowing at 80 ml/min; for mixed column, 120 ml/min

Recorder (1 mv) chart speed 30 in /hr

Sensitivity was adjusted to produce approximately half-scale deflection of the recorder pen with 0.50–ng injection of aldrin. Calculations as ppm were based on heights of the pesticide peaks compared with a pesticide calibration standard of known concentration and purity.

Organophosphate pesticides were also analyzed with a Tracor MT–200 gas chromatograph equipped with a dual flame photometric detector (FPD) with the phosphorus (526 nm) and sulfur (394 nm) interference filters installed for simultaneous recordings of organophosphate and sulfur peaks from thiophosphate pesticides. Instrument parameters and operating conditions follow.

Column	6-ft-by-1.4-in -glass, packed with 3 percent DC–200 on 100–120 mesh Gas-Chrom Q
Temperatures	column 200°C
	injector 225°C
	detector 200°C
Carrier gases	nitrogen flowing at 120 ml/min
	air flowing at 80 ml/min
	hydrogen flowing at 200 ml/min
	oxygen flowing at 20 ml/min
Recorder chart speed	30 in /hr

Sensitivity was adjusted to obtain approximately half-scale deflection with an injection of 1.5–ng ethyl parathion. Calculations were based on peak height as described for organochlorine pesticides.

Methomyl was analyzed as described in the organophosphate analysis section, except the column temperature was 140°C and a 10 percent DC–200 on 100–120 mesh Gas-Chrom Q column was used. Sensitivity of the detector was adjusted to obtain half-scale deflection of the recorder pen with a 20–ng injection of methomyl.

Carbaryl and carbofuran were analyzed by GC as described for the chlorinated hydrocarbons, except that the column

was 6-ft-by 1 4-in glass packed with 5 percent OV–210 on 100 120 mesh Chromosorb-W. Sensitivity was adjusted to obtain approximately half-scale deflection with a 4–ng injection of the carbofuran or carbaryl derivatives.

A series of controls consisting of a solvent check, nonfortified sample, and a sample fortified with known concentrations of pesticides were included with each group of samples. The controls were extracted, stored, and analyzed simultaneously with samples containing unknown concentrations of pesticides in order to monitor possible contamination and analytical and extraction efficiency. The following average range of recoveries was obtained from the various sample types: soil, 58.1 percent for aldrin to 117.0 percent for *p,p'*–DDT; sediment, 71.2 percent for dieldrin to 103.6 percent for *p,p'*–DDE; water, 58.9 percent for carbaryl to 98.4 percent for *p,p'*–DDE; tobacco, 47.6 percent for methyl trithion to 104.9 percent for ethion; frogs, 51.1 percent for malathion to 100.8 percent for *o,p'*–DDT; turtles, 55.8 percent for lindane to 89.7 percent for ethion. All residues were corrected for appropriate recovery values.

Moisture content of all soil and sediment samples was determined by drying a weighed 100–g sample in a 120°C oven for 24 hours, then reweighing the sample to determine moisture loss. Residues were corrected and reported on a dry-weight basis.

All pesticide residues were confirmed by partitioning coefficients (p–values), thin-layer chromatography, chemical derivatization, and multiple-column GC.

Results and Discussion

Table 1 presents pesticide treatment histories for the sampling sites in this project. Pesticides applied were either organophosphates or carbamates except the endosulfan treatments at sites F–3137 and G–1135.

Table 2 presents residue data for soil and sediment in areas inside and outside the program area. Residues ranging from 0.01 to 0.35 ppm *o,p'* TDE and from 0.02 to 1.15 ppm *p,p'* TDE were detected in sediment inside the program area. In sediment from ponds outside these areas, residues ranged from 0.01 to 0.30 ppm *o,p'* –TDE and from 0.02 to 0.87 ppm *p,p'* TDE. No residues of organophosphates, carbaryl, methomyl (Lannate), or carbofuran (Furadan) exceeding the lower limits of sensitivity of 0.01 ppm or 0.05 ppm were detected in any sediment samples.

Generally, soil samples collected within the program area had slightly higher residues than had those outside the program area. Residues ranged from not detectable to 2.00 ppm carbofuran (Furadan). Soil from outside the program area revealed carbofuran residues up to 0.14 ppm, apparently from pesticide treatments in previous years.

TABLE 1. *Record of foliar insecticide treatments of tobacco in sampling sites in pest management areas of North Carolina, 1971* [1]

Sampling Site	Insecticides Applied
A–164	Guthion
A–167	Methomyl (Lannate)
C–541	Carbaryl (Sevin), endosulfan, malathion
C–559	No treatment
C–622	Carbaryl (Sevin)
C–1513	Guthion
D–822	Diazinon
E–3229	Carbaryl (Sevin)
E–3231	Guthion
F–1237	–
F–1281	No treatment
F–3137	Endosulfan, malathion
G–1135	Endosulfan, malathion
G–1348	Azodrin
G–1425	Methomyl (Lannate)
G–1508	Azodrin
G–1509 (site 1)	Carbaryl (Sevin)
G–1509 (site 2)	Azodrin
H–238	Parathion, carbaryl (Sevin)
H–256	Methomyl (Lannate)

[1] Ganyard et al., 1972 (2)

Of the biological samples, tiger beetles showed perhaps the greatest variety of pesticides. Inside and outside the program area, residues of dieldrin, endrin, and all six DDT isomers were detected. No significant differences occurred between samples from inside and those from outside the program area.

Most residues in biological samples from program areas were equal to or less than residues in samples collected outside these areas. Naturally there were some exceptions, such as *p,p'*–DDT in frogs (0.01 ppm outside and 0.02 ppm inside) and *p,p'*–DDE in tiger beetles (4.44 ppm inside and 3.07 ppm outside).

Table 3 shows residue patterns in tobacco samples from this pest management project area. Residues up to 0.31 ppm *o,p'*–TDE, 0.98 ppm *p,p'*–TDE, 0.28 ppm *o,p'*–DDT, and 1.91 ppm *p,p'*–DDT were detected. Carbofuran residues were found in samples from the following areas: Aycock No. 4, 0.96 ppm; Saul's No. 4, 0.47 ppm; R. Marsh No. 4, 0.08 ppm; and W. Harbor No. 4, 0.48 ppm. Carbofuran residues are not shown in the tables.

Residue data for turtles, frogs, fish, and insects within and outside the program area are presented in Table 4. Although measurable amounts of the chlorinated hydrocarbon pesticides were found, the quantities were not unusually high. The highest concentration was 3.81 ppm *p,p'*–DDT in one turtle sample. No organophosphate residues exceeding the lower limits of 0.01 ppm were detected in any biological samples.

TABLE 2 *Organochlorine pesticide residues in soil and sediment from pest management areas in North Carolina, 1971*

Sampling Site	Sampling Series	Residue, ppm dry weight [1,2]					
		o,p'-DDE	*p,p'*-DDE	*o,p'*-TDE	*p,p'* TDE	*o,p'*-DDT	*p,p'* DDT
Inside Program Area							
SEDIMENT							
A-164	1	<0 01	<0 01	0 02	0 07	< 0 01	< 0 01
	2	<0 01	<0 01	0 03	0 16	<0 01	< 0 01
A-167	1	<0 01	<0 01	0 35	1 15	<0 01	<0 01
	2	<0 01	<0 01	0 31	1 03	< 0 01	< 0 01
C-1135	1	<0 01	<0 01	0 05	0 09	< 0 01	< 0 01
	2	<0 01	<0 01	0 05	0 10	<0 01	< 0 01
G-1348	1	<0 01	<0 01	0 05	0 20	<0 01	< 0 01
	2	<0 01	<0 01	0 03	0 07	<0 01	< 0 01
G-1425	1	<0 01	<0 01	0 08	0 15	< 0 01	< 0 01
	2	<0 01	<0 01	0 01	0 02	<0 01	< 0 01
G-1508	1	<0 01	<0 01	0 01	0 04	<0 01	< 0 01
	2	<0 01	<0 01	0 01	0 02	<0 01	< 0 01
G-1509	1	<0 01	<0 01	0 10	0 21	<0 01	<0 01
(site 1)	2	< 0 01	<0 01	0 09	0 16	<0 01	<0 01
G-1509	1	<0 01	<0 01	0 08	0 24	<0 01	< 0 01
(site 2)	2	<0 01	< 0 01	0 15	0 52	<0 01	< 0 01
H-238	1	<0 01	<0 01	0 02	0 07	<0 01	<0 01
	2	<0 01	<0 01	0 04	0 07	<0 01	<0 01
H-256	1	<0 01	< 0 01	0 04	0 15	<0 01	<0 01
	2	<0 01	<0 01	0 04	0 12	<0 01	<0 01
Outside Program Area							
C-541	1	<0 01	<0 01	0 05	0 15	<0 01	<0 01
	2	<0 01	<0 01	0 04	0 11	<0 01	< 0 01
C-559	1	<0 01	<0 01	0 02	0 05	<0 01	<0 01
	2	<0 01	<0 01	0 03	0 08	<0 01	<0 01
C-622	1	<0 01	<0 01	0 02	0 05	<0 01	<0 01
	2	<0 01	<0 01	0 02	0 07	<0 01	< 0 01
C-1513	1	<0 01	<0 01	0 02	0 07	<0 01	<0 01
	2	<0 01	< 0 01	0 01	0 04	<0 01	<0 01
D-822	1	<0 01	<0 01	0 02	0 02	<0 01	< 0 01
	2	<0 01	<0 01	0 01	0 05	<0 01	<0 01
E-3229	1	<0 01	<0 01	0 03	0 08	<0 01	<0 01
	2	<0 01	<0 01	0 03	0 07	<0 01	<0 01
E-3231	1	<0 01	<0 01	0 01	0 03	<0 01	< 0 01
	2	<0 01	< 0 01	0 03	0 07	<0 01	<0 01
F-1237	1	<0 01	<0 01	0 30	0 87	<0 01	< 0 01
	2	<0 01	<0 01	0 19	0 48	<0 01	<0 01
F-1281	1	<0 01	< 0 01	0 09	0 16	<0 01	< 0 01
	2	<0 01	<0 01	0 05	0 10	<0 01	<0 01
F-3137	1	<0 01	<0 01	0 06	0 13	<0 01	<0 01
	2	<0 01	< 0 01	0 05	0 14	<0 01	<0 01
Inside Program Area							
SOIL							
A-164	1	<0 01	<0 01	0 03	0 04	0 04	0 07
A-167	1	<0 01	<0 01	0 05	0 19	0 17	0 50
G-1135	1	<0 01	<0 01	0 03	0 11	0 04	0 12
G-1348	1	<0 01	<0 01	0 05	0 14	0 06	0 54
G-1425	1	<0 01	<0 01	0 04	0 08	0 03	0 07
G-1508	1	<0 01	<0 01	0 01	0 04	0 02	0 06
G-1509 (site 1)	1	<0 01	<0 01	0 02	0 06	0 03	0 09
G-1509 (site 2)	1	<0 01	<0 01	0 03	0 09	0 05	0 14
H-238	1	<0 01	<0 01	0 03	0 08	0 07	0 17
H-256	1	<0 01	<0 01	0 02	0 04	0 03	0 08
Outside Program Area							
C-5541	1	<0 01	<0 01	0 03	0 08	0 02	0 04
C-559	1	<0 01	<0 01	0 04	0 06	0 04	0 09
C-622	1	<0 01	<0 01	0 04	0 06	0 02	0 05
C-1513	1	<0 01	<0 01	0 07	0 14	0 04	0 10
D-822	1	<0 01	<0 01	0 02	0 04	0 03	0 05
E-3229	1	<0 01	<0 01	0 04	0 07	0 01	0 04
F-1237	1	<0 01	<0 01	0 06	0 13	0 02	0 04
F-1281	1	<0 01	<0 01	0 02	0 07	0 03	0 06
F-3137	1	<0 01	<0 01	0 02	0 04	0 10	0 14

[1] Corrected for pesticide recovery from fortified samples
[2] Lower limits of sensitivity = 0 01 ppm

TABLE 3 *Organochlorine pesticide residues in tobacco from pest management areas in North Carolina, 1971* [1]

Sampling Site	RESIDUE, PPM DRY WEIGHT [2,3]					
	o,p' DDE	*p,p'* DDE	*o,p'* TDE	*p,p'* TDE	*o,p'* DDT	*p,p'* DDT
Saul, No. 1	< 0.01	< 0.01	0.09	0.04	0.03	0.22
Saul, No. 2	< 0.01	< 0.01	0.03	0.06	0.04	0.39
Saul, No. 3	< 0.01	< 0.01	0.02	0.05	0.04	0.27
Ward, No. 1	< 0.01	< 0.01	0.31	0.69	0.28	1.91
Ward, No. 2	< 0.01	< 0.01	0.23	0.58	0.24	1.90
Ward, No. 3	< 0.01	< 0.01	0.15	0.54	0.17	1.83
Wilson, No. 1	< 0.01	< 0.01	0.02	0.05	0.04	0.20
Wilson, No. 2	< 0.01	< 0.01	0.02	0.04	0.04	0.19
Wilson, No. 3	< 0.01	< 0.01	0.03	0.05	0.05	0.23
Aycock, No. 1	< 0.01	< 0.01	0.04	0.12	0.04	0.25
Aycock, No. 2	< 0.01	< 0.01	0.02	0.05	0.03	0.15
Aycock, No. 3	< 0.01	< 0.01	0.03	0.98	0.03	0.21
Aycock, No. 4	< 0.01	< 0.01	0.02	0.06	0.04	0.18
W. Saul's	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.22
W. Harbor, No. 1	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.20
W. Harbor, No. 3	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.23
W. Harbor, No. 4	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.20
R. Marsh, No. 1	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.07
R. Marsh, No. 2	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.09
R. Marsh, No. 3	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.09
R. Marsh, No. 4	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.09

[1] Samples collected at harvest in autumn 1971.
[2] Corrected for pesticide recovery from fortified samples.
[3] Lower limits of sensitivity = 0.01 ppm.

TABLE 4 *Organochlorine pesticide residues in biological samples from pest management areas in North Carolina, 1971* [1]

Sampling Site	Species	RESIDUE, PPM [2,3]							
		Dieldrin	Endrin	*o,p'* DDE	*p,p'* DDE	*o,p'* TDE	*p,p'* TDE	*o,p'* DDT	*p,p'* DDT
		Inside Program Area							
TURTLES									
A-164	Snapping	< 0.01	< 0.01	< 0.01	0.29	< 0.01	< 0.01	0.01	< 0.01
A-167	Yellow Bellied	< 0.01	< 0.01	< 0.01	L[4]	< 0.01	0.02	0.01	0.04
G-1135	Musk	< 0.01	< 0.01	< 0.01	0.11	< 0.01	< 0.01	0.01	< 0.01
G-1135	Yellow-Bellied	< 0.01	< 0.01	< 0.01	0.04	< 0.01	< 0.01	0.01	< 0.01
G-1348	Yellow Bellied	< 0.01	< 0.01	< 0.01	0.14	< 0.01	0.01	0.01	0.02
G-1425	Painted	< 0.01	< 0.01	< 0.01	0.05	< 0.01	< 0.01	0.01	< 0.01
G-1425	Yellow Bellied	< 0.01	< 0.01	< 0.01	0.03	< 0.01	< 0.01	0.01	< 0.01
G-1425	Snapping	< 0.01	< 0.01	< 0.01	0.08	< 0.01	< 0.01	0.01	< 0.01
G-1425	Musk	< 0.01	< 0.01	< 0.01	0.33	< 0.01	< 0.01	0.01	< 0.01
G-1508	Yellow Bellied	< 0.01	< 0.01	< 0.01	0.08	< 0.01	< 0.01	0.01	0.02
G-1509	Musk	< 0.01	< 0.01	< 0.01	0.21	< 0.01	< 0.01	0.01	< 0.01
G-1509	Yellow Bellied	< 0.01	< 0.01	< 0.01	0.12	< 0.01	< 0.01	0.01	< 0.01
H-238	Painted	< 0.01	< 0.01	< 0.01	0.09	< 0.01	< 0.01	0.01	< 0.01
H-238	Yellow Bellied	< 0.01	< 0.01	< 0.01	0.24	< 0.01	0.01	0.01	0.04
H-256	Musk	0.01	< 0.01	< 0.01	0.78	< 0.01	< 0.01	0.01	< 0.01
H-256	Painted	< 0.01	< 0.01	< 0.01	1.14	< 0.01	0.02	0.01	0.04
H-256	Yellow Bellie	< 0.01	< 0.01	< 0.01	0.20	< 0.01	0.02	0.01	0.03
		Outside Program Area							
C-54	Musk	< 0.01	< 0.01	< 0.01	0.40	< 0.01	< 0.01	< 0.01	< 0.01
C-541	Painted	0.01	0.01	< 0.01	0.34	< 0.01	0.01	< 0.01	0.02
C-559	Snapping	< 0.01	< 0.01	< 0.01	0.27	< 0.01	< 0.01	< 0.01	< 0.01
C-622	Snapping	< 0.01	< 0.01	< 0.01	0.11	< 0.01	< 0.01	< 0.01	< 0.01
C-1513	Musk	0.01	< 0.01	< 0.01	0.33	< 0.01	< 0.01	< 0.01	< 0.01
C-1513	Painted	< 0.01	< 0.01	< 0.01	0.40	< 0.01	0.01	< 0.01	0.01
D-822	Snapping	0.01	< 0.01	< 0.01	0.18	< 0.01	< 0.01	< 0.01	< 0.01
D-822	Musk	< 0.01	< 0.01	< 0.01	0.26	< 0.01	< 0.01	< 0.01	< 0.01
D-822	Painted	< 0.01	< 0.01	0.01	0.13	< 0.01	0.01	< 0.01	< 0.01
D-822	Yellow Bellied	< 0.01	0.01	0.01	0.03	< 0.01	< 0.01	< 0.01	< 0.01
E-3229	Painted	< 0.01	0.01	0.01	0.26	< 0.01	0.01	< 0.01	0.01
E-3229	Snapping	< 0.01	< 0.01	0.01	0.04	< 0.01	< 0.01	< 0.01	< 0.01
E-3229	Yellow Bellied	< 0.01	< 0.01	0.01	0.11	< 0.01	0.01	< 0.01	0.01
E-3231	Musk	< 0.01	< 0.01	0.01	0.38	< 0.01	< 0.01	< 0.01	< 0.01
E-3231	Painted	< 0.01	0.01	0.01	0.75	< 0.01	0.01	< 0.01	0.06
E-[illegible]3	Snapping	< 0.01	< 0.01	0.01	0.15	< 0.01	< 0.01	< 0.01	< 0.01
[illegible]23	Yellow Bellied	< 0.01	0.01	0.01	0.70	< 0.01	0.01	< 0.01	0.01
E-[illegible]37	Musk	0.01	0.01	0.01	3.81	< 0.01	< 0.01	< 0.01	< 0.01
E-[illegible]37	Painted	0.01	< 0.01	0.01	1.99	< 0.01	0.08	< 0.01	< 0.01
E-1237	Yellow Bellied	< 0.01	< 0.01	0.01	1.99	< 0.01	0.08	< 0.01	0.07

(Continued next page)

TABLE 4 (cont'd.). *Organochlorine pesticide residues in biological samples from pest management areas in North Carolina, 1971*[1]

Sampling Site	Species	Residue, ppm							
		Dieldrin	Endrin	*o,p'*-DDE	*p,p'*-DDE	*o,p'*-TDE	*p,p'*-TDE	*o,p'*-DDT	*p,p'*-DDT
Inside Program Area									
F-1281	Musk	<0.01	<0.01	<0.01	0.17	<0.01	<0.01	<0.01	<0.01
F-1281	Painted	<0.01	<0.01	<0.01	0.18	<0.01	<0.01	<0.01	<0.01
F-1281	Snapping	<0.01	<0.01	<0.01	0.20	<0.01	<0.01	<0.01	<0.01
F-3137	Painted	<0.01	<0.01	<0.01	1.08	<0.01	0.01	<0.01	0.05
Inside Program Area									
FROGS									
A-164		<0.01	<0.01	<0.01	0.04	<0.01	<0.01	<0.01	<0.01
A-167		<0.01	<0.01	<0.01	0.04	<0.01	<0.01	<0.01	<0.01
C-622		<0.01	<0.01	<0.01	0.03	<0.01	<0.01	<0.01	<0.01
G-1135		<0.01	<0.01	<0.01	0.01	<0.01	<0.01	<0.01	<0.01
G-1508		<0.01	<0.01	<0.01	0.10	<0.01	<0.01	<0.01	<0.01
G-1509		<0.01	<0.01	<0.01	0.01	<0.01	<0.01	<0.01	<0.01
H-256		0.05	<0.01	0.01	0.14	<0.01	0.13	<0.01	0.01
Outside Program Area									
C-1513		<0.01	<0.01	<0.01	0.02	<0.01	<0.01	<0.01	<0.01
D-822		<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
E-3229		0.01	<0.01	<0.01	0.06	<0.01	<0.01	<0.01	<0.01
E-3231		<0.01	<0.01	<0.01	0.01	<0.01	<0.01	<0.01	<0.01
F-1237		0.11	<0.01	0.02	0.15	<0.01	0.33	<0.01	0.02
F-1281		<0.01	<0.01	<0.01	0.01	<0.01	<0.01	<0.01	<0.01
Inside Program Area									
FISH									
A-164	Bluegills	0.02	<0.01	0.04	0.19	<0.01	0.15	<0.01	0.03
A-167	Bluegills	0.01	<0.01	0.15	0.87	0.02	0.62	<0.01	0.09
G-1135	Bluegills	0.02	<0.01	0.05	0.11	0.01	0.14	<0.01	0.02
G-1348	Bluegills	<0.01	<0.01	0.03	0.11	<0.01	0.06	<0.01	0.02
G-1425	Bluegills	0.08	<0.01	0.03	0.07	<0.01	0.06	<0.01	0.02
G-1508	Bluegills	0.02	<0.01	0.02	0.16	<0.01	0.05	<0.01	0.04
G-1509	Bluegills	0.01	<0.01	0.03	0.14	<0.01	0.09	<0.01	0.02
H-238	Bluegills	0.01	<0.01	0.03	0.16	<0.01	0.07	<0.01	0.04
H-256	Bluegills	0.16	<0.01	0.09	0.12	0.01	0.15	0.01	0.04
Outside Program Area									
C-541	Bluegills	<0.01	<0.01	0.03	0.10	<0.01	0.05	0.01	0.01
C-559	Bluegills	<0.01	<0.01	0.01	0.09	<0.01	0.02	<0.01	<0.01
C-622	Bluegills	<0.01	<0.01	0.01	0.06	<0.01	0.02	<0.01	0.01
G-1513	Bluegills	<0.01	<0.01	0.06	0.15	<0.01	0.06	0.09	0.03
D-822	Bluegills	<0.01	<0.01	0.02	0.09	<0.01	0.06	<0.01	0.02
E-3229	Bluegills	0.01	<0.01	0.03	0.15	<0.01	0.07	<0.01	0.01
E-3231	Bluegills	<0.01	<0.01	<0.01	0.01	<0.01	<0.01	<0.01	<0.01
F-1237	Bluegills	0.11	<0.01	0.02	0.15	<0.01	0.33	<0.01	0.02
F-1281	Bluegills	0.06	<0.01	0.12	0.80	<0.01	0.32	0.02	0.07
F-3137	Bluegills	0.16	<0.01	0.05	0.12	0.01	0.19	0.01	0.04
G-1435	Bluegills	0.05	<0.01	0.04	0.08	<0.01	0.06	<0.01	0.01
Inside Program Area									
TIGER BEETLES									
A-164		<0.01	0.01	0.11	1.80	<0.01	0.01	0.04	<0.01
A-167		0.01	0.01	0.04	1.02	<0.01	0.02	0.05	0.02
F-1281		0.05	0.05	0.04	1.26	<0.01	0.01	0.06	0.01
F-3137		<0.01	0.02	0.06	2.02	0.01	0.02	0.06	0.03
F-3231		<0.01	<0.01	<0.14	2.44	<0.01	<0.01	0.12	0.01
G-1135		<0.01	<0.01	<0.01	1.58	<0.01	<0.01	0.01	<0.01
G-1348		<0.01	<0.01	<0.01	4.44	<0.01	<0.01	0.01	<0.01
G-1425		<0.01	<0.01	<0.01	0.91	<0.01	<0.01	0.01	<0.01
G-1508		<0.01	<0.01	<0.01	1.38	<0.01	<0.01	0.01	<0.01
G-1509	(site 1)	<0.01	<0.01	<0.01	0.01	<0.01	<0.01	0.01	<0.01
G-1509	(site 2)	<0.01	<0.01	<0.01	2.62	<0.01	<0.01	0.01	<0.01
H-238		<0.01	<0.01	<0.01	2.10	<0.01	0.01	0.01	0.03
H-256		0.04	<0.01	0.01	0.06	0.01	0.01	0.01	0.03

(Continued next page)

TABLE 4 (cont'd.) *Organochlorine pesticide residues in biological samples from pest management areas in North Carolina, 1971* [1]

Sampling Site	Species	Residue, ppm [2,3]							
		Dieldrin	Endrin	*o,p'*-DDE	*p,p'*-DDE	*o,p'*-TDE	*p,p'*-TDE	*o,p'*-DDT	*p,p'*-DDT
Inside Program Area									
Outside Program Area									
C 841		<0.01	<0.01	<0.01	0.46	<0.01	<0.01	<0.01	<0.01
C 889		<0.01	<0.01	<0.01	3.07	<0.01	<0.01	<0.01	<0.01
C 622		<0.01	<0.01	<0.01	1.88	<0.01	<0.01	<0.01	<0.01
G 1813		<0.01	<0.01	<0.01	1.86	<0.01	<0.01	<0.01	<0.01
I 3229		<0.01	<0.01	<0.01	1.03	<0.01	<0.01	<0.01	<0.01
I 3233		<0.01	<0.01	0.14	2.44	<0.01	<0.01	0.12	0.01
I 3231		<0.01	<0.01	<0.01	1.37	<0.01	<0.01	<0.01	<0.01
F 1237		<0.01	<0.01	<0.01	2.39	<0.01	<0.01	<0.01	<0.01
F 1281		0.05	0.05	0.04	1.26	<0.01	0.01	0.06	0.01
I 3137		0.01	0.02	0.06	2.02	<0.01	0.02	0.06	0.03

[1] Samples collected at beginning of crop season.
[2] Corrected for pesticide recovery from fortified samples.
[3] Lower limits of sensitivity = 0.01 ppm.
[4] L - Lost samples.

Conclusions

This one-year study did not produce evidence that pesticide use in Wayne and Wilson Counties is causing significant residue accumulation in the environment. Chemicals used for tobacco pest control are very similar throughout North Carolina, varying only in frequency of use. To project statewide trends and draw conclusions concerning pest management in tobacco and other crops, additional residue information must be accumulated. Plans are in progress to continue these studies for several years, establishing a more reliable baseline for projecting trends.

Acknowledgments

Authors appreciate the expert assistance of persons who helped to collect the samples and of H. Richardson who helped to process the samples. We also thank Dr. Stephen Warnick and staff, Intermountain Laboratories, Inc., Salt Lake City, Utah, for analyzing a portion of the biological samples, and Dr. Don W. Hayne, Professor of Statistics and Zoology, North Carolina State University, for a most beneficial critique.

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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-1,4-5,8-dimethanonaphthalene
AROCLOR	A mixture of chlorinated terphenyls
AZINPHOSMETHYL	*O,O*-dimethyl *S*[4-oxo-1,2,3,-benzotriazin-3(4H)ylmethyl] phosphorodithioate
AZODRIN	3-hydroxy-*n*-methyl-*cis*-crotonamide, dimethyl phosphate
BHC(Benzene Hexachloride)	1,2,3,4,5,6-hexachlorocyclohexane (mixture of isomers) Commercial product contains several isomers of which *gamma* is most active as an insecticide
CARBARYL	1-naphthalenyl methylcarbamate
CARBOFURAN	2,3-dihydro-2,2-dimethyl-7-benzofuranyl methylcarbamate
CHLORDANE	1,2,3,5,6,7,8,8-octachloro-2,3,3a, 4,7,7a-hexahydro-4,7-methanoindene The technical product is a mixture of several compounds, including heptachlor, chlordene, and two isomeric forms of chlordane
DDE	Dichlorodiphenyl dichloro-ethylene (degradation product of DDT) *p,p'*–DDE 1,1-Dichloro-2,2-bis(*p*-chlorophenyl) ethylene *o,p'*–DDE 1,1-Dichloro-2-(*o*-chlorophenyl)-2-(*p*-chlorophenyl) ethylene
DDT	Main component (*p,p'*–DDT) α-Bis(*p*-chlorophenyl) β,β,β-trichloroethane Other isomers are possible and some are present in the commercial product *o,p'*–DDT [1,1,1-Trichloro-2(*o*-chlorophenyl)-2(*p*-chlorophenyl) ethane]
DIELDRIN	Not less than 85% of 1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4-*endo-exo*-5,8 dimethanonaphthalene
ENDOSULFAN	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-*endo-endo*-5,8-dimethanonaphthalene
ETHION	0,0,0',0'-Tetraethyl S,S'-methylene bisphosphorodithioate
ETHYL PARATHION	*O,O*-diethyl-*o-p*-nitrophenyl phosphorothioate
GUTHION	See azinphosmethyl
HEPTACHLOR	1,4,5,6,7,8,8-heptachloro-3a,4,7,7a-tetrahydro-4,7-*endo*-methanoindene
HEPTACHLOR EPOXIDE	1,4,5,6,7,8,8-heptachloro 2,3-epoxy-3a,4,7,7a-tetrahydro-4,7 methanoindane
LINDANE	Gamma isomer of benzene hexachloride (1,2,3,4,5,6-hexachlorocyclohexane) of 99+ % purity
MALATHION	S-[1,2-Bis(ethoxycarbonyl) ethyl] 0,0-dimethyl phosphorodithioate
METHOMYL	methyl *N*-[[(methylamino) carbonyl] oxy] ethanimidothioate
METHYL PARATHION	O,O-Dimethyl O-*p*-nitrophenyl phosphorothioate
METHYL TRITHION	O,O-Dimethyl S-(*p*-chlorophenylthio) methyl phosphorodithioate
MIREX	1,1a,2,2,3,3a,4,5,5,5a,5b,6-dodecachlorooctahydro-1,3,4 metheno-1H-cyclobuta[cd]pentalene
PCB's (Polychlorinated Biphenyls)	Mixtures of chlorinated biphenyl compounds having various percentages of chlorine
STROBANE	Polychlorinates of camphene, pinene, and related terpenes
TDE	2,2-Bis(*p*-chlorophenyl)-1,1-dichloroethane
TOXAPHENE	Chlorinated camphene (67-69% chlorine) Product is a mixture of polychlor bicyclic terpenes with chlorinated camphenes predominating

The *Pesticides Monitoring Journal* welcomes from all sources qualified data and interpretative information on pesticide monitoring. The publication is distributed principally to scientists, technicians, and administrators associated with pesticide monitoring, research, and other programs concerned with pesticides in the environment. Other subscribers work in agriculture, chemical manufacturing, food processing, medicine, public health, and conservation.

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CONTENTS

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PESTICIDES IN PEOPLE

A study of Pesticide Residues in Michigan's General Population, 1968–70 [1]

Arthur W. Bloomer, Stanley I. Nash, Harold A. Price, Robert L. Welch [2]

ABSTRACT

A study undertaken in Michigan from August 1968 to April 1970 showed that the county of residence was the most significant factor for determining pesticide residue levels in humans. Occupation, sex, and location of residence were also determined to be associated with blood residue levels. Residues of ΣDDT and dieldrin were greater in persons 45 years or older. No relationships were detected between blood hemoglobin and blood residue values. In general, as the blood levels for glucose, cholesterol, uric acid, and creatinine increased, so did the levels of pesticide residues. However, when all variables were used, no equation could be developed which would reliably predict a blood residue level given these demographic characteristics.

Introduction

General awareness of pesticide residues in living organisms, including humans, started with the discovery of residues of DDT or its metabolites in humans and other animals in the mid- to late 1940's (*7, 9, 11*). In 1968 the authors helped to initiate a study of segments of the general population in Michigan to assess the relationships of chlorinated hydrocarbon pesticide residues and such variables as blood biochemistries and demographic characteristics.

The Community Study on Pesticides (*8*) was initiated to determine effects on humans of longterm exposure to pesticides. To form as clear a picture as possible concerning the effects of pesticide exposure on humans, both individuals from the general population and individuals who had experienced definite pesticide exposures were selected for evaluation. The present study was undertaken in an effort to provide a clearer picture of pesticide exposure effects in human population.

Sampling Procedures

When the study was initiated, the Michigan Department of Public Health in cooperation with local health departments was conducting an Adult Health Screening Program. Mobile laboratory clinics were used for obtaining blood specimens and medical histories from ambulatory persons, aged 18 and older, who presented themselves voluntarily to these units for screening tests.

Analysis of laboratory capabilities and of the screening program volume indicated that a systematic sample of 1 to 20 persons would provide accurate results. An informed consent was obtained from each participant. In addition to furnishing an extra blood specimen for residue analysis, each person disclosed his/her age, sex, race, marital status, address, and occupation. The study extended from August 15, 1968, through April 7, 1970, during which time blood specimens and demographic data were collected from 1,035 persons. The cut-off date for the sample was determined by a change in analytical procedures in the laboratory which prevented comparison of subsequent pesticide residue values. The sample does not statistically represent Michigan's general population; hence inferences about the general population from these results are not necessarily appropriate.

Analytical Procedures

The following blood biochemistries were determined according to the Technicon Auto Analyzer Methodology (*12*):

Biochemical tests	*Normal Range*
Hemoglobin	Male 14–18 g/100 ml
	Female 12–16 g/100 ml
Glucose	80–120 mg%
Uric Acid	6.3 mg%
Creatine	1.3 mg%
Cholesterol	140–270 mg%

[1] Study supported by Food and Drug Administration, U S Department of Health, Education, and Welfare, Community Studies on Pesticides, under contracts PH 86-65-50 and FDA 70-19

[2] Michigan Department of Public Health, Lansing, Mich 48909

Chlorinated hydrocarbon pesticide residues were determined in blood sera by electron-capture gas chromatography. A modification of the Dale, Curley, and Cueto method (*3*) was used. Two ml serum and 6 ml nanograde hexane were mixed in a 13-ml centrifuge tube at high speed on a Vortex mixer to emulsify the sample. The emulsified serum was centrifuged at 2,000 rpm for 20 minutes to produce a distinct separation of both phases. The hexane layer was transferred to a 25-ml Kuderna-Danish concentrator by means of a disposable pipette. Each sample was extracted three times. Emulsions formed easily on the second and third extractions. After a 3-mm glass bead was added and a modified micro-Snyder column was attached, the combined extracts were concentrated to approximately 1 ml on a 90°C water bath.

Final volume was adjusted to 1 ml by evaporation under a gentle stream of dry nitrogen. A 5-μl aliquot was injected into a Micro-Tek MT 200 gas chromatograph equipped with tritium foil detectors at 210°C and U-shaped glass columns (6 ft. by ¼ in. OD) at 200°C. Columns used were 1.5 percent OV-17/1.95 percent QF-1 on Chromosorb W-HP 80/100 mesh and 4 percent SE30/6 percent QF-1 on Gas-Chrom Q 80/100 mesh with nitrogen flows at 65 ml/min and 80 ml/min, respectively. All qualitative retention times were relative to that of aldrin. Quantitation of pesticide residues was based on relative peak heights. Lower limits of sensitivity for chlorinated hydrocarbon insecticides were as follows: 1.0 ppb (ng/g) for *p,p'*-DDE, dieldrin and β-BHC; 2.0 ppb for *o,p'*-DDT, *p,p'*-DDT, and *p,p'*-TDE. All values less than the sensitivity values were recorded and computed as zero.

The internal quality control program consisted of dividing a human sera composite into culture tubes and storing at −5° to −10°F. Each month controls were removed from the freezer and analyzed along with the routine samples. Results of the controls analyzed indicated the laboratory was in control and within acceptable accuracy and precision limits.

Following are statistics for 24 control sera analyzed during 1970.

	PESTICIDE RESIDUE, ppb		
	p,p'-DDE	*p,p'*-DDT	Dieldrin
Mean	50.15	10.56	1.89
S.D.	2.484	0.933	0.284
Range	45.4–53.9	9.1–12.0	1.4–2.3

Reagent blanks were routinely analyzed along with samples and did not indicate significant levels of the pesticides quantitated in the study.

The laboratory also participated in an interlaboratory quality control program. Several times a year the laboratory received blind controls from the Food and Drug Administration, Primate Research Center in Perrine, Fla. (presently Environmental Toxicology Division, Human Effects Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, N.C.).

After the Perrine Laboratory had checked and compiled the results from all participating laboratories, results and a performance critique were issued to each laboratory.

Results were confirmed by microcoulometry. Pesticide residues in individual sera could not be confirmed because levels were below the detectability limit of this instrument. Therefore, pooled specimen extracts greater than 50 in number were collected for confirmation. Residues of *p,p'*-DDT, *p,p'*-DDE, and dieldrin were confirmed in the pools tested.

Demographic Data Collection

Occupations of individuals sampled were placed in one of three categories: possibly pesticide related, not related to pesticides, and unknown. Farmers and foresters were placed in the first category because the use of pesticides is not implicit for these occupations but there is a strong probability that the subjects have some association with them. Housewives, school teachers, machinists, mechanics, clerks, bankers, and other occupations were descriptive enough to place in the second category. Persons who were self-employed, retired, or unemployed were placed in the third category.

Location of residence was determined by mailing address to fit one of three categories: rural, urban, or unknown. If the mailing address was listed as a rural route, the participant was categorized rural. Residents who had not given a complete address were placed in the unknown category.

Even though information on race was obtained, authors did not use it during analysis of data because fewer than 1 percent of the persons sampled were Negro; all others were Caucasian. Race was excluded as a variable for subsequent analyses.

Several different statistical techniques were employed for analyzing data. Most of the intergroup comparisons used a one-way analysis of variance to test the null hypothesis. If an analysis of variance test was significant, then multiple comparison tests were computed to determine the important means. Linear regression analysis was used in an attempt to detect any linear relationships in the data. Multiple linear regression was used with many of the nonresidue variables as the independent variables and with specific residue variables as the dependent variable in an attempt to explain the sample variance and define an overall predictive equation. In several cases a log transformation was performed on the data prior to the statistical tests to stabilize the variance.

Concerning the levels of statistical significance: small differences can be statistically significant when large samples are involved. These differences may or may not be medically significant. In the present study alpha levels were determined in relation to sample size in the individual statistical test. (An alpha level is defined as the probability that a person rejects the null hypothesis when it is really true, i.e., announces a difference which really does not exist.) In no case was an alpha level greater than 0.1 used. Thus when determining the statistical significance levels, authors used levels which were conservative. That is, when claiming a statistical significance for a particular comparison an alpha level was used which would reduce the probability of finding a false difference. The converse is also true: i.e., by using conservative levels, the authors may have missed some true differences.

Results and Discussion

Statistical analyses of demographic variables indicated that the sample in the present study approximated Michigan's general population as depicted in the 1970 census. Two exceptions were that the study sample had a slightly higher percentage of persons in the older group and that the percentage of males in each age group was greater than expected except for the youngest group.

As stated earlier, race was not used as a controlling variable. Furthermore, urban/rural comparisons were not possible because definitions of the present study were different from those of the Bureau of the Census, U.S. Department of Commerce.

When blood biochemistries were evaluated using both age and sex breakdowns and then compared with published values (*1,2,10,12*), the Michigan sample appeared to represent a normal population. The biochemistries evaluated were cholesterol, creatinine, glucose, hemoglobin, and uric acid.

Mean blood serum pesticide residues of males and females in the general population of the United States have been published by Durham, who reported levels significantly higher in males than in females (*6*). Except for dieldrin, residues reported in the present study are also significantly higher in males and similarly follow the levels for DDT and its congeners (Table 1). Davies et al. (*4*) and Watson et al. (*13*) also demonstrated higher levels of DDT in the blood of males.

Of the many variables analyzed, county of residence is the single most important factor in explaining pesticide residue level. Table 1 was prepared by grouping counties which have geographic and agrarian similarities.

Residents of Benzie, Grand Traverse, and Leelanau counties show markedly higher mean residues than do Michigan residents in general. These are intensive orchard-growing counties in the northwestern part of the lower peninsula. During the growing season trees are often sprayed weekly or more frequently. Although authors know of no reliable variable which can be used to predict exposure, it appears that the sample population in the three orchard counties had had a greater than average exposure.

Genesee, Livingston, and Ogemaw counties have a greater proportion of male and rural residents in their samples, which may explain the greater residues found. Agriculture in these counties is a general type, not requiring heavy use of pesticides.

The Newaygo County sample also had a greater proportion of male and rural residents. Additionally, this county has some large muck soil areas which are intensively cultivated as truck farms. Large quantities of pesticides are applied regularly during the growing season by conventional ground spray rigs and by aircraft. These facts would lead one to expect residues in excess of the ones found.

Dickinson County in Michigan's upper peninsula is oriented toward dairy cattle, and pesticides are used con-

TABLE 1. *Pesticide residues in blood by county of residence, Michigan*—1968-70

County	Sample Size	Average Blood Residue Levels, ppb			
		p,p'-DDE	*p,p'*-DDT	Σ DDT	Dieldrin
Alger	39	6.75	2.36	9.93	0.13
Chippewa	1	—	—	—	—
Delta	4	—	—	—	—
Dickinson	13	22.26	5.44	30.24	0.00
Gogebic	98	14.02	3.02	18.65	0.27
Houghton	1	—	—	—	—
Iron	30	21.29	4.45	28.16	0.15
Luce	31	9.19	3.15	14.54	0.23
Mackinac	24	7.52	3.31	12.10	0.25
Ontonagon	92	14.73	3.88	20.20	0.32
Schoolcraft	6	—	—	—	—
Benzie	63	34.83	7.66	46.70	0.34
Grand Traverse	45	27.80	11.20	43.17	0.32
Leelanau	67	34.23	8.76	46.89	0.33
Antrim	16	22.22	5.52	30.28	0.00
Arenac	6	—	—	—	—
Charlevoix	1	—	—	—	—
Clare	1	—	—	—	—
Genesee	14	30.68	7.77	41.96	0.09
Gladwin	102	11.78	5.71	19.60	0.23
Hillsdale	24	19.54	5.91	27.68	0.42
Ingham	2	—	—	—	—
Iosco	17	22.44	5.06	30.06	0.17
Isabella	9	—	—	—	—
Livingston	140	22.97	6.09	31.67	0.12
Manistee	9	—	—	—	—
Midland	21	18.73	5.57	26.43	0.38
Monroe	1	—	—	—	—
Muskegon	1	—	—	—	—
Newaygo	76	24.06	7.34	34.15	0.24
Oakland	3	—	—	—	—
Ogemaw	35	30.86	6.44	40.83	0.00
Osceola	1	—	—	—	—
Roscommon	13	20.25	5.28	27.85	0.30
Saginaw	1	—	—	—	—
Shiawassee	1	—	—	—	—
Washtenaw	3	—	—	—	—
Wayne	2	—	—	—	—
Overall Sample Average	960	20.56	5.76	28.86	0.23

Note:— = counties which had fewer than 10 residents sampled. Such counties were not included in the intercounty comparisons, but were included in the overall comparisons.

siderably less than in fruit and vegetable production. Rural residents and males comprised a smaller proportion for this county than for any other sampled. All other upper peninsula counties which were sampled showed lower residues than the average for the State. It is thus difficult to understand why the Dickinson County sample had higher residues.

Residue levels of *p,p'*-DDT, *p,p'*-DDE, and ∑DDT increase with age (Table 2). The increases were, however, somewhat inconsistent in age groups under 45. Dieldrin levels also increased with age but were less consistent than the DDT residues.

For analytical purposes, blood chemistry values were grouped as low, normal, and high as shown in Table 3.

No association was found between hemoglobin levels and blood pesticide residues (Table 4). Residues of dieldrin were found in quite small amounts and were often undetectable in individuals of the sample population, but there appeared to be a positive association between blood glucose levels and dieldrin residue levels. Additionally, the mean dieldrin residues for normal and high quantities of glucose were significantly different from mean residues for the low glucose level.

Considerable association was found between increasing cholesterol levels and increasing residues of *p,p'*-DDE, ∑DDT, and dieldrin. In addition, the uric acid levels increased as the mean for these residues increased, even though the significance limit was not achieved for *p,p'*-DDT residues with cholesterol. The Food and Drug Administration, U.S. Department of Health, Education, and Welfare, began an expanded surveillance and compliance food monitoring program in 1963 which has indicated that the combination of meat, fish, poultry, and dairy products accounts for over half the human intake of chlorinated organic pesticides (*5*). The associations which the authors found between cholesterol and residues, and between uric acid levels and residues, tempt one to postulate that increased animal protein intake might cause increased organochlorine pesticide residues in humans (Table 3).

Creatinine levels and residues of *p,p'*-DDT, *p,p'*-DDE, and ∑DDT showed a positive association which may well be explained using the same hypothesis as above (Table 4).

Persons whose occupations were listed as possibly related to pesticide use had significantly higher levels of *p,p'*-

TABLE 2. *Pesticide residues in blood by sex and age of Michigan residents, 1968–70*

		Average Blood Residue Levels, ppb			
	N	*p,p'*-DDT	*p,p'*-DDE	∑DDT [1]	Dieldrin
Male	516	6.52	23.74	33.36 (507)	0.319
Female	494	4.98	17.40	24.65 (478)	0.129
≤29 Male	28	6.62	23.92	33.48 (27)	0.210
≤29 Female	40	4.47	14.54	21.30 (38)	0.110
30–34 Male	45	4.92	19.34	26.47	0.290
30–34 Female	47	4.20	14.78	20.66	—
35–39 Male	48	8.33	24.24	36.05	0.200
35–39 Female	52	4.44	15.42	21.30 (50)	0.020
40–44 Male	67	5.35	22.28	31.37 (65)	0.230
40–44 Female	58	4.19	16.62	23.06 (57)	0.080
45–49 Male	52	5.95	23.00	31.68	0.450
45–49 Female	63	4.66	17.12	23.95 (62)	0.000
50–54 Male	83	6.29	22.62	31.76 (82)	0.310
50–54 Female	64	5.16	16.88	24.71 (58)	0.160
55–59 Male	58	5.92	25.33	34.13	0.290
55–59 Female	62	4.97	18.97	26.17 (60)	0.180
60–64 Male	52	6.85	25.73	36.13 (51)	0.440
60–64 Female	54	5.78	20.15	28.89 (52)	0.040
65+ Male	83	8.03	26.16	37.68 (79)	0.380
65+ Female	54	6.84	20.96	30.23	0.460
Urban Male	278	6.45	22.06	31.42 (275)	0.330
Rural Male	238	6.59	25.69	35.66 (232)	0.300
Urban Female	246	4.79	16.17	23.15 (205)	0.150
Rural Female	248	5.17	18.63	26.11 (243)	0.110
Urban Total	524	5.67	19.29	27.61 (510)	0.240
Rural Total	486	5.87	22.09	30.77 (475)	0.210

[1] Numbers in parentheses indicate sample size different from that given in "N" column

TABLE 3. *Blood chemistry values of Michigan residents sampled for pesticide residues, 1968–70*

	Hemoglobin		Glucose	Cholesterol	Uric Acid	Creatinine
	Males	Females				
	G,%			Mg,%		
Low	14	12	80	140	3	
Normal	14–18	12–16	80–120	140–270	3–6.3	0.5–1.3
High	> 18	> 16	> 120	> 270	> 6.3	> 1.3

TABLE 4. *Pesticide residues in Michigan residents by sex, location, and selected biochemistries, 1968–70*

		Average Blood Residue Levels, ppb			
	N	*p,p'*-DDT	*p,p'*-DDE	∑DDT	Dieldrin
Unknown Occupation	59	5.55	19.37	27.37	0.166
Possible Pesticide	67	6.96	26.93	37.09	0.336
Not Pesticide Related	884	5.69	20.25	28.43	0.222
p-Value		*0.21*	*0.002*	*NC*	*0.250*
Low Hemoglobin	37	7.10	30.51	41.09	0.392
Normal Hemoglobin	590	6.76	25.30	35.05	0.233
High Hemoglobin	16	7.78	30.66	41.93	0.348
p-Value		*0.79*	*0.07*	*0.13*	*0.300*
Low Glucose	143	6.39	25.76	35.08	0.099
Normal Glucose	415	7.32	26.95	37.45	0.244
High Glucose	41	6.45	24.56	34.18	0.358
p-Value		*0.30*	*0.54*	*0.41*	*0.018*
Low Cholesterol	9	5.19	18.91	26.26	0.133
Normal Cholesterol	632	6.08	22.84	31.60	0.207
High Cholesterol	112	6.99	29.01	39.45	0.412
p-Value		*0.32*	*≤0.0005*	*0.001*	*0.005*
Low Uric Acid	31	4.12	16.88	22.93	0.035
Normal Uric Acid	559	5.68	22.77	31.05	0.171
High Uric Acid	169	8.31	28.35	40.26	0.481
p-Value		*<0.0005*	*<0.0005*	*<0.0005*	*<0.0005*
Normal Creatinine	724	6.05	23.28	32.06	0.229
High Creatinine	42	9.40	32.52	45.64	0.348
p-Value		*0.001*	*<0.0005*	*<0.0005*	*0.230*

Note: Results from the Student-Newman-Keuls tests (alpha = 0.05) are indicated by the presence of a vertical line which is interpreted as follows: any two means not connected by the same line are significantly different, and any two means connected by the same line are not significantly different.

The *p*-value is given for the one-way analysis of variance which tests for the equality of means. Where the *p*-value is ≤0.10 then Student-Newman-Keuls tests were computed to determine which means were important.

NC = statistical test not computed.

DDE in their blood than had those whose occupations were not pesticide related. Dieldrin levels were also higher in this group, but statistical significance was not shown (Table 4).

When blood residues of the major DDT constituents and dieldrin are compared with residence, age, and sex, several general patterns emerge. Persons in rural areas tend to have greater residues of DDE; residues of $\sum$DDT and dieldrin tend to increase with age; and males have greater residues than have females. The cause of these relationships is undetermined (Table 2).

In general, the study showed that the county of residence was the most significant factor for determining pesticide residue levels. Occupation, sex, and location of residence were also determined to be associated with blood residue levels. Residues of $\sum$DDT and dieldrin were greater in older persons. No relationships were detected between blood hemoglobin and blood residue values. As the blood levels of glucose, cholesterol, uric acid, and creatinine increased, so generally did the levels of pesticide residues. However, when all variables were used, no equation could be developed which would reliably predict a blood residue level given those demographic characteristics.

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PESTICIDES IN FOOD AND FEED

Pesticide and Other Chemical Residues in Total Diet Samples (XI)

R. D. Johnson and D. D. Manske [1]

ABSTRACT

In the Total Diet study, the Food and Drug Administration has, since 1964, reported residues of pesticides and other chemicals ingested in the average diet of the Nation's largest eater, the young adult male. During the eleventh year of the study, pesticide residues remained at the relatively low levels reported previously. Twenty market baskets were collected in 20 cities which ranged in population from less than 50,000 to 1,000,000 or more. Averages and ranges of residues found are reported for August 1974 through July 1975 by food class. The individual items used in making up the dairy and meat composites in four market baskets were analyzed for pesticides and the results are included. Data for lead, cadmium, selenium, mercury, arsenic, and zinc are also included. Results of recovery studies within various classes of residues are also presented.

Introduction

The Food and Drug Administration Total Diet Program, sometimes called the Market Basket study, began with a program intended for surveillance of fission products for atomspheric tests of thermonuclear weapons in May 1961 (*9*). The program was quickly extended to pesticides and certain nutrients. Although changes have been made in sampling frequency, areas sampled, analytical methods, and types of residues sought, the program has continued in essentially the same form to the present. A market basket of food representing the basic 2-week diet of a 16- to 19-year-old male, statistically the Nation's largest eater, is collected in each of several geographic areas. The various foods are prepared, e.g., by cooking, in the manner in which they would normally be served and eaten. The foods in each of 12 broad classes are then composited into a slurry and analyzed for organochlorine and organophosphorus pesticides, carbaryl, herbicides, certain metals, and polychlorinated biphenyls (PCB's). Methodology includes atomic absorption spectroscopy, fluorometry, gas chromatography, thin-layer chromatography, mass spectroscopy, and established extraction and cleanup techniques. Conditions, techniques, and limits of quantitation have been described in previous reports in this series (*1–5, 12–15, 20*). Amounts and types of residues found from June 1964 through July 1974 have also been described in earlier reports (*6–8, 10, 11, 15–19*). The present report presents the results obtained from August 1974 through July 1975. Samples were collected in 20 different grocery markets in 20 different cities.

In addition, 10 market baskets of an Infant and Toddler series were analyzed for the first time and the data will be presented in a separate paper.

Results

During this reporting period 959 residues of 42 different compounds were found in 240 composites examined. In the previous reporting period, 1,613 residues of 42 different compounds were found in the 360 composites examined. The 42 different residues found are listed in decreasing order of frequency in Table 1. In Table 2, the frequency of occurrence of these residues is broken down according to food class. Table 3 gives the levels of the chemical residues by the food class in which they appeared. The average stated in Table 3 is based on 20 composites examined; no trace residues have been included in calculating the average. For this reason, an average value reported as "T" can be well below the detection limits of the methods for that compound.

The most common residues and their maximum levels are discussed below for each of the 12 food classes. No findings have been corrected for recoveries obtained in recovery experiments.

DAIRY PRODUCTS

Organochlorine compounds were the residues found most frequently in dairy products. The most common organochlorines were dieldrin, 0.005 ppm; BHC, 0.0012 ppm; DDE, 0.017 ppm; and heptachlor epoxide, 0.003 ppm. Other organochlorine residues present were HCB, methoxychlor, octachlor epoxide, lindane, and PCP. Zinc,

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TABLE 1. *Chemical residues found in food composites, August 1974–July 1975*

Chemical Found	No Composites with Residues	No Positive Composites with Residues Reported as Trace[1]	Range, ppm
Zinc	239	0	0 3–33 5
Cadmium	141	113	0 05–0 14
Lead	78	31	0.1–1 6
Selenium	59	19	0 10–0 42
Dieldrin Not less than 85% of 1,2,3,4,10,10–hexachloro–6,7–epoxy–1,4,4a,5,6,7,8,8a–octahydro–1,4–*endo*–*exo*–5,8–dimethanonaphthalene	56	8	0 001–0 015
BHC 1,2,3,4,5,6–hexachlorocyclohexane, mixed isomers except gamma	47	11	0 0004–0 0190
DDE 1,1–dichloro–2,2–bis (*p*–chlorophenyl) ethylene (isomers other than $_{p,p'}$ also included in reportings)	46	10	0 001–0 033
Malathion diethylmercaptosuccinate, *S*–ester with *O,O*–dimethyl phosphorodithioate	34	5	0 003–0 144
Diazinon *O,O*–diethyl *O*–(2–isopropyl–6–methyl–4–pyrimidyl) phosphorothioate	28	12	0 001–0 019
Heptachlor Epoxide 1,4,5,6,7,8,8–heptachloro–2,3–epoxy–3a,4,7,7a–tetrahydro–4,7–methanoindan	25	10	0 0006–0 003
Mercury	22	17	0 02
DDT 1,1,1–trichloro–2,2–bis (*p*–chlorophenyl) ethane (isomers other than $_{p,p'}$ also included in reportings)	20	9	0 003–0 016
HCB hexachlorobenzene	20	10	0 0002–0 017
Lindane 1,2,3,4,5,6–hexachlorocyclohexane, 99% or more gamma isomer	19	10	0 0007–0 0140
Arsenic (As_2O_3)	17	13	0 10–0 16
PCP pentachlorophenol	13	0	0 010–0 040
CIPC isopropyl *n*–(3–chlorophenyl) carbamate	12	0	0 003–0 653
Octachlor Epoxide 1–*exo*–2–*endo*–4,5,6,7,8,8–octachloro–2,3–epoxy–2,3,3a,4,7,7a–hexahydro–4,7–methanoindene	9	8	0 004
TDE 1,1–dichloro–2,2–bis (*p*–chlorophenyl) ethane (isomers other than $_{p,p'}$ also included in reportings)	9	5	0 002–0 004
PCB (polychlorinated biphenyls), calculated as Aroclor® with varied chlorine content	8	8	T
Endosulfan 6,7,8,10,10–hexachloro–1,5,5a,6,9,9a–hexahydro–6,9–methano–2,4,3–benzodioxathiepin 3–oxide (reportings include isomers I, II, and the sulfate)	7	2	0 004–0 022
Parathion *O,O*–diethyl *O*–*p*–nitrophenyl phosphorothioate	6	1	0 002–0 013
PCNB pentachloronitrobenzene	6	3	0 001–0 003
Botran® 2,6–dichloro–4–nitroaniline	5	0	0 001–0 016
PCA pentachloroaniline	5	0	0 002–0 035
Dicofol (Kelthane®) 4,4'–dichloro–a–)trichloromethyl) benzhydrol	3	0	0 006–0 033
Methyl Parathion *O,O*–dimethyl *O*–*p*–nitrophenyl phosphorothioate	3	0	0 0001–0 0010
Pentachlorobenzene	3	0	0 0005–0 0060
Carbaryl 1–naphthyl *n*–methyl carbamate	2	1	0 07

(Continued next page)

TABLE 1 (cont'd). *Chemical residues found in food composites, August 1974–July 1975*

Chemical Found	No Composites with Residues	No Positive Composites with Residues Reported as Trace[1]	Range, ppm
Ethion *O,O O,O* tetraethyl *S,S* -methylene bisphosphorodithioate	2	1	0.061
Methoxychlor 1,1,1-trichloro 2,2-bis (*p*-methoxyphenyl) ethane	2	1	0.007
Orthophenylphenol 2-hydroxydiphenyl	2	1	0.20
TCNB 1,2,4,5-tetrachloro-3-nitrobenzene	2	0	0.001–0.075
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	1	0	0.007
DCPA (Dacthal®) 2,3,5,6-tetrachloroterephthalic acid dimethyl ester	1	0	0.003
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	1	1	T
Leptophos *O*-(2,5-dichloro-4-bromophenyl)-*O*-methyl phenyl-phosphorothioate	1	0	0.040
Nonachlor 1,2,3,4,5,6,7,8,8-nonachloro-3a-4,7,7a-tetrahydro-4,7-*endo*-methanoindane	1	0	0.003
Perthane® 1,1-dichloro, 2,2-bis (*p*-ethylphenyl) ethane	1	0	0.142
Phosalone *O,O*-diethyl *S*-(6-chloro-2-oxobenzoxazolin-3-yl) methyl phosphorodithioate	1	0	0.025
Ronnel *O,O*-dimethyl (*O*-2,4,5-trichlorophenyl) phosphorothioate	1	0	0.004
Toxaphene chlorinated camphene containing 67 to 69% chlorine	1	0	0.118

[1] Chemicals capable of being detected by the specific analytical methodology may be confirmed qualitatively but are not quantifiable when they are present at concentrations below the limit of quantitation. Limit of quantitation varies with residue and food class.

TABLE 2. *Occurrence frequency of chemical residues by food class, August 1974–July 1975*

Chemical	Food Class											
	I	II	III	IV	V	VI	VII	VIII	IX	X	XI	XII
	Number of Occurrences											
Zinc	20	20	20	20	20	20	20	20	19	20	20	20
Cadmium	4	11	19	20	20	3	16	17	5	17	8	1
Lead		4	3	2	11	19	14	14	8		1	2
Selenium	5	20	20	1		6	1	2		2	2	
Dieldrin	17	19		7				12		1		
BHC	19	16	2				1	5		1	3	
DDE	11	20		7	6		1				1	
Malathion			19		1					12	2	
Diazinon		4	8		9		1	3	1	2		
Heptachlor Epoxide	11	13						1				
Mercury		20			1					1		
DDT		13		6							1	
HCB	8	7								4	1	
Lindane	1	6	3					5			4	
Arsenic		13	1	2								1
PCP	1		2		1		2	1	1		5	
CIPC				11					1			
Octachlor Epoxide	2	7										
TDE		9										
PCB		8										
Endosulfan					5			2				
Parathion					4		1	1				
PCNB									1	4	1	
Botran					4				1			
Pentachloroaniline										4	1	
Dicofol									3			
Methyl Parathion					3							
Pentachlorobenzene										2	1	
Carbaryl								1	1			
Ethion									2			

(Continued next page)

TABLE 2 (cont'd.). *Occurrence frequency of chemical residues by food class, August 1974–July 1975*

Chemical	Food Class										
	I	II	III	IV	V	VI	VII	VIII	IX	X	XI	XII
	Number of Occurrences										
Methoxychlor	2										
Orthophenylphenol								1	1			
TCNB			1	1								
Aldrin			1									
DCPA								1				
Endrin		1										
Leptophos								1				
Nonachlor		1										
Perthane					1							
Phosalone									1			
Ronnel			1									
Toxaphene					1							

[1]See Table 3 for description of food class.

TABLE 3. *Levels of chemical residues found by food class, August 1974–July 1975*

Residues, ppm

I. Dairy Products

ZINC		SELENIUM	
Average	4.9	Average	T
Positive Composites		Positive Composites	
Total Number	20	Total Number	5
Number Reported as Trace	0	Number Reported as Trace	5
Range	1.5–8.9	Range	T
BHC		CADMIUM	
Average	0.0005	Average	T
Positive Composites		Positive Composites	
Total Number	19	Total Number	4
Number Reported as Trace	5	Number Reported as Trace	4
Range	0.0004–0.0012	Range	T
DIELDRIN		METHOXYCHLOR	
Average	0.002	Average	T
Positive Composites		Positive Composites	
Total Number	17	Total Number	2
Number Reported as Trace	1	Number Reported as Trace	1
Range	0.001–0.005	Range	T–0.007
DDE		OCTACHLOR EPOXIDE	
Average	0.002	Average	T
Positive Composites		Positive Composites	
Total Number	11	Total Number	2
Number Reported as Trace	3	Number Reported as Trace	2
Range	0.001–0.017	Range	T
HEPTACHLOR EPOXIDE		LINDANE	
Average	0.0004	Average	T
Positive Composites		Positive Composites	
Total Number	11	Total Number	1
Number Reported as Trace	5	Number Reported as Trace	1
Range	0.0006–0.003	Range	T
HCB		PCP	
Average	0.0001	Average	0.0005
Positive Composites		Positive Composites	
Total Number	8	Total Number	1
Number Reported as Trace	5	Number Reported as Trace	0
Range	0.0002–0.0010	Range	0.0100

II. Meat, Fish, and Poultry

DDE		DDT	
Average	0.009	Average	0.004
Positive Composites		Positive Composites	
Total Number	20	Total Number	13
Number Reported as Trace	2	Number Reported as Trace	4
Range	0.003–0.033	Range	0.003–0.016
MERCURY		HEPTACHLOR EPOXIDE	
Average	T	Average	0.001
Positive Composites		Positive Composites	
Total Number	20	Total Number	13
Number Reported as Trace	15	Number Reported as Trace	4
Range	T–0.02	Range	0.001–0.003

(Continued next page)

TABLE 3 (cont'd.) *Level of chemical residues found by food class, August 1974-July 1975*

	Residues, ppm
SELENIUM	
Average	0.25
Positive Composites	
Total Number	20
Number Reported as Trace	0
Range	0.17–0.37
ZINC	
Average	27.6
Positive Composites	
Total Number	20
Number Reported as Trace	0
Range	23.4–33.5
DIELDRIN	
Average	0.004
Positive Composites	
Total Number	19
Number Reported as Trace	1
Range	0.002–0.015
BHC	
Average	0.0005
Positive Composites	
Total Number	16
Number Reported as Trace	4
Range	0.0005–0.0010
ARSENIC	
Average	0.03
Positive Composites	
Total Number	13
Number Reported as Trace	9
Range	0.10–0.16
LINDANE	
Average	0.0002
Positive Composites	
Total Number	6
Number Reported as Trace	4
Range	0.0018–0.0020
DIAZINON	
Average	T
Positive Composites	
Total Number	4
Number Reported as Trace	3
Range	T–0.0020
LEAD	
Average	T
Positive Composites	
Total Number	4
Number Reported as Trace	4
Range	T
CADMIUM	
Average	T
Positive Composites	
Total Number	11
Number Reported as Trace	11
Range	T
TDE	
Average	0.001
Positive Composites	
Total Number	9
Number Reported as Trace	5
Range	0.002–0.004
PCB	
Average	T
Positive Composites	
Total Number	8
Number Reported as Trace	8
Range	T
HCB	
Average	T
Positive Composites	
Total Number	7
Number Reported as Trace	3
Range	0.001–0.002
OCTACHLOR EPOXIDE	
Average	T
Positive Composites	
Total Number	7
Number Reported as Trace	6
Range	T–0.004
ENDRIN	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	1
Range	T
NONACHLOR	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.003

III. Grain and Cereal Products

SELENIUM	
Average	0.24
Positive Composites	
Total Number	20
Number Reported as Trace	0
Range	0.10–0.42
ZINC	
Average	8.5
Positive Composites	
Total Number	20
Number Reported as Trace	0
Range	4.1–11.7
CADMIUM	
Average	T
Positive Composites	
Total Number	19
Number Reported as Trace	17
Range	0.05–0.08
MALATHION	
Average	0.017
Positive Composites	
Total Number	19
Number Reported as Trace	0
Range	0.005–0.043
DIAZINON	
Average	0.001
Positive Composites	
Total Number	8
Number Reported as Trace	4
Range	0.002–0.008
LEAD	
Average	0.01
Positive Composites	
Total Number	3
Number Reported as Trace	2
Range	T–0.18

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TABLE 3 (cont'd.). *Level of chemical residues found by food class, August 1974-July 1975*

	Residues, ppm
LINDANE	
Average	0.0001
Positive Composites	
Total Number	3
Number Reported as Trace	1
Range	0.0007–0.0020
BHC	
Average	T
Positive Composites	
Total Number	2
Number Reported as Trace	1
Range	T–0.0010
PCP	
Average	0.001
Positive Composites	
Total Number	2
Number Reported as Trace	0
Range	0.010–0.013
ALDRIN	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.0070
ARSENIC	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	1
Range	T
RONNEL	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.0040
TCNB	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.001

IV. Potatoes

CADMIUM	
Average	0.04
Positive Composites	
Total Number	20
Number Reported as Trace	7
Range	0.05–0.12
ZINC	
Average	4.9
Positive Composites	
Total Number	20
Number Reported as Trace	0
Range	2.3–7.7
DIELDRIN	
Average	0.001
Positive Composites	
Total Number	7
Number Reported as Trace	2
Range	0.001–0.008
DDT	
Average	T
Positive Composites	
Total Number	6
Number Reported as Trace	4
Range	T–0.004
ARSENIC	
Average	T
Positive Composites	
Total Number	2
Number Reported as Trace	2
Range	T
CIPC	
Average	0.112
Positive Composites	
Total Number	11
Number Reported as Trace	0
Range	0.004–0.653
DDE	
Average	0.002
Positive Composites	
Total Number	7
Number Reported as Trace	2
Range	0.002–0.025
LEAD	
Average	T
Positive Composites	
Total Number	2
Number Reported as Trace	2
Range	T
SELENIUM	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	1
Range	T
TCNB	
Average	0.004
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.075

V. Leafy Vegetables

CADMIUM	
Average	0.05
Positive Composites	
Total Number	20
Number Reported as Trace	7
Range	0.05–0.14
ZINC	
Average	3.1
Positive Composites	
Total Number	20
Number Reported as Trace	0
Range	1.4–10.9
DIAZINON	
Average	0.003
Positive Composites	
Total Number	9
Number Reported as Trace	0
Range	0.001–0.019
DDE	
Average	T
Positive Composites	
Total Number	6
Number Reported as Trace	2
Range	0.003–0.005

(Continued next page)

TABLE 3 (cont'd.). *Level of chemical residues found by food class, August 1974-July 1975*

	Residues, ppm		
LEAD		ENDOSULFAN	
Average	0.06	Average	0.002
Positive Composites		Positive Composites	
Total Number	11	Total Number	5
Number Reported as Trace	4	Number Reported as Trace	1
Range	0.11–0.32	Range	0.004–0.022
BOTRAN		MERCURY	
Average	T	Average	T
Positive Composites		Positive Composites	
Total Number	4	Total Number	1
Number Reported as Trace	0	Number Reported as Trace	1
Range	0.001–0.006	Range	T
PARATHION		PCP	
Average	0.001	Average	T
Positive Composites		Positive Composites	
Total Number	4	Total Number	1
Number Reported as Trace	0	Number Reported as Trace	0
Range	0.002–0.013	Range	0.013
METHYL PARATHION		PERTHANE	
Average	T	Average	0.007
Positive Composites		Positive Composites	
Total Number	3	Total Number	1
Number Reported as Trace	0	Number Reported as Trace	0
Range	0.001–0.010	Range	0.142
MALATHION		TOXAPHENE	
Average	T	Average	0.006
Positive Composites		Positive Composites	
Total Number	1	Total Number	1
Number Reported as Trace	0	Number Reported as Trace	0
Range	0.009	Range	0.118
VI. Legume Vegetables			
ZINC		SELENIUM	
Average	7.7	Average	T
Positive Composites		Positive Composites	
Total Number	20	Total Number	6
Number Reported as Trace	0	Number Reported as Trace	6
Range	2.7–12.0	Range	T
LEAD		CADMIUM	
Average	0.26	Average	T
Positive Composites		Positive Composites	
Total Number	19	Total Number	3
Number Reported as Trace	2	Number Reported as Trace	3
Range	0.10–1.60	Range	T
VII. Root Vegetables			
ZINC		DDE	
Average	4.1	Average	T
Positive Composites		Positive Composites	
Total Number	20	Total Number	1
Number Reported as Trace	0	Number Reported as Trace	1
	1.4–28.6	Range	T
		DIAZINON	
Average	T	Average	T
Positive Composites		Positive Composites	
Total Number	16	Total Number	1
Number Reported as Trace	16	Number Reported as Trace	1
Range	T	Range	T
LEAD		PARATHION	
Average	0.16	Average	T
Positive Composites		Positive Composites	
Total Number	14	Total Number	1
Number Reported as Trace	5	Number Reported as Trace	0
Range	0.17–1.10	Range	0.002
PCP		SELENIUM	
Average	0.001	Average	T
Positive Composites		Positive Composites	
Total Number	2	Total Number	1
Number Reported as Trace	0	Number Reported as Trace	1
	0.010	Range	T

(Continued next page)

TABLE 3 (cont'd.). *Level of chemical residues found by food class, August 1974-July 1975*

	Residues, ppm
BHC	
Average	0.0005
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.010

VIII. Garden Fruits

ZINC		**CADMIUM**	
Average	3.1	Average	T
Positive Composites		Positive Composites	
Total Number	20	Total Number	17
Number Reported as Trace	0	Number Reported as Trace	17
Range	0.9–12.2	Range	T
LEAD		**CARBARYL**	
Average	0.08	Average	T
Positive Composites		Positive Composites	
Total Number	14	Total Number	1
Number Reported as Trace	5	Number Reported as Trace	1
Range	0.10–0.50	Range	T
DIELDRIN		**DCPA**	
Average	0.002	Average	T
Positive Composites		Positive Composites	
Total Number	12	Total Number	1
Number Reported as Trace	3	Number Reported as Trace	0
Range	0.002–0.008	Range	0.003
BHC		**HEPTACHLOR EPOXIDE**	
Average	0.0016	Average	T
Positive Composites		Positive Composites	
Total Number	5	Total Number	1
Number Reported as Trace	0	Number Reported as Trace	1
Range	0.0016–0.0190	Range	T
LINDANE		**LEPTOPHOS**	
Average	0.0004	Average	0.002
Positive Composites		Positive Composites	
Total Number	5	Total Number	1
Number Reported as Trace	3	Number Reported as Trace	0
Range	0.0030–0.0060	Range	0.040
DIAZINON		**ORTHOPHENYLPHENOL**	
Average	T	Average	T
Positive Composites		Positive Composites	
Total Number	3	Total Number	1
Number Reported as Trace	2	Number Reported as Trace	1
Range	T–0.001	Range	T
ENDOSULFAN		**PARATHION**	
Average	T	Average	T
Positive Composites		Positive Composites	
Total Number	2	Total Number	1
Number Reported as Trace	1	Number Reported as Trace	1
Range	T–0.006	Range	T
SELENIUM		**PCP**	
Average	T	Average	T
Positive Composites		Positive Composites	
Total Number	2	Total Number	1
Number Reported as Trace	2	Number Reported as Trace	0
Range	T	Range	0.010

IX. Fruits

ZINC		**CIPC**	
Average	1.2	Average	T
Positive Composites		Positive Composites	
Total Number	19	Total Number	1
Number Reported as Trace	0	Number Reported as Trace	0
Range	0.3–4.4	Range	0.003
LEAD		**DIAZINON**	
Average	0.02	Average	T
Positive Composites		Positive Composites	
Total Number	8	Total Number	1
Number Reported as Trace	5	Number Reported as Trace	1
Range	0.10–0.11	Range	T

(Continued next page)

TABLE 3 (cont'd.). *Level of chemical residues found by food class, August 1974-July 1975*

	Residues, ppm
CADMIUM	
Average	T
Positive Composites	
Total Number	5
Number Reported as Trace	5
Range	T
DICOFOL	
Average	0.002
Positive Composites	
Total Number	3
Number Reported as Trace	0
Range	0.006-0.033
ETHION	
Average	0.003
Positive Composites	
Total Number	2
Number Reported as Trace	1
Range	T-0.061
BOTRAN	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.016
CARBARYL	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.07
ORTHOPHENYLPHENOL	
Average	0.01
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.20
PCNB	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.001
PCP	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.011
PHOSALONE	
Average	0.001
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.025

X. Oils, Fats, and Shortening

ZINC	
Average	5.6
Positive Composites	
Total Number	20
Number Reported as Trace	0
Range	1.0-18.4
CADMIUM	
Average	T
Positive Composites	
Total Number	17
Number Reported as Trace	17
Range	T
MALATHION	
Average	0.023
Positive Composites	
Total Number	12
Number Reported as Trace	4
Range	0.012-0.144
[illegible]	
[illegible]	0.001
[illegible]	
[illegible]	1
[illegible]	2
[illegible]	0.004-0.017
PP'	
[illegible]	0.002
[illegible]	
[illegible]	4
[illegible]	0
[illegible]	0.003-0.025
PCNB	
[illegible]	T
[illegible]	
[illegible]	1
[illegible]	1
[illegible]	T
DIAZINON	
Average	T
Positive Composites	
Total Number	2
Number Reported as Trace	1
Range	T-0.004
PENTACHLOROBENZENE	
Average	0.0003
Positive Composites	
Total Number	2
Number Reported as Trace	0
Range	0.0010-0.006
SELENIUM	
Average	T
Positive Composites	
Total Number	2
Number Reported as Trace	2
Range	T
BHC	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	1
Range	T
DIELDRIN	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	1
Range	T
MERCURY	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	1
Range	T

TABLE 3 (cont'd.). *Level of chemical residues found by food class, August 1974-July 1975*

	Residues, ppm
XI. Sugars and Adjuncts	
ZINC	
Average	3.4
Positive Composites	
Total Number	20
Number Reported as Trace	0
Range	0.6–13.0
CADMIUM	
Average	T
Positive Composites	
Total Number	8
Number Reported as Trace	8
Range	T
PCP	
Average	0.006
Positive Composites	
Total Number	5
Number Reported as Trace	0
Range	0.010–0.040
LINDANE	
Average	0.0008
Positive Composites	
Total Number	4
Number Reported as Trace	1
Range	0.0007–0.0140
BHC	
Average	0.0001
Positive Composites	
Total Number	3
Number Reported as Trace	0
Range	0.0009–0.0010
MALATHION	
Average	T
Positive Composites	
Total Number	2
Number Reported as Trace	1
Range	T–0.003
SELENIUM	
Average	T
Positive Composites	
Total Number	2
Number Reported as Trace	2
Range	T
DDE	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.002
DDT	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	1
Range	T
HCB	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.0008
LEAD	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.12
PENTACHLOROANILINE	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.002
PCNB	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	1
Range	T
PENTACHLOROBENZENE	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	0
Range	0.0005
XII Beverages	
ZINC	
Average	0.8
Positive Composites	
Total Number	20
Number Reported as Trace	0
Range	0.3–4.1
LEAD	
Average	T
Positive Composites	
Total Number	2
Number Reported as Trace	2
Range	T
ARSENIC	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	1
Range	T
CADMIUM	
Average	T
Positive Composites	
Total Number	1
Number Reported as Trace	1
Range	T

NOTE: Average residues are based on 20 composites examined. No trace residues were included; hence average value reported as "T" can be well below detection limits of the method for that compound.

ranging from 1.5 to 8.9 ppm, was found in all 20 composites. Selenium and cadmium were occasionally found in this food class. No organophosphorus residues were detected.

MEAT, FISH, AND POULTRY

Twelve organochlorine compounds occurred in varying combinations in all 20 composites. The most common were DDE, 0.033 ppm; dieldrin, 0.015 ppm; BHC, 0.0010 ppm;

DDT, 0.016 ppm; and heptachlor epoxide, 0.003 ppm. Other residues were TDE, PCB, HCB, octachlor epoxide, lindane, diazinon, endrin, and nonachlor. Mercury, selenium, and zinc, ranging from trace to 0.02 ppm, 0.17 to 0.37 ppm, and 23.4 to 33.5 ppm, respectively, were found in all 20 composites; arsenic, cadmium, and lead were also found.

GRAIN AND CEREAL PRODUCTS

Selenium and zinc, ranging from 0.10 to 0.42 ppm and from 4.1 to 11.7 ppm, respectively, were found in all 20 composites. Malathion, ranging from 0.005 to 0.043 ppm, was found in 19 composites. Other residues included cadmium, diazinon, lead, lindane, BHC, PCP, aldrin, arsenic, ronnel, and TCNB.

POTATOES

Cadmium and zinc, ranging from 0.05 to 0.12 ppm and from 2.3 to 7.7 ppm, respectively, were found in all 20 composites. Of the five organochlorine residues that appeared in this composite, the most common were CIPC, 0.653 ppm; DDE, 0.025 ppm; and dieldrin, 0.008 ppm. Other residues were DDT, arsenic, lead, selenium, and TCNB.

LEAFY VEGETABLES

Organophosphates were the most frequently detected pesticide residue in leafy vegetables. The most common were diazinon, 0.019 ppm; parathion, 0.013 ppm; and methyl parathion, 0.010 ppm. Cadmium and zinc, ranging from 0.05 to 0.14 ppm and from 1.4 to 10.9 ppm, respectively, were found in all 20 composites. Lead, ranging from 0.11 to 0.32 ppm, was found in 11 composites. Less frequently occurring residues were DDT, endosulfan, botran, malathion, mercury, PCP, perthane, and toxaphene.

LEGUME VEGETABLES

Zinc, ranging from 2.7 to 12.0 ppm, was found in all 20 composites. Nineteen composites contained lead ranging from 0.10 to 1.60 ppm. Selenium and cadmium were also found occasionally. No organochlorine or organophosphates were detected in this composite.

ROOT VEGETABLES

Zinc, ranging from 1.4 to 28.6 ppm, was found in all 20 composites. Cadmium was found 16 times at the trace level in this food class. Lead, ranging from 0.17 to 1.10 ppm, was found in 14 composites. Other residues found were PCP, BHC, DDE, diazinon, parathion, and selenium.

GARDEN FRUITS

The most common pesticide residues in garden fruits were dieldrin, 0.008 ppm; BHC, 0.0190 ppm; lindane, 0.0060 ppm; and diazinon, 0.001 ppm. Twenty composites contained zinc ranging from 0.9 to 12.2 ppm. Other residues were cadmium, lead, endosulfan, selenium, carbaryl, DCPA, heptachlor epoxide, leptophos, orthophenylphenol, parathion, and PCP.

FRUITS

Dicofol, 0.033 ppm, and ethion, 0.061 ppm, were the most frequently encountered pesticide residues. Other residues were zinc, lead, cadmium, botran, carbaryl, CIPC, diazinon, orthophenylphenol, PCNB, PCP, and phosalone.

OILS, FATS, AND SHORTENING

The most common nonmetallic residues were malathion, 0.144 ppm; HCB, 0.17 ppm; PCA, 0.035 ppm; and PCNB, 0.003 ppm. Twenty composites contained zinc ranging from 1.0 to 18.4 ppm. Other residues were cadmium, diazinon, pentachlorobenzene, selenium, BHC, dieldrin, and mercury.

SUGARS AND ADJUNCTS

The most frequently found organochlorine residues were PCP, 0.040 ppm; lindane, 0.0140 ppm; and BHC, 0.0010 ppm. Twenty composites contained zinc ranging from 0.6 to 13.0 ppm. Other residues included cadmium, malathion, selenium, DDE, DDT, HCB, lead, pentachloraniline, PCNB, and pentachlorobenzene.

BEVERAGES

No organochlorine or organophosphates were found in any of the 20 beverage composites examined. Zinc, ranging from 0.3 to 4.1 ppm, was found in all 20 composites. Lead, arsenic, and cadmium were also present.

Discussion

Of the 240 composites examined, organochlorine residues were found in 118, or 49 percent. Corresponding findings from previous years were 48 percent, 1973–74; 52 percent, 1972–73; and 54 percent, 1971–72. Organophosphorus residues in the current reporting period were found in 61 composites, or 25 percent. Corresponding findings in previous years were 28, 31, and 28 percent, respectively.

Carbaryl occurred in two composites in this reporting period, once at 0.07 ppm and once at the trace level. This is below the eight findings of carbaryl in the previous reporting period. Orthophenylphenol, which is detected by the method used for carbaryl, was found in two composites, once at trace level and once at 0.20 ppm. Orthophenylphenol was reported five times in the previous reporting period.

No chlorophenoxy acid herbicides appeared in this reporting period. Pentachlorophenol, which is detected by the

method used for chlorophenoxy acids, was found 13 times. In the previous reporting period pentachlorophenol was reported 10 times. Five of the 13 findings were in the sugar and adjuncts composite.

Zinc appeared in 239 of the 240 composites, ranging from 0.3 to 33.5 ppm. The second most commonly occurring metal, cadmium, was found in all 12 food classes. It was detected in 141 of the 240 composites examined at levels ranging from 0.05 to 0.14 ppm.

The highest of the 78 findings of lead was 1.6 ppm, and the highest of the 59 findings of selenium was 0.42 ppm. Mercury was found in 22 composites; meat, fish, and poultry contributed 20 of those findings. The highest residue of mercury was 0.02 ppm. Arsenic was found in 17 composites; meat, fish, and poultry contributed 13 of the findings. The highest arsenic residue was 0.16 ppm As_2O_3.

The individual commodity analysis for chlorinated, organophosphate, and PCP residues on food groups I (dairy) and II (meats) begun in earlier programs was continued on four samples of this period's 20 Total Diet samples. Composites I and II were selected because past data showed that the major occurrence of significant chlorinated residues was in these two groups. Individual commodity analysis results are shown in Table 4 (dairy group) and Table 5 (meat group). Three items from the dairy group, namely, buttermilk, skim milk, and nonfat dry milk, and one item from the meat group, lamb, are not shown because they contained no residues.

Recovery studies were conducted for all classes of chemicals sought throughout the entire year. Table 6 lists the recovery data for this reporting period. Each recovery experiment consisted of a single determination for the unfortified food composite and a single determination for the fortified sample. Since these were performed simultaneously, occasionally the fortification level was below the level present in the sample. In other cases, not enough recoveries were run to permit statistical evaluation. These data are not reported.

At very low fortification levels, recoveries may range from 0 to 200 percent. As the fortification level is raised, however, recovery approaches 100 percent. Recovery data indicate that individual, low-level residues reported may vary from the so-called true value but the overall findings are useful in appraising the national residue picture.

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TABLE 4 *Pesticide residues in individual commodities of dairy composite of four market basket samples, August 1974–July 1975*

Residue Found	Commodity[1,2]							
	Whole Milk (4)	Evaporated Milk (4)	Ice Cream (4)	Cottage Cheese (4)	Processed Cheese (4)	Natural Cheese (4)	Butter (4)	Ice Milk (2)
BHC								
Times Found	2	2	4	1	4	4	4	1
ppm Range	T	0.001	T–0.002	T	0.002–0.007	0.002–0.007	T–0.010	T
p,p'–DDE								
Times Found	3	2	4	2	3	4	4	1
ppm Range	T–0.003	0.005–0.017	0.002–0.030	T–0.005	0.005–0.015	0.003–0.006	T–0.220	T
Dieldrin								
Times Found	1	3	3	1	4	4	4	
ppm Range	T	T–0.004	T–0.004	T	0.004–0.016	0.002–0.008	0.002–0.039	
Heptachlor Epoxide								
Times Found			1		4	4	4	
ppm Range			T		0.002–0.005	T–0.004	T–0.006	
HCB								
Times Found		1	1		2		3	
ppm Range		T	T		T–0.002		0.002–0.006	
Lindane								
Times Found						1	1	
ppm Range						T	0.001	
Octachlor Epoxide								
Times Found							2	
ppm Range							0.005–0.008	
Methoxychlor								
Times Found					1	1		
ppm Range					0.035	0.010		
p,p'–DDT								
Times Found					1			
ppm Range					T			

[1]Buttermilk, skim milk, and nonfat dry milk not shown because no residues were found
[2]Numbers in parentheses show the number of times that commodity was analyzed

TABLE 5. *Pesticide residues in individual commodities of meat composite of four market basket samples, August 1974–July 1975*

Residue Found	Commodity[1,2]														
	Roast Beef (4)	Ground Beef (4)	Pork Chops (4)	Bacon (4)	Chicken (4)	Fish Fillet (4)	Fish Canned (4)	Lunch Meat (4)	Franks (4)	Beef Liver (4)	Eggs (4)	Ham (4)	Round Steak (4)	Veal (1)	Shrimp (2)
BHC															
Times Found	2	3				1	2	3	4	1		1	2	1	
ppm Range	0.001–0.002	0.001–0.002				0.001	0.001–0.004	0.002	T–0.005	0.001		0.001	T–0.001	0.006	
DDE															
Times Found	3	2	4	4	4	2	3	3	4		2	1	2	1	1
ppm Range	T–0.007	0.007–0.021	T–0.139	T–0.007	0.002–0.016	0.008–0.028	0.003–0.023	0.006–0.058	0.006–0.027		0.002–0.003	0.019	0.007–0.020	T	T
TDE															
Times Found			2	1		1	1	1	1			1			
ppm Range			T–0.023	0.003		T	0.008	T	0.005			0.015			
p,p'–DDT															
Times Found			2	2	1	1		3	3			1			
ppm Range			T–0.200	0.019–0.022	0.010	0.084		T–0.012	T–0.032			0.078			
HCB															
Times Found		2				1	2	2					1	1	
ppm Range		T				0.001	T–0.002	T–0.003					T	0.002	
Dieldrin															
Times Found	3	3	3	3	2	2	2	4	4	3	2	1	3		
ppm Range	T–0.021	0.003–0.004	T–0.002	T–0.011	T 0.002	T–0.005	T–0.002	T 0.008	0.003–0.240	T–0.020	T	T	0.001–0.006		
Heptachlor Epoxide															
Times Found	1	2		2	1	1		4	3	2		1	3		
ppm Range	0.003	0.001		T–0.003	T	T		T–0.003	T 0.004	0.003–0.004		T	T–0.051		
Lindane															
Times Found	1	2	1	1	1			3	2						
ppm Range	0.014	T	T	T	0.001			T–0.003	0.001						
PCB															
Times Found						2	1								1
ppm Range						T	0.05								T
Chlordane															
Times Found						1		2		1			1		
ppm Range						T		T		0.002			0.010		
Endrin															
Times Found					1	1									
ppm Range					T	0.003									
Octochlor Epoxide															
Times Found						1									
ppm Range						0.015									

[1]Lamb not shown because no residues found

[2]Numbers in parentheses show the number of times that commodity was analyzed

TABLE 6. *Recovery data on residues found in total diet samples, August 1974—July 1975*

Residue	Type of Food Composites	Spike Level, ppm	Range of Blank Level, ppm[1]	Range of Total Found, ppm[1]	No. of Recovery Studies
Cadmium	Fatty	0.1	0–0.030 (0.007)	0.089–0.171 (0.108)	20
	Nonfatty	0.1	0–0.064 (0.018)	0.068–0.174 (0.116)	37
Lead	Fatty	0.2	0–0.064 (0.014)	0.068–0.272 (0.181)	20
	Nonfatty	0.2	0–0.164 (0.041)	0.036–0.536 (0.226)	36
Selenium	Fatty	0.2	0–0.17 (0.01)	0.09–0.42 (0.19)	36
	Nonfatty	0.2	0–0.18 (0.04)	0.11–0.36 (0.021)	14
Zinc	Fatty	5	3.50–5.00 (4.53)	8.55–10.96 (9.70)	8
	Nonfatty	5	0.31–4.38 (2.43)	5.24–10.42 (7.32)	24
Mercury	Fatty	0.06	0–0.022 (0.005)	0.044–0.078 (0.059)	20
	Nonfatty	0.06	0–0.002 (< 0.001)	0.042–0.067 (0.059)	40
Arsenic	Fatty	0.3	0–0.240 (0.030)	0.165–0.405 (0.305)	20
	Nonfatty	0.3	0–0.070 (0.015)	0.175–0.465 (0.320)	39
Orthophenyl phenol	Nonfatty	0.4	0	0.08–0.40 (0.34)	40
Carbaryl	Nonfatty	0.2	0	0.06–0.20 (0.18)	40
2,4-DB	Fatty	0.02	0	0–0.019 (0.012)	8
	Nonfatty	0.02	0	0.005–0.028 (0.016)	17
2,4-D	Fatty	0.02	0	0–0.020 (0.009)	10
	Nonfatty	0.02	0	0.004–0.029 (0.016)	17
[illegible]	Fatty	0.02	0	0.005–0.013 (0.009)	3
[illegible]	[illegible]	0.02	0	0.006–0.025 (0.016)	8
		0.05	0	0.037–0.058 (0.046)	4
		0.05	0	0.22–0.056 (0.037)	8
		0.01	0–0.0086 (0.00126)	0.0076–0.0212 (0.0124)	6
		0.01	0–0.0013 (0.0002)	0.0014–0.0128 (0.00896)	12

TABLE 6 (cont'd.) *Recovery data on residues found in total diet samples, August 1974—July 1975*

Residue	Type of Food Composites	Spike Level, ppm	Range of Blank Level, ppm[1]	Range of Total Found, Studies	No. of Recovery
CIPC	Fatty	0.05	0	0.036–0.054 (0.042)	4
	Nonfatty	0.05	0–0.029 (0.005)	0.021–0.088 (0.045)	6
Ethion	Fatty	0.005	0	0.0030–0.0053 (0.0038)	4
	Nonfatty	0.005	0	0–0.0070 (0.0039)	8
Octachlor Epoxide	Nonfatty	0.003	0	0.0026–0.0039 (0.0031)	6
Parathion	Nonfatty	0.005	0	0.0031–0.0058 (0.0042)	6
Endrin	Nonfatty	0.01	0	0.0074–0.0107 (0.0092)	6

[1] Numbers in parentheses represent average residue levels

PESTICIDES IN FISH, WILDLIFE, AND ESTUARIES

Concentrations of Total Mercury in Several Fishes from Delaware Bay, 1975 [1]

Ellen Heath Gerhart [2]

ABSTRACT

Mercury levels in eleven species of fishes collected from lower Delaware Bay in April 1975 ranged from 0.018 to 0.321 μg/g and averaged 0.092 μg/g. Raja eglanteria, *the clearnose skate, had the highest mean mercury concentration (0.214* μg/g). *No correlation was found with feeding habits. These assays provide baseline data for future measurement of mercury accumulation by fishes in this region.*

Introduction

Since the Minamata tragedy in Japan in the 1950's-1960's (*8*), reports of mercury levels in the aquatic environment have proliferated (*4, 12*). Environmental mercury monitoring in Sweden has been particularly comprehensive, revealing high mercury concentrations in many regions. Compilation of Swedish data on the subject showed upper levels of 5.0 μg/g in fish tissue (*9*); Holden (*7*) reported even higher concentrations, 9.8 μg/g in pike muscle tissue. Mercury levels up to 5.0 μg/g were found in fishes from the Lake Erie area of the United States (*14*). Total mercury concentrations are reported here for several species of Delaware Bay fishes collected in spring 1975.

Methods

Eleven species of fishes were collected in lower Delaware Bay east of Fowler Beach, Del. The fishes were caught with an otter trawl at depths of 36–220 m on April 10, 1975. Three individuals of each species were frozen separately in polyethylene bags. At the time of analysis the fishes were thawed and their total lengths were measured. From each fish two epaxial muscle samples were removed, each weighing approximately 5 g. In the case of anchovies, whole fish were analyzed.

Each sample was digested in a 250-ml Erlenmeyer flask containing 10 ml distilled water, 10 ml 36 N H_2SO_4 (AR), 10 ml 16 N HNO_3 (AR), and 2 g potassium permanganate crystals. This mixture was allowed to digest, with occasional shaking, for 16–20 hours at 30° C. A control flask containing no tissue was prepared with each set of digestions. After digestion the sample was diluted to approximately 100 ml with distilled water and quantitatively transferred to a standard BOD bottle. The concentration of total mercury was determined by the method of Hatch and Ott (*5*) using Coleman mercury-free reagents. An MAS-50 Perkin-Elmer (Coleman) mercury analyzer was calibrated at each analysis period with known mercury standards. All glassware was thoroughly washed and soaked in 10 percent nitric acid for at least 12 hours prior to analyses.

Results and Discussion

The data indicate detectable concentrations of mercury in all fish analyzed (Table 1). In contrast, mercury had not been detected in samples of water, detritus, algae, and shrimp (*Palaemonetes vulgaris*) collected at Lewes, Del., in spring 1974 (*11*).

No fishes sampled had concentrations higher than 0.5 μg/g, which is the upper permissible level recommended by the Food and Drug Administration, U.S. Department of Health, Education, and Welfare. Total mercury concentrations exceeding 0.20 μg/g are considered elevated (*1*). Only *Raja eglanteria*, the clearnose skate, showed a mean mercury value above 0.20 μg/g. The average mercury concentration of the 35 fishes analyzed was 0.092 μg/g; the range was 0.018–0.321 μg/g.

Many fish species are too mobile to be good indicators of regional mercury contamination. Fishes such as the summer flounder, winter flounder, and clearnose skate have extensive seasonal migrations and spend only part of the year in Delaware Bay. The clearnose skate returns to Delaware Bay in early spring (*3*) and the high mercury value for this fish may indicate exposure to mercury in its more southerly wintering grounds.

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TABLE 1. *Total mercury concentrations in Delaware Bay fishes, 1975*

	Total Mercury Concentrations, μg g		Total Length Ranges, cm
	Mean[1]	Range	
Scophthalmus aquosus (Windowpane)	0.149 ± 0.081	0.666 – 0.251	21–26
Raja Eglanteria (Clearnose Skate)	0.214 ± 0.098	0.119 – 0.321	25–27[2]
Merluccius Bilinearis (Silver Hake)	0.046 ± 0.010	0.036 – 0.060	21–28
Anchoa mitchilli (Bay Anchovy)	0.051 ± 0.029	0.032 – 0.084	5–7
Alosa pseudoharengus (Alewife)	0.065 ± 0.023	0.041 – 0.092	20–29
Urophycis chuss (Spotted Hake)	0.062 ± 0.021	0.047 – 0.077	14
Clupea harengus (Atlantic Herring)	0.104 ± 0.045	0.073 – 0.194	30–34
Paralichthys dentatus (Summer Flounder)	0.096 ± 0.044	0.052 – 0.155	15–19
Pseudopleuronectes americanus (Winter Flounder)	0.057 ± 0.029	0.023 – 0.106	18–20
Urophycis chuss (Red Hake)	0.075 ± 0.031	0.022 – 0.105	21–22
Prionotus carolinus (N. Sea Robin)	0.089 ± 0.075	0.018 – 0.168	9–13

[1] In mean column, numbers following ± represent standard deviation
[2] Wing width

Although mercury content has been shown to be related to feeding habit (*10*), no clear correlation was found here. Plankton feeders such as herrings, alewives, and anchovies were not significantly lower in mercury than predators such as the hakes and flounders.

Studies of other saltwater and estuarine fishes have demonstrated similar or often higher mercury levels. Swedish fishes showed a range of 0.025–0.110 μg g in the 1930's (*13*). More recent studies of Swedish coastal zone fishes report a range of 0.050–4.160 μg/g (*6*). Fishes obtained from Genoa, Italy, showed mercury levels as high as 2.593 μg/g (*2*). Estuarine fishes from Tasmania had some species in the 1.6–2.0 μg/g range although most were assayed at 0.2–0.5 μg/g (*10*). By comparison, values from Delaware Bay fishes reported in this study are relatively low.

Acknowledgments

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Residues of Organochlorine Pesticides and Polychlorinated Biphenyls and Autopsy Data for Bald Eagles, 1973–74 [1]

Richard M. Prouty, William L. Reichel, Louis N. Locke, [2] Andre A. Belisle, Eugene Cromartie, T. Earl Kaiser, Thair G. Lamont, Bernard M. Mulhern, and Douglas M. Swineford

ABSTRACT

Eighty-six bald eagles found sick or dead during 1973–74 in 24 States were analyzed for organochlorine compounds. DDE was detected in all carcasses, polychlorinated biphenyls (PCB's) were found in all but two. Seventy-five carcasses contained TDE and/or dieldrin. Four eagles had possibly lethal levels of dieldrin in the brain. Bald eagles continue to retain high residue levels of organochlorine pollutants. Illegal shooting remained the most common cause of death but accounted for a smaller percentage of the mortalities than in the two previous biennial collections.

Introduction

In a continuing effort to determine the reasons for the decline of the bald eagle (*Haliaeetus leucocephalus*) in the United States and to monitor residues, dead or moribund specimens are collected in the field and sent to Patuxent Wildlife Research Center by Federal, State, and private cooperators for autopsy and analysis. Residue and autopsy data for specimens collected in 1973–74 are reported in this paper; previous years' collections already have been published (*1–4*).

Sampling and Autopsy

[illegible] shipment and storage of these specimens [illegible] have been described (*2*). No systematic col[illegible] bald eagles has been attempted due to the relatively low population and protected status of these birds. Eighty-six specimens were collected in 24 States in 1973–74; the breakdown by State and by year appears in Table 1. Decomposed specimens were not analyzed. Autopsy procedures followed those described previously by Belisle et al. (*1*).

[illegible] U.S. [illegible] Patuxent Wildlife Research [illegible] MD [illegible]

Pr[illegible] U.S. [illegible] Department of the Interior, National [illegible] Laurel, MD [illegible]

Analytical Procedures

After removal of the skin, feet, wings, brain, and gastrointestinal tract, the carcass was ground and homogenized in a Hobart food cutter. A 10-g aliquot and the entire brain were mixed separately with anhydrous sodium sulfate in a blendor and extracted for 7 hours with hexane in a Soxhlet apparatus. Extracts were cleaned by Florisil column chromatography and the pesticides and PCB's were fractionated by Silicar column chromatography. These procedures have been described in detail by Cromartie et al. (*2*).

The fractions were analyzed with a Hewlett-Packard 5753 gas chromatograph (GC) equipped with a Ni^{63} detector, automatic sampler, computing integrator, and a 4 percent SE-30/6 percent QF-1 column at 190°C. Pesticides were quantitated by computer integration of the peak areas, and

TABLE 1. *Distribution of eagles collected by State and year of death, 1973–74*

State	No. Eagles Collected	
	1973	1974
Alaska	6	9
Arkansas		1
Florida	4	
Georgia	1	1
Illinois		2
Indiana		1
Iowa	3	1
Kansas		5
Louisiana		1
Maine	1	
Maryland	1	1
Michigan	4	3
Minnesota	2	12
Missouri	1	1
Montana		1
Nevada		1
New Jersey	1	1
North Dakota	2	
Oklahoma		1
South Carolina	1	
South Dakota	2	
Virginia	1	4
Wisconsin	6	2
Wyoming		2
TOTAL	36	50

PCB's were estimated by comparing total peak area with Aroclor 1254 or 1260. In part of the 1974 specimens, toxaphene was estimated semiquantitatively from the area of two peaks (GC) whose retention times were 1.09 and 1.19, compared to 1.00 for DDT. The area of these peaks represents approximately 5 percent of the cut weight of the total area under the curve traced on a GC chart after an injection of standard toxaphene. Thus toxaphene is reported in terms of the two peaks and is not corrected in the original technical material.

Residues in 36 specimens (42 percent) were confirmed with an LKB 900 gas chromatograph/mass spectrometer (GC-MS). Operating procedures were described previously (2).

Average percentage recoveries from spiked mallard carcass tissue were DDE, 96; TDE, 103; DDT, 110; dieldrin, 101; heptachlor epoxide, 104; mirex, 106; oxychlordane, 98; *cis*-chlordane, 100; *cis*-nonachlor, 98; hexachlorobenzene, 69; and Aroclor 1254, 101. Residue levels reported here are not corrected for recovery. The lower limit of detection for all compounds was 0.05 ppm.

Results and Discussion

RESIDUES

The residues of organochlorine pesticides and PCB's in 86 specimens are summarized in Table 2. Median carcass residues of dieldrin, TDE, DDE, and PCB's showed no decrease from the years 1964–72 (*1–4*). DDE was detected in all carcasses; PCB's were found in all but two. Seventy-five carcasses contained TDE and or dieldrin.

Three specimens had 3.9–7.9 ppm dieldrin in the brain (Table 3); based on the criteria of Stickel et al. (5), they probably died from pesticide poisoning. A fourth possibly died from a combination of disease and of pesticide poisoning; its brain contained 3.6 ppm dieldrin. An adult male from Maryland with 7.9 ppm dieldrin in the brain had an extensive laceration of the right auricle of the heart and resultant hemorrhage in the lungs and pericardial sac; a large clot overlaid the ventral surface of the liver. These lesions suggest that the eagle was killed by a blow to the thoracic cavity, possibly received by falling from a high perch or during flight. In all four birds, coronary fat deposits were either absent or markedly atrophied, which is

TABLE 2. *Pesticide and PCB residues in 86 bald eagles, 1973–74*

Compound	Year	Residues, ppm Wet Weight					
		Carcass			Brain		
		No Specimens[1]	Median	Range	No Specimens	Median	Range
p,p'-DDE	1973	36	12 0	0 15–110 0	35	3 4	0 05– 47 0
	1974	50	6 7	0 14– 85 0	46	8 6	0 05–110 0
p,p'-TDE	1973	31	1 4	0 07– 16 0	19	1 0	0 12– 3 4
	1974	44	0 47	0 05– 7 5	20	0 10	0 05– 1 8
p,p'-DDT	1973	10	0 22	0 07– 1 2	5	0 29	0.07– 0 53
	1974	18	0 12	0 05– 1 7	4	0 07	0 06– 0 16
Dieldrin	1973	34	0 74	0 06– 14 0	24	1 3	0 07– 7 9
	1974	41	0 63	0 07– 9 3	31	0 18	0 05– 4 3
Heptachlor epoxide	1973	22	0 43	0 05– 2 0	17	0 31	0 05– 0 92
	1974	32	0 20	0 05– 1 5	14	0 09	0 05– 0 57
Mirex	1973	10	0 73	0 05– 8 3	8	0 27	0 11– 1 8
	1974	18	0 25	0 07– 2 2	9	0 31	0 06– 0 90
Oxychlordane	1973	19	0 21	0 05– 1 6	12	0 32	0 12– 0 67
	1974	31	0 17	0 05– 1 0	12	0 12	0 05– 0 48
cis-Chlordane[2]	1973	25	0 45	0 08– 3 5	22	0 50	0 05– 2 7
	1974	38	0 37	0 07– 7 8	18	0 18	0 05– 2 8
cis-Nonachlor	1973	20	0 49	0 07– 2 1	16	0 29	0.05– 0 61
	1974	33	0.12	0.05– 1 3	7	0 10	0.07– 0 64
Hexachlorobenzene	1973	15	0 11	0 05– 1 2	16	0 07	0 05– 0 58
	1974	15	0 08	0 05– 0 15	5	0 07	0 05– 0 11
Toxaphene	1974	23[3]	0 28	0 05– 0 95	10	0 22	0 05– 1 2
PCB's	1973	34	23 0	1 4 –820 0	33	7 5	0 24–220 0
	1974	50	9 9	0 30–120 0	46	1 6	0 10– 98 0

[1] Number of specimens containing residues, the median is based on this number
[2] And or *trans*-nonachlor
[3] Only 37 of a total of 50 specimens were analyzed for toxaphene

TABLE 3 *Data on four suspected cases of dieldrin poisoning of bald eagles, 1973–74*

State	Year	Sex	Dieldrin Residues in Brain, ppm	Autopsy Findings [1]
Maryland	1973	M	7.9	Cardiac rupture, no fat deposits
Michigan	1973	M	3.9	Emaciation, no fat deposits
Florida	1973	F	3.6	Enteritis, no fat deposits
Illinois	1974	F	4.3	Open, no fat deposits

[1] Open = no diagnosis could be made on the basis of autopsy findings

characteristic of pesticide poisoning. Although the body burdens of pesticide residues remained high, the percentage of eagles that died from pesticide poisoning in the 1973–74 collection (4.6 percent) decreased to less than half that of the two previous collections.

A minor 9-chlorine component of technical chlordane has been detected and confirmed by GC-MS in specimens containing appreciable chlordane residues. As with other chlordane components, the molecular ion (M), m/e 454, is weaker than the more intense M-Cl ion, m/e 419. This compound has been assigned the empirical formula $C_{11}H_7Cl_9$ by Dr. G. W. Sovocool, Health Effects Research Laboratory, Environmental Toxicology Division, Analytical Branch, U.S. Environmental Protection Agency, Research Triangle Park, N.C. It is probably the result of a side reaction between chlordene and chloroform during chlordane manufacture.

AUTOPSY

Illegal shooting remained the single most frequent cause of death (25 percent) among the eagles examined in this series (Table 4) but it accounted for a smaller percentage of the mortalities than in the two previous biennial collections (*1,2*).

Of seven drowned eagles, an adult male died in the Rappahannock River, Va., while fighting with another eagle. The victim had subcutaneous and deep muscle lacerations probably inflicted by the talons of the other eagle.

[illegible] eagle with multiple puncture wounds and [illegible] was found floundering in the road near Phillips Landing, Va. His puncture wounds were similar to those in the Rappahannock male and in captive raptors killed during fights.

An aged emaciated female was found dead beside a road in Kansas; no evidence of trauma or poisoning was detected. There were obstructing thrombi in the major arteries of the thorax, and the eagle's death appeared to have resulted [illegible] myocardial infarction subsequent to coronary

[illegible] Alaska, was killed when it flew into [illegible] Guard C-130; nonlethal levels of organochlorine pesticides were present in its brain and carcass.

Mortalities associated with the trapping of muskrats and mink for fur appear to have increased in 1973–74. Eagles are frequently captured in steel traps placed on muskrat houses, beaver dams, or pond dikes, and drown when they fall into the water. Six of the seven drowned eagles had such histories. Three eagles that died of fractured skull were apparently killed by fur trappers who found the birds in their traps. Other entrapped eagles are often shot.

Acknowledgments

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TABLE 4 *Probable causes of bald eagle mortality, 1973–74*

Cause of Death	No. Eagles
Shot	21
Drowning	7
Impact injuries	7
Electrocution	6
Emaciation	6
Coccidiosis	5
Dieldrin poisoning	4
Fractured skull	3
Shot, secondary bacterial infection	3
Arteriosclerosis, myocardial infarction	1
Aspergillosis	1
Asphyxiation	1
Bacterial infection	1
Euthanasia	1
Internal hemorrhage	1
Laceration, hemorrhage	1
Nephrosis	1
Multiple puncture wounds	1
Pneumonia	1
Trapping	1
Open [1]	13
Total	86

[1] No diagnosis could be made on the basis of autopsy findings or chemical analysis

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Influence of Environmental Factors on Pesticide Levels in Sport Fish [1]

Michael James Vanderford [2] and Jerry Lee Hamelink [3]

ABSTRACT

The extent of pesticide contamination of sport fish from lakes and reservoirs in Indiana is described. Environmental and water quality factors significantly influenced the concentration of pesticides in fish. The influence of these factors was different between natural lakes and humanmade reservoirs.

*Largemouth bass (*Micropterus salmoides*), sunfish (*Lepomis *sp), and bullheads (*Ictalurus *sp.) were collected and analyzed for dieldrin, aldrin, DDT, DDE, heptachlor, and heptachlor epoxide. The collections and concurrent lake surveys were made during 1972–73.*

Soil and decaying plant particles appeared to influence the levels of dieldrin and Σ DDT taken up by largemouth bass. Strong correlations were observed between lake turbidity and true color and residue levels in the fish. Residue concentrations were also observed to vary between fish species, the seasons of the year, and watershed land uses.

Introduction

The survey reported here was conducted primarily to determine whether sport fish in the heavily agricultural areas of Indiana were grossly contaminated with chlorinated hydrocarbon pesticides. Specifically, the survey was designed to determine the source and causes of variations in pesticide concentrations in fish often observed in previous pesticide monitoring programs.

Materials and Methods

Fish were collected from eight natural lakes and twelve reservoirs in Indiana (Figure 1), using electro-fishing equipment and gillnets. An attempt was made to collect at least three individual bass, sunfish, and bullheads from each lake sampled. Because the age of fish influences residue levels(*1*), authors attempted to collect the same age class (3-4 years) of fish from all areas. Length, weight, sex, condition, maturity, and liver weight of each fish were recorded.

SAMPLE ANALYSIS

Fish were inspected, weighed, measured, and then frozen immediately with dry ice. Fish samples were homogenized with dry ice for fat and residue extraction (*2*). Each whole frozen fish was cut into large pieces, mixed with dry ice, and ground into a coarse meal with a Hobart food chopper. The meal and dry ice were homogenized with a high-speed Sorval Omni-Mixer to produce a free-flowing powder. The grinding and homogenization were performed in a chest

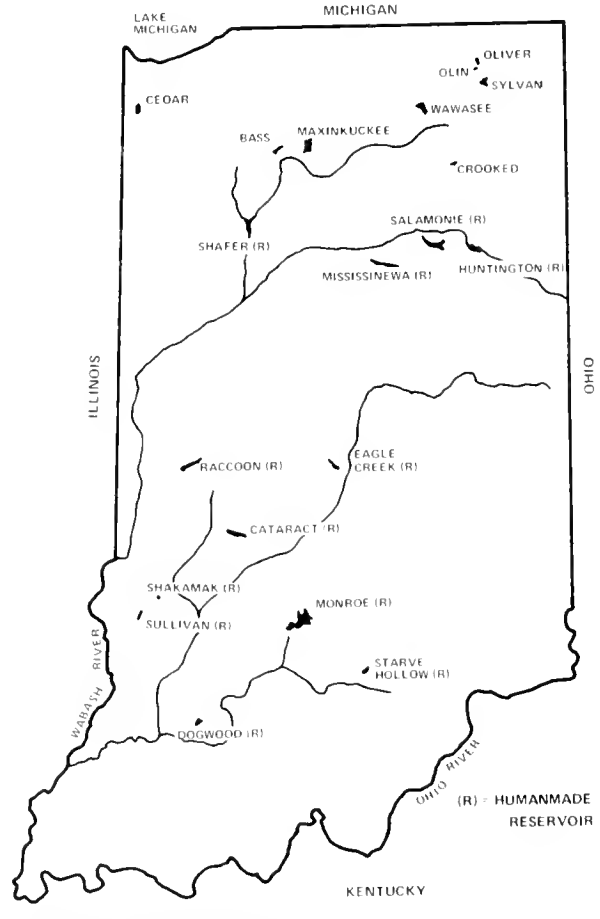

FIGURE 1. *Map of Indiana showing location of 20 lakes and reservoirs sampled.*

[1] ... and Conservation, Purdue University. Study funded by ... Resources.
[2] ... Service, U.S. Department of Interior, 538 Fed ... 55101.
[3] ... P.O. Box 708, D-781, B240, Green ...

freezer with chilled instruments to prevent caking. Separate portions of the whole powdered fish were used for insecticide residue extraction and for determining percentages of fat and water.

The extraction and cleanup procedure employed in the insecticide residue analysis was a modification of methods described by Berg et al., 1972, developed by D. L. Ballee and R. C. Hall of the Purdue University Entomology Department (*4, 18*). Five g fish powder were extracted with 10 g cleaned anhydrous Na_2SO_4 and 40 ml glass distilled methanol in a stainless steel Sorval Omni-Mixer. The mixture and two 10-ml methanol rinses were filtered through a layer of Na_2SO_4 held in a coarse fritted glass funnel into a 125-ml Teflon separatory funnel equipped with a stopcock. The methanol solution was partitioned against three successive 50-ml volumes of hexane. The methanol portion was discarded and the hexane solution was evaporated to dryness before cleanup.

A 4-ft-by-8-mm-OD glass column was packed bottom to top with 2 g sand (45 mm), 1 g Na_2SO_4 (25 mm), 4 g coconut charcoal (310 mm), 1 g Na_2SO_4 (25 mm), and 1 g sand (22 mm). Fisher brand coconut charcoal #5-690, 50–200 mesh, benzene-cleaned and activated at 180°C (*3*), was the only charcoal suitable for the initial fractionation. The hexane-soluble residues were quantitatively transferred into the prewashed column and first eluted with 100 ml 25 percent acetone in ether (fraction A), and then eluted with 100 ml benzene (fraction B) (*3*).

Fraction A was further refined on a 2-g florisil column and successively eluted with 100 ml hexane (fraction A-1) and 100 ml 25 percent ether in hexane (fraction A-2) (*14*). Fraction B was fractionated on a 3-g silica-gel column first eluted with 230 ml hexane (fraction B-1) which contained all PCB's, followed by 40 ml benzene (fraction B-2) (*8,10*). All fractions A–2 and B–2 containing dieldrin and ΣDDT residues, respectively, were analyzed by gas chromatography (GC) with either a Tracor MT-220 or a Packard 7000 series GC equipped with Ni^{63} electron-capture detectors and 6-ft-by-2-mm-ID glass columns packed with 3 percent OV-1 or OV-17 on 100–120 mesh Gas-Chrom Q. No values reported are corrected for efficiency, which was essentially 100 percent, and no interferences were detected in the solvent blanks run with each series of 10 samples.

Results

Sport fish collected from Indiana lakes and reservoirs during 1972–73 were contaminated with pesticide residues (Table 1). Several environmental factors significantly influenced the concentration of pesticide residues found in the fish. Of the seven pesticides monitored, only dieldrin, DDT, and DDE were detected above trace levels. The pesticide residue levels varied significantly among groups of samples, but these variations were correlated with the species of fish, season of the year, watershed land use, and turbidity and true color of the water. The correlations observed with turbidity and true color were positive in reservoirs and negative in natural lakes. However, no correlations existed between pesticide concentrations and any of the other limnological or physiological parameters measured.

Authors found a distinct difference in pesticide levels among different fish species from the same lake (Figure 2). Largemouth bass consistently had higher levels of pesticide residues than had species of sunfish (*Lepomis*). The levels in *Lepomis* were often undetectable. A reason for the relatively lower levels in sunfish was not apparent. However, Henderson et al. also observed this same species difference in Iowa reservoirs (*7*). Although bullheads consistently had measurable residue levels, they could not be obtained in sufficient numbers or from enough lakes to make a statistically valid analysis. Therefore, comparisons between sites and seasons were based solely on data from largemouth bass.

Dieldrin residue levels were relatively high in bass from the flood control reservoirs (Figure 3; Table 1) which drain large areas of corn cropland treated with aldrin (*9,16,17*). Conversely, dieldrin levels were undetectable in samples from Monroe Reservoir and several other smaller reservoirs in southern Indiana which have virtually no corn cropland in their watersheds (*9*). Dieldrin was also absent from Shafer Reservoir in northern Indiana. The watershed of Shafer was not treated with aldrin because it posed a contamination threat to the dairy industry in the basin (personal communication: F. T. Turpin, 1973, Entomology Department, Purdue University, West LaFayette, Ind. 47907).

Dieldrin was rarely detected in fish from natural lakes (Figure 3; Table 1) found only in the northern region of the State where fruit orchards predominate (Figure 1). However, fish from the natural lakes contained significantly higher levels of ΣDDT than did fish from reservoirs (Figure 4).

ΣDDT residue levels in the fish from the natural lakes were negatively correlated to the trophic state of the lake sampled (Figure 5). High ΣDDT residue levels were found in bass from clear, natural lakes which had low levels of turbidity and true color. Conversely, hyper-eutrophic lakes such as Sylvan and Cedar, which had high levels of turbidity and color primarily due to abundant planktonic algae, contained fish with low residue levels (Figure 4).

In contrast to the natural lakes, positive correlations were observed between the trophic state and dieldrin residues in fish from the flood control reservoirs, as demonstrated in Figure 6. (Data on turbidity and true color were not available for two corn crop area reservoirs.) High levels of diel-

TABLE 1. *Pesticide residues in largemouth bass from 20 lakes and reservoirs in Indiana*

Lakes	No.	Residues, ppm wet weight				Turbidity	Color
		Dieldrin		ΣDDT			
		Avg.	SD	Avg.	SD		
Bass	3	0.00	0.00	0.08	0.02	20	18
Cedar	4	0.02	0.014	0.05	0.024	20	25
Crooked	3	0.036	0.017	0.143	0.037	0.28	9
Maxinkukee	3	0.013	0.008	0.157	0.051	2.3	7
Olin	3	0.03	0.005	0.127	0.032	1.1	12
Oliver							
summer '72	2	0.025	0.005	0.112	0.018	1.3	7
fall '72	4	0.00	0.00	0.03	0.008	1.4	7
spring '73	3	0.02	0.015	0.04	0.015	4.5	—
Sylvan							
summer '72	2	0.00	0.00	0.03	0.01	10	25
fall '72	3	0.00	0.00	0.02	0.005	15	30
spring '73	3	0.01	0.00	0.03	0.02	7	—
Wawasee	4	0.01	0.00	0.10	0.04	8	8
Reservoirs							
Cataract	3	0.137	0.092	0.05	0.014	1.25	10
Dogwood	4	0.01	0.00	0.03	0.015	2.5	7
Eagle Creek	5	0.078	0.046	0.036	0.007	—	—
Huntington	2	0.14	0.00	0.035	0.01	44	45
Mississinewa	3	0.075	0.035	0.03	0.00	—	—
Monroe							
spring '72	5	0.00	0.00	0.054	0.027	2.0	8
summer '72	4	0.00	0.00	0.088	0.009	2	8
fall '72	3	0.00	0.00	0.05	0.014	12.3	9
spring '73	2	0.01	0.00	0.01	0.01	7	—
Raccoon	3	0.06	0.06	0.046	0.025	2	13
Salamonie							
summer '72	3	0.23	0.036	0.066	0.017	51	80
fall '72	3	0.14	0.095	0.04	0.017	30	90
winter '73	3	0.09	0.061	0.02	0.00	141	400
spring '73	3	0.23	0.236	0.06	0.018	32	—
Shafer	2	0.00	0.00	0.02	0.00	24.9	30
Sharpsnak	2	0.00	0.00	0.06	0.01	2.3	15
	3	0.01	0.005	0.05	0.015	2.6	15
	3	0.00	0.00	0.05	0.02	5.5	10

drin residue were found in bass from very turbid reservoirs with high true color values, typified by Salamonie Reservoir (Table 1, Figure 5). Correspondingly lower levels of dieldrin residues were consistent with lower levels of turbidity and true color in the reservoirs.

A strong seasonal trend was also observed in the fish sampled both from the natural lakes and the reservoirs (Figure 7). Pesticide levels were high in the summer, but declined during fall and winter.

Discussion

Sediment particles appear to influence significantly the concentration of pesticide contaminants absorbed by fish when residues reach a lake or reservoir. This was partly substantiated by the correlations observed between the dieldrin levels in fish from reservoirs versus the turbidity and true color levels of the water in which the fish lived (Figure 6). The level of pesticide contaminants found in the fish increased as the level of eroded sediment and washed-in debris increased.

A strong seasonal variation in residue concentrations was observed in Salamonie Reservoir (Figure 7). This is a flood control reservoir located in a watershed in northern Indiana which is about 75 percent farmland. Salamonie Reservoir and others more closely resemble wide places in a river than they do lakes because they are continuously flushing. Presumably, the spring rains washed soil particles and decaying plant matter from the cultivated fields, which received spring treatments of aldrin, into the reservoir, introducing large quantities of the recently applied contaminants. The actual concentration in the water would be a function of the amount of pesticide in the sediment and debris washed into the reservoir and the volume of water participating in the exchange (*6*). The fish would subsequently take up the contaminants in proportion to the concentration available in the water (*5*).

The contaminant concentration in the fish would then be expected to decline throughout the growing season for several reasons. Vegetative cover would reduce soil erosion

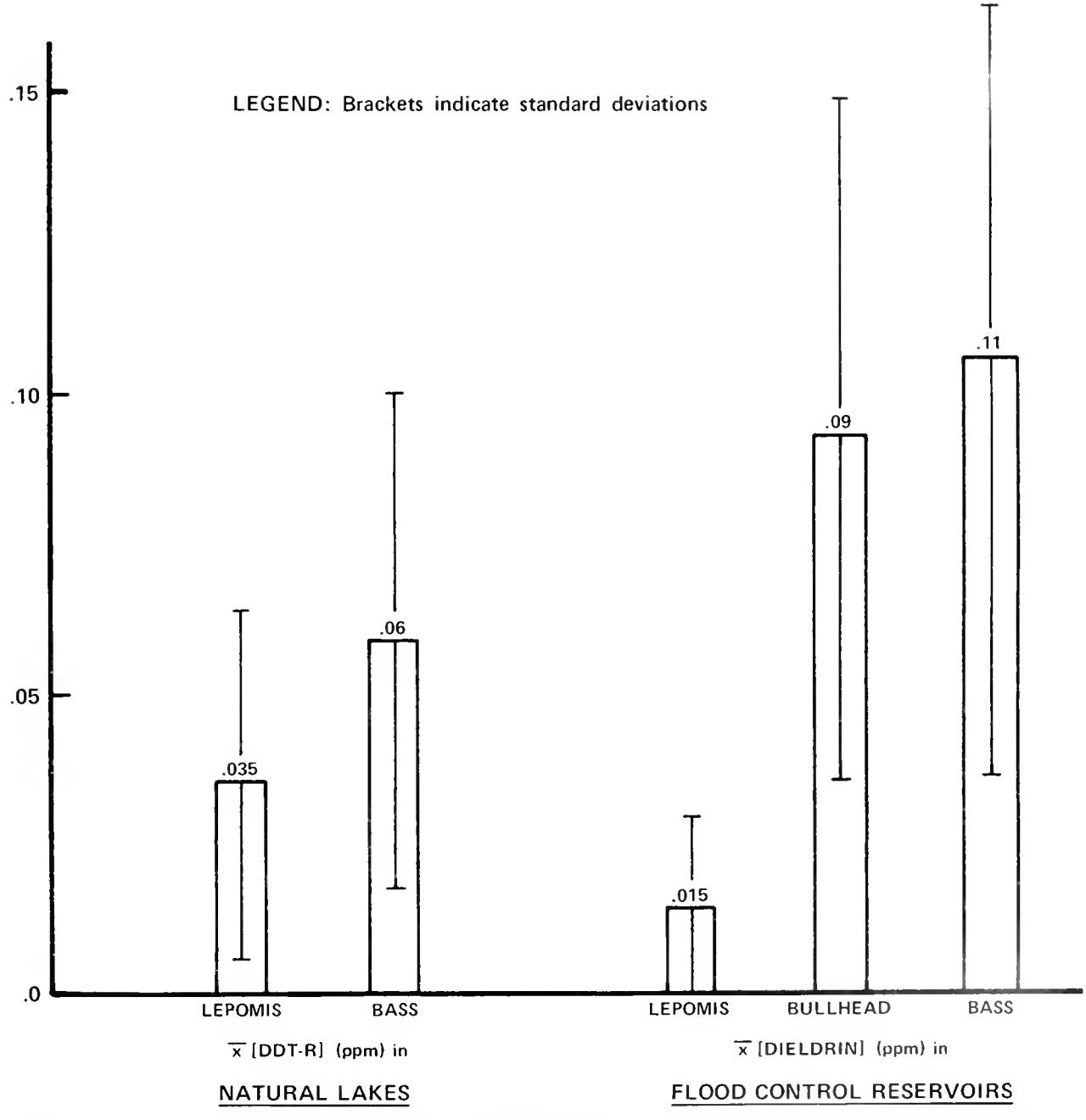

FIGURE 2. *Mean pesticide levels in three fish species combined from eight natural lakes and from six reservoirs in corn crop areas, summer 1972*

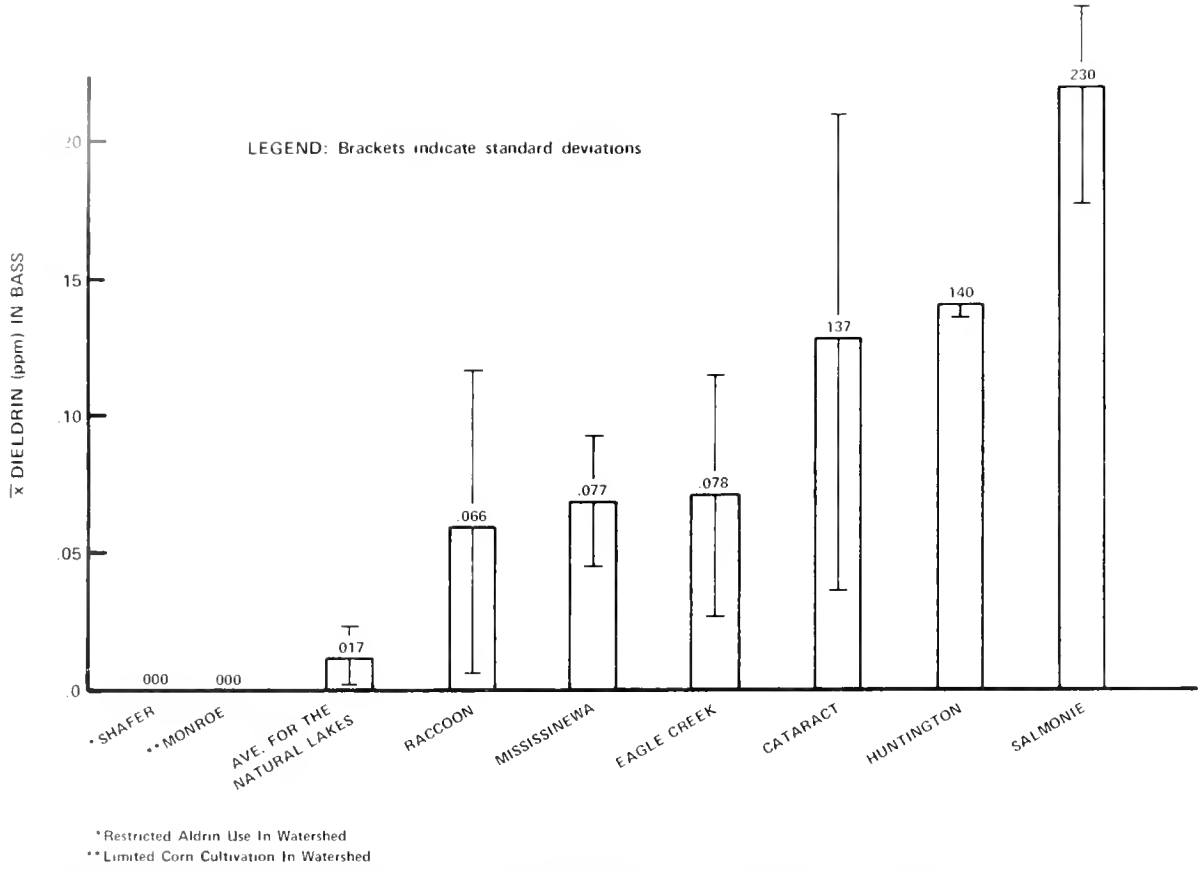

FIGURE 3. *Mean dieldrin levels in bass collected from six reservoirs in areas of heavy corn cultivation and aldrin use, summer 1972*

into the reservoirs. The soil particles washed into the reservoir would contain less residue. The concentration already in the reservoir would be continuously diluted by the flushing of the reservoir itself. The concentration in the fish would decline as the concentration in the water declined (5).

The data from the natural lakes sampled support the idea [illegible] and vegetative debris have a major influence [illegible] fish. In the natural lakes there was a negative correlation between the contaminant levels in fish and the associated turbidity and true color. The soil and plant particles definitely influenced residue levels in the lakes as well as in the reservoirs, but in the lakes, allochthonous particles reduced residue concentrations.

The watersheds of the natural lakes sampled are used extensively for orchard cultivation [illegible] no longer treated with DDT (15). Therefore, any soil and plant particles in runoff received by the lakes should not contain significant ΣDDT residues. Rather, any recently eroded sediment and [illegible] debris should adsorb residues remaining in the lake water from years past. Hence the fish would lose residues to the water, in an equilibrium shift, as the contaminant in the water was reduced. Thus as the amount of turbidity and true color increased, the concentration of residues in the fish decreased (Figure 5). Seasonal fluctuations of the pesticide levels in fish from the natural lakes were also consistent with this concept.

A definite seasonal variation was observed in Oliver Lake, a small natural lake surrounded by summer homes and limited agriculture. The ΣDDT levels in the fish in Oliver were reduced from summer to fall and appeared to increase in the early spring. As previously discussed, these variations would not be caused by seasonal introductions of DDT-laden sediment. Rather, with no new introductions of a pesticide and minimal erosion from the small watershed, this variation in a natural lake would be explained by spring turnover and subsequent sedimentation during thermal stratification.

Spring turnover would reintroduce residue-laden bottom sediment to the water column, thereby making the pesticide residues more available to the water and subsequently to the fish (5). Stratification would permit these sediments, plus any newly generated or introduced sediment from the watershed, to settle onto the lake bottom. The particles

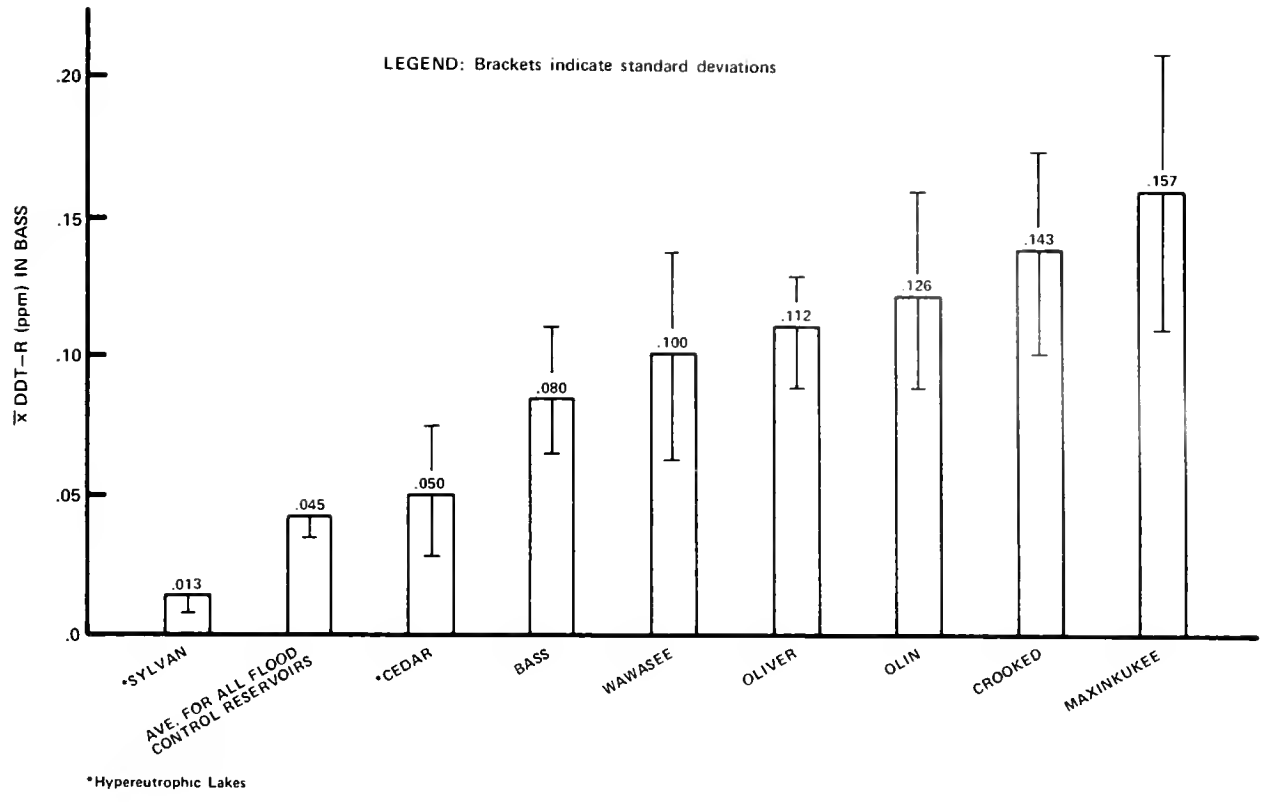

FIGURE 4. *Mean ΣDDT levels in bass collected from natural lakes in summer 1972*

would presumably adsorb residues from the water column which would subsequently reduce residue levels in the fish. Fall turnover probably would not have the same effect as the spring turnover because sedimented organic particles, especially algae, would not have decayed enough to release adsorbed pesticides. Also, the low fall/winter temperatures would promote adsorption rather than desorption (*6*).

The results which authors observed in both natural and humanmade lakes demonstrate that the concentrations of turbidity and true color significantly influence the level of pesticide residues in fish. The contrasting correlations observed between the parameters in natural lakes and reservoirs, and the seasonal variations observed in both, support this contention. Soil and decaying plant particles will cause pesticide residue levels in fish either to increase or decrease according to the amount of residue adsorbed to allocthonous particles and the residue concentration already in the water. The soil and plant particles apparently affect the residue concentration equilibrium between water and the fish.

Acknowledgments

Barbara Macy, a technician in the Purdue University Entomology Department, and Dr. Ronald Waybrant, presently with the Michigan Department of Natural Resources, were essential in training Mr. Vanderford in the chemical and analytical techniques used in this research. Figures were prepared by Michael C. Anderson, artist-illustrator, Control Data Corporation, Bloomington, Minn.

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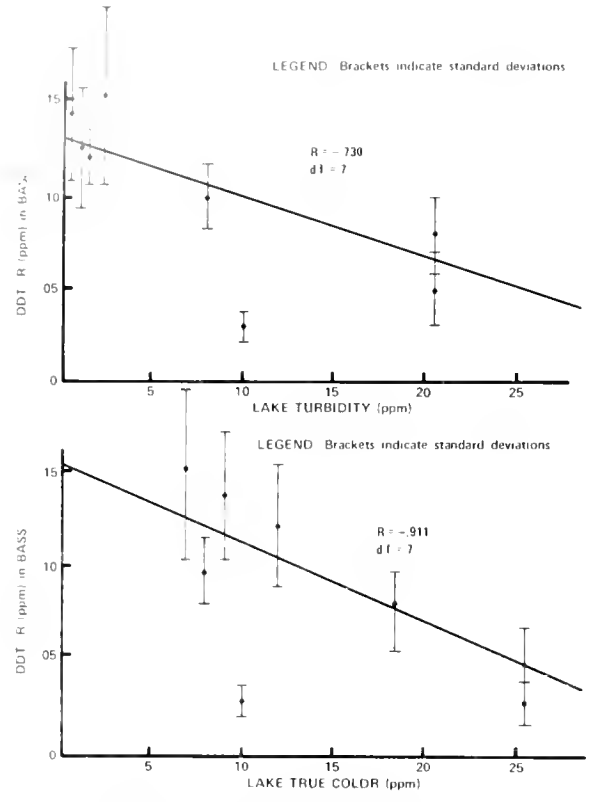

FIGURE 5. *Negative correlation between ΣDDT levels in bass from the natural lakes versus concentration of turbidity and true color in lakes at time of sampling*

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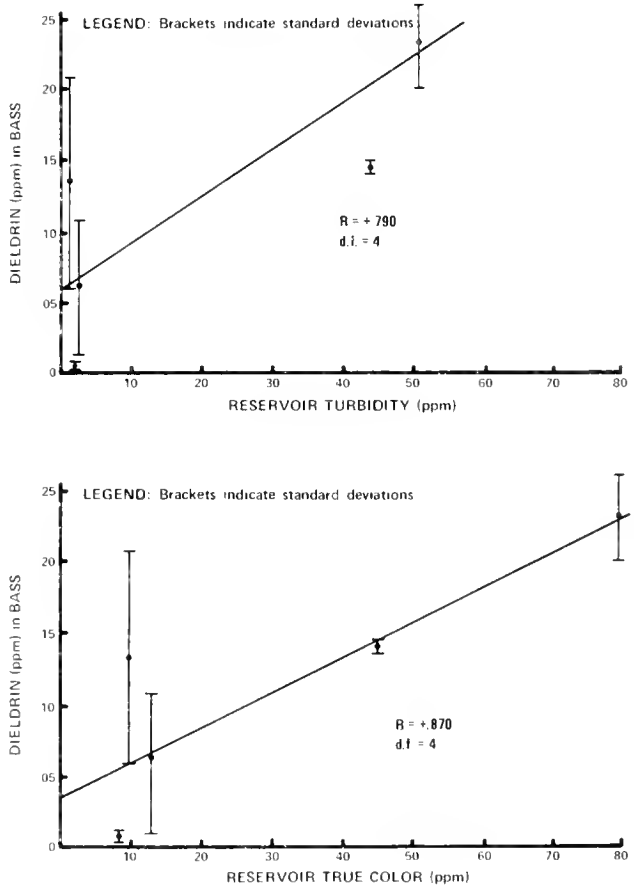

FIGURE 6. *Positive correlation between mean dieldrin levels in bass from Monroe Reservoir and four reservoirs in corn crop areas versus concentration of turbidity and true color in reservoirs at time of sampling.*

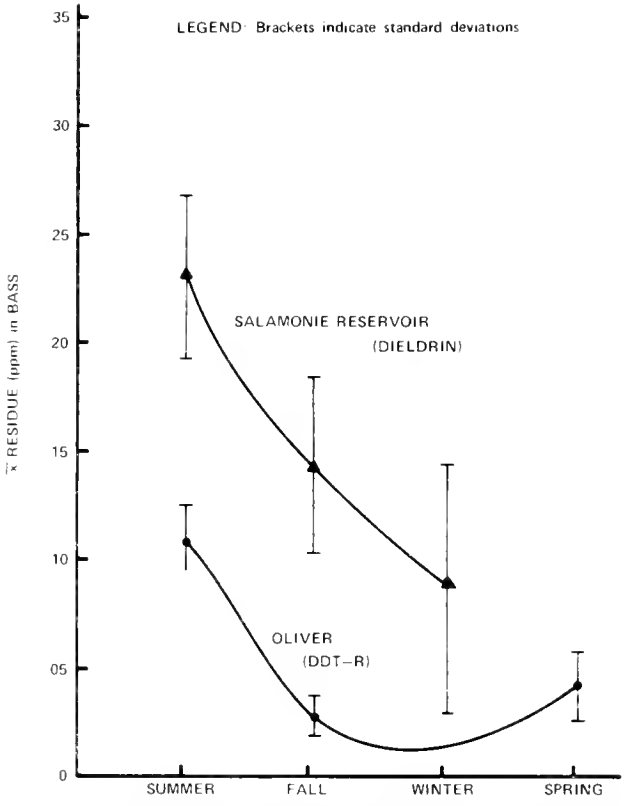

FIGURE 7. *Seasonal change in mean pesticide levels in bass from Oliver Lake and Salamonie Reservoir sampled seasonally, 1972–73*

GENERAL

Mirex Residues in Nontarget Organisms After Application of 10-5 Bait for Fire Ant Control, Northeast Florida—1972–74[1]

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ABSTRACT

The 10–5 bait formulation of mirex insecticide was applied to a 20,000-acre area of northeast Florida. For 24 months after application, samples of a wide variety of fauna were collected and analyzed for mirex content.

Insects accumulated mirex to the greatest extent in the first 6 months after application; most residues had decreased greatly by 12 months. Other invertebrates showed low mirex levels during the first 9 months after application and none after 12 months. Fish possessed low levels of insecticides for 9 months; amphibians showed mirex residues for 12 months after application. In general, reptiles had low levels throughout the 24-month period and mammals had higher levels, particularly in fat tissues. Birds consistently had low to moderate mirex levels. After a single application of 10-5 mirex bait, only relatively low levels of insecticide were detected in exposed fauna. After 24 months, mirex was found infrequently and at low levels.

Introduction

Mirex is the only insecticide approved for area-wide control of the red and black imported fire ants, *Solenopsis invicta* and *Solenopsis richteri*, in the southeastern United States. This chemical has been formulated and applied primarily as the 4X bait composed of 0.3 percent mirex, 14.7 percent soybean oil, and 85 percent corncob grits. Recently, a new 10-5 bait formulation containing 0.1 percent mirex has been described (*2*). This formulation was designed to make more of the toxicant available to the fire ant and leave less behind which might contaminate nontarget organisms and other elements of the environment.

Mirex applications have been shown to cause pesticide residue accumulations in the nontarget species (*1,3–10,12,13*). Most of these studies used the 4X bait formulations although a recent study by some authors of the present paper employed several bait formulations (*12*).

Applications of the 10-5 formulations have been as effective in controlling the fire ants as was the 4X formulation (*2*). No residue data in nontarget organisms have been reported after use of the 10–5 formulation.

This paper reports the results of a 2-year monitoring study of mirex in diverse animals after application of the new 10-5 bait formulation in the animals' habitat.

Methods

EXPERIMENTAL SITE

The experimental site consisted of approximately 20,000 acres (8000 ha.) in Duval and St. Johns Counties near Jacksonville, Fla. Based upon available records, no mirex had been previously applied to this area.

MIREX APPLICATION

Mirex was applied as a 0.1 percent bait consisting of 85 percent corncob grits, 14.9 percent once-refined soybean oil, and 0.1 percent mirex. It was prepared by Allied Chemical Corporation according to the procedures of Banks et al. (*2*).

The bait was applied at a rate of 1.0 lb/acre (1.12 kg/ha.) using a single-engine aircraft at an altitude of 150 feet (45.7 m). The aircraft was guided by kytoons and the bait was distributed using a gravity-fed Piper Pawnee dispersal system. Personnel of the Division of Plant Industry,

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Florida Department of Agriculture and Consumer Services, supervised bait application.

SAMPLE COLLECTION

The sample collection dates are listed in Table 1. Invertebrates and small vertebrates were collected in pitfall traps (*11*) which contained small open jars of technical chloropyrifos as a killing agent. Twenty traps were set in a grid pattern at each of eight locations, two in each of four types of habitat: pasture, pine forest, hardwood forest, and adjacent to swamps. Hand collections were made to supplement pitfall trap collections whenever possible.

Aquatic invertebrates, amphibians, and reptiles were collected by hand. Fish were stunned with an electric shocker or rotenone prior to collection. Mammals and birds were trapped, netted, or shot.

Insectivorous, omnivorous, and predatory brids and mammals were monitored. Certain species were chosen because of their availability and restricted movement, because au-

TABLE 1. *Dates of mirex application and sampling of nontarget organisms, northeast Florida—1972–74*

Pretreatment samples	May 22–June 7, 1972
Bait applied	July 11–12
1-month posttreatment samples	August 4–11
3-month posttreatment samples	October 6–17
6-month posttreatment samples	January 8–29, 1973
9-month posttreatment samples	April 9–30
1-year posttreatment samples	July 9–23
18-month posttreatment samples	January 14–18, 1974
2-year post-treatment samples	August 23–30

TABLE 2. *Animals analyzed for mirex residues after single treatment of 10-5 bait, Dee Dot Ranch, Fla.—1972–74*

Scientific Name	Common Name
Insects	
Pictonemobius ambitiosus	ground cricket
Gryllus rubens	southern field cricket
G. firmus	sand cricket
G. fultoni	southern wood cricket
Miogryllus verticallis	strip-headed cricket
Scapteriscus vicinus	changa
S. acletus	southern mole cricket
Ceuthophilus spp.	camel cricket
Parcoblatta spp.	wood cockroach
Cariblatta lutea	small yellow cockroach
Eurycotis floridana	stinking cockroach
Pycnascelus surinamensis	Surinam cockroach
Gonatista grisea	grizzled mantis
Euborellia annulipes	ringlegged earwig
Labidura riparia	riparian earwig
Marava pulchella	handsome earwig
Prosapia bicincta	twolined spittlebug
Plecia nearctica	lovebug
Spiders	
Latrodectus mactans	black widow spider
Crustaceans	
Uca sp.	fiddler crabs
Mollusks	
Crassostrea virginica	American oyster
----	(mixed unidentified freshwater snails)
Amphibians	
Bufo terrestris	southern toad
Bufo quercicus	oak toad
Rana heckscheri	river frog
Rana sphenacephala	leopard frog
Rana catesbeiana	bull frog
Rana areolata	gopher frog
Rana sp	tadpoles
Gastrophryne carolinensis	narrow-mouth toad
Scaphiopus holbrooki	spadefoot toad
Hyla cinerea	greentree frog
Hyla femoralis	pinewoods tree frog
Reptiles	
Alligator mississippiensis	American alligator
Trionyx ferox	soft-shelled turtle
Kinasternan subrubrum	common mud turtle
Sceloporus undulatus	fence lizard
Scincella laterale	brown skink
Cnemidophorus sexlineatus	six-lined racerunner
Ophisaurus	legless glass lizard
Coluber constrictor priapus	southern black racer
Thamnophis sirtalis	garter snake
Thamnaphis sauritus sackeni	ribbon snake
Worms	
----	mixed unidentified earthworms
Fish	
Gambusia affinis	eastern mosquito fish
Lepomis macrochirus	bluegill
Lepomis microlophus	redear sunfish
Lepomis gulosus	warmouth
Micropterus salmoides	largemouth bass
Notemigonus crysoleucas	golden shiner
Erimyzon sucetta	lake chubsucker
Anguilla rostrata	American eel
Ictalurus nebulosus marmoratus	brown bullhead
Lepisosteus platostomus	shortnose gar
Mammals	
Didelphis virginiana	opossum
Procyon lotor	raccoon
Dasypus novemcinctus	armadillo
Lasiurus borealis	red bat
Nycticeius humeralis	evening bat
Sylvilagus palustris	marsh rabbit
Cryptotis parva	least shrew
Birds	
Anhinga anhinga	anhinga
Buteo jamaicensis	red-tailed hawk
Colinus virginianus	bobwhite
Gallinula chloropus	common gallinule
Melanerpes carolinus	red-bellied woodpecker
Sayornis phoebe	eastern phoebe
Cyanocitta cristata	blue jay
Corvus brachyrhynchos	common crow
Parus carolinensis	carolina chickadee
Sitta pusilla	brown-headed nuthatch
Troglodytes aedon	house wren
Turdus migratorius	robin
Catharus guttata	hermit thrush
Sialia sialis	eastern bluebird
Dendroica pinus	pine warbler
Dondroica coronata	myrtle warbler
Dendroica palmarum	palm warbler
Setophaga ruticilla	American redstart
Agelaius phoeniceus	red-winged blackbird
Quisculus quiscula	common grackle
Pipilo erythrophthalmus	rufous-sided towhee
Melospiza georgiana	swamp sparrow
Melospiza melodia	song sparrow
Spizella passerinu	chipping sparrow
Spizella arborea	tree sparrow
Passerculus sandwichensis	savannah sparrow
Ammodramus henslowii	Henslow's sparrow
Amiophila aestivalis	Bachman's sparrow

thors wanted a cross section of the fauna. Stomach contents of some birds and mammals were analyzed for an indication of dietary habits.

A complete listing of animal species collected is in Table 2.

MIREX ANALYSIS

Sample preparation, extraction, extract cleanup, mirex identification, and quantification have been described by Wojcik et al. (*12*). Mirex residues, reported except as noted in ppm fresh-weight (as the sample was received for analysis), were not corrected for recoveries less than 100 percent.

Results and Discussion

Results are often presented as values measured in composite samples. In Table 3, for example, at the one-month sampling of adult ground crickets, a composite of eight individuals collected in one location had no detectable mirex and another composite of three individuals possessed 0.09 ppm. Occasionally, very high levels were detected in some specimens: for example, one earwig collected at the 3-month sampling listed in Table 4. These values are presented for informational purposes and should not be considered significant.

Mirex residues detected in crickets are shown in Table 3. None of the pretreatment samples contained detectable mirex. By 1 month after application, 24 percent of the samples did possess low levels (0.02–0.37 ppm), but after 3 months the percentage of samples showing mirex was the highest of the entire experimental period: 33 percent. Certain species sampled at 3 and 6 months possessed the highest mirex levels detected during the 24-month period. By 6 months after treatment, the proportion of samples containing mirex had dropped to 15 percent and remained relatively constant through the remainder of the 24-month collection period. The mirex levels detected after 6 months

TABLE 3. *Mirex residues in crickets from Dee Dot Ranch after single treatment of 10-5 bait, 1972–74*

	Residues, ppm Fresh Weight [1]					
Pretreatment	Posttreatment					
	1 month	3 months	6 months	9 months	1 year	2 years
Adult Ground Crickets						
ND (104)	ND (8) 0.09 (3)	ND (25)	ND (13) 0.49 (3) 3.52 (3)	ND (17)	ND (167) 0.04 (4)	ND (23) 0.74 (5)
Nymphal Ground Crickets						
ND (36)	ND (1)	ND (24) 1.54 (7) 3.22 (3)	ND (6)	ND (9)	ND (16) 1.38 (8)	
Adult Southern Field Crickets						
ND (16)	ND (46)	ND (9)	ND (12)	ND (13)	ND (56)	ND (29)
Adult Sand Crickets						
ND (3)	0.09 (2)	ND (9)	ND (1)		ND (1)	ND (6)
Adult Southern Wood Cricket						
ND (1)		ND (4)	ND (1)		ND (1)	ND (2)
Adult Stripe-headed Crickets						
ND (19)	ND (7) 0.13 (3) 0.32 (2) 0.37 (1)				ND (24) 0.02 (4)	ND (1)
Nymphal Stripe-headed Crickets						
ND (23)	ND (1) 0.02 (4)	ND (2) 0.11 (17) 0.61 (8)		ND (4)	ND (1)	ND (1)
Adult and Nymphal Camel Crickets						
ND (27)	ND (2) 0.03 (2)	ND (4)		ND (3) 0.13 (4)	ND (9) 0.03 (2)	
Adult and Nymphal Southern Mole Cricket						
ND (2)						ND (1)
Nymphal Camel Crickets						
ND (9)	0.08 (1) 0.12 (2)	ND (3)	ND (2)	ND (3)	ND (1) 0.02 (1)	ND (3) 0.03 (3)

NOTE: ND = no residues detected at 0.01 ppm level.
Figures in parentheses represent number of specimens in pooled samples.
[1] Fresh weight = as received for analysis. Specimens were not dried.

were substantially reduced, ranging from 0.03 to 0.74 ppm at the 24-month sampling interval. A large percentage of the residues detected were in the ground cricket (*Pictonemobium ambitiosus*) and all the high mirex levels detected were in this species.

Table 4 presents insecticide residues in miscellaneous insects. No mirex was detected in pretreatment samples. Sixty-one percent of the 1-month posttreatment samples contained mirex; one individual had 4.10 ppm. At 3 months, one earwig possessed 19.79 ppm. Sixty other samples had no detectable residue. At 6 months and 12 months 54 percent and 62 percent of the samples had residues: 0.04–7.78 ppm at 6 months and 0.02–1.14 ppm at 12 months. The 7.78 ppm was in five earwigs. No mirex was detected in the 9-month samples. At 24 months, 16 percent of the miscellaneous insects contained mirex at levels ranging from 0.58 to 0.79 ppm.

Residue data from a few miscellaneous invertebrates appear in Table 5. Pretreatment fiddler crabs (*Uca* sp.), which constituted 33 percent of the miscellaneous invertebrate samples, contained 0.33 ppm mirex. Posttreatment percentages of samples containing mirex were 30, 10, 30, 2, 17, and 0 percent for 1, 3, 6, 9, 12, and 24 month samples, respectively. Spiders consistently had detectable mirex levels which ranged from 1.30 ppm in a 1-month sample to 0.06 ppm in a 9-month sample.

Mirex levels in fish are listed in Table 6. Forty-five mosquito fish, 54 percent of the entire fish sample, contained an average of 0.33 ppm mirex before application of the insecticide. One month after application 27 mosquito fish, 44 percent of the 1-month sample, contained an average of 0.08 ppm mirex. At 3, 6, and 9 months, the proportion of samples containing any insecticide ranged from 3 to 13 percent and the levels of mirex, when found, were

TABLE 4. *Mirex residues in insects from Dee Dot Ranch after single treatment of 10-5 bait, 1972–74*

	Residues, ppm Fresh Weight [1]					
Pretreatment	Posttreatment					
	1 month	3 months	6 months	9 months	1 year	2 years
Adult and Nymphal Wood Cockroaches						
ND (8)	ND (5) 4.10 (1)	ND (12)	ND (7) 0.22 (3) 0.04 (22)	ND (20)	ND (6) 0.02 (5) 0.02 (9)	ND (6) 0.58 (5)
Adult and Nymphal Small Yellow Cockroaches						
ND (2)	ND (1)	ND (1)	ND (1)	ND (6)	ND (6)	ND (1)
Adult Dark Wood Cockroaches						
				ND (2)	0.39 (1)	
Adult Stinking Cockroach						
					0.51 (1)	
Adult Surinam Cockroach						
					ND (1)	
Adult Grizzled Mantis						
					0.06 (1)	
Adult and Nymphal Ringlegged Earwigs						
ND (15)	0.48 (10)	ND (41) 19.79 (1)	ND (7)	ND (25)		0.79 (4)
Adult Riparian Earwigs						
					ND (1)	ND (1)
Adult Handsome Earwigs						
			ND (11) 7.78 (5)			
Adult Twolined Spittlebugs						
ND (3)	ND (1)	ND (6)			ND (4) 0.64 (8) 1.14 (9)	ND (1)
Adult Lovebugs						
						ND (39)

NOTE: ND = no residues detected at 0.01 ppm level
Figures in parentheses represent number of specimens in pooled sample
[1] See footnote, Table 3

TABLE 5. *Mirex residues in miscellaneous invertebrates from Dee Dot Ranch after single treatment of 10-5 bait, 1972–74*

Pretreatment	Residues, ppm Fresh Weight [1]					
	Posttreatment					
	1 month	3 months	6 months	9 months	1 year	2 years
Black Widow Spiders						
	1.30 (2)	ND (2) 0.16 (3) 0.49 (7)	0.17 (6)	ND (3) 0.06 (2)		ND (1)
Oysters						
ND (15)	ND (15)	ND (20)	ND (25)	ND (25)	ND (25)	ND (25)
Fiddler Crabs						
0.03 (23)	ND (25)	ND (19)		ND (25)	ND (25)	ND (25)
Miscellaneous Fresh Water Snails						
ND (15)		ND (20)		ND (25)	ND (25)	ND (26)
Earthworms						
ND (16)	0.07 (15)	ND (25)	ND (25)	ND (26)	0.02 (15)	ND (25)

NOTE: ND = no residues detected at 0.01 ppm level.
Figures in parentheses represent number of specimens in pooled sample.
[1] See footnote, Table 3.

TABLE 6. *Mirex residues in fish from Dee Dot Ranch after single treatment of 10-5 bait, 1972–74*

Pretreatment	Residues, ppm Fresh Weight [1]						
	Posttreatment						
	1 month	3 months	6 months	9 months	1 year	18 months	2 years
Eastern Mosquito Fish							
0.33 (45)	0.08 (27)	ND (45)	ND (35)	ND (39)	ND (50)	ND (46)	ND (50)
Bluegills							
ND (16)	ND (12)	ND (40)	ND (30)	ND (35)	ND (17)	ND (25)	ND (35)
Redear Sunfish							
ND (3)	ND (2)	ND (9) 0.21 (1)	ND (25)	ND (20)	ND (7)	ND (2)	ND (14)
Warmouth							
ND (1)	ND (3)	ND (4) 0.25 (1)	ND (7)	0.01 (4)			ND (5)
Largemouth Bass							
ND (6)	ND (6)	ND (1)	ND (27)	ND (35)	ND (23)	ND (15)	ND (30)
Golden Shiners							
	ND (3)	ND (4)	ND (11)	ND (9)	ND (2)	ND (7)	ND (30)
Lake Chubsuckers							
			ND (3)	ND (10)		ND (9)	ND (10)
Eels							
ND (8)		ND (2) 0.07 (1)	0.19 (6)	0.13 (18)	ND (16)	ND (14)	
Brown Bullhead							
ND (5)	0.02 (9)				ND (2)	ND (9)	ND (5)
Shortnosed Gars							
							ND (3)

NOTE: ND = no residues detected at 0.01 ppm level.
Figures in parentheses represent numbers of specimens in pooled sample.
[1] See footnote, Table 3.

0.01–0.25 ppm. After the 9-month sample, no insecticide was detected in fish.

Mirex residues in amphibians are shown in Table 7. Four oak toads contained an average of 0.42 ppm mirex before treatment. Only one other sample in this group had any trace of the insecticide. One month after treatment, 80 percent of the amphibians contained mirex ranging from 0.02 to 0.78 ppm. In general, the percentage of samples containing mirex decreased with time and the levels followed a similar pattern. Mirex was not detected in amphibians 24 months after application.

Mirex levels found in reptiles are listed in Table 8. Although few individuals were sampled, some observations can be made. Only one of six pretreatment samples possessed detectable mirex: 0.01 ppm. From 1 through 9 months after application, more than 85 percent of the samples possessed mirex ranging generally from 0.02 to 0.78 ppm; one alligator fat sample contained 1.19 ppm. At 24 months, the incidence of detectable mirex had dropped to 20 percent of the samples and the levels ranged from 0.01 to 0.06 ppm.

Mirex residues in mammals are shown in Table 9. As with reptiles, only a few individuals were sampled. Prior to treatment, 44 percent of the individuals contained low mirex levels, all below 0.07 ppm. All but one sample contained detectable mirex after application. Insecticide levels varied with time and animal tissue and had an overall range

TABLE 7. *Mirex residues in amphibians from Dee Dot Ranch after single treatment of 10-5 bait, 1972–74*

	Residues, ppm Fresh Weight [1]					
Pretreatment	Posttreatment					
	1 month	3 months	6 months	9 months	1 year	2 years
Southern Toads						
ND (4) 0.03 (1)			ND (1)	0.09 (1)		
Oak Toads						
ND (7) 0.42 (4)	0.78 (4)	0.11 (11)	1.01 (1) 0.20 (1)	0.15 (1)	0.04 (3)	ND (3)
River Frogs						
ND (7)	0.28 (3) 0.02 (12)	ND (5)	ND (2)	ND (5) [a] ND (12) [b] 0.26 (4) [b]	ND (6)	
Leopard Frogs						
ND (5)	ND (1) 0.04 (1)	ND (1) 0.13 (1)		0.01 (3)		
Bull Frogs						
ND (5)	0.04 (8)	ND (3) 0.02 (1)		ND (7)		ND (4)
Gopher Frogs						
	0.14 (4)					
Rana sp Tadpoles						
	ND (7)	ND (12)	ND (10)			
Narrow-mouth Toads						
ND (38)	0.52 (3)	0.09 (3)	0.17 (3)	0.01 (1)	0.06 (1)	ND (10)
Spadefoot Toads						
ND (5)	ND (1)		0.02 (1) 0.02 (1)		0.05 (3)	ND (1)
Green Tree Frog						
	ND (1)	ND (1)		ND (1)		
Pinewoods Tree Frog						
	0.08 (2)		ND (1)			

NOTE: ND = no residues detected at 0.01 ppm level.
Figures in parentheses represent numbers of specimens in pooled samples.
[a] mature adults
[b] recently metamorphosed adults
[1] See footnote, Table 3.

TABLE 8. *Mirex residues in reptiles from Dee Dot Ranch after single treatment of 10-5 bait, 1972–1974*

	Residues, ppm Fresh Weight [1]							
	Pretreatment	Posttreatment						
		1 month	3 months	6 months	9 months	1 year	18 months	2 years
Alligator								
Brain	ND [c]					ND [c]		0.04 [c]
Fat	ND [c]	0.27 [c]	1.19 [c]					0.06 [c]
Liver	ND [c]	ND [c]	ND [c]			ND [c]		0.04 [c]
Muscle	ND [c]	ND [c]	0.02 [c]			ND [c]		0.04 [c]
Stomach Content	0.01 [c]	0.04 [c]	0.13 [c]					
Whole		0.03 [d]						
Soft Shell Turtle								
Fat & Eggs	ND (1)				ND (1)			ND [c]
Whole								0.01 [c]
Mud Turtle								
			ND (1)					
			0.03 (1)					
Fence Lizards								
	ND (1)	0.05 (1)	0.19 (1)	0.12 (1)	0.08 (4)	0.04 (1)	0.30 (3)	ND (7)
			0.51 (1)	0.18 (1)				
Brown Skink								
					0.11 (2)			
Slined Race Runner								
								ND (1)
Legless Glass Lizard								
	ND (1)				ND (1)			
Black Snake								
	ND (1)				0.06 (1)		0.04 (1)	
Garter Snake								
	ND (1)							
Ribbon Snake								
					0.04 (1)			

NOTE: ND = no residues detected at 0.01 ppm level
Figures in parentheses represent numbers of specimens in pooled samples
Residues followed by different letters represent subsamples of different specimens from a sample collection period
[1] See footnote, Table 3

TABLE 9. *Mirex residues in mammals from Dee Dot Ranch after single treatment of 10-5 bait, 1972–74*

	Residues, ppm Fresh Weight [1]							
	Pretreatment	Posttreatment						
		1 month	3 months	6 months	9 months	1 year	18 months	2 years
Opossum								
Brain	ND (1)	0.15 (1)		0.05 (1)			ND (1)	
Fat	ND (1)	1.72 (1)		3.35 (1)			0.55 (1)	
Liver	ND (1)	0.36 (1)		0.35 (1)			0.05 (1)	
Muscle	ND (1)	0.53 (1)		0.06 (1)			0.25 (1)	
Stomach Contents	0.03 (1)			ND (1)			0.02 (1)	
Raccoon								
Brain	ND (1)	0.13 (2)		ND (1)		ND (1)		ND (1)
Fat		2.24 (2)		ND (1)		0.31 (1)		0.06 (1)
Liver	0.07 (1)	0.25 (2)		0.03 (1)		ND (1)		ND (1)
Muscle	0.04 (1)	0.05 (2)		ND (1)		ND (1)		ND (1)
Stomach Contents	ND (1)	0.01 (2)		ND (1)				

(Continued next page)

TABLE 9 (cont'd.). *Mirex residues in mammals from Dee Dot Ranch after single treatment of 10-5 bait, 1972–1974*

	Residues, ppm Fresh Weight[1]						
	Pretreatment	Posttreatment					
		1 month	3 months	6 months	9 months	1 year	18 months	2 years
Armadillo								
Brain	ND (2)	0.20 (4)	0.17 (2)	0.01 (8)	0.10 (5)	ND (1)	ND (3)	ND (1)
Fat	ND (2)	1.10 (4)	0.47 (2)	1.68 (8)	0.08 (4)	0.67 (1)	0.52 (3)	ND (1)
Liver	ND (2)	0.59 (4)	0.13 (2)	0.24 (8)	1.21 (5)	ND (1)	0.04 (3)	0.09 (1)
Muscle	ND (2)	0.02 (4)	0.01 (2)	0.02 (8)	0.04 (5)	ND (1)	ND (3)	ND (1)
Stomach Contents	0.01 (2)	0.01 (3)		ND (8)			ND (3)	
Red Bat	ND (2)							
Evening Bat	ND (2)							
Evening Bat						ND (1)		
Evening Bat								0.09 (2)
Marsh Rabit								
Brain	ND (1)							
Liver	ND (1)							
Muscle	ND (1)							
Least Shrew				1.29 (1)				

NOTE: ND = no residues detected at 0.01 ppm level
Figures in parentheses represent numbers of specimens analyzed to obtain the average residues given
[1] See footnote, Table 3

of 0.01–3.35 ppm. In general, the highest levels were detected in fat tissues from 1 to 6 months after application. From 12 to 18 months after treatment the mirex levels decreased substantially, ranging from 0.02 to 0.67 ppm. At 24 months, the levels were low: 0.06–0.09 ppm.

Mirex levels found in birds are presented in Table 10. Sixty-two percent of the pretreatment samples contained mirex ranging from 0.01 to 0.20 ppm except for one sample which had 1.05 ppm. During the sample collection interval of 1 through 12 months after treatment, 94 percent of the birds contained mirex. The highest levels, approximately 10 ppm, were generally detected in fat tissues 1 to 6 months after mirex application. From 9 to 24 months, the pesticide levels generally decreased, reaching levels similar to those found before mirex application.

Insectivorous species such as armadillos, bluebirds, pine warblers, brown-headed nuthatches, red-bellied woodpeckers, and Bachman's sparrows showed the most dramatic accumulation of mirex. These species had lost most of their mirex burden by the 18-month collection. Omnivorous species such as crows, blue jays, raccoons, and opossums showed a less dramatic accumulation of mirex. Quail and common gallinule, which are mostly herbivorous, showed little accumulation of mirex. Carnivorous species, best represented here by the anhinga, demonstrated a surge in mirex accumulation during the middle collection period, 6 months to 1 year.

Examining the data from a broader viewpoint, one may reach some conclusions. The highest mirex levels were detected within 6 months of application. In general, insects and other invertebrates had low or nondetectable mirex levels 24 months after treatment. Crickets (Table 3), cockroaches, and earwigs (Table 4) seemed to be exceptions to this trend, still possessing detectable insecticide for the entire period, even though this occurred in a low percentage of samples. This is surprising, considering that most of these organisms were not present during bait application when the greatest amount of mirex was available. These organisms are scavengers, however, and must have been exposed to the insecticide some time after application.

The highest mirex levels were detected in birds, except for an earwig reported in Table 4. In most instances the mirex was highest in the fatty tissues rather than in the liver or stomach.

The mammals and birds consistently had a high percentage of individuals with mirex residues. This could be attributed to the fact that they are insectivors and omnivores, and to their place on the food chain and their relatively long life spans.

Acknowledgments

The authors are grateful to J. E. Davis for permitting use of his Dee Dot Ranch for this study. We also thank D. M.

TABLE 10. *Mirex residues in birds from Dee Dot Ranch after single treatment of 10-5 bait, 1972–1974*

	Residues, ppm Fresh Weight [1]						
	Pretreatment	Posttreatment					
		1 month	3 months	6 months	9 months	1 year	18 months	2 years
Anhinga (Piscivorous exclusively)								
Brain	0 03 (3)	ND (3)	ND (1)	0 03 (3)	0 06 (1)	0 88 (1)	ND (1)	0.01 (2)
Fat	0 06 (3)	ND (3)	0 47 (1)	4 98 (2)	1 90 (1)	2.25 (1)	0 03 (1)	ND (2)
Liver	0 06 (3)	ND (3)	0 11 (1)	0 18 (3)	0 23 (1)	0 55 (1)	ND (1)	0 01 (2)
Muscle	0 02 (3)	ND (3)	ND (1)	0 15 (3)	0.19 (1)	0 75 (1)	ND (1)	0.03 (2)
Stomach Content	0 01 (3)	ND (3)		ND (3)				
Red-tailed Hawk (Migratory)								
Brain	0 07 (1)	ND (1)						
Fat	0 01 (1)	0 82 (1)						
Liver	ND (1)	0 02 (1)						
Muscle	0 02 (1)	0 32 (1)						
Stomach Content		ND (1)						
Bobwhite Quail (Herbivorous)								
Brain	ND (3)	0 15 (1)		ND (2)				
Fat	ND (3)							
Liver	ND (3)	0 10 (1)		0 04 (2)				
Muscle	ND (3)	ND (1)		ND (2)				
Stomach Content	ND (3)			ND (2)				
Common Gallinule (adult)								
Brain	ND (2)	ND (2)		0 06 (1)				
Fat	ND (1)	0 29 (2)		0 02 (1)				
Liver	ND (2)	ND (2)		ND (1)				
Muscle	ND (2)	0 03 (2)		ND (1)				
Stomach Content	ND (1)	ND (2)		ND (1)				
Nestling whole	ND (2)	0 11 (1)						
Stomach Content	ND (2)	ND (1)						
Red-Bellied Woodpecker								
Whole	ND (2)	0 21 (2)		0.12 (2)	0 23 (2)	0 11 (2)	0 01 (2)	0 02 (2)
Stomach Content	0 02 (3)	0 01 (2)		0 53 (2)			ND (2)	
Phoebe								
Whole				1 31 (2)				
Stomach Content				0 07 (2)				
Blue Jay								
Whole	ND (3)	0 20 (1)	0 17 (3)	0 10 (6)	0 05 (2)	0 06 (2)	ND (1)	0 04 (3)
Stomach Content	ND (3)	ND (1)					ND (1)	
Common Crow								
Brain		0.18 (2)	0 21 (1)	0.06 (1)	0 35 (1)	ND (1)	0 03 (1)	
Fat	0 05 (1)	0 39 (2)	10 72 (1)			0.39 (1)	0 18 (1)	
Liver	ND (1)	0 39 (2)	0 65 (1)	0 16 (1)	0 58 (1)	0 44 (1)	0 06 (1)	
Muscle	ND (1)	0 15 (2)	0 32 (1)	0 09 (1)	0 20 (1)	ND (1)	0 06 (1)	
Stomach Contents	ND (1)	0.15 (2)		0 09 (1)				
Carolina Chickadee								
Whole	ND (3)	0 20 (3)	0 18 (2)	0 11 (1)	0 09 (3)	0 12 (2)	0 03 (2)	
Stomach Content	ND (3)	ND (2)		ND (1)				
Brown-headed Nuthatch								
Whole	ND (3)	0 96 (3)	0 47 (2)	0 46 (3)	0 24 (3)	0 67 (3)	0 10 (2)	0.03 (4)
Stomach Content	0 20 (3)	0 01 (3)		0 01 (3)			ND (2)	ND (4)
House Wren								
Whole				4 05 (2)				
Stomach Content				0 55 (2)				
Robin								
Whole				0 08 (1)			ND (2)	
Stomach Content				0 05 (1)				
Hermit Thrush								
Whole				1 15 (1)				
Stomach Content				0 14 (1)				
Bluebird								
Whole	ND (3)	8 27 (1)	2 40 (1)	2 34 (2)	3 66 (3)		0 11 (3)	0 21 (2)
Stomach Content	0 01 (3)	1 31 (3)		1 36 (2)			ND (3)	

(Continued next page)

TABLE 10 (cont'd.). *Mirex residues in birds from Dee Dot Ranch after single treatment of 10-5 bait, 1972–1974*

	Residues, ppm Fresh Weight[1]						
	Pretreatment	Posttreatment					
		1 month	3 months	6 months	9 months	1 year	18 months	2 years
Pine Warbler								
Whole	ND (4)	2 57 (7)	0 45 (3)	0 49 (3)	0 19 (2)	0 05 (3)	0.01 (4)	0 02 (1)
Stomach Content	1.05 (4)	0 07 (7)		0 29 (3)				
Myrtle Warbler								
Whole				ND (1)				
Stomach Content				ND (1)				
Palm Warbler								
Whole				0.33 (1)				
Stomach Content				ND (1)				
American Redstart (Spring & Fall Migrant)								
Whole			0 81 (1)					
Stomach Content								
Red-winged Blackbird								
Whole		0 48 (2)		ND (1)	0 05 (3)	ND (1)	0 03 (2)	0 02 (5)
Stomach Content		0 02 (2)		ND (1)			0 01 (2)	
Common Grackle								
Whole	ND (3)					0 68 (2)		
Stomach Content	ND (2)			0 16 (1)				
Brain				0.12				
Fat				10 38				
Liver				0 07				
Muscle				0 14				
Rufous-sided Towee								
Whole							ND (1)	
Stomach Content							ND (1)	
Swamp Sparrow								
Whole				0 11 (2)				
Stomach Content				ND (2)				
Song Sparrow								
Whole				ND (1)				
Stomach Content				ND (1)				
Chipping Sparrow								
Whole				0 08 (2)				
Stomach Content				ND (2)				
Tree Sparrow								
Whole				0 15 (1)				
Stomach Content				0 15 (1)				
Savanna Sparrow								
Whole				ND (1)				
Stomach Content				ND (1)				
Bachman's Sparrow								
Whole	ND (4)	3 65 (5)	1 17 (3)		0 70 (8)	1 05 (3)	0 03 (3)	0 30 (3)
Stomach Content	0.03 (3)	0.25 (5)					ND (3)	

NOTE: All numbers are averages
ND = no residues detected at 0 01 ppm level
Figures in parentheses represent number of specimens analyzed to obtain the average residue given
[1] See footnote, Table 3

Hicks, J. K. Plumley, and Anita Lemire, Agricultural Research Service, for collecting specimens; Dr. Carter Gilbert, State Museum of Florida, for aiding in the identification of fishes; and W. M. Hetrick, Florida Game and Fresh Water Fish Commission, for identifying bird and mammal stomach contents.

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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
AROCLOR 1260	PCB, approximately 60% chlorine
BHC (Benzene Hexachloride)	1,2,3,4,5,6-Hexachlorocyclohexane (mixture of isomers)
CHLORDANE	1,2,3,5,6,7,8,8-Octachloro-2,3,3a,4,7,7a-hexahydro-4 7-methanoindene The technical product is a mixture of several compounds including heptachlor, chlordene, and two isomeric forms of chlordane.
CHLORDENE	4,5,6,7,8,8-hexachloro-3a,4,7,7a-tetrahydro-4,7-methano-1*H*-indene
CHLOROFORM	Trichloromethane
CHLOROPYRIFOS	*0,0*-diethyl *O*-(3,5,6-trichloro-2-pyridinyl) phosphorothioate
DDE	Dichlorodiphenyl dicholoro-ethylene (degradation product of DDT) *p p'*-DDE: 1,1-Dichloro-2 2-bis(*p*-chlorophenyl) ethylene *o p'*-DDE: 1,1-Dichloro-2-(*o*-chlorophenyl)-2-(*p*-chlorophenyl)ethylene
DDT	Main component (*p,p'*-DDT) α-Bis(*p*-chlorophenyl) β β β-trychloroethane Other isomers are possible and some are present in the commercial product. *o p'*-DDT [1,1,1-Trichloro-2-(*o*-chlorophenyl)-2(*p*-chlorophenyl) ethane]
DIELDRIN	Not less than 85% of 1,2,3,4,10,10-Hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4-*endo-exo*-5,8-dimethanonaphthalene
HCB	Hexachlorobenzene
HEPTACHLOR EPOXIDE	1,4,5,6,7,8,8-Heptachloro 2,3-epoxy-3a,4,7,7a-tetrahydro-4,7-methanoindan
LINDANE	*Gamma* isomer of 1,2,3,4,5,6-hexachlorocyclohexane
MIREX	Dodecachlorooctahydro-1,3,4-metheno-1H-cyclobuta[cd]pentalene
NONACHLOR	1,2,3,4,5,6,7,8-Nonachlor-3a,4,7,7a-tetrahydro-4,7-methanoindan
OXYCHLORDANE	2,3,4,5,6,6a,7,7-Octachloro-1a,1b,5,5a,6,6a-hexahydro-2,5-methano-2H-indeno(1,2-β)oxirene
TDE	2,2-Bis(*p*-chlorophenyl)-1,1-dichloroethane

* Compounds mentioned only by Johnson/Manske are defined in Table 1 of that paper

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RESIDUES IN FOOD AND FEED

Organochlorine Insecticide Residues in Bovine Milk and Manufactured Milk Products in Illinois, 1971-76

John L. Wedberg,[1] Stevenson Moore III,[1] Francis J Amore,[2] and Harold McAvoy[3]

ABSTRACT

Monitoring activities were initiated in 1971 to survey the occurrence and levels of organochlorine insecticide residues in bovine milk and manufactured milk products in Illinois. Dieldrin residues were the most prevalent, and were found in 96 percent of the samples. Dieldrin also accounted for the highest average residue concentration (0.09 ppm). Only 0.3 percent of the samples contained illegal insecticide residues. Levels of DDT and lindane were generally declining, but those for dieldrin and heptachlor epoxide tended to remain constant.

Introduction

Use of chlorinated hydrocarbon insecticides on corn soil reached a peak in Illinois during 1967 *(2)* when an estimated 5,601,572 acres of farmland were treated (Table 1).

TABLE 1. *Number of corn acres in Illinois treated with different types of soil insecticides, 1964 through 1976*

Year	Chlorinated Hydrocarbons	Organophosphates and Carbamates
1964	4,009,303	81,822
1965	4,544,432	189,352
1966	5,116,605	326,592
1967	5,601,572	602,721
1968	5,170,726	1,091,143
1969	4,517,931	1,990,138
1970	3,844,740	2,765,547
1971	2,723,119	3,418,920
1972	1,933,089	3,852,239
1973	1,737,510	3,960,543
1974	1,886,042	4,128,300
1975	916,480	4,586,390
1976	935,550	5,428,870

Use of these insecticides has steadily declined since that time because of the development of insect resistance and of farmers' concern about persistence and long-term effects of aldrin, heptachlor, and chlordane residues. The chlorinated hydrocarbons generally have been replaced by organophosphates and carbamates. A study by Moore et al. *(1)* indicated that dieldrin residues were most likely to exceed the action level of the Food and Drug Administration, U.S. Department of Health, Education, and Welfare, on dairy farms which had recently been treated with aldrin. The study showed that hay and oat straw bedding supplied significant amounts of dieldrin to dairy cattle even when this material had been grown on farms with no history of aldrin treatments. Air movement of soil particles containing dieldrin was the reason given for roughage contamination on these farms.

A few Illinois dairy farms have produced milk containing illegal dieldrin residues within recent years *(1)*. Occasionally, this resulted from accidental contamination of feed, but in the majority of cases, the insecticide had been used in accord with the label.

The University of Illinois Cooperative Extension Service and Illinois Natural History Survey (Illinois State Department of Registration and Education) advised against the use of DDT on dairy farms in 1951 because of potential residues in milk. The use of aldrin, chlordane, dieldrin, endrin, heptachlor, or lindane has not been recommended for dairy farms since 1965 or for other farms since 1970. These actions preceded use cancellations by the U.S. Environmental Protection Agency. In September 1971, the Illinois State Department of Public Health announced that it was no longer legal for dairy farmers to store or use these insecticides for agricultural purposes on their farms. The Illinois State Department of Public Health began monitoring milk supplies for insecticide residues in 1971. This report summarizes these findings. All residues expressed in this paper are reported on a fat basis.

[1] College of Agriculture, University of Illinois, Urbana, IL 61801, and Illinois Natural History Survey, Urbana
[2] Illinois Water Survey, formerly, Illinois Department of Public Health, Division of Laboratories, Box 232, Urbana, IL.
[3] Illinois Department of Public Health, Office of Consumer Health Protection, Division of Milk Control, 535 West Jefferson St., Springfield, IL 62761

Sampling Procedure

Samples were drawn at random from tank trucks of milk or from manufactured milk products in the plant and were analyzed for insecticide residues at either the Springfield or Chicago laboratories of the Illinois Department of Public Health. Each sample of raw milk represents at least eight farms; a manufactured milk product sample could include milk from a few to several hundred farms.

Method

EXTRACTION

Butterfat for residue analysis was extracted by placing 100 ml of milk, 30 g of grated cheese, or 50 g of melted ice cream in a 1-liter separatory funnel and adding 50 ml of 20 percent potassium carbonate, 100 ml of ethanol, and 50 ml of diethyl ether. This mixture was shaken briefly and 80 ml of Nanograde hexane was added. The resulting mixture was shaken approximately 200 times and allowed to stand until the phases separated. The lower aqueous phase was drained and discarded. For cheese samples both phases were transferred to another separatory funnel before separation of layers to remove the liquid from cheese solids. The lower aqueous phase was drained and discarded. The ether-hexane (organic) phase was washed three times with 100 ml of water to remove ethanol. Water was poured through the organic phase without shaking and the wash was discarded. Any emulsion formed was broken by adding about 5 ml of ethanol. The ether-hexane phase was transferred to a 250-ml beaker and placed on a sand bath maintained at 58 C in a fume hood; the volume was reduced to approximately 10 ml. Approximately 10 g of anhydrous granular sodium sulfate was added to remove water, the solution was transferred to a 100-ml beaker, and the remaining solvent was removed by passing a gentle stream of air over the solution.

ACETONITRILE PARTITIONING

A 1-g portion of butterfat was weighed into a 25-ml Erlenmeyer flask. The fat was dissolved in 15 ml of Nanograde hexane and transferred with rinsing to a 250-ml separatory funnel. The hexane was extracted four times with 30-ml portions of Nanograde acetonitrile saturated with hexane. The acetonitrile (lower layer) was drained into a 1-liter separatory funnel; after the fourth acetonitrile extraction, the hexane was discarded. The 120 ml of acetonitrile was transferred to a 250-ml separatory funnel and shaken with 10 ml of Nanograde hexane. After the lower acetonitrile layer was returned to the 1-liter separatory funnel, 500 ml of distilled water and 100 ml of saturated sodium chloride solution were added to salt out pesticides. The acetonitrile-water was extracted with 100 ml of 10 percent diethyl ether in hexane. The lower acetonitrile-water phase was discarded after separation. The ether-hexane phase was washed three times by passing 100 ml portions of distilled water through the ether-hexane to wash out traces of acetonitrile. Water washes were discarded. The ether-hexane was quantitatively transferred to a 250-ml beaker, with the aid of a hexane rinse, and placed on a sand bath to evaporate to about 10 ml. Approximately 5 g of sodium sulfate was added to remove traces of water.

COLUMN CHROMATOGRAPHY

Pesticides were separated and further cleaned by using glass columns, 250 × 20 or 330 × 22 mm, containing 80–100-mesh Florisil. The Florisil was heated at 450°C for 8 hours and stored at 130°C before use. Thirty g of Florisil was placed in the column and 0.5 inch of anhydrous sodium sulfate was added. The column was pre-eluted with 50 ml of acetone, 25 ml of anhydrous diethyl ether, 25 ml of water-saturated diethyl ether, and 25 ml of petroleum ether.

The pesticide extract was transferred to the Florisil column with three 1-ml portions of hexane. α-BHC, lindane, heptachlor, aldrin, heptachlor epoxide, γ-chlordane, α-chlordane, *p,p'*-DDE, *p,p'*-TDE, and *p,p'*-DDT were eluted with 120 ml of 6 percent diethyl ether in petroleum ether. Dieldrin and endrin eluted with 120 ml of 15 percent diethyl ether in petroleum ether. Eluates were collected in 250-ml beakers and reduced to approximately 5 ml by air evaporation in a fume hood. Samples were dried with about 2 g of sodium sulfate, transferred with washings of the eluting solvent to a 10-ml volumetric flask, and diluted to volume.

GAS LIQUID CHROMATOGRAPHY

Pesticides were determined by injecting 5-μl samples into a Bendix Model 2500 gas chromatograph equipped with a Ni-63 electron-capture detector and a 6-ft X ¼-inch id glass column containing 1.95 percent QF-1/1.25 percent OV-17 on 100–120-mesh Supelcoport. Instrument operating conditions were:

Injection port temperature	225°C
Column temperature	190°C
Transfer line temperature	225°C
Detector temperature	275°C
Nitrogen flow rate	30 ml/minute

Sensitivity so that 100 pg of dieldrin gave a 30 percent recorder response with a noise level of about 0.5 percent

A standard solution containing 0.01 ng/μl each of α-BHC, lindane, heptachlor, aldrin, heptachlor epoxide, γ-chlordane, and α-chlordane and 0.02 ng/μl each of *p,p'*-DDE, dieldrin, endrin, *p,p'*-TDE, and *p,p'*-DDT was used for retention time comparison and quantitation. Quantitation was based on peak height comparison between a standard and a sample.

EFFICIENCY MONITORING

Recovery efficiency of the 12 pesticides listed above was

continually monitored by using a sample of previously extracted butterfat to which all 12 pesticides were added to give final concentrations equivalent to those in the quantitation standard. Whenever recovery efficiencies decreased below 80 percent, final results were corrected for percent recovery. Procedural adjustments were made whenever recoveries decreased to less than 60 percent. This usually consisted of Florisil column modification. Aldrin had consistently lower recoveries than did the other insecticides; this was expected from its unfavorable partition coefficient from hexane to acetonitrile. Column packing was changed and the detector was cleaned as needed, based on reduced resolution and sensitivity.

PCBs, if present at levels above 0.5 μg/g fat, could be detected without procedure modification. No PCBs were detected in any samples. If PCBs had been present, it would have been necessary to modify the procedure to separate the pesticides and PCBs and prevent errors in the calculated pesticide levels.

Results and Discussion

Detailed results of the milk monitoring program for insecticides in 1971-76 are given in Table 2. Dieldrin was the most prevalent insecticide: it was found in 96 percent of the samples. Other insecticides which occurred in many of the samples were heptachlor epoxide, 93 percent; lindane, 73 percent; chlordane, 69 percent; and DDT, 48 percent. No other insecticides were found in significant amounts (>0.01 ppm) in these samples. Dieldrin also accounted for the highest average residue concentration in milk: 0.09 ppm; followed by heptachlor epoxide, 0.04 ppm; chlordane, 0.03 ppm; DDT, 0.01 ppm; and lindane, 0.01 ppm. DDT is the only insecticide with an official tolerance (1.25 ppm fat basis, 0.05 ppm whole milk basis) established in milk. Dieldrin was found in quantities above the action level in one sample during 1973, lindane exceeded the action level in one sample during 1974, and chlordane exceeded the action level in one sample during 1976. Therefore, in 6 years of sampling milk and manufactured milk products (1169 samples), only 0.3 percent of the samples contained illegal insecticide residues.

Too few samples were obtained in 1971 and 1972 for use in determining trends. However, the large number of samples obtained during 1973-76 makes it possible to discuss trends for insecticide levels in milk. Levels of DDT and lindane have generally declined since 1973, but dieldrin and heptachlor epoxide levels have not declined (Fig. 1). No organochlorine insecticide showed a definite upward trend in milk. During this study, combined chlorinated hydrocarbon residues in milk and manufactured milk products averaged 0.18 ppm (Table 2). The level of combined residues declined after 1973 (Fig. 1).

These data show that the occurrence of illegal residues of organochlorine insecticides in Illinois milk is rare. Levels are below the tolerance established by the Environmental Protection Agency or action levels established by the Food and Drug Administration. This trend is expected to continue as the use of insecticides such as aldrin, dieldrin, chlordane, and heptachlor for agricultural purposes is discontinued.

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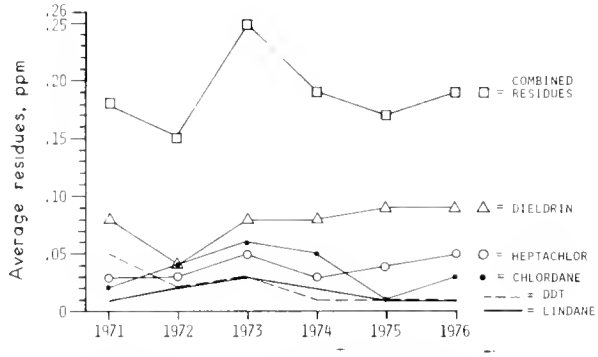

FIGURE 1. *Trends in insecticide residues in milk and in manufactured milk products, Illinois—1971-76*

TABLE 2. *Insecticide residues in samples of bovine milk and manufactured milk products, by year, Illinois—1971-76*

Insecticide	No. Positive	Percent Positive	Avg. ppm	Percent Samples 0.01–0.10 ppm	Percent Samples 0.11–0.20 ppm	Percent Samples 0.21–0.30 ppm	Percent Samples 0.31+ 0.01–0.10 ppm
1971 (13 SAMPLES)							
Chlordane	5	39	0.02	100	0	0	0
DDT	12	92	0.05	92	8	0	0
Dieldrin	13	100	0.08	85	15	0	0
Heptachlor epoxide	13	100	0.03	100	0	0	0
Lindane	1	8	trace	100	0	0	0
Total	13	100	0.18	—	—	—	—
1972 (34 SAMPLES)							
Chlordane	34	100	0.04	100	0	0	0
DDT	31	91	0.02	97	3	0	0
Dieldrin	34	100	0.04	100	0	0	0
Heptachlor epoxide	30	88	0.03	100	0	0	0
Lindane	30	88	0.02	100	0	0	0
Total	34	100	0.15	—	—	—	—
1973 (153 SAMPLES)							
Chlordane	135	88	0.06	90	10	0	0
DDT	141	92	0.03	98	2	0	0
Dieldrin	141	92	0.08	65	31	3	1
Heptachlor epoxide	142	93	0.05	89	11	0	0
Lindane	130	85	0.03	96	2	2	0
Total	153	100	0.25	—	—	—	—
1974 (186 SAMPLES)							
Chlordane	143	76	0.05	84	16	0	0
DDT	122	64	0.01	98	2	0	0
Dieldrin	173	92	0.08	72	25	3	0
Heptachlor epoxide	162	86	0.03	98	2	0	0
Lindane	157	83	0.02	97	1	1	1
Total	186	98	0.19	—	—	—	—
1975 (283 SAMPLES)							
Chlordane	69	24	0.01	93	7	0	0
DDT	48	17	0.01	77	17	6	0
Dieldrin	271	96	0.09	72	24	4	0
Heptachlor epoxide	265	94	0.04	99	1	0	0
Lindane	259	92	0.01	100	0	0	0
Total	283	100	0.17	—	—	—	—
1976 (500 SAMPLES)							
Chlordane	421	84	0.03	98	1	0	1
DDT	213	43	0.01	98	2	0	0
Dieldrin	494	99	0.09	65	34	1	0
Heptachlor epoxide	481	96	0.05	97	3	0	0
Lindane	280	56	0.01	99	1	0	0
Total	500	100	0.19	—	—	—	—
SUMMARY—1971–1976 (1,169 SAMPLES)							
Chlordane	807	69	0.03	94	6	0	0
DDT	566	48	0.01	96	4	0	0
Dieldrin	1126	96	0.09	69	29	2	0
Heptachlor epoxide	1093	93	0.04	97	3	0	0
Lindane	857	73	0.01	99	1	0	0
Total	1169	100	0.18	—	—	—	—

* cis- and trans-chlordane residues are combined as chlordane. DDD, DDE, and TDE are combined as DDT. Lindane and BHC are combined as lindane.

RESIDUES IN FISH, WILDLIFE, AND ESTUARIES

Variations in DDT Concentration in Muscle Tissue of Channel Catfish, Ictalurus punctatus, *from the Des Moines River, 1971*[1]

Ross Vivian Bulkley[2]

ABSTRACT

Concentrations of DDT in muscle tissue of channel catfish, Ictalurus punctatus, *from the Des Moines River, Iowa, were compared in relation to length, age, sex, and fat content. Residue concentrations were not correlated with sex. Although residue concentrations were significantly correlated with size-related factors and fat content, they varied widely. Therefore, predictions of DDT concentrations in individual catfish on the basis of length, age, or muscle fat content may be grossly inaccurate.*

Introduction

Accumulation of pesticides in fish at a given exposure is often envisioned as a simple function of age, body size, or fat content. Old, large, or fat fish are expected to contain higher residue concentrations than are young, small, or lean fish. The many exceptions and wide variations involved in the relation of pesticide concentrations to size-related factors in individual fish are frequently overlooked. Differences in pesticide concentrations sometimes exceed 100 percent among fish of similar size, age, or fat content *(3,7)*, and pesticide levels may decrease with increased age or fat content *(1, 6)*. Data on concentrations of DDT in muscle tissue of individual channel catfish, *Ictalurus punctatus*, obtained during a study of dieldrin contamination *(5)* provided an additional example of the variability in pesticide concentrations in relation to age, length, and fat content of the fish.

Methods

Channel catfish were collected from the Des Moines River near Fraser, Iowa, from April to October 1971, with baited hoop nets and electrofishing gear. The length of each fish was measured at capture. Fish were wrapped in aluminum foil and frozen until chemical analysis. Dorsal muscle tissue of all catfish collected in June was analyzed individually. On each collection date in other months, the fish in each of two length groups, 200-299 mm and 300-399 mm, were pooled for analyses. Percentage fat content of muscle tissue was determined by the rapid modified Babcock method *(2)* on individual samples. Sex was determined by examination of gonads, and age was determined from pectoral spine sections.

Muscle tissue samples were analyzed for DDT content according to the *Pesticide Analytical Manual (4)*. A sample of 10-40 g of tissue was ground with 350 ml of 35 percent distilled water-acetonitrile solution in a high-speed blender for 10 minutes. The sample was filtered through fluted number 40 Whatman paper. The filtrate (260 ml) was transferred to a 1-liter separatory funnel; 10 ml of petroleum ether was added, and the mixture was shaken for 2 minutes. Six hundred ml of distilled water and 10 ml of a saturated saline solution were added to the sample and mixed thoroughly by vigorous tumbling for 15 seconds. The layers were allowed to separate, and the aqueous layer was discarded. The solvent layers were gently washed with two 100-ml portions of water. The washings were dis-

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[2] Iowa Cooperative Fishery Research Unit, Iowa State University, Ames, IA 50011

carded, and the solvent layer was filtered through a 50-mm column of anhydrous sodium sulfate into a 100-ml graduated cylinder. The volume was recorded, and the sample was filtered through a 115-mm column of Florisil topped by a 25-mm column of anhydrous sodium sulfate. DDT was eluted from the column with 200 ml of a 6 percent mixture of diethyl ether in petroleum ether. The solvent was evaporated to 10 ml for injection into the chromatograph.

A Beckman Model GC-5 gas chromatograph equipped with a discharge electron-capture detector was used to separate and quantitate the compounds of interest. A 5 percent OV-210 column at 180°C and 1.5 percent OV-17/QF-1 column at 200°C were used. Helium flow was about 100 ml/min and attenuation was $2x10^3$. A column of different polarity, 4 percent SE-30/6 percent QF-1, with a gas flow of 120-ml/min, a temperature of 200°C, and attenuation of $2x10^3$ was used as a qualitative check. Identities of compounds were confirmed by comparing retention times on the two columns with that of a DDT standard. Recovery of DDT and its metabolites ranged from 85 to 95 percent. Values were not corrected for percent recovery. Concentrations were expressed in wet weight.

All data were transformed to a $\log_{10}$ base for calculating regression and correlation coefficients of DDT in relation to fat content, length, and weight.

Results

ANALYSES OF INDIVIDUAL FISH

In June, 32 fish (14 males and 18 females, 155 to 602 mm long) were collected just before the spawning season. Differences in concentration of total DDT (DDT, TDE, and DDE) in males and females were not significant. Males averaged 355 mm long and contained an average of 1221 μg/kg of ΣDDT (78-6336 μg/kg) in muscle tissue; females averaged 325 mm long and contained an average of 799 μg/kg (154-4117 μg/kg). The regression lines expressing $\log_{10}$ DDT versus $\log_{10}$ body length were not significantly different ($P=0.05$) between males and females in terms of slope b ($t=0.25$, $df=30$) or intercept a ($t=0.28$, $df=30$). For all subsequent comparisons, therefore, data on male and female fish were combined.

Mean concentrations of DDT, DDE, TDE, and ΣDDT increased with length of fish in the June sample, with minor exceptions (Table 1). All DDT values given hereafter refer to ΣDDT. The correlation between length and DDT concentration ($r=0.63$) was significant at the 0.01 level of probability. The absolute and relative variation in DDT concentration, however, increased with size of the mean. The standard deviation ranged from 26 percent of the mean (coefficient of variation) in the 100-199-mm length group to 73 percent in the 300-399 group. Although the mean DDT concentration in muscle was greater in large than in small fish, the range in concentrations was also greater. Confidence limits about the mean DDT concentration for the large fish included the mean of all small fish. For example, the 500-599-mm group contained an average of 3389 μg/kg ΣDDT, with a standard deviation of 2317. The 95-percent confidence interval was -296 to 7974 μg/kg. The mean of all other length groups fell within this interval. Concentrations of DDT in the four fish in the 500-599-mm length group ranged from 1285 to 6336 μg/kg, and both values were obtained from fish of equal length (515 mm). Within the six length groups, the DDT concentration in individual fish was 0.6 to 6.4 times greater than the concentration in other fish the same length or shorter. The geometric mean DDT concentration more closely indicated the central tendency of the data for each length group than did the arithmetic mean (Table 1).

Inasmuch as one source of variation in pesticide extraction and quantitation is the person conducting the analysis, the June sample was divided into two groups for analysis by two chemists (Fig. 1). Although the author would have preferred having the chemists analyze duplicate samples of the same fish, this was precluded by the expense of analysis and the need for a sizeable series of samples. Analyses conducted by chemist A provided a relation between DDT concentration and body length, using $\log_{10}$ transformed data, expressed by the equation: $\log_{10}$ DDT (μg/kg) = -1.50292 + 0.34265 $\log_{10}$ total length (mm). Chemist B obtained a relation expressed by the equation: $\log_{10}$ DDT (μg/kg) = -2.06662 + 0.17648 $\log_{10}$ total length (mm). Data from both analysts fit a single regression line ($P<0.05$) on the basis of slope b ($t=0.18$, $df=30$) and intercept a ($t=0.30$, $df=30$). Correlation coefficients for length versus ΣDDT, using $\log_{10}$ transformed data, were 0.74 for chemist A and 0.58 for chemist B; the average value for all 32 fish was 0.63. All other samples in the study were analyzed by chemist A.

Fish in the June sample ranged from 2 to 12 years (Table 2, Fig. 1). Mean concentration of DDT did not increase with age as uniformly as with body length, although the correlation coefficient was close to that for length ($r=0.65$) and was also significant at the 0.01 probability level. The range in DDT levels was greatest (1288-6336 μg/kg) in fish of 7 years. The coefficient of variation expressed as a percentage of the mean was relatively low for ages 2 and 3 (13 and 24 percent, respectively), and values increased up to age 7 (95 percent), the oldest age group represented by more than one fish.

Fat content is frequently considered a major determinant in how much pesticide residue a fish will contain because of the lipophilic properties of many pesticides. Muscle fat ranged from less than 0.1 to 11.9 percent in the 32 fish. Fat content was correlated ($P=0.01$) with both length ($r=0.73$) and age ($r=0.63$). Fat content was highest in fish of 6 and 7 years and 450-525 mm long. Fish in this group were in prime spawning condition, which probably ex-

TABLE 1 *Residues of DDT, TDE, DDE, and ΣDDT in channel catfish from Des Moines River, June 1971*

Length Group, mm	No. Fish	Residues, μg/kg				ΣDDT			
		DDT	TDE	DDE	ΣDDT	C.V., Percent[1]	Confidence Limit[2]		G.M.[3]
100-199	6	119 (28)	70 (30)	170 (44)	363 (96)	26	253	to 473	348
200-299	9	126 (89)	62 (28)	143 (87)	332 (151)	45	217	447	290
300-399	6	166 (131)	137 (89)	300 (234)	604 (443)	73	139	1069	459
400-499	6	297 (236)	256 (190)	447 (255)	1001 (659)	66	310	1692	772
500-599	4	1211 (926)	695 (673)	1483 (578)	3389 (2317)	68	296	7074	2794
600-699	1	1182	994	1675	3851	—	—	—	—
	Mean	333	222	450	1006	56	—	—	—

Note: Standard deviations are shown in parentheses.
[1] Coefficient of variation = 100 (standard deviation) mean.
[2] 95-percent confidence limits.
[3] Geometric mean.

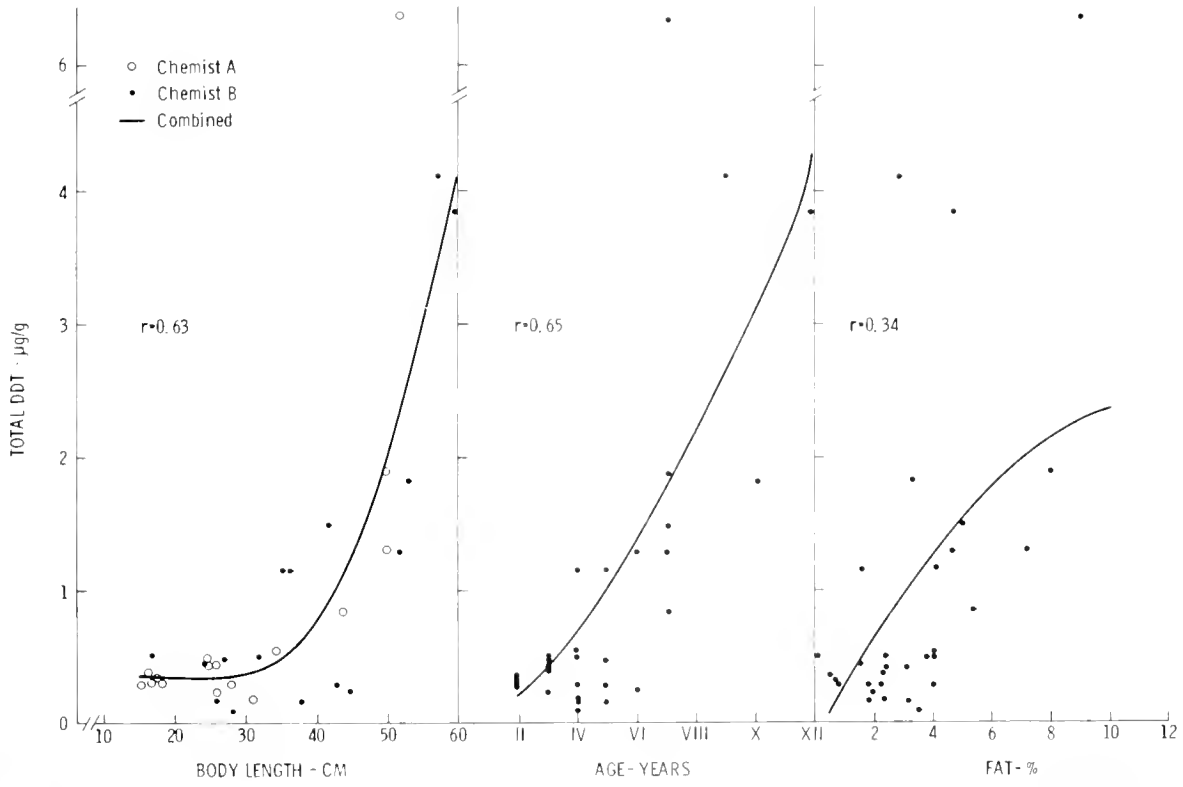

FIGURE 1 *Relation of ΣDDT to body length, age, and muscle fat content of channel catfish collected from Des Moines River, June 1971*
(Lines on each graph express a second degree polynomial regression for the data.)

plained the high fat content in muscle tissue. Fat buildup in conjunction with gonad development is common in some species *(8)*. Fish larger and older than 6 and 7 years contained less fat, and fish of 2 years and shorter than 200 mm contained the least. Correlation of muscle fat with DDT levels (r=0.34) was much lower than DDT versus length or age (Table 2) and the correlation value obtained was not significant at the 0.05 level of probability. To illustrate the low correlation, the fish with the highest fat content (11.9 percent) contained less DDT in the muscle tissue (261 μg/kg) than did four fish each containing less than 1 percent muscle fat. The range in DDT concentrations within each fat group (Table 2) was also large; in four groups the standard deviation exceeded the mean. Thus percentage fat content was of little value in predicting the amount of DDT present in muscle tissue of catfish of different lengths and ages.

TABLE 2. *ΣDDT in channel catfish of different ages and muscle fat content collected in June 1971*

Group	No. Fish	Mean	S.D.	Confidence Limits[2]	
		Residues, μg/kg			
Age, yr					
2	5	324 (322)	42	271	to 377
3	6	420 (407)	101	315	525
4	7	406 (291)	363	71	741
5	4	511 (388)	444	-195	1217
6	2	733 (579)	724	-5732	7278
7	5	2361 (1799)	2253	434	5156
9	1	4117	-	-	-
10	1	1819	-	-	-
12	1	3851	-	-	-
Fat, percent					
0.1-0.9	4	364 (355)	104	199	529
1.0-1.9	5	411 (351)	436	-130	952
2.0-2.9	6	978 (505)	1541	639	2595
3.0-3.9	5	500 (338)	708	-287	1467
4.0-4.9	6	1260 (854)	1331	-136	2656
5.0-5.9	2	1156 (1112)	450	-2884	5196
7.0-7.9	1	1288	-	-	-
8.0-8.9	1	1871	-	-	-
9.0-9.9	1	6336	-	-	-
11.0-11.9	1	261	-	-	-

[1] Geometric means in parentheses.
[2] 95 percent confidence limits.

TABLE 3. *Correlation of ΣDDT with length, age, and muscle fat content in channel catfish, 1971*

Variable	r Value	R Value
JUNE (ALL LENGTHS)		
Length	0.63	
Age	0.65	
Fat	0.34[1]	
Length, age		0.65
Length, fat		0.66
Age, fat		0.66
Length, age, fat		0.67
APRIL-OCTOBER (300-399 mm)		
Length	0.30[1]	
Age	0.34[2]	
Fat	0.31[1]	
Length, age		0.36[2]
Length, fat		0.37[2]
Age, fat		0.44
Length, age, fat		0.44

NOTE: Values are significant at the 0.01 level of probability unless otherwise noted.
[1] Nonsignificant at 0.05 level of probability.
[2] Significant at 0.05 level of probability.

Comparison of two or more size related factors with DDT concentration accounted for little additional variation in the relationships (Table 3). Length, age, and fat accounted for 40, 42, and 11 percent, respectively, of the variation in DDT levels, but the amounts were not additive. When two, or all three, factors were considered together, a maximum of only 45 percent (R=0.67) of the variation in DDT could be accounted for.

SEASONAL SAMPLES (APRIL-OCTOBER)

Samples collected from April to October provided additional observations on the relation of DDT to length. Pooled samples of fish 200-299 mm long contained the highest DDT concentrations in April and May and the lowest in August and September (Table 4). In fish 300-399 mm long analyzed individually, the DDT concentration was also greatest in April. When one specimen containing 3218 μg/kg DDT was omitted from the July samples of fish measuring 300-399 mm, the trend in DDT concentration in both size groups was similar, ranging from highest in April to lowest in August. No significant difference was evident between seasonal mean DDT levels in fish of the two length groups (t=0.253, df=12); however, as in the June sample, fish 300-399 mm long averaged slightly more DDT than did those measuring 200-299 mm long (569 versus 515 μg/kg).

Comparison of DDT levels in the two length groups within each month from April to October revealed only minor differences, with one exception. The 95-percent confidence limits for the mean of the 300-399-mm group included the mean DDT levels of the 200-299-mm group in all months except April. The mean concentration (1448 μg/kg) of DDT in the shorter length group in April was unusually high. Because tissues of the April sample of 15 fish were combined for DDT analysis, variation among individual fish within the sample could not be measured. Even though the mean monthly DDT concentration in muscle tissue of the 300-mm fish which were anlyzed individually ranged from 798 μg/kg in April to 179 μg/kg in August, monthly means for the April-October period were not significantly different (F=1.83; df=6, 33).

Discussion

Pesticide concentrations in fish frequently do not bear a simple and direct relation to body condition, size, age, or fat content. Kleinert et al. *(6)* reported that the relation of DDT levels to percentage fat was not significant in fish from most Wisconsin water samples. Anderson and Fenderson *(1)* found significantly greater concentrations of DDT in high-fat Atlantic salmon *(Salmo salar)* than in low-fat fish at ages 3+ and 4+ but not at age 5+. They recommended stratification of sampling by sex, age, and fat content for pesticide analysis. Bulkley et al. *(3)* found that dieldrin concentrations in muscle tissue of the same channel catfish used in this paper were significantly correlated with body length (r=0.46) and age (r=0.60) only if fish of all lengths and ages were included in the analyses. When only fish shorter than 400 mm and younger than age 6 were considered, dieldrin concentration was not related to length and age. Dieldrin was significantly correlated

TABLE 4 *Seasonal concentrations of ΣDDT (DDT, TDE, DDE) in two length groups of channel catfish from Des Moines River, April-October 1971*

Month	200-299 mm Long			300-399 mm Long			C.V., Percent[1]	Confidence Limits[2]		
	No. Fish	Mean	SD	No. Fish	Mean	SD				
April	15	1448	-	9	798	518	65	407	to	1289
May	7	884	-	8	689	769	112	46		1332
June	9	332	151	6	604	443	73	139		1069
July	6	231	-	7	780	1083	139	-221		1781
August	4	101	-	3	179	78	43	-16		374
September	3	224	-	4	696	303	43	212		1180
October	3	385	-	3	239	74	31	54		424

[1] Coefficient of variation = 100 (standard deviation)/mean
[2] 95-percent confidence limits

with muscle fat content ($r=0.68$, $P=0.01$) only if the longest and oldest catfish was removed from the sample. When all fish were included, the correlation coefficient was 0.27. The correlation of ΣDDT concentration with length, age, or fat content was slightly higher than the correlation between dieldrin and these variables in the same fish ($r=0.68$, 0.68, 0.45, respectively), but exceptions to these relationships were common. Other size-related variables, such as metabolic rate or individual variations in diet or in development of detoxifying enzyme systems, are undoubtedly involved in DDT and dieldrin accumulation in wild catfish. Hence predictions of concentrations of DDT or dieldrin in muscle of individual channel catfish on the basis of length, age, or muscle fat content may be grossly inaccurate, and generalities about any relationship should be made with caution.

Acknowledgment

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p,p'-DDE, Polychlorinated Biphenyls, and Endrin in Oldsquaws in North America, 1969-73

Steven R. Peterson[1] and Robert S. Ellarson[2]

ABSTRACT

Organochlorinated compounds were monitored in oldsquaws (Clangula hyemalis) *and their food from Lake Michigan between October and May, 1969-72; in adult oldsquaws, eggs, young, and food from northwest Hudson Bay in 1971; and in oldsquaws from five wintering areas other than Lake Michigan in 1971-73. Analyses were conducted on 300 carcasses, 14 wings, 29 gullet samples, and 11 clutches. Average residues in carcasses from Lake Michigan ranged from 4 to 107 ppm PCBs, 2 to 42 ppm DDE, and < 0.1 to 0.7 ppm endrin. Differences in DDE levels occurred between several sex and age classes during December on Lake Michigan; these differences were not apparent in the spring. Increases in DDE and PCB residues for oldsquaws occurred on Lake Michigan between December and May. DDE residues in the wing and carcass were significantly correlated. Residues were relatively low in oldsquaw foods from Lake Michigan, concentration factors between the food and the ducks varied between 1 × and 22 ×, depending on the date and compound. Organochlorinated residues were lower in Arctic than in Lake Michigan food samples. DDE in paired male and female oldsquaws was highly correlated, as was DDE in females and clutches. Eggshell thickness had declined 4.5 percent compared with eggs collected before 1947. Residues were highest in oldsquaws wintering on the Great Lakes and lowest in oldsquaws from coastal areas.*

Introduction

An exploratory study on Lake Michigan indicated that several trophic levels had been contaminated *(16)*. This paper reports residues of *p,p'* DDE, endrin, and polychlorinated biphenyls (PCBs) in oldsquaw *(Clangula hyemalis)* adults, eggs, young, and food. Various organochlorinated compounds were measured in this study, but because residue identities could not be confirmed by mass spectrometry, and because PCB levels were relatively high, results are not included for TDE, DDT, dieldrin, BHC, and heptachlor epoxide. These data are available from the senior author on request.

Oldsquaws wintering on Lake Michigan are suitable for monitoring contamination on the lake. The species arrives on Lake Michigan in November and remains there until April or May *(7)*. Presumably, then, any detectable increase in pesticide body burdens after November would come from within the lake.

While on the lake the species feeds almost entirely on a bottom dwelling deep-water amphipod, *Pontoporeia affinis (7, 25)*. This invertebrate is also the predominant food of the commercially important lake whitefish *(Coregonus clupeaformis) (7, 20)*; thus this food organism in common with both consumers allowed authors to measure concentration in a bird and to compare findings with other studies on a fish at the same trophic level.

Several age and sex classes are discernible in the oldsquaw *(1, 7, 24)*. This allowed authors to test for differences in body burdens between sex, age, and time.

Since the oldsquaw is an Arctic nester, the Lake Michigan population spends about half the year in a relatively polluted environment and the remainder in a relatively clean environment. Thus shifts in body burdens caused by changes in diet contamination were investigated, and the transfer of pesticides from a wintering to a breeding area was explored.

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Methods

SAMPLE COLLECTION

Samples collected on Lake Michigan were obtained from commercial fishing operators, who found the birds drowned in gill nets. All other birds were killed by shotgun except those collected on Lake Huron. The latter samples were found dead on a beach; an autopsy performed by the Rose Lake Laboratory of the Michigan Department of Natural Resources indicated that the birds had died of hemorrhagic enteritis of undetermined cause.

Samples were catalogued between October 30 and August 10, 1969-73. They were either examined and prepared immediately or frozen until dissection and preparation. Sex and age were confirmed during dissection. Hanson's *(10)* classification was adopted for age categories: juvenile oldsquaws are less than fully grown and less than 1 year old; subadults are more than 1 year old and essentially fully grown although the majority of the cohort has not completed its first breeding season; adults are at least 2 years old, fully grown, and most members of the cohort have completed one or more breeding seasons. In this paper, adults and subadults combined in one class are called matures.

Carcasses were prepared for chemical analysis according to the following procedure: the head was severed, wings were clipped at the distal end of the humerus, feet were removed at the tarso-metatarsal joint, and feathers were sheared with scissors as close to the skin as possible. After the gastrointestinal and reproductive tracts were removed, the carcass was wrapped in aluminum foil and frozen until chemical analysis.

Food samples were removed from the esophagi of oldsquaws. Only *Pontoporeia affinis*, with some adhering inorganic material, was analyzed from the Lake Michigan samples. Since little food was noted in oldsquaws collected in the Arctic, all food found in the esophagi was composited. Food samples were placed in glass jars previously rinsed with acetone, and sealed with aluminum foil, capped, and frozen until analysis.

Eggs from clutches incubated less than 1 week were wrapped in aluminum foil and frozen for shipment. In the laboratory, the eggs were slightly thawed, and the contents were placed in glass jars previously rinsed with acetone. The jars were then sealed with aluminum foil, capped, and frozen until analysis of the contents.

Wings were prepared for analysis by clipping the feathers as close to the skin as possible. The samples were then wrapped in aluminum foil and frozen for analysis.

EGGSHELL MEASUREMENTS

Weight, length, and breadth of all fresh eggs were measured before freezing. After the contents had been removed, the shells were washed and air-dried to a constant weight; then shell thickness was measured at the equator with a modified Starrett Model 1010M micrometer. Measurements on pre-1947 eggs were obtained from clutches deposited in the Field Museum of Natural History in Chicago and the Museum of Vertebrate Zoology, University of California, Berkeley, California.

CHEMICAL ANALYSIS

All samples were anlyzed at WARF Institute, Inc., Madison, Wisconsin, by a gas chromatographic procedure. The entire carcass, wing, egg contents of a composite clutch, a composite brood, or composite food sample was homogenized in a Hobart food chopper. A sample was removed, weighed, dried at 40°C for 72-96 hours, reweighed, ground with sodium sulfate, and extracted for 8 hours on a Soxhlet extractor using 70 ml of ethyl ether and 170 ml of petroleum ether. An aliquot of the sample was cleaned up on a column of previously standardized Florisil. Typical elutions were 150 ml of 5 percent ethyl ether in petroleum ether, followed by 240 ml of 15 percent ethyl ether in petroleum ether. Compounds were separated and detected on a Barber-Colman Pesticide Analyzer Model 5360 with two columns: one 4-ft x 4-mm column was packed with 5 percent DC-200 on 60–70-mesh Cromport XXX. The column temperature was 200°C, and the nitrogen flow was one which gave *p,p'*-DDT a retention time of 6-8 minutes. The other similar-size column was packed with 3 percent OV-17 on 100–120-mesh Gas-Chrom Q with a column temperature of 195°C and a nitrogen flow which gave lindane a retention time of 1 minute.

The following procedure was used to quantitate PCBs: the samples were injected, and peaks on the DC-200 column which had retention times of TDE and DDT were measured and quantitated. The extract was then subjected to alkaline hydrolysis and re-injected, and the PCBs were quantitated against Aroclor 1254, using the peaks between TDE and DDT and the peak at DDT. Endrin was separated from the PCBs by the initial Florisil chromatography.

Results

LAKE MICHIGAN

Relationship of Body Burden to Age/Sex Class and Time—Of the compounds measured, DDE and PCB residues were generally the highest; endrin residues were relatively low (Table 1). In December, mean DDE levels varied from 4.9 ppm in juvenile males and females to 13.8 ppm in mature males. Adult females averaged 7.0 ppm; this was significantly lower than the average residue level recorded from subadult females ($\bar{x}$ = 10.1 ppm; $P<0.05$) and mature males ($\bar{x}$ = 13.8 ppm; $P<0.01$).

Average December PCB levels generally were higher than December DDE levels but otherwise exhibited trends similar to DDE. Mature males contained the highest residues,

averaging 19 ppm; averages for juvenile males were the lowest, 5 ppm. Statistical tests similar to those performed on DDE showed similar levels of significance for PCBs between different age/sex classes.

No apparent trend was noted for endrin. Overall F-tests among age/sex classes were nonsignificant, and t-tests between selected groups did not yield any statistically significant information.

DDE and PCB residues were much higher ($P<0.05$) in March–April than in the previous December in all age/sex classes (Table 1). Significant differences between age/sex classes which had been apparent in early winter were not present in the spring.

In 1970–71, collections were expanded to monthly intervals in order to check yearly differences in residues, to confirm early winter differences in DDE and PCB residues between

TABLE 1. *Mean and standard deviation of organochlorine pesticide and PCB residues in oldsquaws collected on Lake Michigan, 1969–70*

Sample Size	Age/Sex Class	Residues, ppm Wet Weight		
		p,p' DDE	PCBs	Endrin
	1–15 DECEMBER 1969			
8	Mature male	13.8 ±5.3	19 ±14	0.2 ±0.1
8	Juvenile male	4.9 ±2.5	5 ±2	0.2 ±0.1
10	Adult female	7.0 ±3.6	9 ±4	0.1 ±0.1
4	Subadult female	10.1 ±0.8	14[1] ±4	0.2 ±0.1
7	Juvenile female	4.9 ±2.2	9 ±13	0.2 ±0.1
	22 MARCH – 22 APRIL			
13	Mature male	42.2 ±21.5	75 ±64	0.2 ±0.1
9	Juvenile male	34.7 ±13.4	69 ±38	0.4 ±0.3
12	Adult female	34.1 ±14.5	72 ±57	0.3 ±0.2
6	Subadult female	24.1 ±15.4	46 ±32	0.3 ±0.1
4	Juvenile female	18.5 ±5.3	30 ±14	0.2 ±0.1

[1] One value in the mean was 41, without this value, $\bar{x} = 4 \pm 2$

TABLE 2. *Mean and standard deviation of organochlorine pesticide and PCB residues in oldsquaws collected on Lake Michigan, 1970–71*

Sample Size	Age/Sex Class	Residues, ppm Wet Weight		
		p,p' DDE	PCBs	Endrin
	DECEMBER			
4	Mature male	12.5 ±6.0	20 ±7	0.1 ±0.1
4	Juvenile male	2.9 ±1.0	4 ±1	0.9 ±1.1
4	Adult female	6.0 ±1.1	11 ±2	0.1 ±0.1
6	Subadult female	8.2 ±4.7	18 ±12	0.2 ±0.1
6	Juvenile female	2.1 ±0.9	8 ±10	0.2 ±0.1
	JANUARY			
	Mature male	12.6 ±2.8	23 ±2	0.5 ±0.2
4	Juvenile male	5.0 ±2.8	14 ±17	0.4 ±0.2
4	Adult female	8.5 ±4.8	15 ±11	0.5 ±0.2
	Subadult female	7.7 0.1	14 ±4	0.6 ±0.1
2	Juvenile female	2.4 ±0.6	4 ±1	0.4 ±0.3
	FEBRUARY			
0	Mature male			
4	Juvenile male	5.9	12 ±5	0.5 ±0.1
	Adult female			
	Subadult female			
3	Juvenile female		9 ±2	0.5 ±0.2
	MARCH			
4	Mature male	19.1 ±8.7	39 ±16	0.4 ±0.1
4	Juvenile male	10.1 ±2.7	25 ±14	0.4 ±0.1
4	Adult female	12.2 ±3.9	27 ±6	0.3 ±0.2
4	Subadult female	12.8 ±5.9	34 ±24	0.3 ±0.2
4	Juvenile female	7.8 ±3.8	25 ±22	0.2 ±0.1
	APRIL			
4	Mature male	14.8 ±10.1	89 ±55	0.2 ±0.2
4	Juvenile male	13.5 ±6.0	38 ±23	0.3 ±0.1
4	Adult female	15.0 ±7.8	52 ±12	0.8 ±0.7
5	Subadult female	25.4 ±7.9	71 ±41	0.3 ±0.1
4	Juvenile female	9.8 ±4.7	36 ±38	0.3 ±0.1
	MAY			
4	Mature male	30.9 ±11.6	86 ±69	0.2 ±0.1
6	Juvenile male	21.5 ±6.4	54 ±25	0.2 ±0.1
4	Adult female	32.1 ±18.0	85 ±40	0.3 ±0.1
1	Subadult female	30.9	115	0.4
4	Juvenile female	18.7 ±5.2	37 ±22	0.2 ±0.1

[illegible] one-half the stated [illegible] less than [illegible] value and residues not detected were treated as zero.

age/sex classes, and to graph the apparent accumulation in DDE and PCB residues over the winter.

Early winter and spring levels for 1970-71 were similar to those recorded the previous winter (Table 2). For a particular age/sex class, average December DDE (except in juvenile females) and PCB levels were not significantly different between 1969 and 1970 ($P>0.05$). In March-April 1970-71, DDE and PCB levels were not significantly different ($P>0.05$) for any particular age/sex class.

The increases ($P<0.01$) of DDE and PCB residues in oldsquaws wintering on Lake Michigan between October and May (Fig. 1) are similar, but the increase is greater for PCBs. In birds starting the winter with relatively low residue levels (juveniles and adult females), the data fit the equation for a curved line ($Y = aX^b$) better than for a straight line ($Y = a + bX$). For DDE, juveniles had a linear coefficient of determination of 69 percent and a power coefficient of determination of 85 percent. For PCBs, these coefficients were 48 percent and 70 percent, respectively. Similarly, in adult females, these coefficients increased from 43 to 58 percent for DDE, and from 52 to 71 percent for PCBs. The data for subadult females and mature males did not suggest a better fit to a curved line equation because the coefficients decreased from 59 to 55 percent and from 29 to 26 percent in DDE for these respective age/sex classes.

These results suggest two relationships: that DDE and PCBs increase at a relatively constant rate throughout the winter; and that coefficients of determination are higher in younger birds than in older birds, which indicates that as a cohort ages, its members are exposed to varying amounts of contamination and consequently the variability in the data increases.

During January and February, it is often difficult to obtain sufficient samples of oldsquaws because ice makes it difficult to tend the nets. In 1972, the previous collection was augmented with some additional samples from these months (Table 3). These data largely confirm the results discussed in Table 2.

Comparison of Milwaukee Harbor and Lake Michigan Oldsquaws—A population of 5,000-10,000 oldsquaws apparently winters in Milwaukee Harbor *(29)*. A sewage treatment plant is located in the center of the harbor, and the bottom mud contains high levels of PCBs *(33)*. It was hypothesized that the oldsquaws feeding on the bottom organisms would reflect these high levels of PCBs.

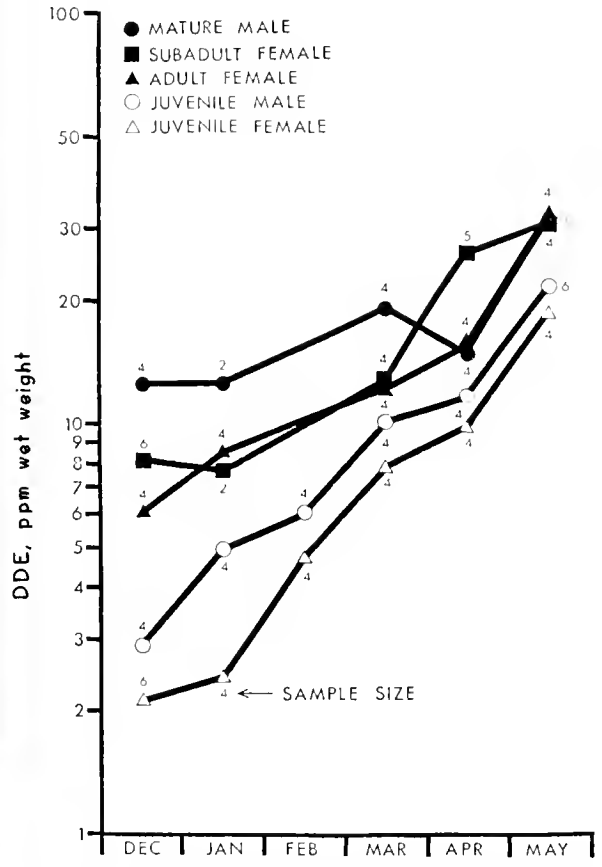

FIGURE 1. *Semilog plot of DDE residues in different age/sex classes of oldsquaws collected on Lake Michigan, 1970-71*

TABLE 3. *Mean and range of organochlorine pesticide and PCB residues in oldsquaws collected on Lake Michigan, 1972*

Sample Size	Age/sex class	Residues, ppm Wet Weight		
		p,p'-DDE	PCBs	Endrin
		JANUARY		
2	Mature male	19 2 13 0-26 4	50 30-70	0 4 0 2-0 5
2	Subadult female	9 7 6 3-13 0	18 11-25	0 7 0 4-1 0
		FEBRUARY		
2	Mature male	21 3 19 5-23 1	43 33-54	0 7 0 6-0 8
2	Adult female	8 2 5 3-11 1	18 13-23	0 6 0 5-0 8

TABLE 4. *Mean and range of organochlorine pesticide and PCB residues in oldsquaws collected in Milwaukee Harbor, January 28, 1972*

Sample Size	Age-Sex Class	Residues, ppm Wet Weight		
		p,p'-DDE	PCBs	Endrin
4	Mature male	12.2 4.5-27.3	97 60-122	<0.1 ND-0.1
2	Juvenile male	9.5 9.2-9.8	82 80-84	0.3 0.1-0.5
4	Adult female	6.9 3.1-11.5	89 78-130	<0.1 ND-<0.1
2	Subadult female	14.3 13.0-15.6	107 69-145	0.2 ND-0.3

Note: ND = not detected = no peak observed. All values were used in computing means; trace values were used as one-half the stated "less than" value and ND was used as zero.

Twelve birds were collected in January 1972 (Table 4); PCB residues averaged 5-8 times higher in these oldsquaws than in oldsquaws obtained from Lake Michigan during January 1971, and 2-6 times higher than in oldsquaws collected in January 1972. DDE residues were about the same as in Lake Michigan, and mature males and subadult females contained higher concentrations than juvenile males and adult females, as in the Lake Michigan data. Endrin occurred in relatively low concentrations in the Milwaukee Harbor samples, as in Lake Michigan samples.

Correlation of DDE in Wings and Carcasses—Several investigators have determined a significant relationship between organochlorinated compounds or mercury in wings and various tissues of birds (*5, 11, 34*). Authors hypothesized that if a smiliar relationship could be established in oldsquaws between the wing and carcass, average monthly residues could be more precisely measured by pooling large numbers of wings of a given age/sex class for a particular month. In addition, the number of determinations could be reduced if authors assumed that sufficient variability was incorporated into the sample.

Figure 2 illustrates the positive correlation ($P<0.001$) between DDE in oldsquaw wings and carcasses. Residues in the carcasses selected ranged from 0.4 to 32.0 ppm. The relatively high correlation coefficient ($r = 0.91$) indicates that oldsquaw wings could be used to monitor pesticide burdens in the carcasses.

Residues in Food—Table 5 presents monthly averages of DDE, PCB, and endrin residues in amphipods taken from oldsquaw gullets. The data suggest that residues are relatively constant between December and May when old-

TABLE 5. *Mean and range of organochlorine pesticide and PCB residues in amphipod samples taken from the gullets of oldsquaws collected on Lake Michigan*

Month	Year	Residues, ppm Wet Weight		
		p,p'-DDE	PCBs	Endrin
December	1969	0.62 0.56-0.68	1.26 1.18-1.37	0.04 0.04-0.04
December	1970	1.74 1.59-1.91	3.36 3.18-3.45	0.08 0.05-0.10
January	1971	0.87 0.60-1.23	1.66 1.15-2.38	0.06 0.05-0.08
February	1971	2.66 1.95-4.01	5.60 4.91-6.44	0.17 0.04-0.33
March	1970	1.21 1.04-1.50	2.84 2.02-3.69	0.08 0.07-0.10
March	1971	1.07 1.04-1.13	2.29 2.02-2.49	0.08 0.05-0.11
April	1971	0.81 0.12-1.28	2.57 2.23-2.82	0.06 0.05-0.08
May	1971	1.22 1.07-1.51	3.23 2.94-3.51	0.08 0.06-0.11
Overall average and range	1969-1971	1.27 0.12-4.01	2.85 1.15-6.44	0.08 0.04-0.33

NOTE: $n = 3$ for each monthly sample

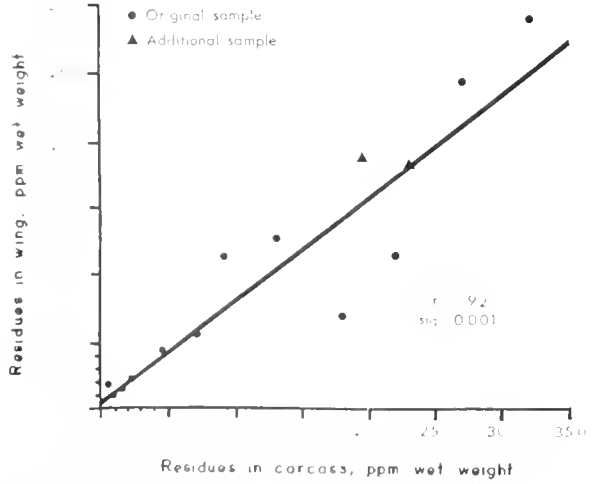

FIGURE 2. *DDE residues in wings and carcasses of mature male oldsquaws collected on Lake Michigan, 1970-71*

TABLE 6. *Biological magnification of organochlorine pesticide and PCB residues between amphipods and oldsquaws collected on Lake Michigan, 1970-71*

Month	Sample Size[1]	*p,p'*-DDE	PCBs	Endrin
December	A = 3 OS = 24	4	4	3
January	A = 3 OS = 16	8	8	8
February	A = 3 OS = 8	2	2	3
March	A = 3 OS = 20	12	13	4
April	A = 3 OS = 21	20	22	6
May	A = 3 OS = 19	21	21	3

[1]A = amphipods, OS = oldsquaws

squaws were collected for analyses. The apparent buildup of DDE and PCBs in oldsquaws throughout the winter is not reflected in the food sample data. Where there is replicate sampling for a certain month in successive years, the concentration of a particular toxicant varies from 0 to 280 percent.

Biological magnification factors between oldsquaws and their food were computed for each compound by dividing monthly averages of the residues in oldsquaws with corresponding averages for the food (Table 6). Since samples of each age/sex class of oldsquaws were not constant within and between months, the magnification factors should be considered rough approximations.

Several interesting trends appear in these data. Biological magnification from amphipods to oldsquaws is not constant but can vary from 1× to 22×, depending on the compound or the time of year. Magnification factors for DDE and PCBs are similar and appear to increase through the winter and spring from 4× in December to 21× in May, but residues of these compounds remained relatively constant in the food during the same period. The magnification factor computed for endrin did not vary so widely over time. Thus these data suggest that biological magnification factors computed for PCBs and DDE are not static, but dynamic, and it may be more appropriate to determine rates of increase as a function of time for specific species, compounds, and localities.

NORTHWEST HUDSON BAY

Ducks were collected during the breeding season at Rankin Inlet and Eskimo Point, Northwest Terriories (NWT) in 1972 to help interpret difference in DDE and PCB residues in December oldsquaws on the wintering grounds; to measure any loss in body-burden residues that may occur on the breeding grounds; and to examine the passage of pesticides from the female to the young. Even though comparisons were made between levels observed on the breeding ground and those recorded on Lake Michigan, it should not be construed that the same populations were sampled. Since oldsquaws winter in several areas and breed throughout the Arctic, breeding populations may be composed of birds from one or several wintering areas.

Breeding Pairs—Breeding pairs were collected on tundra ponds as soon as the ice thawed around the edges and birds moved inland from the sea. Average residues for each compound were similar in paired males and females (Table 7). PCBs averaged 25 ppm in males and 18 ppm in females; DDE averaged 6.4 ppm in males and 6.5 ppm in females. Endrin averaged less than 0.1 ppm in both paired males and females.

Figure 3 indicates a strong relationship ($r = 0.86$; $P<0.01$) betweeen the levels of DDE in the male and female of a given pair. Alison *(1)* showed that oldsquaws pair early on the wintering grounds. Pesticide data from the Arctic reported here indicate that pair bonds maintained throughout the winter and migratory periods expose members of a pair to similar levels of contamination. Lake Michigan data collected just before spring migration also indicate similar residue levels in mature males and females (Fig. 1).

DDE residues in breeding pairs suggest that most of those birds obtained in the Arctic probably did not come from Lake Michigan. Of the 20 birds collected on Lake Michigan in May 1971, the lowest level of DDE recorded was

TABLE 7. *Mean and range of organochlorine pesticide and PCB residues in oldsquaws collected at Eskimo Point, Diana River, and Rankin Inlet, Northwest Territory, 1971*

Class	Sample Size	Date	Residues, ppm Wet Weight		
			p,p'-DDE	PCBs	Endrin
Paired males	10	7-10 June	6.4 0.7-21.9	25 3-81	<0.1 ND-0.1
Paired females	10	7-10 June	6.5 0.6-19.8	18 3-44	<0.1 Tr-0.2
Females with clutches	11	29 June-11 July	4.7 0.1-16.0	24 1-95	<0.1 Tr-0.1
Composite clutches[1]	11	29 June-11 July	7.6 0.2-19.1	48 <1-172	0.1 ND-0.2
Females with broods	3	25 July-2 August	2.8 0.3-7.6	14 2-32	ND -
Composite broods[2]	3	25 July-2 August	2.1 0.2-3.1	25 1-63	<0.1 ND-<0.1
Molting females without broods	5	8 August	2.6 0.3-7.7	21 1-57	ND -
Subadult males[3]	4	10 July-8 August	2.9 0.6-6.8	15 <1-43	<0.1 ND-<0.1

Note. ND = not detected = no peak observed, - = range not calculated because no residue detected, all values were used in computing means, trace values were used as one-half the stated "less than" value and ND was used as zero

[1] Clutches contained 1-8 eggs

[2] Broods contained 3-5 young

[3] These males still retained their juvenile plumage from the preceding year. After the summer molt, they could not be distinguished from males 2 years or older

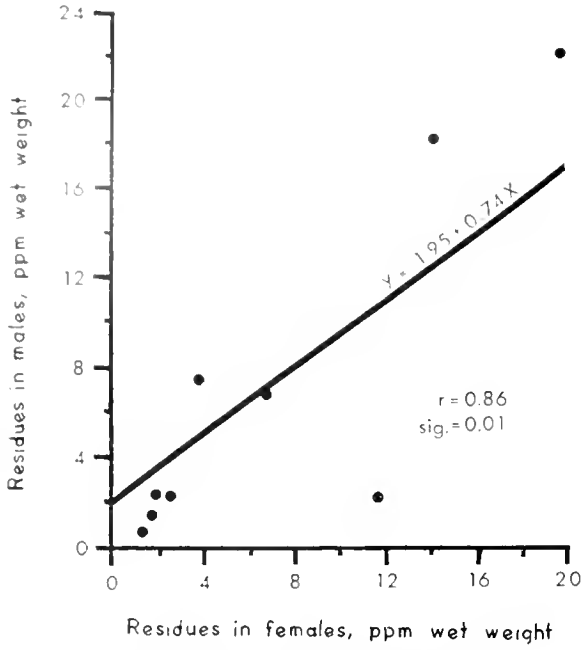

FIGURE 3. *DDE residues in pairs of oldsquaws collected at Rankin Inlet, Northwest Territory, June 1971*

13.7 ppm in a juvenile male, and the lowest in an adult was 16.4 ppm. Of the 20 birds collected as breeding pairs, only 4 (2 pairs) had DDE residues higher than 14 ppm, and 11 of the 20 paired oldsquaws had less than 5 ppm DDE. In addition, the breeding pair data exhibit a much wider spread between the highest and lowest DDE residues recorded (37×) than between mature oldsquaws collected on Lake Michigan during May 1971 (4×) or for the entire period from December to May (11×). This suggests that the breeding pair population collected at Rankin Inlet may have wintered in more than one area and these subpopulations were exposed to differing levels of DDE contamination

Females with Clutches—Female oldsquaws with their clutches were collected approximately 1 month after the breeding pairs had been obtained. Averages for DDE and endrin in the females with clutches were lower than for the paired females but these differences were not significant (Table 7; $P > 0.05$). Residues in composite clutches averaged higher than in the females that laid the clutches ($P < 0.05$); laying or incubating females averaged 4.7 ppm DDE, and their clutches contained an average of 7.6 ppm DDE. PCB residues averaged 24 ppm in the females and twice that amount in the eggs. For all three compounds, average residues in the clutches were higher than in the laying or incubating hens, paired hens, or paired males.

Figure 4 indicates a strong relationship ($r = 0.78$, $P < 0.01$) between DDE present in the clutch and the female collected with that clutch. Females with high levels of DDE tended to lay clutches with high DDE content, whereas hens with low DDE residues tended to transfer little of that compound to the egg.

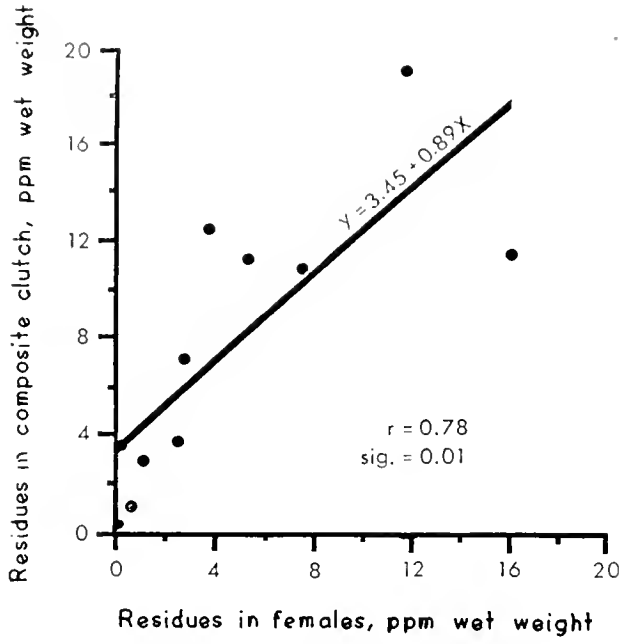

FIGURE 4. *DDE residues in female oldsquaws and their clutches collected at Rankin Inlet and Eskimo Point, Northwest Territory, June-July 1971*

The eggshell thickness index discussed by Ratcliffe *(26)* was used to measure differences between eggs reported here and those eggs collected before the heavy use of DDT. The mean eggshell thickness index for 106 oldsquaw eggs collected before 1947 was 1.55±0.01; the average eggshell index for 75 eggs collected in 1971 was 1.48±0.02. This was a decline of 4.5% ($P < 0.005$).

Females with Broods—Three broods with accompanying females were collected approximately 1 month after incubating and laying females had been collected. Average residue content in females with broods was lower than in females collected previously on nests (Table 7), but again the differences were not significant. The limited brood sample did not show a positive correlation between duckling residues and the female as did the clutch and female data, but average residues were lower in broods than in clutches.

Other Collections—On August 8, 1971, authors collected five adult females molting on the sea near the mouth of the Diana River, NWT. These females were part of a group of approximately 200 other oldsquaws, all adult females. Average organochlorine pesticide and PCB residues in this sample fell between those in females with broods collected 1-2 weeks earlier and those in females with clutches collected 1 month earlier (Table 7). Since no broods were observed in the flock, either these females had not bred that year, or they had lost their clutches to predation. By this time, the ovaries of the molting females had regressed to normal winter sizes recorded on Lake Michigan.

A few yearling males were observed throughout the summer, and they were collected when the opportunity arose (Table 7). Average residues in these males were similar to those of adults collected in the Arctic but lower than residues in juvenile males collected on Lake Michigan in May 1971.

Little food was found in the gullets of recently arrived breeding pairs and females with clutches. The liver and entire gastrointestinal tract were quite small, indicating reduced use. Esophageal contents from birds collected in the late summer provided material for comparing organochlorine residues in the Arctic summer foods of oldsquaws with those found in the primary food of oldsquaws wintering on Lake Michigan. PCB residues were the highest (Table 8), but only one-tenth as high as the average residues found in Lake Michigan food samples. No endrin was detected. These data indicate that there is some contamination of the oldsquaw food base in the Arctic habitat sampled, but the level is much lower than the exposure level on Lake Michigan during the winter.

OTHER WINTERING AREAS

Oldsquaws were obtained from five wintering areas other than Lake Michigan. These collections were made in different years or at different times in the same year, depending on when cooperators could obtain specimens. Not all age/sex classes analyzed on Lake Michigan are represented in these samples. Consequently, only broad generalizations have been made from the data presented in Table 9.

Specimens collected on Lake Huron and Lake Ontario exhibit pesticide profiles similar to those recorded for oldsquaws from Lake Michigan; the orders of magnitude are also similar. Oldsquaws from Maine and Newfoundland averaged lower levels of each pesticide than did similar samples from the Great Lakes during the same time period, and those birds secured from Alaska in February contained the lowest levels of any given pesticide for all areas sampled. Endrin was not found in the eastern collections and occurred as a trace in one of the 10 specimens analyzed from Alaska.

In general, these data suggest a continuum in contamination of wintering oldsquaw populations. Those populations wintering in heavily polluted waters such as the Great Lakes contain high residues of PCBs and DDE. Populations that winter in coastal areas near highly developed urban and industrial centers contained lower residues. Oldsquaws wintering in an essentially pelagic environment far from major sources of pollution had low or undetected residue levels.

TABLE 8. *Mean and range of organochlorine pesticide and PCB residues in gullet samples of oldsquaws collected at Eskimo Point and Rankin Inlet, Northwest Territory, June-July 1971*

Compound	Residues, ppm Wet Weight	
	Mean ($n=5$)	Range
p,p'-DDE	0.02	0.006-0.060
PCBs	0.28	0.160-0.580
Endrin	0.00	

Discussion

Few studies have reported pesticides in oldsquaws or other ducks from Arctic areas. Five oldsquaws collected on February 24, 1964 from Lake Michigan had pesticide residues similar to those described here *(16)*. Low levels of DDE and PCBs were recorded in the neck fat of ducks off Greenland: oldsquaws contained 0.8-1.3 ppm, eiders *(Somateria spectabilis* and *S. mollissima)* contained 0.8-2.8 ppm; and harlequin ducks *(H. histrionicus)* contained 0.7-1.9 ppm DDE and PCBs *(3)*.

Various investigations present conflicting evidence as to whether DDE reaches an asymptote or continues to increase with continuing exposure. Dindal *(6)* found that DDE leveled off in mallard *(Anas platyrhynchos)* and lesser scaup *(Aythya affinia)*. Ludke *(23)* found that DDE peaked in *C. coturnix* if this was the only compound present, but if fed in conjuction with dieldrin, neither compound reached equilibrium by the end of the 56-day experiment. Data collected here indicate that DDE continued to accumulate in oldsquaws as long as they remained on Lake Michigan.

Studies on mallards and black ducks have shown that a diet containing 10 or more ppm DDE (dry weight) increased the incidence of cracked eggs, thin shells, and embryo and duckling mortality. In mallards, the ingestion of 40 ppm DDE caused the same phenomena, and they occurred earlier than in the ducks that had ingested lower levels of DDE *(13)*. Black duck females dosed with 10 ppm and 30 ppm DDE (dry weight) produced similarly detrimental effects, and the eggs averaged 46.3 ppm and 144.1 ppm DDE, respectively *(22)*. These data are similar to those reported for the brown pelicans *(Pelecanus occidentalis)* on Anacapa Island, California, when this species was in serious reproductive trouble *(21)*.

Data concerning detrimental reproductive effects in the oldsquaws can be compared with the above studies on mallards and black ducks only indirectly because residues in females that produced the observed levels in eggs were not determined and since dosages less than 10 ppm DDE were not used, the point at which statistically measurable detrimental effects appear in waterfowl is unknown. The eggs collected in the Arctic in this study had relatively thick shells; the thickness index averaged only 4.5% less than in eggs collected before the heavy use of DDT. Eggs collected in 1971 averaged only 7.6 ppm DDE. These data suggest that the species is not in any serious reproductive

TABLE 9. *Mean and range of organochlorine pesticide and PCB residues in oldsquaws collected from wintering ground in the United States and Canada, other than Lake Michigan, 1971-73*

Date	Age-Sex Class	n	Residues, ppm Wet Weight		
			p,p' DDE	PCBs	Endrin
LAKE HURON (SAGINAW BAY)					
11 May 1971	Mature male	2	19.4 7.4-31.3	62 47-77	0.3 0.2-0.4
	Mature female	1	21.9	50	0.4
	Juvenile male	2	8.2 5.7-10.7	21 20-22	0.2 0.2-0.3
	Juvenile female	0	-	-	-
LAKE ONTARIO (PRESQU'ILE POINT)					
23 Nov-13 Dec 1972	Mature male	1	5.7	32	<0.1
	Mature female	4	1.9 0.6-2.2	21 3-35	<0.1 <0.1-<0.1
	Juvenile male	4	1.2 0.9-2.2	9 3-20	0.1 <0.1-0.2
	Juvenile female	4	1.6 1.0-2.5	9 5-15	0.1 <0.1-0.3
MAINE (PENOBSCOT BAY)					
11 Mar 1972	Mature male	6	1.7 0.4-5.5	9 2-25	ND -
	Mature female	5	0.6 0.3-0.8	4 2-5	ND -
	Juvenile male	0	-	-	-
	Juvenile female	0	-	-	-
NEWFOUNDLAND (ST. JOHN'S)					
1-5 May 1971	Mature male	2	0.3 0.2-0.4	1 1-1	ND -
	Mature female	2	0.2 0.2-0.3	1 1-1	ND -
	Juvenile male	2	0.2 0.2-0.2	1 1-1	ND -
	Juvenile female	2	0.1 0.1-0.1	1 1-1	ND -
ALASKA (POINT BAKER)					
18-23 Feb 1973	Mature male	4	0.1 0.1-0.2	1 <1-1	Tr ND-Tr
	Mature female	4	0.1 0.1-0.2	<1 <1 - 1	ND
	Juvenile male	1	- 0.1	- 1	ND
	Juvenile female	1	0.1	- 1	ND

NOTE: ND = not detected; no peak observed. Tr = trace = 0.005 ppm; - = no specimens collected. All values were used in computing means, trace values were used as one-half the stated le- than- value and ND was used as zero.

trouble. However, few females collected in the Arctic had DDE levels similar to those in ducks collected on Lake Michigan in May. If it is assumed that detrimental reproductive effects are similar in black ducks and oldsquaws with similar body burdens, and that the data in Figure 4 correlating DDE in the female and clutch are similarly proportional for hens with higher concentrations of DDE, then the equation of the line predicts that it would take a female oldsquaw with approximately 48 ppm DDE in her body to produce a clutch averaging 44 ppm DDE. Adverse

effects on black duck reproduction have been recorded at this level *(22)*. Since one of the four adult female oldsquaws collected on Lake Michigan in May contained 58.3 ppm DDE, these data suggest that some of the oldsquaws wintering on Lake Michigan may have reproductive problems.

PCBs were relatively high in Lake Michigan oldsquaws. Several studies have shown seriously adverse effects of PCBs on reproduction in chickens, but up to 50 ppm PCBs have not caused any measurable reproductive effects or eggshell thinning in mallards *(14, 28)*.

Body fat stores various organochlorine compounds and reduces the possibility of immediate toxic effects *(32)*. If lipid stores remain high, these chemicals can be eliminated by gradual mobilization and excretion. Studies on changes in oldsquaw body weights indicate that body lipids in Lake Michigan oldsquaws are highest (19-24 percent of carcass weight) in December-January when the pesticide body burden is relatively low, and also in May (23-25 percent of carcass weight) just before migration when pesticide body burdens are relatively high *(7, 24)*. There are two periods when oldsquaws have relatively little fat. During April, oldsquaws on Lake Michigan average only 7-9 percent lipids, and in early August, adult females with newly hatched broods averaged less than 2 percent fat. Residues in brain tissue were not measured, but if high concentrations of pesticide are present and mobilized when lipids are reduced, then April and August could be critical months. Massive mobilization of lipids and the stored pesticides can be similar to the experimental situation in which a high acute oral dose of DDT induced 25 percent thinning in mallard eggshells *(30)*. If, for lack of food or other physiological reasons, oldsquaws do not accumulate high lipid levels before migration from Lake Michigan, stored residues could be lethal to the breeding female as well as to the developing embryo.

Studies on food habits of the oldsquaw indicate that the diet of this species is 99 percent animal matter and that this is primarily an amphipod, *Pontoporeia affinis (7,25)*. Studies relating pesticide concentration to diet have generally shown that animal feeders and those species high on the food chain exhibit high concentrations of organochlorine compounds *(6, 8)*. Oldsquaws wintering on Lake Michigan between 1969 and 1972 were on a continuous diet of amphipods containing an average of 1.27 ppm DDE, and adult female oldsquaws averaged 32.1 ppm DDE just before spring migration in 1971. Hickey et al. *(16)* found that DDE residues in *P. affinis* varied between 0.19 ppm and 0.24 ppm in July 1964. This is considerably lower than the estimates of average levels in amphipods reported here, but their calculated concentration factor of 15× in February between *P. affinis* and oldsquaws is within the range of 4× (December) to 21× (May) reported in this paper.

Even though oldsquaws and whitefish in Lake Michigan eat the same organisms and are, therefore, on the same trophic level, residues reach much higher levels in the birds than in the fish. DDE residues in whitefish reported by Hickey et al. *(16)*, Reinert (DDE-TDE-DDT) *(27)*, and the Wisconsin Department of Natural Resources (WDNR; unpublished data) were 3.00, 0.78, and 0.64 ppm, respectively, whereas DDE averaged 6.1 ppm for all oldsquaws collected in December 1970, and 25.6 ppm in May 1971. PCBs averaged 4.3 ppm in the Wisconsin whitefish, and December and May averages for all oldsquaws were 12.5 ppm and 67.2 ppm, respectively. It seems reasonable to expect higher levels in birds than in fish that eat the same food, because birds are warm-blooded and would require more energy than would fish.

The relationship of age and sex with the accumulation of pesticides in avian species has been little studied. Young nonbreeding birds show only minimal sex-dependent differences in susceptibility, but this might not hold for mature birds during the breeding season *(31)*. Experimental LD_{50} values for DDT have been shown to vary with age in mallards *(9)*, and DDE has appeared to be higher in male than in female mallard wings although the differences have not been significant *(15)*. It has been hypothesized that the greater resistance of adult females versus adult males to pesticide toxicity was partly a result of the females' ability to excrete pesticides through their eggs *(9, 17)*. Early winter differences in DDE residues among different age and sex classes of oldsquaws, as well as the positive relationship between DDE in the female and clutch, suggest that breeding female oldsquaws can eliminate pesticides through the egg and return to wintering areas with a lower body burden than can nonbreeding females or males that presumably have bred.

A number of studies have pointed out regional differences in the pesticide burdens of various avian species and populations. Because of differences in age, dietary habits, and possibly winter migration to more heavily polluted areas, the amount of polychlorinated hydrocarbons may vary from one species to another *(3)*. Keith *(19)* summarized regional differences in pesticides when he stated: "Early results show truly astonishing regional within-species variations in residue loads, and these require explanation presumably in terms of local pesticide use patterns and local food preference." The nationwide pesticide monitoring program conducted on mallard and black duck wings also showed State and regional differences in DDE and PCB levels *(12, 15)*. Data from the present study concerning the Great Lakes as well as Atlantic and Pacific coastal areas support these conclusions concerning regional differences. The relatively low levels in oldsquaws wintering in estuaries or pelagic areas confirm an early prediction by Butler and Springer *(4)* that ". . . because of rapid dilution rates, high pesticidal concentrations are not to be anticipated in estuaries."

Only about 200 oldsquaws have been banded in North

America outside Alaska (unpublished data, Migratory Bird Population Station, Laurel, Maryland, 2). Consequently, little is known of the migratory movements of this species. Oldsquaws primarily winter on the Great Lakes and in coastal areas of the northern United States and southern Canada (18). Since marked differences in pesticide residues appear in oldsquaws collected from these areas, it may be possible to use characteristic pesticide profiles of these compounds in place of leg bands to trace movements of the birds. Thus the wintering grounds of different breeding birds might be determined, at least approximately, by comparing the amounts, kinds, and ratios of pesticides found in various populations.

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Mercury, Cadmium, Lead, and Arsenic in Sediments, Plankton, and Clams from Lake Washington and Sardis Reservoir, Mississippi, October 1975-May 1976 [1]

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ABSTRACT

Mercury, cadmium, lead, and arsenic concentrations were measured in clams, plankton, and sediments from Lake Washington, Washington County, an oxbow lake off the Mississippi River, and Sardis Reservoir, Lafayette and Panola Counties, a flood control reservoir on the Little Tallahatchie River. Mercury, cadmium, and lead were measured by atomic absorption spectrophotometric methods, arsenic was measured by the silver diethyldithiocarbamate method. Correlation coefficients for concentrations of metals in the sediments, plankton, and clams were derived. Mean concentrations for all metals were higher in plankton and clams than in sediment, although levels were only slightly higher in clams than in sediments. Plankton contained the highest levels of all four metals. There were no significant correlations between the metals for sediments or clams. A t-test used to compare concentrations of the four metals in the sediments of the two bodies of water showed a significant difference only for arsenic concentration. A high correlation of mercury between plankton and clams indicates the food web relationships. There was also a positive correlation of cadmium between sediments and clams, and a correlation of arsenic between sediments and plankton. Comparisons between clam size and trace metal concentrations showed higher concentrations of mercury in the smaller clams.

Introduction

During the past 23 years, trace metal pollution in aquatic systems has been researched extensively. This was brought about by the tragedy in Minamata City, Japan in 1953 in which 41 persons died of what was subsequently shown to be mercury poisoning (*19, 15, 16*). Effluent from a factory where $HgCl_2$ was used as a catalyst in the production of vinyl chloride was discharged into Minamata Bay; as a result, fish and shellfish from the bay, which were a major source of food for the villagers, accumulated high levels of mercury. Since the Minamata City tragedy and the realization of the toxicity of low concentrations of heavy metals, data concerning mercury pollution have increased, and other elements such as lead, cadmium, and arsenic are being investigated.

The occurrence of trace elements in the biosphere at higher levels than in the hydrosphere has been recognized for some time (*23, 25, 6*), but the order of magnitude among organisms varies considerably. The mechanisms by which trace metals are concentrated by organisms are still not fully understood, but there is evidence that accumulation occurs through the food chain (*23, 12*).

Because of their potential toxicity to humans and their persistence in the environment, certain heavy metals which accumulate through the food chain are of particular interest. The magnifying effects at the higher trophic levels make higher organisms particularly vulnerable to these elements. Radionuclides, chlorinated hydrocarbons, and trace metals pose a serious problem in the aquatic environment because they are concentrated from the water into sediments and the food web (*18*).

It has been shown that Pelecypoda feed on plankton (*1, 20*) and can selectively concentrate pollutants against a gradient (*23*). Although certain materials are sorted before entering the stomach, all particles, including great quantities of sediment, are retained by the gills (*8*); stomach contents also contain detectable quantities of sediments (*13*).

In this study, mercury, cadmium, lead, and arsenic were measured and compared in sediments, clams, and plankton from Lake Washington and Sardis Reservoir, Mississippi, to study food web relationships.

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Description of Sardis Reservoir and Lake Washington

Sardis Reservoir is part of a long-range plan for flood control of the Yazoo River Basin. The dam and appurtenances were developed by the U.S. Army Corps of Engineers in 1940. At full capacity, the reservoir has a surface area of 237 km^2 with an average depth of 5.5 m. Water level fluctuates considerably as water is periodically drained; at the lowest water level, the conservation pool is about 43 km^2.

Lake Washington is an oxbow off the Mississippi River, located 32 km south of Greenville, Mississippi. It is a large shallow lake occupying 20 km^2 in Washington County. One-fourth of the lake contains cypress trees in about 1 m of water. Water level fluctuates no more than 2 m; the high level occurs in the spring and the low level occurs in the fall.

Fish from Lake Washington contained pesticide residues at concentrations above the action level of the Food and Drug Administration, U.S. Department of Health, Education and Welfare (9), thus the lake is presently closed to commercial fishing and the public has been warned against consuming fish from the lake.

Materials and Methods

Samples were collected monthly from October 1975 through May 1976. Three stations on each body of water were sampled each month; a fourth station was sampled intermittently (Fig. 1, 2). Samples of the upper 6 cm of sediment were collected as near as possible to the sites of the plankton samples.

Clam, sediment, and plankton samples were placed in appropriate containers and frozen on the day of collection to ensure that no trace metals would be lost to volatilization.

Plankton samples were collected by using a pump and hose apparatus and were concentrated by using a No. 25 plankton net. The pump consisted of five individual marine and camping 12-volt dc pumps arranged radially symmetrical on a 3.1-mm-thick Plexiglas disk 40 cm in diameter.

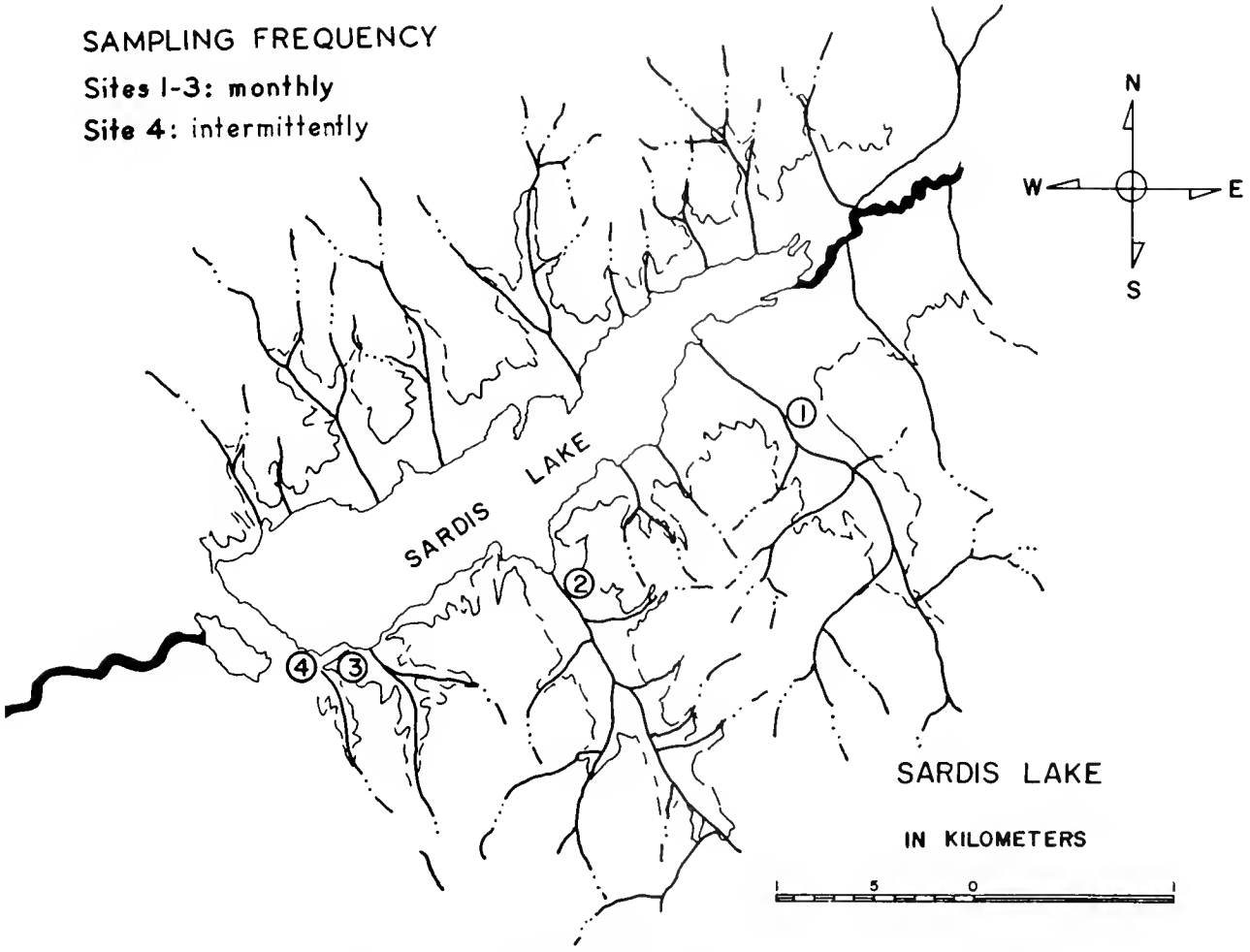

FIGURE 1. *Sardis Reservoir (34° 30' north latitude, 89° 40' west longitude), showing selected sites for collecting sediments, plankton, and clams for heavy metal determinations*

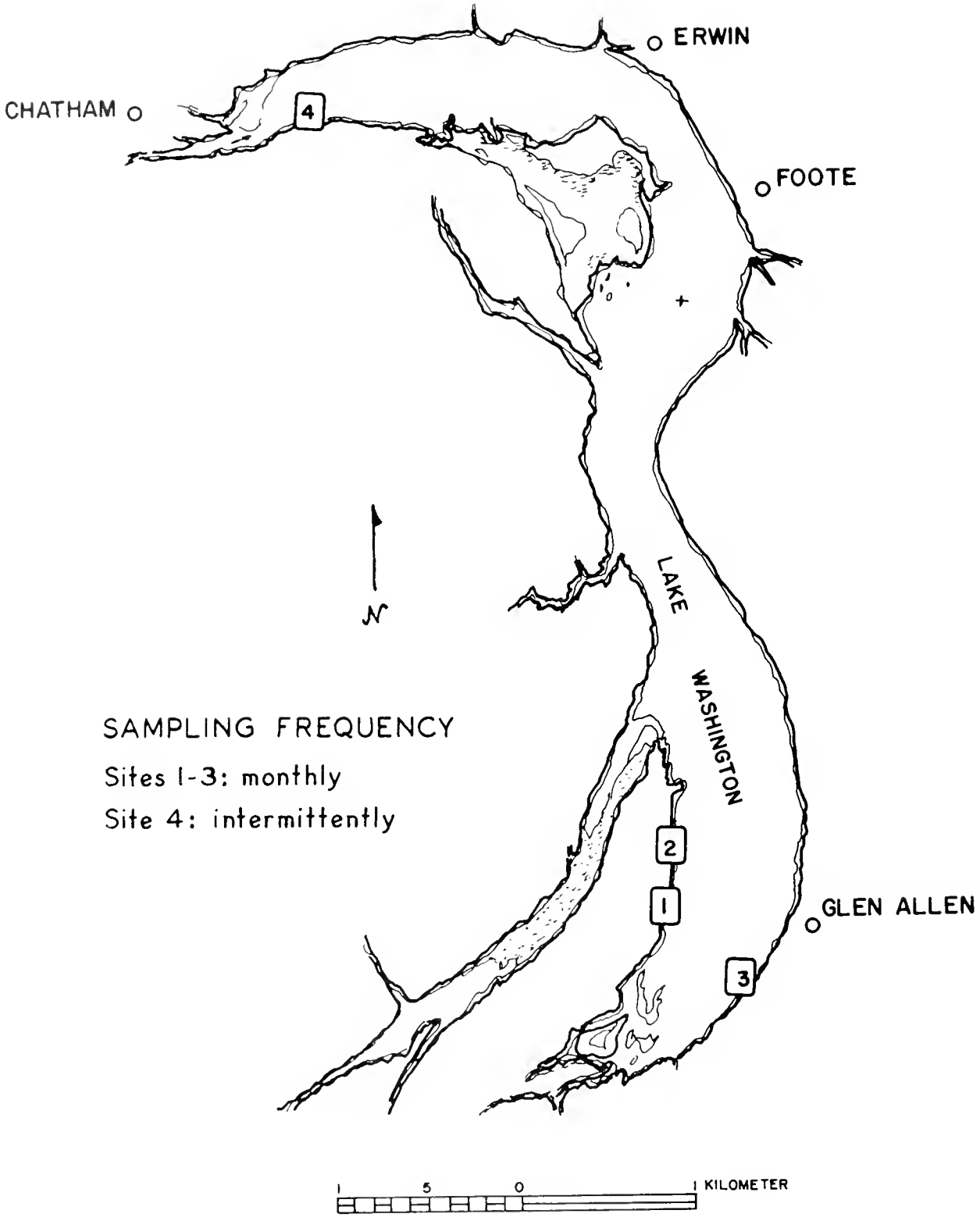

FIGURE 2. *Lake Washington (33° 05' north latitude, 91° 05' west longitude), showing sites selected for collecting sediments, plankton, and clams for heavy metal determinations*

Incurrent holes of the pumps were located 11.7 cm from the periphery of the disk. Another disk of the same size was placed 1.9 cm below the first disk and mounted on feet which were 9.5 cm long. Thus plankton samples were obtained 10.0 cm above the bottom. Because the disk was parallel to the bottom during operation, the water was pulled in horizontally. All pumps were joined in the center to a 3.8-cm hose which led to the surface.

The plankton pump was calibrated before and after each run in the time required to fill a container with 20 liters of water; then the amount of water filtered by the net was calculated.

A compound microscope equipped with a draw tube, 10×-ocular, and 16-mm objective was used in combination with a Whipple ocular micrometer for plankton enumeration. An aliquot was placed in a Sedgwick-Raftar counting cell, and field counts were made. For filamentous forms, a length of 300 μ was considered a unit. All other forms, including individual cells and colonies, were counted as single units. Plankton counts are summarized in Table 1.

Clams were collected by hand with a garden rake or by a dredge constructed after a model from Burch (*7*).

Samples were analyzed for mercury by the flameless atomic absorption technique developed by Hatch and Ott (*14*). Samples were digested with 5 ml of concentrated nitric acid and 5 ml of concentrated sulfuric acid for 12 hours, and then reduced with 5 ml of potassium permanganate and 5 ml of potassium persulfate for 12 hours. The reaction flask was placed in the closed system, and stannous sulfate in a sodium chloride-hydroxylamine medium was added to reduce the mercury to its elemental state. Once maximum absorbance was obtained, the mercury vapor was vented to the hood. Standards were analyzed by the same procedure. The standard values were statistically corrected by the method of least squares (*2*) and were used to obtain a calibration curve. A Beckman Model 1301 atomic absorption spectrophotometer equipped with a 10-inch potentiometric recorder was used for the analysis.

Sediment samples for lead, cadmium, and arsenic measurements were dried in a vacuum oven at 105°C for 24 hours. Plankton samples were dried only for arsenic determinations.

In order to use larger sample sizes of plankton, mercury, cadmium, and lead analyses were all conducted on the same sample. Samples were first analyzed for mercury, and then filtered and prepared for cadmium and lead analyses by the method of additions (*22*). Blanks were used to compensate for contamination by chemicals. Final results were obtained by the method of least squares and are reported as ppm wet weight.

Clams were also analyzed for cadmium and lead by the method of additions to eliminate matrix effects. Clam soft tissues were digested with nitric acid on a hot plate until dry. Charring of samples was minimized by adding more nitric acid. After each sample cooled, 5 ml of concentrated hydrochloric acid was added. Samples were then diluted to 25 ml with distilled water, and filtered. Five-ml aliquots were used for the method of additions.

Sediment samples for cadmium and lead analyses were digested in nitric acid to dryness on a hot plate. The residue was dissolved in concentrated hydrochloric acid, diluted to volume, and aspirated into the atomic absorption apparatus. Absorbance was compared to a standard curve obtained by the method of least squares. Results are expressed as ppm wet weight.

For arsenic analysis, various combinations of acids were tried for digestion of samples, but a 4:1:1 ratio of nitric, sulfuric, and perchloric acids (*10*) provided the best results. This mixture was added to the sample and heated until fumes of SO_3- appeared. The sample was allowed to cool and then 5 ml of concentrated hydrochloric acid was added and diluted to 35 ml with distilled water The samples were then subjected to arsine generation as described by the American Public Health Association (*2*). Standards were analyzed with each set of samples. Percent transmittances obtained from the spectrophotometer were converted to absorbance to obtain a working curve. The method of least squares was used to plot a straight line from which concentrations could be obtained. Results are expressed as ppm wet weight.

Methods, instrumentation, sensitivities, ranges, mean recoveries, and total number of analyses are presented in Table 2.

Concentration data for the four metals were arranged in three groups: one group of sediment data only (N=53, where N represents sample size), one group of sediment and plankton data (N=25), and one group of sediment, plankton, and clam data (N=19). Correlation coefficients and a *t*-test were run on each group to ascertain relationships and significant differences between the two bodies of water.

TABLE 1 *Ranges and means for net plankton counts from Sardis Reservoir and Lake Washington, Mississippi*

	Net Plankton Counts	
Statistic	Sardis Reservoir	Lake Washington
Range	493,083 - 1603	1432 - 45
Mean	106,495	1308
N	21	23

TABLE 2. *Instrumental and analytical parameters for determining mercury, cadmium, lead, and arsenic in sediments, plankton, and clams*

Parameter	Metal			
	Hg	Cd	Pb	As
Method	Flameless AAS	Flame AAS	Flame AAS	Silver diethyldithiocarbamate
Instrument	Beckman Model 1301	Perkin-Elmer Model 303	Perkin-Elmer Model 303	Spectronic 20
Sensitivity, ppm	0.0005	0.20	0.40	0.5
Range, ppm	0.0005-0.020	0.05-5.0	4-40	1-10
Mean recovery (percent)[1]	97.0	99.8	101.6	76.5
Number of analyses	275	203	203	284

[1] Data are not corrected for percent recoveries.

TABLE 3. *Mean concentration of heavy metals in sediment, plankton, and clams from Lake Washington and Sardis Reservoir, Mississippi*

Sample	Metal			
	Hg	Cd	Pb	As
Sediment	0.0481	0.3614	8.4847	2.99
Plankton	0.4551	14.4587	501.5964	21.74
Clams	0.1073	0.2123	11.5685	0.41

NOTE: Concentrations in ppm wet weight. N = 19

Results and Discussion

A *t*-test on the group containing 19 cases with complete samples, i.e., sediment, plankton, and clam data, showed no significant difference between the two bodies of water. These results were pooled to obtain one value to show any food web relationships (Table 3). In all cases, mean concentrations were higher in the plankton and clams than in the sediment.

Mercury may have been biomagnified, with increased concentrations from sediments to plankton and clams. The higher levels in plankton compared to clams may be due to the trophic conditions of the water (*17*). According to Smith and Green (*25*), water must be severely polluted before mercury levels increase in clams. The habitats in the present study are relatively uncontaminated; this could explain why the clams contained only slightly higher levels of mercury than did the sediments.

Frazier stated that cadmium uptake by oysters is direct and independent of the food web and that clams also have been used as indicators of cadmium levels in their environment (*11*). This suggestion of bioconcentration is illustrated by the similar concentrations of cadmium in sediments and clams. Plankton are known to accumulate cadmium; this is supported by the results of the present study.

Background levels of lead have been widely reported, but no single value is universally accepted. Uptake of lead by plankton through surface adsorption has been demonstrated (*21*), and this is shown in the present study. Remarkably uniform concentrations of lead in mussels have been reported (*4*) and are representative of the particular environment in which they are found (*23, 5, 24*). Clams in Sardis Reservoir and Lake Washington appear to concentrate lead only to levels barely exceeding those in sediment.

Plankton are known to accumulate and magnify arsenic to concentrations above that found in the water (*36*). The large amounts of arsenic in plankton in the present study illustrate this point. Arsenic data for Mollusca are scarce, and higher organisms are not known to accumulate arsenic (*26*). The low value in this study indicates little concentration of arsenic by Mollusca.

A *t*-test with only sediment values for the two bodies of water showed a significant difference between arsenic in the sediments: 3.6 ppm in Sardis Reservoir and only 2.4 ppm in Lake Washington.

When arsenic enters the aquatic environment, it settles and accumulates in the sediments (*26*); levels in sediments have been used for water appraisal (*3*). The difference between the two lakes may be explained by the large amount of runoff which Sardis Reservoir receives.

With sediment data (Table 4), a correlation matrix used to ascertain relationships among the four metals showed one significant value ($P > 0.50$): mercury with date of sample. This may indicate some seasonal cycle. Nonsignificant values for the rest of the matrix indicate no correlations between the metals in the sediments.

A *t*-test for heavy metals in clams from the two lakes revealed that there were no significant differences between the two groups. The absence of significant differences suggests that the mode of uptake of the metals examined for the clams from the two bodies of water must be similar.

TABLE 4. *Correlation matrix for heavy metals in sediment*

Metal	Hg	Cd	Pb	As
Hg	1 00			
Cd	-0 16	1 00		
Pb	0 15	0 23	1 00	
As	0 32	0 06	0 34	1 00
Date	0 61	-0 30	0 06	0 29

NOTE N=53

TABLE 5. *Correlation matrix for heavy metals in clams*

Metal	Hg	Cd	Pb	As
Hg	1 00			
Cd	-0 09	1 00		
Pb	-0 22	-0 09	1 00	
As	-0 10	-0 00	0 02	1 00
Date	0 33	-0 19	0 02	-0 24

NOTE N=19

TABLE 6. *Correlation matrix for heavy metals in plankton*

Metal	Hg	Cd	Pb	As
Hg	1 00			
Cd	0 98	1 00		
Pb	0 98	0 96	1 00	
As	-0 12	-0 12	-0 06	1 00
Date	-0 28	-0 26	-0 24	0 07

NOTE N=25

TABLE 7. *Correlation coefficients for heavy metals in sediments, plankton, and clams*

Material	Hg	Cd	Pb	As
Sediment/plankton	-0 08	-0 17	-0 06	0 45
Plankton/clams	0 45	-0 20	-0 12	0 31
Sediment/clams	-0 39	0 16	-0 07	0 13

NOTE N=19

Concentrations of heavy metals in clams from the two bodies of water were then pooled for correlation analysis of the heavy metals. The correlation matrix (Table 5) for the clams showed no significant values, indicating that there were no reciprocal relations between the metals in the clams.

The correlation matrix for heavy metals in the plankton (Table 6) shows some high correlations between cadmium and mercury, cadmium and lead, and mercury and lead. These unexpected high values are due to experimental design: all three analyses were conducted on the same sample. A *t*-test on this group of data revealed no significant difference in metal content in the plankton from either body of water.

The correlation coefficients for heavy metals in sediments, plankton, and clams (Table 7) further illustrate some trends. A high correlation of mercury between plankton and clams indicates the food web relationships of the two. The positive correlation of cadmium between the sediments and clams indicates their relationship as mentioned earlier. Lead shows no particular trends with this matrix. Arsenic displays the sediment/plankton relationship as discussed earlier.

Data were regrouped to test for possible relationships of metal concentration with the various species of clams (Table 8). Eight species were analyzed: *Corbicula manilensis* (Philippi), *Proptera purpurata* (Lamarck), *Anodonta grandis corpulenta* (Say), *Anodonta suborbiculata* (Say), *Anodonta grandis* (Say), *Carunculina parva* (Barnes), *Quadrula pustulosa* (Lea), and *Lampsilis anodontoides* (Lea). Of these, *A. grandis*, *Q. pustulosa*, and *L. anodontoides* came from Lake Washington. *A. g. corpulenta*, *C. manilensis*, and *P. purpurata* came from Sardis Reservoir. *A. suborbiculata* and *C. parva* were obtained from both lakes. This provided an even distribution of species comparison.

Smaller clams, i.e., *C. parva*, *Q. pustulosa*, *C. manilensis*, and some *L. anodontoides*, contained higher levels of mercury than did the larger clams. Since smaller organisms have higher surface-to-volume ratios, they would be expected to have greater uptake.

Cadmium appeared to be concentrated in approximately the same amounts regardless of species.

Clams from Lake Washington contained higher levels of lead than did *A. g. corpulenta*, *C. manilensis*, or *P. purpurata* from Sardis Reservoir. These higher levels may be due to the richer trophic condition and the age of Lake Washington.

Arsenic levels were consistently low in all species, reinforcing the theory of little or no arsenic accumulation by Mollusca.

Summary

A significant correlation of mercury in the sediments with date of samples indicates some seasonal trends.

The *t*-tests revealed no significant differences between Lake Washington, an oxbow lake, and Sardis Reservoir, a flood control reservoir, with respect to the metals in the

TABLE 8. *Ranges and means of heavy metals in clams from Lake Washington and Sardis Reservoir, Mississippi*

Species of clam	Residues, ppm wet weight			
	Hg	Cd	Pb	As
Proptera purpurata				
Range	0.000-0.034	0.000-0.800	0.000-6.910	0.00-0.72
Mean	0.017	0.311	3.396	0.19
Carunculina parva				
Range	0.000-0.463	0.000-1.170	0.000-89.262	0.00-1.03
Mean	0.043	0.288	9.433	0.22
Anodonta suborbiculata				
Range	0.000-0.012	0.031-0.380	0.000-1.023	0.00-1.93
Mean	0.001	0.175	0.544	0.30
Anodonta grandis corpulenta				
Range	0.000-0.011	0.000-0.521	0.000-1.167	0.00-0.36
Mean	0.005	0.120	0.330	0.11
Quadrula pustulosa				
Range	0.000-0.432	0.000-0.243	0.000-31.857	0.00-1.39
Mean	0.087	0.086	8.777	0.36
Lampsilis anodontoides				
Range	0.000-0.149	0.000-0.212	0.585-6.360	0.00-0.54
Mean	0.058	0.160	3.313	0.06
Anodonta grandis				
Range	0.000-0.166	0.000-0.547	0.000-33.847	0.00-0.32
Mean	0.008	0.230	5.082	0.10
Corbicula manilensis				
Range	0.000-0.218	0.000-0.235	0.000-1.429	0.30-0.33
Mean	0.063	0.151	0.614	0.31

biosphere, although there was a significant difference in arsenic content of the sediments.

Small clams contained higher levels of mercury than did large ones.

Clams from Lake Washington contained higher levels of lead than did Sardis clams.

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RESIDUES IN WATER

Residues of Polychlorinated Biphenyls, DDT, and DDT Metabolites in Pennsylvania Streams, Community Water Supplies, and Reservoirs, 1974–76 [1]

David A. Kurtz [2]

ABSTRACT

Pennsylvania streams, community water supplies, and open water reservoirs were analyzed for PCB mixtures, DDT, and DDT metabolites. Streams were sampled in 1974 and again in 1975. Only 4 of 19 stream locations were contaminated. Maximum concentrations in community supplies of Aroclor 1242 (2 locations), Aroclor 1254 (3 locations), and ΣDDT (2 locations) were 350, 260, and 620 ng/kg, respectively. Of the 110 community water supplies sampled in 1975 and 1976, only 7 contained residues. Maximum concentrations in community supplies of Aroclor 1242 (4 locations) and ΣDDT (2 locations) were 460 ng/kg and 75 ng/kg, respectively. The seventh contained 0.7 ng/kg of dieldrin. None of the three open water reservoirs contained detectable residues of the compounds of interest.

Essentially no correlation was found between PCB and DDT analogs in streams and those in fish from streams which had been sampled at similar locations in a related study in 1976.

Introduction

Polychlorinated biphenyls (PCBs) and residual DDT and DDT metabolites (ΣDDT) are ubiquitous environmental contaminants not only in the United States but in other parts of the world (*8*). Contamination has spread from localized points to broad areas by many modes of transport, one of which is water.

The history of pesticide analysis in water can be traced to the 1950s in publications of the Public Health Service, U.S. Department of Health, Education, and Welfare, and the Geological Survey, U.S. Department of Interior (*7*). The Geological Survey began monitoring under the National Water Quality Network in 1965 and focused on the analysis of western streams (*21*).

Only within the past few years has there been a directive to study the nationwide contamination of water systems by PCBs and DDT. Federal drinking water limits for chlorinated hydrocarbons were first released in 1973 (*17*). In 1975, the U.S. Environmental Protection Agency (EPA) published national interim primary drinking water standards pursuant to requirements set by the Safe Drinking Water Act (Pub. L. 93–523) (*23*). Requirements of this act included the study of PCB contamination for the first time. In 1977, EPA set toxic effluent standards for DDT analogs (*24*) and for PCBs (*25*).

A summary of the residue studies conducted nationwide from January 1971 through June 1972 has been published (*8*). At that time, PCBs were found in the water of 12 States. Eight States were sampled moderately (25–50 samples) and three States were sampled heavily (greater than 160 samples), but Pennsylvania was sampled only twice.

Recent individual studies relating to environmental contamination of these long-lasting compounds have been reported. The occurrence of chlorinated hydrocarbon insecticides in southern Florida in 1968–72 was determined (*15*). Seasonal variations were found in residues of chlorinated hydrocarbons in the water of the Utah Lake drainage system (*4*). DDT analogs and PCBs were found in all stream tributaries to San Francisco Bay (*14*). Selected western streams frequently contained DDT analogs in 1967–71 but PCBs were found only twice in 1969–71 (*20*). Organochlorine pesticides were found in the Arkansas River in the vicinity of Tulsa, Oklahoma, in 1970–72, although no DDT analogs were found (*16*). Insecticide residues were studied (*12*) in the Tuttle Creek Reservoir ecosystem, Kansas, in 1970–71. Highly sensitive analyses

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were made for chlorinated hydrocarbon residues in the lower Mississippi River (*6*). Most samples of drinking water studied on the Island of Oahu, Hawaii, in early 1971 contained *p,p'*-DDT (*3*). In a 1969–71 study of four small drainage basins in Pennsylvania (*22*), DDT and analogs were found in water as well as soil, streambed material, and fish. In that study, PCBs were not determined.

Ground water has also been analyzed. The movement of DDT in ground water was measured in the Ogallala Aquifer in Texas (*18, 19*). In Georgetown County, South Carolina, 27 ground water wells from the lower coastal region bordered by the Atlantic Ocean were analyzed and all contained DDT in the parts per thousand range (*1*).

These reports indicate many studies of contamination in streams but few studies of community water supplies or open water reservoirs. The present study has helped to delineate the extent of contamination in community water supplies and open water reservoirs by PCB mixtures and DDT analogs. This is the first broad study of such sources in Pennsylvania. Streams were included in the study.

Analytical Technique

FIELD PROCEDURES

Stream samples were taken at Pennsylvania Water Quality Network (WQN) Stations (*9*). They were sampled at two-to-four points along a cross section or from both sides as described in the publication. Community supplies were sampled either as finished water at the main distribution point or as source water supply points. The grab samples were collected in new bottles that had been cleaned with solvent and rinsed with sample. Caps had Teflon liners. Samples were refrigerated until tested. The samples were not preserved with formaldehyde as has been described (*2*) because some batches of formaldehyde were contaminated by Aroclor 1242.

INITIAL EXTRACTION AND CLEANUP

Chemicals and laboratory equipment have been described (*13*). Water samples were extracted by adsorption to a cellulose triacetate membrane filter (*13*). Any sediment collected was included in the analysis. Occasionally several filters were required to shorten filtration time due to clogging. PCBs and DDT analogs present in water samples collected in 1974–75 were recovered from the filters by shaking once for 30 minutes with hexane and anhydrous sodium sulfate and then twice for 10 minutes with hexane alone. The combined hexane layers were concentrated, passed through a Florisil column, and analyzed by gas-liquid chromatography (GLC).

Water samples collected in 1976 were subjected to an improved recovery procedure. The filters were precleaned with ethyl ether and rinsed with distilled water before samples were filtered. Adsorbed compounds were removed with two portions of ethyl ether. The combined ethyl ether extracts were dried, concentrated, and transferred to isooctane by successive evaporations. The samples were then passed through a Florisil column and analyzed by GLC.

Control samples of laboratory double-distilled water were analyzed at regular intervals.

ANALYSIS

GLC analyses (*13*) were performed on a 5.5-foot × ¼-inch OD glass U column packed with 1.5 percent SP-2250/1.95 percent SP-2401 on 100–120-mesh Supelcoport. The column was maintained at 215°C; the nitrogen flow was 60 ml/min.

The ^{63}Ni electron-capture detector temperature was 330°C. Electrometer sensitivity was 1.6×10^{-9} amp full scale. Peaks of interest were measured with an Infotronics Model CRS-100 area integrator.

Samples which had peaks at retention times for PCBs and/or DDT and its metabolites were subjected to silicic acid column chromatography to separate PCBs from the DDT analogs: 430-mm × 7-mm ID column containing 5 g of silicic acid (Mallinckrodt, CC-7 Special), activated for 4 hours at 190°C, was used for the separations. The packing was prewashed with toluene. PCBs were eluted with petroleum ether and the DDT analogs were eluted with toluene. Samples were rechromatographed on the above GLC column, and peak identities were confirmed on a 6-foot column of 3 percent DEGS on 80–100-mesh Chromosorb WHP.

Compounds were quantitated by comparing GLC peak areas with those obtained from standards analyzed at regular intervals. PCB mixtures were quantitated by comparing the sums of the peak areas of mixture. When two mixtures of PCB types were apparent, peaks were quantitated by using the sum of the areas of nonoverlapping peaks.

STORAGE RECOVERY

Aroclor 1254 was added to double-distilled water at two levels and stored in the refrigerator with the other samples. The storage recovery samples were analyzed after 13 months; the stream and water supply samples were analyzed after 1–9 months of storage.

Average percent recoveries of triplicate analyses were:

NG AROCLOR 1254 ADDED	PERCENT RECOVERED ($\bar{x}$)
500	97
50,000	93

RECOVERIES

PCB- and DDT-spiked distilled water samples were passed through the membrane filter and recoveries were determined after hexane extraction of the filter disks. Complete details of this procedure are described elsewhere (*13*). This

scheme was used for the analysis of samples collected in 1975. The summary of triplicate analyses at three spiking levels is

Compound	µg Added	Percent Recovered (±SD, N=9)
Aroclor 1242	0.40, 2.0, 10	40 (±14)
Aroclor 1254	0.60, 3.0, 15	26 (±16)
p,p' DDE	0.02, 0.1, 0.5	24 (±14)
p,p' DDT	0.04, 0.2, 1.0	16 (±8.5)

The PCBs, DDT analogs, and others were recovered from samples collected in 1976 by ethyl ether extraction of the filter disks. Triplicate analyses at three spiking levels produced the recoveries below:

Compound	µg Added	Percent Recovered (±SD, N=9)
Aroclor 1242	0.40, 2.0, 10	98 (±14)
Aroclor 1254	0.60, 3.0, 15	83 (± 2)
p,p' DDE	0.03, 0.15, 0.75	72 (± 7)
p,p' TDE	0.06, 0.3, 1.5	74 (± 9)
p,p' DDT	0.06, 0.3, 1.5	76 (±15)
Mirex	0.3, 7.5	105 (±12) (n=6)

The results reported in this paper were corrected for these recoveries.

Results

Analysis of Pennsylvania water samples collected in 1974–76 showed the presence of PCBs or DDT analogs only in isolated locations (Tables 1–3). Only four stream locations of 19 Pennsylvania Water Quality Network stations sampled showed the presence of these pollutants. Of the community water supplies, only six of 110 sampled areas contained these compounds; a seventh contained dieldrin. None of the three open water reservoir samples contained any of these residues.

Stream stations were sampled twice, once in 1974 and again in 1975. Most of the stream contamination was found in Philadelphia, where the Delaware and Schuylkill Rivers contained both PCBs and DDT analogs. The Delaware River contained Aroclors 1242 and 1254, and DDE, TDE, and DDT at both sampling times. The concentrations of Aroclors in the normal flow sample taken in 1974 were 70 ng/kg (ppt) and 260 ng/kg (ppt), respectively; ΣDDT (*p,p'*-DDE + *p,p'*-TDE + *p,p'*-DDT) concentration was 620 ppt. At higher flows in fall 1975, the concentrations of Aroclors and DDT analogs were reduced, respectively, to 180 ppt and 60 ppt. At the lower end of the Schuylkill River in fall 1974, the PCB concentration was 500 ppt, but in fall 1975, no PCBs were found.

Only trace quantities of the compounds of interest were found at the other WQN stations. DDT was found at the Lehigh (Easton) station, and Aroclor 1254 was found at the Monongahela (Rankin) station upstream from Pittsburgh. The amount of stream flow may indicate a dilution effect. The Delaware samples in Philadelphia contained fewer contaminants with increased water flow; the Lehigh samples may have been affected similarly. The dilution effect was not observed in one study (*10*), however, of a uniformly treated area where DDT residues were distributed evenly over varied flow rates.

Of the community water supplies, the 1975 Lewiston sample contained 460 ppt Aroclor 1242. Subsequent sampling in 1976 of the two runs leading to the reservoir and of one location in the distribution system did not indicate any residues. Similarly, the 1975 Lock Haven sample contained a trace of Aroclor 1242, but further sampling of this reservoir and two distribution sites in 1976 showed no residues.

The same situation occurred in two communities in southwestern Pennsylvania. The 1975 Waynesburg sample contained 75 ppt ΣDDT, yet in 1976 no DDT was found in the water supplies in four different locations of that community. Trace DDT was found in 1975 in Point Marion, but in 1976 none was found in supplies in five different locations.

Two of the community water supplies sampled in 1976 contained residues. A total of 19 communities were sampled at that time. The Masontown sample contained 20 ppt Aroclor 1242, and the Pottstown sample contained a trace of Aroclor 1242. The community water supply of Centre Township, also in that area, contained 0.7 ppt dieldrin.

Sulfur was not observed as an industrial contaminant in any of these samples.

Discussion and Conclusions

PCB AND DDT LEVELS OF CONTAMINATION

Although PCBs are currently reported to be environmentally ubiquitous (*8*), they were detected in only a few locations in Pennsylvania at any significant level: three of 19 stream locations and four of 110 community water supplies, comprising a total of 5.4 percent. The nationwide study (*8*) conducted from January 1971 through June 1972 found a similar percentage of PCB contamination: 83 of 1627, comprising 5.1 percent. In the earlier study, one of two locations sampled in Pennsylvania contained PCBs. Neither study, unfortunately, was broad enough to indicate any trend in the concentration of these pollutants.

Truhlar and Reed (*22*) studied in detail DDT and DDT analogs in four Pennsylvania streams in 1969–71. ΣDDT in samples from a forest stream (Young Womens Creek near Renovo), a farm stream (Bixler Run near Loysville), a residential stream (Spring Creek near Dauphin), and an orchard stream (Latimore Creek in Adams County) were 0.00, 0.03, 0.60, and 0.47 µg/liter, respectively (Table 4). Data from the two studies cannot be compared statistically because they represent only four areas in the Common-

TABLE 1. *Chlorinated hydrocarbon residues in Pennsylvania waters—1974-76*

WQN No	Location[1]	Date Sampled	Sample Size, g	Stream Flow,[2] cfs	Flow Condition[3,4]	Chlorinated Hydrocarbon Residues, Corrected for Recovery, ng/kg (ppt)					
						Aroclor 1242	Aroclor 1254	*p,p'*-DDE	*p,p'*-TDE	*p,p'*-DDT	ΣDDT
		STREAM SOURCES									
PCB S-1	Schuylkill R Pottstown	9-23-74	3535	450		-[4]	-	-	-	-	-
	(Douglasville Br)	6-5-75	3625[5]	900							
PCB S-2	Delaware R Easton	9-23-74	3525	2000		-	-	-	-	-	-
	(Northampton St Br)	6-5-75	3625[5]	3200		-	-	-	-	-	-
MDA-1	Delaware R Phila	10-17-74	3595		B	70	260	420	90	110	620
	(below metal bank)	9-24-75	1745		A	70	110	40	90	11	60
104	Delaware R Hancock, NY	9-19-74	3585		B	-		-	-	-	-
	Bucking Twp (Rt 191 Br)	5-8-75	3530		A	-	-	-	-	-	-
123	Lehigh R Easton	9-23-74	3503	1100		-	-	-	-	tr[6]	tr
	(Third St Br)	6-5-75	3630[7]	1550		-	-	-	-	-	-
124	Lehigh R Bethlehem	9-23-74	3525	800		-	-	-	-	-	-
	(Hill-to-Hill Br)	6-5-75	3605[5]	1200		-			-	-	-
143	Schuylkill R Phila	10-1-74	3513		A	350	150	-	-	-	-
	(Strawberry Mansion Br)	9-29-75	1790		A	-	-		-	-	-
202	Susquehanna R Harrisbg	9-12-74	3610		C	-		-	-	-	-
	(Walnut St Br)	5-2-75	3630		A				-	-	-
214	Juniata R Newport	9-12-74	3624		C		-	-	-	-	-
	(Rt 34 Br)	4-25-75	3640		A		-		-	-	-
232	Susquehanna R Holtwood	9-11-74	3600					-	-	-	-
	(Rt 372 bridge)	5-2-75	3615		A				-	-	-
301	N Br Sesq Danville	10-2-74	3585		B				-	-	-
	(Rt 54 bridge)	12-1-75	1791						-	-	-
305	N Br Sesq Towanda	9-19-74	3595		B				-	-	-
	(U S 6 bridge)	5-8-75	3583		A		-	-	-	-	-
401	W Br Sesq Lewisburg	10-2-74	3370		B			-	-	-	-
	(Rt 45 bridge)	12-1-75	1814						-	-	-
601	Lake Erie Mill Creek Twp	9-10-74	3439					-	-	-	-
	(Erie Waterworks intake)	10-1-75	1795		B	-		-	-	-	-
701	Monongahela R Rankin	9-23-74	3475		B		tr		-	-	-
		11-25-75	1775							-	-
801	Allegheny R Kensington	10-4-74	3159		C					-	-
	(left & right sides)	11-21-75	1702		A	-				-	-
803	Allegheny R Parker	9-24-74	3360	10,000	A				-	-	-
902	Ohio R Sewickley	9-23-74	3540		B				-	-	-
	(Sewickley Br)	11-25-75	1802		B				-	-	-
904	Beaver R Rochester	10-1-74	3410		B		-	-	-	-	-
		11-13-75	1817		B				-	-	-
		OPEN WATER RESERVOIRS									
	Prince Gallitzen St Park (breast of dam)	8-5-75	1755		B				-	-	-
	Quemahoning Res (breast of dam)	10-1-75	1790			-	-	-		-	-
	Lake Wallenpawpack, Wolfs Cove	10-7-75	1640			-	-	-		-	-
		COMMUNITY WATER SUPPLIES									
	Centre Township (Greene Co)[8]	8-13-76	3830			-	-	-		-	-
	Lewistown (Mifflin Co) (see below)	9-29-75	1715			460			-	-	-
	Lock Haven (Clinton Co) (see below)	8-26-75	1783			tr	-	-	-	-	-
	Masontown (Fayette Co, gas station) (see below)	8-25-76	3872			20				-	-
	Point Marion (Fayette Co)	9-3-75	1750				-		-	tr	tr
	Pottstown (Montgomery Co)	7-29-76	3853			tr			-	-	-
	Waynesburg (Greene Co) (see below)	9-3-75	1791			-		6	9	60	75

[1] WQN = Water Quality Network. See literature reference 9.
[2] Instantaneous measurement taken at time of sampling, cfs = cubic feet per second.
[3] A = above, B = normal, C = below.
[4] No detectable residue.
[5] Rain in morning.
[6] Trace, see Table 3.
[7] Rain in morning; stream had slight turbidity.
[8] South fork of Ten Mile Creek above McCourtney Run contained 0.7 ppt dieldrin.

TABLE 2 *Community water supplies which had no detectable PCB or DDT analog residues, Pennsylvania, 1974-76*

Allentown
Altoona
Ambridge (Beaver Co.)
Bedford
Belfast Twp. (Fulton Co.)
Bellefonte (Centre Co.)
Biglerville (Adams Co.)
Birdsboro (Berks Co.)
Bloomsburg (Columbia Co.)
Blossburg (Tioga Co.)
Blythe (Schuylkill Co.)
Bradford (McKean Co.)
Brockway (Jefferson Co.)
Brookhaven (Delaware Co.)
Butler City
Cambridge Springs (Crawford Co.)
Canonsburg (Washington Co.)
Carmichaels Boro (Greene Co., 1976, plant influent, plant effluent at Amoco, distrib. system at Amoco on S. Vine and at 106 S. Vine)
Center Township (Greene Co., 1976, McCourtney Run, Lightner Run, Pursley Cr., Rush Run)
Chambersburg (Franklin Co.)
Charleroi (Washington Co.)
Columbia (Lancaster Co.)
Conemaugh (Somerset Co.)
Cumberland Twp. (Greene Co., 1976)
Danville (Montour Co.)
Delaware Water Gap (Monroe Co.)
Derry (Westmoreland Co.)
Derry Twp. (Dauphin Co., 1975 and 1976, 1883 g)
Duncannon (Perry Co.)
Dunkard (Greene Co., 1976, Dunkard Cr., Brunley Res., Gogallatto Res., plant influent, plant effluent, Monongahela R. up from Cheat R.)
Eaglesmere (Sullivan Co.)
East Berlin (Adams Co.)
Ellwood City (Lawrence Co.)
Emlenton (Venango Co.)
Emporium (Cameron Co.)
Erie
Fayette City (Fayette Co.)
Franklin Twp. (Greene Co., 1976, Brown's Cr., first and second unnamed tribs. to Ten Mile Cr., Clear Run)
Gallitzin (Cambria Co.)
German Twp. (Fayette Co., 1976, Cat's Cr., Big Run filtration plant, plant effluent, distrib. system at Kolenick's Sunoco at the Pizza Parlor)
Gettysburg (Adams Co.)
Greensboro (Greene Co., 1976)
Greenville (Mercer Co.)
Hallstead (Sesquehanna Co.)
Harrisburg
Hazelton
Hershey
Hollidaysburg (Blair Co.)
Homer City (Indiana Co.)
Honesdale (Wayne Co.)
Houtzdale (Clearfield Co.)
Jersey Shore (Lycoming Co., 1975 and 1976)
Johnstown
Kingston (Lucerne Co.)
Lancaster
Lansford (Carbon Co.)
Lebanon
Lewistown (Mifflin Co., 1976, Laurel Run, Mutterbaugh Gap Run, distrib. system at Burnham M. A. STP)
Lititz (Lancaster Co.)
Littlestown (Adams Co.)
Lock Haven (1976, Castanea Res., Keller Res., Rosecranes Res., distrib. system at Swinehart residence in McElhatten and at R and K Grocery)
Lower Paxton Twp. (Dauphin Co., 1976)
Masontown Boro (Fayette Co., 1976, plant influent, plant effluent, distrib. system at 305 S. Main St.)
McKeesport
Media (Delaware Co.)
Middleburg (Snyder Co.)
Mifflinburg (Union Co.)
Milesburg (Centre Co.)
Milford (Pike Co.)
Milton (Northumberland Co.)
Monongahela Twp. (Greene Co., 1976)
Morrisville (Bucks Co.)
Moscow (Lackawana Co.)
Mount Joy (Lancaster Co.)
New Bethelem (Clarion Co.)
New Freedom (York Co.)
Nicholson (Wyoming Co.)
Nicholson Twp. (Fayette Co., 1976)
North East (Erie Co.)
North Shenango (Crawford Co.)
Northampton (Northampton Co.)
Oakmont (Allegheny Co., 1976)
Parker City (Armstrong Co.)
Philadelphia (Belmont Res., Queen Lane Res., Torresdale Res., Phila. suburban at Lower Merion pump station)
Pittsburgh
Point Marion (Fayette Co., 1976, first unnamed trib. to Cheat R., Grassy Run, Conrad Res., Rose Res., plant influent, plant effluent)
Rauchtown (Clinton Co., 1976)
Reading
Ridgway (Elk Co.)
Roulette (Potter Co.)
Saltsburg (Indiana Co., 1976)
Scranton
Sharon (Mercer Co., 1976)
Shawnee State Park (Bedford Co.)
Sheffield (Warren Co.)
Shrewsbury (York Co.)
Springhill Twp. (Fayette Co., 1976)
Stewartstown (York Co.)
Stroudsburg (Monroe Co.)
Tionesta (Forest Co.)
Towanda (Bradford Co.)
Union Twp. (Mifflin Co., 1976)
Wagontown (Chester Co.)
Waynesburg (Greene Co., 1976, plant influent, plant effluent, distrib. system at Gulf Station and at McDonald's Rest.)
West Penn Power (Greene Co., 1976, intake, Monongehela R. below discharge)
Wilkes-Barre
Williamsport
Woolrich (Clinton Co., 1976)
Wormsleyburg (Cumberland Co.)
York

NOTE: Samples collected August-November 1975 were all approx. 1800 grams. Samples collected July-August 1976 are marked "1976" and were all approx. 3600 grams, unless designated otherwise.

TABLE 3 *Detectability limits of method of analysis for water samples*

Limit	Sample Size, g	Aroclor 1242	Aroclor 1254	*p,p'* DDE	*p,p'* TDE	*p,p'*-DDT	∑DDT
No detectable residue	3600	< 10	< 10	< 1	< 1	< 1	—
	1800	< 20	< 20	< 2	< 2	< 2	—
Trace	3600	10–25	10–25	1–2.5	1–2.5	1–2.5	—
	1800	20–50	20–50	2–5	2–5	2–5	—

TABLE 4 *Comparison of mean ΣDDT in Pennsylvania waters, 1969–76*

Study Area	Residues, μg/liter N	Means
FROM TRUHLAR AND REED, 1969–71[1]		
Forest stream	10	0.00
Farm stream	14	0.03
Residential stream	24	0.60
Orchard stream	32	0.47
FROM THIS WORK, 1975–76	Residues, μg/kg	
Rivers	192	0.018
Communities	110	0.0007

[1] See literature reference 22
[2] Sampled twice

wealth. One can say only that DDT concentration in water in forest and farm areas is similar to that in rivers across Pennsylvania: 0.018 μg/kg. DDT concentration in water in the forest area is similar to that in areas serving the communities of Pennsylvania: 0.0007 μg/kg.

DETERMINING STREAM POLLUTION

Direct point source sampling of water in streams gives the analyst only limited information regarding the presence of contaminants over a broad geographic area such as a state. Unless a large number of samples are taken at a high cost, other sampling means must be used.

Several averaging methods are available. These include continuous or intermittent direct sampling at one location, or indirect sampling. The content of lypophobic compounds, e.g., chlorinated hydrocarbons such as PCBs, in water can be estimated by analyzing the fish in that water or by analyzing the organic sediment in the active stream system. Since both media are organic, the equilibria of these compounds in water are in the direction of the former.

Truhlar and Reed analyzed bottom sediment separately for DDT analogs (22). They found a positive correlation of ΣDDT between the bottom sediment and the water of Lattimore Creek.

Another time-averaged method would be the analysis of fish, an animal high on the biomagnification chain. Selecting the fish based on its feeding habits or range of habitat enables the researcher to evaluate the concentration of PCBs or DDT analogs present over the fish's lifetime.

The Pennsylvania Department of Environment Resources (DER) conducted a survey of the levels of PCBs and other chlorinated hydrocarbons in fish from selected Pennsylvania waters (5) in 1976. Many of the collection points correspond closely to those for water-stream sources in the Pennsylvania Water Quality Network. Pertinent results of that study are summarized in Table 5.

In the DER study, fish were generally sampled 6 or 18 months after water sampling times. The range in PCB concentrations (ΣPCB expressed as Aroclor 1254) for sunfish (carnivorous feeders living on zooplankton and aquatic invertebrates) was 0.05–0.86 μg/g (wet weight, whole-tissue). PCBs were found in nine stations corresponding to WQN sampling points. ΣDDT and analog concentration in sunfish ranged from 0.01 to 0.10 μg/g and were found in only seven of these stations.

The range in PCB concentrations for carp, which are bottom feeders, was greater: 0.04–1.63 μg/g in 12 stations. For DDT the range was also greater: 0.01–1.27 μg/g in nine stations.

An examination of the points of correspondence for the fish and water studies showed little correlation of data. Correlation coefficients (Pearson product moment) for water versus sunfish for PCBs and DDT were undefined and −0.156, respectively. Those for water versus carp for PCBs and DDT were 0.132 and 0.891, respectively. Such poor correlations are probably due to the fact that most water samples contained less than a determinable amount of contaminant. The correlation of DDT in water versus carp (0.891) is misleading, even though the hypothesis of no correlation is rejected at the 1 percent level since 12 of the 16 values lie essentially at the origin of the plot.

MODEL OF PCB FLOW IN STREAMS

Several models could be designed to determine how PCB's, DDT, polybrominated biphenyls, and similar pollutants are transported in waterways. In one such model, compounds are present in more or less constant concentrations. This could result from a slow dissolution of a concentrated source into the water flowing by the source. The absolute level could vary from one point to another, but would remain at a given level for several days or weeks or would change slowly from level to level.

Another model that could describe this information is a slug-flow model. In this type, the compounds are released within a matter of hours as a result of some human or natural activity. This contamination moves downstream at a rate close to the water movement. At any point along the stream the concentration of contaminants could vary widely from moment to moment.

The slug-flow model would have to be modified to account for the effects of sediment usually present in stream flows. PCBs moving downstream adsorb to organic matter up to the capacity of the matter. Given only a small amount of lyphophobic contaminant and enough sediment and stream turbulence, the entire amount could be adsorbed by the organic material. The rate of transfer from water to sediment would depend on the concentration of organic matter and turbulence. The rate would probably be relatively high due to the minimal solubility in water. Hence if small amounts of PCB enter a flowing stream, soon they could be ad-

TABLE 5. *PCB and ΣDDT residues in fish taken from locations corresponding to water sampling locations, Pennsylvania, April–July, 1976* [1]

Fish Sampling Location	WQN Station at Water Sampling Location[2]	Residues, μg/g Wet Weight, Whole Fish					
		Sunfish[3]			Carp[4]		
		No Fish	PCB	ΣDDT	No Fish	PCB	ΣDDT
I-10	PCB S-1	10	0.56	0.02	2	1.63	ND [5]
I-19	PCB S-2	5	ND	ND	5	ND	ND
I-1	MDA-1				4	0.70	1.27
I-14	123	6	ND	ND	7	0.61	ND
I-15	124	13	ND	ND	5	ND	ND
I-5	143				5	0.61	0.18
III-1	202	5	0.86	0.08	5	ND	ND
III-2	232	5	0.06	0.01	5	0.07	0.01
III-4	301	5	0.72	ND	5	0.66	0.04
III-9	305	5	0.05	0.01	5	1.16	0.07
III-10	401	6	0.07	0.02	5	ND	ND
IV-7	601	5	0.19	ND	5	0.93	0.33
IV-21W	701				5	ND	0.36
IV-21E					5	0.63	0.73
IV-8	801	5	0.10	0.10	5	0.31	0.01
IV-9	803	5	ND	ND	5	0.04	ND
IV-4	902	5	0.81	0.03	5	0.76	0.02
IV-4	904	5	0.81	0.03	5	0.76	0.02

[1] See literature reference 5
[2] WQN = Water Quality Network. See literature reference 9
[3] Includes smallmouth, largemouth, and rock bass, black and white crappie, bluegill, and pumpkinseed and redbreast sunfish
[4] Includes goldfish, channel catfish, brown bullhead, white sucker, and silver redhorse
[5] ND = no detectable residue, < 0.01 μg/g

sorbed to sediment. If the sediment settles, the PCB would not be found in the analysis of the sample taken downstream from that point.

The present study included 19 major stream stations sampled twice, 7 to 14 months apart. From this collection of data, there were seven examples of residues in the first sampling (PCBs or DDT analogs) and not in the second. Only one station had residues (Station MDA-1, Delaware River, Philadelphia) in both samplings. These data, although limited, support slug-flow movement.

The Truhlar and Reed (22) data support this conclusion more strongly. There were numerous occasions when the authors analyzed two to four samples taken the same day. Considering only those that contained significant amounts of DDT analogs, there were five examples that could support a slug model of transport. In these cases the levels found varied widely. Some were also inversely proportional to sediment concentration; the Truhlar and Reed data as well as the data reported here included sediment concentrations in the water samples. Only three examples supported a constant content model, i.e., the levels found were relatively constant or they varied proportionally with sediment concentration.

The sampling procedures of this study and the Truhlar and Reed study were different. Truhlar and Reed picked a few sites and took many samples for two years. In this study, many sites across the State were sampled, but none more than twice. Both studies appear to support a slug-flow model for contamination.

Both models could be operating at the same time. Slugs of contamination could be imposed on a continuous level of the material. The concentration at the continuous level may be lower than has been routinely detected; if so, greater analytical sensitivity is needed.

In this study, most samples were 1.8 kg which is a convenient size to collect and ship. The detection limit for this size was 20 ppt (ng/kg) for PCBs and 2 ppt for each DDT analog. These compounds are insoluble in water at levels slightly above two powers of ten higher for Aroclor 1254 and three to four powers of ten higher for Aroclor 1242 (*11*). In a recent ruling to improve sensitivity in water analyses, the U.S. Environmental Protection Agency set the ambient water criterion (25) for PCBs in navigable waters at 1 ng/liter. This would require a detection limit of at least 1/10 as much or 0.1 ng/liter, which is 1/200 of the limit obtained in this work. To lower this detection limit, the sample size would need to be increased, the background levels would need to be decreased, and the detection technique would have to be improved.

By examining the bioaccumulation of PCBs in fish, one can gain insight in this matter. One could assume that the PCB level in water was constant at the upper end of the nondetectable level, i.e., 10 ppt in this work. At a bioaccumulation factor of 274,000 for fathead minnows (*25*), fish would contain 2.7 ppm PCBs. Nine stations in the Pennsylvania study yielded carp containing 0.13–1.63 ppm (*5*), and no stations yielded carp containing greater than 1.63 ppm. Since no samples of fish contained as much as 2.7 ppm, it is reasonable that no water samples contained even detectable amounts. This was, in fact, the case. Excluding input by sediment or other means this would indicate that the minimum detectable levels in water were too

high by a factor of ten to produce these concentrations in fish. Hence the detectability limit must be decreased from 10 to 0.1. When this is done, the question of whether the PCB levels are constant or whether they appear in slugs as suggested in this paper can be approached.

A single sample analysis of stream water would be appropriate if the source of contamination were continuous. If the source were variable, however, either multigrab samples or a time-averaged method would be required to determine the contamination profile.

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GENERAL

DDT Residues in Rainwater in New Brunswick and Estimate of Aerial Transport of DDT into the Gulf of St. Lawrence, 1967-68

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ABSTRACT

Residues of DDT were detected in 76 of 101 samples of rainwater collected during spring and summer at several sites in the Province of New Brunswick in 1967 and 1968, and at one site in the Magdalen Islands, Quebec, in 1968. The dominant residue was p,p'-DDT. Levels of DDT and metabolites combined ranged from < 0.01 to 1.33 μg/kg. Levels of DDT and metabolites in the pollen of four species of forest trees in New Brunswick ranged from 0.544 to 1.01 mg/kg; such contaminated pollen possibly contributed to residues in rainwater. Residue data for rainwater from two sites were used to estimate the amount of DDT aerially transported into the Gulf of St. Lawrence during July to October 1968

Introduction

Despite the interest in transport of organochlorines through the atmospheric environment, few residue data collected in areas remote from pesticide use have been related to specific spray programs. Levels of DDT and its metabolites in rainwater sampled at a number of sites in New Brunswick in 1967 and 1968, and at one station in Quebec in 1968, are reported. Also recorded are concentrations of DDT in the pollen of four species of trees common in the New Brunswick forest, since contaminated pollen could have contributed to residues in rainwater samples. Investigations were conducted at a time when there was still a substantial regional use of DDT in forestry and agriculture. By determining residue levels in some rainwater samples, one may estimate the amount of DDT transported through the atmosphere into the adjacent Gulf of St. Lawrence.

Materials and Methods

SAMPLE COLLECTION AND ANALYTICAL PROCEDURE

Fifty-eight rainwater samples were collected at seven sites in New Brunswick from mid-May to late July 1967. The sites were selected to encircle the area of forest in the center of the Province to be sprayed with DDT and the organophosphates phosphamidon and fenitrothion. In 1968, 43 samples were collected from mid-April to mid-August from four sites in New Brunswick and one in the Magdalen Islands, Quebec, all downwind from the treatment area. New Brunswick is about 90 percent forested. Most collection sites in that Province were in or near the forest. The Magdalen Islands are located in the middle of the Gulf of St. Lawrence, about 250 km from the eastern shore of New Brunswick. The geographic positions of the collection sites are given in Tables 1 and 2.

Water was collected in glass bottles through a large glass funnel supported in a section of stove pipe firmly placed in the ground. In 1967, authors attempted to keep the sample size to about 1 liter and used both clear and amber bottles as well as a small fiber-glass pad to prevent possible clogging of the funnel stem by leaves and large insects. The following year, authors modified the procedure slightly to exclude the fiber-glass pad, since there had been no obvious accumulation of extraneous organic matter in bottles or funnel stems. In 1968, amber bottles were used at all stations except one, which was supplied with clear bottles wrapped in aluminum foil. Precautions were taken whenever possible to prevent contamination of the samples. All glassware was washed in detergent, and rinsed several times with water and twice with redistilled acetone before use.

In May 1968, before the forest had been sprayed, authors obtained samples of pollen of jack pine (*Pinus banksiana*), red spruce (*Picea rubens*), white birch (*Betula papyrifera*), and red maple (*Acer rubrum*) from a region in central New

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[3] Canadian Wildlife Service, Department of Fisheries and the Environment, Ottawa, Ontario, Canada K1A OH3

TABLE 1. *DDT residue levels in rainwater samples from New Brunswick, Canada—1967*

Site	Collection Period	Total Volume, ml	Residues, μg/kg			
			p,p'-DDE	*p,p'*-TDE	*o,p'*-DDT	*p,p'*-DDT
Allardville	15-19 May	1150	ND	ND	ND	ND
47 28	19-27 May	2000	ND	ND	ND	ND
65 29	27-28 May	1180	0 09	0 06	0 09	0 14
	28 May-8 June	1280	0 10	0 08	0 06	0 14
	8-22 June	1220	0 03	0 08	ND	0 09
	22-26 June	1910	0 06	0 05	ND	0 12
	26 June-4 July	1590	0 02	< 0 01	ND	< 0 01
	4-10 July	1180	ND	ND	ND	ND
	10-13 July	154	0 19	0 21	0 14	0 36
St. Louis	15-18 May	1140	ND	ND	ND	ND
46 44	18-19 May	1400	ND	ND	ND	ND
64 58	19-26 May	1070	ND	ND	ND	ND
	26-29 May	1110	ND	ND	ND	ND
	29 May-15 June	1070	0 03	0 06	ND	0 14
	15-19 June	1150	0 03	0 05	0 04	0 09
	19-24 June	1100	0 37	0 18	2 78[1]	20 0[1]
	24 June-1 July	1170	0 19	0 24	0 72[1]	12 5[1]
	1-5 July	1060	ND	0 06	ND	0 81[1]
	5-8 July	1040	ND	< 0 01	0 07	0 53[1]
	8-13 July	1380	0 06	0 07	ND	0 78[1]
Canaan	12-13 May	1850	ND	ND	ND	ND
46 05	13-18 May	1570	ND	ND	ND	ND
65 22	18-26 May	1380	ND	ND	ND	ND
	26-29 May	1230	< 0 01	ND	ND	ND
	29 May-12 June	1120	ND	ND	ND	ND
	12-18 June	2176	ND	ND	ND	ND
	18-25 June	1560	0 04	0 05	0 02	0 06
	25 June-3 July	1210	ND	ND	ND	ND
	3-4 July	1600	0 06	0 06	ND	0 08
	4-14 July	710	ND	ND	ND	ND
Fredericton	12-18 May	1140	0 06	0 06	0 07	0 17
45 58	18-26 May	1340	ND	0 05	0 02	0 08
66 39	26 May-17 June	1370	0 04	0 06	ND	ND
	17-19 June	1130	0 04	0 05	0 02	0 09
	19-24 June	1640	0 03	0 03	ND	0 10
	24 June-4 July	1010	0 21	0 17	0 09	0 41
	4-26 July	980	0 03	0 05	ND	0 08
Parker	13-20 May	1520	< 0 01	< 0 01	ND	< 0 01
Ridge	20-27 May	1200	ND	< 0 01	ND	< 0 01
46 27	27 May-12 June	1210	ND	ND	ND	0 27
66 31	12-18 June	1360	0 03	0 06	0 05	0 31
	18-25 June	1130	ND	ND	ND	0 50
	25-26 June	1040	ND	ND	ND	ND
	26 June-2 July	1240	ND	ND	ND	ND
	2-9 July	1400	ND	ND	ND	ND
	9-24 July	1000	ND	ND	ND	ND
Gordonsville	16-20 May	970	ND	ND	ND	ND
46 29	20 May-8 June	1020	ND	ND	ND	ND
67 31	8-17 June	1180	ND	ND	ND	ND
	17-24 June	1100	0 03	0 03	0 01	0 08
	24-26 June	1930	ND	ND	ND	ND
	26 June-10 July	1940	0 06	ND	ND	ND
Kedgwick	16-20 May	1210	0 04	0 06	0 04	0 09
47 39	20 May-2 June	1030	< 0 01	0 03	ND	0 09
67 21	2-23 June	970	0 02	ND	ND	0 17
	23-26 June	1930	ND	ND	ND	ND
	26 June-14 July	1780	0 04	0 06	ND	0 29
	14-18 July	425	0 06	0 12	0 06	0 22

NOTE: ND = not detected (0.001 μg/kg)
[1] Local contamination

Brunswick which had been aerially sprayed with DDT each year from 1956 to 1967, except 1966. The collection procedure was simply to shake staminate flowers carefully in thoroughly cleaned glass jars.

Pesticide residues were determined by gas-liquid chromatography (GLC). The organochlorine compounds were detected with an electron-capture (EC) detector; the EC detector was used in conjuction with the phosphate (P) detector for the organophosphates. A 500-ml aliquot of each sample was extracted with ether-hexane in the presence of sodium sulfate, the phases were allowed to separate, and the aqueous portion was re-extracted with hexane. The combined organic phases were rinsed with distilled water, dried over anhydrous sodium sulfate, and analyzed.

TABLE 2. *DDT residue levels in rainwater samples from New Brunswick and Quebec, Canada—1968*

Site	Collection Period	Total Volume, ml	Residues, μg/kg			
			p,p'-DDE	*p,p'*-TDE	*o,p'*-DDT	*p,p'*-DDT
Miscou	7-27 May	690	0.01	ND	ND	0.01
47 01	27 May-4 June	1050	ND	ND	ND	0.02
64 31	4-19 June	1000	< 0.01	ND	ND	0.01
	19-24 June	1110	0.08	ND	ND	0.01
	24 June-7 July	1640	0.13	ND	ND	0.04
	7-24 July	960	< 0.01	ND	0.01	0.11
	24 July-9 August	700	< 0.01	ND	<0.01	0.08
Escuminac	10-29 May	380	ND	ND	0.42	0.91
47 04	29 May-1 June	615	ND	ND	0.04	0.08
64 48	1-5 June	640	ND	ND	ND	0.03
	5-16 June	1015	< 0.01	ND	0.02	0.06
	16-20 June	860	0.01	ND	0.04	0.06
	20-22 June	1260	0.01	ND	0.03	0.01
	22 June-1 July	1550	ND	ND	ND	0.01
	1-14 July	430	ND	ND	ND	ND
	14-20 July	730	ND	ND	ND	0.05
	20 July-9 August	380	ND	ND	ND	0.05
Clairville	19-29 April	1925	< 0.01	0.01	ND	0.01
46 23	29 April-9 May	400	0.02	ND	ND	0.02
65 06	9-22 May	960	0.03	<0.01	ND	0.01
	22 May-11 June	1700	0.03	< 0.01	ND	0.01
	11-12 June	530	0.02	< 0.01	ND	0.01
	12-17 June	760	0.01	0.01	ND	0.01
	17-20	760	0.01	0.01	ND	0.01
	20-23 June	1590	0.01	ND	ND	0.01
	23 June-1 July	1675	ND	ND	ND	ND
	1-26 July	555	0.31	0.21	0.66[1]	10.4[1]
Fredericton	17-23 April	580	0.01	ND	ND	0.06
45 58	23-28 April	950	< 0.01	ND	ND	0.06
66 39	28 April-14 May	480	0.01	ND	ND	0.06
	14-22 May	1015	0.01	0.02	0.02	0.13
	22 May-3 June	715	0.01	0.03	0.03	0.16
	3-5 June	620	< 0.01	0.02	0.02	0.09
	5-17 June	487	0.01	0.02	0.01	0.11
	17-22 June	1700	0.01	0.01	0.01	0.08
	22 June-6 July	1980	0.01	0.01	ND	0.02
	6 July-11 August	1960	0.01	0.01	ND	0.07
Magdalen	18 May-2 June	1120	ND	ND	ND	0.01
Islands	2-15 June	980	0.01	< 0.01	ND	0.03
47 23	15-16 June	970	< 0.01	ND	ND	<0.01
61 52	16-17 June	1130	< 0.01	< 0.01	ND	<0.01
	17-23 June	990	0.02	<0.01	ND	0.01
	23 June-13 August	1925	ND	ND	ND	0.06

NOTE: ND = not detected = <0.001 μg/kg
[1] Local contamination

Reagent grade chemicals were used. Ether was redistilled but hexane (Nanograde) was used as supplied. Varian Aerograph Models 1200 (equipped with an EC detector) and 204 (with EC and P detectors) were used. The column effluent from the Model 204 was split 50:50 so that half the injected sample passed through each detector, and the responses were recorded by an attached dual-pen recorder. Both glass helix GLC columns, 152 cm × 0.32 cm, were packed with 4 percent SE-30/60 percent QF-1 on 60-80 mesh acid-washed Chromosorb W.

An investigation was made to ascertain what portion, if any, of the pesticides adhered to the bottle surface and therefore was not quantitatively recovered. The water sample was removed from each of four bottles and the bottle surfaces were thoroughly rinsed with 200 ml of acetone. The acetone extracts were analyzed. The results prompted a modification of the method in order to recover the residues remaining on the bottle surfaces. Each bottle was thereafter emptied and rinsed with 200 ml acetone, and an appropriate proportion of the rinsings was added to the 500-ml aliquot to be analyzed. All residues reported here include those adsorbed on bottle surfaces. No attempt was made to determine what residues remained on the funnel surfaces. A jar cleaned in the same manner as those used for collecting the pollen samples was also rinsed with acetone, and no additional residues were recovered.

The recoveries of added pesticides including lindane, heptachlor, aldrin, heptachlor epoxide, DDE, dieldrin, TDE, *p,p'*-DDT, and fenitrothion were essentially 100 percent, and although the confidence level for the method is taken as 0.01 ppb (μg/kg), residues as low as 0.001 ppb were detected. Confirmatory analyses were conducted with thin-layer chromatography (TLC). A small peak with a retention time similar to fenitrothion was observed with the

EC detector in some samples, but the P detector proved the absence of any organophosphate. During the analyses, authors were aware of the possibility of interferences from polychlorinated biphenyls (PCBs). The GLC peaks eluting after DDT were small, indicating that if PCBs (Aroclor 1254 and 1260 types) were present, the amounts were small. The TLC analyses confirmed that the organochlorine pesticides were responsible for at least the major portion of the residues.

TABLE 3 *Precipitation in the Magdalen Islands, Canada—1968*

Month	Precipitation, cm
January	11 0
February	6 6
March	7 6
April	6 1
May	3 5
June	9 4
July	3 4
August	12 0
September	2 8
October	8 1
November	11 4
December	12 4

CALCULATION OF DDT DEPOSITION INTO THE GULF OF ST. LAWRENCE

The concentrations of ΣDDT in the rainwater at the two downwind sites most remote from the spray program and from possible local usage, viz., Miscou and the Magdalen Islands, were used to estimate deposition of DDT into the Gulf of St. Lawrence. Monthly precipitation in the Magdalen Islands in 1968 is shown in Table 3. The initial assumption made was that the falloff of concentration with the distance from source was exponential, and the geometry of the region was reduced to a wedge-shaped area (Fig. 1).

Thus the concentration is considered to be related to the distance from the area of forest spraying by the equation $y = Ae^{-bx}$, where y = concentration of DDT, x = distance, and A and b are constants. Each segment of the wedge has an area of $Kx.dx$, where $K = 2\ \pi a/360$ and a = the angle of the wedge. L is the approximate distance between central New Brunswick and the eastern coastline of that province, and U is the distance between central New Brunswick and the arc of a circle running through southwestern Newfoundland. The amount of DDT deposited per centimeter of rainfall in the truncated wedge-shaped area extending from the coastline of New Brunswick to southwestern Newfoundland is:

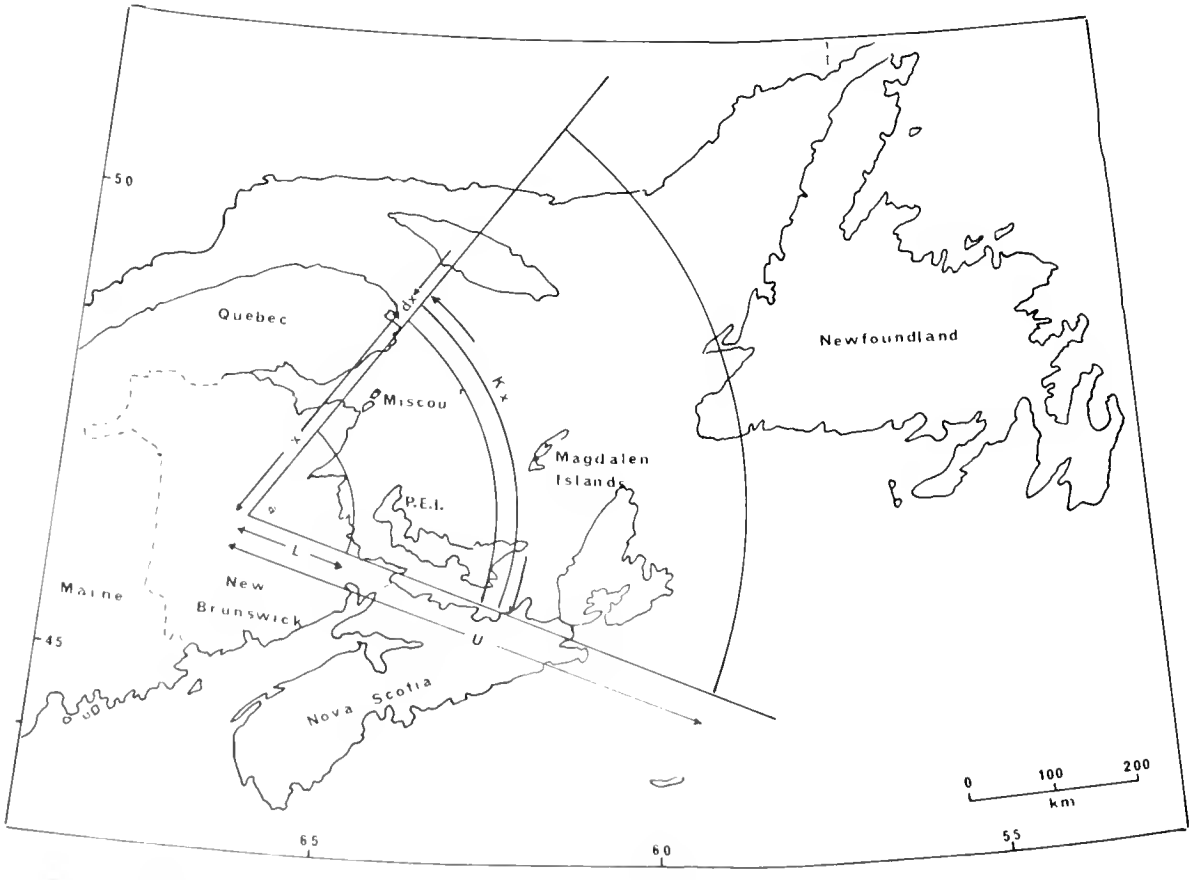

FIGURE 1 *Map of the Atlantic Provinces showing method of estimating DDT deposition into the Gulf of St. Lawrence*

$$\int_L^U f(x) . Kx.dx =$$

$$\int_L^U Ae^{-bx} Kx.dx = KA \int_L^U xe^{-bx} dx$$

Integrating by parts gives:

$$\text{Amount of DDT} = KA/b^2(Lb + 1)e^{-Lb} - (Ub + 1)e^{-Ub}$$

Results and Discussion

The 1967 and 1968 analysis results for the DDT group residues in rainwater are shown in Tables 1 and 2, respectively. DDT and/or its metabolites were detected in 76 of the 101 samples analyzed. In 1967, ΣDDT ranged from <0.01 to 0.90 μg/kg. In 1968, ΣDDT ranged from <0.01 to 1.33 μg/kg.

The dominant residue in almost all rainwater samples was *p,p'*-DDT. Using the Friedman two-way analysis of variance, *p,p'*-DDT has a rank sum of 344.5, compared to 245 for DDE, 228 for TDE, and 192.5 for *o,p'*-DDT. The ranking difference for *p,p*-DDT was significant at $P < 0.0001$ over all other compounds. Similiar rankings were found at individual sites; *p,p'*-DDT was the dominant residue in 11 of 12 groups. The differences in the ranking between the remaining residues were not significant. The percentage of *o,p'*-DDT was variable, but in half the groups, the percentage was between 10 and 20. The relatively small proportion of DDE shows that little DDT has been incorporated in biological tissue to be subsequently released to the atmosphere. The occurence of TDE may be due to photodecomposition of DDT *(8)*. The ratios of the various compounds of the DDT group are similar to those reported from Great Britain in the mid-1960s *(1, 10)*. The levels reported here range higher than do other published values for that period (Table 4).

The only other organochlorine compound detected was dieldrin; small concentrations (0.01 μg/kg) were found in five rainwater samples from two sites in 1967 and in 11 samples from four sites in 1968. Airborne dieldrin could have originated as dieldrin or from epoxidation of aldrin. The only potential source in the region was the agricultural use of aldrin in New Brunswick and adjacent areas for the control of root maggots (*Hylemya* spp.) in cruciferous crops.

Central New Brunswick was sprayed June 10-29, 1967, and June 4-28, 1968, to protect the forest against damage by spruce budworm (*Choristoneura fumiferana*). The aircraft operated 15 – 45 m above the tree canopy. Approximate ΣDDT and organophosphate emissions in thousands of kg active ingredient were, respectively, 190 and 57 in 1967 and 20 and 52 in 1968. About 40 thousand kg DDT was used in northern Maine in 1967 for spruce budworm control. None was used for that purpose in 1968. In western and northwestern New Brunswick and also in neighboring Prince Edward Island and Aroostook County, Maine, potato-growing is an important industry. Among the many pests of that crop, the flea beetle (*Epitrix cucumeris*) and, to a lesser extent, the Colorado potato beetle (*Leptinotarsa decemlineata*) were, at the time of the study, controlled by ground application of DDT in late June and early July. About 27 thousand kg DDT was used annually in New Brunswick for that purpose.

The prevailing wind during the collection periods was from the west and southwest. Meteorological data taken at the collection sites did not indicate a relation between peak residue concentrations in rainwater samples and periods of maximum insecticide use. The periods in June during which the samples were collected were likely out of phase

TABLE 4 *Comparison of DDT and metabolite levels found in precipitation in other investigations, 1965-74*

Region	Year	Residues, μg/kg			Reference
		p,p'-DDE	*p,p'*-TDE	*p,p'*-DDT	
Great Britain	1965	0.002-0.017	0.016-0.034	0.113-0.195	Abbott et al. *(1)*
U.S.A. (Ohio)	1965	0.005-0.030	not given	0.070-0.340	Cohen and Pinkerton *(3)*
Great Britain	1966-1967	0.007-0.028	0.007-0.034	0.018-0.066	Tarrant and Tatton *(10)*
Antarctica	1966-1967	ND	ND	0.040	Peterle *(7)*
Eastern Canada	1967-1968	ND-0.21[1]	ND-0.21[1]	ND-0.91[1]	This study
U.S.A. (Hawaii)	1970-1971	ND	not given	0.001-0.13	Bevenue et al. *(2)*
U.S.A. (New York)	1974	ND-trace	ND	ND-0.002	Peakall *(6)*

NOTE: ND = not detected
[1] Excludes known local contamination

with those during which forest spray aircraft were active, since no spraying took place when it was raining. It is possible, therefore, that insecticide drifting downwind from spray zones was not intercepted by rain at the collection sites. The occurrence of measurable amounts of DDT in rainwater before the spray programs started suggests more distant sources of contamination and also suggests that sprayed forest and agricultural areas in the region may contaminate the atmosphere for many months after spray programs are completed. Spencer et al. *(9)* concluded that a high percentage of organochlorine pesticides lost from plant surfaces is lost by volatilization. Lloyd-Jones *(5)* examined the rate of evaporation of DDT from planchets and screens and concluded that about half the DDT applied to field crops may enter the atmosphere. He found that the rate of evaporation was highly temperature-dependent. Thus the amount of DDT lost from surfaces during the cold New Brunswick winter months presumably would be very small. The dispersion of tree pollen over considerable distances has been documented by several investigators and reviewed by Lanner *(4)*. Concentrations of DDT and metabolites ranged from 0.544 to 1.01 mg/kg in tree pollen from the forest of central New Brunswick in 1968 (Table 5). Pollen was taken when some of the earliest, pre-spray samples of rainwater were collected that year. If present, such contaminated pollen may have contributed residues to the levels reported for those rainwater samples, since pollen was not separated from water before extraction and analysis.

It seems likely that DDT used in forestry and agriculture in the Maritime Provinces and Maine contributed to fallout of that pesticide in rain in the region. However, for the reasons outlined and since there was no clear spatial or temporal pattern of residue peaks in the rainwater samples, the contamination cannot be related directly to regional use of DDT during the period of investigation.

By using the equations developed above and mean values of DDT concentration in rain of 0.11 μg/kg at Miscou and 0.06 μg/kg at the Magdalen Islands, it can be calculated that about 5,000 kg DDT were deposited in the wedge-shaped area covering the Gulf of St. Lawrence from July to October in 1968. The time frame was chosen on the assumption that the evaporation of DDT from ground and plant surfaces would be severely reduced after the onset of cold weather in November. The calculation of DDT deposition is a rough one only, and completely depends on a very small number of measurements. The assumption that 0.06 μg/kg is typical of DDT fallout at the Magdalen Islands during that period cannot be tested. Although the calculations presented are only first approximations, they do support the current view that the aerial route is important in the transport of organochlorines in the environment.

TABLE 5. *DDT residue levels in tree pollen from central New Brunswick, Canada, 1968.*

Species	Residues, mg/kg			
	p,p' DDE	*p,p'* TDE	*o,p'* DDT	*p,p'* DDT
Jack pine	0.025	0.043	0.050	0.88
Red spruce	0.109	0.083	0.117	0.568
White birch	0.108	0.056	0.110	0.270
Red maple	0.110	0.077	0.110	0.564

Acknowledgments

We are much indebted to J. A. Keith, Canadian Wildlife Service, Department of Fisheries and the Environment, who conceived the project and was involved in technical discussions during this study. G.E.J. Smith, also of that agency, provided valuable advice on analysis and application of the residue data. Information on precipitation was supplied by the Atmospheric Environment Service, Department of Fisheries and the Environment.

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APPENDIX

Chemical Names of Compounds Discussed in This Issue

AROCLOR 1242	PCB, approximately 42% chlorine
AROCLOR 1254	PCB, approximately 54% chlorine
CHLORDANE	1,2,3,5,6,7,8,8-Octachloro-2,3,3a,4,7,7a-hexahydro-4,7-methanoindene. The technical product is a mixture of several compounds including heptachlor, chlordene, and two isomeric forms of chlordane
DDE	Dichlorodiphenyl dichloro-ethylene (degradation product of DDT) *p, p'*-DDE 1,1-Dichloro-2,2-bis(*p*-chlorophenyl) ethylene *o,p'*-DDE 1,1-Dichloro-2-(*o*-chlorophenyl)-2-(*p*-chlorophenyl) ethylene
DDT	Main component (*p,p'*-DDT) α-Bis(*p*-chlorophenyl) β β β-trichloroethane Other isomers are possible and some are present in the commercial product *o p'*-DDT [1,1,1-Trichloro-2-(*o*-chlorophenyl)-2-(*p*-chlorophenyl) ethane]
DIELDRIN	Not less than 85% of 1,2,3,4,10,10-Hexachloro-6,7-epoxy-1 4 4a,5,6,7 8,8a-octahydro-1,4-*endo-exo*-5,8-dimethanonaphthalene
ENDRIN	Hexachloroepoxyoctahydro-*endo,endo*-dimethanonaphthalene
HEPTACHLOR EPOXIDE	1,4,5,6,7,8,8-Heptachloro 2,3-epoxy-3a,4,7,7a-tetrahydro-4,7-methanoindan
LINDANE	*Gamma* isomer of 1,2,3,4,5,6-hexachlorocyclohexane
PCBs (Polychlorinated Biphenyls)	Mixtures of chlorinated biphenyl compounds having various percentages of chlorine
TDE	2,2-Bis(*p*-chlorophenyl)-1,1-dichloroethane (including isomers and dehydrochlorination products)

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SUBJECT AND AUTHOR INDEXES

Volume 11, June 1977—March 1978

Preface

Primary headings in the subject index include pesticide compounds, media in which pesticide residues are monitored, and major concepts related to the monitoring of pesticides in the environment. Pesticide compounds are listed by common names; trade names are used for those which have no common names.

Secondary headings cross-reference the primary headings. For a paper which discusses five or more organochlorines the compounds are grouped by class under media and concept headings but each compound appears individually under the primary headings for pesticide compounds.

In the author index all information on a paper appears in the senior author's citations: associate authors, title of the paper, and volume, issue, and pages where the article was published. Names of associate authors are cross-referenced as minor headings, but the reader is referred to the senior author's entry for the paper's complete citation.

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The *Pesticides Monitoring Journal* welcomes from all sources qualified data and interpretative information on pesticide monitoring. The publication is distributed principally to scientists, technicians, and administrators associated with pesticide monitoring, research, and other programs concerned with pesticides in the environment. Other subscribers work in agriculture, chemical manufacturing, food processing, medicine, public health, and conservation.

Articles are grouped under seven headings. Five follow the basic environmental components of the National Pesticide Monitoring Program: Pesticide Residues in People; Pesticide Residues in Water; Pesticide Residues in Soil; Pesticide Residues in Food and Feed; and Pesticide Residues in Fish, Wildlife, and Estuaries. The sixth is a general heading; the seventh encompasses briefs.

Monitoring is defined here as the repeated sampling and analysis of environmental components to obtain reliable estimates of levels of pesticide residues and related compounds in these components and the changes in these levels with time. It can include the recording of residues at a given time and place, or the comparison of residues in different geographic areas. The Journal will publish results of such investigations and data on levels of pesticide residues in all portions of the environment in sufficient detail to permit interpretations and conclusions by author and reader alike. Such investigations should be specifically designed and planned for monitoring purposes. The Journal does not generally publish original research investigations on subjects such as pesticide analytical methods, pesticide metabolism, or field trials (studies in which pesticides are experimentally applied to a plot or field and pesticide residue depletion rates and movement within the treated plot or field are observed).

Authors are responsible for the accuracy and validity of their data and interpretations, including tables, charts, and references. Pesticides ordinarily should be identified by common or generic names approved by national or international scientific societies. Trade names are acceptable for compounds which have no common names. Structural chemical formulas should be used when appropriate. Accuracy, reliability, and limitations of sampling and analytical methods employed must be described thoroughly, indicating procedures and controls used, such as recovery experiments at appropriate levels, confirmatory tests, and application of internal standards and interlaboratory checks. The procedure employed should be described in detail. If reference is made to procedures in another paper, crucial points or modifications should be noted. Sensitivity of the method and limits of detection should be given, particularly when very low levels of pesticide residues are being reported. Specific note should be made regarding correction of data for percent recoveries. Numerical data, plot dimensions, and instrument measurements should be reported in metric units.

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The Journal 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 short papers which present timely and informative data. These papers must be limited to two published pages (850 words) and should conform to the format for regular papers accepted by the Journal.

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