

A Survey of Chlorinated Hydrocarbon Residues in Menhaden Fishery Products

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Introduction

Menhaden (*Brevoortia tyrannus* and *B. patronus*) is the largest volume (but probably the least-known) fishery in the United States because it is not used directly as food for humans. Before Europeans arrived in the Western Hemisphere, menhaden played an active role in American Indian agriculture. Historians state that the Indian Squanto showed the colonists of New England how to catch and use these fish when they schooled inshore in the springtime. A fish was placed in every hill along with the seeds of corn to provide food for the heavily feeding plants, an early example of organic gardening. Future American farmers who did not follow the Indians' lead eventually found the

soil too impoverished to sustain continuous growth of corn, cotton, and tobacco, all of which place heavy demands on the nutrients in the soil.

Menhaden is an oily fish, and, shortly after the War of 1812, menhaden oil found use as a substitute for whale oil in lamps. Autolysis (self-digestion) was the original method for reducing the fish into oil. When cooking was introduced into the reduction process, the leftover cooked flesh was sold as fertilizer. By the 1880's, Europeans were adding fish meal, the dried

residual flesh, to animal feeds, a practice adopted in the United States only after World War I.

Since 1935, fish meal has become too valuable for use as fertilizer. Virtually all menhaden meal is now used in animal feed, indirectly an important source of protein for humans. In the years 1971-75, menhaden constituted more than 40 percent by weight of all fish landings in the United States (Robinson, 1977).

Few people are aware of the contribution of menhaden products to the human diet. The oil is a common raw material processed for use in margarine and shortening in many countries, but it has not been used for that purpose in the United States since 1951 (Stansby, 1973). Poultry feeds often contain up to 5 percent menhaden meal as a source of high quality protein as well as unknown growth factors. Menhaden oil also is used in animal feed as a source of calories.

Starting in 1969, the fishing industry became aware that a class of chemicals called chlorinated hydrocarbons were accumulating in menhaden oil in much the same way as they were in agricultural products and freshwater fisheries. The insecticide DDT¹, which had prevented the typhus and plague epidemics after

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ABSTRACT— Menhaden is the largest volume but, because it is rarely consumed by humans, the least-known fishery in the United States. Oil from menhaden is a component of animal feeds and after processing, in some countries, of margarine and shortening. Menhaden meal is a source of protein and unknown growth factors in poultry feeds. We have determined the chlorinated hydrocarbon content of whole menhaden (*Brevoortia tyrannus* and *B. patronus*) and its reduction products: menhaden meal, oil, solubles, and stickwater, in the 9 years 1969-77. We analyzed 610 samples for Σ DDT (1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane, DDT; and its metabolites 1,1-dichloro-2,2-bis(p-chlorophenyl)ethylene, DDE; and 1,1-dichloro-2,2-bis(p-chlorophenyl)ethane, TDE), 314 samples for PCB's (polychlorinated biphenyls),

and 253 samples for dieldrin (1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4:5,8-endo-exo-dimethanonaphthalene) and endrin (the endo-endo isomer of dieldrin). Both the arithmetic means and the high values for chlorinated hydrocarbons decreased in most types of samples from most areas over the period studied. Nonetheless, statistical analyses of the 541 data on menhaden oils using the F-test and Scheffe's method for multiple comparison (s-method) only partially confirmed the hypothesis that chlorinated hydrocarbon levels were decreasing with time. Comparisons among the four areas of the menhaden fishery showed significant differences in levels in oils in some years. These differences probably related to variations in size, age distribution, and life history of sample groups.

¹Abbreviations used in this manuscript: DDT = 1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane; DDE = 1,1-dichloro-2,2-bis(p-chlorophenyl)ethylene; TDE = 1,1-dichloro-2,2-bis(p-chlorophenyl)ethane; Σ DDT = DDT and its metabolites DDE and TDE; and PCB's = polychlorinated biphenyls.

World War II, was the first such problem chemical. Early in the 1970's, an industrial chlorinated hydrocarbon group called polychlorinated biphenyls (PCB's) was similarly identified in menhaden oil. PCB's boil at high temperatures, do not burn or degrade chemically, and possess excellent electrical insulating properties. PCB's became widely distributed in the environment from uses in water-resistant coatings, marine lubricants, and transformers and capacitors. During the past decade, other chlorinated hydrocarbon insecticides, including toxaphene, chlordane, mirex, kepone, endrin, and dieldrin have also been identified in menhaden oil. All these chlorinated hydrocarbons have common properties. They are resistant to degradation both by chemicals and by the environment. They are insoluble in water and highly soluble in oil and fat. They accumulate in fatty tissues of organism both at the beginning of the food chain and at the end, in plankton and in carnivores, in sharks and in man. Unfortunately, they are also toxic to a variety of organisms besides insects.

Over the the past 20 years, numerous authors have documented the damaging effects of chlorinated hydrocarbons on the environment. Rachel Carson (1962) described in depth the effects of DDT accumulation. Burdick et al. (1964) correlated the death of lake trout fry with elevated concentrations of DDT in the eggs. Kuratsune et al. (1972) reported about the PCB-contaminated rice bran oil that caused the 1968 epidemic of the condition called Yusho in Japan. Another incident related to contaminated rice bran oil occurred in Taiwan in 1979². Several episodes of poor hatchability and deformities in chickens in the United States occurred when PCB's leaked into fish meal during pasteurization (Cecil et al., 1972). Aulerich et al. (1973) determined that mink, fed Lake Michigan coho salm-

on, failed to whelp because of the high level of PCB's in the fish. Dieldrin came to the fore in 1974 when high levels of dieldrin in meat products were traced back to soybean foots accidentally added to animal rations. (Soybean foots, the colored and odoriferous substances removed from soybean oil during alkali refining, are considered inedible and normally are directed to industrial uses. Furthermore, dieldrin was present in the foots at a concentration that was unexpectedly high, because dieldrin was not applied directly to soybean plants. Dieldrin did find widespread use in the control of insect pests in corn, and soybeans often follow corn in the usual rotation of crops. Presumably dieldrin was absorbed by the soybean plants from the soil either directly through the roots or indirectly by evaporation and subsequent transpiration into the leaves.)

The continuing problems of chlorinated hydrocarbon contamination of foodstuffs for both animal and human consumption led to a variety of regulatory actions throughout the world. The U.S. Food and Drug Administration established an administrative guideline of 5 ppm (parts per million) for Σ DDT in the edible portion of fish (the only product of marine origin for which a DDT guideline was ever established). The tolerance for dieldrin in fish oil was set at 0.3 ppm, an indication of the greater toxicity of this compound. Temporary PCB tolerances were established in 1974 (Schmidt) at 5 ppm in the edible portion of fish, 2 ppm in animal feed components including fish meal and oil, and 0.2 ppm in finished animal feed. Sweden banned DDT in the late 1960's as did the United States in 1972. The Japanese were the first to stop manufacture and use of PCB's because of the 1968 Yusho episode which affected more than 1,000 people (Kuratsune et al., 1972). PCB usage in the United States was restricted in stages. Currently, PCB's may be used legally for only a few specialty purposes in the United States.

When this project began in 1969,

fish from the Great Lakes were being seized for excess Σ DDT and the possibility of elevated levels in menhaden oil was of great concern to the industry. Menhaden oil is used in paints and lubricants, but is more valuable as a component of animal feeds. Initially we wanted to determine the levels of Σ DDT present in menhaden fishery products from various geographical locations. In addition, if elevated levels were found, we wanted to investigate methods for removing Σ DDT, especially from menhaden oil. Fish oil contains all the chlorinated hydrocarbons initially present in the fish, undiluted by the protein and water. Obviously the oil would contain higher concentrations of the chlorinated hydrocarbons than either the original fish or the other reduction products: Meal and solubles. Our studies (Stout et al., 1970) showed that removal of chlorinated hydrocarbons was not practical except under the conditions used for refining fish oil for production of margarine and shortening.

Subsequent work concentrated on determining the chlorinated hydrocarbon levels, initially Σ DDT, in samples from throughout the fishery. In 1971, we first included PCB's in our investigations. We added dieldrin and endrin analysis to our regimen in 1974 when a tankcar of menhaden oil was detained by regulatory officials, because the oil contained more than 0.3 ppm dieldrin. For historical perspective, we assayed the dieldrin content of some of our older samples as well.

This paper reports our findings on Σ DDT, PCB's, dieldrin, and endrin in whole menhaden and menhaden meal, oil, and solubles, from all areas of the fishery in the years 1969-77.

Materials and Methods

Atlantic menhaden, *Brevoortia tyrannus*, occurs along the Atlantic coast from Maine to Florida; Gulf menhaden, *B. patronus*, occurs in the Gulf of Mexico (Fig. 1). For those unfamiliar with fish reduction, a brief description will serve to define the substances studied in this paper.

²Anonymous. 1980. PCB widespread poisoning incident in Taiwan reported. Food Chem. News, Jan. 7, p. 23.

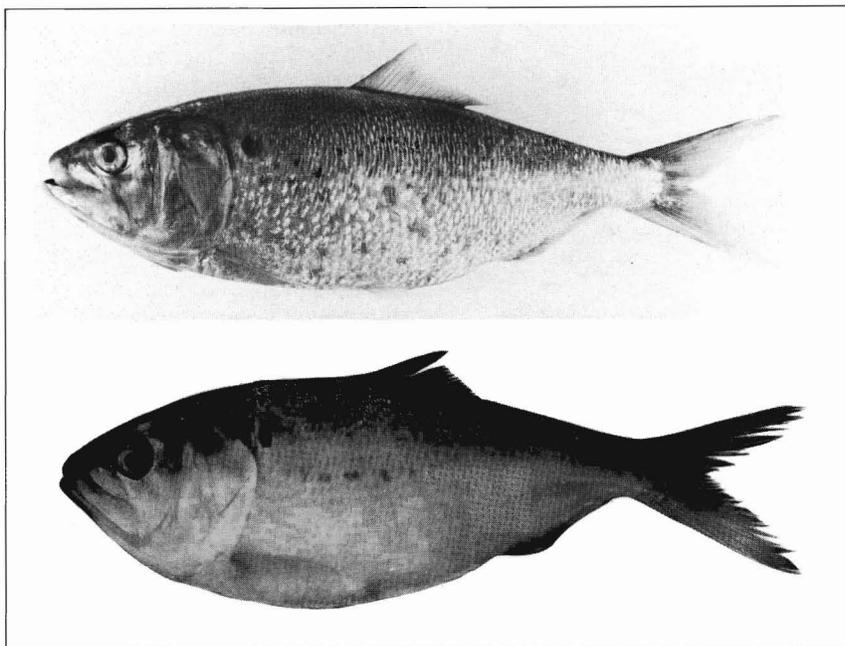


Figure 1.—Atlantic menhaden, *Brevoortia tyrannus*, fork length 25 cm (top). Gulf menhaden, *Brevoortia patronus*, fork length 27 cm (bottom). Photographs by J. Reintjes.

Whole fish are heated with steam in a large cooker and transferred to a press. The solid portion or presscake is dried to give fish meal. The liquid, which is expressed by the press, is centrifuged to separate the fish oil from the aqueous fraction, the stickwater. Evaporation of water from the latter yields solubles, which are sometimes sprayed onto the presscake prior to drying. Reduction plants cooperated in collecting samples from all regions of the fishery (Fig. 2). Fish were obtained just as they were being unloaded and the collection site noted. Collection of samples of menhaden meal, oil, and solubles was timed so that the samples of products represented the same fish as those from which the samples of fish themselves had been drawn. Obviously, this system was only approximate.

Fish were wrapped in aluminum foil and frozen at the time of collection. Solubles and oil were placed in glass bottles with aluminum foil or

Teflon³ lining the lids. Samples of meal were stored in double brown paper bags which contained no contaminating substances. Bottles, bags, and aluminum foil were shipped to all samplers to assure uniformity of receivers. Bottles were washed and baked at 260°-270°C before shipment.

After shipment of the frozen samples to this center, the fish were partially thawed, ground whole, mixed thoroughly, and stored in aluminum tubs sealed with tape at -18°C until analysis. Meals were ground in a meat grinder to break up scales and bones to provide a more homogeneous sample.

Chlorinated hydrocarbon analysis was evolving during the course of these studies. For the 1969 samples, the AOAC method for determination of chlorinated pesticides in nonfatty

³Mention of trade names does not imply endorsement by the National Marine Fisheries Service, NOAA.

foods (Horowitz, 1970a) was used for fish and meal by adjusting the sample size to limit the oil content to 2 g, and adding water to make a total water volume of 80 ml. The oil was analyzed by extending the backwash procedure of Porter et al. (1970) of the AOAC method for fatty foods (Horowitz, 1970b). Oil (3 g), diluted to 15 ml with petroleum ether, was extracted with 30 ml of acetonitrile saturated with petroleum ether. The acetonitrile fraction was backwashed with 15 ml of petroleum ether. Five additional 30 ml portions of acetonitrile were shaken successively with the two petroleum ether solutions. This extensive partitioning process was necessary to obtain quantitative extraction of the chlorinated hydrocarbons from the fish oils and to separate them from interfering substances present in high concentration in the oils. The combined acetonitrile fractions were treated with 2 percent (weight/volume) aqueous NaCl and 100 ml of petroleum ether in the usual manner (Stout, 1978).

In all later years, the procedures of Reinert (1970) were used, because they gave cleaner extracts of marine fishery samples; Stout and Beezhold (1979) detailed our methods. The technique for fish oil was especially efficient for samples of marine origin which form intractable emulsions during the AOAC extraction procedure.

PCB separation from Σ DDT was introduced in 1971. Consequently, in samples from 1969 and 1970, part of the PCB's was quantitated as Σ DDT because of the overlap between the gas chromatographic peaks of certain components of PCB's and the DDT family. Although no effort was made to correct for PCB interference, the levels of Σ DDT were high enough at that time that the data give a fairly accurate indication of the extent of Σ DDT contamination. Ironically, by the end of the study 5 years after DDT was banned, the levels of Σ DDT had decreased so much that the small gas chromatographic peaks of DDE, TDE, and DDT were sometimes obscured by unidentified substances.

Figure 2.—Areas of commercial menhaden fisheries and location of menhaden plants participating in the survey of chlorinated hydrocarbons.



For dieldrin and endrin analyses the alkaline saponification technique of Reinert (1970) was used. This procedure eliminated the contaminants remaining in extracts prepared by the AOAC method (Horowitz, 1970a).

Extracts were quantitated by electron-capture gas chromatography with a titanium-tritide detector. Details of our quantitation procedures have been published (Stout, 1980). The quantifiable limit for DDE, TDE, and DDT was 0.01 ppm in fish, meal, solubles, and stickwater and 0.05 ppm in menhaden oil. The quantifiable limit for PCB was 0.05 ppm in fish, meal, solubles, and stickwater, and 0.25 ppm in the oil. For dieldrin and endrin, the limit was 0.005 ppm in fish and meal and 0.01 ppm in the

oil. Residue values were calculated on the basis of μg chlorinated hydrocarbon per gram wet tissue or parts per million (ppm).

The data are tabulated by areas of commercial menhaden fishing (Dryfoos et al., 1973) (Fig. 2). Along the Atlantic coast, the New York and New Jersey areas were combined into the Middle Atlantic area, and the North Carolina area of Dryfoos et al. (1973) was designated as the South Atlantic area. Data for all plants in the fishery in the Gulf of Mexico were combined. In every area, grouping relates to the location of the reduction plant where the samples were collected. In earlier years, the Atlantic menhaden fisheries rarely overlapped. With the introduction of shipboard

refrigeration, vessels extended their territories into adjacent areas, but in tabulating data, no effort was made to segregate samples by area of catch.

The goals of the project and, therefore, the types of relevant samples, evolved over the years. The fishery declined for a time because the fish disappeared from some areas. Furthermore, collection, storage, and shipment of samples from remote areas were often difficult. Consequently, data for all contaminants are not available for all types of samples from all areas in every year. Table 1 summarizes the type of menhaden samples collected and the chlorinated hydrocarbons determined in each area of the fishery by year.

Results and Discussion

DDT and its metabolites

Overall, the ΣDDT levels declined in all products in all areas from the beginning of this survey in 1969 until the last year, 1977. Initially, the levels of ΣDDT were highest in all types of samples from the Middle Atlantic area (Fig. 2). In 1969, fish contained 0.966 ppm; meal, 0.399 ppm; and oil, 5.93 ppm ΣDDT . By 1976, the last year in which fish were collected, however, the concentration in fish in the Gulf of Mexico area (0.504 ppm) was three times that in the Middle Atlantic area (0.163 ppm). Although the concentration of ΣDDT in fish from the Gulf of Mexico declined between 1970 and 1974, the 1976 level exceeded that in 1970 (0.402 ppm), the first year in which fish were collected from the Gulf fishery. Since the seven samples of menhaden oil from the 1976 fishery in the Gulf of Mexico did not show a similar marked increase in ΣDDT level and the 16 samples of oil from the 1977 season contained less ΣDDT than in any previous year, the elevated level in the four 1976 fish samples may reflect a localized problem rather than a general trend. In all types of samples from the Middle Atlantic area, the levels of ΣDDT declined substantially during the 9 years of the survey. Similar, though less

Table 1.—Types of menhaden samples collected and chlorinated hydrocarbons analyzed by area and year.

Area	Type of chlorinated hydrocarbon			
	Σ DDT	PCB	Dieldrin	Endrin
Middle Atlantic				
1969	FMO	F ¹	O	O
1970	—	—	—	—
1971	FMO	FMO	O	O
1972	FMOS	FMOS	O	O
1973	FMOS	FMOS	O	O
1974	FMOS	FMOS	MO	MO
1975	—	—	MO	MO
1976	F O	F O	F O	F O
1977	O	O	O	O
Chesapeake Bay				
1969	MO	—	O	O
1970	FMO	M ¹	—	—
1971	FM	FM	—	—
1972	MOS	MOS	O	O
1973	MOS	MOS	O	O
1974	FMOS	FMOS	MO	MO
1975	—	—	MO	MO
1976	F O	F O	F O	F O
1977	O	O	O	O
South Atlantic				
1969	M	—	O	O
1970	FMO	—	—	—
1971	FMO	FMO	O	O
1972	—	—	—	—
1973	MOS	MOS	O	O
1974	MOS	MOS	MO	MO
1975	—	—	—	—
1976	F O	F O	F O	F O
1977	O	O	O	O
Florida				
1970	FM	M ²	—	—
Gulf of Mexico				
1969	M	—	O	O
1970	FMO	—	—	—
1971	—	—	—	—
1972	FMOSsw	FMOSsw	O	O
1973	FMOS	FMOS	O	O
1974	FMOS	FMOS	MO	MO
1975	—	—	MO	MO
1976	F O	F O	F O	F O
1977	O	O	O	O

F = whole fish, M = meal, O = oil, S = solubles, Sw = stickwater, — = no samples of any type collected in that year. Blanks indicate certain types of samples not collected or not analyzed for the indicated contaminant.

¹ One sample.
² Two samples.

dramatic, declines occurred in other areas as well, as shown graphically for menhaden oil from the South Atlantic fishery in Figure 3. The trend in samples from the Gulf of Mexico was more erratic (Fig. 4). Perhaps that behavior related to the lower initial levels in the area. Alternatively, the reservoir of ΣDDT in the Mississippi River drainage may be greater or its release more readily influenced by fluctuations in precipitation and other

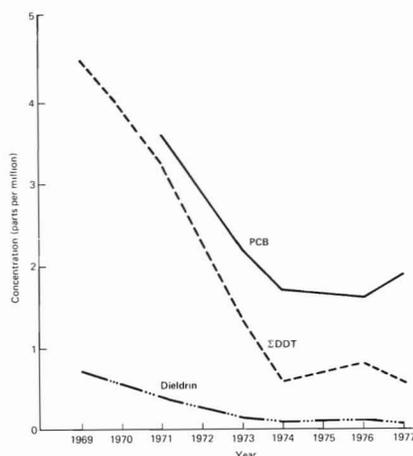


Figure 3.—Chlorinated hydrocarbon levels in oil from menhaden taken from the South Atlantic fishery, 1969-77.

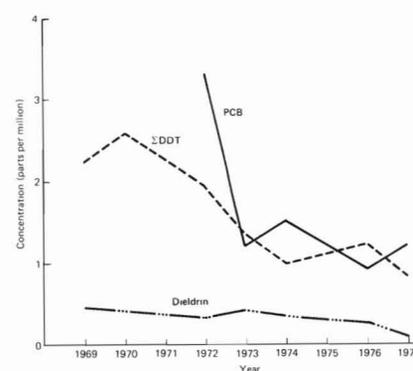


Figure 4.—Chlorinated hydrocarbon levels in oil from menhaden taken from the Gulf of Mexico fishery, 1969-77.

conditions. The data for ΣDDT in fish, meal, oil, solubles, and stickwater are summarized in Tables 2, 3, 4, and 5, respectively. Also shown are the relative proportions of DDT and its metabolites DDE and TDE. The most consistent change was a decrease in the parent compound DDT in menhaden oils. In oils from the Middle Atlantic states and the Gulf of Mexico, this decrease was accompanied by an increase in the proportion of TDE, the metabolite formed by a reductive process, involving replacement of one chlorine atom by a hydrogen atom. Since DDT usage declined in the late 1960's and was banned entirely in 1972 (and DDT does degrade slowly), decreases both in the amount of ΣDDT and the proportion of DDT might be anticipated. Also to be noted was the increased proportion of DDE in menhaden meal (Table 3). This might result from degradation of DDT to DDE during the reduction process. Iron and other chemicals catalyze the dehydrochlorination of DDT to DDE. This is the typical terrestrial mode of degradation of DDT, in contrast to the anaerobic degradation of DDT to TDE, which occurs in the marine environment in the presence of microorganisms. Dehy-

drochlorination may occur more rapidly at the elevated temperature of the meal drier.

PCB, the Industrial Chemicals

The initial findings about PCB's were most startling: 6.59 ppm in fish and 9.9 and 13.4 ppm in two samples of oil. Further analyses confirmed that nearly all samples contained much higher concentrations of PCB's than ΣDDT. Although PCB's were introduced a decade before DDT, PCB's were not identified as environmental pollutants until long after ΣDDT, because of the complexity of PCB analysis. The method for analyzing chlorinated hydrocarbons, gas chromatography with an electron-capture detector, produces one or more peaks for each individual compound present. PCB's are a mixture of up to 210 compounds resulting from the chlorination of biphenyl. The PCB's commonly found in samples of marine origin produce a series of more than ten broad peaks, several of which overlap the three narrow peaks produced by the DDT family, i.e., DDE, TDE, and DDT. Once recognized as ubiquitous environmental pollutants, PCB's still presented the challenge of developing a satisfactory method for separation from the DDT family. Furthermore, the electron-capture detector is less sensitive to

Table 2.—ΣDDT in menhaden fish by area (1969-76).

Area and year	No. of samples (total no. of fish)	Mean (ppm)	SD	Low (ppm)	High (ppm)	Percent of ΣDDT as			PCB / ΣDDT ratio
						DDE	TDE	DDT	
Middle Atlantic									
1969	2 (24)	0.966	0.161	0.852	1.08	23.6 ± 14.8	58.4 ± 9.8	18.1 ± 4.9	¹ 3.20
1971	1 (12)	1.59				48.8	40.3	10.8	2.04
1972	5 (88)	0.820	0.128	0.644	0.944	42.9 ± 7.9	43.1 ± 5.4	14.0 ± 3.8	4.46 ± 1.83
1973	5 (81)	0.569	0.125	0.400	0.722	46.9 ± 11.3	38.7 ± 5.9	14.4 ± 6.3	2.73 ± 1.01
1974	4 (75)	0.283	0.042	0.237	0.334	43.8 ± 7.8	45.2 ± 5.7	11.0 ± 3.5	3.64 ± 0.95
1976	4 (73)	0.163	0.156	<0.02	0.336	35.2 ± 9.9	52.9 ± 11.7	11.8 ± 2.3	4.30 ± 0.71
Chesapeake Bay									
1970	6 (61)	0.795	0.826	0.250	2.18	46.7 ± 17.0	34.9 ± 10.1	18.4 ± 10.3	n.a.
1971	3 (28)	0.440	0.165	0.312	0.626	38.0 ± 12.7	32.2 ± 2.8	29.8 ± 10.3	0.92 ± 0.06
1974	3 (58)	0.069	0.031	0.037	0.098	40.6 ± 2.3	50.6 ± 7.0	8.8 ± 9.0	3.96 ± 1.38
1976	2 (31)	0.090	0.047	0.056	0.123	44.6 ± 4.9	40.2 ± 8.7	15.2 ± 3.8	2.94 ± 0.36
South Atlantic									
1970	21(177)	0.189	0.176	0.026	0.853	45.6 ± 17.4	22.8 ± 13.4	32.5 ± 10.1	n.a.
1971	6 (72)	0.282	0.192	0.086	0.539	37.0 ± 4.9	33.1 ± 12.2	30.1 ± 10.4	2.56 ± 2.40
1976	2 (36)	0.158	0.155	0.049	0.268	34.4 ± 9.1	50.8 ± 8.3	14.8 ± 0.7	4.62 ± 3.37
Florida									
1970	10 (75)	0.071	0.080	0.019	0.283	45.9 ± 15.5	12.8 ± 20.8	41.7 ± 12.0	¹ 1.25
Gulf of Mexico									
1970	32(100)	0.402	0.220	0.145	1.12	27.7 ± 10.3	39.9 ± 4.7	32.2 ± 9.8	n.a.
1972	3 (42)	0.272	0.047	0.229	0.322	34.0 ± 11.6	35.6 ± 4.3	30.5 ± 9.9	1.86 ± 1.11
1973	8(155)	0.265	0.038	0.197	0.306	29.3 ± 10.8	39.3 ± 7.7	31.4 ± 6.7	1.28 ± 0.28
1974	9(184)	0.198	0.064	0.085	0.314	22.9 ± 9.0	52.8 ± 20.6	24.4 ± 15.1	1.85 ± 0.87
1976	4 (77)	0.504	0.043	0.460	0.563	28.2 ± 3.4	37.8 ± 2.8	34.1 ± 1.0	0.81 ± 0.11

¹One sample only.

When year omitted, either samples not obtained or analyses not performed.

n.a. = PCB not analyzed.

Table 3.—ΣDDT in menhaden meal by area (1969-74).

Area and year	No. of samples	Mean (ppm)	SD	Low (ppm)	High (ppm)	Percent of ΣDDT as			PCB / ΣDDT ratio
						DDE	TDE	DDT	
Middle Atlantic									
1969	6	0.399	0.098	0.324	0.591	39.6 ± 10.0	53.3 ± 9.7	7.2 ± 2.4	n.a.
1971	1	0.811				61.9	34.8	3.3	1.86
1972	5	0.273	0.041	0.226	0.334	53.8 ± 9.5	40.9 ± 5.0	5.3 ± 4.8	3.94 ± 1.02
1973	5	0.152	0.041	0.083	0.190	52.4 ± 10.7	34.7 ± 10.6	12.9 ± 6.4	3.26 ± 1.16
1974	4	0.104	0.025	0.083	0.135	55.5 ± 11.6	44.5 ± 11.6	0 ± 0	4.72 ± 2.26
Chesapeake Bay									
1969	2	0.228	0.022	0.213	0.244	26.0 ± 0.4	64.8 ± 13.3	9.2 ± 12.9	n.a.
1970	17	0.364	0.277	0.080	0.993	52.0 ± 15.2	30.6 ± 6.9	17.4 ± 10.2	¹ 0.82
1971	3	0.304	0.046	0.255	0.347	46.8 ± 4.5	36.0 ± 2.9	17.2 ± 3.2	1.29 ± 0.28
1972	4	0.226	0.040	0.166	0.250	40.1 ± 5.0	31.1 ± 5.0	28.8 ± 1.2	2.03 ± 0.41
1973	3	0.176	0.027	0.148	0.201	46.9 ± 3.8	36.4 ± 5.0	16.6 ± 7.4	1.97 ± 0.39
1974	5	0.067	0.006	0.058	0.075	55.3 ± 5.2	44.7 ± 5.2	0 ± 0	3.58 ± 0.78
South Atlantic									
1969	12	0.235	0.074	0.098	0.315	35.9 ± 5.9	37.1 ± 4.8	26.8 ± 3.2	n.a.
1970	36	0.168	0.068	0.069	0.358	47.2 ± 14.6	26.9 ± 13.3	26.1 ± 7.8	n.a.
1971	8	0.331	0.108	0.233	0.492	38.6 ± 9.7	41.9 ± 7.4	19.5 ± 11.8	1.62 ± 0.64
1973	3	0.117	0.052	0.076	0.175	45.3 ± 1.8	29.1 ± 7.1	25.7 ± 8.8	1.03 ± 0.55
1974	2	0.054	0.006	<0.05	0.059	¹ 59.3	¹ 40.7	0 ± 0	² 2.75
Florida									
1970	18	0.130	0.039	0.057	0.196	44.3 ± 11.8	16.1 ± 13.2	39.6 ± 7.1	² 0.71 ± 0.01
Gulf of Mexico									
1969	2	0.102	0.006	0.097	0.106	32.4 ± 4.9	35.6 ± 2.2	32.6 ± 3.5	n.a.
1970	2	0.169	0.079	<0.03	0.369	33.8 ± 9.8	33.1 ± 9.3	33.1 ± 8.7	n.a.
1972	4	0.164	0.126	0.088	0.353	34.3 ± 6.6	25.8 ± 10.5	40.2 ± 12.8	2.20 ± 0.68
1973	8	0.117	0.022	0.087	0.146	33.2 ± 6.2	37.8 ± 4.4	29.0 ± 5.9	1.42 ± 0.57
1974	9	0.059	0.038	0.028	0.155	33.2 ± 8.3	57.7 ± 10.6	9.2 ± 11.8	2.82 ± 0.98

When year omitted, either samples not obtained or analyses not performed.

n.a. = PCB not analyzed.

¹ One sample only.

² Two samples only.

Table 4.—ΣDDT in menhaden oil by area (1969-77).

Area and year	No. of samples	Mean (ppm)	SD	Low (ppm)	High (ppm)	Percent of ΣDDT as			PCB ΣDDT ratio
						DDE	TDE	DDT	
Middle Atlantic									
1969	6	5.93	1.78	4.42	9.36	37.5 ± 15.1	42.6 ± 10.1	19.8 ± 8.9	n.a.
1971	2	4.99	0.283	4.79	5.19	37.2 ± 5.4	44.7 ± 5.9	18.1 ± 0.6	2.36 ± 0.63
1972	5	4.40	1.06	2.73	5.49	43.6 ± 3.9	42.6 ± 3.7	13.9 ± 3.1	3.50 ± 0.19
1973	5	2.30	0.625	1.35	2.89	34.8 ± 13.2	43.3 ± 8.3	21.8 ± 9.2	2.62 ± 1.54
1974	4	1.11	0.332	0.743	1.54	34.7 ± 12.5	48.6 ± 7.5	16.6 ± 5.8	7.48 ± 4.19
1976	4	1.56	0.360	1.24	2.07	36.6 ± 10.1	52.0 ± 9.7	11.1 ± 1.2	5.56 ± 2.99
1977	6	0.978	0.207	0.745	1.21	28.8 ± 13.8	61.2 ± 12.6	10.0 ± 3.1	6.13 ± 1.84
Chesapeake Bay									
1969	2	3.41	0.834	2.82	4.00	19.0 ± 1.8	61.6 ± 7.3	19.2 ± 8.8	n.a.
1970	18	5.92	3.64	1.59	12.1	47.2 ± 17.0	33.0 ± 6.1	19.9 ± 13.2	n.a.
1972	4	3.78	0.519	3.01	4.16	40.2 ± 3.2	31.8 ± 3.4	27.9 ± 1.0	1.46 ± 0.30
1973	3	1.73	0.688	1.22	2.51	43.2 ± 8.5	36.8 ± 3.8	19.8 ± 6.0	1.57 ± 0.15
1974	5	1.02	0.136	0.902	1.24	38.7 ± 9.7	45.1 ± 4.0	16.3 ± 6.2	2.58 ± 0.47
1976	4	1.13	0.564	0.716	1.96	39.6 ± 15.3	48.8 ± 12.6	11.5 ± 3.1	1.59 ± 0.34
1977	4	0.708	0.355	0.380	1.21	54.4 ± 10.2	34.5 ± 9.1	11.2 ± 2.8	2.76 ± 0.59
South Atlantic									
1969	12	4.51	1.80	2.09	7.21	29.2 ± 4.2	43.9 ± 3.4	26.9 ± 2.0	n.a.
1970	29	3.93	1.20	2.47	7.14	41.0 ± 4.9	27.9 ± 4.2	31.1 ± 6.1	n.a.
1971	7	3.26	0.476	2.72	4.06	36.2 ± 4.3	41.6 ± 8.7	22.2 ± 7.0	1.18 ± 0.85
1973	3	1.34	0.263	1.04	1.51	34.6 ± 2.7	34.4 ± 2.8	30.6 ± 0.6	1.69 ± 0.45
1974	2	0.581	0.332	0.346	0.816	44.0 ± 5.4	46.2 ± 19.3	9.9 ± 13.9	2.73 ± 0.88
1976	6	0.819	0.339	0.496	1.44	42.3 ± 7.1	40.8 ± 4.0	17.0 ± 8.1	2.38 ± 1.82
1977	10	0.601	0.242	0.354	1.07	37.8 ± 10.3	49.1 ± 11.8	13.2 ± 4.6	2.70 ± 1.23
Gulf of Mexico									
1969	2	2.24	0.106	2.16	2.31	26.9 ± 6.6	36.8 ± 3.1	36.3 ± 3.7	n.a.
1970	42	2.55	1.05	< 1.46	6.28	27.8 ± 6.6	44.1 ± 8.2	28.1 ± 8.3	n.a.
1972	4	1.94	0.677	1.39	2.89	28.8 ± 9.8	37.7 ± 9.7	33.5 ± 3.5	1.84 ± 0.61
1973	9	1.36	0.245	1.05	1.80	24.6 ± 5.2	39.7 ± 3.8	35.8 ± 4.7	0.96 ± 0.40
1974	9	0.992	0.348	0.557	1.71	23.3 ± 9.8	46.5 ± 6.5	30.3 ± 9.8	1.58 ± 0.91
1976	7	1.23	0.469	0.673	1.67	29.9 ± 5.0	48.2 ± 4.2	21.9 ± 7.6	0.94 ± 0.17
1977	16	0.838	0.304	0.290	1.38	37.2 ± 13.6	47.9 ± 15.5	14.9 ± 6.5	1.54 ± 0.10

When year omitted, either samples not obtained or analyses not performed.

n.a. = PCB not analyzed.

¹North Carolina oil, January 1972 production, considered part of New Jersey fishery because of distinctly different chlorinated hydrocarbon characteristics from North Carolina early fall fishery.

Table 5.—ΣDDT in menhaden solubles and stickwater by area (1972-74).

Area and year	No. of samples	Mean (ppm)	SD	Low (ppm)	High (ppm)	Percent of ΣDDT as			PCB ΣDDT ratio
						DDE	TDE	DDT	
Middle Atlantic									
1972	5	0.368	0.140	0.234	0.580	55.0 ± 10.3	44.2 ± 9.7	0.9 ± 1.7	3.50 ± 0.80
1973	5	0.216	0.083	0.140	0.346	55.2 ± 12.3	41.3 ± 9.6	3.5 ± 3.4	3.10 ± 1.34
1974	4	0.125	0.064	0.069	0.182	50.5 ± 3.7	49.5 ± 3.7	0 ± 0	4.05 ± 0.62
Chesapeake Bay									
1972	4	0.212	0.068	0.130	0.297	46.0 ± 6.0	37.6 ± 9.5	16.4 ± 4.7	1.43 ± 0.43
1973	3	0.100	0.038	0.063	0.139	58.4 ± 1.2	41.6 ± 1.2	0 ± 0	2.53 ± 0.45
1974	5	0.053	0.018	0.033	0.070	54.8 ± 13.5	45.2 ± 13.5	0 ± 0	3.36 ± 1.62
South Atlantic									
1973	3	0.101	0.018	0.080	0.112	40.4 ± 3.7	42.1 ± 12.5	17.5 ± 13.2	1.67 ± 0.72
1974	2	0.022	0.005	0.019	0.026	51.3 ± 1.8	48.7 ± 1.8	0 ± 0	2.16 ± 0.59
Gulf of Mexico									
1972	1	0.232				46.6	53.4	0	1.55
1972	3	0.038	0.022	< 0.02	0.062	26.8 ± 12.2	44.2 ± 14.7	29.0 ± 2.3	14.55 ± 17.21
1973	9	0.150	0.070	0.042	0.230	45.2 ± 9.7	51.8 ± 8.4	3.2 ± 7.0	1.71 ± 0.94
1974	8	0.092	0.024	0.039	0.123	38.1 ± 12.8	61.9 ± 12.8	0 ± 0	1.82 ± 1.06

When year omitted, either samples not obtained or analyses not performed.

¹ Stickwater; all other samples solubles.

Table 6.—PCB in menhaden fish by area (1969-76).

Area and year	No. of samples (total no. of fish)	Mean (ppm)	SD	Low (ppm)	High (ppm)
Middle Atlantic					
1969	1 (12)	6.59			
1971	1 (12)	3.25			
1972	5 (88)	3.67	1.54	1.58	5.7
1973	5 (81)	1.61	0.782	0.73	2.4
1974	4 (75)	1.05	0.390	0.61	1.5
1976	4 (73)	1.02	0.645	0.15	1.7
Chesapeake Bay					
1971	3 (28)	0.409	0.177	0.26	0.60
1974	3 (58)	0.271	0.169	0.17	0.46
1976	2 (31)	0.271	0.171	0.15	0.39
South Atlantic					
1971	6 (72)	0.510	0.313	0.14	0.94
1976	2 (36)	0.995	1.25	<0.11	1.88
Florida					
1970	1 (12)	0.454			
Gulf of Mexico					
1972	3 (42)	0.493	0.288	0.30	0.82
1973	8(155)	0.343	0.105	0.24	0.51
1974	9(184)	0.325	0.107	0.18	0.50
1976	6(139)	0.301	0.160	0.06	0.43

When year omitted, samples either not obtained or analyses not performed.

Table 7.—PCB in menhaden meal by area (1970-74).

Area and year	No. of samples	Mean (ppm)	SD	Low (ppm)	High (ppm)
Middle Atlantic					
1971	1	1.51			
1972	5	1.06	0.238	0.83	1.46
1973	5	0.518	0.274	0.18	0.91
1974	4	0.502	0.259	0.14	0.75
Chesapeake Bay					
1970	1	0.71			
1971	3	0.383	0.031	0.35	0.41
1972	4	0.447	0.010	0.44	0.46
1973	3	0.353	0.107	0.23	0.42
1974	5	0.242	0.074	0.19	0.37
South Atlantic					
1971	8	0.552	0.287	0.08	1.01
1973	3	0.116	0.066	0.05	0.18
1974	2	0.124	0.053	0.09	0.16
Florida					
1970	2	<0.1	0		
Gulf of Mexico					
1972	4	0.309	0.124	0.19	0.47
1973	8	0.172	0.087	0.06	0.34
1974	9	0.147	0.061	0.09	0.28

When year omitted, samples either not obtained or analyses not performed.

Table 8.—PCB in menhaden oil by area (1971-77).

Area and year	No. of samples (total no. of fish)	Mean (ppm)	SD	Low (ppm)	High (ppm)
Middle Atlantic					
1971	2	11.7	2.46	9.9	13.4
1972	5	15.4	3.93	9.5	19.5
1973	5	6.2	3.53	1.4	9.9
1974	4	7.6	4.05	3.3	12.5
1976	5	8.0	3.54	3.6	12.0
1977	6	6.1	2.26	2.7	9.0
Chesapeake Bay					
1972	4	5.4	0.561	4.9	6.2
1973	3	2.7	0.856	1.9	3.6
1974	5	2.6	0.234	2.3	3.0
1976	5	1.8	0.596	1.2	2.6
1977	5	2.0	1.31	1.0	4.2
South Atlantic					
1971	7	3.6	2.23	1.3	7.0
1973	3	2.2	0.433	1.8	2.7
1974	2	1.7	1.42	0.73	2.7
1976	8	1.6	1.19	0.40	3.7
1977	10	1.9	1.64	0.59	5.8
Gulf of Mexico					
1972	4	3.3	0.561	2.7	4.0
1973	9	1.3	0.369	0.69	2.0
1974	9	1.5	0.888	0.90	3.6
1976	13	0.92	0.388	0.51	1.6
1977	18	1.22	0.302	0.50	1.6

When year omitted, samples either not obtained or analyses not performed.

¹ North Carolina oil, January 1972 production, considered part of New Jersey fishery because of distinctly different chlorinated hydrocarbon characteristics from North Carolina early fall fishery.

many of the components of PCB's than to the DDT family. Recognition and identification of those strange PCB peaks which appeared in the gas chromatograms used to quantify ΣDDT required complex analytical techniques, which are now used routinely in environmental studies, but which were just being developed in research laboratories during the late 1960's. Even today quantification of PCB's is a complex process, requiring much skill, experience, and attention to detail. Accurate and precise PCB analyses are still much more difficult to achieve than those for ΣDDT, dieldrin, or endrin.

In general, PCB levels decreased over the interval of this study, 1969-77, but the declines were less dramatic than for ΣDDT. The insecticide DDT was diluted substantially and sprayed thinly over wide areas. In this way it was readily absorbed on dust and soil particles, subjected to degradation by sunlight, air, and water, and buried by silt and sediment. When use stop-

ped, the reservoir of ΣDDT generally disappeared rapidly. In contrast, in Alabama and California at two sites where DDT was manufactured and the wastes discharged without substantial dilution (O'Shea et al., 1980; Stout and Beezhold, 1981), large reservoirs of ΣDDT still remain and fauna collected nearby contained elevated levels of this contaminant long after manufacture of DDT ended. PCB's were most commonly used in large quantities in undiluted form as a dielectric fluid (insulator) in transformers and capacitors. Many old electrical systems are still in operation. Dispersal of PCB's from this equipment results from slow evaporation and spillage. More PCB's are slowly dissipating by seepage from equipment buried at waste-disposal sites. Thus, a huge reservoir of PCB's remains in the environment. It will take a long time to disappear. For example, in oil the mean for PCB's in the South Atlantic area went from 3.6 ppm in 1971 to 1.9 ppm in 1977,

whereas for ΣDDT the value decreased from 3.26 ppm to 0.601 ppm during the same period. The PCB data are summarized in Tables 6-9.

Dieldrin and Endrin

The levels of dieldrin in the menhaden fishery have decreased over the years studied, as is most obvious from an examination of the high values for each set of data. For instance, in the South Atlantic fishery, the highest dieldrin level in 1969 was 1.53 ppm, compared with 0.16 ppm in 1977. The levels of dieldrin and endrin in menhaden fish and fishery products were always lower than those for ΣDDT and PCB's. One sample of oil collected in the Middle Atlantic area in 1969 contained 3.17 ppm dieldrin; another from the same area in 1972 contained 2.62 ppm. These were the exceptions. Even

Table 9.—PCB in menhaden solubles and stickwater by area (1972-74).

Area and year	No. of samples (total no. of fish)	Mean (ppm)	SD	Low (ppm)	High (ppm)
Middle Atlantic					
1972	5	1.29	0.527	0.58	1.8
1973	5	0.640	0.344	0.43	1.2
1974	4	0.513	0.289	0.23	0.85
Chesapeake Bay					
1972	4	0.285	0.068	0.22	0.38
1973	3	0.268	0.118	<0.13	0.36
1974	5	0.180	0.129	0.07	0.39
South Atlantic					
1973	3	0.160	0.036	0.13	0.20
1974	2	0.050	0.024	0.03	0.07
Gulf of Mexico					
1972	1	0.36			
1972	13	0.362	0.281	<0.2	0.68
1973	9	0.246	0.183	0.08	0.68
1974	8	0.170	0.116	0.05	0.43

When year omitted, samples either not obtained or analyses not performed.

¹ Stickwater; all other samples solubles.

Table 10.—Dieldrin in menhaden fish and meal by area (1974-76).

Area and year	Type of sample	No. of samples ¹	Mean (ppm)	SD	Low (ppm)	High (ppm)
Middle Atlantic						
1974	Meal	4	0.015	0.006	0.007	0.021
1975	Meal	6	0.024	0.009	0.009	0.033
1976	Fish	5 (92)	0.066	0.010	0.058	0.080
Chesapeake Bay						
1974	Meal	5	0.011	0.003	0.006	0.013
1975	Meal	4	0.008	0.000	0.007	0.008
1976	Fish	5 (74)	0.017	0.005	0.013	0.023
South Atlantic						
1974	Meal	2	0.005	0		
1976	Fish	8(181)	0.010	0.004	0.003	0.015
Gulf of Mexico						
1974	Meal	9	0.029	0.014	0.008	0.059
1975	Meal	14	0.012	0.007	0.005	0.025
1976	Fish	12(249)	0.061	0.025	0.012	0.088

When year omitted, samples either not obtained or analyses not performed.

¹Number in parentheses indicates total number of fish.

in the early years of the survey, fewer than 20 percent of the samples of menhaden oil contained more than 1 ppm dieldrin. Since the initial levels of

Table 11.—Dieldrin in menhaden oil by area (1969-77).

Area and year	No. of samples (total no. of fish)	Mean (ppm)	SD	Low (ppm)	High (ppm)
Middle Atlantic					
1969	6	0.91	1.11	0.37	3.17
1971	2	0.90	0.046	0.87	0.94
1972	5	1.1	0.866	0.43	2.6
1973	5	0.51	0.207	0.24	0.78
1974	4	0.36	0.196	0.18	0.62
1975	6	0.36	0.149	0.17	0.58
1976	5	0.25	0.141	0.12	0.45
1977	6	0.20	0.109	0.11	0.41
Chesapeake Bay					
1969	2	0.54	0.081	0.48	0.60
1972	4	0.39	0.032	0.36	0.44
1973	3	0.31	0.008	0.30	0.31
1974	5	0.20	0.034	0.16	0.25
1975	5	0.20	0.046	0.15	0.28
1976	5	0.14	0.036	0.09	0.19
1977	5	0.11	0.043	0.07	0.16
South Atlantic					
1969	12	0.72	0.404	0.15	1.53
1971	7	0.40	0.163	0.26	0.67
1973	3	0.14	0.029	0.11	0.17
1974	2	0.09	0.025	0.07	0.11
1976	8	0.11	0.047	0.04	0.21
1977	10	0.08	0.048	0.03	0.16
Gulf of Mexico					
1969	2	0.46	0.257	0.28	0.64
1972	4	0.33	0.022	0.30	0.35
1973	9	0.42	0.149	0.18	0.56
1974	9	0.35	0.110	0.12	0.47
1975	14	0.31	0.139	0.12	0.54
1976	13	0.26	0.115	0.08	0.41
1977	18	0.11	0.077	0.01	0.24

When year omitted, samples either not obtained or analyses not performed.

¹North Carolina oil, January 1972 production, considered part of New Jersey fishery because of distinctly different chlorinated hydrocarbon characteristics from North Carolina early fall fishery.

dieldrin and endrin in menhaden products were lower, the dramatic declines observed for ΣDDT and PCB's would not be anticipated. Our findings are summarized in Tables 10 and 11.

Endrin is an insecticide related to dieldrin, but with much more restricted usage, mainly in cotton production. Endrin levels were the lowest of all chlorinated hydrocarbons studied. The highest levels, e.g., 0.259 ppm in 1974, were found in menhaden oils from plants in the Gulf of Mexico area (Tables 12, 13).

Statistical Evaluations

The 719 data on chlorinated hydro-

Table 12.—Endrin in menhaden fish and meal by area (1974-76).

Area and year	Type of sample	No. of samples ¹	Mean (ppm)	SD	Low (ppm)	High (ppm)
Middle Atlantic						
1974	Meal	4	<0.01	0.008	0.003	<0.01
1975	Meal	6	<0.01	0		
1976	Fish	5 (92)	<0.009	0.002	0.005	<0.01
Chesapeake Bay						
1974	Meal	5	0.004	0.003	0.002	0.010
1975	Meal	4	0.01	0		
1976	Fish	5 (74)	0.002	0.001	0.001	0.004
South Atlantic						
1974	Meal	2	0.001	0.000	0.001	0.002
1976	Fish	8(181)	<0.005	0.004	0.001	<0.01
Gulf of Mexico						
1974	Meal	9	0.008	0.007	0.002	0.020
1975	Meal	14	0.006	0.004	0.001	0.009
1976	Fish	12(249)	0.012	0.005	0.004	0.020

When year omitted, samples either not obtained or analyses not performed.

¹Number in parentheses indicates total number of fish.

carbons in menhaden oil provided the basis for statistical studies. Among the many possible questions, the most critical to answer were: "What happened to the levels of chlorinated hydrocarbons during the period studied?" and "Were the levels in the four areas of the menhaden fishery distinguishable?"

In the comparison for differences among years using the *F*-test, the null hypothesis of equality could be rejected for ΣDDT in all areas (Table 14), for PCB's except in the South Atlantic States (Table 15), and for dieldrin except in the Middle Atlantic States (Table 16). Where the null hypothesis was rejected, in order to identify differing groups, the data were further tested by Scheffe's method for multiple comparison (*s*-method), because this method can be applied when the sample sizes are not equal and more is known about the behavior of data (Scheffe, 1959). Examples of our findings follow. For ΣDDT in menhaden oils from the Middle Atlantic States, there were significant differences ($P<0.05$) between 1969 and 1973, 1974, 1976, or 1977,

Table 13.—Endrin in menhaden oil by area (1969-77).

Area and year	No. of samples	Mean (ppm)	SD	Low (ppm)	High (ppm)
Middle Atlantic					
1969	6	< 0.04	0.004	< 0.04	< 0.05
1971	2	< 0.03	0		
1972	5	0.028	0.004	0.021	0.031
1973	5	0.028	0.010	< 0.02	0.045
1974	4	< 0.05	0.007	0.036	< 0.05
1975	6	0.021	0.005	0.016	0.031
1976	5	< 0.023	0.005	< 0.02	< 0.03
1977	6	0.018	0.008	0.013	0.033
Chesapeake Bay					
1969	2	< 0.05	0.014	< 0.04	< 0.06
1972	4	0.027	0.008	0.017	0.036
1973	3	0.040	0.029	0.023	0.074
1974	5	0.043	0.016	0.026	0.065
1975	5	0.023	0.009	0.013	0.032
1976	5	0.028	0.015	0.018	0.054
1977	5	0.016	0.006	0.010	0.025
South Atlantic					
1969	12	0.106	0.046	< 0.05	0.197
1971	7	< 0.031	0.002	< 0.03	0.035
1973	3	< 0.037	0.004	0.033	< 0.04
1974	2	0.028	0.005	0.025	0.032
1976	8	0.025	0.010	0.012	0.040
1977	10	0.015	0.005	0.009	0.027
Gulf of Mexico					
1969	2	0.100	0.071	< 0.05	0.150
1972	4	0.064	0.006	0.058	0.071
1973	9	0.076	0.032	< 0.03	0.121
1974	9	0.097	0.078	0.028	0.259
1975	14	0.074	0.037	0.017	0.147
1976	12	0.059	0.023	0.028	0.095
1977	18	0.049	0.041	< 0.01	0.127

When year omitted, samples either not obtained or analyses not performed.

¹ North Carolina oil, January 1972 production, considered part of New Jersey fishery because of distinctly different chlorinated hydrocarbon characteristics from North Carolina early fall fishery.

between 1971 and 1974, 1976, or 1977, and between 1972 and 1974, 1976, or 1977. There were differences in PCB levels in both the Chesapeake Bay and Gulf of Mexico areas between 1972 and 1973, 1974, 1976, and 1977. For menhaden oils collected from plants in Chesapeake Bay significant differences in dieldrin levels existed between 1969 and 1972, 1973, 1974, 1975, 1976, or 1977, between 1972 and 1974, 1975, 1976, or 1977, and between 1973 and 1976 or 1977. The details are shown in Table 17.

Comparison of data for differences among areas within a single year showed that ΣDDT in menhaden oils differed significantly ($P < 0.05$) in the years 1970 through 1973, but not in 1969 or 1974 through 1977. During all the years that PCB's were analyzed, samples of oils from the Middle Atlantic States differed from those from other areas. In 5 of 8 years, namely 1971, 1973, 1974, 1976, and 1977, dieldrin levels differed among areas. In 1971, 1973, and 1977, dieldrin levels in the Middle Atlantic area differed from those in the South Atlantic area. In 1973 the dieldrin level in the South Atlantic area also differed from that in the Gulf of Mexico area, and in 1976 the only difference was be-

Table 14.—Analysis of variance (ANOVA) tables for ΣDDT in menhaden oil among years within specific areas.

Source and area	Sample size	df	Sum of squares	Mean square	F-value
Middle Atlantic					
Years	7	6	118.911	19.818	21.701*
Within	25	25	22.832	0.913	
Total	32	31	141.742	4.572	
Chesapeake Bay					
Years	7	6	198.826	33.138	4.775*
Within	33	33	228.992	6.939	
Total	40	39	427.818	10.970	
South Atlantic					
Years	7	6	158.897	26.483	20.883*
Within	62	62	78.627	1.268	
Total	69	68	237.524	3.493	
Gulf of Mexico					
Years	7	6	48.924	8.154	13.201*
Within	82	82	50.651	0.618	
Total	89	88	99.574	1.132	

* $P < 0.05$.

Table 15.—ANOVA tables for PCB in menhaden oil among years within specific areas.

Source and area	Sample size	df	Sum of squares	Mean square	F-value
Middle Atlantic					
Years	6	5	323.960	64.792	5.615*
Within	21	21	242.308	11.538	
Total	27	26	566.268	21.780	
Chesapeake Bay					
Years	5	4	35.856	8.964	14.016*
Within	17	17	10.872	0.640	
Total	22	21	46.728	2.225	
South Atlantic					
Years	5	4	18.627	4.657	1.755*
Within	25	25	66.345	2.654	
Total	30	29	84.972	2.930	
Gulf of Mexico					
Years	5	4	18.015	4.504	18.459*
Within	48	48	11.711	0.244	
Total	53	52	29.726	0.572	

* $P < 0.05$.

Table 16.—ANOVA tables for dieldrin in menhaden oil among years within specific areas.

Source and area	Sample size	df	Sum of squares	Mean square	F-value
Middle Atlantic					
Years	8	7	4.272	0.610	1.953
Within	31	31	9.686	0.312	
Total	39	38	13.958	0.367	
Chesapeake Bay					
Years	7	6	0.437	0.073	45.364*
Within	22	22	0.035	0.002	
Total	29	28	0.473	0.017	
South Atlantic					
Years	6	5	3.114	0.623	11.263*
Within	36	36	1.991	0.055	
Total	42	41	5.105	0.125	
Gulf of Mexico					
Years	7	6	0.859	0.143	10.392*
Within	62	62	0.854	0.014	
Total	69	68	1.714	0.025	

* $P < 0.05$.

Table 17.—Test (*F*-test) of null hypothesis for differences in chlorinated hydrocarbon levels among years in menhaden oil collected from plants in four areas.

Area and year	Test of null hypothesis		
	ΣDDT	PCB	Dieldrin
Middle Atlantic			
1969	de	n.a.	n
1971	df	d	n
1972	dg	de	n
1973	de'	de'	n
1974	de'f'g'	d	n
1975	n.a.	n.a.	n
1976	de'f'g'	d	n
1977	de'f'g'	de'	n
Chesapeake Bay			
1969	ds	n.a.	de
1970	ds	n.a.	n.a.
1972	ds	de	de'f
1973	ds	de'	de'g
1974	ds	de'	de'f'
1975	n.a.	n.a.	de'f'
1976	ds	de'	de'f'g'
1977	ds	de'	de'f'g'
South Atlantic			
1969	de	n.a.	de
1970	df	n.a.	n.a.
1971	dg	n	d
1973	de'f'	n	de'
1974	de'f'	n	d
1976	de'f'g'	n	de'
1977	de'f'g'	n	de'
Gulf of Mexico			
1969	d	n.a.	de
1970	de	n.a.	n.a.
1972	d	de	d
1973	de'	de'	de
1974	de'	de'	de
1975	n.a.	n.a.	de
1976	de'	de'	d
1977	de'	de'	de'

When year omitted, samples either not obtained or analyses not performed.

n Cannot reject the null hypothesis of no difference among years (*F*-test), within a single area.

n.a. Not analyzed.

d Reject the null hypothesis of no difference among years, within a single area.

e Years marked "e" differ from years marked "e'", within a single area.

f Years marked "f" differ from years marked "f'", within a single area.

g Years marked "g" differ from years marked "g'", within a single area.

s Although the *F*-test rejected the hypothesis of equality of means for the Scheffe multiple comparison procedure, no confidence intervals for mean differences failed to contain the point zero.

tween those two areas. The details of the interarea comparisons are shown in Tables 18-21.

Examination of Tables 17 and 21 shows two instances in which the null hypothesis was rejected, but no groups differed. In the case of ΣDDT in menhaden oils collected from plants in the Chesapeake Bay area, although the *F*-test rejected the

Table 18.—ANOVA tables for ΣDDT in menhaden oil among areas for a specific year.

Source and area	Sample size	df	Sum of squares	Mean square	<i>F</i> -value
1969					
Areas	4	3	24.761	8.254	2.861
Within	18		51.936	2.885	
Total	22	21	76.696	3.652	
1970					
Areas	3	2	145.618	72.809	20.152*
Within	86		310.719	3.613	
Total	89	88	456.337	5.186	
1971					
Areas	2	1	4.679	4.679	22.760*
Within	7		1.439	0.206	
Total	9	8	6.118	0.765	
1972					
Areas	3	2	13.983	6.991	10.505*
Within	10		6.655	0.666	
Total	13	12	20.638	1.720	
1973					
Areas	4	3	3.152	1.051	5.380*
Within	16		3.125	0.195	
Total	20	19	6.277	0.330	
1974					
Areas	4	3	0.393	0.131	1.415
Within	16		1.483	0.093	
Total	20	19	1.876	0.099	
1976					
Areas	4	3	1.363	0.454	2.386
Within	17		3.238	0.190	
Total	21	20	4.601	0.230	
1977					
Areas	4	3	0.630	0.210	2.680
Within	32		2.507	0.078	
Total	36	35	3.137	0.090	

**P* < 0.05.

Table 19.—ANOVA tables for PCB in menhaden oil among areas for a specific year.

Source and area	Sample size	df	Sum of squares	Mean square	<i>F</i> -value
1971					
Areas	2	1	100.018	100.018	19.543*
Within	7		35.825	5.118	
Total	9	8	135.844	16.980	
1972					
Areas	3	2	385.654	192.827	30.265*
Within	10		63.713	6.371	
Total	13	12	449.367	37.447	
1973					
Areas	4	3	79.095	26.365	8.015*
Within	16		52.634	3.290	
Total	20	19	131.729	6.933	
1974					
Areas	4	3	108.790	36.263	10.066*
Within	16		57.639	3.602	
Total	20	19	166.430	8.759	
1976					
Areas	4	3	190.459	63.486	27.105*
Within	27		63.240	2.342	
Total	31	30	253.699	8.457	
1977					
Areas	4	3	108.006	36.002	21.656*
Within	35		58.185	1.662	
Total	39	38	166.191	4.373	

**P* < 0.05.

hypothesis of equality of means among years, for the Scheffe (1959) multiple comparison procedure, no confidence intervals for mean differences failed to contain the point zero (Table 17). This behavior of data is uncommon, but occurred in the test between areas for 1974 dieldrin data as well (Table 21).

Interpretation of Results

The levels of chlorinated hydrocarbons decreased in most types of samples during the period studied, but statistical analyses did not always confirm this observation. Several factors contributed to this discrepancy: 1) The number of samples of a specific type from a single area was sometimes extremely limited; 2) the number of samples varied widely from year to year; 3) marine fish from even a single catch vary widely in chlorinated hy-

drocarbon content (Stout and Beezhold, 1981); 4) the size and age distribution of menhaden catches fluctuated broadly, in response to conditions in the fishery; 5) Atlantic menhaden migrate a long distance, north in the spring and south in the fall (Dryfoos et al., 1973); 6) in later years, especially in the Atlantic fishery, fish contained exceptionally small amounts of oil; and 7) with the introduction of refrigeration, fishing vessels extended their range, so that previously discrete fishing areas lost some definition.

To explain in more detail, we collected a total of 268 samples of menhaden oil during the 9 years of the survey, but some years we collected no samples from a specific area because no fish appeared there. When fishing did occur, some years we received only two samples from an area. At the other extreme, in 1970 we collected more than 40 samples each of meal and oil from the Gulf of Mexico (Tables 3, 4). Furthermore, sometimes the fish were small, other times

Table 20.—ANOVA tables for dieldrin in menhaden oil among areas for a specific year.

Source and area	Sample size	df	Sum of squares	Mean square	F-value
1969					
Areas	4	3	0.417	0.139	0.312
Within		18	8.011	0.445	
Total	22	21	8.427	0.401	
1971					
Areas	2	1	0.392	0.392	16.966*
Within		7	0.162	0.023	
Total	9	8	0.554	0.069	
1972					
Areas	3	2	1.842	0.921	3.065
Within		10	3.006	0.301	
Total	13	12	4.848	0.404	
1973					
Areas	4	3	0.292	0.097	4.433*
Within		16	0.351	0.022	
Total	20	19	0.644	0.034	
1974					
Areas	4	3	0.168	0.056	4.121*
Within		16	0.218	0.014	
Total	20	19	0.386	0.020	
1975					
Areas	3	2	0.072	0.036	2.153
Within		22	0.370	0.017	
Total	25	24	0.443	0.018	
1976					
Areas	4	3	0.130	0.043	4.494*
Within		27	0.260	0.010	
Total	31	30	0.390	0.013	
1977					
Areas	4	3	0.065	0.022	4.030*
Within		35	0.188	0.005	
Total	39	38	0.253	0.007	

*P < 0.05.

they were very large. These were the normal fluctuations in the menhaden fishery. When fish were scarce, smaller fish, more commonly allowed to mature further, were taken for reduction. The chlorinated hydrocarbon content of samples was dependent on the age structure of the catches. Menhaden live in different environments at different stages of development. In the spring, maturing Atlantic menhaden, which constitute one single population (Dryfoos et al., 1973) migrate northward from an unknown wintering area offshore in the Atlantic. The youngest fish apparently spawn farthest south, in Florida, North Carolina, and Chesapeake Bay. Older fish travel as far as New Jersey, New York, and in some years even Massachusetts and Maine before spawning. Menhaden spawn just on the edge of the shore and the eggs

Table 21.—Test (F-test) of null hypothesis for differences in chlorinated hydrocarbon levels menhaden oils among areas.

Chlorinated hydrocarbon and year	Area			
	Middle Atlantic	Chesapeake Bay	South Atlantic	Gulf of Mexico
Σ DDT				
1969	n	n	n	n
1970	n.a.	de	de'f	de'f'
1971	de	n.a.	de'	n.a.
1972	de	df	n.a.	de'f'
1973	de	d	d	de'
1974	n	n	n	n
1976	n	n	n	n
1977	n	n	n	n
Polychlorinated biphenyls				
1971	de	n.a.	de'	n.a.
1972	de	de'	n.a.	de'
1973	de	d	d	de'
1974	de	de'	de'	de'
1976	de	de'	de'	de'
1977	de	de'	de'	de'
Dieldrin				
1969	n	n	n	n
1971	de	n.a.	de'	n.a.
1972	n	n	n	n
1973	de	d	de'f	df'
1974	ds	ds	ds	ds
1975	n	n	n	n
1976	d	d	de	de'
1977	de	d	de'	d

When year omitted, samples either not obtained or analyses not performed.

n Cannot reject the null hypothesis of no difference among areas (F-test), within a single year.

n.a. Not analyzed.

d Reject the null hypothesis of no difference among areas, within a single year.

e Areas marked "e" differ from areas marked "e'", within a single year.

f Areas marked "f" differ from areas marked "f'", within a single year.

s Although the F-test rejected the hypothesis of equality of means for the Scheffe multiple comparison procedure, no confidence intervals for mean differences failed to contain the point zero.

drift into estuaries to develop. Juvenile menhaden spend many months in the estuaries before migrating southward in the fall along with the older fish from farther north. Larger fish often contain higher concentrations of chlorinated hydrocarbons, whether because of longer exposure, greater pollution in more northerly waters or other factors is not known. When taken on their first southward migration, juvenile fish coming from highly polluted estuaries presumably also contain higher levels of contaminants than their surviving cohorts after wintering over in the cleaner environment of the open sea.

Unrelated to size and migration, the concentration of chlorinated hydrocarbons in the oil of a fish depends directly on the oil content of the animal. In other words, independent of the absolute amount of contaminant (total micrograms) present, the more oil in the fish the lower the concentration of chlorinated hydrocarbons (i.e., micrograms per gram oil). Once deposited in fish, these contaminants tend to remain there for life. The chlorinated hydrocarbons are very soluble in oil and nearly insoluble in water and proteins. In live animals these contaminants are associated with the fatty tissue. During reduction they concentrate in the oil and the oil remaining in the meal, stickwater, and solubles. Contrary to possible expectations, the oil content of menhaden varies markedly from year to year.

In the mid-1970's, for unknown reasons, Atlantic menhaden contained extremely small amounts of oil. The levels of chlorinated hydrocarbons in menhaden products, therefore, may have been deceptive. Anticipated decreases with restriction in use of chlorinated hydrocarbons may have been masked by the marked decrease in menhaden oil in this marine resource. More extensive sampling of fish would have eliminated most of these problems, but because of difficulties in handling fresh fish, they were not always collected. In spite of all these complexities in the sampling, both the means and the high values did decrease regularly in most types of samples from most areas, indicating that the amounts of chlorinated hydrocarbons to which menhaden were exposed were decreasing at least to a limited extent.

Conclusions

We monitored the levels of chlorinated hydrocarbons throughout the menhaden fisheries in both the Atlantic Ocean and the Gulf of Mexico from 1969, when DDT was still common, until 1977, when DDT had been banned and PCB's drastically curtailed. DDT and its metabolites

TDE and DDE, PCB's as well as dieldrin and endrin, were found in most samples of whole menhaden and the reduction products, meal, oil, solubles, and stickwater, from all areas of the fisheries. The very high levels found in the earlier years (a maximum of 12 ppm Σ DDT, 20 ppm PCB's and 3 ppm dieldrin in menhaden oil) declined sharply with increasing restriction in usage of chlorinated hydrocarbons. By 1977 the highest levels in the oil were 1.4 ppm Σ DDT, 9 ppm PCB's, and 0.4 ppm dieldrin. In 1977 the highest means, 1.0 ppm Σ DDT, 6 ppm PCB's, and 0.2 ppm dieldrin, occurred in the Middle Atlantic fishery. This fishery represents a very small proportion of the total catch. The mean levels of PCB's and dieldrin in the other three areas were much lower. Since the Middle Atlantic fishery involves the largest proportion of older fish, with time the levels in this area may drop as well. Both the means and the high values for chlorinated hydrocarbons decreased in most types of samples from most areas during the period of this study. Nonetheless statistical analyses, using the *F*-test and Scheffe's (1959) method for multiple comparison (*s*-method) on 541 data for menhaden oil, only partially confirmed these findings. Possible reasons for these observations include, among others, limitation and variation in sample size, variability of chlorinated hydrocarbon level in fish from a single catch, and the broad migration of menhaden, at least in the Atlantic fishery.

Over the years studied following the restriction in use of DDT, the proportion of Σ DDT present in samples as DDT itself decreased. The anaerobic metabolite of DDT that forms in the marine environment, TDE, increased during the same interval. The proportions of DDT, TDE, and DDE were generally similar in all types of samples except meal, which contained a greater proportion of DDE. Degradation of DDT to DDE,

which commonly occurs on land and in the freshwater environment, may be accelerated at the elevated temperatures of the meal drier.

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Literature Cited

- Aulerich, R. J., R. K. Ringer, and S. Iwamoto. 1973. Reproductive failure and mortality in mink fed on Great Lakes fish. *J. Reprod. Fert., Suppl.* 19, 365-376.
- Burdick, G. E., E. J. Harris, H. J. Dean, T. M. Walker, J. Skea, and D. Colby. 1964. The accumulation of DDT in lake trout and the effect on reproduction. *Trans. Am. Fish. Soc.* 93:127-136.
- Carson, R. L. 1962. *Silent spring*. Houghton-Mifflin Co., Boston, Mass., 368 p.
- Cecil, H. C., J. Bitman, G. F. Fries, L. W. Smith, and R. J. Lillie. 1972. PCB's in laying hens. *In* Symposium on PCB's - still prevalent - still persistent, p. 86-90. American Chemical Society, Wash., D. C.

- Dryfoos, R. L., R. P. Cheek, and R. L. Kroger. 1973. Preliminary analyses of Atlantic menhaden, *Brevoortia tyrannus*, migrations, population structure, survival and exploitation rates, and availability as indicated from tag returns. *Fish. Bull., U.S.* 71:719-734.
- Horowitz, W. (editor). 1970a. Official methods of analysis of the Association of Official Analytical Chemists. 11th ed. Assoc. Off. Anal. Chem., Wash., D.C. No. 29.002 and 29.009, p. 475.
- _____. 1970b. Official methods of analysis of the Association of Official Analytical Chemists. 11th ed. Assoc. Off. Anal. Chem., Wash., D.C. No. 29.010, p. 480.
- Kuratsune, M., T. Yoshimura, J. Matsuzaka, and A. Yamaguchi. 1972. Epidemiologic study on yusho, a poisoning caused by ingestion of rice oil contaminated with a commercial brand of polychlorinated biphenyls. *Environ. Health Perspec.* 1:119-128.
- O'Shea, T. J., W. J. Fleming III, and E. Cromartie. 1980. DDT contamination at Wheeler National Wildlife Refuge. *Science* 209: 509-510.
- Porter, M. L., S. J. V. Young, and J. A. Burke. 1970. A method for the analysis of fish, animal, and poultry tissue for chlorinated pesticide residues. *J. Assoc. Off. Anal. Chem.* 53:1300-1303.
- Reinert, R. E. 1970. Pesticide concentrations in Great Lakes fish. *Pestic. Monit. J.* 3:233-240.
- Robinson, L. A. (editor). 1977. Fisheries of the United States, 1976. U.S. Dep. Commer., NOAA, Natl. Mar. Fish. Serv. *Curr. Fish. Stat.* 7200, 96 p.
- Scheffe, H. 1959. *The analysis of variance*. John Wiley & Sons, Inc., N.Y., 447 p.
- Schmidt, A. M. 1974. Poisonous or deleterious substances in food. *Fed. Regist.* 39 (236): 42743-42748.
- Stansby, M. E. 1973. Problems discouraging use of fish oil in American-manufactured shortening and margarine. *J. Am. Oil Chem. Soc.* 50:220A-225A.
- Stout, V. F. 1978. Fate of dichlorodiphenyltrichloroethane and its metabolites during the preparation of fish protein concentrate. *J. Agric. Food Chem.* 26:610-612.
- _____. 1980. Organochlorine residues in fishes from the northwest Atlantic Ocean and Gulf of Mexico. *Fish. Bull., U.S.* 78:51-58.
- _____, and F. L. Beezhold. 1979. Analysis of chlorinated hydrocarbon pollutants: A simplified extraction and cleanup procedure for fishery products. *Fish. Bull., U.S.* 76:880-886.
- _____, 1981. Chlorinated hydrocarbon levels in fishes and shellfishes of the north-eastern Pacific Ocean, including the Hawaiian Islands. *Mar. Fish. Rev.* 43(1):1-12.
- _____, F. L. Beezhold, and C. R. Houle. 1970. DDT residue levels in some U.S. fishery products and some treatments in reducing them. *In* Mario Ruivo (editor) FAO Technical conference on marine pollution and its effects on living resources and fishing. FIR: MP/70/E-106, 8 p. (Also, condensed form *in* Mario Ruivo (editor), *Marine pollution and sea life*, p. 550-553. Fishing News (Books) Ltd., Surrey, Engl., 1972.