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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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SOIL

DDT Moratorium in Arizona—Agricultural Residues After Seven Years [1]

George W. Ware, Betty J. Estesen, Norman A. Buck, and William P. Cahill

ABSTRACT

The moratorium on agricultural use of DDT in Arizona that began in January 1969 proved very effective during the first 7 years of enforcement. Residues on green alfalfa declined significantly to a probable inherent level of 0.02 ppm wet weight. Soil residues of ΣDDT-related degradation products declined significantly, averaging 23 percent; residues in desert soils declined 60 percent. The ΣDDT half-life in irrigated soils was about 7 years; it decreased to 2.5 years in nonirrigated soils.

Introduction

The moratorium on agricultural use of DDT in Arizona began in January 1969 (*2, 4, 5*). This is the fourth and probably last report on the status of DDT residues and ΣDDT-related degradation products, after 18 years of unrestricted use and 4 years of restricted use in Arizona.

Analytical Methods

Alfalfa and soil samples were collected as described in previous reports (*2, 4, 5*) from the three major irrigated areas in Arizona: Salt River Valley, which surrounds Phoenix; Pinal County; and the Yuma mesa and valley in Yuma County. Desert soil samples, but only the top 0.25 inch, adjacent to these areas were also collected. In addition an earlier study (*3*) was continued to provide reference standards and continuity for the seven-year period (Table 1). The sampling sites are located on a 60-mile Maricopa County east-west transect along Baseline Road, much of which is now residential.

Alfalfa and soil samples were extracted and cleaned by procedures previously described (*2–5*).

TABLE 1. *ΣDDT residues in green alfalfa, Baseline Rd., Maricopa Co., Arizona, 1967–75*

	ΣDDT Residues, ppm						
Sample	1967 Aug.	1968 Sept.	1969 Sept.	1970 Sept.	1971 Sept.	1972 Sept.	1975 Oct.
2	—	0.220	0.038	0.050	0.020	0.023*	0.009*
3	0.283	—	0.027	0.030	—	0.025*	0.007*
4	0.170	0.120	0.038	0.037	0.031	0.022	0.016*
5	—	0.060	0.020	0.024	0.011	0.029*	0.009*
6	0.277	—	0.035	0.022	—	0.008*	—
8	0.794	—	—	0.027	0.038	0.013*	0.023
9	—	0.076	0.034	0.042	0.020	0.029*	0.027
10	0.350	0.092	0.054	0.162	0.027	0.031	0.022*
11	0.453	0.580	0.064	0.047	0.085	0.056	0.027*
12	0.299	0.077	0.025	0.038	—	0.023*	0.014*
13	0.606	—	—	0.021	0.027	—	0.008*
Means	0.404d	0.175c	0.037b	0.045b	0.032b	0.026b	0.016a

NOTE: — = no sample analyzed.
* = substitute adjacent fields
Means with same letter are not significantly different at the 0.05 level.

Samples were analyzed by electron-capture gas-liquid chromatography (EC-GLC). Recovery standards and analytical reagent blanks were also extracted and cleaned each day. Recoveries were consistently above 90 percent; however, the data presented have not been corrected. The minimum sensitivity of the method was arbitrarily set at 0.02 ng for *p, p'*- and *o, p'*-DDT, DDE, and TDE. Standard curves extended from 0.03 ng to 0.10 ng. The sensitivities were 0.001 ppm for alfalfa and 0.003 ppm for soil. Results are based on a minimum sample size and 6 μl extract injected into the chromatograph.

Analytical EG-GLC confirmatory tests were conducted randomly using a double-length GLC column at the same temperatures as those used in the previous study (*2*). Because of low levels of ΣDDT and interfering peaks of toxaphene which may have drifted from nearby cotton fields, all alfalfa extracts were dehydrohalogenated after cleanup on Florisil and residues were measured only as *o,p'*- and *p,p'*-DDE as described by Cahill et al. (*2*); results were combined when measurable levels of *o,p'*-DDE were found.

[1] Department of Entomology, The University of Arizona, Tucson, AZ 85721. This paper submitted to Regional Project W-45, "Residues of Pesticides and Related Chemicals in the Agricultural Environment—Their Nature, Distribution, Persistence, and Toxicological Implications." University of Arizona Agricultural Experiment Station Journal Series No. 2759

Results and Discussion

Residues observed in alfalfa and soil samples during the past 7 years are presented in Tables 1–3 as ΣDDT. The Student-Newman Keul's test was used to analyze differences among residue means for the various sampling dates. Comparisons were made on least-square means in the soil samples (Table 3) because there were too few samples. Residues on alfalfa from all four areas shown in Tables 1 and 2 appear to have leveled off at about 0.02 ppm. September values for Yuma County alfalfa were consistently high from 1969 through 1972. However, these values were well below 0.02 ppm in 1975.

Residue levels in alfalfa soils declined from the previous sampling period, September 1972 (Table 3). In the past, yearly examination of these soil residues indicated almost imperceptible changes. After 3 years, however, the residues had declined significantly, an average of 23 percent. Residues in the desert soils declined 60 percent. This suggests that the ΣDDT half-life in the irrigated soils of Arizona is about 7 years, and decreases to about 2½ years in the desert or nonirrigated soils.

ΣDDT residues now found in the agricultural soils of Arizona are shifting steadily toward higher proportions of DDE. The ratio of DDE:DDT in these soils shifted from 56:44 in 1972 to 62:38 in 1975. In the desert soils, the shift was approximately the same: from 65:35 in 1972 to 71:29 in 1975. These data suggest that ΣDDT residues are declining at a predictable rate, probably both by volatility and conversion to metabolites not measured with the analytical methods used in this study.

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TABLE 2. *ΣDDT residues in green alfalfa during 1969–75 DDT moratorium, Arizona*

	ΣDDT Residues, ppm						
Sample	1969 Jan	1969 Sept	1970 Sept.	1971 Sept	1972 Jan	1972 Sept	1975 Oct
Maricopa County, Arizona							
1	0.087	0.042	0.057	—	—	—	0.019*
2	0.303	0.062	0.050	0.025	—	0.039*	0.037*
3	0.102	0.078	0.093	0.038	—	—	0.011*
4	0.107	0.047	0.076	0.037	—	0.046*	0.017
5	0.049	0.030	0.025	0.007	—	0.011	0.015*
6	0.113	0.064	0.060	0.051	—	0.045*	0.041*
7	0.082	0.034	0.023	—	—	0.055	0.21
8	0.125	0.056	—	—	—	—	—
9	0.085	0.044	0.101	—	—	—	—
10	—	—	0.080	0.059	—	—	—
Means	0.117c	0.051b	0.063b	0.036b	—	0.39b	0.023a
Pinal County, Arizona							
1	0.047	0.042	0.034	0.055	—	0.041*	—
2	0.047	0.031	0.059	0.036	—	—	0.068*
3	0.142	0.187	—	—	—	—	0.006*
4	0.231	0.076	0.071	0.072	—	0.025	—
5	0.092	0.130	0.045	—	—	0.025*	—
6	0.038	0.058	0.045	—	—	—	—
7	0.079	0.118	0.059	0.038	—	0.044	0.023*
8	0.068	0.071	0.031	0.034	—	0.018	0.077*
9	0.054	0.068	0.057	0.060	—	—	0.006*
Means	0.088b	0.086b	0.050a	0.049a	—	0.031a	0.036a
Yuma County, Arizona							
1	0.047	0.373	—	0.120	0.025	0.032	0.016*
2	0.039	0.098	—	—	0.010*	0.017*	0.008*
3	0.049	0.256	0.084	0.270	0.073*	0.040*	0.040
4	0.057	0.093	—	—	0.055*	0.075*	0.025
5	0.057	0.545	0.063	0.340	0.047*	0.290*	0.030*
6	0.044	0.317	—	—	0.035*	0.300*	0.032*
7	0.059	0.241	—	—	0.026*	0.190*	0.034*
8	0.036	0.045	0.034	0.031	0.039*	—	0.005*
9	0.021	0.056	—	—	0.015*	—	—
10	0.046	0.074	0.051	0.050	0.028	0.045	0.006
Means	0.46a	0.210b	0.058a	0.162b	0.035a	0.123b	0.022a

NOTE: — = no samples analyzed
* = substitute adjacent fields.
Means with same letter are not significantly different at the 0.05 level

TABLE 3. *ΣDDT residues in soils during 1969–75 DDT moratorium, Arizona*

Field No	1969 Jan., ppm Residues				1972 Sept., ppm Residues				1975 Oct., ppm Residues			
	DDE	*o,p'*-DDT	*p,p'*-DDT	Total	DDE	*o,p'*-DDT	*p,p'*-DDT	Total	DDE	*o,p'*-DDT	*p,p'*-DDT	Total
Maricopa County												
1	0.35	0.04	0 12	0.54	0 40	0 04	0 11	0.55	0 43	0 10	0 13	0 66
2	0.48	0.17	0.78	1 54	0 98	0.18	0.47	1 63				
3	0.33	0.07	0.16	0 59	1 24	0 13	0.32	1 69	1 0	0.10	0.23	1.33
4	0.49	0.05	0 17	0 74	0 58	0.05	0.23	0 86	0 40	0.03	0 12	0.55
5	0.29	0.05	0.09	0.44	0.17	0.01	0 05	0.23	0 17	0.01	0.02	0.21
6	2 10	0.43	1 10	3 93	2 58	0 28	0 96	3.82	2 24	0 31	0.58	3.13
7	0.84	0.11	0 23	1 22	0 92	0.09	0 29	1.30	0 68	0.06	0.14	0.88
8	2 22	0.38	1.29	4.00	2.37	0 27	1 21	3.85	1 96	0 24	0 98	3.18
9	1.18	0.21	0 91	2 41	1 12	0 17	0 77	2.06	0 83	0 14	0.55	1.51
10	—	—	—	(0.24)	0.31	0 04	0 07	0 42	0.24	0 02	0.06	0.32
Means	0.92	0.17	0.54	1 57a	1 07	0 13	0 45	1 64a	0.883	0 11	0.31	1.31b
Desert												
1	0.08	<0.01	0.03	0 13	0 43	0.07	0 09	0 59	0 04	0.00	0 02	0 06
2	0.24	0 02	0.06	0.35	0.28	0 03	0.58	0 89	0 04	0.01	0.02	0 06
3	0.44	0 04	0 15	0 67	0.18	0 02	0 04	0.24	0.06	0 01	0.02	0.10
4	—	—	—	(2.39)	0.54	0 08	0 06	0.68	0 69	0.10	0 19	0 98
Means	—	—	—	0 89a	0.36	0.05	0.19	0 60a	0.21	0 03	0.06	0.30b
Pinal County												
1	0.64	0 48	2.43	3.77	0 74	0 34	2 64	3.72	0 59	0 24	2.51	3.34
2	0.27	0 15	1.03	1.52	0 41	0.13	0 96	1 50	0 37	0.09	1 03	1 49
3	1.05	0 32	1.38	2.75	1 16	0 16	0 80	2.12	0 64	0.09	0 42	1 16
4	0.99	0 27	1 04	2.30	1.40	0 18	0.74	2.32	1.60	0 21	0.25	2.05
5	0.16	0 02	0 21	0 41	0 25	0 02	0 16	0 43	0 19	0.02	0 10	0 31
6	0.06	0.01	0.07	0 14	0 07	0.01	0 04	0 12	0 04	0.00	0 05	0 08
7	1 09	0.28	1 37	2 74	1 63	0.20	0 80	2 63	1.32	0 16	0.31	1.79
8	0.09	<0.01	0.04	0 14	0 08	0.01	0 02	0 11	0 05	0.00	0 01	0 06
9	0.67	0 09	0 29	1 06	0 74	0.03	0 06	0 83	0.59	0 05	0 08	0 72
10	0.66	0 14	0 36	1 16	1 19	0 15	0 39	1.73	0.91	0.09	0.08	1.08
Means	0.57	0 18	0 82	1 60a	0 69	0 12	0 66	1 55a	0 63	0.10	0.48	1 21b
Desert												
1	0.09	<0.01	0.06	0.16	0.17	0.02	0 12	0.31	0 04	0.00	0.01	0 06
2	0.18	0 01	0.11	0 32	0 21	0.02	0 21	0 44	0 07	0 00	0.02	0 09
3	0.05	0 03	0 10	0 21	0 06	0.01	0 02	0 09	0 04	0 01	0 01	0 06
4	0.09	0 03	0 10	0 25	0 77	0.07	0 09	0 93	0 49	0 05	0 07	0.61
Means	0.10	0.02	0.09	0.24a	0 30	0.03	0 11	0 44a	0 16	0.02	0 03	0 20b
Yuma County												
1	0.10	<0.01	0.07	0 17	0 12	0 02	0 03	0 17	0 06	0.01	0.01	0.08
2	0.24	0.05	0 25	0 54	0 20	0.03	0 07	0 30	0 24	0.03	0.06	0 33
3	0.72	0 16	0.72	1 60	0 79	0 16	0.49	1 44	0 71	0.15	0 29	1.15
4	0.59	0.11	0.47	1 17	0 98	0.15	0.46	1 59	0.84	0.13	0.22	1.18
5	0.48	0.05	0 30	0 83	0 75	0.12	0.34	1.21	0 54	0.10	0.15	0 79
6	0.29	0.16	0 74	1 19	0 48	0.10	0 43	1.01	0 45	0 08	0.29	0 81
7	1.29	0.07	0.37	1.73	1.11	0 09	0 60	1 80	0 91	0.06	0 01	0 97
8	0.06	0.01	0.01	0.08	0 05	<0 01	<0.01	0 07	0.02	0.00	0.00	0 02
9	0 00	0.00	0.00	0 00	<0.01	<0 01	<0.01	0 03	0 00	0.00	0.00	0.00
10	0.26	0.02	0.03	0 31	0 17	0 01	0 02	0.20	0.08	0.00	0 01	0 10
Means	0 40	0 06	0.30	0.76a	0 47	0.07	0.25	0 78a	0 39	0.06	0.10	0 54b
Desert												
1	0.27	0.02	0 07	0.36	0 24	0.06	0 09	0 39	0 10	0.03	0.03	0 16
2	0 03	0 01	0 02	0.06	0 02	0.01	0 02	0 05	0 02	0.00	0.01	0 03
3	0.02	0.01	0 03	0.06	0 02	0.01	0.02	0.05	0 02	0.00	0 02	0.05
4	0 00	0 00	0.01	0.01	0 79	0.07	0 15	1 04	0.00	0.00	0.00	0 01
Means	0.08	0.01	0.03	0.12a	0.27	0.04	0.08	0.38b	0.035	0.01	0 02	0.06a

NOTE: — = no sample analyzed
Figures in parentheses are missing values calculated by randomized blocks missing value formula
Means with same letter are not significantly different at the 0.05 ppm level

FISH, WILDLIFE, AND ESTUARIES

Organochlorine Insecticide, Polychlorinated Biphenyl, and Metal Residues in Some South Dakota Birds, 1975–76 [1]

Yvonne A. Greichus, Brian D. Gueck, and Barbara D. Ammann

ABSTRACT

Chlorinated hydrocarbon insecticide, polychlorinated biphenyl (PCB), and metal residues were measured in tissues of common crows (Corvus brachyrhynchos), *American coots* (Fulica americana), *starlings* (Sturnus vulgaris), *and Franklin's gulls* (Larus pipixcan), *of South Dakota in 1975–76. Insecticides and PCBs were analyzed by column, thin-layer, and gas-liquid chromatography. Metals were analyzed by atomic absorption spectrophotometry.*

DDE was the most prevalent residue; it was detected in 93 percent of all samples and averaged 66 percent of the total residues in the carcass. Average values ranged from 0.04 ppm to 0.54 ppm. Dieldrin was detected in 61 percent of all samples and averaged < 0.01 ppm to 0.15 ppm. TDE and DDT were found in 27 percent and 15 percent, respectively, of all samples, and the averages for both ranged from < 0.01 ppm to 0.06 ppm. Heptachlor epoxide and lindane were detected in some samples. PCBs were not found above the minimum detectable level, 0.1 ppm, in any sample.

Gulls had higher insecticide and metal residues than had coots, starlings, or crows. Arsenic values averaged 1.4 ppm dry weight in carcass samples from the four species of birds. Cadmium, copper, manganese, lead, and zinc averaged 0.10, 0.94, 4.8, 1.0, and 69 ppm dry weight, respectively, and were no higher than values reported in some birds from other areas.

Introduction

Organochlorine insecticides have been used in South Dakota since 1946 for the control of noxious insects (*4*). Although many of these insecticides have been banned or limited, residues of some of the more persistent compounds such as DDT, dieldrin, and lindane are still commonly found in birds of South Dakota (*6, 7*).

Four common species of South Dakota birds with distinctly different feeding habits were analyzed in 1975–76 for eleven insecticide residues, six metals, and polychlorinated biphenyls (PCBs) to determine present levels of these chemicals so that comparisons could be made in future studies.

Methods and Materials

INSECTICIDE AND PCB ANALYSIS

Seven common crows (*Corvus brachyrhynchos*), six American coots (*Fulica americana*), six starlings (*Sturnus vulgaris*), and six Franklin's gulls (*Larus pipixcan*) were analyzed. Organochlorine insecticide and PCB residue levels were measured on a wet-weight basis in brain, liver, feather, and carcass samples from each bird. Metal levels were measured on a dry-weight basis for each bird. Samples were analyzed for lindane, heptachlor, heptachlor epoxide, dieldrin, aldrin, methoxychlor, endrin, toxaphene, DDE, TDE, DDT, zinc, cadium, lead, copper, arsenic, and manganese.

All birds were killed by shotgun. Gulls were collected September 2, 1975, approximately three miles west of Nunda, South Dakota, while feeding in a freshly plowed field. Coots were collected September 15, 1975, approximately five miles southeast of Arlington, in a marsh. Starlings were obtained February 14, 1976, near Crocker. South Dakota Game, Fish, and Parks personnel collected crows April 6, 1976, near Richmond Lake in Brown County. All specimens collected appeared to be normal and healthy.

Authors had intended to use only adults for the study but could find no literature on estimating the age of crows and starlings. They selected the seven heaviest crows for study and they analyzed all starlings collected because only six had been taken. Coots were aged by leg color (*9*) and gulls by plumage (*15*). All coots and five of six gulls analyzed were judged to be adults.

[1] Station Biochemistry Section, Chemistry Department, South Dakota State University, Brookings, SD 57007. This paper is being published with the approval of the Director of the South Dakota Agricultural Experiment Station as Publication No 1515 of the journal article series.

Each specimen was necropsied to remove tissue samples and to determine sex, stomach contents, and general body condition. Technicians removed 5 g of feathers, finely cut them, and wrapped them with aluminum foil. Brains and livers were removed, weighed, and stored in glass jars. Carcass samples consisted of the entire body minus beak, legs, stomach contents, and the samples of feather, brain, and liver previously removed. After necropsy, the carcasses were wrapped and frozen in aluminum foil; several days later they were homogenized by grinding with a Toledo meat chopper, and frozen in glass jars for later analysis. All glassware used for storage and later insecticide analysis was washed in detergent, rinsed with distilled water, and baked at 425°C for at least 3 hours to remove organic contamination.

Samples were extracted and purified for chlorinated hydrocarbon residues analysis by a Florisil column method (*16*) as modified by Greichus et al. (*8*). Methods for separating PCBs and insecticides and quantitating PCBs have been described by Greichus et al. (*5*). One gram of carcass and liver and 0.5 g of brain and feathers were analyzed.

Gas chromatograph:	Varian Aerograph Model 2100
Detectors:	^{63}Ni and Sc^{3}H electron-capture
Recorders:	Beckman Ten Inch, 1 mv
Columns:	6-ft × 1/16-inch borosilicate glass
Packing:	15 percent QF-1 silicone (Fluoro) or 1:1 mixture of 15 percent QF-1 and 10 percent DC-200 silicone, both on 60–100-mesh Chromosorb W (HP), acid-washed and dimethylchlorosilane-treated
Carrier gas:	Nitrogen at 40 ml/minute
Column temp.:	210° C
Injector temp.:	220° C
Detector temp.:	280° C

Identity of individual insecticides was verified by using thin-layer chromatography (*2*, *4*). Insecticides and PCBs were recovered at 89 percent and 95 percent, respectively. Minimum detection limits were set at 0.01 ppm and 0.1 ppm for insecticides and PCBs and were corrected for percent recovery but values for metals were not corrected.

METAL ANALYSIS

Zinc was determined with a Perkin-Elmer Model 303 flame atomic absorption spectrophotometer. Lead, arsenic, cadmium, copper, and manganese were determined with a Perkin-Elmer Model 503 atomic absorption spectrophotometer equipped with a heated HGA-2100 graphite furnace and a Sargent-Welch Model SRLG recorder. A Perkin-Elmer deuterium arc power supply Model 560 background corrector was used in conjunction with the spectrophotometer when necessary. Operating conditions of the instrument were essentially the same as those given by the manufacturer. Before analysis, 0.5 g dry weight of each sample was digested in 10 ml of concentrated nitric acid on a micro-Kjeldahl digestion apparatus until 2 ml of solution remained. An additional 5 ml of nitric acid was added, and the solution was boiled until 1 ml remained. Samples were reconstituted to 10 ml with distilled water and analyzed directly. Average recoveries for metals were copper 87, cadmium 91, manganese 82, arsenic 73, lead 79, and zinc, 94 percent.

Minimum detection limits used for heavy metals were 0.01 ppm for cadmium, 0.1 ppm for arsenic and lead, 0.5 ppm for copper and manganese, 1.0 ppm for zinc. In calculations of averages and totals, less than (<) values were included and given one-half the stated value; that is, a value of less than 0.1 ppm is recorded as 0.05 ppm.

Results and Discussion

INSECTICIDES AND PCBs

Average insecticide residue concentrations for common crows, starlings, American coots, and Franklin's gulls are given in Table 1. Endrin, heptachlor, methoxychlor, aldrin, and PCBs were not detected above the minimum detectable levels in any of the 100 samples analyzed. Toxaphene detected in starling feathers was judged to have been an inadvertent contaminant from a container used to carry the birds. Lindane was found in only two crows and was not used in the calculation of average total insecticides. One crow had carcass and liver residues of 0.01 ppm and 0.11 ppm lindane, respectively; another crow had a carcass residue of 0.01 ppm. Heptachlor epoxide was detected in crow carcass and liver samples and in one crow brain. Dieldrin residues were found in all species, all four tissue types, and in 61 percent of all samples, except the coot. Dieldrin was either absent from the tissues of the coot, or present in the liver at the limit of detection, 0.01 ppm. Dieldrin concentrations in the brain and feathers of the four species were usually below or slightly above the 0.01 ppm lower analytical limit.

DDT and its metabolites were the residues found most consistently. DDE was the most prevalent of the DDT complex and was found in 93 percent of all samples. TDE and DDT were detected in 27 percent and 15 percent, respectively, of all samples.

Starlings reflect the general environmental levels of organochlorine insecticides and metals available to them in South Dakota because they are often year-around terrestrial residents. Coots and Franklin's gulls do not reflect true South Dakota contamination levels because they are summer resident only and are subject to migratory contamination in other areas. The low levels of TDE and DDT may reflect the decreased use and eventual banning of DDT in

TABLE 1. *Organochlorine insecticide residues in South Dakota birds, 1975–76*

Bird	Average Residues, ppm (μg/g) Wet Weight					
	Heptachlor Epoxide	Dieldrin	DDE	TDE	DDT	Total Insecticides
CARCASS						
Crow	0.06	0.13	0.54	0.04	0.06	0.84
Coot	<0.01	<0.01	0.04	<0.01	<0.01	0.06
Starling	<0.01	0.02	0.06	<0.01	<0.01	0.10
Gull	<0.01	0.15	0.44	0.06	<0.01	0.66
LIVER						
Crow	0.10	0.05	0.41	0.04	0.02	0.61
Coot	<0.01	0.01	0.02	<0.01	<0.01	0.05
Starling	<0.01	0.04	0.06	<0.01	<0.01	0.12
Gull	<0.01	0.04	0.10	0.04	<0.01	0.20
BRAIN						
Crow	<0.01	0.07	0.05	<0.01	<0.01	0.13
Coot	<0.01	<0.01	0.01	<0.01	<0.01	0.04
Starling	<0.01	<0.01	0.02	<0.01	<0.01	0.04
Gull	<0.01	<0.01	0.02	<0.01	<0.01	0.04
FEATHERS						
Crow	<0.01	0.06	0.04	<0.01	<0.01	0.11
Coot	<0.01	<0.01	0.02	<0.01	<0.01	0.04
Starling[1]	—	—	—	—	—	—
Gull	<0.01	0.02	0.05	<0.01	<0.01	0.08

Note: Seven crows and six each of coots, starlings, and gulls were analyzed.
[1] Starling feathers were contaminated with toxaphene at bird-collection site; no residues are reported here.

the United States in 1973, although DDE is still common in the environment.

Nationwide monitoring of mallard and black duck wings by the Fish and Wildlife Service, U.S. Department of the Interior, since 1965 has shown DDE to the the predominant residue (*10, 11*). Results of the monitoring in 1965–66 showed DDE to be the predominant residue, followed by DDT, TDE, dieldrin, and heptachlor expoxide; in 1969, DDE was followed by PCBs, DDT, dieldrin, TDE, and heptachlor epoxide. In both studies, organochlorine residues were generally highest in the Atlantic and Pacific flyways and lowest in the Central flyway of which South Dakota is a part, and in the Mississippi flyway.

Total insecticide residues were consistently higher in crows than in other species. Franklin's gulls had the second highest total residue level, followed by starlings and American coots. In brain samples, however, all three species had approximately equal concentrations. Carcass samples usually had the highest insecticide levels, followed by livers; brains and feathers were about equal.

Martin (*13*) analyzed carcasses of starlings from 128 areas of the United States in 1967–68 and found DDT, its metabolites, and dieldrin in all sites. At four South Dakota sites, the average residues for 1967–68 ranged from 0.103 ppm to 1.925 ppm DDE, 0.013 ppm to 0.018 ppm TDE, 0.018 ppm to 0.030 ppm DDT, and 0.012 ppm to 0.080 ppm dieldrin. Heptachlor epoxide and lindane were occasionally found at all South Dakota sites. Average total insecticide residues were 0.234, 0.201, 2.054, and 0.334 ppm at the four sites (*13*). Starlings monitored for the present study in 1976 had lower average total insecticides, 0.10 ppm, than had birds in any of the four South Dakota sites studied by Martin (*13*).

METALS

Average concentrations of metals in carcasses of crows, coots, gulls, and starlings are reported in Table 2. Values are given on a dry-weight basis but can be converted to the approximate wet weight by multiplying the value by 0.43, which was the average dry weight of 1 g of bird carcass. Arsenic levels were similar in all four types of birds. Converted to wet weight, arsenic residues were greater than those reported by Martin and Nickerson (*14*). Starlings collected from 50 sites in the United States contained < 0.05 ppm wet weight arsenic except for one sample with 0.21 ppm arsenic (*14*). Gulls averaged 0.21 ppm cadmium, higher than residues in other birds of this study but lower than some values reported for starlings by Martin and Nickerson (*14*).

TABLE 2. *Metal residues in South Dakota bird carcasses, 1975–76*

Metal	Average Residues, ppm (μg/g) Dry Weight[1]			
	Gulls	Coots	Starlings	Crows
Arsenic	1.6	1.5	1.6	1.0
Cadmium	0.31	0.08	0.10	0.03
Copper	1.8	0.75	0.51	0.75
Manganese	4.5	9.8[2]	4.0	4.2
Lead	3.2[2]	0.86	0.77	0.72
Zinc	82.0	71.0	75.0	52.0

Note: Seven crows and six each of coots, starlings, and gulls were analyzed.
[1] Residues can be converted to wet weight by multiplying each value by 0.43, the average dry weight of 1 g of bird carcass.
[2] Two birds were analyzed.

Gulls also had higher concentrations of lead than had coots, starlings, or crows. The gulls could have been contaminated in areas other than South Dakota because they are migratory. A possible cause could be the ingestion of shot. Waterfowl are susceptible to shot ingestion in wetland areas; upland birds are susceptible to a lesser extent in terrestrial areas (*1*). Lead residues in South Dakota starlings averaged 0.36 ppm in 1971 (*14*), which is close to 0.33 ppm wet weight found among starlings in the present study.

Manganese, copper, and zinc are essential dietary elements and are not usually considered contaminants. Levels of copper and zinc (Table 2) reported for the four types of birds were not unusual. Considerably higher levels of copper (21 ppm wet weight) and zinc (76 ppm wet weight) have been found in livers of white pelicans (*12*). Manganese concentrations of 9.8 ppm were more than twice as great in coots than in other birds, possibly because their diet contains aquatic plants rich in this element. Some aquatic plants have comparatively high levels of manganese (660 ppm dry weight) (*3*).

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Organochlorine Pesticide Residues in Florida Birds of Prey, 1969–76

David. W. Johnston [1]

ABSTRACT

Chlorinated hydrocarbon pesticide residues, especially DDT and its metabolites, were determined in 71 individuals of 14 species of predatory birds obtained in Florida between 1969 and 1976. Of the 71 birds, 68 contained p,p'*-DDE or another DDT metabolite; 34 contained dieldrin. DDE was found in 93 percent of the 57 adipose tissue samples, all the 9 brain samples, and 89 percent of the 62 uropygial gland samples. Of the 65 birds taken since 1972, 61 contained DDE in at least one of these three tissues. The annual average of ΣDDT in adipose tissue and uropygial gland over the 6-year span was approximately 5 ppm wet weight. From 1973 to 1976, no significant increase or decrease in pesticide burdens was detected. Some birds had no DDE whereas others contained up to 76 ppm ΣDDT. None of the data suggest that any of the birds of prey had died of DDT or DDT metabolite poisoning.*

Introduction

For approximately two decades in North America, much public and scientific interest has been focused on population declines of various birds of prey including eagles, osprey (*Pandion haliaetus*), and peregrine falcon (*Falco peregrinus*). In some species, correlations have been made or suspected between pesticide burdens, especially DDE, and mortality, population declines, or altered physiological processes resulting in impaired reproductive performances (*8, 18*). Eggshell thinning is now believed to be a result of high DDE burdens, both in captive and feral birds of prey (*13, 14, 17*). One might anticipate high pesticide burdens in birds of prey because they are usually terminal members of food chains, and thus can concentrate the fat-soluble chlorinated hydrocarbon pesticides. In most published accounts dealing with these birds, pesticide residues were extracted from eggs or nestling birds or from birds experimentally fed DDT (*4, 13, 14*); there are few published accounts of body burdens in adults except for a limited number of autopsied birds found dead and suspected of pesticide poisoning. In fact, virtually nothing has been published on body burdens in feral adult birds of prey which reportedly produced thin eggshells. Thus, to date, pesticide burdens at levels presumably not impairing reproduction are poorly documented (*2*). In the present report, some organochlorine pesticide residues extracted from birds of prey obtained recently in Florida are quantitated.

Sampling Methods

The birds analyzed were obtained between 1969 and 1976, chiefly in northcentral Florida near Gainesville. Most birds were fresh roadkills or were illegally shot by hunters. A few were picked up alive in a weakened condition or were having convulsions; they were kept in an aviary, and died within 24 hours. With the possible exception of the latter birds, the present report includes birds dying accidentally, that is, there was no a priori suggestion that any pesticide burden contributed to death.

The sample includes two orders (Falconiformes: vultures, kites, hawks, falcons, osprey, caracara; Strigiformes: owls). In all, 6 families, 12 genera, 14 species, and 71 individuals were analyzed.

Analytical Procedures

From each specimen, whether fresh or previously frozen in individual plastic bags, samples of subcutaneous adipose tissue (fat) and/or the entire uropygial gland and/or the cerebrum were removed for analysis. Recently, a number of investigators have indicated the possibility of using the unique avian uropygial, or preen, gland as an indicator of pesticide burdens in birds (*3, 4, 11*). In feral, migratory songbirds, Johnston (*11*) reported a high correlation,

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$r = 0.7568$, of ΣDDT between adipose tissue and uropygial gland. In the present study, birds varied in the degree of obesity; in some, essentially no fat could be located, so only the gland or brain was used for analysis. For 59 samples of fat, the mean sample weight was 1.1224 g; for 62 samples of uropygial glands, the mean weight was 0.5590 g; and the mean brain weight taken from 9 birds was 4.1375 g. Each sample was individually thoroughly homogenized in sodium sulfate, and extracted for at least 12 hours with petroleum ether in a Soxhlet apparatus. The lipid extract was evaporated to dryness, weighed, and partitioned with acetonitrile and hexane.

All samples were analyzed on a Model 600-D Varian gas chromatograph with the following instrument parameters and operating conditions:

Detector:	electron-capture
Column:	6-ft × ¼-inch glass, packed with a mixture of 6.4 percent OV-210 and 1.6 percent OV-17 (1+1) on Chromosorb W
Temperatures:	injection port 210° C column 212° C detector 215° C
Carrier gas:	nitrogen flowing at 45 ml/minute

Recoveries for the organochlorine compounds ranged from 75 percent to 95 percent. Sensitivity was approximately 0.01 ppm.

Results

Table 1 contains the results of analyses for the 71 birds of prey. Tissues analyzed for the individual birds were not

TABLE 1. *Chlorinated pesticide burdens in Florida birds of prey, 1969–76*

County	Date	Sex Age [1]	Tissue [2] and Sample Weight, g	Residues, ppm Wet Weight		
				p,p'-DDE	ΣDDT	Dieldrin
CATHARTES AURA (TURKEY VULTURE)						
Alachua	Nov 71	UNK	A (2.0751)	3.37	3.55	0
Levy	Apr 73	M	A (3.0238)	1.32	3.00	0.07
			B (6.0752)	0.17	0.17	0
Alachua	Apr 73	F	A (4.0514)	0.78	1.32	0.46
			B (5.5159)	0.11	0.11	0
Levy	May 73	M	A (2.5833)	1.59	2.28	0.07
			B (6.1278)	0.01	0.01	0
Levy	May 73	M	A (2.6192)	2.86	4.21	0.19
			B (4.7010)	0.04	0.04	0
Gilchrist	May 73	M	A (1.9826)	4.04	5.05	0.16
			B (5.7562)	0.69	0.87	0.02
Leon	July 74	F	A (1.8175)	3.03	3.74	0.74
			U (0.3899)	0.64	1.03	0
Leon	Sept 74	M	A (1.9901)	1.18	1.41	0.05
			U (0.3023)	0	0	0
Leon	Oct 74	F	A (1.4910)	9.05	13.35	0.30
			U (0.3417)	6.44	6.44	0
Leon	Oct 74	F	A (1.5841)	7.89	9.72	1.11
			U (0.4800)	5.00	5.00	0.16
CORAGYPS ATRATUS (BLACK VULTURE)						
Alachua	Jan. 72	M	F (2.0323)	6.39	11.75	1.18
Marion	May 73	F	A (2.8724)	3.83	6.87	0.22
			U (1.0003)	10.50	15.25	0.50
Alachua	May 73	F	A (2.2551)	1.26	1.49	0.04
			B (5.6169)	0.10	0.11	0

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TABLE 1 (continued). *Chlorinated pesticide burdens in Florida birds of prey, 1969–76*

County	Date	Sex/ Age [1]	Tissue [2] and Sample Weight, g	Residues, ppm Wet Weight		
				p,p'-DDE	ΣDDT	Dieldrin
CORAGYPS ATRATUS (BLACK VULTURE)—Continued						
Alachua	May 73	M	A (3.0264)	3.06	7.68	0.66
			U (0.8266)	5.44	6.84	0.24
Alachua	June 74	M	A (0.9518)	11.56	25.43	2.02
			U (0.8682)	9.33	14.74	0.69
ELANOIDES FORFICATUS (SWALLOW-TAILED KITE)						
Marion	May 75	M	U (0.6269)	0.48	0.48	0
ACCIPITER STRIATUS (SHARP-SHINNED HAWK)						
Alachua	Nov. 75	F	A (0.3009)	16.62	17.12	0
			U (0.0637)	19.62	19.62	0
ACCIPITER COOPERII (COOPER'S HAWK)						
Alachua	Sept. 73	F	U (0.2763)	12.12	13.02	0
BUTEO JAMAICENSIS (RED-TAILED HAWK)						
Alachua	Jan. 73	IM	A (1.8650)	6.25	6.25	0
			U (0.4830)	0.10	0.10	0
Alachua	Jan. 73	IMF	A (1.0422)	3.49	3.73	0.89
			U (0.4788)	0.11	0.11	0
Alachua	July 73	IM	A (0.7617)	0.37	0.48	0.04
			U (0.7570)	0.39	0.50	0
Alachua	July 74	IM	A (1.6963)	0.59	1.32	0
			U (0.4846)	0.21	0.21	0
Alachua	Dec. 74	F	A (0.5357)	0.37	0.37	0
			U (0.4527)	0	0	0
Alachua	Jan. 76	M	A (0.4694)	4.26	5.86	0.85
			U (0.3936)	0.38	0.38	0
Madison	Jan. 76	IM	A (0.5699)	6.14	7.19	2.37
			U (0.2852)	1.05	1.05	0.18
BUTEO LINEATUS (RED-SHOULDERED HAWK)						
Alachua	Jan. 72	F	A (1.3596)	0.64	1.04	0.06
			U (0.2853)	0.18	0.18	0
Alachua	Sept. 73	IM	A (0.3310)	0.45	1.21	0
			U (0.3104)	0.24	0.60	0
Alachua	Jan. 76	AD	A (0.1888)	7.15	7.15	0
			U (0.2552)	0.39	0.39	0
Pinellas	Jan. 76	F	A (0.2615)	34.42	61.76	38.24
			U (0.2514)	0.80	1.80	0.80
Baker	May 76	F	U (0.6186)	1.21	1.21	0

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TABLE 1 (continued). *Chlorinated pesticide burdens in Florida birds of prey, 1969–76*

County	Date	Sex/ Age [1]	Tissue [2] and Sample Weight, g	Residues, ppm Wet Weight		
				p,p'-DDE	ΣDDT	Dieldrin
			PANDION HALIAETUS (OSPREY)			
Marion	—	JUV	A (0.3864)	0.13	0 26	0
			U (2.4581)	1.55	1 79	0
Monroe	Apr. 73	JUV M	A (2 8327)	0.55	1 29	0
			U (4 1778)	0 41	0.62	0
Pinellas	Sept. 74	UNK	A (0.3840)	0	0	0
			U (1.1643)	0 09	0.09	0
Pinellas	Oct 74	M	A (0 7767)	0 33	0.33	0
			U (0 6309)	0 32	0.32	0
Alachua	Apr 75	F	U (0 4922)	13.21	15.85	0.91
Pinellas	May 76	F	A (0 4861)	1 65	1 87	0
			U (3 7545)	0.53	0 71	0
Pinellas	May 76	M	A (0.4264)	1.52	2.46	0
			U	1 39	1.39	0
			CARACARA CHERIWAY (CARACARA)			
Glades	July 75	AD F	A (0.1416)	2.47	2.47	0
			U (0.5854)	1.25	1.25	0
Highlands	July 75	IM F	A (0.0805)	1.24	1.24	0
			U (0.6235)	0.48	0.48	0
Highlands	Apr 76	IM F	A (0.1229)	3 25	3 25	0
			U (0.5734)	2.44	2.44	0
			FALCO SPARVERIUS (AMERICAN KESTREL)			
Indian Riv	Mar 73	M	A (0 2810)	14 59	16.37	0.36
			U (0 0954)	3.15	3 15	0
Indian Riv	Mar 73	F	A (1 1978)	1 77	2 09	0
Indian Riv	Mar 73	F	A	20 57	22.71	0
			U	4 63	4 63	0
Broward	Mar 73	F	A (0 1588)	0.79	0 79	0
			U (0.0788)	0 63	0.63	0
Indian Riv	Mar 73	F	A (0.1399)	2 14	2 14	0
			U (0 0602)	1.66	1.66	0
Indian Riv.	Mar 73	F	A (0 0551)	9.07	9 07	0
			U (0 0833)	1 80	1 80	0
Indian Riv	Mar 73	M	U (0 0653)	0	0	0
Indian Riv	Jan 74	M	A (0.0980)	0	0	0
			U (0.0258)	1 94	1 94	0
Indian Riv.	Oct. 74	M	A (0.0262)	0	0	0
			U (0.0302)	0	0	0
Leon	Jan 75	M	A	7.61	8.03	0
			U	1.44	2.20	0
Pinellas [3]	Nov. 75	M	U (0.0486)	4.12	4 12	3 09
			B (1.0756)	0.42	0 42	0.70
Pinellas [3]	Nov. 75	M	U (0.0459)	7.63	18.53	4 36

(Continued next page)

TABLE 1 (continued). *Chlorinated pesticide burdens in Florida birds of prey, 1969–76*

County	Date	Sex/ Age [1]	Tissue [2] and Sample Weight, g	Residues, ppm Wet Weight		
				p,p'-DDE	ΣDDT	Dieldrin
FALCO SPARVERIUS (AMERICAN KESTREL)						
			B (1 1574)	0 30	0 74	0 82
Pinellas [3]	Nov 75	F	U (0.0486)	2 06	2.06	1.03
			B (1 2114)	1 03	1 03	0.37
Pinellas [3]	Feb. 76	M	A (0 0790)	0	0	0
			U (0.0505)	0	0	0
TYTO ALBA (BARN OWL)						
Indian Riv	Mar 69	M	A (0 9538)	8 28	9 27	1.68
			U (2.1913)	1 31	1.31	0
Indian Riv	May 76	M	U (0 3550)	0	0	0
OTUS ASIO (SCREECH OWL)						
Alachua	Oct. 71	F	A (1 1317)	6.19	6 19	0
			U (0 1201)	1.17	1 17	0
Levy	Sept 73	UNK	A (0.3585)	0.26	0 26	0
			U (0 1454)	0	0	0
Alachua	Dec 75	UNK	A (0 6573)	0.30	0 30	0
			U (0 1435)	3 48	3 48	0
Indian Riv	May 75	UNK	U (0 0378)	10 58	10 58	0
Pinellas [3]	Jan. 76	M	U (0 0502)	49 80	49 80	0
Pinellas [3]	May 76	M	U (0 0748)	1 34	1.34	0
BUBO VIRGINIANUS (GREAT HORNED OWL)						
Leon	Mar 73	UNK	A (3 0278)	2 06	3 59	0.08
			U (0 6823)	8 24	8 68	0
Alachua	Nov 75	F	A (0 6460)	5.42	9 21	3 02
			U (0 7466)	3 28	4 28	0.47
Dixie	Dec 75	UNK	U (0 4400)	17.05	17.05	0
Marion	May 76	MF	A (0 2821)	9 75	12 05	6 20
			U (0.5537)	0 81	0 81	0
STRIX VARIA (BARRED OWL)						
Dixie	Apr 73	UNK	A (1 5201)	5.76	6.90	0 21
			U (0 7385)	5.08	5 69	0 14
Alachua	May 74	F	U (0.5460)	74 18	76 93	0
Alachua	Dec 75	F	A (0.2286)	1 09	1 09	0
			U (0.5321)	0.37	0.37	0 40
Leon	Jan 75	M	A (0 1183)	7 61	8 03	0
			U (0 6588)	1 44	2.20	0
Pasco	Mar 76	AD	A (0.1830)	1 09	1 09	0
			U (0.5843)	0 73	0 73	0

[1] M = adult male, F = adult female, AD = adult of undetermined sex, UNK = bird of unknown sex or age, JUV = juvenile, IM = immature. Sex of juvenile and immature birds was not always recorded.
[2] Tissue abbreviations: A = adipose tissue, U = uropygial gland, B = brain.
[3] Birds that reportedly died in captivity and exhibited convulsions.

always perfectly uniform because birds were obtained in different ways by different persons, and it was frequently inconvenient or impossible to take samples of brain, fat, and the uropygial gland from every bird. Furthermore, due to its relatively superficial position, the uropygial gland was sometimes damaged, and quite often a specimen was so lean that no fat could be found for pesticide analysis.

Even so, a number of important features emerge from the data in Table 1. All taxa (family, genus, and species) had some birds containing *p,p'*-DDE or other DDT metabolite. Dieldrin, on the other hand, was not present in all taxa. Of the 71 birds, 68 (96 percent) contained DDE but only 34 (48 percent) contained dieldrin in at least one of the three tissues. In the three tissues analyzed 93 percent of the fat samples contained DDE, 100 percent of the brains contained DDE, and 89 percent of the uropygial glands contained DDE. These values indicate a nearly universal occurrence of DDE in the birds studied and in the three tissues sampled.

In the 45 birds of prey in which both adipose tissue and uropygial gland were analyzed and in which one or both samples contained DDE, 40 (89 percent) had DDE in both tissues, 3 (7 percent) had DDE in adipose tissue only, and only 2 (4 percent) had DDE in the uropygial gland alone. However, Figure 1 shows a poor correlation ($r = 0.3398$) of ΣDDT, in ppm wet weight, between these two tissue types. Of 46 birds, 40 had higher concentrations of ΣDDT in the adipose tissue than in the uropygial gland. For the species analyzed, the mean ratio of ΣDDT in adipose tissue to uropygial gland was 2.6:1, a higher ratio than the 2.2:1 reported by Johnston (*11*) for a sample of other feral species such as loons, cormorants, herons, and gulls.

Figures 2 and 3 show ΣDDT found, respectively, in the adipose tissue and uropygial gland through the sampling period. In both samples, median values were calculated for all the species in a given year; these values are indicated by

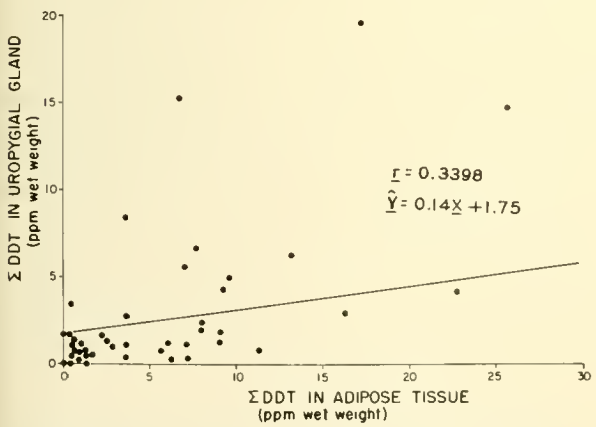

FIGURE 1. *Relationship of ΣDDT in adipose tissue and uropygial gland in Florida birds of prey, 1969–76*

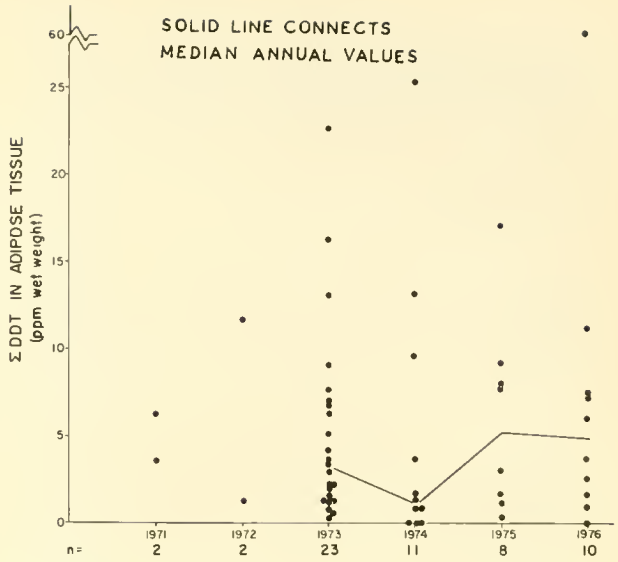

FIGURE 2. *ΣDDT in adipose tissue of Florida birds of prey, 1971–76*

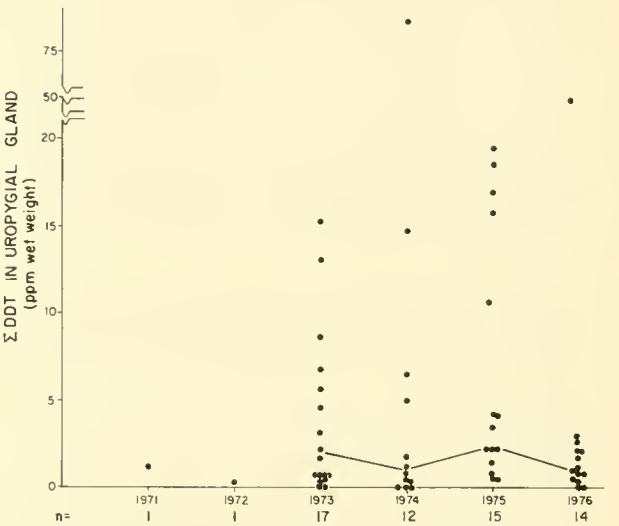

FIGURE 3. *ΣDDT in uropygial glands of Florida birds of prey, 1971–76*

a solid line. The lines might not indicate realistic trends because the numbers of a given species available for analysis varied from year to year, as indicated by raw data in Table 1.

Discussion

Of the 14 species examined here, there is no assurance that any bird was a permanent resident where collected except for the caracaras and juvenile osprey. Any or all the birds might have been transient or migratory at some time during their lives, so pesticide burdens determined for these birds

were not necessarily accumulated in Florida, but could have come from prey consumed on a wintering area south of the state, from a breeding area in the north, or in intervening areas during migratory flights. It is likely, however, that most of the four owl species were Florida residents because they tend to migrate less than the other birds of prey studied here.

Subspecific information was determined only for the American kestrel. These small falcons were all representative of the northern subspecies (*Falco s. sparverius*) which migrates into Florida from the northern United States and winters in large numbers in the state. The author was unable to obtain samples of the local resident Florida subspecies, *F. s. paulus*, for analysis. Data on the vultures, *Cathartes* and *Coragyps*, are presented in Table 1 and appear to be the first residue findings published on these species.

Because vultures are terminal members of food chains, they might be expected to have exceptionally high DDT levels, but this does not appear to be true of *Cathartes*. Only 1 of 10 turkey vulture fat samples exceeded 10 ppm ΣDDT; the mean was 4.76 ppm. However, the mean for the fat of 5 *Coragyps* was 10.64 ppm ΣDDT; one bird had 25.43 ppm. Both vultures scavenge road-killed animals such as the nine-banded armadillo, Virginia opossum, dogs, and various smaller mammals, birds, and reptiles in Florida, most of which are nonmigratory. Why *Coragyps* should have a higher mean burden of ΣDDT than do *Cathartes* is unclear.

The *Accipiter* hawks, also called bird hawks, have pesticide burdens as high as or higher than most other species studied (Table 1). The small sample size precludes generalizations, but it is noteworthy that in 1972 Henny reported that the Cooper's hawk "is in serious jeopardy in the northeastern U.S." (*6*).

Some species listed in Table 1 are largely insectivorous (*5*), namely, *Elanoides forficatus*, *Falco sparverius*, and *Otus asio*. At least three of the 14 specimens of *Falco* had ΣDDT burdens exceeding 10 ppm; one contained 18.53 ppm in the uropygial gland, and the ΣDDT burden in adipose tissue probably exceeded 50 ppm. Two of six *Otus* specimens had exceptionally high levels in their uropygial glands: 10.58 ppm and 49.80 ppm. For this species, 10 ppm DDE dry weight in the diet produced thin eggshells (*13*). Although dietary levels of DDT may not be directly related to levels in the adipose tissue or uropygial gland, it is significant that 5 ppm DDT wet weight in the diet of *Falco sparverius* resulted in the classical eggshell-thinning syndrome (*17*). However, carcass (*17*) and breast muscle (*8*) analyses of dead or dying American kestrels in the northern United States had generally higher DDT burdens than those found in the present study (*1*). For this species, it is significant that three individuals contained no DDT or metabolite (Table 1).

Henny et al. presented data on eggshell thicknesses and populations of red-shouldered hawks (*Buteo lineatus*) from a refuge in Maryland (*7*). The authors thought it "doubtful that the relatively low pesticide levels in the eggs had a detrimental effect on the reproductive performance of the population." Except for a single bird containing 61.76 ppm ΣDDT and 38.24 ppm dieldrin, organochlorine residues in this species were generally low (Table 1).

The osprey (*Pandion haliaetus*) was studied intensively in the 1960s because its population had declined precipitously in some areas (*6*). As with other species discussed in this paper, presticide levels in eggs and nestlings have been published but data for adults are scarce. Wiemeyer et al. reported brain and carcass analyses of dead birds in Connecticut and Virginia (*18*). DDE residues in carcasses averaged 23 ppm wet weight, generally, exceeding the levels in adipose tissue and uropygial gland in Florida birds reported here. Because different tissues were analyzed, it is difficult to compare previously published data on red-tailed hawks and great horned owls with those reported here. For three nestling red-tailed hawks, Seidensticker found an average of 21.50 ppm ΣDDT wet weight, in breast muscle (*15*). Seidensticker and Reynolds reported 1.40 ppm wet weight ΣDDT in nestling red-tail hawk muscle and 9.29 ppm ΣDDT in the muscle of a great horned owl (*16*).

Two generalities emerge from the data in Table 1 and Figures 2 and 3. There is no firm evidence for this sample of birds of prey from Florida that DDE and dieldrin burdens diminished in 1971–76. In both the adipose tissue and uropygial glands, the data indicate an approximate average of 5 ppm over the 4–6 year span. Small migratory songbirds, on the other hand, showed a dramatic decrease of DDE in adipose tissue from 1964 to 1973 (*9*, *10*). That decrease was correlated with the decreased use of DDT in the United States during the same time. Presumably, the ban on DDT use in the United States imposed by the U.S. Environmental Protection Agency (EPA) December 31, 1972, should have reduced the amount of DDT in natural ecosystems. The birds of prey studied here are significant, especially those analyzed after 1972, because a large proportion did contain DDT or a metabolite. How would a hawk, owl, or vulture obtain significant quantities of DDT in 1976? It is plausible that a long-lived bird could have accumulated small pesticide quantities for years and simply stored them in adipose tissue. Unless these deposits were totally depleted for energy resources, the pesticides might not have been mobilized into the bird's bloodstream or eliminated except in very small quantities. The data on uropygial glands presented in Table 1 indicate that birds of prey eliminate smaller quantities of pesticides through this gland than do other types of birds (*3*, *4*).

A second possible explanation for the DDT burden in birds of prey after the EPA ban in 1972 is that at least eight species analyzed here might have migrated to Florida from

the West Indies or Central America where they could have obtained DDT-contaminated foods. This is probably similar to the situation of the migratory American kestrels discussed by Lincer and Sherburne *(12)*. They suggested that this species obtained pesticide-laden foods chiefly from the wintering grounds rather than from nesting sites in the northern United States. They state: "The disastrous role played by the far-removed, but inordinately contaminated, winter prey once again dramatically points out the global nature of the biocide problem." Still, the presence of DDT in tissues of the caracara, which is a resident of southcentral Florida, is an enigma.

Since 1973, the ΣDDT burdens in adipose tissue of two species examined here, osprey and American kestrel, were very low (0–2 ppm).

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Shell Thinning and Pesticide Residues in Texas Aquatic Bird Eggs, 1970

Kirke A. King,[1] Edward L. Flickinger,[1] and Henry H Hildebrand [2]

ABSTRACT

Significant decreases in eggshell thickness were found in 15 of 22 species of aquatic birds in Texas in 1970. Shell thickness reductions of 9 to 15 percent were found in white pelicans (Pelecanus erythrorhynchos), *brown pelicans* (P. occidentalis), *and great blue herons* (Ardea herodias). *DDT family compounds were found in all eggs, and mean residues ranged from 0.4 ppm in white ibis* (Eudocimus albus) *to 23.2 ppm in great egrets* (Casmerodius albus). *ΣDDT residues were negatively correlated with shell thickness in five species; PCBs were negatively correlated in two. Residues in marine birds were generally lower and more uniform than levels in birds feeding in fresh and brackish water. DDT and dieldrin residues were higher in eggs from colonies near agricultural areas where these insecticides were heavily used; higher PCB residues were consistently associated with urban and industrial areas. Populations of five species have declined and deserve continued study: brown pelican, reddish egret* (Dichromanassa rufescens), *white-faced ibis* (Plegadis chihi), *laughing gull* (Larus atricilla), *and Forster's tern* (Sterna forsteri). *Population trends of four other species were undetermined and should be followed closely in future years.*

Introduction

Eggshell thinning has been noted in a number of declining populations of fish-eating birds in the United States (*2, 6, 19, 20*). Laboratory investigations show that the DDT family compounds, ΣDDT, primarily DDE, induce shell thinning in some wild birds and their eggs (*15, 16, 28*). The recent decline in brown pelicans, reddish egrets, and an apparent decline in white-faced ibis on the Texas Gulf Coast prompted the present study to determine the extent of eggshell thinning and the impact of pesticide contamination on these and other fish-eating birds breeding in Texas. The authors present information on shell thickness changes and chemical residues in eggs of 22 species of aquatic birds. Sources of contamination and species threatened by exposure to pesticides are identified.

Study Area and Methods

From March through July 1970, 1,043 eggs were collected in 30 locations on the Texas Coast. One egg was taken randomly from each nest sampled in a pattern distributed as evenly as possible throughout each colony. Whole eggs were weighed and measured, wrapped in aluminum foil, and frozen. Contents were later removed, stored in jars prerinsed with acetone, and immediately refrozen until analysis. Five to 20 eggs of each species were analyzed at the Denver Wildlife Research Center Laboratory, Denver, Colorado. Chemical analyses were completed in 1970 and 1971. Except for brown pelican eggs which were addled, only fresh eggs were analyzed for pesticide residues.

The authors biased selection of eggs for chemical analysis by singling out thin-shelled eggs from each species. Random samples of white-faced ibis, black-crowned night heron, and Forster's tern eggs were also analyzed. Mercury levels were determined in 10 white-faced ibis and 10 great blue heron eggs.

Organochlorine residues and polychlorinated biphenyls (PCBs) were determined by using methods described by Peterson et al. (*25*). The methods measure ΣDDT, aldrin, dieldrin, endrin, heptachlor epoxide, and lindane at 0.1 ppm wet weight, and chlordane and toxaphene at 0.5 ppm wet weight. PCBs were not separated from pesticides before measurement. When found, PCBs were identified on two separate columns and by visual comparison of chromatograms with standard Aroclors. The PCB residues were quantitated by averaging peak responses and comparing them with Aroclor 1254 standards. Detection limit of the procedure for PCBs was 0.5 ppm. Mercury residues

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were determined by using methods described by Okuno et al. (*24*). No corrections were made for possible moisture loss.

The authors compared shell thicknesses of eggs collected in 1970 with those of museum eggs collected before widespread use of DDT. Data on white and brown pelican eggshells collected before 1947 are from Anderson and Hickey (*2*); data on white-faced ibis eggshells were supplied by A. J. Smith and J. O. Keith (personal communication, 1971). All other measurements of eggshells collected before 1943 were obtained from the Western Foundation of Vertebrate Zoology, Los Angeles, California, and Welder Wildlife Foundation, Sinton, Texas. Anderson and Hickey (*1*), showed that eggshell thickness for a particular species varies significantly over broad geographic areas particularly with latitude. Whenever possible, museum eggs from the Texas Coast and other southern latitudes were selected for shell thickness measurement.

Results and Discussion

EGGSHELL CHANGES

Fifteen of 22 species sampled showed a signficant negative change in eggshell thicknesses from their museum mean (Table 1). The species with the greatest average thinning were the white pelican (15 percent), great blue heron (13 percent), and brown pelican (11 percent). No collapsed eggs were found in the nests of these species. Although the average thinning of white-faced ibis eggshells was only 4 percent, numerous collapsed, dented, and cracked eggs were found in and around ibis nests. In 1971, continued sampling showed that about 3.5 percent of the white-faced ibis eggs in marked nests had dented or cracked shells; the incidence of cracked eggs of other species was less than 1 percent. Numerous field studies have shown that eggshell thinning of less than 10 percent seldom incurs egg breakage (*3, 6, 10*). Egg loss becomes evident with thinning of 10–15 percent (*19*), and serious breakage, usually accompanied by population decline, occurs when eggshell thinning exceeds 15 percent (*2, 20*). The degree of shell thinning among the white and brown pelicans and great blue heron approaches that found in other populations in which shell thinning adversely affected reproduction.

Average shell thinning was greatest in the Lower Laguna Madre–Green Island region (Figure 1). Shell thickness did not vary significantly among heronries sampled elsewhere on the Texas Coast.

ORGANOCHLORINE RESIDUES

Residues of ΣDDT, primarily DDE, were found in all samples. The highest averages were in eggs of the great egret, 23.2 ppm; Caspian tern, 15.1 ppm; and laughing gull, 10.4 ppm (Table 2). ΣDDT in the eggs of the remaining species ranged from 0.4 ppm in white ibis to 9.7 ppm in black skimmer. Consistently higher levels of ΣDDT and the greatest amount of shell thinning were found in eggs from the lower coast near the intensively cultivated Rio Grande Valley. ΣDDT compounds were found in eggs of species that feed in all habitats: freshwater, brackish, and marine.

Dieldrin residues, found in 14 species, were highest in the snowy egret, white-faced ibis, and great egret (Table 2), species that feed primarily in freshwater and brackish marshes. Little dieldrin was found in eggs of ocean-feeding birds such as brown pelican, royal tern, and Sandwich tern. Greatest dieldrin residues were in eggs from colonies adjacent to the Texas rice belt where aldrin had often been used to treat rice seed.

TABLE 1. *Eggshell changes of several Texas fish-eating birds, pre-1943 and in 1970*

Species	Period[1,2]	No	Shell Thickness, mm: Mean±SE	Change, %
White Pelican	pre-1947	102	0.676±0.005	
Pelecanus erythrorhynchos	1970	28	0.577±0.008[3]	−15
Great Blue Heron	pre-1943	32	0.413±0.005	
Ardea herodias	1970	74	0.359±0.003[3]	−13
Brown Pelican	pre-1947	43	0.557±0.006	
P. occidentalis	1970	14	0.497±0.013[3]	−11
Snowy Egret	pre-1943	38	0.241±0.003	
Egretta thula	1970	79	0.220±0.002[3]	−9
Royal Tern	pre-1943	18	0.358±0.004	
Thalasseus maximus	1970	12	0.330±0.007[3]	−8
Olivaceous Cormorant	pre-1943	30	0.347±0.005	
Phalacrocorax olivaceus	1970	24	0.323±0.006[4]	−7
Louisiana Heron	pre-1943	31	0.238±0.003	
Hydranassa tricolor	1970	58	0.225±0.002[3]	−5
Little Blue Heron	pre-1943	31	0.243±0.002	
Florida caerulea	1970	32	0.232±0.003[4]	−5
Great Egret	pre-1943	30	0.295±0.004	
Casmerodius albus	1970	113	0.282±0.002[3]	−4
White Ibis	pre-1943	38	0.363±0.004	
Eudocimus albus	1970	48	0.347±0.003[3]	−4
White-faced Ibis	pre-1943	18	0.312±0.006	
Plegadis chihi	1970	86	0.301±0.002[4]	−4
Black-crowned Night Heron	pre-1943	79	0.278±0.003	
Nycticorax nycticorax	1970	74	0.266±0.003[4]	−4
Black Skimmer	pre-1943	28	0.249±0.004	
Rynchops nigra	1970	48	0.240±0.002[5]	−4
Gull-billed Tern	pre-1943	31	0.239±0.002	
Gelochelidon nilotica	1970	58	0.231±0.002[4]	−3
Laughing Gull	pre-1943	27	0.270±0.003	
Larus atricilla	1970	65	0.263±0.002[5]	−3
Sandwich Tern	pre-1943	25	0.286±0.004	
Sterna sandvicensis	1970	19	0.277±0.005	−3
Anhinga	pre-1943	31	0.328±0.004	
Anhinga anhinga	1970	8	0.318±0.007	−3
Roseate Spoonbill	pre-1943	32	0.426±0.008	
Ajaia ajaja	1970	53	0.415±0.004	−3
Reddish Egret	pre-1943	47	0.270±0.002	
Dichromanassa rufescens	1970	54	0.267±0.003	−1
Least Tern	pre-1943	22	0.156±0.003	
S. albifrons	1970	15	0.154±0.004	−1
Forster's Tern	pre-1943	26	0.219±0.003	
S. forsteri	1970	41	0.218±0.003	0
Caspian Tern	pre-1943	15	0.336±0.005	
S. caspia	1970	32	0.339±0.003	+1

[1] Pre-1947 white and brown pelican data are from Anderson and Hickey (*2*).
[2] All pre-1943 eggs are from the Texas Coast except white pelican, western United States; black-crowned night heron, South Carolina, Florida, and California; snowy egret, little blue heron, great egret, and anhinga, Gulf Coast, Florida, and South Carolina
[3] $p<0.001$ (Student's t-test)
[4] $p<0.01$
[5] $p<0.05$

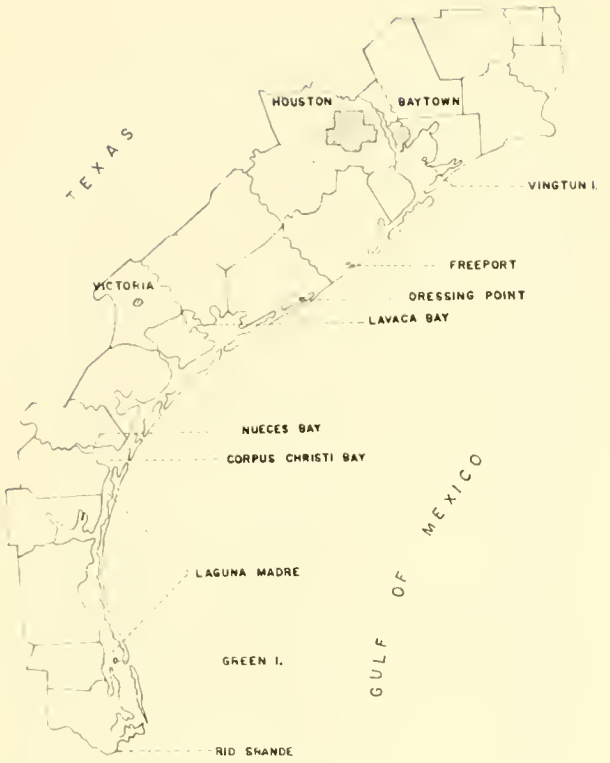

FIGURE 1. *Location of colonies of wading birds sampled for eggshell thinning, Texas Gulf Coast—1970*

PCB residues were found in all but two species; highest levels occurred in the olivaceous cormorant, Caspian, Forster's, and royal terns (Table 2). Except for the royal tern, these birds feed most frequently in freshwater and estuarine areas. The colonies associated with highest PCB contamination are Vingtun Island near the sprawling urban-industrial complex of Houston-Baytown, Texas, and Dressing Point, south of Freeport, Texas; both areas have numerous oil refineries and petrochemical plants.

Insecticide and PCB residues in marine birds were generally lower and more uniform than levels in birds feeding in freshwater and brackish habitats. ΣDDT and dieldrin residues were higher in eggs from colonies near agricultural areas where insecticides were heavily used. Higher PCBs were consistently associated with urban and industrial areas.

RESIDUE CORRELATIONS WITH EGGSHELL THICKNESS

ΣDDT or DDE was negatively correlated with shell thickness for the great blue heron ($r = -0.66$; $p < 0.01$), white-faced ibis ($r = -0.64$; $p < 0.01$), gull-billed tern ($r = -0.936$; $p < 0.02$), reddish egret ($r = -0.74$; $p < 0.05$), and brown pelican ($r = -0.61$; $p < 0.05$). PCB residues were negatively correlated only for the reddish egret ($r = -0.72$; $p < 0.05$) and the brown pelican ($r = -0.53$; $p < 0.1$); no correlation was found between any of the remaining insecticide residues and eggshell thickness.

Other insecticides and industrial pollutants may affect shell thickness because many pollutants are capable of altering food chain composition, ecosystem energy flow, and ultimately the bioenergetics of individual populations of birds. The many environmental factors and physiological processes that result in eggshell thinning are not well understood. However, the chemical pollutant most frequently identified with shell thinning is DDE. The authors' data support the findings of others who have reported that DDE is the principal agent correlated with eggshell thinning in wild birds (*3, 7, 16, 26*).

SOURCES OF CONTAMINATION

This study indicates that DDE and dieldrin levels detected in egg samples are related to food habits of adult birds. Flickinger and King (*12*) found wet-weight residues of ΣDDT from 0.2 to 1.6 ppm and dieldrin from 0.4 to 2.8 ppm in three species of freshwater fish that are commonly consumed by fish-eating birds. Maximum ΣDDT residues of 9.3 ppm were found in menhaden (*Brevoortia* sp.) and 6.4 ppm in anchovies (*Anchoa* sp.) collected from 1967 through 1969 from rivers, bays, and estuaries in Texas (*9*). Potential effects of these residue levels in food items are evident from results of other studies showing that 3–4 ppm wet-weight DDE in the diet will cause eggshell thinning in certain species of birds (*16, 22, 23, 30*).

DDT was found in the eggs of six species: great egret, white-faced ibis, Sandwich tern, least tern, gull-billed tern, and roseate spoonbill. Low DDT residues, less than 0.8 ppm, were found in all roseate spoonbill eggs. Frequency of contamination in the other five species ranged from 4 percent in the white-faced ibis to 40 percent (two eggs) in the Sandwich tern. The highest DDT residue found was 1.3 ppm in a Sandwich tern egg. Local contamination through the food chain is possible since DDT residues have been found in a pooled sample of 76 sailfin molly (*Poecilia latipinna*) and in crawfish (*Procambarus clarki*), two common foods of aquatic birds in Texas (*12*). Birds migrating to Mexico have been contaminated also since DDT still was widely used there in 1970. DDT residues occurred in all five species that regularly migrate to Mexico: roseate spoonbill, white-faced ibis, snowy egret, Sandwich tern, and least tern.

SIGNIFICANCE OF RESIDUES

DDE—DDE-induced shell thinning has been summarized for numerous birds (*2, 28*). Residues in eggs reported in the present study are comparable to levels found in wild populations that have experienced reproductive failures. Some laboratory studies indicate that harmful effects other than shell thinning are possible. Longcore (*22*) found reduced survival of ducklings (*Anas rubripes*) hatched from

TABLE 2. *Insecticide and PCB residues in eggs of Texas wading birds, 1970*

Species	No	Mean Residues±SE Wet Weight			Lipid, %
		ΣDDT [1]	Dieldrin	PCBs	
Great Egret	10	23.24±3.61	0.63±0.14 (*10*)	ND	5.6
Caspian Tern	10	15.13±2.25	ND	16.50±4.51 (*10*)	8.5
Laughing Gull	10	10.35±3.90	0.52±0.34 (*5*)	3.00±2.13 (*2*)	10.6
Black Skimmer	5	9.68±3.02	ND	5.40±1.89 (*5*)	11.0
Least Tern	5	6.94±3.52	ND	2.60±0.81 (*4*)	17.2
Louisiana Heron	5	6.50±2.17	0.16±0.12 (*2*)	2.40±0.81 (*4*)	8.5
Olivaceous Cormorant	5	6.22±2.08	0.30 (*1*)	32.00±5.83 (*5*)	4.7
Great Blue Heron	20	5.55±1.05	0.14±0.09 (*3*)	5.54±1.02 (*20*)	5.4
White-faced Ibis	16	5.33±2.92	0.81±0.22 (*12*)	3.00±2.13 (*8*)	6.2
Gull-billed Tern	10	4.89±2.73	0.18±0.15 (*4*)	1.25±0.33 (6)	9.3
Royal Tern	5	4.28±0.88	ND	11.60±2.84 (*5*)	12.7
Roseate Spoonbill	10	3.85±0.88	TR (*2*)	2.10±0.28 (*10*)	5.4
Snowy Egret	10	3.26±1.30	1.06±0.67 (*5*)	2.03±1.24 (*7*)	6.2
Brown Pelican	11	3.23±0.20	ND	9.73±1.38 (*10*)	4.8
Reddish Egret	10	2.52±0.60	ND	1.50±0.29 (*10*)	5.9
Black-crowned Night Heron	10	1.76±0.58	TR (*4*)	ND	5.4
Forster's Tern	10	1.74±0.20	0.47 (*1*)	12.50±4.76 (*7*)	9.1
White Pelican	5	1.38±0.30	ND	0.98±0.97 (*5*)	4.7
Little Blue Heron	5	1.20±0.75	0.12±0.05 (*4*)	1.40±0.37 (*5*)	6.5
Sandwich Tern	5	1.12±0.36	0.72 (*1*)	1.40±0.24 (*5*)	15.2
White Ibis	5	0.41±0.12	TR (*5*)	ND	11.0

NOTE: ND=not detected. TR=trace
Numbers in parentheses represent number of eggs with residues.
[1] ΣDDT residues found in all eggs sampled

eggs of hens which had consumed food treated with 10 ppm and 30 ppm DDE. Haegele and Hudson (*15*) also reported increased mortality of young and reduced clutch size in ring doves (*Streptopelia risoria*) fed 40 ppm DDE. DDE fed at 10 ppm and 40 ppm to mallards (*Anas platyrhynchos*) reduced hatching of eggs, although survival of hatchlings to 14 days was unaffected (*16*).

Dieldrin—Dieldrin levels found in the present study are lower than those reported in several studies investigating reproductive success and survival of young birds. Fowler et al. (*13*) reported normal hatching success of purple gallinule (*Porphyrula martinica*) and common gallinule (*Gallinula chloropus*) eggs containing average dieldrin residues of 3.8–17.5 ppm. Pheasants (*Phasianus colchicus*), fed varying amounts of dieldrin, showed no effects on fertility, hatching, or survival associated with yolk residues of up to 52 ppm (*4*). Dieldrin residues in whole eggs would normally average about 26 ppm. Chickens fed up to 5 ppm dieldrin showed no effects on clutch size, hatching, or survival of young associated with egg residues of 4–5 ppm (*14*). In contrast, Baxter et al. (*5*) found second-generation effects: fertility and hatching were significantly lower in eggs of hens that received dieldrin through the egg. Dieldrin above 1 ppm in the eggs of golden eagles (*Aquila chrysaetos*) may cause reproductive problems (*28*), and dieldrin residues of 0.54 ppm are lethal to brown pelican embryos (*7*). In view of the great variation in toxicity of dieldrin to different wildlife species, egg residues greater than 1 ppm must be viewed as hazardous.

PCBs—Laboratory experiments indicate that PCB levels found in the present study do not reflect acute exposure of fish-eating birds, but results of reproductive studies are not so conclusive (*17, 26*). One important consideration is the wide range in species sensitivity to PCBs; Heath et al. (*17*) found a fourfold difference in sensitivity between two gallinaceous species. The complex problems associated with the wide range of sensitivity to PCBs and the varying toxicities of different Aroclors were reviewed by Stendell (*27*). These differences emphasize the difficulties in drawing conclusions about the meaning of residues in eggs of fish-eating birds. But at least five species in this study have sufficiently high egg levels of PCBs to warrant additional research: olivaceous cormorant, Caspian tern, Forster's tern, royal tern, and brown pelican.

Mercury—A pooled sample of 30 white-faced ibis eggs contained 0.18 ppm wet-weight mercury, and 10 great blue heron eggs averaged 0.30 ppm. Fimreite (*11*) reported significantly lowered hatching success in pheasant eggs containing 0.5–1.5 ppm mercury, and Borg et al. (*8*) found similar effects at levels of 1.3–2.0 ppm. However, Heinz (*18*) found no significant effects on mallard reproduction associated with egg residue levels of 1.0 ppm. Herring gull (*Larus argentatus*) chicks hatched from each of 24 clutches that contained mercury between 0.5 ppm and 2.0 ppm (*29*). Thus it seems unlikely that mercury residues in white-faced ibis and great blue heron eggs were high enough to affect reproduction adversely.

White-faced ibis eggs were expected to contain high mercury residues because ibis feed in flooded rice fields where mercury-based fungicides were used on seed, but levels were low compared with those found in other studies (*8, 11, 18, 29*). Great blue heron feed in various freshwater and brackish habitats and had slightly but not significantly greater mercury residues than had ibis. This indicates that mercury is found throughout the coastal environment, at least in feeding areas of both species in the Texas rice belt.

THREATENED SPECIES

One objective of the present study was to identify populations possibly threatened by pesticide contamination. On the basis of recent population trends, residue levels, and shell thinning, the authors believe that the brown pelican, white-faced ibis, reddish egret, laughing gull, and Forster's tern warrant immediate attention. Populations of white pelican, olivaceous cormorant, great blue heron, and great egret showed weak or undetermined population trends and should be watched closely in future years. Results of a Texas brown pelican study were recently published (*21*) and ibis data are being prepared for publication.

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Organochlorine Insecticide and Polychlorinated Biphenyl Residues in Woodcock Wings, 1971–72

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ABSTRACT

Pesticide residues in wings of adult woodcock (Philohela minor) *were used to monitor regional differences in a 1970–71 survey of DDT, DDE, TDE, dieldrin, mirex, and PCBs in Maine, New Hampshire, New York, New Jersey, Pennsylvania, North Carolina, South Carolina, Georgia, Lousiana, Michigan, and Wisconsin. In 1971–72, wings were sampled again to compare levels of organochlorine insecticide residues with those of the previous survey and to delineate differences in residue values between adult and immature woodcock. Three additional states, Massachusetts, Minnesota, and Vermont, and one additional organochlorine insecticide, heptachlor epoxide, were included in the second survey.*

Residue levels in the 1971–72 wings showed the same pattern as that observed in 1970–71: organochlorine insecticide residues were highest in wings collected in the southern states and in New Jersey; residues were lowest in samplings taken in the northern and midwestern states. Residues of DDT, TDE, and dieldrin in the 1971–72 wings were slightly lower than those found in 1970–71. DDE, PCB, and mirex residues were significantly lower ($P < 0.05$, $P < 0.01$, and $P < 0.01$, respectively) in 1971–72. Wings of immature woodcock in Louisiana had significantly lower ($P < 0.05$) mirex residues than did adult wings.

Introduction

The woodcock is well suited for monitoring environmental pollutants because it is a migratory upland game bird distributed throughout the eastern United States from the Mississippi River to the Atlantic Ocean and from Michigan to Florida. Personnel from the Fish and Wildlife Service, U.S. Department of the Interior, monitor reproductive success of woodcock by annually inspecting wings submitted by cooperating hunters. Thus wings are in ample quantity for other studies. The same wings can be used to assess quantities of pollutants which the birds have acquired, largely from their food. The woodcock occurs near or at the top of a terrestrial food chain and subsists on animal material, primarily earthworms (*7, 10*). Earthworms concentrate an array of persistent environmental pollutants in their tissues and are important in the diets of a number of avian species (*2, 3, 4, 5, 6*).

Woodcock wings were first monitored for environmental pollutants in 1970–71. Regional differences were clearly demonstrated and baseline measurements were obtained for later comparisons (*8*). An expanded sampling of wings was undertaken in 1971–72 to compare residues with those found in 1970–71, and to determine whether residues in the wings of adult and immature woodcock differed. This paper reports the findings of the 1971–72 survey.

Methods

Wings were collected in 15 states: Connecticut, Georgia, Louisiana, Maine, Massachusetts, Michigan, Minnesota, New Hampshire, New Jersey, New York, North Carolina, Pennsylvania, South Carolina, Vermont, and Wisconsin. These states provided a suitable geographic distribution and offered the best chance for collecting adequate numbers of wings. Because wings from North Carolina, South Carolina, and Georgia were too few to provide a sample from each state, the wings from these states were combined into one tri-state area sample. Wings from adult and immature woodcock from each state and from the tri-state area were sorted into groups of 25. Five of these groups from each state and five from the tri-state area were randomly selected for analysis.

Wings were plucked and the distal joint was removed. The part remaining was ground in a hand grinder and

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homogenized with the group of 25 which made up the complete sample. A 20-g aliquot was taken for analysis.

Organochlorine pesticides and polychlorinated biphenyls (PCBs) were determined at WARF Institute, Inc., Madison, Wisconsin, by the following procedures:

The 20-g aliquot was dried at 40° C for 96–120 hours, and then ground with sodium sulfate and extracted for 8 hours on a Soxhlet extractor with 105 ml of ethyl ether and 250 ml of petroleum ether. The extract was concentrated on a steam bath and diluted to 50 ml with petroleum ether. A 10-ml aliquot of the extract was cleaned and separated into two fractions by elution through a Florisil column with mixtures of ethyl ether and petroleum ether (5+95 and 15+85). An aliquot of the final elution was passed through a standardized silicic acid column as described by Armour and Burke (*1*).

The pesticides and PCBs were determined by electron-capture gas chromatography under the following conditions:

Chromatograph:	Barber-Coleman Model 5360 Pesticide Analyzer
Column:	4-ft × 3-mm glass, packed with 5 percent DC-200 on 80–100-mesh Gas-Chrom Q
Temperatures:	injector 225° C column 205° C detector 245° C
Carrier gas:	purified nitrogen flowing at 80 ml/minute
Chromatograph:	Barber-Coleman Model 5000
Column:	4-ft × 4-mm glass, packed with 3 percent OV-17 on 100–120-mesh Gas-Chrom Q
Temperatures	injector 215° C column 200° C detector 250° C
Carrier gas.	purified nitrogen flowing at 80 ml/minute

The sensitivity level of this method was 0.05 ppm organochlorine pesticide and 0.10 ppm PCBs on a lipid basis. Recovery for organochlorine pesticides ranged between 80 and 95 percent, and PCB recoveries ranged between 75 and 85 percent. None of the residue data has been adjusted for rates of recovery.

Results and Discussion

Table 1 shows the ranges and the means as ppm lipid weight for DDT, DDE, TDE, PCBs, dieldrin, mirex, and heptachlor epoxide residues in adult woodcock wings from 12 states and the tri-state area arranged in approximate geographic order from south to north. DDT and its

TABLE 1. *Ranges and geometric means of organochlorine insecticide residues in adult woodcock wings from 15 eastern/midwestern states, 1971–72*

State	Residues, ppm Lipid Weight						
	DDT	DDE	TDE	PCB	Dieldrin	Mirex	Heptachlor Epoxide
Louisiana	1.88–5.45 2.74	5.74–13.09 9.20	0.52–1.40 0.91	1.65–4.10 2.21	1.27–5.56 1.90	4.70–8.49 6.20	0.52–1.13 0.70
Tri-state area	2.35–11.12 5.90	6.99–27.00 18.69	0.64–2.02 1.42	2.63–4.22 3.24	1.22–4.04 1.88	1.66–5.27 3.14	0.21–1.48 0.58
New Jersey	3.27–8.20 4.90	10.11–25.80 16.96	0.76–2.34 1.25	1.97–4.04 2.92	0.27–0.77 0.43	ND–2.12 0.58	ND —
Pennsylvania	0.29–1.37 0.60	2.11–4.71 3.59	ND–0.17 0.03	0.94–2.07 1.39	0.12–2.99 0.30	0.24–0.78 0.48	ND —
Connecticut	1.28–5.61 2.36	3.38–7.12 6.23	0.16–0.65 0.35	1.52–4.38 2.66	0.12–1.12 0.36	ND–0.38 0.50	ND —
New York	0.29–2.87 1.12	4.16–13.07 6.32	0.14–0.26 0.19	1.37–1.84 1.60	0.15–0.21 0.18	0.28–0.96 0.54	ND —
Massachusetts	0.51–5.41 2.16	8.28–22.65 15.63	0.11–0.73 0.33	4.03–9.58 5.84	0.05–0.91 0.15	ND–0.91 0.24	ND —
New Hampshire	0.68–8.47 1.92	5.96–11.56 8.47	ND–0.47 0.31	1.44–1.90 1.69	0.14–0.59 0.27	ND–0.92 0.45	ND —
Vermont	0.25–0.67 1.36	2.63–3.57 3.33	0.07–0.13 0.12	1.54–2.02 1.75	0.08–0.11 0.09	0.24–1.25 0.54	ND —
Maine	0.36–0.94 0.77	3.24–7.20 5.13	0.06–0.25 0.18	0.96–1.26 1.12	0.06–0.12 0.08	0.34–1.44 0.87	ND —
Michigan	0.24–0.68 0.50	2.28–6.96 3.53	0.06–0.32 0.18	1.02–2.21 1.39	0.07–0.11 0.09	0.59–5.01 1.34	ND —
Wisconsin	ND–0.18 0.10	2.60–4.23 3.15	ND —	0.46–1.22 0.77	0.09–0.82 0.18	ND–1.78 0.85	ND —
Minnesota	0.16–0.47 0.30	1.12–3.34 1.74	ND —	ND–0.48 0.08	0.05–0.06 0.05	ND–0.74 0.21	ND —

NOTE: Tri-state area=North Carolina, South Carolina, and Georgia. Wings from three states were combined because not enough were available from any one state
ND=not detected.

metabolites are distributed in a similar pattern: geometric means of DDT and its metabolites were highest in the tri-state area (DDT, 5.90 ppm; DDE, 18.69 ppm; TDE, 1.42 ppm) and second highest in New Jersey (DDT, 4.90 ppm; DDE, 16.96 ppm; TDE, 1.25 ppm). Differences in contaminant residues levels were determined by one-way analysis of variance with Duncan's multiple range test. Average TDE residues in woodcock wings from the tri-state area and New Jersey were significantly higher ($P<0.01$) for the tri-state area than for all other states except New Jersey, Massachusetts, and Louisiana. The average level of DDE was significantly lower ($P<0.01$) for Minnesota than for all other states.

The average PCB residue in woodcock wings (5.84 ppm) was significantly higher ($P<0.01$) for Massachusetts than for all other states; PCBs in wings were higher ($P<0.01$) for the tri-state area than for all other states except New Jersey, Connecticut, and Louisiana. The average PCB level was significantly lower ($P<0.01$) in Minnesota than in all other states.

Average dieldrin residues in wings from Louisiana and the tri-state area (1.90 ppm and 1.88 ppm, respectively) were significantly higher than those in all other states. Minnesota had the lowest average residues (0.05 ppm).

Heptachlor epoxide residues were found in adult wings in only two areas: Louisiana and the tri-state area. These two areas were included in the fire ant (*Solenopsis saevissima*) eradication program which used heptachlor in the 1950s. Mirex was substituted for heptachlor in the early 1960s. Heptachlor epoxide residues found in adult wings from Louisiana ranged from 0.52 to 1.13 ppm; the geometric mean was 0.70 ppm. Residues in adult wings from the tri-state area ranged from 0.21 to 1.48 ppm; the geometric mean was 0.58 ppm.

Woodcock wings from the two southern areas, Louisiana and the tri-state area, had consistently higher organochlorine residues other than PCBs. PCB residues were highest in Massachusetts and second highest in the southern areas. Wings from Minnesota had the lowest organochlorine residues except for DDT. Wisconsin had the lowest DDT residues; Minnesota had the second lowest.

Eleven of the 13 states, including those in the tri-state area, were sampled in both 1970–71 and 1971–72 (Table 2). Generally, residues were lower in the second sampling period. DDE, mirex, and PCB residues were significantly lower in 1971–72 than in 1970–71 ($P<0.05$, $P<0.01$, and $P<0.01$, respectively).

The relationship of residue levels among states for the two years was tested by a two-way analysis of variance (Table 3). Residues in both sampling periods were consistently highest in the southern states and in New Jersey. Residues were lowest in the northern and midwestern states.

Table 4 shows ranges and geometric means of organochlorine insecticide residues found in immature woodcock wings. Immature wing residues follow the same pattern as residues in adult wings in all but three instances. Mirex residues were higher in immature wings from the tri-state area than in immature wings from Louisiana. Average PCB residues in immature wings were lowest in New Jersey, Louisiana, and the tri-state area; this is the opposite order of residues in adult wings. Heptachlor epoxide residues were found in adult and immature wings from Louisiana and the tri-state area; heptachlor epoxide was also found in two pools of immature woodcock wings from New Jersey.

TABLE 2. *Geometric means of organochlorinated insecticide residues in woodcock wings from eastern/midwestern states, 1970–71 and 1971–72*

Residue	Geom Mean, ppm Lipid Weight	
	1970–71	1971–72
DDT	1.48	1.26
DDE [1]	8.79	6.82
TDE	1.41	1.42
Dieldrin	0.31	1.31
Mirex [1]	1.54	1.09
PCB [2]	5.58	1.64

NOTE: See Table 3 for list of states sampled
[1] Significant at $P<0.05$
[2] Significant at $P<0.01$

TABLE 3. *Comparison of organochlorine insecticide residues in adult woodcock wings, 1970–71 and 1971–72*

State	Geom Mean of Residues, ppm Lipid Weight					
	DDE	DDT	TDE	PCB	Dieldrin	Mirex
Maine	4.71d	0.78de	0.19c	2.18c	0.08b	1.04c
New Hampshire	7.58cd	1.61cd	0.25c	3.08bc	0.19b	0.63c
New York	5.92cd	0.77de	0.15c	3.27b	0.19b	1.04c
Pennsylvania	4.07d	0.70de	0.11c	2.51bc	0.17b	0.46c
New Jersey	16.01ab	5.35ab	0.81b	4.16a	0.53ab	0.63c
Tri-state area [1]	28.56a	9.19a	2.27a	5.24a	2.25a	3.11b
Louisiana	10.83bc	2.33bc	0.66b	3.36b	1.98a	10.25a
Michigan	4.65d	0.63e	0.16c	2.43bc	0.15b	1.39c
Wisconsin	5.05d	0.33e	0.51c	2.14c	0.15b	1.08c

NOTE, Values with the same letter are not significantly different
[1] See Table 1 for explanation

TABLE 4. *Ranges and geometric means of organochlorine insecticide residues in immature woodcock wings from seven eastern/midwestern states, 1971–72*

State	Residues, ppm Lipid Weight						
	DDT	DDE	TDE	PCB	Dieldrin	Mirex	Heptachlor Epoxide
Maine	0 51–2 28 1 19	2 56–5 28 4 07	0 16–0 41 0.25	0 75–1 07 0 89	0.06–0.83 0 16	ND —	ND —
Michigan	0 46–4 33 0 92	1 90–9 77 3 16	0 12–2.04 0 23	0 95–1.52 1 18	0.07–0.35 0 20	ND —	ND —
New Jersey	3 10–27 04 6 41	9 40–18 01 13 64	0 46–2 84 1 11	1 93–4 28 2 55	0 61–1 07 0 88	ND —	ND–0 42 0.13
Tri-state area [1]	2 89–18 10 6 82	15 29–47 47 26.03	0 64–4 09 1 46	ND–3 93 2 04	0.76–2 70 1 64	1 80–3.98 2 87	0 26–1 23 0 51
Louisiana	1 93–4 01 2 97	7 42–12 53 9 80	0 46–0 95 0 72	1 27–3 68 2 23	1 32–10 20 2 46	1 43–3 72 2 48	0.45–0.96 0 69

NOTE. ND = not detected
[1] See Table 1 for explanation

Mirex levels in wings of adult and immature woodcock from Louisiana are clearly different; the residues in wings from adults were significantly higher ($P < 0.05$). Mirex residues in adult wings ranged from 4.70 to 8.49 ppm; the geometric mean was 6.20 ppm. In immature wings, mirex residues ranged from 1.43 to 3.72 ppm; the geometric mean was 2.48 ppm. Mirex residue levels from all other states were very low. No significant difference in residue levels were found between adult and immature woodcock in other states, nor among other organochlorine insecticides.

The authors conclude that woodcock wings can be used to help determine the levels and trends of a variety of environmental pollutants in the eastern United States. Periodic assessment of residues in the wings of this species will provide important monitoring information at nominal cost.

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Chlorinated Hydrocarbons and Mercury in Birds of Lake Päijänne, Finland—1972–74[1]

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ABSTRACT

The levels of mercury, PCBs, DDT and its analogs, lindane, and dieldrin were examined in aquatic birds nesting on the shores of Lake Päijänne, the second largest lake in Finland, which is polluted by a wood-processing industry and urban sewages. The primary food of the 10 species examined was fish. In muscle of about 350 individuals, the highest average residues were PCBs; in livers, mercury was the highest. Lindane was found in some individuals; dieldrin appeared in none. The differences among levels in 1972, 1973, and 1974 were not significant. Some regional differences were found, particularly for mercury. Some PCB contamination was observed near the town of Jyväskylä. DDT was distributed evenly. A stronger correlation existed between residues of PCBs and DDT than between residues of any other compounds. In some gulls, males had higher average residues than had females. The DDT:PCB ratio generally corresponded to that of the North Atlantic Ocean, but the difference among species was great. Higher mercury, PCB, and DDT values existed in adults than in juveniles; higher mercury values existed in livers than in muscles. Black-throated divers had highest mercury residues; in herring gulls, PCBs and DDT were highest. The levels generally correspond to those found in other studies.

Introduction

Authors undertook the present study to discover the levels of chlorinated hydrocarbons and mercury in the aquatic birds of Lake Päijänne, Finland. Simultaneously, the methods of chemical analysis and the chemical structures of the compounds were developed. Data on the birds were collected by the University of Jyväskylä as part of a monitoring study of the chlorinated hydrocarbons and mercury in the food webs of Lake Päijänne, in which residues were analyzed in the higher aquatic plants, plankton, bottom fauna, sediment, fishes, and aquatic birds.

Adults and juvenile birds were analyzed separately. Juveniles were birds of the same summer, ranging in age from a few days to several weeks. Muscle and liver tissues were analyzed separately.

Concentrations of different residues were analyzed according to age, location on the lake, and species. Attention was also paid to the differences between and ratios of residues in liver and muscle, and to the correlations of different residues to muscle:liver ratios, differences of residue load between the sexes, and the ΣDDT:PCB ratios. The significances of the differences were statistically tested.

Sampling and Collection

Lake Päijänne, the second largest lake of Finland (1100 km^2), has been the object of limnological investigations since 1968 (*32*). It receives waste principally from the three origins shown in Figure 1. The sulphite and sulphate pulp mill wastes of Äänekoski come from the north in the upper part of the watercourse, approximately 40 km upstream from Lake Päijänne. Wastes are also discharged into the northern part of the lake from the town of Jyväskylä via Lake Jyväsjärvi (station 1); these effluents contain urban sewages and paper mill wastes. The third source of wastes is in the center of the lake near station 4, which receives effluents from a sulphite pulp mill and two paper mills of Jamsa, as well as a minor amount of domestic waste. At the northern end of the lake, the content of human sewages is greater than in the center which is contaminated almost exclusively by the wood-processing industry. When flowing from the north to the center (station 3), the water becomes cleaner. Water extending from the central part of the lake (station 4) to the southern part (station 6) is quite clean.

The main sampling sites of the study were stations 1, 4, 5, and 6. From stations 2 and 3, a few birds were obtained for supplementary study. Station 1 is polluted by domestic

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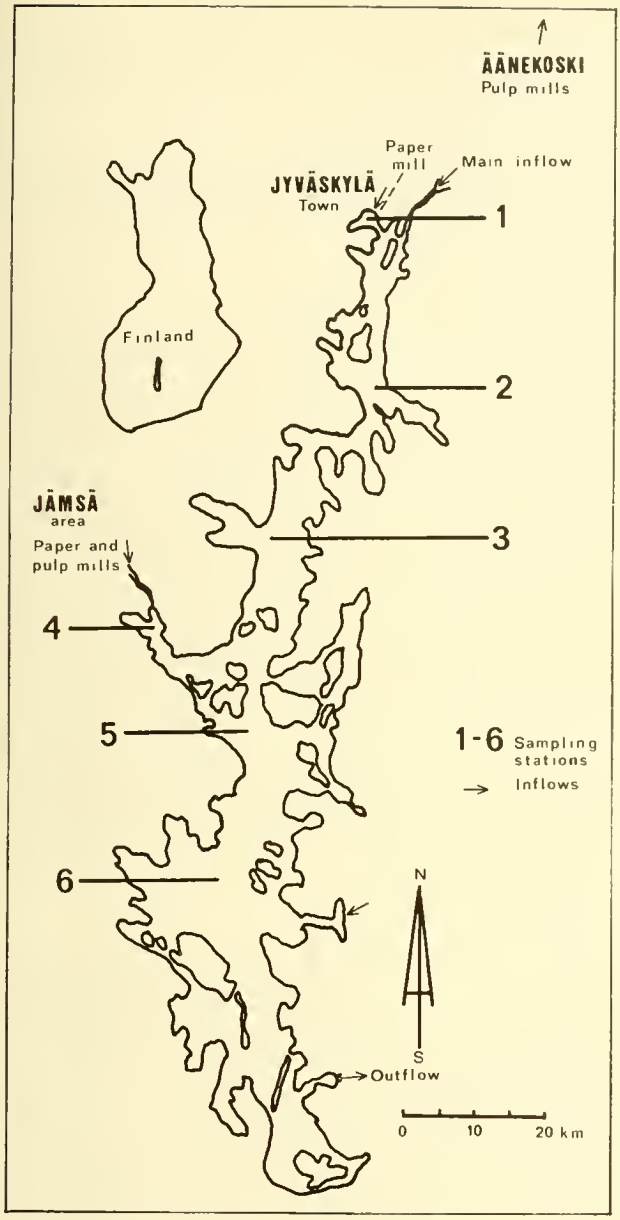

FIGURE 1. *Lake Päijänne with sampling stations.*

sewages and paper mill effluents. Until 1968, effluents from the paper mills and the Äänekoski pulp mills contained mercury originating from slime-preventing chemicals (*12*). Water at stations 2 and 3 gradually becomes cleaner as it moves south. Water at station 4 is affected by a wood-processing industry whose effluents contained mercury until 1968. Water at station 5 becomes cleaner as it approaches station 6, which is almost limnologically pure (*28*).

Adult birds were collected by shooting and young birds were caught live. No individuals were found dead. The adults were all caught after eggs had been laid. The whole birds were conserved by freezing in plastic bags which did not contain PCBs.

Species analyzed were black-throated diver (*Gavia arctica* L.), great crested grebe (*Podiceps cristatus* L.), goldeneye (*Bucephala clangula* L.), redbreasted merganser (*Mergus serrator* L.), sandpiper (*Tringa hypoleucos* L.), lesser black-backed gull (*Larus fuscus* L.), herring gull (*Larus argentatus* L.), common gull (*Larus canus* L.), black-headed gull (*Larus ridibundus* L.), and common tern (*Sterna hirundo* L.).

Species were chosen to represent aquatic birds, especially those which feed at Lake Päijänne in the summer. This is why such species as mallard (*Anas platyrhynchos*) and other common game birds were not sampled. All species, however, are migratory, spending only about one third of the year in Finland.

The number of birds analyzed for total mercury was 344; for methyl mercury, 32; and for chlorinated hydrocarbons, 301.

Analytical Procedures

CHLORINATED HYDROCARBONS

The frozen sample was thawed and 5-10 g breast muscle or liver was weighed. The sample was ground in a mortar with acid-washed sand (Merck) and anhydrous sodium sulphate, 4 g of the latter for each gram of wet tissue. The homogenized mixture was transferred to a glass container and dried at room temperature for 48 hours.

The extraction was performed by Soxhlet in thimbles which had been washed ultrasonically in a 1:1 mixture of acetone and diethyl alcohol. The homogenate was transferred to the thimble and extracted for 6 hours in a mixture of diethyl ether, petroleum ether (boiling point 40°–60° C), n-hexane, and acetone in quantities of 1:9:2, 5:5, 5 (v/v). All solvents were pesticide analytical (p.a.) grade and redistilled. This solvent system has been statistically proved to be the most effective for extracting animal tissue (*14*).

The extracted fat was weighed and cleaned by the following methods: shaking with concentrated sulphuric acid (*2*), thin-layer chromatography (*15*), and a column chromatographic method (*16*). In routine analyses, if extracted fat exceeded 20 mg, it was made into a 1 percent solution in n-hexane and divided into halves. One half was shaken with concentrated sulphuric acid for determining total PCBs, lindane, and DDE. The residues were extracted in hexane which was ready for gas chromatography. The hexane was shaken again with chromic acid for determining DDE (*35*). The second half was applied on a thin-layer plate for determining TDE, DDT, dieldrin, and endrin. When extracted fat was 10 mg or less, thin-layer

chromatography was the only cleanup method used. The column chromatographic method was used mainly for analyzing bird material because residues were greater than in the rest of the samples and required dilution from 10 mg fat, which is the maximum amount accommodated by the column, to 10 ml fat for proper gas chromatography. The cleanup methods have been tested to determine the highest values of the chlorinated hydrocarbons per fresh weight of tissue (*15*). The highest value of PCBs is the only recovery criterion available at present. The sulphuric acid cleanup produced a satisfactory measurement of PCBs in a fat reference sample of the Organization for Economic Cooperation and Development (OECD).

The equipment used in determining the residues was a Varian Model 600 D gas chromatograph with an H^3 electron-capture detector. The length of the glass column was 1.5 m and the inside diameter was 1.5 mm. In the routine analyses the column filling was a mixture of 65 parts of 8 percent QF-1 and 35 parts of 4 percent SF-96 on Chromosorb W 100-120 mesh. Occasionally SF-96 on Chromosorb W 100-120 mesh was also used for control purposes. The carrier gas was nitrogen (99.999 percent). The column temperature was 180° C, the detector and injector were 190° and 225° C, respectively.

The following pesticide standards, all 100 percent pure, were used: aldrin, *p,p'*-TDE, *p,p'*-DDE, *p,p'*-DDT, *o,p'*-DDT, dieldrin, endrin, and lindane. The PCB standard was Clophen A 60 by Bayer because the PCB contamination in Finland had been statistically tested and proved to be that type (*13*). The final concentration in chromatographing was 10 ng/ml for pesticides and 100 ng/ml for the PCBs. The calculation was carried out as described by Gaul (*10*) and the PCBs were calculated by summing nine peaks (total 13 peaks) which did not interfere with the pesticides. Injection of 50 pg pesticides or 500 pg PCBs produced peak heights of approximately 50 percent of full-scale deflection.

TOTAL MERCURY

Total mercury was determined by cold vapor atomic absorption using a Coleman MAS-50 mercury analyzer. A sample of 0.5-1 g was homogenized in an Erlenmeyer flask with 0.5 ml water, and 10 ml concentrated sulphuric acid was added while the flask was kept in an ice bath. The flask was then covered with plastic film and kept in a 60° C water bath for 4 hours. After cooling, 15 ml 6 percent solution of $KMnO_4$ was added from a buret, the bottle was kept in an ice bath and shaken well, and the sample was diluted to 100 ml. To reduce mercury II ions to mercury metal, 2 ml 20 percent hydroxylamine hydrochloride and 1 ml stannous chloride (40 percent solution in 5 percent sulphuric acid) were added and the measurement was taken immediately. The standard was $HgCl_2$ and a standard curve was made daily after treating the standard as described above.

METHYL MERCURY

Methyl mercury was identified by gas chromatography using the following conditions:

Chromatograph Varian Aerograph 2400
Detector H^3 tritium
Column glass, 1.8 m long and 6 mm ID, packed with 10 percent Carbowax 20M on Chromosorb W 80–100 mesh
Temperatures column 140° C
injector, 180° C
detector, 210° C

In a Sorvall Omnimixer, 1–5 g material was homogenized in 26 ml 29 percent KBr. Then 3.5 ml 47 percent HBr that had been prewashed with benzene was added to the homogenate which was then centrifuged and the liquid was decanted. The homogenate was treated again with KBr and HBr. The liquid phases were combined in a 250-ml separatory funnel and 50 ml redistilled benzene was added. Methylmercury bromide was added to the benzene. The water phase was extracted again with 25 ml benzene and the extracts were combined; then 8 ml 20 percent cysteine acetate (dried with Na_2SO_4) was added and the solution was shaken to bind the methylmercury bromide to cysteine. Five ml of the water phase was shaken with 1 ml 47 percent HBr and 10 ml benzene to extract the methylmercury bromide in benzene. The benzene phase was chromatographed and the peak heights of the sample and the standard were calculated. Injection of 50 μg Hg as methylmercury bromide produced a peak height of full-scale deflection.

Results

Table 1 shows the average levels of the residues studied in muscles and livers of both adult and juvenile birds. Differences among species, areas, and years are not considered in this table. The table shows that in muscles, the residues of highest concentration are PCBs; in the livers, mercury appears at the highest levels. Concentrations of TDE and DDT are very small compared with those of DDE; all three are combined in subsequent tables as ΣDDT. Lindane was present in only a few individuals, accounting for minute

TABLE 1. *Average chlorinated hydrocarbon and mercury concentrations in muscles and livers of aquatic birds, Lake Päijänne, Finland—1972–74*

Compound	Average Concentration, mg/kg Wet Weight			
	Muscles		Livers	
	Adults	Juveniles	Adults	Juveniles
Total Hg	2.729	0.777	7.900	2.312
Methyl Hg	2.697	0.275	—	—
PCB	4.970	1.135	5.734	0.961
DDE	3.373	0.708	4.187	0.821
TDE	0.012	0.000	0.015	0.000
DDT	0.002	0.000	0.007	0.000
ΣDDT	3.387	0.708	4.209	0.821
Lindane	0.002	0.001	0.000	0.000
Dieldrin	0.000	0.000	0.000	0.000

residue averages. Dieldrin was not present in any individual at concentrations above 0.0005 mg/kg wet weight.

The material of each year of study consists of different numbers of samples from different sampling areas, so results for the different years were not compared with parametric statistical tests. The yearly differences of the average concentrations of total Hg, PCBs, and ΣDDT were examined separately in different bird species for the muscles and livers of the adults and juveniles, using nonparametric Friedman two-way analysis of variance or Wilcoxon matched-pair signed-rank tests (*29*). No significant differences among the years were observed.

Tables 2–5 present the corresponding residues in birds at different areas of the lake. Because residues vary broadly

TABLE 2. *Average concentrations of total Hg, PCBs, and ΣDDT in muscles of adult birds, Lake Päijänne, Finland*

Species	Statistic	Study area 1			Area 2			Area 3			Area 4			Area 5			Area 6		
		Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT
		Residues, mg/kg wet weight																	
Black-throated diver	M	—	—	—	—	—	—	—	—	—	—	—	—	12 80	3.53	5.66	14.57	1 81	6.81
	SD													0.00	2 88	2.59	0.00	0.37	4.34
	N													1	3	3	1	3	3
Great crested grebe	M	2 88	3.99	3.74	—	—	—	—	—	—	1 78	1.33	3.54	—	—	—	—	—	—
	SD	1.13	3.11	2 76							0.80	1 02	3 11						
	N	8	8	8							4	4	4						
Goldeneye	M	—	—	—	—	—	—	—	—	—	0.24	0 22	0.14	—	—	—	—	—	—
	SD										0.06	0 17	0.03						
	N										4	3	3						
Merganser	M	—	—	—	—	—	—	—	—	—	—	—	—	5 48	2 16	1 28	5 42	1 85	3 22
	SD													0.00	0.00	0 00	1 41	1 41	4 36
	N													1	1	1	3	3	3
Sandpiper	M	—	—	—	—	—	—	—	—	—	—	—	—	0.31	0.28	0 71	0 63	3 30	5 41
	SD													0.00	0.00	0.00	0 00	0.00	0 00
	N													1	1	1	1	1	1
Black-backed gull	M	3.00	7.97	1.11	3.44	14 32	5.87	2.43	—	—	2 74	3 64	5 22	3 27	7.15	7.02	3.66	5 10	6 60
	SD	0.00	0.00	0 00	1 24	4 72	1 93	0.60	—	—	1 58	3.09	3 79	1 06	4 91	3 54	1.29	3 29	4 46
	N	1	1	1	5	5	5	3			9	9	9	17	17	17	15	12	12
Herring gull	M	0.10	6 69	1 79	—	—	—	—	—	—	4.00	19 54	20.38	2.80	20 49	6 90	2.97	8 46	6 89
	SD	0.00	0.00	0.00							1.25	2.06	2.36	1.10	12.31	5 10	1 92	5 33	3.92
	N	1	1	1							2	2	2	5	5	5	12	8	8
Common gull	M	2.64	8.98	5.88	2.05	7 16	2 61	3 16	11.77	14 29	2 03	4 04	2.41	2 01	3.33	2.58	1.70	3 18	2.40
	SD	0.00	0.00	0.00	0.15	3 15	2 86	0.00	0.00	0 00	1 15	4 54	2 18	1 48	2.57	2 41	1.01	3 26	1 84
	N	1	1	1	5	5	5	1	1	1	20	20	20	14	14	14	16	16	16
Black-headed gull	M	1 78	4 67	1 58	2 73	4 89	0 87	—	—	—	0 96	2.28	0 69	1 17	2 60	0 48	—	—	—
	SD	0 88	4 17	3 38	1.55	2 53	0.32				0 54	5.84	1.07	0 35	0.14	0.13			
	N	17	16	16	4	4	4				18	18	18	2	2	2			
Common tern	M	3 01	6 18	2.36	2 68	4 53	0 66	5 94	—	—	3 48	2.91	1.37	4.38	2 92	1.32	5.08	4 27	1.53
	SD	1 60	4.38	5 18	2 19	3 03	0 29	0.51			2.28	1 92	1 50	1.32	1 52	0.94	1.61	3.92	1 56
	N	13	12	12	5	5	5	2			16	16	16	7	6	6	7	6	6

NOTE: See Figure 1 for location of study areas.
M=mean, SD=standard deviation, N=number of observations

TABLE 3. *Average concentrations of total Hg, PCBs, and ΣDDT in muscles of juvenile birds, Lake Päijänne, Finland*

Species	Statistic	Study Area 1			Area 2			Area 4			Area 5			Area 6		
		Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT
		Residues, mg/kg wet weight														
Black-throated diver	M	—	—	—	—	—	—	—	—	—	—	—	—	8 15	—	—
	SD													0.00		
	N													1		
Great creasted grebe	M	0.53	1.25	0.30	—	—	—	0.38	0.19	0.11	—	—	—	—	—	—
	SD	0.19	0.81	0 15				0 12	0.15	0 04						
	N	6	3	3				3	2	2						
Goldeneye	M	—	—	—	—	—	—	0.07	—	—	0.24	0.15	0.16	—	—	—
	SD							0.00			0.00	0.00	0.00			
	N							1			1	1	1			
Merganser	M	—	—	—	—	—	—	—	—	—	—	—	—	1.32	0.18	0.78
	SD													0.48	0.00	0.00
	N													4	1	1
Black-backed gull	M	1 98	7.70	4 39	0.87	4 06	0.82	—	—	—	0.92	1.10	0.51	0 69	2.28	5 00
	SD	0.00	0.00	0.00	0.00	0 00	0 00				0 23	1 14	0.44	0.41	2.30	5.31
	N	1	1	1	1	1	1				7	3	3	4	3	3
Herring gull	M	0.04	0.67	0.36	—	—	—	0.94	5.44	2.57	0 79	2.97	0 96	0.64	2.85	1.52
	SD	0.00	0.00	0.00				0.00	6 86	3 25	0.00	0.00	0.00	0.41	2.47	0.84
	N	1	1	1				2	2	2	1	1	1	11	3	3
Common gull	M	1 45	1.03	0 40	—	—	—	0 76	0 33	0 13	1 00	0 43	0.51	0.85	1 32	0.59
	SD	0.00	0.00	0.00				0 32	0 00	0 00	0.39	0 51	1 08	0.20	1 90	0.79
	N	1	1	1				3	1	1	13	12	12	6	4	4
Black-headed gull	M	0.45	0.52	0.10	—	—	—	0 35	0.36	0.15	—	—	—	—	—	—
	SD	0.28	0.33	0.05				0.32	0.37	0 11						
	N	9	8	8				11	11	11						
Common tern	M	0.50	0 95	0.16	—	—	—	1.12	1 06	0 67	0.34	1 00	0.41	0.45	0.68	0.25
	SD	0.09	0 22	0.04				0 65	0.59	0 62	0 07	0.35	0 12	0.02	0 00	0.00
	N	5	4	4				4	4	4	3	3	3	2	2	2

NOTE: See Figure 1 for location of study areas
M=mean, SD=standard deviation, N=number of observations

TABLE 4. *Average concentrations of total Hg, PCBs, and ΣDDT in livers of adult birds, Lake Päijänne, Finland*

Species	Statistic	Study area 1			Area 2			Area 3			Area 4			Area 5			Area 6		
		Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT
											Residues, mg/kg wet weight								
Black-throated diver	M	—	—	—	—	—	—	—	—	—	—	—	—	45.80	3.79	—	82.33	6.10	24.91
	SD													0.00	0.00	0.00	0.00	0.00	0.00
	N													1	1	1	1	1	1
Great creasted grebe	M	5.62	6.37	5.23	—	—	—	—	—	—	6.01	2.82	8.71	—	—	—	—	—	—
	SD	2.19	3.99	4.03							2.90	1.85	7.59						
	N	8	8	8							4	4	4						
Goldeneye	M	—	—	—	—	—	—	—	—	—	2.19	0.30	0.12	2.00	0.35	0.13	—	—	—
	SD										1.32	0.05	0.04	0.00	0.00	0.00			
	N										4	3	3	1	1	1			
Merganser	M	—	—	—	—	—	—	—	—	—	—	—	—	24.00	2.16	1.28	22.97	1.80	3.78
	SD													0.00	0.00	0.00	8.54	0.39	1.93
	N													1	1	1	3	3	3
Sandpiper	M	—	—	—	—	—	—	—	—	—	—	—	—	0.38	0.16	0.44	1.25	0.44	0.25
	SD													0.00	0.00	0.00	0.00	0.00	0.00
	N													1	1	1	1	1	1
Black-backed gull	M	7.20	6.83	3.19	8.74	19.97	16.40	8.42	4.60	4.97	7.98	9.59	9.53	9.74	5.91	8.54	10.62	5.72	7.82
	SD	0.00	0.00	0.00	2.09	16.28	10.99	0.78	2.86	3.55	4.36	8.64	6.96	4.10	5.32	5.22	4.35	3.78	6.54
	N	1	1	1	5	5	5	3	3	3	9	9	9	17	17	17	15	12	12
Herring gull	M	0.21	0.77	2.49	—	—	—	—	—	—	10.65	25.16	19.96	7.61	13.62	6.53	7.59	5.46	4.48
	SD	0.00	0.00	0.00							3.61	6.11	1.23	2.80	9.87	4.96	5.11	2.66	3.62
	N	1	1	1							2	2	2	5	5	5	12	5	5
Common gull	M	6.75	5.33	1.05	6.29	14.16	5.82	10.00	10.07	10.99	5.65	4.45	4.08	6.17	2.48	2.67	5.36	4.58	3.32
	SD	0.00	0.00	0.00	0.72	13.91	2.87	0.00	0.00	0.00	2.74	3.98	4.75	4.58	3.97	3.36	4.45	4.15	3.73
	N	1	1	1	5	5	5	1	1	1	20	19	19	14	14	14	16	16	16
Black-headed gull	M	4.65	6.86	1.11	6.04	4.59	2.34	—	—	—	2.65	2.03	0.66	2.28	2.21	0.65	—	—	—
	SD	2.72	5.17	0.72	3.51	1.94	2.21				1.56	1.74	0.61	0.74	0.28	0.07			
	N	17	17	17	4	4	4				18	18	18	2	2	2			
Common tern	M	7.79	6.19	1.14	6.34	10.35	2.09	15.30	7.10	1.38	8.36	3.87	1.62	15.52	2.62	1.04	14.6	5.35	1.80
	SD	5.06	4.01	0.87	4.68	8.03	1.27	0.28	0.40	0.26	6.32	2.65	1.61	9.52	1.45	0.55	4.32	4.17	1.92
	N	13	13	13	5	5	5	2	2	2	16	15	15	7	7	7	7	7	7

NOTE: See Figure 1 for location of study areas.
M=mean, SD=standard deviation, N=number of observations.

TABLE 5. *Average concentrations of total Hg, PCBs, and ΣDDT in livers of juvenile birds, Lake Päijänne, Finland*

Species	Statistic	Study area 1			Area 2			Area 4			Area 5			Area 6		
		Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT	Hg	PCB	ΣDDT
								Residues, mg/kg wet weight								
Black-throated diver	M	—	—	—	—	—	—	—	—	—	—	—	—	19.20	12.62	6.42
	SD													0.00	0.00	0.00
	N													1	1	1
Great crested grebe	M	1.00	1.40	0.29	—	—	—	2.21	0.71	3.55	—	—	—	—	—	—
	SD	0.11	0.34	0.10				0.93	0.48	3.31						
	N	2	2	2				3	3	3						
Goldeneye	M	—	—	—	—	—	—	0.22	0.24	0.13	0.49	0.40	0.30	—	—	—
	SD							0.09	0.05	0.08	0.01	0.17	0.36			
	N							2	2	2	3	3	3			
Merganser	M	—	—	—	—	—	—	—	—	—	—	—	—	2.78	1.81	1.77
	SD													0.63	2.60	2.70
	N													2	3	3
Sandpiper	M	—	—	—	—	—	—	—	—	—	—	—	—	1.11	—	—
	SD													0.04		
	N													2		
Black-backed gull	M	—	—	—	2.30	1.91	0.63	—	—	—	—	—	—	2.10	—	—
	SD				0.00	0.00	0.00							0.00		
	N				1	1	1							1		
Herring gull	M	0.26	0.46	0.14	—	—	—	—	—	—	1.32	0.34	0.18	1.77	—	—
	SD	0.00	0.00	0.00							0.00	0.00	0.00	1.64		
	N	1	1	1							1	1	1	5		
Common gull	M	—	—	—	—	—	—	3.11	1.20	0.42	3.50	0.43	0.72	3.43	0.96	0.37
	SD							1.43	0.00	0.00	1.22	0.48	1.04	1.01	1.33	0.31
	N							4	1	1	8	7	7	4	3	3
Black-headed gull	M	1.45	0.58	0.12	—	—	—	0.56	0.14	0.07	—	—	—	—	—	—
	SD	0.77	0.36	0.09				0.48	0.13	0.07						
	N	4	4	4				6	6	6						
Common tern	M	—	—	—	—	—	—	—	—	—	—	—	—	—	041	0.15
	SD														0.02	0.02
	N														2	2

NOTE: See Figure 1 for location of study areas.
M=mean, SD=standard deviation, N=number of observations

amont different species, locations, and sampling years, the nonparametric Friedman two-way analysis of variance was used here, too, for comparing the different areas. In areas 1, 4, 5, and 6 (Figure 1) and in the black-backed gull, herring gull, common gull, and common tern, significant regional differences occurred with ΣDDT in the livers of

the adult birds but with no other compounds. The greatest concentration of ΣDDT was in area 4 and the smallest concentration was in area 1.

Table 6 presents averages and standard deviations of total mercury, PCBs, and ΣDDT for the muscles and livers of adults and juveniles. Table 7 presents means and ranges of concentrations in different bird species. From these tables, comparisons between different species, between muscles and livers, and between adults and juveniles can be made. Because lindane was present in only three individuals, the data on this compound appear separately in Table 8.

TABLE 6. *Average Hg, PCB, and ΣDDT concentrations in muscles and livers of aquatic birds, Lake Päijänne, Finland*

Residue		Muscles		Livers	
		Adults	Juveniles	Adults	Juveniles
Total Hg	M	2.73	0.78	7.90	2.31
	SD	1 95	0.86	7 64	2.83
	N	242	101	243	50
PCB	M	4 97	1.14	5 73	0.96
	SD	5.32	1.75	6.33	2.08
	N	229	72	230	40
ΣDDT	M	3.39	0.71	4.21	0.82
	SD	3.94	1 58	5.41	1 68
	N	229	72	230	40

NOTE: M=mean, SD=standard deviation, N=number of observations.

The ratios of residues in muscle to residues in liver were compared with those of other studies (*1, 4, 9, 17, 18*). For mercury, this ratio varied in different bird species between 0.112 and 0.577 in adults, and between 0.278 and 0.573 in juveniles. The muscle:liver ratio for PCBs in adults was between 0.540 and 5.917; in juveniles the ratio was between 0.100 and 8.258. The ΣDDT ratio in adults ranged from 0.383 to 8.884, and in juveniles, from 0.044 to 9.919. These values are approximately the same as those found in the investigations cited above.

Table 9 presents the correlation coefficients of different residues. Compounds whose residues correlated most fre-

TABLE 7. *Concentrations of total Hg, PCBs, and ΣDDT in muscles and livers of aquatic bird species, Lake Päijänne, Finland*

Species	Residue	Muscles				Livers			
		Adults		Juveniles		Adults		Juveniles	
		Mean	Range	Mean	Range	Mean	Range	Mean	Range
		Residues, mg/kg wet weight							
Black-throated diver	total Hg	13.69	12.80–14.57	8.15	8.15	64.07	45.80–82 33	19.20	19.20
	PCB	2.67	1.32– 6.79	—	—	4 94	3.79– 6.10	12.62	12.62
	ΣDDT	6.24	2.90–11.82	—	—	16.27	7.63–24.91	6.42	6.42
Great crested grebe	total Hg	2 51	0.90– 4.76	0.48	0.30– 0 75	5.75	1 15– 8.50	1.73	0.92– 2.80
	PCB	3.10	0.54–10.68	0.83	0.09– 2.18	5.18	0.41–10.00	0.99	0.16– 1 65
	ΣDDT	3.67	0.19– 7.58	0.22	0.08– 0 46	6.39	0.06–17.55	2.25	0.22– 6.89
Goldeneye	total Hg	0 24	0.16– 0.28	0.16	0.07– 0.24	2.15	0.72– 3.42	0.38	0.15– 0.50
	PCB	0.22	0.06– 0.40	0.15	0 15	0.31	0 25– 0.36	0.34	0.21– 0.60
	ΣDDT	0 14	0.13– 0.17	0.16	0 16	0.12	0 09– 0.16	0.23	0.08– 0.71
Merganser	total Hg	5.44	3 80– 6.40	1.32	0.93– 1 97	23.23	16 00–32.50	2.78	2.33– 3.22
	PCB	1.93	0.59– 3.38	0.18	0.18	1.89	1.45– 2 23	1.81	0.30– 4.82
	ΣDDT	2.73	0.56– 8.25	0.08	0.08	3 16	1.28– 5.95	1.77	0.20– 4.89
Sandpiper	total Hg	0.47	0.31– 0.63	—	—	0.82	0.38– 1 25	—	—
	PCB	1 79	0.28– 3 30	—	—	0.30	0.16– 0.44	—	—
	ΣDDT	3.06	0.71– 5 41	—	—	0.34	0.25– 0.44	—	—
Black-backed gull	total Hg	3.25	1.32– 6.56	0.93	0.10– 1.98	9.46	4.50–22.20	2.20	2.10– 2.30
	PCB	6 71	0.27–18.87	2.74	0.38– 7.70	8.00	0.84–47.83	1.91	1.91
	ΣDDT	6.27	0.08–16.83	2.75	0.23–10.97	9.04	1.54–34.41	0.63	0.63
Herring gull	total hg	2.89	0 10– 6.55	0.65	0.04– 1.74	7.53	0 21–18.00	1 49	0.26– 4 62
	PCB	13.49	0.68–37.71	3.30	0.59–19.29	11.27	0.77–29.48	0.40	0.34– 0.46
	ΣDDT	8.26	1.04–22.05	1 60	0.27– 4.87	7.50	0.21–20.83	0.16	0.14– 0.18
Common gull	total Hg	1 97	0.45– 5.36	0 95	0.30– 1 59	5.85	1.22–16.60	3.38	1 42–5.56
	PCB	4 12	0.19–17.45	0.66	0.15– 4 15	4 98	0.33–37.90	0.64	0.09–2.50
	ΣDDT	2.73	0.12–14.29	0.50	0.11– 3.93	3.73	0.02–16.28	0.60	0.09–2.41
Black-headed gull	total Hg	1.48	0.18– 4 36	0.40	0.10– 1.22	3.79	0.64– 9.90	0.92	0 19– 2 52
	PCB	3.51	0.09–24.52	0.43	0.06– 1.34	4 29	0.36–19 13	0.32	0.05– 1.09
	ΣDDT	1.06	0.02–14.13	0.13	0.04– 0.42	1.01	0.09– 5.52	0.09	0.02– 0.21
Common tern	total Hg	3.73	0.69– 8.00	0.64	0.30– 1.92	10.10	0.68–35.60	1 11	1.08– 1.14
	PCB	4 15	0 43–15.92	0.95	3.24– 1.77	5 31	0.76–24.38	0.41	0.40– 0.43
	ΣDDT	1.57	0.16–18.75	0.39	0.12– 1.55	1.47	0.18– 6.55	0.15	0.13– 0.16
Average of all species	total Hg	2.73	0 10–14 57	0.78	0.04– 1.98	7.90	0.21–82.33	2.31	0.15–19.20
	PCB	4 97	0.06–37.71	1 14	0.06–10.29	5.73	0.16–47.83	0.96	0 05–12.62
	ΣDDT	3.39	0.02–22.05	0.71	0.04–10.97	4.21	0.02–34.41	0.82	0.02– 6.89

TABLE 8. *Lindane residues in muscles of three individual birds, Lake Päijänne, Finland*

Species/age	Study Area	Date	mg/kg Wet Weight
Common gull, juvenile	5	1-8-73	0.019
Common gull, juvenile	5	1-8-73	0.058
Merganser, adult (male)	6	5-6-73	0.362

NOTE: See Figure 1 for location of study areas.

TABLE 9. *Correlation coefficients (r) of different residues in muscles and livers of adult and juvenile birds, Lake Päijänne, Finland*

Tissue	Residue	Total	Methyl Hg	PCBs
Muscles Adult	methyl Hg	+0.287***		
	PCBs	+0.214***	+0 049	
	ΣODT	+0 237***	+0.026	+0.565***
Muscles Juvenile	methyl Hg	+0.039		
	PCBs	+0.114	−0.018	
	ΣDDT	+0.074	−0 012	+0.700***
Livers Adult	PCBs	+0.131*		
	ΣDDT	+0.317***		+0 644***
Livers Juvenile	PCBs	+0.819***		
	ΣDDT	+0.543***		+0 689***

NOTE: * = p < 5 percent.
*** = p < 0.1 percent.

quently in the greatest number of birds were PCBs and ΣDDT.

Table 10 presents the percentage of total mercury which is methyl mercury. Percentages varied in different species between 91 and 117, indicating inaccuracy of analytical methods, since the correct value must be below 100 percent.

TABLE 10. *Ratios of methyl mercury to total mercury in muscles of adult birds from study areas 1 and 4, Lake Päijänne, Finland—1972*

Species	N[1]	Methyl Hg: Total Hg,%[2]	Ratios, Range
Great crested grebe	6	91	67-110
Black-backed gull	4	117	107-126
Herring gull	1	100	100
Common gull	2	111	107-115
Black-headed gull	7	93	66-127
Common tern	10	107	69-160

NOTE: See Figure 1 for location of study areas.
[1] N=number of individuals sampled
[2] Percentages over 100 indicate inaccuracy in analytical methods

Table 11 lists t-test findings which indicate that average concentrations of residues in males and females differed significantly. Muscles and livers in adults of each species were tested. In some gull species significant differences were found, and in all cases the average residue concentration in males was higher than in females.

Table 12 presents the ratio of ΣDDT:PCBs among different bird species for comparison with corresponding values in earlier studies (*4, 5, 23, 27, 34*). Generally, birds which have been nesting in industrial areas contain more PCBs in relation to DDT than do individuals nesting far from such areas (*27*). In Lake Päijänne, these ratios never reached the high levels of 9:10 found in more remote areas of the globe, but the average levels do correspond to those of the North Atlantic (*4*). In many species the ratio ΣDDT:PCBs parallels the values for birds in Greenland (*5*). Great differences exist in the ΣDDT:PCB ratios of the different bird species of Lake Päijänne.

Discussion

The nonparametric tests showed no regional differences of concentration patterns among the birds sampled except for ΣDDT in liver. If the material sampled from the different areas is combined on this ground and t-tests are used to search the yearly differences for every species, only the PCB contents of the black-backed gull seem to have decreased. If the absence of any significant variations between the sampling years is regarded as a basic fact, then the regional differences can be examined for every species from material in which the results of the different years are combined. Such an examination indicates that the different gull species and the common tern contain significantly more mercury at areas 3, 5, and 6 than elsewhere; PCBs appear most often at area 1; and in all species, ΣDDT appears in comparatively even amounts at the different sampling areas. Thus for mercury, the regional maximums are not found in the locations of greatest pollution, areas 1 and 4. The same paradox applies to certain other trophic levels of the lake (*24*). An explanation of this might be that mercury is retained within the sediment at the low-oxygenated areas 1 and 4; it does not pass through the food chain as effectively as it would in sediment which is farther from the sources of pollution.

TABLE 11. *Significant t-test differences between residues in adult male and female birds, Lake Päijänne, Finland*

Species	Content	Tissue	Males			Females			Signif.[1] of Diff.
			M	SD	N	M	SD	N	
Black-backed gull	total Hg	muscle	3.51	1 30	25	2 80	1 05	22	*
	total Hg	liver	10.19	4 24	25	8 19	3.36	22	*
Herring gull	total Hg	liver	10 19	4 41	8	5 76	3 98	12	*
Common gull	total Hg	muscle	2.28	1 13	35	1 46	0 98	22	* *
	total Hg	liver	6 99	3 75	36	3 88	2 46	21	* *
	DDE	liver	4 63	4 24	35	2 24	3 26	21	*
	ΣDDT	liver	4 63	4 24	35	2 24	3.26	21	*

NOTE: M=mean, SD=standard deviation, N=number of observations
[1] Significances *=p<10%, *=p<5%, **=p<1%

TABLE 12. *ΣDDT:PCB ratios in muscles and livers of aquatic bird species, Lake Päijänne, Finland*

Species	ΣDDT:PCB Ratio			
	Muscles		Livers	
	Adults	Juveniles	Adults	Juveniles
Black-throated diver	2.339	—	3 292	0.509
Great crested grebe	1 185	0.267	1 233	2.281
Goldeneye	0.658	1.073	0.397	0 688
Merganser	1.418	0 429	1 671	0 976
Sandpiper	1.710	—	1 139	—
Black-backed gull	0.935	1.004	1 130	0.329
Herring gull	0.612	0.479	0 665	0.404
Common gull	0.663	0.763	0.750	0.933
Black-headed gull	0.300	0.301	0.235	0.277
Common tern	0.379	0.409	0.278	0.353
Average	0.681	0.624	0.734	0 854

PCBs seem to enter the watercourse from the town of Jyväskylä but their exact origin is unknown. Otherwise there is little regional difference of PCB and ΣDDT contamination in the waterways around Lake Päijänne, indicating that the residues detected in the birds originate primarily in the wintering regions or along the migration routes, or that they reflect the global levels of contamination. The differences between the average residues in adults and those in juveniles show that bioaccumulation occurs as individuals age (Tables 6, 7). Within each species the contents of mercury, PCBs, and DDT were significantly higher in adults than in juveniles. This was seen in all species that had sufficient material for statistical comparison.

Tables 6 and 7 also show that mercury levels are higher in liver than in muscle, and these differences are significant in all species having sufficient material for statistical comparison according to t-tests. Conversely, PCBs and ΣDDT do not accumulate in the liver more than in pectoral muscles.

Correlations between the different pesticide contaminants (Table 9) do not reveal any causes but they do, to a certain degree, illustrate the possible common origin of the different residues, possible similar bioaccumulation in the food chains, or possible similar behavior in metabolism. The high significant positive correlation between the levels of PCBs and ΣDDT indicates that these fat-soluble contaminants behave similarly.

Authors referred to the published literature to compare residue levels of Lake Päijänne birds with levels in the same species in other countries (*1, 3–7, 9, 17–23, 25–27, 33, 34, 36*). It must be remembered, however, that material from Lake Päijänne did not contain birds that were dead. This excluded from the sample those individuals that may have been fatally poisoned by pesticides.

For the goldeneye, merganser, herring gull, and common tern, mercury levels were lower in Lake Päijänne than in Canada; and for the common gull, mercury content was lower than in Norway. For the grebe, PCB levels were lower in Lake Päijänne than in Great Britain; and for the black-backed gull, levels were lower than in the Faeroe Islands north of Scotland. For the merganser, juvenile herring gull, and common tern, ΣDDT levels were lower in Lake Päijänne than in the United States; and for the black-headed gull, ΣDDT residues were lower than in the Po Delta of northern Italy.

In Lake Päijänne, mercury residues for the black-throated diver were greater than in Aberdeen in eastern Scotland, and Canada; and for mergansers and herring gulls, residues were greater than in Canada. In Lake Päijänne, mercury content was higher for mergansers than elsewhere in Finland; higher for black-backed gulls and herring gulls than in the Faeroes; higher for the herring gulls than at Fife, Scotland; and higher for the black-headed gulls than in Norway and Great Britain. Mercury was present in equal concentrations among mergansers in Lake Päijänne and goosanders in the Baltic Sea.

For the black-backed gull in the Faeroes and the black-headed gull of the Po Delta, PCB concentrations were lower than in Lake Päijänne.

For the black-headed gull and the common tern, ΣDDT concentrations were greater in Lake Päijänne than in the Po Delta. For the herring gulls from the Faeroes and the common tern from the Po Delta, PCB levels were equal to those of Lake Päijänne.

Lindane occurred at about the same concentrations in many individual birds from Lake Päijänne as in those from other locations. Fat of aquatic birds of Greenland averaged 0.40 mg/kg of lindane (*5*); aquatic bird eggs of Ireland averaged 0.045 mg/kg (*8*); cormorants in the United States averaged 0.05 mg/kg in liver and whole bodies (*11*). Black-headed gulls in the Po Delta averaged 0.049 mg/kg in the muscle and 0.495 mg/kg in the liver; for the same individuals, maximum values were 0.110 mg/kg for muscle and 1.87 mg/kg for liver (*34*). Although average concentrations of lindane in Lake Päijänne birds were almost zero, the maximum levels were similar to those in the other countries mentioned.

Dieldrin, which did not appear at all in Lake Päijänne birds, has been reported in aquatic birds elsewhere (*8, 11, 18, 22, 30 34, 37*). Values as high as 0.348 mg/kg have been observed in aquatic birds of Utah, although maximum levels range generally from 0.01 to 0.10 mg/kg (*30*).

Comparison of concentrations in various bird species shows that mercury residues are highest in the diver, merganser, common tern, and common gull. PCB contents are highest in the herring gull and common gull, and ΣDDT is highest in the herring gull, black-backed gull, and diver. Considering ΣDDT concentrations in liver

alone, residues are highest in the diver. Differences among the bird species may depend principally on feeding habits, although duration of life, migration routes, and wintering regions also cause differences. The gulls, especially the black-headed gull and the herring gull, feed on garbage as well as fish, and the black-headed gull also eats terrestrial animals living in arable lands.

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Dieldrin, DDT, PCBs, and Mercury Levels in Freshwater Mullet from the Upper Great Lakes, 1975–76 [1]

Mary E. Zabik,[2] Barbara Olson,[2] and Teiko M. Johnson [2]

ABSTRACT

Freshwater mullet harvested commercially during various seasons of 1975–76 from the upper Great Lakes were analyzed for organochlorine pesticides, PCBs, and mercury. Species analyzed were Catostomus commersoni, C. catostomus, *and* Moxostoma erythruran. *Whole ground fish, mechanically deboned flesh, head, middle, and tail steaks, and various muscles were analyzed for pesticides and PCBs; only edible flesh was analyzed for mercury. Dieldrin ranged from none detected to 0.23 ppm in deboned and whole ground samples, the DDT range was a trace to 0.30 ppm, and PCBs ranged from 0.06 ppm to 0.79 ppm. Levels were also higher in head sections and in high fat-containing medial muscle and belly flap. Mercury levels ranged from 0.03 ppm to 0.28 ppm in the flesh of mullet from Lake Michigan.*

Introduction

Freshwater mullet from the lakes surrounding Michigan have received little attention as significant sources of human food. In their native form, these fish are frequently considered unattractive to consumers because of their intramuscular bony structure and/or their muddy flavor which is characteristic of fish with their particular eating habits. Estimates indicate, however, that mullet could be harvested from Michigan waters at an annual rate approaching one million kg. Two species of the genus *Catostomus* comprise most of the mullet population in Lakes Huron, Michigan, and Superior. The white mullet *(Catostomus commersoni)* is widespread in Lakes Huron and Michigan, the longnose mullet (*C. catostomus*) predominates in Lake Superior, and the golden redhorse mullet (*Moxostoma erythruran*) is available in commercially harvestable quantities from Lake Huron.

In addition to their muddy flavor, these fish have been unpopular with consumers because of the numerous Y bones throughout the fleshy portion of the fish. Recently, however, mechanical means have been developed for separating meat from bone, yielding a boneless minced flesh product. This minced flesh can be used in various consumer products. However, before commercial products can be developed, it has been necessary to determine the levels of environmental contaminants, their seasonal variation, variation of environmental contaminants within different muscles, and location of the fish in representative species from the three lakes concerned.

Sampling Procedures

Mullet were harvested by commercial anglers from Lakes Huron (Saginaw Bay, Standish, and Au Gres, Michigan), Michigan (Epoufette Bay, Epoufette, Michigan), and Superior (Whitefish Bay, Brimley, Michigan) during different seasons of 1975–76. They were readily available from commercial anglers in Saginaw Bay. The fish were less readily available in the upper Lakes Superior and Michigan, so seasonal variation could not be determined specifically. Fish were ice-packed and transported to the laboratory for processing and analyses, usually arriving the day after the catch. Following heading and gutting, fish to be deboned by machine were split into halves and run through the Bibun deboner (Type SD × 13, 5-mm holes), resulting in a minced flesh product separated from bone, skin, and scales. Whole headed and gutted mullet (35–40 cm long) were coarsely ground three times in a Hobart food cutter fitted with chopper attachment. Other whole dressed mullet were filleted into the ventral, dorsal, medial, and belly flap muscles or sectioned into head, midsection, and tail cross slices. Two mullet, 35–40 cm long, were used for each muscle or section study for each catch date for each lake. Muscles or sections were homogenized separately in an Osterizer blender and all samples were frozen

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and held at −23° C in glass jars before being thawed overnight at 4°–5° C for residue analyses.

Analytical Procedures

PESTICIDES AND POLYCHLORINATED BIPHENYLS (PCBs)

Two samples of each fish variable were extracted separately with hexane–acetone (2:1), partitioned with acetonitrile, and subjected to Florisil-Celite column cleanup according to the method of Yadrick et al. (*7*). Solids were determined by drying 2-g samples under vacuum at 90° C to constant weight; lipid was estimated by evaporating an aliquot of the hexane extract to dryness at 70° C under vacuum.

Gas chromatographic analyses were performed with a Tracor 560 gas-liquid chromatograph (GLC) equipped with a ^{63}Ni electron-capture detector and interfaced with a Digital PDP-8e-Pamila GC data system. Instrument parameters and operating conditions follow.

Column:	1.83-m × 4.0-mm ID Pyrex, packed with 3 percent OV-1 on 80–100-mesh Chromosorb W-HP
Temperatures:	column 190° C injection port 230° C detector 300° C
Carrier gas:	nitrogen flowing at 40 ml/minute

Standards were prepared with 99+ percent pure recrystallized dieldrin, *p,p'*-DDT, and *p,p'*-TDE, and Aroclor 1248 in Nanograde hexane. Quantitations were based on peak area for pesticides; the area of three peaks was used to quantitate the PCBs. Standards were run every morning and after every eight or nine samples. Recoveries with this method of extraction and quantitation were 85±2 percent for PCBs and 92±1 percent for dieldrin and DDT compounds; limits of detection were 0.01 ppm for PCBs and 0.001 ppm for dieldrin and DDT compounds. Data presented in this paper are not corrected for recoveries.

Presence of these residues was confirmed by mass spectrometric analysis on a pool of all extracted samples from each lake. The chromatograph used was a Beckman GC-65 interfaced with a DuPont 21–490 mass spectrometer which in turn was interfaced with a Digital PDP-12-LDP computer. Mass spectra were obtained at an ionizing voltage of 70 eV with a source temperature of 210° C.

MERCURY

Mercury was determined from duplicate edible flesh samples for each catch from each lake as total elemental mercury by using flameless atomic absorption spectrophotometry as described by Gomez and Markakis (*2*). Concentrated sulfuric acid was used to digest the samples as described in their Digestion 1 procedure. Recovery was 95±1 percent, and the limit of detection was 0.005 ppm. Values presented are not corrected for recovery data.

Results

Fat, solids, pesticides, and PCBs in whole ground and mechanically deboned mullet from the upper Great Lakes are presented in Tables 1 and 2. Dieldrin content ranged from none to 0.23 ppm. ΣDDT in white mullet caught in Lake Superior in June ranged from a trace to 0.30 ppm. PCBs varied from 0.06 ppm to 0.79 ppm. All levels are below the tolerances for these environmental contaminants established by the Food and Drug Administration (FDA), U.S. Department of Health, Education, and Welfare, although dieldrin levels in the mullet from Lake Michigan are closest to their tolerance level, 0.3 ppm.

Seasonal variation appears to be minor. As much variation occurred in the levels of contaminants themselves as in the levels as they related to the different catch dates.

The Great Lakes Environmental Contaminant Survey analyzed two freshwater mullet under 16 inches long from Lake Huron in 1974 and four in 1975 (*3*, *4*). Values reported there are similar to those in the current study. An earlier analysis of a freshwater mullet revealed 1.14 ppm DDT (*5*). Thus DDT levels may be decreasing. Similar DDT levels were reported in freshwater mullet from Lakes

TABLE 1. *Fat, solids, pesticides, and PCBs in whole ground freshwater mullet, Upper Great Lakes, 1975–76*

Lake	Type	Date of Catch	Fat, %	Solids, %	Dieldrin		ΣDDT		PCBs as Aroclor 1248	
					Wet Tissue	Fat	Wet Tissue	Fat	Wet Tissue	Fat
					Residues, ppm					
Huron	White	February 75	2.63	23.43	0.03	1.10	0.06	2.03	0.54	10.03
	White	May 75	1.20	21.30	0.10	8.84	0.06	4.26	0.54	43.40
	White	August 75	2.70	24.40	0.09	3.15	0.08	2.89	0.79	29.36
	Redhorse	August 75	7.90	31.70	0.11	1.45	0.08	0.98	0.70	8.86
	White	December 75	2.30	21.30	0.16	3.97	0.30	13.31	0.12	5.30
	White	February 76	2.30	23.90	0.04	1.92	0.08	3.57	0.15	6.28
Michigan	Longnose	June 75	4.20	25.25	0.21	5.01	0.23	3.12	0.62	14.05
	Longnose	August 75	5.55	25.65	0.23	4.31	0.27	4.49	0.71	12.67
	White	June 76	1.15	24.00	0.03	2.77	0.03	2.82	0.16	12.99
Superior	White	June 75	2.05	22.35	—	—	Tr[1]	—	0.06	3.12
	Longnose	December 75	3.95	24.25	0.09	2.33	0.14	3.46	0.26	6.55

[1] Tr = 0.005–0.009 ppm

Ontario and Erie, although dieldrin levels were less than 0.01 ppm (*1*).

Variation in levels of environmental contaminants from head to tail is summarized in Table 3. The head slices which contained the most fat had the highest levels of environmental contaminants. On a fat basis, however, the distribution was more uniform.

Variation in contamination according to muscle content is shown in Table 4. The high-fat medial muscle and belly flap contained the highest amounts of residues. Because the residues are fat-soluble, trimming would be a feasible method of reducing contaminants if the deboned flesh ever exceeded FDA tolerances. Reinert and Bergman (*6*) also found that these areas had higher levels of contaminants in Coho salmon, but they concluded that trimming would have little benefit because residues in the loin muscles were also high.

Mercury levels in the edible flesh (Table 5) were highest in fish from Lake Michigan. Values reported for fish from Lake Huron are close to those reported by the Great Lakes Environmental Contaminants Survey (*3, 4*).

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TABLE 2. *Fat, solids, pesticides, and PCBs in mechanically deboned freshwater mullet, upper Great Lakes, 1975–76*

Lake	Type	Date of Catch	Fat, %	Solids, %	Dieldrin		ΣDDT		PCBs as Aroclor 1248	
					Wet Tissue	Fat	Wet Tissue	Fat	Wet Tissue	Fat
					Residues, ppm					
Huron	White	February 75	2.07	22.37	0.01	0.62	0.03	1.84	0.29	14.13
	White	May 75	1.50	19.83	0.06	4.27	0.06	4.14	0.50	33.39
	White	August 75	1.60	19.75	0.07	4.16	0.08	4.63	0.41	25.01
	Redhorse	August 75	5.50	24.85	0.05	0.96	0.04	0.69	0.18	3.22
	White	December 75	2.75	20.70	0.15	5.22	0.20	6.99	0.70	24.38
	White	February 76	2.95	20.25	0.07	2.47	0.10	3.26	0.17	5.88
Michigan	Longnose	August 75	5.23	23.90	0.13	2.37	0.16	2.93	0.49	9.29
	White	June 76	1.83	19.75	0.03	1.90	0.03	1.87	0.26	14.06
Superior	White	June 75	2.15	18.20	Tr[1]	—	0.01	0.56	0.06	2.93
	Longnose	December 75	3.00	21.15	0.07	2.28	0.12	3.88	0.70	23.32

[1] Tr = 0.005–0.009 ppm

TABLE 3. *Pesticides and PCBs in sections of freshwater mullet, upper Great Lakes, 1975–76*

Lake	Section	Mean Fat (Range), %	Mean Solids (Range), %	Mean Dieldrin		Mean ΣDDT		Mean PCBs as Aroclor 1248	
				Wet Tissue (Range)	Fat (Range)	Wet Tissue (Range)	Fat (Range)	Wet Tissue (Range)	Fat (Range)
				Residues, ppm					
Huron[1]	Head	5.80 (3.65–9.55)	27.77 (23.65–33.55)	0.16 (0.02–0.68)	3.35 (0.45–13.31)	0.24 (0.03–0.98)	4.84 (1.75–19.25)	0.86 (Tr[2]–1.92)	15.06 (Tr–32.40)
	Middle	3.14 (1.72–7.60)	24.13 (22.65–30.00)	0.06 (0.01–0.21)	2.66 (0.58–9.40)	0.07 (0.02–0.19)	2.91 (0.36–8.09)	0.39 (0.14–1.10)	15.54 (3.02–37.36
	Tail	2.04 (0.65–5.25)	26.21 (23.00–30.00)	0.03 (Tr–0.09)	2.34 (Tr–6.68)	0.04 (Tr–0.13)	2.97 (Tr–6.85)	0.18 (Tr–0.34)	12.94 (Tr–23.29)
Michigan[3]	Head	4.77 (2.25–8.20)	26.22 (23.20–30.10)	0.09 (0.05–0.11)	2.25 (1.33–3.10)	0.10 (0.06–0.17)	2.18 (1.94–2.53)	0.56 (0.49–0.61)	15.09 (8.07–18.78)
	Middle	3.82 (1.15–7.05)	24.30 (21.05–26.70)	0.09 (0.02–0.14)	2.17 (1.62–2.94)	0.12 (0.03–0.17)	2.13 (1.94–2.43)	0.29 (0.23–0.35)	11.61 (8.56–16.90)
	Tail	2.13 (1.24–3.80)	23.07 (21.10–24.70)	0.06 (0.02–0.08)	2.58 (1.58–3.95)	0.06 (0.02–0.08)	2.09 (1.94–2.25)	0.26 (0.10–0.46)	15.55 (7.40–30.58)
Superior[4]	Head	2.65 (2.05–3.25)	23.70 (21.90–25.50)	0.06 (Tr–0.11)	1.68 (Tr–3.36)	0.13 (Tr–0.23)	3.34 (Tr–6.68)	0.24 (0.08–0.39)	7.29 (3.16–11.41)
	Middle	2.10 (1.70–2.50)	24.15 (22.20–26.10)	0.02 (0.00–0.04)	0.84 (0.00–1.68)	0.03 (Tr–0.06)	1.17 (Tr–2.34)	0.15 (0.08)–0.22)	6.66 (4.42–8.89)
	Tail	1.70 (1.20–2.20)	22.48 (20.70–24.25)	0.03 (Tr–0.05)	1.21 (Tr–2.42)	0.06 (0.00–0.11)	2.56 (0.00–5.11)	0.14 (0.07–0.21)	7.80 (5.76–9.84)

[1] Based on six catches from Feburary 1975 to February 1976.
[2] Tr = 0.005–0.009 ppm
[3] Based on three catches from June 1975 to June 1976
[4] Based on two catches from June 1975 to December 1975

TABLE 4. *Pesticides and PCBs in muscles of freshwater mullet, upper Great Lakes, 1975–76*

Lake	Muscle	Mean Fat (Range), %	Mean Solids (Range), %	Mean Dieldrin		Mean ΣDDT		Mean PCBs as Aroclor 1248	
				Wet Tissue (Range)	Fat (Range)	Wet Tissue (Range)	Fat (Range)	Wet Tissue (Range)	Fat (Range)
				Residues, ppm					
Huron[1]	Ventral	0.83 (0.55–1.05)	19 94 (17 25–22.20)	0.02 (Tr[2]–0.05)	3.39 (Tr–11 20)	0.07 (0 01–0 28)	10.06 (1.22–38 69)	0 18 (Tr–0 52)	32.20 (Tr–120.02)
	Lateral line	5.44 (1.50–8 25)	24.26 (17.25–29.45)	0 08 (0.01–0.18)	2 18 (0.36–5.78)	0.10 (0.05–0.18)	2 43 (0 62–5.39)	0 80 (0.19–1 17)	16.93 (9 44–23 45)
	Dorsal	1.10 (0.50–1 90)	20.11 (17.05–21 65)	0.02 (Tr–0 06)	2 45 (Tr–7.10)	0.06 (Tr–0.20)	5.27 (0 63–20 47)	0 09 (Tr–0.17)	10.83 (Tr–16.43)
	Belly flap	3.51 (1.15–7.05)	21.55 (19 00–26 90)	0 10 (0.01–0.24)	3.24 (0.70–6.36)	0.13 (0.03–0.36)	3 91 (1 67–6.29)	0.69 (0.24–1 53)	21 03 (7.06–41.19)
Michigan[3]	Ventral	1.04 (0.60–1 45)	20 59 (19.85–21 30)	0.06 (0.02–0.10)	6.00 (4 04–9 83)	0.06 (0.03–0 11)	6.32 (3.28–11.11)	0.30 (0 09–0.46)	29 20 (16.59–40 42)
	Lateral line	8.13 (2.49–13 95)	26.70 (21.30–31 45)	0.22 (0.15–0.28)	3.92 (1 57–5 52)	0.28 (0 16–0.41)	4 32 (1 99–6.00)	1 22 (1.13–1.31)	22.10 (9 47–40.84)
	Dorsal	1.32 (0.52–2.30)	20 53 (18 85–21.40)	0.05 (0.02–0.10)	4 50 (1.60–8.51)	0.22 (0.16–0.34)	6 91 (1 80–13.36)	0.21 (0.02–0 34)	15.12 (3 44–23.38)
	Belly flap	6.13 (1.85–11.90)	23.88 (19 45–29.40)	0.26 (0.19–0.34)	5 98 (2 13–7 92)	0.37 (0.13–0.55)	7.77 (3 62–11 95)	1.33 (0.35–2.44)	24 49 (10.22–49 20)
Superior[4]	Ventral	1.59 (0.87–2.30)	19.68 (17 90–21.45)	0.04 (Tr–0 08)	1.33 (Tr–2.66)	0.07 (Tr–0 14)	2.37 (Tr–4 74)	0.08 (0 06–0.10)	7 85 (4 92–10.77)
	Lateral line	8 50 (6 80–10 20)	28 95 (26 75–31 15)	0 12 (0 02–0.22)	1 24 (0.35–2.13)	0.37 (0.03–0.70)	3 74 (0 56–6 91)	0.42 (0.27–0.57)	5 71 (2 75–8.66)
	Dorsal	1 45 (0 85–2.05)	19 73 (18 60–20 85)	0.02 (Tr–0.04)	1 06 (Tr–2 12)	0.04 (0.01–0.06)	2.22 (1.52–2 87)	0 08 (0.08–0 08)	7.32 (4 73–9 91)
	Belly flap	3.93 (2.00–5 85)	24 13 (20 00–28 25)	0.09 (0 02–0 15)	1 86 (0.71–2.95)	0 13 (0.01–0 24)	2 48 (0.62–4 34)	0.21 (0.13–0 28)	6.92 (6 70–7.13)

[1] Based on six catches from February 1975 to February 1976.
[2] Tr = 0 005–0.009 ppm
[3] Based on three catches from June 1975 to June 1976
[4] Based on two catches from June 1975 to December 1975

TABLE 5. *Mecury levels in freshwater mullet, upper Great Lakes, 1975–76*

Lake	Type	Date of Catch	Mercury, ppm
Huron	White	February 75	0.03
	White	May 75	0.06
	White	August 75	0.09
	Redhorse	August 75	0 07
	White	December 75	0 06
	White	February 76	0 05
Michigan	Longnose	June 75	0 21
	Longnose	August 75	0.12
	White	June 76	0 28
Superior	White	June 75	0.10
	White	December 75	0 06

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General

Mirex Incorporation in Estuarine Animals, Sediment, and Water, Mississippi Gulf Coast—1972–74 [1]

Armando A. de la Cruz [2] and Kuang Yang Lue [3]

ABSTRACT

Analysis of mirex residues in estuarine animals, sediments, and waters collected from the Mississippi Gulf Coast in 1972–74 showed the following ranges of concentrations: seston, 200–3000 ppb; molluscs, 36–500 ppb; fish, 0–259 ppb; sediment, 3–5 ppb; and water, 0–0.01 ppb. These data indicate that mirex in aquatic environments is localized in animal tissues and bottom substrate and that only a negligible amount is incorporated in the water

Introduction

In 1971–74, the authors conducted a series of studies on the toxicity and ecological and physiological effects of mirex on nontarget organisms. The three areas of study included residue monitoring and toxicity, effects of mirex on certain ecological processes of plants and animals, and physiological effects on enzyme systems. The results of these studies are cited in a literature review by Lue (*4*).

The ecological aspect of this project emphasizes the incorporation of mirex in the environment through leaching of the insecticide from decaying fire ant bait in the field (*2, 10*). Mirex residues were recovered from seafood from the Atlantic and Gulf Coastal states (*7*), in terrestrial and aquatic invertebrates from Louisiana (*8*), and in other selected organisms (*11*). During these studies, therefore, the authors routinely collected samples from different habitats (*9*). This paper reports mirex residues detected in samples collected from an estuarine environment on the Mississippi Gulf Coast. The animal samples were collected in the fall of 1972, the sediment samples during summer 1973, and the water samples in 1972 and 1974.

Materials and Procedures

COLLECTION OF SAMPLES

The animals were collected manually from the substrate in St. Louis Bay marsh during low tide. Those from Mississippi Sound were collected by using a shrimp trawl. The specimens were rinsed of mud or debris, blotted dry, wrapped in aluminum foil, and frozen until analysis. Water samples were collected in clean, hexane-rinsed 10-liter jugs by directly filling the jugs a few centimeters beneath the water surface. Water samples for mirex analysis were refrigerated when not immediately processed. Waters intended for seston analysis were promptly filtered through AA millipore filters (0.8-μm porosity) in a millipore vacuum-filtration apparatus. Seston is particulate matter suspended in water including plankton, organic detritus, and inorganic silt. Sediments were collected by an Ekman dredge from St. Louis Bay and by a Petersen dredge from Mississippi Sound. The samples were placed in clean, hexane-rinsed wide-mouth specimen jars and refrigerated until extraction.

EXTRACTION OF SAMPLES

Single or pooled (2–10 specimens) whole-body samples of animals were extracted for residue analysis according to the procedure of Naqvi and de la Cruz (*9*). Only the fleshy tissue of molluscs was extracted. Specimens were rinsed with distilled water to remove salt and briefly dipped in hexane to remove any external insecticide contamination. Samples were ground in nanograde hexane and shaken vigorously, and the decanted solvent was evaporated to dryness. Prior to gas-liquid chromatography, the extracts were cleaned by using activated alumina.

Seston samples were extracted according to the procedure in the *Pesticide Analytical Manual* (*3*) for small samples. The filter paper holding the seston was ground in a tissue grinder with acetonitrile. The filter paper was free of mirex when checked for contamination. The extract was concentrated and reduced to a suitable volume for analysis.

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Water was extracted with nanograde hexane in 250-ml separatory funnels; 150 ml samples were shaken vigorously with 50 ml hexane three successive times, 3 minutes each time. The three hexane extracts were combined and evaporated to a volume suitable for gas chromatographic analysis.

Samples of 150 g sediment were extracted with 300 ml hexane-isopropanol mixture (3:1) according to the procedure of Markin et al. (*6*). The extract was filtered through Na_2SO_4 and concentrated to 10 ml.

CHROMATOGRAPHY

Extracts of all samples were analyzed in a Barber-Colman Pesticide Analyzer Model 5360 equipped with an electron-capture detector. A 152.4 mm × 3.2 mm glass column was used. Standard injection techniques were used consistently for all samples. Extract volumes (2 μl) were injected. Information about operating parameters of the analyzer can be obtained from the Physiological Laboratory, Department of Zoology, Mississippi State University, Mississippi State, Mississippi 39762. The concentration of mirex was calculated with the following formula:

$$\text{mirex residue} = Vwd_2/Wvd_1$$

where W = weight of the sample in grams, V = volume of final extract in milliliters, v = volume of extract injected in μl, w = weight of the standard injection in nanograms, d_1 = peak height of standard solution, d_2 = peak height of extract. A second column (1.5 percent SP-250, dimethylchlorosilane-treated and acid-washed) was used to confirm the mirex residues recovered from the field samples.

Results and Discussion

Mirex residues in seston and animals collected from St. Louis Bay and Mississippi Sound are summarized in Tables 1 and 2. Concentrations in seston filtered from Mississippi Sound water (1000–3000 ppb) is one order of magnitude higher than in seston from St. Louis Bay (200–800 ppb). Residues in the animals were all below 1 ppm except in the fiddler crab *Uca* (1.3 ppm). The molluscs, i.e., snails, clams, and mussels, from St. Louis Bay, which are basically filter feeders, had slightly higher levels of mirex (36–500 ppb) than did the other invertebrates from Mississippi Sound (0–133 ppb). In an earlier study, Naqvi and de la Cruz (9) found 70–410 ppb mirex in snails and clams collected from a similar estuarine habitat. Residues in the fish ranged from 0 to 259 ppb.

The residue levels of sediments from bay and sound were essentially similar (Table 3) and fairly low (2.8–4.6 ppb). These values are, however, much higher than the residue levels detected in the water samples (0.001–0.010 ppb) from Mississippi Sound, St. Louis Bay, and from the

TABLE 1. *Mirex residues in seston[1] and animals[2] from St. Louis Bay marsh-estuary, November 1972*

Specimen	Biomass Extracted, g	Residues, ppb[3] Col. I	Col. II
Seston	0.07	817.7	920.6
	0.26	204.1	235.0
	0.19	215.9	199.7
	0.10	408.8	376.9
Rangia cuneata (Clam)	8.10	331.3	247.5
	3.00	490.2	450.0
Modiolus demissus (Ribbed mussel)	4.60	183.8	159.8
	3.00	36.7	71.3
Melampus bidentatus (Snail)	3.80	339.2	265.2
	2.90	471.4	415.2
	0.65	81.8	0.0
	0.45	118.2	0.0
Littorina irrorata (Snail)	0.40	130.9	0.0
	0.80	499.9	31.3
	0.70	75.9	89.3
	0.70	37.9	0.0
	0.60	66.5	35.7
Uca sp. (Fiddler crab)	0.30	1302.0	2661.0
Strongylura marina (Atlantic needlefish)	13.80	50.9	47.4

[1] Seston includes suspended particulate matter consisting of plankton organisms, organic detritus, and inorganic sediment filtered from 300 ml of water with 0.8 µm Millipore acetate filter.
[2] Animals were pooled from 2–10 individuals of about the same size. Biomass represents whole tissue, excluding shells and molluscs.
[3] All analyses were done with two columns to verify the mirex residue.

TABLE 2. *Mirex residues in seston[1] and animals[2] from Mississippi Sound, September 1972*

Specimen	Biomass Extracted, g	Residues, ppb Col. I	Col. II
Seston	0.01	3038.4	2396.8
	0.03	1507.8	2629.8
	0.03	1001.4	4150.5
	0.23	1172.7	1321.1
	0.01	3260.7	2007.4
	0.02	2291.8	2814.9
	0.01	2677.2	2677.2
	0.01	3243.4	3003.7
Sponge	0.61	133.5	231.0
Luidia clathrata (Starfish)	7.24	28.1	37.0
	8.68	24.0	0.0
Lolliguncula brevis (Squid)	5.40	0.0	0.0
	5.97	13.6	0.0
Palaemonetes sp. (Grass shrimp)	2.77	0.0	0.0
	3.39	0.0	0.0
Callinectes sapidus (Blue crab)	15.84	7.6	106.0
	20.80	3.7	0.0
	18.00	6.4	6.4
Squilla empusa (Mantis shrimp)	1.10	128.0	207.8
	1.30	22.0	0.0
Bairdiella chrysura (Silver perch)	8.30	4.8	0.0
Bagre marinus (Gafftopsail catfish)	6.60	1.1	1.8
Porichthys porisissimus (Atlantic midshipman)	11.70	81.6	97.0
	9.70	15.9	9.6
Etropus crossotus (Fringed flounder)	9.20	7.2	—
Symphurus plagiusa Blackcheek tonguefish)	11.30	12.5	11.7
	12.80	11.0	16.5
Cynoscion arenarius (Sand seatrout)	45.00	0.0	0.0
Strongylura marina (Atlantic needlefish)	19.30	259.1	245.4
	18.00	179.9	132.0

[1] Seston includes suspended particulate matter consisting of plankton organisms organic detritus, and inorganic sediment filtered from 300 ml of water with 0.8 µm Millipore acetate filter.
[2] Animals were single speciments; whole-body tissue was analyzed.
[3] All analyses were done with two columns to verify the mirex residue.

Jordan and Wolf Rivers that empty into the bay (Table 4). Spence and Markin (*10*) found that the highest mirex level in natural water was 0.02 ppb. In a separate study (*5*), the authors found 0.01 ppb residue in samples of water collected from a farm pond. The residue data reported in this paper indicate that mirex in aquatic environments is localized in bottom sediments, animal tissues, and in particulate matter, i.e., seston, suspended in the water, and that only negligible amounts of mirex are incorporated in the water (*1*, *10*).

TABLE 3. *Mirex residues in estuarine sediment, Mississippi Gulf Coast—1973*

Sampling Location	Collection Date	Amount Extracted, g	Residues, ppb [1]	
			Col. I	Col. II
St. Louis Bay [2]	5/29/73	100	2.8	5.0
	6/18/73	100	3.9	5.9
	8/26/73	100	3.5	5.3
Mississippi Sound [3]	7/17/73	100	4.6	2.2
	7/19/73	100	3.5	5.2

[1] All analyses were done with two columns to verify the mirex residue
[2] Collected by an Ekman dredge from the mouth of Catfish Bayou on the western side of the bay
[3] Collected by a Petersen dredge about 3 km off the Biloxi–Ocean Spring coastline

TABLE 4. *Mirex residues in estuarine water, Mississippi Gulf Coast—1972–74*

Sampling Site	Collection date	Amount Extracted, ml	Residues, ppb [1]	
			Col. I	Col. II
Jourdan River [2]	3/1/74	4,000	0.007	0.000
	6/20/74	4,000	0.005	0.000
			0.009	0.003
			0.004	0.000
Wolf River [2]	5/1/74	4,000	0.001	0.001
	7/31/74	4,000	0.004	0.001
			0.007	0.000
St. Louis Bay	5/1/72	4,000	0.000	0.000
	4/4/72	500	0.000	0.000
	11/15/72	500	0.000	0.000
	5/15/73	4,000	0.030	0.001
	6/12/73	4,000	0.000	0.000
	2/22/74	4,000	0.010	0.003
			0.000	0.001
			0.004	0.000
			0.000	0.000
	3/1/74	4,000	0.004	0.002
	4/6/74	4,000	0.000	0.000
Mississippi Sound	9/23/72	4,000	0.000	0.000
			0.004	0.000
	4/6/74	4,000	0.000	0.000

[1] All analyses were done with two columns to verify the mirex residue.
[2] Samples collected a few kilometers inland from St. Louis Bay

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APPENDIX

Chemical Names of Compounds Discussed in This Issue

ALDRIN	Hexachlorohexahydro-endo, exo-dimethanonaphthalene 95% and related compounds 5%
AROCLOR 1248	PCB, approximately 48% chlorine
DDD	See TDE.
DDE	Dichlorodiphenyldichloroethylene (degradation product of DDT)
DDT	Dichlorodiphenyltrichloroethane
DIELDRIN	Hexachloroepoxyoctahydro-endo, exo-dimethanonaphthalene 85% and related compounds 15%
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-methanoindane
LINDANE	Gamma isomer of benzene hexachloride (1,2,3,4,5,6-hexachlorocyclohexane) of 99+% purity
MIREX	Dodecachlorooctahydro-1,3,4-metheno-1H-cyclobuta[cd]pentalene
PCBs (polychlorinated biphenyls)	Mixtures of chlorinated biphenyl compounds having various percentages of chlorine
TDE	Dichlorodiphenyldichloroethane

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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SOIL

Bromacil and Diuron Residue Levels in Florida Citrus Soils[1]

David P. H. Tucker[2]

ABSTRACT

The widespread use of herbicides in Florida citrus groves raises the possibility of residue accumulation following repeated applications. To determine residue levels of commonly used herbicides, soil samples were taken from large experimental plots in commercial groves in Polk and Hardee Counties. Bromacil and diuron had been applied in combination at both locations for 7–8 years. Analyses of samples showed low levels of both herbicides at various soil depths to 60 cm. Only a small amount of bromacil was detectable one year after application, but diuron levels were higher. Continuous applications at recommended rates and frequencies have resulted in maximum bromacil and diuron levels of 3.9 percent and 13.1 percent, respectively, of their total application.

Introduction

Integrated weed control programs used on large acreages of citrus in Florida include herbicides, various cultivation practices, limited hand labor, and naturally occurring weed pathogens and insect pests. Herbicides have been widely used for the past decade, and have been applied annually to a large percentage of nonbearing and young-bearing acreage. Herbicides are now used on older groves to control rapidly increasing annual and perennial vines which thrive under tree canopies.

This widespread use of predominantly soil-sterilant herbicides has caused concern about their accumulation with repeated application. Therefore, continued monitoring of their residual levels in major citrus-growing soil types is warranted.

Bromacil and diuron are degraded in the soil by biological and nonbiological means, and they may be altered by one or more mechanisms including microbial decomposition, adsorption, volatilization, leaching, chemical degradation, and plant uptake (*2, 5, 7, 8*). A number of review papers on this general subject have been presented (*3, 4*). The persistence of soluble herbicides in soils in forms toxic to plants is likely to be less serious in humid areas such as Florida than in more arid citrus-growing regions. The amount, frequency, and intensity of rainfall is important to herbicide longevity in soil since moisture affects herbicide efficacy and mode of dissipation.

Tucker and Phillips (*9*) sampled the major citrus-growing soil types which had received repeated applications of herbicides. Analyses of these samples for bromacil, terbacil, dichlobenil, and trifluralin showed a fairly predictable annual rate of dissipation from the top 45 cm of the soil profile. The results precluded the possibility of any substantial toxicity to citrus trees due to accumulation in the soils following repeated applications at recommended rates. The present paper presents additional data showing levels of bromacil and diuron following their commercial application to two soil types at two grove locations over 7–8 years. Residue levels are shown at different locations under the tree canopy and at various depths.

Sampling and Analysis

In 1969 and 1970, paired 10-acre blocks of citrus were selected in commercial groves in Polk and Hardee Counties. Soil types were Astatula fine sand (95 percent sand, 0.42 percent organic matter, pH 7.8) and Mayakka fine sand (99 percent sand, 0.38 percent organic matter, pH 7.3), respectively. Annual rainfall at both locations averaged 114–127 cm. The Hardee County grove has a permanent overhead irrigation system with supplemental irrigation averaging 30–50 cm/year. The Polk County grove receives only occasional supplemental irrigation. At each site, weeds were controlled by tillage in one block and by broadcast

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herbicides in the other. Generally, weed control was satisfactory with one application of herbicide each year. However, in some years, herbicides were re-applied when weed growth resumed before the end of the season.

Herbicides were sprayed by a machine-mounted boom to the entire grove floor area rather than in strips down tree rows. Wettable powder formulations of bromacil (5-bromo-3-sec-butyl-6-methyluracil) and diuron [3-(3,4-dichlorophenyl)-1,1-dimethylurea] in tank mixes or as a chemically blended combination were used throughout the experimental period. Soil samples were collected with a 2.2-cm-ID soil tube from 0- to 15-cm and 15- to 30-cm depths at both locations except at one sampling time when samples were also taken from 30- to 45-cm and 45- to 60-cm depths. Each sample was a composite of 10 subsamples. Samples were taken in row middles between trees, at the drip line or tree canopy edge, and under the tree canopy. There were three separate sampling times in Polk County and two in Hardee County. Care was taken in obtaining the lower depth samples to avoid the top soil layers falling into the holes. To assure this, samples were taken during optimum soil moisture conditions. Samples were stored at -10°F before shipment for residue analyses by the Dupont Company. Samples were analyzed for bromacil by the microcoulometric gas chromatographic method of Pease (*6*), and for diuron colorimetrically after chromatographic cleanup by the method of Dalton and Pease (*1*).

Results and Discussion

The data in Table 1 show that concentrations of bromacil and diuron at depths sampled are very low in both locations compared to the total amounts applied over the 7–8-year experimental period. The levels, as percentages of the total amounts applied, range from 0.3 to 3.9 for bromacil and from 3.7 to 13.1 for diuron. As percentages of the last application only, they range from 2.5 to 31.0 for bromacil and 33.6 to 84.6 for diuron. This indicates that a substantial part of the residues remains from the latest application within one year of sampling.

Residues of diuron remained at considerably higher levels in the soil than did those of bromacil. This is influenced primarily by their relative water solubilities: 800 ppm for bromacil and 42 ppm for diuron. Residue levels do not appear to be influenced by the location of sampling. Since precipitation is greater on the tree drip line due to the umbrella effect of the tree canopy, leaching would also be greater, resulting in an earlier breakdown in weed control.

Other factors which may influence residue levels at various sampling locations include photodecomposition of diuron, probably greatest in the row middles due to the high light intensity. Under tree canopies, where sunlight breakdown and precipitation would be less, adsorption of herbicides by organic matter and breakdown by microorganisms would be greater. Another factor to consider is that spray coverage is frequently poorer in areas where tree canopies hinder equipment movement.

Inadequate spray coverage in the tree row also is frequently due to poor overlap of spray patterns. In most cases, bromacil was more evenly distributed throughout the profile depth sampled than was diuron where higher concentrations were consistently found in the surface layers. Again, this is a reflection of the much lower solubility of diuron and hence its slower movement through leaching. Overall residue levels of both herbicides were higher in the Mayakka fine sand of Hardee County than in the Astatula fine sand of Polk County.

Bromacil levels in control samples taken from cultivated plots are at or very close to the lower end of the detection limit of the test procedure. Such background levels are not unusual in analyses of soils for herbicide residues. The levels of diuron are, however, more finite, and an explanation of these levels in the nontreated soil sample is more difficult. Contamination of soil in the cultivated blocks may have occurred when sandy soils were blown in during the dry windy season or washed in during heavy rains. Equipment movement throughout the experimental areas may also account for some movement of herbicides in the surface soil. The fact that diuron remains in the surface of the soil profile for longer periods would allow for greater movement than bromacil which is more rapidly moved into the lower soil profile.

From the data presented, it is evident that bromacil and diuron levels are relatively low in the 0–60-cm layers of the soil types sampled. Since soil was not sampled below 60 cm, the extent of residue movement through leaching into the lower soil profile is unknown. However, the data suggest that residue levels do decrease with depth. Although soil samples were not collected yearly, the data indicate that the degree of accumulation would not lead to cumulative levels toxic to citrus at rates used in commercial practice. This statement is supported by the fact that the tree foliage has not exhibited phytotoxicity symptoms throughout the experimental period. Rather, residues are steadily dissipating through leaching and degradation.

TABLE 1. *Bromacil and diuron residue levels in two Florida soil types*

Herbicide	Rate, kg/ha	Total Applied, kg/ha	Interval Between Last Application and Sampling, Months	Soil Type	Sample Location	Sample Depth, cm	Residue: Bromacil kg/ha–15 cm (ppm)	Residue: Diuron kg/ha–15 cm (ppm)	Residue, % of: Last Application[1] Bromacil	Last Application[1] Diuron	Total Application Bromacil	Total Application Diuron
Location 1—Polk County												
Bromacil + Diuron	3.6 + 1.8	25.1 + 12.5 (5 years)	11	Astatula fine sand	Row middle	0–15	<0.09 (<0.04)	2.90 (1.30)				
						15–30	<0.09 (<0.04)	1.07 (0.48)				
					Drip line	0–15	<0.09 (<0.04)	1.70 (0.76)				
						15–30	<0.09 (<0.04)	0.51 (0.23)				
					Under canopy	0–15	0.11 (0.05)	1.16 (0.52)	2.5	72.9	0.3	10.5
						15–30	0.09 (0.04)	0.54 (0.24)				
Control						0–15	<0.09 (<0.04)	0.36 (0.16)				
Bromacil + Diuron	3.6 + 1.8	28.7 + 14.3 (6 years)	1	Astatula fine sand	Row middle	0–15	0.36 (0.16)	2.24 (1.00)				
						15–30	0.20 (0.09)	0.50 (0.22)				
					Drip line	0–15	1.28 (0.57)	2.22 (1.00)				
						15–30	0.72 (0.32)	0.60 (0.27)				
					Under canopy	0–15	2.46 (1.10)	2.91 (1.30)	26.6	80.4	3.3	10.1
						15–30	0.73 (0.33)	<0.22 (<0.10)				
Control						0–15	<0.09 (<0.04)	<0.22 (<0.10)				
Bromacil + Diuron	3.6 + 1.8	32.3 + 16.1 (7 years)	10	Astatula fine sand	Row middle	0–15	0.11 (0.05)	1.84 (0.82)				
						15–30	0.90 (0.04)	0.69 (0.31)				
						30–45	0.07 (0.03)	0.34 (0.15)				
						45–60	0.15 (0.06)	0.27 (0 12)				
					Drip line	0–15	0.07 (0.03)	1.34 (0.60)				
						15–30	0.04 (0.02)	0.54 (0.24)				
						30–45	<0.04 (<0.02)	<0.22 (<0.10)	4.4	33.6	0.5	3.7
						45–60	0.04 (0.02)	<0.22 (<0.10)				
					Under canopy	0–15	0.18 (0.08)	1.14 (0.51)				
						15–30	0.16 (0.07)	<0.22 (<0.10)				
						30–45	0.09 (0.04)	<0.22 (<0.10)				
						45–60	0.07 (0.03)	<0.22 (<0.10)				
Control						0–15	0.09 (0.04)	0.69 (0.31)				
Location 11—Hardee County												
Bromacil + Diuron	3.6 + 1.8	28.6 + 11.6 (7 years)	8	Myakka fine sand	Row middle	0–15	0.18 (0.08)	3.36 (1.50)				
						15–30	0.07 (0.03)	0.52 (0.23)				
					Drip line	0–15	0.74 (0.33)	2.69 (1.20)				
						15–30	1.46 (0.65)	0.38 (0.17)	18.2	84.6	2.3	13.1
					Under canopy	0–15	0.63 (0.28)	1.88 (0.84)				
						15–30	0.37 (0.39)	0.31 (0.14)				
Control						0–15	0.07 (0.03)	1.03 (0.46)				
Bromacil + Diuron	3.6 + 1.8	28.6 + 11.6 (7 years)	12	Myakka fine sand	Row Middle	0–15	2.24 (1.00)	2.69 (1.20)				
						15–30	0.60 (0.27)	0.29 (0.30)				
					Drip line	0–15	2.04 (0.91)	2.69 (1.20)				
						15–30	0.67 (0.30)	<0.22 (<0.10)	31.0	71.4	3.9	11.0
					Under canopy	0–15	0.85 (0.38)	1.61 (0.72)				
						15–30	0.31 (0.14)	<0.22 (<0.10)				
Control						0–15	<0.04 (<0.02)	0.65 (0.29)				

[1] Values represent an average of residue levels from all depths.

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FISH, WILDLIFE, AND ESTUARIES

Residues of Pesticides and PCBs in Estuarine Fish, 1972–76—National Pesticide Monitoring Program

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ABSTRACT

This report summarizes 1524 analyses of juvenile fish collected semiannually in 144 estuaries nationwide from July 1972 through June 1976. Pooled samples of 25 whole fish were screened for 20 common pesticides and polychlorinated biphenyls (PCBs). The three most common residues, DDT, PCBs, and dieldrin, were found in 39, 22, and 5 percent of the samples, respectively. Data indicate that estuarine pollution levels continue to decline.

Introduction

The economic and aesthetic importance of estuaries prompts many investigations to determine the causes and effects of imbalances in these sensitive ecosystems. The most comprehensive program was the monthly surveillance in 1965–72 for pesticide pollution of molluscan populations (*4*). The nationwide study identified the widespread contamination of estuarine fauna with DDT and demonstrated that DDT levels had peaked and were declining.

The persistence of DDT and other synthetic organochlorines made it desirable to continue monitoring estuarine areas, but it was necessary to reduce the analytical workload of the monitoring program. Unfortunately, residue data from molluscan populations are best understood when obtained continually. The animals purge themselves rapidly when pollution loading is intermittent (*3*).

The literature on accumulation and long storage of synthetic compounds by fish indicated that fish could be sampled less frequently than mollusks. However, little information was available on the sensitivity or selectivity of different species of fish in acquiring residues of specific pollutants or combinations of pollutants. Also, it was difficult to determine when and where migratory species acquired residues.

Sample Selection and Collection

Many species of estuary fish spend only their first year within a single estuary; other species may spend their lifetime in an estuary. Presumably, fish less than a year old would reflect pollution levels during the preceding few months at or near where they were caught. So, each estuary was monitored at 6-month intervals in the spring and fall.

The geographic extent of this program meant that comparisons of residues in different species would be questionable. Consequently, in a given estuary, the same two species of fish were collected for the duration of the program. The two species represented different food webs, e.g., a carnivore and a particle feeder. This manner of sampling made it possible to detect pollution trends over the 4-year period.

Fish were collected with trawls and beach seines in 144 primary and secondary estuaries in 19 coastal states, Puerto Rico, and the Virgin Islands. Monitoring in Alaska, Hawaii, and Mississippi was limited to one year, but in most areas, six to eight semiannual collections were made during five calendar years. The 154 species collected represent 52 of the 175 families of marine fishes of North America (*1*). Some species and estuaries were monitored only once to identify possible problem areas. More than 60 species were sampled at least three times, and 22 species were collected in the estuaries of three or more states (Tables 1, 2). About 38,000 fish were analyzed in groups which made up 1524 samples.

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TABLE 1. *Summary of estuarine fish collections, July 1972–June 1976*

Coastal Area	Number of Years Monitored	Number of Estuaries	Number of Fish Species	Number of Samples[1]
Alabama	3	3	4	13
Alaska	1	8	17	37
California	4	7	17	82
Connecticut	4	4	3	39
Delaware	4	3	11	57
Florida	3	11	22	66
Georgia[2]	4	9	15	74
Hawaii	1	8	14	22
Louisiana	2	14	14	51
Maryland	4	8	8	140
Mississippi	1	4	6	21
New York	4	3	4	46
North Carolina	4	19	28	251
Oregon	3	5	13	178
Puerto Rico[2]	3	5	14	25
Rhode Island	4	1	2	32
South Carolina[2]	4	6	5	99
Texas	4	9	8	51
Virginia	3	3	5	55
Virgin Islands[2]	2	8	19	28
Washington state	4	6	3	157
TOTAL		144	154[3]	1524

[1] Each sample consisted of 25 fish less than one year old.
[2] Some monitoring data for 1972–74 have also been published for these four coastal areas (see literature references *11, 12*).
[3] Different species, some species were collected in more than one state.

Sample Preparation

Earlier laboratory investigations indicated that analyses of 15 randomly selected fish would cover the range of individual variations in pesticide concentrations in experimentally exposed fish populations (*2*). In the present study, 50 yearling fish were collected semi-annually and analyzed in pools of 25 each. Whole fish samples were homogenized, and an aliquot was blended with a desiccant as described in the molluscan program (*4*). The prepared samples were shipped unrefrigerated to the Pesticides Monitoring Laboratory, U.S. Environmental Protection Agency, Bay St. Louis, Mississippi, for analysis.

Analytical Procedure

Desiccated samples were shaken with acetonitrile for 4 hours, and partitioned and cleaned by the Mills method (*8*); methylene chloride and hexane were used to elute the Florisil column (*9*). The extract was analyzed by flame photometric detector before Florisil cleanup to avoid possible loss of organophosphorus compounds (*6*). Polychlorinated biphenyls (PCBs) were separated from other chlorinated compounds by the silicic acid method (*7*). Instrument parameters and operating conditions used for gas chromatographic analysis and confirmation are given in Table 3. Samples were routinely screened for residues of the synthetic compounds listed in Table 4. The recovery range for organochlorines was 75–85 percent, and for organophosphates, 85–95 percent.

Results and Discussion

ΣDDT

DDT is persistent in sediments with high organic content; its presence long after its use has been terminated is not surprising. However, DDT residues found recently in fish a few months old are not so easily explained. Of the states and territories monitored, DDT was absent only from Alaska, Hawaii, and the Virgin Islands (87 samples). In 595 samples, 39 percent, DDT was detected at levels of 10+ μg/kg (Table 5). In many areas, DDT residues were consistently present in small amounts in fish only a few months old. However, these low levels probably are biologically insignificant. Some samples from California, Delaware, Florida, and New York had DDT residues in the 1000–4000-μg/kg range. DDT burdens this high could cause physiological stress and lessen reproductive capacity in fish populations (*5*). The larger residues surpass levels observed in oysters in the same estuaries in 1965–72 when DDT was still being used. The fact that the half-life of pesticide residues is much shorter in mollusks than in fish may explain this paradox.

Coastal areas are ranked in the order of frequency and magnitude of ΣDDT residues in Table 6. Not surprisingly, the 10 areas with the highest frequency of positive fish samples are essentially the same coastal areas which had the highest frequency of ΣDDT-positive molluscan samples during 1965–72. However, there was a 30 percent decline in the overall frequency of DDT-positive samples of fish compared to mollusks in the 13 states where both were monitored. This decline was not uniform; in Delaware, the frequency remained at 75 percent, and in Washington state it declined from 11 to 4 percent.

Examination of the percentage distribution of DDT and its metabolites, TDE and DDE, in residues indicates to some extent the movement of DDT in the estuarine environment in recent years (Table 7). There has been a well defined shift from the large proportion of DDT in 1972 to its absence from fish samples collected in 1976 and the concomitant increase in levels of DDE. Yet, there has been no significant change in the mean residues of ΣDDT present during the 4-year period (Table 8). This suggests that DDT is continually recycled in the food web since it occurs in juvenile fish, and, in moving along biological pathways, DDT is gradually metabolized to the more stable compound. More important, it indicates that DDT is no longer being introduced into the estuarine environment and that a pollutant can be controlled nationwide by enforcing legislation.

POLYCHLORINATED BIPHENYLS (PCBs)

PCBs were identified in 331 samples, 22 percent of the total analyzed. Residues were quantitated by comparison with standards of Aroclors 1242, 1254, and 1260. In the data tabulations, PCBs are reported as a single entity regardless of the standard used to quantitate them. Thus, residues consisting of more than one PCB are not fully identified, and reported data of the actual amounts may vary.

TABLE 2. *Geographic distribution of fish species collected three or more times, 1972–76*

Species[1]	Scientific Name	Coastal Area																			
		AL	AK	CA	CT	DE	FL	GA	HI	LA	MD	MS	NY	NC	OR	PR	RI	SC	TX	VA	VI	WA
Alewife	(*Alosa pseudoharengus*)					X					X			X						X		
American shad	(*Alosa sapidissima*)														X				X			
Atlantic croaker	(*Micropogon undulatus*)	X					X	X		X	X			X				X	X			
Atlantic menhaden	(*Brevoortia tyrannus*)					X		X			X			X								
Atlantic needlefish	(*Strongylura marina*)						X															
Atlantic silverside	(*Menidia menidia*)				X	X	X						X	X						X		
Bay anchovy	(*Anchoa mitchelli*)	X		X		X	X	X		X	X	X		X					X	X		
Blackcheek tonguefish	(*Symphurus plagiusa*)							X				X		X								
Blueback herring	(*Alosa aestivalis*)													X								
Bluefish	(*Pomatomus saltatrix*)				X									X								
Blenny	(*Cirripectes variolus*)								X													
Buffalo sculpin	(*Enophrys bison*)														X							
California halibut	(*Paralichthys californicus*)			X																		
Capelin	(*Mallotus villosus*)		X																			
Cunner	(*Tautogolabrus adspersus*)				X																	
English sole	(*Parophrys vetulus*)			X											X							X
Eulachon	(*Thaleichthys pacificus*)		X																			
Flathead sole	(*Hippoglossoides elassodon*)		X																			
Gafftopsail catfish	(*Bagre marinus*)									X												
Gizzard shad	(*Dorosoma cepedianum*)									X				X								
Great barracuda	(*Sphyraena barracuda*)															X					X	
Gulf killifish	(*Fundulus grandis*)						X					X										
Gulf menhaden	(*Brevoortia patronus*)									X		X							X			
Hogchoker	(*Trinectes maculatus*)									X	X			X								
Iao	(*Pranesus insularum*)								X													
Little skate	(*Raja erinacea*)																X					
Lizardfish	(*Saurida gracilis*)								X													
Longfin smelt	(*Spirinchus thaleichthys*)			X											X							
Longnose killifish	(*Fundulus similis*)						X												X			
Pacific halibut	(*Hippoglossus stenolepsis*)			X																		
Pacific herring	(*Clupea harengus pallasi*)		X												X							
Pacific sanddab	(*Citharichthys sordidus*)			X																		
Pacific sandlance	(*Ammodytes hexapterus*)		X												X							
Pacific staghorn sculpin	(*Leptocottus armatus*)														X							X
Pacific tomcod	(*Microgadus proximus*)														X							
Pinfish	(*Lagodon rhomboides*)						X							X					X			
Queenfish	(*Seriphus politus*)			X																		
Queen triggerfish	(*Balistes vetula*)																				X	
Rockhind	(*Epinephelus adscensionis*)																				X	
Sand seatrout	(*Cynoscion arenarius*)									X				X								
Sand sole	(*Psettichthys melanostictus*)														X							
Sea catfish	(*Arius felis*)							X		X												
Shiner perch	(*Cymatogaster aggregata*)			X											X							
Silver jenny	(*Eucinostomus gula*)						X									X			X			
Silver perch	(*Bairdiella chrysura*)						X							X				X				
Southern kingfish	(*Menticirrhus americanus*)						X	X						X								
Speckled sanddab	(*Citharichthys stigmaeus*)														X							
Spot	(*Leiostomus xanthurus*)	X				X	X	X		X	X			X				X				
Spotfin mojarra	(*Eucinostomus argenteus*)						X															
Spotted sand bass	(*Paralabrax maculatofasciatus*)			X																		
Stardrum	(*Stellifer lanceolatus*)							X		X				X				X				
Starry flounder	(*Platichthys stellatus*)		X												X							X
Striped anchovy	(*Anchoa hepsetus*)													X								
Striped bass	(*Morone saxatilis*)			X							X									X		
Striped mullet	(*Mugil cephalus*)	X					X		X			X		X		X			X		X	
Thread herring	(*Opisthonema oglinum*)						X							X								
Tidewater silverside	(*Menidia beryllina*)						X															

TABLE 2 (cont'd.). *Geographic distribution of fish species collected three or more times, 1972–76*

Species[1]	Scientific Name	Coastal Area																					
		AL	AK	CA	CT	DE	FL	GA	HI	LA	MD	MS	NY	NC	OR	PR	RI	SC	TX	VA	VI	WA
Walleye pollock	(*Theragra chalcogramma*)		X																			
Weakfish	(*Cynoscion regalis*)					X	X	X										X	X			
White croaker	(*Genyonemus lineatus*)			X										X		X					X	
White mullet	(*Mugil curema*)										X			X						X		
White perch	(*Morone americana*)					X								X								
Windowpane flounder	(*Scophthalmus aquosus*)					X																
Winter flounder	(*Pseudopleuronectes americanus*)			X		X							X				X					
Yellowfin sole	(*Limanda aspera*)		X																			

[1]See Literature Cited, ref. *1*.

PCBs were not found in samples from Alaska and Mississippi. In 11 states, Puerto Rico, and the Virgin Islands, Aroclor 1254 was the only standard used. In the remaining six coastal areas, standards of Aroclors 1242 and 1260 were occasionally required as well for the quantitation of residues (Table 5). The annual incidence of PCB-positive samples is summarized in Table 8. Data indicate a gradual decline in both the maximum residues observed in most years and the average concentration of the PCB residues. The changes were expected in view of the general curtailment in production and use of the compounds. Their chemical persistence suggests, however, that they will continue to contaminate the environment for several years.

Only at one station each in Delaware and Washington state did PCB residues frequently exceed 1000 μg/kg. Such data do not indicate high PCB levels in the ambient water since residues are cumulative and fish may have had up to one year of exposure. However, controlled experiments show that PCB concentrations as low as 1.0 μg/kg are sufficient to cause fin rot and increased mortality in chronically exposed fish (*10*).

Coastal areas are ranked in order of the frequency and magnitude of PCB residues in estuarine fish (Table 9). These residues were found in 19 of the 21 areas monitored, but in only four states were they present in more than half the samples. In contrast, DDT residues were found in 18 areas and were present in more than half the samples from nine states. This indicates a much broader contamination of the environment with DDT than with PCBs.

The incidence of PCB residues in fish cannot be compared with the much lower frequency observed in mollusks in 1970–72. PCBs are an industrial pollutant and are not usually found where shellfish are harvested.

DIELDRIN

Residues of dieldrin were detected in 74 samples, 5 percent of the total samples, ranging from 10 μg/kg to 145 μg/kg. Positive samples were collected in some of the estuaries of 12 states and the Virgin Islands (Table 10). About half the positive samples were collected in secondary estuaries in the Maryland section of Chesapeake Bay. Samples from this area contained dieldrin in 1972–74, but not in 1975. Dieldrin was found in a variety of fish species, but its presence had no apparent correlation with their different feeding patterns. In 1972–74, dieldrin was found in about 7 percent of the fish samples; but in 1975–76, less than 1 percent of the samples contained detectable levels (Table 8). During the 1965–72 monitoring of mollusks, dieldrin was found in 15 percent of the samples at levels approximately double those detected in the juvenile fish.

TABLE 3. *Operating parameters for analyzing estuarine fish for pesticide and PCB residues—1972–76*

Detector	Column	Temperatures, °C			Carrier Gas, Flow Rate
		Column	Detector	Injector	
Electron-capture	Glass, 1.8 m long × 4 mm ID, packed with 3 percent DC-200 on 80–100-mesh Supelcoport	188	300	250	Argon/methane 50 ml/minute
Electron-capture	Glass, 1.8 m long × 2 mm ID, packed with a mixture of 1.5 percent OV-17 and 1.95 percent OV-210 on 80–100-mesh Supelcoport	193	200	230	Nitrogen 30 ml/minute
Electron-capture	Glass, 1.8 m long × 2 mm ID, packed with 5 percent OV-210 on 80–100-mesh Supelcoport	173	200	230	Nitrogen 30 ml/minute
Flame photometric	Glass, 1.8 m long × 4 mm ID, packed with 3 percent OV-101 on 80–100-mesh Chromosorb W-HP	177	184	230	Nitrogen 50 ml/minute

TABLE 4. *Compounds detected by gas chromatographic analysis of estuarine fish tissue—1972–76*[1]

Organochlorine	Organophosphate
Aldrin	Azinphosmethyl
Chlordane	Carbophenothion
DDT	DEF
Dieldrin	Demeton
Endosulfan	Diazinon
Heptachlor	Ethion
Lindane	Malathion
Methoxychlor	Parathion
Mirex	Phorate
PCBs	
Toxaphene	
Trifluralin	

NOTE: See appendix for chemical names of compounds.
[1]Lower detection limit is 10 μg/kg for all compounds except the following: endosulfan, 20 μg/kg; methoxychlor and ethion, 30 μg/kg; mirex, PCBs, toxaphene, carbophenothion, and DEF, 50 μg/kg.

PESTICIDES OCCASIONALLY DETECTED

Despite the fact that all samples were routinely screened for 21 synthetic hydrocarbons and their oxygen analogs, few were detected. DDT and its metabolites, dieldrin, and PCBs were the most common residues. Only six other pesticides were found in measurable amounts (Table 11). These were detected in 48 samples or about 3 percent of the total. A majority of these residues occurred in fish from the upper end of Chesapeake Bay and along the Texas coast. The insecticide endrin and the herbicide Dacthal (DCPA) were also identified in fish from a heavily farmed area in the Texas Rio Grande river basin. This area was monitored monthly and the data will be presented in a separate publication.

DATA INTERPRETATION

The data are organized on a seasonal and geographic basis, i.e., by state boundaries, in an effort to make the large group of heterogenous samples more manageable. Unfortunately, some details of localized pollution patterns are lost in the process. For example, data from only one river basin in Rhode Island can be compared with data from 3–19 river basins in other states. Or, as in Washington state, data from one polluted estuary were averaged with five other relatively clean areas in the state. In Table 9, the frequency of PCB residues is shown as 17 percent in Washington. Actually, all 27 samples from the Duamish River were contaminated, but none of the 128 samples from the other five estuaries contained PCB residues during the 4-year period.

PCB residue data from the Duamish River samples illustrate the importance of sampling continuity to determine localized pollution patterns and trends. The Pacific staghorn sculpin and English sole were both collected seven times in the 4-year period. Quantitation of the PCB residues required three different standards (Table 12). The residues were probably mixtures of two or more PCBs, but the data indicate both a shift in the kind of pollution and a decline in pollution levels.

There must always be some ambiguity in the comparison of residue data from different species in the absence of controlled experiments on their ability to accumulate pesticides. In the Duamish River samples, the consistently higher residue levels in English sole probably were due to a difference in age rather than in species. Sole populations sampled were usually about 6 months older than the sculpins.

Comparisons of residue data in a single fish species distributed over a wide geographic range permit valid judgments of regional pollution differences. The bay anchovy was the most widely distributed species in the present program. It was collected in 37 estuaries in the 11 states from Delaware to Texas over a 3-year period. Samples from three estuaries in Georgia and three in Louisiana contained no detectable DDT or PCBs. In contrast, 42 bay anchovy samples collected in Delaware and Chesapeake Bay during this 3-year period contained residues of DDT (10–467 μg/kg, mean 77) and PCBs (90–996 μg/kg, mean 340). On the basis of such data, it is possible to identify regional pollution patterns when juvenile fish of the same species are monitored periodically.

In general, residue data from all the estuaries in a single state were strongly skewed because only one or two estuaries were highly polluted. In Washington state, less

TABLE 5. *Residues of ΣDDT and PCBs in whole-body samples of juvenile estuarine fish, 1972–76*

Coastal Area, Year	Number of Samples	Residues, μg/kg Wet Weight					
		ΣDDT			PCBs		
		Number Positive	Maximum Residue	Geometric $\bar{X}$ of Positive Samples	Number Positive	Maximum Residue	Geometric $\bar{X}$ of Positive Samples
Alabama							
1972	2	2	82	67	0		
1973	2	1	17	17	0		
1975	3	2	35	20	0		
1976	6	4	49	35	3	174	163
Alaska							
1972	7	0			0		
1973	30	0			0		
California							
1972	6	4	213	69	0		
1973	21	19	667	75	2	270	229
1974	17	15	1422	69	5	512	224
1975	18	15	1349	79	6	432	210
1976	20	18	2588	95	12	400	254
Connecticut							
1972	4	3	63	43	4	592	313
1973	7	1	68	68	7	678	321
1974	15	4	43	26	14	1065	406
1975	5	4	97	25	5	497	252
1976	8	0			4	289	172
Delaware							
1972	6	6	1425	220	4	4504	1469
1973	12	12	636	85	7	2671	802
1974	14	13	1194	109	6	823	258
1975	9	8	1146	181	8	1566	720
1976	16	4	1015	471	4	1258	649
Florida							
1972	25	8	170	25	0		
1973	15	4	18	13	0		
1974	19	17	1640	36	16	614	62
1975	7	5	23	21	1	104	104
Georgia							
1972	12	3	65	26	1	508	508
1973	17	2	14	13	0		
1974	10	1	32	32	1	137	137
1975	18	1	16	16	0		
1976	17	0			0		
Hawaii							
1972	8	0			3	305	244
1973	14	0			0		
Louisiana							
1975	24	5	108	52	1	256	256
1976	27	1	23	23	0		
Maryland							
1972	22	14	184	55	14	788	351
1973	45	26	345	73	12	1046	318
1974	45	35	694	51	16	878	287
1975	28	6	714	251	9	940	267
Mississippi							
1972	5	4	16	14			
1973	16	2	159	135			
New York							
1972	6	3	174	71	5	310	231
1973	10	5	115	49	2	235	149
1974	12	10	106	34	10	301	165
1975	6	5	4082	188	2	694	471
1976	12	10	104	39	10	447	295
North Carolina							
1972	30	29	140	43	15	786	258
1973	80	34	357	39	1	120	120
1974	70	26	322	39	3	174	131
1975	41	18	78	24	1	173	173
1976	30	18	140	33	2	538	527
Oregon							
1973	77	21	125	29	4	277	130
1974	66	22	221	32	7	247	179
1975	35	3	12	11	6	288	236

(Continued next page)

TABLE 5 (cont'd.). *Residues of ΣDDT and PCBs in whole-body samples of juvenile estuarine fish, 1972–76*

Coastal Area, Year	Number of Samples	Residues, μg/kg Wet Weight					
		ΣDDT			PCBs		
		Number Positive	Maximum Residue	Geometric $\bar{X}$ of Positive Samples	Number Positive	Maximum Residue	Geometric $\bar{X}$ of Positive Samples
Puerto Rico							
1972	4	2	157	100	2	201	181
1973	8	1	172	172	2	416	316
1974	4	0			4	579	238
1976	9	5	86	28	0		
Rhode Island							
1972	4	0					
1973	8	0			4	477	451
1974	8	5	78	24	4	797	464
1975	8	4	20	17	8	524	231
1976	4	0			4	241	230
South Carolina					4	356	275
1972	12	7	60	29			
1973	25	13	33	16	1	182	182
1974	21	6	29	19	0		
1975	22	2	12	11	0		
1976	19	0			0		
Texas					0		
1972	7	5	52	38			
1973	9	5	188	82	3	267	136
1974	11	8	223	65	0		
1975	18	12	59	23	4	240	95
1976	6	4	70	37	4	265	150
Virginia					1	157	157
1973	26	20	124	39			
1974	11	10	60	39	8	438	214
1975	18	7	821	115	8	456	254
Virgin Islands					5	2549	850
1972	6	0					
1973	13	0			3	166	142
1974	9	0			0		
Washington state					2	809	615
1972	21	0					
1973	48	1	25	25	4	4903	2552
1974	48	1	11	11	8	3363	1577
1975	24	0			7	2028	1515
1976	16	4	38	32	4	2639	2057
					4	900	668

NOTE: Samples from Alaska contained no PCBs. Aroclor 1254 was used as the standard in all other coastal areas with the following occasional additions: Aroclor 1260: California, Connecticut, Delaware, Maryland, North Carolina, and Washington state; Aroclor 1242: Delaware, Maryland, North Carolina, and Washington state.

TABLE 6. *Frequency and average concentration of ΣDDT residues in juvenile estuarine fish by coastal area, 1972–76*

Coastal Area	Frequency of Residues, %	Coastal Area	Average Concentration, μg/kg[1]
California	87	Delaware	213
Delaware	75	Maryland	108
New York	72	Puerto Rico	100
Alabama	69	California	77
Virginia	67	New York	76
Texas	67	Mississippi	75
Maryland	58	Virginia	64
Florida	52	Texas	49
North Carolina	48	Connecticut	41
Puerto Rico	32	Louisiana	38
Connecticut	31	North Carolina	36
Mississippi	29	Alabama	35
South Carolina	29	Florida	24
Rhode Island	28	Oregon	24
Oregon	26	Washington state	23
Louisiana	12	Georgia	22
Georgia	10	Rhode Island	21
Washington state	4	South Carolina	19
Alaska	0		
Hawaii	0		
Virgin Islands	0		

NOTE: Comparisons are limited in that the number of samples, number of sampling stations, period (years) of sampling, and species of fish differ for each coastal area.

[1] Arithmetic average of geometric means of positive samples in all collection years.

than 4 percent of the samples collected in 5 years contained measurable residues of DDT. The geometric means of the positive samples, along with the maximum residue detected and the number of positive samples, is the best summary of actual pollution levels. Conversely, the geometric means of the residue data from year to year in a given state were normally distributed, and the arithmetic means were used to compare pollution levels in different geographic areas (Tables 6, 8, 9). Plans are under way to store sample and analytical data in a computer data bank to provide more precise data analyses in studies of localized pollution problems.

TABLE 7. *Percentage distribution of metabolites in ΣDDT residues in juvenile estuarine fish by coastal area, 1972–76*

Year	Number of Positive Samples	Distribution, %		
		DDT	TDE	DDE
1972	90	23	37	40
1973	167	12	30	58
1974	173	5	36	59
1975	97	1	21	78
1976	68	0	14	86

TABLE 8. *Annual incidence of ΣDDT, PCB, and dieldrin residues in juvenile whole fish samples, 1972–76*

Year	No. of Samples	Residues, μg/kg								
		DDT			PCBs			Dieldrin		
		% Positive	Maximum Residue	Average Residue[1]	% Positive	Maximum Residue	Average Residue[1]	% Positive	Maximum Residue	Geometric $\bar{X}$
1972	187	48	1425	62	34	4903	540	7	140	21
1973	483	34	667	58	12	3363	429	6	140	30
1974	380	46	1640	42	29	2028	320	8	145	12
1975	284	34	4082	69	20	2639	460	1	15	14
1976	190	36	2588	88	22	1258	351	0	—	—

[1] Arithmetic average of the geometric means of positive samples from each coastal area.

Conclusions

Juvenile fish are satisfactory tools for gauging pesticide pollution trends in estuaries provided at least 25 individuals, 6–12 months old, of the same species are sampled annually at a specific location. Analyses of the same species of fish at different geographic locations permit valid comparisons of pollution levels.

Existing ΣDDT residues are the result of biotic recycling, and probably little, if any, DDT has been introduced recently into the estuarine systems monitored in this study.

The magnitude and frequency of biotic residues of DDT, dieldrin, endrin, and toxaphene declined substantially between 1965–70 and 1972–76.

Data from this study warrant annual monitoring of juvenile fish in the nation's estuaries.

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TABLE 9. *Frequency and average concentration of PCB residues in juvenile estuarine fish by coastal area, 1972–76*

Coastal Area	Frequency of Residues, %	Coastal Area	Average Concentration, μg kg[1]
Connecticut	87	Washington state	1674
Rhode Island	75	Delaware	780
New York	63	Virginia	439
Delaware	51	Virgin Islands	379
Virginia	38	Rhode Island	330
Maryland	36	Connecticut	323
Puerto Rico	32	Georgia	323
California	31	Maryland	306
Florida	26	New York	262
Texas	24	Louisiana	256
Alabama	23	Puerto Rico	245
Virgin Islands	18	Hawaii	244
Washington state	17	North Carolina	242
Hawaii	14	California	229
Oregon	10	Oregon	182
North Carolina	9	South Carolina	182
Georgia	3	Alabama	163
Louisiana	2	Texas	135
South Carolina	1	Florida	83
Alaska	0		
Mississippi	0		

NOTE: Comparisons are limited in that the number of samples, number of sampling stations, periods (years) of sampling, and species of fish differ for each coastal area.

[1] Arithmetic average of geometric means of positive samples in all collection years.

TABLE 10. *Geographic incidence of dieldrin residues in juvenile estuarine fish, 1972–76*

Coastal Area	Number of Samples	Number Positive	Mean Residue, μg/kg
California	82	2	34
Connecticut	39	3	15
Delaware	57	2	59
Florida	66	12	10
Georgia	74	2	60
Louisiana	51	1	15
Maryland	140	35	30
Mississippi	21	2	17
New York	46	2	24
North Carolina	251	4	20
Texas	51	6	20
Virginia	55	2	10
Virgin Islands	28	1	10

TABLE 11. *Pesticide residues occasionally detected in juvenile estuarine fish, 1972–76*

State	Chlordane	Heptachlor Epoxide	Toxaphene	Ethyl Parathion	Methyl Parathion	Carbophenothion	Ethion
Alabama	1–13–133						
Connecticut				1–39–10			
Hawaii	6–22–290						
Louisiana			1–51–504				
Maryland	22–140–118	3–140–15				2–140–109	19–140–169
Mississippi			2–21–388				
New York	2–46–207						
North Carolina				1–251–12			
Texas			3–51–75	3–51–75	2–51–47	1–51–103	1–51–83

NOTE: Data in columns represent incidence, number of samples, and mean residue, μg/kg, respectively.

lina Wildlife and Marine Resources Department, M. H. Shealy, Jr.; Texas Parks and Wildlife Department, R. Childress; Virginia Institute of Marine Science, R. J. Huggett; State of Washington Department of Fisheries, B. Pattie; and University of Washington Fisheries Research Institute. B. Miller.

TABLE 12. *Trends in PCB residues in English sole and Pacific staghorn sculpin, Duamish River, Washington state, fall 1972–spring 1976*

Date	Fish Species	Most Similar Aroclor Standard[1]		
		1254	1260	1242
Fall 1972	E	3346		
	P	2202		
Spring 1973	E	2111		
	P	2065		
Fall 1973	E	1683		
	P	1129		
Spring 1974	E		1927	
	P		1477	
Fall 1974	E		1733	
	P		825[2]	
Spring 1975	E		2541	
	P		1832	
Spring 1976	E		888	1241
	P		506	492

NOTE: E = English sole, P = Pacific staghorn sculpin.
[1]Data represent average of two sample pools of 25 fish each (wet weight, μg/kg).
[2]Only one sample.

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Residues of Organochlorine Insecticides and Polychlorinated Biphenyls in Fish from Lakes Huron and Superior, Canada—1968–76 [1]

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ABSTRACT

Five species of fish from Lake Superior and 12 species from Lake Huron were analyzed for organochlorine pesticides and polychlorinated biphenyls (PCBs) between 1968 and 1975. Mean residues of ΣDDT peaked at 1.72 ppm and 7.60 ppm in lake trout (Salvelinus namaycush) *from Lakes Superior and Huron, respectively. By 1975, the mean level of ΣDDT had decreased in lake trout and was highest in bloaters* (Coregonus hoyi) *from both lakes: 1.06 ppm and 1.87 ppm, respectively. Dieldrin levels in fish from Lake Superior changed little over the same period. However, in 1969–70, dieldrin levels in fish from Lake Huron exceeded the 0.3 ppm tolerance level set by Health and Welfare Canada or the Food and Drug Administration, U.S. Department of Health, Education, and Welfare in 5 percent of lake whitefish* (Coregonus clupeaformis) *and 10 percent of bloaters. By 1975, 50 percent of bloaters caught in Georgian Bay and North Channel had dieldrin levels above 0.3 ppm. PCB residues declined in lake trout and lake whitefish caught in Lake Superior between 1971 and 1975, but increased slightly in bloaters and white sucker* (Catostomus commersoni). *Mean PCB residues in bloaters caught in Lake Huron in 1969–71 and 1975–76, and splake* (Salvelinus fontinalis *and* S. namaycush) *and cisco* (Coregonus artedii) *caught in 1975 exceeded the 2 ppm tolerance level.*

Introduction

The Great Lakes are surrounded by land that is highly developed for urban, industrial, agricultural, and recreational activities. Since outflow of the Great Lakes is limited, chemical discharges into the lakes are very persistent. For the past decade organochlorines have been identified as a serious contaminant in fish, resulting in long-range detrimental effects to private and commercial fishing.

Organochlorine insecticides and polychlorinated biphenyls (PCBs) have been identified in fish caught in Lakes Huron and Superior. Reinhert reported residues of 0.2–7.4 ppm ΣDDT and 0.01–0.05 ppm dieldrin in several species of fish caught in Lake Superior in 1967–68 *(7)*. Reinke et al. reported that two fish species caught in 1970 from the same lake had mean residues of 0.2 ppm and 1.3 ppm ΣDDT and 0.06 ppm dieldrin *(9)*. Four species, also caught in Lake Superior in 1974–75, cited by the Upper Great Lakes Reference Group, contained mean residues of 0.2–4.4 ppm ΣDDT and 0.01–0.15 ppm dieldrin *(11)*. Residues of chlordane, lindane, and PCBs were also reported in these four species.

Reinhert found mean residues of 0.8–6.9 ppm ΣDDT and 0.02–0.11 ppm dieldrin in nine species of fish from Lake Huron in 1967–68 *(7)*. Reinke et al. reported mean residues of 0.5–16.4 ppm ΣDDT and 0.01–0.31 ppm dieldrin in the same major fish species in Lake Huron in 1970 *(9)*. The Upper Great Lakes Reference Group cited considerably lower residues of ΣDDT in three fish species caught in 1974–75 *(11)*, but levels of dieldrin, lindane, chlordane, and PCBs were similar to those found in other studies.

Studies on the distribution of organochlorines in water, sediment, and seston in Lakes Superior and Huron reveal that these compounds are widespread in the Great Lakes ecosystem *(3)*. Miles and Harris reported that the Muskoka River discharged large amounts of ΣDDT to Georgian Bay *(6)*. Peak discharges of 5.4 kg/week occurred in May 1971, but the quantity declined rapidly from May to October, averaging 0.9 kg ΣDDT/week. Frank et al. found that fish in the Muskoka Lake–Muskoka River system contained some of the highest residue levels found in fish from inland lakes of Ontario *(2)*. Fourteen species had mean residues of 0.22–22.4 ppm ΣDDT; sediments in this lake–river system contained ΣDDT residues as high as 2.9 ppm.

The present study, begun in 1968, was originally intended to identify and measure organochlorine residues

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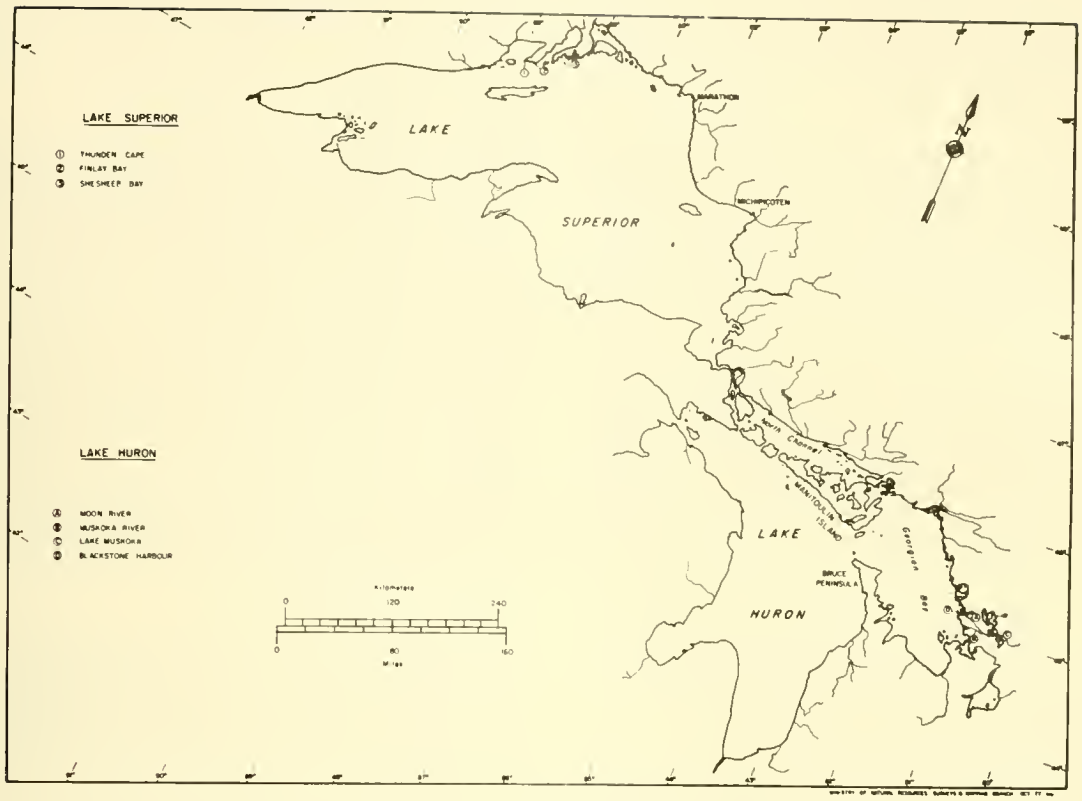

FIGURE 1. *Map of Lakes Huron and Superior showing fish collection areas*

in fish from the Great Lakes. However, it was broadened following restrictions on the use of aldrin, dieldrin, and heptachlor in Canada in 1969, DDT in 1970, and the voluntary restrictions on the use of PCBs in 1971 within the Province of Ontario. Authors wished to determine whether these use restrictions were significantly reflected in organochlorine residues in fish from Lakes Huron and Superior.

Methods and Materials

FIELD COLLECTION

Fifteen species (843 fish) were caught by net, line, or trap between 1968 and 1976 from Lakes Huron and Superior; many of the larger fish were obtained from commercial catches. Five species (115 fish) were caught in the Canadian waters of eastern Lake Superior between Michipicoten and the entrance to the North Channel (Figure 1). Between 1968 and 1976, 14 species (728 fish) were caught in Lake Huron. Of these, 481 fish of 12 species were from the Canadian waters of Lake Huron, 142 fish of five species were from Georgian Bay, and 105 fish of five species were from the North Channel. Bloaters *(Coregonus hoyi)*, coho salmon *(Oncorhynchus kisutch)*, and walleye *(Stizostedion vitreum vitreum)* were caught in southern Lake Huron, walleye caught in Georgian Bay at the mouth of the Moon River, and rainbow trout *(Salmo gairdneri)* and lake trout *(Salvelinus namaycush)* came from the south shore of Georgian Bay. Other species were caught between the Bruce Peninsula and Manitoulin Island.

Fish species were identified and named according to the nomenclature of the American Fisheries Society *(1)*.

SAMPLE PREPARATION

Fish were measured, weighed, and where possible, the sex was determined. Heads and viscera were removed and the remainder of the fish was macerated in a Hobart meat grinder. A 150–200-g subsample was stored in a sealed glass jar at −20° C; storage time varied from a few days to four months. Individual fish were analyzed when the sample size was not limiting. Alewife, shiners, smelt, and other small fish were prepared as composites of similar sized fish. They were weighed and measured individually before being ground.

ANALYTICAL PROCEDURE

Ten grams of tissue homogenate was ground with 100 g anhydrous sodium sulfate and 25 g Ottawa sand. The mixture was extracted with 300 ml hexane for 7 hours in a Soxhlet extractor. Solvent was evaporated by rotary vacuum and the percentage fat was determined gravimetrically.

A one-step Florisil column cleanup method described by Langlois et al. *(5)* was used to isolate organochlorine insecticides and PCBs. A maximum of 1 g fat was mixed with conditioned Florisil and placed above another layer of Florisil. The column was eluted with a 300-ml 1:4 mixture of dichloromethane–hexane. Solvent was evaporated by rotary vacuum.

PCBs, Hexachlorobenzene (HCB), and organochlorine insecticides were separated on a charcoal column according to the method described by Holdrinet *(4)*. Analyses were performed with a Tracor Model 550 gas-liquid chromatograph (GLC). Instrument parameters and operating conditions follow.

Detector:	^{63}Ni
Column:	15 cm × 0.64 cm OD glass, packed with a mixture of 4 percent SE-30 and 6 percent QF-1 on 80–100-mesh Chromosorb W
Temperature:	180°C
Carrier gas:	nitrogen flowing at 60 ml/minute
Injection volume:	5 µl was equivalent to 1 ng fat sample

Two-dimensional thin-layer chromatography was used on random samples for confirmation. Samples were removed, redissolved, and re-injected into the GLC column.

Recoveries were checked periodically by fortification of tissue homogenates prior to extraction. Average recoveries were:

Residue	%	Residue	%
o,p'-DDT	91	Dieldrin	89
p,p'-DDT	89	*cis*-Chlordane	98
p,p'-TDE	94	*trans*-Chlordane	90
p,p'-DDE	96	PCBs	85–90

The data were not corrected for recoveries. Detection limits were 0.005 ppm for organochlorines and 0.05 ppm for PCBs. PCBs were identified by comparing them with mixtures of Aroclors 1254 and 1260 and checking for a resemblance to peaks VII, VIII, and X on sample chromatograms according to Reynolds *(10)*.

Analysis was begun in 1968 when the known main contaminants in fish were *p,p'*-DDT and its analogs plus dieldrin and heptachlor epoxide; PCB values prior to 1970 were estimated. With the introduction of a column fractionation technique in 1970 for the separation of PCBs from organochlorine insecticides, the measurement of PCB residues became more precise. Analysis for HCB was included in 1973 but was discontinued because of the low level and incidence of HCB found in the samples. The analysis and confirmation for *cis*- and *trans*-chlordane was refined in 1975; analyses for mirex and oxychlordane were introduced in 1976.

Results

LAKE SUPERIOR

ΣDDT—None of the five fish species caught in Lake Superior contained annual mean residues in excess of the 5 ppm tolerance level established by Health and Welfare Canada or the Food and Drug Administration, U.S. Department of Health, Education, and Welfare. The highest mean residue of 2.7 ppm was found in lake trout caught in 1968. However, of 18 lake trout analyzed, three contained residues of ΣDDT that exceeded 5 ppm (Table 1): a 1544-g fish caught in Shesheep Bay contained 14.1 ppm; a 2906-g fish caught off Thunder Cape contained 7.9 ppm; and a 3314-g fish caught in Finlay Bay contained 5.2 ppm (Figure 1). Lake trout caught in 1971 and bloaters caught in 1971 and 1975 contained the second highest mean ΣDDT residues of 1.16 ppm and 1.06 ppm, respectively, but no individuals exceeded the tolerance level.

Residues of ΣDDT declined in both lake trout and lake whitefish *(Coregonus clupeaformis)* between 1971 and 1975, but no trend was apparent in either bloater or white sucker *(Coregonus commersoni)*. The ratio of DDE plus TDE to ΣDDT increased in lake trout and lake whitefish from 1971 to 1975, indicating a metabolic breakdown of *o,p'*- and *p,p'*-DDT; this was not so apparent in bloaters and white sucker (Table 2). The decline is more evident in lake trout when similar weight classes are compared (Table 3). In spite of higher fat content in fish caught in 1975, ΣDDT is only a fraction of the residue found in 1968–70.

Dieldrin—No fish species contained mean residues that exceeded 0.08 ppm dieldrin, and no individual fish contained residues which exceeded the 0.3 ppm guideline set by FDA. The highest level of dieldrin found in an individual fish was 0.26 ppm in a lake trout caught in 1968. In general, levels of dieldrin were low, but the rate of disappearance of dieldrin since 1971 also has been slow. On the basis of a ΣDDT/dieldrin ratio, ΣDDT declined more rapidly than dieldrin between 1971 and 1975 (Table 2). Lake trout exhibited a decline in the ratio between 1968 and 1975 of 91 to 5. The ratio of PCBs to dieldrin changed little between 1971 and 1975. This was borne out when similar weight classes of lake trout were compared (Table 3).

PCBs—None of the five fish species caught in Lake Superior contained mean residues of PCBs greater than the 2 ppm tolerance level set by Health and Welfare Canada (Table 1). However, two individual trout caught

TABLE 1. *Organochlorine residues in five fish species caught in the Canadian waters of eastern Lake Superior, 1969–75*

Species	Year	No. of Analyses	Mean and Range Weight, g	Mean and Range Fat, %	Mean Content and Range of Contaminants in Fish Puree, ppm[1,2] DDE	TDE	DDT	ΣDDT	Dieldrin	PCBs
Catostomidae										
White sucker	1971	5	1102	2.2	0.08	0.01	0.04	0.13	0.01	0.2
			988–1202	0.9–5.0	<0.01–0.15	<0.01–0.02	<0.01–0.07	0.01–0.24		<0.1–0.5
	1975	8	946	3.1	0.14	0.01	0.05	0.20	0.02	0.3
			696–1154	0.7–7.1	0.03–0.46	<0.01–0.03	<0.01–0.15	0.08–0.59	<0.01–0.06	0.1–0.7
Esocidae										
Northern pike	1971	5	2044	1.2	0.23	0.03	0.14	0.40	<0.01	0.3
			1474–2752	0.8–1.8	0.08–0.48	0.01–0.07	0.02–0.41	0.11–0.96		0.1–0.6
Salmonidae										
Bloater	1971	4(19)[3]	149	9.7	0.68	0.07	0.41	1.16	0.02	0.6
			145–175	9.4–10.0	0.56–0.75	0.06–0.08	0.34–0.45	0.96–1.36	0.01–0.06	0.5–0.7
	1975	10	169	10.2	0.52	0.07	0.47	1.06	0.04	1.0
			112–268	3.1–18.7	0.07–1.76	0.02–0.16	0.12–1.39	0.22–3.23	0.01–0.09	0.3–3.7
Lake trout	1968	18	2016	8.0	1.44	0.24	1.04	2.72	0.08	0.7
			455–5506	1.3–14.7	0.16–7.11	0.01–1.32	0.02–5.68	0.27–14.1	0.01–0.26	<0.1–2.0
	1969	20	734	6.4	0.43	0.12	0.43	0.98	0.03	0.3
			409–1700	1.7–14.4	0.20–0.75	0.04–0.20	0.19–0.77	0.43–1.69	<0.01–0.05	0.1–0.6
	1971	5	1901	17.4	0.98	0.09	0.65	1.72	0.03	1.8
			1572–2728	15.7–22.1	0.59–1.25	0.06–0.11	0.38–0.82	1.03–2.18	0.02–0.05	1.1–2.3
	1975	10	1121	20.7	0.11	0.01	0.05	0.17	0.04	0.4
			555–1432	14.7–29.4	0.09–0.16	<0.01–0.03	0.02–0.09	0.10–0.24	0.03–0.05	0.3–0.6
Lake whitefish	1971	5	959	12.0	0.35	0.04	0.35	0.74	0.04	0.8
			895–1060	8.5–14.2	0.29–0.45	0.03–0.05	0.30–0.43	0.63–0.93	0.03–0.05	<0.1–1.0
	1975	10	1135	10.8	0.16	0.02	0.06	0.24	0.07	0.3
			766–1400	6.2–12.2	0.09–0.29	0.01–0.03	<0.01–0.16	0.12–0.48	0.04–0.11	0.1–0.7

[1] In 1975 traces (0.004 ppm) of *cis*- and *trans*-chlordane were detected in some bloater, white sucker, lake trout, and lake whitefish.
[2] <0.01 ppm represents a trace of contaminant above the level of detection (0.001 ppm) but below 0.010 ppm.
[3] Composite of 19 fish.

off Grass Cap Point in 1971 had residues of 2.2 ppm and 2.3 ppm PCBs and two bloaters caught commercially in 1975 had residues of 2.1 ppm and 3.7 ppm. Mean residues for lake trout in 1971 and bloaters in 1975 were 1.8 ppm and 1.0 ppm, respectively.

TABLE 2. *Ratios of organochlorine contaminants in four species of fish caught in Lake Superior, Lake Huron, and Georgian Bay, 1968–76*

Species	Year	DDE+TDE / ΣDDT	ΣDDT / PCBs	ΣDDT / Dieldrin	PCBs / Dieldrin
Lake Superior					
Bloater	1971	0.65	2.0	50	30
	1975	0.56	1.1	30	25
White sucker	1971	0.69	0.5	21	20
	1975	0.75	0.8	10	15
Lake trout	1968	0.62	3.9	91	9
	1969	0.56	3.3	33	10
	1971	0.62	0.9	50	60
	1975	0.70	0.4	5	10
Lake whitefish	1971	0.53	0.9	19	20
	1975	0.75	0.8	3	4
Lake Huron (Main Lake)					
Bloater	1969	0.74	3.5	69	20
	1970	0.52	1.8	29	16
	1971	0.64	2.1	94	44
Cisco	1969	0.66	6.1	61	10
	1976	0.95	1.0	6	7
Coho salmon	1968	0.54	0.5	26	50
	1969	0.68	2.5	51	20
	1970	0.60	1.6	25	15
	1971	0.61	1.2	19	17
	1975	0.91	0.4	7	16
Lake whitefish	1969	0.36	3.6	9	2
	1972	0.60	1.4	9	7
	1973	0.60	1.2	8	7
	1976	0.80	0.6	2	3
Georgian Bay					
Bloater	1971	0.66	1.0	24	24
	1975	0.61	0.7	5	7
Cisco	1969	0.38	3.2	159	50
	1976	0.62	0.7	8	12

Mean PCB residues declined in lake trout and lake whitefish between 1971 and 1975 but increased in bloaters over the same period. Comparison of lake trout by weight class revealed no significant decline in PCB residues (Table 3). The ΣDDT/PCB ratio in all species declined, suggesting the disappearance of ΣDDT. The PCB/dieldrin ratio indicates that dieldrin is more persistent in fish tissues than are PCBs.

Other organochlorines—Trace quantities (<0.01 ppm) of *cis*- and *trans*-chlordane were detected in some bloaters, white sucker, lake trout, and lake whitefish caught in 1975, but no oxychlordane, endrin, or heptachlor epoxide was detected in fish caught in 1968–75.

LAKE HURON

ΣDDT—Three fish species caught in Lake Huron and Georgian Bay contained mean residues that exceeded 5 ppm. These included walleye (5.05 ppm) caught in southern Lake Huron in 1970, lake trout (7.60 ppm) caught in Georgian Bay in 1969, and bloaters (5.18 ppm) caught in 1971 in Georgian Bay (Table 4). Individual fish of five species contained ΣDDT residues in excess of 5 ppm including: bloaters (1970 and 1971), coho salmon (1970), and walleye (1970), caught in the southern half of Lake Huron; and bloaters (1971), rainbow trout (1968), lake trout (1969), and walleye (1969 and 1970) caught in Georgian Bay (Table 4, Figure 1).

ΣDDT residues declined noticeably between 1968–71 and 1975–76 in six species including alewife (*Alosa pseudoharengus*), smallmouth bass (*Micropterus dolomieui*), cisco (*Coregonus artedii*), coho salmon, rainbow

TABLE 3. *Comparison of organochlorine residues in two weight classes of splake, lake trout, and lake whitefish caught in Lake Huron and Lake Superior, 1969–76*

Species and Location	Year	0.5–1.0 kg Class						1.0–1.5 kg Class					
		No. of Fish	Weight, g	Fat, %	ΣDDT, ppm	Dieldrin, ppm	PCBs, ppm	No. of Fish	Weight, g	Fat, %	ΣDDT, ppm	Dieldrin, ppm	PCBs, ppm
Splake													
Lake Huron	1969	3	821	6.8	1.61	0.06	0.2	5	1351	6.9	0.86	0.03	0.3
	1970	3	784	13.2	1.16	0.06	1.6	8	1220	17.6	1.35	0.07	1.5
	1972	3	787	10.8	0.87	0.05	0.7						
	1973	10	690	6.6	0.77	0.03	0.6	4	1108	12.2	0.75	0.06	0.9
	1974	1	526	3.3	0.11	<0.01	0.1	4	1271	4.4	0.15	0.02	0.3
Georgian Bay	1975	6	747	11.9	0.78	0.14	1.4	6	1185	14.1	0.87	0.16	1.9
Whitefish													
Lake Superior	1971	3	910	12.5	0.71	0.04	0.8	2	1032	11.3	0.78	0.04	0.8
	1975	1	766	8.6	0.28	0.04	0.2	9	1176	11.0	0.22	0.07	0.3
Lake Huron	1969	22	730	5.4	0.40	0.05	0.1	1	1180	3.7	0.25	0.03	<0.1
	1972	12	813	8.2	0.55	0.07	0.3	7	1142	12.3	0.87	0.09	0.6
	1973							7	1172	17.1	0.64	0.08	0.4
	1976	2	850	3.7	0.08	0.03	0.1	10	1237	6.3	0.12	0.07	0.2
North Channel	1969	6	936	3.2	0.14	0.01	<0.1	8	1187	6.1	0.89	0.10	0.1
	1970	1	980	8.6	0.80	0.05	0.4	2	1285	10.6	0.71	0.07	0.4
Georgian Bay	1969	4	939	4.5	0.44	0.01	0.1	6	1131	3.7	0.54	0.01	0.2
Lake Trout													
Lake Superior	1968	8	698	4.8	0.818	0.046	0.21	4	1566	10.6	4.94	0.128	1.19
	1969	10	619	3.8	0.731	0.024	0.25	10	1308	8.8	1.25	0.040	0.34
	1970							4	1694	17.9	1.75	0.033	1.88
	1975	3	768	18.6	0.192	0.037	0.33	7	1272	21.6	0.17	0.037	0.49

smelt (*Osmerus mordax*), and walleye from the main waters of Lake Huron, and bloaters from Georgian Bay. ΣDDT mean residues were erratic or unchanged in cisco, splake (*Salvelinus fontinalis* and *S. namaycush*), and walleye caught in Georgian Bay and in splake and lake whitefish caught in the main lake.

To determine whether ΣDDT residues in splake and lake whitefish had declined, similar weight classes were compared (Table 3). ΣDDT levels in splake with an average weight of 1250 g declined between 1971 and 1974 from 1.35 ppm to 0.15 ppm. A similar decline in ΣDDT residues in lake whitefish was noted between 1972 and 1976. Cisco, coho salmon, and lake whitefish all showed a marked increase in the DDE+TDE/ΣDDT ratio during the present study (Table 2), suggesting a lower intake of the parent compound and/or degradation to metabolites; this decline was not evident in bloaters.

Dieldrin—Mean residues for all species investigated did not exceed the 0.3 ppm tolerance level set by FDA. However, individual fish of three species exceeded the level. One of 20 lake whitefish caught in the North Channel in 1969 contained 0.58 ppm dieldrin; one of 10 bloaters caught in Lake Huron in 1970 had a residue of 0.44 ppm dieldrin; five of 10 bloaters caught in Georgian Bay in 1975 contained dieldrin levels of 0.34–0.50 ppm; 10 of 20 bloaters caught in the North Channel in 1975 contained residues of 0.3–0.6 ppm dieldrin; and two large splake caught in Lake Huron contained residues of 0.43 ppm and 0.53 ppm dieldrin. The 10 bloaters caught in the North Channel during 1975, which had residues above the tolerance level, weighed an average of 333 g and contained an average of 0.40 ppm dieldrin. The remaining 10 bloaters, which averaged 236 g, contained a mean residue of 0.19 ppm dieldrin. In this instance, and in the case of the splake, higher dieldrin residues were associated with larger fish, but this relationship was not apparent in the 10 bloaters caught in Georgian Bay in 1975 (Table 4).

Dieldrin levels increased in alewife, bloaters, cisco, yellow perch (*Perca flavescens*), coho salmon, and splake during 1968–71 and 1975–76; levels in other species showed little change. Assessment of dieldrin levels on the basis of similar weight classes of lake whitefish and splake indicate that residues declined in lake whitefish and increased in splake (Table 3). A marked decline was noted in the ΣDDT/dieldrin ratio in four species: in cisco, for example, the ratio declined from 61 to 6 between 1969 and 1976. The PCB/dieldrin ratio also declined in the same four species suggesting declining PCB residues and static or increasing dieldrin residues (Table 2).

PCBs—Three fish species contained mean PCB residues which exceeded the 2 ppm tolerance level set by Health and Welfare Canada. Bloaters from the main lake (1970 and 1971), from Georgian Bay (1971 and 1975), and from the North Channel (1975) contained mean residues of 2.2–5.2 ppm. Individual bloaters had residues as high as 5.0 ppm and 6.4 ppm (Table 4). Cisco netted in Georgian Bay during 1975 contained a mean PCB residue of 2.2 ppm and a high level of 4.6 ppm in individual fish. Two large splake taken from the main waters of Lake Huron in 1975 contained levels of 5.5 ppm and 6.4 ppm PCBs.

TABLE 4. *Organochlorine residues in 14 fish species caught in the North Channel, Georgian Bay, and Canadian waters of Lake Huron, 1968–76*

Species	Year	Location	No. of Analyses[1]	Mean and Range Weight, g	Mean and Range Fat, %	Mean Content and Range of Contaminants in Fish Puree, ppm[2] DDE	TDE	DDT	ΣDDT	Dieldrin	PCBs
Catostomidae											
White sucker	1972	Huron	5	723	2.5	0.08	0.01	0.02	0.11	<0.01	0.1
				550–909	1.8–3.3	0.05–0.13	<0.01–0.03	<0.01–0.06	0.06–0.22		<0.1–0.2
	1973	Georgian Bay	4	131	0.7	<0.01	<0.01	<0.01	0.01	<0.01	0.1
				66–212	0.2–1.0	<0.01–0.02			<0.01–0.03		<0.1–0.2
	1976	Huron	10	977	0.6	0.06	<0.01	0.02	0.09	<0.01	0.1
				738–1837	0.1–1.1	<0.01–0.20		<0.01–0.14	<0.01–0.37		<0.1–0.2
Centrachidae											
Smallmouth bass	1968	Huron	3	499	3.1	0.68	0.76	0.53	1.97	<0.01	0.9
				429–630	2.0–4.9	0.12–1.69	0.15–2.02	0.12–1.23	0.30–4.94		0.2–2.0
	1972	Huron	5	353	3.7	0.12	0.01	0.03	0.16	0.01	0.4
				298–437	2.6–4.5	0.11–0.13		0.02–0.04	0.15–0.18	<0.01–0.03	0.3–0.5
	1972	Georgian Bay	6	281	2.8	0.05	0.01	<0.01	0.07	<0.01	0.01
				270–300	1.6–4.0	0.04–0.07	0.01–0.02		0.06–0.10		0.1–0.1
	1975	Georgian Bay	9	364	3.2	0.17	0.01	0.03	0.21	0.03	0.6
				275–562	1.7–4.5	0.09–0.28	<0.01–0.04	<0.01–0.08	0.12–0.36	<0.01–0.09	0.4–0.9
Clupeidae											
Alewife	1970	Huron	8(21)	33	7.5	0.76	0.23	0.64	1.63	0.08	1.1
				26–40	1.5–13.2	0.16–1.40	0.04–0.52	0.22–1.48	0.27–3.40	0.01–0.22	0.5–2.0
	1976	Huron	5(23)	23	10.7	0.44	0.10	0.26	0.80	0.14	0.3
				3–49	5.8–16.9	0.04–1.08	0.01–0.12	0.01–0.54	0.06–1.74	<0.01–0.25	0.1–0.6
Osmeridae											
Rainbow smelt	1970	Huron	8(21)	22	6.5	0.36	0.12	0.32	0.80	0.04	0.7
				12–67	4.0–8.4	0.06–0.97	0.01–0.25	0.02–0.80	0.11–1.86	<0.01–0.15	0.2–1.0
	1970	N. Channel	5(24)	26	3.6	0.12	0.04	0.15	0.31	0.02	0.1
				18–44	2.8–4.4	0.05–0.20	0.03–0.05	0.08–0.20	0.15–0.45	<0.01–0.03	
	1976	Huron	7(32)	14	2.7	0.11	0.02	0.02	0.15	0.01	0.01
				7–30	1.2–3.9	0.05–0.19	0.01–0.02	<0.01–0.03	0.08–0.23	<0.01–0.02	<0.1–0.2
Percidae											
Yellow perch	1968	Huron	5	335	0.8	0.20	0.12	0.20	0.52	<0.01	0.2
				118–426	0.5–1.0	0.06–0.61	0.02–0.47	0.08–0.51	0.16–1.59		<0.1–0.5
	1969	N. Channel	20	201	1.4	0.03	0.01	0.03	0.07	<0.01	<0.1
				167–341	0.5–2.4	<0.01–0.08	<0.01–0.03	<0.01–0.05	<0.01–0.18		
	1972	Huron	5	67	4.4	0.07	0.01	0.03	0.11	0.01	0.1
				64–74	3.8–5.3	0.06–0.08	0.01	0.02–0.03	0.09–0.12	<0.01–0.02	
	1975	N. Channel	10	175	6.1	0.36	0.03	0.09	0.48	0.05	0.9
				150–197	3.5–8.6	0.13–0.57	<0.01–0.05	<0.01–0.17	0.13–0.72	0.02–0.09	0.4–1.4
	1976	Huron	17	236	2.5	0.21	0.03	0.08	0.32	0.02	0.2
				66–481	0.7–5.4	0.06–0.68	0.01–0.08	0.01–0.55	0.07–1.31	<0.01–0.05	<0.1–0.4
Walleye	1968	Huron	3	409	0.8	0.12	0.04	0.13	0.29	<0.01	0.1
				390–426	0.6–0.9	0.06–0.22	0.02–0.07	0.08–0.21	0.16–0.50		<0.1–0.1
	1969	Georgian Bay	15	2073	2.6	1.05	0.24	1.08	2.37	0.02	1.5
				792–4190	0.6–6.0	0.23–3.53	0.06–0.81	0.23–4.03	0.54–8.36	<0.01–0.07	0.5–2.1
	1970	Huron	2	2083	10.1	2.37	0.64	2.04	5.05	0.08	1.3
				1910–2255	9.7–10.4	1.80–2.91	0.43–0.84	1.65–2.47	3.88–6.22	0.06–0.08	0.7–1.9
	1970	Georgian Bay	21	3236	4.7	0.94	0.23	0.98	2.15	0.04	1.4
				1721–4760	1.2–10.6	0.23–3.70	0.04–0.85	0.18–3.88	0.45–8.33	<0.01–0.16	0.5–2.3
	1970	N. Channel	3	605	2.1	0.19	0.04	0.16	0.39	<0.01	0.1
				526–715	1.3–3.5	0.16–0.23	0.04–0.05	0.13–0.21	0.34–0.49		<0.1–0.1
	1971	Georgian Bay	10	2859	5.8	1.74	0.21	1.11	3.06	0.03	1.8
				1132–4756	2.9–11.6	0.08–4.08	0.04–0.37	0.04–2.32	0.16–6.93	<0.01–0.07	0.1–3.9
	1975	Huron	10	2539	4.7	0.38	0.05	0.27	0.70	0.03	0.5
				722–5218	1.5–13.3	0.16–1.01	<0.01–0.15	0.03–1.01	0.27–2.17	0.01–0.13	0.3–2.5
Salmonidae											
Bloater	1969	Huron	15	97	3.0	0.39	0.12	0.18	0.69	0.01	0.2
				55–143	1.4–7.7	0.06–1.54	0.02–0.50	0.04–0.66	0.20–2.71	<0.01–0.02	<0.1–0.7
	1970	Huron	10	260	16.0	1.95	0.49	2.24	4.68	0.16	2.6
				148–307	8.1–26.7	1.00–3.69	0.20–1.03	1.01–5.17	2.48–9.88	0.04–0.44	1.5–5.0
	1971	Huron	6	71	15.0	2.73	0.29	1.69	4.71	0.05	2.2
				61–101	7.2–21.1	2.02–4.34	0.05–0.78	1.14–2.40	3.44–7.52	0.03–0.07	1.1–3.2
	1971	Georgian Bay	4(12)	259	20.1	2.90	0.53	1.75	5.18	0.22	5.2
				140–433	18.0–23.0	2.34–3.83	0.29–0.75	1.63–1.85	4.26–6.43	0.18–0.28	4.3–6.4
	1975	Georgian Bay	10	219	16.3	0.71	0.16	0.56	1.43	0.30	2.2
				179–250	8.1–20.1	0.33–1.15	0.01–0.30	0.15–1.11	0.49–2.56	0.10–0.50	0.8–4.4
	1975	N. Channel	20	285	22.9	1.16	0.17	0.54	1.87	0.29	2.6
				134–560	15.5–29.8	0.56–2.34	<0.01–0.41	0.01–1.48	0.74–4.18	<0.01–0.60	0.6–5.2
Cisco	1969	Huron	2	180	5.4	0.35	0.05	0.21	0.61	0.01	0.1
				100–260	2.0–8.8	0.18–0.52	0.02–0.07	0.07–0.35	0.27–0.94	<0.01–0.02	<0.1–0.2
	1969	Georgian Bay	10	820	7.2	0.41	0.19	0.99	1.59	0.01	0.5
				337–937	4.8–9.4	0.22–1.01	0.08–0.40	0.54–2.58	0.83–3.99	<0.01–0.02	0.2–1.1
	1975	Georgian Bay	6	543	18.0	0.78	0.15	0.58	1.51	0.19	2.2
				352–710	11.3–24.7	0.47–1.14	0.09–0.29	0.26–1.22	0.83–2.65	0.12–0.30	1.3–4.6
	1976	Huron	9(11)	138	5.0	0.14	0.05	0.01	0.20	0.03	0.2
				47–242	3.0–7.5	0.11–0.19	0.03–0.09	<0.01–0.02	0.14–0.29	0.01–0.08	0.1–0.4

(*continued next page*)

TABLE 4. (cont.d.) *Organochlorine residues in 14 fish species caught in the North Channel, Georgian Bay, and Canadian waters of Lake Huron, 1968–76*

Species	Year	Location	No. of Analyses[1]	Mean and Range Weight, g	Mean and Range Fat, %	Mean Content and Range of Contaminants in Fish Purée, ppm[2] DDE	TDE	DDT	ΣDDT	Dieldrin	PCBs
Coho salmon	1968	Huron	8	81	3.9	0.11	0.03	0.12	0.26	<0.01	0.5
				39–163	1.2–5.4	0.04–0.42	0.01–0.12	0.04–0.43	0.09–0.97	0.01–0.03	0.2–1.0
	1969	Huron	5	1885	5.8	0.88	0.16	0.48	1.52	0.03	0.6
				1138–3335	5.1–6.5	0.31–2.01	0.13–0.21	0.35–0.62	0.87–2.84	0.01–0.05	0.3–1.2
	1970	Huron	41	1031	4.9	0.48	0.11	0.39	0.98	0.04	0.6
				754–1595	0.8–11.1	0.9–5.2	0.03–0.66	0.08–1.89	0.22–7.75	0.01–0.24	0.1–7.0
	1971	Huron	10	936	8.0	0.50	0.20	0.45	1.15	0.06	1.0
				475–1395	4.9–13.0	0.23–1.15	0.07–0.62	0.16–0.80	0.48–2.09	0.02–0.13	0.2–2.1
	1975	Huron	11	2284	5.8	0.43	0.05	0.05	0.53	0.08	1.3
				280–4356	3.9–8.4	0.04–0.94	<0.01–0.14	<0.01–0.19	0.05–1.22	0.01–0.16	0.1–3.3
Kokanee salmon	1968	Huron	2	95	3.5	0.44	0.12	0.59	1.15	0.04	0.3
				94–96	2.8–4.2	0.08–0.80	0.02–0.22	0.10–1.08	0.20–2.00	<0.01–0.07	0.1–0.4
	1969	Huron	11	98	3.3	0.10	0.03	0.10	0.23	0.01	<0.1
				58–375	1.4–5.3	0.02–0.80	<0.01–0.06	0.03–0.18	0.06–0.57	<0.01–0.03	
	1970	Huron	15	512	4.1	0.38	0.12	0.45	0.95	0.04	0.6
				204–1098	0.9–7.8	0.17–0.67	0.05–0.24	0.16–1.03	0.78–1.76	<0.01–0.12	0.2–1.0
Splake	1969	Huron	20	1113	6.3	0.43	0.14	0.46	1.03	0.04	0.2
				25–2354	1.8–10.1	0.03–0.85	0.02–0.33	0.03–0.85	0.08–2.11	<0.01–0.09	<0.1–0.5
	1970	Huron	23	810	10.2	0.37	0.13	0.50	1.00	0.04	1.0
				208–1420	3.2–15.7	0.12–0.82	0.04–0.20	0.01–1.03	0.19–1.81	<0.01–0.12	0.1–1.5
	1972	Huron	5	544	8.5	0.30	0.04	0.28	0.62	0.03	0.5
				138–877	5.1–11.7	0.14–0.44	0.01–0.04	0.08–0.56	0.23–1.10	0.01–0.05	0.2–0.9
	1973	Huron	26	556	8.8	0.38	0.03	0.06	0.47	0.03	0.4
				96–450	3.6–16.1	0.03–1.20	<0.01–0.13	<0.01–0.16	0.04–1.31	<0.01–0.10	<0.1–1.5
	1974	Huron	7	1238	4.1	0.11	0.01	0.03	0.15	0.02	0.2
				526–1540	2.7–5.9	0.06–0.19	<0.01–0.02	<0.01–0.05	0.08–0.25	<0.01–0.04	<0.1–0.7
	1975	Huron	2	2127	17.2	1.80	0.42	0.46	2.68	0.48	6.0
				1997–2256	13.6–20.8	1.58–2.02	0.40–0.44	0.37–0.55	2.39–2.97	0.43–0.53	5.5–6.4
	1975	Georgian Bay	17	1048	13.3	0.52	0.08	0.18	0.78	0.15	1.6
				458–1798	8.5–17.4	0.18–0.88	0.03–0.11	0.10–0.37	0.28–1.12	0.08–0.18	0.6–2.3
Rainbow trout	1968	Georgian Bay	12	857	5.5	0.74	0.17	0.84	1.75	0.04	0.3
				284–1850	3.2–7.8	0.12–6.17	0.02–1.27	0.13–5.70	0.27–13.1	<0.01–0.19	<0.1–1.5
Lake trout	1969	Georgian Bay	4	6328	13.4	4.04	0.50	3.06	7.60	0.07	0.7
				4200–8740	12.9–13.8	3.14–5.51	0.45–0.63	2.66–3.71	6.28–9.85	0.06–0.09	0.4–0.9
Lake whitefish	1969	N. Channel	20	1430	4.8	0.13	0.07	0.36	0.56	0.06	0.1
				854–2785	0.8–8.9	0.03–1.58	0.01–0.31	0.66–2.66	0.10–4.75	<0.01–0.58	<0.1–0.7
	1969	Huron	26	711	5.0	0.13	0.05	0.18	0.36	0.04	0.1
				386–1080	2.9–9.9	0.07–0.24	0.02–0.10	0.04–0.36	0.16–0.73	<0.01–0.08	<0.1–0.3
	1969	Georgian Bay	10	1054	4.0	0.20	0.09	0.22	0.51	0.01	0.2
				881–1241	3.2–6.4	0.10–0.31	0.03–0.13	0.09–0.33	0.22–0.78	<0.01–0.02	<0.1–0.3
	1970	N. Channel	3	1183	9.9	0.27	0.07	0.40	0.74	0.06	0.4
				980–1361	8.6–11.7	0.23–0.29	0.07	0.33–0.45	0.63–0.80	0.05–0.09	0.2–0.5
	1972	Huron	25	761	8.2	0.28	0.65	0.22	0.55	0.06	0.04
				80–1393	1.8–21.9	0.07–1.02	0.01–0.16	0.03–0.42	0.11–1.47	0.01–0.11	0.1–0.8
	1973	Huron	19	804	14.2	0.23	0.05	0.19	0.47	0.06	0.04
				108–1747	6.9–26.5	0.03–1.48	0.01–0.21	0.02–0.92	0.07–2.61	<0.01–0.25	<0.1–0.6
	1976	Huron	15	1323	5.8	0.08	0.02	0.03	0.13	0.06	0.2
				802–2495	2.5–11.6	0.04–0.15	0.01–0.03	0.02–0.05	0.07–0.23	0.03–0.14	0.1–0.5

[1]Most analyses were performed on single fish; where composites were analyzed, the number of fish is given in parentheses. Composites were of similar weight.

[2]<0.01 ppm represents a trace of contaminant above the level of detection (0.001 ppm) but below 0.010 ppm.

Alewife, rainbow smelt, and walleye caught in the main waters of Lake Huron were the only species in which PCB levels declined between 1968–71 and 1975–76. Residues in cisco from Georgian Bay, yellow perch from the North Channel, and coho salmon and splake from the main lake increased during these periods. In the above species caught in other locations and in other species, no trend was evident: PCB levels were static. Analysis of splake and lake whitefish on the basis of similar weight classes indicates that PCB levels peaked in splake in 1970 and then declined between 1970 and 1974. PCB levels in lake whitefish declined between 1972 and 1976 (Table 3).

In general, the ΣDDT/PCB and PCB/dieldrin ratios declined between 1968–71 and 1975–76 in four species: bloaters, cisco, coho salmon, and lake whitefish; this supported the finding that ΣDDT residues were declining, dieldrin residues were increasing, and PCB residues were static or declining slowly (Table 2).

Chlordane—Residues of chlordane were detected in smallmouth bass and walleye caught in Georgian Bay in 1975 at mean levels of 0.01 ppm and 0.05 ppm, respectively (sum of *cis*- and *trans*-isomers). Trace levels (<0.01 ppm) were suspected in bloaters, cisco, coho salmon, and splake in 1975. Oxychlordane analysis was included in 1976. Total chlordane levels found in 1976 ranged from traces in yellow perch to 0.039 ppm in alewife; both species were caught in the open part of Lake Huron (Table 5).

TABLE 5. *Residues of chlordane and heptachlor epoxide in fish from the main waters of Lake Huron and Georgian Bay, 1975–76*

Location and Species	No. of Fish[1]	Average Weight, g	Average Fat, %	Residue, ppm Chlordane[2]	Heptachlor Epoxide
1975 Georgian Bay					
Smallmouth bass	9	364	3.2	0.01 <0.01–0.04	ND
Walleye	10	2539	4.7	0.05 <0.01–0.19	ND
1976 Lake Huron					
Alewife	23(5)[3]	23	10.7	0.039 0.004–0.060	0.026 ND–0.100
Cisco	11(9)	138	5.0	0.025 0.015–0.040	0.013 ND–0.044
Yellow perch	17	236	2.5	<0.001 ND–0.013	0.002 ND–0.009
Rainbow smelt	32(7)	14	2.7	0.002 ND–0.013	0.004 ND–0.007
White sucker	10	977	0.6	0.001 ND–0.013	0.002 ND–0.006
Lake whitefish	15	1323	5.8	0.047 0.025–0.087	0.026 0.013–0.065

NOTE: ND=not detected to less than 0.005 ppm.
[1]Number in brackets represents number of analysis.
[2]1975 analyses included *cis*- and *trans*-isomers; 1976 analyses included *cis*- and *trans*-chlordane and oxychlordane.

Heptachlor epoxide—No heptachlor epoxide was identified in fish caught prior to 1976. Mean residues in alewife caught in 1976 ranged from 0.002 ppm in yellow perch to 0.026 ppm in alewife caught in the main lake (Table 5).

Hexachlorobenzene (HCB)—Analyses for HCB in fish tissues were not routinely carried out during the study period. An indication of the extent of HCB in fish was obtained from samples caught in 1972 and 1973 from Lake Huron. One of five splake caught in the open lake contained 0.001 ppb HCB, and smelt caught off Black Stone Harbour in Georgian Bay contained 0.03 ppm. HCB was not detected in a limited number of smallmouth bass, yellow perch, or lake whitefish from either Georgian Bay or the main lake.

Discussion

Lake Superior water analyzed by Glooschenko et al. *(3)* was free of DDT, dieldrin, and PCBs down to the detection limit. However, residues of these contaminants were found in sediment and seston. Sediment samples taken from various sites in the Canadian waters of Lake Superior had measurable amounts (0.005 ppm) of dieldrin and ΣDDT in 14 percent and 5 percent, respectively. PCB residues were present in all sediments at all sites; highest level reported was 1.3 ppm in samples collected near Marathon. Seston contained only traces of ΣDDT and dieldrin, but the mean level of PCBs was 1.3 ppm, identical to that in the sediments.

Levels of ΣDDT and dieldrin in lake trout caught in 1970 in Lake Superior correspond closely with those reported by Reinhert *(7)* in 1966–67. Residues in lake trout reported in the present study did not agree with those cited in the Upper Great Lakes Reference Group report *(11)*. However, bloaters contained similar residues in two studies.

Measurable levels of ΣDDT were reported by Glooschenko et al. in 29 percent of sediments taken from various sites in the Canadian waters of Lake Huron and in 14 percent of sediments taken from Georgian Bay *(3)*; maximum levels in both Lake Huron and Georgian Bay were 0.02 ppm. Dieldrin was present at trace levels, and PCBs ranged up to 0.02 ppm. ΣDDT and dieldrin in sediments from the North Channel were below detection levels, but traces of PCBs were found. Organochlorines were highest in seston from the open lake, ranging from 0.8 to 8.1 ppm compared to 0.7 to 6.7 ppm in Georgian Bay and a high level of 1.0 ppm in the North Channel.

Residue levels of ΣDDT and dieldrin in fish from Lake Huron and Georgian Bay reported in this paper correspond closely to the levels reported previously by Reinhert *(7)* and Reinke et al. *(9)* for alewife, bloaters, kokanee *(Oncorhynchus nerka)*, rainbow smelt, and walleye, but discrepancies are evident in yellow perch, lake whitefish, and rainbow trout. ΣDDT mean residues of 2.44 ppm are reported by Reinhert for alewife caught in 1966–67 *(7)*; the present study reveals a decline to 1.63 ppm ΣDDT mean residues in 1970 and a further decline to 0.80 ppm ΣDDT by 1976; conversely, dieldrin levels were slightly higher in 1976 (0.14 ppm) than in 1966–67 (0.05 ppm). Levels of ΣDDT in rainbow trout show little change between the 1966–67 study and those detected in 1970 in the present study, 0.75 and 0.8 ppm, respectively. However, a marked decline to a mean residue of 0.15 ppm by 1976 occurred in rainbow trout caught in Lake Huron. A mean level of 4.7 ppm ΣDDT in bloaters caught in 1970–71 in the present study is similar to levels of 3.6 ppm and 3.08 ppm reported respectively by Reinhert *(7)* in 1966 and Reinke et al. *(9)* in 1970. Mean levels of ΣDDT, dieldrin, and PCBs in bloaters in the present study closely parallel those reported by the Upper Great Lakes Reference Group *(11)* for 1975–76.

Reinke et al. found 6.02 ppm ΣDDT in walleye caught in 1970 in the main waters of Lake Huron *(9)*; this is close to the mean level of 5.05 ppm reported here. Reinke et al. reported 0.47 ppm ΣDDT in walleye caught in 1970 in Georgian Bay *(9)*, but the present study reports mean levels of 2.2 ppm and 3.1 ppm, respectively, for 1970 and 1971. This discrepancy may be partly explained by the fact that the walleye in the present study were obtained at the mouth of the Moon River, an area where high DDT residues were reported *(2, 6)*.

Residues in kokanee from Lake Huron reported here are similar to those reported by Reinke et al. *(9)* but ΣDDT

residues of 0.52 ppm in yellow perch reported in the present study are considerably lower than the mean values of 1.59 ppm in 1966–67 and 1.46 ppm in 1970 reported by Reinhert *(7)* and Reinke et al. *(9)*, respectively. Mean ΣDDT residues in lake whitefish in the present study are also markedly lower than those reported previously *(7, 9)*.

Although there are differences in the data for ΣDDT levels in coho salmon between the present study and earlier reports, there is more similarity among coho salmon from the same location. Reinke et al. reported a mean of 1.26 ppm ΣDDT and 0.08 ppm dieldrin for fish caught in northern Lake Huron *(9)*; the present study shows mean levels of 0.98 ppm ΣDDT and 0.04 ppm dieldrin for 41 coho salmon caught in the same area. ΣDDT levels in rainbow trout caught in southern Georgian Bay vary considerably from those reported previously. Reinke et al. reported a mean of 8.7 ppm ΣDDT in rainbow trout caught in 1970 *(9)*, but only 1.75 ppm ΣDDT was found in the same species caught in the same location in 1968 for the present survey. This discrepancy may be due to local differences in ΣDDT use.

Despite the number of variables which are associated with a sampling study of this kind, it is remarkable that such close agreement is found between different studies in different time frames for such large bodies of water as Lakes Superior and Huron. Other factors that cause fluctuations in contaminant concentrations in fish tissues are spawning times and changes in fat content.

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Residues of Organochlorine Insecticides and Polychlorinated Biphenyls in Fish from Lakes Saint Clair and Erie, Canada—1968–76 [1]

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ABSTRACT

Eighteen species of fish from Lake Saint Clair and 19 species from Lake Erie were analyzed for organochlorine pesticides and polychlorinated biphenyls (PCBs) between 1968 and 1976. Mean residues of ΣDDT peaked at 1.19 ppm in longnose gar (Lepisosteus osseus) *caught in Lake Saint Clair in 1970–71, but had declined in all species by 1975–76. Dieldrin levels in fish tissues increased over the same period. White bass* (Morone chrysops), *caught in 1975 in Lake Erie, had the highest mean residue of dieldrin at 0.17 ppm. PCB residues increased in some species and decreased in others. PCB residues exceeding the tolerance level of Health and Welfare Canada were found in the following: from Lake Saint Clair, smallmouth bass* (Micropterus dolomieui) *in 1975 and channel catfish* (Ictalurus punctatus) *in 1971; from Lake Erie, coho salmon* (Oncorhynchus kisutch) *in 1970, smallmouth bass, alewife* (Alosa pseudoharengus), *freshwater drum* (Aplodinotus grunniens), *and gizzard shad* (Dorosoma cepedianum) *in 1971, and white bass in 1971 and 1976.*

Sediments in Lake Erie were five to ten times more highly contaminated with ΣDDT, dieldrin, and PCBs than were sediments from Lake Saint Clair. ΣDDT and dieldrin residues in fish tissues did not necessarily reflect this trend, but PCBs were higher in fish from Lake Erie.

Introduction

DDT, dieldrin, and PCBs have been identified in fish from Lake Erie and Lake Saint Clair. Reinert reported residues of ΣDDT in 14 species caught in 1967–68 that ranged from 0.25 ppm in spottail shiner (*Notropis hudsonius*) to 1.89 ppm in white bass (*Morone chrysops*) (*11*). Dieldrin was not detected in nine species; maximum dieldrin levels found in alewife (*Alosa pseudoharengus*) were 0.15 ppm. Reinke et al. found similar residues in six species caught in 1970 (*13*). The highest residues of ΣDDT were 0.56 ppm in alewife. Carr et al. reported on six species caught in 1970–71 (*2*). Coho salmon (*Oncorhynchus kisutch*) contained the highest mean residues of ΣDDT and dieldrin, 0.90 ppm and 0.07 ppm, respectively; channel catfish (*Ictalurus punctatus*) had the highest mean PCB residues: 4.4 ppm. Kelso and Frank found that ΣDDT and dieldrin residues varied with time of catch in three species from the eastern basin of Lake Erie (*7*). Residues were generally low; higher residue levels were associated with fish having a higher fat content.

Watersheds on the Canadian side of Lakes Erie and Saint Clair drain the most intensive agricultural belt in Ontario (Figure 1). Before restrictions on the use of aldrin, dieldrin, and heptachlor in 1969 and ΣDDT in 1970–71, this area accounted for 90 percent of organochlorine insecticides used in Ontario. Miles and Harris (*9, 10*) and Frank et al. (*3, 4*) reported that DDT and dieldrin were deposited in Lake Erie by creeks draining areas of intensive pesticide use. Frank et al. found that fish caught in the streams and creeks had residues of ΣDDT and dieldrin that were one order of magnitude higher than those caught in the adjoining waters of Lake Erie (*4*).

The present study was initiated in 1968 to determine organochlorine residues in fish before legislative restriction of the use of these materials. After use of the materials was restricted, monitoring of fish tissue was continued to determine the impact of these actions. At the same time, PCBs were identified in fish in both lakes, and monitoring for these contaminants was included to determine whether the voluntary restrictions on their use since 1971 were reflected in residue levels in fish tissue.

Methods and Materials

Twenty-eight species of fish were caught by gill net or trap net between 1968 and 1976 in Lakes Saint Clair and Erie (Table 1). Most were obtained from the field

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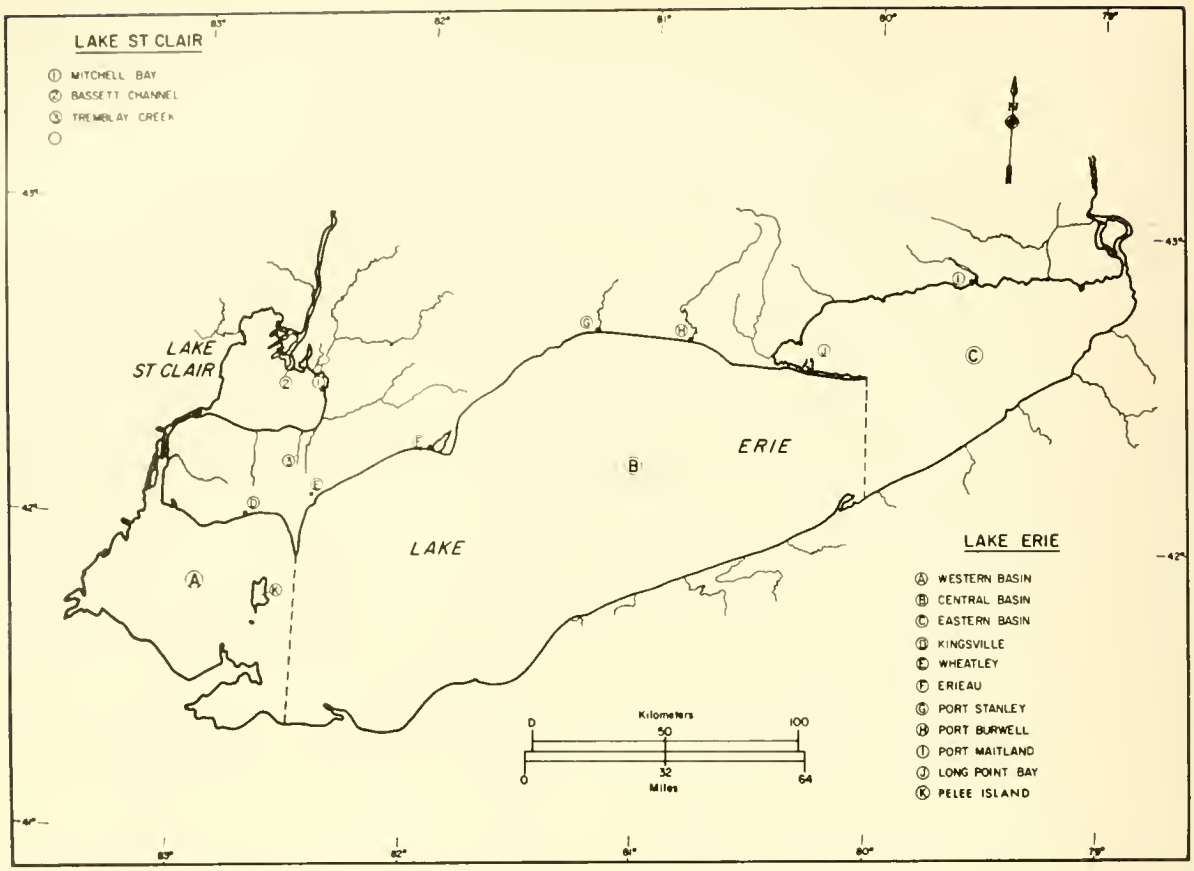

FIGURE 1. *Map of Lakes Erie and Saint Clair showing fish collection areas.*

staff of the Ontario Ministry of Natural Resources. Some of the larger fish were obtained from commercial gill net catches. Eighteen species (278 fish) were caught in Lake Saint Clair in or around Mitchell Bay, Tremblay Creek, and Bassett Channel (Figure 1). Nineteen species (1,023 fish) were caught in Canadian waters of Lake Erie. These came from onshore and offshore locations in all three basins, as defined by Thomas et al. (*16*). Eleven species (429 fish) from the western basin were caught off Kinsville and west of Pelee Island. Nine species (287 fish) were caught in the central basin off Wheatley, Erieau, Port Stanley, and Port Burwell. Fifteen species (307 fish) from the eastern basin were netted in Long Point Bay and off Port Maitland (Figure 1).

Fish species were identified and named according to the nomenclature of the American Fisheries Society (*1*).

TABLE 1. *Analyses of fish caught in Lakes Saint Clair and Erie, 1968–76*

	Lake Saint Clair			Lake Erie		
Year	Fish Species	Fish Caught	Analyses Performed	Fish Species	Fish Caught	Analyses Performed
1968	6	25	25	14	115	106
1970	6	45	45	1	11	11
1971	15	183	183	11	137	119
1972	0	0	0	3	78	78
1973	0	0	0	1	10	1
1974	0	0	0	1	5	5
1975	1	6	6	11	636	181
1976	2	19	19	3	31	31
TOTAL	18	278	278	19	1023	532

NOTE: Purees of eviscerated, headless samples were analyzed.

Analytical Procedure

Fish were eviscerated, their heads were removed, and the remaining flesh was minced in a Hobart food chopper to a homogeneous consisteney from which a representative subsample was selected. Tissue homogenates were stored at −20°C until analysis; storage time rarely exceeded 4 months. Ten grams of tissue homogenate was ground with 100 g anhydrous sodium sulfate and 25 g Ottawa sand. This mixture was extracted in hexane for 7 hours in a Soxhlet extractor. The solvent was removed by rotary vacuum, and the percentage of fat or oil was determined gravimetrically.

A one-step Florisil column cleanup method described by Langlois et al. was used to isolate organochlorines and PCBs (*8*). Florisil (60–100 mesh), activated commercially at 650°C, was reheated at 135°C for at least 24

hours; after the adsorbent cooled, it was equilibrated with 5 weight percent water. A maximum of 1 g fat from the fish extracts was thoroughly mixed with 25 g of conditioned Florisil; this was placed on top of a second 25-g portion of conditioned Florisil in a 25-mm ID cleanup column. The column was eluted with 300 ml 1:4 (v/v) mixture of dichloromethane–hexane. The eluate was evaporated to dryness with rotary vacuum, and the residue was dissolved in 5 ml acetone.

PCBs were separated from organochlorine insecticides and HCB on a charcoal column as described by Holdrinet (*6*). Charcoal (Fisher No. 5-690, 50–200 mesh) was washed with acetone, filtered by suction, dried, and stored at 135°C. Columns (9-mm ID) were prepared by sandwiching a 7.5-cm layer of charcoal between 1.3-cm layers of sand and prewashing with a 1:3 (v/v) mixture of acetone–diethyl ether. The acetone solution from the Florisil cleanup was quantitatively transferred to the charcoal column and eluted successively with 180 ml of 1:3 (v/v) mixture of acetone–diethyl ether and 80 ml benzene; the organochlorine insecticides were contained in the first eluate, and PCBs were in the second eluate. Eluates were concentrated to dryness by rotary vacuum and dissolved in measured amounts of hexane.

Extracts were analyzed on a Tracor Model 550 gas chromatograph with the following instrument parameters and operating conditions:

Detector:	^{63}Ni electron-capture
Column:	glass, 15 cm × 0.64 cm OD packed with a mixture of 4 percent SE-30 and 6 percent QF-1 on 80–100-mesh Chromosorb W
Temperature:	180°C
Carrier gas:	nitrogen flowing at 60 ml/minute
Injection volume:	5 μl equivalent to 1 ng fat

Residue identity was confirmed on random samples by thin-layer chromatography (TLC); appropriate areas of the chromatogram were removed, redissolved, and re-examined by gas-liquid chromatography (GLC). This confirmation was essential for the positive identification of *cis*- and *trans*-chlordane which, when analyzed by GLC alone, are subject to misidentification because of co-extractive interferences.

Recoveries of pesticides and PCBs were checked periodically by fortification of fish tissue homogenate before the Soxhlet extraction. Average recoveries were as follows: *p,p'*-DDT, 89 percent; *p,p'*-DDE, 96 percent; *p,p'*-TDE, 94 percent; *o,p'*-DDT, 91 percent; dieldrin, 89 percent; *cis*-chlordane, 92 percent; *trans*-chlordane, 90 percent; and PCBs, 85–90 percent. The data do not include corrections for recovery. Quantitation limits, below which values were designated as either trace or not detected, were set at 0.005 ppm in fat for all organochlorine insecticides and 0.05 ppm in fat for PCBs.

PCB estimations were based on comparison with standard mixtures of Aroclors 1254 and 1260 and were quantitated by comparison of the sum of peak heights of peaks VII, VIII, and X according to the Reynolds numbering system (*14*). The ratio of Aroclor 1254 to Aroclor 1260 in the standard mixture varied from 5:1 to 4:1.

Analysis began in 1968 when the known main contaminants in fish were *p,p'*-DDT and its analogs and dieldrin and heptachlor epoxide; PCB values before 1970 were estimated. With the introduction of a column fractionation technique in 1970 for the separation of PCBs from organochlorine insecticides, the measurement of PCB residues became more precise. Analysis for hexachlorobenzene (HCB) was included in the procedure in 1973 but was subsequently discontinued because of the low levels and incidence of HCB found in the samples. Analysis and confirmation for *cis*- and *trans*-chlordane was refined in 1975, and the analyses for mirex and oxychlordane were introduced in 1976.

Results

LAKE SAINT CLAIR

ΣDDT—None of the 18 species caught in Lake Saint Clair contained mean residues of ΣDDT that exceeded the 5 ppm action level established by both the Canadian and United States governments. Longnose gar (*Lepisosteus osseus*) caught in 1971 had the highest mean residue of 1.19 ppm and was the only species with a mean residue above 1.0 ppm (Table 2). Eight of 12 longnose gar caught off Tremblay Creek contained ΣDDT residues of 1.10–2.35 ppm. Individual fish from three other species contained residues that exceeded 1.0 ppm. In 1971, two of eight carp (*Cyprinus carpio*) from Mitchell Bay contained 1.19 ppm and 1.26 ppm ΣDDT. Four of 12 mooneye (*Hiodon tergisus*) caught in 1970 off Tremblay Creek had 1.12–2.38 ppm ΣDDT.

Three of six smallmouth bass (*Micropterus dolomieui*) caught in 1975 had ΣDDT residues of 1.02–1.15 ppm.

Eight of the 18 species from Lake Saint Clair were caught in 1968–71. In seven of the species, residues of ΣDDT showed a decline by 1971 (Tables 2, 3). Only quillback (*Carpiodes cyprinus*) showed no apparent change. In all years, however, residues of ΣDDT were below 0.5 ppm.

Smallmouth bass, freshwater drum (*Aplodinotus grunniens*), and walleye (*Stizostedion vitreum vitreum*) were the only three species caught in 1968–71 and again in 1975–76. In smallmouth bass, mean ΣDDT residues were higher in 1976 (0.76 ppm) than in 1968 (0.42 ppm); however, the mean weight of fish was 853 g as opposed to 453 g (Table 2). When residues of similar weight classes were compared, the residue declined slightly between the two periods (Table 3). A mean

TABLE 2. *Organochlorine residues in 18 fish species caught in Canadian waters of Lake Saint Clair, 1968–76*

Species	Year	No. of Analyses[1]	Mean and Range Weight, g	Mean and Range Fat, %	Mean Content and Range of Contaminants in Fish Purée, ppm[2] DDE	TDE	DDT	ΣDDT	Dieldrin	PCBs
Amiidae										
Bowfin	1971	10	1367 955–2050	0.2 0.1–0.4	<0.01 <0.01–0.01	<0.01	<0.01	0.01 <0.01–0.02	<0.01	<0.1
Catostomidae										
Quillback	1970	6	1319 300–1775	4.2 1.6–7.6	0.03 0.01–0.04	0.03 0.01–0.06	0.02 0.01–0.03	0.08 0.03–0.13	0.01 <0.01–0.02	0.3 0.2–0.4
	1971	9	1244 350–1935	2.1 1.1–4.5	0.03 <0.01–0.07	0.03 <0.01–0.08	0.02 <0.01–0.06	0.08 0.02–0.17	<0.01	0.2 <0.1–0.3
Redhorse	1970	8	928 695–1235	2.6 0.3–5.8	0.07 <0.01–0.25	0.03 <0.01–0.11	0.03 <0.01–0.13	0.13 <0.01–0.49	0.01 <0.01–0.04	0.7 <0.1–2.6
	1971	8	698 375–985	0.7 0.3–1.3	0.01 <0.01–0.03	0.01 <0.01–0.02	<0.01 <0.01–0.02	0.03 0.01–0.07	<0.01	0.2 <0.1–0.4
White sucker	1968	2	547 306–787	2.8 2.2–3.4	0.07 0.01–0.12	0.04 0.01–0.07	0.08 0.02–0.14	0.19 0.04–0.33	<0.01	0.1 <0.1–0.2
	1970	10	1298 1035–2050	1.3 0.7–2.5	0.01 <0.01–0.03	0.02 <0.01–0.04	0.01 <0.01–0.03	0.04 <0.01–0.06	<0.01	0.3 0.2–0.4
Centrarchidae										
Largemouth bass	1970	6	564 315–685	3.5 1.8–7.2	0.22 0.12–0.40	0.09 0.04–0.22	0.10 0.03–0.26	0.41 0.19–0.88	0.03 <0.01–0.08	1.3 0.3–4.3
	1971	5	632 250–1100	2.6 1.4–4.2	0.18 0.14–0.21	0.07 0.03–0.13	0.06 0.02–0.11	0.31 0.19–0.45	0.02 <0.01–0.04	0.8 0.6–1.2
Rock bass	1971	10	230 145–335	0.4 0.1–0.7	<0.01 <0.01–0.02	<0.01	<0.01 <0.01–0.02	0.01 <0.01–0.04	<0.01	0.1 <0.1–0.3
Smallmouth bass	1968	5	453 283–748	2.9 2.0–3.4	0.20 0.13–0.31	0.11 0.08–0.18	0.21 0.14–0.32	0.52 0.38–0.69	<0.01	0.3 0.2–0.6
	1975	6	853 264–1491	2.5 1.1–3.6	0.60 0.09–0.92	0.11 0.02–0.16	0.05 0.02–0.09	0.76 0.13–1.15	0.09 0.03–0.14	2.1 0.4–3.1
Bluegill	1971	25	172 85–250	0.4 0.1–1.8	<0.01 <0.01–0.02	<0.01	<0.01	0.01 <0.01–0.04	<0.01	<0.1 <0.1–0.2
Black crappie	1968	6	174 116–212	2.3 0.4–6.2	0.08 0.03–0.17	0.05 0.01–0.12	0.10 0.04–0.71	0.23 0.11–0.60	<0.01	0.2 <0.1–0.5
	1971	13	199 35–455	0.3 0.2–0.6	<0.01 <0.01–0.01	<0.01	<0.01	0.01 <0.01–0.02	<0.01	<0.1 <0.1–0.2
Pumpkinseed	1968	5	118 97–137	2.8 2.3–3.7	0.03 0.02–0.03	0.03 0.02–0.04	0.05 0.04–0.06	0.11 0.08–0.13	0.01 <0.01–0.02	0.1 <0.1–0.2
	1971	22	104 40–265	0.4 0.1–0.8	0.02 0.01–0.02	0.01 0.01–0.01	0.01 0.01–0.02	0.04 0.03–0.05	<0.01 <0.01–0.01	<0.1 <0.1–0.1
Cyprinidae										
Carp	1971	8	3676 1410–9710	10.1 3.1–22.0	0.30 0.04–0.84	0.19 0.05–0.53	0.04 <0.01–0.10	0.53 0.09–1.26	0.04 <0.01–0.13	0.7 0.3–1.5
Hiodontidae										
Mooneye	1970	12	306 100–485	10.7 6.8–14.9	0.72 0.31–1.20	0.09 0.05–0.29	0.12 0.01–0.89	0.93 0.10–2.38	0.03 0.01–0.13	1.9 0.7–7.2
Ictaluridae										
Brown bullhead	1971	12	427 240–580	0.5 0.1–1.4	<0.01 <0.01–0.02	<0.01	<0.01	0.01 <0.01–0.03	<0.01	0.1 <0.1–0.3
Channel catfish	1971	6	2016 465–5275	5.2 2.6–10.2	0.36 0.14–0.75	0.10 0.03–0.14	0.08 <0.01–0.17	0.54 0.22–0.89	0.02 <0.01–0.04	2.3 0.9–3.9
Lepisosteidae										
Longnose gar	1971	12	723 320–1310	3.7 0.7–9.1	0.79 0.28–1.82	0.27 0.12–0.46	0.13 <0.01–0.21	1.19 0.48–2.35	0.02 <0.01–0.03	1.5 0.5–4.0
Percidae										
Yellow perch	1968	3	201 135–236	1.1 0.5–1.4	0.08 0.04–0.16	0.05 0.02–0.10	0.10 0.04–0.23	0.23 0.10–0.49	<0.01	<0.1
	1970	3	108 80–155	0.4 0.1–0.7	0.02 <0.01–0.2	<0.01	<0.01 <0.01–0.01	0.03 <0.01–0.05	<0.01	0.1 <0.1–0.2
	1971	11	59 35–75	0.3 0.1–0.5	0.01 <0.01–0.02	<0.01	<0.01 <0.01–0.01	0.02 <0.01–0.03	<0.01	0.1 <0.1–0.2
Walleye	1968	4	410 250–539	1.1 0.8–1.3	0.08 0.02–0.14	0.04 0.01–0.06	0.08 0.02–0.14	0.20 0.05–0.34	<0.01	<0.1
	1971	20	401 120–1990	2.2 0.1–5.1	0.05 <0.01–0.17	0.05 <0.01–0.25	0.01 <0.01–0.04	0.11 <0.01–0.42	0.02 <0.01–0.15	0.4 <0.1–2.0
	1976	9	1726 203–3311	0.8 0.3–2.5	0.06 <0.01–0.21	0.01 ND–0.05	0.01 ND–0.04	0.08 <0.01–0.28	0.01 ND–0.01	0.2 <0.1–0.8
Sciaenidae										
Freshwater drum	1971	12	519 225–1130	1.6 0.3–6.1	0.01 <0.01–0.04	<0.01 <0.01–0.02	0.01 <0.01–0.05	0.03 <0.01–0.09	<0.01	0.2 <0.1–0.6
	1976	10	259 86–521	1.7 0.6–4.4	0.02 <0.01–0.04	<0.01 <0.01–0.02	<0.01 ND–0.01	0.03 0.01–0.05	<0.01	0.2 <0.1–0.3

NOTE: ND = none detected.
[1] Number of analyses represents number of individual fish (278).
[2] Eviscerated fish with heads and tails removed.

residue of 0.03 ppm ΣDDT was present in freshwater drum caught in 1971 and 1976, and little change occurred among different weight classes (Tables 2, 3).

Walleye caught in three separate years had steadily declining ΣDDT residues: 0.20 ppm in 1968, 0.11 ppm in 1971, and 0.08 ppm in 1976. By weight class, a

TABLE 3. *Comparison of residues by weight class of six fish species caught in Lakes Saint Clair and Erie, 1968–76*

Species	Lake[1]	Year	Weight Class, kg	Number of Analyses[2]	Mean Weight, g	Mean Fat, %	Mean Content of Residues in Tissue, ppm ΣDDT	Dieldrin	PCBs
Smallmouth bass	Saint Clair	1968	0.25–0.50	4	378	2.8	0.48	0.006	0.38
			0.50–0.75	1	748	3.3	0.67	ND	0.26
		1975	0.25–0.50	1	264	2.5	0.13	0.030	0.40
			0.50–0.75	1	698	1.1	0.28	0.030	0.90
			0.75–1.00	2	826	3.5	0.93	0.125	2.60
			1.00+	2	1252	2.1	1.13	0.120	3.00
	Erie	1968	0–0.25	9	206	1.6	0.52	0.004	0.28
	(E)		0.25–0.50	7	374	2.0	1.24	0.009	0.41
		1971	1.25–1.50	2	1449	7.5	1.20	0.002	5.80
		1972	0–0.25	8	89	2.2	0.12	0.006	0.35
			0.25–0.50	8	412	3.5	0.25	0.019	0.86
			0.50–0.75	2	624	4.0	0.33	0.025	1.00
		1975	0.25–0.50	1	480	3.8	0.12	0.050	0.40
			0.50–0.75	1	707	3.5	0.09	0.20	0.30
			0.75–1.00	3	838	3.9	0.15	0.027	0.30
			1.00+	1	1226	5.4	0.30	0.020	0.40
White bass	Erie	1968	0–0.25	7	127	2.7	0.32	0.010	0.06
	(E,W)		0.25–0.50	1	295	2.1	1.51	0.007	0.31
	(E,C,W)	1971	0–0.25	29	156	5.3	0.11	ND	1.44
			0.25–0.50	2	388	8.0	0.15	ND	3.10
	(E)	1972	0–0.25	27	124	3.2	0.15	0.014	0.65
			0.25–0.50	2	361	7.2	0.55	0.025	3.30
			0.75–1.00	1	755	9.0	0.71	0.020	4.70
	(C,W)	1975	0–0.25	3	77	7.5	0.16	0.153	0.70
			0.25–0.50	1	319	8.6	0.42	0.160	2.40
			0.50–0.75	1	607	7.3	0.56	0.190	3.20
	(E)	1976	0–0.25	6	93	3.5	0.05	0.005	0.12
			0.25–0.50	1	274	3.8	0.06	0.006	0.10
Freshwater drum	Saint Clair	1971	0–0.25	2	235	1.0	0.01	0.004	0.09
			0.25–0.50	5	405	2.0	0.03	0.005	0.22
			0.50–0.75	3	560	1.1	0.02	0.002	0.13
			1.00–1.25	2	1023	1.9	0.05	0.004	0.21
		1976	0–0.25	4	138	0.8	0.03	0.004	0.17
			0.25–0.50	5	303	2.3	0.03	0.004	0.17
			0.50–0.75	1	521	1.0	0.04	ND	0.18
	Erie	1968	0–0.25	8	120	2.6	0.23	0.005	0.05
	(W)		0.25–0.50	3	344	2.8	0.27	0.008	0.05
	(E,C,W)	1971	0–0.25	18(14)	144	5.9	0.03	ND	1.75
			0.25–0.50	7	387	9.0	0.12	ND	2.17
			0.50–0.75	3	570	6.3	0.23	ND	3.87
	(E,C,W)	1975	0–0.25	9(16)	85	3.1	0.03	0.017	0.28
			0.25–0.50	13	387	4.7	0.10	0.038	0.85
			0.50–0.75	10	606	4.9	0.17	0.036	0.57
			0.75–1.00	2	832	3.7	0.10	0.015	0.25
Yellow perch	Saint Clair	1968	0–0.25	3	201	1.1	0.24	0.003	0.09
		1970	0–0.25	3	108	0.4	0.28	0.003	0.12
		1971	0–0.25	11	59	0.3	0.02	0.003	0.11
	Erie								
	(C,W)	1968	0–0.25	23	123	1.0	0.11	0.006	0.06
	(E,C,W)	1971	0–0.25	29	112	2.1	0.04	ND	0.64
	(E)	1972	0–0.25	29	87	2.6	0.08	0.011	0.25
			0.25–0.50	1	449	4.4	0.07	0.010	0.23
	(E,C,W)	1975	0–0.25	42(111)	63	1.9	0.06	0.023	0.38
			0.25–0.50	2	384	3.4	0.11	0.035	0.45
			0.50–0.75	2	594	2.9	0.06	0.035	0.30
	(E)	1976	0–0.25	15	112	1.6	0.04	0.012	0.20
Coho salmon	Erie								
	(C)	1968	0–1.0	2	471	5.4	0.51	0.029	0.33
		1970	1.0–2.0	2	1795	12.6	4.53	0.100	5.80
			2.0–3.0	9	2263	11.6	2.40	0.080	2.10
	(C)	1971	0–1.0	3	806	11.5	1.76	0.010	1.70
	(C,W)	1975	0–1.0	1	932	0.1	0.12	0.020	0.70
			1.0–2.0	6	1823	1.1	0.15	0.035	0.90
			2.0–3.0	10	2501	0.9	0.11	0.035	0.63
			3.0–4.0	8	3369	1.3	0.20	0.050	1.06
			5.0–6.0	1	5300	2.5	0.76	0.070	2.70
		1976	0–1.0	1	515	1.7	0.09	0.014	0.26
			1.0–2.0	1	1625	2.7	0.09	0.012	0.52
			2.0–3.0	3	2641	1.9	0.11	0.011	0.28
			3.0–4.0	1	3125	1.4	0.04	0.004	0.11
Walleye	Saint Clair	1968	0–0.5	3	366	1.1	0.14	0.004	0.07
			0.5–1.0	1	539	1.2	0.34	0.004	0.12
		1971	0–0.5	15	225	2.6	0.13	0.026	0.50
			0.5–1.0	4	660	0.7	0.02	0.001	0.13
			1.5–2.0	1	1990	1.2	0.06	0.003	0.17
		1976	0–0.5	1	203	2.5	0.18	0.011	0.75
			0.5–1.0	2	678	0.8	0.04	ND	0.11
			1.5–2.0	3	1690	0.3	0.02	ND	0.09
			2.0–2.5	1	2496	0.7	0.12	0.008	0.28
			3.0–3.5	2	3204	0.8	0.15	0.055	0.32

(Continued next page)

TABLE 3 (cont'd.). *Comparison of residues by weight class of six fish species caught in Lakes Saint Clair and Erie, 1968–76*

Species	Lake[1]	Year	Weight Class, kg	Number of Analyses[2]	Mean Weight, g	Mean Fat, %	Mean Content of Residues in Tissue, ppm ΣDDT	Dieldrin	PCBs
	Erie								
	(W)	1968	0–0.5	3	265	2.1	0.24	0.005	0.08
			0.5–1.0	3	773	3.7	0.42	0.010	0.24
	(E,W)	1971	0–0.5	4	369	3.7	0.04	ND	0.89
			0.5–1.0	1	725	3.0	0.02	ND	1.10
	(W)	1975	0–0.5	1(5)	72	3.7	0.13	0.054	0.66
			0.5–1.0	1	906	3.9	0.15	0.060	0.70
			2.0–2.5	2	2175	21.3	1.32	0.360	4.60

[1] E = eastern basin; C = central basin; W = western basin.
[2] Analyses performed on single fish in most cases; in some cases composite samples were analyzed, and the number of fish is in parentheses.

marked drop was noted for ΣDDT between 1968 and 1971, but thereafter the decline was small (Tables 2, 3).

Dieldrin—Mean residues in all species were less than 0.10 ppm. In addition, 12 species had mean residues at or below 0.01 ppm dieldrin. The highest mean residue of 0.09 ppm was present in smallmouth bass caught in 1975 (Table 2). By weight class, smallmouth bass exhibited an increase in dieldrin residues between 1968 and 1976 (Table 3). Only three other species, carp, mooneye, and walleye, had individual fish with residues of 0.10–0.15 ppm dieldrin (Table 2). Dieldrin residues in walleye peaked in 1971 and declined by 1976 (Table 3). Freshwater drum, the only other species caught in the early and late years, showed no change in dieldrin residues (Tables 2, 3).

PCBs—Only one fish from Lake Saint Clair, a 435-g mooneye, exceeded the 5.0 ppm tolerance level for PCB residues in fish tissues set by the Food and Drug Administration, U.S. Department of Health, Education, and Welfare. However, several species and individual fish exceeded the 2.0 ppm Health and Welfare Canada tolerance level (Table 2). Smallmouth bass caught in 1975 and channel catfish caught in 1971 had mean residues of 2.1 ppm and 2.3 ppm PCBs, respectively (Table 2). In 1970, one of six largemouth bass (*Micropterus salmoides*), four of 12 mooneye, and one of eight redhorse (*Moxostoma* sp.) contained 2.1–7.2 ppm PCBs. In 1971, three of 11 longnose gar, and four of six channel catfish caught in Tremblay Creek and in Mitchell Bay, respectively, had residues of 2.0–4.0 ppm PCBs. In 1975, PCB levels in four of six smallmouth bass ranged from 2.2 to 3.1 ppm.

PCB residues increased in smallmouth bass between 1968 and 1975. However, freshwater drum and walleye showed little change even by weight class (Tables 2, 3).

HCB—Forty-eight fish of 17 species caught in 1970–71 and analyzed in 1973 had detectable HCB residues below 0.1 ppm. Redhorse mullet had the highest mean residue, 0.024 ppm, and the highest residue in a single fish, 0.08 ppm. Carp, channel catfish, and yellow perch (*Perca flavescens*) had the second highest residues of 0.013 ppm HCB (Table 4).

Chlordane and heptachlor epoxide—The same 48 fish caught in 1970–71 were analyzed for *cis*- and *trans*-chlordane and heptachlor epoxide. Interfering compounds prevented confirmation of chlordane below 0.05 ppm. Smallmouth bass caught in 1975 contained low levels of chlordane but these could not be satisfactorily separated from interfering compounds. By 1976, both chlordane and heptachlor epoxide were identified at low levels in freshwater drum and walleye (Table 5).

TABLE 4. *Hexachlorobenzene residues in 17 species of fish (48 fish) caught in Lake Saint Clair, 1970–71*

Fish Species	Year	No. of Fish	Average Weight, g	HCB, ppm Mean	HCB, ppm Range	Location
Largemouth bass	1970–71	2	683	0.005	0.002–0.008	Mitchell Bay
Rock bass	1971	3	220	0.008	0.002–0.013	Tremblay Creek
Bluegill	1971	6	170	0.002	<0.001–0.004	Mitchell Bay
Bowfin	1971	2	1630	0.008	0.005–0.015	Mitchell Bay
Brown bullhead	1971	3	412	0.003	0.002–0.003	Mitchell Bay
Carp	1971	2	2890	0.013	0.006–0.020	Mitchell Bay
Channel catfish	1971	2	1910	0.013	0.005–0.020	Tremblay Creek, Mitchell Bay
Black crappie	1971	3	243	0.002	0.001–0.003	Mitchell Bay
Freshwater drum	1971	3	623	0.006	0.002–0.008	St. Lukes Bay
Longnose gar	1970	1	1195	0.007		Tremblay Creek
Mooneye	1970	2	158	0.009		Tremblay Creek
Yellow perch	1971	2	55	0.013	0.007–0.019	Mitchell Bay
Pumpkinseed	1971	5	123	0.001	<0.001–0.002	Mitchell Bay
Quillback	1970–71	4	1670	0.008	0.005–0.010	Mitchell Bay
Redhorse mullet	1970–71	4	670	0.024	0.002–0.080	Bassett Channel, Mitchell Bay
White sucker	1970	3	1185	0.004	0.003–0.006	Bassett Channel
[illegible]	1971	1	120	0.002		Mitchell Bay

TABLE 5. *Chlordane and heptachlor epoxide residues in fish species caught in Lakes Saint Clair and Erie, 1972–76*

Lake	Fish Species	Year	No. of Fish[1]	Mean Weight, g	Mean Fat, %	Mean Content of Residues in Fish Tissues, ppm Chlordane[2]	Heptachlor Epoxide
Saint Clair	Freshwater drum	1976	10	259	1.7	0.011	0.003
						ND–0.080	ND–0.013
	Walleye	1976	9	1726	0.8	0.008	0.004
						ND–0.028	ND–0.013
Lake Erie							
Central basin	Rainbow trout[3]	1974	5	642	4.4	ND	0.006
							ND–0.033
Eastern basin	White bass	1972	11	156	5.9	0.023	ND
						0.010–0.050	
		1976	7	118	3.6	0.010	0.004
						0.008–0.011	0.002–0.007
	Yellow perch	1972	10	133	3.2	0.011	ND
						<0.001–0.020	
		1975	15	60	2.1	0.007	0.001
						0.002–0.016	ND–0.006
		1976	15	121	1.6	0.007	0.003
						<0.001–0.014	<0.001–0.007
	Coho salmon	1976	6	2198	11.5	0.037	0.007
						0.011–0.045	0.002–0.010
	Emerald shiner	1976	4(12)	5.6	5.6	0.038	0.012
						0.011–0.050	0.006–0.016
	Rainbow smelt	1975	5	74	8.9	0.015	0.006
						0.004–0.021	0.001–0.009
		1976	10	20	5.1	0.046	0.015
						0.022–0.134	0.009–0.033

[1] See footnote 1, Table 3.
[2] NOTE: Chlordane present as *cis*- and *trans*-isomers in all species except white bass and yellow perch caught in 1972. Then, only *cis*-chlordane was confirmed.
[3] Three rainbow trout caught in Silver Creek also contained endosulfan with mean residue of 0.025 ppm (0.007–0.050 ppm). NOTE: ND=not detected.

Other organochlorines—No endrin or methoxychlor was detected in fish caught in Lake Saint Clair. Samples were analyzed for mirex in 1975–76, but no residues were detected in smallmouth bass, freshwater drum, or walleye caught in those years.

LAKE ERIE

ΣDDT—No mean residues of ΣDDT for any species caught in Lake Erie in 1968–76 exceeded the 5.0 ppm United States and Canadian tolerance levels. Three coho salmon caught in 1970 in the central basin contained levels of 8.23, 7.67, and 7.61 ppm ΣDDT, and the whole catch of 11 fish averaged 2.80 ppm (Table 6). These three fish were the largest, weighing 1,963, 2,276, and 2,640 g, respectively. Three coho salmon caught in 1971 from the same basin and weighing an average of 806 g contained only 1.76 ppm ΣDDT.

Smallmouth bass caught in 1971 from the eastern basin was the only other species with mean residues above 1.0 ppm; mean residues were 1.2 ppm ΣDDT. Smallmouth bass caught in 1968 from the same basin averaged 0.83 ppm; however, two of 16 fish had 1.53 ppm and 4.28 ppm ΣDDT. White bass and walleye had individual fish with residues above 1.0 ppm.

Five species were caught in all three basins during the same year, and one of these, coho salmon, was caught in three basins over two years (Tables 3, 6, 7). Emerald shiner (*Notropis atherinoides*) and yellow perch, which are localized species, contained residues of ΣDDT that were not significantly different among the three basins. In migrating species of white bass, freshwater drum, coho salmon, and rainbow smelt (*Osmerus mordax*), ΣDDT residues were similar for catches in the three basins. Where differences occurred, the higher residues correlated with fish size rather than with basin. The highest residues of ΣDDT from the central, eastern, and western basins, respectively, were freshwater drum caught in 1971 and 1975 and coho salmon caught in 1975. In all three cases, the individual fish were 1.5–4 times heavier than members of the same species from the other basins, and a correlation was evident between increasing weight and increasing ΣDDT residue; these differences virtually disappear when similar weight classes are compared among the basins (Tables 3, 6, 7).

Six species were divided into weight classes to determine the extent of decline in ΣDDT residues between 1968 and 1976 (Tables 3, 7). In the eastern basin, smallmouth bass, which were caught in four separate years, offered the best example. ΣDDT mean residues for the species peaked in 1971 and declined thereafter (Table 6); when compared by weight class, however, species showed a decline in ΣDDT from 1968 to 1976 (Table 3). Declining residues of ΣDDT in the eastern basin were evident in rock bass (*Ambloplites rupestris*), white bass, and yellow perch but not in rainbow smelt or freshwater drum (Tables 3, 6).

In the central basin, ΣDDT residues in coho salmon peaked in 1971 and declined thereafter. Residues also declined in freshwater drum and rainbow smelt but not

TABLE 6. *Organochlorine residues in 19 fish species caught in Canadian waters of Lake Erie (1968–76) and segregated into western, central, and eastern basins*

Fish Species	Year	Basin	No. of Analyses[1]	Mean and Range Weight, g	Mean and Range Fat, %	Mean Content and Range of Contaminants in Fish Tissue, ppm DDE	TDE	DDT	ΣDDT	Dieldrin	PCBs
Centrarchidae											
Largemouth bass	1975	East	12	409	3.9	0.13	0.04	<0.01	0.18	0.02	0.1
				254–835	2.3–7.1	0.03–0.33	0.01–0.14	<0.01–0.03	0.07–0.50	<0.01–0.08	0.1–0.3
Rock bass	1968	East	7	91	1.7	0.06	0.03	0.03	0.12	0.01	0.2
				84–113	0.8–2.6	0.01–0.14	<0.01–0.07	<0.01–0.07	0.02–0.28	<0.01–0.02	<0.1–0.5
	1971	East	8	180	1.9	0.09	ND	ND	0.09	ND	0.3
				101–239	1.2–2.7	0.02–0.13			0.02–0.13		0.2–0.6
Smallmouth bass	1968	East	16	280	1.8	0.31	0.11	0.41	0.83	<0.01	0.3
				162–478	0.9–4.2	0.11–1.60	0.04–0.44	0.15–2.24	0.32–4.28	<0.01–0.03	0.2–0.84
	1971	East	2	1449	7.5	0.90	0.13	0.17	1.20	<0.01	5.8
				1376–1522	5.7–9.3	0.50–1.30	0.03–0.23	0.05–0.28	0.55–1.81		2.3–9.3
	1972	East	18	292	3.0	0.12	0.03	0.05	0.20	0.01	0.7
				76–697	1.9–5.0	0.07–0.27	0.01–0.07	0.01–0.13	0.01–0.42	<0.01–0.03	0.4–1.2
	1975	East	6	821	4.1	0.11	0.03	0.02	0.16	0.03	0.3
				480–1226	2.5–5.4	0.04–0.20	0.01–0.25	<0.01–0.05	0.05–0.30	0.01–0.05	0.2–0.4
Bluegill	1968	East	4	209	0.5	<0.01	<0.01	<0.01	0.02	<0.01	<0.1
				97–341	0.4–0.5	<0.01–0.03	<0.01–0.01	<0.01–0.02	<0.01–0.06		
Black crappie	1968	East	5	111	1.2	0.06	0.04	0.04	0.14	<0.01	<0.1
				80–173	0.7–1.8	0.02–0.10	0.02–0.07	0.01–0.07	0.05–0.21		<0.1–0.1
Pumpkinseed	1968	East	6	95	1.4	0.02	0.01	0.01	0.04	<0.01	<0.1
				79–113	0.9–1.9	0.01–0.02	<0.01–0.01		0.03–0.05	<0.01–0.01	<0.1–0.1
Clupeidae											
Alewife	1971	East	7	101	23.2	0.24	ND	ND	0.24	<0.01	3.0
				93–108	19.4–25.5	0.12–0.29			0.12–0.29		1.9–3.7
	1975	West	2(21)	40	8.6	0.05	0.08	ND	0.13	0.07	0.5
				34–51	8.1–8.9	0.04–0.06	0.07–0.09		0.11–0.15		0.4–0.5
		Central	5(22)	39	21.8	0.05	0.09	ND	0.14	0.08	0.4
				20–66	14.1–31.9	0.03–0.07	0.03–0.15		0.06–0.22	0.02–0.15	0.3–0.4
Gizzard shad	1968	West	6	234	9.4	0.06	0.17	0.09	0.32	0.02	0.3
				37–302	4.4–12.6	0.04–0.08	0.10–0.25	0.06–0.15	0.20–0.47	<0.01–0.04	<0.1–0.6
	1971	West	3(6)	92	15.5	0.07	ND	ND	0.07	ND	2.6
				74–105	13.8–16.7	0.06–0.07			0.06–0.07		2.1–3.5
		Central	3(9)	72	15.3	0.14	ND	ND	0.14	ND	3.4
				67–77	11.8–18.6	0.08–0.19			0.08–0.19		2.4–4.7
	1975	West	4(27)	136	11.1	0.04	0.09	ND	0.13	0.08	0.7
				110–157	10.1–12.0	0.03–0.06	0.06–0.11		0.09–0.17	0.06–0.10	0.6–0.9
		Central	2(7)	63	12.0	0.05	0.09	ND	0.14	0.08	0.5
				47–69	4.7–12.6		0.09–0.10		0.14–0.15	0.07–0.09	0.4–0.6
Cyprinidae											
Emerald shiner	1975	West	3(60)	4.2	6.7	0.06	0.06	ND	0.12	0.05	0.6
				4.0–4.5	5.6–7.7	0.05–0.07	0.05–0.07		0.10–0.14	0.05–0.06	0.5–0.7
		Central	3(60)	6.4	7.7	0.04	0.05	ND	0.09	0.04	0.3
				4.5–8.9	5.3–9.0	0.03–0.05	0.04–0.08		0.07–0.13	0.03–0.06	
		East	4(12)	5.6	5.6	0.08	0.02	0.02	0.12	0.02	0.4
				2.5–10.0	4.0–8.0	0.07–0.10	0.01–0.04	0.01–0.02	0.10–0.16	ND–0.03	0.3–0.6
Spottail shiner	1975	West	3(60)	11.1	3.8	0.05	0.06	ND	0.11	0.04	0.06
				5.7–16.0	3.5–4.0	0.03–0.07	0.04–0.08		0.07–0.15	0.03–0.06	0.04–0.07
Ictaluridae											
Brown bullhead	1968	East	4	149	0.9	0.02	0.02	0.01	0.05	<0.01	<0.1
				95–183	0.2–1.4	<0.01–0.04	<0.01–0.03	<0.01–0.02	0.01–0.10		
Channel catfish	1968	West	4	105	3.5	0.13	0.18	0.15	0.46	0.01	0.2
				74–135	2.1–4.5	0.09–0.18	0.13–0.26	0.12–0.20	0.34–0.64	<0.01–0.01	<0.1–0.2
	1971	West	2	518	19.3	0.16	ND	ND	0.16	ND	5.0
				356–680	17.7–20.9	0.14–0.18			0.14–0.18		4.2–5.7
Rainbow smelt	1968	Central	4(13)	24	2.2	0.05	0.04	0.09	0.18	0.01	0.2
				16–30	1.7–3.5	0.03–0.07	0.03–0.06	0.07–0.10	0.13–0.22	<0.01–0.02	0.2–0.3
	1971	East	2(7)	36	5.9	0.09	ND	ND	0.09	ND	1.3
				33–41	3.8–7.5	0.07–0.12			0.07–0.12		1.2–1.4
	1973	West	1(10)	26	1.1	0.13	0.14	0.04	0.31	0.06	0.5
	1975	West	9(60)	29	3.4	0.04	0.04	ND	0.08	0.03	0.4
				22–45	3.0–4.4	0.02–0.07	0.04–0.08		0.05–0.15	0.02–0.06	0.2–0.6
		Central	6(70)	16	3.5	0.03	0.03	ND	0.06	0.03	0.1
				13–20	3.1–3.8	0.02–0.05	0.03–0.04		0.05–0.08	0.02–0.03	
		East	8(23)	16	3.2	0.05	0.04	<0.01	0.10	0.03	0.3
				13–18	2.1–4.1	0.02–0.08	0.01–0.06	ND–0.03	0.03–0.12	0.01–0.06	0.1–0.6
	1976	East	10	30	5.1	0.05	0.04	0.02	0.11	0.05	0.3
				23–55	3.4–10.4	0.02–0.19	0.02–0.16	<0.01–0.13	0.05–0.48	0.03–0.10	<0.1–1.4
Percidae											
Yellow perch	1968	West	12	141	1.3	0.04	0.06	0.04	0.14	<0.01	<0.1
				105–216	0.4–3.4	0.01–0.11	0.01–0.14	0.01–0.11	0.06–0.36		
		East	11	108	0.8	0.03	0.02	0.05	0.10	<0.01	<0.1
				87–137	0.5–1.8	<0.01–0.06	<0.01–0.05	0.02–0.08	0.03–0.16		<0.1–0.1
	1971	West	10	116	2.0	0.02	ND	ND	0.02	ND	1.0
				82–137	1.4–2.9	<0.01–0.06			<0.01–0.06		0.2–2.6
		Central	10	102	1.7	0.03	ND	ND	0.03	ND	0.3
				79–139	1.3–2.7	0.02–0.04			0.02–0.04		0.2–0.6
		East	9	122	2.7	0.08	ND	ND	0.08	ND	0.6
				99–137	1.5–5.8	0.04–0.14			0.04–0.14		0.3–1.0

(Continued next page)

TABLE 6 (cont'd.). *Organochlorine residues in 19 fish species caught in Canadian waters of Lake Erie (1968–76) and segregated into western, central, and eastern basins*

Fish Species	Year	Basin	No. of Analyses[1]	Mean and Range Weight, g	Mean and Range Fat, %	Mean Content and Range of Contaminants in Fish Tissue, ppm DDE	TDE	DDT	ΣDDT	Dieldrin	PCBs
	1972	East	30	98	2.6	0.06	0.01	0.01	0.08	0.01	0.3
				39–449	1.0–5.8	0.03–0.10	<0.01–0.03	<0.01–0.03	0.04–0.15	<0.01–0.03	0.1–0.4
	1975	West	10(59)	40	1.7	0.03	0.04	ND	0.07	0.03	0.6
				7–84	1.4–2.0	0.01–0.07	0.02–0.07		0.03–0.14	0.02–0.07	0.4–0.9
	1975	Central	15(30)	118	2.9	0.02	0.02	<0.01	0.05	0.02	0.2
				32–605	0.8–3.9	<0.01–0.05	ND–0.07	ND–<0.01	<0.01–0.11	ND–0.05	<0.1–0.8
		East	21(26)	85	2.6	0.03	<0.01	<0.01	0.05	<0.01	0.1
				32–210	0.8–3.9	<0.01–0.13	ND–0.03	ND–0.01	<0.01–0.15	ND–0.02	0.1–0.8
	1976	East	15	121	1.6	0.02	0.01	<0.01	0.04	0.01	0.2
				69–212	0.6–3.5	0.01–0.04	<0.01–0.03	ND–0.01	0.02–0.07	<0.01–0.03	<0.1–0.8
Walleye	1968	West	6	519	2.9	0.11	0.12	0.10	0.33	<0.01	0.2
				256–923	1.7–4.1	0.06–0.16	0.07–0.21	0.06–0.15	0.19–0.46	<0.01–0.2	<0.1–0.3
	1971	West	4	460	4.0	0.03	ND	ND	0.03	ND	1.0
				158–362	1.9–5.4	0.02–0.03			0.02–0.03		0.5–1.6
		East	1	362	1.9	0.06	ND	ND	0.06	ND	0.6
	1975	West	8(14)	430	6.1	0.15	0.12	0.02	0.29	0.10	1.3
				57–2275	1.8–22.2	0.05–0.99	0.05–0.66	<0.01–0.19	0.10–1.84	0.03–0.45	0.3–5.1
Salmonidae											
Coho salmon	1968	Central	2	471	5.4	0.19	0.12	0.20	0.51	0.03	0.3
				410–531	4.0–6.8	0.17–0.20	0.10–0.14	0.18–0.22	0.49–0.53	0.02–0.04	0.2–0.4
	1970	Central	11	2178	11.8	1.05	0.92	0.83	2.80	0.09	4.0
				1627–2640	9.7–13.6	0.31–3.16	0.25–2.70	0.21–2.37	0.77–8.23	0.03–0.20	1.0–14.0
	1971	Central	3	806	11.5	0.81	0.53	0.42	1.76	0.01	1.7
				748–908	11.0–12.1	0.77–0.85	0.24–0.74	0.36–0.45	1.45–1.99	0.01–0.02	1.5–2.0
	1975	West	9	3081	2.7	0.24	0.10	ND	0.34	0.08	1.4
				1798–5300	0.6–4.9	0.08–0.73	0.02–0.15		0.10–0.76	0.02–0.12	0.6–2.7
		Central	17	2436	0.3	0.09	0.02	ND	0.11	0.02	0.7
				1773–3520	0.1–1.0	0.05–0.21	<0.01–0.10		0.06–0.30	0.01–0.07	0.4–2.0
	1976	East	6	2198	1.9	0.05	0.02	0.02	0.09	0.01	0.3
				515–3125	1.3–2.7	0.03–0.07	0.01–0.03	<0.01–0.03	0.04–0.13	<0.01–0.02	0.1–0.5
Rainbow trout	1974	Central	5	642	4.3	0.13	0.06	<0.01	0.20	0.07	0.3
				93–1691	2.7–6.1	0.02–0.43	<0.01–0.26		0.03–0.69	<0.01–0.26	<0.1–0.8
Sciaenidae											
Freshwater drum	1968	West	11	181	2.7	0.05	0.10	0.07	0.22	<0.01	<0.1
				41–380	0.5–6.4	0.03–0.11	0.05–0.17	0.04–0.11	0.12–0.38	<0.01–0.01	
	1971	West	5(9)	106	5.8	0.04	ND	ND	0.04	ND	1.4
				82–208	3.8–7.3	0.01–0.12			0.01–0.12		0.7–3.5
		Central	9	407	7.8	0.17	ND	ND	0.17	ND	3.7
				173–688	3.8–11.2	0.07–0.39			0.07–0.39		2.2–4.7
		East	10	239	7.3	0.03	ND	ND	0.03	ND	1.3
				139–390	5.5–10.2	<0.01–0.07			<0.01–0.07		0.6–1.8
	1975	West	16(23)	255	4.4	0.03	0.03	ND	0.06	0.03	0.6
				14–674	1.5–8.3	0.01–0.07	<0.01–0.07		0.02–0.14	0.01–0.07	0.2–1.8
		Central	8	345	5.5	0.05	0.06	ND	0.11	0.05	0.7
				123–575	2.1–9.1	0.02–0.12	0.04–0.10		0.07–0.19	0.03–0.08	0.4–1.4
		East	10	612	4.3	0.12	0.03	0.01	0.16	0.03	0.4
				399–856	1.9–7.2	0.06–0.30	<0.01–0.09	<0.01–0.05	0.06–0.42	0.01–0.04	0.2–0.6
Serranidae											
White bass	1968	West	6	161	3.0	0.17	0.24	0.16	0.57	<0.01	0.1
				117–295	2.1–4.2	0.04–0.41	0.12–0.60	0.08–0.40	0.23–1.41		<0.1–0.3
		East	2	110	1.3	0.04	0.02	0.12	0.18	0.02	<0.1
				107–113	1.2–1.5	0.03–0.05	0.01–0.02	0.07–0.16	0.11–0.23	0.01–0.02	
	1971	West	10	230	6.5	0.09	ND	ND	0.09	ND	2.2
				163–401	3.2–10.0	0.03–0.19			0.03–0.19		1.1–4.8
		Central	11	160	6.5	0.13	ND	ND	0.13	ND	1.6
				110–232	4.3–11.2	0.10–0.17			0.10–0.17		0.9–2.2
		East	10	127	3.6	0.10	ND	ND	0.10	ND	0.8
				92–199	2.6–4.8	0.05–0.17			0.05–0.17		0.5–1.4
	1972	East	30	161	3.7	0.12	0.04	0.03	0.19	0.01	1.0
				54–755	0.5–9.0	0.07–0.44	<0.01–0.33	<0.01–0.17	0.08–0.84	<0.01–0.04	0.5–5.4

NOTE: Fish eviscerated, heads and tails removed; alewife, shiner and smelt analyzed whole.
[1] See footnote 1, Table 3.

in yellow perch or gizzard shad (*Dorosoma cepedianum*) (Tables 3, 6, 7).

In the western basin, good examples were not available to show trends, and decline of ΣDDT residues were not so obvious. ΣDDT generally declined in channel catfish, freshwater drum, yellow perch, and rainbow smelt, but not in white bass (Table 6). To observe a decline in ΣDDT for walleye, similar weight classes must be compared (Table 3).

Dieldrin—Only white bass and walleye caught in 1975 contained mean residues of dieldrin at or above 0.1 ppm. Three white bass from the western basin had dieldrin levels of 0.12–0.19 ppm, and two from the central basin had 0.17 ppm dieldrin. Two walleye in a catch of 14 fish from west of Pelee Island had the highest resdues, 0.27 ppm and 0.45 ppm. These two fish were the largest of the catch (2.0 kg and 2.3 kg) and contained 20–22 percent fatty tissue (Table 6).

TABLE 7. *Six species of fish caught in all three basins of Lake Erie in either the same year or in a two-year period (1971, 1975–76)*

Fish Species (Year)	Weight Class, kg	Basin[1]	Average Weight, g	Mean Fat, %	Mean Content in Tissue, ppm			Number of Fish[3]
					ΣDDT	Dieldrin[2]	PCBs	
White bass	0–0.25	W	191	4.9	0.07	ND	1.73	8
(1971)		C	219	6.5	0.13	ND	1.62	11
		E	127	3.6	0.10	ND	0.80	10
Freshwater drum	0–0.25	W	106	5.0	0.04	ND	1.40	5(9)
(1971)		C	176	8.5	0.11	ND	5.50	2
		E	183	6.4	0.02	ND	1.14	7
(1975)	0.25–0.50	W	371	4.7	0.09	0.041	0.09	7
		C	439	2.8	0.11	0.020	0.40	3
		E	372	6.8	0.11	0.050	1.20	3
	0.50–0.75	W	592	4.3	0.07	0.033	0.60	3
		C	628	5.5	0.23	0.034	0.40	5
		E	574	4.4	0.16	0.045	0.95	2
Yellow perch	0–0.25	W	116	2.0	0.02	ND	0.96	10
(1971)		C	102	1.7	0.03	ND	0.34	10
		E	122	2.7	0.08	ND	0.64	9
(1975)	0–0.25	W	40	1.7	0.07	0.030	0.60	10(59)
		C	95	1.8	0.05	0.025	0.18	11(26)
		E	85	2.6	0.05	0.007	0.10	21(26)
Emerald shiners	0–0.25	W	4.2	6.7	0.12	0.053	0.63	3(60)
(1975)		C	6.4	7.7	0.09	0.043	0.30	3(60)
		E	5.6	5.6	0.12	0.021	0.41	4(12)
Rainbow smelt	0–0.25	W	29	3.4	0.08	0.030	0.45	9(60)
(1975)		C	16	3.5	0.06	0.029	0.10	6(70)
		E	16	3.2	0.10	0.029	0.34	8(23)
Coho salmon	1.0–2.0	W	1.9	2.9	0.31	0.075	1.35	2
(1975–76)		C	1.8	0.3	0.08	0.015	0.70	4
		E	1.6	2.7	0.09	0.012	0.52	1
	2.0–3.0	W	2.8	2.1	0.25	0.068	1.05	4
		C	2.3	0.3	0.08	0.013	0.55	6
		E	2.6	1.9	0.11	0.011	0.28	3
	3.0–4.0	W	3.8	4.3	0.32	0.095	1.40	2
		C	3.2	0.5	0.15	0.035	0.95	6
		E	3.1	1.4	0.04	0.004	0.11	1

[1] W=western, C=central, E=eastern.
[2] ND=not detected.
[3] See footnote 1, Table 3.

Three species, rainbow trout (*Salmo gairdneri*) caught in 1974, and alewife and coho salmon caught in 1975, had mean residues of dieldrin below 0.1 ppm. Only a few members of these species had levels above 0.1 ppm.

Although differences in dieldrin residues among basins are not apparent, dieldrin residues did increase in 1968–71 and 1975–76, as exhibited by alewife, smallmouth bass, white bass, freshwater drum, yellow perch, gizzard shad, and walleye (Tables 3, 6, 7).

PCBs—In the eastern basin, only smallmouth bass caught in 1971 had mean residues of PCBs above the 5.0 ppm U.S. tolerance limit; channel catfish from the western basin averaged 5.0 ppm in the same year (Table 6). In addition, species of white bass caught in the eastern basin in 1972, coho salmon caught in the central basin in 1970, and walleye caught in the western basin in 1975 had individual members whose PCB residues exceeded 5.0 ppm.

Species other than smallmouth bass and channel catfish which had PCB mean residues exceeding the 2.0 ppm Canadian tolerance limit were: alewife from the eastern basin (1971), white bass from the western basin (1971, 1975), freshwater drum from the central basin (1971), coho salmon from the central basin (1970), and gizzard shad from both western and central basins (1971). Among other catches in which the mean residue was below 2.0 ppm PCBs but individual fish exceeded the 2.0 ppm tolerance limit were white bass from the eastern basin (1972), freshwater drum from the western basin (1971), yellow perch from the western basin (1971), and coho salmon from the western basin (1975).

There was no correlation between highest mean residue of PCBs in a species and the basin in which it was caught. In 1971, three species were caught in all three basins. Alewife had the highest mean residues (3.0 ppm) of the eastern basin species; freshwater drum had the highest mean residues (3.7 ppm) of the central basin species; and yellow perch had the highest mean residues (1.0 ppm) of western basin species (Table 6).

In the western basin, white bass and walleye showed increased residues of PCBs between 1968 and 1975 both for mean residues and residues by weight class (Tables 3, 6). Freshwater drum and gizzard shad contained residues of PCBs that increased between 1968 and 1971 and declined in 1975. PCB residues also declined in yellow perch between 1971 and 1975 (Tables 3, 6, 7).

In the central basin, PCBs in coho salmon peaked in

1970 and then declined until 1975. This was true for similar weight classes. PCB residues declined in tissues of white bass, freshwater drum, and gizzard shad in the central basin. No species showed increasing PCB residues, but yellow perch and rainbow smelt, which had low residues in 1968, showed little change in tissue residues by 1975.

In the eastern basin, mean residues of PCBs in smallmouth bass and yellow perch reached a maximum level in 1971 and then declined (Table 6). However, a comparison of fish by weight classes showed that residues of PCBs peaked in 1972 and have shown little change since (Table 3). White bass, freshwater drum, and rainbow smelt had their highest residues in 1971–72 and declined by 1975.

Chlordane and heptachlor epoxide—Residues of *cis*- and *trans*-chlordane were first determined in 1972. In that year, *cis*-chlordane was positively identified in only two species, white bass and yellow perch from Long Point Bay, Lake Erie (Table 5). The presence of *trans*-chlordane in the two species was suspected but not confirmed. Chlordane residues in other fish caught between 1972 and 1974 were not confirmed because of the interference of other compounds on the chromatogram.

In 1975–76, both *cis*- and *trans*-isomers of chlordane were detected in white bass, yellow perch, coho salmon, emerald shiner, and rainbow smelt; highest residues were found in rainbow smelt in 1976. Chlordane was also suspected in other species. However, levels were either too low to be confirmed or interfering substances made separation and identification difficult. In 1976, several species were analyzed for oxchlordane but it was not detected.

Heptachlor epoxide was first positively identified in rainbow trout caught in Silver Creek draining into the central basin (Table 5). In 1975 and 1976, residues of heptachlor epoxide were also identified in white bass, yellow perch, coho salmon, emerald shiner, and rainbow smelt. As with chlordane, the highest residues of heptachlor epoxide were found in rainbow smelt.

Other organochlorine compounds—Endosulfan was identified in rainbow trout caught in Silver Creek in 1974 (Table 5). Neither endrin nor methoxychlor was identified in any fish caught in Lake Erie. Mirex analysis was added in 1975–76, but no measurable residues were detected.

Discussion

SEDIMENT AND FISH RESIDUES

Sediments in Lake Erie were five to ten times more highly contaminated with ΣDDT, dieldrin, and PCBs than were sediments from Lake Saint Clair, mostly because sediment is transitory through Lake Saint Clair but accumulates in the basins of Lake Erie (*5*). Fish tissue residues of ΣDDT and dieldrin did not necessarily show this trend, but PCBs were higher in fish from Lake Erie. For example, rock bass and smallmouth bass caught in Lake Erie in 1971 had higher residues of ΣDDT than did those caught in Lake Saint Clair. The reverse was true of channel catfish caught the same year. Dieldrin residues were generally at the trace level in fish from both bodies of water. Residues of PCBs were higher in rock bass, channel catfish, freshwater drum, yellow perch, and walleye caught in Lake Erie than those caught in Lake Saint Clair during the same year. Smallmouth bass were an exception; residues in fish caught in Lake Saint Clair were higher. Frank et al. reported that the parent compound *p,p'*-DDT was low or absent from sediments in Lake Erie (*5*). In the present study, *p,p'*-DDT was not found in many fish caught in Lake Erie.

Sediments collected from the western basin of Lake Erie contained ΣDDT and PCB residues two to three times higher than did sediments in either the central or eastern basins (*5*). Differences in residues among the same species caught during the same year in all three basins were not apparent.

FISH RESIDUES

Residues of ΣDDT were considerably higher in 1971 than those reported by Reinke et al. in Lake Saint Clair in 1970 (*13*). ΣDDT and dieldrin residues in 13 species of fish caught in 1965–68 in Lake Erie (*11*) were two to nine times higher than those in the same species caught in 1968 in the present study. Samples of gizzard shad in the two studies were similar (0.50 ppm and 0.32 ppm, respectively), but yellow perch samples were different (0.9 ppm and 0.1 ppm, respectively).

Reinke et al. (*13*) reported on ΣDDT and dieldrin residues in six species of fish from Canadian waters of Lake Erie in 1970 which were 1.5–10 times higher than those in similar species reported herein. ΣDDT residues in alewife were similar for the two studies (0.34 ppm and 0.24 ppm, respectively), but residues in freshwater drum and yellow perch were an order of magnitude different.

The site of catch can have a significant bearing on the contaminant residue level (*4*). In fish of the same species, ΣDDT and dieldrin residues were 10–15 times higher in fish caught in streams than in those caught in the lakes. Bluegill (*Lepomis macrochirus*), brown bullhead (*Ictalurus nebulosus*), pumpkinseed (*Lepomis gibbosus*), and rock bass all exhibited ΣDDT and dieldrin residues an order of magnitude higher in fish from creeks draining the tobacco belt of Ontario than in fish caught in Long Point Bay, Lake Erie (*4*).

Residues of ΣDDT, dieldrin, and PCBs reported by Carr et al. (2) in fish caught in 1970–71 correspond more closely with the residue levels reported in the present study, especially in the six species common to both surveys.

Residues of ΣDDT in coho salmon caught in 1970 during the present study correspond with those reported by Reinert and Bergman (12) for the same species caught in 1969. Coho salmon (1970) weighing 2.0 kg contained 2.2 ppm ΣDDT; coho salmon (1969) weighing 2.2 kg had 2.8 ppm ΣDDT.

Suns and Rees documented residues in spottail shiners from both the western and the eastern basins of Lake Erie (15). ΣDDT and dieldrin levels in spottail shiners from the western basin reported in the present study are similar to those of Suns and Rees (15), but PCB residues are lower by an order of magnitude. Emerald shiners caught close to the same location, however, contained similar PCB levels (0.6 ppm). Suns and Rees reported that spottail shiner are good indicators of specific site effluents of PCBs (15).

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Organochlorine Residues in Aquatic Environments in Iran, 1974

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ABSTRACT

Organochlorine pesticide residues in various organisms from different aquatic ecosystems in Iran were investigated in spring 1974. DDT levels were high in fish taken from two rivers in southern Iran, whereas low levels were detected in samples obtained from a freshwater lake in the same area. Fish from two of the reservoirs supplying Tehran with potable water contained moderate levels of DDT. The low residue level in pike collected in the Bandar-Pahlavi Mordab in northwest Iran indicates that only a small amount of organochlorine pesticides used in this area enters the pelagic food chain.

Sturgeon collected at different places in the Caspian Sea showed similar accumulations of DDT in the muscles and in the eggs. Polychlorinated biphenyls (PCBs) were detected only in samples of sediment from the drainage systems in Tehran.

Introduction

Although reports on the widespread distribution of organochlorine pesticide residues in the global ecosystem are increasing *(4, 5, 8, 10, 11, 17)*, very little is known of their occurrence, distribution, effects, and ecological significance in many developing countries. Because large quantities of pesticides are used in such countries for agriculture and in vector control programs, information is needed to evaluate the full effects and benefits of pesticidal applications.

Iran imported about 2,720 tons/year of organochlorine pesticides during 1966–75; DDT was the main import (Table 1). Consumption increased considerably during that period, and the amount of DDT compounds imported during 1974–75 was about 10 times that imported in 1966–67. Far-reaching ecological implications may be foreseen regarding the stability of the pesticides and their readiness to accumulate in food chains, especially in areas subjected to regular, intense applications.

During 1970–72, Higgins *(3)* analyzed various samples from the Caspian Sea for DDT and heavy metals. Hashemy-Tonkabony and Asadi Langaroodi *(2)* studied organochlorine pesticide residues in 14 species of fish from the Caspian Sea. The levels found by Higgins were not regarded as hazardous, but a closer study of the occurrence, distribution, and possible effects of these pesticides in selected biota was recommended.

The purpose of the present study was to monitor certain areas in Iran to evaluate the level of contamination and its significance.

Substances Investigated

Samples were analyzed for benzene hexachloride (BHC), lindane, aldrin, dieldrin, DDT, and polychlorinated biphenyls (PCBs). All except PCBs are widely used in Iran as insecticides.

The form of DDT most used in pesticide formulations contains approximately 70 percent *p,p'*-DDT and 20 percent *o,p'*-DDT; the remaining 10 percent contains at least seven different substances *(1)*. Therefore, it is assumed that DDT enters the environment mainly as *p,p'*-DDT or *o,p'*-DDT. In a study of the distribution of DDT and its metabolites in the environment, the pattern of degradation may be used to evaluate DDT input to the ecosystem.

PCBs include at least 50 different compounds, homologs, or isomers. They are not spread as pesticides, but are

TABLE 1. *Amount of chlorinated hydrocarbons imported to Iran, 1966–75*[1]

Year	Total Chlorinated Hydrocarbons, Tons	DDT Compounds, Tons
1966–67	1005	514
1967–68	850	585
1968–69	2137	1214
1969–70	1799	1168
1970–71	1965	1142
1971–72	3347	2967
1972–73	1247	517
1973–74	5841	3620
1974–75	6291	5786

[1] SOURCE: Department of the Environment and the Plant Protection Department, Tehran, Iran.

[1] Present address: Institute of Limnology, University of Lund, Sweden.

[2] Department of the Environment, P.O. Box 1430, Tehran, Iran.

used in industry as heat-transfer media, lubricants, waxes, and synthetic resins to improve chemical resistance, adhesiveness, and flexibility *(9)*. The sources of PCBs and their modes of transport into the environment are poorly understood.

Materials and Methods

SAMPLING

Samples of various organisms collected in spring 1974 were frozen and brought to the laboratory in Tehran. Sturgeon and their eggs were sampled at three places along the Iranian Caspian Sea coast. Birds and fish from Parishan Lake and the Shapour and Kupor Rivers, situated in the Shiraz area in southern Iran, were sampled. Fish from water reservoirs near Tehran were also analyzed, as well as pike from the Bandar Pahlavi region, 300 km northwest of Tehran (Figure 1).

From sturgeon, a section of the dorsal musculature just behind the gills was excised. The skin was removed, and the sample was wrapped in aluminum foil and frozen until processed. The sturgeon eggs were removed and frozen in a similar manner. From other fish, a section of the lateral body muscle from the left side of the fish, anterior to the anal openings, was taken for analysis. From the birds, the breast muscle was sampled.

1. Bandar Pahlavi (sturgeon)
2. Babulsar (sturgeon)
3. Miankaleh and Tazze Abad (sturgeon)
4. Bandar Pahlavi Mordab (pike)
5. Parishan Lake, Shapour and Kupor Rivers (fish and birds)

FIGURE 1. *Location of sampling areas in Iran*

ANALYTICAL METHODS

Organochlorine residues were extracted, cleaned, and separated and quantitated by gas chromatography by the method of Södergren *(12)*.

Samples (1–3 g) were homogenized in a 1:1 solution of acetone–hexane. After acetone was removed, the hexane extract was evaporated to 1 ml and divided into thirds for subsequent cleanup and fat determination.

Two cleanup processes, one acidic and one involving basic hydrolysis, were performed simultaneously for each sample. The compounds were chemically derivatized, and the conversion products were used to confirm the identity of the original compounds. *p,p'*-DDT and *p,p'*-TDE were treated with potassium hydroxide and quantitatively converted to *p,p'*-DDE and *p,p'*-DDMU [1-chloro-2,2-bis(*p*-chlorophenylethylene)], respectively. In the acidic treatment, dieldrin is degraded but is recovered in the potassium hydroxide–treated extract. On the other hand, lindane and benzene hexachloride (BHC) are lost in the KOH procedure, but are recovered in the acidic treatment. Neither treatment affects the PCBs.

Two hundred μl of the extract was taken for gravimetric determination of extractable lipids in the sample.

The hexane extracts were analyzed by gas-liquid chromatography on a Model 2700 Varian Aerograph equipped with a Hoechst Oxysorb filtering unit. A modified electron-capture detector was used *(13)*. The system, all glass from the injector to the detector, diminishes the risk of pyrolysis. Sensitivity was also increased over that of conventional Kovar cells. Instrument parameters and operating conditions follow.

Column:	205 cm long × 1.5 mm ID glass, packed with a 3:1 mixture of 4 percent SF-96 and 8 percent QF-1 on 100–120-mesh Chromosorb W AW/DMCS
Resolution:	approximately 1700 theoretical plates for *p,p'*-DDT
Temperatures:	column 185°C injector 225°C detector 220°C
Carrier gas:	nitrogen flowing at 25 ml/minute

The quantity of organochlorines in the samples was estimated by comparing peak heights of aliquots of purified extracts with peak heights of a known quantity of a standard solution. The results were not corrected for recovery. For the PCBs, a commercially available mixture, Clophen A50, was used as a reference.

TABLE 2. *Organochlorine residues in organisms from Parishan Lake, Kupor and Shahpour Rivers—1974*

Location	Species	Fat, %	Fresh Weight, ng/g				Fat Weight, mg/kg			
			DDE	TDE	DDT	ΣDDT	DDE	TDE	DDT	ΣDDT
Parishan Lake	*Barbus* sp.	0.4	7	ND	ND	7	1.7	ND	ND	1.7
		0.4	7	ND	ND	7	1.8	ND	ND	1.8
		0.3	3	ND	ND	3	1.2	ND	ND	1.2
	Coot, *Fulica atra*	2.4	37	ND	ND	37	1.6	ND	ND	1.6
Kupor River	*Barbus* sp.	0.8	1425	ND	180	1605	174.0	ND	22.0	196.0
		0.4	1161	261	241	1662	72.9	16.4	15.1	104.4
		0.1	251	30	39	320	482.3	57.6	74.9	614.8
Shahpour River	*Varichorhinus* sp.	2.5	3030	118	910	4058	121.2	4.7	36.4	162.3
	Barbus sp.	0.5	250	18	30	298	50.8	3.7	6.1	60.6

NOTE: ND=not detected.

Results

Only small amounts of *p,p'*-DDE were detected in fish from Parishan Lake and in a coot which was found dead (Table 2). Fish obtained from the Shahpour and Kupor Rivers contained appreciable amounts of DDT and its metabolites DDE and TDE (Table 2). The Shahpour and Kupor Rivers flow through malaria-infected areas, and DDT is used for indoor spraying.

In fish and fish eggs from two reservoirs supplying Tehran with potable water, various amounts of DDT were detected (Tables 3, 4). The levels in cyprinide fish *(Varichorhinus nikolskii)* from the Latian reservoir far exceeded those found in fish from the Karadj reservoir. The main metabolite accumulated was *p,p'*-DDE.

In samples from the Latian reservoir, the levels of DDT compounds in rainbow trout *(Salmo gairdneri)* were similar to those found in *V. nikolskii*. The *Varichorhinus* species has a shorter food chain than the *Salmo* species, resulting in a deviation from the usual pattern of biomagnification of persistent compounds.

Low levels of DDT were found in pike *(Esox lucius)* collected from the Bandar Pahlavi Mordab (Table 5). Again, the principal metabolite found was *p,p'*-DDE. The presence of only small proportions of *p,p'*-DDT suggests that the accumulation occurred over considerable time, and that the input is not recent.

In May 1974, more than 100 samples of sturgeon and their eggs were collected from two species *(Accipencer*

TABLE 3. *Organochlorine residues in fish and fish eggs from the Latian Dam, 1974*

Species	Fat, %	Fresh Weight, ng/g				Fat Weight, mg/kg			
		DDE	TDE	DDT	ΣDDT	DDE	TDE	DDT	ΣDDT
Salmo gairdneri	0.7	13	2	13	28	1.6	0.3	1.6	3.2
	0.2	650	ND	ND	650	88	ND	ND	88
	1.4	97	ND	ND	97	41	ND	ND	41
	1.0	340	ND	ND	340	24	ND	ND	24
	2.4	75	ND	ND	75	7.9	ND	ND	7.9
Varichorhinus nikolskii	1.9	178	81	8	267	9.5	4.3	0.4	14.2
	2.2	245	17	ND	262	11	0.8	ND	11.8
	0.7	92	10	ND	102	10.6	1.2	ND	11.8
	1.7	129	ND	10	139	7.6	0	0.6	8.2
	1.4	77	ND	ND	77	5.7	ND	ND	5.7
Alburnoides bipantatur	0.8	185	19	ND	204	24.2	2.5	ND	26.7
	0.5	410	26	ND	436	262.7	16.4	ND	279.1
Coregonus sp.	0.8	13	2	13	28	1.6	0.3	1.6	3.5
Eggs from *S. gairdneri*[1]	1.6	235	21	ND	256	14.3	1.3	ND	15.6

NOTE: ND=not detected.
[1]Pooled sample from six individuals.

TABLE 4. *Organochlorine residues in cyprinide,* Varichorhinus nikolskii, *from Karadj Reservoir, 1974*

Fat, %	Fresh Weight, ng/g				Fat Weight, mg/kg			
	DDE	TDE	DDT	ΣDDT	DDE	TDE	DDT	ΣDDT
0.3	11	3	ND	14	4.7	1.1	ND	5.8
0.6	23	ND	ND	23	3.8	ND	ND	3.8
0.6	11	ND	ND	11	1.7	ND	ND	1.7
0.2	8	ND	ND	8	3.7	ND	ND	3.7
0.9	22	ND	ND	22	2.4	ND	ND	2.4

NOTE: ND=not detected.

TABLE 5. *Organochlorine residues in pike,* Esox lucius, *from Bandar Pahlavi Mordab, 1974*

Age, Years	Fat, %	Fresh Weight, ng/g				Fat Weight, mg/kg			
		DDE	TDE	DDT	ΣDDT	DDE	TDE	DDT	ΣDDT
3+	0.7	3	ND	ND	3	0.4	ND	ND	0.4
3+	1.1	6	ND	ND	6	0.6	ND	ND	0.6
3+	0.6	5	ND	ND	5	1.2	ND	ND	1.2
3+	0.7	17	ND	ND	17	2.3	ND	ND	2.3
3+	0.6	3	ND	ND	3	0.6	ND	ND	0.6
3+	0.5	9	ND	ND	9	2.0	ND	ND	2.0
3+	0.7	5	ND	ND	5	0.7	ND	ND	0.7
3+	0.5	8	ND	ND	8	1.3	ND	ND	1.3
3+	0.5	2	ND	ND	2	0.4	ND	ND	0.4
3+	0.4	4	ND	ND	4	0.9	ND	ND	0.9

NOTE: ND = not detected.

TABLE 6. *Organochlorine residues in sturgeon,* Accipenser stellatus, *from Miankaleh and Tazze Abad at the Caspian Sea, 1974*

Sample	Weight, kg	Fat, %	Fresh Weight, ng/g					Fat Weight, mg/kg				
			Lindane	DDE	TDE	DDT	ΣDDT	Lindane	DDE	TDE	DDT	ΣDDT
Muscle	9.5	2.6	4	14	7		21	0.2	0.5	0.3		0.8
Eggs		17.5	13	67	16	8	91	0.1	0.4	0.1	0.1	0.6
Muscle	9.0	3.0	2	16	4	4	24	0.1	0.5	0.1	0.1	0.7
Eggs		17.3	ND	84	31	18	133	ND	0.5	0.2	0.1	0.8
Muscle	7.0	6.6	ND	276	46	149	471	ND	4.2	0.7	2.3	7.2
Eggs		16.1	23	494	484	224	1202	0.2	3.1	3.0	1.4	7.5
Muscle	8.0	4.8	7	96	19	44	159	0.2	2.0	0.4	0.9	3.3
Eggs		19.4	26	471	54	141	666	0.2	2.4	0.3	0.7	3.4
Muscle	8.5	2.0	ND	25	7	10	42	ND	1.2	0.4	0.5	2.1
Eggs		16.6	15	193	37	55	285	0.2	1.2	0.2	0.3	1.7
Muscle	10.5	3.1	4	208	97	126	431	0.2	6.7	3.1	4.1	13.9
Eggs		16.6	17	996	125	662	1783	0.1	6.0	0.8	4.0	10.8
Muscle	9.5	4.1	5	27	14	10	51	0.1	0.7	0.3	0.3	1.3
Eggs		16.9	17	79	38	27	144	0.1	0.5	0.2	0.2	0.9
Muscle	8.0	5.0	6	26	19	11	56	0.1	0.5	0.4	0.2	1.1
Eggs		19.6	23	79	44	34	157	0.2	0.4	0.2	0.2	0.8
Muscle	9.0	6.0	7	92	19	46	157	0.2	1.5	0.3	0.8	2.6
Eggs		14.3	13	204	38	77	319	0.1	1.4	0.3	0.5	2.2
Muscle	8.5	2.5	ND	55	12	20	87	ND	2.2	0.5	0.8	3.5
Eggs		14.0	16	300	48	114	462	0.2	2.1	0.3	0.8	3.2

NOTE: ND = not detected.

guldenstadti and *A. stellatus)* from three different places along the Iranian coast of the Caspian Sea (Figure 1). From these, 20 samples of *A. stellatus* of similar size and weight were analyzed (Table 6). Fat content in the muscle was 2.0–6.6 percent; corresponding range for the eggs was 12.0–19.6 percent. Calculated on the extractable lipid fraction, the average levels of DDT in muscle and egg were 3.7 ppm and 3.2 ppm, respectively. BHC and lindane were detected, but no PCBs were found.

No significant differences in the distribution of DDT and its metabolites in egg and muscle were revealed (Table 7). The range of DDT found in muscles of four species of sturgeon sampled at Babolsar in March 1974 was 1.0–13.1 ppm (Table 8). For *A. stellatus*, the mean level of DDT was 4.7 ppm.

The only samples in which PCBs were detected came from Tehran. Sediment from the drainage system along the streets contained appreciable amounts of DDT and PCBs (Table 9).

TABLE 7. *Distribution of DDT and its metabolites in muscle and eggs of sturgeon,* Accipenser stellatus*—1974*

Sample	% DDE	% TDE	% DDT
Muscle	82	38	
	72	14	14
	58	10	32
	61	12	27
	57	19	24
	48	22	30
	54	23	23
	45	36	19
	58	12	30
	63	14	23
Mean	58	20	22
Eggs	80	20	
	82	25	13
	41	40	19
	71	9	20
	71	12	17
	56	7	37
	56	22	22
	50	25	25
	64	14	22
	66	9	25
Mean	64	16	20

TABLE 8. *Organochlorine residues in sturgeon from Babolsar at the Caspian Sea, 1974*

Species	Fat, %	Fresh Weight, ng/g					Fat Weight, mg/kg				
		Lindane	DDE	TDE	DDT	ΣDDT	Lindane	DDE	TDE	DDT	ΣDDT
Accipenser guldenstadti	2.6	1	95	11	56	162	0.1	3.5	0.4	2.1	6.0
	1.7	ND	41	ND	15	56	ND	2.5	ND	0.9	3.4
	1.5	2	18	ND	7	25	0.1	1.3	ND	0.9	1.8
	0.5	1	51	1	19	71	0.1	1.0	ND	0.4	1.4
	2.4	1	37	5	21	63	0.1	1.5	0.2	0.9	2.6
	2.0	1	23	ND	1	24	0.1	1.1	ND	0.1	1.1
	3.7	1	24	3	22	49	0.1	0.6	0.1	0.6	1.2
	2.7	16	220	ND	142	362	0.5	8.0	ND	5.1	13.1
	4.8	ND	106	10	85	201	ND	2.2	0.2	1.7	4.1
A. stellatus	1.0	ND	12	ND	7	19	ND	2.2	ND	0.7	1.9
	1.2	ND	84	3.6	51	139	ND	6.7	0.3	4.1	11.1
	4.6	ND	239	ND	119	368	ND	4.9	ND	2.5	7.7
	3.7	ND	310	40	240	59	ND	3.9	0.5	3.0	7.4
	5.7	9	141	22	155	318	1.1	2.5	0.4	2.7	5.6
	5.0	ND	64	32	71	167	ND	1.2	0.7	1.3	3.2
	2.9	ND	23	ND	18	41	ND	0.8	ND	0.6	1.4
	10.4	ND	242	ND	84	326	ND	2.3	ND	0.8	3.1
	0.5	ND	15	ND	3	18	ND	2.7	ND	0.6	3.3
	1.4	ND	56	ND	31	87	ND	3.9	ND	2.1	6.0
A. nudiventris	2.9	1	27	2	8	37	0.1	0.9	0.1	0.3	1.3
	12.3		76	12	45	133		0.6	0.1	0.4	1.0
	2.7	5	28	5	18	56	0.2	1.1	0.2	0.7	2.0
Huso huso	0.6		26	ND	9	35		4.0	ND	1.3	5.4

NOTE: ND=not detected.

TABLE 9. *Organochlorine residues in sediment from street drainage systems in Tehran, 1974*

Street	Wet Weight, ng/g	
	ΣDDT	PCB
Karim Kahn Zand	85	138
Fisherabad	112	155
ShahAbbas	35	ND

NOTE: ND=not detected.

Discussion

Fish are exposed to pesticide residues not only in the water but in food and sediments. Some fish continue to accumulate residues over a period of years. Therefore, the levels in the fish may reflect their integrated history of exposure and can be used to assess the degree of pesticide contamination in a freshwater ecosystem.

Food can be a significant source of residues if the prey species has had a greater exposure in its physical environment than has its predator. However, biomagnification of persistent residues does not depend simply on position in the food chain but is basically determined by the rate at which the residue is taken up and eliminated. Although of limited statistical significance, the results from the Latian reservoir show that despite a lower trophic position *the Varichorhinus* species accumulated about the same amount of DDT as did the *Salmo* species.

DDT and its metabolites were the principal organochlorine residues detected. Aldrin or dieldrin was not found, and PCBs occurred in significant quantities only in samples collected in Tehran.

Very high levels of DDT were found in fish from the Kupor and Shahpour Rivers in southern Iran. The proportions of the DDT not metabolized, 12 percent in Kupor River samples and 16 percent in Shahpour River samples, indicate that the input of DDT to the rivers is of recent origin and/or is still occurring. DDT probably originates from the mosquito-spraying operations in these areas.

Only the indoors are sprayed. The results of this study, however, suggest a more direct contamination. Interviews with villagers indicate that, at several places, the spraying equipment was cleaned in the rivers after spraying was completed.

Very low levels of residues were found in organisms from the Parishan Lake. The levels are comparable to those found in areas subjected only to airborne contamination *(14)*. However, due to the limited number of samples processed from Parishan Lake and the Kupor and Shahpour Rivers, the results are only tentative. The distribution within these areas requires further studies. However, the high levels found in the Kupor and Shahpour Rivers may adversely affect reproduction of certain fish species.

Comparison of DDT levels in cyprinide fish from the Latian and Karadj reservoirs shows that the Latian reservoir is more exposed to pesticide contamination than is the Karadj reservoir.

Pike collected from Bandar Pahlavi Mordab show remarkably low levels of DDT in the muscles. The pike is a predatory fish and usually accumulates persistent substances readily. DDT has been used in the area for agriculture and in vector control programs. However, due to runoff, the amount of clay and soil particles in the water is extremely high. So, most DDT probably

enters the lake attached to these particles, settles to the bottom, and is not directly incorporated in the pelagic food chain.

The amounts of DDT found in the muscle and eggs of sturgeon from the Caspian Sea were similar to those reported by Higgins *(3)* but higher than those found by Hashemy-Tonkabony and Asadi Langaroodi *(2)*.

The magnitude and pattern of accumulation of DDT in sturgeon muscle and eggs is elosely related to the fat content. When calculated on a fat-weight basis, the amount accumulated in muscle and eggs of individual fish is not significantly different. Thus, the accumulation of DDT and its metabolites in muscle and eggs of the sturgeon seems to be of a similar magnitude.

It is well known that even if the DDT accumulated by fish does not harm the individual, it might be disastrous for the population. This is because DDT may, even at low levels, interfere with the reproduction of certain species *(6)*. Present levels of DDT found in the sturgeon eggs may be a threat to the sturgeon population. However, different species respond differently to the influence of accumulated compounds. Lack of experimental information on the sensitivity of sturgeon to organochlorine pesticide residues make it impossible to evaluate the present threat.

The occurrence of PCBs in various components of the global ecosystem is well documented *(4, 5, 7, 10, 11)*. In Europe, and especially in industrialized areas, PCBs are frequently found in the biota and in airborne fallout *(15, 16)*.

In Iran, however, there is not yet any sign of a widespread contamination by PCBs as indicated by the absence of these compounds in the organisms analyzed. PCBs have only been found in samples collected in Tehran, presumably originating from local runoff. Strict regulation of the PCBs and PCB-containing products might prevent their accumulation in food chains and reduce their impact on the environment.

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Chlorinated Hydrocarbon Pesticide Residues in Pacific Oysters (Crassostrea gigas) *from Tasmania, Australia—1973*

Colin Edward Sumner[1]

ABSTRACT

Pacific oysters (Crassostrea gigas *Thunberg*) *from 19 sites in Tasmania were surveyed for pesticide residues. All samples were analyzed for dieldrin and DDT, and five samples were analyzed for hexachlorobenzene (HCB) and lindane. Only DDT was found in all samples. Dieldrin levels were high in oysters from the Tamar River, but were highest (0.39 mg/kg wet weight) in samples from Ruffin's Bay. In contrast, other residue levels were low. Distribution of pesticides in Tamar River samples differed: dieldrin could be correlated with industrial uses upstream and DDT could be correlated with low-level widespread agricultural use.*

Introduction

Pacific oysters (*Crassostrea gigas* Thunberg), imported from Japan for cultivation trials, successfully reproduced themselves and colonized estuarine areas in the Tamar River, northern Tasmania (*15, 16, 17*). They represent the only commercial breeding stocks of Pacific oysters in Australia, and an oyster industry has evolved using annual spatfalls. Stick and shell cultch are set in January and later relaid on growing areas around the state. Oyster spat from the river are also sold to growers in South Australia and are being used in cultivation trials in Tongan saltwater lagoons (P. Dinamani, Fisheries Research Division, New Zealand. 1977. Personal communication).

Wild oysters abound on the shores of the Tamar River within easy access of the general public. In contrast, oyster farms are located on intertidal mud/sand flats leased from the state for private use by individuals and companies.

In February 1973, dieldrin and DDT residues in Tamar River oysters were surveyed to assess the risk of spatfall failure resulting from pesticide accumulation by adult oysters (*3, 7, 9*). The results indicated that significant levels of pesticides were present in oyster tissues, and a complete survey of major oyster beds in the Tamar River and other oyster-growing areas was commissioned to investigate more fully the risk of spatfall failure and to establish pesticide levels in oysters available to the general public.

Sampling and Analytical Methods

Oysters were collected from 14 sites in the Tamar River: four oyster farms and ten natural reefs. Samples were also taken from five farms in other areas of the state (Fig. 1). Tamar sampling sites were identical to those chosen for a heavy metal survey (*1*). Samples of 12 oysters were considered representative of the local population (*2*). Ages of cultivated oysters were noted and, when available, year classes were sampled independently.

All samples were routinely screened for dieldrin and ΣDDT residues. Five samples were analyzed also for hexachlorobenzene (HCB) and lindane. DDT here includes ΣDDT resdues and *p,p'*-TDE and *p,p'*-DDE. Analyses were performed at the Public Health Service Analysts Laboratory, Hobart, Tasmania.

Shucked undrained oyster meats were stored at −18°C, in mason jars. Before analysis, they were homogenized in an electric blender. Oyster meats were combined with a desiccant, anhydrous sodium sulfate (1:3, wet weight), and alternately blended and chilled until smooth flowing. Standard procedures were followed for cleaning high-moisture nonfatty foods (*18*). Aliquots were extracted with acetonitrile and were diluted with water before hexane partitioning. The hexane extract was back-washed with distilled water and filtered through a Florisil column.

The column was packed with activated magnesium silicate and topped with 1 cm of anhydrous sodium sulfate. Residues were eluted from the column with 6 percent and 15 percent ethyl ether in petroleum ether. The 6 percent eluate was used directly to determine DDT residues, HCB, and lindane. The 15 percent eluate was concentrated and subjected to additional cleanup through a new Florisil column.

Samples eluted from the Florisil columns were identified and quantitated by using a Varian Model 1400 gas

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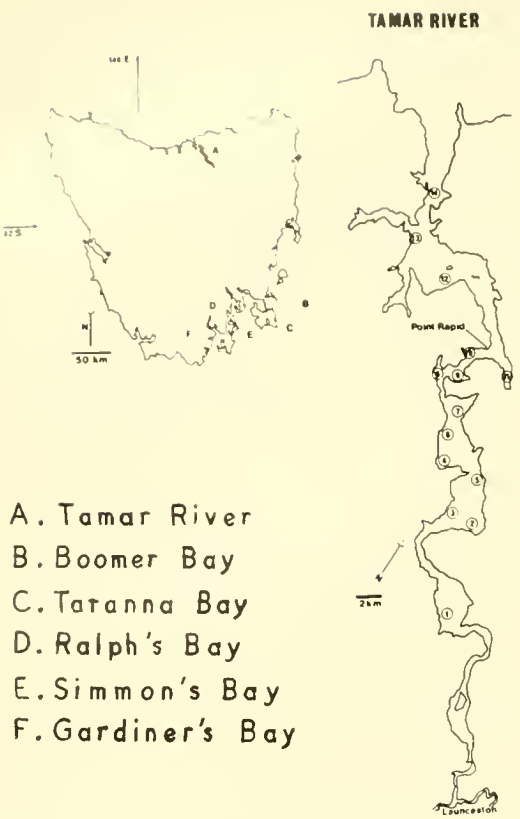

FIGURE 1. *Oyster sampling sites, Tasmania, Australia with map of Tamar River area—1973 (A: Tamar R.; B. Boomer Bay; C. Taranna Bay; D. Ralph's Bay; E. Simmons Bay; F. Gardners Bay)*

chromatograph equipped with an electron-capture detector. Instrument parameters and operating conditions follow.

Columns:	Pyrex, 5-ft × ⅛-inch diameter, packed with a mixture of 3 percent DC-200 and 5 percent QF-1 on 80–100-mesh Gas-Chrom Q
Temperatures:	detector 200°C injector 210°C oven 185°C
Carrier gas:	nitrogen flowing at 40 ml/minute

Thin-layer chromatography was used to check results obtained by gas chromatography and to check for possible interference from the presence of polychlorinated biphenyls (PCBs). Samples fortified with 1 μg of each compound produced average recoveries of 88 percent ΣDDT, 93 percent HCB and lindane, and 90 percent dieldrin. All data reported are corrected for recovery. The lower limit of quantitation was 10 ppb (10 μg/kg); values less than this but for positively identified peaks were recorded as trace.

Results

Residue levels of dieldrin, ΣDDT, HCB, and lindane in whole oyster meats are presented in Table 1. Dieldrin was detected in all but three samples. Elevated levels in oysters from the Tamar River were reflected in a high of 0.39 μg/g in the Ruffins Bay sample. ΣDDT residues were positively identified from all samples but were of an order of magnitude lower than dieldrin levels, ranging from trace to 0.06 μg/g. Traces of HCB were found only in the Gardners Bay oysters, and traces

TABLE 1. *Pesticide levels in Pacific oysters* (Crassostrea gigas), *Tasmania, Australia*

	Sampling Station	Sampling Date	Age, Year	No. Bulked	Residues, mg/kg, Whole Oyster			
					Dieldrin	ΣDDT	HCB	Lindane
TAMAR RIVER								
1	Rosevears	March 1973	—	18	0.20	0.03	—	—
2	Swan Bay	March 1973	—	12	0.10	0.02	—	—
3	Gravelly Beach	March 1973	—	15	0.21	0.01	—	—
4	Supply River[1]	February 1973	2	17	0.20	0.02	—	—
		March 1973	2	15	0.09	0.01	—	—
5	Hillwood Jetty	March 1973	—	22	0.10	0.02	—	—
6	Devoit	March 1973	—	12	0.10	0.02	—	—
7	Craigburn	March 1973	—	22	0.19	0.01	—	—
8	Devils Elbow	March 1973	—	20	0.10	0.01	—	—
9	Redwood Bay	February 1973	—	16	0.19	0.03	—	—
		March 1973	—	12	0.09	0.01	—	—
10	Ruffins Bay[1]	February 1973	2	13	0.39	0.01	—	—
		March 1973	3	22	0.27	0.06	—	—
11	East Arm[1]	February 1973	1	34	0.10	0.02	—	—
			2	15	0.09	0.02	—	—
			3	19	0.19	0.01	—	—
		March 1973	2	19	0.08	0.01	—	—
			3	13	0.20	0.01	—	—
12	Middle Island[1]	February 1973	3	16	0.11	0.03	—	—
	Flats	March 1973	2	21	0.09	0.02	—	—
			3	19	0.10	0.01	—	—
13	West Arm	March 1973	—	14	0.09	0.01	—	—
14	Bryants Bay	March 1973	—	14	0.08	0.01	—	—
	Boomer Bay[1]	August 1973	1.5	15	ND	T	ND	T
	Taranna Bay[1]	July 1973	1.5	15	T	T	ND	T
	Ralphs Bay[1]	January 1972	2	24	ND	T	—	—
	Simmons Bay[1]	August 1973	1.5	15	ND	0.01	ND	T
	Gardners Bay[1]	July 1973	1.5	15	T	0.06	T	T

NOTE: — not analyzed, ND not detected, T <0.01 mg/kg.
[1] Oyster farms.

of lindane were identified in samples from three of the leased farms.

The limited sampling of oysters of different ages from growing areas in the Tamar River suggests few differences in pesticide concentrations among the groups; this agrees with Butler's observations (*2*).

Tamar River samples taken at increasing distances downriver from Launceston showed differences in pesticide concentrations in oyster fats (Fig. 2). Dieldrin levels were inversely correlated with distance from Launceston ($r = 0.900$; $P = 0.001$), whereas DDT levels showed a more general spread suggestive of wide-scale low-level use of the pesticide ($r = 0.490$; $0.05 < P < 0.10$).

Discussion

Levels of pesticides other than dieldrin were generally low and probably of negligible significance. Because oysters are extremely sensitive to organochlorine pollutants, these levels indicate little contamination of the waterways (*4*).

Dieldrin levels were higher and indicated a serious level of contamination of the Tamar River. Levels are comparable to those reported by Clegg for the Sydney rock oyster (*C. commercialis*) in the Brisbane River (*6*). Butler, reporting on the U.S. National Pesticide Monitoring Program (NPMP), noted similar levels of dieldrin in oysters from a few locations in Georgia, New York, South Carolina, and Washington, but these were the exception (*5*). Dieldrin was detected in only 15 percent of all NPMP samples.

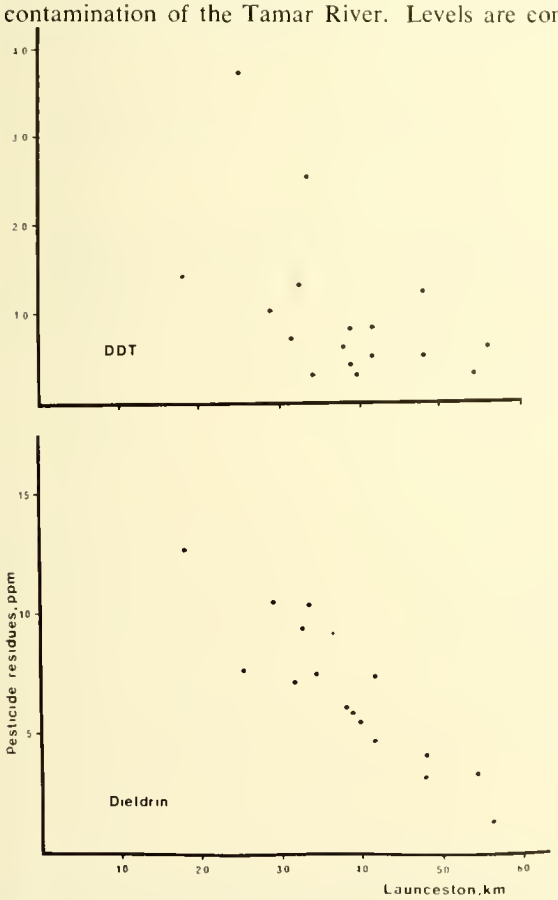

FIGURE 2. *Pesticide concentrations in oyster fats with increasing distance from Launceston, from sampling stations in Tamar River, Tasmania, Australia—March 1973*

Uptake of dieldrin by eastern oysters (*C. virginica*) was studied for a short term by Mason and Rowe (*11*) and over a longer period by Parrish (*14*). Concentration ratios for the pesticide were $2–8 \times 10^3$ for oysters exposed to ambient water concentrations of 0.1–9 μg/liter. If similar concentration factors apply to *C. gigas*, dieldrin levels in the Tamar River should range from 0.3 μg/liter to 0.075 μg/liter. This agrees with 0.18–0.02 μg/liter reported in a 1972–73 survey of the Tamar River by the State Department of the Environment (*8*).

Such levels would not affect embryonic development or larval growth and survival if Pacific oysters exhibit tolerances similar to those reported for eastern oysters. In the latter, Davis and Hidu (*7*) found little difference between controls and experimental cultures at dieldrin concentrations of 25 μg/liter.

Levels of DDT, HCB, and lindane in these oysters are within Australian tolerance standards for food (*13*) and represent little risk to public health. At present there is no published tolerance for dieldrin residues in fish, but if limits of the Food and Drug Administration, U.S. Department of Health, Education and Welfare, are applied (0.3 mg/kg, shellfish meats), then only one sample in the February survey exceeded these limits.

Results of the heavy-metal investigation mentioned earlier revealed widespread contamination of oysters in the Tamar River. Subsequently, oysters cannot be taken for human consumption from any point upstream of Point Rapid. This effectively removes any risk of consumption of oysters with high dieldrin concentrations because those downstream of Point Rapid exhibited much lower concentrations of the residue than did upstream samples. Distribution of pesticide levels throughout the Tamar River suggested that the minute amounts of DDT are probably attributable to agricultural runoff. Dieldrin levels suggested an upstream source of contamination for the pollutant. Industrial sources in Launceston were implicated in the Annual Report of the Department of the Environment (*8*), and it seems likely that the dieldrin was used to insect-proof woolen fabrics produced by a woolen mill in Launceston. Similar instances were recorded about mills in the United States (*10, 12*). Since this survey was conducted, the State Department of the Environment has attempted to limit the disposal of a number of pesticides in effluents. (The Launceston-

based woolen mill was prosecuted for illegal discharge of dieldrin residues.) Continued monitoring of Tamar River water samples for dieldrin and DDT has reflected the success of these moves. In 1972–73, dieldrin was detected in 89 percent of samples with a maximum concentration of 0.39 μg liter; DDT was found in 14 percent of samples with a high of 0.04 μg/liter. Comparable figures for 1975–76 were: dieldrin, 70 percent, 0.13 μg liter; DDT, 5 percent, trace (0.01 μg/liter) (B. O. Healey, Water Pollution Officer, Department of the Environment, Hobart, Tasmania, 1977. Unpublished data.)

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FOOD AND FEED

DDT Residues in Butter and Infant Formula in India, 1977[1]

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ABSTRACT

Samples of commercial brands of butter and infant formula from different parts of India were examined for DDT residues. All 18 samples of butter representing nine brands were contaminated. Levels of DDT residues ranged from 0.42 to 11.36 ppm and exceeded the Food and Agriculture Organization/World Health Organization practical residue limit of 1.25 ppm in 90 percent of the samples. All four brands of infant formula contained DDT residues above the practical residue limit. Most DDT residues were in the form of p,p'-TDE in both commodities. This contamination of milk with excessive amounts of DDT residues seems to be widespread in India.

Introduction

The proportions of DDT and its metabolites present in cows' milk indicate possible sources of these residues (*5*). Different routes of animal exposure result in secretion of DDT in different forms (*11*). Animal uptake by aspiration or intravenous injection results in secretions of DDT; ingestion leads to secretions in the form of DDT metabolites.

Limited information is available in India on the nature of DDT residues in bovine milk. Milk samples from Delhi contained only residues of *p,p'*-DDT (*1*). On the other hand, most DDT residues in milk from Ludhiana were in the form of *p,p'*-TDE (*2*). Because milk is an important food commodity, particularly for children, it is necessary to know the extent and sources of its contamination with DDT. Samples of commercial brands of butter and infant formula from different parts of India were analyzed for DDT residues. These commodities were chosen because of their availability.

Materials and Methods

BUTTER

Different commercial brands of butter manufactured in Punjab, Haryana, Delhi, Rajasthan, and Gujarat were purchased from the local market in 100-g packages February and March 1977. Three butter samples weighing 100 g each were also purchased during the same period from local dairies situated in different parts of Ludhiana city. Laboratory extractions were made within 2 days.

The method described by Faubert Maunder et al. (*4*) was modified slightly and used to extract and isolate DDT residues. The butter was warmed at about 50°C to separate the fat which was decanted through dry filter paper. A 5-g sample of the clarified fat was dissolved in 10 ml of hexane and transferred quantitatively to a 125-ml separatory funnel by using additional small portions of hexane totaling 15 ml. The hexane extract was partitioned three times into hexane-saturated dimethylformamide, using 10 ml of solvent each time. The dimethylformamide fraction was backwashed with 10 ml of dimethylformamide-saturated hexane, diluted with 250 ml of water and 50 ml of sodium chloride-saturated aqueous solution, and extracted twice with 100 ml of hexane. The combined herane extracts were concentrated to about 5–10 ml for subsequent column cleanup. Silica gel, 60–200 mesh, was thoroughly washed with acetone and methanol and activated 1 hour at 130°C. It was packed in a 50-cm × 2-cm glass column to a height of 10 cm between layers of anhydrous sodium sulfate. The column was prewashed with 100 ml of hexane. The sample extract in hexane was added to the column and eluted with 150 ml of 50 percent benzene in hexane. The eluate was concentrated to 1–10 ml and was analyzed by thin-layer and gas-liquid chromatography.

Thin-layer chromatography was done by the method of Thompson et al. (*7*) on $AgNO_3$-incorporated, alumina-

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G-coated glass plates. *n*-Hexane was used as the developing solvent. The *Rf* values were: *p,p'*-DDT, 0.65; *p,p'*-DDE, 0.88; *p,p'*-TDE, 0.35; *o,p'*-DDT, 0.77; *o,p'*-TDE, 0.42; α-BHC, 0.52; β-BHC, 0.1; γ-BHC, 0.32; and δ-BHC, 0.1.

GLC determinations were made by injecting 1–10 μl of the sample solution into a Model 7624 Packard gas chromatograph. Two columns were used: (A) was the working column and (B) was used for confirmation. Instrument parameters and operating conditions follow:

Detector:	Tritium electron-affinity
Columns:	(A) Pyrex, 102 cm long × 0.4 cm ID, packed with 5 percent DC-200 on 80–100-mesh Gas-Chrom Q
	(B) Pyrex, 1.84 m long × 0.4 cm ID, packed with 2 percent DEGS on 80–100-mesh Gas-Chrom Q
Temperature:	Column 190°C
	Detector 200°C
	Inlet 210°C
Carrier gas:	Nitrogen
Flow rate:	70 ml/minute for Column A 100 ml/minute for Column B

Retention times, in minutes, are listed below:

	Column A	Column B
p,p'-DDE	2	3.5
p,p'-TDE	2.5	10
p,p'-DDT	3	8
o,p'-DDT	2.5	5
o,p'-TDE	2	6.5
α-BHC	1	1.5
γ-BHC	1.10	2
β-BHC	1	5.5

On column A, the half-scale deflection was obtained with 0.5 ng of *p,p'*-DDE, 0.8 ng of *p,p'*-TDE, and 1.0 ng of *p,p'*-DDT. Quantitative estimations were made by comparing peak heights of the unknown with the standards treated similarly. Recoveries of DDT and its metabolites at the fortification levels of 0.5 ppm were 80–90 percent. Results were expressed as such and were not corrected for recovery. The limit of detection of *p,p'*-DDT in butter was 0.01 ppm.

The nature of DDT residues was confirmed by a micro-alkali dehydrohalogenation procedure in the *Manual of Analytical Methods for Analysis of Pesticide Residues in Human and Environmental Samples* (*10*).

INFANT FORMULA

Four brands of infant formula manufactured in Punjab, Bombay, and Gujarat were purchased from a local market in 500-g packages February-April 1977. Ten g of infant formula was weighed and diluted to 80 ml with distilled water. Each sample was blended with 160 ml of acetone and 160 ml of hexane in a vortex beaker for 3 minutes. The extract was centrifuged at 3000 rpm for 10 minutes. The hexane layer was removed by pipet, concentrated to about 25 ml, and partitioned into dimethylformamide three times, using 15 ml of solvent each time. The combined dimethylformamide fractions were cleaned and analyzed by the procedures described for butter.

Results and Discussion

DDT residues in butter occurred mainly in the form of *p,p'*-DDT, *p,p'*-DDE, and *p,p'*-TDE. Small amounts of *o,p'*-DDT and *o,p'*-TDE were also detected. Some samples had BHC residues in the form of α-, β-, and γ-isomers. Only traces of BHC were found. The maximum residue, 1 ppm BHC, was found in a sample of butter from Gujarat.

Levels of DDT residues in eighteen samples of butter representing six commercial and three local brands are given in Table 1. All but one brand of butter contained DDT residues higher than the practical residue limit of 1.25 ppm established by the United Nations Food and Agriculture Organization (FAO)/World Health Organization (WHO) (*9*). The level of DDT residues varied from 0.42 to 11.36 ppm with an average of 4.77 ppm. In a study at Uttar Pradesh Agricultural University, Pantnager, India (*8*), two of five butter samples were contaminated with DDT at an average level of 0.4 ± 0.14 ppm. The highest level of DDT detected was 0.5 ppm. Agnihotri et al. (*1*) reported that seven of eight samples of butter collected from Delhi contained DDT residues higher than the practical residue limit. The concentration of residues varied from 1.1 to 8.0 ppm with an average level of 3.8 ppm. The present study shows that most of the commercial brands of butter manufactured in Punjab, Haryana, Delhi, Rajasthan, and Gujarat contained DDT residues higher than the practical residue limit, and suggests widespread contamination in India of milk with high levels of DDT residues.

TABLE 1. *Residues of DDT and its metabolites in commercial butter samples, India, 1977*

Butter	Sample Number	Origin	Residues, ppm DDT	DDE	TDE	ΣDDT
Brand I	1	Gujarat	1.88	1.48	8.00	11.36
	2		2.54	1.44	6.53	10.51
	3		1.62	1.44	6.35	9.41
Brand II	1	Haryana	1.16	0.74	3.74	5.64
	2		1.18	0.73	3.51	5.42
	3		0.50	0.30	1.36	2.16
Brand III	1	Punjab	0.75	0.58	3.54	4.87
	2		0.73	0.42	3.25	4.40
	3		0.63	0.41	2.53	3.57
Brand IV	1	Rajasthan	0.75	0.73	3.73	5.21
	2		0.68	0.49	2.63	3.80
	3		0.70	0.42	2.50	3.62
Brand V	1	Delhi	0.35	0.25	1.55	2.15
Brand VI	1	Gujarat	0.02	0.17	0.33	0.52
	2		0.02	0.19	0.21	0.42
Locale I	1	Ludhiana	0.81	0.58	4.47	5.86
Locale II	1	Ludhiana	0.70	0.42	2.84	3.96
Locale III	1	Ludhiana	0.57	0.38	2.16	3.11

TDE is the predominant metabolite detected in all brands of butter (Table 1). Milk collected recently from Ludhiana and surrounding areas showed similar results (*2*). Since TDE is not being used in India for crop protection or mosquito control, then TDE residues must arise as a result of metabolism of DDT. However, milk and butter samples from Delhi did not show residues of any metabolite. The residues were detected as DDT only (*1*). The other two studies carried out in India on the DDT contamination of milk and milk products did not consider the metabolites (*6, 8*). The high level of TDE found in butter samples suggests that cattle ingest DDT mainly through contaminated feed. Witt et al. found a 1:1 relation between levels of DDT residues in cattle feed and the concentration of DDT secreted in bovine milkfat (*12*). If this relationship were true in the present study, DDT residues in cattle feed would be expected to vary between 0.42 and 11.36 ppm, averaging 4.77 ppm. The sources of such high DDT contamination of cattle feed must be determined particularly because the use of DDT for plant protection is limited in India. DDT is used mainly for malaria control; indoor residual spraying on the walls and roofs is carried out at the rate of 1 g/m^2. Dhaliwal and Kalra suggested that the indoor spraying might contaminate stored feed, and thereby contribute partly toward the ingestion of DDT by cattle (*2*). However, the contribution of this and other sources of contamination of milk needs further investigation.

All four popular brands of infant formula contained DDT residues above the tolerance level of 1.25 ppm, usually in the form of TDE (Table 2). The concentration of DDT varied from 1.52 to 2.72 ppm, averaging 1.90 ppm. Apparently, no other study has been carried out in India on the DDT contamination of commercial infant formula. The present study shows that even the spray drying process in the manufacture of infant formula, does not reduce residues of DDT to below the FAO/WHO tolerance level. This corresponds with the observation of Engst et al. (*3*).

The average level of DDT residues found in infant formula is 1.90 ppm. The consumption of this milk by a three-month-old child weighing approximately 5 kg at the normal feeding rate of 135 g/day would result in a daily intake of 47 μg of DDT. This value is about twice the acceptable daily intake of 0.005 mg/kg of baby weight (25 μg for an infant weighing 5 kg) established for DDT by the Joint Pesticides Committee of FAO and WHO (*9*).

TABLE 2. *Residues of DDT and its metabolites in commercial infant formula samples, India, 1977*

Infant Formula	Origin	Fat Content, %	Residues on Fat Basis, ppm			
			DDT	DDE	TDE	ΣDDT
Brand I	Punjab	19	0.63	0.33	1.76	2.72
Brand II	Bombay	19	0.40	0.25	1.04	1.69
Brand III	Gujarat	18	0.26	0.36	1.03	1.65
Brand IV	Bombay	18	0.33	0.17	1.02	1.52

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GENERAL

Organochlorine Pesticides and Polychlorinated Biphenyls on Sediments from a Subarctic Salt Marsh, James Bay, Canada—1976

W. A. Glooschenko[1] and R. C. J. Sampson[2]

ABSTRACT

Sediment samples were collected from a subarctic salt marsh on James Bay, Ontario in May 1976. Of 15 organochlorine compounds analyzed, trace amounts mainly of p,p′-DDE and polychlorinated biphenyls (PCBs) were detected, but could not be quantitated.

Introduction

Organochlorine pesticides and polychlorinated biphenyls (PCBs) have been detected in subarctic and arctic marine food chains. PCBs and ΣDDT have been found in polar bears, seals, and fish in the Canadian arctic (*1*) and in fish in a landlocked lake in northwestern Quebec (*5*). The authors wished to determine levels of these organochlorine compounds in sediments of a subarctic wetland since this part of the ecosystem would be the ultimate sink of many of the compounds.

Sediment samples were collected in May 1976 from a subarctic salt marsh at North Point, Ontario (51°29′N, 80°27′W), on the western shore of James Bay, approximately 27 km northeast of Moosonee at the southern end of James Bay. A sample was collected in Moosonee from a drainage ditch to check the possibility of local sources of contamination.

Methods and Materials

Sediment samples were collected by hand with a stainless steel trowel from the top 5 cm of five salt marsh sites, two freshwater creek sediments, and a drainage ditch in the Moosonee settlement. Samples were placed in aluminum cans which had been carefully cleaned with interference-free solvents and were frozen until analysis within two months of collection.

Thawed wet-sediment samples (10 g) were extracted by using an ultrasonic probe. Each sample was extracted three times with 75 ml of acetonitrile for 2 minutes each time and filtered through Celite and sodium sulfate. The combined filtrate and washings were partitioned into petroleum ether, washed with water, dried with sodium sulfate, and evaporated with a rotary evaporator to 1 ml, using isooctane as a keeper. Recovery was 80–100 percent (*2*).

The concentrate was analyzed by high-pressure liquid chromatography. Four fractions were collected, evaporated to 1 ml, and analyzed by computerized gas chromatography (GC) with automatic sampling. Identification was based on quantitative reproducibility (±20 percent) on four columns of varying polarity with a 2 percent retention time variability window. Instrument parameters and operating conditions follow.

Detectors:	linearized ^{63}Ni electron-capture
Columns:	(1) 2 m × 3.5 mm I.D., pyrex, packed with mixture of 1.5 percent OV-17 and 1.95 percent QF-1 on 100–120-mesh Gas-Chrom Q (2) 1.86 m × 4 mm I.D., packed with mixture of 4 percent OV-101 and 6 percent OV-210 or QF-1 on 80–100-mesh Gas-Chrom Q (3) 1.86 × 4 mm I.D., packed with 3 percent OV-101 on 80–100-mesh Chromosorb W-HP (4) 2 m × 3.5 mm I.D., packed with 3 percent OV-225 on 100–120-mesh Gas-Chrom Q
Temperatures:	column 200° C injector 225° C detector 325° C
Carrier gases:	mixture of 5 percent methane and 95 percent argon flowing at 50–75 ml/minute

Quantitation limits are given in Table 1. Detection limits for the pesticides analyzed are approximately one-tenth the quantitation limit. Authors were unable to confirm identities of residues by mass spectrometry because of the low levels of compounds.

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TABLE 1. *Distribution of organochlorines in sediments from North Point salt marsh complex*

Compound	Quantitation Limit, μG/G	Sample Site							
		Salt Marsh					Creek Beds		Moosonee
		1	2	3	4	5	1	2	1
		Residues, μG/G dry weight							
Lindane	0.001	< 0.001	ND	ND	ND	ND	ND	ND	< 0.001
Heptachlor	0.001	ND	ND	ND	ND	ND	ND	ND	ND
Aldrin	0.001	ND	ND	ND	ND	ND	ND	ND	ND
Heptachlor epoxide	0.001	ND	ND	ND	ND	ND	ND	ND	ND
p,p'-DDE	0.001	< 0.001	< 0.001	ND	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001
Dieldrin	0.001	ND	ND	ND	ND	ND	ND	ND	ND
p,p'-DDT	0.001	ND	ND	ND	ND	< 0.001	ND	ND	ND
o,p'-DDT	0.001	ND	ND	ND	ND	ND	ND	ND	ND
Endrin	0.001	ND	ND	ND	< 0.001	ND	ND	ND	ND
α-Chlordane	0.005	ND	ND	ND	ND	ND	ND	ND	ND
γ-Chlordane	0.005	ND	ND	ND	ND	ND	ND	ND	ND
α-Endosulfan	0.01	ND	ND	ND	ND	ND	ND	ND	ND
β-Endosulfan	0.01	ND	ND	ND	ND	ND	ND	< 0.01	ND
p-p'-Methoxychlor	0.05	ND	ND	ND	ND	ND	ND	ND	< 0.05
Total PCBs	0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1

Results and Discussion

Results are in Table 1. Of the 15 organochlorine compounds, none could be quantitated. However, *p,p'*-DDE and PCBs were detected in nearly all the samples. Traces of lindane, *p,p'*-DDT, endrin β-endosulfan, and *p,p'*-methoxychlor were noted.

No river entering James Bay drains regions of agriculture, nor is there intensive recreational use of the area, a source of pesticide input in southern Ontario (*2–4*). Therefore, it appears that traces of organochlorine compounds have been transported to the area by air.

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APPENDIX

Chemical Names of Compounds Discussed in This Issue

ALDRIN	Hexachlorohexahydro-*endo, exo*-dimethanonaphthalene 95% and related compounds 5%
AROCLOR 1242	PCB, approximately 42% chlorine
AROCLOR 1254	PCB, approximately 54% chlorine
AROCLOR 1260	PCB, approximately 60% chlorine
AZINPHOSMETHYL	*O,O*-Dimethyl *S*-[(4-oxo-1,2,3-benzotriazin-3(4*H*)-yl)methyl] phosphorodithioate
BENZENE HEXACHLORIDE (BHC)	1,2,3,4,5,6-Hexachlorocyclohexane
BROMACIL	5-Bromo-3-*sec*-butyl-6-methyluracil
CARBOPHENOTHION	*S*-[[(*p*-Chlorophenyl)thio]methyl] *O,O*-diethyl phosphorodithioate
CHLORDANE	Octachloro-4,7-methanotetrahydroindane 60% and related compounds 40%
DDE	Dichlorodiphenyldichloroethylene
DDT	Dichorodiphenyltrichloroethylene
DACTHAL (DCPA)	Dimethyl tetrachloroterephthalate
DEF	*S,S,S*-Tributyl phosphorotrithioate
DEMETON	*O,O*-Diethyl *O*-[2-(ethylthio)ethyl] phosphorothioate and *O,O*-diethyl *S*-[2-(ethylthio)ethyl] phosphorothioate
DIAZINON	*O,O*-Diethyl *O*-(2-isopropyl-6-methyl-4-pyrimidinyl) phosphorothioate
DIELDRIN	Hexachloroepoxyoctahydro-*endo, exo*-dimethanonaphthalene 85% and related compounds 15%
DIURON	3-(3,4-Dichlorophenyl)-1,1-dimethylurea
ENDOSULFAN	Hexachlorohexahydromethano-2,4,3-benzodioxathiepin-3-oxide
ENDRIN	Hexachloroepoxyoctahydro-*endo, endo*-dimethanonaphthalene
ETHION	*O,O,O',O'*-Tetraethyl *S,S'*-methylene bisphosphorodithioate
HEPTACHLOR	Heptachlorotetrahydro-4,7-methanoindene and related compounds
LINDANE	*Gamma* isomer of 1,2,3,4,5,6-hexachlorocyclohexane
MALATHION	*O,O*-Dimethyl dithiophosphate of diethyl mercaptosuccinate
METHOXYCHLOR	2,2-Bis(*p*-methoxyphenyl)-1,1,1-trichloroethane 88% and related compounds 12%
MIREX	Dodecachlorooctahydro-1,3-methano-1*H*-cyclobuta[cd]pentalene
PARATHION	*O,O*-Diethyl *O*-*p*-nitrophenyl phosphorothioate
PHORATE	*O,O*-Diethyl *S*-[(ethylthio)methyl] phosphorodithioate
PCBs	Polychlorinated biphenlys, mixtures of chlorinated biphenyl compounds having various percentages of chlorine
TDE	Dichlorodiphenyldichloroethane
TOXAPHENE	Technical chlorinated camphene 67-69% chlorine
TRIFLURALIN	α,α,α-Trifluoro-2,6-dinitro-*N,N*-dipropyl *p*-toluidine

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.

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FISH, WILDLIFE, AND ESTUARIES

Pesticide Residues in Estuarine Mollusks, 1977 versus 1972—National Pesticide Monitoring Program

Philip A. Butler,[1] Charles D. Kennedy,[2] and Roy L. Schutzmann[2]

ABSTRACT

Bivalve mollusks were monitored for residues of 20 organochlorine and organophosphate pesticides and polychlorinated biphenyls in spring 1977 in 87 of the 181 estuaries routinely monitored on a monthly basis during 1965–72. DDT, the only pesticide detected in 1977, occurred at low levels in one estuary each on the Atlantic and Pacific coasts.

Introduction

In 1965 the U.S. Bureau of Commercial Fisheries initiated a program to monitor shellfish populations for organochlorines. In cooperation with local laboratories, about 180 permanent monitoring stations in 15 coastal states were sampled for any one of 10 species of mollusks monthly. The eastern oyster, *Crassostrea virginica*, was the principal species collected on the Atlantic coast, and *C. gigas* was the species usually monitored on the Pacific coast. The program continued until 1972, but not all areas were monitored for the entire period. About 8,100 samples containing 15 pooled individuals were analyzed. DDT was found in almost all samples. Dieldrin was the next most commonly detected pesticide; residues of endrin, mirex, toxaphene, and polychlorinated biphenyls (PCBs) were detected occasionally. By 1972, there was a clearly defined trend toward fewer and smaller residues of DDT and its metabolites (*1*).

Early in 1977, the U.S. Environmental Protection Agency monitored mollusks at some of the same sites to determine further trends in pollution levels after the 5–7-year lapse.

Materials and Methods

The original cooperating laboratories agreed to collect the new samples. About half the former stations where pesticides had been found consistently a decade ago were monitored again. Single collections of 30 bivalves at each site were made just before or during early stages of the spawning cycle so that tissue lipid levels presumably would approach the maximum.

There were 178 samples; replicate collections were made at 89 stations in 87 estuaries. Depending on the availability, seven species of mollusks were used including the freshwater Asiatic clam, *Corbicula manilensis;* eastern oyster, *Crassostrea virginica;* Pacific oyster, *C. gigas;* Atlantic ribbed mussel, *Geukensia demissa;* northern quahog, *Mercenaria mercenaria;* soft-shell clam, *Mya arenaria;* and blue mussel, *Mytilus edulis*. Oysters were sampled in 63 estuaries, mussels in 14, and clams in 10 estuaries. However, clams are the least satisfactory as biomonitors (*2*).

Two samples of 15 bivalves each were collected at each station. They were shucked but were not drained, and were homogenized in an electric blender. A single aliquot of about 50 g from each pooled sample was preserved with 50 ml reagent grade methanol and mailed in a methylpentene vial to the EPA Pesticides Monitoring Laboratory in Bay St. Louis, Mississippi, for analysis. Analytical procedures, detailed elsewhere (*3*), permitted the detection of 20 organochlorine and organophosphate pesticides and PCBs (Table 1). In the 1965–72 program, samples were screened routinely for only 11 of the more persistent organochlorine pesticides.

Results and Discussion

The salient feature of the 1977 monitoring data was the absence of detectable pesticide residues in 85 of the 87

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TABLE 1. *Compounds detected by chemical procedures used in monitoring mollusks*

Organochlorines	Organophosphates
Aldrin	Azinphosmethyl
Chlordane	Carbophenothion
ΣDDT	DEF
Dieldrin	Demeton
Endosulfan	Diazinon
Heptachlor	Ethion
Lindane	Malathion
Methoxychlor	Parathion
Mirex	Phorate
PCBs	
Toxaphene	
Trifluralin	

NOTE: Lower detection limit is 10 μg/kg for all compounds except endosulfan, 20 μg/kg; methoxychlor and ethion, 30 μg/kg; mirex, PCBs, toxaphene, carbophenothion, and DEF, 50 μg/kg.

estuaries sampled and the complete absence of PCBs. On the Atlantic coast, oysters from two adjacent New Jersey reefs and one reef on the Delaware side of upper Delaware Bay contained DDE. Average residue in the six samples was 33 ± 15 μg/kg. Oysters from reefs closer to the mouth of the estuary did not contain detectable residues. As recently as 1972, every monthly oyster sample on the New Jersey side of the Bay contained about three times as much DDT as did samples collected in 1977, as well as residues of dieldrin and PCBs. The fauna in Delaware Bay were presumably contaminated by the hundreds of tons of DDT sprayed aerially between 1950 and 1966 to control New Jersey marsh mosquitoes (*4*).

On the Pacific coast, bivalves in only one of the 14 estuaries monitored in California and Washington state contained pesticide residues. Replicate samples of blue mussels from Muga Lagoon, about 35 miles north of Los Angeles, contained DDT and its metabolites, TDE and DDE, at the average level of 122 μg/kg. A decade earlier, monthly samples of mussels from this station contained ΣDDT residues of 500–1,800 μg/kg, as well as traces of dieldrin and endrin.

The reliability of these isolated data in documenting the virtual disappearance of pesticide pollution from estuarine water is dependent on knowledge gained from the earlier program of the seasonal aspects of waterborne pesticide pollution. Monthly samples in that study showed that pesticide residues in intermittently polluted areas were typically present in the spring, and, if continuously present, were usually larger in the spring, presumably the result of increased river runoff.

The decline in pollution is emphasized by comparison of the present data with pesticide residue levels and incidence in bivalves from the same estuaries during the final 12 months of the earlier program (Table 2). This table shows the number of stations monitored in each state in 1977 but does not repeat the 1977 residue data.

Since filter-feeding bivalves purge themselves of organic residues within a few weeks in the absence of continuing pollution (*2*), the 1977 data show essentially the disappearance of pesticides from the water mass. However, there is evidence that persistent pesticides have not disappeared entirely from most of these estuarine ecosystems. During 1972–76, yearling fish of several species were monitored in many of the same estuaries from which bivalves were collected in 1977 (*3*). Samples consisted of 25 whole fish captured twice yearly. In 1976, 68 samples or 36 percent of the 190 samples analyzed contained DDT residues at levels up to 2,500 μg/kg; 22 percent of the samples also contained PCBs.

TABLE 2. *Summary of pesticide residues in estuarine mollusks during the final 12 months of the 1965–72 program in those estuaries re-monitored in 1977*

State	Final 12 Months	No. of Stations	No. of Samples	% of Samples with DDT	Arith. Mean of DDT, μg/kg	Other Residues Detected [1]	Species Monitored [2]
Alabama	1968–69	2	10	100	102	D	2
California	1971–72	14	68	96	81	D,E,P	1,3,4,7
Delaware	1968–69	5	58	74	44	D	2,4,5
Florida	1968–69	6	61	85	308	D	2
Georgia	1971–72	5	60	20	14	D,T,P	2
Maine	1969–70	5	36	14	29	—	6,8
Maryland	1969–70	6	11	64	25	D	2
Mississippi	1971–72	3	30	63	31	—	2
New Jersey	1971–72	3	15	100	74	D,P	2
New York	1971–72	6	67	88	40	D	2,5,7
North Carolina	1971–72	9	88	35	46	D	2
South Carolina	1968–69	7	83	37	24	D,M	2
Texas	1971–72	6	56	73	72	D,E,T,P	2
Virginia	1971–72	6	24	96	36	D,P	2
Washington state	1967–68	6	72	18	20	—	3

[1] D=dieldrin, E=endrin, M=mirex, P=PCB, T=toxaphene.
[2] 1, *Corbicula manilensis*, Asiatic clam; 2, *Crassostrea virginica*, eastern oyster; 3, *C. gigas*, Pacific oyster; 4, *Geukensia demissa*, Atlantic ribbed mussel; 5, *Mercenaria mercenaria*, northern quahog; 6, *Mya arenaria*, soft-shell clam; 7, *Mytilus edulis*, blue mussel.

The residues in fish are probably the result of storage and recycling of synthetic pesticides in different links of the food web. The filter-feeding mollusks present a more realistic picture of the current input of pesticides into the marine environment. However, bivalves must be monitored more frequently to reflect fluctuating pollution patterns.

Acknowledgment

The authors are grateful to the staffs of the many federal, state, and university agencies, all of whom participated enthusiastically in this project.

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Chlorinated Insecticide and PCB Residues in Fish and Mussels of East Coastal Waters of the Middle and North Adriatic Sea, 1974–75 [1]

Mladen Picer, Nena Picer, and Marijan Ahel[2]

ABSTRACT

Concentrations of chlorinated pesticides and polychlorinated biphenyls (PCBs) were determined in mussels (Mytilus galloprovincialis) *and goby fish* (Gibius *sp.) collected in four areas located in eastern coastal waters of the middle and north Adriatic Sea. Most samples were collected in early spring and late summer of 1974 and 1975.*

The compounds p,p'-*DDT*, p,p'-*DDE*, p,p'-*TDE, and PCBs were detected most frequently. In about 60 percent of the samples dieldrin was also detected.*

Average wet-weight concentrations of ΣDDT and PCBs in mussels from the four areas sampled were: Istrian coast, 65 and 76 ppb; Rijeka Bay, 58 and 75 ppb; Zadar, 36 and 128 ppb; Losinj Island, 167 and 133 ppb. Average concentrations in fish samples were: Istrian coast, 124 and 144 ppb; Rijeka Bay, 37 and 82 ppb; Losinj Island, 166 and 157 ppb. Dieldrin concentrations were in the low ppb range.

Although major Italian rivers discharge chlorinated hydrocarbons into the north Adriatic, sampling of biota from Istrian coastal waters indicates no significant effect on the pollution level. However, waste waters from small coastal settlements evidently do contribute significantly to chlorinated hydrocarbon contamination of that ocean.

Marine samples from Losinj Island had high chlorinated hydrocarbon concentrations, indicating uptake of pollutants from the north Adriatic.

Introduction

Many chlorinated insecticides and industrial aromatic chlorinated hydrocarbons such as polychlorinated benzenes, naphthalenes, biphenyls, and terphenyls are extremely resistant to degradation in the environment (*12, 22*). On the other hand, toxicological and other harmful effects of these compounds on aquatic and terrestrial ecosystems are well documented (*2, 8*). Thus worldwide research has focused on the occurrence and fate of chlorinated hydrocarbons in terrestrial, freshwater, and marine ecosystems (*4, 11, 19*).

The most delicate and endangered parts of world oceans are semiclosed formations such as the Mediterranean Sea and the Adriatic Sea. The Adriatic Sea is shallow and small, and its northernmost extension, the Gulf of Trieste, lies virtually in the heart of Middle Europe; hence it is among the most jeopardized marine ecosystems in the world (*18*).

As part of the United Nations Development Program assisted project "Protection of the Human Environment in the Yugoslav Adriatic Region," chlorinated hydrocarbons were measured in mussels (*Mytilus galloprovincialis*) and in some benthic fishes (*Gobius* sp.) of the eastern coastal water of the north and middle Adriatic and near Losinj Island (Figure 1).

The mussel was chosen for monitoring chlorinated hydrocarbons because it is a well-known filter feeder recommended for monitoring many organic and inorganic pollutants (*6*). The goby fish was selected for its restricted living area and high tolerance for polluted seawater, which makes it a logical indicator of polluted marine environments. Other fish species were chosen for their popularity as food among local populations.

By analyzing chlorinated hydrocarbon contamination in mussels and fish from the eastern waters of the north and middle Adriatic, authors hoped to measure regional pollution caused by intensive agricultural and industrial discharges into the northern Adriatic, and local pollution of two nuclei, the Bay of Rijeka and the town of Zadar. Losinj Island south of the Bay of Rijeka was chosen as a clean reference area because it has no significant industry or agriculture and it is not heavily populated.

Sampling and Analysis

Mussels were collected manually or by dredging in intertidal or very shallow water. Soft tissue was removed

[1] Study supported in part by Self-Management Community of Interest for Scientific Research of S. R. Croatia.

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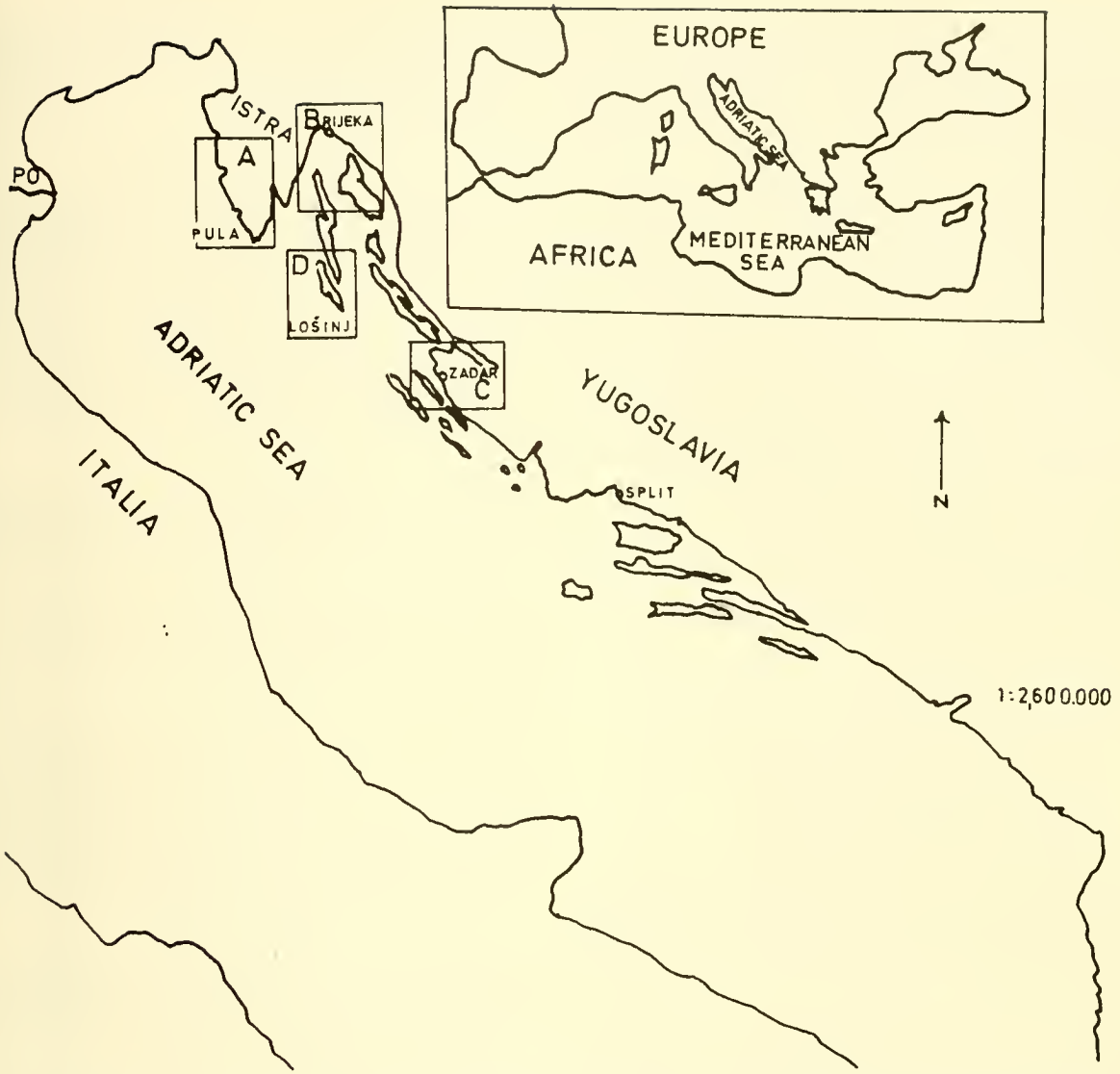

FIGURE 1. *Adriatic Sea, with areas sampled for chlorinated insecticide and PCB residues in marine biota*

from the shell, placed in aluminum foil, and frozen. The foil had been cleaned with redistilled petroleum ether and heated at 200°C for 12 hours. Samples consisted of 20–30 individual animals with shells 3–5 cm long. For the extraction of chlorinated hydrocarbons a subsample of 10 g was taken by a clean scalpel. Sample remains were frozen for analysis.

Goby fish were taken from the sea by angle, placed in clean aluminum foil, and frozen within a few hours of capture. Each sample consisted of six individual fishes 8–12 cm long. Samples of single fish were obtained from commercial catches in local markets. The specimen was measured and weighed, its dorsolateral surface was scraped clean, and 10 g of epaxial white muscle tissue was removed by a clean scalpel.

Ten g of muscle tissue and 10 g of anhydrous Na_2SO_4 were concurrently homogenized and extracted twice with 75 ml petroleum ether in a Lourdes blender for 3 minutes. Each extract was decanted into an Erlenmeyer flask and left overnight for settling of fine particles and then filtered through a 3-cm-high column of anhydrous Na_2SO_4. The aliquot of extract was evaporated to dryness and the residue of extracted organic matter was weighed and recorded.

Samples were cleaned as recommended by Holden and Marsden (*9*). Mirex was added as an internal standard prior to concentration of the sample extract with 50–100 mg lipid residue. The sample extract was concentrated to 1 ml under vacuum by means of a rotary evaporator and applied to a 6-mm-ID column holding

2 g alumina. The alumina had been prepared by heating activated alumina (Brockmann activity 1) at 500°C for 12 hours and partly deactivated by adding 5 percent distilled water by weight. Elution was performed with 15 ml hexane.

PCBs were separated from organochlorine insecticides on a miniature silica gel column according to the modified method of Snyder and Reinert (*13, 17*). Hexane eluate was evaporated to 1 ml and applied to a 10-mm column holding 100 mm silica gel. The gel was activated for 18 hours at 200°C. After cooling to room temperature, *n*-pentane was added and column was filed with a mixture of *n*-pentante and silica gel. Elution started with 32 ml *n*-pentane and was completed with 40 ml benzene. The first eluate contained PCBs and mirex; the second contained *p,p'*-DDE, *p,p'*-DDT, *p,p'*-TDE, and dieldrin.

A Hewlett-Packard 7620 model gas chromatograph (GC) equipped with ^{63}Ni electron-capture detector was used. Operating parameters for GC analysis were:

Columns:	(A) 1.8-m-by-4-mm glass packed with 1.5 percent SP-2250 + 1.95 percent SP-2401 on 100/120 mesh Supelcon AW-DMCS
	(B) 1.5-m-by-4-mm glass packed with 4 percent SE-30 + 6 percent OV-210 on 100/120 mesh Gas-Chrom Q
Temperatures:	Injector 240°C
	Column 210°C
	Detector 250°C
Carrier gas:	5 percent methane in argon
Flow rate:	30 ml/minute

Organochlorine compounds were quantitated by comparing peak areas in sample and standard chromatograms. PCBs were determined by using a standard solution of Aroclor 1254.

Experiments comparing aldrin and mirex as internal standards showed mirex to be superior. Mirex was used as an internal standard throughout the analyses because it is rather easily separated from PCBs on a GC column. Its loss was used as a measure of recovery in this study; in fact, recovery of chlorinated hydrocarbons varied between 68 and 87 percent.

For the confirmatory test samples with higher contents of DDT were hydrolyzed by KOH (*10*).

Sensitivity of DDT and its metabolites is 1 ppb wet weight and for PCBs it is 10 ppb.

In some samples low concentrations of dieldrin were found but the data are not reported in this paper.

The method of organochlorine determination was intercalibrated within the International Intercalibration Program on Chlorinated Hydrocarbons in Marine Materials funded by the United Nations Environmental Program (UNEP). Results obtained in the Centre for Marine Research were relatively close to the mean values after excluding disproportionately high residues according to criteria of Chauvenet (*5, 14*).

Results and Discussion

Concentrations of chlorinated hydrocarbons in mussels and fish from coastal waters of the eastern Adriatic are presented in Table 1. Distribution frequencies of ΣDDT and PCBs in mussel and fish samples are presented in Figure 2. The level of organochlorine concentrations varied widely, which is not unreasonable considering the unusual pollution pattern and hydrography of the Adriatic Sea and the complexity of the biotic samples analyzed.

Figures 3 and 4 present arithmetic means and ranges of DDT and its metabolites, dieldrin, and PCBs in mussels, goby fishes, and several species of benthic fishes. Although 14 species of benthic fishes were analyzed in the present investigation, results are presented only for those species which had three or more valid samples analyzed. Except for gobies, fish species are presented by decreasing order of summed pesticide and PCB concentrations. Comparing these two decreasing orders shows that the position of fish species differs according to whether the concentrations of pollutants are presented as wet weight or as extracted organic matter. But both figures indicate that fish species living in similar environments and eating similar food have similar concentrations of pollutants.

Most specimens of goby fishes were caught in highly polluted coastal waters, especially semiclosed harbors polluted with industrial and domestic wastes, but concentrations of chlorinated hydrocarbons in these fishes are not significantly higher than in other commercial fishes such as mullet, annular gilthead, and black tail sea bream. However, these differences become significant when concentrations of pollutants are compared as extracted organic matter (Figure 4).

Stations for monitoring chlorinated hydrocarbon pollution of eastern coastal waters of the north and the middle Adriatic Sea are located in four different areas. The Istrian coastal area belongs to the northern region of the Adriatic Sea; Rijeka, Zadar, and Losinj areas belong to the so-called Region of Islands (*18*). The northern region of the Adriatic is predominantly affected by river waters from northern Italy which create the most severe pollution problem in the whole Adriatic. Intensive urban, tourist, agricultural, and industrial development in both coastal areas contributes to the problem. The Region of Islands includes water surrounding nearly 1000 islands along the eastern Adriatic coast and semiseparated waters between islands and main-

TABLE 1. *Chlorinated hydrocarbon concentrations in fish and mussels of east coastal waters of middle and north Adriatic Sea, 1974–75*

Station No.	Species[1]	Sampling Date	*p,p'*-DDT EOM, %	*p,p'*-DDT wet weight	*p,p'*-DDT EOM, ppm	*p,p'*-DDE wet weight	*p,p'*-DDE EOM, ppm	*p,p'*-TDE wet weight	*p,p'*-TDE EOM, ppm	Dieldrin wet weight	Dieldrin EOM, ppm	PCBs wet weight	PCBs EOM, ppm
						ISTRIAN COAST							
1	M.G.	March 1974	1.63	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1	M.G.	March 1974	1.25	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
2	M.G.	March 1974	2.79	ND	ND	8	1.40	ND	ND	—	—	ND	ND
2	M.G.	Sept. 1974	0.71	29	4.10	21	3.00	26	3.60	ND	ND	85	11.97
2	M.G.	Sept. 1974	0.37	13	3.50	13	3.40	14	3.80	1	0.32	34	9.20
2	M.G.	March 1974	2.94	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
2	G.	Sept. 1974	0.84	16	1.90	15	1.80	13	1.60	2	0.24	ND	ND
3	M.G.	March 1974	1.16	23	1.94	18	1.51	10	1.81	3	0.26	ND	ND
3	M.G.	March 1974	2.53	41	1.65	15	0.60	13	0.53	—	—	25	0.98
3	M.G.	March 1974	8.03	45	0.56	39	0.48	17	0.21	—	—	115	1.43
3	M.G.	Sept. 1974	0.62	15	2.41	19	3.05	20	3.22	4	0.58	ND	ND
3	M.G.	Sept. 1974	0.62	34	5.48	29	4.68	38	6.13	9	1.45	168	27.10
3	M.G.	Sept. 1974	1.37	27	1.93	49	3.28	28	2.12	4	0.26	85	6.20
4	M.G.	Oct. 1972	3.79	105	2.78	30	0.80	53	1.40	—	—	367	9.68
4	M.G.	March 1973	1.52	35	2.30	65	4.30	44	2.90	—	—	256	16.80
5	M.G.	Oct. 1972	2.26	ND	ND	1	0.06	ND	ND	—	—	4	0.16
5	M.G.	March 1973	1.22	ND	ND	ND	ND	ND	ND	—	—	ND	ND
5	M.G.	Oct. 1975	0.41	2	0.44	1	0.16	1	0.16	ND	ND	ND	ND
	D.A.	Sept. 1974	4.05	130	3.21	80	1.98	40	0.99	13	0.31	195	4.80
	O.M.	Sept. 1974	1.30	46	3.53	27	2.07	29	2.22	4	0.34	45	3.50
	B.B.	Sept. 1974	2.75	133	4.85	43	1.87	30	1.09	—	—	422	15.36
	P.E.	Sept. 1974	0.91	9	0.99	14	1.54	5	0.48	1	0.12	45	4.90
	M.A.	Oct. 1973	1.57	24	1.5	29	1.84	15	0.95	ND	ND	ND	ND
	O.M.	Sept. 1974	2.76	60	2.17	36	1.30	18	0.65	4	0.15	80	2.90
	P.E.	Sept. 1974	2.59	4	0.15	16	0.62	13	0.50	2	0.08	ND	ND
	M.B.	Sept. 1974	2.64	30	1.13	20	0.76	14	0.53	6	0.24	ND	ND
	M.A.	Sept. 1974	3.67	64	1.95	48	1.31	76	2.07	15	0.40	520	14.20
						RIJEKA AREA							
1	M.G.	March 1974	0.52	ND	ND	ND	ND	ND	ND	—	—	ND	ND
1	M.G.	March 1974	0.80	15	1.85	7	0.87	ND	ND	—	—	8	0.94
1	M.G.	Sept. 1974	0.68	5	0.74	4	0.53	4	0.53	ND	ND	11	1.60
1	M.G.	Sept. 1974	1.25	12	0.96	5	0.40	13	1.02	ND	ND	192	14.20
1	G.	Sept. 1974	0.89	8	0.84	9	0.96	3	0.35	1	0.07	27	3.10
2	M.G.	March 1974	2.20	131	6.05	49	2.22	32	1.45	ND	ND	23	1.03
2	M.G.	March 1974	0.74	7	1.25	5	0.65	10	1.28	0.3	0.09	ND	ND
2	M.G.	March 1974	1.20	83	6.90	21	1.77	10	0.79	ND	ND	75	6.20
2	M.G.	March 1974	1.05	28	2.52	9	0.90	7	0.65	ND	ND	8	0.84
2	M.G.	March 1974	1.35	15	1.10	8	0.61	7	0.54	—	—	9	0.65
2	M.G.	March 1974	1.20	63	5.20	28	2.30	38	3.15	—	—	128	9.80
2	M.G.	Sept. 1974	0.74	8	1.02	4	0.47	12	1.57	ND	ND	168	22.50
2	M.G.	Sept. 1974	0.82	7	0.85	2	0.25	8	1.02	ND	ND	75	9.10
2	M.G.	Sept. 1974	0.63	23	3.65	4	0.55	22	3.42	ND	ND	83	13.20
2	M.G.	Sept. 1974	0.56	42	7.50	5	0.80	48	8.60	ND	ND	77	13.70
2	M.G.	Sept. 1974	0.58	22	3.80	8	1.29	36	6.20	ND	ND	164	28.20
3	M.G.	March 1974	1.70	23	1.32	5	0.28	11	0.64	3	0.18	63	3.70
3	M.G.	Sept. 1974	0.56	13	2.20	6	1.03	9	1.60	2	0.29	64	11.40
3	G.	Sept. 1974	1.06	8	0.72	5	0.43	9	0.85	1	0.12	168	15.90
4	M.G.	March 1973	1.10	49	4.50	17	1.54	6	0.52	—	—	234	21.40
4	M.G.	Oct. 1975	0.24	2	0.79	1	0.42	1	0.59	ND	ND	26	10.60
5	G.	Oct. 1975	3.29	49	1.48	86	2.60	14	0.44	ND	ND	159	4.90
	D.A.	Sept. 1974	2.03	4	0.17	2	0.10	2	0.10	1	0.05	16	0.76
	M.A.	Oct. 1975	4.15	35	0.85	32	0.78	28	0.67	8	0.20	356	8.60
	B.B.	Oct. 1975	14.13	30	0.21	29	0.20	10	0.07	10	0.07	174	1.20
	M.B.	Sept. 1974	1.95	ND	ND	9	0.46	10	0.52	ND	ND	115	5.85
	P.E.	Sept. 1974	0.53	4	0.72	6	1.10	1	0.15	—	—	4	0.71
	M.Mer.	Sept. 1974	0.32	14	4.40	14	4.40	2	0.77	—	—	8	2.35
	M.Mer.	Oct. 1975	1.75	20	1.11	8	0.47	8	0.47	ND	ND	98	5.57
	G.M.	Oct. 1975	0.92	ND	ND	ND	ND	ND	ND	ND	ND	25	2.70
	M.B.	Oct. 1975	2.52	ND	ND	ND	ND	ND	ND	ND	ND	53	2.08
	P.D.	Sept. 1974	1.20	11	0.92	22	1.84	5	0.35	2	0.13	28	2.30
	L.C.	Sept. 1974	0.66	12	1.82	19	2.80	7	1.14	2	0.24	20	3.00
						ZADAR AREA							
1	M.G.	March 1974	3.20	14	0.42	9	0.28	ND	ND	—	—	ND	ND
1	M.G.	March 1974	2.60	17	0.64	9	0.33	ND	ND	—	—	ND	ND
1	M.G.	Sept. 1974	0.93	13	0.13	1	0.07	19	2.04	1	0.07	ND	ND
1	M.G.	Sept. 1974	0.87	33	3.80	3	0.32	ND	ND	2	0.26	80	9.20
1	G.	March 1974	1.69	ND	ND	ND	ND	ND	ND	—	—	ND	ND
1	G.	Sept. 1974	1.17	8	0.68	6	0.51	7	0.60	2	0.14	ND	ND
2	M.G.	March 1974	1.10	7	0.59	7	0.59	ND	ND	—	—	ND	ND
2	M.G.	March 1974	1.40	11	0.81	4	0.28	6	0.43	ND	ND	ND	ND
2	M.G.	March 1974	1.70	63	3.70	28	1.62	11	0.62	—	—	200	11.60
2	M.G.	March 1974	1.76	37	2.25	14	0.78	23	1.32	ND	ND	345	19.50
2	M.G.	March 1974	1.07	33	3.10	10	0.97	20	1.87	4	0.37	390	36.50

(*Continued next page*)

TABLE 1 (cont'd.). *Chlorinated hydrocarbon concentrations in fish and mussels of east coastal waters of middle and north Adriatic Sea, 1974–75*

Station No.	Species[1]	Sampling Date	*p,p'*-DDT		*p,p'*-DDE		*p,p'*-TDE		Dieldrin		PCBs	
			EOM, %	Wet weight	EOM, ppm	Wet weight	EOM, ppm	Wet weight	EOM, ppm	Wet weight	EOM, ppm	Wet weight	EOM, ppm
2	M.G.	Sept. 1974	0.49	5	1.02	2	0.45	3	0.65	ND	ND	11	2.30
2	M.G.	Sept. 1974	0.72	6	0.76	4	0.50	5	0.69	ND	ND	36	5.00
2	M.G.	Sept. 1974	0.71	18	2.55	18	2.55	24	3.38	1	0.20	326	46.00
2	M.G.	Sept. 1974	1.66	7	0.42	6	0.34	14	0.80	1	0.07	336	22.20
2	M.G.	Sept. 1974	0.76	6	0.79	7	0.92	7	0.92	ND	ND	36	4.80
2	G.	March 1974	0.70	43	6.07	113	16.07	68	9.64	2	0.29	148	21.10
2	G.	Sept. 1974	1.60	17	1.06	14	0.85	2	0.10	3	0.16	11	0.68
						LOSINJ ISLAND							
1	M.G.	March 1974	1.58	375	23.80	88	5.50	134	8.50	—	—	200	12.70
1	M.G.	March 1974	1.60	46	2.90	25	1.56	44	2.70	7	0.43	138	8.60
1	M.G.	March 1974	1.60	138	8.60	61	3.85	75	4.70	—	—	ND	ND
1	M.G.	Sept. 1974	0.78	8	0.98	5	0.50	8	0.98	ND	ND	120	15.40
1	M.G.	Sept. 1974	0.73	25	3.42	7	0.95	46	6.30	ND	ND	222	30.40
1	M.G.	Sept. 1974	0.57	8	1.32	5	0.79	22	3.80	ND	ND	157	27.50
1	M.G.	Oct. 1975	0.84	33	3.86	16	1.88	33	3.86	ND	ND	94	11.20
1	G.	March 1974	1.41	90	6.40	45	3.20	98	6.91	ND	ND	ND	ND
1	G.	March 1974	1.30	78	6.00	30	2.30	83	6.40	4	0.31	43	3.30
1	G.	Sept. 1974	0.73	44	6.04	68	9.32	870	119.00	3	0.40	152	20.80
2	M.G.	March 1974	1.40	119	8.50	44	3.13	38	2.73	—	—	ND	ND
2	M.G.	March 1974	2.20	128	5.80	39	1.76	41	1.85	—	—	112	5.40
2	M.G.	Sept. 1974	1.13	27	2.38	27	2.38	159	14.10	ND	ND	130	11.30
2	M.G.	Sept. 1974	1.20	13	1.08	9	0.71	78	6.50	ND	ND	202	16.90
2	M.G.	Oct. 1975	0.52	30	5.80	24	4.62	27	5.20	ND	ND	220	42.30
2	G.	March 1974	1.60	94	5.61	20	1.20	19	1.13	—	—	724	45.10
2	G.	Sept. 1974	1.48	17	1.15	59	3.95	250	17.00	ND	ND	112	7.60
	M.A	Oct. 1975	9.41	172	1.83	107	1.14	130	1.38	ND	ND	295	3.12
	D.A.	Sept. 1974	15.40	215	1.39	158	1.03	120	0.78	13	0.08	360	2.34
	D.A.	Oct. 1975	1.57	7	0.44	3	0.18	1	0.07	1	0.07	14	0.86
	O.M.	Sept. 1974	3.11	82	3.65	70	2.25	50	1.60	7	0.22	624	20.00
	O.M.	Oct. 1975	1.65	25	1.50	15	0.91	10	0.61	ND	ND	90	5.50
	B.B.	Sept. 1974	2.00	43	2.12	38	1.87	14	0.69	—	—	151	7.50
	M.B.	Oct. 1975	8.61	42	0.49	85	0.99	37	0.43	5	0.06	128	1.50
	P.E.	Oct. 1975	0.60	17	0.88	16	2.63	6	1.00	ND	ND	54	9.00
	B.S.	Sept. 1974	1.68	8	0.65	7	0.39	8	0.45	5	0.27	ND	ND
	B.S.	Oct. 1975	2.00	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	S.S.	Oct. 1975	3.78	15	0.36	21	0.50	ND	ND	4	0.10	102	2.40
	M.Ma.	Oct. 1975	1.31	90	6.90	62	4.75	30	2.30	2	0.13	40	3.05

Note: ND = not detectable; — = not measured; EOM = extracted organic matter.

[1] Names of species in Latin, English, and Croatoserbian: M.G. = *Mytilus galloprovincialis*, Mediterranean mussel, Dagnja; G. = *Gobius* (several species), Goby, Glavoc; D.A. = *Diplodus annularis* L., Annular gilthead, Spar; O.M. = *Oblada melanura* L., Saddled bream, Usata; B.B. = *Boops boops* L., Bogue, Bukva; P.E. = *Pagellus erythrinus* L., Pandora, Rumenac; M.A. = *Mugil auratus risso*, Golden grey mullet, Skocac zlatac; M.B. = *Mullus barbatus*, Red mullet, Barbun; M.Mer. = *Merlucius merlucius* L., Hake, Oslic; G.M. = *Gadus merlangus* L., Whiting, Mol; L.C. = *Lepidotrigla cavilone* Lac., unknown, Cucin; B.S. = *Boops salpa* L., Saupe, Salpa; S.S. = *Serranus scriba* L., Painted comber, Pirka; M.Ma. = *Maena maena* L., Caockarel, Modrak; T.D. = *Trachinus draco* L., Greater weever, Pauk bijelac.

land. Sparsely populated karstic islands and mountains, with modest agriculture and almost no industry, constitute the hinterland of these waters. But also in this region are several pollution nuclei: the Bay of Rijeka and nearby towns of Bakar, Zadar, and Sibenik; the Bay of Kastela and the neighboring town of Split.

Chlorinated hydrocarbon pollutants of marine environments can originate from such land-based sources as direct industrial discharges, sewage, and rubbish. But indirect discharges of these pollutants, especially as agricultural runoff of pesticides and farm wastes into rivers, also contribute significantly to their concentration in marine environments (*4, 7, 16*). These direct and indirect discharges are the most common sources of local pollution. Air is an important secondary source of chlorinated hydrocarbon pollution (*1*); wet and dry fallout contributes to the regional or even global pollution of the marine environment.

Concentrations of ΣDDT, dieldrin, and PCBs in mussels, goby fishes, and other benthic fishes according to their sampling areas are presented in Figures 5–7. Stationary species of mussels and goby fishes, which are indicators of local pollution, were often sampled near the source of waste discharges. Other benthic fishes indicate broader areas of pollution. Data in the figures show differences in arithmetic means of residues in mussels, goby fishes, and benthic fishes between the areas investigated. Since concentrations vary considerably, ΣDDT and PCB residues in mussels and benthic fishes were analyzed in order to find whether arithmetic means differ significantly among the areas investigated (Table 2). Mussels from the Losinj area had significantly higher concentrations of ΣDDT than had those from any other area investigated. Significantly higher PCB concentrations were found in the Losinj area than along the Istrian coast and Bay of Rijeka, but PCB residues were lower than were DDT concentrations. In fish

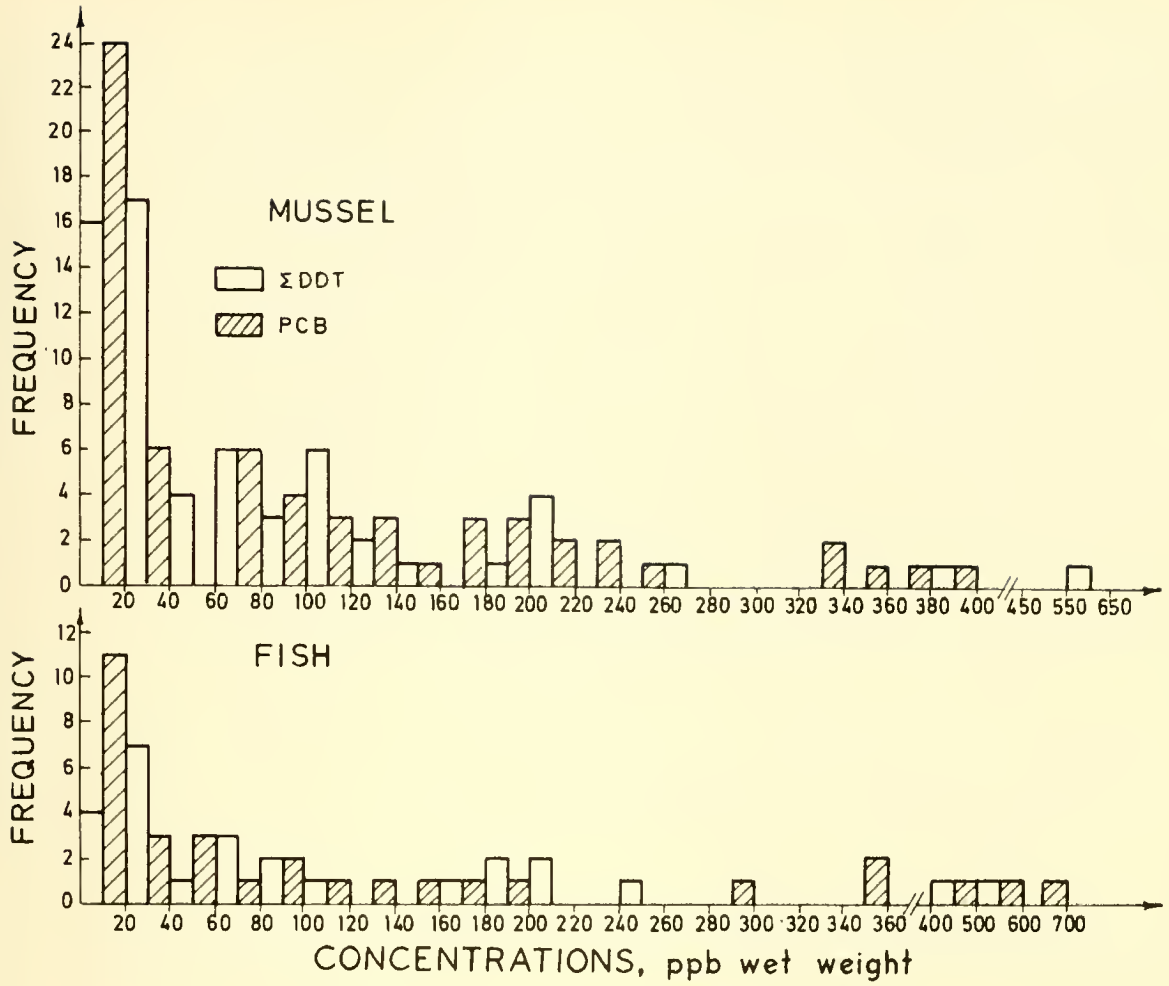

FIGURE 2. *Distribution frequencies of ΣDDT and PCBs in mussels and fish from east coastal waters of middle and north Adriatic Sea*

samples, the only concentrations that differed significantly by area were ΣDDT concentrations in samples from Rijeka Bay versus those from the Losinj area and in samples from the Istrian coast versus those from the Rijeka area. PCB concentrations did not differ significantly.

Table 3 shows significant differences in arithmetic means of ΣDDT and PCB concentrations in fish and mussel samples from the same area. No major difference between ΣDDT and PCB concentrations is indicated in mussels and benthic fishes from the same area. Significant difference appears only in ΣDDT concentrations in fish from the Istrian coastal area.

The ratio of PCB and pesticide concentrations frequently is used for identifying chlorinated hydrocarbon pollution of marine areas. If this ratio is higher than 1, the source of pollution is more likely industrial than agricultural. The ratios of PCB and ΣDDT concentrations in samples investigated during the present monitoring program are given in Figure 8. Only in the Rijeka area is this ratio significantly higher than 1 for all the indicator organisms investigated.

To determine main sources of chlorinated hydrocarbon pollution in eastern Adriatic coastal waters, correlation between ΣDDT and PCB concentrations in mussel and fish samples was investigated (Figure 9). Statistical results of the analysis are presented in Table 4 as Pearson's correlation coefficients.

Significant correlation between concentrations of ΣDDT and PCBs existed only in mussels from the Istrian coastal area and fish from the Rijeka area. This suggests two possibilities: different sources of DDT and PCB residues in the areas investigated, or different uptake and loss pathways of ΣDDT and PCBs for mussels and fish.

Several papers have been published on investigations of chlorinated hydrocarbons in Adriatic biota and sedi-

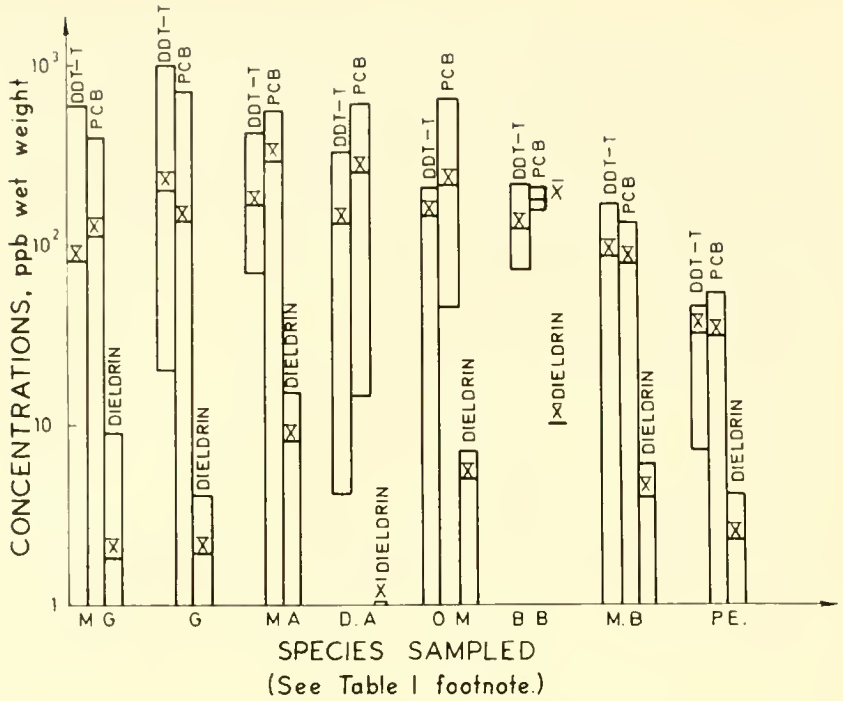

FIGURE 3. *Concentrations (wet weight) of ΣDDT, dieldrin, and PCBs in mussels and fish from east coastal waters of middle and north Adriatic Sea*

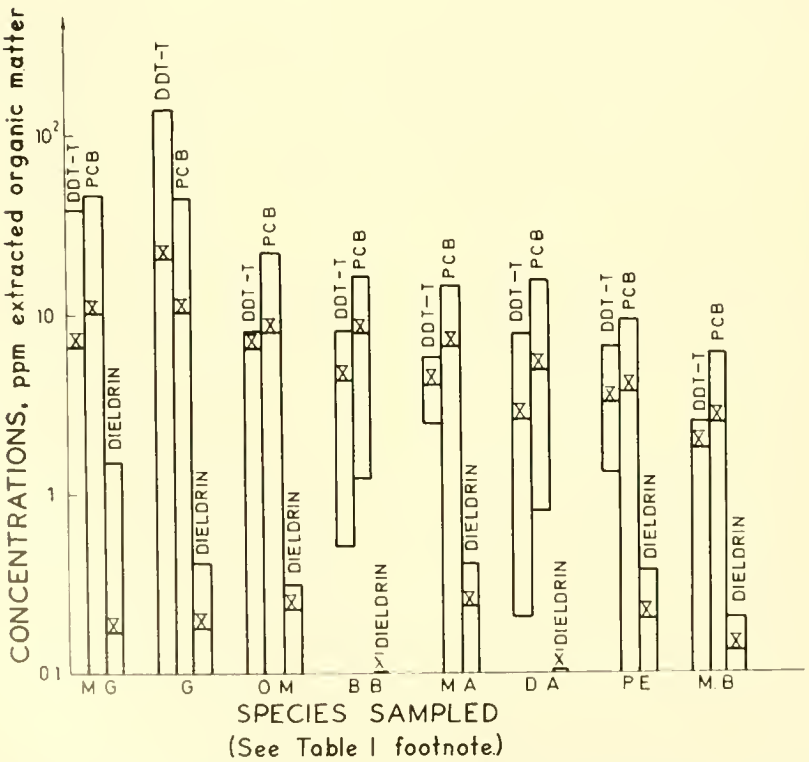

FIGURE 4. *Concentrations (extracted organic matter) of ΣDDT, dieldrin, and PCBs in mussels and fish from east coastal waters of middle and north Adriatic Sea*

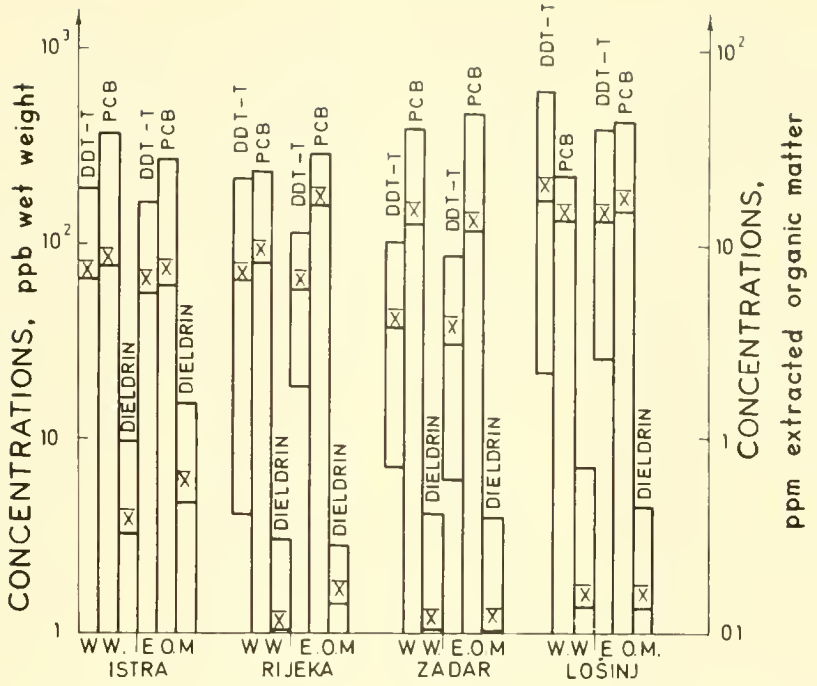

FIGURE 5. *Comparison of ΣDDT, dieldrin, and PCB concentrations in mussels from east coastal waters of middle and north Adriatic Sea*

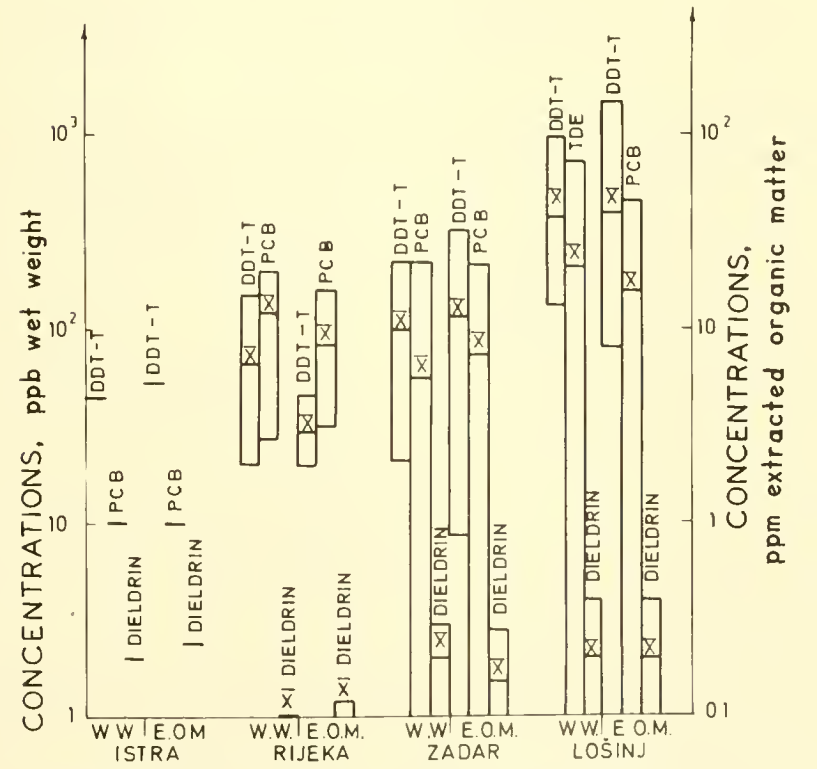

FIGURE 6. *Comparison of ΣDDT, dieldrin, and PCB concentrations in goby fishes from east coastal waters of middle and north Adriatic Sea*

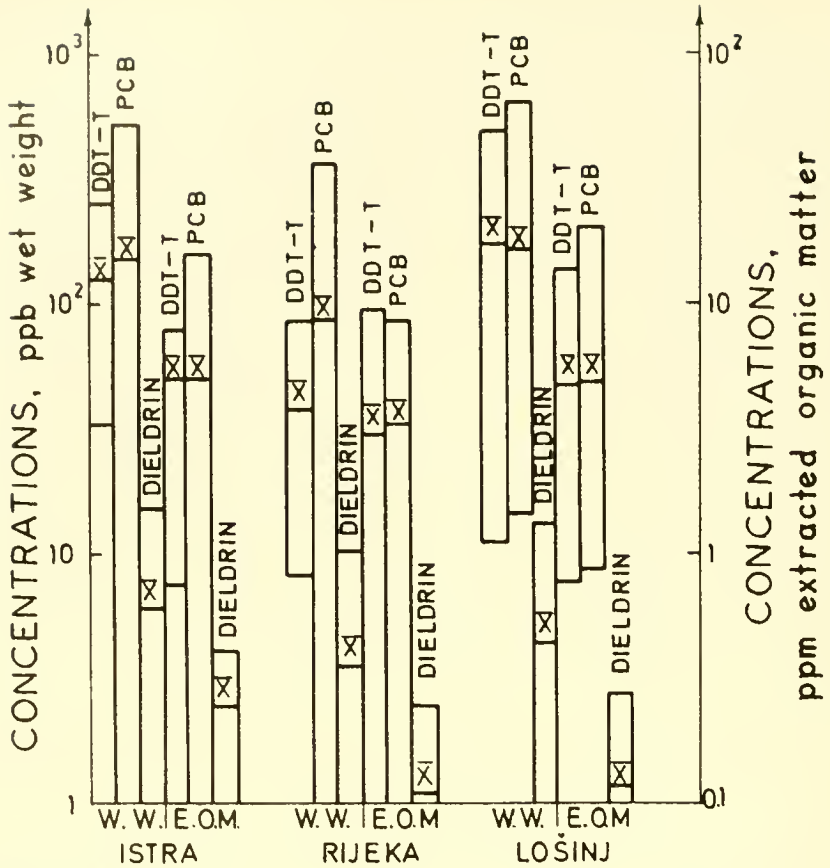

FIGURE 7. *Comparison of ΣDDT, dieldrin, and PCB concentrations in benthic fishes from east coastal waters of middle and north Adriatic Sea*

TABLE 2. *Results of Student's t-test for ΣDDT and PCB concentrations in mussels and fish from same areas of middle and north Adriatic Sea, 1974–75*

Compared Areas	Significant Difference of Arithmetic Means			
	ΣDDT		PCBs	
	Mussels	Fishes	Mussels	Fishes
Istrian coast—Rijeka	None	0.01	None	None
Istrian coast—Zadar	0.1	NC	None	NC
Istrian coast—Losinj Island	0.1	None	0.1	None
Rijeka—Zadar	0.1	NC	0.1	NC
Rijeka—Losinj Island	0.01	0.05	0.1	None
Zadar—Losinj Island	0.01	NC	None	NC

Note: NC = not calculated.

TABLE 3. *Results of Student's t-test for ΣDDT and PCB concentrations in mussels and fish from same areas of middle and north Adriatic Sea, 1974–75*

Area	Significant Difference of Arithmetic Means	
	ΣDDT	PCBs
Istrian coast	0.05	None
Rijeka	None	None
Losinj Island	None	None

ments (*3, 15, 18, 20, 21*). But difficulties of analyzing chlorinated hydrocarbons in marine samples are numerous (*5, 14*), and results of the present study were not compared with published results because analytical methods of the various studies have not been intercalibrated.

Conclusions

Analyses of chlorinated hydrocarbons in biota from eastern coastal waters of the middle and north Adriatic sea lead authors to several conclusions.

Although major north Italian rivers polluted with chlorinated hydrocarbons discharge their loads into the North Adriatic, samples from Istrian coastal waters did not have significantly higher concentrations of these pollutants than did other waters.

High concentrations of chlorinated hydrocarbons in marine organisms from Losinj Island indicate a probable uptake of pollutants in North Adriatic waters.

Chlorinated hydrocarbon levels often differ dramatically in samples collected at stations which are close together,

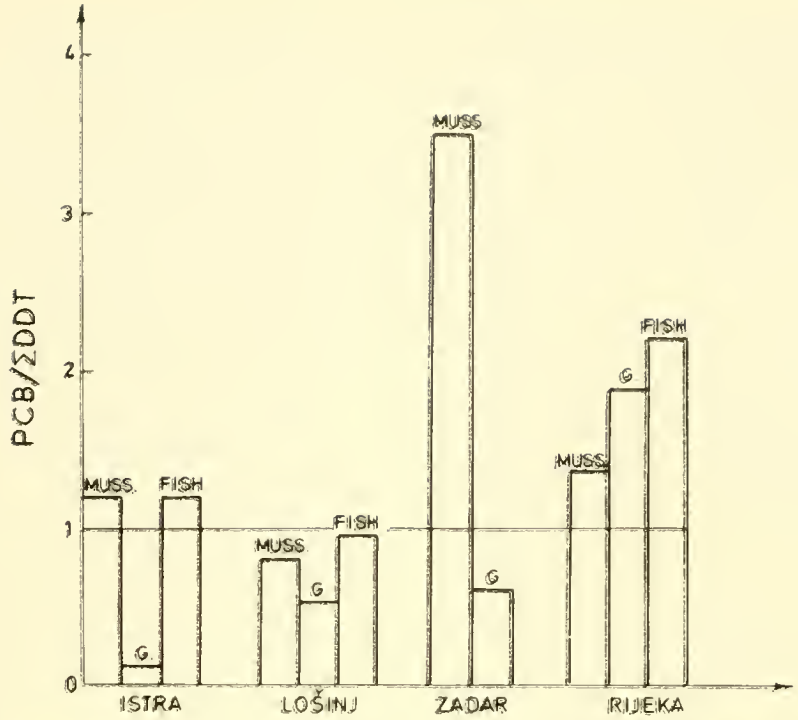

FIGURE 8. *Comparison of ratios of PCBs to ΣDDT concentrations in mussels, goby fishes, and benthic fishes from east coastal waters of middle and north Adriatic Sea*

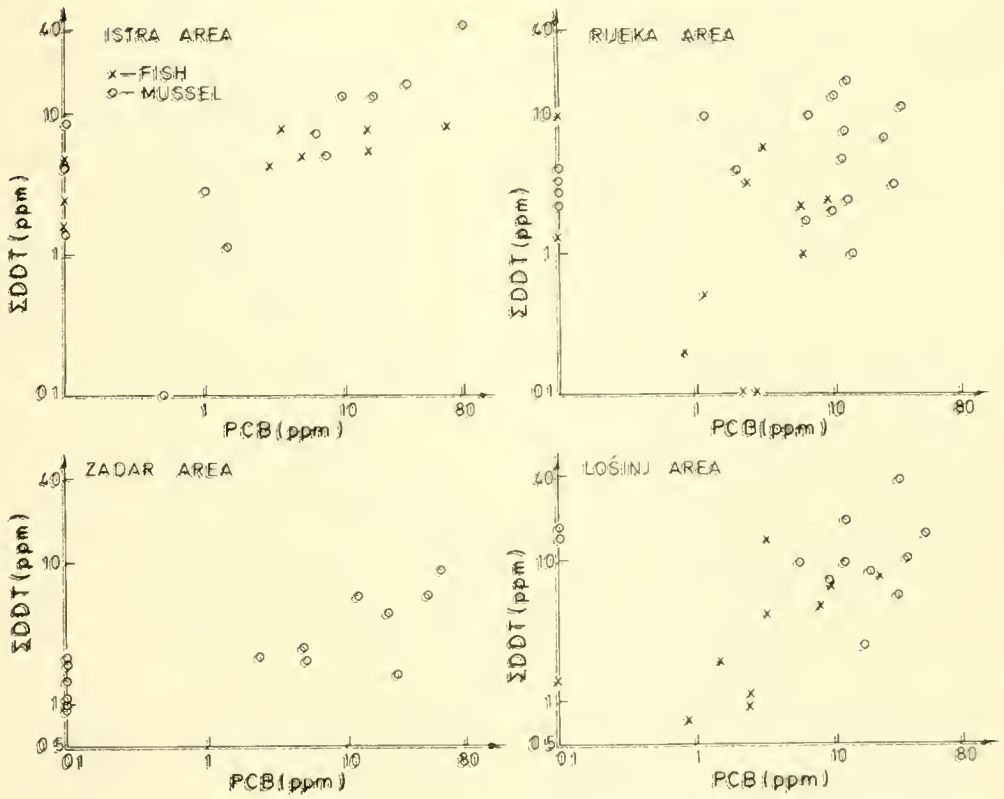

FIGURE 9. *Correlation of ΣDDT and PCB concentrations in mussels and benthic fishes from east coastal waters of middle and north Adriatic Sea*

TABLE 4. *Pearson's coefficient of correlation between ΣDDT and PCB concentrations in mussels and fish from coastal waters of middle and north Adriatic Sea, 1974–75*

Area	Mussels	Fish
Istrian coast	0.927	0.740
Rijeka	0.205	0.815
Zadar	0.712	NC
Losinj Island	−0.069	0.578

Note: NC = not calculated.

possibly because the first station waters had been contaminated with waste waters and the second station had not. Evidently urban waste waters even from small settlements contribute significantly to the contamination of Adriatic coastal waters by chlorinated hydrocarbon pollutants.

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Organochlorine Residues and Reproduction in the Little Brown Bat, Laurel, Maryland—June 1976

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ABSTRACT

Twelve of 43 pregnant little brown bats (Myotis lucifugus) *collected at Montpelier Barn, Laurel, Maryland, gave birth to dead young. Eleven of these 12 dead neonates were abnormally small. Most of the stillbirths were attributable to unknown reproductive difficulties associated with first pregnancies, but four may have been due to high concentrations of polychlorinated biphenyls (PCB) in the newborn. Residues of the PCB, DDE, and oxychlordane crossed the placenta at similar rates.*

Introduction

A study of wild-caught, pregnant big brown bats *(Eptesicus fuscus)* suggested that Aroclor 1260, a polychlorinated biphenyl (PCB), caused young to be stillborn (*3*). However, experimental elevation of Aroclor 1260 levels produced no additional stillbirths (*2*). The results indicated only that both stillbirths and high levels of Aroclor 1260 were characteristic of young adult female big brown bats.

The present study was undertaken after dead neonate little brown bats *(Myotis lucifugus)* were observed at Laurel, Maryland, roosts. Authors wished to determine whether high organochlorine residues are associated with stillbirths of little brown bats and, if so, whether this association resembles that found in big brown bats.

Materials and Methods

On June 3, 1976, 45 pregnant little brown bats were collected in Montpelier Barn at the Montpelier Mansion State Historical Site, Laurel, Prince Georges County, Maryland. Bats were confined individually at Patuxent Wildlife Research Center in stainless steel wire mesh cages, 18 cm × 22 cm × 37 cm, equipped with rodent watering bottles. Laboratory temperature averaged 28.2°C. Subdued sunlight entered two draped windows.

Before being caged, the bats were anesthetized individually with the inhalant anesthetic Metofane (Pittman-Moore, Inc., Fort Washington, Pennsylvania) and the occlusal tip width of the upper canine (canine tip width, CTW) was measured with an ocular micrometer in a 30× dissecting microscope. This measurement is an indicator of relative age (*1*).

Pregnant bats were fed mealworms, larvae of the beetle *Tenebrio molitor*, samples of which had been found free of organochlorine residues.

Parturition began June 3, and the last young was born June 13. All pregnancies produced single young. After parturition, each female and her young were killed by freezing. Two females never gave birth: one died of unknown causes June 8, and the other was frozen June 8 because she apparently was not pregnant although a small embryo (0.564 g) was found during dissection.

ANALYTICAL PROCEDURES

Adults were prepared for analysis as carcasses; young were analyzed whole, except for removal of the gastrointestinal tract, according to procedures described previously (*2*). Gastrointestinal tracts were left in several small fetuses (0.9 g or less) where removal would have been difficult.

Samples were ground with anhydrous sodium sulfate. The dried mixture was extracted with hexane in a paper extraction thimble on a Soxhlet extractor for about 7 hours. The extract was cleaned by Florisil column chromatography, and the eluate containing the pesticides and PCB was fractionated by Silicar column chromatography (*5*). The fractions were analyzed with a Hewlett-Packard Model 5753 gas-liquid chromatograph equipped with a ^{63}Ni detector, automatic sampler, and computing integrator. Instrument parameters and operating conditions follow:

Column:	glass, 1.83 m, packed with a mixture of 1.5 percent OV-17 and 1.95 percent QF-1
Temperatures:	column 200°C, detector 300°C, injection port 250°C
Carrier gas:	5 percent methane in argon flowing at 60 ml/minute; purge flow, 40 ml/minute

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Samples were analyzed for *p,p'*-DDE, *p,p'*-TDE, *p,p'*-DDT, dieldrin, endrin, heptachlor epoxide, mirex, oxychlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor, hexachlorobenzene (HCB), toxaphene, and PCBs. The PCB that was recovered resembled Aroclor 1260 in all cases.

Recoveries from spiked mallard duck (*Anas platyrhynchos*) tissues ranged from 80 to 104 percent. Residue data were not adjusted on the basis of these recoveries. The lower limit of sensitivity was 0.1 ppm. Residues in 10 percent of the samples were confirmed on an LKB Model 9000 gas-liquid chromatograph–mass spectrometer operated as described previously (*5*). Samples for one adult and two young were lost during analysis. Results are given as ppm wet weight.

Geometric means are given for residues because the data were positively skewed. Arithmetic means are given with standard errors; geometric means are given with 95 percent confidence intervals (CI). Residue levels reported as not detected (ND) were entered as zeros. To allow conversion to logs and/or machine plotting of the data, a constant was added to each value in those data series that included zeros (Fig. 1). Regression lines were fitted by the least-squares method.

Results and Discussion

CONDITION OF NEWBORN LITTLE BROWN BATS

Of 43 bats that gave birth, 12 (27.9 percent) produced dead young. Eleven of the 12 dead young weighed less (0.048–0.869 g) than the smallest liveborn bat (1.072 g). The twelfth dead neonate weighed 1.541 g. Six of the 12 dead young were partly eaten by their mothers: one young was missing its wing tips; a second, one wing and one foot; and a third, both wings and both feet. Only the head and the vertebral column of the fourth remained, and only heads remained of the other two.

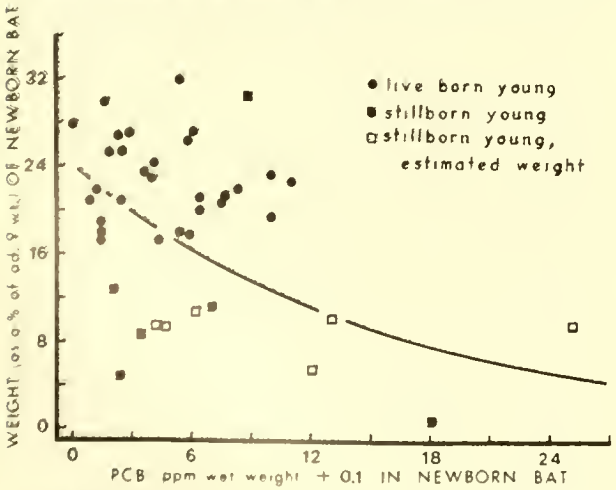

FIGURE 1. *Relationship of weight as a percent of adult female weight to Aroclor 1260 concentration among 41 neonatal little brown bats*
(Sample includes all neonates except two whose extracts were lost during chemical analysis.)

Total weights of the six young were estimated from the remaining portions. Estimations for the latter three young were based on a head-length-to-body-weight relationship derived from the undersized dead young that were recovered intact. The incompleteness of these six specimens probably did not seriously bias the results of the chemical analyses except perhaps for the latter three, which may actually have contained higher concentrations of chemicals than were estimated because most of the young bats' fat, and, therefore, residues, was in the body portions eaten by the mother. Nevertheless, residues of the PCB for these three bats (6.1, 12, and 25 ppm) exceeded the mean (Table 1) and included the maximum.

Wimsatt (*6*) observed several times that a majority of a group of females of *Myotis lucifugus* in advanced pregnancy aborted their fetuses, usually stillborn, within a few hours of removal from a colony. He attributed this result to handling or confinement. In the present study, dead young tended to be more common among later births, but beyond this tendency there was no clear pattern. When all 43 births were divided into four groups of 11, 11, 11, and 10 according to chronological order, the incidences of dead young were 9.1, 18.2, 54.5, and 30.0 percent, respectively. So, the possible roles of handling and confinement in stillbirths were not clarified by the present study.

TABLE 1. *Principal organochlorine residues in adult female little brown bats and their young, Laurel, Maryland—June 1976*

Chemical	Residues, ppm Wet Weight	
	Adults (n = 44)	Young (n = 43)
PCB (Aroclor 1260)		
Geometric mean	11.38	4.16[1]
95% CI	9.68–13.38	3.08–5.61
Range	3.6–24	ND–25
DDE		
Geometric mean	1.65	0.50[2]
95% CI	1.50–1.82	0.36–0.69
Range	0.72–3.4	ND–2.2
DDT		
Geometric mean	0.08[3]	—[4]
95% CI	0.05–0.13	—
Range	ND–1.0	—
Oxychlordane		
Geometric mean	0.45[2]	—[4]
95% CI	0.33–0.60	—
Range	ND–1.6	—
Dieldrin		
Geometric mean	0.13[5]	—[4]
95% CI	0.08–0.19	—
Range	ND–0.94	—

NOTE: CI = confidence interval; ND = not detected.
[1] Residue was not detected in 1 sample.
[2] Residue was not detected in 2 samples.
[3] Residue was not detected in 12 samples.
[4] Residue was not detected in 20 or more samples.
[5] Residue was not detected in 7 samples.

GENERAL LEVELS OF RESIDUES

Except for the PCB, levels of organochlorines in females and their young were generally low (Table 1) and similar to those found in big brown bats from Montpelier Barn (*3*). Levels of the PCB in adult little brown bats were 5.8 times greater than those found in the June 1974 collections of big brown bats; the amounts in newborn little brown bats were 3.5 times greater than those in newborn big brown bats (*3*).

Eighteen pregnant big brown bats that had been dosed with Aroclor 1260 (*2*) contained 1.8 times the concentration found in little brown bats in the present study when their carcasses were analyzed after parturition. The young of big brown bats contained a mean residue of 4.38 ppm, similar to the mean residue of 4.16 ppm found in neonates in the present study.

PLACENTAL TRANSFER OF RESIDUES

Amounts in micrograms of the PCB, DDE, and oxychlordane in young were computed as percentages of the amounts in adults, using the 29 females whose live-born young appeared to be full-term. The results were 13.2 ± 1.3 percent, 14.3 ± 1.5 percent, and 8.6 ± 1.7 percent, respectively. Paired t tests showed that the average percentage for oxychlordane was significantly less than that of either of the other chemicals. However, 13 of the values for oxychlordane in newborns were zero (not detected), and when zero values were eliminated ($n = 16$) the respective averages became 15.6 ± 1.9 percent, 17.0 ± 2.2 percent, and 15.5 ± 1.6 percent and there were no significant differences. Elimination of zeros was probably justified for this comparison because the small absolute amounts of oxychlordane made their detection less likely. These percentages resembled those for both control and dosed big brown bats when Aroclor 1260 was fed experimentally (*2*), but they were lower than one of two percentages for Aroclor 1260 and higher than both percentages for DDE found earlier in big brown bats that had not been dosed (*3*).

RESIDUES AND DEAD YOUNG

Dead young averaged more than twice as much PCB (mean = 6.68 ppm, $n = 12$) as did live young (mean = 3.04 ppm, $n = 29$), but the difference was not significant at the 95 percent level ($t = 1.91$, $0.1 > p > 0.05$). Levels of DDE and oxychlordane were almost identical in dead and live young.

Possible effects of the PCB on weight of the young were calculated by correlating the ppm PCB in the young with the weight of the young expressed as a percentage of adult female weight; the result (Fig. 1) was significant ($r = -0.47$, $0.01 > p > 0.001$). When the six data points based on estimated weights were eliminated, the relationship remained significant ($r = -0.46$, $0.01 > p > 0.001$). Although this relationship suggests that the PCB may have caused some neonates to be small, the plotted data in Figure 1 also indicate that neonates may at the same time be small and contain little PCB. A similar analysis for DDE produced no significant correlations.

To determine whether weight of the young was related to residues in adult females, a correlation was made between weight of the young as a percentage of adult female weight, and ppm of the PCB, DDE, DDT, oxychlordane, and dieldrin in adult females. No significant relationships were found. Also, females that produced dead young did not contain residues significantly higher than those of females that produced live young.

RESIDUES IN FEMALES COMPARED WITH RESIDUES IN YOUNG

The relationships between total micrograms of the PCB, DDE, and oxychlordane in adult females and in their newborn young were tested using all 29 pairs of females and young in which the neonates were entire and of normal size. Micrograms of residues in the young were dependent in a positive, linear fashion on the amount in the adult female: PCB $r = 0.74$, $p < 0.001$; DDE $r = 0.60$, $p < 0.001$; oxychlordane $r = 0.48$, $0.01 > p > 0.001$. Similar relationships were found in other bat species (*3*, *4*).

RESIDUES COMPARED WITH DAYS IN CAPTIVITY

Micrograms of residues of the PCB, DDE, DDT, oxychlordane, dieldrin, and *trans*-nonachlor in carcasses of adult females were compared to days in captivity for all 44 females in which residues were measured. Only oxychlordane declined significantly, from an average 2.6 μg to 1.0 μg. The 11-day interval was probably too short to produce any major declines such as that for PCBs found earlier in big brown bats confined for 43 days (*3*).

RESIDUES COMPARED WITH AGE OF FEMALE

No correlations were found between age estimated by CTW and residues (total μg in females plus young, $n = 44$) of the PCB, DDE, DDT, oxychlordane, dieldrin, and *trans*-nonachlor, whereas PCB residues declined significantly with age in big brown bats (*2*, *3*).

CAUSE OF STILLBIRTHS

Aroclor 1260 did not cause stillbirths in big brown bats, but high PCB levels and stillbirths were associated because both occurred more often in younger parent female bats (*2*, *3*). In the present study, CTW and PCB concentrations were not correlated. Furthermore, when CTW for females with dead young (mean = 0.1169 ± 0.0213 mm, $n = 12$) was compared with CTW for females with live young (mean = 0.1217 ± 0.0130 mm,

$n = 31$), the difference was highly significant among big brown bats (*2*). Nevertheless, there appears to be an association between age and incidence of stillbirths. Among the neonates represented in Figure 1, there were seven small dead bats, less than 16 percent of the female parent's weight that had PCB concentrations equal to or less than 7 ppm. Five of the seven female parents of these bats showed no wear on their canines and were probably yearlings producing their first offspring. Among the 30 neonates that were heavier than 16 percent of the female's weight (Fig. 1), only nine showed no canine wear. The difference between these ratios is significant ($x^2 = 4.14$, $0.05 > p > 0.01$). Threfore, unknown reproductive difficulties associated with first pregnancies probably accounted for most of the young that were born dead. Beyond these, however, there remain the four dead young with the largest amounts of the PCB (12, 13, 18, and 25 ppm); none of their female parents was a yearling.

Therefore, high levels of the PCB may have caused four young bats to be born dead, but feeding studies with captive bats are needed to confirm this conclusion.

Acknowledgment

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SOILS

Pesticide Residue Levels in Soils and Crops, 1971— National Soils Monitoring Program (III)

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ABSTRACT

Data from the 1971 National Soils Monitoring Program are summarized. Composite samples of soil and mature crops were scheduled for collection from 1,533 4-hectare sites in 37 states. Analyses were performed on 1,486 soil samples for organochlorines, organophosphates, PCBs, and elemental arsenic; samples were analyzed for atrazine only when pesticide application data indicated current-year use. Organochlorine pesticides were detected in 45 percent of the soil samples in the following order of frequency: dieldrin, ΣDDT, aldrin, chlordane, and heptachlor epoxide. Most pesticide levels ranged from 0.01 to 0.25 ppm. Crop samples were collected from 729 sites, and all were analyzed for organochlorines. Crop samples were analyzed for organophosphates and atrazine only when pesticide application data indicated current-year use. Organochlorines were detected in 42 percent of the crop samples analyzed, organophosphates in 13 percent, and atrazine in 1 percent.

Introduction

The National Soils Monitoring Program is an integral part of the National Pesticide Monitoring Program (NPMP). The NPMP was initiated at the recommendation of the President's Science Advisory Committee in 1963 to determine levels and trends of pesticides and their degradation products in the environment (*4*). The Committee recommended that appropriate federal agencies "develop a continuing network to monitor residue levels in air, water, soil, man, wildlife and fish" (*1*). The U.S. Department of Agriculture (USDA) began monitoring agricultural soils in 1964. After a series of short-term monitoring projects (*5–7*), a nationwide agricultural soil monitoring program was designed (*9*) and tested (*10*). The USDA initiated widespread monitoring in 1968 (*11*) and 1969 (*3*).

The National Soils Monitoring Program was transferred to the U.S. Environmental Protection Agency (EPA), when EPA was created in 1970. The present report summarizes soil and crop pesticide concentration data collected in 1971 (fiscal year 1972) at 1,486 sampling sites in 37 states. Data were not collected from some larger western states because of budgetary limitations and because either those states have little widespread agriculture or they grow wheat and other small grains which require fewer pesticides than do nongrain crops.

Sampling Procedures

Site selection criteria and statistical design for the present study have been described by Wiersma et al. (*9*). During late summer and fall 1971, 1,486 sites in 37 states were sampled (Fig. 1). At each 4-hectare (10-acre) site, a composite soil sample and a composite mature crop sample were collected according to procedures described in the U.S. EPA Sample Collection Manual (*8*). Information on cropping practices and a history of pesticide application for the current cropping season were obtained in interviews with landowners or operators. These data have been summarized and published separately (*2*).

Analytical Procedures

ORGANOCHLORINES AND ORGANOPHOSPHATES

Sample Preparation, Soil—A 300-g subsample was taken from a thoroughly mixed field sample. The subsample was moistened with 80 ml water and extracted with 600 ml 3:1 hexane–isopropanol by concentric rotation for 4 hours. The isopropanol was removed by

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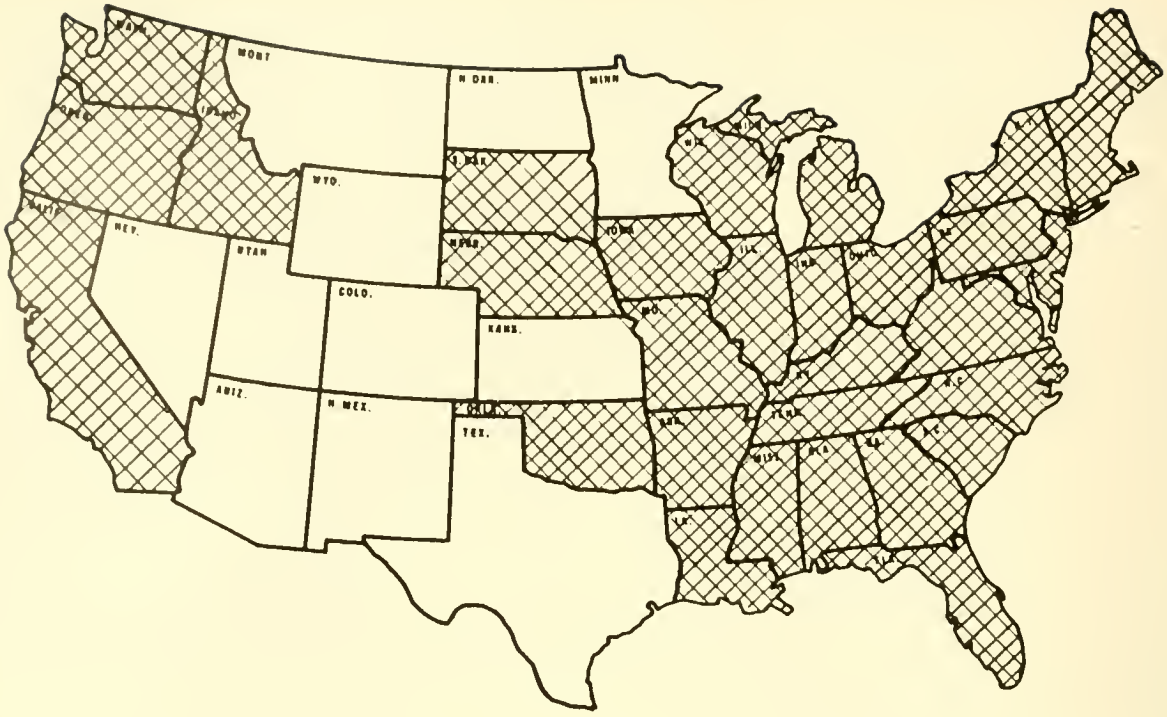

FIGURE 1. *States where agricultural soils and crops were sampled for the 1971 National Soils Monitoring Program, U.S. Environmental Protection Agency*

three distilled water washes, and the hexane extract was dried with anhydrous sodium sulfate. The sample extract was stored at low temperature for subsequent gas-liquid chromatographic analysis.

Sample Preparation, Crops—For samples containing less than 2 percent fat, e.g., alfalfa, bur clover, cornstalks, cotton stalks, green bolls, miscellaneous hay, a 100-g sample of the crop was blended with 25 ml distilled water for 3 minutes in 800 ml acetonitrile. An aliquot of the sample extract, representing 10 g of the original sample, was decanted into a 500-ml Erlemeyer flask. The sample extract was concentrated under a three-ball Snyder column to approximately 10 ml; 100 ml hexane was added, and the hexane–acetonitrile azeotrope was again concentrated to 10 ml. This process was carried out three times to remove the acetonitrile. The hexane extract was dried with anhydrous sodium sulfate, the volume was adjusted to 50 ml, and the extract was stored at low temperature until partitioning.

For crop samples containing more than 2 percent fat, e.g., corn kernels, cottonseed, soybeans, a 100-g sample was prewashed with 100 ml isopropanol and then with 100 ml hexane. Both prewashes were discarded. The prewashes were used to remove surface residues which may have contaminated the grain during removal of shells, husks, or pods, thus assuring that residues detected were actually contained in the grain. The sample was dried, dry blended, added to 100 ml isopropanol, and blended again. After 300 ml hexane was added, the isopropanol was removed by two washes with saturated aqueous NaCl solution and one wash with distilled water. The water–alcohol layers were discarded; the hexane layer was concentrated, adjusted to 50 ml, and held at low temperature until partitioning.

After extraction, crop samples were partitioned with hexane–acetonitrile as follows: 50 ml of the hexane sample extract, representing 10 g, was shaken with 100 ml acetonitrile in a 500-ml separatory funnel. The bottom acetonitrile layer was set aside. Another 100 ml acetonitrile was added to the hexane extract and the separation step described above was repeated twice; the hexane was discarded and the three acetonitrile layers were combined. The 300-ml acetonitrile extract, which contained essentially all the pesticides from the original hexane extract, was backwashed with 25 ml acetonitrile-saturated hexane, and the hexane layer was discarded. The acetonitrile sample extract was concentrated to approximately 10 ml under a three-ball Snyder column, and 100 ml hexane was added. This process was carried out three times to remove the acetonitrile. The

hexane extract was adjusted to 7.5 ml and stored at low temperature for subsequent Florisil column cleanup and fractionation.

A separate aliquot of the extract not subjected to Florisil cleanup was reserved for analysis for organophosphates by flame photometric detection.

Florisil Cleanup—An extract equivalent to 5 g original crop sample was fractionated through a 15-g Florisil column by use of 100 ml 10 percent methylene chloride in hexane and 100 ml methylene chloride for fractions one and two, respectively.

Methylene chloride was removed by concentration of each extract to low volume under a three-ball Snyder column, addition of 100 ml hexane, and concentration again to low volume. After two additions of hexane, the methylene chloride was essentially removed. Each extract volume was adjusted to 2.5 ml for separate injection on the gas-liquid chromatograph.

Gas-Liquid Chromatography—Gas chromatographs were equipped with tritium foil electron-affinity detectors for organochlorines and thermionic or flame photometric detectors for organophosphates. A multiple-column system with polar and nonpolar columns was used to identify compounds. Instrument parameters and operating conditions follow:

Gas chromatographs:	Hewlett Packard 402A Hewlett Packard 402B Tracor MT-220
Columns:	glass, 6 mm OD × 4 mm ID, 183 cm long, packed with 9 percent QF-1 on 100–120-mesh Gas-Chrom Q 3 percent DC-200 on 100–120-mesh Gas-Chrom Q a mixture of 1.5 percent OV-17 and 1.95 percent QF-1 on 100–120-mesh Supelcoport
Carrier gases:	5 percent methane-argon flowing at 80 ml/minute, prepurified nitrogen flowing at 80 ml minute
Temperatures:	thermionic detector housing 250°C detector (EC and FPD) 200–210 C injection port 250°C columns 166°C 170–175°C 185–190°C

Minimum detection levels for organochlorines and trifluralin were 0.002–0.03 ppm except for combinations of polychlorinated biphenyls (PCBs), chlordane, toxaphene, and other chemicals which had minimum detectable levels of 0.05–0.1 ppm. Minimum detectable levels for organophosphates were approximately 0.01–0.03 ppm. The compounds detectable by the methodology of the present study are listed in Table 1. Trifluralin is detected by the organochlorine methodology and, for that reason, appears with the organochlorine analyses in the tables.

Recovery Studies—Pesticide recovery values from soil were 80–110 percent, but usually were close to 100 percent. Values from crops ranged from 70 to 100 percent, and varied with amount and type of pesticide and type of crop involved. Residues in both crop and soil samples were corrected for recovery. Soil samples were also corrected to a dry-weight basis.

TABLE 1. *Compounds detectable by chemical methodology of the present study, 1971—National Soils Monitoring Program*

ORGANOCHLORINES	
Alachlor	Endrin
Aldrin	Heptachlor
Chlordane	Heptachlor epoxide
o,p'-DDT	Isodrin
p,p'-DDT	Lindane (γ-BHC)
o,p'-DDE	Methoxychlor
p,p'-DDE	Ovex
o,p'-TDE	PCBs
p,p'-TDE	PCNs
Dieldrin	Propachlor
Endosulfan (I)	Toxaphene
Endosulfan (II)	
Endosulfan sulfate	
ORGANOPHOSPHATES	
DEF	Parathion, ethyl
Diazinon	Parathion, methyl
Ethion	Phorate
Malathion	Trithion
OTHER HALOGENS	
Trifluralin	

NOTE: Although trifluralin is a dinitroaniline compound, it is detected in the methodology used in the present study, and appears in Tables 1–7 under the Organochlorines heading.

ATRAZINE

A 50-g subsample was taken from a thoroughly mixed field sample. The subsample was extracted with 25 ml water and 300 ml methanol by concentric rotation for 4 hours. The sample extract was then decanted into a 1-liter separatory funnel and 200 ml water was added. The extract was partitioned with 150 ml Freon 113 three times. The Freon 113 fractions were combined and concentrated to incipient dryness. The extract was dissolved in hexane and adjusted to 5 ml for injection into a gas-liquid chromatograph equipped with a thermionic flame detector with a rubidium sulfate coating on a helix coil. Instrument parameters and operating conditions follow:

Column:	glass, 183 cm long × 6 mm OD × 4 mm ID, packed with 3 percent Versamid 900 on 100–120-mesh Gas-Chrom Q
Carrier gas:	helium
Detector fuel gases:	oxygen flowing at 200–300 ml/minute; hydrogen flowing at 20–30 ml/minute
Temperatures:	detector 200°C injection port 240°C column 240°C

Confirmatory analyses were performed on a DC-200 column at 180°C and a Coulson detector in the reductive mode at the following temperatures: pyrolysis tube, 850°C; transfer line, 220°C; and block, 220°C. Recovery was 90–110 percent with a minimum detection level of 0.01 ppm.

ARSENIC

Arsenic was determined by atomic absorption spectrophotometry. The soil sample was extracted with 9.6*N* HCl and arsenic was reduced to As^{+3} with $SnCl_2$. As^{+3} was partitioned from the acid to benzene, and then further partitioned from benzene into water for the absorption measurement. A Perkin-Elmer Model 303 spectrophotometer was used, and absorbance was measured with an arsenic cathode lamp at 1972 A with argon as an aspirant to an air–hydrogen flame. Minimum detection limit was 0.1 ppm, and recovery averaged 70 percent.

Results from all analyses were corrected for recovery and are expressed as ppm dry weight.

Results and Discussion

Tables presented in this report can be divided into two groups: those showing concentrations of pesticides in soil samples by all sites and states, and those showing concentrations of pesticides in mature agricultural crops. Most tables list the number of analyses, the number of times a compound was detected, the percent occurrence of the compound, the arithmetic mean, the estimated geometric mean, and the minimum and maximum positive concentrations detected.

The estimated geometric mean is routinely presented in the tables as an alternative to the arithmetic mean as a measure of central tendency for the data evaluation. Pesticide residue data frequently contain a large number of zero values, resulting either from the absence of pesticides or their presence at levels below the analytical sensitivity. Such data are seldom distributed normally, as shown by tests for skewness and kurtosis, but often approximate a log-normal distribution. After repeated tests for significant kurtosis and/or skewness, the $\log(X + 0.01)$ transformation was used to determine the logarithmic means. The antilogs of these figures minus 0.01 were taken to estimate the geometric mean in the untransformed dimension. The estimated geometric mean was calculated only for those compounds with more than one positive detection.

COMPOUND CONCENTRATIONS IN CROPLAND SOIL

All Sites—A total of 1,486 soil samples were received from 1,533 sites in 37 states, resulting in a 97 percent design completion. Results of analyses for organochlorines, organophosphates, triazines, and elemental arsenic are presented in Table 2. The most frequently detected pesticide was dieldrin, found in 27 percent of all samples analyzed. Next were ΣDDT, aldrin, chlordane, and

TABLE 2. *Compound concentrations in cropland soils for all sample sites in 37 states, 1971 (FY 1972)—National Soils Monitoring Program*

Compound	No. of Positive Detections	% of Positive Detections	Residues, ppm Dry Weight			
			Arithmetic Mean	Estimated Geometric Mean[1]	Extremes of Detected Values	
					Min.	Max.
ORGANOCHLORINES, 1,486 SAMPLES						
Aldrin	144	9.7	0.02	0.002	0.01	1.88
Chlordane	119	8.0	0.06	0.003	0.01	6.98
o,p'-DDE	21	1.4	<0.01	<0.001	0.01	0.34
p,p'-DDE	334	22.5	0.11	0.007	0.01	54.98
o,p'-DDT	198	13.3	0.07	0.004	0.01	32.75
p,p'-DDT	305	20.5	0.37	0.010	0.01	245.18
o,p'-TDE	10	0.7	0.01	<0.001	0.02	16.79
p,p'-TDE	116	7.8	0.05	0.002	0.01	38.46
ΣDDT	356	24.0	0.61	0.013	0.01	388.16
Dieldrin	408	27.5	0.35	0.009	0.01	9.83
Endosulfan (I)	2	0.1	<0.01	<0.001	0.05	0.23
Endosulfan (II)	3	0.2	<0.01	<0.001	0.07	1.24
Endosulfan sulfate	3	0.2	<0.01	<0.001	0.16	2.07
Endrin	14	0.9	<0.01	<0.001	0.02	1.00
Heptachlor	73	4.9	0.01	0.001	0.01	1.37
Heptachlor epoxide	103	6.9	<0.01	0.001	0.01	0.43
Isodrin	3	0.2	<0.01	<0.001	0.01	0.02
Ovex	1	0.1	<0.01	—	1.13	—
Propachlor	3	0.2	<0.01	<0.001	0.07	0.10
Toxaphene	92	6.2	0.27	0.004	0.18	36.33
Trifluralin	52	3.5	<0.01	0.001	0.01	1.29
ORGANOPHOSPHATES, 1,141 SAMPLES						
DEF	4	0.4	<0.01	<0.001	0.15	0.66
Diazinon	4	0.4	<0.01	<0.001	0.02	0.05
Ethion	2	0.2	<0.01	<0.001	0.06	0.24
Malathion	1	0.1	<0.01	—	0.19	—
Parathion, ethyl	4	0.4	<0.01	<0.001	0.05	0.19
Phorate	1	0.1	<0.01	—	0.08	—
TRIAZINES, 213 SAMPLES						
Atrazine	152	71.4	0.23	0.052	0.01	16.73
HEAVY METALS, 1,474 SAMPLES						
Arsenic	1461	99.1	5.92	3.522	0.09	180.42

[1] Not calculated when fewer than two positive detections were present.

heptachlor epoxide, found in 24, 10, 8, and 7 percent of all samples analyzed, respectively.

Table 3 gives the occurrence of pesticide residues in the agricultural soil samples collected in 1971. The frequency of detection varied widely among the states surveyed. Atrazine detection frequencies are not comparable to the detection frequencies of other compounds because atrazine analyses were performed only when site application records indicated atrazine use during the current growing season.

Table 4 gives the percent incidence of residues of selected organochlorines at specific levels. For most compounds, the highest percentages of positive detections were in the 0.01–0.25-ppm category. Toxaphene was the exception; highest incidence of positive residues occurred in the >10.00-ppm category.

By State—Pesticide concentrations in soils of specific states or state groups are presented in Table 5. Because some of the smaller eastern states had very few sites, those with similar geographic locations and/or agricultural characteristics were combined to obtain more representative data. State groups used were: Mid-Atlantic: Delaware, Maryland, New Jersey; New England: Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, Vermont; Virginia and West Virginia.

Comparisons of the percent occurrence of aldrin, dieldrin heptachlor epoxide, ΣDDT, chlordane, and arsenic

TABLE 3. *Occurrence of organochlorine, organophosphate, and triazine residues in cropland soil, by state, 1971—National Soils Monitoring Program*

State	Organochlorines			Organophosphates			Triazines[1]		
	No. of Analyses	No. of Positive Detections	% of Positive Detections	No. of Analyses	No. of Positive Detections	% of Positive Detections	No. of Analyses	No. of Positive Detections	% of Positive Detections
Alabama	23	20	87	11	0	—	1	0	—
Arkansas	46	34	74	33	1	3	1	0	—
California	64	49	77	48	2	4	—	—	—
Florida	18	9	50	15	2	13	—	—	—
Georgia	30	3	10	15	0	—	—	—	—
Idaho	33	8	24	25	0	—	—	—	—
Illinois	142	102	72	93	3	3	23	20	87
Indiana	58	28	48	38	0	—	11	7	64
Iowa	152	108	71	104	0	—	54	44	81
Kentucky	31	3	10	31	0	—	6	5	83
Louisiana	26	20	77	12	0	—	—	—	—
Michigan	55	22	40	50	0	—	11	10	91
Mid-Atlantic	18	7	39	18	2	11	2	1	50
Mississippi	31	31	100	15	3	20	—	—	—
Missouri	80	31	39	67	0	—	20	13	65
Nebraska	106	32	30	99	2	2	21	17	81
New England	20	8	40	19	1	5	1	0	—
New York	38	12	32	35	0	—	6	6	100
North Carolina	31	27	87	7	0	—	—	—	—
Ohio	57	13	23	49	0	—	10	5	50
Oklahoma	64	7	11	58	0	—	1	1	100
Oregon	38	14	37	18	0	—	—	—	—
Pennsylvania	36	8	22	35	0	—	5	2	40
South Carolina	17	17	100	3	0	—	—	—	—
South Dakota	106	7	7	101	0	—	3	3	100
Tennessee	27	12	44	16	0	—	1	0	—
Virginia/West Virginia	27	12	44	25	0	—	—	—	—
Washington State	45	11	24	37	0	—	—	—	—
Wisconsin	67	7	10	64	0	—	36	18	50
TOTAL	1486	662	45	1141	16	1	213	152	71

[1] Samples analyzed only when application records indicated atrazine use during the current growing season.

TABLE 4. *Percent incidence of selected pesticides in cropland soil from all sampling sites in 37 states, 1971—National Soils Monitoring Program*

Concentration, ppm Dry Wt	ΣDDT[1]	Aldrin	Dieldrin	Chlordane	Heptachlor	Heptachlor Epoxide	Toxaphene	Trifluralin
Not Detected	76.0	90.3	72.5	92.0	95.1	93.1	93.8	96.5
0.01–0.25	11.2	7.9	22.3	3.4	4.4	6.7	0.1	3.2
0.26–1.00	6.3	1.3	4.8	3.1	0.4	0.2	1.1	0.2
1.01–5.00	5.1	0.5	0.3	1.4	0.1	—	3.4	0.1
5.01–10.00	0.7	—	0.1	0.1	—	—	1.0	—
>10.00	0.6	—	—	—	—	—	6.7	—
TOTAL	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0

[1] ΣDDT = *o,p'*-DDT + *p,p'*-DDT + *o,p'*-DDE + *p,p'*-DDE + *o,p'*-TDE + *p,p'*-TDE.

TABLE 5. *Compound concentrations in cropland soil, by state, 1971—National Soils Monitoring Program*

Compound	No. of Positive Detections	% of Positive Detections	Residues, ppm Dry Weight			
			Arithmetic Mean Concentration	Geometric Mean Concentration[1]	Extremes of Detected Values	
					Min.	Max.
ALABAMA, 23 SITES						
Organochlorines, 23 samples						
Chlordane	1	4.4	0.02	—	0.45	—
p,p'-DDE	18	78.3	0.10	0.044	0.01	0.33
o,p'-DDT	10	43.5	0.05	0.014	0.01	0.38
p,p'-DDT	16	69.6	0.26	0.065	0.01	1.39
ΣDDT	18	78.3	0.41	0.106	0.01	2.07
Dieldrin	4	17.4	<0.01	0.002	0.01	0.02
Endrin	1	4.4	0.02	—	0.42	—
Heptachlor	1	4.4	<0.01	—	0.07	—
Toxaphene	5	21.7	0.76	0.022	0.18	6.78
Organophosphates, 11 samples: no residues detected						
Triazines, 1 sample: no residues detected						
Heavy Metals, 23 samples						
Arsenic	23	100.0	2.84	1.855	0.39	8.13
ARKANSAS, 46 SITES						
Organochlorines, 46 samples						
Aldrin	2	4.4	<0.01	<0.001	0.01	0.02
o,p'-DDE	1	2.2	<0.01	—	0.03	—
p,p'-DDE	26	56.5	0.10	0.028	0.01	0.94
o,p'-DDT	14	30.4	0.07	0.099	0.01	0.95
p,p'-DDT	28	60.9	0.34	0.054	0.02	4.82
p,p'-TDE	15	32.6	0.07	0.012	0.01	1.03
ΣDDT	28	60.9	0.57	0.079	0.03	7.14
Dieldrin	15	32.6	0.02	0.009	0.02	0.18
Endrin	1	2.2	<0.01	—	0.10	—
Toxaphene	9	19.6	0.50	0.017	0.47	6.67
Trifluralin	2	4.4	<0.01	<0.001	0.05	0.06
Organophosphates, 33 samples						
Diazinon	1	3.0	<0.01	—	0.02	—
Triazines 1 sample: no residues detected						
Heavy Metals, 46 samples						
Arsenic	46	100.0	8.32	6.448	0.65	24.74
CALIFORNIA, 64 SITES						
Organochlorines, 64 samples						
Chlordane	1	1.6	0.04	—	2.45	—
o,p'-DDE	4	6.3	0.01	0.001	0.02	0.34
p,p'-DDE	45	70.3	0.15	0.050	0.01	0.87
o,p'-DDT	30	46.9	0.09	0.024	0.02	0.71
p,p'-DDT	39	60.9	0.33	0.064	0.01	2.53
o,p'-TDE	2	3.1	<0.01	0.001	0.01	0.20
p,p'-TDE	10	15.6	0.02	0.004	0.02	0.93
ΣDDT	47	73.4	0.61	0.123	0.01	3.88
Dieldrin	3	4.7	0.01	0.001	0.09	0.19
Endosulfan II	1	1.6	<0.01	—	0.18	—
Endosulfan sulfate	1	1.6	0.01	—	0.39	—
Heptachlor	1	1.6	<0.01	—	0.02	—
Heptachlor epoxide	1	1.6	<0.01	—	0.07	—
Ovex	1	1.6	0.02	—	1.13	—
Toxaphene	13	20.3	0.61	0.020	0.73	8.30
Trifluralin	3	4.7	0.03	0.002	0.10	1.29
Organophosphates, 48 samples						
Ethion	1	2.1	<0.01	—	0.24	—
Parathion, ethyl	1	2.1	<0.01	—	0.17	—
Heavy Metals, 54 samples						
Arsenic	54	100.0	5.26	3.802	0.72	19.14
FLORIDA, 18 SITES						
Organochlorines, 18 samples						
Aldrin	3	16.7	0.01	0.003	0.01	0.11
Chlordane	1	5.6	0.01	—	0.10	—
p,p'-DDE	6	33.3	0.04	0.011	0.02	0.42
o,p'-DDT	3	16.7	0.03	0.006	0.02	0.33
p,p'-DDT	6	33.3	0.10	0.015	0.04	1.14
p,p'-TDE	3	16.7	0.01	0.004	0.03	0.09
ΣDDT	7	38.9	0.19	0.025	0.02	1.89
Dieldrin	4	22.2	0.15	0.014	0.19	1.70
Endrin	2	11.1	0.06	0.004	0.02	1.00
Toxaphene	1	5.6	0.13	—	2.35	—
Organophosphates, 15 samples						
Ethion	1	6.7	<0.01	—	0.06	—
Parathion, ethyl	1	6.7	0.01	—	0.19	—
Heavy Metals, 18 samples						
Arsenic	16	88.9	1.49	0.575	0.12	10.11

(*Continued next page*)

TABLE 5 (cont'd.). *Compound concentrations in cropland soil, by state, 1971—National Soils Monitoring Program*

Compound	No. of Positive Detections	% of Positive Detections	Residues, ppm Dry Weight: Arithmetic Mean Concentration	Geometric Mean Concentration[1]	Extremes of Detected Values: Min.	Max.
GEORGIA, 30 SITES						
Organochlorines, 30 samples						
Chlordane	3	10.0	0.02	0.003	0.14	0.21
o,p'-DDE	1	3.3	<0.01	—	0.02	—
p,p'-DDE	25	83.3	0.14	0.062	0.01	0.83
o,p'-DDT	14	46.7	0.07	0.019	0.01	0.63
p,p'-DDT	22	73.3	0.35	0.093	0.01	2.70
o,p'-TDE	1	3.3	<0.01	—	0.03	—
p,p'-TDE	11	36.7	0.03	0.010	0.02	0.26
ΣDDT	25	83.3	0.59	0.172	0.01	4.42
Dieldrin	7	23.3	0.04	0.007	0.01	0.45
Heptachlor epoxide	2	6.7	<0.01	0.001	0.01	0.04
Toxaphene	9	30.0	1.25	0.046	1.06	10.20
Trifluralin	1	3.3	0.01	—	0.21	—
Organophosphates, 15 samples: no residues detected						
Heavy Metals, 30 samples						
Arsenic	30	100.0	1.64	1.116	0.20	6.99
IDAHO, 33 SITES						
Organochlorines, 33 samples						
p,p'-DDE	9	27.3	0.03	0.008	0.02	0.41
o,p'-DDT	4	12.1	0.01	0.002	0.02	0.27
p,p'-DDT	8	24.2	0.13	0.009	0.01	3.23
p,p'-TDE	1	3.0	<0.01	—	0.08	—
ΣDDT	9	27.3	0.18	0.013	0.04	3.99
Dieldrin	4	12.1	<0.01	0.002	0.01	0.03
Toxaphene	1	3.0	0.15	—	4.96	—
Trifluralin	2	6.1	<0.01	0.001	0.06	0.07
Organophosphates, 25 samples: no residues detected						
Heavy Metals, 31 samples						
Arsenic	31	100.0	2.17	1.785	0.30	4.99
ILLINOIS, 142 SITES						
Organochlorines, 142 samples						
Aldrin	54	38.0	0.06	0.011	0.01	1.83
Chlordane	46	31.7	0.47	0.027	0.04	6.98
p,p'-DDE	2	1.4	<0.01	<0.001	0.01	0.06
p,p'-DDT	4	2.8	<0.01	0.001	0.04	0.10
ΣDDT	5	3.5	<0.01	0.001	0.01	0.16
Dieldrin	96	66.9	0.14	0.050	0.01	0.75
Heptachlor	39	27.5	0.04	0.008	0.01	1.37
Heptachlor epoxide	45	31.7	0.02	0.008	0.01	0.34
Propachlor	2	1.4	<0.01	<0.001	0.10	—
Trifluralin	7	4.9	<0.01	0.001	0.02	0.15
Organophosphates, 93 samples						
Diazinon	1	1.1	<0.01	—	0.05	—
Malathion	1	1.1	<0.01	—	0.19	—
Phorate	1	1.1	<0.01	—	0.08	—
Triazines, 23 samples						
Atrazine	20	87.0	0.22	0.102	0.03	0.92
Heavy Metals, 141 samples						
Arsenic	141	100.0	7.8	5.950	0.86	28.22
INDIANA, 58 SITES						
Organochlorines, 58 samples						
Aldrin	14	24.1	0.08	0.009	0.01	1.64
Chlordane	6	10.3	0.12	0.006	0.16	4.10
o,p'-DDE	1	1.7	<0.01	—	0.02	—
p,p'-DDE	6	10.3	0.01	0.002	0.02	0.25
o,p'-DDT	2	3.5	<0.01	0.001	0.02	0.08
p,p'-DDT	4	6.9	0.01	0.002	0.02	0.56
p,p'-TDE	2	3.5	0.01	0.001	0.06	0.27
ΣDDT	6	10.3	0.03	0.004	0.04	0.89
Dieldrin	22	37.9	0.10	0.019	0.01	0.85
Endosulfan	1	1.7	<0.01	—	0.05	—
Endosulfan II	1	1.7	<0.01	—	0.07	—
Endosulfan sulfate	1	1.7	<0.01	—	0.16	—
Heptachlor	5	8.6	0.01	0.002	0.01	0.20
Heptachlor epoxide	5	8.6	0.01	0.002	0.01	0.43
Isodrin	1	1.7	<0.01	—	0.01	—
Trifluralin	3	5.2	<0.01	0.001	0.03	0.13
Organophosphates, 38 samples: no residues detected						
Triazines, 11 samples						
Atrazine	7	63.6	0.05	0.020	0.01	0.27
Heavy Metals, 58 samples						
Arsenic	58	100.0	4.66	3.478	0.42	15.93

(*Continued next page*)

TABLE 5 (cont'd.). *Compound concentrations in cropland soil, by state, 1971—National Soils Monitoring Program*

Compound	No. of Positive Detections	% of Positive Detections	Residues, ppm Dry Weight: Arithmetic Mean Concentration	Geometric Mean Concentration[1]	Extremes of Detected Values: Min.	Max.
IOWA, 152 SITES						
Organochlorines, 152 samples						
Aldrin	43	28.3	0.04	0.008	0.01	1.01
Chlordane	20	13.2	0.06	0.006	0.02	1.63
o,p'-DDE	1	0.7	<0.01	—	0.01	—
p,p'-DDE	17	11.2	0.01	0.002	0.01	0.19
o,p'-DDT	5	3.3	<0.01	0.001	0.02	0.24
p,p'-DDT	19	12.5	0.02	0.003	0.02	1.42
p,p'-TDE	3	2.0	<0.01	<0.001	0.01	0.04
ΣDDT	22	14.5	0.03	0.004	0.01	1.59
Dieldrin	97	63.8	0.09	0.033	0.01	0.79
Heptachlor	13	8.6	<0.01	0.001	0.01	0.15
Heptachlor epoxide	18	11.8	0.01	0.002	0.01	0.16
Isodrin	2	1.3	<0.01	<0.001	0.02	—
Toxaphene	1	0.7	0.04	—	5.97	—
Trifluralin	15	9.87	0.01	0.002	0.01	0.40
Organophosphates, 104 samples: no residues detected						
Triazines, 54 samples						
Atrazine	44	81.5	0.62	0.135	0.02	16.73
Heavy Metals, 152 samples						
Arsenic	151	99.3	6.3	4.574	0.24	26.05
KENTUCKY, 31 SITES						
Organochlorines, 31 samples						
Aldrin	1	3.2	<0.01	—	0.01	—
Chlordane	1	3.2	0.08	—	2.47	—
p,p'-DDE	2	6.5	<0.01	0.001	0.01	0.05
p,p'-DDT	1	3.2	<0.01	—	0.01	—
p,p'-TDE	1	3.2	<0.01	—	0.02	—
ΣDDT	2	6.5	<0.01	0.001	0.02	0.07
Dieldrin	1	3.2	0.02	—	0.48	—
Endosulfan	1	3.2	0.01	—	0.23	—
Endosulfan II	1	3.2	0.04	—	1.24	—
Endosulfan sulfate	1	3.2	0.07	—	2.07	—
Toxaphene	1	3.2	0.06	—	1.80	—
Organophosphates, 31 samples: no residues detected						
Triazines, 6 samples						
Atrazine	5	83.3	0.03	0.022	0.02	0.05
Heavy Metals, 31 samples						
Arsenic	31	100.0	9.25	5.608	0.74	29.31
LOUISIANA, 26 SITES						
Organochlorines, 26 samples						
Aldrin	1	3.9	<0.01	—	0.03	—
Chlordane	2	7.7	0.01	0.002	0.06	0.26
o,p'-DDE	1	3.9	0.01	—	0.25	—
p,p'-DDE	11	42.3	0.25	0.033	0.03	2.23
o,p'-DDT	10	38.5	0.24	0.020	0.01	3.66
p,p'-DDT	11	42.3	0.79	0.046	0.01	7.41
p,p'-TDE	8	30.8	0.12	0.014	0.02	1.67
ΣDDT	11	42.3	1.41	0.067	0.05	15.22
Dieldrin	7	26.9	0.02	0.006	0.01	0.15
Toxaphene	8	30.8	3.02	0.057	0.68	36.33
Trifluralin	2	7.7	0.02	0.003	0.11	0.37
Organophosphates, 12 samples: no residues detected						
Heavy Metals, 26 samples						
Arsenic	25	96.2	4.09	2.541	0.41	10.77
MICHIGAN, 55 SITES						
Organochlorines, 55 samples						
Aldrin	4	7.3	0.02	0.002	0.07	0.52
Chlordane	7	12.7	0.02	0.004	0.02	0.37
p,p'-DDE	9	16.4	0.11	0.006	0.01	4.35
o,p'-DDT	6	10.9	0.04	0.004	0.02	1.45
p,p'-DDT	8	14.6	0.22	0.007	0.02	8.20
p,p'-TDE	2	3.6	0.01	0.001	0.04	0.72
ΣDDT	9	16.4	0.38	0.008	0.02	14.72
Dieldrin	16	29.1	0.02	0.007	0.01	0.34
Heptachlor	1	1.8	<0.01	—	0.01	—
Heptachlor epoxide	3	5.5	<0.01	0.001	0.01	0.06
Trifluralin	1	1.8	<0.01	—	0.10	—
Organophosphates, 50 samples: no residues detected						
Triazines, 11 samples						
Atrazine	10	90.9	0.09	0.068	0.02	0.25
Heavy Metals, 55 samples						
Arsenic	55	100.0	8.26	4.763	0.55	73.65

(*Continued next page*)

TABLE 5 (cont'd.). *Compound concentrations in cropland soil, by state, 1971—National Soils Monitoring Program*

Compound	No. of Positive Detections	% of Positive Detections	Residues, ppm Dry Weight			
			Arithmetic Mean Concentration	Geometric Mean Concentration[1]	Extremes of Detected Values	
					Min.	Max.
MID-ATLANTIC[2], 18 SITES						
Organochlorines, 18 samples						
Aldrin	1	5.6	<0.01	—	0.05	—
Chlordane	1	5.6	<0.01	—	0.06	—
o,p'-DDE	1	5.6	<0.01	—	0.03	—
p,p'-DDE	3	16.7	0.04	0.005	0.04	0.61
o,p'-DDT	1	5.6	0.01	—	0.16	—
p,p'-DDT	2	11.1	0.04	0.004	0.08	0.71
o,p'-TDE	1	5.6	0.01	—	0.11	—
ΣDDT	3	16.7	0.10	0.007	0.04	1.62
Dieldrin	5	27.8	0.02	0.007	0.03	0.09
Heptachlor epoxide	2	11.1	0.01	0.001	0.01	—
Organophosphates, 18 samples						
Diazinon	1	5.6	<0.01	—	0.03	—
Parathion, ethyl	1	5.6	<0.01	—	0.05	—
Triazines, 2 samples						
Atrazine	1	50.0	0.03	—	0.07	—
Heavy Metals, 18 samples						
Arsenic	18	100.0	3.83	2.652	0.43	18.01
MISSISSIPPI, 31 SITES						
Organochlorines, 31 samples						
p,p'-DDE	30	96.8	0.29	0.152	0.01	1.26
o,p'-DDT	26	83.9	0.41	0.203	0.01	1.73
p,p'-DDT	30	96.8	1.98	0.611	0.01	16.07
p,p'-TDE	11	35.5	0.08	0.015	0.02	1.16
ΣDDT	30	96.8	2.68	0.922	0.02	19.97
Dieldrin	6	19.4	0.01	0.003	0.01	0.10
Endrin	2	6.5	0.02	0.002	0.02	0.64
Toxaphene	22	71.0	3.82	0.579	0.46	21.00
Trifluralin	9	29.0	0.01	0.006	0.02	0.15
Organophosphates, 15 samples						
DEF	3	20.0	0.08	0.010	0.15	0.66
Heavy Metals, 31 samples						
Arsenic	31	100.0	9.65	7.726	1.10	20.15
MISSOURI, 80 SITES						
Organochlorines, 80 samples						
Aldrin	7	8.8	0.03	0.002	0.01	1.88
Chlordane	5	6.3	0.03	0.002	0.09	1.09
o,p'-DDE	1	1.3	<0.01	—	0.09	—
p,p'-DDE	4	5.0	<0.01	0.001	0.01	0.06
o,p'-DDT	1	1.3	<0.01	—	0.05	—
p,p'-DDT	5	6.3	0.01	0.002	0.03	0.33
ΣDDT	7	8.8	0.02	0.003	0.01	0.47
Dieldrin	25	31.3	0.07	0.014	0.01	0.78
Heptachlor	4	5.0	<0.01	0.001	0.01	0.07
Heptachlor epoxide	6	7.5	<0.01	0.001	0.02	0.10
Propachlor	1	1.3	<0.01	—	0.07	—
Trifluralin	3	3.8	<0.01	0.001	0.02	0.13
Organophosphates, 67 samples: no residues detected						
Triazines, 20 samples						
Atrazine	13	65.0	0.06	0.026	0.01	0.34
Heavy Metals, 80 samples						
Arsenic	80	100.0	5.02	3.739	0.88	21.86
NEBRASKA, 106 SITES						
Organochlorines, 106 samples						
Aldrin	1	0.9	<0.01	—	0.02	—
Chlordane	8	7.6	0.02	0.002	0.02	0.71
o,p'-DDE	1	0.9	<0.01	—	0.02	—
p,p'-DDE	4	3.8	0.01	0.001	0.02	0.55
o,p'-DDT	2	1.9	<0.01	0.001	0.02	0.41
p,p'-DDT	5	4.7	0.02	0.002	0.01	1.35
ΣDDT	5	4.7	0.03	0.002	0.03	2.33
Dieldrin	32	30.2	0.02	0.008	0.01	0.31
Endrin	2	1.9	<0.01	<0.001	0.06	0.08
Heptachlor	3	2.8	0.01	<0.001	0.01	—
Heptachlor epoxide	9	8.5	0.01	0.001	0.01	0.07
Organophosphates, 99 samples						
DEF	1	1.0	<0.01	—	0.20	—
Diazinon	1	1.0	<0.01	—	0.03	—
Triazines, 21 samples						
Atrazine	17	81.0	0.07	0.042	0.02	0.28
Heavy Metals, 106 samples						
Arsenic	104	98.1	5.24	3.282	0.41	18.37

(*Continued next page*)

TABLE 5 (cont'd.). *Compound concentrations in cropland soil, by state, 1971—National Soils Monitoring Program*

Compound	No. of Positive Detections	% of Positive Detections	Residues, ppm Dry Weight			
			Arithmetic Mean Concentration	Geometric Mean Concentration[1]	Extremes of Detected Values	
					Min.	Max.
NEW ENGLAND[2], 20 SITES						
Organochlorines, 20 samples						
Aldrin	1	5.0	0.01	—	0.28	—
Chlordane	1	5.0	0.11	—	2.20	—
p,p'-DDE	6	30.0	0.07	0.014	0.06	0.44
o,p'-DDT	4	20.0	0.02	0.006	0.05	0.22
p,p'-DDT	6	30.0	0.16	0.019	0.03	0.90
o,p'-TDE	1	5.0	<0.01	—	0.05	—
p,p'-TDE	5	25.0	0.07	0.010	0.02	0.92
ΣDDT	6	30.0	0.32	0.026	0.09	2.16
Dieldrin	3	15.0	0.17	0.006	0.03	3.26
Heptachlor	1	5.0	<0.01	—	0.04	—
Heptachlor epoxide	1	5.0	<0.01	—	0.03	—
Organophosphates, 19 samples						
Parathion, ethyl	1	5.3	0.01	—	0.14	—
Triazines, 1 sample: no residues detected						
Heavy Metals, 20 samples						
Arsenic	19	95.0	8.56	2.841	0.60	69.10
NEW YORK, 38 SITES						
Organochlorines, 38 samples						
Chlordane	2	5.3	0.01	0.002	0.11	0.40
o,p'-DDE	1	2.6	<0.01	—	0.10	—
p,p'-DDE	11	29.0	1.74	0.016	0.01	54.98
o,p'-DDT	7	18.4	1.31	0.013	0.01	32.75
p,p'-DDT	10	26.3	7.69	0.022	0.02	245.18
o,p'-TDE	2	5.3	0.45	0.003	0.23	16.79
p,p'-TDE	5	13.2	1.07	0.008	0.11	38.46
ΣDDT	11	29.0	12.26	0.028	0.02	388.16
Dieldrin	4	10.5	0.28	0.005	0.03	9.83
Heptachlor epoxide	1	2.6	<0.01	—	0.03	—
Trifluralin	1	2.6	<0.01	—	0.14	—
Organophosphates, 35 samples: no residues detected						
Triazines, 6 samples						
Atrazine	6	100.0	0.18	0.136	0.04	0.38
Heavy Metals, 38 samples						
Arsenic	38	100.0	11.63	5.466	0.41	180.42
NORTH CAROLINA, 31 SITES						
Organochlorines, 31 samples						
Aldrin	1	3.2	<0.01	—	0.04	—
Chlordane	2	6.5	0.05	0.003	0.37	1.06
o,p'-DDE	2	6.5	<0.01	0.001	0.01	0.05
p,p'-DDE	25	80.7	0.08	0.043	0.01	0.50
o,p'-DDT	18	58.1	0.05	0.022	0.01	0.51
p,p'-DDT	25	80.7	0.27	0.087	0.01	2.62
p,p'-TDE	18	58.1	0.05	0.024	0.03	0.23
ΣDDT	26	83.9	0.46	0.169	0.02	3.63
Dieldrin	14	54.2	0.04	0.015	0.01	0.13
Endrin	1	3.2	<0.01	—	0.03	—
Heptachlor	1	3.2	0.01	—	0.34	—
Heptachlor epoxide	1	3.2	<0.01	—	0.08	—
Toxaphene	7	22.6	0.65	0.022	0.51	12.00
Organophosphates, 7 samples: no residues detected						
Heavy Metals, 31 samples						
Arsenic	28	90.3	2.41	0.996	0.38	17.95
OHIO, 57 SITES						
Organochlorines, 57 samples						
Aldrin	4	7.0	0.01	0.002	0.02	0.33
Chlordane	3	5.3	0.02	0.002	0.05	0.86
p,p'-DDE	4	7.0	0.12	0.003	0.04	4.55
o,p'-DDT	2	3.5	0.07	0.002	0.15	3.79
p,p'-DDT	4	7.0	0.51	0.004	0.06	23.70
o,p'-TDE	1	1.8	<0.01	—	0.05	—
p,p'-TDE	3	5.3	0.06	0.002	0.05	2.07
ΣDDT	5	8.8	0.76	0.005	0.04	34.11
Dieldrin	6	10.5	0.02	0.004	0.06	0.46
Heptachlor	1	1.8	<0.01	—	0.14	—
Heptachlor epoxide	2	3.5	<0.01	<0.001	0.01	0.04
Trifluralin	1	1.8	<0.01	—	0.10	—
Organophosphates, 49 samples: no residues detected						
Triazines, 10 samples						
Atrazine	5	50.0	0.25	0.047	0.05	1.38
Heavy Metals, 57 samples						
Arsenic	57	100.0	14.17	9.858	1.11	48.97

(*Continued next page*)

TABLE 5 (cont'd.). *Compound concentrations in cropland soil, by state, 1971—National Soils Monitoring Program*

Compound	No. of Positive Detections	% of Positive Detections	Residues, ppm Dry Weight: Arithmetic Mean Concentration	Geometric Mean Concentration[1]	Extremes of Detected Values: Min.	Max.
OKLAHOMA, 65 SITES						
Organochlorines, 64 samples						
o,p'-DDE	1	1.6	<0.01	—	0.03	—
p,p'-DDE	5	7.8	0.03	0.002	0.01	1.72
o,p'-DDT	1	1.6	<0.01	—	0.03	—
p,p'-DDT	3	4.7	0.01	0.001	0.01	0.85
p,p'-TDE	1	1.6	<0.01	—	0.12	—
ΣDDT	6	9.4	0.05	0.002	0.01	3.02
Dieldrin	2	3.1	<0.01	0.001	0.08	0.23
Endrin	1	1.6	<0.01	—	0.05	—
Organophosphates, 58 samples: no residues detected						
Triazines, 1 sample						
Atrazine	1	100.0	0.05	—	0.05	—
Heavy Metals, 65 samples						
Arsenic	64	98.5	2.66	1.872	0.32	10.08
OREGON, 38 SITES						
Organochlorines, 38 samples						
Aldrin	2	5.3	<0.01	0.001	0.02	—
o,p'-DDE	1	2.6	<0.01	—	0.01	—
p,p'-DDE	12	31.6	0.45	0.008	0.01	16.69
o,p'-DDT	6	15.8	0.12	0.003	0.01	4.51
p,p'-DDT	6	15.8	0.49	0.006	0.03	18.20
p,p'-TDE	2	5.3	<0.01	0.001	0.01	0.10
ΣDDT	12	31.6	1.07	0.011	0.01	39.40
Dieldrin	6	15.8	0.07	0.006	0.06	2.15
Endrin	2	5.3	<0.01	0.009	0.03	—
Heptachlor epoxide	1	2.6	<0.01	—	0.01	—
Organophosphates, 18 samples: no residues detected						
Heavy Metals, 38 samples						
Arsenic	38	100.0	5.04	2.830	0.38	61.81
PENNSYLVANIA, 36 SITES						
Organochlorines, 36 samples						
Aldrin	1	2.8	<0.01	—	0.15	—
p,p'-DDE	3	8.3	0.01	0.002	0.05	0.14
o,p'-DDT	1	2.8	<0.01	—	0.01	—
p,p'-DDT	3	8.3	0.01	0.002	0.02	0.15
ΣDDT	3	8.3	0.01	0.003	0.07	0.30
Dieldrin	5	13.9	0.02	0.003	0.01	0.49
Endrin	1	2.8	<0.01	—	0.06	—
Organophosphates, 35 samples: no residues detected						
Triazines, 5 samples						
Atrazine	2	40.0	0.02	0.009	0.03	0.05
Heavy Metals, 36 samples						
Arsenic	36	100.0	6.83	5.979	1.96	17.19
SOUTH CAROLINA, 17 SITES						
Organochlorines, 17 samples						
Aldrin	1	5.9	<0.01	—	0.01	—
p,p'-DDE	17	100.0	0.24	0.182	0.01	0.47
o,p'-DDT	15	88.2	0.23	0.127	0.02	0.91
p,p'-DDT	17	100.0	0.85	0.544	0.05	3.38
p,p'-TDE	4	23.5	0.08	0.012	0.12	0.55
ΣDDT	17	100.0	1.40	0.908	0.06	4.65
Dieldrin	6	35.3	0.11	0.014	0.02	1.42
Toxaphene	13	76.5	3.17	0.636	0.49	18.10
Organophosphates, 3 samples: no residues detected						
Heavy Metals, 17 samples						
Arsenic	17	100.0	1.75	1.085	0.13	9.59
SOUTH DAKOTA, 106 SITES						
Organochlorines, 106 samples						
Aldrin	1	0.9	<0.01	—	0.08	—
Chlordane	4	3.8	0.01	0.001	0.03	0.36
p,p'-DDE	1	0.9	<0.01	—	0.01	—
p,p'-DDT	1	0.9	<0.01	—	0.01	—
ΣDDT	1	0.9	<0.01	—	0.02	—
Dieldrin	5	4.7	<0.01	0.001	0.01	0.27
Heptachlor epoxide	4	3.8	<0.01	0.001	0.01	0.05
Organophosphates, 101 samples: no residues detected						
Triazines, 3 samples						
Atrazine	3	100.0	0.19	0.166	0.09	0.31
Heavy Metals, 106 samples						
Arsenic	105	99.1	6.04	5.231	0.90	31.05

(*Continued next page*)

TABLE 5 (cont'd.). *Compound concentrations in cropland soil, by state, 1971—National Soils Monitoring Program*

Compound	No. of Positive Detections	% of Positive Detections	Residues, ppm Dry Weight: Arithmetic Mean Concentration	Geometric Mean Concentration[1]	Extremes of Detected Values: Min.	Max.
TENNESSEE, 27 SITES						
Organochlorines, 27 samples						
Chlordane	1	3.7	0.02	—	0.49	—
p,p'-DDE	9	33.3	0.09	0.013	0.01	1.28
o,p'-DDT	7	25.9	0.03	0.009	0.01	0.23
p,p'-DDT	10	37.0	0.17	0.022	0.01	1.59
p,p'-TDE	4	14.8	0.01	0.003	0.02	0.15
ΣDDT	11	40.7	0.30	0.030	0.01	2.29
Dieldrin	1	3.7	<0.01	—	0.02	—
Heptachlor	1	3.7	<0.01	—	0.02	—
Heptachlor epoxide	1	3.7	<0.01	—	0.12	—
Toxaphene	2	7.4	0.16	0.005	2.06	2.14
Trifluralin	2	7.4	<0.01	0.001	0.01	—
Organophosphates, 16 samples: no residues detected						
Triazines, 1 sample: no residues detected						
Heavy Metals, 27 samples						
Arsenic	27	100.0	8.52	7.114	1.53	16.78
VIRGINIA/WEST VIRGINIA[2], 27 SITES						
Organochlorines, 27 samples						
Chlordane	1	3.7	0.03	—	0.83	—
o,p'-DDE	2	7.4	0.01	0.001	0.01	0.14
p,p'-DDE	9	33.3	0.21	0.010	0.02	5.41
o,p'-DDT	3	11.1	0.02	0.003	0.03	0.36
p,p'-DDT	5	18.5	0.16	0.007	0.03	3.78
o,p'-TDE	2	7.4	0.05	0.003	0.12	1.35
p,p'-TDE	5	18.5	0.29	0.006	0.02	7.47
ΣDDT	9	33.3	0.74	0.017	0.02	18.51
Dieldrin	5	18.5	0.01	0.003	0.01	0.08
Endrin	1	3.7	0.02	—	0.51	—
Heptachlor	1	3.7	<0.01	—	0.12	—
Heptachlor epoxide	1	3.7	<0.01	—	0.08	—
Organophosphates, 25 samples: no residues detected						
Heavy Metals, 27 samples						
Arsenic	27	100.0	3.48	2.081	0.41	16.66
WASHINGTON STATE, 45 SITES						
Organochlorines, 45 samples						
Aldrin	2	4.4	<0.01	0.001	0.01	0.03
Chlordane	2	4.4	0.01	0.001	0.01	0.45
o,p'-DDE	2	4.4	<0.01	0.001	0.01	0.06
p,p'-DDE	10	22.2	0.11	0.007	0.01	2.74
o,p'-DDT	4	8.9	0.02	0.003	0.10	0.48
p,p'-DDT	5	11.1	0.21	0.008	0.53	3.21
p,p'-TDE	2	4.4	0.04	0.002	0.07	1.83
ΣDDT	10	22.2	0.39	0.011	0.01	7.46
Dieldrin	4	8.9	<0.01	0.001	0.02	0.04
Organophosphates, 37 samples: no residues detected						
Heavy Metals, 45 samples						
Arsenic	45	100.0	3.29	2.279	0.64	32.07
WISCONSIN, 67 SITES						
Organochlorines, 67 samples						
Chlordane	1	1.5	<0.01	—	0.03	—
p,p'-DDE	5	7.5	0.01	0.002	0.01	0.32
o,p'-DDT	2	3.0	<0.01	0.001	0.13	0.20
p,p'-DDT	2	3.0	0.02	0.001	0.51	0.94
ΣDDT	5	7.5	0.04	0.002	0.01	1.46
Dieldrin	3	4.5	<0.01	0.001	0.07	0.15
Heptachlor	1	1.5	<0.01	—	0.01	—
Organophosphates, 64 samples: no residues detected						
Triazines, 36 samples						
Atrazine	18	50.0	0.06	0.020	0.01	0.63
Heavy Metals, 67 samples						
Arsenic	66	98.5	1.53	1.039	0.09	12.66

[1] Not calculated when fewer than two positive detections were present.

[2] Some smaller eastern states with few sites, but which have similar geographic locations and/or agricultural characteristics were combined to obtain more representative data including: Mid-Atlantic states: Delaware, Maryland, New Jersey. New England states: Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, Vermont; Virginia/West Virginia.

are presented in Figures 2–7. The key for each figure is based on the arithmetic average percent occurrence ($\bar{x}$) of the compound for all sites. The four classes are described as: greater than $2\bar{x}$; greater than $\bar{x}$ but less than $2\bar{x}$; greater than $\frac{1}{2}\bar{x}$ but less than $\bar{x}$; and less than $\frac{1}{2}\bar{x}$.

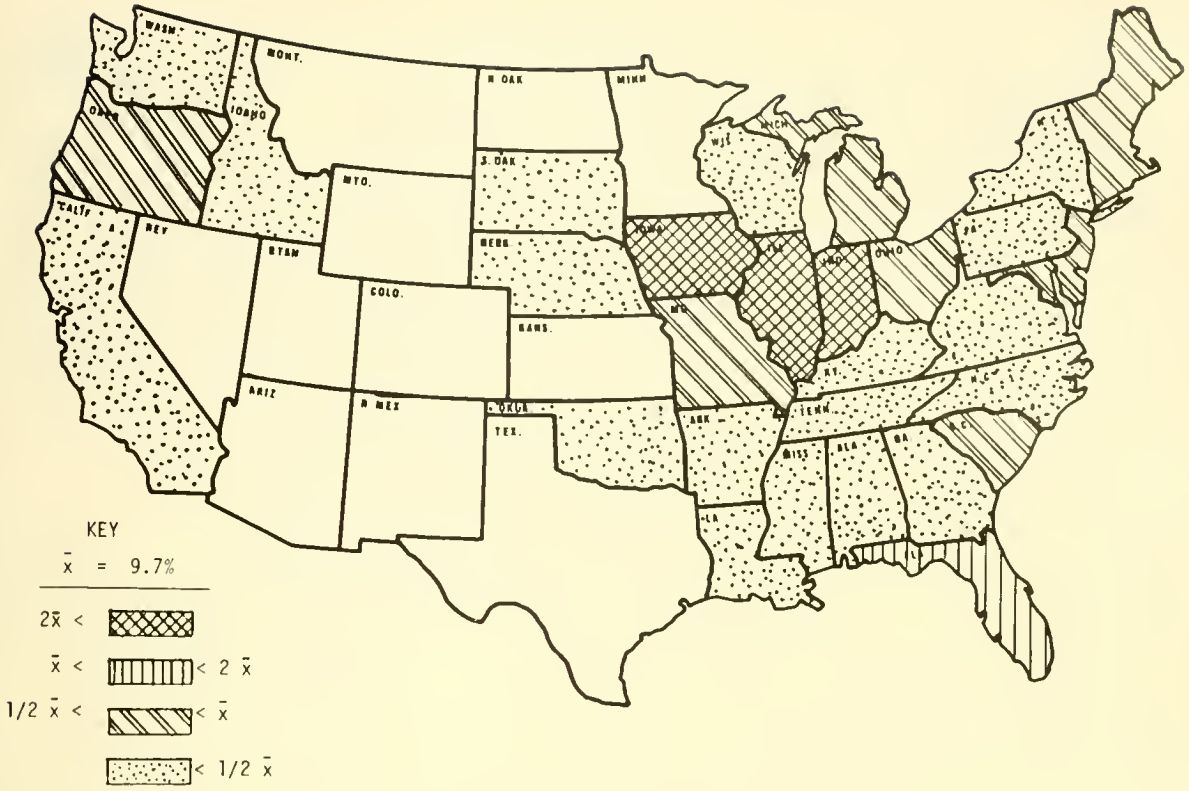

FIGURE 2. *Percent occurrence of aldrin residue detections in cropland soil, by state, 1971, National Soils Monitoring Program, U.S. Environmental Protection Agency*

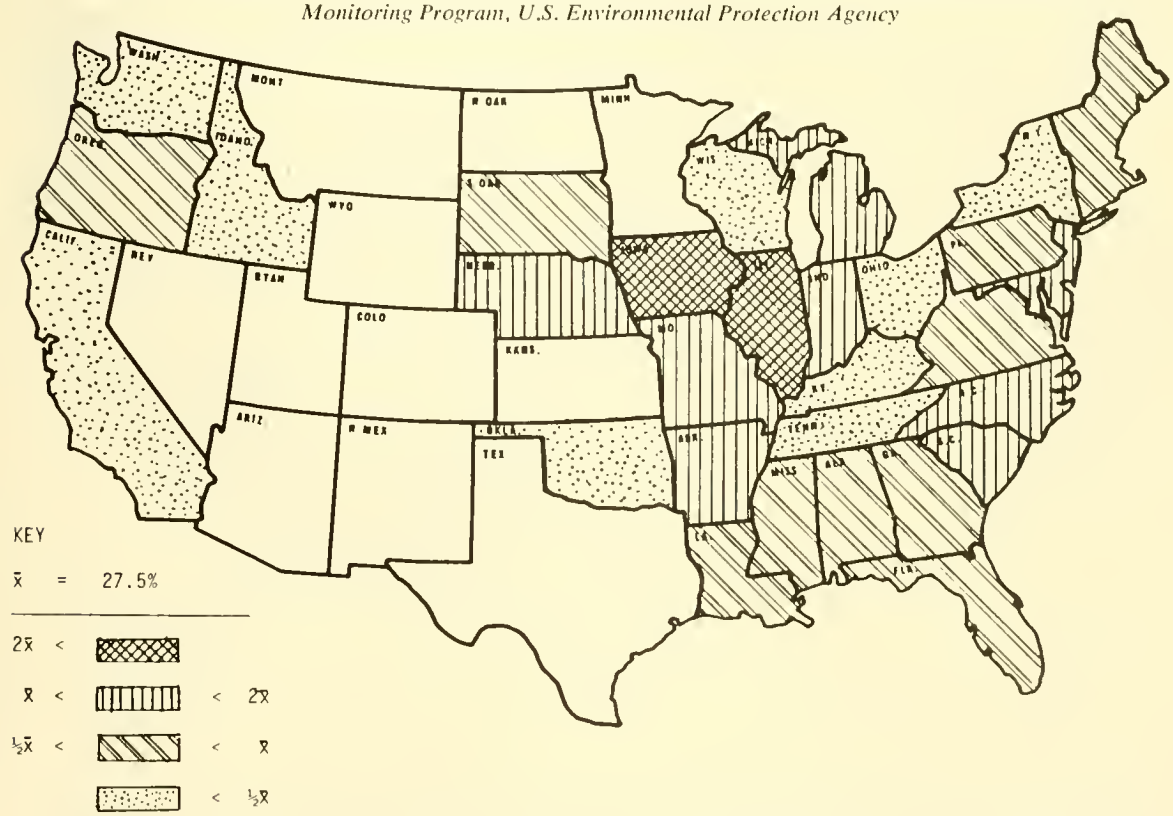

FIGURE 3. *Percent occurrence of dieldrin residue detections in cropland soil, by state, 1971, National Soils Monitoring Program, U.S. Environmental Protection Agency*

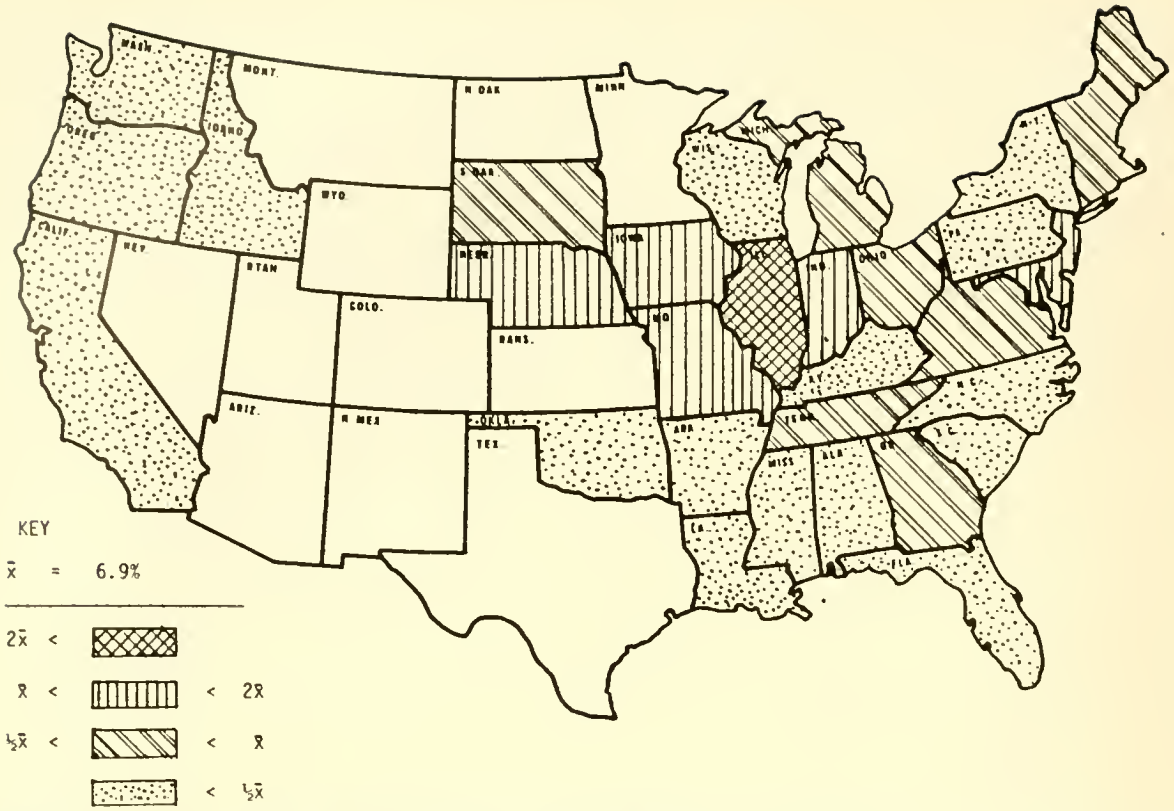

FIGURE 4. *Percent occurrence of heptachlor epoxide residue detections in cropland soil, by state, 1971, National Soils Monitoring Program, U.S. Environmental Protection Agency*

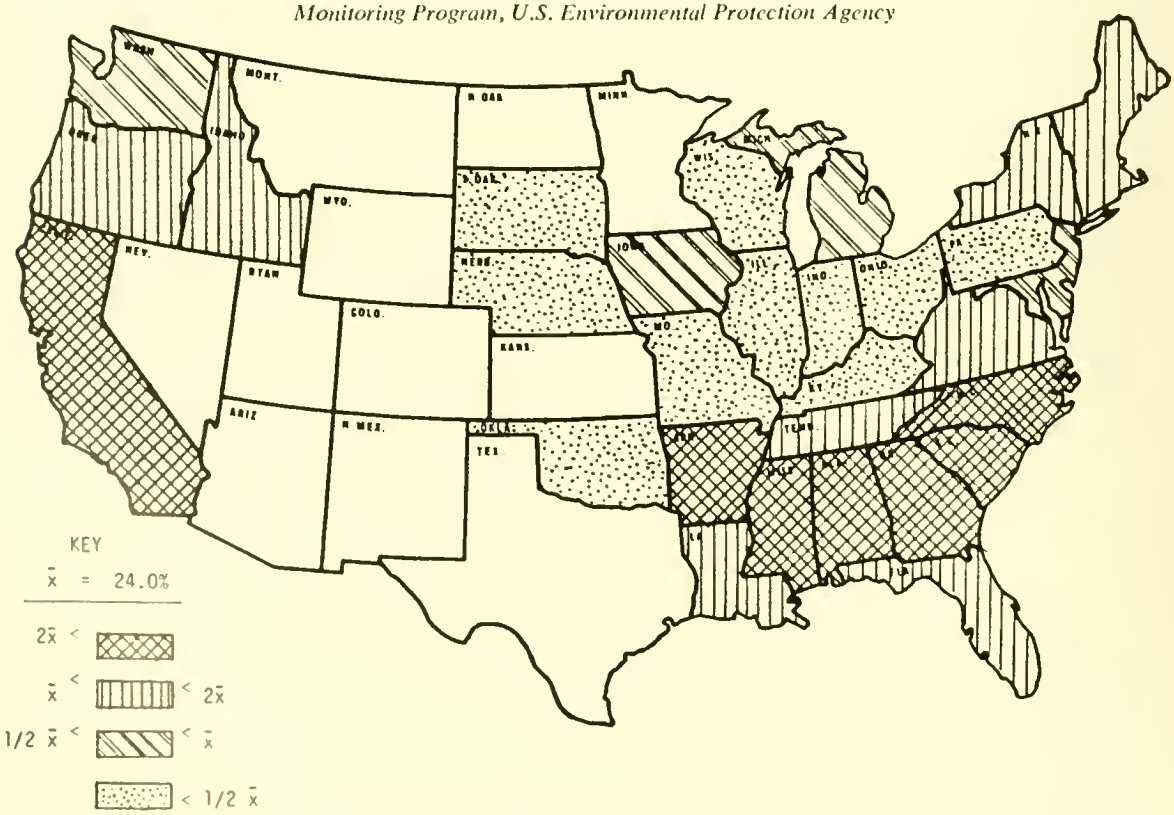

FIGURE 5. *Percent occurrence of ΣDDT residue detections in cropland soil, by state 1971, National Soils Monitoring Program, U.S. Environmental Protection Agency*

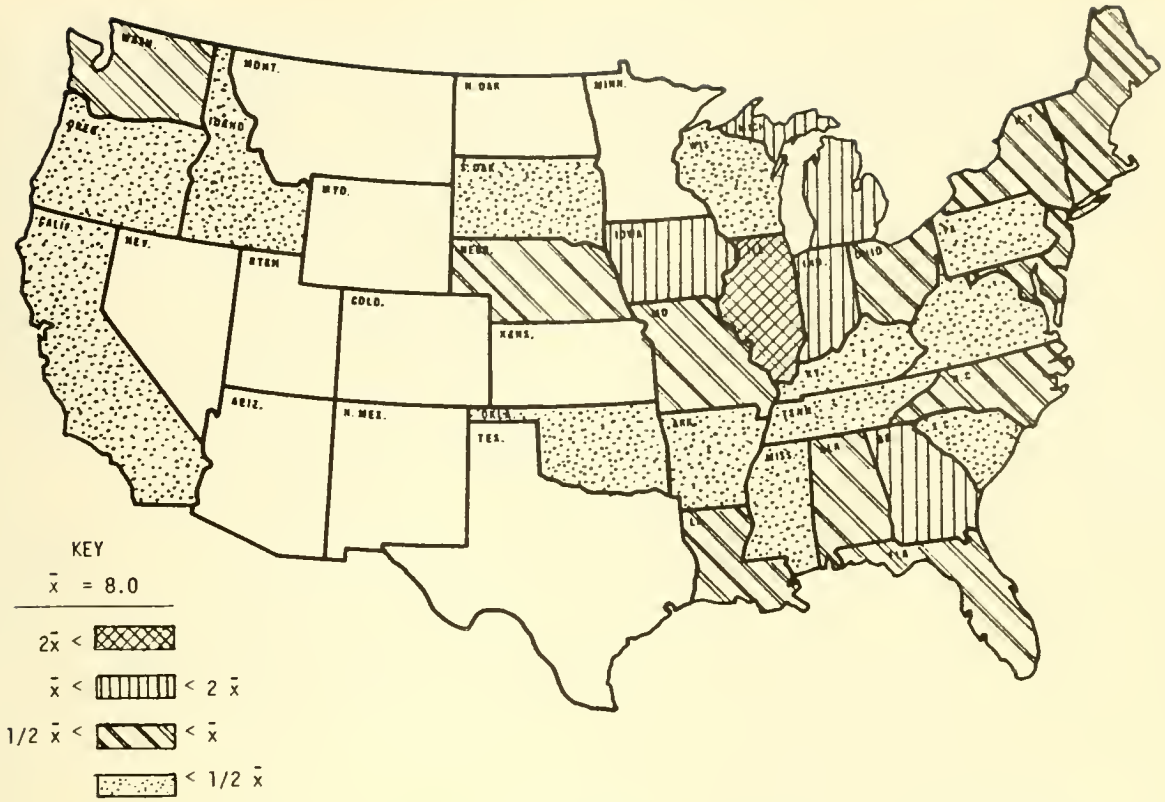

FIGURE 6. *Percent occurrence of chlordane residue detections in cropland soil, by state, 1971, National Soils Monitoring Program, U.S. Environmental Protection Agency*

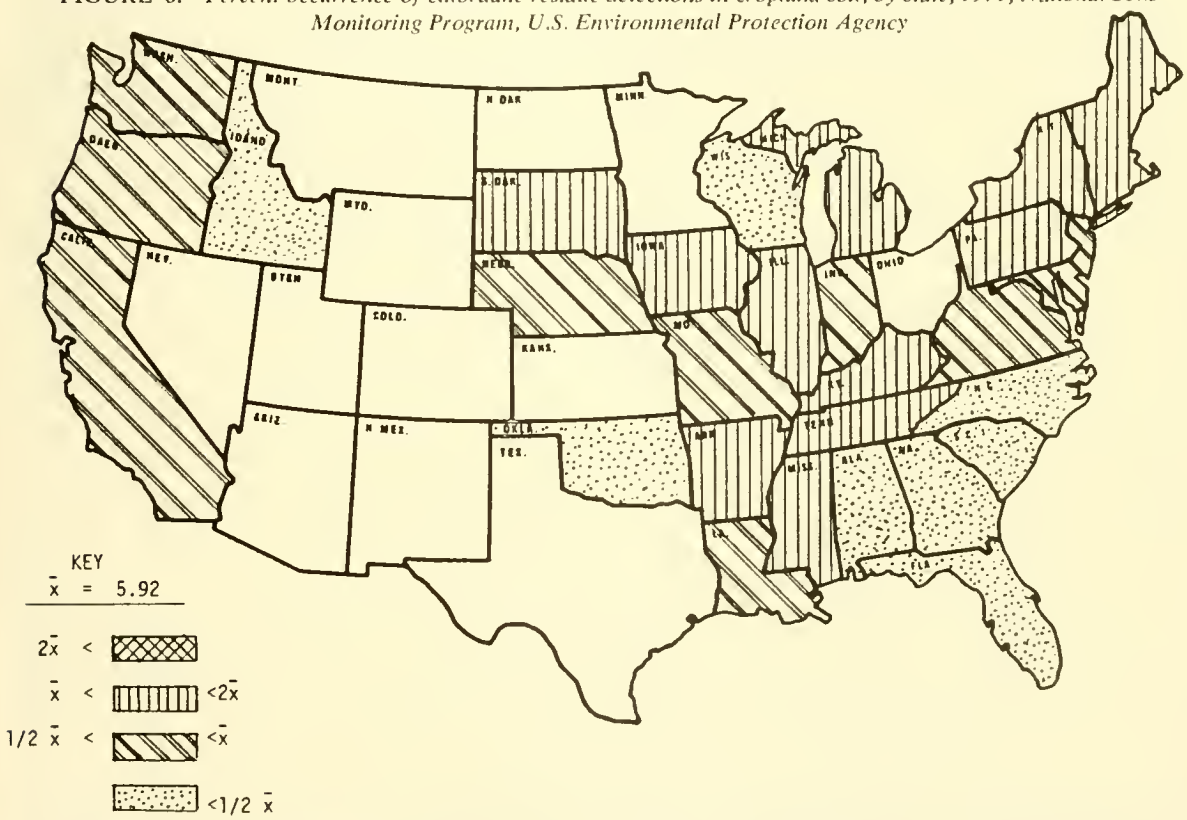

FIGURE 7. *Percent occurrence of elemental arsenic detections in cropland soil, by state, 1971, National Soils Monitoring Program, U.S. Environmental Protection Agency*

Illinois showed the highest percent occurrence of aldrin, dieldrin, chlordane, and heptachlor epoxide (Fig. 2–4, 6). The compounds are soil insecticides or their degradation products used in corn production. ΣDDT residues were concentrated in the southeastern states and California (Fig. 5). Generally, Oklahoma, Oregon, Pennsylvania, and Wisconsin had pesticide levels below the all-sites average detection frequency.

COMPOUND CONCENTRATIONS IN CROPS

Crop samples were collected from 729 sites, or 48 percent of the scheduled 1,533 sites. Samples were collected only from those sites where crops were mature and/or ready for harvest. All crop samples were analyzed for organochlorines. In addition, samples were analyzed for organophosphates and atrazine when pesticide application records indicated their use during the current growing season. Thus, the organophosphate and atrazine concentration data could result in higher occurrence frequencies than might occur if all samples had been analyzed.

Table 6 gives the occurrence of pesticide residues in the crop materials sampled. For all crops, 42 percent of the samples analyzed contained detectable concentrations of organochlorines, 13 percent contained detectable concentrations of organophosphates, and only 1 percent contained detectable concentrations of atrazine. In general, crops with known patterns of heavy pesticide application, or animal feed crops (alfalfa, hay, field corn, soybeans) grown in rotation with these crops, had the highest frequencies of detectable pesticides.

Table 7 presents the compound concentrations detected in each crop sampled. ΣDDT occurred most frequently in all crops analyzed, with the exception of cornstalks, in which dieldrin residues predominated. The high frequency of occurrence of ΣDDT is probably the result of prior, widespread use of DDT.

Acknowledgments

It is not possible to list by name all persons who contributed to this study. The authors are especially grateful to the staff of the Pesticides Monitoring Laboratory, Bay St. Louis, Mississippi, who received, processed and analyzed the samples for compound residues, and to the inspectors of the Animal and Plant Health Inspection Service, USDA, who collected the samples.

TABLE 6. *Occurrence of pesticide residues in standing agricultural crops, 1971—National Soils Monitoring Program*

Crop	Organochlorines			Organophosphates			Triazines		
	No. of Analyses	No. of Positive Detections	% of Positive Detections	No. of Analyses	No. of Positive Detections	% of Positive Detections	No. of Analyses	No. of Positive Detections	% of Positive Detections
Alfalfa bur clover	61	33	54	17	2	12	—	—	—
Beans, dry	5	0	0	4	0	0	—	—	—
Clover	4	2	50	1	0	0	—	—	—
Corn, field (kernels)	304	40	13	46	1	2	1	1	100
Cornstalks	286	164	57	125	1	1	73	0	0
Cotton	28	15	54	26	8	31	—	—	—
Cottonseed	19	12	63	18	5	28	—	—	—
Cotton stalks	44	40	91	35	27	77	—	—	—
Cowpeas	1	0	0	—	—	—	—	—	—
Grass hay	11	6	55	3	0	0	—	—	—
Milo	2	1	50	—	—	—	—	—	—
Mint	1	1	100	—	—	—	—	—	—
Mixed hay	51	26	51	17	1	6	—	—	—
Oats	1	0	0	—	—	—	—	—	—
Oats, straw	4	4	100	2	0	0	—	—	—
Pasture	18	10	56	3	0	0	—	—	—
Peanuts	8	2	25	1	0	0	—	—	—
Pecans	1	0	0	—	—	—	—	—	—
Rice	2	2	100	—	—	—	—	—	—
Rice straw	1	1	100	—	—	—	—	—	—
Sorghum (grain)	18	6	33	3	0	0	2	0	0
Sorghum stalks	23	14	61	4	0	0	2	0	0
Soybeans	177	69	39	45	0	0	9	0	0
Soybean hay	8	8	100	—	—	—	—	—	—
Sweet sorghum	1	0	0	—	—	—	—	—	—
Timothy	1	0	0	—	—	—	—	—	—
Tobacco	2	2	100	—	—	—	—	—	—
Wheat	1	0	0	—	—	—	—	—	—
Wheat straw	1	0	0	—	—	—	—	—	—
TOTAL	1,084	458	42	350	45	13	87	1	1

TABLE 7. *Compound concentrations in standing agricultural crops, 1971—National Soils Monitoring Program*

Compound	No. of Positive Detections	% of Positive Detections	Residues, ppm Dry Weight			
			Arithmetic Mean	Estimated Geometric Mean[1]	Detected Values	
					Min.	Max.
ALFALFA/BUR CLOVER						
Organochlorines, 61 samples						
Chlordane	2	3.3	0.01	0.001	0.17	0.42
p,p'-DDE	20	32.8	0.01	0.005	0.01	0.09
o,p'-DDT	15	24.6	0.01	0.004	0.01	0.14
p,p'-DDT	27	44.3	0.04	0.014	0.01	0.66
p,p'-TDE	1	1.6	<0.01	—	0.01	—
ΣDDT	28	45.9	0.06	0.018	0.01	0.88
Dieldrin	11	18.0	<0.01	0.002	0.01	0.05
Toxaphene	1	1.6	0.01	—	0.38	—
Organophosphates, 17 samples						
Parathion, ethyl	2	11.8	2.32	0.013	3.20	36.20
Parathion, methyl	1	5.9	0.27	—	4.57	—
BEANS, DRY (All Varieties)						
Organochlorines, 5 samples: no residues detected						
Organophosphates, 4 samples; no residues detected						
CLOVER (*Trifolium* sp.)						
Organochlorines, 4 samples						
p,p'-DDT	1	25.0	<0.01	—	0.02	—
ΣDDT	1	25.0	<0.01	—	0.02	—
Dieldrin	1	25.0	<0.01	—	0.01	—
Organophosphates, 1 sample: no residues detected						
FIELD CORN (Kernels)						
Organochlorines, 304 samples						
Chlordane	3	1.0	<0.01	<0.001	0.08	0.48
p,p'-DDE	2	0.7	<0.01	<0.001	0.01	0.03
o,p'-DDT	1	0.3	<0.01	—	0.05	—
p,p'-DDT	2	0.7	<0.01	<0.001	0.01	0.26
ΣDDT	3	1.0	<0.01	<0.001	0.01	0.34
Dieldrin	38	12.5	<0.01	0.001	0.01	0.07
Heptachlor	1	0.3	<0.01	—	0.05	—
Heptachlor epoxide	1	0.3	<0.01	—	0.01	—
Organophosphates, 46 samples						
Parathion, methyl	1	2.2	<0.01	—	0.09	—
Triazines, 99 samples						
Atrazine	1	1.0	<0.01	—	0.01	—
CORNSTALKS						
Organochlorines, 286 samples						
Chlordane	16	5.6	0.02	0.002	0.05	1.26
p,p'-DDE	37	12.9	<0.01	0.001	0.01	0.06
o,p'-DDT	49	17.1	<0.01	0.002	0.01	0.16
p,p'-DDT	105	36.7	0.02	0.006	0.01	0.55
p,p'-TDE	17	5.9	<0.01	0.001	0.01	0.10
ΣDDT	107	37.1	0.03	0.008	0.01	0.78
Dieldrin	114	39.9	0.01	0.006	0.01	0.17
Endrin	1	0.4	<0.01	—	0.06	—
Heptachlor	3	1.1	<0.01	<0.001	0.01	0.03
Heptachlor epoxide	22	7.7	<0.01	0.001	0.01	0.51
Toxaphene	15	5.3	0.04	0.002	0.07	2.83
Organophosphates, 125 samples						
Parathion, ethyl	1	0.8	<0.01	—	0.36	—
Triazines, 73 samples: no residues detected						
COTTON						
Organochlorines, 28 samples						
p,p'-DDE	7	25.0	0.07	0.006	0.01	1.86
o,p'-DDT	2	7.1	0.25	0.004	0.21	6.87
p,p'-DDT	15	53.6	0.95	0.039	0.01	22.99
ΣDDT	15	53.6	1.27	0.043	0.03	31.72
Dieldrin	1	3.6	<0.01	—	0.02	—
Endrin	1	3.6	<0.01	—	0.09	—
Endrin ketone	1	3.6	<0.01	—	0.06	—
Toxaphene	6	21.4	1.22	0.019	0.18	28.89
Organophosphates, 26 samples						
DEF	6	23.1	0.08	0.012	0.08	0.62
Parathion, ethyl	2	7.7	0.04	0.004	0.49	0.53
Parathion, methyl	1	3.8	0.01	—	0.18	—
COTTONSEED						
Organochlorines, 19 samples						
p,p'-DDE	7	36.8	0.06	0.010	0.01	0.82
o,p'-DDT	6	31.6	0.28	0.019	0.02	3.32

(*Continued next page*)

TABLE 7 (cont'd.). *Compound concentrations in standing agricultural crops, 1971—National Soils Monitoring Program*

Compound	No. of Positive Positive	% of Positive Detections	Residues, ppm Dry Weight			
			Arithmetic Mean	Estimated Geometric Mean[1]	Detected Values Min.	Max.
p,p'-DDT	9	47.4	0.87	0.040	0.03	14.09
ΣDDT	9	47.4	1.21	0.053	0.04	18.23
Toxaphene	5	26.3	1.12	0.031	0.55	13.54
Organophosphates, 18 samples						
DEF	5	27.8	0.07	0.013	0.10	0.63
COTTON STALKS						
Organochlorines, 44 samples						
Chlordane	1	2.3	0.01	—	0.40	—
o,p'-DDE	1	2.3	<0.01	—	0.10	—
p,p'-DDE	34	77.3	0.30	0.062	0.01	4.06
o,p'-DDT	34	77.3	1.48	0.153	0.01	28.10
p,p'-DDT	40	90.9	7.67	0.691	0.02	114.63
p,p'-TDE	17	38.6	0.83	0.032	0.01	17.78
ΣDDT	40	90.9	9.15	0.916	0.04	160.51
Dieldrin	4	8.9	<0.01	0.001	0.01	0.08
Endrin	1	2.3	0.14	—	6.26	—
Endrin ketone	1	2.3	0.01	—	0.37	—
Heptachlor epoxide	1	2.3	<0.01	—	0.01	—
Toxaphene	31	70.5	10.21	0.628	0.15	150.00
Organophosphates, 35 samples						
DEF	17	48.6	2.01	0.085	0.11	37.13
Parathion, ethyl	5	14.3	0.23	0.006	0.04	7.32
Parathion, methyl	21	60.0	0.30	0.068	0.04	1.53
COWPEAS						
Organochlorines, 1 sample: no residues detected						
GRASS HAY						
Organochlorines, 11 samples						
Chlordane	1	9.1	0.01	—	0.19	—
p,p'-DDE	5	45.4	0.02	0.007	0.01	0.12
o,p'-DDT	4	36.4	0.03	0.008	0.01	0.32
p,p'-DDT	5	45.4	0.08	0.015	0.02	0.73
ΣDDT	5	45.4	0.13	0.021	0.03	1.17
Dieldrin	2	18.2	<0.01	0.002	0.01	0.02
Toxaphene	2	18.2	0.21	0.012	0.26	2.00
Organophosphates, 3 samples: no residues detected						
MILO						
Organochlorines, 2 samples						
Dieldrin	1	50.0	0.05	—	0.11	—
MINT						
Organochlorines, 1 sample						
p,p'-DDE	1	100.0	0.05	—	0.05	—
o,p'-DDT	1	100.0	0.01	—	0.01	—
p,p'-DDT	1	100.0	0.15	—	0.15	—
ΣDDT	1	100.0	0.21	—	0.21	—
MIXED HAY						
Organochlorines, 51 samples						
Chlordane	4	7.8	0.07	0.004	0.25	1.68
p,p'-DDE	12	23.5	0.01	0.003	0.01	0.48
o,p'-DDT	8	15.7	0.03	0.003	0.01	1.23
p,p'-DDT	17	33.3	0.26	0.011	0.01	12.24
p,p'-TDE	1	2.0	<0.01	—	0.01	—
ΣDDT	17	33.3	0.31	0.012	0.02	13.95
Dieldrin	15	29.4	0.01	0.004	0.01	0.05
Toxaphene	6	11.8	0.36	0.007	0.16	15.73
Organophosphates, 17 samples						
DEF	1	5.9	<0.01	—	0.06	—
Parathion, methyl	1	5.9	<0.01	—	0.02	—
OATS						
Organochlorines, 1 sample: no residues detected						
OAT HAY/STRAW						
Organochlorines, 4 samples						
Chlordane	1	25.0	0.01	—	0.03	—
p,p'-DDE	2	50.0	<0.01	0.004	0.01	—
o,p'-DDT	2	50.0	<0.01	0.004	0.01	—
p,p'-DDT	3	75.0	0.02	0.019	0.02	0.04
ΣDDT	3	75.0	0.03	0.026	0.04	0.06
Dieldrin	2	50.0	0.01	0.009	0.01	0.05
Organophosphates 2 samples: no residues detected						

(*Continued next page*)

TABLE 7 (cont'd.). *Compound concentrations in standing agricultural crops, 1971—National Soils Monitoring Program*

Compound	No. of Positive Detections	% of Positive Detections	Residues, ppm Dry Weight			
			Arithmetic Mean	Estimated Geometric Mean[1]	Detected Values Min.	Max.
PASTURE						
Organochlorines, 18 samples						
Chlordane	3	16.7	0.05	0.009	0.37	0.63
p,p'-DDE	3	16.7	<0.01	0.001	0.01	0.02
o,p'-DDT	2	11.1	<0.01	0.001	0.02	0.03
p,p'-DDT	6	33.3	0.01	0.005	0.01	0.08
o,p'-TDE	1	5.6	<0.01	—	0.45	—
p,p'-TDE	1	5.6	<0.01	—	0.07	—
ΣDDT	6	33.3	0.04	0.008	0.01	0.63
Dieldrin	6	33.3	<0.01	0.006	0.02	0.05
Endrin	1	5.6	<0.01	—	0.01	—
Heptachlor epoxide	1	5.6	<0.01	—	0.01	—
Toxaphene	1	5.6	0.01	—	0.23	—
Organophosphates, 3 samples: no residues detected						
PEANUTS						
Organochlorines, 8 samples						
Dieldrin	2	25.0	0.01	0.004	0.02	0.03
Organophosphates, 1 sample: no residues detected						
PECANS						
Organochlorines, 1 sample: no residues detected						
RICE						
Organochlorines, 2 samples						
p,p'-DDE	2	100.0	0.02	0.018	0.01	0.03
o,p'-DDT	1	50.0	0.03	—	0.06	—
p,p'-DDT	2	100.0	0.15	0.096	0.03	0.27
ΣDDT	2	100.0	0.20	0.126	0.04	0.36
Heptachlor	1	50.0	<0.01	—	0.01	—
RICE STRAW						
Organochlorines, 1 sample						
p,p'-DDE	1	100.0	0.04	—	0.04	—
o,p'-DDT	1	100.0	0.11	—	0.11	—
p,p'-DDT	1	100.0	0.12	—	0.12	—
ΣDDT	1	100.0	0.27	—	0.27	—
Toxaphene	1	100.0	0.52	—	0.52	—
SORGHUM						
Organochlorines, 18 samples						
Chlordane	3	16.7	0.03	0.005	0.02	0.42
p,p'-DDE	2	11.1	0.01	0.003	0.07	0.14
o,p'-DDT	1	5.6	0.02	—	0.30	—
p,p'-DDT	3	16.7	0.06	0.007	0.02	0.64
p,p'-TDE	1	5.6	<0.01	—	0.05	—
ΣDDT	3	16.7	0.04	—	—	—
Dieldrin	4	22.2	0.02	0.004	0.01	0.28
Endrin	1	5.6	<0.01	—	0.02	—
Heptachlor epoxide	1	5.6	<0.01	—	0.02	—
Toxaphene	1	5.6	0.05	—	0.84	—
Organophosphates, 3 samples: no residues detected						
Triazines, 2 samples: no residues detected						
SORGHUM STALKS						
Organochlorines, 23 samples						
Chlordane	4	17.4	0.08	0.009	0.11	0.81
p,p'-DDE	6	26.1	0.01	0.005	0.01	0.12
o,p'-DDT	10	43.5	0.03	0.008	0.01	0.33
p,p'-DDT	15	65.2	0.09	0.023	0.01	1.07
p,p'-TDE	4	17.4	<0.01	0.002	0.02	0.03
ΣDDT	16	69.6	0.13	0.031	0.01	1.52
Dieldrin	6	26.1	0.05	0.010	0.02	0.51
Endrin	1	4.3	0.03	—	0.60	—
Endrin ketone	1	4.3	0.01	—	0.19	—
Heptachlor	1	4.3	<0.01	—	0.02	—
Heptachlor epoxide	2	8.7	<0.01	0.001	0.03	0.04
Toxaphene	4	17.4	0.09	0.009	0.24	0.91
Organophosphates, 4 samples: no residues detected						
Triazines, 2 samples: no residues detected						
SWEET SORGHUM						
Organochlorines, 1 sample: no residues detected						

(*Continued next page*)

TABLE 7 (cont'd.). *Compound concentrations in standing agricultural crops, 1971—National Soils Monitoring Program*

Compound	No. of Positive Detections	% of Positive Detections	Residues, ppm Dry Weight			
			Arithmetic Mean	Estimated Geometric Mean[1]	Detected Values Min.	Max.
SOYBEANS						
Organochlorines, 177 samples						
p,p'-DDE	5	2.8	<0.01	<0.001	0.01	0.02
o,p'-DDT	1	0.6	<0.01	—	0.01	—
p,p'-DDT	4	2.3	<0.01	<0.001	0.01	0.05
ΣDDT	5	2.8	<0.01	<0.001	0.02	0.07
Dieldrin	55	31.1	<0.01	0.003	0.01	0.05
Endrin	7	3.9	<0.01	<0.001	0.01	0.03
Heptachlor epoxide	2	1.1	<0.01	<0.001	0.01	0.03
Toxaphene	3	1.7	0.01	0.001	0.10	0.66
Organophosphates, 45 samples: no residues detected						
Triazines, 9 samples: no residues detected						
SOYBEAN HAY						
Organochlorines, 8 samples						
Chlordane	1	12.5	0.02	—	0.17	—
p,p'-DDE	4	50.0	0.01	0.006	0.01	0.02
o,p'-DDT	2	25.0	<0.01	0.002	0.01	—
p,p'-DDT	6	75.0	0.02	0.017	0.01	0.05
p,p'-TDE	3	37.5	0.01	0.005	0.01	0.04
ΣDDT	6	75.0	0.04	0.027	0.01	0.09
Dieldrin	5	62.5	0.01	0.006	0.01	0.02
Endrin	1	12.5	<0.01	—	0.01	—
TIMOTHY						
Organochlorines, 1 sample: no residues detected						
TOBACCO						
Organochlorines, 2 samples						
o,p'-DDE	1	50.0	0.25	—	0.50	—
p,p'-DDE	1	50.0	0.39	—	0.78	—
o,p'-DDT	1	50.0	1.10	—	2.20	—
p,p'-DDT	2	100.0	3.87	0.384	0.01	7.74
o,p'-TDE	1	50.0	2.87	—	5.74	—
p,p'-TDE	1	50.0	8.54	—	17.09	—
ΣDDT	2	100.0	17.03	0.815	0.01	34.05
Dieldrin	1	50.0	0.34	—	0.69	—
Endosulfan	1	50.0	0.66	—	1.33	—
Endosulfan II	1	50.0	2.63	—	5.26	—
Endosulfan sulfate	1	50.0	3.28	—	6.57	—
WHEAT						
Organochlorines, 1 sample: no residues detected						
WHEAT STRAW						
Organochlorines, 1 sample: no residues detected						

[1]Not calculated when fewer than two positive detections were present.

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Pesticide Application and Cropping Data from 37 States, 1971— National Soils Monitoring Program

Ann E. Carey,[1] Jeanne A. Gowen,[2] and G. Bruce Wiersma [3]

ABSTRACT

This report summarizes pesticide application and cropping data collected in 1971 from 1,473 agricultural sampling sites in 37 states as part of the National Soils Monitoring Program. Pesticide application data are summarized by all sites, state, and crop. Tables generally give the number of reporting sites, the number of times a compound was applied, the percent occurrence, and the arithmetic mean total application rate.

Pesticides applied most frequently to sampling sites were atrazine, 2,4-D, captan, and malathion. Pesticides were most frequently applied to field corn and cotton, least frequently to alfalfa/bur clover and mixed hay.

Introduction

In 1963, the report of the President's Science Advisory Committee recommended that appropriate federal agencies "develop a continuing network to monitor residue levels in air, water, soil, man, wildlife and fish" (*1*). As a result of this recommendation, the National Pesticide Monitoring Program (NPMP) was established to determine levels and trends of pesticides and their degradation products in the environment (*3*). Federal responsibility for monitoring pesticides was officially mandated in Section 20 of the amended Federal Insecticide, Fungicide and Rodenticide Act of 1972 (PL 92–516).

The National Soils Monitoring Program is an integral part of the NPMP, monitoring agricultural soils and raw agricultural crops. It was initiated in 1968 by the U.S. Department of Agriculture and is administered by the U.S. Environmental Protection Agency. The present report summarizes pesticide application and cropping data collected in 1971 from 1,473 sampling sites in 37 states. Composite soil and crop samples were also collected from these sites for pesticide residue analyses, the results of which are published separately (*2*).

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Sampling

The site selection criteria and statistical design of the National Soils Monitoring Program have been described (*4*). In 1971, 1,533 sites in 37 states were scheduled for sampling (Fig. 1). At each 4-hectare (10-acre) site, landowners or operators supplied information on the crops grown and the kinds and amounts of pesticides applied during 1971.

Results and Discussion

COMPOUNDS APPLIED TO CROPLAND

Pesticide use data were received from 1,473 or 96 percent of the scheduled 1,533 sites. Of these, 784 or 53 percent of the sites had one or more pesticides applied during 1972. Tables summarizing the application data show the number and percent of sites with reported pesticide application and the average rate of total pesticide application for each site, expressed both in pounds per acre and kilograms per hectare.

Table 1 gives the frequency of pesticide use on sample sites in various states and state groups. Because some of the smaller eastern states had very few sites, those with similar geographic location and/or agricultural characteristics were combined to obtain more representative data. States were grouped as follows: Mid-Atlantic: Delaware, Maryland, New Jersey; New England: Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, Vermont; and Virginia and West Virginia. Among the individual states and state groups, frequency of pesticide use ranged from 23 percent in Pennsylvania to 77 percent in Mississippi.

ALL SITES

Applications of 132 compounds were recorded for all reporting sites. The compounds included 50 herbicides, including defoliants, 48 insecticides and/or acaricides, 28 fungicides, 4 nematocides, 1 soil fumigant, and 1 growth retardant (Table 2). The most frequently applied compounds were atrazine, 2,4-D, captan, and malathion, which were reported from 14, 10, 9, and 8 percent of the reporting sites, respectively.

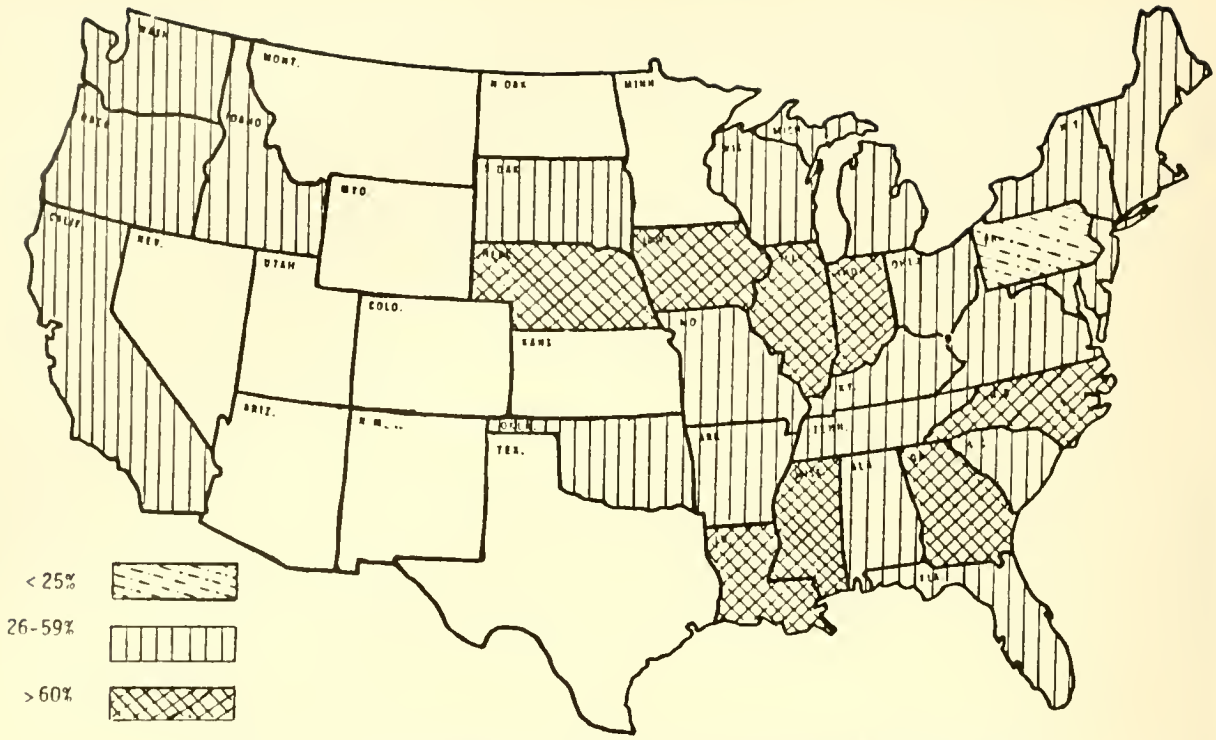

FIGURE 1. *States scheduled for sampling, 1971, National Soils Monitoring Program*

TABLE 1. *Occurrence of pesticide applications by state, 1971—National Soils Monitoring Program*

State or State Group[1]	Total No. of Sites Reporting	Pesticides Applied		Pesticides Not Applied	
		No. of Sites	%	No. of Sites	%
Alabama	22	9	41	13	59
Arkansas	45	24	53	21	47
California	61	29	48	32	52
Florida	18	8	44	10	56
Georgia	29	19	66	10	34
Idaho	33	11	33	22	67
Illinois	142	100	70	42	30
Indiana	74	50	68	24	32
Iowa	152	103	68	49	32
Kentucky	28	11	39	17	61
Louisiana	25	17	68	8	32
Michigan	54	25	46	29	54
Mid-Atlantic	16	7	44	9	56
Mississippi	31	24	77	7	23
Missouri	79	37	46	42	54
Nebraska	106	65	61	41	39
New England	21	6	29	15	71
New York	36	17	47	19	53
N. Carolina	30	18	60	12	40
Ohio	57	31	54	26	46
Oklahoma	60	22	37	38	63
Oregon	37	13	35	24	65
Pennsylvania	35	8	23	27	77
S. Carolina	15	8	53	7	47
S. Dakota	106	51	48	55	52
Tennessee	24	11	46	13	54
Virginia and W. Virginia	25	8	32	17	68
Washington state	45	22	49	23	51
Wisconsin	67	30	45	37	55
Total	1473	784	53	689	47

[1] Some smaller eastern states which had few sites but similar geographic locations and/or agricultural characteristics were combined to obtain more representative data, including: Mid-Atlantic states: Delaware, Maryland, New Jersey; New England states: Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, Vermont; and Virginia and West Virginia.

BY STATE

Table 3 presents the application data by state or state grouping. Because of the number of states sampled, it is not feasible to discuss in detail the pesticide data from each state. However, the pesticide application information from each state reflects both the crops grown and the intensity of agricultural land use in the state.

In Figure 2, the frequency of reported pesticide applications in each state is designated as follows: low, states where less than 25 percent of the sites reported pesticide applications; medium, states where 25–59 percent of the sites reported applications; and, high, where over 60 percent of the sites in a state reported pesticide applications.

BY CROP

Table 4 lists crops grown on sample sites in 1972 as well as the pesticide application status for each crop. Application data for selected major crops are presented in Table 5. Pesticide use varied widely among these crops.

Table 6 shows the pesticide applications in 1971 for selected major crops, by state.

Acknowledgment

It is not possible to list by name all the persons who contributed to this study. However, the authors are especially grateful to the inspectors from the Plant Protection and Quarantine Programs, Animal and Plant Health Inspection Service, U.S. Department of Agriculture, who collected the data.

TABLE 2. *Compounds applied to 1,473 cropland sites, 1971—National Soils Monitoring Program*

Compound	Trade Name, If Noted	Pesticides Applied		Average Total Application	
		No. of Sites	% of Sites	lb./Acre	kg/Hectare
Alachlor	Lasso	65	4.4	1.58	1.77
Aldrin		45	3.0	1.15	1.29
Arsenic pentoxide		2	0.1	0.50	0.56
Atrazine	AAtrex	214	14.1	1.78	1.99
Azinphosmethyl	Guthion	6	0.4	0.60	0.67
Bacillus thuringiensis	B.T.	1	0.1	0.11	0.12
Barban	Carbyne	1	0.1	0.25	0.28
Benefin	Balan	3	0.2	1.11	1.24
BHC		3	0.2	0.02	0.02
Bordeaux mixture		1	0.1	1.25	1.40
Bromacil	Hyvar	3	0.2	0.62	0.70
Butylate	Sutan	18	1.2	1.74	1.95
Bux		17	1.1	1.26	1.41
Captafol	Difolatan	1	0.1	1.50	1.68
Captan		138	9.3	0.11	0.12
Carbaryl	Sevin	18	1.2	2.12	2.37
Carbophenothion	Trithion	1	0.1	3.20	3.58
Carbofuran	Furadan	20	1.3	1.01	1.13
Chevron RE-5353		4	0.3	0.85	0.95
Chloramben	Amiben	41	2.8	1.39	1.56
Chlordane		1	0.1	2.50	2.80
Chlorobenzilate	Acaraben	2	0.1	1.38	1.55
Chloroneb	Demosan	9	0.6	0.02	0.02
Chloropropham	Chloro-IPC	1	0.1	2.50	2.80
Chlorothalonil	Bravo	1	0.1	3.00	3.36
Copper carbonate (basic)		1	0.1	3.90	4.37
Copper hydroxide		1	0.1	1.08	1.21
Copper oxide		1	0.1	1.70	1.90
Copper sulfate		3	0.2	13.97	15.65
Cypromid	Clobber	1	0.1	0.75	0.84
2,4-D	Decamine	145	9.8	0.87	0.97
Dalapon	Dowpon	4	0.3	2.60	2.91
2,4-DB	Butyrac	7	0.5	0.64	0.72
DCPA	Dacthal	1	0.1	3.75	4.20
DDT		33	2.2	3.83	4.29
DEF		9	0.6	1.03	1.16
Demeton	Systox	2	0.1	1.56	1.75
Diallate	Avadex	1	0.1	0.12	0.13
Diazinon		15	1.0	0.75	0.84
Dicamba	Banvel D	12	0.8	0.31	0.34
Dichlofenthion	Nemacide	1	0.1	6.00	6.72
Dichloropropene	Telone	3	0.2	28.00	31.36
Dichlorprop	2,4-DP	1	0.1	3.00	3.36
Dicofol	Kelthane	1	0.1	1.00	1.12
Dieldrin		6	0.4	0.09	0.10
Dinitrocresol		3	0.2	1.64	1.84
Diphenamid	Enide	1	0.1	0.25	0.28
Disulfoton	Di-Syston	24	1.6	1.21	1.35
Diuron	Karmex	9	0.6	1.56	1.75
DNBP	Premerge	16	1.1	1.35	1.51
Dodine		2	0.1	0.83	0.93
DSMA		11	0.7	2.00	2.24
Dyfonate		1	0.1	0.90	1.00
EMTS	Ceresan M	9	0.6	0.06	0.06
Endosulfan	Thiodan	8	0.5	1.44	1.61
Endrin		3	0.2	2.20	2.46
EPTC	Eptam	10	0.7	2.09	2.34
Ethoprop	Mocap	1	0.1	1.00	1.12
Ethylmercury chloride	Ceresan Red	8	0.5	0.03	0.03
Fensulfothion	Dasanit	5	0.3	1.15	1.28
Fentin hydroxide		1	0.1	2.25	2.52
Ferbam		2	0.1	2.59	2.90
Fluometuron	Cotoran	22	1.5	0.95	1.06
Folex		5	0.3	1.05	1.18
Folpet	Phaltan	1	0.1	1.00	1.12
Furethrin		1	0.1	8.00	8.97
Heptachlor		8	0.5	1.27	1.42
Hexachloro-benzene	HCB	7	0.5	0.01	0.01
Isodrin		1	0.1	0.01	0.01
Lead arsenate		3	0.2	7.07	7.91
Lindane		1	0.1	0.02	0.02
Linuron	Lorox	23	1.6	0.89	1.00
Londax		1	0.1	0.50	0.56
Malathion		111	7.5	0.16	0.18
Maleic hydrazide	MH	3	0.2	3.00	3.36
Mancozeb	Dithane M-45	2	0.1	12.40	13.89
Maneb		3	0.2	2.34	2.62
MCPA	MCP	5	0.3	0.70	0.78
Mercury		2	0.1	0.06	0.06
Metham	Vapam	1	0.1	2.16	2.42
Methomyl	Lannate	1	0.1	1.13	1.27
Methoxychlor		24	1.6	0.17	0.19
Methylmercury acetate	Ceresan L	6	0.4	0.01	0.01
Methylmercury dicyandiamide	Panogen	18	1.2	0.08	0.09
Methyl trithion		1	0.1	3.00	3.36
Mevinphos	Phosdrin	2	0.1	0.75	0.84
Mirex		6	0.4	0.07	0.08
Monocrotophos	Azodrin	4	0.3	0.33	0.36
Monuron	Telvar	2	0.1	1.30	1.46
MSMA		17	1.1	1.77	1.99
Nabam		1	0.1	5.00	5.60
Naptalam	Alanap	4	0.3	1.94	2.17
Nitralin	Planavin	5	0.3	1.05	1.18
Oil Spray		2	0.1	60.00	67.20
Ovex		1	0.1	0.50	0.56
Oxydemeton-methyl	Metasystox-R	2	0.1	0.50	0.56
Paraquat		4	0.3	0.86	0.97
Parathion, ethyl		21	1.4	3.32	3.72
Parathion, methyl		48	3.2	2.81	3.15
PCNB		2	0.1	3.51	3.93
Pebulate	Tillam	1	0.1	0.12	0.13
Pentachloro-phenol	PCP	2	0.1	3.02	3.38
Phenylmercury actate	PMA	2	0.1	0.01	0.01
Phenylmercury urea		3	0.2	0.01	0.01
Phorate	Thimet	21	1.4	1.71	1.91
Phosalone	Zolone	1	0.1	4.00	4.48
Phosphamidon	Dimecron	2	0.1	0.27	0.30
Prolate	Imidan	2	0.1	11.60	13.00
Prometryn	Caparol	2	0.1	1.08	1.21
Propachlor	Ramrod	44	3.0	1.64	1.83
Propanil	Stam	6	0.4	3.46	3.87
Propargite	Omite	2	0.1	2.58	2.88
Pyrazon	Pyramin	1	0.1	1.25	1.40
Silvex		3	0.2	0.42	0.47
Simazine	Princep	9	0.6	4.00	4.48
Sodium chlorate		5	0.3	1.74	1.95
Solan		1	0.1	1.00	1.12
Sulfur		12	0.8	34.27	38.38
2,4,5-T		2	0.1	0.30	0.34
TCA		2	0.1	2.50	2.80
TEPP		1	0.1	4.00	4.48
Terbacil	Sinbar	1	0.1	1.40	1.56
Terbutryn	Igran	1	0.1	1.75	1.96
Terrazole		1	0.1	0.15	0.16
Tetradifon	Tedion V-18	1	0.1	0.75	0.84
Thiram		13	0.9	0.01	0.01
Toxaphene		33	2.2	7.00	7.84
Trichlorfon	Dylox	2	0.1	0.88	0.98
Trietazine		1	0.1	0.70	0.78
Trifluralin	Treflan	64	4.3	0.95	1.06
Vernolate	Vernam	2	0.1	2.25	2.52
Zineb		1	0.1	7.50	8.40

TABLE 3. *Compounds applied to cropland sites by state, 1971—National Soils Monitoring Program*

Compound	Trade Name, If Noted	Pesticides Applied: No. of Sites	Pesticides Applied: % of Sites	Average Total Application: lb./Acre	Average Total Application: kg/Hectare
ALABAMA, 22 SITES					
Atrazine	AAtrex	3	13.6	2.67	2.98
Benefin	Balan	1	4.5	0.75	0.84
Butylate	Sutan	2	9.0	0.25	0.28
DDT		3	13.6	3.67	4.10
Disulfoton	Di-Syston	1	4.5	7.00	7.84
Diuron	Karmex	1	4.5	0.34	0.38
DSMA		1	4.5	1.50	1.68
EMTS	Ceresan M	1	4.5	0.01	0.01
Endrin		1	4.5	1.40	1.56
Fluometuron	Cotoran	3	13.6	0.92	1.02
MSMA		1	4.5	2.00	2.24
Parathion, ethyl		1	4.5	1.40	1.56
Parathion, methyl		3	13.6	2.23	2.50
PCNB		1	4.5	7.00	7.84
Toxaphene		3	13.6	4.55	5.09
Trifluralin	Treflan	1	4.5	0.50	0.56
ARKANSAS, 45 SITES					
Aldrin		1	2.2	0.75	0.84
Atrazine	AAtrex	1	2.2	0.75	0.84
Captan		1	2.2	0.01	0.01
Chloroneb	Demosan	2	4.4	0.01	0.01
2,4-D		1	2.2	0.50	0.56
2,4-DB		1	2.2	1.50	1.68
DDT		3	6.6	0.57	0.63
Disulfoton	Di-Syston	1	2.2	0.25	0.28
DNBP	Premerge	5	11.1	0.94	1.05
DSMA		2	4.4	1.20	1.34
EMTS	Ceresan M	3	6.6	0.15	0.16
Fluometuron	Cotoran	5	11.1	0.84	0.92
Linuron	Lorox	2	4.4	0.50	0.56
Methylmercury dicyandiamide	Panogen	3	6.6	0.25	0.28
Monuron	Telvar	1	2.2	1.00	1.12
MSMA		5	11.1	1.20	1.34
Nitralin	Planavin	3	6.6	1.17	1.30
Parathion, ethyl		1	2.2	7.00	7.84
Parathion, methyl		5	11.1	2.00	2.24
Propanil	Stam	2	4.4	5.50	6.16
Solan		1	2.2	1.00	1.12
Thiram		1	2.2	0.01	0.01
Toxaphene		3	6.6	1.00	1.12
Trifluralin	Treflan	9	20.0	1.11	1.24
CALIFORNIA, 61 SITES					
Aldrin		1	1.6	0.01	0.01
Azodrin		1	1.6	0.50	0.56
Bacillus thuringiensis		1	1.6	0.11	0.12
Captan		1	1.6	0.01	0.01
2,4-D		2	3.2	0.31	0.34
DCPA	Dacthal	1	1.6	3.75	4.20
Diazinon		2	3.2	0.08	0.08
Dicofol	Kelthane	1	1.6	1.00	1.12
Diphenamid	Enide	1	1.6	0.25	0.28
Diuron	Karmex	1	1.6	2.40	2.68
EPTC	Eptam	1	1.6	3.00	3.36
Ethylmercury chloride	Ceresan Red	1	1.6	0.01	0.01
Malathion		4	6.5	1.71	1.91
MCPA	MCP	2	3.2	1.25	1.40
Mercury		2	3.2	0.06	0.06
Methomyl	Lannate	1	1.6	1.13	1.26
Mevinphos	Phosdrin	1	1.6	1.00	1.12
Oil Spray		1	1.6	50.00	56.04
Ovex		1	1.6	0.50	0.56
Oxydemeton-methyl	Metasystox-R	1	1.6	0.50	0.56
Paraquat		2	3.2	0.22	0.25
Parathion, ethyl		4	6.5	2.08	2.32
Parathion, methyl		2	3.2	1.38	1.54
PCNB		1	1.6	0.01	0.01
Pebulate	Tillam	1	1.6	0.12	0.13
Phenylmercury acetate	PMA	1	1.6	0.01	0.01
Propargite	Omite	1	1.6	0.15	0.16
Simazine	Princep	3	4.9	8.00	8.96
Sodium chlorate		1	1.6	5.00	5.60
Sulfur		5	8.1	15.34	17.19
Tetradifon	Tedion V-18	1	1.6	0.75	0.84
Toxaphene		1	1.6	0.40	0.44
Trichlorfon	Dylox	2	3.2	0.88	0.98
Trifluralin	Treflan	2	3.2	0.20	0.22
FLORIDA, 18 SITES					
Atrazine	AAtrex	2	11.1	2.75	3.08
Carbaryl	Sevin	1	5.5	5.00	5.60
Carbofuran	Furadan	1	5.5	4.00	4.48
Chlorobenzilate	Acaraben	2	11.1	1.38	1.54
Copper carbonate (basic)		1	5.5	3.90	4.37
2,4-D		3	16.6	5.33	5.97
Dalapon	Dowpon	1	5.5	1.50	1.68
Endrin		1	5.5	1.00	1.12
EPTC	Eptam	1	5.5	0.20	0.22
Malathion		1	5.5	3.17	3.55
Maneb		1	5.5	0.08	0.08
Oil Spray		1	5.5	70.00	78.45
Sulfur		3	16.6	78.67	88.16
Zineb		1	5.5	7.50	8.40
GEORGIA, 30 SITES					
Benefin	Balan	1	3.3	1.50	1.68
Butylate	Sutan	1	3.3	0.75	0.84
Captan		5	16.6	0.02	0.02
Carbaryl	Sevin	2	6.6	2.56	2.86
Chlorothalonil	Bravo	1	3.3	3.00	3.36
Copper oxide		1	3.3	1.70	1.90
Copper sulfate		1	3.3	30.00	33.62
2,4-D		2	6.6	0.75	0.84
DDT		5	16.6	2.61	2.93
DNBP		2	6.6	1.50	1.68
Ethylmercury chloride	Ceresan Red	2	6.6	0.01	0.01
Folex		1	3.3	1.50	1.68
Malathion		2	6.6	0.01	0.01
Maleic hydrazide		1	3.3	3.00	3.36
Methoxychlor		2	6.6	0.02	0.02
Methyl trithion		1	3.3	3.00	3.36
Mirex		2	6.6	0.04	0.04
Parathion, ethyl		3	10.0	7.88	9.52
Parathion, methyl		5	16.6	3.45	3.86
Sulfur		2	6.6	25.00	28.02
Thiram		3	10.0	0.01	0.01
Toxaphene		4	13.3	4.00	4.48
Trifluralin	Treflan	5	16.6	0.39	0.44
Vernolate	Vernam	1	3.3	2.50	2.80
IDAHO, 33 SITES					
Captan		1	3.0	0.08	0.08
2,4-D		3	9.0	0.67	0.75
DDT		2	6.0	3.25	3.64
Diallate	Avadex	1	3.0	0.12	0.13
Ethylmercury chloride	Ceresan Red	2	6.0	0.10	0.11
Malathion		1	3.0	1.00	1.12
PCP		1	3.0	0.03	0.03
Trifluralin	Treflan	1	3.0	1.00	1.12
ILLINOIS, 142 SITES					
Alachlor	Lasso	15	10.5	1.93	2.16
Aldrin		13	9.1	1.15	1.29
Atrazine	AAtrex	22	15.4	1.74	1.95
Butylate	Sutan	3	2.1	1.47	1.64
Bux		2	1.4	1.40	1.56
Captan		59	41.5	0.01	0.01
Carbofuran	Furadan	2	1.4	0.33	0.36
Chloramben	Amiben	18	12.6	1.47	1.64
2,4-D		6	4.2	1.11	1.24
Demeton	Systox	1	0.7	0.12	0.13
Fensulfothion	Dasanit	1	0.7	0.90	1.00
Ferbam		1	0.7	2.00	2.24

(Continued next page)

TABLE 3 (cont'd.). *Compounds applied to cropland sites by state, 1971—National Soils Monitoring Program*

Compound	Trade Name, If Noted	Pesticides Applied		Average Total Application	
		No. of Sites	% of Sites	lb./Acre	kg/Hectare
Heptachlor		4	2.8	1.75	1.96
Linuron	Lorox	6	4.2	0.89	0.99
Malathion		52	36.6	0.01	0.01
Methoxychlor		15	10.5	0.01	0.01
MSMA		1	0.7	0.25	0.28
Paraquat		1	0.7	2.00	2.24
PCP		1	0.7	6.00	6.72
Phorate	Thimet	7	4.9	0.62	0.69
Propachlor	Ramrod	18	12.6	1.22	1.36
Simazine	Princep	1	0.7	2.00	2.24
Trifluralin	Treflan	8	5.6	1.20	1.34
2,4,5-T		1	0.7	0.25	0.28
INDIANA, 76 SITES					
Alachlor	Lasso	9	11.8	1.56	1.74
Aldrin		9	11.8	1.35	1.51
Atrazine	AAtrex	20	26.3	1.92	2.15
Azinphosmethyl	Guthion	1	1.3	0.22	0.24
Butylate	Sutan	1	1.3	1.00	1.12
Chloramben	Amiben	7	9.2	1.44	1.61
Captan		1	1.3	0.01	0.01
Carbaryl	Sevin	1	1.3	0.61	0.68
Chlordane		1	1.3	2.50	2.80
Copper hydroxide		1	1.3	1.08	1.21
Copper sulfate		1	1.3	1.42	1.59
2,4-D		4	5.2	0.63	0.70
Diazinon		1	1.3	0.40	0.44
DNBP	Premerge	1	1.3	2.25	2.52
Endosulfan		1	1.3	0.54	0.60
EPTC	Eptam	1	1.3	2.00	2.24
Linuron	Lorox	2	2.6	0.55	0.61
Maneb		1	1.3	2.14	2.39
Propachlor	Ramrod	2	2.6	1.20	1.34
Silvex		1	1.3	0.25	0.28
Simazine	Princep	3	3.9	2.00	2.24
Trifluralin	Treflan	5	6.5	2.37	2.66
2,4,5-T		1	1.3	0.35	0.39
IOWA, 152 SITES					
Alachlor	Lasso	15	9.8	0.93	1.04
Aldrin		10	6.5	0.83	0.93
Atrazine	AAtrex	39	25.6	1.39	1.56
Butylate	Sutan	7	4.6	2.46	2.75
Bux		6	3.9	0.82	0.92
Captan		1	0.6	0.01	0.01
Carbaryl	Sevin	1	0.6	1.60	1.79
Carbofuran	Furadan	7	4.6	0.92	1.03
Chloramben	Amiben	12	7.8	1.10	1.23
2,4-D		19	12.5	0.54	0.61
DDT		2	1.3	1.00	1.12
Diazinon		6	3.9	0.54	0.61
Dicamba	Banvel D	3	1.9	0.75	0.84
DNBP	Premerge	2	1.3	0.44	0.49
Dyfonate		1	0.6	0.90	1.00
Ethoprop	Mocap	1	0.6	1.00	1.12
Fensulfothion		1	0.6	1.02	1.14
Lindane		1	0.6	0.02	0.02
Linuron	Lorox	3	1.9	1.00	1.12
Phorate	Thimet	6	3.9	0.93	1.04
Propachlor	Ramrod	14	8.5	1.50	1.68
Toxaphene		1	0.6	2.73	3.05
Trifluralin	Treflan	14	9.2	0.69	0.77
KENTUCKY, 31 SITES					
Atrazine	AAtrex	2	6.4	1.02	1.14
Dalapon	Dowpon	2	6.4	1.05	1.17
2,4-D		1	3.2	0.05	0.05
2,4-DB	Butyrac	1	3.2	0.80	0.89
Paraquat		1	3.2	1.00	1.12
LOUISIANA, 25 SITES					
Alachlor	Lasso	1	4.0	1.00	1.12
Aldrin		3	12.0	0.15	0.16
Azinphosmethyl	Guthion	1	4.0	0.75	0.84
2,4-D		4	16.0	0.81	0.91
2,4-DB	Butyrac	1	4.0	1.95	2.18
Dalapon	Dowpon	1	4.0	6.80	7.62
DDT		5	20.0	6.62	7.41
DEF		1	4.0	1.12	1.25
Diuron	Karmex	1	4.0	0.70	0.78
DSMA		5	20.0	2.24	2.51
EMTS	Ceresan M	1	4.0	0.01	0.01
Fluometuron	Cotoran	6	24.0	0.99	1.11
Linuron	Lorox	1	4.0	0.50	0.56
Methylmercury dicyandiamide	Panogen	3	12.0	0.08	0.08
Monocrotophos	Azodrin	3	12.0	0.27	0.29
Monuron		1	4.0	1.60	1.79
MSMA		2	8.0	1.46	1.63
Nitralin	Planavin	1	4.0	0.75	0.84
Parathion, methyl		7	28.0	3.56	3.99
Prometryn	Caparol	2	8.0	1.08	1.21
Propanil	Stam	4	16.0	2.44	2.73
Silvex		1	4.0	0.50	0.56
Sodium chlorate		1	4.0	0.05	0.05
TCA		1	4.0	4.00	4.48
Toxaphene		6	24.0	13.45	15.07
Trifluralin	Treflan	2	8.0	0.63	0.70
MICHIGAN, 54 SITES					
Aldrin		1	1.8	2.00	2.24
Atrazine	AAtrex	14	25.9	2.00	2.24
Captan		1	1.8	5.00	5.60
Carbaryl		1	1.8	1.00	1.12
2,4-D		1	1.8	1.00	1.12
Demeton		1	1.8	3.00	3.36
Endosulfan		1	1.8	6.00	6.72
EPTC	Eptam	3	5.5	1.67	1.86
Fentin hydroxide		1	1.8	2.25	2.52
Isodrin		1	1.8	0.01	0.01
Lead arsenate		1	1.8	16.00	17.93
Mancozeb	Dithane M-45	1	1.8	12.00	13.44
Parathion, ethyl		2	3.7	3.50	3.92
Phosalone		1	1.8	4.00	4.48
Prolate	Imidan	2	3.7	11.60	13.00
Pyrazon	Pyramin	1	1.8	1.25	1.40
Silvex		1	1.8	0.50	0.56
TCA		1	1.8	1.00	1.12
TEPP		1	1.8	4.00	4.48
MID-ATLANTIC,[1] 16 SITES					
Alachlor	Lasso	1	6.2	2.00	2.24
Atrazine	AAtrex	4	25.0	0.94	1.05
Butylate	Sutan	2	12.5	1.63	1.82
Captan		2	12.5	0.01	0.01
Carbofuran	Furadan	1	6.2	1.00	1.12
2,4-D		1	6.2	0.50	0.56
Diazinon		1	6.2	0.80	0.89
Malathion		2	12.5	0.01	0.01
Parathion, ethyl		1	6.2	2.00	2.24
Sulfur		1	6.2	48.00	53.79
MISSISSIPPI, 31 SITES					
Alachlor	Lasso	1	3.2	0.75	0.84
Captan		1	3.2	0.03	0.03
Chloroneb	Demosan	7	22.5	0.03	0.03
DDT		8	25.8	3.81	4.27
DEF		4	12.9	0.90	1.00
Disulfoton	Di-Syston	7	22.5	0.01	0.01
Diuron	Karmex	2	6.4	2.75	3.08
DNBP		4	12.9	1.19	1.33
DSMA		1	3.2	1.86	2.08
Endrin		1	3.2	4.20	4.70
Ethylmercury chloride	Ceresan Red	1	3.2	0.01	0.01
Fluometuron	Cotoran	5	16.1	0.76	0.85
Folex		3	9.6	0.75	0.84
Linuron	Lorox	1	3.2	1.00	1.12
Malathion		1	3.2	2.40	2.68
Methylmercury acetate	Ceresan L	6	19.3	0.01	0.01
Mirex		4	12.9	0.08	0.09
MSMA		7	22.5	2.48	2.77
Nitralin	Planavin	1	3.2	1.00	1.12
Parathion, methyl		13	41.9	3.36	3.77

(Continued next page)

TABLE 3 (cont'd.). *Compounds applied to cropland sites by state, 1971—National Soils Monitoring Program*

Compound	Trade Name, If Noted	Pesticides Applied: No. of Sites	Pesticides Applied: % of Sites	Average Total Application: lb./Acre	Average Total Application: kg/Hectare
Sodium chlorate		2	6.4	1.07	1.20
Terrazole		1	3.2	0.15	0.16
Thiram		1	3.2	0.04	0.04
Toxaphene		10	32.2	6.90	7.73
Trifluralin	Treflan	7	22.5	0.75	0.84
MISSOURI, 80 SITES					
Alachlor	Lasso	7	8.7	1.82	2.03
Aldrin		3	3.7	0.63	0.70
Atrazine	AAtrex	17	21.2	2.24	2.50
Chloramben	Amiben	2	2.5	0.88	0.98
2,4-D		2	2.5	0.50	0.56
2,4-DB	Butyrac	1	1.2	0.22	0.24
DSMA		1	1.2	3.00	3.36
Linuron	Lorox	2	2.5	0.88	0.98
MSMA		1	1.2	1.65	1.84
Naptalam	Alanap	2	2.5	2.00	2.24
Propachlor	Ramrod	3	3.7	3.13	3.51
Trifluralin	Treflan	3	3.7	0.75	0.84
NEBRASKA, 105 SITES					
Alachlor	Lasso	2	1.9	1.50	1.68
Atrazine	AAtrex	22	20.9	1.38	1.54
Bux		5	4.7	0.91	1.01
Captan		31	29.5	0.01	0.01
Carbaryl	Sevin	2	1.9	1.17	1.31
Carbofuran	Furadan	6	5.7	0.89	0.99
Chevron RE-5353		4	3.8	0.85	0.95
2,4-D		21	20.0	0.72	0.81
Diazinon		1	0.9	1.30	1.45
Dichloropropene		1	0.9	17.00	19.05
Dieldrin		2	1.9	0.01	0.01
Disulfoton	Di-Syston	1	0.9	1.00	1.12
EPTC	Eptam	1	0.9	3.00	3.36
Fensulfothion		1	0.9	0.61	0.68
Heptachlor		1	0.9	0.01	0.01
Londax		1	0.9	0.50	0.56
Malathion		26	24.7	0.03	0.03
Methoxychlor		2	1.9	0.01	0.01
Methylmercury dicyandiamide	Panogen	2	1.9	0.01	0.01
Parathion, ethyl		2	1.9	1.00	1.12
Phorate	Thimet	5	4.7	0.87	0.97
Propachlor	Ramrod	4	3.8	2.59	2.90
Thiram		1	0.9	0.01	0.01
NEW ENGLAND,[3] 18 SITES					
Alachlor	Lasso	1	5.5	2.00	2.24
Atrazine	AAtrex	1	5.5	1.00	1.12
Azinphosmethyl	Guthion	1	5.5	0.50	0.56
Carbaryl	Sevin	1	5.5	1.25	1.40
Dinitrocresol		1	5.5	0.75	0.84
Endosulfan		1	5.5	0.75	0.84
EPTC	Eptam	1	5.5	4.00	4.48
Maneb		1	5.5	4.80	5.37
Parathion, methyl		1	5.5	1.25	1.40
NEW YORK, 37 SITES					
Atrazine	AAtrex	11	29.7	1.38	1.54
Azinphosmethyl	Guthion	2	5.4	0.56	0.62
Butylate	Sutan	1	2.7	3.00	3.36
Captan		4	10.8	0.66	0.73
Carbaryl	Sevin	2	5.4	3.20	3.58
2,4-D		3	8.1	0.37	0.41
Dieldrin		2	5.4	0.25	0.28
Dinitrocresol		1	2.7	2.67	2.99
Disulfoton	Di-Syston	2	5.4	0.70	0.78
DNBP	Premerge	2	5.4	2.97	3.33
Dodine		1	2.7	0.33	0.36
Endosulfan		1	2.7	0.50	0.56
Folpet	Phaltan	1	2.7	1.00	1.12
Lead arsenate		1	2.7	4.00	4.48
Malathion		2	5.4	0.01	0.01
Mancozeb	Dithane M-45	1	2.7	12.80	14.34
Methoxychlor		1	2.7	0.01	0.01
Phosphamidon	Dimecron	1	2.7	0.50	0.56
Propargite	Omite	1	2.7	5.00	5.60
Simazine	Princep	1	2.7	2.04	2.28
Sulfur		1	2.7	0.50	0.56
NORTH CAROLINA, 30 SITES					
Alachlor	Lasso	3	10.0	3.00	3.36
Atrazine	AAtrex	7	23.3	1.86	2.08
Carbaryl	Sevin	2	6.6	3.00	3.36
2,4-D		5	16.6	2.20	2.46
DEF		1	3.3	0.75	0.84
Dichlofenthion	Nemacide	1	3.3	6.00	6.72
Dichloropropene		1	3.3	67.00	75.09
Disulfoton	Di-Syston	2	6.6	0.90	1.00
Fensulfothion		1	3.3	2.00	2.24
Fluometuron	Cotoran	1	3.3	1.25	1.40
Malathion		1	3.3	0.50	0.56
Maleic hydrazide		2	6.6	3.00	3.36
Naptalam	Alanap	1	3.3	3.00	3.36
Toxaphene		1	3.3	0.09	0.10
Trifluralin	Treflan	2	6.6	1.00	1.12
OHIO, 59 SITES					
Alachlor	Lasso	3	5.0	0.92	1.02
Aldrin		3	5.0	3.67	4.10
Atrazine	AAtrex	8	13.5	2.01	2.25
Azinphosmethyl	Guthion	1	1.6	1.00	1.12
Bordeaux mixtures		1	1.6	1.25	1.40
Captan		1	1.6	5.20	5.82
Carbaryl	Sevin	2	3.3	1.13	1.26
Carbophenothion	Trithion	1	1.6	3.20	3.58
Chloramben	Amiben	2	3.3	2.75	3.08
Chloropropham	CIPC	1	1.6	2.50	2.80
Cypromid	Clobber	1	1.6	0.75	0.84
2,4-D		12	20.3	0.50	0.56
Dicamba	Banvel D	5	8.4	0.20	0.22
Dodine		1	1.6	0.50	0.56
Ferbam		1	1.6	3.17	3.55
Heptachlor		2	3.3	1.57	1.76
Lead arsenate		1	1.6	1.20	1.34
Linuron	Lorox	4	6.7	1.25	1.40
Methoxychlor		1	1.6	2.00	2.24
Parathion, ethyl		1	1.6	0.50	0.56
Phosphamidon	Dimecron	1	1.6	0.03	0.03
Simazine		1	1.6	2.00	2.24
OKLAHOMA, 62 SITES					
Alachlor	Lasso	1	1.6	5.00	5.60
Arsenic pentoxide		2	3.2	0.50	0.56
Atrazine	AAtrex	1	1.6	13.00	14.57
Captan		2	3.2	0.01	0.01
2,4-D		3	4.8	3.08	3.45
Disulfoton		2	3.2	0.65	0.72
EMTS	Ceresan M	4	6.4	0.01	0.01
Ethylmercury chloride	Ceresan Red	1	1.6	0.01	0.01
Furethrin		1	1.6	8.00	8.96
Methylmercury dicyandiamide	Panogen	2	3.2	0.01	0.01
Nabam		1	1.6	5.00	5.60
Parathion, ethyl		3	4.8	3.17	3.54
Parathion, methyl		5	8.0	0.50	0.56
Phorate		1	1.6	15.00	16.81
Thiram		3	4.8	0.01	0.01
OREGON, 37 SITES					
Atrazine	AAtrex	1	2.7	4.00	4.48
Bromacil	Hyvar	1	2.7	0.37	0.41
Captafol	Difolatan	1	2.7	1.50	1.68
2,4-D		3	8.1	0.50	0.56
Dicamba	Banvel D	1	2.7	0.06	0.06
Dichlorprop	2,4-DP	1	2.7	3.00	3.36
Dichloropropene		1	2.7	0.01	0.01
Disulfoton	Di-Syston	2	5.4	2.00	2.24
Endosulfan	Thiodan	2	5.4	0.75	0.84
EPTC	Eptam	1	2.7	3.00	3.36
Heptachlor		1	2.7	0.01	0.01
Hexachloro-benzene		1	2.7	0.01	0.01
Linuron	Lorox	1	2.7	0.75	0.84

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TABLE 3 (cont'd.). *Compounds applied to cropland sites by state, 1971—National Soils Monitoring Program*

Compound	Trade Name, If Noted	Pesticides Applied: No. of Sites	Pesticides Applied: % of Sites	Average Total Application: lb./Acre	Average Total Application: kg/Hectare
Malathion		1	2.7	1.00	1.12
Methylmercury dicyandiamide	Panogen	3	8.1	0.14	0.15
Mevinphos	Phosdrin	1	2.7	0.50	0.56
Oxydemeton-methyl	Metasystox-R	1	2.7	0.50	0.56
Parathion, ethyl		2	5.4	3.25	3.64
PENNSYLVANIA, 36 SITES					
Alachlor	Lasso	2	5.5	0.88	0.98
Atrazine	AAtrex	5	13.8	1.65	1.84
Butylate	Sutan	1	2.7	1.20	1.34
2,4-D		2	5.5	0.50	0.56
Malathion		1	2.7	1.00	1.12
Methoxychlor		1	2.7	1.00	1.12
SOUTH CAROLINA, 15 SITES					
Benefin	Balan	1	6.6	1.08	1.21
BHC		1	6.6	0.03	0.03
Carbaryl	Sevin	2	13.3	2.25	2.52
Copper sulfate		1	6.6	10.50	11.76
2,4-DB		1	6.6	0.25	0.28
DDT		3	20.0	8.94	9.24
DEF		2	13.3	1.16	1.30
Disulfoton	Di-Syston	2	13.3	0.59	0.65
Diuron	Karmex	2	13.3	1.00	1.12
Naptalam	Alanap	1	6.6	0.75	0.84
Parathion, methyl		5	33.3	4.76	5.34
Thiram		1	6.6	0.01	0.01
Toxaphene		3	20.0	13.16	14.75
Trifluralin	Treflan	2	13.3	1.00	1.12
SOUTH DAKOTA, 106 SITES					
Atrazine	AAtrex	5	4.7	1.59	1.78
Barban	Carbyne	1	0.9	0.25	0.28
Bux		3	2.8	0.70	0.78
Captan		24	22.6	0.01	0.01
2,4-D		32	30.1	0.45	0.50
Diazinon		3	2.8	1.64	1.83
Dicamba	Banvel D	2	1.8	0.12	0.13
Dieldrin		2	1.8	0.01	0.01
Ethylmercury chloride	Ceresan Red	1	0.9	0.01	0.01
Fensulfothion		1	0.9	1.20	1.34
Malathion		17	16.0	0.01	0.01
MCPA	MCP	2	1.8	0.25	0.28
Methoxychlor		1	0.9	0.01	0.01
Methylmercury dicyandiamide	Panogen	4	3.7	0.01	0.01
Parathion, methyl		1	0.9	0.50	0.56
Phorate	Thimet	1	0.9	0.60	0.67
Propachlor	Ramrod	3	2.8	2.40	2.68
Thiram		2	1.8	0.01	0.01
Atrazine	AAtrex	2	8.3	1.85	2.07
2,4-DB	Butyrac	1	4.1	0.29	0.32
DDT		1	4.1	3.00	3.36
TENNESSEE, 24 SITES					
DEF		1	4.1	1.50	1.68
Disulfoton	Di-Syston	3	12.5	3.00	3.36
Diuron	Karmex	2	8.3	1.55	1.73
DSMA		1	4.1	2.00	2.24
Fluometuron	Cotoran	2	8.3	1.50	1.68
Folex		1	4.1	1.50	1.68
Parathion, methyl		1	4.1	1.50	1.68
Sodium chlorate		1	4.1	1.50	1.68
Toxaphene		1	4.1	6.00	6.72
Trietazine		1	4.1	0.70	0.78
Trifluralin		3	12.5	0.98	1.09
VIRGINIA/WEST VIRGINIA,[1] 26 SITES					
Atrazine	AAtrex	2	7.6	2.00	2.24
Captan		1	3.8	0.08	0.08
Carbaryl	Sevin	1	3.8	2.00	2.24
Diazinon		1	3.8	0.40	0.44
Dinitrocresol		1	3.8	1.50	1.68
Endosulfan		1	3.8	1.20	1.34
EPTC	Eptam	1	3.8	0.70	0.78
Metham	Vapam	1	3.8	2.16	2.42
Methoxychlor		1	3.8	0.80	0.89
Vernolate	Vernam	1	3.8	2.00	2.24
WASHINGTON STATE, 45 SITES					
Aldrin		1	2.2	0.43	0.48
BHC		2	4.4	0.01	0.01
Bromacil		2	4.4	0.75	0.84
Captan		2	4.4	0.06	0.06
2,4-D		13	28.8	1.32	1.48
DDT		1	2.2	0.75	0.84
Dicamba	Banvel D	1	2.2	0.13	0.14
HCB		6	13.3	0.01	0.01
Methylmercury dicyandiamide		1	2.2	0.01	0.01
Parathion, ethyl		1	2.2	1.50	1.68
Phenylmercury acetate		1	2.2	0.01	0.01
Phenylmercury urea		3	6.6	0.01	0.01
Terbacil	Sinbar	1	2.2	1.40	1.56
Terbutryn	Igran	1	2.2	1.75	1.96
WISCONSIN, 66 SITES					
Alachlor	Lasso	4	6.0	1.44	1.61
Atrazine	AAtrex	25	37.8	1.83	2.04
Bux		1	1.5	7.00	7.84
Carbofuran	Furadan	3	4.5	0.90	1.01
2,4-D		2	3.0	1.50	1.68
2,4-DB	Butyrac	1	1.5	0.50	0.56
Disulfoton	Di-Syston	1	1.5	2.00	2.24
Endosulfan	Thiodan	1	1.5	1.00	1.12
Linuron	Lorox	1	1.5	1.00	1.12
MCPA	MCP	1	1.5	0.50	0.56
Phorate	Thimet	1	1.5	6.00	6.72
Thiram		1	1.5	0.01	0.01

[1] See Table 1.

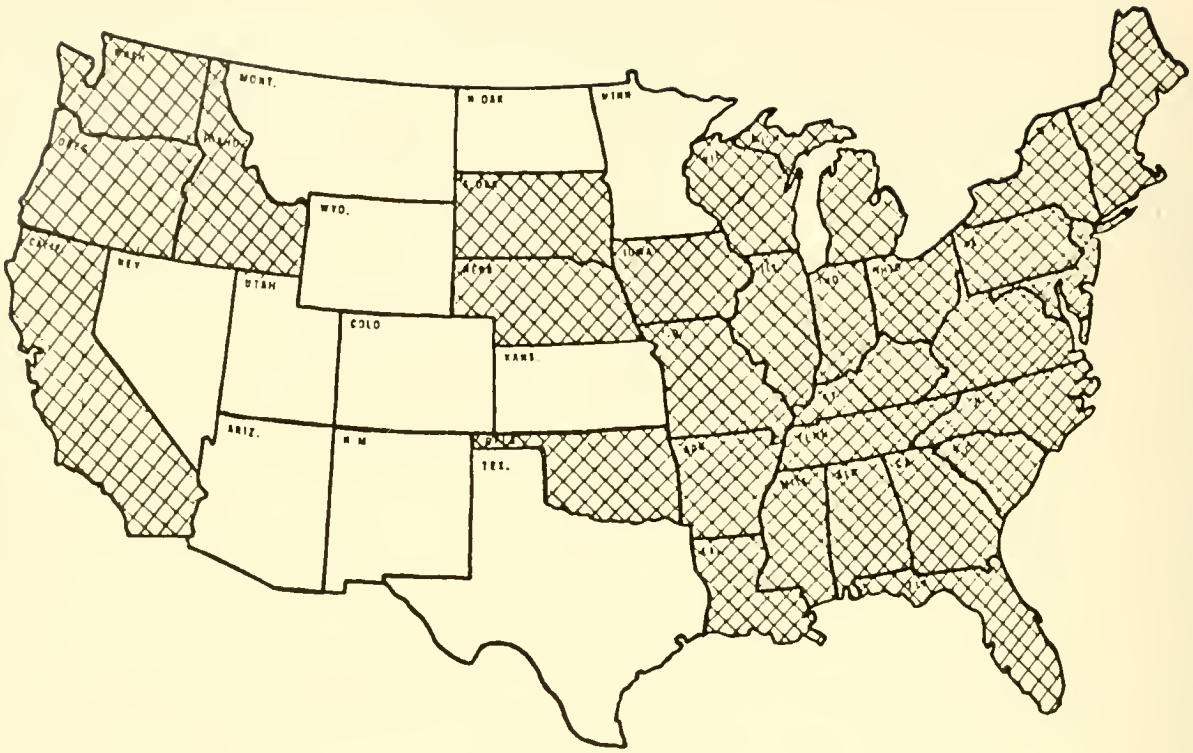

FIGURE 2. *Percent of sites reporting pesticide applications, 1971, National Soils Monitoring Program*

TABLE 4. *Crop and pesticide application data for sampling sites, 1971—National Soils Monitoring Program*

Crop	Total No. of Sites	Pesticides Applied No. of Sites	%	Pesticides Not Applied No. of Sites	%	Pesticides Application Unknown No. of Sites	%
Corn, field	445	366	82	70	16	9	2
Soybeans	251	147	59	100	40	4	1
Wheat	115	56	49	59	51		
Hay, mixed	112	3	3	108	96	1	1
Alfalfa and/or bur clover	108	10	9	97	90	1	1
Cotton	63	55	87	6	10	2	3
Sorghum, sweet sorghum, milo	52	32	62	19	37	1	1
Oats	47	16	34	30	64	1	2
Pasture	41	2	5	39	95		
Hay, grass	25			25	100		
Barley	16	4	25	12	75		
Peanuts	11	9	82			2	18
Potatoes, white	11	9	82	2	12		
Clover	9	1	11	8	89		
Rice	9	8	89	1	11		
Beans, dry	8	5	63	3	37		
Grapes	8	7	88	1	12		
Apples	7	7	100				
Oranges	7	5	71	2	29		
Sugarbeets	6	5	83	1	17		
Sugarcane	6	6	100				
Peas	5	2	40	3	60		
Rye	5			5	100		
Tobacco	5	4	80	1	20		
Safflower	3	1	33	2	67		
Almonds	2	2	100				
Blueberries	2			2	100		
Cabbage	2	2	100				
Figs	2			2	100		
Peaches	2	2	100				
Timothy	2			2	100		
Tomatoes	2						
Apricots	1	1	100				
Broccoli	1	1	100				
Carrots	1	1	100				
Cherries	1			1	100		
Cowpeas	1			1	100		
Cucumbers	1	1	100				
Flax	1			1	100		
Grapefruit	1	1	100				
Lemons	1	1	100				
Lentils	1	1	100				
Lettuce	1	1	100				
Mint	1	1	100				
Pecans	1			1	100		
Plums/prunes	1					1	100
String beans	1	1	100				
Watermelon	1	1	100				
Other	11	3	27	8	73		
Fallow sites	83	3	4	80	96		

TABLE 5. *Compounds applied to cropland sites by crop, 1971—National Soils Monitoring Program*

Compound	Pesticides Applied		Average Total Application		Reported Total Application Rate, kg/Hectare	
	No. of Sites	% of Sites	lb./Acre	kg/Hectare	Min.	Max.
ALFALFA and BUR CLOVER, 106 SITES						
Carbaryl	1	0.9	1.00	1.12	—	1.12
Diazinon	1	0.9	0.40	0.45	—	0.45
EPTC	2	1.9	1.85	2.07	0.78	3.36
Malathion	3	2.8	1.13	1.27	1.12	1.57
Methoxychlor	2	1.9	0.90	1.01	0.90	1.12
Mevinphos	2	1.9	1.50	1.68	1.12	2.24
Parathion, ethyl	3	2.8	2.50	2.80	0.56	6.72
Trichlorfon	1	0.9	0.75	0.84	—	0.84
COTTON, 61 SITES						
Aldrin	1	1.6	0.01	0.01	0.01	
Arsenic pentoxide	2	3.3	0.50	0.56	0.56	0.56
Azodrin	4	6.6	0.33	0.37	0.06	0.56
Cacodylic acid	1	1.6	0.01	0.01	0.01	
Captan	1	1.6	0.01	0.01	0.01	
Chloroneb	9	14.8	0.01	0.01	0.01	0.01
2,4-D	1	1.6	0.44	0.49	0.49	
DDT	25	41.0	4.28	4.80	0.09	13.45
DEF	9	14.8	1.03	1.16	0.67	1.68
Dicofol	1	1.6	1.00	1.12	1.12	
Disulfoton	14	23.0	1.31	1.46	0.01	7.85
Diuron	8	13.1	1.45	1.63	0.38	5.04
DNBP	2	3.3	1.62	1.82	1.12	2.52
DSMA	11	18.0	2.00	2.24	0.24	4.48
EMTS	4	6.6	0.04	0.04	0.01	0.11
Endrin	2	3.3	2.80	3.14	1.57	4.71
Ethylmercury chloride	1	1.6	0.01	0.01	0.01	
Fluometuron	21	34.4	0.96	1.07	0.56	2.24
Folex	5	8.2	1.05	1.18	0.84	1.68
Linuron	2	3.3	0.63	0.70	0.56	0.84
Malathion	2	3.3	1.55	1.74	0.78	2.69
MCPA	1	1.6	0.50	0.56	0.56	
Mercury	2	3.3	0.06	0.07	0.01	0.12
Methyl trithion	1	1.6	3.00	3.36	3.36	
Methylmercury acetate	6	9.8	0.01	0.01	0.01	0.01
Methylmercury dicyandiamide	2	3.3	0.01	0.01	0.01	0.01
Mirex[1]	1	1.6	0.01	0.01	0.01	
Monuron	2	3.3	1.30	1.46	1.12	1.79
MSMA	15	24.6	1.86	2.08	0.75	5.60
Nitralin	1	1.6	1.00	1.12	1.12	
Paraquat	1	1.6	0.25	0.28	0.28	
Parathion, ethyl	5	8.2	6.78	7.60	0.84	21.02
Parathion, methyl	36	59.0	3.26	3.65	0.06	11.21
PCNB	2	3.3	3.51	3.93	0.01	7.85
Prometryn	2	3.3	1.08	1.21	0.18	2.24
Sodium chlorate	4	6.6	1.80	2.02	0.06	5.60
Terrazole	1	1.6	0.15	0.17	0.17	
Thiram	3	4.9	0.01	0.01	0.01	0.01
Toxaphene	27	44.3	7.95	8.91	0.10	40.35
Trifluralin	21	34.4	0.74	0.83	0.28	1.24
FIELD CORN, 427 SITES						
Alachlor	37	8.7	1.66	1.86	0.28	6.72
Aldrin	37	8.7	1.37	1.54	0.11	5.60
Atrazine	199	46.6	1.72	1.93	0.16	4.48
Butylate	18	4.2	1.74	1.95	0.28	3.36
Bux	17	4.0	1.26	1.41	0.50	7.85
Captan	116	27.2	0.01	0.01	0.01	0.01
Carbaryl	3	0.7	1.32	1.47	0.84	1.79
Carbofuran	18	4.2	0.81	0.90	0.28	2.58
Chevron RE-5353	4	0.9	0.85	0.95	0.78	1.12
Chloramben	2	0.5	1.12	1.26	0.28	2.24
Chlordane	1	0.2	2.50	2.80	2.80	
Cypromid	1	0.2	0.75	0.84	0.84	
2,4-D	72	16.9	0.73	0.81	0.06	3.36
Dalapon	2	0.5	1.05	1.18	1.18	1.18
DDT	2	0.5	1.00	1.12	1.12	1.12
Demeton	1	0.2	0.12	0.13	0.13	
Diazinon	11	2.6	0.93	1.05	0.01	2.80
Dicamba	9	2.1	0.37	0.42	0.13	1.12
Dieldrin	3	0.7	0.01	0.01	0.01	0.01
DNBP	1	0.2	3.20	3.59	3.59	
Disulfoton	2	0.5	0.65	0.73	0.56	0.90
Dyfonate	1	0.2	0.90	1.01	1.01	
EPTC	1	0.2	2.00	2.24	2.24	
Ethoprop	1	0.2	1.00	1.12	1.12	

(*Continued next page*)

TABLE 5 (cont'd.). *Compounds applied to cropland sites by crop, 1971—National Soils Monitoring Program*

Compound	Pesticides Applied		Average Total Application		Reported Total Application Rate, kg/Hectare	
	No. of Sites	% of Sites	lb./Acre	kg/Hectare	Min.	Max.
Ethylmercury chloride	1	0.2	0.01	0.01	0.01	
Fensulfothion	4	0.9	0.93	1.04	0.68	1.34
Ferbam	1	0.2	2.00	2.24	2.24	
Furethrin	1	0.2	8.00	8.97	8.97	
Heptachlor	6	1.4	1.67	1.87	0.01	3.36
Isodrin	1	0.2	0.01	0.01	0.01	
Lindane	1	0.2	0.02	0.02	0.02	
Linuron	2	0.5	0.52	0.59	0.33	0.84
Londax	1	0.2	0.50	0.56	0.56	
Malathion	96	22.5	0.01	0.01	0.01	0.01
Methoxychlor	20	4.7	0.01	0.01	0.01	0.02
Mirex[1]	1	0.2	0.01	0.01	0.01	
MSMA	1	0.2	0.25	0.28	0.28	
Paraquat	1	0.2	1.00	1.12	1.12	
Parathion, ethyl	3	0.7	0.87	0.97	0.56	1.23
PCP	1	0.2	6.00	6.72	6.72	
Phorate	20	4.7	0.81	0.91	0.19	16.81
Propachlor	38	8.9	1.42	1.57	0.10	6.72
Silvex	2	0.5	0.38	0.42	0.28	0.56
Simazine	5	1.2	2.00	2.24	1.40	2.80
2,4,5-T	2	0.5	0.30	0.34	0.28	0.39
Thiram	1	0.2	0.01	0.01	0.01	
Toxaphene	1	0.2	2.73	3.06	3.06	
MIXED HAY, 111 SITES						
2,4-D	2	1.8	0.80	0.90	0.67	1.12
Mirex[1]	1	0.9	0.01	0.01	0.01	
SOYBEANS, 243 SITES						
Alachlor	27	11.1	1.33	1.49	0.22	6.16
Captan	3	1.2	0.04	0.04	0.01	0.08
Carbaryl	5	2.1	1.88	2.11	0.90	4.30
Chloramben	38	15.6	1.42	1.59	0.25	4.48
Chloropropham	1	0.4	2.50	2.80	2.80	
Dalapon	1	0.4	6.80	7.62	7.62	
2,4-DB	6	2.5	0.84	0.94	0.25	2.19
DDT	2	0.8	2.50	2.80	1.12	4.48
Dichloropropene	1	0.4	67.00	75.09	75.09	
DNBP	10	4.1	1.08	1.21	0.43	2.52
Fluometuron	1	0.4	1.00	1.12	1.12	
Linuron	16	6.6	0.96	1.08	0.28	2.24
Mirex[1]	1	0.4	0.01	0.01	0.01	
MSMA	1	0.4	2.00	2.24	2.24	
Naptalam	3	1.2	2.33	2.61	1.12	3.36
Nitralin	4	1.6	1.06	1.19	0.84	1.68
Paraquat	1	0.4	2.00	2.24	2.24	
Parathion, methyl	3	1.2	2.55	2.86	1.12	5.77
Propachlor	1	0.4	2.80	3.14	3.14	
Solan	1	0.4	1.00	1.12	1.12	
Thiram	1	0.4	0.04	0.04	0.04	
Toxaphene	2	0.8	3.82	4.29	2.24	6.33
Trifluralin	38	15.6	1.11	1.24	0.25	5.60
Vernolate	3	1.2	0.80	0.90	0.78	1.01
WHEAT, 113 SITES						
Aldrin	1	0.9	0.01	0.01	0.01	
Azinphosmethyl	1	0.9	0.22	0.25	0.25	
Barban	1	0.9	0.25	0.28	0.28	
BHC	2	1.8	0.02	0.02	0.01	0.02
Bromacil	2	1.8	0.56	0.63	0.41	0.84
Captan	1	0.9	0.25	0.28	0.28	
2,4-D	28	24.8	0.83	0.93	0.13	4.48
Dicamba	3	2.7	0.11	0.12	0.07	0.15
Dichlorprop	1	0.9	3.00	3.36	3.00	
Disulfoton	2	1.8	0.36	0.40	0.40	0.40
EMTS	4	3.5	0.01	0.01	0.01	0.01
Ethylmercury chloride	4	3.5	0.06	0.06	0.01	0.11
Hexachlorobenzene	6	5.3	0.02	0.02	0.01	0.03
Methylmercury dicyandiamide	9	8.0	0.01	0.01	0.01	0.01
Parathion, ethyl	1	0.9	8.00	8.97	8.97	
Parathion, methyl	4	3.5	0.50	0.56	0.56	0.56
Phenylmercury acetate	3	2.7	0.01	0.01	0.01	0.01
Terbutryne	1	0.9	1.75	1.96	1.96	
Thiram	2	1.8	0.01	0.01	0.01	0.01

[1] Aerially applied for control of the imported fire ant.

TABLE 6. *Pesticide application information on selected crops, by state, for sampling sites, 1971—National Soils Monitoring Program*

State	Total No. of Sites	Pesticides Applied	Pesticides Not Applied	Pesticides Use Unknown	Total No. of Sites	Pesticides Applied	Pesticides Not Applied	Pesticides Use Unknown
	ALFALFA AND/OR BUR CLOVER				COTTON			
Alabama	0				4	4		
Arkansas	1		1		9	7	1	1
California	5	3	2		7	6	1	
Georgia	0				5	5		
Illinois	4		4		0			
Indiana	1		1		0			
Iowa	19		19		0			
Louisiana	0				7	7		
Michigan	7	1	6		0			
Mississippi	0				13	12	1	
Missouri	3	1	2		1	1		
Nebraska	10		10		0			
New England	2		2		0			
New York	4		4		0			
N. Carolina	0				1	1		
Ohio	2		2		0			
Oklahoma	2	1	1		5	2	2	1
Oregon	5	1	4		0			
Pennsylvania	7	1	5	1	0			
S. Carolina	0				5	4	1	
S. Dakota	16		16		0			
Tennessee	0				6	6		
Va./W. Va.	2	2			0			
Washington state	2		2		0			
Wisconsin	16		16		0			
	FIELD CORN				SOYBEANS			
Alabama	5	3	2		7	1	6	
Arkansas	1		1		24	13	11	
California	1		1		0			
Florida	1		1		2		2	
Georgia	13	5	8		3	2	1	
Illinois	67	65	2		58	36	22	
Indiana	34	31	2	1	21	17	3	1
Iowa	81	70	11		42	34	8	
Kentucky	16	10	4	2	3	1	1	1
Louisiana	1		1		5	2	3	
Michigan	21	14	7		1		1	
Mid-Atlantic	9	5	2	2	1	1		
Mississippi	1	1			14	9	5	
Missouri	18	16	2		22	13	8	1
Nebraska	46	40	4	2	3		3	
New England	3	2	1		0			
New York	15	11	2	2	0			
N. Carolina	13	9	4		8	5	3	
Ohio	23	19	4		14	8	5	1
Oklahoma	3	2	1		2		1	1
Pennsylvania	8	5	3		1		1	
S. Carolina	2		1	1	7	2	5	
S. Dakota	27	26	1		1	1		
Tennessee	5	2	3		9	2	7	
Va./W. Va.	3	3			1		1	
Washington state	2	1	1		0			
Wisconsin	24	23	1		0			
	WHEAT				MIXED HAY			
Alabama	0				1		1	
Arkansas	1		1		2		2	
California	4	1	3		1		1	
Florida	0				1		1	
Idaho	13	5	8		0			
Illinois	6	1	5		1		1	
Indiana	5	1	4		2		2	
Iowa	1		1		5		5	
Kentucky	0				2		2	
Michigan	1		1		9	1	8	
Mid-Atlantic	0				1		1	
Mississippi	0				1	1		
Missouri	1		1		20		20	
Nebraska	3	1	2		0			
New England	0				5		5	
New York	0				9		8	1
N. Carolina	0				1		1	
Ohio	5	1	4		6		6	
Oklahoma	34	13	21		0			
Oregon	3	3			3		3	

(Continued next page)

TABLE 6 (cont'd.). *Pesticide application information on selected crops, by state, for sampling sites, 1971—National Soils Monitoring Program*

	Total No. of Sites	Pesticides Applied	Pesticides Not Applied	Pesticides Use Known	Total No. of Sites	Pesticides Applied	Pesticides Not Applied	Pesticides Use Known
Pennsylvania	0				13	1	12	
S. Dakota	20	15	5		8		8	
Va., W. Va.	0				5		5	
Washington state	18	15	3		1		1	
Wisconsin	0				15		15	

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WATER

Organochlorines, Cholinesterase Inhibitors, and Aromatic Amines in Dutch Water Samples, September 1969–December 1975

Ronald C. C. Wegman and Peter A. Greve [1]

ABSTRACT

The Dutch aquatic environment was monitored from September 1969 to December 1975 for organochlorine pesticides and their metabolites, cholinesterase inhibitors, and aromatic amines. The 1,492 samples analyzed included surface water, rainwater, groundwater, and drinking water.

The highest concentrations of hexachlorobenzene (HCB) and α- and β-benzene hexachloride (BHC) were found in the Rhine River and its tributaries. Concentrations of the compounds in the Dutch part of the Rhine River decreased downstream. Other organochlorine pesticides and their metabolites, heptachlor, heptachlor epoxide, aldrin, dieldrin, endrin α- and β-endosulfan, and ΣDDT were detected occasionally, but only in low concentrations. Cholinesterase inhibitors and aromatic amines were always present in the Rhine River and its tributaries.

Introduction

Preliminary investigations before 1969 of organochlorine pesticides and related substances in the Dutch aquatic environment indicated the necessity of a long-term investigation. Endosulfan levels found in the Rhine River later in 1968 (*6*) underlined the need for such an investigation.

Samples were taken from surface water, rainwater, groundwater, and drinking water prepared from surface water. Presently, about one third of the Dutch population is at least partly supplied with drinking water prepared from surface water. Sampling sites varied every year, except for a few fixed sites including the Maas and Rhine Rivers, so that after 7 years all parts of The Netherlands were investigated for at least 1 year. Special interest was paid to large agricultural areas such as the IJsselmeerpolders.

During the study, the number of sampling sites at drinking water stations was gradually decreased as the stations acquired equipment and expertise to analyze their own samples.

Levels of organochlorine pesticides were determined because they are persistent and accumulate in the food chain. Analyses were performed for cholinesterase inhibitors including phosphates, thiophosphates, dithiophosphates, and carbamates (e.g., dichlorvos, parathion, malathion, carbaryl, respectively). From the herbicide group, urea compounds were chosen because of their great application rate. This group of compounds was determined as their aromatic amine moiety.

During the present investigation, papers were published on endosulfan in the Rhine River (*6*), cholinesterase inhibitors in Dutch surface waters (*8*), pesticides in the Rhine River (*9*), aromatic amines and their derivatives in Dutch surface waters (*10*), and the fate of pesticides during drinking water preparation (*7*). In cooperation with the Federal Health Office in Berlin, the concentrations of cholinesterase inhibitors in the German and Dutch parts of the Rhine River were compared and the main source was determined (*5*). From these papers, only the primary results are repeated here.

Methods and Materials

The 1,492 samples were collected by means of a bail and were transported in acetone-washed bottles to the National Institute of Public Health, Bilthoven, The Netherlands. Surface water was taken from a depth of about 1 m. Locations of the 92 sampling sites are given in Figure 1.

The methods mentioned in the present report include improvements introduced during the study. They had no

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FIGURE 1. *Sampling sites for study of organochlorines, cholinesterase inhibitors, and aromatic amines in Dutch water samples*

significant influence on the results, except for the C_6 compounds which could be determined separately only from May 1970.

ORGANOCHLORINE COMPOUNDS

Water samples of 1000 ml, including silt, were extracted successively with 200, 100, and 100 ml of petroleum ether (boiling range, 40°–60°C). The combined extracts were dried over anhydrous sodium sulfate and concentrated to about 5 ml in a Kuderna-Danish evaporative concentrator. The last few milliliters of solvent were evaporated to exactly 1 ml by a gentle stream of nitrogen at room temperature. The concentrated extract was added to a microcolumn containing 2.00 g basic alumina (W-200, activity Super I, Woelm). Before use, the microcolumn was activated for 16 hours at 150°C, and then deactivated with 11 percent water (11 g water + 89 g alumina).

The column was eluted with 5 ml of petroleum ether to produce Eluate A containing HCB, α- and γ-BHC, heptachlor epoxide (about 10 percent), *p,p'*-DDE, *o,p'*-DDT, TDE, *p,p'*-DDT, telodrin, isodrin, aldrin, and heptachlor. The receiving tube was changed and a

second elution was carried out with 10 ml of a 20:80 (v/v) mixture of ethyl ether–petroleum ether to produce Eluate B containing β-BHC, heptachlor epoxide (about 90 percent), dieldrin, and endrin. The eluates were concentrated to exactly 1 ml by a gentle stream of nitrogen at room temperature.

To determine α- and β-endosulfan, a microcolumn containing 2.00 g 60–200-mesh silica gel (Fisher S 661) activated for 2–3 hours at 140°C was used. The column was eluted first with 8 ml of a 80:20 (v/v) mixture of hexane–toluene and next with 8 ml of a 40:60 (v/v) mixture of hexane–toluene and 8 ml toluene; α- and β-endosulfan were present in the second eluate. One-μl portions of the concentrated eluates were injected into the gas chromatographs. Instrument parameters and operating conditions follow:

(1) Model 1800 Varian Aerograph

Detector:	tritium electron-capture
Column:	180 cm × 0.3 cm ID Pyrex, packed with a mixture of 5 percent OV-210 and 5 percent OV-17 (4+1) on 80–100-mesh Chromosorb W-HP
Temperatures:	injection port 205°C oven 190°C detector 200°C
Carrier gas:	nitrogen flowing at 40 ml/minute

(2) Perkin-Elmer Model F 22 gas chromatograph

Detector:	^{63}Ni electron-capture
Column:	40 m × 0.35 mm ID Pyrex capillary, coated with SE-30 (GC grade)
Temperatures:	injection port 215°C oven 155°–225°C at 3°C/minute with a linear temperature programmer detector 250°C
Carrier gas:	helium flowing at 2–3 ml/minute; helium splitting gas flow of 0–60 ml/minute; nitrogen purge gas flow of 80 ml/minute

The practical lower limit of detectability was 0.01 ppb. Recovery data, obtained by spiking river water samples with the pesticides and carrying them through the entire analytical procedure, were over 90 percent. Results are not corrected for recovery. To confirm the identity of the pesticides, *p*-values or chemical conversions were used, such as the quantitative conversion of *o,p'*-DDT and *p,p'*-DDT to, respectively, *o,p'*-DDE and *p,p'*-DDE by treatment with MgO, the disappearance of dieldrin and endrin by treatment with concentrated sulfuric acid, and the peak shift for endosulfan under the influence of alkali (*6*).

AROMATIC AMINES

The sums of aromatic amines and their derivatives were determined colorimetrically (*10*). Concentrations are expressed as 3,4-dichloroaniline. The practical lower limit of detectability was 0.5 ppb.

CHOLINESTERASE INHIBITORS

Colorimetric determination of cholinesterase inhibitors was performed in a methylene chloride extract of the sample on an AutoAnalyzer (*9*). The enzyme source was freeze-dried human plasma. Concentrations were calculated as paraoxon equivalents. The practical lower limit of detection was 0.2 ppb.

Results

The 20,000 data points collected in the monitoring program during 1969–75 are summarized in Tables 1–7. In view of the low frequency of occurrence and the low concentrations found, the concentrations of β-BHC, aldrin, heptachlor, heptachlor epoxide, endrin, TDE, *o,p'*-DDT, *p,p'*-DDE, and *p,p'*-DDT are not given in the tables. Unless stated otherwise, all extracts of water samples included silt.

The Rhine River was studied in more detail than the other Dutch surface waters. Samples were taken weekly near Lobith at sampling site 45 (Fig. 1). The geographical distribution of HCB, and α- and γ-BHC in the Rhine River is illustrated in Figures 2–4 for the southern branch of the river, Rhine–Boven Merwede–Nieuwe Waterweg.

Discussion

The data in Tables 1–7 indicate that the highest concentrations of pesticides and related substances are found in the Rhine River and its tributaries. The highest concentrations in the Maas River, compared below, are much lower.

Pesticide	Residue, ppb	
	Rhine River	Maas River
HCB	0.55	0.29
α-BHC	0.60	0.07
γ-BHC	0.42	0.18
Dieldrin	0.06	0.03
Endosulfan	0.81	0.09
Cholinesterase inhibitors	56	1.7
Aromatic amines	10	2.4

Levels in other waters were lower still or not detected.

HCB and α- and γ-BHC were almost always present in the Rhine water samples. Median values in ppb varied during 1969–75 as follows: HCB, 0.06–0.14; α-BHC, 0.06–0.22; and γ-BHC, 0.04–0.13. Concentrations of the by-product, α-BHC, are higher than those of the commercial product, γ-BHC. This means either that significant amounts of α-BHC-containing products, which have been banned for years, are still used along the Rhine or that industry, rather than agriculture, is the main source of pollution. Because the source of contamination is located across the German border, it was not possible to determine the exact source of the BHC discharge. BHC has had only limited use as a fungicide. Since July 1974, the concentrations of α- and γ-BHC in the Rhine have decreased considerably. Median values of α- and γ-BHC in 1974 were 0.22 ppb and 0.13 ppb, respectively; in 1975, 0.06 ppb and 0.04 ppb, respectively. The levels of α-BHC in the Rhine and its tributaries are considered harmful to the reproduction of *Daphnia magna* (water flea) (*3*).

TABLE 1. *Concentrations of BHC, dieldrin, endosulfan, and cholinesterase inhibitors in Dutch samples, 1969*

Sampling Site	No.	Types of Water	No. of Samples	Residues, ppb							
				γ-BHC		Dieldrin		α- and β-Endosulfan		Cholinesterase Inhibitors[1]	
				Max	Med	Max	Med	Max	Med	Max	Med
Surface water for drinking water preparation											
Braakman	1	raw water	2	—	—	0.01	—	—	—		
Berenplaat	2	raw water	4	0.16	0.06	0.01	—	0.03	—	3.02	1.03
Berenplaat	2	treated water	4	0.02	—	0.01	—	—	—	0.17	0.17
Drentse A	3	raw water	3	—	—	—	—	—	—		
Loenerveense Plas	4	raw water	3	—	—	0.01	0.01	—	—		
Wantij	6	raw water	3	0.09	0.05	0.01	—	0.11	0.09	5.20	1.82
IJsselmeer, Andijk	7	raw water	4	—	—	0.04	—	0.01	—		
IJsselmeer, Andijk	7	treated water	4	—	—	0.04	—	0.01	—		
Surface water for infiltration											
Amsterdam-Rijnkanaal	8	raw water	2	0.02	0.01	—	—	0.17	0.14	1.42	1.06
Amsterdam-Rijnkanaal	8	raw water[2]	1	—	—	—	—	0.06	0.06	1.20	1.20
Lek	9	raw water	2	0.03	0.02	—	—	0.10	0.09	1.38	1.09
Lek	9	raw water[2]	1	—	—	—	—	—	—	1.52	1.52
Enschede	10	raw water[3]	3	0.15	0.02	0.01	—	—	—	0.05	0.05
St. Jansteen	11	raw water	3	—	—	0.02	0.01	—	—		
St. Jansteen	11	treated water	1	0.01	0.01	0.03	0.03	0.03	0.03		
Valkenburgse Watering	15	raw water	4	0.03	0.02	0.02	—	0.05	—	0.32	0.22
IJsselmeer region											
IJsselmeer, Staveren	25	surface water	1	0.03	0.03	—	—	—	—		
IJsselmeer, Y-2	27	surface water	1	—	—	—	—	—	—		
IJsselmeer, Steile Bank	28	surface water	1	0.02	0.02	—	—	—	—		
Maas and tributaries											
Maas, Eijsden	35	surface water	7	0.08	0.02	—	—	0.09	—	0.44	0.22
Roer	42	surface water	2	0.02	0.01	0.01	—	—	—		
Niers	43	surface water	3	0.11	0.03	0.02	0.01	0.13	0.06	0.19	0.18
Rhine and tributaries											
Rhine	45	surface water	17	0.24	0.18	0.04	—	0.81	0.24	10.67	2.46
Kromme Rijn	47	surface water	6	0.08	0.02	0.02	—	0.04	—	2.04	1.00
Other surface waters											
Oostermoerse Vaart	57	surface water	4	0.01	—	0.02	—	—	—		
Boomawetering	76	surface water	8	0.09	—	0.02	0.01	0.01	—	0.57	0.42
Rijnbeek	82	surface water	4	—	—	0.01	—	—	—	0.52	0.52
Lage Vaart, Colijn	85	surface water	12	0.05	—	0.06	0.01	0.09	—		
Hoge Vaart, Colijn	86	surface water	24	0.08	—	0.08	—	0.10	—		
Lage Vaart, Wortman	89	surface water	29	0.10	—	0.14	—	0.09	—		
Larser Vaart	90	surface water	13	—	—	0.08	0.02	—	—		
Wortmanvaart	92	surface water	12	—	—	0.03	0.01	—	—		

NOTE: β-BHC, aldrin, heptachlor, heptachlor epoxide, endrin, and ΣDDT were detected occasionally in low concentrations; — = not detected. Unless stated otherwise, all water samples included silt.

[1] As paraoxon-equivalents.

[2] After rapid filtration.

[3] Before infiltration.

TABLE 2. *Concentrations of HCB, BHC, dieldrin, endosulfan, and cholinesterase inhibitors in Dutch water samples, 1970*

Sampling Site	No.	Types of Water	No. of Samples	Residues, ppb											
				HCB		α-BHC		γ-BHC		Dieldrin		α- and β-Endosulfan		Cholinesterase Inhibitors[1]	
				Max	Med	Max	Med	Max	Med	Max	Med	Max	Med	Max	Med
Surface water for drinking water preparation															
Braakman	1	raw water	2	0.02	—	0.02	0.01	0.02	0.01	0.02	0.01	0.03	—	—	—
Berenplaat	2	raw water	2	—	—	0.17	0.12	0.09	0.07	0.03	—	0.07	—	0.80	0.30
Drentse A	3	raw water	2	—	—	—	—	0.02	0.01	—	—	0.02	—	0.18	0.06
Loenerveense Plas	4	raw water	1	—	—	—	—	—	—	—	—	0.03	—	—	—
Oud-Beijerland	5	raw water	2	0.03	—	0.17	0.12	0.06	0.05	0.01	—	0.12	—	1.08	0.83
Oud-Beijerland	5	treated water	2	0.01	—	0.13	0.09	0.07	0.06	0.02	—	0.04	—	0.79	0.50
Wantij	6	raw water	3	0.08	0.05	0.18	0.15	0.12	0.11	—	—	0.04	—	2.00	0.63
Wantij	6	treated water	2	0.01	—	0.06	0.06	0.05	0.05	0.02	—	0.03	—	0.45	0.20
IJsselmeer, Andijk	7	raw water	3	—	—	0.02	0.02	0.02	0.01	0.01	—	0.07	—	0.27	0.06
IJsselmeer, Andijk	7	treated water	3	—	—	0.02	0.01	0.02	0.01	0.02	—	0.05	—	0.17	0.07
Surface water for infiltration															
Amsterdam-Rijnkanaal	8	raw water	3	0.05	0.03	0.13	0.08	0.13	0.08	0.01	—	0.05	—	0.82	0.52
Amsterdam-Rijnkanaal	8	raw water[2]	3	0.03	0.02	0.15	0.10	0.18	0.11	0.01	0.01	0.04	—	0.82	0.42
Lek	9	raw water	3	0.04	0.02	0.16	0.08	0.20	0.10	0.02	0.01	0.07	—	1.10	0.36
Lek	9	raw water[2]	3	0.03	0.02	0.17	0.10	0.20	0.11	0.01	—	0.05	—	1.05	0.40
Enschede	10	raw water[3]	3	—	—	0.50	0.32	0.05	0.02	0.02	0.02	0.05	—	0.07	0.06
Enschede	10	raw water[2]	3	—	—	0.14	0.12	—	—	0.01	—	0.04	—	—	—
St. Jansteen	11	raw water	3	0.01	—	0.01	—	0.01	—	0.02	—	0.03	—	—	—
Valkenburgse Watering	15	raw water	6	—	—	—	—	—	—	0.01	—	0.03	—	0.75	0.34
Groundwater															
Bilthoven	18	groundwater	2	—	—	—	—	—	—	—	—	—	—	—	—
Coastal waters															
Waddenzee	22	surface water	3	—	—	0.01	—	0.01	—	0.01	—	0.10	—	0.15	0.08
IJsselmeer region															
IJsselmeer, Y-1	26	surface water	2	0.01	—	0.03	0.02	0.04	0.03	0.01	—	0.05	0.02	0.34	0.21
Ketelmeer, Y-14	31	surface water	1	0.02	0.02	0.23	0.23	0.13	0.13	—	—	0.04	0.02	0.63	0.49
IJsselmeer, Y-104	34	surface water	2	0.01	—	0.03	0.03	0.04	0.04	0.01	—	0.01	—	0.24	0.22
Maas and tributaries															
Maas, Eysden	35	surface water	8	0.04	—	0.03	—	0.06	0.02	0.01	—	0.03	—	0.50	0.22
Roer	42	surface water	5	0.02	—	0.02	0.01	0.05	0.04	0.01	—	0.03	—	0.12	0.06
Niers	43	surface water	6	0.01	—	0.06	0.02	0.05	0.03	0.01	—	0.04	—	0.11	0.06
Rhine and tributaries															
Rhine	45	surface water	51	0.39	0.08	0.26	0.14	0.16	0.08	0.04	—	0.40	0.03	4.01	0.72
Kromme Rijn	47	surface water	6	0.02	—	0.15	0.05	0.11	0.05	0.03	—	0.03	—	2.08	0.40
Other surface waters															
Ruiten A	52	surface water	5	0.01	—	—	—	—	—	0.01	—	0.02	—	0.10	0.05
Overijsselse Vecht	60	surface water	4	—	—	—	—	—	—	0.01	—	0.03	—	0.09	—
ditch, A.Paulowna	68	surface water	5	—	—	0.01	—	—	—	0.02	0.01	—	—	0.33	0.10
ditch, Hillegom	69	surface water	2	—	—	0.03	0.02	0.01	—	—	—	—	—	0.22	0.18
ditch, Hillegom	70	surface water	2	—	—	0.04	0.02	0.01	—	0.04	0.04	—	—	0.37	0.18
ditch, Hillegom	71	surface water	2	—	—	0.01	—	—	—	—	—	—	—	0.34	0.17
ditch, Hoogeveen	72	surface water	2	—	—	—	—	—	—	0.02	0.01	—	—	0.32	0.22
Leidse Vaart, Lisse	73	surface water	2	—	—	—	—	0.02	0.02	0.03	0.02	—	—	0.26	0.16
ditch, Noordwijkerhout	74	surface water	2	0.01	—	—	—	—	—	0.08	0.06	—	—	0.21	0.10
Leidse Vaart, De Zilk	75	surface water	2	—	—	0.01	—	0.05	0.02	0.04	0.03	—	—	0.22	0.16
Boomawetering	76	surface water	5	—	—	0.03	0.01	0.04	0.02	0.03	—	0.15	0.01	0.40	0.12
Rijnbeek	82	surface water	6	0.01	—	0.02	—	0.03	0.01	0.01	—	0.05	0.03	0.53	0.10
Lage Vaart, Colijn	85	surface water	5	—	—	0.02	0.01	0.02	0.01	0.02	0.01	0.02	—	0.23	0.06
Hoge Vaart, Colijn	86	surface water	5	0.01	—	0.06	0.02	0.04	0.02	0.02	0.01	0.02	0.01	0.24	
Lage Vaart, De Block van Kuffeler	87	surface water	5	0.04	—	0.02	0.01	0.03	0.02	0.03	0.01	0.04	—	0.11	0.05
ditch, N.O.polder	91	surface water	1	—	—	—	—	—	—	0.07	0.07	0.07	0.07	0.14	0.14
Wortman	92	surface water	5	0.01	0.01	0.03	0.02	0.03	0.03	0.01	—	0.03	—	0.13	0.12

NOTE: See NOTE, Table 1.
[1] As paraoxon-equivalents.
[2] After rapid filtration.
[3] Before infiltration.

TABLE 3. *Concentrations of HCB, BHC, dieldrin, endosulfan, and cholinesterase inhibitors in Dutch water samples, 1971*

Sampling Site	No.	Types of Water	No. of Samples	Residues, ppb											
				HCB		α-BHC		γ-BHC		Dieldrin		α- and β-Endosulfan		Cholinesterase Inhibitors[1]	
				Max	Med	Max	Med	Max	Med	Max	Med	Max	Med	Max	Med
Surface water for drinking water preparation															
IJsselmeer	7	raw water	12	0.01	—	0.13	0.02	0.20	0.03	0.03	—	—	—	0.20	0.08
Surface water for infiltration															
Enschede	10	raw water[2]	5	—	—	0.15	0.06	0.03	0.01	0.02	—	—	—	—	—
Enschede	10	raw water[3]	5	—	—	0.38	0.06	0.02	—	0.03	—	—	—	—	—
Valkenburgse Watering	15	raw water	6	0.01	—	0.03	0.02	0.08	0.02	—	—	0.02	—	1.18	0.40
Groundwater															
Bilthoven	18	groundwater	1	—	—	—	—	—	—	—	—	—	—	—	—
Haarlem	19	groundwater	1	0.01	0.01	—	—	—	—	—	—	—	—	—	—
Hillegom	20	groundwater	1	—	—	—	—	—	—	—	—	—	—	—	—
IJsselmeer region															
Ketelmeer, Y-14	31	surface water	8	0.01	—	0.13	0.05	0.10	0.03	0.03	0.01	0.01	—	0.40	0.19
Ketelhaven	32	surface water	9	0.03	0.02	0.18	0.12	0.14	0.06	0.06	—	0.04	—	1.26	0.33
Maas and tributaries															
Maas, Eysden	35	surface water	10	0.05	0.01	0.01	0.01	0.03	0.01	0.03	—	—	—	0.25	0.08
Maas, Urmond	36	surface water	2	0.02	0.01	—	—	0.01	—	0.05	0.04	0.01	—	0.08	0.04
Maas, Maasbracht	37	surface water	6	0.01	—	0.01	0.01	0.12	0.01	0.02	0.01	—	—	0.20	—
Maas, Kessel	38	surface water	6	0.01	—	0.01	—	0.13	—	0.03	0.02	0.07	—	0.18	—
Roer	42	surface water	6	0.01	—	0.02	0.01	0.02	0.01	0.02	—	0.02	—	0.12	0.08
Niers	43	surface water	5	0.01	—	0.08	0.02	0.03	0.02	0.06	—	0.02	—	0.08	0.08
Rhine and tributaries															
Rhine	45	surface water	52	0.52	0.14	0.48	0.16	0.34	0.10	0.06	—	0.25	—	2.00	0.16
Other surface waters															
Winschoterdiep	51	surface water	5											0.56	0.12
Bagmolenbeek	58	surface water	1	—	—	0.06	0.06	0.02	0.02	—	—	—	—	0.74	0.38
Regge	63	surface water	3											0.14	0.12
Twentekanaal, Almelo	64	surface water	3	—	—	0.01	0.01	0.01	—	0.02	0.02	—	—	0.50	0.46
Twentekanaal, bovenpand	65	surface water	3	—	—	0.13	0.10	0.20	0.01	—	—	—	—	—	—
Lage Vaart, Colijn	85	surface water	6	0.01	—	0.04	0.01	0.02	0.01	0.02	—	—	—	0.40	0.09
Hoge Vaart, Colijn	86	surface water	6	—	—	0.08	0.04	0.05	0.04	0.02	—	—	—	0.46	0.23
Lage Vaart, Wortman	89	surface water	6	—	—	0.03	0.02	0.02	0.01	0.04	—	0.03	—	0.32	—

NOTE: See NOTE, Table 1.
[1] As paraoxon-equivalents.
[2] Before infiltration.
[3] After rapid filtration.

TABLE 4. *Concentrations of HCB, BHC, dieldrin, endosulfan, and cholinesterase inhibitors in Dutch water samples, 1972*

Sampling Site	No.	Types of Water	No. of Samples	Residues, ppb											
				HCB		α-BHC		γ-BHC		Dieldrin		α- and β-Endosulfan		Cholinesterase Inhibitors[1]	
				Max	Med	Max	Med	Max	Med	Max	Med	Max	Med	Max	Med
Surface water for drinking water preparation															
IJsselmeer, Andijk	7	raw water	12	0.01	—	0.05	0.02	0.04	0.02	0.05	—	0.01	—	0.44	0.24
Surface water for infiltration															
Enschede	10	raw water[2]	6	0.01	—	0.17	0.10	0.02	0.02	0.05	—	—	—	—	—
Enschede	10	raw water[3]	6	0.01	—	0.09	0.06	0.01	0.01	—	—	—	—	—	—
Valkenburgse Watering	15	raw water	9	0.03	—	0.04	0.02	0.03	0.02	0.02	—	—	—	3.52	0.76
Rainwater															
Bilthoven	17	rainwater	8	0.01	—	0.50	0.02	0.06	0.02	0.01	—	—	—	—	—
IJsselmeer region															
IJsselmeer, Y-2	27	surface water	11	0.01	—	0.05	0.02	0.05	0.03	0.02	—	—	—	0.42	0.13
IJsselmeer, Y-2	27	surface water[3]	4	0.03	—	0.04	0.03	0.03	0.02	0.02	—	—	—		
IJsselmeer, Y-10	29	surface water	11	0.05	—	0.25	0.03	0.20	0.03	0.03	—	—	—	1.10	0.25
IJsselmeer, Y-10	29	surface water[3]	4	0.02	0.01	0.24	0.06	0.20	0.05	0.02	—	—	—		
IJsselmeer, Y-12	30	surface water	11	0.20	0.01	0.20	0.07	0.20	0.05	0.02	—	—	—	2.44	0.48
IJsselmeer, Y-12	30	surface water[3]	4	0.03	0.01	0.12	0.12	0.13	0.08	0.04	0.02	—	—		
Ketelhaven	32	surface water	11	0.08	0.04	0.20	0.12	0.22	0.10	0.02	—	—	—	1.18	0.76
Ketelhaven	32	surface water[3]	4	0.04	—	0.16	0.09	0.19	0.14	0.02	—	—	—		
IJsselmeer, Y-20	33	surface water	11	0.06	0.01	0.08	0.02	0.08	0.03	0.03	—	—	—	1.94	0.17
IJsselmeer, Y-20	33	surface water[3]	4	0.01	—	0.02	0.02	0.03	0.02	0.05	0.02	—	—		
Maas and tributaries															
Maas, Eijsden	35	surface water	11	0.03	0.01	0.07	0.01	0.07	0.02	0.01	—	—	—	0.44	—
Maas, Grave	39	surface water	12	0.02	—	0.08	0.01	0.13	0.02	0.01	—	0.01	—	0.14	—
Maas, Keizersveer	41	surface water	12	0.01	—	0.06	0.01	0.18	0.02	0.02	—	0.01	—	0.16	0.07
Roer	42	surface water	12	0.01	—	0.09	—	0.04	0.02	0.02	—	0.01	—	0.50	—
Niers	43	surface water	12	0.08	0.01	0.15	0.08	0.06	0.04	0.08	—	0.09	—	0.12	—
Dieze	44	surface water	10	0.05	—	0.06	0.01	0.07	0.03	0.02	—	0.02	—	0.32	0.14
Rhine and tributaries															
Rhine	45	surface water	52	0.37	0.13	0.57	0.16	0.28	0.11	0.02	—	0.03	—	2.36	0.73
Other surface waters															
Zuidlaardermeer	53	surface water	6	0.01	—	0.01	—	0.01	—	0.01	—	—	—	0.12	—
Lauwersmeer	54	surface water	6	0.03	—	0.01	—	0.01	—	0.01	—	—	—	—	—
Van Starkenborghkanaal	55	surface water	6	0.03	—	0.02	—	0.02	0.01	0.01	—	—	—	—	—
Meppelerdiep	56	surface water	6	0.02	—	0.02	—	0.02	—	0.01	—	—	—	—	—
Regge, bovenloop	61	surface water	3											0.22	0.07
Regge, benedenloop	62	surface water	3	0.02	0.02	0.11	0.06	0.05	0.04	0.01	—	—	—	0.11	—
Twentekanaal, bovenpand	65	surface water	2	—	—	0.44	0.22	0.02	0.01	0.01	—	—	—	0.06	—
Eem	66	surface water	6	0.01	—	0.08	0.05	0.06	0.04	0.01	—	—	—	1.72	0.17
Vecht	67	surface water	6	0.01	—	0.06	0.03	0.05	0.02	—	—	—	—	0.64	0.06
Lage Vaart, Colijn	85	surface water	7	0.01	—	0.01	—	0.02	0.01	0.03	—	—	—	0.06	—
Hoge Vaart, Colijn	86	surface water	7	0.01	—	0.08	0.02	0.09	0.01	0.01	—	—	—	0.52	0.06
Lage Vaart, De Block van Kuffeler	87	surface water	7	0.03	—	0.01	—	0.02	—	—	—	—	—	—	—
Hoge Vaart, De Block van Kuffeler	88	surface water	7	0.01	—	0.03	0.01	0.04	0.02	0.01	—	—	—	0.14	—
Lage Vaart, Wortman	89	surface water	7	0.01	—	0.01	0.01	0.02	0.02	0.01	—	—	—	0.36	

NOTE: See NOTE, Table 1.
[1] As paraoxon-equivalents.
[2] Before infiltration.
[3] After rapid filtration.

TABLE 5. *Concentrations of HCB, BHC, dieldrin, endosulfan, and cholinesterase inhibitors in Dutch water samples, 1973.*

Sampling Site	No.	Types of Water	No. of Samples	Residues, ppb											
				HCB		α-BHC		γ-BHC		Dieldrin		α- and β-Endosulfan		Cholinesterase Inhibitors[1]	
				Max	Med	Max	Med	Max	Med	Max	Med	Max	Med	Max	Med
Surface water for drinking water preparation															
IJsselmeer, Andijk	7	raw water	9	0.01	—	0.10	0.03	0.08	0.03	—	—	0.01	—	1.10	0.12
Surface water for infiltration															
Enschede	10	raw water[2]	6	—	—	0.09	0.04	—	—	—	—	—	—	1.90	—
Rainwater															
Bilthoven	17	rainwater	13	—	—	0.03	0.01	0.05	0.02	0.02	—	—	—		
Coastal waters															
Bocht van Watum	21	surface water	3	—	—	0.01	—	0.05	0.01	—	—	—	—	0.06	—
Westerschelde, Schaar van Ouden Doel	23	surface water	13	0.03	0.01	0.10	0.01	0.12	0.04	0.01	—	—	—	0.68	0.32
Westerschelde, Hansweert	24	surface water	13	0.07	—	0.03	—	0.14	0.03	0.01	—	—	—	0.60	0.08
IJsselmeer region															
IJsselmeer, Y-2	27	surface water	12	0.01	—	0.14	0.04	0.13	0.03	—	—	—	—	2.64	0.20
IJsselmeer, Y-10	29	surface water	13	0.01	—	0.10	0.05	0.06	0.04	—	—	—	—	1.65	0.50
Ketelhaven	32	surface water	11	0.08	0.02	0.23	0.10	0.19	0.09	—	—	0.07	—	5.10	1.88
Maas and tributaries															
Maas, Eijsden	35	surface water	13	0.29	0.01	0.02	0.01	0.05	0.01	—	—	0.01	—	1.65	0.06
Maas, Grave	39	surface water	12	0.03	0.01	0.19	0.02	0.12	0.02	—	—	—	—	1.26	—
Maas, Keizersveer	41	surface water	13	0.04	0.01	0.02	0.01	0.06	0.02	—	—	—	—	1.62	0.06
Roer	42	surface water	1									—	—		
Niers	43	surface water	1									—	—		
Dieze	44	surface water	1									—	—		
Rhine and tributaries															
Rhine	45	surface water	52	0.55	0.08	0.45	0.19	0.42	0.12	0.02	—	0.10	—	15.80	2.42
Boven Merwede	48	surface water	24	0.10	0.03	0.36	0.15	0.23	0.11	0.01	—	0.02	—	4.40	1.46
Nieuwe Waterweg	50	surface water	13	0.06	0.02	0.35	0.13	0.21	0.09	0.01	—	0.01	—	4.45	1.24
Other surface waters															
Twentekanaal, Almelo	64	surface water	5	—	—	0.03	0.01	0.01	0.01	—	—	0.01	—	0.60	—
Twentekanaal, bovenpand															
Roosendaalse Vliet	81	surface water	1	0.01	—	—	—	—	—	—	—	—	—	0.18	0.06
Hoge Vaart, Colijn	86	surface water	6	0.01	—	0.09	—	0.07	—	—	—	0.01	—	1.16	0.25
Hoge Vaart, De Block van Kuffeler	88	surface water	6	0.01	—	0.02	—	0.01	—	—	—	0.01	—	—	—

NOTE: See NOTE, Table 1.
[1] As paraoxon-equivalents.
[2] After rapid filtration.

TABLE 6. *Concentrations of HCB, BHC, dieldrin, endosulfan, cholinesterase inhibitors, and aromatic amines in Dutch water sample, 1974*

Sampling Site	No.	Types of Water	No. of Samples	Residues, ppb: HCB Max	HCB Med	α-BHC Max	α-BHC Med	γ-BHC Max	γ-BHC Med	Dieldrin Max	Dieldrin Med	α- and β-Endosulfan Max	α- and β-Endosulfan Med	Cholinesterase Inhibitors[1] Max	Cholinesterase Inhibitors[1] Med	Aromatic Amines[2] Max	Aromatic Amines[2] Med
Surface water for drinking water preparation																	
Enschede	10	surface water[3]	5	0.01	—	0.15	0.07	0.02	—								
Isabella Wetering	12	surface water	5	—	—	0.01	—	0.03	0.01	—	—	—	—	—	—	—	—
Pieters v.d.Endevaart	13	surface water	6	—	—	0.01	—	0.01	—	0.01	—	—	—	—	—	—	—
canal near Valkenburg	14	raw water	6	—	—	0.11	0.06	0.08	0.04					1.32	0.05	1.0	—
Wijde A	16	raw water	6	—	—	0.22	0.07	0.09	0.05					1.54	0.26	1.8	0.5
Rainwater																	
Bilthoven	17	rainwater	12	—	—	0.08	0.02	0.10	0.04	—	—						
Coastal waters																	
Bocht van Watum	21	surface water	5	—	—	0.01	0.01	0.05	0.01					1.60	—	0.8	—
IJsselmeer region																	
IJsselmeer, Y-10	29	surface water	12	0.01	—	0.12	0.06	0.10	0.04					1.36	0.70	4.6	0.8
Ketelhaven	32	surface water	11	0.09	0.04	0.57	0.14	0.26	0.07					3.34	0.56	15	3.4
Maas and tributaries																	
Maas, Eijsden	35	surface water	12	0.05	0.01	0.02	0.01	0.04	0.02					0.50	—	0.8	—
Maas, Keizersveer	41	surface water	13	0.02	—	0.03	0.01	0.05	0.02					1.64	0.12	1.0	—
Rhine and tributaries																	
Rhine	45	surface water	50	0.39	0.10	0.60	0.22	0.33	0.13	0.05	—	0.02	—	3.64	1.36	8.6	4.5
Boven Merwede	48	surface water	12	0.12	0.06	0.55	0.28	0.26	0.12					3.36	0.79	16	3.8
Hollandse IJssel	49	surface water	6	0.01	—	0.10	0.03	0.05	0.04					1.16	0.09	1.0	0.6
Nieuwe Waterweg	50	surface water	11	0.05	0.03	0.36	0.21	0.23	0.11					2.40	0.60	5.8	2.6
Other surface waters																	
Twentekanaal, bovenpand	65	surface water	7	0.05	—	2.1	0.58	0.12	0.05					0.83	—	—	—
ditch, Ouddorp	78	surface water	6	0.01	—	0.01	—	0.01	—					1.60	—	3.7	—
Gentse Vaart	79	surface water	6	0.01	—	0.01	—	0.02	0.01	—	—			0.12	—	0.7	—
Roosendaalse Vliet	81	surface water	6	—	—	0.01	—	0.02	0.01					0.05	—	—	—
Zwarte Water I	83	surface water	6	—	—	0.01	0.01	0.16	0.01	0.06	—			0.10	—	8.1	—
Zwarte Water II	84	surface water	6	—	—	0.09	0.02	0.04	0.02	—	—			—	—	3.0	0.6
Hoge Vaart, Colijn	86	surface water	6	0.02	—	0.14	0.01	0.10	—					0.58	—	4.4	0.7
Hoge Vaart, De Block van Kuffeler	88	surface water	6	0.01	—	0.02	0.01	0.03	—					—	—	1.0	

NOTE: See NOTE, Table 1.
[1] As paraoxon-equivalents.
[2] As 3,4-dichloroaniline-equivalents.
[3] Before infiltration.

TABLE 7. *Concentrations of HCB, BHC, dieldrin, endosulfan, cholinesterase inhibitors, and aromatic amines in Dutch water samples, 1975*

Sampling Site	No.	Types of Water	No. of Samples	Residues, ppb: HCB Max	HCB Med	α-BHC Max	α-BHC Med	γ-BHC Max	γ-BHC Med	Dieldrin Max	Dieldrin Med	α- and β-Endosulfan Max	α- and β-Endosulfan Med	Cholinesterase Inhibitors[1] Max	Cholinesterase Inhibitors[1] Med	Aromatic Amines[2] Max	Aromatic Amines[2] Med
Rainwater																	
Bilthoven	17	rainwater	10	0.01	—	0.03	0.02	0.04	0.03	—	—						
Maas and tributaries																	
Maas, Eijsden	35	surface water	13	0.02	—	0.01	0.01	0.03	0.02					0.44	—	1.5	0.6
Maas, Lith	40	surface water	13	0.02	—	0.03	0.01	0.07	0.02					0.18	—	2.4	0.7
Rhine and tributaries																	
Rhine	45	surface water	44	0.21	0.06	0.21	0.06	0.14	0.04	0.02	—	0.02	—	56.0	7.80	10	3.7
IJssel	46	surface water	11	0.06	0.03	0.09	0.03	0.06	0.03					21.0	8.70	14	2.8
Boven Merwede	48	surface water	12	0.10	0.03	0.13	0.05	0.07	0.03					18.0	7.20	9.5	3.8
Nieuwe Waterweg	50	surface water	12	0.02	0.01	0.09	0.04	0.09	0.03					10.0	6.00	4.0	2.6
Other surface waters																	
Overijsselse Vecht	59	surface water	6											0.12	—		
Twentekanaal, Almelo	64	surface water	5	—	—	0.06	0.04	0.04	0.02					0.04	—	1.9	0.7
Twentekanaal, bovenpand	65	surface water	6	0.01	—	1.40	0.47	0.04	0.04					2.10	—	1.0	0.6
polder ditch	77	surface water	6	—	—	0.02	—	0.01	—					—	—	0.5	—
Grote Kreek	80	surface water	6	—	—	0.02	0.01	0.02	0.01	—	—			0.34	—	0.7	—
Zwarte Water I	83	surface water	6	0.01	—	0.01	—	0.02	0.01	—	—			—	—	1.4	—
Zwarte Water II	84	surface water	6	0.08	—	0.30	0.04	0.09	0.02	—	—			0.14	—	6.8	2.3

NOTE: See NOTE, Table 1.
[1] As paraoxon-equivalents.
[2] As 3,4-dichloroaniline-equivalents.

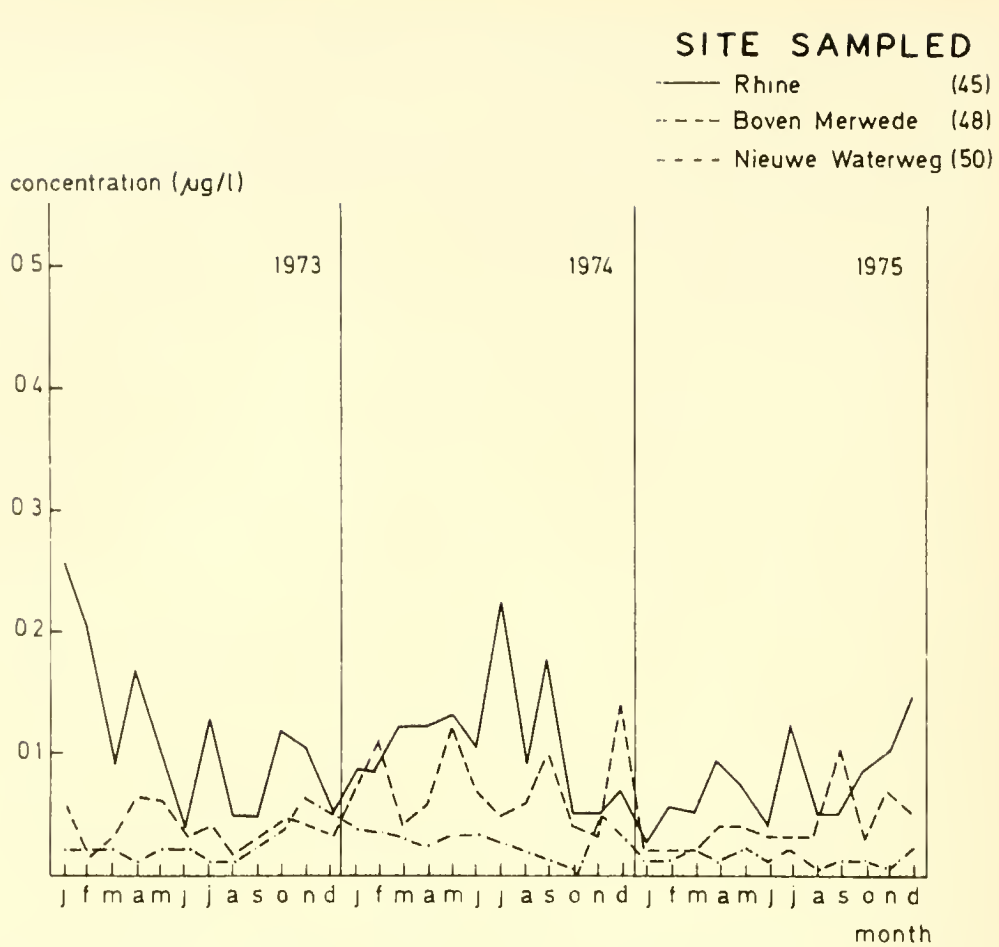

FIGURE 2. *Concentrations of HCB in the southern region of the Rhine River (sites 45, 48, and 50 in Fig. 1)*

High concentrations of α-BHC were also found in the Twentekanaal. The source of the contamination was a chemical plant which produces γ-BHC. The α-BHC, a worthless by-product of the synthesis of γ-BHC, was dumped beside the canal. Removal of the dumped material led to a gradual decrease of concentrations in the canal and in drinking water removed from canal water.

Concentrations of HCB have also decreased, but gradually and less drastically (Figs. 2–4). HCB is a low-polarity compound which is volatile with water and readily adsorbed by the solid particles which settle in fluvial transport.

Concentrations of cholinesterase inhibitors have gradually increased since 1972 and significantly in 1975. Concentrations of α- and β-endosulfan have decreased greatly following the first sensational wave in June–July 1969 (*9*) and a second, less important one in autumn of the same year.

In Table 8, maximum and median or mean concentrations of α-BHC, γ-BHC, ΣBHC, dieldrin, ΣDDT, and DDE from nine nations are summarized (*1, 2, 4, 12–23*). Levels of α- and γ-BHC, ΣBHC, dieldrin, ΣDDT, and DDE in Dutch surface waters are of the same order of magnitude as are the concentrations in other industrialized countries. Concentrations of aromatic amines are comparable in Dutch and German parts of the Rhine River (*11*).

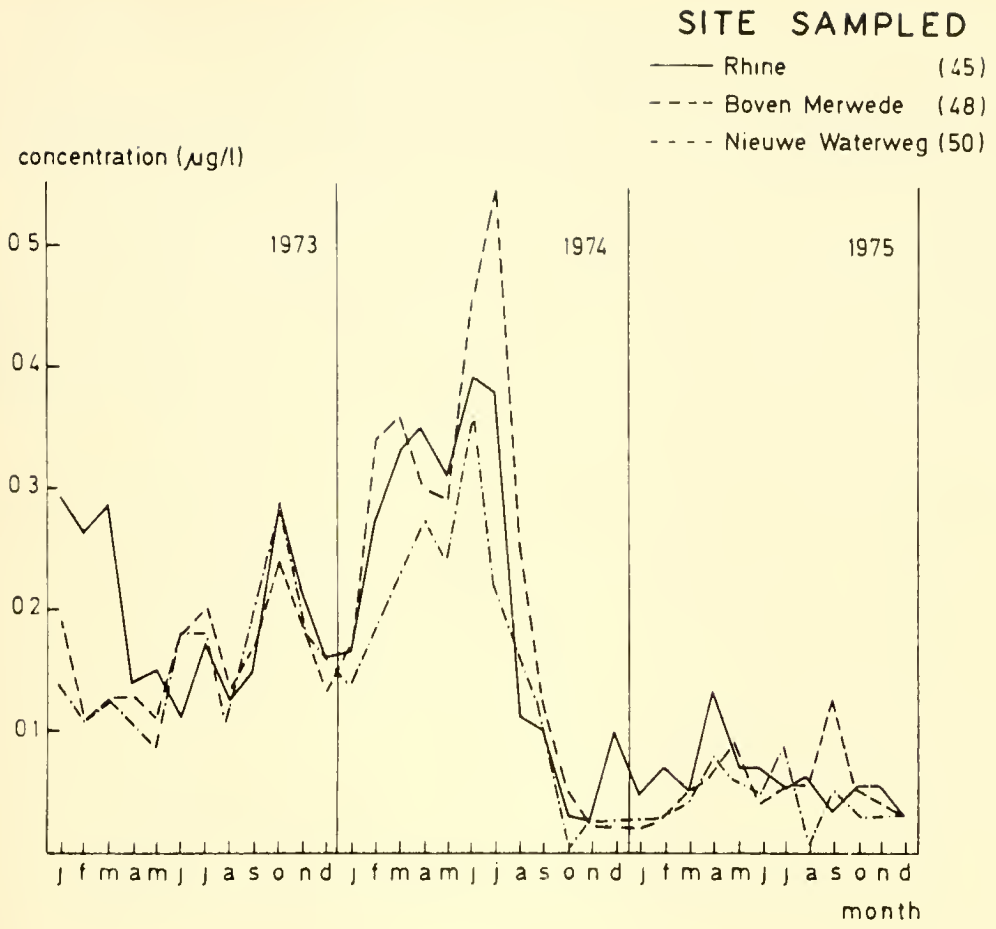

FIGURE 3. *Concentrations of α-BHC in the southern region of the Rhine River (sites 45, 48, and 50 in Fig. 1)*

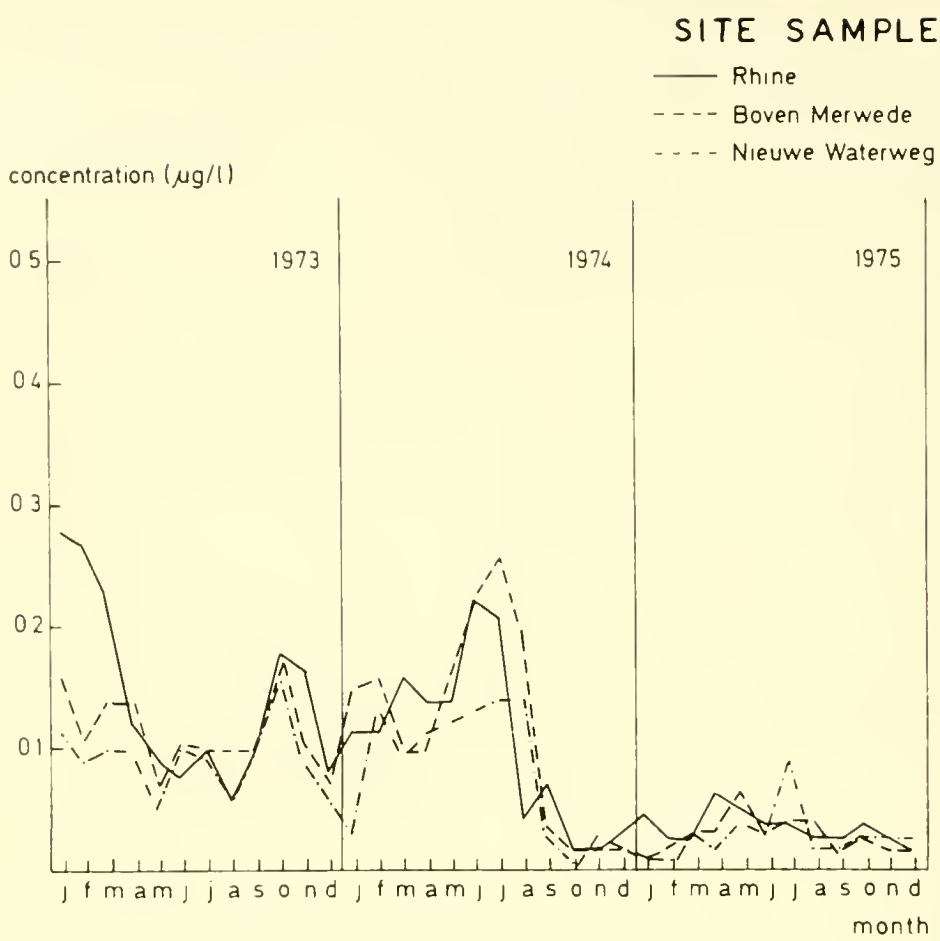

FIGURE 4. *Concentrations of γ-BHC in the southern region of the Rhine River (sites 45, 48, and 50 in Fig. 1)*

TABLE 8. *Concentrations of organochlorine pesticides in worldwide surface waters, 1968–75*

Location	No. of Sites	Types of Water	Residues, ppb												Literature References
			α-BHC		γ-BHC		ΣBHC		Dieldrin		DDT		DDE		
			Max	Med	Max	Med	Max	Med	Max	Med	Max	Med	Max	Med	
Brazil	9	surface water					4	<1			<1	<1			Lara and Barreto, 1972 (*15*)
Canada	3	surface water	—	—[1]	—	—[1]			0.04	0.01[1]	0.07	0.01[1]	0.01	—[1]	Miles and Harris, 1973 (*16*)
Czechoslovakia	150	surface water 1971–72	0.52		0.81						0.60				Uhnáh et al., 1974 (*23*)
Federal Republic of Germany (FRG)	8	surface water 1970	1.90	0.10[1]	7.10	0.10[1]			0.04	—[1]	0.25	—[1]			Herzel, 1972 (*12*)
	27	surface water 1971	2.40	0.07[1]	1.75	0.17[1]			—	—[1]	0.84	—[1]			Herzel, 1972 (*12*)
German Democratic Republic (GDR)	26	surface water			0.67	0.15					3.2	0.34	0.98	0.15	Engst and Knoll, 1973 (*4*)
Japan	130	river water 1970–73			3.43	0.20	14.15	0.92							Suzuki et al., 1974 (*21*)
Hungary	4	Balaton Lake 1973			0.04	0.04[1]					0.01	—[1]	—[1]	—[1]	Pásztor et al., 1975 (*18*)
Netherlands	16	surface water 1969					0.24	—[1]	0.14	—[1]	0.20	—[1]	0.16	—[1]	the present report
	26	surface water 1970	0.50	0.03[1]	0.20	0.05[1]			0.08	—	0.11	—[1]	—	—[1]	
	17	surface water 1971	0.48	0.04[1]	0.34	0.03[1]			0.06	—[1]	0.11	—[1]	—	—[1]	
	26	surface water 1972	0.57	0.04[1]	0.28	0.03[1]			0.08	—[1]	0.17	—[1]	0.15	—[1]	
	21	surface water 1973	0.45	0.07	0.42	0.04[1]			0.02	—	0.11	—	0.01	—	
	17	surface water 1974	0.60	0.17	0.33	0.07			0.06	—	0.04	—	0.01	—	
	13	surface water 1975	1.40	0 03	0.14	0.03			0.02	—	0.03	—	0.01	—	
United States of America		Utah Lake 1970–71					1.3				4.1				Bradshaw et al., 1972 (*1*)
	1	Mississippi River 1974			—	—			0.01	—					Brodtmann, 1976 (*2*)
	6	Iowa Rivers 1968							0.01	—	0.01	—	0.01	—	Johnson and Morris, 1971 (*13*)
	10	Iowa Rivers 1969							0.06	—	0.01	—	0.01	—	
	10	Iowa Rivers 1970							0.06	—	0.02	—	0.02	—	
	1	Des Moines River Iowa 1971							0.05	0.03					Kellog and Bulkley, 1976 (*14*)
		Iowa 1972							0.04	—					
		Iowa 1973							0.02	0.01					
	10	Iowa rivers 1968							0.01		0.01		0.01		Morris et al., 1972 (*17*)
	10	Iowa rivers 1969							0.06		0.02		0.01		
	10	Iowa rivers 1970							0.06		0.02		0.02		
	10	Iowa rivers 1971							0.04		0.22		0.03		
	19	surface water 1974							0.07				3.92		Richard et al., 1975 (*19*)
	20	rivers 1968			0.07				0.03		0.46		0.10		Schulze et al., 1973 (*20*)
		rivers 1969			0.04				0.02		0.05		0.06		
		rivers 1970			0.16				0.02		0.09		0.05		
		rivers 1971			0.05				0.01		0.09		0.08		
	4	streams 1969							0.33	—	2.50	0.01	0.71	—	Truhlar and Reed, 1975 (*22*)
	4	streams 1970							0.16	—	11.0	0.02	0.21	—	
	4	streams 1971							—	—	0.12	—	0.05	—	

[1] Mean value.

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BRIEF

Organochlorine Pesticide Levels in Ottawa Drinking Water, 1976

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ABSTRACT

Duplicate samples of Ottawa drinking water were collected once a month during 1976 and analyzed for organochlorine pesticides. The samples were analyzed by gas chromatography–mass spectrometry, and pesticides were identified by comparing their retention times, coupled with selected ion monitoring, with those of known standards. The pesticides detected and their mean concentrations in parts per trillion were aldrin (0.9), heptachlor epoxide (3), heptachlor (0.6), α-BHC (6), γ-BHC (3), endrin (4), dieldrin (1), o,p'-TDE (1), o,p'-DDT (3), and o,p'-DDE (0.2).

Introduction

Ottawa drinking water was monitored for organochlorine pesticides by a simple new method using Amberlite XAD-2 macroreticular resin for the analysis of potable water at the parts per trillion (ppt) level.

Sampling and Analysis

In 1976, duplicate 200-liter samples per month, except July, of Ottawa drinking water was passed through Amberlite XAD-2 macroreticular resin during a 10-day period according to the procedure of McNeil et al. (*1*). The resin was eluted with 250 ml hexane, and the eluates were dried with sodium sulfate and concentrated to 1 ml. The concentrated hexane eluates were then analyzed with a Finnigan Model 4000 gas chromatograph–mass spectrometer coupled to a Model 6000 Data System with the following instrument parameters and operating conditions:

Column:	1.8 m × 2 mm ID glass, packed with 3 percent OV-17 on 80–100-mesh Chromosorb 750
Temperatures:	oven from 200°C (0.1 minute hold) to 250°C (hold) at 5°C/minute; injection port 225°C
Carrier gas:	helium flowing at 25 ml/minute

The mass spectrometer, operating in the selected ion mode, was programmed to monitor four ions (m/q 66, 81, 100, 109) for the first 4 minutes and four other ions (m/q 67, 79, 235, 246) for 10 minutes more. Analyses were performed on a standard pesticide mixture, including the 10 pesticides detected under identical GC–MS conditions to permit identification and quantitation. The lower limit of detection was about 0.1 ppt of pesticide in the original 200-liter water sample.

Results and Discussion

Results of the pesticide analyses are presented in condensed form in Table 1, including the relative retention time and specific ion monitored for each pesticide.

There was no consistent seasonal trend for any of the 10 pesticides detected. The monthly pesticide values varied considerably with the mean as shown by the high standard deviations in Table 1. This is expected since the levels of many of the pesticides were close to the detection limit, and the use of selected ion monitoring, although more selective than simple gas chromatography, is still subject to interference, particularly at the trace levels found.

Authors concluded that organochlorine pesticides detected in Ottawa drinking water exist as background levels which are consistently present in trace amounts in the environment.

TABLE 1. *Organochlorine pesticide residue levels in Ottawa drinking water, 1976*

Pesticide	Selected Ion Monitored	Relative Retention Time	Range min.–max., ppt	Mean ± Std Dev.	Median
α-BHC	109	1.00	0.1–15	6±4	6
γ-BHC	109	1.30	0.4–11	3±3	2
Heptachlor	100	1.63	0.1–1	0.6±0.3	0.7
Aldrin	66	1.97	0.1–6	0.9±1	0.5
Heptachlor epoxide	81	2.70	0.2–9	3±3	1
o,p'-DDE	246	3.72	0.1–0.5	0.2±0.2	0.2
Dieldrin	79	3.77	0.1–4	1±1	0.7
o,p'-TDE	235	4.17	0.1–3	1±1	0.8
Endrin	67	4.35	1–7	4±4	4
o,p'-DDT	235	4.72	0.2–8	3±3	2

LITERATURE CITED

(*1*) *McNeil, E. E., R. Otson, W. F. Miles, and F. J. M. Rajabalee. 1977.* Determination of chlorinated pesticides in potable water. J. Chromatogr. 132(2):277–286.

[1] Bureau of Chemical Hazards, Environmental Health Directorate, Health and Welfare Canada, Ottawa, Ontario, Canada K1A OL2.

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 1254	PCB, approximately 54% chlorine
AROCLOR 1260	PCB, approximately 60% chlorine
BHC (BENZENE HEXACHLORIDE)	1,2,3,4,5,6-Hexachlorocyclohexane (mixture of isomers)
CARBARYL	1-Naphthyl *N*-methylcarbamate
CHLORDANE	1,2,3,4,5,6,7,8,8-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]
DICHLORVOS	2,2-Dichlorovinyl dimethyl phosphate
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	Hexachloroepoxyoctahydro-*endo*, *endo*-dimethanonaphthalene
HCB	Hexachlorobenzene
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
ISODRIN	Hexachlorohexahydro-*exo,exo*-dimethanonaphthalene
MALATHION	S-[1,2-Bis(ethoxycarbonyl) ethyl] *O,O*-dimethyl phosphorodithioate
MIREX	1,1a,2,2,3,3a,4,5,5,5a,5b,6-Dodecachlorooctahydro-1,3,4-metheno-1*H*-cyclobuta[cd]pentalene
NONACHLOR	1,2,3,4,5,6,7,8-Nonachlor-3a,4,7,7a-tetrahydro-4,7-methanoindan
PARATHION	*O,O*-Diethyl *O*-*p*-nitrophenyl phosphorothioate
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)
TELODRIN	Octachlorohexahydro-4,7-methanoisobenzofuran
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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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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FOOD AND FEED

Acephate and Methamidophos Residue Behavior in Florida Citrus, 1976 [1]

Herbert N. Nigg,[2] James A. Reinert,[3] and George E. Fitzpatrick[3]

ABSTRACT

The half-life of acephate and its hydrolysate, methamidophos, in the rind of Temple and Valencia oranges, and grapefruit, lemons, and tangerines was 10.3 days and 10.5 days, respectively. Half-lives of acephate and methamidophos in citrus pulp were 15.0 days and 6.1 days, respectively, based on 7-, 14-, and 21-day data. Seven days after treatment, acephate and methamidophos reached maximum levels in rind and pulp. Acephate residue levels in rind were less than 3.0 ppm 14 days after treatment; acephate residues in pulp were less than 3.0 ppm throughout the experiment. Methamidophos residue levels averaged less than 0.25 ppm after 21 days.

Introduction

In 1937, the citrus blackfly, *Aleurocanthus woglumi* Ashby (Homoptera: Aleyrodidae), was eradicated from the Florida Keys by use of petroleum oil (*7*). Early in 1976, the citrus blackfly was again discovered in Fort Lauderdale, Florida, and surrounding Broward County (*8*). Infestations are currently found in Broward, Collier, Dade, Indian River, Martin, Okeechobee, Palm Beach, and Saint Lucie Counties (G. E. Fitzpatrick, University of Florida Institute of Food and Agricultural Sciences, October 1978: personal communication). After discovery of the infestations, an intensive state and federally sponsored eradication program was begun, but it was complicated by the urban nature of the citrus blackfly infestation.

Based on chemical efficacy and citrus blackfly life-cycle data, three treatments of acephate at 3-week intervals were necessary for eradication (*8*). Treatments were applied to all Florida citrus owned by individual homeowners in the heavily urbanized area under an emergency exemption granted by the United States Environmental Protection Agency (EPA). The homeowner was advised by the Florida and U.S. Departments of Agriculture to wait 7 days before consuming treated fruit.

It was not known whether acephate and its environmental metabolite, methamidophos (Monitor), would reach their respective action levels of 3.0 ppm and 0.25 ppm in whole fruit within 7 days. In addition, acephate and methamidophos are systemic chemicals and might readily penetrate fruit rind into the edible pulp.

The purpose of the present study was to monitor levels of acephate and methamidophos in common Florida citrus to determine half-lives and tolerances of these materials.

Materials and Methods

Each experimental unit consisted of one city block. Within each city block, a random 8-fruit sample was taken from 3–10 trees of Temple and Valencia oranges, and grapefruit, lemons, and tangerines on each sample date. Treatments were replicated four times in a completely random design including four unsprayed check blocks. Acephate at 0.6 g active ingredient(AI)/liter (ca 38 liters/tree) was applied with a hydraulic sprayer at 29 kg/cm^2 and with a mist blower at 2.4 g AI/liter (ca 0.8 liter/tree). The hydraulic sprayer was a standard, truck-mounted unit with two 100-m hoses and attached handheld sprayguns. The mist blower was a gasoline-driven backpack unit (KWH Whirlwind, Holland). Three separate treatments were applied at 3-week intervals. Dual samples of each variety were taken after the third application on days 1, 3, 5, and 7, and single samples of each variety were taken on days 14 and 21 by clipping the fruit into plastic bags. Each sample consisted of eight fruits. One set of the dual samples was washed in a weak soap solution of Ivory liquid to simulate homeowner washing. Samples were frozen at −20°C and transported frozen to the laboratory for analysis.

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Samples were stored approximately one month at −10°C prior to analysis. Valencia oranges were 0.8 mature when harvested; other varieties were completely mature.

The method of Leary (*4*) was modified for extraction and analyses of acephate and methamidophos. Fruits were thawed, the rind was removed from one half of each fruit, and the pulp was sliced into a Waring blender. The pulp was blended for 3 minutes, and a 10-g subsample was removed for analysis. The rind was diced, blended for 3 minutes, and a 10-g subsample was taken for analysis. Separate fruit knives were used for all operations, and between samples all equipment was washed thoroughly with hot soapy water, rinsed in tap water, deionized water, isopropanol, and again in deionized water.

The 10-g sample of either rind or pulp was homogenized in 100 ml ethyl acetate and 15 g sodium sulfate for 5 minutes in a Sorvall blending cup in an ice bath. The blender cup top was loosened upon removal from the mixer, and particulate matter was allowed to settle for 1 minute. A 20-ml aliquot was evaporated to dryness under a nitrogen atmosphere at 40°C, and transferred to brown glass bottles over sodium sulfate in 10 ml methyl isobutyl ketone (MIBK) for gas chromatographic (GC) analysis. No further cleanup was performed on the extractions, and they were stored at −20°C until analysis. The effect of storage on the hydrolysis of acephate to methamidophos was not determined.

For acephate and methamidophos, GC was conducted on a Hewlett-Packard Model 5730A gas chromatograph equipped with dual nitrogen-phosphorus detectors. Instrument parameters and operating conditions follow:

Column:	glass, 30 inches long × 1/8-inch ID, packed with 1 percent Reoplex 400 on 80–100-mesh Gas-Chrom Q
Temperatures, °C:	detector 300
	injector 210
	program 150–200 at 8°/minute, 8-minute final hold, 45-second delay after injection
Carrier gas:	helium flowing at 30 ml/minute

Compounds were quantified by comparing peak heights of standard materials chromatographed at the same attenuation. Unsprayed fruit extracts fortified with standard acephate and methamidophos (Chevron Chemical Co., Richmond, Virginia) were linear at each GC attenuation setting. However, at the attenuation setting of 8, standard materials chromatographed in MIBK alone produced a 10–20 percent lower response than in fortified fruit extracts. Fruit extracts alone were blank, apparently as a result of an unexpected synergistic effect of some component in the fruit extract on the nitrogen–phosphorus detector response. Consequently, fruit extracts fortified with acephate and methamidophos were used for quantification. Standards were chromatographed every fourth injection. All injections were 5 μl.

All solvents were assessed for interferences by evaporating 100 ml of each solvent to 1 ml and chromatographing 5 μl.

Recoveries of standard materials from fortified homogenates were 73.1 percent methamidophos and 77.8 percent acephate at 1 ppm and 82.6 percent methamidophos and 85.4 percent acephate at 5 ppm for both peel and pulp. There were no varietal differences in recovery of standard materials. Variations in recovery averaged 16.8 percent for methamidophos and 15.3 percent for acephate at 1 ppm and 4.6 percent for methamidophos and 5.5 percent for acephate at 5 ppm. Lowest accurate level of detection for both standards was 0.01 ppm; lower levels are reported as trace. The data in Tables 1 and 2 are not corrected for recovery. No analyses were performed on either the formulated acephate or tank mixes. The equation for decay was:

$$y_t = y_o e^{-bt} \quad (1)$$

or

$$\ln(y_t/y_o) = -bt \quad (2)$$

Half-life, $t_{1/2}$, was calculated as

$$t_{1/2} = \ln(0.5)/(-b) \quad (3)$$

Residue levels were compared among varieties on individual sampling days and among sampling days for individual varieties with a t-test (*10*). Degrees of freedom were 14 for days 1–7 and 6 for days 14 and 21 [$df = 2(n - 1)$] (*10*). Comparison of residue levels are significant at the 0.01 level.

Results and Discussion

There was no statistical difference between residues of acephate and methamidophos on or in washed and un-

TABLE 1. *Acephate residues in rind and pulp of Florida citrus, 1976*

	Day, Post Application					
	1	3	5	7	14	21
	Residues (Mean ± Std Dev.), ppm					
Temple orange						
Rind	2.3 ± 0.7	4.6 ± 1.6	2.8 ± 2.8	7.9 ± 5.8	2.6 ± 1.8	2.0 ± 1.2
Pulp	1.3 ± 0.5	0.8 ± tr	0.8 ± 0.5	2.6 ± 1.3	1.6 ± 0.8	1.2 ± 2.0
Grapefruit						
Rind	2.7 ± 2.1	1.9 ± 1.1	2.3 ± 2.1	3.9 ± 2.5	1.4 ± 0.2	1.9 ± 0.8
Pulp	0.3 ± 0.2	0.4 ± 0.3	0.4 ± 0.4	0.9 ± 0.6	0.5 ± tr	0.3 ± 0.3
Valencia orange						
Rind	3.9 ± 1.5	3.1 ± 2.5	4.1 ± 2.8	4.2 ± 1.5	1.8 ± 1.2	2.1 ± 1.3
Pulp	1.1 ± 0.7	0.6 ± 0.8	1.4 ± 0.5	0.7 ± 0.5	0.8 ± 0.3	1.0 ± 1.0
Lemon						
Rind	3.8 ± 2.2	5.7 ± 4.1	2.9 ± 1.8	6.2 ± 2.9	2.6 ± 1.8	1.4 ± 1.0
Pulp	1.5 ± 0.7	1.9 ± 1.0	1.0 ± 0.6	2.4 ± 1.6	1.3 ± 1.2	1.4 ± 0.9
Tangerine						
Rind	3.8 ± 2.3	4.9 ± 3.8	4.9 ± 2.1	4.9 ± 1.8	2.1 ± 1.2	4.9 ± 4.6
Pulp	0.7 ± 0.3	2.0 ± 1.3	1.3 ± 1.1	2.0 ± 0.7	0.9 ± 0.4	1.0 ± 0.6

NOTE: tr = trace = <0.01 ppm.

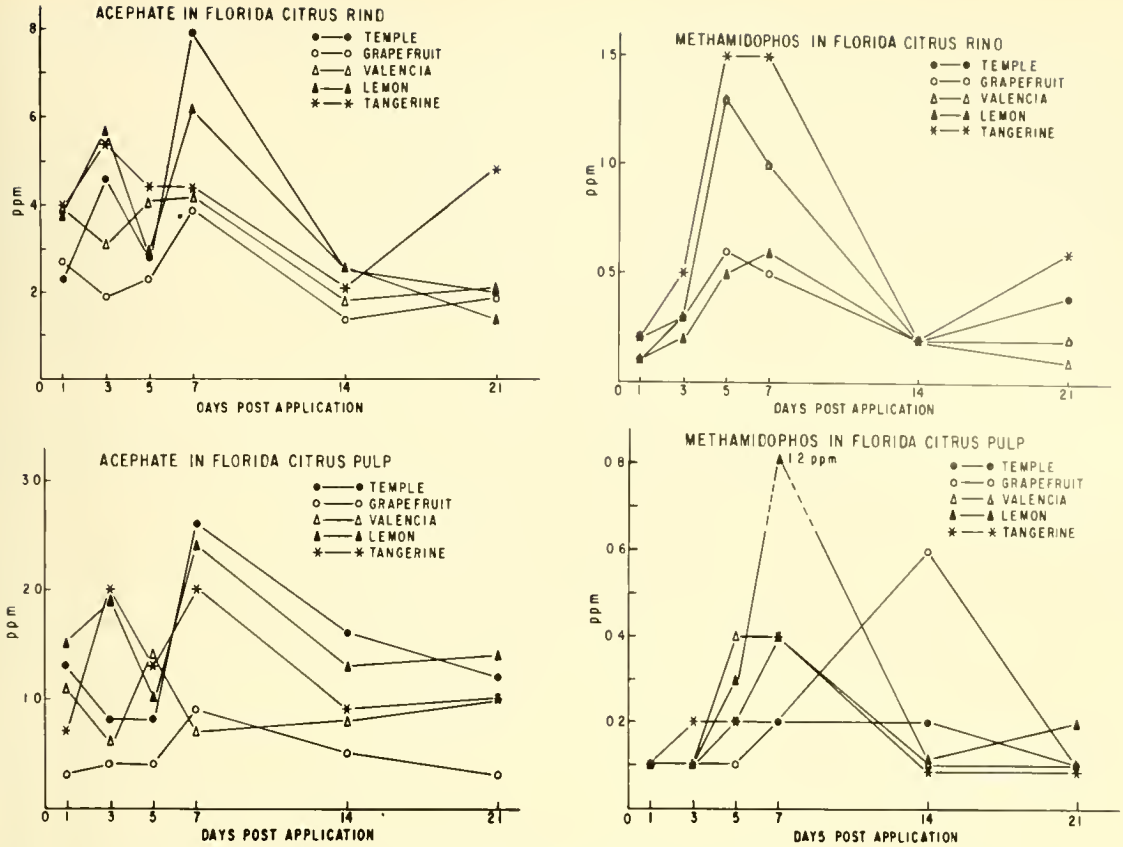

FIGURE 1. *Acephate and methamidophos residue in rind and pulp of Florida citrus. Points for days 1, 3, 5, and 7 are averages of eight determinations. Days 14 and 21 are averages of four determinations.*

TABLE 2. *Methamidophos residues in rind and pulp of Florida citrus, 1976*

	Day, Post Application					
	1	3	5	7	14	21
	Residues (Mean ± Std Dev.), ppm					
Temple orange						
Rind	0.2 ± tr	0.3 ± 0.4	1.3 ± 1.1	1.0 ± 0.5	0.2 ± 0.1	0.4 ± 0.2
Pulp	0.1 ± tr	0.1 ± tr	0.2 ± 0.2	tr	0.2 ± 0.1	0.1 ± 0.2
Grapefruit						
Rind	0.1 ± 0.1	0.3 ± 0.2	0.6 ± 0.5	0.5 ± 0.3	0.2 ± 0.1	0.2 ± tr
Pulp	ND	0.1 ± 0.1	0.1 ± 0.2	0.2 ± 0.3	0.6 ± tr	0.1 ± 0.1
Valencia orange						
Rind	0.2 ± 0.1	0.3 ± 0.3	1.3 ± 0.6	1.0 ± 0.8	0.2 ± 0.1	0.2 ± 0.1
Pulp	0.1 ± 0.2	0.1 ± 0.1	0.4 ± 0.1	tr	0.1 ± 0.1	0.1 ± 0.1
Lemon						
Rind	0.1 ± 0.1	0.2 ± 0.2	0.5 ± 0.3	0.6 ± 0.6	0.2 ± 0.1	0.1 ± 0.1
Pulp	tr	0.1 ± 0.1	0.3 ± 0.6	1.2 ± 1.8	0.1 ± 0.1	0.2 ± 0.1
Tangerine						
Rind	0.2 ± 0.2	0.5 ± 0.4	1.5 ± 1.2	1.5 ± 1.2	0.2 ± 0.1	0.6 ± 0.4
Pulp	tr	0.2 ± 0.2	0.2 ± 0.2	0.4 ± 0.4	0.1 ± tr	0.1 ± 0.1

NOTE: tr = trace = <0.01 ppm. ND = not detected.

washed fruit (days 1, 3, 5, 7), and data for washed and unwashed fruit were combined for statistical analyses. This result may be due to the method of handling samples (*3*). In the present study, frozen fruits were thawed before being peeled. Condensation on the fruits collected in the bottom of the bag; this condensate was not added to the extract because only half of each fruit was peeled. The fruits were thus washed by condensation prior to peeling. This accounts for the absence of statistical difference between washed and unwashed fruits. The data presented here can only properly be considered penetrated residues. Also, no residues of acephate or methamidophos were detected in fruit which had been misted. Only the results of the hydraulic application are reported here.

Both acephate and methamidophos are systemic insecticides, and the data in Figure 1 indicate that at least acephate readily penetrates the rind of all citrus varieties. Because methamidophos can be produced from acephate by hydrolysis, internal methamidophos could have come from acephate.

The peak of penetrated residues of both compounds occurs on days 5 and 7. Acephate residues in rind are significantly higher (0.01 ppm) on day 7 than on days 5 and 14 for Temple oranges, grapefruit, and lemons. For Valencia oranges and tangerines, day 14 residues are significantly lower than are day 7 residues, but due to the variability of the data, the peak of penetrated residues may have occurred on day 5 (Table 1). Had data been taken on day 9, higher methamidophos resi-

dues might have been found, indicating additional conversion of acephate. The data do show, however, that the residues are above the EPA action levels of 3.0 ppm acephate and 0.25 ppm methamidophos on day 7 (Tables 1, 2; Fig. 1).

The acephate-in-pulp pattern is similar to that in rind. Day 7 residues are significantly higher than are residues on days 5 or 14 in Temple oranges, grapefruit, lemons, and tangerines. For Valencia oranges, the peak of acephate in pulp may have occurred on day 5 when residues of acephate were significantly higher than on days 3 or 7. Acephate in pulp was never above the action level of 3.0 ppm. The maximum level of acephate in pulp was 2.0 ppm in tangerines on day 14. Most pulp acephate residues averaged 1.0 ppm or less (Table 1).

The pattern of methamidophos residues was similar to that of acephate (Table 2). For Temple orange, grapefruit, Valencia orange, lemon, and tangerine rind, day 7 residues were significantly higher than were day 14 residues. However, methamidophos levels in rind were the same on days 5 and 7, so residues may have peaked on day 5. In pulp, no peak of methamidophos residues was apparent in Temple oranges, but statistically significant peaks occurred on day 7 in lemons and tangerines, on day 5 in Valencia oranges, and on day 14 in grapefruit.

The pattern of penetration of acephate and methamidophos in both rind and pulp of these varieties was statistically significant and consistent. The peak penetrated residues of acephate and methamidophos in rind and in pulp occurred on or before day 7 with decreasing residues thereafter.

The statistical comparison of varieties in Table 3 indicates that by day 14 there are no differences in acephate residue levels in rind among varieties. Before day 14, no consistent pattern of residue levels is evident. The same comparison for acephate in pulp (Table 4) points to significantly lower residues in grapefruit pulp than in lemon and tangerine pulp. With this exception, there were no differences in acephate residues in pulp by day 21. Residues in Temple oranges were significantly higher than were residues in grapefruit until day 21 (Tables 1, 4).

By day 14 there were no significant differences in methamidophos levels in rind among varieties, yet differences appear on day 21 (Table 5). There were no differences in methamidophos residues in pulp by day 21 (Table 6).

There is a nonrandom source of variation in the comparison of residue levels in citrus which has been noted in greenhouse tomato studies with acephate, surface area–to-weight ratios (*5*).

Confounded with fruit size is varietal rind thickness. Valencia orange rind thickness has been reported as 4.0 mm (*2*), 4.1 mm (*11*), and 3.0 mm (*9*). Marsh grapefruit rind thickness has been noted as 5.5 mm (*11*) and 12.0 mm (*9*). Lemon rind thickness has been reported as 7.3 mm (*11*), 3.6 mm (*1*), and 5.0 mm (*9*). In addition to genetic differences in rind thickness, many climatic and cultural practices affect rind thickness (*1, 2, 9, 11*). In the present experiment, thick grapefruit rind with a low surface area–to-weight ratio appears to account for low pesticide residues in grapefruit. Future experiments to compare citrus variety differences in residue behavior should include rind thickness and surface area measurements to determine

TABLE 3. *Statistical comparison of acephate residue levels in citrus rind, 1976*

	Day, Post Application					
	1	3	5	7	14	21
Temple orange vs. grapefruit	no	yes	no	yes	no	no
Temple vs. Valencia oranges	yes	yes	no	no	no	no
Temple orange vs. lemon	yes	no	no	no	no	no
Temple orange vs. tangerine	yes	no	yes	yes	no	no
Grapefruit vs. Valencia orange	yes	yes	yes	no	no	no
Grapefruit vs. lemon	yes	yes	no	yes	no	no
Grapefruit vs. tangerine	no	yes	yes	no	no	no
Valencia orange vs. lemon	no	yes	no	yes	no	no
Valencia orange vs. tangerine	no	yes	no	no	no	no
Lemon vs. tangerine	no	no	yes	yes	no	yes

NOTE: Yes = means are statistically different at 0.01 level; no = means are not statistically different at 0.01 level (*10*).

TABLE 4. *Statistical comparison of acephate residue levels in citrus pulp, 1976*

	Day, Post Application					
	1	3	5	7	14	21
Temple orange vs. grapefruit	yes	yes	yes	yes	yes	no
Temple vs. Valencia oranges	no	no	yes	yes	yes	no
Temple orange vs. lemon	no	yes	no	no	no	no
Temple orange vs. tangerine	yes	yes	yes	yes	yes	no
Grapefruit vs. Valencia orange	yes	no	yes	no	yes	no
Grapefruit vs. lemon	yes	yes	yes	yes	yes	yes
Grapefruit vs. tangerine	yes	yes	yes	yes	yes	yes
Valencia orange vs. lemon	no	yes	yes	yes	no	no
Valencia orange vs. tangerine	yes	yes	no	yes	no	no
Lemon vs. tangerine	yes	no	no	no	no	no

See NOTE, Table 3.

TABLE 5. *Statistical comparison of methamidophos residue levels in citrus rind, 1976*

	Day, Post Application					
	1	3	5	7	14	21
Temple orange vs. grapefruit	yes	no	yes	yes	no	yes
Temple vs. Valencia oranges	no	no	no	no	no	yes
Temple orange vs. lemon	yes	no	yes	yes	no	yes
Temple orange vs. tangerine	no	no	no	yes	no	no
Grapefruit vs. Valencia orange	yes	no	yes	yes	no	no
Grapefruit vs. lemon	no	no	no	no	no	yes
Grapefruit vs. tangerine	yes	yes	yes	yes	no	yes
Valencia orange vs. lemon	yes	no	yes	yes	no	no
Valencia orange vs. tangerine	no	yes	no	no	no	yes
Lemon vs. tangerine	yes	yes	yes	yes	no	yes

See NOTE, Table 3.

TABLE 6. *Statistical comparison of methamidophos residue levels in citrus pulp, 1976*

	Day, Post Application					
	1	3	5	7	14	21
Temple orange vs. grapefruit	yes	no	no	yes	yes	no
Temple vs. Valencia oranges	no	no	yes	no	no	no
Temple orange vs. lemon	yes	no	no	yes	no	no
Temple orange vs. tangerine	yes	yes	no	yes	yes	no
Grapefruit vs. Valencia orange	yes	no	yes	yes	yes	no
Grapefruit vs. lemon	yes	no	no	yes	yes	no
Grapefruit vs. tangerine	yes	yes	no	yes	yes	no
Valencia orange vs. lemon	yes	no	no	yes	no	no
Valencia orange vs. tangerine	yes	yes	yes	yes	no	no
Lemon vs. tangerine	no	yes	no	yes	no	no

See NOTE, Table 3.

whether any differences in residue levels could be due to fruit structure.

Penetration of both compounds into rind and pulp complicates data analyses. The overall data actually show that the appearance of residue is due to penetration. The fit to a first-order disappearance model is correspondingly poor, ranging from a low of $r = 0.02$ for acephate in Valencia pulp to a high of $r = -0.79$ for acephate in lemon rind. However, when data from days 7 (maximum concentration), 14, and 21 are used, disappearance is clearer (Fig. 1).

There are still positive correlations for methamidophos in Temple and Valencia orange pulp which reflect an appearance of the compound in the pulp, and the tangerine rind data for acephate do not fit a first-order model. Based on 7-, 14-, and 21-day data the half-life averages are 10.5 days and 10.3 days for methamidophos and acephate, respectively, in fruit rind, and 6.1 days and 15.0 days for methamidophos and acephate, respectively, in pulp (Table 7).

The data presented for acephate and methamidophos show that both compounds disappear under Florida conditions after reaching maximum penetrated residues on day 7. Acephate was below 3 ppm in rind 14 days after application and never reached 3 ppm in pulp. Penetrated residues of methamidophos reached an average level of less than 0.25 ppm 21 days after application.

TABLE 7. *Acephate and methamidophos first-order disappearance in Florida citrus, 7–21-day data, 1976*

	Slope		$t_{1/2}$[1] (half-life, days)		r	
	Metha-midophos	Ace-phate	Metha-midophos	Ace-phate	Metha-midophos	Ace-phate
Temple orange						
Rind	−0.07	−0.10	9.9	6.9	−0.57[2]	−0.94[3]
Pulp	0.32	−0.06	2.2	11.6	−0.79[2]	−0.98[3]
Grapefruit						
Rind	−0.07	−0.05	9.9	13.9	−0.86[2]	−0.68[2]
Pulp	−0.05	−0.08	13.9	8.7	−0.38	−0.99[3]
Valencia orange						
Rind	−0.05	−0.05	13.9	13.9	−0.68[2]	−0.77[2]
Pulp	0.32	0.03	2.2	23.1	0.86[3]	0.99[3]
Lemon						
Rind	−0.08	−0.11	8.7	6.3	−0.99[3]	−0.99[3]
Pulp	−0.13	−0.04	5.3	17.3	−0.69[2]	−0.81[2]
Tangerine						
Rind	−0.07	—	9.9	—	−0.45	—
Pulp	−0.10	−0.05	6.9	13.9	−0.87[2]	−0.79[2]
Averages						
Rind			10.5	10.3		
Pulp			6.1	15.0		

[1] $t_{1/2} = \ln(0.5)/\text{slope}$.

[2] Significant at 5 percent level (6).

[3] Significant at 1 percent level or higher (6).

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FISH, WILDLIFE, AND ESTUARIES

Effects of Organochlorine Residues on Eggshell Thickness, Reproduction, and Population Status of Brown Pelicans (Pelecanus occidentalis) *in South Carolina and Florida, 1969–76*

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ABSTRACT

Shells of brown pelican (Pelecanus occidentalis) *eggs collected in South Carolina from 1969 through 1975 and in Florida during 1969, 1970, and 1974 were significantly thinner ($P > 0.05$) than eggshells collected before 1947. Thickness of South Carolina eggshells increased in 1975, and mean thickness of eggshells collected in Florida during 1974 was greater than that of eggshells collected during 1969 and 1970, primarily in Gulf Coast colonies.*

Residues of 13 organochlorines were found in eggs and tissues of pelicans found dead during 1974 and 1975, although residues in brains of these specimens were not high enough to cause death. Residues of organochlorines, except PCBs, declined through 1975. PCBs increased in eggs from Atlantic Coast colonies.

Reproductive success and population status of brown pelicans in South Carolina have improved markedly since authors began their studies in 1969. Good reproductive success was reported in 3 of 5 years from 1973 through 1977.

Introduction

This is part of a series of papers on the effects of environmental pollutants on the brown pelican *(Pelecanus occidentalis)*. In previous papers, organochlorine residues in brown pelicans have been related to eggshell thinning *(6, 7)*, reproductive success *(9)*, adult mortality *(5, 10)*, population decline *(4)*, and possible extirpation of a population in Louisiana *(8)*. The objective of the present study is to further explore effects of organochlorines on brown pelicans, particularly the significance of declining residues. Emphasis is placed on data gathered during 1974–76, but data from 1969 onward are used to show trends over 8 years.

Procedures for Sampling, Necropsy, and Field Study

Most procedures have been described in previous papers *(4, 10)*. Brief visits were made to brown pelican colonies in South Carolina in 1969, 1970, and 1976 and to Florida colonies in 1969, 1970, and 1974. The two brown pelican nesting colonies in South Carolina, Deveaux Bank and Marsh Island, Cape Romain National Wildlife Refuge (CRNWR), were studied intensively in the spring and summer each year from 1971 through 1975. Censuses were made of total nests and fledged young in both South Carolina colonies from 1969 through 1976. However, most accurate data were collected during 1971–75 when a number of visits were made to each colony during each nesting season. Addled and viable eggs in all stages of incubation were collected. One egg was usually taken from each nest selected for sampling. Eggs were weighed and measured, and their contents were placed in chemically cleaned glass bottles and frozen. Eggshells were thoroughly washed with tap water and allowed to dry. Shell thickness (shell plus shell membranes) was measured at three sites on the waist of the egg with a micrometer graduated in units of 0.01 mm.

Nests with full clutches and nests from which one egg was collected were marked on Marsh Island to determine their success. Marked nests were checked for eggs or young on each visit to the colony; colonies were visited twice a week for up to 1 hour.

Several dead pelicans and samples of fish regurgitated by pelicans were collected and frozen. The pelicans were removed from the freezer several months later,

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thawed, and subsequently necropsied. 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 cleaned glass bottle, and the carcass, except for skin, feet, wings, liver, kidney, and gastrointestinal tract, was wrapped in foil and refrozen. Brains and carcasses were later analyzed for organochlorine residues.

Analytical Procedures

The contents of eggs collected during 1969–71 were homogenized. A 20-g portion was mixed with anhydrous sodium sulfate in a blender and extracted for 7 hours with hexane in a Soxhlet apparatus. The extract was cleaned by acetonitrile partitioning and was eluted on partly deactivated Florisil. For pesticide analyses, residues in the cleaned extract were separated and removed in four fractions from a silica gel thin-layer plate (*17*). 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 W-HP. ΣDDT in fractions III or IV was confirmed on a column of 3 percent XE-60 or 3 percent QF-1 Gas-Chrom Q. Polychlorinated biphenyls (PCBs) were identified and measured semiquantitatively by thin-layer chromatography (*16*). Average recoveries of organochlorine pesticides and their metabolites were 75–112 percent.

Methodology was modified for eggs collected from 1972 to 1975 (*11*). The extract of the 10-g portion was cleaned on a Florisil column. Pesticides and PCBs were separated into three fractions on a Silicar column and analyzed by GC on a column packed with a mixture of 4 percent SE-30 and 6 percent QF-1. This methodology enabled authors to detect toxaphene, *cis*-chlordane, and/or *trans*-nonachlor, and *cis*-nonachlor. Until 1973, there was neither a *cis*-nonachlor standard for quantification nor a procedure to estimate toxaphene levels. Lipids were removed from the eggs collected during 1974–75 either by Florisil cleanup or by automated gel permeation chromatography. In 1974, *cis*-chlordane and *trans*-nonachlor were separated and quantified by changing the column packing to a mixture of 1.5 percent OV-17 and 1.95 percent QF-1.

Residues in about 10 percent of the samples were confirmed by combined gas chromatography–mass spectrometry (GC–MS). Average recoveries from spiked chicken eggs were 81–110 percent; residues are not corrected for recovery values. The lower limit of detection for pesticides or their metabolites was 0.01 μg/g in fish and 0.10 μg/g in other samples (0.01 μg/g for hexachlorobenzene). The lower limit for PCBs was 0.05 μg/g in fish and 0.50 μg/g in other samples.

Results

REPRODUCTIVE SUCCESS AND POPULATION STATUS

From 1969 through 1972 (*10*) and for previous years (*3*), reproductive success of South Carolina pelicans was below the recruitment standard of 1.2–1.5 fledged young per breeding female per year that is necessary to maintain a stable population (*14*). Following a successful reproductive season in 1973, pelicans experienced poor success in 1974 and 1975, then had successful reproductive seasons in 1976 (Table 1) and 1977 (Vivian Mendenhall, Fish and Wildlife Service, U.S. Department of the Interior, 1977: personal communication).

Except in 1969, reproductive success was higher on Deveaux Bank than on Marsh Island (Table 1). However, there was a significant positive correlation ($r = 0.797$, $P < 0.05$) between young fledged per nest in the two colonies over the 8 years considered in the present report. Thus reproductive success in one colony paralleled that in the other colony. Lower reproduction on Marsh Island was attributed to tidal flooding of nests each year, a rare occurrence on Deveaux Bank. Many of the pelicans with flooded nests laid a second clutch, but replacement clutches also were frequently laid in low areas that were eventually flooded.

The size of the breeding population of brown pelicans in South Carolina slowly increased from 1969 through 1974 and then increased 41 percent from 1974 to 1975 as follows: 1,266 pairs in 1969; 1,670 pairs in 1974; 2,400 pairs in 1975; and 3,300 pairs in 1977.

TABLE 1. *Reproductive success of brown pelicans in South Carolina, 1969–76*

Year	Colony	No. of Nests	No. of Young Fledged	Young Fledged per Nest
1969	Cape Romain	1016	900[1]	0.82[1]
	Deveaux Bank	250[1]	80	0.32[1]
	Both Colonies	1266	980	0.78
1970	Cape Romain	637	500[1]	0.78[1]
	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
1974	Cape Romain	920	825	0.90
	Deveaux Bank	750	800	1.07
	Both Colonies	1670	1625	0.97
1975	Cape Romain	900	500	0.56
	Deveaux Bank	1500	1300	0.87
	Both Colonies	2400	1800	0.75
1976	Cape Romain	1440	1399	0.97
	Deveaux Bank	1100[1]	1738[1]	1.58[1]
	Both Colonies	2540	3137	1.23

[1] Estimated numbers—all other figures are based on actual counts.

TABLE 2. *Probable causes of brown pelican mortality, South Carolina, 1974–75*

Year	Sex	Age	Probable Cause of Mortality
1974	F	4 weeks	sacrificed, had subcutaneous emphysema
	F	6 weeks	hemorrhagic enteritis in combination with severe pecking injuries
	F	12 weeks	respiratory problems—apparent air saculitis
	M	8 weeks	sacrificed, bird was near death of diarrhea and excessive fluid in lungs, air sacs, and pericardium
1975	M	adult	hemorrhagic enteritis
	M	adult	hemorrhagic enteritis

MORTALITY

Pelicans died of possible starvation and several diseases. Hemorrhagic enteritis caused the death of at least two of the six adults found dead on Deveaux Bank April 9, 1975 (Table 2). These pelicans apparently had recently migrated to South Carolina. Many brown pelicans that breed in South Carolina winter on the Atlantic Coast of Florida where hemorrhagic enteritis was responsible for many deaths of the birds in 1972 (*10, 20*).

In 1974, a 6-week-old pelican apparently died of hemorrhagic enteritis and severe pecking; the pecking probably occurred when the sick young was attacked by hostile young and adults. A 12-week-old pelican apparently died of respiratory problems including air saculitis. One of two young sacrificed in 1974 (Table 2) was near death, and the other had subcutaneous emphysema, a condition that is rarely fatal (*13*).

Several hundred downy young were found dead on Deveaux Bank in 1974. Little regurgitated food was observed during visits to the colony compared to visits in other years, and except for the usual heavy mortality after hatching, the deaths involved young at least 4 weeks old, an age when food demand rapidly increases.

EGGSHELL THICKNESS

Mean eggshell thickness of brown pelican eggs collected in South Carolina (Table 3) was 10–17 percent less than the pre-1947 mean of 0.557 mm (*1*). The significant increase ($P < 0.05$) in mean shell thickness in 1975, compared to the 6 preceding years, initiated an upward trend extending to 1977 (Vivian Mendenhall: personal communication).

Overall eggshell thickness of pelican eggs in Florida increased slightly from 1969–70 to 1974 (Table 3); it increased markedly in the Gulf Coast colonies and remained unchanged in the Atlantic Coast colonies (Tables 4, 5). Shell thickness of Gulf Coast pelican eggs collected in 1974 averaged just 2 percent less than the pre-1947 mean, whereas Atlantic Coast eggs averaged 11 percent less. There were insufficient data to compare trends in shell thickness in Florida Bay colonies (Table 4). In addition to South Carolina and Florida, eggshell thickness of brown pelicans has been increasing in California (*2*) and Louisiana (*5*).

RESIDUES IN EGGS

PCB and DDE residues made up the bulk of the 13 organochlorines identified in eggs of brown pelicans (Tables 6–8). Residues in pelican eggs in 1974–75 followed the same pattern in each of the two South Carolina colonies: there was a similarity in mean residues of each organochlorine in a given year, there was much individual variation in residues of each organochlorine, and there was a general decline in residues of most organochlorines (Table 9). These patterns and trends were also evident in samples collected from 1969 through 1973 (*4, 10*). Residues of DDE, DDT, and ΣDDT declined steadily from 1969 through 1975, whereas TDE declined steadily to 1973 and then increased somewhat. Dieldrin declined until 1971 and then remained essentially stable through 1975. PCB residues were erratic and followed no definite trend.

From 1969–70 to 1974 (Table 10), there were significant declines ($P < 0.05$) in DDE, TDE, DDT, and ΣDDT in brown pelican eggs from four regions of the southeastern United States; dieldrin decreased significantly ($P < 0.05$) in South Carolina and along the At-

TABLE 3. *Shell thickness of brown pelican eggs, 1969–75, compared to pre-1947 levels*

Eggshell Thickness, mm[1]							
Pre-1947	1969	1970	1971	1972	1973	1974	1975
South Carolina							
0.557 ± A[2]	0.463 ± D	0.461 ± D	0.480 ± C	0.470 ± CD	0.463 ± D	0.469 ± CD	0.499 ± B
0.012 (23)	0.006 (49)	0.007 (38)	0.005 (65)	0.005 (67)	0.003 (104)	0.004 (116)	0.004 (95)
Florida							
0.557 ± A	0.516 ± B	0.511 ± B				0.521 ± B	
0.003 (169)	0.005 (89)	0.004 (144)				0.004 (122)	

[1] Mean ± standard error; sample size in parentheses.
[2] A significant difference among thickness means ($P < 0.05$) is indicated for those means not sharing a common letter. Means were separated by multiple range tests (*12, 15*).

TABLE 4. *Shell thickness of brown pelican eggs from Florida colonies, 1969–70, 1974*

Colony	Eggshell Thickness, mm[1] 1969	1970	1974
ATLANTIC COAST			
Port Orange	0.488 ± 0.012 (9)	0.497 ± 0.009 (9)	0.476 ± 0.013 (14)
Crane Island	—	0.491 ± 0.009 (10)	—
Cocoa Beach	0.497 ± 0.011 (10)	0.482 ± 0.019 (10)	0.499 ± 0.010 (15)
Pelican Island	0.499 ± 0.012 (10)	0.498 ± 0.017 (9)	0.499 ± 0.010 (14)
Fort Pierce	0.513 ± 0.012 (6)	0.504 ± 0.009 (9)	0.508 ± 0.011 (8)
FLORIDA BAY			
Nest Key	—	0.532 ± 0.012 (10)	—
Buchanan Key	0.530 ± 0.015 (3)	0.545 ± 0.013 (10)	—
Fanny Key	—	0.523 ± 0.019 (7)	—
Marquesas Key	—	0.541 ± 0.012 (10)	0.523 ± 0.016 (9)
GULF COAST			
Seahorse Key	0.530 ± 0.015 (6)	0.531 ± 0.016 (10)	0.547 ± 0.009 (15)
Tarpon Key	0.509 ± 0.015 (8)	0.487 ± 0.015 (10)	—
Cortez	—	0.502 ± 0.012 (10)	0.534 ± 0.010 (15)
Bird Key	0.559 ± 0.014 (10)	0.517 ± 0.014 (10)	0.549 ± 0.013 (15)
Matlacha Pass	0.522 ± 0.023 (9)	0.504 ± 0.019 (10)	—
Hemp Island	0.516 ± 0.012 (10)	0.519 ± 0.015 (10)	0.549 ± 0.012 (15)

[1] See footnote 1, Table 3.

TABLE 5. *Shell thickness of brown pelican eggs from the Gulf and Atlantic Coasts of Florida, 1969–70, 1974*

Eggshell Thickness, mm[1] 1969	1970	1974
GULF COAST		
0.528 ± AB[2] 0.007 (43)	0.510 ± A 0.006 (60)	0.545 ± B 0.006 (60)
ATLANTIC COAST		
0.498 ± A 0.006 (35)	0.494 ± A 0.006 (47)	0.494 ± A 0.006 (51)

[1] See footnote 1, Table 3.
[2] See footnote 2, Table 3.

lantic Coast of Florida, remained stable in Florida Bay, and increased slightly on the Gulf Coast. In contrast, PCBs increased significantly ($P < 0.05$) in two areas and showed little change in the other two areas. The most striking change was on the Atlantic Coast of Florida where the PCB residues more than doubled from 1969–70 to 1974. The DDE:PCB ratio changed dramatically in most areas. For example, the ratio was approximately 1:1 on the Atlantic Coast of Florida in 1969–70 and 1:6 in 1974. DDT residues were rarely found in 1974 samples. The order of decreasing organochlorine contamination, by area, in pelican eggs during each sampling period was: South Carolina > Florida Atlantic Coast > Florida Gulf Coast > Florida Bay (Table 10). Eggs collected from the Gulf Coast and Florida Bay colonies in 1974 were essentially devoid of organochlorine residues.

RESIDUES IN TISSUES

Birds found dead were analyzed for organochlorine residues. Residues in tissues of four pre-fledgling pelicans found dead in South Carolina in 1974 were as low as those reported previously in other young pelicans (*4, 10*). Six freshly dead adult pelicans were found on Deveaux Bank April 9, 1975.

Residues in three male adults were much higher than in the young birds collected in 1974, but residues in their brains were below lethal levels (Table 11).

RESIDUES IN FISH

Breeding brown pelicans in South Carolina feed almost exclusively on young-of-the-year Atlantic menhaden (*Breevoortia tyrannus*) that hatch off the coast from October through April and migrate into the estuaries as larvae where they usually remain for 6–8 months (*19*). Residues of DDE in menhaden in 1974 and 1975 were much lower than those reported in 1973 (*10*); DDT and dieldrin were found in most 1973 samples but were not detected in 1974–75 samples (Table 12). PCB residues averaged about the same in 1973 and 1974 but declined substantially in 1975.

Discussion

Because trips to Deveaux Bank were infrequent, it could not be established that starvation was responsible for the deaths of downy young in 1974. Both young that were necropsied exhibited signs of disease that may or may not have been related to starvation (Table 2). There were no apparent deaths of downy young on the CRNWR, about 65 km northeast of Deveaux Bank, although the pelicans there had poor reproductive success and, judging from regurgitated boluses, they preyed on a greater variety of fish than usual. Therefore, poor food supply was probably responsible for the deaths of downy young on Deveaux Bank.

The authors previously suggested that migration of Atlantic menhaden complicate interpretation of biomagnification of residues from fish to pelican eggs (*10*) because adult menhaden are exposed to varying levels of organochlorine residues during migration. However, authors have since determined that breeding pelicans in South Carolina feed almost exclusively on young-of-the-year menhaden that apparently accumulate nearly all their residues from local estuaries. The interpretation of biomagnification is still complicated by the migratory behavior of the brown pelican that exposes it to several habitats with differing degrees of organochlorine pollution.

TABLE 6. *Organochlorine residues in brown pelican eggs, South Carolina, 1974*

	Residues, µg/g fresh wet weight												
	DDE	TDE	DDT	Dieldrin	Heptachlor Epoxide	Mirex	Oxychlordane	*cis*-Chlordane	*trans*-Nonachlor	*cis*-Nonachlor	HCB	Toxaphene	PCBs
MARSH ISLAND													
	1.89	0.60	—	0.44	—	—	—	0.18	0.19	0.11	—	0.27	5.60
	1.43	0.48	—	0.33	—	—	—	0.15	0.15	—	—	0.12	4.15
	1.37	0.48	—	0.27	—	—	—	—	—	—	—	0.18	5.25
	1.56	0.46	—	0.39	—	0.11	—	0.11	—	—	—	0.11	6.87
	1.70	0.48	—	0.55	—	—	—	0.16	0.15	—	0.10	1.88	7.57
	2.39	0.58	—	0.46	—	—	—	0.19	0.25	0.12	—	0.12	7.98
	1.35	0.46	—	0.30	—	—	—	0.16	0.10	—	—	—	4.34
	1.51	0.38	—	0.40	—	—	—	—	—	—	—	0.21	8.30
	1.65	0.37	—	0.40	—	—	—	0.13	—	—	—	0.22	4.19
	1.65	0.38	—	0.36	—	—	—	0.16	0.13	0.10	—	0.19	7.02
	1.67	0.30	—	0.42	—	—	—	—	—	—	—	0.18	6.90
	1.00	0.32	—	0.27	—	—	—	0.15	0.13	—	—	0.15	5.40
	7.03	1.48	—	1.46	0.14	0.20	—	0.41	0.63	0.35	—	0.83	18.09
	2.46	0.36	—	0.46	—	—	—	0.14	0.21	0.16	—	0.15	13.80
	1.33	0.39	—	0.36	—	—	—	0.15	0.16	—	—	0.21	9.21
	3.83	0.91	—	0.96	0.10	—	—	0.31	0.34	0.22	—	0.38	13.88
	2.36	0.47	—	0.57	—	0.10	—	0.21	0.24	0.18	—	0.29	11.80
	1.37	0.35	—	0.13	—	—	—	0.12	0.15	—	—	0.20	7.66
	1.75	0.42	—	0.28	—	—	—	0.11	0.14	—	—	0.17	17.00
	4.69	1.80	0.58	2.89	0.32	—	0.11	0.61	0.71	0.63	—	—	13.00
	1.95	0.57	—	0.62	—	—	—	0.19	0.17	0.15	—	—	6.49
	2.75	0.60	—	0.73	—	—	—	0.19	0.19	0.25	—	—	10.50
	1.86	0.45	—	0.42	—	—	—	0.16	0.10	0.10	—	—	5.69
	3.42	0.64	—	0.73	—	—	—	0.21	0.11	0.22	—	—	11.70
	1.40	—	—	0.46	—	—	—	0.17	0.13	—	—	—	8.15
	2.94	0.34	—	0.74	—	—	—	—	—	0.15	—	—	12.50
	3.79	—	—	0.88	—	—	—	0.22	0.17	0.31	—	—	7.10
	5.85	0.14	—	1.27	—	—	—	0.36	0.33	0.30	—	—	22.11
	5.51	—	—	1.03	—	—	—	0.35	0.38	0.25	—	—	21.80
	1.22	0.16	—	0.49	—	—	—	—	—	—	—	—	7.34
	2.45	0.41	0.15	0.61	—	—	—	0.13	0.17	0.15	—	—	8.53
	2.82	—	—	0.86	—	—	—	0.14	0.20	0.27	—	—	12.15
	3.80	0.78	—	0.90	—	—	—	0.24	0.24	0.22	—	—	11.65
	4.13	0.59	—	0.83	—	—	—	0.17	0.19	0.18	—	—	9.80
	2.21	0.47	—	0.64	—	—	—	0.17	0.16	0.23	—	—	8.19
	2.38	0.40	—	0.71	—	—	—	0.14	0.13	0.16	—	—	7.38
	5.91	1.25	—	1.21	—	—	0.10	0.40	0.42	0.45	—	—	17.72
	0.81	0.19	—	0.18	—	—	—	0.15	0.11	0.11	—	—	6.39
	3.90	0.68	—	0.71	—	—	—	0.23	0.19	0.11	—	—	8.28
	1.40	0.34	—	0.31	—	—	—	—	—	—	—	0.21	0.70
	3.86	0.63	—	0.84	—	—	—	0.22	0.24	0.17	—	0.46	11.33
	5.57	1.09	0.73	1.26	0.16	0.20	—	0.44	0.39	0.31	—	0.82	14.04
	2.41	0.63	0.34	0.56	—	—	—	0.24	0.22	0.15	—	0.46	7.05
	1.53	0.26	—	0.26	—	—	—	0.11	0.16	—	—	0.17	8.02
	2.38	0.59	—	0.36	—	—	—	0.10	—	0.10	—	0.17	5.47
	5.00	0.90	—	1.10	—	—	—	0.45	0.47	0.45	—	0.57	27.48
	3.04	0.78	—	0.71	—	—	—	0.28	0.28	0.18	—	0.58	14.43
	3.68	0.83	—	0.71	—	—	—	0.27	0.32	0.17	—	0.39	11.77
	2.67	0.49	—	0.36	—	—	—	0.16	0.16	0.14	—	0.23	5.10
	2.99	0.67	—	0.57	—	—	—	0.22	0.30	0.15	—	0.37	9.90
	1.95	0.53	—	0.49	—	—	—	0.14	0.12	0.22	—	0.35	2.30
	1.95	0.36	—	0.38	—	—	—	0.16	0.20	0.12	—	—	8.80
	2.08	0.47	—	0.59	0.10	—	—	0.25	0.23	0.16	—	—	8.00
	1.07	0.23	—	0.26	—	—	—	0.11	0.15	—	—	0.35	5.80
GM	2.35	0.41		0.55				0.17	0.16	0.12		0.13	8.32
CL	2.04–2.70	0.34–0.51		0.47–0.63				0.14–0.20	0.13–0.19	0.10–0.15		0.10–0.17	7.10– 9.76
Range	0.81–7.03	ND–1.80	ND–0.73	0.17–2.89	ND–0.32	ND–0.20	ND–0.11	ND–0.61	ND–0.71	ND–0.63	ND–0.10	ND–1.88	0.70–27.48
DEVEAUX BANK													
	1.50	0.50	—	0.36	—	—	—	0.11	0.09	—	—	0.32	5.01
	1.27	0.38	—	0.33	—	—	—	0.10	—	—	—	0.20	5.16
	0.65	0.36	—	0.34	—	—	—	—	—	—	—	0.22	4.07
	1.36	0.37	—	0.29	—	—	—	0.14	0.13	0.09	—	0.21	5.00
	1.48	0.38	—	0.33	—	0.30	—	—	—	—	—	—	1.90
	1.84	0.48	—	0.45	—	—	—	0.09	0.21	0.09	—	—	8.14
	1.48	0.47	—	0.45	—	—	—	—	0.18	—	—	—	7.53
	1.93	0.56	—	0.59	—	—	—	0.22	0.12	—	—	—	12.00
	0.89	0.22	—	0.26	—	—	—	—	0.14	—	—	—	4.24
	2.44	0.76	—	0.66	—	0.20	—	0.23	0.19	0.15	—	—	11.07
	2.70	0.61	—	0.81	—	—	—	0.18	0.19	0.15	—	—	9.82
	2.18	0.40	0.11	0.62	—	—	—	0.17	0.23	—	—	—	12.16
	1.20	0.34	—	0.26	—	—	—	—	0.17	—	—	—	4.80
	2.30	0.59	—	0.54	0.16	—	—	0.11	0.22	0.10	—	—	7.58

(Continued next page)

TABLE 6 (Cont'd.) *Organochlorine residues in brown pelican eggs, South Carolina, 1974*

	Residues, µg/g fresh wet weight												
	DDE	TDE	DDT	Dieldrin	Heptachlor Epoxide	Mirex	Oxychlordane	*cis*-Chlordane	*trans*-Nonachlor	*cis*-Nonachlor	HCB	Toxaphene	PCBs
	1.40	0.34	—	0.36	—	—	—	0.11	0.12	—	—	—	5.37
	2.50	—	—	0.73	—	—	—	—	0.13	—	—	—	8.84
	3.53	0.95	—	0.95	—	0.13	—	0.31	0.24	0.24	—	—	16.76
	2.29	0.37	—	0.56	—	—	—	0.12	0.16	0.14	—	—	12.90
	2.42	0.59	—	0.61	—	—	—	0.16	0.16	0.14	—	—	8.27
	2.99	0.69	0.10	0.83	—	—	—	0.23	0.41	0.21	—	—	17.00
	1.70	0.46	—	0.50	—	—	—	0.21	0.26	0.18	—	—	7.91
	1.72	0.37	—	0.39	—	—	—	0.12	0.27	0.11	—	—	9.58
	1.60	0.31	—	0.39	—	—	—	0.15	0.19	0.13	—	—	7.50
	2.19	0.50	—	0.59	0.26	—	0.53	0.12	0.83	0.22	—	—	12.71
	1.65	0.43	—	0.43	—	—	—	0.16	0.19	0.11	—	0.17	4.74
	2.08	0.45	—	0.61	—	—	—	0.20	0.28	0.19	—	—	3.07
	2.02	0.52	—	0.47	—	—	—	0.19	0.25	0.13	—	—	7.18
	1.15	0.42	—	0.44	—	—	—	0.21	0.24	0.14	—	—	5.51
	1.43	0.42	—	0.42	—	—	—	0.17	0.17	0.10	—	—	8.02
	2.87	0.48	—	0.60	—	—	—	0.24	0.29	0.22	—	0.24	6.47
	2.11	0.37	—	0.55	—	—	—	0.16	0.15	0.16	—	0.76	5.70
	1.36	0.21	—	0.29	—	—	—	0.18	0.23	0.14	—	—	5.96
	1.37	0.31	—	0.60	—	—	—	0.31	0.24	0.20	—	—	1.32
	0.74	0.23	—	0.22	—	—	—	0.11	0.10	0.10	—	—	0.62
	0.78	0.16	—	0.32	—	—	—	—	—	—	—	—	0.95
	0.76	0.21	—	0.19	—	—	—	0.10	—	—	—	—	2.18
	1.41	0.22	0.20	0.27	—	—	—	0.14	0.16	0.10	—	—	3.17
	2.35	0.10	—	1.28	0.11	—	—	0.45	0.25	0.23	—	0.24	2.20
	0.80	0.27	—	0.27	—	—	—	0.14	0.13	0.13	—	0.39	5.35
	2.18	0.39	—	0.44	—	—	—	0.18	0.15	0.14	—	0.41	6.04
	2.05	0.37	—	0.56	—	—	—	0.16	0.12	0.13	—	0.41	2.68
	2.16	0.96	0.18	0.86	—	0.10	—	—	0.18	0.19	—	0.49	4.91
	1.84	0.48	—	0.39	—	—	—	0.29	0.14	0.24	—	0.31	6.54
	4.51	0.95	—	1.10	0.13	0.17	—	0.41	0.32	0.33	—	0.74	9.90
	3.04	0.84	—	1.14	0.12	—	—	0.41	0.30	0.25	—	0.48	14.18
	3.76	0.73	0.19	0.80	—	—	—	0.33	0.27	0.29	—	0.57	15.48
	3.11	0.62	—	0.53	—	—	—	0.29	0.31	0.20	—	—	5.27
	1.98	0.45	—	0.54	—	—	—	0.17	0.14	0.10	—	0.26	3.30
	2.12	0.31	—	0.33	—	—	—	0.12	0.12	0.10	—	0.14	8.11
	1.96	0.51	—	0.54	—	—	—	0.24	0.12	0.13	—	0.22	5.88
	1.92	0.62	0.10	0.67	0.11	—	—	0.29	0.24	0.15	—	—	5.80
	2.33	0.44	—	0.61	—	—	—	0.15	0.18	0.14	—	0.21	7.70
	3.32	0.59	—	0.88	0.12	—	—	0.20	0.21	0.19	—	—	9.50
	4.62	0.92	0.11	1.43	0.23	—	—	0.44	0.42	0.32	—	—	19.40
	4.94	1.22	—	1.22	0.14	3.01	—	0.45	0.40	0.35	0.02	0.73	21.40
	3.59	0.76	—	0.84	—	0.33	—	0.26	0.33	0.23	0.02	0.41	12.40
	3.67	0.37	—	0.85	0.11	—	—	0.22	0.22	0.18	0.02	—	12.60
	2.98	0.69	—	0.71	0.10	0.21	—	0.30	0.25	0.20	0.01	0.16	11.50
	1.59	0.38	—	0.40	—	—	—	0.22	0.14	0.13	0.01	0.18	8.80
	4.48	0.96	—	0.85	0.10	0.25	—	0.36	0.34	0.25	0.02	0.49	14.30
	2.30	0.80	—	0.96	0.13	—	—	0.50	0.21	0.37	0.04	0.41	24.80
GM	1.96	0.45		0.53				0.17	0.18	0.13		0.11	6.59
CL	1.74–2.21	0.40–0.52		0.47–0.60				0.14–0.19	0.16–0.21	0.11–0.15		0.09–0.14	5.48– 7.94
Range	0.65–4.94	ND–1.22	ND–0.20	0.19–1.43	ND–0.26	ND–3.01	ND–0.53	ND–0.50	ND–0.83	ND–0.37	ND–0.04	ND–0.76	0.62–24.80
MARSH ISLAND AND DEVEAUX BANK													
GM	2.13	0.44		0.54				0.17	0.17	0.13		0.12	7.36
CL	1.95–2.34	0.39–0.49		0.49–0.59				0.15–0.19	0.15–0.19	0.11–0.14		0.10–0.14	6.50– 8.32
Range	0.65–7.03	ND–1.80	ND–0.73	0.17–2.89	ND–0.32	ND–3.01	ND–0.53	ND–0.61	ND–0.83	ND–0.63	ND–0.10	ND–1.88	0.62–27.48

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

TABLE 7. *Organochlorine residues in brown pelican eggs, South Carolina, 1975*

	Residues, μg/g fresh wet weight											
	DDE	TDE	DDT	Dieldrin	Heptachlor Epoxide	Mirex	Oxy-chlordane	*cis*-Chlordane	*trans*-Nona-chlor	*cis*-Nona-chlor	Toxaphene	PCBs
MARSH ISLAND												
	1.41	0.38	—	0.50	—	—	—	0.13	—	0.15	0.38	5.02
	1.04	0.30	—	0.22	—	—	—	0.11	0.13	—	0.28	3.01
	1.91	0.75	—	0.58	—	0.10	—	0.29	0.27	0.24	0.27	4.35
	1.68	0.37	—	0.44	—	—	—	0.15	0.14	0.10	0.32	3.95
	1.15	0.27	—	0.34	—	—	—	0.15	0.13	0.11	0.24	4.64
	1.84	0.33	0.13	0.36	0.10	—	0.12	0.33	0.23	0.29	0.57	7.40
	1.00	0.35	—	0.32	—	—	—	0.17	0.19	0.13	0.24	3.28
	1.61	0.53	—	0.50	—	—	—	0.15	—	0.09	0.43	3.36
	3.10	0.69	—	0.72	—	—	—	0.26	0.24	0.19	0.31	10.03
	1.53	0.26	—	0.36	—	—	—	—	0.10	—	0.48	3.10
	1.20	0.33	—	0.35	—	—	—	0.17	0.18	0.11	0.48	4.50
	1.10	0.18	—	0.23	—	—	—	—	0.13	—	—	5.31
	1.22	0.31	—	0.31	—	—	—	0.12	0.16	—	0.14	6.45
	2.59	0.49	—	0.66	—	—	—	0.18	0.26	0.19	0.49	11.05
	1.64	0.41	—	0.48	—	—	—	0.20	0.26	0.18	0.55	6.20
	1.20	0.34	—	0.27	—	—	—	0.12	0.16	—	0.14	5.20
	0.81	0.19	—	0.22	—	—	—	0.10	0.10	—	0.11	8.80
	1.44	0.59	—	0.38	—	—	—	0.25	0.25	0.13	0.18	6.98
	1.09	0.34	—	0.27	—	—	—	0.14	0.18	0.10	0.16	5.02
	1.42	0.50	—	0.34	—	—	—	0.18	0.25	0.13	0.15	6.31
	1.03	0.34	—	0.27	—	—	—	0.19	0.15	0.11	0.21	5.95
	1.10	0.34	—	0.24	—	—	—	—	0.15	—	0.22	4.87
	0.75	0.30	—	0.16	—	—	—	0.11	—	—	0.16	7.06
	0.65	0.21	—	0.17	—	—	—	0.10	0.11	—	0.41	6.92
	0.96	0.31	—	0.26	—	—	—	0.16	0.18	0.12	0.16	8.73
	0.88	0.21	—	0.22	—	—	—	—	—	—	0.16	7.33
	1.61	0.57	—	0.38	—	—	—	0.25	0.14	0.14	0.26	8.38
	1.73	0.64	—	0.45	—	—	—	0.31	0.24	0.15	0.22	10.89
	1.13	0.34	—	0.34	—	—	—	0.16	0.14	0.11	0.21	7.96
	1.50	0.37	—	0.37	—	—	—	0.17	0.18	0.14	0.21	14.40
	1.91	0.58	—	0.53	—	0.10	—	0.31	0.22	0.20	0.34	12.78
	1.34	0.38	—	0.30	—	—	—	0.17	0.19	0.12	0.20	9.84
	1.57	0.54	—	0.51	—	0.14	—	0.24	0.16	0.14	0.42	12.96
	1.64	0.47	—	0.55	—	—	—	0.27	0.17	0 23	0.27	13.46
	1.12	0.41	—	0.32	—	—	—	0.21	0.19	0.13	0.20	10.72
	0.70	0.34	—	0.22	—	—	—	0.18	0.16	0.11	0.15	9.85
	1.57	0.41	—	0.51	—	—	—	0.22	0.16	0.16	0.28	12.91
	0.36	0.10	—	0.10	—	—	—	—	—	—	—	5.23
	0.87	0.29	—	0.23	—	—	—	0.13	0.14	—	—	5.81
	1.76	0.62	—	0.41	—	—	—	0.25	0.21	0.18	0.27	14.89
	1.15	0.45	—	0.34	—	—	—	0.23	0.13	0.14	0.12	6.46
	0.70	0.20	—	0.19	—	—	—	—	—	—	—	4.70
	0.95	—	—	0.23	—	—	—	0.14	0.10	0.14	0.11	11.53
	1.71	0.39	—	0.43	—	—	—	0.19	0.15	0.19	0.21	12.32
	1.85	0.60	—	0.53	—	—	—	0.25	0.14	0.20	0.37	14.53
	2.76	0.74	—	0 76	0.11	0.27	—	0.31	0.29	0.31	0.27	13.91
	1.65	0.57	—	0.42	—	—	—	0.26	0.18	0.18	0.21	9.67
	0.89	0.37	—	0.28	—	—	—	0.15	—	0.11	0.23	7.57
	1.72	0.67	—	0.92	—	—	0.13	0.67	0.35	0.40	0.40	12.93
	0.95	0.21	—	0.26	—	—	—	0.16	0.98	0.16	0.14	12.31
	1.80	0.38	—	0.40	—	—	—	0.17	0.16	0.18	0.29	17.99
	1.08	0.42	—	0.28	—	—	—	0.17	0.15	0.11	0.14	6.57
	2.51	0.58	—	0.71	0.10	0.38	—	0.39	0.23	0.33	0.33	20.08
	2.36	0.65	—	0.67	—	—	—	0.28	0.19	0.19	0.31	10.81
	1.58	0.43	—	0.49	—	0.10	—	0.21	0.12	0.13	0.27	7.43
	1.60	—	—	0.50	—	—	—	—	—	—	—	1.40
	0.18	—	—	—	—	—	—	—	—	—	—	0.38
	2.00	0.81	—	0.50	—	—	—	0.31	0.33	0.19	0.50	7.47
	1.59	0.58	—	0.40	—	—	—	0.28	0.28	0.17	0.29	10.95
GM	1.30	0.36		0.35				0.16	0.15	0.12	0.21	7.23
CL	1.15–1.46	0.30–0.42		0.31–0.40				0.14–0.19	0.13–0.18	0.10–0.14	0.18–0.25	6.13– 8.52
Range	0.18–3.10	ND–0.81	ND–0.13	ND–0.92	ND–0.11	ND–0.38	ND–0.13	ND–0.67	ND–0.98	ND–0.40	ND–0.57	0.38–20.08
DEVEAUX BANK												
	1.24	0.36	—	0.39	—	—	—	0.19	0.10	0.12	0.22	1.77
	3 03	0.68	—	0.63	—	0.19	—	0.15	0.11	0.15	0.54	1.84
	1.70	0.37	—	0.40	—	—	—	0.15	0.17	0.12	0.22	3.49
	2.51	0.91	—	0.55	—	—	—	0.25	0.17	0.13	0.50	7.32
	1.34	0.37	—	0.41	—	—	—	0.16	0.15	0.12	0.13	2.60
	1.35	0.32	—	0.34	—	—	—	0.13	0.14	0.12	0.24	2.69
	2.03	0.54	—	0.59	—	—	—	0.19	0.19	0.16	0.40	3.96
	0.91	0.33	—	0.21	—	—	—	0.14	—	—	0.11	2.51
	1.34	0.41	—	0.29	—	—	—	0.17	—	0.10	0.21	2.43
	0.39	—	—	—	—	—	—	—	0.09	—	—	1.70

(Continued next page)

TABLE 7 (cont'd.). *Organochlorine residues in brown pelican eggs, South Carolina, 1975*

	Residues, μg/g fresh wet weight											
	DDE	TDE	DDT	Dieldrin	Heptachlor Epoxide	Mirex	Oxy-chlordane	*cis*-Chlordane	*trans*-Nona-chlor	*cis*-Nona-chlor	Toxaphene	PCB
	0.65	0.11	—	0.11	—	—	0.13	0.13	0.11	0.07	0.09	6.82
	—	0.29	—	0.34	—	—	—	0.17	0.24	0.16	0.22	3.92
	0.10	0.80	—	0.64	—	—	—	0.29	0.29	0.18	0.31	4.20
	1.02	0.26	—	0.28	—	—	—	0.14	0.19	—	0.11	3.07
	1.62	0.45	—	0.40	—	—	—	0.16	0.20	0.14	0.38	3.60
	0.88	0.31	—	0.21	—	—	—	0.17	—	—	0.18	1.54
	3.69	0.10	—	0.97	—	—	—	0.59	—	0.35	0.35	5.93
	1.00	0.34	—	0.27	—	0.10	—	0.12	—	—	0.22	1.98
	1.52	0.41	—	0.97	—	0.14	—	0.15	0.27	0.11	0.36	3.63
	0.50	—	—	0.11	—	—	—	—	—	—	0.21	4.37
	1.78	0.60	—	0.44	—	—	—	0.24	0.33	0.18	0.58	13.56
	1.48	0.35	—	0.29	—	—	—	0.16	0.22	0.14	0.31	9.66
	0.97	0.35	—	0.33	—	—	—	0.17	0.19	0.10	0.20	5.55
	1.99	0.52	—	0.39	—	—	—	0.18	0.24	0 14	0.37	7.88
	1.19	0.30	—	0.35	—	—	—	0.13	0.18	0.11	0.40	10.81
	1.02	0.42	—	0.35	0.10	—	—	0.17	0.22	0.11	0.14	8.40
	1.23	0.31	—	0.33	—	—	—	0.15	0.22	0.14	0.10	6.86
	1.59	0.54	—	0.45	—	—	—	0.20	0.27	0.14	0.25	5.19
	1.40	0.38	—	0.34	—	—	—	0.19	0.26	0.14	0.15	9.88
	1.99	0.51	—	0.51	0.12	—	—	0.19	0.27	0 16	0.12	9.86
	2.48	0.72	—	0.46	0.10	—	—	0.23	0.34	0.18	0.27	10.25
	1.00	0.20	—	0.21	—	—	—	0.10	0.12	0.09	—	7.92
	1.73	0.22	—	0.43	—	—	—	0.14	0.19	0.12	—	10.43
	1.28	0.20	—	0.35	—	—	0.10	0.25	0.16	0.09	—	7.22
	0.76	0.23	—	0.20	—	—	—	0.10	0.13	—	0.23	7.93
	2.09	0.38	—	0.53	0.10	—	0.13	0.26	0.28	0.13	0.26	10.03
	3.04	0.58	—	0.40	0.14	—	—	0.96	—	0.20	1.27	11.46
	1.90	0.41	—	0.63	—	—	—	0.46	0.16	0.12	0.09	3.90
	2.91	0.78	—	0.70	0.20	—	—	0.24	—	0.20	1.02	6.06
	3.62	1.38	—	1.04	0.50	—	0.10	0.61	0.68	0.22	0.38	6.11
	1.86	0.44	—	0.46	0.14	—	—	0.13	0.28	0.14	0.51	4.88
	3.13	0.96	—	0.96	0.31	—	—	0.36	0.53	0.31	0.67	10.10
	2.22	0.38	—	0.68	0.21	—	—	0.28	0.40	0.17	0.57	6.37
GM	1.29	0.38		0.38				0.18	0.16	0.12	0.23	5.07
CL	0.99–1.67	0.31–0.47		0.31–0.46				0.16–0.22	0.13–0.20	0.10–0.14	0.18–0.30	4.20– 6.1
Range	ND–3.69	ND–1.38	ND	ND–1.04	ND–0.50	ND–0.19	ND–0.13	ND–0.96	ND–0.68	ND–0.35	ND–1.27	1.54–13.5
	MARSH ISLAND AND DEVEAUX BANK											
GM	1.29	0.36		0.36				0.17	0.15	0.12	0.22	6.24
CL	1.14–1.47	0.32–0.41		0.32–0.40				0.15–0.19	0.14–0.17	0.11–0.13	0.19–0.25	5.50– 7.0
Range	ND–3.69	ND–1.38	ND–0.13	ND–1.04	ND–0.50	ND–0.38	ND–0.13	ND–0.96	ND–0.98	ND–0.40	ND–1.27	0.38–20.0

NOTE: ND or — = no residues detected.
GM = geometric mean.
CL = 95 percent confidence limits.

TABLE 8. *Organochlorine residues in brown pelican eggs, Florida, 1974*

Colony	Residues, μg/g fresh wet weight									
	DDE	TDE	PCBs	Heptachlor Epoxide	Mirex	*cis*-Chlordane	*trans*-Nonachlor	*cis*-Nonachlor	Toxaphene	Dieldrin
GULF COAST										
Cedar Key	—	0.11	0.91	—	—	—	—	—	—	—
	0.16	—	0.36	—	—	—	—	—	—	—
	0.23	—	0.86	—	—	—	—	—	—	—
	0.52	—	1.60	—	—	—	—	—	—	0.10
	0.47	—	1.20	—	—	—	—	—	—	—
	0.29	—	0.56	—	—	—	—	—	—	—
	0.99	0.31	0.75	—	—	0.10	—	—	—	0.14
	0.20	—	0.47	—	—	—	—	—	—	—
	0.64	0.18	1.40	—	—	—	—	—	—	—
	0.42	—	0.65	—	—	0.14	0.12	0.10	—	0.21
	0.24	—	0.69	—	—	—	—	—	—	—
	—	—	0.69	—	—	—	—	—	—	0.16
	—	0.14	0.79	—	—	—	—	—	—	0.58
	—	—	0.44	—	—	—	—	—	—	0.44
	0.24	—	1.10	—	—	—	—	—	—	0.19
GM	0.29		0.80							
CL	0.16–0.43		0.62– 1.00							
Range	ND–0.64	ND–0.31	0.36– 1.60	ND	ND	ND–0.14	ND–0.12	ND–0.10	ND	ND–0.58
Cortez	0.51	0.10	2.10	—	—	0.20	—	0.13	—	0.18
	0.33	—	1.00	—	—	—	—	—	—	0.14
	0.57	0.18	2.00	—	—	0.27	—	0.15	—	0.19
	1.00	0.12	1.80	—	—	—	—	—	—	0.12
	0.64	0.14	1.10	—	—	—	1.00	—	—	0.15
	0.39	0.13	1.10	—	—	0.10	0.13	—	—	0.15
	0.37	0.10	1.00	—	—	0.15	0.19	—	—	0.11
	0.12	0.10	10.30	—	—	—	—	—	—	—
	0.31	—	1.50	—	—	0.33	0.23	0.18	—	0.38
	1.47	0.20	2.20	—	—	0.14	0.17	0.10	—	0.23
	0.35	0.11	0.99	—	—	0.15	0.11	—	—	0.18
	—	0.11	3.90	—	—	0.17	—	—	—	0.10
	0.66	0.22	0.75	—	—	0.19	0.17	0.12	—	0.19
	—	0.15	1.40	—	—	0.29	0.20	0.10	—	0.17
	0.52	0.14	1.20	—	—	0.23	0.19	0.11	—	0.20
GM	0.45	0.12	1.74			0.14	0.14	0.06		0.16
CL	0.28–0.65	0.08–0.15	1.11– 2.57			0.08–0.20	0.04–0.26	0.02–0.10		0.12–0.20
Range	ND–1.47	ND–0.22	0.75–10.30	ND	ND	ND–0.33	ND–1.00	ND–0.18	ND	ND–0.38
Bird Key	0.15	—	0.60	—	—	—	—	—	0.60	—
	0.29	—	0.41	—	—	—	—	—	—	0.11
	0.26	0.10	0.15	—	—	—	—	—	—	0.12
	0.55	0.16	1.70	—	—	0.15	0.16	0.10	—	0.18
	0.57	0.11	1.60	—	—	0.17	0.22	0.13	—	0.25
	—	0.19	0.25	—	—	0.19	—	—	—	—
	0.59	0.13	1.20	—	—	0.10	—	—	—	0.17
	0.30	—	1.00	—	—	0.10	—	—	—	0.13
	0.22	—	0.33	—	—	0.10	—	—	—	0.11
	0.33	0.13	3.20	—	—	—	—	—	—	0.15
	0.20	—	1.20	—	0.31	0.17	—	—	—	0.10
	—	—	1.20	—	0.10	0.09	—	—	—	—
	0.31	0.14	2.80	—	—	—	—	—	—	0.19
	0.22	—	1.50	—	0.19	—	—	0.11	—	—
	0.12	—	0.51	—	—	—	—	—	0.44	—
GM	0.27	0.06	1.02			0.07				0.10
CL	0.18–0.37	0.02–0.10	0.63– 1.51			0.03–0.11				0.07–0.14
Range	ND–0.59	ND–0.19	0.15– 2.80	ND	ND–0.31	ND–0.19	ND–0.22	ND–0.13	ND–0.60	ND–0.25
Hemp Island	1.05	0.41	4.10	0.13	0.10	0.83	0.48	0.44	—	0.65
	0.16	—	0.94	—	—	—	—	—	—	—
	0.70	0.30	1.50	—	—	0.45	0.33	0.18	—	0.30
	0.28	—	0.61	—	—	—	—	—	—	0.10
	—	—	0.25	—	—	—	—	—	—	—
	0.52	—	1.10	—	—	—	—	—	—	—
	0.58	—	0.60	—	—	—	—	—	—	0.22
	0.16	—	0.80	—	—	—	—	—	—	—
	0.73	—	3.30	—	—	0.15	0.18	—	—	0.26
	0.60	0.13	1.40	—	—	0.13	0.15	—	—	0.19
	0.63	0.11	1.80	—	—	0.13	0.14	—	0.23	0.15
	0.29	—	1.70	—	—	—	—	—	—	0.12
	0.23	—	0.50	—	—	—	—	—	—	—
	0.31	0.13	1.30	—	—	—	—	—	—	0.11
	0.40	—	1.30	—	—	—	0.10	—	—	0.10
GM	0.42		1.24							0.12
CL	0.29–0.58		0.82– 1.77							0.08–0.18
Range	ND–1.05	ND–0.41	0.50–4.10	ND–0.13	ND–0.10	ND–0.83	ND–0.48	ND–0.44	ND–0.23	ND–0.65

(Continued next page)

TABLE 8 (Cont'd.). *Organochlorine residues in brown pelican eggs, Florida, 1974*

Colony	Residues, μg/g fresh wet weight									
	DDE	TDE	PCBs	Heptachlor Epoxide	Mirex	*cis*-Chlordane	*trans*-Nonachlor	*cis*-Nonachlor	Toxaphene	Dieldrin
FLORIDA BAY										
Marquesas Key	0.13	—	—	—	—	—	—	—	—	—
	0.64	—	—	—	—	—	—	—	0.12	0.14
	0.23	—	0.41	—	—	—	—	—	—	—
	0.11	—	—	—	—	—	—	—	—	—
	0.42	—	—	—	—	—	—	—	—	—
	0.44	—	1.29	—	—	—	—	—	—	0.10
	1.05	0.14	1.60	—	—	—	—	—	0.14	0.14
	0.25	—	0.83	—	—	—	—	—	—	—
	0.42	—	1.06	—	—	—	—	—	—	—
GM	0.39		0.47							
CL	0.19–0.61		0.08– 1.00							
Range	0.11–1.05	ND–0.14	ND– 1.60	ND	ND	ND	ND	ND	ND–0.14	ND–0.14
Fanny Key	0.37	0.10	2.41	—	—	—	—	—	—	0.12
	0.19	—	0.85	—	—	—	—	—	—	—
ATLANTIC COAST										
Fort Pierce	1.61	0.31	10.90	—	—	0.16	0.18	0.11	—	0.26
	0.91	0.12	4.66	—	—	—	—	—	—	0.19
	1.47	0.34	8.89	—	—	0.10	—	0.13	—	0.42
	1.16	0.31	6.63	—	—	0.14	—	0.11	—	0.40
	0.60	0.19	3.98	—	—	—	—	—	—	0.24
	1.15	0.22	7.79	—	—	0.21	—	0.12	—	0.28
	2.15	0.65	12.92	—	—	0.21	0.11	0.20	0.20	0.41
	1.19	0.38	10.47	0.12	—	0.22	0.14	0.16	—	0.40
GM	1.24	0.31	7.79			0.13		0.10		0.31
CL	0.89–1.66	0.19–0.44	5.49–10.91			0.05–0.21		0.04–0.16		0.24–0.40
Range	0.60–2.15	0.12–0.65	3.98–12.92	ND–0.12	ND	ND–0.22	ND–0.18	ND–0.20	ND–0.20	0.19–0.42
Cocoa Beach	0.67	0.14	2.98	—	—	—	—	—	—	0.20
	1.81	0.45	7.80	—	—	0.16	0.12	0.14	0.25	0.35
	1.39	0.29	6.10	—	—	0.16	—	0.18	0.18	0.44
	1.13	0.32	4.77	—	—	0.15	0.17	0.13	0.14	0.34
	0.72	0.13	2.40	—	—	—	—	—	—	0.14
	0.44	0.19	5.16	—	—	—	—	—	0.21	0.27
	0.74	0.18	5.43	—	—	0.16	0.13	0.15	—	0.25
	0.85	0.23	3.00	—	—	0.13	—	0.11	0.10	0.41
	1.73	0.40	5.58	—	—	0.30	0.15	0.23	0.21	0.70
	1.72	0.40	8.35	—	—	—	0.13	—	—	0.38
	1.20	0.27	4.76	—	—	0.15	0.15	0.16	0.22	0.41
	1.20	0.28	7.77	—	—	0.16	0.10	0.13	0.21	0.33
	3.40	0.78	9.22	—	—	0.24	0.18	0.20	0.46	0.78
	0.94	0.29	3.38	—	—	—	—	—	0.44	0.23
	0.49	0.17	2.83	—	—	0.12	—	0.10	—	0.18
GM	1.13	0.29	4.94			0.11	0.07	0.10	0.15	0.32
CL	0.82–1.51	0.21–0.38	3.89– 6.24			0.06–0.16	0.03–0.11	0.05–0.15	0.07–0.24	0.25–0.42
Range	0.44–1.81	0.13–0.78	2.40– 9.22	ND	ND	ND–0.30	ND–0.18	ND–0.23	ND–0.46	0.14–0.78
Pelican Island	0.99	0.25	2.48	—	—	—	—	—	0.22	0.15
	1.33	0.25	5.77	—	—	0.19	0.12	0.20	—	0.33
	1.25	0.28	4.18	—	—	0.15	—	0.13	0.14	0.23
	1.01	0.16	3.07	—	—	—	—	—	—	0.26
	0.72	0.21	4.67	—	—	0.13	0.11	—	—	0.23
	1.11	0.35	6.36	—	—	0.13	0.16	—	—	0.31
	1.40	0.46	7.25	—	—	0.16	0.16	0.13	0.27	0.46
	1.77	0.62	9.73	0.11	—	0.18	0.20	0.15	0.30	0.60
	1.03	0.49	9.06	—	—	0.14	0.13	0.13	0.12	0.42
	1.40	0.60	9.52	—	—	0.35	0.30	0.25	—	0.65
	1.40	0.35	9.24	—	—	0.19	0.17	0.12	—	0.45
	0.49	—	2.71	—	—	—	—	—	—	0.13
	1.72	0.50	9.26	—	—	0.15	0.17	0.14	0.11	0.48
	0.66	0.13	1.98	—	—	—	0.12	—	—	0.15
GM	1.13	0.32	5.47			0.12	0.11	0.09		0.31
CL	0.92–1.37	0.22–0.43	3.96– 7.45			0.07–0.18	0.06–0.17	0.04–0.14		0.23–0.42
Range	0.49–1.77	ND–0.62	1.98– 9.73	ND–0.11	ND	ND–0.35	ND–0.30	ND–0.25	ND–0.30	0.13–0.60
Port Orange	1.67	0.43	10.01	—	—	0.18	0.17	0.10	0.26	0.43
	0.91	0.14	4.97	—	—	0.10	0.10	—	—	0.46
	1.80	0.65	7.80	—	—	0.24	0.21	0.14	0.23	0.54
	2.11	0.59	11.27	—	—	0.18	0.22	0.13	0.33	0.47
	2.64	0.74	11.78	0.10	—	0.53	0.34	0.26	0.29	0.62
	1.55	0.38	8.10	—	—	0.17	0.16	0.11	0.24	0.38

(Continued next page)

TABLE 8 (Cont'd.). *Organochlorine residues in brown pelican eggs, Florida, 1974*

Colony	Residues, μg/g fresh wet weight									
	DDE	TDE	PCBs	Heptachlor Epoxide	Mirex	*cis*-Chlordane	*trans*-Nonachlor	*cis*-Nonachlor	Toxaphene	Dieldrin
	1.02	0.12	5.45	—	—	—	—	—	—	0.28
	1.16	0.34	6.05	—	—	0.24	0.15	0.11	0.12	0.31
	1.04	0.38	4.90	—	—	—	—	—	0.13	0.30
	0.45	0.10	4.17	—	—	—	—	—	—	0.18
	1.51	0.45	8.42	—	—	0.47	0.14	0.11	0.23	0.42
	1.00	0.37	8.30	—	—	—	—	—	0.41	0.83
	1.94	0.46	7.43	—	—	0.31	0.33	0.33	1.53	0.46
	1.34	0.22	9.70	—	—	—	0.12	—	0.18	0.28
GM	1.32	0.33	7.39			0.14	0.12	0.09	0.19	0.40
CL	1.03–1.71	0.23–0.47	6.13–8.91			0.08–0.23	0.08–0.19	0.06–0.13	0.11–0.32	0.32–0.50
Range	0.45–2.64	0.10–0.74	4.27–11.78	ND–0.10	ND	ND–0.53	ND–0.34	ND–0.33	ND–1.53	0.18–0.83

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

TABLE 9. *Trends for organochlorine residues in brown pelican eggs, Deveaux Bank and Marsh Island, South Carolina, 1969–75*

Year	Sample Size	Residues, μg/g fresh wet weight					
		DDE	TDE	DDT	Σ DDT	Dieldrin	PCBs
1969	15	5.45[1] A[2] (4.44–6.70)	1.65 A (1.30–2.10)	0.45 A (0.15–0.83)	7.81 A (6.48–9.40)	1.16 A (1.03–1.52)	6.11 AB (5.00–7.45)
1970	13	3.58 B (2.23–5.72)	0.79 B (0.53–1.20)	0.55 A (0.42–0.69)	5.27 B (3.49–7.77)	0.82 B (0.52–1.32)	5.25 AB (3.92–7.04)
1971	65	2.48 C (2.27–2.71)	0.48 C (0.43–0.53)	0.17 B (0.13–0.21)	3.20 D (2.94–3.48)	0.46 C (0.40–0.52)	6.49 A (5.44–7.73)
1972	72	3.03 B (2.70–3.40)	0.36 C (0.31–0.42)	0.18 B (0.15–0.21)	3.69 C (3.31–4.12)	0.45 C (0.39–0.52)	7.51 A (6.68–8.46)
1973	104	2.09 D (1.91–2.29)	0.19 D (0.17–0.22)	0.17 B (0.15–0.20)	2.56 E (2.35–2.78)	0.45 C (0.41–0.50)	4.75 B (4.26–5.31)
1974	115	2.22 CD (2.03–2.43)	0.49 C (0.44–0.54)	0.02 C (0.01–0.04)	2.72 E (2.49–2.96)	0.58 C (0.53–0.64)	7.63 A (6.80–8.55)
1975	102	1.40 E (1.27–1.54)	0.41 C (0.37–0.46)	0.004 C (0.002–0.007)	1.80 F (1.64–1.97)	0.40 C (0.36–0.43)	6.45 A (5.75–7.24)

[1] Geometric mean; 95 percent confidence limits are in parentheses.
[2] See Footnote 2, Table 3.

The factors underlying the large population increase were not evident. The excellent reproductive success in 1973 cannot account for the large population increase just 2 years later. It is possible that many South Carolina adults did not breed before 1975 because of insufficient food. Many adult brown pelicans in Mexico and California apparently do not breed when the food supply is poor (*2*). The breeding population in South Carolina showed only a slight increase in 1973 when pelicans had an excellent reproductive season and menhaden were apparently readily available. Thus it is doubtful that large numbers of adult pelicans in South Carolina failed to breed from 1969 to 1974. There is no evidence from banding studies that large numbers of pelicans migrated from natal areas in Florida to South Carolina to breed. Although the population increase was probably caused by a combination of factors, the most likely factor seems to be the decline in organochlorine residues that resulted in improved reproductive success and probable increased longevity after fledging.

DDE is the organochlorine exerting most influence on reproductive success. However, little is known about adult mortality from organochlorines except that several

TABLE 10. *Organochlorine residue trends in brown pelican eggs from four regions, 1969–70, 1974*

Pollutant	Region[1]	Mean residues, μg/g fresh wet weight	
		1969–70	1974
DDE	SC	4.65 A[2]	2.22 B
	AC	2.32 B	1.21 C
	FB	1.04 C	0.37 D
	GC	1.48 C	0.36 D
TDE	SC	1.29 A	0.49 C
	AC	0.91 B	0.32 D
	FB	0.18 E	0.03 E
	GC	0.55 C	0.07 E
DDT	SC	0.49 A	0.02 C
	AC	0.43 A	0.01 C
	FB	0.07 C	ND C
	GC	0.27 B	ND C
Σ DDT	SC	6.52 A	2.72 C
	AC	3.68 B	1.52 D
	FB	1.25 D	0.39 E
	GC	2.27 C	0.42 E
Dieldrin	SC	1.09 A	0.58 B
	AC	0.51 B	0.36 C
	FB	0.06 D	0.04 D
	GC	0.11 D	0.13 D
PCBs	SC	5.77 B	7.63 A
	AC	2.68 C	6.12 AB
	FB	0.75 D	0.62 D
	GC	0.70 D	1.18 D

[1] SC = South Carolina, AC = Florida Atlantic Coast, FB = Florida Bay, and GC = Florida Gulf Coast.
[2] See Footnote 2, Table 3.

TABLE 11. *Organochlorine residues in tissues of brown pelicans found dead, South Carolina, 1974–75*

Year	Sex	Age	Tissue	Residues, μg/g fresh wet weight										
				DDE	TDE	DDT	Dieldrin	Heptachlor Epoxide	*cis*-Chlordane	*trans*-Nonachlor	*cis*-Nonachlor	Toxaphene	Mirex	PCBs
1974	F	4 wk	Carcass	0.14	—	—	—	—	—	—	—	—	—	0.25
			Brain	—	—	—	—	—	—	—	—	—	—	0.25
	F	6 wk	Carcass	0.16	—	—	—	—	—	—	—	—	—	1.44
			Brain	0.52	—	—	0.14	—	—	—	—	—	—	2.46
	F	12 wk	Carcass	0.46	0.16	—	0.13	—	—	—	—	—	—	1.27
			Brain	—	—	—	—	—	—	—	—	—	—	0.74
	M	8 wk	Carcass	0.15	—	—	—	—	—	—	—	—	—	1.28
			Brain	0.20	—	—	—	—	—	—	—	—	—	1.58
1975	M	AD	Carcass	3.09	1.25	0.14	1.67	0.22	0.44	0.56	0.47	0.56	1.40	25.28
			Brain	3.43	0.55	—	0.99	0.10	0.26	0.20	0.22	0.48	0.64	14.22
	M	AD	Carcass	3.24	1.56	0.14	1.87	0.26	0.62	0.92	0.71	0.73	1.80	38.80
			Brain	1.31	0.54	—	0.91	—	—	0.19	0.23	0.54	0.87	12.83
	M	AD	Brain	1.46	0.61	—	0.99	0.10	0.27	0.31	0.27	0.47	0.65	2.92

NOTE: — = no residues detected.
AD = adult.

TABLE 12. *Organochlorine residues in Atlantic menhaden regurgitated by brown pelicans, South Carolina, 1974–75*

Year		Residues, μg/g fresh wet weight							
		DDE	TDE	Heptachlor Epoxide	*cis*-Chlordane	*cis*-Nonachlor	*trans*-Nonachlor	Toxaphene	PCBs
1974		0.01	—	—	—	—	—	—	0.23
		0.04	—	—	—	—	—	—	0.19
		0.01	—	—	—	—	—	—	0.02
		0.06	0.04	0.02	—	0.01	—	—	0.36
		—	0.01	—	—	—	—	—	0.22
	GM	0.016							0.147
	CL	0.004–0.060							0.036–0.608
1975		—	—	—	—	—	—	—	—
		0.03	0.04	0.03	0.02	0.02	—	0.06	0.03
		0.01	0.03	—	—	—	—	0.02	0.03
		0.02	0.06	—	0.02	—	—	0.02	0.09
		0.03	0.06	0.03	—	0.01	—	—	0.08
		0.01	0.01	—	—	—	—	—	0.02
		0.02	0.03	—	0.01	—	0.01	—	0.10
		0.01	—	—	—	—	—	—	0.10
		0.02	0.03	—	0.01	—	0.01	0.02	0.15
		0.01	0.01	—	—	—	—	0.01	0.11
	GM	0.014	0.020						0.050
	CL	0.009–0.022	0.010–0.039						0.024–0.107

NOTE: — = no residues detected.
GM = geometric mean.
CL = 95 percent confidence limit.

pelicans have died of endrin and dieldrin poisoning. An increase in adult survival would have a marked effect on the breeding population and on the recruitment standard necessary to maintain a stable population. There are no data to support the theory of increased adult longevity, but it may be investigated in the future by analyzing banding data.

The South Carolina brown pelican population formerly numbered about 6,000 breeding pairs (*3, 10*), and if the present rate of reproductive success continues, the population should reach 6,000 breeding pairs within the next 5 years. The pelican population in Florida has been essentially stable since aerial surveys of nesting colonies were initiated in 1968 (*18, 21*).

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Pesticide Contamination of Water Rats in the Murrumbidgee Irrigation Areas, New South Wales, Australia, 1970–72

Penny Olsen [1] and Harry Settle [2]

ABSTRACT

Organochlorine pesticides were found in all samples of livers, kidneys, mammary glands, and fetuses of eastern water rats (Hydromys chrysogaster) *collected in the Murrumbidgee irrigation areas of New South Wales in 1970 and 1972. DDE was the predominant residue. Livers contained 0.01–3.10 ppm ΣDDT air-dried weight; kidneys, < 0.01–1.12 ppm; mammary glands, 0.14–23.75 ppm; and fetal liver, 0.28–0.66 ppm. Variations in residue levels are discussed in relation to the possible effects of environmental and physiological factors.*

Introduction

Large amounts of water are used in the Murrumbidgee irrigation areas of New South Wales for flood irrigation of rice crops. Drainage water from these crops and from irrigated orchards, vineyards, and cereal and vegetable crops enters Mirrool Creek. A weir, Willow Dam, controls entry of the creek's water into a storage swamp or diverts it for further irrigation use.

Several pesticides are used on area farms, although DDT predominates. About 1–4.5 kg/ha. is used annually (*2*), largely to control the bloodworm (*Chironomus* sp.) which damages rice seedlings. Eastern water rats (*Hydromys chrysogaster*), common in the irrigation area, were collected monthly from Mirrool Creek and Willow Dam as part of a study of the biology of the species.

Little is known of pesticide contamination of Australian fauna (*2*). The present study is a preliminary examination of the degree of exposure of water rats to pesticides.

Materials and Methods

SAMPLE COLLECTION

Eastern water rats were live-trapped from Mirrool Creek at Willow Dam near Griffith, New South Wales, between January 1970 and January 1973. Livers, kidneys, mammary glands, and fetuses were removed from the freshly killed rats and preserved in 10 percent formalin. A small number of samples taken during 1970 and 1972 were analyzed for pesticides as follows: in 1970, January (7), April (3), October (3), November (4); in 1972, February (2), May (2), July (7), August (6). Sampling pattern is illustrated in Figure 1.

ANALYSIS

In the laboratory, samples were drained and air dried, cut into small pieces, mixed with sodium sulfate, and extracted with hexane in a Soxhlet thimble for 4 hours. Extraction for a longer period did not increase residue recovery. The hexane extracts were concentrated to about 10 ml and partitioned three times with 25 ml acetonitrile as a preliminary cleanup. The acetonitrile phase was passed into 300 ml 2 percent sodium sulfate and shaken with 100 ml hexane. The hexane layer was dried by passing it through anhydrous sodium sulfate and was concentrated to 5 ml. The concentrate was mixed with 20 g 2 percent deactivated Florisil, poured into a chromatographic column containing 20 g 2 percent deactivated Florisil, and eluted in three fractions (*5*, *6*) as follows:

Fraction A, eluted with 200 ml 20 percent methylene chloride–hexane, was analyzed for lindane, HCB, aldrin, heptachlor, heptachlor epoxide, DDE, TDE, DDT, and polychlorinated biphenyls (PCBs).

Fraction B, eluted with 200 ml 20 percent methylene chloride–hexane, was analyzed for dieldrin, dursban, and trithion.

Fraction C, eluted with 200 ml acetone, was analyzed for malathion, ethion, delnav, and diazinon.

The eluates were concentrated to 1 ml. Fractions A and B were examined by injection into a Varian Model 2700 gas-liquid chromatograph fitted with a tritium electron-capture detector. Fraction C was injected into

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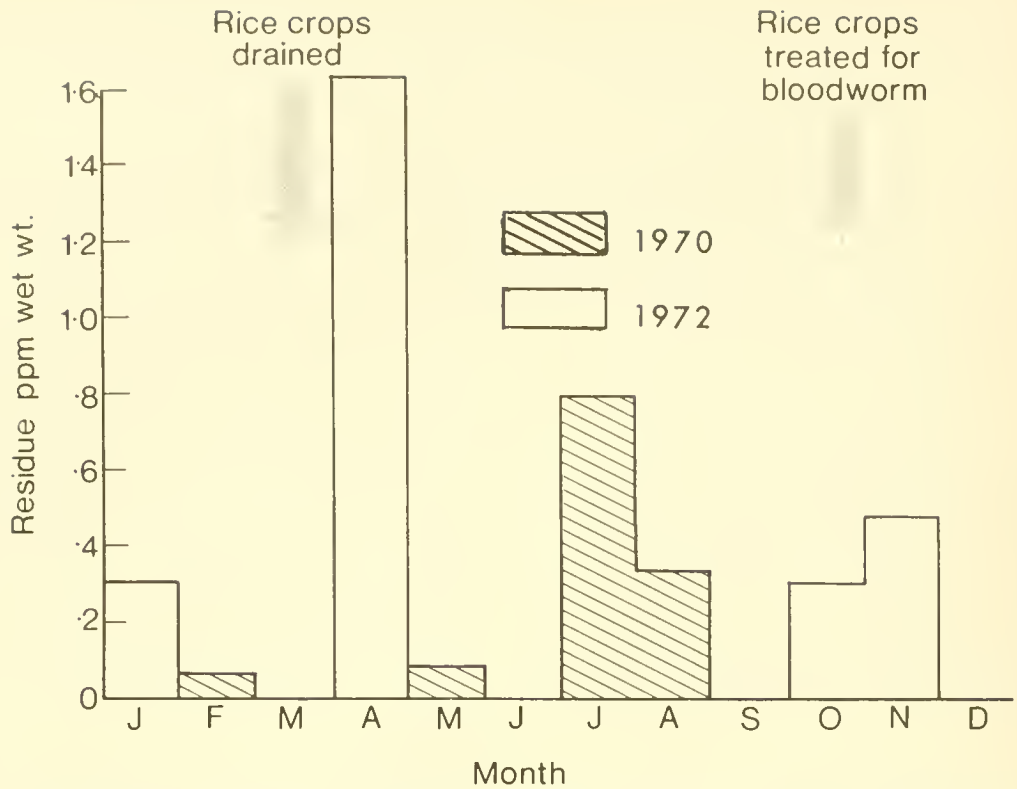

FIGURE 1. *Mean organochlorine content of eastern water rat livers by month sampled, Murrumbidgee irrigation area, New South Wales, Australia, 1970–72*
(ΣDDT represented at least 94 percent of residues in each month. Number of samples analyzed each month was 7, 2, 3, 2, 7, 6, 3, 4, respectively.)

a Tracor gas-liquid chromatograph fitted with a phosphorus-mode flame photometric detector (Table 1). Residues detected at 0.005 ppm and above were reported to the nearest 0.01 ppm.

CONFIRMATION OF RESIDUES AND RECOVERIES

All samples having organochlorine residues greater than 0.1 ppm were spotted on a thin-layer chromatographic plate for confirmation. Blank analyses were carried out at frequent intervals from the sodium sulfate/Soxhlet step. Replicate recoveries (Table 2) were carried out by adding known amounts of organochlorine and organophosphorus pesticides to sodium sulfate in the Soxhlet thimble and treating the recovery as in the sample procedure.

TABLE 1. *Parameters for gas-liquid chromatographic analyses for pesticides in eastern water rats, 1970–72*

	Varian 2700	Tracor 550
Detector	tritium	FPD (P mode)
Columns	glass ⅛-inch I.D., 6 foot effective length	
Column packing	a mixture of 0.2% DC-200 and 0.8% QF-1 on Varaport 30	3% OV-1 on Gas-Chrom Q
Temperatures, °C		
column	200	220
inlet	220	230
detector	220	170
Carrier gas flow (ml minute)		
nitrogen	30	60
hydrogen		50
air		100

Because a one-step cleanup was not sufficient, the acetonitrile–hexane partition method was used (*6*). This results in low HCB recoveries; consequently HCB results were corrected for recovery as follows:

$$\text{HCB (reported)} = \text{HCB found in determination} \times (100/33)$$

Results and Discussion

Pesticide residues detected in the water rats are listed in Table 3. All samples contained organochlorines and an unidentified organophosphorus compound. There were no significant differences in residue levels between males and females. Mammary glands, because of their fatty composition, contained the highest levels, and residues tended to increase as parturition approached. Mammary TDE positively correlated with fetal weight

TABLE 2. *Results of replicate recoveries of organochlorines and organophosphates in eastern water rats, 1970–72*

Pesticide	Amount added, μg	No. of replicates	Mean % recovery ± std dev
HCB	0.25	6	33.0 ± 4.49
DDE	0.25	4	76.5 ± 6.70
TDE	0.25	6	89.0 ± 8.61
DDT	0.25	only 2 recoveries measured (71% and 92%)	
Dieldrin	0.25	7	83.0 ± 6.36
Malathion	1	4	98.8 ± 8.93
Diazinon	1	5	75.4 ± 13.31
Delnav	1	4	85.8 ± 4.91
Dursban	1	6	86.7 ± 7.49
Ethion	1	5	84.6 ± 4.03
Trithion	1	5	86.2 ± 7.68

($P < 0.05$). Liver TDE was correlated with mammary TDE ($P < 0.001$). Fetal residues tended to reflect maternal liver residues and were positively correlated with fetal weight ($P < 0.01$).

No significant differences in residues were found between younger and older animals and breeding and nonbreeding animals (Table 4). However, younger animals tended to carry lower levels than older animals. The nonbreeding female group was the only one which showed a positive correlation between age and residue level ($P < 0.05$). Breeding females had the highest liver pesticide loads, and nonbreeding females, mature and immature, had the lowest. Kidney residues were lower in breeders than in nonbreeders.

Stomachs of pregnant females contained more food items, particularly insects, than did those of males or nonbreeding females (*8*). This suggests that breeders may have a greater opportunity for contamination through greater food consumption and may consume more dead and dying nontarget arthropods weakened by insecticides, as demonstrated by Stehn in small mammal scavengers (*7*). Lower liver residues in nonbreeding females and increasing residues in mammary glands as parturition approached suggested a lowering of body burdens through mobilization of fat during pregnancy and lactation; this phenomenon is thought to occur in harbor porpoises (*3*) and Arctic ringed seals (*1*).

Seasonal changes in residue levels may be related to irrigation and pesticide application practices in the area. Peak residues occurred in animals in April after water had been drained from the rice fields in March (Fig. 1). Because DDT has a low water solubility and deposits out of suspension to be adsorbed on organic matter, plants, and sediments (*4*), increased amounts may be available to water rat prey in the dry soil of drained rice fields and, particularly, through flushing of water with suspended clay, organic matter, and plant material into the creek.

Up to 8 ppm DDT has been found in sediments of drainage channels adjacent to the rice bays, indicating considerable movement of the pesticide from the site of application (K. H. Bowmer, Division of Irrigation Research, 1974, personal communication). Fish and aquatic insects may also be flushed from the bays or may be stranded in drained fields, becoming easy prey. A smaller peak in residue levels in November coincides with the treatment of rice for bloodworm.

Corresponding with the April peak residue levels, there was a seasonal decline in weight of the rats which may indicate a breakdown of body fats and consequent release of stored pesticides. Because trophic level is thought to be one factor in biomagnification of residues,

TABLE 3. *Pesticide residues in liver, kidney, mammary glands, and fetal liver samples from water rats, Murrumbidgee irrigation areas, New South Wales, Australia, 1970–72*

Tissue	No. Samples	Residues, ppm air-dried wt (± std dev.) (Range) DDE	TDE	DDT	Dieldrin	HCB	ΣDDT
Females							
Livers[1]	17	0.40 ± 0.14 (2.10–0.01)	0.09 ± 0.05 (0.85–ND)	0.01 (0.06–ND)	0.01 ± 0.01 (0.09–ND)	0.03 ± 0.02 (0.40–ND)	0.49 ± 0.19 (3.10–0.01)
Kidneys	12	0.17 ± 0.03 (0.46–ND)	0.03 ± 0.01 (0.15–ND)	ND	ND	0.01 ± 0.01 (0.11–ND)	0.20 ± 0.03 (0.57–0.06)
Mammary glands	6	5.07 ± 2.22 (12.20–0.13)	2.11 ± 1.68 (10.40–ND)	0.64 ± 0.37 (2.23–ND)	0.01 ± 0.01 (0.05–ND)	ND	7.82 ± 3.86 (23.75–0.14)
Fetal liver	2	0.30 (0.38–0.22)	0.13 (0.23–0.02)	0.05 (0.05–0.04)	0.04 (0.05–0.02)	ND	0.47 (0.66–0.28)
Males							
Livers	17	0.44 ± 0.10 (1.57–0.02)	0.05 ± 0.02 (0.22–ND)	ND (0.04–ND)	0.01 (0.04–ND)	ND (0.08–ND)	0.49 ± 0.11 (1.61–0.02)
Kidneys	13	0.31 ± 0.06 (0.89–ND)	0.02 ± 0.01 (0.20–ND)	ND	0.01 (0.04–ND)	0.01 (0.08–ND)	0.33 ± 0.07 (1.12–ND)

NOTE: PCBs not detected in any sample; ND = <0.01 ppm.
[1]One liver with 0.01 ppm malathion.

TABLE 4. *Differences in organochlorine residues in livers and kidneys of eastern water rats, Murrumbidgee irrigation areas, New South Wales, Australia, 1970–72*

	Mean total residues, ppm wet wt	
	Liver	Kidney
Females		
Est. age < 6 months	0.39 (5)	0.10 (2)
Est. age ≥ 6 months	0.64 (12)	0.26 (10)
Nonbreeding	0.40 (11)	0.24 (8)
Pregnant only	0.83 (4)	0.18 (3)
Pregnant and lactating	0.67 (2)	0.06 (1)
Males		
Est. age < 6 months	0.33 (5)	0.36 (4)
Est. age ≥ 6 months	0.56 (12)	0.33 (9)

NOTE: Age of all animals was estimated by use of dry eye lens weights. Tests were scrotal in males 6 months or older and nonscrotal in those younger than 6 months. Number of animals used in samples is in parentheses.

it is of interest that in the months of high residue levels, April–August, vertebrates were more important in the diet and insects were less important (*8*). Although stomach and rectal contents revealed food intake over a limited period, they may represent individual preference and reflect seasonal trends. Higher residues were found in those animals with fish, mammal, bird, and crustacean remains in their guts than in those with insects and spiders ($P < 0.01$). Mean liver residues and corresponding stomach contents were as follows: mammals ($n = 3$), 0.99 ppm; fish ($n = 3$), 1.29 ppm; birds ($n = 5$), 1.04 ppm; crustaceans ($n = 2$), 1.89 ppm; spiders ($n = 4$), 0.33 ppm; and insects ($n = 9$), 0.51 ppm.

There was no significant difference between residue levels in 1970 and 1972. HCB was found in 1970 samples only. Dieldrin, found in 4 of 17 liver samples (0.01–0.03 ppm) in 1970, occurred in 7 of 17 samples in 1972 (0.01–0.09 ppm). DDT and dieldrin sales were unchanged during the study. However, in 1972, the organophosphate abate was used in more rice-growing areas for bloodworm treatment, and HCB was no longer recommended for use as a fungicide.

Data from other studies on water rats are scarce. The Australian Academy of Science (*2*) reports in its appendices that residues of ΣDDT in a water rat in Victoria were: fat, 0.50; muscle, 0.23; kidney, 0.19 ppm wet weight. However, no biological information or locality is given.

Although DDT is no longer recommended for bloodworm control, the moderate degree of contamination found in water rats and the continuing use of potentially harmful pesticides in the area point to the need for a more detailed study on the fate and ecological effect of these substances, with particular emphasis on more sensitive species.

Acknowledgments

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Organochlorine Residues in Harp Seal (Phagophilus groenlandicus) *Tissues, Gulf of St. Lawrence, 1971, 1973*[1]

K. T. Rosewell, D. C. G. Muir, and B. E. Baker

ABSTRACT

Levels of p,p'-DDT, p,p'-TDE, p,p'-DDE, dieldrin, polychlorinated biphenyls (PCBs), and HCB were determined in certain tissues of 31 harp seals (Phagophilus groenlandicus) *taken from the Gulf of St. Lawrence during 1971 and 1973. The seals ranged in age from less than two weeks to 18 years. Mean concentrations of PCBs and ΣDDT in the various tissues were about the same. ΣDDT levels were 1.64–9.88 ppm in adult seal blubber and 1.08–3.73 ppm in seal pup blubber. Organochlorine levels in harp seal samples taken in 1973 were similar to those reported by other workers for samples collected in the Gulf of St. Lawrence during 1967–71.*

Introduction

Seals occupy a top position in long food chains, and because they carry large quantities of subcutaneous fat which can store organochlorines, they have been used as indicators of pollution in the marine environment (*1, 3, 5, 7, 8, 11, 13*). Organochlorine concentrations in seals collected in 1967 and 1968 in the Gulf of St. Lawrence indicated a degree of marine pollution similar to that in European coastal waters (*10*). In the present study, harp seals (*Phagophilus groenlandicus*) from the Gulf of St. Lawrence region were examined for organochlorines to determine whether 1967–68 marine pollution levels still existed and to measure organochlorine residue levels in various tissues of adult and young harp seals.

Materials and Methods

SAMPLE COLLECTION

Tissue samples were obtained from 11 harp seals (age 1–18 years) caught in the Gulf of St. Lawrence in 1971, and 20 harp seal pups caught in the same region in 1973. All samples were frozen immediately after collection and transported to the laboratory where they were stored at −20°C until analysis. Blubber, kidney, liver, muscle, spleen, brain, and gonad tissues were taken for analysis.

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ANALYTICAL METHODS

Tissue samples obtained in 1971 were analyzed as described by Porter et al. (*16*) for their fat content in order to estimate how much tissue would contain the 1–3 g of fat required for organochlorine analysis.

An appropriate weight of each sample was dried with sodium sulfate and then extracted with petroleum ether (*16*). The petroleum ether extracts were cleaned by acetonitrile–petroleum ether partitioning and Florisil column chromatography (*17*). The 6:94 (v/v) diethyl ether:petroleum ether eluate from the Florisil column was transferred to a 4:1 (by weight) silica–Celite column (*4*) in order to separate PCBs from ΣDDT. The 15 percent eluate from the Florisil column, which contained dieldrin residues, was subjected to further cleanup in which concentrated eluate was refluxed with 2:92 (v/v) methanolic KOH (*17*).

Tissue samples from harp seal pups caught in 1973 were analyzed for fat content by the method of Holdrinet (*12*). An appropriate weight of each sample was mixed with sodium sulfate and sand and then extracted with hexane on a Soxhlet extractor. The hexane extracts were cleaned on a deactivated (2 percent) Florisil column (*12, 15*), and then were passed through a charcoal column (*12*) in order to separate PCBs and HCB from ΣDDT.

Pesticides and PCBs were determined by (^{3}H) electron-capture gas chromatography under the following conditions:

Chromatograph:	Varian Model 600D	
Columns:	(1) glass, 1.08 m × 3 mm OD, packed with a mixture of 6 percent QF-1 and 4 percent SE-30 on Chromosorb W-HP	
	(2) glass, 1.68 m × 3 mm OD, packed with 1 percent OV-1 on Chromosorb W-HP	
Temperatures, °C:	column (1)	195
	column (2)	185
Chromatograph:	Varian Model 1400	
Columns:	glass, 1.83 m × 3 mm ID packed with:	
	(1) a mixture of 6 percent QF-1 and 4 percent SE-30 on Chromosorb W-HP	
	(2) 3 percent OV-225 on Chromosorb W-HP	
Temperatures, °C:	column (1)	215
	column (2)	185

Known quantities of pesticides (*p,p'*-DDT, *p,p'*-TDE, *p,p'*-DDE, and dieldrin) and PCBs (Aroclors 1242 and

1260) were added to a sample of the sodium sulfate used to dehydrate the tissues. Extraction by the method of Porter et al. (*16*) produced recoveries of 69–102 percent for organochlorine pesticides and 69–84 percent for PCBs. The following recoveries were obtained using the method of Holdrinet (*12*): *p,p'*-DDT, *p,p'*-TDE, and *p,p'*-DDE, 85–112 percent; dieldrin, 81–89 percent; PCBs (Aroclor 1254), 84–85 percent; HCB, 78–89 percent.

Gas-liquid chromatography results were confirmed by use of two columns of different polarity, by thin layer chromatography, and by chemical derivatization. In all instances, the results were confirmed by at least two of the three procedures.

Results and Discussion

The fat content of seal tissues is shown in Table 1. Tables 2 and 3 show the results, not corrected for recovery, of analyses of the various tissues for organochlorines. ΣDDT and PCBs were detected in all samples. Dieldrin was detected in all but five tissue samples analyzed. Forty of 42 tissue samples from harp seal pups contained HCB. Blubber contained the highest levels of organochlorines. The mean PCB and ΣDDT concentrations in various tissues were about the same.

Mean HCB levels, determined only in seal pups, and mean dieldrin levels were similar in all tissues analyzed.

Brain tissue contained more extractable lipid (8.3 percent) than did liver (3.5–4.0 percent), kidney (4.2 percent), muscle (2.6 percent), and spleen (2.8 percent). Mean levels of ΣDDT and PCBs in the brain, however, were lower than in other tissues. The results suggest that a brain barrier to PCB- and DDT-type compounds may exist in the harp seal as reported by Frank et al. (*7*). This may result from a difference between the constitution of brain lipids and the lipids of depot fat. The authors suggest that a similar phenomenon may exist with dieldrin, but it was not observed in the present work.

TABLE 1. *Fat content of tissues of harp seals, Gulf of St. Lawrence—1971, 1973*

Tissue	No. Samples Analyzed	Average Fat Content, %
Blubber (adults)	5	82.5
(pups)	7	86.2
Liver (adults)	2	3.5
(pups)	3	4.0
Kidney (adults)	3	4.2
Muscle (adults)	2	2.6
Spleen (adults)	2	2.8
Brain (pups)	3	8.3
Gonad (male pups)	1	1.7
(female pups)	1	7.3

Since the types of residues in tissues of the harp seal pups were similar to those in the same tissues of older seals, it is probable that the residues in the adult seals are passed along to the fetus as well as to nursing seal pups. Holden concluded that organochlorine residues in nursing gray seal pups were derived solely from the parent seals, since the pups were still being fed by the adult females at the time of capture (*11*). This conclusion is supported by the fact that organochlorines have been found in the milk of fur seals (*2*) and harp seals (*6*).

Organochlorine levels in harp seal pups in the present study are similar to those reported previously in harp seals taken from the Gulf of St. Lawrence (*9, 10, 14*). In the present study, blubber, liver, and brain tissues of young harp seals contained PCB levels similar to and dieldrin levels higher than those found by Frank et al. (*7*). ΣDDT levels were slightly higher in the blubber and liver, but similar in brain tissue to those of pups studied by Frank et al. (*7*).

In the present study, the blubber of adult harp seals contained slightly lower levels of ΣDDT and PCBs than did those reported by Addison et al. (*1*) and Frank et al. (*7*). Muscle tissue of adult seals contained higher levels of PCBs but similar levels of ΣDDT and dieldrin. Liver tissue had lower levels of ΣDDT but higher concentrations of PCBs than did the corresponding tissue analyzed by the above authors (*1, 7*). Dieldrin concentrations in tissues analyzed for the present study were similar to those reported previously (*1, 7, 14*).

The ratio of ΣDDT to PCBs (Table 4) was close to 1.0 in all tissues except the liver, muscle, and spleen of the adult seals. This may reflect heavy use of DDT for spraying forests in areas drained by rivers flowing directly into the Gulf of St. Lawrence, as well as the high degree of urban industrial pollution which is the major source of PCBs in the environment.

Acknowledgments

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TABLE 2. *Organochlorine residues in tissues of adult harp seals, Gulf of St. Lawrence—March 1971*

Seal Number	Sex	Age, years	Tissue	Residues, ppm wet weight					
				p,p'-DDE	*p,p'*-TDE	*p,p'*-DDT	ΣDDT	Dieldrin	PCBs
1	M	11	blubber	0.680	0.359	1.096	2.135	0.320	2.05
			kidney	0.070	0.036	0.212	0.318	0.012	0.26
			liver	0.105	0.043	0.291	0.439	0.006	1.45
			muscle	0.138	0.039	0.102	0.279	0.005	0.47
			spleen	0.039	0.016	0.076	0.131	<0.002	0.16
2	F	1	blubber	0.918	0.433	8.530	9.881	0.244	0.49
			kidney	0.268	0.194	2.197	2.659	0.002	1.54
			liver	0.147	0.088	0.079	0.314	0.004	0.65
			muscle	0.048	0.017	0.063	0.128	0.002	1.10
			spleen	0.358	0.292	0.759	1.409	0.004	2.57
3	M	3	blubber	2.056	0.683	2.684	5.423	0.011	2.45
			kidney	0.145	0.060	0.206	0.411	0.005	0.86
			liver	0.060	0.075	0.147	0.282	0.007	0.37
			muscle	0.052	0.031	0.090	0.173	0.003	0.18
			spleen	0.108	0.059	0.086	0.253	0.004	0.55
4	M	5	blubber	0.726	0.631	2.550	3.907	0.024	13.30
			kidney	0.057	0.020	0.082	0.159	0.005	0.62
			liver	0.086	0.116	0.106	0.308	0.009	0.76
			muscle	0.089	0.018	0.097	0.204	0.002	0.34
5	M	6	blubber	1.187	0.459	1.280	2.926	0.096	1.96
			kidney	0.048	0.023	0.047	0.118	0.002	0.25
			liver	0.039	0.071	0.036	0.146	<0.002	0.11
			muscle	0.076	0.057	0.084	0.217	0.002	0.46
			spleen	0.056	0.038	0.156	0.250	<0.002	0.17
6	M	6	blubber	0.835	0.316	1.501	2.652	0.012	3.51
			kidney	0.097	0.041	0.039	0.177	0.004	0.45
			liver	0.170	0.086	0.055	0.311	<0.002	0.48
			muscle	0.094	0.040	0.106	0.240	0.005	0.32
			spleen	0.112	0.062	0.103	0.277	0.009	0.30
7	M	2–3	blubber	0.610	0.287	1.551	2.448	0.124	3.46
			kidney	0.034	0.016	0.042	0.092	<0.002	0.04
			liver	0.052	0.030	0.087	0.169	0.016	0.82
			spleen	0.051	0.021	0.059	0.131	0.002	0.07
8	M	1–2	blubber	1.063	0.732	3.391	5.186	0.010	1.53
			kidney	0.074	0.036	0.070	0.180	0.006	0.19
			liver	0.077	0.114	0.057	0.248	0.002	0.36
			muscle	0.091	0.087	0.130	0.308	0.003	0.77
			spleen	0.094	0.095	0.152	0.341	0.007	0.30
9	M	13	blubber	0.556	0.354	0.731	1.641	0.022	1.20
			kidney	0.071	0.022	0.128	0.221	0.005	0.18
			liver	0.217	0.149	0.455	0.821	0.018	0.36
			muscle	0.098	0.038	0.068	0.204	0.004	0.07
			spleen	0.075	0.023	0.091	0.189	0.010	0.33
10	M	13	blubber	1.849	0.770	2.300	4.919	0.022	2.77
			kidney	0.125	0.077	0.366	0.568	0.008	0.20
			liver	0.347	0.215	0.121	0.683	0.026	0.64
			muscle	0.060	0.033	0.112	0.205	0.008	0.06
			spleen	0.049	0.023	0.047	0.119	0.003	0.09
11	M	18	blubber	1.108	0.680	1.326	3.114	0.011	2.83
			kidney	0.061	0.047	0.064	0.172	0.005	0.31
			muscle	0.066	0.054	0.200	0.320	0.009	0.41
			spleen	0.043	0.161	0.090	0.294	0.016	0.07

NOTE: Detection limit = 0.002 ppm.

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TABLE 3. *Organochlorine residues in tissues of harp seal pups, Gulf of St. Lawrence—March 1973* [1]

Seal Number	Tissue	*p,p'*-DDT	*p,p'*-TDE	*p,p'*-DDE	ΣDDT	Dieldrin	HCB	PCBs
1	blubber	0.833	0.132	2.019	2.984	0.087	0.054	1.812
	liver	0.041	0.007	0.096	0.144	0.005	0.003	0.116
	brain	0.026	0.003	0.021	0.050	0.006	0.002	0.097
2	blubber	0.602	0.119	1.044	1.765	0.150	0.130	1.869
	liver	0.027	0.004	0.025	0.056	0.007	0.007	0.063
	brain	0.028	0.003	0.010	0.041	0.010	0.005	0.022
3	blubber	0.830	0.209	1.314	2.353	0.117	0.061	2.984
	liver	0.031	0.008	0.037	0.076	0.007	0.003	0.116
	brain	0.034	0.006	0.014	0.054	0.008	<0.002	0.037
4	blubber	0.460	0.079	0.690	1.229	0.092	0.109	1.392
	liver	0.038	0.006	0.036	0.080	0.006	0.007	0.112
	brain	0.022	0.003	0.011	0.036	0.006	0.004	0.029
5	blubber	0.599	0.100	0.787	1.486	0.075	0.106	1.601
	liver	0.032	0.004	0.019	0.055	0.004	0.005	0.043
	brain	0.007	0.002	0.007	0.016	0.004	0.003	0.019
6	blubber	0.811	0.096	1.206	2.113	0.103	0.114	1.476
	liver	0.023	0.004	0.024	0.051	0.004	0.005	0.039
	brain	0.015	0.002	0.010	0.027	0.006	0.003	0.017
7	blubber	0.750	0.129	1.713	2.592	0.082	0.062	2.908
	liver	0.023	0.005	0.040	0.068	0.003	0.003	0.071
	brain	0.023	0.004	0.017	0.044	0.005	0.002	0.039
8	blubber	0.670	0.144	1.294	2.108	0.096	0.034	2.623
	liver	0.031	0.008	0.060	0.099	0.009	0.004	0.145
	brain	0.019	0.003	0.016	0.038	0.009	0.002	0.033
9	blubber	0.729	0.115	1.294	2.138	0.096	0.055	2.664
	liver	0.021	0.006	0.041	0.068	0.004	0.003	0.099
	brain	0.034	0.005	0.014	0.053	0.007	<0.002	0.041
10	blubber	0.660	0.086	1.079	1.825	0.095	0.085	1.810
	liver	0.037	0.007	0.056	0.100	0.007	0.006	0.115
	brain	0.006	0.003	0.007	0.016	0.005	0.002	0.022
11	blubber	0.578	0.117	1.137	1.832	0.088	0.121	2.020
12	blubber	0.536	0.086	0.757	1.379	0.075	0.065	2.268
13	blubber	0.468	0.053	0.562	1.083	0.076	0.083	1.150
	gonad	0.023	0.003	0.012	0.038	0.002	0.002	0.045
14	blubber	0.634	0.132	1.327	2.093	0.093	0.119	2.225
	gonad	0.079	0.014	0.116	0.209	0.008	0.011	0.211
15	blubber	0.735	0.152	0.994	1.881	0.104	0.067	2.074
16	blubber	0.760	0.159	1.362	2.821	0.144	0.097	2.416
17	blubber	0.460	0.071	0.830	1.361	0.073	0.068	1.926
18	blubber	1.188	0.404	2.138	3.730	0.179	0.042	6.226
19	blubber	0.475	0.100	0.708	1.283	0.087	0.050	2.313
20	blubber	0.626	0.070	0.875	1.571	0.074	0.028	1.512

NOTE: Detection limit = 0.002 ppm.
[1] Age of pups <2 weeks.

TABLE 4. *Ratios of DDT to DDE and ΣDDT to PCBs in harp seal tissues, Gulf of St. Lawrence—1971–1973*

Tissue	DDT/DDE	ΣDDT/PCB
Blubber (adults)	2.32	1.24
(pups)	0.58	0.86
Liver (adults)	1.10	0.62
(pups)	0.70	0.92
Kidney (adults)	3.30	1.04
Muscle (adults)	1.30	0.54
Spleen (adults)	1.65	0.74
Brain (pups)	1.62	1.03

NOTE: Ratios calculated from mean concentrations of each residue.

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Nationwide Residues of Organochlorine Compounds in Starlings (Sturnus vulgaris), *1976*

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ABSTRACT

Organochlorine pesticide and PCB residues in starlings from 126 sites within the contiguous 48 states were monitored during fall 1976. The average nationwide level of DDE and PCBs has increased significantly since 1974, but the number of sites reporting PCB residues has decreased fivefold. Dieldrin residues have remained unchanged since 1974. Highest DDE levels occurred in samples from parts of Arizona, Arkansas, California, Louisiana, and New Mexico.

Introduction

The Fish and Wildlife Service, U.S. Department of the Interior, began nationwide monitoring of organochlorine residues in starlings *(Sturnus vulgaris)* in 1967–68 as part of the National Pesticides Monitoring Program. Residue data from the original collections were to serve as a baseline against which future residue levels might be compared. Initially, organochlorine compounds were to be monitored at 2-year intervals. However, in 1976, starling collections were scheduled at 3-year intervals to coincide with waterfowl wing collections which also are monitored nationwide for organochlorine residues. Starlings were selected because their range is the continental United States, they are considered expendable, and their omnivorous feeding habits should reflect residues from a wide range of food sources (*1*). The present report presents results of the 1976 starling collections including residue levels from each collection site, a comparison of nationwide averages of DDE, dieldrin, and polychlorinated biphenyls (PCBs) in the four collection periods since 1970, and the distribution of DDE, dieldrin, and PCBs by frequency of occurrence at collection sites.

Collection Methods

Sampling design and collection procedures have been reported previously (*1–3*). The sample area lies within the continental United States and consists of 40 blocks of 5° latitude and longitude. In the initial 1967–68 study, 139 collection sites were randomly selected within these blocks and were to be used for starling collections thereafter. During September–December 1976, samples were obtained from 126 of the sites. Table 1 lists collection sites for 1976 by state and county; Figure 1 shows their actual locations within sampling blocks.

Starling samples consist of pools of 10 birds taken by trapping or shooting, although some samples may be smaller; those samples with fewer than 10 birds are identified in Table 1. Each pool is wrapped in aluminum foil, placed in a polyethylene bag, frozen as soon as possible, and shipped to Raltech Scientific Services, Inc., Madison, Wisconsin, for chemical analysis. A total of 227 pools were analyzed for organochlorine residues.

Analytical Procedures

The feet, beaks, wing tips, and skins were removed from birds in each composite sample and the sample was weighed and ground in a food grinder. Twenty grams of the homogenate was ground with 150 g anhydrous sodium sulfate and allowed to air dry overnight in a hood. The dried sample was placed in a 43 mm × 123 mm Whatman extraction thimble and extracted for 8 hours on a Soxhlet apparatus with 150 ml ethyl ether and 150 ml petroleum ether. The resulting solution was concentrated to near dryness on a steam bath, and the remaining solvent was removed with nitrogen at room temperature. The residue was transferred to a 25-ml volumetric flask with 93:1 toluene–ethyl acetate solution and diluted to volume.

Five ml of the extract was placed on an Auto-Prep Model 1001 gel permeation chromatograph, standardized for chlorinated insecticides and PCBs, with the following operating conditions:

Packing:	80 g Bio-Beads (SX-3), 200–400 mesh
Column:	600 mm × 25 mm ID
Solvent:	3:1 toluene–ethyl acetate solution
Flow rate:	5.5 ml/minute
Dump time:	30 minutes
Collect time:	14 minutes
Wash time:	4 minutes

The resulting solution was concentrated on a flash evaporator to approximately 5 ml in the presence of 5 ml isooctane and diluted to 25 ml with petroleum ether. A 4-μl sample was injected into a gas chromatograph equipped with an electron-capture detector. If PCBs were not detected, the results were quantified.

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TABLE 1. *Organochlorine residues in starlings, continental United States, 1976*

State	County[1]	Site	Residues, ppm wet weight						
			DDE	DDT	Dieldrin	PCBs[2]	Heptachlor Epoxide	HCB	Chlordane Isomers
Alabama	Marion	3-H-1	0.28	ND	0.14	ND	0.05	ND	0.04
	Calhoun	4-H-3	0.31	ND	0.01	0.85	0.04	ND	0.04
Arizona	Navajo	3-C-3	0.13	ND	ND	ND	ND	ND	ND
	Yavapai	3-C-4	0.27	ND	0.03	ND	0.01	ND	ND
	Maricopa	4-C-1	5.00	ND	0.01	ND	0.01	ND	ND
	Graham (3)	4-C-2	3.41	ND	0.01	ND	ND	ND	ND
Arkansas	Yell	3-G-2	0.31	ND	ND	0.35	0.13	ND	0.06
	Lonoke	3-G-3	11.10	ND	0.09	0.15	0.17	ND	0.06
California	Colusa (9)	2-A-1	0.39	ND	0.02	ND	ND	ND	ND
	Shasta	2-A-2	0.16	ND	0.01	ND	ND	ND	ND
	Modoc	2-A-3	0.13	ND	0.01	ND	0.01	ND	ND
	Ventura	3-A-1	1.26	ND	0.04	ND	ND	ND	ND
	Monterey (9)	3-A-3	2.20	0.02	0.08	0.39	ND	ND	0.01
	Kern	3-B-4	3.14	ND	0.03	ND	ND	ND	ND
	Imperial	4-B-1	7.41	ND	0.02	ND	ND	ND	ND
	Los Angeles	4-B-2	1.37	ND	0.04	ND	ND	ND	ND
Colorado	Weld	2-D-4	1.36	ND	0.06	ND	0.01	ND	ND
	Montrose	3-D-1	0.26	ND	ND	ND	ND	ND	ND
	Crowley	3-D-2	0.15	ND	ND	ND	0.01	ND	ND
Connecticut	New London	2-K-2	0.54	ND	0.03	0.39	0.09	ND	0.17
Florida	Bay	4-H-1	0.23	0.04	0.09	0.28	0.04	ND	0.07
	Madison	4-I-3	0.90	ND	0.11	ND	0.07	ND	0.18
	Highlands	5-I-2	0.67	ND	0.01	ND	ND	ND	ND
Georgia	Upson	4-H-4	1.03	ND	0.16	0.44	0.30	ND	0.20
	Wayne	4-I-2	0.35	0.03	0.11	0.26	0.03	ND	0.13
Idaho	Nez Perce	1-B-1	0.16	ND	ND	ND	ND	0.01	0.01
	Owyhee	2-B-1	1.15	ND	0.03	ND	0.01	ND	ND
	Franklin	2-C-3	1.12	ND	0.05	ND	0.02	ND	ND
	Minidoka	2-C-4	2.06	ND	0.06	0.16	0.01	ND	ND
Illinois	Stephenson	2-G-1	0.49	ND	0.17	0.21	0.06	ND	0.05
	Adams	2-G-3	0.04	ND	0.22	ND	0.36	0.56	0.09
	Kane	2-H-2	0.65	ND	0.12	ND	0.04	ND	0.01
Indiana	Henry	2-H-3	0.02	ND	0.03	ND	0.03	0.03	0.01
Iowa	Fremont	2-F-3	0.05	ND	0.23	ND	0.12	ND	0.04
	Jasper (8)	2-G-2	0.08	ND	0.28	ND	0.17	0.01	0.06
	Marshall (9)	2-G-4	0.09	ND	0.07	ND	0.11	ND	0.02
Kansas	Rawlins	2-E-1	0.29	ND	0.02	0.15	ND	ND	ND
	Phillips (7)	2-E-2	0.05	ND	0.02	ND	ND	0.03	ND
	Kearny (9)	3-E-1	0.03	ND	0.02	ND	0.01	ND	0.03
	Nemaha	2-F-4	0.07	ND	0.16	ND	0.04	0.01	0.02
	Marion	3-F-2	0.04	ND	0.06	ND	0.03	0.06	0.02
Kentucky	Ohio	3-H-2	0.15	ND	0.04	ND	0.02	ND	0.03
	Hopkins (9)	3-H-4	1.04	ND	0.04	ND	0.02	ND	0.11
Louisiana	Jefferson	4-G-3	0.93	ND	0.04	0.42	0.08	0.02	0.10
	Rapides	4-G-4	10.70	ND	0.04	0.63	0.03	ND	0.01
Maine	Penobscot	1-K-2	0.13	0.06	0.01	0.24	0.01	ND	0.01
Michigan	Chippewa	1-H-1	0.03	ND	0.03	ND	ND	ND	ND
	Grand Traverse	1-H-2	0.47	ND	0.02	ND	ND	ND	ND
	Kent	2-H-1	0.17	ND	ND	0.11	ND	ND	ND
	Ingham	2-H-4	0.51	ND	0.02	ND	0.03	ND	0.01
Minnesota	Aitkin	1-G-1	0.05	ND	ND	ND	ND	ND	ND
	Renville	1-F-2	0.04	ND	0.03	ND	ND	ND	ND
Mississippi	Leake	4-G-1	0.42	ND	0.18	ND	0.26	ND	0.09
	Harrison	4-G-2	0.67	0.04	0.24	ND	0.11	ND	0.07
	Jackson	4-H-2	1.43	ND	0.07	ND	0.04	ND	0.03
Missouri	Butler (7)	3-G-1	0.12	ND	0.06	0.11	0.03	0.23	ND
	Bollinger	3-G-4	0.06	ND	0.02	ND	ND	ND	ND
Montana	Meagher (9)	1-C-1	0.03	ND	ND	0.14	ND	ND	ND
	Missoula	1-C-4	0.06	0.04	0.01	ND	0.02	0.02	ND
	Richland (6)	1-D-1	0.01	ND	ND	ND	ND	ND	ND
	Yellowstone	1-D-4	ND	ND	ND	ND	ND	ND	ND
Nebraska	Keith (7)	2-E-3	0.04	ND	0.02	ND	ND	ND	ND
	Brown	2-E-4	0.04	ND	0.02	ND	0.03	ND	ND
	Lancaster (6)	2-F-1	0.25	0.07	ND	0.14	0.05	0.01	0.04
	Clay	2-F-2	0.10	ND	0.07	ND	ND	ND	ND
Nevada	White Pine	2-B-3	0.07	0.04	ND	ND	ND	ND	ND
	Humboldt	2-B-4	0.53	ND	0.02	ND	0.02	ND	0.04
	Nye	3-B-2	0.17	ND	0.04	ND	ND	ND	ND
	Clark	3-B-3	0.20	0.04	0.06	0.32	0.10	ND	0.21
New Mexico	Bernalillo	3-D-3	0.60	ND	ND	ND	ND	ND	ND
	Santa Fe	3-D-4	2.20	ND	0.03	ND	ND	ND	ND
	Luna	4-D-1	0.63	ND	ND	ND	ND	ND	ND
	Otero	4-D-2	1.71	ND	0.02	ND	ND	ND	ND
	Chaves	4-D-3	12.40	ND	0.03	ND	0.03	ND	0.01
	Quay	3-E-2	0.15	ND	ND	ND	0.01	ND	ND

(Continued next page)

TABLE 1 (Cont'd.). *Organochlorine residues in starlings, continental United States, 1976*

State	County[1]	Site	Residues, ppm wet weight						
			DDE	DDT	Dieldrin	PCBs[2]	Heptachlor Epoxide	HCB	Chlordane Isomers
New York	Jefferson (5)	2-J-4	0.09	ND	ND	ND	0.03	ND	0.04
	Rensselaer (8)	2-K-1	0.99	0.03	0.02	ND	ND	ND	ND
North Carolina	Wilkes	3-I-1	0.08	ND	0.02	ND	ND	ND	ND
	Macon	3-I-3	0.51	ND	0.03	ND	ND	ND	ND
	Pender	3-J-1	1.21	ND	0.20	0.18	0.03	ND	0.11
North Dakota	McLean	1-E-3	0.03	ND	0.01	ND	ND	ND	ND
	Grand Forks	1-F-1	0.43	ND	ND	ND	ND	ND	ND
	Ransom	1-F-4	0.07	0.01	ND	0.16	ND	ND	ND
Ohio	Pickaway	2-I-1	0.05	ND	0.05	ND	0.04	0.73	0.05
	Wood	2-I-2	0.08	ND	0.15	ND	0.05	0.06	0.01
	Noble	2-I-3	0.08	0.04	0.01	ND	0.02	ND	0.03
Oklahoma	Beckham	3-E-4	0.14	ND	0.03	ND	0.02	ND	ND
	Canadian	3-F-1	0.06	ND	0.03	ND	0.05	0.03	ND
	Nowata (9)	3-F-3	1.52	ND	0.05	ND	0.02	ND	0.03
	Okmulgee	3-F-4	0.12	0.05	0.10	ND	0.01	ND	0.01
Oregon	Yamhill	1-A-3	0.67	ND	0.10	ND	0.15	0.10	0.01
	Lane	1-A-4	0.32	ND	0.05	ND	0.05	ND	ND
	Benton	1-A-5	0.27	ND	0.06	ND	0.02	ND	ND
	Klamath	2-A-4	0.20	ND	ND	ND	0.02	ND	ND
	Baker (9)	1-B-4	0.06	ND	ND	ND	ND	ND	ND
	Harney	2-B-2	0.15	ND	ND	ND	ND	ND	ND
Pennsylvania	Somerset (6)	2-J-2	0.46	ND	0.05	ND	0.06	ND	0.10
	Luzerne	2-J-3	0.59	0.04	0.06	0.48	0.06	ND	0.13
South Dakota	Potter	1-E-1	0.07	ND	0.02	ND	ND	ND	ND
	Butte	1-E-2	0.02	ND	ND	ND	ND	ND	ND
	Hughes	1-E-4	0.03	ND	0.02	ND	ND	ND	ND
	Brown	1-F-3	0.03	ND	0.02	ND	ND	ND	ND
Tennessee	Davidson	3-H-3	0.09	0.02	0.14	0.22	0.01	ND	0.05
Texas	Kinney	4-E-3	1.05	ND	0.05	ND	0.89	ND	0.05
	Cochran	4-E-4	0.11	ND	0.04	ND	0.04	0.01	ND
	Bexar (7)	4-F-1	0.15	ND	0.02	ND	0.04	0.02	0.04
	Clay	4-F-3	0.97	ND	0.04	ND	0.47	0.07	0.07
	San Patricio	5-F-1	0.23	ND	ND	ND	0.01	ND	ND
Utah	Weber	2-C-1	0.91	0.04	0.04	0.55	ND	ND	ND
	Duschesne	2-C-2	0.10	ND	ND	ND	0.02	ND	ND
	Millard	3-C-1	0.42	ND	0.01	ND	ND	ND	ND
	Grand	3-C-2	0.93	ND	ND	ND	0.08	ND	0.02
Vermont	Addison	1-K-1	0.14	ND	0.01	0.11	0.04	ND	0.10
Virginia	Amherst (8)	3-I-4	0.52	ND	0.02	ND	ND	0.02	ND
	Prince George (9)	3-J-2	0.38	ND	0.02	0.11	0.02	0.04	0.05
	Caroline	3-J-3	0.11	ND	0.06	ND	0.07	0.20	0.07
Washington	Yakima	1-A-2	0.26	ND	0.12	ND	0.03	0.54	ND
	Spokane (5)	1-B-2	0.38	ND	0.24	ND	ND	2.01	ND
	Whitman	1-B-3	0.27	ND	ND	ND	ND	0.51	ND
Wisconsin	Trempealeau (9)	1-G-3	1.16	ND	0.01	ND	0.01	ND	ND
	Marathon (9)	1-G-2	0.07	ND	ND	ND	ND	ND	ND
Wyoming	Big Horn	1-D-2	0.02	ND	0.01	ND	0.03	ND	ND
	Crook (9)	1-D-3	ND	ND	ND	ND	ND	ND	ND
	Goshen	2-D-1	0.21	ND	ND	ND	ND	ND	ND
	Washakie	2-D-2	0.07	ND	0.03	ND	0.02	ND	ND

NOTE: ND = not detected.
[1]Most samples consist of a pool of 10 birds. Numbers in parentheses indicate samples made up of fewer birds.
[2]PCBs were quantified on the basis of Aroclor 1254.

If PCBs were detected, the extracts were subjected to silicic acid separation. Ten ml of the extract from the gel permeation chromatograph was placed on a 15-g standardized Silicar CC-4 column. Typical elutions were as follows:

Fraction I: 60 ml petroleum ether, contains HCB and mirex
Fraction II: 350 ml petroleum ether, contains PCBs and some DDE
Fraction III: 150-ml mixture of 1 percent acetonitrile, 19 percent hexane, and 80 percent methylene chloride, contains the remaining organochlorine compounds

Fractions I and II were concentrated on a steam bath to 1–2 ml; Fraction III was concentrated on a flash evaporator to 1–2 ml. All were diluted to 10 ml with petroleum ether. Quantities of 4 μl per solution were injected into a gas chromatograph equipped with an electron-capture detector.

Determinations were made on a Hewlett-Packard Model 5710A gas chromatograph equipped with a linear Ni^{63} detector and automatic injector, attached to a Hewlett-Packard Model 3352C data acquisition system. Instrument parameters and operating conditions for determining chlorinated insecticides and PCBs follow:

Column:	glass, 1219 mm × 4 mm ID, packed with a mixture of 1.95 percent OV-17 and 1.5 percent QF-1 on 80–100-mesh Supelcoport
Temperatures, °C:	column 200 injector 250 detector 300
Carrier gas:	a mixture of 95 percent argon and 5 percent methane flowing at 33 ml/minute

Instrument parameters and operating conditions for determining chlordane isomers were:

Column:	glass, 1219 mm × 4 mm ID, packed with 3 percent OV-1 on 80–100-mesh Gas-Chrom Q
Temperatures, °C:	column 190 injector 250 detector 300
Carrier gas:	a mixture of 95 percent argon and 5 percent methane flowing at 32 ml/minute

Residues in 5 percent of the samples were confirmed by mass spectrometry. Recoveries were 74–120 percent; analytical results were not corrected.

All residues are expressed as ppm wet weight. They may be converted to dry or lipid weight by dividing a given wet-weight value by 0.30 or 0.05, the mean proportions of dry and lipid material in the samples. Quantification limit was 0.01 ppm for organochlorine compounds. Trace residues were not reported.

Results and Discussion

Residues of DDE, DDT, dieldrin, PCBs, heptachlor epoxide, hexachlorobenzene (HCB), and chlordane isomers in starlings collected in 1976 are shown in Table 1. Since collections were made in the fall, residues do not necessarily reflect year-round levels. Also, findings should not be interpreted strictly on a statewide basis because some starlings are migratory. However, samples from certain localities consistently contain fairly high residues, suggesting that samples reflect local environmental contamination. For example, when results from previous monitorings (*1–4*) are compared, samples from certain parts of Arizona, Arkansas, California, Louisiana, and New Mexico usually contain higher DDE levels than do those from other states.

A summary of DDE, dieldrin, and PCB residues in starlings from 1970 through 1976 is shown in Table 2. The average DDE level in 1976 was similar to the 1970 level, before the use of technical DDT had been suspended. In fact, DDE residues were significantly higher nationwide ($P<0.001$) in 1976 than in 1974 (Table 2). It is difficult to explain why DDE residues have increased sharply since 1974, when residues were at their lowest level in 7 years. Possibly, DDT or its related

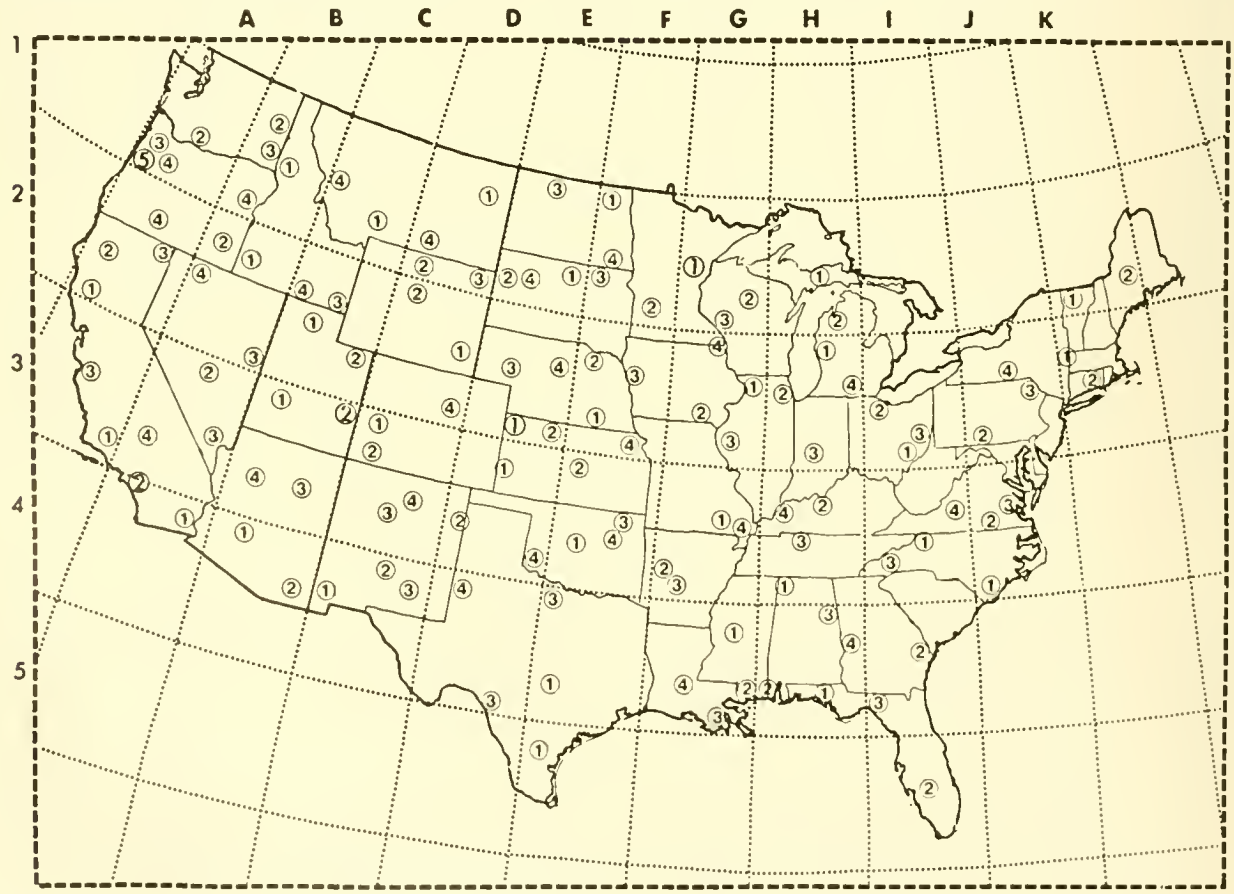

FIGURE 1. *Starling collection sites, continental United States, 1976*

TABLE 2. *Comparison of DDE, dieldrin, and PCB residues in starlings, continental United States, 1970–76*

Year	No. Pools	Residues, ppm wet weight								
		DDE			Dieldrin			PCBs		
		$\bar{x} \pm SE$[1]	Range	Geom. $\bar{x}$	$\bar{x} \pm SE$	Range	Geom. $\bar{x}$	$\bar{x} \pm SE$	Range	Geom. $\bar{x}$
1970	125	0.839 ± 0.138 (125)	0.037–48.2	0.355	0.117 ± 0.038 (125)	0.005–3.59	0.036	0.663 ± 0.196 (125)	0.09–24.3	0.358
1972	130	0.788 ± 0.124 (130)	0.047–14.8	0.387	0.098 ± 0.018 (130)	0.005–1.56	0.035	0.425 ± 0.153 (130)	0.04–19.9	0.215
1974	126	0.617 ± 0.118 (126)	0.007– 9.1	0.229	0.057 ± 0.011 (122)	0.005–1.01	0.019	0.112 ± 0.016 (126)	0.01– 1.9	0.068
1976	126	0.827 ± 0.174[2] (124)	0.010–12.4	0.254	0.059 ± 0.006 (96)	0.010–0.28	0.039	0.290 ± 0.036[2] (26)	0.11– 0.85	0.243

[1] Figure in parentheses represents number of pools having detectable residues.
[2] Residues in 1976 significantly higher than in 1974 ($P<0.001$, Students *t-test*, log-transformed data).

compounds may have been used, especially in certain geographical regions of the country.

Dieldrin residues declined steadily between 1970 and 1974, but the average dieldrin level in 1976 was almost identical to the 1974 average (Table 2), indicating no further decline of dieldrin during the 2-year period.

PCBs have increased significantly nationwide ($P<0.001$) since 1974, although 1976 residues remained below those reported for 1970 and 1972 (Table 2). Only 26 samples contained PCBs in 1976 compared to 126 in 1974; although the average PCB level was higher in 1976 than in 1974, the number of sites reporting PCB residues decreased fivefold in 1976.

The distribution of DDE, dieldrin, and PCBs by frequency of occurrence at collection sites for 1976 is shown in Table 3. In general, residues were low; most values were between 0 and 1.0 ppm for the three compounds. Dieldrin and PCBs were not detected in starlings at levels greater than 1.0 ppm.

In addition to organochlorine compounds in Table 1, certain other chemicals were detected in starlings less frequently. TDE occurred in six samples, ranging from 0.01 to 0.10 ppm; mirex was found in 13 samples, mostly from southeastern states, ranging from 0.01 to 1.24 ppm; lindane was detected in six samples, ranging from 0.01 to 0.15 ppm; and endrin occurred in only three samples, ranging from 0.02 to 0.18 ppm.

TABLE 3. *Distribution of residues in starlings by frequency of occurrence, continental United States, 1976*

Range, ppm	Number of Sites with Residues		
	DDE	Dieldrin	PCBs
ND– 0.01	3	43	99
>0.01– 0.10	36	65	0
>0.10– 1.0	63	17	26
>1.0 –13.0	23	0	0

NOTE: ND = not detected.

Conclusions

Nationwide, residues of DDE in starlings have increased significantly since 1974 to approximately the level reported in 1970 samples. Average PCB levels also increased, but the actual number of samples containing PCB residues declined. Dieldrin levels have remained unchanged since 1974.

These data indicate that starlings can serve as indicators of environmental contamination and thus provide information on residue trends over time. Geographical differences in residue levels also were detected.

Acknowledgments

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SOILS

Pesticide Application and Cropping Data from 37 States, 1972— National Soils Monitoring Program

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ABSTRACT

This report summarizes pesticide application and cropping data collected in 1972 from 1,402 agricultural sampling sites in 37 states as part of the National Soils Monitoring Program. Pesticide application data are summarized by all sites, state, and crop. Tables generally give the number of sites reporting, number of times a compound was applied, percent occurrence, and arithmetic mean application rate.

Pesticides applied most frequently were atrazine, 2,4-D, captan, and trifluralin. Among selected major crops, pesticides were most frequently applied to sites growing field corn and cotton, least frequently to sites growing alfalfa/bur clover and mixed hay.

Introduction

The increasing use of chemical pesticides in agriculture in the past 30 years has helped fewer farmers feed more people than at any other time in history. Today, the American farmer not only feeds and clothes this Nation's population, but also contributes significantly to the rest of the world. Yet the sensible use of toxic compounds also carries the responsibility to minimize their effects on nontarget components of the environment.

In 1963, the President's Science Advisory Committee recommended that appropriate federal agencies "develop a continuing network to monitor residue levels in air, water, soil, man, wildlife and fish" (*1*). As a result of the recommendation, the National Pesticide Monitoring Program (NPMP) was established to determine levels and trends of pesticides and their degradation products in various components of the environment (*2*). The federal responsibility for monitoring pesticides was officially codified in Section 20 of the amended Federal Insecticide, Fungicide and Rodenticide Act of 1972 (PL 92-516).

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The National Soils Monitoring Program (NSMP) is an integral part of the NPMP and monitors residues in agricultural soils and raw agricultural crops. It was established in 1968 by the U.S. Department of Agriculture and is administered by the U.S. Environmental Protection Agency. The present report summarizes pesticide application and cropping data collected during 1972 (FY–73) from 1,402 sampling sites in 37 states. Data for composite soil and crop samples, collected from the sites for pesticide residue analysis, are presented in a separate report (*3*).

Sampling

The site selection criteria and statistical design of the NSMP have been described (*4*). In 1972, 1,533 sites in 37 states were scheduled for sampling (Fig. 1). At each 4-hectare (10-acre) site, the landowner or operator was interviewed concerning crops grown and the kinds and amounts of pesticides applied during the 1972 growing season.

Results and Discussion

COMPOUNDS APPLIED TO CROPLAND

Cropping and pesticide use data were received from 1,402 of the scheduled 1,533 sites or 91 percent. Of these, 742 or 53 percent of the sites had one or more pesticides applied during the 1972 growing season. Tables summarizing the application data show the number of sites reporting a pesticide application, the percent of sites reporting the pesticide application, and the average rate of application, expressed in pounds per acre and kilograms per hectare.

Table 1 lists the frequency of pesticide use on sample sites in various states and state groups. Because some small eastern states had very few sites, those with similar geographic location and/or agricultural characteristics were combined to obtain more representative data. State groups used were Mid-Atlantic: Delaware, Maryland,

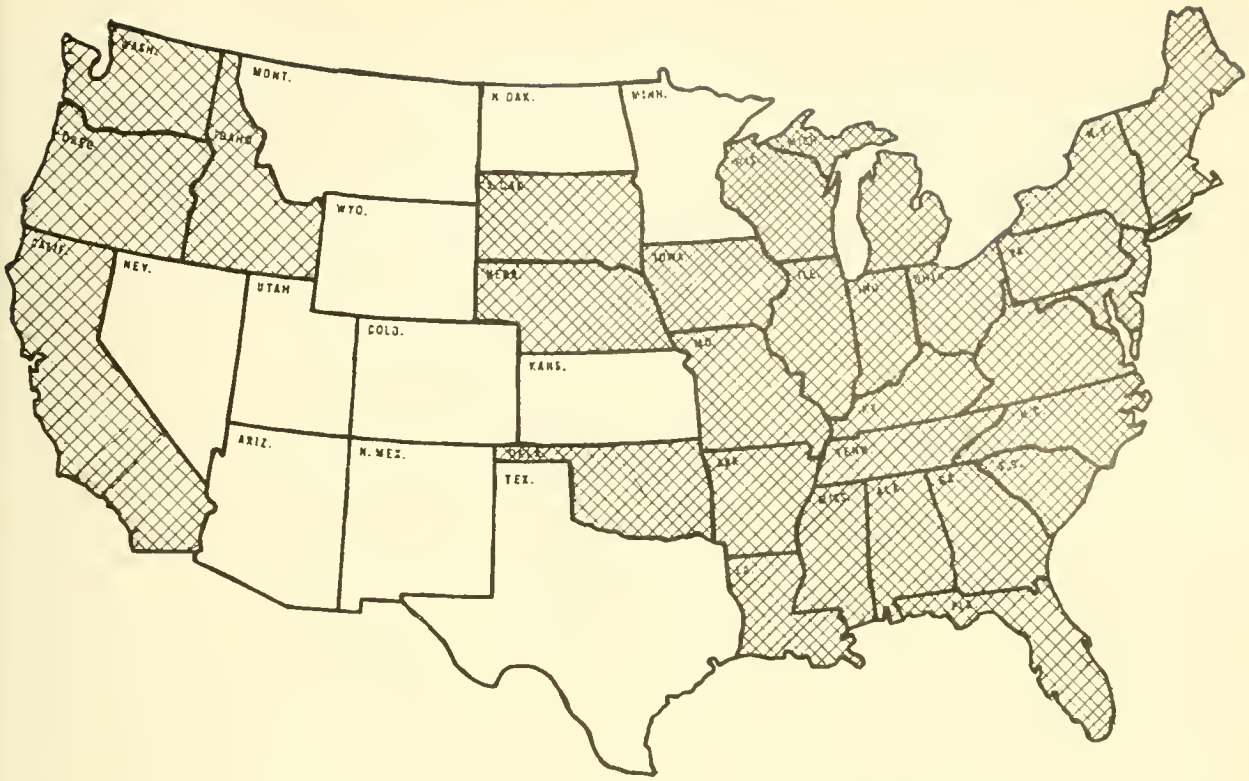

FIGURE 1. *States scheduled for sampling, 1972—National Soils Monitoring Program*

TABLE 1. *Pesticide application data from 1,402 reporting sites in 37 states, 1972—National Soils Monitoring Program*

State	No. of Sites Reporting	Pesticides Used		No Pesticides Used	
		No.	%	No.	%
Alabama	20	9	45	11	55
Arkansas	47	29	62	18	38
California	52	22	42	30	58
Florida	15	7	47	8	53
Georgia	27	13	48	14	52
Idaho	30	15	50	15	50
Illinois	139	94	68	45	32
Indiana	74	45	61	29	39
Iowa	149	106	71	43	29
Kentucky	16	7	44	9	56
Louisiana	27	18	67	9	33
Michigan	50	26	52	24	48
Mid-Atlantic[1]	18	10	56	8	44
Mississippi	27	24	89	3	11
Missouri	81	39	48	42	52
Nebraska	97	40	41	57	59
New England[1]	11	1	9	10	91
New York	31	13	42	18	58
North Carolina	31	17	55	14	45
Ohio	67	31	46	36	54
Oklahoma	43	27	63	16	37
Oregon	37	15	41	22	59
Pennsylvania	34	14	41	20	59
South Carolina	16	10	63	6	37
South Dakota	106	45	42	61	58
Tennessee	22	10	45	12	55
Virginia/West Virginia[1]	24	4	17	20	83
Washington	45	26	58	19	42
Wisconsin	66	25	38	41	62
TOTAL	1,402	742	53	660	47

[1]Because some small eastern states had very few sites, those with similar geographic location and/or agricultural characteristics were combined to obtain more representative data. State groups used were Mid-Atlantic: Delaware, Maryland, and New Jersey; New England: Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, and Vermont; and Virginia and West Virginia. Among individual states and state groups, frequency of pesticide use ranged from 9 percent in the New England states to 89 percent in Mississippi.

ALL SITES

The 121 compounds applied to all sites included 54 herbicides, 38 insecticides, 20 fungicides, 4 acaricides, 2 defoliants, 2 soil fumigants, and 1 growth retardant (Table 2). The most commonly applied compounds were atrazine, 2,4-D, captan, and trifluralin, which were used on 14, 10, 8, and 7 percent of the sites, respectively.

BY STATE

Table 3 presents the application data by state or state group. Because of the number of states sampled, it is not feasible to discuss in detail the pesticide data from each state. However, pesticide application data from each state tended to reflect both the crops grown and the intensity of agricultural land use in the state. For example, Iowa, predominantly a corn- and soybean-producing state, recorded the use of 17 compounds on 149 sites. California, a fruit and vegetable producer, recorded 29 compounds used on 52 sites.

In Figure 2, the frequency of reported pesticide applications in each state was arbitrarily classified as follows: low, less than 25 percent of the sites reported pesticide application; medium, 25–59 percent reported applica-

TABLE 2. *Summary of compounds applied to 1,402 cropland sites in 37 states, 1972—National Soils Monitoring Program*

Compound	Trade Name If Noted	Sites Reporting Application		Average Total Application	
		No.	%	Lb/Acre	Kg/Ha
Alachlor	Lasso	86	6.1	1.38	1.55
Aldicarb	Temik	2	0.1	0.40	0.45
Aldrin		33	2.4	1.57	1.76
Amitrole		1	0.1	0.15	0.17
Ancrack		4	0.3	1.08	1.20
Atrazine	AAtrex	200	14.3	1.56	1.75
Azinphosmethyl	Guthion	4	0.3	1.23	1.37
Benefin	Balan	6	0.4	0.83	0.93
Benomyl	Benlate	3	0.1	2.58	2.90
Benzene hexachloride		2	0.1	1.25	1.40
Bromacil	Hyvar	2	0.1	0.42	0.47
Bromoxynil		1	0.1	1.25	1.40
Butylate	Sutan	17	1.2	1.68	1.89
Bux		25	1.8	0.88	0.98
Captafol	Difolatan	3	0.1	3.83	4.29
Captan		106	7.6	0.19	0.22
Carbaryl	Sevin	23	1.6	2.49	2.79
Carbofuran	Furadan	17	1.2	1.07	1.19
Carbophenothion	Trithion	3	0.2	0.78	0.87
Chloramben	Amiben	51	3.6	1.38	1.55
Chlorobenzilate	Acaraben	4	0.3	3.45	3.87
Chlordane		5	0.4	3.18	3.57
Chloroneb	Demosan	8	0.6	0.02	0.02
Chloropropham	Chloro-IPC	1	0.1	0.59	0.66
Chloropropylate	Acarolate	1	0.1	3.50	3.92
Chloroxuron	Tenoran	1	0.1	2.00	2.24
Copper carbonate (basic)		1	0.1	3.50	3.92
Cyanazine	Bladex	2	0.1	2.15	2.41
Cycloate	Ro-Neet	3	0.2	1.95	2.19
2,4-D		136	9.7	0.69	0.77
Dalapon	Dowpon	2	0.1	7.80	8.74
2,4-DB	Butyrac	7	0.5	0.91	1.02
DDT		21	1.5	5.83	6.53
DEF		6	0.4	0.99	1.11
Diazinon		8	0.6	0.52	0.59
Dibromochloropropane	Nemagon	1	0.1	0.50	0.56
Dicamba	Banvel D	12	0.9	0.34	0.38
Dichlone	Phygon	1	0.1	0.50	0.56
Dichloropropene	Telone	1	0.1	60.00	67.25
Dichlorprop	2,4-DP	1	0.1	2.00	2.24
Dicofol	Kelthane	2	0.1	0.75	0.84
Dicrotophos	Bidrin	2	0.1	0.08	0.09
Dimethoate	Cygon	6	0.4	0.58	0.65
DNBP	Premerge	16	1.1	1.24	1.39
Dinitrocresol		2	0.1	1.63	1.82
Diphenamid	Enide	1	0.1	1.00	1.12
Disulfoton	Di-Syston	13	0.9	0.38	0.43
Diuron	Karmex	11	0.8	0.71	0.80
Dodine	Cyprex	1	0.1	0.98	1.09
DSMA		8	0.6	2.51	2.81
Dyfonate		3	0.2	0.97	1.08
EMTS	Ceresan M	9	0.6	0.01	0.01
EPN		1	0.1	3.00	3.36
EPTC	Eptam	10	0.7	2.19	2.45
Ethion		3	0.2	3.35	3.75
Ethoprop	Mocap	1	0.1	1.00	1.12
Ethylmercury chloride	Ceresan Red	5	0.4	0.01	0.01
Fenac		1	0.1	1.25	1.40
Fenaminosulf	Dexon	1	0.1	0.01	0.01
Fensulfothion	Dasanit	4	0.3	2.79	3.13
Fentin hydroxide		2	0.1	8.75	9.81
Fluometuron	Cotoran	23	1.6	0.93	1.04
Folex		5	0.4	1.10	1.23
Heptachlor		5	0.4	1.26	1.41
Hexachlorobenzene	No-Bunt	11	0.8	0.04	0.04
Lead arsenate		1	0.1	4.00	4.48
Lindane		1	0.1	0.01	0.01
Linuron	Lorox	39	2.8	1.11	1.24
Malathion		83	5.9	0.04	0.04
Maleic hydrazide	MH	5	0.4	2.25	2.52
Maneb		3	0.2	0.70	0.78
MCPA	MCP	5	0.4	1.40	1.56
MCPB		5	0.4	0.85	0.95
Mercury		10	0.7	0.04	0.04
Methomyl	Lannate	1	0.1	0.34	0.38
Methoxychlor		11	0.7	0.19	0.21
Methylmercury acetate	Ceresan L	7	0.5	0.01	0.01
Methylmercury dicyandiamide	Panogen	3	0.2	0.01	0.01
Methyl trithion		1	0.1	0.25	0.28
Metribuzin	Sencor	1	0.1	0.50	0.56
Mevinphos	Phosdrin	1	0.1	0.25	0.28
Mirex		7	0.5	0.01	0.01
Molinate	Ordram	2	0.1	3.00	3.36
Monocrotophos	Azodrin	3	0.2	1.67	1.87
MSMA		21	1.5	2.36	2.65
Naled	Dibrom	1	0.1	1.00	1.12
Naptalam	Alanap	8	0.6	1.35	1.52
Nitralin	Planavin	9	0.6	1.16	1.30
Norea	Herban	3	0.2	1.57	1.76
Oil spray		2	0.1	55.00	61.64
Oxythioquinox	Morestan	1	0.1	0.08	0.09
Paraquat		7	0.5	0.43	0.48
Parathion, ethyl		17	1.1	2.29	2.57
Parathion, methyl		40	2.9	2.99	3.35
PCNB		7	0.5	0.02	0.02
Pebulate	Tillam	1	0.1	4.00	4.48
Pentachlorophenol	PCP	1	0.1	0.05	0.06
Phenylmercury acetate	PMA	4	0.3	0.02	0.02
Phorate	Thimet	26	1.9	1.79	2.01
Picloram	Borolin	1	0.1	0.75	0.84
Polyram		1	0.1	1.00	1.12
Prolate	Imidan	3	0.2	3.92	4.39
Prometryn	Caparol	4	0.3	0.87	0.98
Propachlor	Ramrod	40	2.9	1.93	2.16
Propanil	Stam	2	0.1	3.50	3.92
Propargite	Omite	2	0.1	1.59	1.78
Propham	IPC	2	0.1	1.75	1.96
Pyrazon	Pyramin	1	0.1	0.94	1.05
Simazine	Princep	5	0.4	2.82	3.16
Sodium chlorate		2	0.1	1.00	1.12
Sulfur		10	0.7	27.85	31.21
TCA		2	0.1	5.63	6.30
TCBC		1	0.1	8.00	8.97
TEPP		1	0.1	0.25	0.28
Terbacil	Sinbar	2	0.1	1.75	1.96
Thiram		14	0.9	0.03	0.03
Toxaphene		30	2.1	9.36	10.49
Trietazine		1	0.1	0.25	0.28
Trifluralin	Treflan	97	6.9	0.86	0.96
Vernolate	Vernam	6	0.4	1.20	1.35

TABLE 3. *Compounds applied to cropland sites by state, 1972—National Soils Monitoring Program*

Compound	Trade Name if Noted	Sites Reporting Application		Average Total Application	
		No.	%	Lb/Acre	Kg/Ha
ALABAMA, 20 SITES					
Atrazine	AAtrex	1	5.0	1.00	1.12
Benefin	Balan	3	15.0	0.75	0.84
Benomyl	Benlate	2	10.0	3.50	3.92
Captan		1	5.0	0.01	0.01
Disulfoton	Di-Syston	1	5.0	1.00	1.12
2,4-D		1	5.0	1.00	1.12
DNBP	Premerge	1	5.0	1.00	1.12
Linuron	Lorox	1	5.0	1.00	1.12
Naptalam	Alanap	1	5.0	2.00	2.24
Parathion, methyl		1	5.0	13.00	14.57
Toxaphene		2	10.0	8.50	9.53
Trifluralin	Treflan	2	10.0	2.00	2.24
Vernolate	Vernam	3	15.0	0.75	0.84
ARKANSAS, 47 SITES					
Alachlor	Lasso	2	4.3	3.25	3.64
Ancrack		1	2.1	0.50	0.56
Captan		3	6.4	0.01	0.01
Chloroxuron	Tenoran	1	2.1	2.00	2.24
DEF		1	2.1	0.50	0.56
DDT		5	10.6	3.90	4.37
Dicrotophos	Bidrin	2	4.3	0.08	0.09
Disulfoton	Di-Syston	1	2.1	0.01	0.01
Diuron	Karmex	2	4.3	1.25	1.40
DNBP	Premerge	4	8.5	0.94	1.05
DSMA		2	4.3	2.50	2.80
2,4-DB	Butyrac	4	8.5	0.88	0.98
EMTS	Ceresan M	2	4.3	0.01	0.01
EPN		1	2.1	3.00	3.36
Fluometuron	Cotoran	7	14.9	0.96	1.08
Folex		1	2.1	1.50	1.68
Linuron	Lorox	2	4.3	0.50	0.56
Mercury		7	14.9	0.05	0.05
Metribuzin	Sencor	1	2.1	0.50	0.56
MSMA		9	19.1	1.94	2.18
Naptalam	Alanap	3	6.4	0.83	0.93
Nitralin	Planavin	2	4.3	1.00	1.12
Paraquat		1	2.1	0.02	0.02
Parathion, ethyl		1	2.1	3.00	3.36
Parathion, methyl		9	19.1	2.69	3.02
Prometryn	Caparol	3	6.4	1.08	1.21
Thiram	Stimusoy	2	4.3	0.12	0.13
Toxaphene		8	17.0	4.84	5.43
Trifluralin	Treflan	12	25.5	0.77	0.86
CALIFORNIA, 52 SITES					
Alachlor	Lasso	1	1.9	0.50	0.56
Carbophenothion	Trithion	1	1.9	0.09	0.10
Chloroneb	Demosan	1	1.9	0.01	0.01
2,4-D		2	3.8	0.50	0.56
DNBP	Premerge	1	1.9	0.50	0.56
Dibromochloro-propane	Nemagon	1	1.9	0.50	0.56
Dicofol	Kelthane	2	3.8	0.75	0.84
Disulfoton	Di-Syston	1	1.9	1.00	1.12
EPTC	Eptam	1	1.9	3.00	3.36
Fenaminosulf	Dexon	1	1.9	0.01	0.01
Malathion		1	1.9	1.00	1.12
MCPA	MCP	2	3.8	2.00	2.24
Methomyl	Lannate	1	1.9	0.34	0.38
Mevinphos	Phosdrin	1	1.9	0.25	0.28
Molinate	Ordram	2	3.8	3.00	3.36
Naled	Dibrom	1	1.9	1.00	1.12
Nitralin	Planavin	1	1.9	0.75	0.84
Paraquat		2	3.8	0.63	0.70
Parathion, ethyl		5	9.6	0.76	0.85
Parathion, methyl		2	3.8	0.36	0.40
Phorate	Thimet	2	3.8	1.00	1.12
Prolate	Imidan	1	1.9	0.75	0.84
Propanil	Stam	1	1.9	4.00	4.48
Propargite	Omite	1	1.9	1.68	1.88
Simazine	Princep	2	3.8	2.75	3.08
Sulfur		2	3.8	0.80	0.90
TEPP		1	1.9	0.25	0.28
Toxaphene		1	1.9	3.00	3.36
Trifluralin	Treflan	2	3.8	0.38	0.42
FLORIDA, 15 SITES					
Atrazine	AAtrex	1	6.7	2.00	2.24
Azinphosmethyl	Guthion	1	6.7	2.00	2.24
Carbophenothion	Trithion	1	6.7	1.50	1.68
Chlorobenzilate	Acaraben	4	26.7	3.45	3.87
Copper carbonate (basic)		1	6.7	3.50	3.92
Ethion	Ethodan	3	20.0	3.35	3.76
Fensulfothion	Dasanit	1	6.7	7.50	8.41
Oil spray		2	13.3	55.00	61.64
Sulfur		4	26.7	36.88	41.33
GEORGIA, 27 SITES					
Alachlor	Lasso	1	3.7	2.50	2.80
Atrazine	AAtrex	1	3.7	4.00	4.48
Benefin	Balan	1	3.7	1.13	1.27
Captan		1	3.7	0.01	0.01
Carbaryl	Sevin	5	18.5	2.40	2.69
DDT		1	3.7	4.50	5.04
Captafol	Difolatan	1	3.7	10.00	11.21
Disulfoton	Di-Syston	1	3.7	1.00	1.12
Fentin hydroxide	Du-Ter	2	7.4	8.75	9.81
Mirex		2	7.4	0.01	0.01
Maleic hydrazide	MH-30	1	3.7	6.00	6.72
Parathion, ethyl		2	7.4	2.75	3.08
Parathion, methyl		1	3.7	4.50	5.04
Sulfur		1	3.7	34.00	38.11
Toxaphene		2	7.4	5.25	5.88
Trifluralin	Treflan	1	3.7	1.00	1.12
IDAHO, 30 SITES					
Atrazine	AAtrex	2	6.7	0.75	0.84
Bromoxynil		1	3.3	1.25	1.40
2,4-D		7	23.3	1.21	1.36
DDT		1	3.3	1.00	1.12
EMTS	Ceresan M	2	6.7	0.01	0.01
EPTC	Eptam	1	3.3	0.25	0.28
MCPB		1	3.3	2.00	2.24
Sulfur		1	3.3	20.00	22.42
Trifluralin	Treflan	2	6.7	0.25	0.28
ILLINOIS, 139 SITES					
Alachlor	Lasso	10	7.2	1.24	1.38
Aldrin		6	4.3	1.10	1.23
Atrazine	AAtrex	23	16.5	1.33	1.49
Butylate	Sutan	9	6.5	0.94	1.06
Bux		2	1.4	1.30	1.46
Captan		45	32.4	0.01	0.01
Carbofuran	Furadan	3	2.2	0.75	0.84
Chloramben	Amiben	20	14.4	1.25	1.40
Chlordane		3	2.2	0.97	1.09
2,4-D		14	10.1	0.43	0.49
2,4-DB	Butyrac	1	0.7	0.50	0.56
Diazinon		2	1.4	2.01	2.25
Dicamba	Banvel-D	2	1.4	0.17	0.18
Dyfonate		1	0.7	0.50	0.56
EPTC	Eptam	1	0.7	0.42	0.47
Ethylmercury chloride	Ceresan Red	1	0.7	0.01	0.01
Heptachlor		2	1.4	1.65	1.85
Lindane		1	0.7	0.01	0.01
Linuron	Lorox	2	1.4	1.25	1.40
Malathion		44	31.6	0.01	0.01
Methoxychlor		5	3.6	0.01	0.01
Nitralin	Planavin	1	0.7	2.80	3.14
Phorate	Thimet	3	2.2	0.80	0.90
Propachlor	Ramrod	15	10.8	1.71	1.92
TCBC	Randox-T	1	0.7	8.00	8.97
Simazine	Princep	1	0.7	3.00	3.36
Trietazine	Gesaftoc	1	0.7	0.25	0.28
Toxaphene		1	0.7	0.40	0.45
Trifluralin	Treflan	12	8.6	0.73	0.82
Vernolate	Vernam	1	0.7	0.45	0.50
INDIANA, 74 SITES					
Alachlor	Lasso	15	20.3	1.86	2.08
Aldrin		7	9.5	1.33	1.49

(*Continued next page*)

TABLE 3 (cont'd.). *Compounds applied to cropland sites by state, 1972—National Soils Monitoring Program*

Compound	Trade Name if Noted	Sites Reporting Application		Average Total Application	
		No.	%	Lb/Acre	Kg/Ha
Atrazine	AAtrex	20	27.0	1.83	2.05
Captan		6	8.1	0.01	0.01
Chloramben	Amiben	5	6.8	1.20	1.35
2,4-D		4	5.4	0.50	0.56
EPTC	Eptam	1	1.4	10.00	11.21
Linuron	Lorox	8	10.8	1.38	1.55
Malathion		6	8.1	0.01	0.01
Trifluralin	Treflan	3	4.1	1.00	1.12
IOWA, 149 SITES					
Alachlor	Lasso	10	6.7	0.86	0.97
Aldrin		8	5.4	1.20	1.35
Atrazine	AAtrex	38	25.5	1.38	1.55
Butylate	Sutan	6	4.0	2.75	3.08
Bux		13	8.7	0.90	1.01
Carbofuran	Furadan	4	2.7	0.98	1.09
Chloropropham	Chloro-IPC	1	0.7	0.59	0.66
Chloramben	Amiben	16	10.7	0.96	1.08
2,4-D		18	12.1	0.51	0.57
Diazinon		1	0.7	0.07	0.08
Dicamba	Banvel-D	6	4.0	0.25	0.28
Dyfonate		1	0.7	1.40	1.57
Ethoprop	Mocap	1	0.7	1.00	1.12
Heptachlor		1	0.7	1.00	1.12
Phorate	Thimet	8	5.4	1.06	1.19
Propachlor	Ramrod	14	9.4	1.74	1.95
Trifluralin	Treflan	22	14.8	0.80	0.89
KENTUCKY, 16 SITES					
Atrazine	AAtrex	5	31.3	1.00	1.12
Carbaryl	Sevin	2	12.5	1.50	1.68
2,4-D		1	6.3	1.00	1.12
Linuron	Lorox	1	6.3	0.75	0.84
Malathion		1	6.3	1.00	1.12
Methoxychlor		1	6.3	2.00	2.24
Trifluralin	Treflan	1	6.3	1.00	1.12
LOUISIANA, 27 SITES					
Alachlor	Lasso	1	3.7	1.00	1.12
Aldrin		1	3.7	0.01	0.01
Azinphosmethyl	Guthion	1	3.7	1.50	1.68
Chloramben	Amiben	1	3.7	1.50	1.68
2,4-D		3	11.1	1.12	1.25
2,4-DB	Butyrac	1	3.7	2.00	2.24
DCPA	Dacthal	1	3.7	0.75	0.84
DDT		5	18.5	11.30	12.67
DSMA		4	14.8	3.15	3.53
Dalapon	Dowpon	1	3.7	2.00	2.24
DEF		1	3.7	1.50	1.68
Dichlorprop	2,4-DP	1	3.7	2.00	2.24
Diphenamid	Enide	1	3.7	1.00	1.12
Diuron	Karmex	3	11.1	1.25	1.40
DNBP	Premerge	2	7.4	1.25	1.40
EMTS	Ceresan M	1	3.7	0.01	0.01
Fenac		1	3.7	1.25	1.40
Fluometuron	Cotoran	4	14.8	1.03	1.15
Folex		1	3.7	1.00	1.12
MSMA	Ansar	3	11.1	2.50	2.80
Norea	Herban	1	3.7	0.60	0.67
Parathion, methyl		8	29.6	3.66	4.10
Propanil	Stam	1	3.7	3.00	3.36
TCA		1	3.7	11.00	12.33
Terbacil	Sinbar	1	3.7	2.00	2.24
Thiram		1	3.7	0.01	0.01
Toxaphene		5	18.5	23.40	26.23
Trifluralin	Treflan	8	29.6	1.41	1.58
Vernolate	Vernam	1	3.7	2.50	2.80
MICHIGAN, 50 SITES					
Alachlor	Lasso	1	2.0	0.50	0.56
Aldrin		1	2.0	1.40	1.57
Atrazine	AAtrex	15	30.0	2.09	2.35
Captan		10	20.0	0.01	0.01
2,4-D		6	12.0	1.29	1.45
Dicamba	Banvel D	1	2.0	1.00	1.12
EPTC	Eptam	2	4.0	2.00	2.24
Linuron	Lorox	3	6.0	1.67	1.87
Malathion		10	20.0	0.01	0.01
Pyrazon	Pyramin	1	2.0	0.94	1.05
TCA		1	2.0	0.25	0.28
Trifluralin	Treflan	1	2.0	1.00	1.12
MID-ATLANTIC STATES,[1] 18 SITES					
Alachlor	Lasso	5	27.8	2.06	2.31
Atrazine	AAtrex	2	11.1	1.75	1.96
Azinphosmethyl	Guthion	1	5.6	0.90	1.01
Captan		4	22.2	0.01	0.01
Carbaryl	Sevin	2	11.1	1.92	2.15
Chlordane		1	5.6	5.00	5.60
2,4-D		1	5.6	0.50	0.56
Dichlone	Phygon	1	5.6	0.50	0.56
Dieldrin		2	11.1	0.26	0.29
Dimethoate	Cygon	1	5.6	0.66	0.74
Dinitrocresol		1	5.6	3.00	3.36
Linuron	Lorox	1	5.6	0.38	0.43
Maneb		1	5.6	1.44	1.61
Malathion		2	11.1	0.01	0.01
Parathion, ethyl		1	5.6	1.30	1.46
Prolate	Imidan	1	5.6	2.00	2.24
Sulfur		1	5.6	37.00	41.47
Thiram	Arasan	1	5.6	0.01	0.01
Trifluralin	Treflan	1	5.6	1.20	1.35
MISSISSIPPI, 27 SITES					
Alachlor	Lasso	1	3.7	2.00	2.24
Aldicarb	Temik	2	7.4	0.40	0.45
Ancrack		3	11.1	1.27	1.42
Azinophosmethyl	Guthion	1	3.7	0.50	0.56
Captan		1	3.7	0.03	0.03
Carbaryl	Sevin	1	3.7	1.00	1.12
Chloroneb	Demosan	7	25.9	0.03	0.03
2,4-DB	Butyrac	1	3.7	0.40	0.45
DDT		7	25.9	5.71	6.40
DEF		4	14.8	0.98	1.10
DSMA		2	7.4	1.24	1.39
DNBP	Premerge	7	25.9	1.70	1.91
Disulfoton	Di-Syston	5	18.5	0.01	0.01
Diuron	Karmex	2	7.4	0.30	0.34
Fluometuron	Cotoran	7	25.9	0.70	0.78
Folex		2	7.4	0.75	0.84
Linuron	Lorox	3	11.1	1.83	2.05
MSMA		7	25.9	2.75	3.08
Methylmercury acetate	Ceresan L	6	22.2	0.01	0.01
Mirex		4	14.8	0.01	0.01
Monocrotophos	Azodrin	3	11.1	1.67	1.87
Naptalam	Alanap	1	3.7	3.00	3.36
Nitralin	Planavin	3	11.1	1.33	1.49
Norea	Herban	1	3.7	1.60	1.79
Parathion, methyl		10	37.0	4.38	4.90
Sodium chlorate		2	7.4	1.00	1.12
Toxaphene		8	29.6	10.25	11.49
Trifluralin	Treflan	10	37.0	0.85	0.95
MISSOURI, 81 SITES					
Alachlor	Lasso	11	13.6	1.42	1.59
Atrazine	AAtrex	13	16.0	1.67	1.87
Aldrin		3	3.7	1.67	1.87
Chloramben	Amiben	2	2.5	3.01	3.37
2,4-D		3	3.7	0.42	0.47
Diuron	Karmex	1	1.2	0.25	0.28
Fluometuron	Cotoran	3	3.7	1.31	1.46
Linuron	Lorox	6	7.4	0.87	0.97
MSMA		1	1.2	3.40	3.81
Norea	Herban	1	1.2	2.50	2.80
Trifluralin	Treflan	11	13.6	0.73	0.82
NEBRASKA, 97 SITES					
Alachlor	Lasso	4	4.1	1.11	1.25
Atrazine	AAtrex	18	18.6	1.40	1.57
Bux		7	7.2	0.73	0.82
Carbofuran	Furadan	5	5.2	0.69	0.78

(Continued next page)

TABLE 3 (cont'd.). *Compounds applied to cropland sites by state, 1972—National Soils Monitoring Program*

Compound	Trade Name if Noted	Sites Reporting Application		Average Total Application	
		No.	%	Lb/Acre	Kg/Ha
Chloramben	Amiben	1	1.0	1.50	1.68
Cyanazine	Bladex	1	1.0	2.80	3.14
Cycloate	Ro-Neet	1	1.0	0.40	0.45
2,4-D		8	8.2	0.61	0.68
Diazinon		1	1.0	0.98	1.10
Dyfonate		1	1.0	1.00	1.12
EPTC	Eptam	1	1.0	1.75	1.96
Fensulfothion	Dasanit	1	1.0	0.90	1.01
Linuron	Lorox	2	2.1	0.94	1.05
Parathion, ethyl		2	2.1	0.65	0.73
Propachlor	Ramrod	5	5.2	2.13	2.39
Phorate	Thimet	3	3.1	0.88	0.99
Simazine	Princep	1	1.0	4.00	4.48
NEW ENGLAND,[1] 11 SITES					
Captan		1	9.1	19.20	21.52
Carbophenothion	Trithion	1	9.1	0.75	0.84
Chloropropylate	Acarolate	1	9.1	3.50	3.92
Dodine	Cyprex	1	9.1	0.98	1.10
Prolate		1	9.1	0.38	0.43
Propargite	Omite	1	9.1	9.00	10.09
NEW YORK, 31 SITES					
Alachlor	Lasso	2	7.1	0.88	0.98
Atrazine	AAtrex	10	32.3	2.03	2.27
Benomyl	Benlate	1	3.6	0.75	0.84
Bux		1	3.6	0.70	0.78
Captan		3	10.7	0.01	0.01
Carbaryl	Sevin	2	7.1	4.25	4.76
Carbofuran	Furadan	1	3.6	1.00	1.12
2,4-D		1	3.6	0.25	0.28
Diazinon		2	7.1	0.51	0.57
Dinitrocresol		1	3.6	0.25	0.28
DNBP	Premerge	1	3.6	0.21	0.24
EPTC	Eptam	1	3.6	0.25	0.28
Methoxychlor		4	12.9	0.01	0.01
Parathion, ethyl		1	3.6	0.33	0.37
Thiram		4	12.9	0.01	0.01
NORTH CAROLINA, 31 SITES					
Alachlor	Lasso	3	9.7	1.00	1.12
Atrazine	AAtrex	4	12.9	1.63	1.82
Carbaryl	Sevin	4	12.9	3.38	3.78
2,4-D		1	3.2	1.00	1.12
Dichloropropene	Telone	1	3.2	60.00	67.25
Fensulfothion	Dasanit	1	3.2	2.00	2.24
Lead arsenate		1	3.2	4.00	4.48
Linuron	Lorox	1	3.2	1.50	1.68
Maleic hydrazide		4	12.9	1.31	1.47
Maneb		1	3.2	0.41	0.46
Naptalam	Alanap	2	6.5	0.92	1.03
Nitralin	Planavin	1	3.2	0.50	0.56
Parathion, ethyl		2	6.5	10.50	11.77
Paraquat		2	6.5	0.38	0.42
Pebulate	Tillam	1	3.2	4.00	4.48
Phorate	Thimet	1	3.2	0.50	0.56
Pentachlorophenol	PCP	1	3.2	0.05	0.06
Toxaphene		1	3.2	10.00	11.21
Trifluralin	Treflan	1	3.2	0.80	0.90
OHIO, 67 SITES					
Alachlor	Lasso	4	6.0	1.14	1.27
Aldrin		6	9.0	3.33	3.74
Atrazine	AAtrex	13	19.4	1.89	2.12
Butylate	Sutan	1	1.5	2.00	2.24
Bux		1	1.5	0.80	0.90
Captan		1	1.5	0.01	0.01
Carbofuran	Furadan	1	1.5	1.00	1.12
Chloramben	Amiben	5	7.5	2.60	2.91
2,4-D		5	7.5	1.00	1.12
Dicamba	Banvel D	2	3.0	1.00	1.12
Linuron	Lorox	5	7.5	0.92	1.03
Methylmercury acetate	Ceresan L	1	1.5	0.01	0.01
Picloram	Borolin	1	1.5	0.75	0.84
Propachlor	Ramrod	1	1.5	8.00	8.97
OKLAHOMA, 43 SITES					
Alachlor	Lasso	2	4.7	2.00	2.24
Benefin	Balan	1	2.3	1.00	1.12
Captan		2	4.7	0.01	0.01
Carbaryl	Sevin	3	7.0	2.17	2.43
2,4-D		4	9.3	0.50	0.56
EMTS	Ceresan M	4	9.3	0.01	0.01
Ethylmercury chloride	Ceresan Red	1	2.3	0.01	0.01
MCPB		2	4.7	0.50	0.56
Parathion, methyl		8	18.6	0.50	0.56
PCNB	Terrachlor	6	14.0	0.02	0.02
Polyram		1	2.3	1.00	1.12
Thiram	Arasan	5	11.6	0.01	0.01
Trifluralin	Treflan	1	2.3	0.50	0.56
OREGON, 37 SITES					
Amitrole		1	2.7	0.15	0.17
Bromacil		1	2.7	0.09	0.10
Captan		2	5.4	0.01	0.01
Cycloate	Ro-Neet	1	2.7	5.20	5.83
2,4-D		11	29.7	0.72	0.81
Diazinon		1	2.7	0.10	0.11
Diuron	Karmex	1	2.7	0.10	0.11
Ethylmercury chloride	Ceresan Red	2	5.4	0.01	0.01
Hexachloro-benzene	HCB	2	5.4	0.04	0.04
Malathion		1	2.7	0.50	0.56
Maneb		1	2.7	0.25	0.28
Methylmercury dicyandiamide	Panogen	1	2.7	0.01	0.01
Parathion, ethyl		1	2.7	0.09	0.10
Phenylmercury acetate	PMA	2	5.4	0.02	0.02
Propham	IPC	1	2.7	0.50	0.56
PENNSYLVANIA, 34 SITES					
Alachlor	Lasso	3	8.8	0.75	0.84
Atrazine	AAtrex	9	26.5	1.60	1.79
Butylate	Sutan	1	2.9	1.60	1.79
Captafol	Difolatan	1	2.9	1.00	1.12
2,4-D		3	8.8	0.58	0.65
Disulfoton	Di-Syston	1	2.9	0.50	0.56
Linuron	Lorox	1	2.9	0.50	0.56
Phorate	Thimet	1	2.9	2.50	2.80
SOUTH CAROLINA, 16 SITES					
Benefin	Balan	1	6.25	0.60	0.67
Captan		1	6.25	0.01	0.01
Carbaryl	Sevin	1	6.25	1.00	1.12
DDT		2	12.50	0.50	0.56
Methyl trithion		1	6.25	0.25	0.28
Mirex		1	6.25	0.01	0.01
Nitralin	Planavin	1	6.25	0.35	0.39
Parathion, ethyl		1	6.25	2.40	2.69
Parathion, methyl		1	6.25	0.25	0.28
Sulfur		1	6.25	38.40	43.04
Toxaphene		2	12.50	1.00	1.12
Trifluralin	Treflan	3	18.75	0.58	0.65
Vernolate	Vernam	1	6.25	2.00	2.24
SOUTH DAKOTA, 106 SITES					
Alachlor	Lasso	4	3.8	1.06	1.19
Atrazine	AAtrex	5	4.7	0.90	1.01
Bux		1	0.9	1.00	1.12
Captan		20	18.9	0.01	0.01
Carbofuran	Furadan	2	1.9	0.25	0.28
Chloramben	Amiben	1	0.9	2.00	2.24
2,4-D		27	25.5	0.41	0.45
Dieldrin		2	1.9	0.01	0.01
Dimethoate	Cygon	4	3.8	0.21	0.23
Disulfoton	Di-Syston	2	1.9	0.31	0.35
Malathion		18	17.0	0.01	0.01
MCPA		1	0.9	0.50	0.56

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TABLE 3 (cont'd.). *Compounds applied to cropland sites by state, 1972—National Soils Monitoring Program*

Compound	Trade Name if Noted	Sites Reporting Application		Average Total Application	
		No.	%	Lb/Acre	Kg/Ha
Methoxychlor		1	0.9	0.01	0.01
Methylmercury dicyandiamide	Panogen	2	1.9	0.01	0.01
Parathion, ethyl		1	0.9	0.25	0.28
Propachlor	Ramrod	5	4.7	1.69	1.89
Phorate	Thimet	1	0.9	0.70	0.78
TENNESSEE, 22 SITES					
Alachlor	Lasso	1	4.5	2.00	2.24
Atrazine	AAtrex	3	13.6	1.60	1.79
Dimethoate	Cygon	1	4.5	2.00	2.24
Diuron	Karmex	1	4.5	0.11	0.12
Disulfoton	Di-Syston	1	4.5	0.72	0.81
Ethylmercury chloride	Ceresan Red	1	4.5	0.01	0.01
Folex		1	4.5	1.50	1.68
Fluometuron	Cotoran	2	9.1	0.91	1.01
Linuron	Lorox	2	9.1	0.63	0.70
MSMA		1	4.5	2.00	2.24
Paraquat		1	4.5	0.50	0.56
PCNB	Terrachlor	1	4.5	0.01	0.01
Prometryn		1	4.5	0.24	0.27
Trifluralin	Treflan	4	18.2	1.04	1.16
VIRGINIA, WEST VIRGINIA,[1] 24 SITES					
Atrazine	AAtrex	1	4.2	1.60	1.79
Carbaryl	Sevin	2	8.3	3.17	3.55
Dalapon		1	4.2	13.60	15.24
Naptalam	Alanap	1	4.2	1.50	1.68
Paraquat		1	4.2	0.50	0.56
Phorate	Thimet	1	4.2	0.70	0.78
Simazine		1	4.2	1.60	1.79
WASHINGTON STATE, 45 SITES					
Aldrin		1	2.2	0.05	0.06
Benzene hexachloride	BHC	2	4.4	1.25	1.40
Bromacil	Hyvar	1	2.2	0.75	0.84
Captan		5	11.1	0.03	0.03
Cycloate	Ro-Neet	1	2.2	0.25	0.28
2,4-D		12	26.7	1.31	1.47
Dicamba	Banvel D	1	2.2	0.25	0.28
Dieldrin		1	2.2	0.04	0.04
Diuron	Karmex	1	2.2	0.50	0.56
EPTC	Eptam	1	2.2	0.25	0.28
Hexachlorobenzene	No-Bunt	9	20.0	0.05	0.05
Heptachlor		1	2.2	0.01	0.01
MCPA		2	4.4	1.25	1.40
Mercury		3	6.7	0.01	0.01
Oxythioquinox	Morestan	1	2.2	0.08	0.09
Phenylmercury acetate	PMA	2	4.4	0.03	0.03
Phorate	Thimet	1	2.2	0.03	0.03
Propham	IPC	1	2.2	3.00	3.36
Propargite	Omite	1	2.2	1.50	1.68
Terbacil	Sinbar	1	2.2	1.50	1.68
WISCONSIN, 66 SITES					
Alachlor	Lasso	5	7.6	0.95	1.06
Atrazine	AAtrex	16	24.2	1.33	1.49
Captafol	Difolatan	1	1.5	0.50	0.56
Carbaryl	Sevin	1	1.5	1.50	1.68
Carbofuran	Furadan	1	1.5	6.00	6.72
Chlordane		1	1.5	8.00	8.97
Cyanazine	Bladex	1	1.5	1.50	1.68
2,4-D		4	6.1	0.69	0.78
Diazinon		1	1.5	0.01	0.01
EPTC	Eptam	1	1.5	2.00	2.24
Fensulfothion	Dasanit	1	1.5	0.75	0.84
Heptachlor		1	1.5	2.00	2.24
Linuron	Lorox	1	1.5	1.25	1.40
MCPB		2	3.0	0.63	0.70
Phorate	Thimet	5	7.6	5.32	5.96
Thiram	Arasan	1	1.5	0.01	0.01

[1] Because some small eastern states had very few sites, those with similar geographic location and/or agricultural characteristics were combined to obtain more representative data. State groups used were: Mid-Atlantic: Delaware, Maryland, and New Jersey; New England: Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, and Vermont; and Virginia and West Virginia.

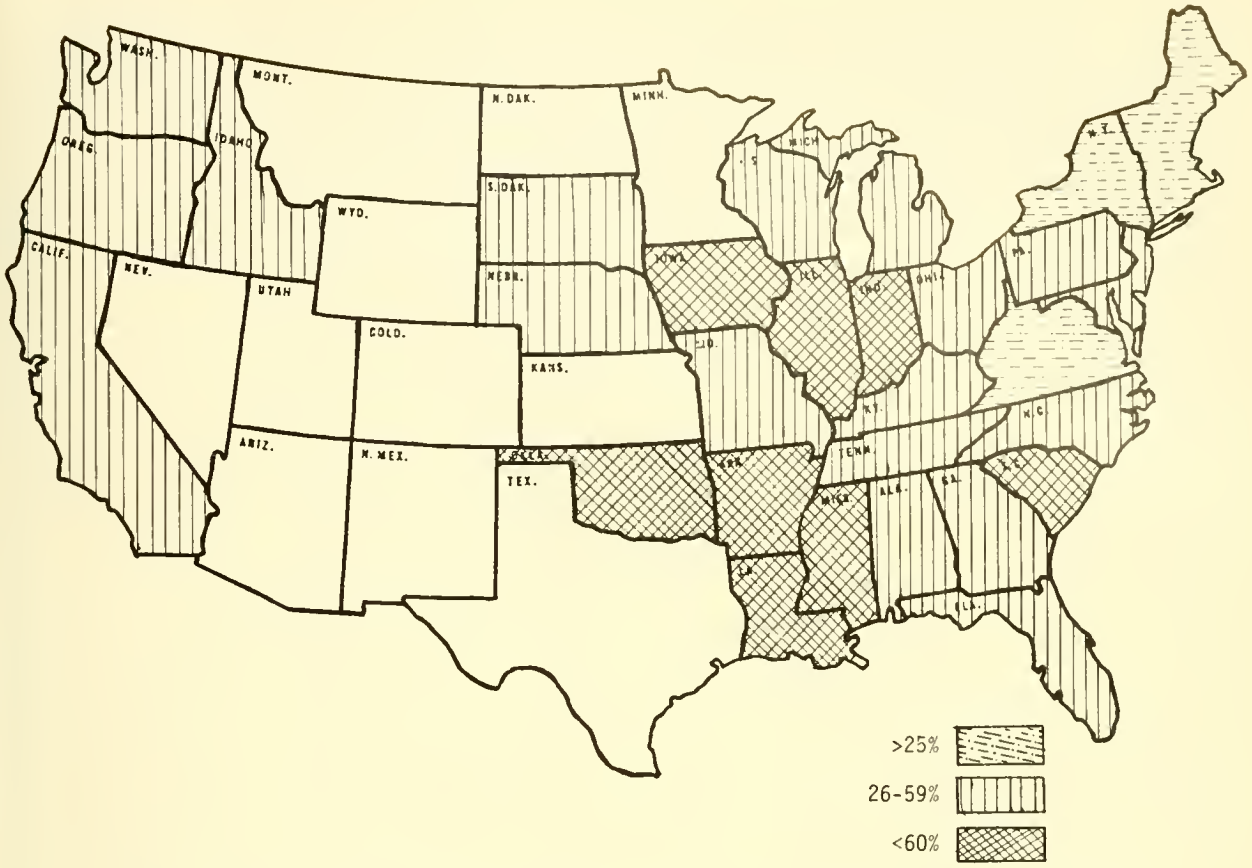

FIGURE 2. *Percent of sites reporting pesticide applications, 1972—National Soils Monitoring Program*

tion; and high, states where more than 60 percent of the sites reported pesticide application.

BY CROP

Table 4 lists crops grown on sample sites in 1972, and illustrates the diversity of crops grown in the United States. Application data for several major crops are presented in Table 5. Pesticide use varied widely among these crops. Thirty-nine different compounds were applied to field corn sites but only five compounds were applied to more than 10 percent of the sites. Cotton-growing sites also received applications of 39 compounds, but only 11 compounds were applied to more than 10 percent of the sites.

Table 6 shows pesticide applications on several crops by state. Differences in pesticide use among selected crops are apparent. For example, only 10.6 percent of the sites growing alfalfa and/or bur clover reported any pesticide applications, but 81.5 percent of the cotton sites did.

Acknowledgments

It is not possible to list all the persons who contributed to this study. However, the authors are especially grate-

TABLE 4. *List of crops grown on 1,402 sampling sites, 1972—National Soils Monitoring Program*

Crop	No. of Sites	Crop	No. of Sites
Field corn	364	Potatoes	3
Soybeans	266	Blueberries	2
Wheat	111	Apples	2
Mixed hay	105	Peaches	2
Alfalfa and/or bur clover	104	Turf	2
Pasture	66	Almonds	2
Cotton	54	Chick peas	2
Grass hay	42	Range	2
Oats	41	Sweet corn	2
Sorghum	24	Apricots	1
Barley	12	Plums	1
Oranges	9	*Lespedeza sericea*	1
Dry beans	9	Sweet clover	1
Silage (corn or sorghum)	8	Mint	1
Peas	7	Hops	1
Grapes	6	Sweet sorghum	1
Rye	6	Celery	1
Tobacco	5	Green peppers	1
Sugar beets	5	Lettuce	1
Rice	4	Pumpkins	1
Milo	4	Tomatoes	1
String beans	4	Millet	1
Pecans	3	Sunflowers	1
Flax	3	Other	9
Sugarcane	3	Fallow sites	129
Asparagus	3		

TABLE 5. *Compounds applied to cropland sites, by most common crop, 1972—National Soils Monitoring Program*

Compound	Sites Reporting Application		Average Total Application	
	No.	%	Lb/Acre	Kg/Ha
ALFALFA and BUR CLOVER, 104 SITES				
Carbaryl	3	2.9	2.33	2.61
Carbofuran	1	1.0	0.25	0.28
EPTC	1	1.0	2.00	2.24
IPC	1	1.0	3.00	3.36
Malathion	1	1.0	1.00	1.12
Methoxychlor	1	1.0	2.00	2.24
Parathion, ethyl	2	1.9	0.38	0.43
Picloram	1	1.0	0.75	0.84
Prolate	1	1.0	0.75	0.84
COTTON, 54 SITES				
Aldicarb	2	3.7	0.40	0.45
Azinphosmethyl	1	1.9	0.50	0.56
Captan	3	5.6	0.02	0.02
Carbaryl	1	1.9	1.00	1.12
Chloroneb	8	14.8	0.2	0.03
DDT	16	29.6	7.44	8.34
DEF	6	11.1	0.99	1.11
Dibromochloropropane	1	1.9	0.50	0.56
Dicrotophos	2	3.7	0.08	0.09
Dimethoate	1	1.9	2.00	2.24
Disulfoton	7	13.0	0.11	0.12
Diuron	9	16.7	0.80	0.90
DNBP	4	7.4	1.06	1.19
DSMA	8	14.8	2.51	2.81
EMTS	3	5.6	0.01	0.01
EPN	1	1.9	3.00	3.36
Ethylmercury chloride	1	1.9	0.01	0.01
Fenaminosulf	1	1.9	0.01	0.01
Fluometuron	22	40.7	0.98	1.09
Folex	5	9.3	1.10	1.23
Linuron	3	5.6	1.33	1.49
MCPB	2	3.7	0.50	0.56
Mercury	7	13.0	0.05	0.05
Methylmercury acetate	5	9.3	0.01	0.01
Monocrotophos	3	5.6	1.66	1.87
MSMA	20	37.0	2.38	2.67
Naled	1	1.9	1.00	1.12
Nitralin	2	3.7	0.88	0.98
Norea	2	3.7	1.10	1.23
Paraquat	1	1.9	0.02	0.02
Parathion, methyl	24	44.4	4.61	5.17
PCNB	1	1.9	0.01	0.01
Phorate	1	1.9	1.00	1.12
Prometryn	4	7.4	0.87	0.98
Propargite	1	1.9	1.68	1.88
Sodium chlorate	2	3.7	1.00	1.12
Thiram	1	1.9	0.01	0.01
Toxaphene	21	38.9	12.76	14.30
Trifluralin	3	55.6	0.91	1.02
FIELD CORN, 364 SITES				
Alachlor	36	9.9	1.19	1.33
Aldrin	31	8.5	1.67	1.88
Atrazine	188	51.8	1.56	1.74
Butylate	17	4.7	1.68	1.89
Bux	25	6.9	0.92	1.03
Captan	82	22.6	0.01	0.01
Carbaryl	1	0.3	1.33	1.49
Carbofuran	14	3.9	0.78	0.87
Chloramben	2	0.6	0.75	0.84
Chlordane	4	1.1	3.79	4.25
Cyanazine	1	0.3	2.80	3.14
2,4-D	73	20.1	0.62	0.69
Dalapon	1	0.3	13.60	15.24
Diazinon	5	1.4	1.10	1.13
Dicamba	10	2.8	0.38	0.43
Dicofol	1	0.3	1.00	1.12
Dieldrin	1	0.3	0.01	0.01
Disulfoton	1	0.3	0.50	0.56
Dyfonate	2	0.6	0.95	1.06
EPTC	2	0.6	5.21	5.84
Ethoprop	1	0.3	1.00	1.12
Fensulfothion	2	0.6	0.83	0.92
Heptachlor	4	1.1	1.58	1.77
Lindane	1	0.3	0.01	0.01
Linuron	3	0.8	0.95	1.06
Malathion	75	20.7	0.01	0.01
Methoxychlor	8	2.2	0.01	0.01
Methylmercury acetate	1	0.3	0.01	0.01
Mirex	2	0.6	0.01	0.01
Naptalam	1	0.3	0.83	0.93
Paraquat	1	0.3	0.50	0.56
Pentachlorophenol	1	0.3	0.05	0.06
Phorate	19	5.2	1.67	1.88
Propachlor	35	9.6	1.83	2.05
Simazine	2	0.5	2.80	3.14
TCBC	1	0.3	8.00	8.97
Thiram	4	1.1	0.01	0.01
Toxaphene	1	0.3	0.40	0.45
Trietazine	1	0.3	0.25	0.28
MIXED HAY, 105 SITES				
Carbofuran	1	1.0	1.00	1.12
Chlordane	1	1.0	0.75	0.84
2,4-D	2	1.9	0.42	0.47
EPTC	1	1.0	3.00	3.36
Malathion	1	1.0	0.50	0.56
Propham	1	1.0	0.50	0.56
SOYBEANS, 266 SITES				
Alachlor	44	16.5	1.54	1.72
Ancrack	4	1.5	1.08	1.20
Butyrac	1	0.4	0.40	0.45
Captan	5	1.9	0.01	0.01
Carbaryl	7	2.6	1.83	2.06
Chloramben	49	18.4	1.38	1.54
Chloropropham	1	0.4	0.59	0.66
Chloroxuron	1	0.4	2.00	2.24
2,4-D	4	1.5	0.96	1.07
2,4-DB	6	2.3	1.00	1.12
DDT	3	1.1	0.66	0.74
Dimethoate	1	0.4	0.66	0.74
Dinitrocresol	1	0.4	3.00	3.36
DNBP	9	3.4	1.54	1.73
Fluometuron	1	0.4	0.50	0.56
Linuron	32	12.0	1.11	1.24
Methyl trithion	1	0.4	0.25	0.28
Metribuzin	1	0.4	0.50	0.56
Mirex	1	0.4	0.01	0.01
MSMA	1	0.4	2.00	2.24
Naptalam	5	1.9	1.30	1.46
Nitralin	6	2.3	1.36	1.52
Paraquat	2	0.8	0.38	0.42
Parathion, methyl	6	2.3	0.71	0.79
Phorate	1	0.4	0.70	0.78
Propachlor	2	0.8	1.90	2.13
Simazine	1	0.4	3.00	3.36
Thiram	3	1.1	0.08	0.09
Toxaphene	5	1.9	1.00	1.12
Trifluralin	59	22.2	0.87	0.97
Vernolate	3	1.1	1.65	1.85
WHEAT, 111 SITES				
Aldrin	1	0.9	0.05	0.06
Benzene hexachloride	2	1.8	1.25	1.40
Bromacil	1	0.9	0.75	0.84
Captan	5	4.5	0.03	0.03
2,4-D	27	24.3	0.84	0.94
Dicamba	1	0.9	0.25	0.28
Diuron	1	0.9	0.50	0.56
EMTS	3	2.7	0.01	0.01
Ethylmercury chloride	4	3.6	0.01	0.01
Heptachlor	1	0.9	0.01	0.01
Hexachlorobenzene	10	9.0	0.05	0.06
Malathion	1	0.9	0.01	0.01
Mercury	2	1.8	0.01	0.01
Methylmercury dicyandiamide	1	0.9	0.01	0.01
Parathion, methyl	7	6.3	0.50	0.56
PCNB	6	5.4	0.02	0.03
Phenylmercury acetate	2	1.8	0.03	0.03
Thiram	2	1.8	0.01	0.01
Trifluralin	1	0.9	0.50	0.56

TABLE 6. *Pesticide applications on selected crops, by state, 1972—National Soils Monitoring Program*

State	ALFALFA/BUR CLOVER				COTTON			
	No. of Sites	Pesticides Applied	No Pesticides Applied	Pesticide Use Unknown	No. of Sites	Pesticides Applied	No Pesticides Applied	Pesticide Use Unknown
Alabama	0	—	—	—	3	3	—	—
Arkansas	0	—	—	—	13	12	1	—
California	8	2	4	2	4	2	1	1
Florida	0	—	—	—	0	—	—	—
Georgia	0	—	—	—	2	1	—	1
Idaho	5	1	4	—	0	—	—	—
Illinois	4	—	4	—	0	—	—	—
Indiana	4	—	4	—	0	—	—	—
Iowa	10	—	10	—	0	—	—	—
Kentucky	1	1	—	—	0	—	—	—
Louisiana	0	—	—	—	6	6	—	—
Michigan	5	—	5	—	0	—	—	—
Mid-Atlantic[1]	0	—	—	—	0	—	—	—
Mississippi	0	—	—	—	10	9	—	1
Missouri	2	—	2	—	6	6	—	—
Nebraska	10	—	9	1	0	—	—	—
New England[1]	0	—	—	—	0	—	—	—
New York	2	1	1	—	0	—	—	—
N. Carolina	0	—	—	—	0	—	—	—
Ohio	4	1	3	—	0	—	—	—
Oklahoma	3	2	1	—	7	2	5	—
Oregon	2	—	2	—	0	—	—	—
Pennsylvania	3	—	3	—	0	—	—	—
S. Carolina	0	—	—	—	0	—	—	—
S. Dakota	14	1	13	—	0	—	—	—
Tennessee	0	—	—	—	3	3	—	—
Virginia/W. Virginia[1]	0	—	—	—	0	—	—	—
Washington state	6	1	5	—	0	—	—	—
Wisconsin	21	1	20	—	0	—	—	—
Total	104	11	90	3	54	44	7	3
%	100.0	10.6	86.5	2.9	100.0	81.5	13.0	5.5

State	FIELD CORN				SOYBEANS			
	No. of Sites	Pesticides Applied	No Pesticides Applied	Pesticide Use Unknown	No. of Sites	Pesticides Applied	No Pesticides Applied	Pesticide Use Unknown
Alabama	7	1	6	—	2	2	—	—
Arkansas	0	—	—	—	25	16	9	—
California	2	1	—	1	0	—	—	—
Florida	1	1	—	—	1	—	1	—
Georgia	5	2	3	—	5	5	—	—
Idaho	1	1	—	—	0	—	—	—
Illinois	56	54	2	—	50	36	14	—
Indiana	27	25	2	—	24	19	5	—
Iowa	73	65	7	1	48	41	5	2
Kentucky	8	5	—	3	3	1	1	1
Louisiana	2	1	1	—	8	7	1	—
Michigan	22	18	4	—	4	4	—	—
Mid-Atlantic[1]	6	4	2	—	5	5	—	—
Mississippi	1	1	—	—	13	11	2	—
Missouri	16	14	2	—	23	18	4	1
Nebraska	31	26	5	—	3	2	1	—
New England[1]	0	—	—	—	0	—	—	—
New York	13	9	4	—	0	—	—	—
N. Carolina	8	4	4	—	9	4	5	—
Ohio	23	19	4	—	16	10	6	—
Oklahoma	0	—	—	—	0	—	—	—
Oregon	2	1	—	1	0	—	—	—
Pennsylvania	14	12	2	—	1	1	—	—
S. Carolina	1	1	—	—	10	7	3	—
S. Dakota	16	16	—	—	4	3	1	—
Tennessee	4	3	1	—	6	4	2	—
Virginia/W. Virginia[1]	1	1	—	—	5	2	2	1
Washington state	0	—	—	—	0	—	—	—
Wisconsin	24	17	5	2	1	1	—	—
Total	364	302	54	8	266	199	62	5
%	100.0	82.9	14.9	2.2	100.0	74.8	23.3	1.9

State	WHEAT				MIXED HAY			
	No. of Sites	Pesticides Applied	No Pesticides Applied	Pesticide Use Unknown	No. of Sites	Pesticides Applied	No Pesticides Applied	Pesticide Use Unknown
Alabama	0	—	—	—	1	—	1	—
Arkansas	0	—	—	—	1	—	1	—
California	2	—	1	1	1	1	—	—
Florida	0	—	—	—	0	—	—	—
Georgia	0	—	—	—	0	—	—	—
Idaho	4	4	—	—	3	—	3	—
Illinois	7	2	5	—	6	1	5	—
Indiana	9	—	9	—	0	—	—	—
Iowa	0	—	—	—	1	—	1	—
Kentucky	0	—	—	—	1	—	1	—

(Continued next page)

TABLE 6 (cont'd.). *Pesticide applications on selected crops, by state, 1972—National Soils Monitoring Program*

State	WHEAT				MIXED HAY			
	No. of Sites	Pesticides Applied	No Pesticides Applied	Pesticide Use Unknown	No. of Sites	Pesticides Applied	No Pesticides Applied	Pesticide Use Unknown
Louisiana	0	—	—	—	0	—	—	—
Michigan	0	—	—	—	8	—	8	—
Mid-Atlantic[1]	0	—	—	—	0	—	—	—
Mississippi	0	—	—	—	0	—	—	—
Missouri	5	—	5	—	11	—	11	—
Nebraska	14	—	12	2	0	—	—	—
New England[1]	0	—	—	—	2	—	2	—
New York	0	—	—	—	12	—	12	—
N. Carolina	0	—	—	—	0	—	—	—
Ohio	7	1	6	—	12	—	12	—
Oklahoma	25	16	9	—	0	—	—	—
Oregon	6	6	—	—	10	3	7	—
Pennsylvania	0	—	—	—	8	—	8	—
S. Carolina	0	—	—	—	1	—	1	—
S. Dakota	14	9	5	—	7	—	7	—
Tennessee	2	—	2	—	1	—	1	—
Virginia/W. Virginia[1]	0	—	—	—	8	—	8	—
Washington state	0	—	—	—	0	—	—	—
Wisconsin	16	15	1	—	12	—	12	—
Total	111	53	55	3	106	5	101	—
%	100.0	47.8	49.5	2.7	100.0	5.0	95.0	0

ful to the inspectors from the Plant Protection and Quarantine Programs, Animal and Plant Health Inspection Service, U.S. Department of Agriculture, for collecting the data.

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Pesticide Residue Levels in Soils and Crops from 37 States, 1972— National Soils Monitoring Program (IV)

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ABSTRACT

Residue data from the 1972 (FY–73) National Soils Monitoring Program are summarized. Composite samples of agricultural soil and mature crops were collected from 1,483 of the 1,533 selected 4-hectare sites in 37 states. Analyses were performed for organochlorine and organophosphorus compounds, trifluralin, and polychlorinated biphenyls (PCBs); analysis for atrazine was performed only when pesticide application data indicated current-year use. Organochlorine pesticides were detected in 45 percent of the soil samples. The most frequently detected compound was dieldrin, found in 27 percent of all soil samples. Other compounds detected, in order of frequency, included DDT, aldrin, chlordane, and heptachlor epoxide, found, respectively, in 21, 9, 8, and 7 percent of all soil samples. Crop samples were collected from 727 sites. All were analyzed for organochlorines; analyses were performed for organophosphates and atrazine only when pesticide application data indicated current-year use. For all crops, 40 percent of the samples contained detectable levels of organochlorines and 10 percent contained detectable levels of organophosphates. Atrazine was not detected.

Introduction

The National Pesticide Monitoring Program (NPMP) was initiated at the recommendation of the President's Science Advisory Committee in 1963 to "develop a continuing network to monitor residue levels in air, water, soil, man, wildlife and fish" (*8*). The primary objective of the NPMP is to determine levels and trends of pesticides and their degradation products in various components of the environment (*5*). The National Soils Monitoring Program (NSMP) was established in 1968 as an integral part of NPMP to monitor residues in agricultural soils and raw agricultural crops.

The present report summarizes soil and crop pesticide concentration data collected from 1,483 sampling sites in 37 states during 1972 (FY–73). Data were not collected from all conterminous states because of budgetary limitations. The states omitted from the survey were generally large, western states either having little widespread agriculture or growing primarily wheat and other small grains, which require fewer pesticides than do other nongrain crops.

Sampling Procedures

A total of 1,533 sites in 37 states were scheduled for sampling during late summer and fall of 1972 (Fig. 1). Site selection criteria, statistical design, and sampling techniques involved in the present study have been described (*3, 8*). At each 4-hectare (10-acre) site, a composite soil sample and a composite mature crop sample, if available, were collected according to established procedures (*6*). In addition, information on cropping practices and a history of pesticide applications for the current cropping season were obtained in a personal interview with the landowner or operator. These data have been summarized and published separately (*1*).

Analytical Procedures

ORGANOCHLORINES AND ORGANOPHOSPHATES

Sample Preparation, Soil—A 100-g subsample was taken from a thoroughly mixed field sample. The subsample was moistened with 25 ml distilled water and extracted with 200 ml 3:1 hexane:isopropanol solvent by shaking for 4 hours on a reciprocating shaker. The isopropanol was removed by three distilled water washes and the hexane extract was dried through anhydrous sodium sulfate. The sample extract was then stored at low temperature for subsequent gas-liquid chromatographic (GLC) analysis.

Crops—For samples containing less than 2 percent fat (e.g., alfalfa, bur clover, corn stalks, cotton stalks, green bolls, miscellaneous hay), a 100-g sample of the crop was dry blended for 3 minutes and then blended for 5

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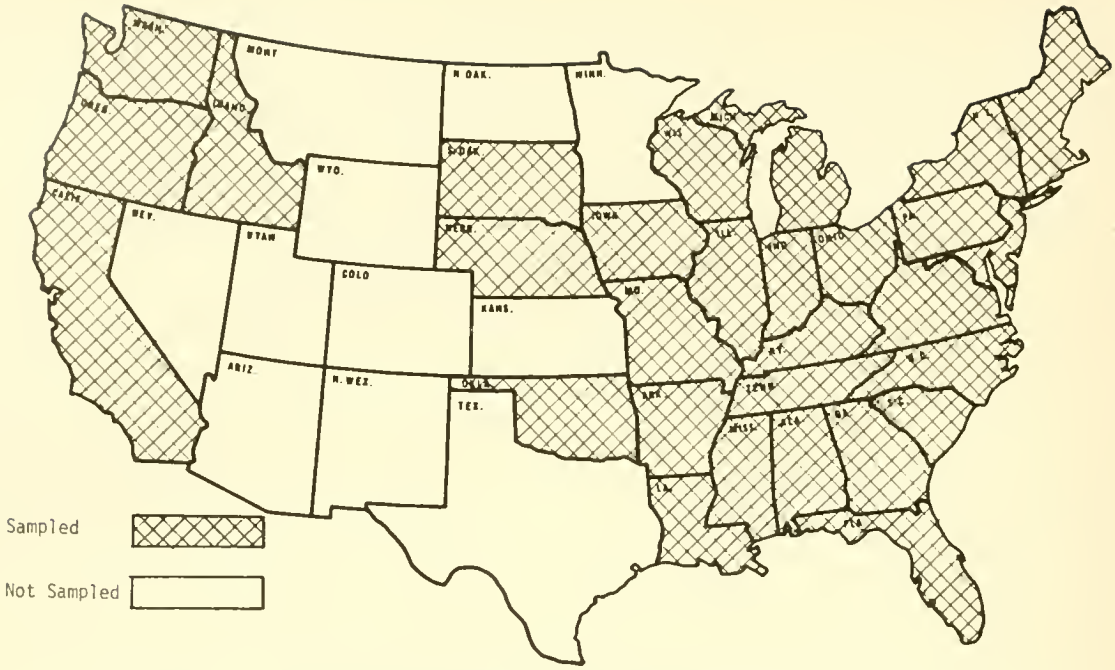

FIGURE 1. *States where agricultural soils and crops were sampled, 1972 (FY 1973) —National Soils Monitoring Program*

minutes in 800 ml acetonitrile. An aliquot of the sample extract, representing 10 g of the original sample, was decanted into a 500-ml Erlenmeyer flask. The extract was concentrated under a three-ball Snyder column to approximately 10 ml, 100 ml hexane was added, and the hexane–acetonitrile azeotrope was again concentrated to 10 ml. The process was carried out three times to remove essentially all acetonitrile. The hexane extract was dried through anhydrous sodium sulfate, the volume was adjusted to 50 ml, and the extract was stored at low temperature.

For crop samples containing more than 2 percent fat (e.g., corn kernels, cottonseed, soybeans), a 100-g sample was prewashed with 100 ml isopropanol and then with 100 ml hexane. Both prewashes were discarded. The sample was extracted as described in the preceding paragraph. A separate aliquot of the extract, not subjected to Florisil cleanup, was reserved for flame photometric analysis for organophosphates.

Florisil Cleanup—An extract equivalent to 5 g original crop sample was fractionated through a 15-g Florisil column into two fractions by use of 100 ml 10 percent methylene chloride in hexane and 100 ml methylene chloride for fractions 1 and 2, respectively.

Methylene chloride was removed by concentrating each extract to low volume under a three-ball Snyder column, adding 100 ml hexane, and concentrating again to low volume. After two additions of hexane, the methylene chloride was essentially removed. Each extract volume was adjusted to 2.5 ml for separate injection on the gas-liquid chromatograph.

GLC—Analyses were performed on gas chromatographs equipped with tritium foil electron-affinity detectors for organohalogens and thermionic or flame photometric detectors for organophosphates. A multiple-column system of polar and nonpolar columns was used to identify compounds. Instrument parameters and operating conditions follow:

Gas chromatographs:	Hewlett-Packard Model 402A Hewlett-Packard Model 402B Tracor Model MT-220	
Columns:	glass, 6 mm OD × 4 mm ID, 183 cm long, packed with one of the following: 5 percent OV-210 on 80–100-mesh Chromosorb W-HP; 3 percent DC-200 on 100–120-mesh Gas-Chrom Q; a mixture of 1.5 percent OV-17 and 1.95 percent QF-1 on 100–120-mesh Supelcoport	
Temperatures, °C:	thermionic detector housing	250
	detector (EC and FPD)	200
	injection port	250
	column OV-210	166
	column DC-200	170–175
	mixed column	185–190
Carrier gases:	5 percent methane–argon flowing at 80 ml/minute; prepurified nitrogen flowing at 80 ml/minute	

Sensitivity or minimum detection levels for organochlorines and trifluralin were 0.002–0.03 ppm except for combinations of polychlorinated biphenyls (PCBs), chlordane, toxaphene, and other chemicals which had minimum detectable levels of 0.05–0.1 ppm. Minimum detectable levels for organophosphates were approximately 0.01–0.03 ppm. Compounds detectable by this

methodology are listed in Table 1. When necessary, residues were confirmed on a Dohrmann microcoulometric detector or a Coulson electrolytic conductivity detector. Because trifluralin is detected by the organochlorine methodology, it appears with the organochlorine analyses in the tables.

TABLE 1. *Compounds detectable by chemical methodology of the present study*

Organochlorines	
Alachlor	Endrin ketone
Aldrin	Heptachlor
Benzene hexachloride	Heptachlor epoxide
Chlordane	Hexachlorobenzene
ΣDDT	Isodrin
Dieldrin	Lindane (γ-BHC)
DCPA	Methoxychlor
Dicofol	Ovex
Endosulfan I	PCBs
Endosulfan II	PCNs
Endosulfan sulfate	Propachlor
Endrin	Toxaphene
Organophosphates	
DEF	Parathion, ethyl
Diazinon	Parathion, methyl
Ethion	Ronnel
Malathion	Trithion
Phorate	
Other Halogenated Hydrocarbons	
Trifluralin[1]	

[1] Although trifluralin is a dinitroaniline compound, it is detected by the organochlorine methodology and thus appears with organochlorines in Tables 2–7.

Recovery Studies—Pesticide recovery values from soil were 80–110 percent, but usually were close to 100 percent. Values from crops ranged from 70 to 100 percent, depending on the amount of pesticide present, the individual pesticide, and the type of crop involved. Residue concentrations detected in both soil and crop samples were corrected for recovery. Soil samples were also converted to a dry-weight basis.

ATRAZINE

To analyze soil samples for atrazine, a 50-g subsample was taken from a thoroughly mixed field sample. The subsample was placed in the Soxhlet thimble and moistened with 40 ml 1:1 distilled water:methanol. After addition of 250 ml nanograde methanol, the sample was extracted for 4 hours. The extract in the Soxhlet flask was evaporated to about 50 ml on a hot plate and by use of a three-ball Snyder column. The sample extract was then decanted into a 1-liter separatory funnel. The extract was partitioned three times with 150 ml Freon 113 each time. The Freon 113 fractions were combined and concentrated to incipient dryness on a rotary evaporator. The extract was dissolved in isooctane and adjusted to 5 ml for injection into a gas-liquid chromatograph.

GLC—A Coulson electrolytic conductivity cell detector in the nitrogen mode was used for detection and quantification of the atrazine. Positive samples were confirmed by alkali flame detection. Recovery rate was 90–110 percent; minimum detection level was 0.01 ppm.

Results and Discussion

Tables 2–5 show concentrations of pesticides in soil samples, and Tables 6–8 show concentrations of pesticides in mature agricultural crops. Soil concentration data are also summarized by all sites and by state or state groups. Most tables list the number of analyses, the number of times a compound was detected, percent occurrence of the compound, the arithmetic mean, the estimated geometric mean, and the minimum and maximum positive concentrations detected.

The estimated geometric mean is routinely presented in the tables as an alternative to the arithmetic mean as a measure of central tendency for the data evaluation. Pesticide residue data frequently contain a large number of zero values, resulting either from the absence of pesticides or their presence at levels below the analytical sensitivity. Such data are seldom distributed normally, as shown by tests for skewness and kurtosis, but often tend to approximate a log-normal distribution. After repeated tests for significant skewness and/or kurtosis, the log $(X + 0.01)$ transformation was used to determine the logarithmic means. The antilogs of these figures, minus 0.01, were taken to obtain the estimates of the geometric mean in the untransformed dimension. The estimated geometric mean was calculated only for those compounds with more than one positive detection.

COMPOUND CONCENTRATIONS IN CROPLAND SOIL

All Sites—Soil samples were received from 1,483 of the scheduled 1,533 sites in 37 states. Results of analyses for organochlorine and organophosphorus pesticides and atrazine are presented in Table 2. The most frequently detected chemical was dieldrin, found in 27 percent of all samples analyzed. Other compounds, in order of frequency, included ΣDDT, aldrin, chlordane, and heptachlor epoxide found, respectively, in 21, 9, 8, and 7 percent of all samples analyzed.

Table 3 lists the occurrence of pesticide residues in the agricultural soil samples collected during 1972. The frequency of detection varied widely among the states surveyed. The detection frequencies of atrazine appear to be much higher for individual states than in other analyses because atrazine analyses were performed only when site application records indicated its use during the current growing season.

Table 4 presents the percent incidence of residues of selected pesticides at specific levels. For most of the compounds listed, the highest percentage of positive

TABLE 2. *Compound concentrations in cropland soil for all sample sites in 37 states, 1972 (FY 1973) —National Soils Monitoring Program*

Compound	Positive Detections		Residues, ppm dry weight			
			Arithmetic Mean	Estimated Geometric Mean[1]	Extremes of Detected Values	
	No.	%			Min.	Max.
ORGANOCHLORINES (1,483 samples)						
Aldrin	129	8.7	0.03	0.002	0.01	13.28
Benzene hexachloride	1	0.1	<0.01	—	0.02	
Chlordane	117	7.9	0.05	0.003	0.01	7.89
DCPA	1	0.1	<0.01	—	0.18	
o,p'-DDE	10	0.7	<0.01	<0.001	0.01	0.09
p,p'-DDE	299	20.2	0.05	0.006	0.01	7.16
o,p'-DDT	161	10.9	0.03	0.003	0.01	5.62
p,p'-DDT	275	18.5	0.13	0.007	0.01	18.93
o,p'-TDE	1	0.1	<0.01	—	0.31	
p,p'-TDE	46	3.1	0.01	0.001	0.01	8.20
ΣDDT	314	21.2	0.22	0.010	0.01	29.45
Dicofol	7	0.5	<0.01	<0.001	0.06	2.15
Dieldrin	403	27.2	0.04	0.008	0.01	6.18
Endosulfan I	1	0.1	<0.01	—	0.08	
Endosulfan II	1	0.1	<0.01	—	0.25	
Endosulfan sulfate	1	0.1	<0.01	—	0.31	
Endrin	10	0.7	<0.01	<0.001	0.01	2.13
Endrin ketone	2	0.1	<0.01	<0.001	0.02	0.38
Heptachlor	57	3.9	<0.01	0.001	0.01	0.60
Heptachlor epoxide	97	6.6	<0.01	0.001	0.01	0.72
Hexachlorobenzene	11	0.7	<0.01	<0.001	0.01	0.44
PCB	2	0.1	<0.01	<0.001	0.80	1.49
PCNB	3	0.2	<0.01	<0.001	0.22	2.61
Propachlor	1	0.1	<0.01	—	0.10	
Ronnel	1	0.1	<0.01	—	0.19	
Toxaphene	76	5.1	0.24	0.003	0.22	46.58
Trifluralin[2]	81	5.5	0.01	0.001	0.01	1.86
ORGANOPHOSPHATES (1,246 samples)						
DEF	4	0.3	<0.01	<0.001	0.06	0.67
Diazinon	3	0.2	<0.01	<0.001	0.07	0.17
Malathion	2	0.2	<0.01	<0.001	0.08	0.13
Parathion, ethyl	7	0.6	<0.01	<0.001	0.02	0.19
Parathion, methyl	1	0.1	<0.01	—	0.01	
Phorate	13	1.0	<0.01	<0.001	0.01	0.04
TRIAZINE (151 samples)						
Atrazine	134	88.7	0.10	0.051	0.01	0.77

[1] Not calculated when fewer than two positive detections present.
[2] See footnote, Table 1.

TABLE 3. *Occurrence of pesticide residues in cropland soils from 37 states, 1972—National Soils Monitoring Program*

State	Organochlorines[1]			Organophosphates			Atrazine[2]		
	No. of Analyses	Positive Detections		No. of Analyses	Positive Detections		No. of Analyses	Positive Detections	
		No.	%		No.	%		No.	%
Alabama	22	18	82	22	1	5	—		
Arkansas	43	37	86	43	0	—	—		
California	64	45	70	53	4	8	—		
Florida	17	12	71	17	0	—	1	0	—
Georgia	29	22	76	28	4	14	—		
Idaho	29	15	52	25	0	—	—		
Illinois	139	100	72	87	2	2	18	17	94
Indiana	78	27	35	59	0	—	4	4	100
Iowa	150	101	67	113	1	1	34	34	100
Kentucky	28	10	36	15	0	—	3	2	67
Louisiana	27	21	78	26	3	11	—		
Michigan	53	9	17	44	0	—	14	14	100
Mid-Atlantic[3]	14	7	50	14	0	—	1	0	—
Mississippi	30	25	83	25	3	12	—		
Missouri	82	33	40	66	0	—	13	13	100
Nebraska	101	39	39	86	0	—	19	17	90
New England[3]	20	7	35	20	0	—	—		
New York	36	13	36	35	0	—	6	6	100
N. Carolina	31	19	61	28	3	11	—		
Ohio	67	20	30	53	0	—	8	7	88
Oklahoma	64	7	11	64	0	—	—		
Oregon	37	11	30	33	0	—	—		
Pennsylvania	37	11	30	37	0	—	7	5	71
S. Carolina	17	15	88	17	1	6	—		
S. Dakota	106	12	11	90	2	2	—		
Tennessee	25	15	60	21	0	—	2	2	100
Virginia/W. Virginia[3]	25	6	24	25	0	—	2	0	—
Washington state	45	9	20	43	0	—	—		
Wisconsin	67	8	12	57	0	—	16	15	94

[1] Although trifluralin is a dinitroaniline compound, it is detected by the organochlorine methodology and thus appears with organochlorines in Tables 2–7.

[2] Samples analyzed only when application records indicated atrazine use during the current growing season.

[3] Because some small eastern states had very few sites, those with similar geographic location and/or agricultural characteristics were combined to obtain more representative data. State groups used were Mid-Atlantic: Delaware, Maryland and New Jersey; New England: Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, and Vermont; and Virginia and West Virginia.

TABLE 4. *Percent incidence of selected pesticides in cropland soil from all sampling sites in 37 states, 1972 —National Soils Monitoring Program*

Concentration, ppm dry wt	ΣDDT	Aldrin	Dieldrin	Chlordane	Heptachlor	Heptachlor Epoxide	Toxaphene	Trifluralin
Not detected	78.8	91.3	72.8	92.1	96.2	93.5	94.9	94.5
0.01– 0.25	11.7	7.3	23.6	3.3	3.6	6.4	0.1	5.0
0.26– 1.00	5.3	1.0	3.2	3.2	0.2	0.1	1.1	0.4
1.01– 5.00	3.1	0.3	0.3	1.3	—	—	2.6	0.1
5.01–10.00	0.7	—	0.1	0.1	—	—	0.9	—
>10.00	0.4	0.1	—	—	—	—	0.4	—
TOTAL	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0

TABLE 5. *Compound concentrations in cropland soils, by state, 1972—National Soils Monitoring Program*

Compound	Positive Detections		Residues, ppm dry weight			
	No.	%	Arithmetic Mean Concentration	Estimated Geometric Mean[1]	Extremes of Detected Values Min.	Max.
ALABAMA						
Organochlorines,[2] 22 samples						
Chlordane	1	4.6	0.01	—	0.16	
p,p'-DDE	14	63.6	0.08	0.028	0.01	0.58
o,p'-DDT	8	36.4	0.03	0.009	0.01	0.19
p,p'-DDT	15	68.2	0.16	0.042	0.01	1.24
ΣDDT	15	68.2	0.27	0.062	0.01	1.97
Dieldrin	2	9.1	<0.01	0.001	0.01	0.01
Endrin	1	4.6	<0.01	—	0.10	
Ronnel	1	4.6	<0.01	—	0.19	
Toxaphene	7	31.8	0.67	0.038	0.22	5.94
Trifluralin	4	18.2	0.02	0.006	0.07	0.17
Organophosphates, 22 samples						
Phorate	1	4.6	<0.01	—	0.04	
ARKANSAS						
Organochlorines,[2] 43 samples						
Chlordane	2	4.6	<0.01	0.001	0.03	0.08
o,p'-DDE	1	2.3	<0.01	—	0.03	
p,p'-DDE	25	58.1	0.16	0.036	0.01	1.87
o,p'-DDT	22	51.2	0.13	0.027	0.01	0.92
p,p'-DDT	27	62.8	0.54	0.083	0.01	4.49
p,p'-TDE	5	11.6	0.02	0.002	0.01	0.45
ΣDDT	27	62.8	0.85	0.114	0.03	7.35
Dieldrin	10	23.3	0.02	0.005	0.01	0.24
Endrin	2	4.6	0.01	0.001	0.02	0.24
Toxaphene	11	25.6	1.01	0.033	0.48	9.11
Trifluralin	17	39.5	0.04	0.015	0.01	0.31
Organophosphates, 43 samples: no residues detected						
CALIFORNIA						
Organochlorines,[2] 64 samples						
Chlordane	2	3.1	0.02	0.001	0.02	1.02
o,p'-DDE	3	4.7	<0.01	0.001	0.01	0.03
p,p'-DDE	44	68.7	0.16	0.042	0.01	2.72
o,p'-DDT	23	35.9	0.06	0.011	0.01	1.38
p,p'-DDT	32	50.0	0.26	0.033	0.02	5.62
p,p'-TDE	7	10.9	0.01	0.002	0.01	0.27
ΣDDT	45	70.3	0.49	0.074	0.01	9.72
Dicofol	4	6.3	0.05	0.003	0.38	2.15
Dieldrin	7	10.9	0.01	0.002	0.01	0.36
Hexachlorobenzene	1	1.6	0.01	—	0.44	
PCBs	1	1.6	0.02	—	1.49	
Toxaphene	9	14.1	0.25	0.010	0.46	6.45
Trifluralin	1	1.6	<0.01	—	0.05	
Organophosphates, 53 samples						
DEF	1	1.9	<0.01	—	0.10	
Malathion	1	1.9	<0.01	—	0.13	
Parathion, ethyl	4	7.6	0.01	0.002	0.02	0.19
FLORIDA						
Organochlorines, 17 samples						
Aldrin	1	5.9	0.01	—	0.16	
Chlordane	4	23.5	0.03	0.007	0.02	0.22
p,p'-DDE	10	58.8	0.08	0.017	0.01	0.66
o,p'-DDT	2	11.8	0.03	0.004	0.02	0.56
p,p'-DDT	10	58.8	0.21	0.022	0.01	2.16
p,p'-TDE	1	5.9	0.04	—	0.74	
ΣDDT	11	64.7	0.37	0.035	0.01	3.38
Dicofol	3	17.6	0.03	0.006	0.06	0.23
Dieldrin	3	17.6	0.08	0.009	0.15	1.09
Heptachlor epoxide	1	5.9	<0.01	—	0.01	
Toxaphene	3	17.6	0.83	0.019	2.04	9.00
Organophosphates, 17 samples: no residues detected						
Triazines, 1 sample: no residues detected						
GEORGIA						
Organochlorines,[2] 29 samples						
Benzene hexachloride	1	3.4	<0.01	—	0.02	
Chlordane	1	3.4	<0.01	—	0.01	
o,p'-DDE	1	3.4	<0.01	—	0.01	
p,p'-DDE	20	69.0	0.11	0.031	0.01	1.30

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TABLE 5 (Cont'd.). *Compound concentrations in cropland soils, by state, 1972—National Soils Monitoring Program*

Compound	Positive Detections		Residues, ppm dry weight			
			Arithmetic Mean Concentration	Estimated Geometric Mean[1]	Extremes of Detected Values	
	No.	%			Min.	Max.
o,p'-DDT	6	20.7	0.08	0.008	0.04	1.71
p,p'-DDT	20	69.0	0.33	0.043	0.01	6.11
p,p'-TDE	2	6.9	0.01	0.002	0.03	0.15
ΣDDT	22	75.9	0.52	0.072	0.01	9.12
Dieldrin	4	13.8	<0.01	0.001	0.01	0.02
Endrin	1	3.4	<0.01	—	0.01	
Toxaphene	8	27.6	2.22	0.036	0.65	46.58
Trifluralin	2	6.9	<0.01	0.001	0.01	0.09
Organophosphates, 28 samples						
Phorate	4	14.3	<0.01	0.002	0.02	0.04
IDAHO						
Organochlorines, 29 samples						
Chlordane	1	3.4	0.01	—	0.20	
p,p'-DDE	11	37.9	0.02	0.008	0.01	0.13
o,p'-DDT	4	13.8	0.01	0.003	0.02	0.29
p,p'-DDT	10	34.5	0.05	0.009	0.01	0.96
p,p'-TDE	2	6.9	<0.01	0.001	0.01	0.04
ΣDDT	12	41.4	0.09	0.015	0.01	1.38
Dieldrin	11	37.9	0.01	0.006	0.01	0.04
Heptachlor epoxide	1	3.4	<0.01	—	0.04	
Hexachlorobenzene	1	3.4	<0.01	—	0.01	
Organophosphates, 25 samples: no residues detected						
ILLINOIS						
Organochlorines,[2] 139 samples						
Aldrin	51	36.7	0.14	0.009	0.01	12.69
Chlordane	38	27.3	0.22	0.020	0.04	3.97
o,p'-DDE	1	0.7	<0.01	—	0.03	
p,p'-DDE	10	7.2	<0.01	0.001	0.02	0.06
p,p'-DDT	10	7.2	<0.01	0.001	0.03	0.11
ΣDDT	12	8.6	0.01	0.002	0.02	0.16
Dieldrin	93	66.9	0.16	0.051	0.01	6.18
Endrin ketone	1	0.7	<0.01	—	0.02	
Heptachlor	31	22.3	0.01	0.004	0.01	0.60
Heptachlor epoxide	37	26.6	0.02	0.007	0.01	0.26
Trifluralin	9	6.5	0.01	0.001	0.02	0.27
Organophosphates, 87 samples						
Diazinon	2	2.3	<0.01	<0.001	0.15	0.17
Phorate	1	1.2	<0.01	—	0.40	
Triazines, 18 samples						
Atrazine	17	94.4	0.11	0.074	0.01	0.33
INDIANA						
Organochlorines,[2] 78 samples						
Aldrin	13	16.7	0.02	0.004	0.01	0.40
Chlordane	5	6.4	0.08	0.003	0.26	3.95
p,p'-DDE	1	1.3	<0.01	—	0.05	
p,p'-TDE	2	2.6	<0.01	<0.001	0.01	0.06
ΣDDT	2	2.6	<0.01	<0.001	0.01	0.11
Dieldrin	22	28.2	0.05	0.010	0.01	1.11
Heptachlor	4	5.1	<0.01	0.001	0.03	0.21
Heptachlor epoxide	4	5.1	<0.01	0.001	0.04	0.12
Trifluralin	4	5.1	0.01	0.002	0.06	0.60
Organophosphates, 59 samples: no residues detected						
Triazines, 4 samples						
Atrazine	4	100.0	0.08	0.075	0.03	0.11
IOWA						
Organochlorines,[2] 150 samples						
Aldrin	28	18.7	0.04	0.004	0.01	2.07
Chlordane	29	19.3	0.10	0.009	0.02	3.44
p,p'-DDE	16	10.7	0.01	0.002	0.01	0.33
o,p'-DDT	6	4.0	<0.01	0.001	0.01	0.24
p,p'-DDT	14	9.3	0.02	0.003	0.01	0.93
p,p'-TDE	2	1.3	<0.01	<0.001	0.01	0.01
ΣDDT	17	11.3	0.03	0.003	0.02	1.50
Dieldrin	85	56.7	0.09	0.029	0.01	1.62
Heptachlor	13	8.7	0.01	0.001	0.01	0.44
Heptachlor epoxide	27	18.0	0.01	0.003	0.01	0.20
PCBs	1	0.7	0.01	—	0.80	
Propachlor	1	0.7	<0.01	—	0.10	
Trifluralin	22	14.7	0.01	0.004	0.01	0.18

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TABLE 5 (Cont'd.). *Compound concentrations in cropland soils, by state, 1972—National Soils Monitoring Program*

Compound	Positive Detections		Residues, ppm dry weight			
			Arithmetic Mean Concentration	Estimated Geometric Mean[1]	Extremes of Detected Values	
	No.	%			Min.	Max.
Organophosphates, 113 samples						
Diazinon	1	0.9	<0.01	—	0.07	
Triazines, 34 samples						
Atrazine	32	94.1	0.21	0.114	0.01	0.77
KENTUCKY						
Organochlorines, 28 samples						
Chlordane	2	7.1	0.01	0.002	0.14	0.18
p,p'-DDE	3	10.7	<0.01	0.001	0.01	0.03
p,p'-DDT	3	10.7	<0.01	0.002	0.02	0.05
p,p'-TDE	2	7.1	<0.01	0.001	0.01	0.01
Σ DDT	4	14.3	0.01	0.002	0.01	0.08
Dieldrin	7	25.0	0.01	0.004	0.01	0.12
Heptachlor epoxide	2	7.1	<0.01	0.001	0.01	0.02
Organophosphates, 15 samples: no residues detected						
Triazines, 3 samples						
Atrazine	2	66.7	0.01	0.010	0.01	0.03
LOUISIANA						
Organochlorines,[2] 27 samples						
Aldrin	2	7.4	<0.01	0.001	0.01	0.05
p,p'-DDE	14	51.9	0.43	0.046	0.01	6.21
o,p'-DDT	10	37.0	0.41	0.030	0.01	5.62
p,p'-DDT	14	51.9	1.26	0.072	0.01	15.86
Σ DDT	14	51.9	2.09	0.100	0.03	27.69
Dieldrin	11	40.7	0.03	0.012	0.01	0.27
Endrin	1	3.7	0.02	—	0.48	
Toxaphene	8	29.6	3.51	0.065	2.08	29.99
Trifluralin	2	7.4	0.01	0.002	0.11	0.12
Organophosphates, 26 samples						
DEF	1	3.9	<0.01	—	0.08	
Phorate	2	7.7	<0.01	0.001	0.04	0.04
MICHIGAN						
Organochlorines,[2] 53 samples						
Aldrin	2	3.8	0.25	0.002	0.04	13.28
Chlordane	1	1.9	0.02	—	1.24	
DCPA	1	1.9	<0.01	—	0.18	
p,p'-DDE	6	11.3	0.24	0.005	0.02	7.16
o,p'-DDT	4	7.6	0.13	0.004	0.09	3.36
p,p'-DDT	5	9.4	0.67	0.006	0.05	18.93
p,p'-TDE	1	1.9	<0.01	—	0.10	
Σ DDT	6	11.3	1.04	0.007	0.02	29.45
Dieldrin	4	7.6	0.06	0.003	0.09	2.26
Hexachlorobenzene	1	1.9	<0.01	—	0.07	
Trifluralin	1	1.9	0.01	—	0.31	
Organophosphates, 44 samples: no residues detected						
Triazines, 14 samples						
Atrazine	14	100.0	0.09	0.062	0.01	0.41
MID-ATLANTIC[3]						
Organochlorines, 14 samples						
Chlordane	2	14.3	0.07	0.007	0.16	0.83
o,p'-DDE	1	7.1	0.01	—	0.09	
p,p'-DDE	4	28.6	0.15	0.013	0.04	1.83
o,p'-DDT	3	21.4	0.02	0.006	0.04	0.23
p,p'-DDT	4	28.6	0.12	0.014	0.02	1.17
p,p'-TDE	1	7.1	<0.01	—	0.07	
Σ DDT	4	28.6	0.30	0.020	0.06	3.32
Dieldrin	4	28.6	0.03	0.008	0.04	0.26
Endrin	1	7.1	0.02	—	0.25	
Heptachlor	1	7.1	<0.01	—	0.01	
Heptachlor epoxide	1	7.1	<0.01	—	0.07	
Organophosphates, 14 samples: no residues detected						
Triazines, 1 sample: no residues detected						
MISSISSIPPI						
Organochlorines,[2] 30 samples						
p,p'-DDE	24	80.0	0.23	0.087	0.02	1.54
o,p'-DDT	18	60.0	0.27	0.057	0.01	1.79
p,p'-DDT	24	80.0	1.12	0.239	0.02	8.76
p,p'-TDE	3	10.0	0.04	0.003	0.01	1.25

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TABLE 5 (Cont'd.). *Compound concentrations in cropland soils, by state, 1972—National Soils Monitoring Program*

Compound	Positive Detections		Residues, ppm dry weight			
			Arithmetic Mean Concentration	Estimated Geometric Mean[1]	Extremes of Detected Values	
	No.	%			Min.	Max.
ΣDDT	24	80.0	1.66	0.337	0.05	12.33
Dieldrin	3	10.0	<0.01	0.001	0.01	0.05
Toxaphene	16	53.3	2.21	0.185	0.49	12.77
Trifluralin	1	3.3	<0.01	—	0.05	
Organophosphates, 25 samples						
DEF	2	8.0	0.03	0.003	0.06	0.67
Phorate	1	4.0	<0.01	—	0.03	
MISSOURI						
Organochlorines,[2] 82 samples						
Aldrin	14	17.1	0.05	0.006	0.01	1.55
Chlordane	3	3.7	0.01	0.001	0.26	0.62
p,p'-DDE	6	7.3	<0.01	0.001	0.01	0.10
o,p'-DDT	3	3.7	<0.01	0.001	0.01	0.12
p,p'-DDT	6	7.3	0.01	0.002	0.06	0.51
ΣDDT	6	7.3	0.02	0.002	0.08	0.73
Dieldrin	26	31.7	0.05	0.012	0.01	0.60
Heptachlor	2	2.4	<0.01	0.001	0.05	0.07
Heptachlor epoxide	3	3.7	<0.01	0.001	0.01	0.06
Toxaphene	2	2.4	0.04	0.001	1.01	1.99
Trifluralin	6	7.3	0.02	0.003	0.04	0.68
Organophosphates, 66 samples: no residues detected						
Triazines, 13 samples						
Atrazine	13	100.0	0.07	0.055	0.01	0.17
NEBRASKA						
Organochlorines,[2] 101 samples						
Aldrin	2	2.0	<0.01	<0.001	0.01	0.06
Chlordane	8	7.9	0.01	0.002	0.01	0.19
p,p'-DDE	7	6.9	<0.01	0.001	0.01	0.36
o,p'-DDT	1	1.0	<0.01	—	0.12	
p,p'-DDT	6	5.9	0.01	0.001	0.01	0.50
ΣDDT	7	6.9	0.01	0.002	0.02	0.98
Dieldrin	34	33.7	0.03	0.009	0.01	0.29
Endrin	1	1.0	<0.01	—	0.01	
Heptachlor epoxide	5	5.0	<0.01	0.001	0.01	0.07
Organophosphates, 86 samples: no residues detected						
Triazines, 19 samples						
Atrazine	17	89.5	0.06	0.035	0.01	0.31
NEW ENGLAND[3]						
Organochlorines, 20 samples						
Chlordane	2	10.0	0.03	0.004	0.30	0.31
p,p'-DDE	6	30.0	0.24	0.012	0.02	4.34
o,p'-DDT	3	15.0	0.01	0.004	0.02	0.18
p,p'-DDT	5	25.0	0.20	0.015	0.04	2.49
p,p'-TDE	3	15.0	0.44	0.009	0.09	8.20
ΣDDT	6	30.0	0.90	0.022	0.03	15.03
Dieldrin	2	10.0	0.24	0.006	0.19	4.64
Endosulfan I	1	5.0	<0.01	—	0.08	
Endosulfan II	1	5.0	0.01	—	0.25	
Endosulfan sulfate	1	5.0	0.02	—	0.31	
Heptachlor epoxide	2	10.0	<0.01	0.002	0.03	0.06
Organophosphates, 20 samples: no residues detected						
NEW YORK						
Organochlorines, 36 samples						
Chlordane	1	2.8	0.03	—	1.02	
o,p'-DDE	2	5.6	<0.01	0.001	0.03	0.04
p,p'-DDE	9	25.0	0.06	0.007	0.01	1.26
o,p'-DDT	7	19.4	0.02	0.005	0.02	0.44
p,p'-DDT	9	25.0	0.13	0.012	0.05	3.14
o,p'-TDE	1	2.8	0.01	—	0.31	
p,p'-TDE	2	5.6	0.02	0.002	0.18	0.52
ΣDDT	9	25.0	0.24	0.016	0.08	5.06
Dieldrin	6	16.7	0.01	0.004	0.01	0.21
Endrin	1	2.8	0.01	—	0.24	
Heptachlor	1	2.8	<0.01	—	0.01	
Organophosphates, 35 samples: no residues detected						
Triazines, 6 samples						
Atrazine	6	100.0	0.06	0.045	0.01	0.21

(Continued next page)

TABLE 5 (Cont'd.). *Compound concentrations in cropland soils, by state, 1972—National Soils Monitoring Program*

Compound	Positive Detections		Residues, ppm dry weight			
			Arithmetic Mean Concentration	Estimated Geometric Mean[1]	Extremes of Detected Values	
	No.	%			Min.	Max.
NORTH CAROLINA						
Organochlorines,[2] 31 samples						
Aldrin	1	3.2	<0.01	—	0.10	
Chlordane	2	6.4	0.05	0.002	0.02	1.39
p,p'-DDE	17	54.8	0.18	0.027	0.02	3.92
o,p'-DDT	13	41.9	0.04	0.013	0.01	0.26
p,p'-DDT	17	54.8	0.18	0.044	0.02	1.58
p,p'-TDE	4	12.9	0.01	0.003	0.02	0.28
ΣDDT	17	54.8	0.41	0.065	0.06	5.76
Dieldrin	10	32.3	0.03	0.007	0.01	0.32
Endrin	1	3.2	0.07	—	2.13	
Endrin ketone	1	3.2	0.01	—	0.38	
Heptachlor	1	3.2	<0.01	—	0.01	
Heptachlor epoxide	1	3.2	<0.01	—	0.03	
PCNB	1	3.2	0.03	—	0.98	
Toxaphene	4	12.9	0.47	0.010	1.07	11.03
Trifluralin	3	9.7	0.06	0.003	0.02	1.86
Organophosphates, 28 samples						
Parathion, ethyl	1	3.6	<0.01	—	0.12	
Phorate	3	10.7	<0.01	0.001	0.01	0.06
Triazines, 2 samples: no residues detected						
OHIO						
Organochlorines, 67 samples						
Aldrin	12	17.9	0.03	0.005	0.01	0.61
Chlordane	2	3.0	0.09	0.002	0.87	4.99
p,p'-DDE	2	3.0	<0.01	<0.001	0.02	0.04
p,p'-TDE	2	3.0	<0.01	0.001	0.05	0.06
ΣDDT	2	3.0	<0.01	0.001	0.08	0.09
Dieldrin	18	26.9	0.02	0.007	0.01	0.27
Heptachlor	2	3.0	0.01	0.001	0.16	0.29
Heptachlor epoxide	1	1.5	<0.01	—	0.07	
Organophosphates, 53 samples: no residues detected						
Triazines, 8 samples						
Atrazine	7	87.5	0.07	0.050	0.02	0.19
OKLAHOMA						
Organochlorines,[2] 64 samples						
p,p'-DDE	4	6.3	0.01	0.001	0.01	0.41
o,p'-DDT	2	3.1	<0.01	0.001	0.03	0.18
p,p'-DDT	3	4.7	0.01	0.001	0.05	0.30
ΣDDT	4	6.3	0.02	0.002	0.01	0.89
Dieldrin	1	1.6	<0.01	—	0.04	
Hexachlorobenzene	2	3.1	<0.01	0.001	0.03	0.12
PCNB	1	1.6	0.04	—	2.61	
Trifluralin	1	1.6	<0.01	—	0.08	
Organophosphates, 64 samples: no residues detected						
OREGON						
Organochlorines, 37 samples						
Chlordane	2	5.4	<0.01	0.001	0.02	0.05
p,p'-DDE	8	21.6	0.03	0.005	0.01	0.65
o,p'-DDT	3	8.1	0.01	0.002	0.03	0.35
p,p'-DDT	5	13.5	0.04	0.005	0.03	1.05
p,p'-TDE	1	2.7	<0.01	—	0.03	
ΣDDT	8	21.6	0.08	0.007	0.01	2.08
Dieldrin	4	10.8	0.01	0.003	0.03	0.19
Heptachlor epoxide	2	5.4	<0.01	<0.001	0.01	0.01
Toxaphene	2	5.4	0.03	0.003	0.55	0.64
Organophosphates, 33 samples: no residues detected						
PENNSYLVANIA						
Organochlorines,[2] 37 samples						
Chlordane	2	5.4	0.01	0.002	0.21	0.26
p,p'-DDE	7	18.9	0.04	0.006	0.01	0.72
o,p'-DDT	3	8.1	0.01	0.002	0.02	0.16
p,p'-DDT	6	16.2	0.03	0.005	0.02	0.61
ΣDDT	7	18.9	0.07	0.007	0.01	1.49
Dieldrin	6	16.2	0.01	0.004	0.03	0.23
Heptachlor epoxide	2	5.4	<0.01	0.001	0.01	0.05
Trifluralin	1	2.7	<0.01	—	0.13	
Organophosphates, 37 samples: no residues detected						
Triazines, 7 samples						
Atrazine	5	71.4	0.03	0.022	0.01	0.10

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TABLE 5 (Cont'd.). *Compound concentrations in cropland soils, by state, 1972—National Soils Monitoring Program*

Compound	Positive Detections		Residues, ppm dry weight			
	No. of	%	Arithmetic Mean Concentration	Estimated Geometric Mean[1]	Extremes of Detected Values	
					Min.	Max.
SOUTH CAROLINA						
Organochlorines,[2] 17 samples						
o,p'-DDE	1	5.9	<0.01	—	0.03	
p,p'-DDE	15	88.2	0.16	0.088	0.02	0.38
o,p'-DDT	12	70.6	0.08	0.032	0.01	0.39
p,p'-DDT	15	88.2	0.40	0.159	0.02	1.11
p,p'-TDE	4	23.5	0.01	0.004	0.01	0.14
ΣDDT	15	88.2	0.64	0.263	0.04	1.88
Dieldrin	2	11.8	<0.01	0.002	0.01	0.05
Toxaphene	6	35.3	1.17	0.062	0.82	6.16
Trifluralin	4	23.5	0.01	0.003	0.01	0.05
Organophosphates, 17 samples						
Phorate	1	5.9	<0.01	—	0.04	
SOUTH DAKOTA						
Organochlorines, 106 samples						
Aldrin	3	2.8	<0.01	<0.001	0.01	0.05
Chlordane	2	1.9	<0.01	0.001	0.15	0.31
p,p'-DDE	1	0.9	<0.01	—	0.13	
o,p'-DDT	1	0.9	<0.01	—	0.03	
p,p'-DDT	1	0.9	<0.01	—	0.33	
p,p'-TDE	1	0.9	<0.01	—	0.01	
ΣDDT	1	0.9	<0.01	—	0.50	
Dieldrin	11	10.4	0.01	0.002	0.01	0.21
Endrin	1	0.9	<0.01	—	0.04	
Heptachlor epoxide	2	1.9	<0.01	<0.001	0.01	0.03
Organophosphates, 90 samples						
Malathion	1	1.1	<0.01	—	0.08	
Parathion, ethyl	2	2.2	<0.01	0.001	0.06	0.10
Parathion, methyl	1	1.1	<0.01	—	0.01	
TENNESSEE						
Organochlorines,[2] 25 samples						
Chlordane	1	4.0	0.32	—	7.89	
p,p'-DDE	8	32.0	0.02	0.008	0.01	0.25
o,p'-DDT	3	12.0	0.01	0.003	0.06	0.20
p,p'-DDT	8	32.0	0.04	0.011	0.03	0.32
p,p'-TDE	1	4.0	0.01	—	0.17	
ΣDDT	9	36.0	0.08	0.017	0.02	0.51
Dieldrin	6	24.0	0.04	0.007	0.02	0.41
Heptachlor epoxide	1	4.0	0.03	—	0.72	
PCNB	1	4.0	0.01	—	0.22	
Toxaphene	1	4.0	0.13	—	3.37	
Trifluralin	2	8.0	<0.01	0.002	0.05	0.07
Organophosphates, 2 samples: no residues detected						
Triazines, 2 samples						
Atrazine	2	100.0	0.02	0.018	0.01	0.03
VIRGINIA/WEST VIRGINIA[3]						
Organochlorines, 25 samples						
Chlordane	1	4.0	0.03	—	0.70	
p,p'-DDE	3	12.0	0.01	0.002	0.01	0.09
o,p'-DDT	2	8.0	<0.01	0.001	0.01	0.02
p,p'-DDT	3	12.0	0.01	0.002	0.01	0.13
ΣDDT	4	16.0	0.02	0.003	0.01	0.23
Dieldrin	3	12.0	0.01	0.003	0.03	0.15
Heptachlor epoxide	2	8.0	0.01	0.001	0.01	0.12
Organophosphates, 25 samples: no residues detected						
Triazines, 3 samples: no residues detected						
WASHINGTON STATE						
Organochlorines,[2] 45 samples						
p,p'-DDE	2	4.4	<0.01	0.001	0.05	0.06
o,p'-DDT	1	2.2	<0.01	—	0.01	
p,p'-DDT	1	2.2	<0.01	—	0.04	
ΣDDT	2	4.4	<0.01	0.001	0.05	0.11
Dieldrin	2	4.4	<0.01	0.001	0.02	0.03
Hexachlorobenzene	6	13.3	<0.01	0.002	0.01	0.03
Trifluralin	1	2.2	<0.01	—	0.06	
Organophosphates, 43 samples: no residues detected						

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TABLE 5 (Cont'd.). *Compound concentrations in cropland soils, by state, 1972—National Soils Monitoring Program*

Compound	Positive Detections		Residues, ppm dry weight			
	No.	%	Arithmetic Mean Concentration	Estimated Geometric Mean[1]	Extremes of Deteced Values Min.	Max.
WISCONSIN						
Organochlorines, 67 samples						
Chlordane	3	4.5	0.04	0.002	0.69	1.19
p,p'-DDE	3	4.5	0.01	0.001	0.05	0.42
o,p'-DDT	1	1.5	<0.01	—	0.08	
p,p'-DDT	2	3.0	0.01	0.001	0.06	0.41
Σ DDT	3	4.5	0.02	0.001	0.05	0.91
Dieldrin	6	9.0	0.01	0.002	0.03	0.13
Heptachlor	2	3.0	<0.01	0.001	0.02	0.06
Heptachlor epoxide	3	4.5	<0.01	0.001	0.01	0.06
Organophosphates, 57 samples: no residues detected						
Triazines, 16 samples						
Atrazine	15	93.8	0.04	0.030	0.01	0.13

[1]Not calculated when fewer than two positive detections present.
[2]See footnote 1, Table 1.
[3]See footnote 3, Table 3.

TABLE 6. *Occurrence of pesticide concentrations in standing agricultural crops from 1,483 sampling sites, 1972 —National Soils Monitoring Program*

Crop Materials	Organochlorines			Organophosphates			Triazines		
	No. of Analyses	Positive Detections No.	%	No. of Analyses	Positive Detections No.	%	No. of Analyses	Positive Detections No.	%
Alfalfa/bur clover	43	25	58	39	3	7	—		
Asparagus	1	1	100	1	0	—	—		
Beans, dry	3	1	33	3	0	—	—		
Clover (*Trifolium*)	8	5	63	8	0	—	—		
Corn, sweet (kernels)	2	0	—	2	0	—	—		
Corn, field (kernels)	288	31	11	167	0	—	12	0	—
Corn stalks	283	132	47	247	6	2	16	0	—
Cotton stalks	40	39	98	40	32	80	—		
Cotton	2	0	—	2	0	—	—		
Cotton seed	38	31	82	32	13	41	—		
Grass hay	21	14	67	21	6	29	—		
Lespedeza	1	1	100	1	1	100	—		
Mixed hay	47	31	66	43	3	7	—		
Oat hay	1	0	—	1	0	—	—		
Pasture forage	10	5	50	9	1	11	—		
Peanut vines	2	2	100	2	0	—	—		
Soybean hay	1	1	100	—			—		
Sugar beet tops	1	0	—	—			—		
Silage (corn or sorghum)	3	1	33	2	1	50	—		
Milo	3	1	33	2	0	—	—		
Peanuts	9	6	67	3	0	—	—		
Peas	1	0	—	1	0	—	—		
Pecans	1	0	—	1	0	—	—		
Rye	1	1	100	1	0	—	—		
Sorghum (grain)	14	5	36	11	2	18	—		
Sorghum (stalks)	18	8	44	15	3	20	—		
Soybeans	199	73	37	66	0	—	—		
Sugarcane	2	0	—	2	0	—	—		
Sweet sorghum	2	2	100	—			—		
Tobacco	2	2	100	2	1	50	—		

TABLE 7. *Occurrence of organochlorine concentrations in selected, mature crops, from 1,483 sites by state or state group, 1972—National Soils Monitoring Program*

State	Field Corn, Kernels			Soybeans			Mixed Hay		
	No. of Analyses	Positive Detections		No. of Analyses	Positive Detections		No. of Analyses	Positive Detections	
		No.	%		No.	%		No.	%
Alabama	7	0	—	4	4	100	1	0	—
Arkansas	—			19	10	53	—		
California	—			—			—		
Florida	—			1	0		—		
Georgia	4	0	—	5	2	40	—		
Idaho	—			—			—		
Illinois	41	1	2	28	16	57	1	1	100
Indiana	24	4	17	14	6	43	—		
Iowa	71	1	1	41	17	41	1	1	100
Kentucky	6	0	—	3	0	—	1	1	100
Louisiana	1	0	—	7	3	43	—		
Michigan	21	1	5	4	0	—	10	10	100
Mid-Atlantic[1]	3	1	33	3	0	—	—		
Mississippi	—			15	1	7	—		
Missouri	10	0	—	15	6	40	1	1	100
Nebraska	27	2	7	3	0	—	1	1	100
New England[1]	—			—			8	1	13
New York	11	8	73	—			1	1	100
N. Carolina	6	0	—	9	1	11	—		
Ohio	18	3	17	9	0	—	11	7	64
Oklahoma	1	0	—	—			1	1	100
Oregon	—			—			—		
Pennsylvania	10	9	90	1	1	100	1	1	100
S. Carolina	—			7	1	14	1	1	100
S. Dakota	11	0	—	—			4	2	50
Tennessee	2	0	—	4	3	75	1	1	100
Virginia/W. Virginia[1]	1	1	100	4	0	—	3	1	33
Washington state	—			—			—		
Wisconsin	13	0	—	1	0	—	—		

[1] See footnote 3; Table 3.

TABLE 8. *Compound concentrations in standing agricultural crops, 1972—National Soils Monitoring Program*

Compound	Positive Detections		Residues, ppm dry weight			
			Arithmetic Mean Concentration	Estimated Geometric Mean[1]	Extremes of Detected Values	
	No.	%			Min.	Max.
ALFALFA/BUR CLOVER						
Organochlorines, 43 samples						
Chlordane	7	16.3	0.02	0.005	0.04	0.24
p,p'-DDE	12	27.9	0.01	0.003	0.01	0.05
o,p'-DDT	12	27.9	0.01	0.004	0.01	0.09
p,p'-DDT	15	34.9	0.02	0.009	0.02	0.23
ΣDDT	15	34.9	0.04	0.012	0.03	0.28
Dieldrin	16	37.2	0.01	0.007	0.01	0.09
Heptachlor epoxide	1	2.3	<0.01	—	0.01	
Toxaphene	2	4.6	0.01	0.002	0.17	0.19
Organophosphates, 39 samples						
DEF	1	2.6	<0.01	—	0.02	
Diazinon	1	2.6	<0.01	—	0.01	
Malathion	3	7.7	0.01	0.002	0.03	0.26
ASPARAGUS						
Organochlorines, 1 sample						
p,p'-DDE	1	100.0	0.11	—	0.11	
o,p'-DDT	1	100.0	0.03	—	0.03	
p,p'-DDT	1	100.0	0.33	—	0.33	
ΣDDT	1	100.0	0.47	—	0.47	
Organophosphates, 1 sample: no residues detected						
BEANS, DRY (all varieties)						
Organochlorines, 3 samples						
Dicofol	1	33.3	0.05	—	0.15	
Organophosphates, 3 samples: no residues detected						
CLOVER (*Trifolium* sp.)						
Organochlorines, 8 samples						
Chlordane	2	25.0	0.02	0.008	0.07	0.10
p,p'-DDE	3	37.5	0.02	0.009	0.01	0.08
o,p'-DDT	4	50.0	0.03	0.014	0.01	0.07
p,p'-DDT	4	50.0	0.05	0.022	0.03	0.14
ΣDDT	4	50.0	0.10	0.031	0.04	0.29
Dieldrin	5	62.5	0.03	0.018	0.02	0.11
Organophosphates, 8 samples: no residues detected						
CORN, SWEET (kernels)						
Organochlorines, 2 samples: no residues detected						
Organophosphates, 2 samples: no residues detected						
CORN STALKS						
Organochlorines, 283 samples						
Alachlor	1	0.3	<0.01	—	0.09	
Chlordane	17	6.0	0.01	0.002	0.02	0.41
p,p'-DDE	28	9.9	<0.01	0.001	0.01	0.16
o,p'-DDT	37	13.1	<0.01	0.002	0.01	0.25
p,p'-DDT	62	21.9	0.02	0.004	0.01	2.33
p,p'-TDE	2	0.7	<0.01	<0.001	0.01	0.01
ΣDDT	62	21.9	0.03	0.005	0.01	2.74
Dieldrin	99	35.0	0.01	0.005	0.01	0.29
Endrin	3	1.1	<0.01	<0.001	0.01	0.04
Heptachlor	1	0.3	<0.01	—	0.01	
Heptachlor epoxide	14	5.0	<0.01	<0.001	0.01	0.06
Hexachlorobenzene	1	0.3	<0.01	—	0.02	
Toxaphene	9	3.2	0.04	0.002	0.19	4.14
Organophosphates, 247 samples						
Diazinon	2	0.8	<0.01	<0.001	0.04	0.10
Malathion	3	1.2	<0.01	<0.001	0.06	0.25
Phorate	4	1.6	<0.01	<0.001	0.01	0.02
Triazines, 16 samples: no residues detected						
FIELD CORN (kernels)						
Organochlorines, 288 samples						
Chlordane	2	0.7	<0.01	<0.001	0.01	0.15
o,p'-DDT	1	0.3	<0.01	—	0.03	
p,p'-DDT	2	0.7	<0.01	<0.001	0.03	0.07
ΣDDT	2	0.7	<0.01	<0.001	0.03	0.10
Dieldrin	8	2.8	<0.01	0.001	0.01	0.21

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TABLE 8 (Cont'd.). *Compound concentrations in standing agricultural crops, 1972—National Soils Monitoring Program*

Compound	Positive Detections No.	%	Residues, ppm dry weight: Arithmetic Mean Concentration	Estimated Geometric Mean[1]	Extremes of Detected Values Min.	Max.
Endrin	2	0.7	<0.01	<0.001	0.01	0.02
Heptachlor	1	0.3	<0.01	—	0.02	
Heptachlor epoxide	23	8.0	<0.01	0.001	0.01	0.02
PCNB	1	0.3	<0.01	—	0.01	
Organophosphates, 167 samples: no residues detected						
Triazines, 12 samples: no residues detected						
COTTON STALKS						
Organochlorines,[2] 40 samples						
Chlordane	6	15.0	0.05	0.006	0.15	1.00
o,p'-DDE	1	2.5	<0.01	—	0.13	
p,p'-DDE	29	72.5	0.67	0.089	0.01	8.89
o,p'-DDT	28	70.0	0.79	0.116	0.01	13.40
p,p'-DDT	38	95.0	7.36	0.739	0.02	102.00
p,p'-TDE	12	30.0	0.04	0.010	0.01	0.41
ΣDDT	38	95.0	8.87	0.913	0.02	115.79
Dieldrin	14	35.0	0.02	0.008	0.01	0.19
Endosulfan sulfate	1	2.5	0.07	—	2.70	
Endrin	2	5.0	0.01	0.002	0.15	0.15
Heptachlor epoxide	3	7.5	<0.01	0.001	0.01	0.02
Toxaphene	28	70.0	25.44	1.078	0.66	462.30
Trifluralin	1	2.5	<0.01	—	0.02	
Organophosphates, 40 samples						
Carbophenothion	1	2.5	<0.01	—	0.08	
DEF	25	62.5	1.20	0.069	0.01	24.19
Diazinon	1	2.5	<0.01	—	0.02	
Malathion	4	10.0	0.03	0.003	0.01	0.94
Parathion, ethyl	5	12.5	0.01	0.003	0.01	0.12
Parathion, methyl	16	40.0	0.15	0.026	0.02	1.39
Phorate	1	2.5	<0.01	—	0.01	
COTTON SEED						
Organochlorines, 38 samples						
p,p'-DDE	16	42.1	0.01	0.008	0.01	0.12
o,p'-DDT	15	39.5	0.03	0.012	0.02	0.19
p,p'-DDT	31	81.6	0.22	0.082	0.01	1.40
p,p'-TDE	2	5.3	<0.01	0.001	0.04	0.14
ΣDDT	31	81.6	0.27	0.091	0.01	1.79
Dieldrin	2	5.3	<0.01	0.001	0.01	0.03
Toxaphene	20	52.6	0.49	0.082	0.20	3.71
Organophosphates, 32 samples						
DEF	13	40.6	0.09	0.016	0.02	0.71
Parathion, methyl	2	6.3	<0.01	0.001	0.04	0.05
COTTON						
Organochlorines, 2 samples: no residues detected						
Organophosphates, 2 samples: no residues detected						
SILAGE						
Organochlorines, 3 samples						
Chlordane	1	33.3	0.05	—	0.16	
p,p'-DDT	1	33.3	<0.01	—	0.01	
ΣDDT	1	33.3	<0.01	—	0.01	
Organophosphates, 2 samples						
Diazinon	1	50.0	0.05	—	0.11	
Malathion	1	50.0	1.32	—	2.64	
GRASS HAY						
Organochlorines, 21 samples						
Chlordane	2	9.5	0.01	0.003	0.09	0.09
o,p'-DDT	12	57.1	0.02	0.014	0.01	0.08
p,p'-DDT	13	61.9	0.07	0.033	0.01	0.23
p,p'-DDE	9	42.9	0.02	0.009	0.01	0.08
ΣDDT	13	61.9	0.11	0.044	0.01	0.30
Dieldrin	7	33.3	0.01	0.006	0.01	0.11
Toxaphene	6	28.6	0.15	0.020	0.30	1.19
Organophosphates, 21 samples						
DEF	1	4.8	0.01	—	0.12	
Diazinon	6	28.6	0.04	0.011	0.02	0.34
Malathion	5	23.8	0.03	0.007	0.02	0.22

(Continued next page)

TABLE 8 (Cont'd.). *Compound concentrations in standing agricultural crops, 1972—National Soils Monitoring Program*

Compound	Positive Detections		Residues, ppm dry weight			
	No.	%	Arithmetic Mean Concentration	Estimated Geometric Mean[1]	Extremes of Detected Values Min.	Max.
LESPEDEZA SERICEA						
Organochlorines, 1 sample						
p,p'-DDT	1	100.0	0.15	—	0.15	
ΣDDT	1	100.0	0.15	—	0.15	
Dieldrin	1	100.0	0.03	—	0.03	
Endrin	1	100.0	0.02	—	0.02	
Toxaphene	1	100.0	0.48	—	0.48	
Organophosphates, 1 sample						
DEF	1	100.0	0.15	—	0.15	
MILO						
Organochlorines, 3 samples						
p,p'-DDT	1	33.3	0.02	—	0.06	
p,p'-DDE	1	33.3	<0.01	—	0.01	
ΣDDT	1	33.3	0.02	—	0.07	
Toxaphene	1	33.3	0.04	—	0.13	
Organophosphates, 2 samples: no residues detected						
PASTURE FORAGE						
Organochlorines, 10 samples						
Chlordane	1	10.0	0.05	—	0.48	
o,p'-DDT	3	30.0	0.01	0.006	0.02	0.07
p,p'-DDT	4	40.0	0.08	0.021	0.08	0.40
p,p'-DDE	3	30.0	0.01	0.004	0.01	0.03
ΣDDT	4	40.0	0.10	0.026	0.17	0.40
Dieldrin	4	40.0	0.01	0.007	0.01	0.04
Toxaphene	2	20.0	0.15	0.014	0.59	0.86
Organophosphates, 9 samples						
Diazinon	1	11.1	<0.01	—	0.01	
MIXED HAY						
Organochlorines, 47 samples						
Chlordane	10	21.3	0.03	0.008	0.05	0.44
o,p'-DDE	1	2.1	<0.01	—	0.04	
p,p'-DDE	21	44.7	0.02	0.008	0.01	0.13
o,p'-DDT	23	48.9	0.02	0.009	0.01	0.12
p,p'-DDT	26	55.3	0.04	0.019	0.01	0.44
p,p'-TDE	1	2.1	<0.01	—	0.02	
ΣDDT	26	55.3	0.08	0.027	0.02	0.69
Dieldrin	22	46.8	0.02	0.012	0.01	0.11
Endrin	2	4.3	<0.01	<0.001	0.01	0.01
Heptachlor epoxide	1	2.1	<0.01	—	0.05	
Organophosphates, 43 samples						
Diazinon	2	4.6	<0.01	<0.001	0.01	0.02
Malathion	3	7.0	<0.01	0.001	0.02	0.09
Parathion, methyl	1	2.3	<0.01	—	0.01	
Phorate	1	2.3	<0.01	—	0.02	
PEANUTS						
Organochlorines, 9 samples						
p,p'-DDE	2	22.2	<0.01	0.003	0.02	0.02
ΣDDT	2	22.2	<0.01	0.003	0.02	0.02
Toxaphene	6	66.7	0.25	0.100	0.17	0.65
Organophosphates, 3 samples: no residues detected						
PEANUT VINES						
Organochlorines, 2 samples						
p,p'-DDT	1	50.0	0.42	—	0.85	
ΣDDT	1	50.0	0.42	—	0.85	
Dieldrin	2	100.0	0.21	0.102	0.02	0.41
Toxaphene	1	50.0	106.82	—	213.65	
Organophosphates, 2 samples: no residues detected						
PEAS (all varieties)						
Organochlorines, 1 sample: no residues detected						
Organophosphates, 1 sample: no residues detected						
PECANS						
Organochlorines, 1 sample: no residues detected						
Organophosphates, 1 sample: no residues detected						

(Continued next page)

TABLE 8 (Cont'd.). *Compound concentrations in standing agricultural crops, 1972—National Soils Monitoring Program*

Compound	Positive Detections No.	Positive Detections %	Residues, ppm dry weight: Arithmetic Mean Concentration	Residues, ppm dry weight: Estimated Geometric Mean[1]	Residues, ppm dry weight: Extremes of Detected Values Min.	Residues, ppm dry weight: Extremes of Detected Values Max.
RYE						
Organochlorines, 1 sample						
Chlordane	1	100.0	0.08	—	0.08	
Dieldrin	1	100.0	0.02	—	0.02	
Organophosphates, 1 sample: no residues detected						
SORGHUM (grain)						
Organochlorines, 14 samples						
o,p'-DDT	1	7.1	<0.01	—	0.02	
p,p'-DDT	5	35.7	0.01	0.006	0.01	0.07
p,p'-DDE	2	14.3	<0.01	0.001	0.01	0.02
ΣDDT	5	35.7	0.01	0.006	0.01	0.10
Dieldrin	2	14.3	<0.01	0.001	0.01	0.01
Organophosphates, 11 samples						
Malathion	2	18.2	0.03	0.006	0.04	0.29
Parathion, ethyl	1	9.1	<0.01	—	0.03	
Parathion, methyl	1	9.1	<0.01	—	0.01	
Phorate	1	9.1	<0.01	—	0.01	
SORGHUM STALKS						
Organochlorines, 18 samples						
Chlordane	1	5.6	0.01	—	0.15	
o,p'-DDT	5	27.8	0.01	0.004	0.01	0.03
p,p'-DDT	8	44.4	0.02	0.012	0.03	0.11
p,p'-DDE	6	33.3	0.01	0.004	0.01	0.04
p,p'-TDE	1	5.6	<0.01	—	0.07	
ΣDDT	8	44.4	0.04	0.017	0.04	0.20
Dieldrin	5	27.8	0.01	0.004	0.01	0.05
Toxaphene	1	5.6	0.01	—	0.25	
Organophosphates, 15 samples						
Malathion	3	20.0	0.02	0.006	0.06	0.13
Parathion, ethyl	1	6.7	<0.01	—	0.02	
SUGAR BEET TOPS						
Organochlorines, 1 sample: no residues detected						
SOYBEANS						
Organochlorines, 199 samples						
Chlordane	1	0.5	<0.01	—	0.07	
o,p'-DDT	1	0.5	<0.01	—	0.02	
p,p'-DDT	12	6.0	<0.01	0.001	0.01	0.07
p,p'-TDE	1	0.5	<0.01	—	0.18	
ΣDDT	13	6.5	<0.01	0.001	0.01	0.18
Dieldrin	47	23.6	<0.01	0.002	0.01	0.04
Endrin	16	8.0	<0.01	0.001	0.01	0.21
Heptachlor	1	0.5	<0.01	—	0.01	
Heptachlor epoxide	8	4.0	<0.01	<0.001	0.01	0.03
Toxaphene	12	6.0	0.01	0.002	0.14	0.38
Organophosphates, 66 samples: no residues detected						
SOYBEAN HAY						
Organochlorines, 1 sample						
o,p'-DDT	1	100.0	0.32	—	0.32	
p,p'-DDT	1	100.0	1.43	—	1.43	
p,p'-DDE	1	100.0	0.18	—	0.18	
ΣDDT	1	100.0	1.93	—	1.93	
Dieldrin	1	100.0	0.05	—	0.05	
Toxaphene	1	100.0	3.52	—	3.52	
SUGARCANE						
Organochlorines, 2 samples: no residues detected						
Organophosphates, 2 samples: no residues detected						
SWEET SORGHUM (grain)						
Organochlorines, 2 samples						
p,p'-DDT	1	50.0	0.07	—	0.14	
ΣDDT	1	50.0	0.07	—	0.14	
Dieldrin	1	50.0	<0.01	—	0.01	
Toxaphene	1	50.0	0.18	—	0.37	

(Continued next page)

TABLE 8 (Cont'd.). *Compound concentrations in standing agricultural crops, 1972—National Soils Monitoring Program*

Compound	Positive Detections		Residues, ppm dry weight			
	No.	%	Arithmetic Mean Concentration	Estimated Geometric Mean[1]	Extremes of Detected Values Min.	Max.
TOBACCO						
Organochlorines, 2 samples						
o,p'-DDT	1	50.0	0.03	—	0.07	
p,p'-DDT	2	100.0	0.48	0.385	0.19	0.77
p,p'-DDE	1	50.0	0.02	—	0.05	
ΣDDT	2	100.0	0.54	0.490	0.31	0.77
Dieldrin	2	100.0	0.04	0.043	0.03	0.06
Endrin	1	50.0	0.01	—	0.02	
Toxaphene	2	100.0	2.62	2.520	1.89	3.36
Organophosphates, 1 sample						
Diazinon	1	100.0	0.01	—	0.01	

[1] Not calculated when fewer than two positive detections present.
[2] Although trifluralin is a dinitroaniline compound, it is detected by the organochlorine methodology and thus appears with organochlorines in Tables 2–7.

detections was in the 0.01–0.25-ppm category, except for toxaphene, which was in the 1.01–5.00-ppm category.

By State—Pesticide concentrations in soils, by states or state groups, are presented in Table 5. Because some small eastern states had very few sites, those with similar geographic locations and/or agricultural characteristics were combined to obtain more representative data. State groups used were Mid-Atlantic: Delaware, Maryland, and New Jersey; New England: Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, and Vermont; and Virginia and West Virginia.

Comparisons of the percent occurrence of aldrin, dieldrin, heptachlor epoxide, ΣDDT, and chlordane are presented in Figures 2–6. The key for each figure is based on the arithmetic mean percent occurrence ($\bar{x}$) of the compound for all sites. The four classes are: greater than $2\bar{x}$, greater than $\bar{x}$ but less than $2\bar{x}$, greater than $\frac{1}{2}\bar{x}$ but less than $\bar{x}$, and less than $\frac{1}{2}\bar{x}$.

Illinois sites had the highest percent occurrence of aldrin, dieldrin, chlordane, and heptachlor epoxide (Figures 2–6). These compounds are soil insecticides, or their degradation products, used in corn production. The 1972 results generally correspond with results of the previous years for this Program (*2, 4, 7*). ΣDDT detections were concentrated in the southeastern states and California (Fig. 5). Oklahoma, Pennsylvania, South Dakota, and Wisconsin were generally below the all-sites average detection frequency for the compounds.

The detection of ronnel in soil from one site in Alabama (Table 5) was unusual. Ronnel is used to control flies, ticks, and gnats on domestic animals and in animal quarters. A thorough examination of the cropping and pesticide application record for that site revealed no pesticide applications during the growing season. However, the site was being used as a cattle pasture, and the chemical was probably transferred to the soil by treated cattle.

COMPOUND CONCENTRATIONS IN CROPS

Mature crop samples were collected from 737 sites, or 48 percent of the scheduled 1,533 sites. All crop samples were analyzed for organochlorines, including trifluralin. In addition, samples were analyzed for organophosphates and atrazine when pesticide application records indicated their use. Thus the organophosphate and atrazine concentration data samples are biased, and yield higher occurrence frequencies than might otherwise occur if all samples had been analyzed.

Table 6 lists the occurrence of pesticide residues in crop materials sampled. For all crops, 40 percent of the 1,045 samples analyzed contained detectable concentrations of organochlorines and 10 percent contained detectable amounts of organophosphates. Atrazine was not detected. In general, crops with known patterns of heavy pesticide application, or animal feed crops such as alfalfa, hay, field corn, or soybeans grown in rotation with these crops, had the highest detection frequencies.

Table 7 presents the occurrence of organochlorines in field corn kernels, soybeans, and mixed hay for each state or state group sampled. Residue detections varied most in field corn. Not enough samples were available to draw broad conclusions about mixed hay.

Table 8 presents the compound concentrations detected in each crop sampled. ΣDDT occurred most frequently in all crops except corn stalks, where dieldrin residues were predominant. The high frequency of occurrence of ΣDDT is probably the result of its prior, widespread use.

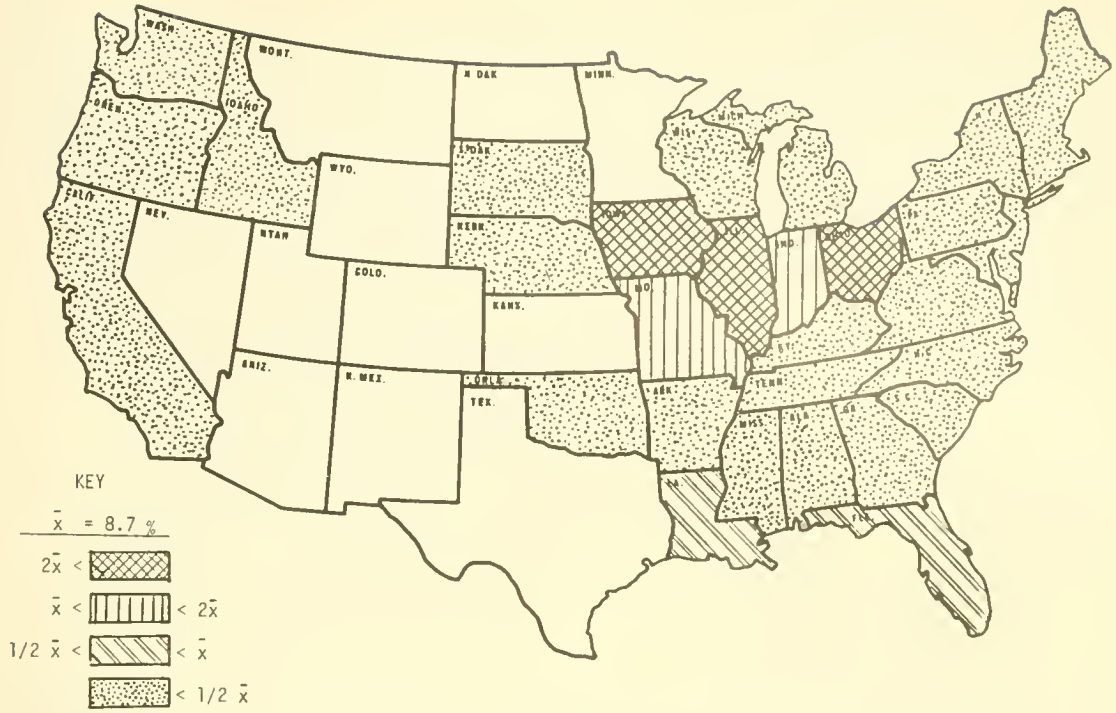

FIGURE 2. *Percent occurrence of aldrin residue detections in cropland soil of 37 states, by state, 1972 —National Soils Monitoring Program*

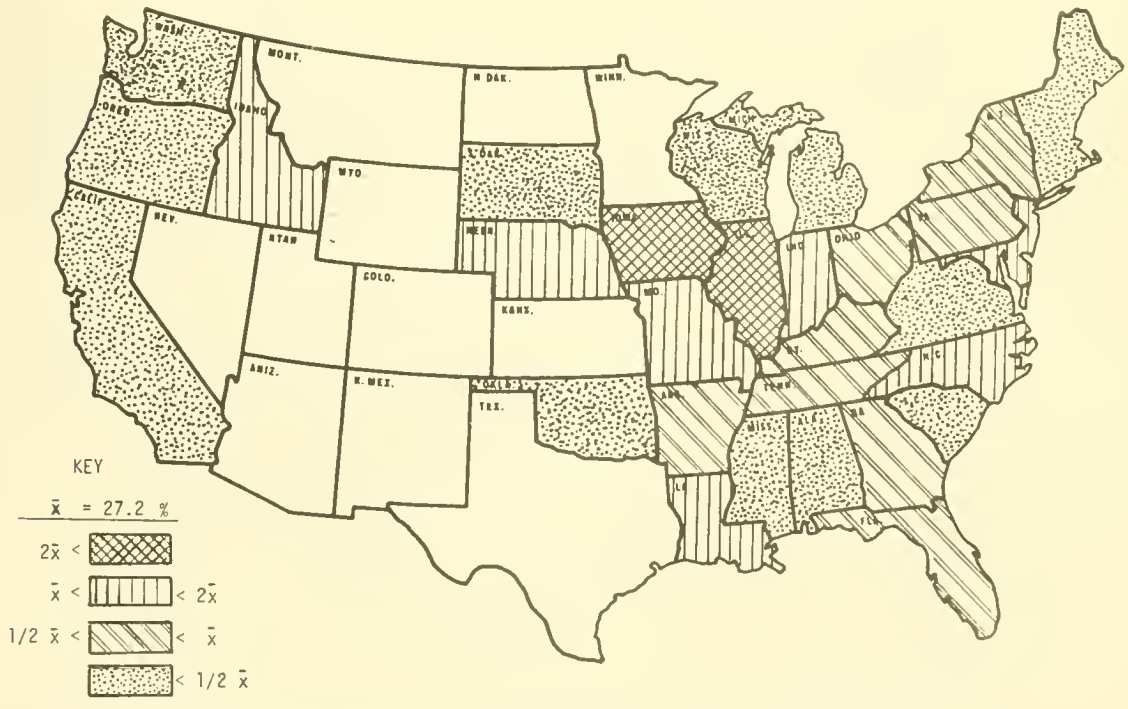

FIGURE 3. *Percent occurrence of dieldrin residue detections in cropland soil of 37 states, by state, 1972 —National Soils Monitoring Program*

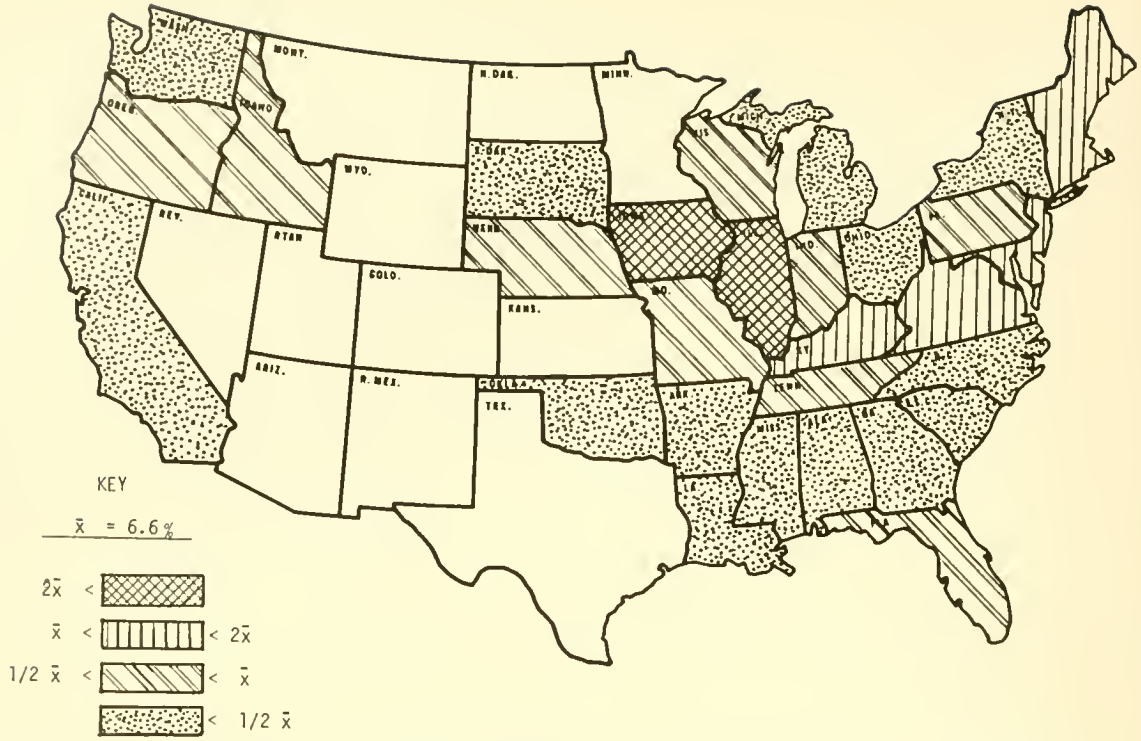

FIGURE 4. *Percent occurrence of heptachlor epoxide residue detections in cropland soil of 37 states, by state, 1972 —National Soils Monitoring Program*

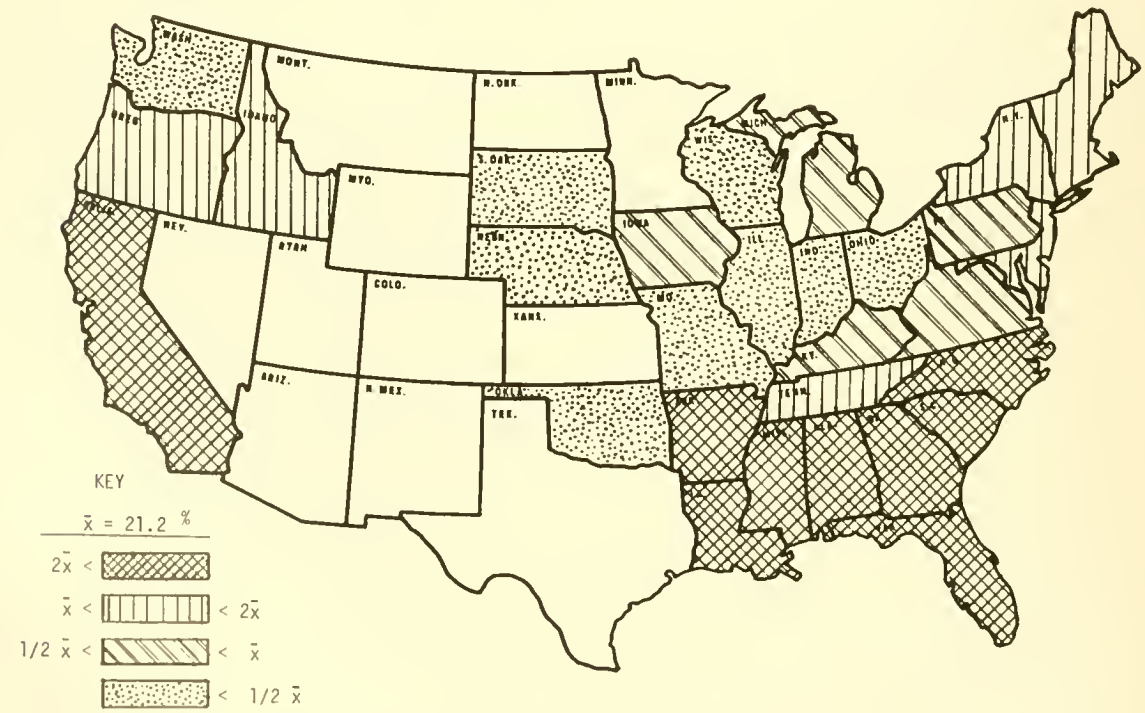

FIGURE 5. *Percent occurrence of ΣDDT residue detections in cropland soil of 37 states, by state, 1972 —National Soils Monitoring Program*

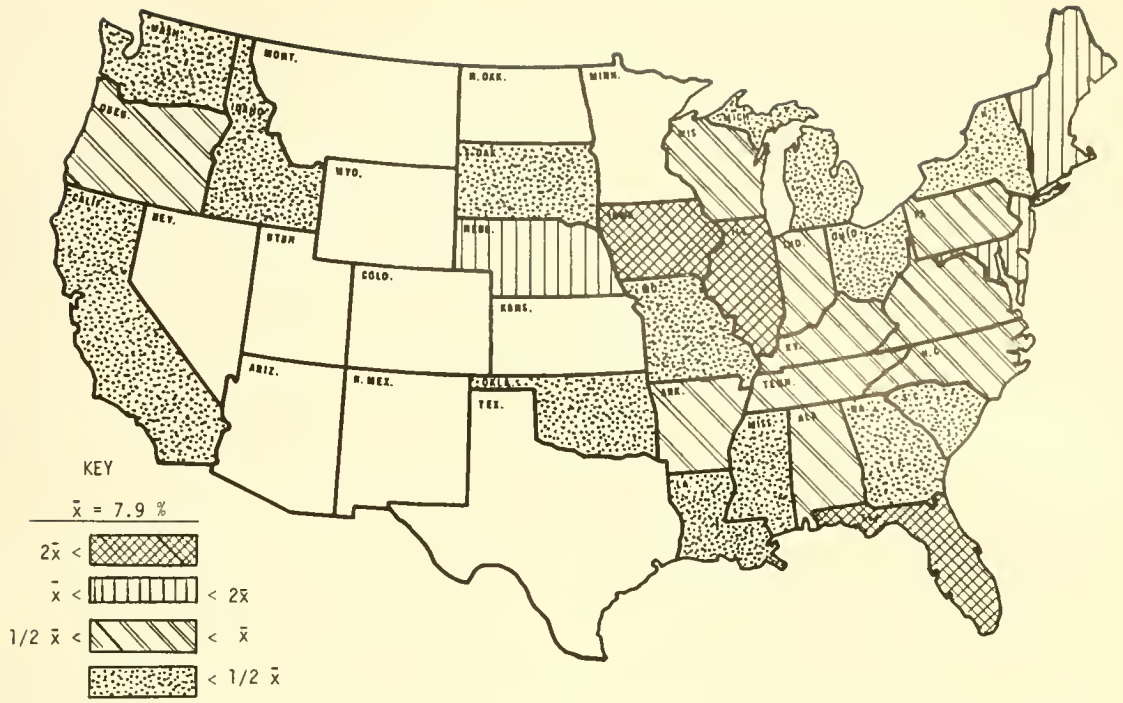

FIGURE 6. *Percent occurrence of chlordane residue detections in cropland soil of 37 states, by state, 1972 —National Soils Monitoring Program*

Acknowledgments

It is not possible to list by name all persons who contributed to this study. The authors are especially grateful to the staff of the Pesticides Monitoring Laboratory, Bay St. Louis, Mississippi, who received, processed, and analyzed samples for compound residues, and to the inspectors of the Animal and Plant Health Inspection Service, U.S. Department of Agriculture, who collected the samples.

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Organochlorine Pesticide Residues in Soils from Six U.S. Air Force Bases, 1975–76

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ABSTRACT

Soil samples collected during 1975 and 1976 from United States Air Force installations in California, Georgia, Ohio, Oklahoma, Texas, and Utah were analyzed for organochlorine pesticide residues. ΣDDT, chlordane, and dieldrin were the pesticides most commonly found. In 1975, ΣDDT residues were significantly higher in samples from residential areas than in samples from golf courses or areas free of pesticide application. Chlordane residues in 1975 were significantly higher in both residential and golf course areas than in areas where pesticides had not been used. No significant differences were found in 1976 in residue levels of any pesticide monitored among various land use areas.

Introduction

In 1975, the United States Air Force Occupational and Environmental Health Laboratory at Kelly Air Force Base, Texas, initiated a two-year pilot pesticides monitoring program to gather preliminary data on organochlorine residues in soils and sediments from Air Force bases and to determine the feasibility of developing a full-scale Air Force pesticides monitoring program. Only the baseline data on soil samples are discussed here. The feasibility study and the baseline data for sediment samples have been discussed elsewhere by Lang (*4*).

Sample Collection and Preparation

Six Air Force Logistics Command bases were sampled, including Hill AFB, Utah; Kelly AFB, Texas; McClellan AFB, California; Robins AFB, Georgia; Tinker AFB, Oklahoma; and Wright-Patterson AFB, Ohio. All bases represent urban environments with substantial industrialization and histories of considerable pesticide use.

Soil samples were collected from residential, open or nonuse, and golf course areas. Core samples from each use stratification were taken from the top 3 inches (7.6 cm) with a 3-inch (7.6-cm)-diameter bulb planter. Twenty core samples from each site were composited in a plastic bucket, thoroughly mixed by hand, and poured back and forth into a similar bucket. The composite sample was sieved through ¼-inch (6.4-mm) hardware cloth to remove large particles and debris. A subsample of the composite sample was placed in a clean hexane-rinsed 8-oz (240-ml) amber glass salve jar. Salve jars were capped with aluminum foil-lined lids and subsamples were kept frozen until being prepared for analysis. All sampling equipment was thoroughly rinsed with water after each stratum was sampled to avoid cross contamination.

At each residential sampling site, 10 individual core samples were taken from both sides of randomly selected streets. At those sites with sidewalks, all samples were taken within 1 ft (30.5 cm) of the sidewalk in the direction of the house. At sites without sidewalks, samples were taken approximately 4 ft (1.37 m) from the street. At each open sampling site, 10 core samples were collected at 45-ft (13.7-m) intervals along two parallel straight lines 45 ft (13.7 m) apart which originated at a randomly selected point. Golf course samples were collected from random starting points at 45-ft (13.7-m) intervals along both sides of the fairway at the edge of the rough.

Analytical Procedures

PREPARATION OF SAMPLES

Two grams of dry-sieved subsample (sieve size No. 14) were placed in a 15-ml test tube with a Teflon-lined screw cap, and 10 ml 3:1 hexane–isopropanol was added. Tubes were rotated for 4 hours, and the subsample was centrifuged. The solution was transferred to a 60-ml separatory funnel and washed three times with water to remove the alcohol. The solution was dried with anhydrous sodium sulfate, the solvent was reduced by evaporation, and the sample was cleaned by passage through a Florisil microcolumn. Subsample extracts were stored at low temperature for subsequent gas-chromatographic (GC) analysis.

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GAS CHROMATOGRAPHY

The analytical procedures were basically the same as those described by Wiersma et al. (*6*). Samples were analyzed for organochlorines and PCBs with a Tracor Model 222 gas chromatograph equipped with two Ni-63 electron-capture detectors (EC) and four glass columns. Two sets of polar and nonpolar columns were used to identify and confirm the organochlorine pesticides and PCBs. The gas chromatograph was equipped with a Model 8000 Varian Auto Sampler and interfaced with a Model 3354 Hewlett-Packard Data System. Instrument parameters and operating conditions follow:

Columns:	glass, 6 ft long, 6 mm OD × 4 mm ID, packed with (1) a mixture of 1.5 percent SP-2250 and 1.95 percent SP-2401 on 100–120-mesh Supelcon, AW, DMCS (2) a mixture of 4 percent SE-30 and 6 percent SP-2401 on 100–120-mesh Supelcon, AW, DMCS
Temperatures, °C:	detector 300 injection port 225 column 200
Carrier gas:	5–10 percent methane-argon flowing at 60 ml/minute

Compounds and their quantitative detectable levels are listed in Table 1. Minimum detectable levels of organochlorine pesticides were 0.01–2.00 mg/kg.

RECOVERY STUDIES

Recovery of the components listed in Table 1 ranged from 91 to 102 percent. Data presented in Tables 2 and 3 were not corrected for recovery.

Results and Discussion

Because a similar data pattern emerged on each base, data for a given year on the same pesticide on the same land use area were combined from all six bases (Table 2). ΣDDT residues were the most ubiquitous organochlorines on the six bases (Table 2). ΣDDT residues were also quantitatively higher overall than were residues of any other organochlorine except chlordane, which in 1975 had arithmetic mean levels consistently

TABLE 1. *Quantitative detection limits of organochlorines found in soils of six U.S. Air Force bases, 1975–76*

Compound	Residue, ppm
ΣDDT	0.05
Aldrin	0.01
Heptachlor	0.01
Lindane	0.01
Toxaphene	2.00
Chlordane	0.20
Dieldrin	0.02
Endrin	0.02
Heptachlor epoxide	0.01
Methoxychor	0.04
PCBs	0.40

TABLE 2. *Organochlorine residues in pooled soil samples from six U.S. Air Force installations, 1975–76*

Statistic	Residues, ppm dry wt															
	ΣDDT		Chlordane		Dieldrin		Heptachlor epoxide		Lindane		Heptachlor		Endrin		PCBs	
	75	76	75	76	75	76	75	76	75	76	75	76	75	76	75	76
Residential (20) (21)[1]																
Range	ND–3.83	ND–7.60	ND–52.11	ND–1.20	ND–0.04	ND–0.02	ND–0.03	ND–0.01	Tr	Tr	ND–0.16	Tr	ND–0.01	—	—	—
Average	0.86	0.63	5.43	0.16	0.01	<0.01	<0.01	<0.01	Tr	Tr	0.01	Tr	<0.01	—	—	—
% Pos. Sites	80.0	66.7	65.0	9.5	55.0	47.6	15.0	35.0	10.0	4.8	5.9	4.8	5.0	—	—	—
Nonuse (29) (29)[1]																
Range	ND–0.32	ND–13.93	ND–1.76	ND–3.44	ND–0.31	ND–0.10	ND–0.03	ND–0.06	—	Tr	—	ND–0.01	—	—	—	—
Average	0.06	0.94	0.09	0.18	0.01	0.01	<0.01	<0.01	—	Tr	—	<0.01	—	—	—	—
% Pos. Sites	48.0	44.8	24.0	14.0	17.4	24.0	17.0	14.3	—	3.5	—	7.0	—	—	—	—
Golf Course (17) (17)[1]																
Range	Tr–1.07	ND–0.69	ND–4.57	ND–3.05	ND–0.05	ND–0.03	ND–0.02	ND–0.01	—	Tr	—	—	—	ND–0.04	ND–3.20	ND–4.33
Average	0.19	0.16	0.67	0.56	0.01	0.01	<0.01	<0.01	—	Tr	—	—	—	<0.01	0.18	0.24
% Pos. Sites	70.6	58.8	58.8	35.3	23.5	23.5	11.8	17.7	—	5.9	—	—	—	5.9	5.9	5.9

NOTE: ND = nondetectable, Tr = trace.
[1]Number of samples collected from the indicated land use area in 1975 and 1976, respectively.

TABLE 3. *Geometric means and 95 percent confidence intervals for pooled ΣDDT, chlordane, and dieldrin residue data from use-stratified areas on six U.S. Air Force bases—1975–76*

Area	ΣDDT 95% CI Lower	ΣDDT Mean	ΣDDT 95% CI Upper	Chlordane 95% CI Lower	Chlordane Mean	Chlordane 95% CI Upper	Dieldrin 95% CI Lower	Dieldrin Mean	Dieldrin 95% CI Upper
				1975 Residues, ppm					
Residential	0.0791	0.2276[a]	0.6549	0.0440	0.1875[a]	0.7153	0.0100	0.0246[a]	0.0606
Open	0.0124	0.0235[b]	0.0443	0.0091	0.0158[b]	0.0275	0.0089	0.0119[a]	0.0159
Golf course	0.0347	0.0599[b]	0.1033	0.0338	0.1049[a]	0.3253	0.0091	0.0122[a]	0.0163
				1976 Residues, ppm					
Residential	0.0257	0.0782[a]	0.2382	0.0033	0.0150[a]	0.0694	0.0094	0.0117[a]	0.0145
Open	0.0134	0.0361[a]	0.0960	0.0100	0.0182[a]	0.0329	0.0099	0.0129[a]	0.0167
Golf course	0.0110	0.0437[a]	0.1729	0.0084	0.0320[a]	0.1218	0.0089	0.0110[a]	0.0136

NOTE: For a given year, means in a vertical column followed by the same letter are not significantly different at the 5 percent level.

higher than ΣDDT. The high arithmetic mean for chlordane residues in residential areas during 1975 was mainly attributable to the high levels found at Wright-Patterson AFB. This finding is notable in light of past problems with chlordane contamination in Capehart housing units on Wright-Patterson (*1, 2*). Except for chlordane levels found in residential soils in 1975, the arithmetic means shown in Table 2 closely approximate mean levels of the same pesticides in various nonmilitary urban areas of the United States (*3*).

Since residue data are not normally distributed, the arithmetic means in Table 2 are useful for comparison only in a relative sense. Therefore, the more statistically useful geometric means and associated 95 percent confidence limits based on data normalized with the ln $(X + 0.01)$ transformation discussed by Carey et al. (*3*) are given in Table 3 for the three most ubiquitous pesticides: ΣDDT, chlordane, and dieldrin.

To obtain an overall picture of the data, a three-factor analysis of variance was used to evaluate pesticide by land use by year interactions. The only significant interaction was for land use areas between the two years. Further examination showed that only the residential area means for 1975 and 1976 differed significantly ($P < 0.001$). There was no significant difference for the open area means ($P < 0.10$) and only an indication of a difference for the golf course means ($P < 0.10$).

One-way analyses of variance were used to evaluate data on a particular pesticide during a given year. Significant F values were found only for ΣDDT and chlordane in 1975. Bartlett's test (*5*) was used to check homogeneity of variances in the two cases. Variances were homogeneous for the ΣDDT data but not for the chlordane data. Therefore, standard t-tests were used to compare mean differences in ΣDDT data, and t′-tests (*5*) were used on the chlordane data.

In 1975, ΣDDT residues were significantly higher in residential areas than in open and golf course areas. Chlordane levels were significantly higher in both residential and golf course areas than in open areas. There was no significant difference in chlordane levels between residential and golf course areas.

Large differences between 1975 and 1976 ΣDDT and chlordane data (Table 3) are puzzling. From what is known generally of organochlorine degradation rates, microbial or other forms of degradation could not account for the decreases in ΣDDT and chlordane levels between 1975 and 1976. The most likely explanation for the rather drastic reduction in ΣDDT residues in residential areas and chlordane residues in residential and golf course areas between 1975 and 1976 is the irregular distribution of pesticide residues in the environment and the relatively small number of samples collected from each land use area.

Conclusions

Organochlorine residues on the six Air Force installations generally were the same generic type and quantity as those found in nonmilitary urban environments. ΣDDT residues were the most abundant followed by chlordane and dieldrin. Residential areas generally were contaminated more heavily with organochlorines than were open or nonuse and golf course areas. Large variations between 1975 and 1976 data on some pesticides indicate that, if the Air Force program is continued, more samples should be taken from each sampling site and increased emphasis should be placed on sampling protocol to ensure the gathering of comparative data.

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APPENDIX[1]

Chemical Names of Compounds Discussed in This Issue

ACEPHATE	*O,S*-Dimethyl acetylphosphoramidothioate
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
CHLORDANE	1,2,3,4,5,6,7,8,8-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	Dichlorophenyl 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]
DELNAV	2,3-*p*-Diozanedithiol *S,S*-bis (*O,O*-diethyl phosphorodithioate)
DIAZINON	*O,O*-Diethyl *O*-(2-isopropyl-6-methyl-4-pyrimidinyl) phosphorothioate
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
DURSBAN	*O,O*-Diethyl *O*-(3,5,6-trichloro-2-pyridyl)
ENDRIN	Hexachlorocepoxyoctahydro-*endo,endo*-dimethano-naphthalene
ETHION	*O,O,O',O'*-Tetraethyl *S,S'*-methylene bisphosphorodithioate
HCB	Hexachlorobenzene
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-methanoindene
LINDANE	*Gamma* isomer of 1,2,3,4,5,6-hexachlorocyclohexane
MALATHION	*O,O*-Dimethyl dithiophosphate of diethyl mercaptosuccinate
METHAMIDOPHOS	*O,S*-Dimethyl phosphoramidothioate
MIREX	1,1a,2,2,3,3a,4,5,5,5a,5b,6-Dodecachlorooctahydro-1,3,4-metheno-1*H*-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-2*H*-indeno(1,2-β)oxirene
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)
TOXAPHENE	Chlorinated camphene (67-69% chlorine). Product is a mixture of polychlor bicyclic terpenes with chlorinated camphenes predominating.
TRITHION	*S*-[[(*p*-Chlorophenyl)thio]methyl] *O,O*-diethyl phosphorodithioate

[1] Does not include compounds listed only in Carey and Gowen and in Carey et al.

ERRATA

PESTICIDES MONITORING JOURNAL, Volume 12, Number 3

Page 99: Charles D. Kennedy and Roy L. Schutzmann, coauthors of the paper "Pesticide Residues in Estuarine Mollusks, 1977 versus 1972—National Pesticide Monitoring Program" are employed by the Ecological Monitoring Branch, Pesticides Monitoring Laboratory, U.S. Environmental Protection Agency, Bay St. Louis, MS 39520.

Pages 137–148: In the paper "Pesticide Application and Cropping Data from 37 States, 1971—National Soils Monitoring Programs," maps for Figures 1 and 2 were transposed.

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SUBJECT AND AUTHOR INDEXES

Volume 12, June 1978—March 1979

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 or organophosphates 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.

* Note: With the exception of 12(3):137-148 and 12(4):198-208 in which no compounds are used as secondary headings. Each compound is listed as a primary heading with application as its only secondary heading.

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