

# ORGANOCHLORINE RESIDUES IN FISHES FROM THE NORTHWEST ATLANTIC OCEAN AND GULF OF MEXICO

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## ABSTRACT

Residues of  $\Sigma$ DDT (*p,p'*-DDT and its metabolites *p,p'*-TDE and *p,p'*-DDE), PCB (polychlorinated biphenyls), dieldrin, and endrin were determined in the flesh of 700 specimens of fishes caught between 1973 and 1975 in the northwestern Atlantic Ocean and northern Gulf of Mexico off the coasts of the southeastern United States. Species with lowest oil content (<3%)—gag, *Mycteroperca microlepis*; black grouper, *M. bonaci*; red grouper, *Epinephelus morio*; and red snapper, *Lutjanus campechanus*—contained the least amounts of chlorinated hydrocarbon residues. Species with higher oil content—king mackerel, *Scomberomorus cavalla*, (3.5%) and Spanish mackerel, *S. maculatus*, (4.6%)—more consistently contained residues, but still at low levels. Significant correlations ( $P < 0.05$ ) were found in red snapper and king mackerel between lipid and size and between lipid and chlorinated hydrocarbon content. The correlations between lipid and PCB in gag and between lipid and DDT in black grouper were also significant. The highest mean values for any species were 0.18 ppm  $\Sigma$ DDT, 0.32 ppm PCB, 0.007 ppm dieldrin, and 0.008 ppm endrin. The highest level in any one composite sample of 10 fish was 1.0 ppm  $\Sigma$ DDT, 1.8 ppm PCB, 0.026 ppm dieldrin, and 0.026 ppm endrin.

Half a century ago manufacturers of surface coatings and of electrical equipment found a common interest in a newly introduced group of organochlorine chemicals, the PCB.<sup>2</sup> These compounds dissolve the inks in carbonless carbon paper, which duplicates without the use of carbon paper. They plasticize the waterproof coatings for dairy silos and fish tanks, and marine antifouling paint. The thermal and electrical properties of PCB are highly desirable in dielectric fluids, the electrical insulators in transformers and capacitors. The PCB are also highly resistant to chemical and biological degradation, and these properties, too, are valued by industrial users.

Immediately following World War II, another organochlorine chemical, DDT, became the magic tool of the medical profession, deeply concerned with preventing outbreaks of infectious, insect-borne diseases. Enormous quantities of DDT were used to prevent epidemics of typhus and plague in war-torn Europe. DDT was dramatically effective in controlling lice and fleas, which carried these

diseases. The use of this and related compounds spread rapidly for mosquito and agricultural pest control. Before insect resistance developed, DDT reduced the incidence of malaria to a very low level.

Since PCB and the organochlorine pesticides are not only stable but also easily dispersed in the air and through the water, it is not surprising in retrospect that they are now found even in the polar regions (Sladen et al. 1966; Risebrough et al. 1976; Bowes and Jonkel 1975) and that chlorinated hydrocarbon pollution has become a worldwide problem. Not until the 1960's did appreciation of the adverse effects of these chlorinated hydrocarbons begin to develop. "Silent Spring" (Carson 1962) described the effects on the environment of the accumulation of DDT, and Jensen (Anonymous 1966; Jensen et al. 1969) found PCB in marine animals. Burdick et al. (1964) noted reproductive failure in lake trout when the eggs contained a high level of  $\Sigma$ DDT. Aulerich et al. (1973) traced the reproductive failure and mortality in ranch-grown mink back to coho salmon, *Oncorhynchus kisutch*, from Lake Michigan and ultimately to the PCB they contained. Only recently fin erosion, a disease associated with municipal and industrial discharges, was related to  $\Sigma$ DDT (Mearns and Sherwood 1977). Montrose Chemical Company released wastes from DDT manufacture directly into

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<sup>2</sup>Abbreviations used in this manuscript: DDE = *p,p'*-dichlorodiphenyldichloroethylene; DDMU = *p,p'*-dichlorodiphenylchloroethylene; DDT = *p,p'*-dichlorodiphenyltrichloroethane; PCB = polychlorinated biphenyls;  $\Sigma$ DDT = DDT and its metabolites DDE and TDE; TDE = *p,p'*-dichlorodiphenyldichloroethane.

## METHODS

the Los Angeles County sewer system which flows via sewer outfalls into the marine environment of southern California. From this unanticipated source of  $\Sigma$ DDT pollution, high levels of  $\Sigma$ DDT developed throughout the southern California marine environment from mollusks (Young et al. 1976), crustaceans (Burnett 1971), and fishes (McDermott-Ehrlich et al. 1978) to marine birds (Anderson et al. 1975) and cetaceans (Le Boeuf and Bonnell 1971). Henderson et al. (1969, 1971) found organochlorine residues in freshwater fishes throughout the United States. Some species of freshwater fishes, especially those from the Great Lakes, contained levels of chlorinated hydrocarbons that exceeded the U.S. Food and Drug Administration guidelines (Reinert 1970; Veith 1975). In recent years, the use of chlorinated hydrocarbons has been drastically curtailed, but concern remains about the continuing occurrence of these toxic compounds in the marine environment.

Fishes and shellfishes are excellent organisms for monitoring chlorinated hydrocarbons. Shellfishes have been used as indicators of short-term pollution (Butler 1973; Goldberg et al. 1978) because they accumulate and depurate these substances readily. Fishes, on the other hand, reflect long-term exposure since they lose chlorinated hydrocarbons slowly, if at all (Lieb et al. 1974). Butler and Schutzmann (1978) have reported on pesticides and PCB in yearling estuarine fishes of the United States. Outside of their study, however, few data on fishes for human consumption from the western Atlantic Ocean have been published except on fishes from Canadian waters (Sims et al. 1977). The study reported here was undertaken to obtain information about levels of  $\Sigma$ DDT, PCB, dieldrin, and endrin in fillets from fishes caught in the northwestern Atlantic Ocean and northern Gulf of Mexico. Six marine species of both commercial and sport value have been examined.

Samples of gag, *Mycteroperca microlepis*; black grouper, *M. bonaci*; red grouper, *Epinephelus morio*; red snapper, *Lutjanus campechanus*; king mackerel, *Scomberomorus cavalla*; and Spanish mackerel, *S. maculatus*, were collected from the northwestern Atlantic Ocean and northern Gulf of Mexico, from Beaufort, N.C., south to the Florida Keys, and west to Aransas Pass, Tex. Sampling occurred between October 1973 and October 1975, but mainly in 1975. Specimens were frozen and held at  $-18^{\circ}$  C. They were thawed for filleting, grinding, compositing, and refrozen until analysis. In the aggregate, 70 samples each containing 10 fish of the same species and of similar size were obtained. Each sample was a composite of equal weights of ground skinless fillets from the 10 individual fish. At most sites, two or three samples from different size fish were selected. The collection sites, size, and lipid content of the specimens are listed in Table 1.

Extracts were prepared by the procedures of Reinert (1970). Samples for  $\Sigma$ DDT and PCB analysis were extracted with propanol-2/benzene (1:1), and the extracted materials transferred to hexane by repeated codistillation of the propanol-2, benzene, and water with hexane. One aliquot of the hexane extract was evaporated to minimum weight for determination of the lipid content. Another aliquot was cleaned up on Florisil.<sup>3</sup> PCB were separated from TDE, DDT, and most of the DDE on activated silica gel (Snyder and Reinert 1971), which also separates the interfering hydrocarbons, phenanthrene, fluoranthrene, and pyrene from the PCB (Zitko 1978). At least 90% of the PCB was contained in the pentane fraction, which also contained part of

<sup>3</sup>Mention of specific products or companies in this paper does not imply endorsement by the National Marine Fisheries Service, NOAA.

TABLE 1.—Collection sites, size, and lipid content of fishes from the northwest Atlantic Ocean and Gulf of Mexico.

Species	Sites <sup>1</sup>	No. <sup>2</sup>	Fork length (cm)		Weight (kg)		Lipid content (%)	
			Mean	Range <sup>3</sup>	Mean	Range <sup>3</sup>	Mean	Range <sup>3</sup>
Gag	1, 2, 3	8	86	64-108	9.77	3.79-18.32	2.9	0.2-5.9
Black grouper	5, 6	6	77	54- 96	7.01	1.98-12.70	0.6	0.2-1.2
Red grouper	1, 4, 5, 6	10	68	52- 82	6.08	2.42-11.50	0.5	0.1-1.0
Red snapper	1, 2, 3, 4, 5, 6, 7, 8	18	65	45- 83	5.94	1.65-11.35	1.5	0.4-3.9
King mackerel	1, 2, 3, 4, 5, 6, 9	18	87	55-119	6.01	1.20-12.91	3.5	0.4-7.5
Spanish mackerel	3, 4, 5, 6, 7	10	54	45- 64	1.41	0.48- 2.34	4.6	1.7-9.4

<sup>1</sup>1 - Beaufort, N.C.; 2 - Savannah, Ga.; 3 - Florida, East Coast; 4 - Florida Keys; 5 - Tampa/St. Petersburg, Fla.; 6 - Panama City, Fla.;

<sup>2</sup>7 - Mobile, Ala.; 8 - Pascagoula, Miss.; 9 - Aransas Pass, Tex.

<sup>3</sup>Number of composites, each consisting of 10 fish.

<sup>3</sup>Range of means of individual composites.

the DDE. All of the TDE and DDT eluted into the benzene fraction. For dieldrin and endrin analysis, tissues were saponified with KOH in aqueous ethanol, extracted with hexane, and cleaned up on Florisil. Since the specimens, originally collected for trace-metal analysis, were stored in polypropylene containers (Falcon No. 4014) with polyethylene lids (Falcon No. 4017), the containers and lids were also analyzed to assure freedom from interfering substances. The details of our procedures were published previously (Stout and Beezhold 1979). Blanks carried through the whole procedure for either PCB and  $\Sigma$ DDT or dieldrin and endrin showed no chromatographic peaks which interfered with quantitation of the chlorinated hydrocarbons.

The extracts of the fishes were quantitated by electron-capture gas chromatography. A Varian 600 C gas chromatograph with a titanium tritide detector was fitted with a 1.5 m (5 ft)  $\times$  0.32 cm (0.125 in) o.d. glass column containing a mixture of equal parts of 15% QF-1 on 80-100 mesh Gas Chrom Q and 10% DC-200 on the same support (Burke and Holswade 1966). Reference standards of *p,p'*-DDE, *p,p'*-DDMU, *p,p'*-TDE, *p,p'*-DDT, dieldrin, and endrin were obtained from the U.S. Environmental Protection Agency, Health Effects Research Laboratory, Research Triangle Park, N.C. Aroclor 1254 obtained from the Monsanto Company, St. Louis, Mo., was the standard for the PCB because the residues in the fishes matched this Aroclor most closely. Standard curves of peak height versus concentration were used to determine the concentrations of components in the extracts. The sensitivity throughout each run was assured by frequent injections of standard solutions. The quantifiable limit was about 0.003 ppm for DDE, TDE, DDT, dieldrin, and endrin, and about 0.04 ppm for PCB depending on daily sensitivity. The mean relative standard deviation for samples analyzed in duplicate was 11%. The average recovery of samples spiked with standards was 85%. The values reported were not corrected for recovery. Residue values were calculated on the basis of micrograms chlorinated hydrocarbon per gram wet tissue or parts per million (ppm).

The PCB were quantitated by summing the peak heights corresponding to the five major peaks in Aroclor 1254 after omitting the twin peak with a retention time similar to that of *p,p'*-DDE. As Veith (1975) also concluded, use of five peaks increased the accuracy of PCB measurement by reducing the effect of minor variations in concen-

tration of individual components in the samples (Figure 1).

Since part of the DDE eluted from silica gel with the PCB fraction and one of the major peaks in Aroclor 1254 overlapped the DDE peak in the gas-chromatographic traces, a special technique was needed to quantitate the DDE in the PCB fraction. First the gross DDE concentration was determined in the usual way from the peak height versus DDE concentration curve. Next, the contribution of PCB to that overlapping peak was calculated based on the assumption that the height of Aroclor peak 3, the peak which overlapped DDE, was proportional to the heights of the five PCB peaks used to calculate the concentration of PCB. To make this calculation, the sum of the peak heights of the five major PCB peaks excluding the "DDE" peak was plotted against the peak height of the "DDE" peak in PCB standards of increasing concentrations. From the sum of the five PCB peaks in the sample, the peak height of the PCB portion of peak 3 in that sample was determined. This peak height was converted to concentration of DDE via the peak height versus concentration curve for DDE. The apparent concentration of DDE from PCB peak 3 was subtracted from the gross DDE concentration in the "DDE" peak to obtain the net concentration of DDE in the PCB fraction. The electron-capture detector is so much more sensitive to DDE than PCB that this method of calculation affected the accuracy of DDE determination only to a small extent (Figure 1). Use of a minicomputer expedited these calculations.

Confirmation of DDT and its metabolites and PCB was effected by saponifying separate portions of tissue (Reinert 1970). DDT is dehydrochlorinated by base to DDE, and TDE similarly to DDMU. PCB are stable to base. The levels of dieldrin and endrin were too low to warrant confirmation studies.

## RESULTS AND DISCUSSION

The marine fishes from the northwestern Atlantic Ocean and northern Gulf of Mexico analyzed in this study contained relatively low levels of  $\Sigma$ DDT and PCB. Of the 70 composite samples, only 29 contained more than 0.05 ppm  $\Sigma$ DDT and 0.1 ppm PCB in the edible portion (skinless fillets), and only one as much as 1 ppm  $\Sigma$ DDT and PCB. Red grouper contained the lowest levels of both chlorinated hydrocarbons. The mean  $\Sigma$ DDT content for

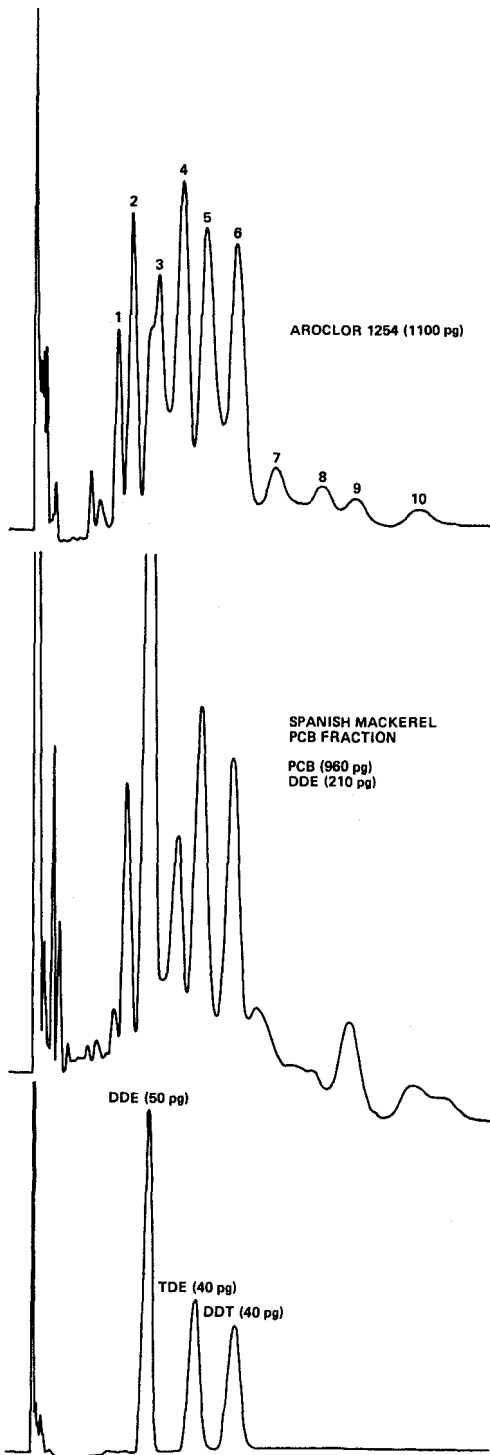


FIGURE 1.—Gas chromatographic curves of Aroclor 1254, the PCB fraction of an extract from Spanish mackerel, and DDT and its metabolites DDE and TDE; pg = picograms or  $10^{-12}$  g.

10 sets of red grouper was 0.008 ppm; for most samples, PCB were not detectable. Black grouper and gag contained slightly higher levels of  $\Sigma$ DDT and PCB. Red snapper also contained little  $\Sigma$ DDT (mean 0.039 ppm), but the PCB level in six samples exceeded 0.1 ppm. Only the two species of mackerel consistently contained quantifiable amounts of both  $\Sigma$ DDT and PCB. The mean levels of  $\Sigma$ DDT were 0.144 ppm in Spanish mackerel and 0.177 ppm in king mackerel. The mean PCB level in both species of mackerel was 0.32 ppm. The highest levels of both chlorinated hydrocarbons were found in one composite sample of king mackerel from the Florida Keys, 0.996 ppm  $\Sigma$ DDT and 1.8 ppm PCB. The data are summarized in Table 2.

The limited data in the literature convey a similar picture. Groupers of the genera *Epinephelus* and *Mycteroperca* from the Gulf of Mexico and the Grand Bahamas contained 0-0.10 ppm  $\Sigma$ DDT and 0.003-0.22 ppm PCB in muscle (Giam et al. 1974). Red snapper from Mobile Bay, Ala., contained 0.086 ppm  $\Sigma$ DDT and 0.14 ppm PCB in the whole animal; gray snapper, *Lutjanus griseus*, from Jacksonville, Fla., 0.007 ppm  $\Sigma$ DDT and no PCB in the whole animal (Markin et al. 1974). Small Spanish mackerel (306 g) from the Savannah River estuary in Georgia contained neither  $\Sigma$ DDT nor PCB in muscle. (Butler<sup>4</sup>) Although somewhat larger than those fish, the smallest fish in the current study (475 g) contained barely detectable amounts of these substances (0.008 ppm  $\Sigma$ DDT and 0.034 ppm PCB). Markin et al. (1974) found 0.04-0.16 ppm  $\Sigma$ DDT and <0.01-0.18 ppm PCB in seven whole Spanish mackerel from the southeastern United States. A single sample of king mackerel muscle from the Gulf of Mexico off Mexico contained 0.024 ppm  $\Sigma$ DDT and 0.034 ppm PCB (Giam et al. 1972). Atlantic mackerel, *Scomber scombrus*, collected in 1971-72 in Canadian waters (Sims et al. 1977) contained more  $\Sigma$ DDT (0.26 ppm) and PCB (0.41 ppm) in the "edible portion" (which may have contained skin and/or bones) than did skinless fillets of either species of mackerel examined in my study. On the other hand, muscle from Atlantic mackerel from the Bay of Fundy-Gulf of Maine contained the same level of PCB (0.35 ppm) (Zitko et al. 1972).

The proportions of *p,p'*-DDT and its metabolites were very similar in the king and Spanish mackerel examined in our study. *p,p'*-DDE is the

<sup>4</sup>Butler, P. A. 1978. EPA-NOAA Cooperative Estuarine Monitoring Program, Final Report, Gulf Breeze, Fla., 108 p.

TABLE 2.— $\Sigma$ DDT and PCB in fishes from the northwest Atlantic Ocean and Gulf of Mexico. nq = not quantifiable; nd = none detected.

Species	No. <sup>1</sup>	$\Sigma$ DDT (ppm)		PCB (ppm)	
		Mean $\pm$ SD	Range	Mean $\pm$ SD	Range
Gag	8	0.036 $\pm$ 0.013	0.017-0.050	0.087 $\pm$ 0.023	nq-0.129
Black grouper	6	0.009 $\pm$ 0.007	0.003-0.020	nq	nq-0.059
Red grouper	10	0.008 $\pm$ 0.007	n.d.-0.025	nd	nd-nq
Red snapper	18	0.039 $\pm$ 0.076	n.d.-0.322	0.121 $\pm$ 0.108	nd-0.464
King mackerel	18	0.177 $\pm$ 0.239	0.009-0.996	0.322 $\pm$ 0.414	0.060-1.78
Spanish mackerel	10	0.144 $\pm$ 0.097	0.008-0.319	0.319 $\pm$ 0.263	0.034-0.821

<sup>1</sup>Number of composites, each consisting of 10 fish.

major component (~65%) accompanied by about 25% of the parent compound and 10% *p,p'*-TDE. Although the composite picture for red snapper looked markedly different (Table 3), in fact the DDT residues in that species actually fell into two categories. In the first group, *p,p'*-TDE and *p,p'*-DDT were not quantifiable, and the maximum content of *p,p'*-DDE was 0.029 ppm. In the second group, five samples containing >0.029 ppm *p,p'*-DDE, both *p,p'*-TDE and *p,p'*-DDT were quantifiable. In those five samples, the proportions of the three components were the same as in the mackerel. Finfish from the Atlantic coast of Canada (Sims et al. 1977) contained proportionately much less *p,p'*-DDE (45%) and more *p,p'*-DDT (40%) and *p,p'*-TDE (15%). The increase in the proportion of *p,p'*-DDE in the present samples may reflect degradation of the parent compound in the environment before it accumulated in the fishes. Samples for the Canadian study were collected in 1971 and 1972, soon after usage of DDT had been drastically curtailed (around 1970) as the result of increasing insect resistance, problems with indirect contamination of foodstuffs, and concern about effects of DDT on nontarget species. Several years elapsed before the samples for the present study were collected, mainly in 1975. In the interval, DDT was degrading aerobically to DDE and anaerobically in the marine environment to TDE. Alternatively, DDT may metabolize more rapidly to DDE in the more temperate climate of the region studied and somewhat less rapidly to TDE.

TABLE 3.—Mean proportions of  $\Sigma$ DDT present as *p,p'*-DDE, *p,p'*-TDE, and *p,p'*-DDT in fishes from the northwest Atlantic Ocean and Gulf of Mexico.

Species	<i>p,p'</i> -DDE (%)		<i>p,p'</i> -TDE (%)		<i>p,p'</i> -DDT (%)	
	Mean $\pm$ SD	Range	Mean $\pm$ SD	Range	Mean $\pm$ SD	Range
Red snapper, all	88 $\pm$ 18	58-100	3 $\pm$ 5	0-15	9 $\pm$ 13	0-30
Red snapper <sup>1</sup>	62 $\pm$ 6	58-72	10 $\pm$ 3	7-15	28 $\pm$ 4	21-32
King mackerel	64 $\pm$ 8	48-77	8 $\pm$ 6	0-17	28 $\pm$ 6	16-40
Spanish mackerel	66 $\pm$ 19	48-100	12 $\pm$ 8	0-24	21 $\pm$ 12	0-37

<sup>1</sup>Five samples which contained >0.029 ppm DDE.

The concentration of PCB, when present, was higher than that of  $\Sigma$ DDT in all samples but one. The mean ratio of PCB to  $\Sigma$ DDT was 1.8 for gag, 2.2 for king and Spanish mackerel, and 2.6 for six sets of red snapper. In the two other samples of red snapper with quantifiable levels of PCB, the PCB/ $\Sigma$ DDT ratios were 22.7 and 24.3. The concentrations of  $\Sigma$ DDT were below 0.01 ppm in both cases. The one sample of red snapper in which the PCB/ $\Sigma$ DDT ratio was below 1 contained a relatively large amount of  $\Sigma$ DDT, 0.096 ppm, the second highest  $\Sigma$ DDT value in the 18 samples of red snapper. In contrast, the PCB concentration was low both in absolute amount, <0.06 ppm, and in rank, 14th out of 18 samples.

The chlorinated-hydrocarbon content of the specimens in this study was, in general terms, directly related to the lipid content. The groupers, which contained <1% lipid, contained the least  $\Sigma$ DDT and PCB. King and Spanish mackerel had the highest lipid contents, 3.5 and 4.6%, respectively, and the highest levels of both  $\Sigma$ DDT and PCB. In three of the six species, the correlation between lipid and  $\Sigma$ DDT was significant, i.e.,  $P < 0.05$ . Similarly, in three of the four species for which PCB were quantifiable the correlation between lipid and PCB was significant. Possible relationships between size and chlorinated hydrocarbon content were also examined. Although length, weight,  $\Sigma$ DDT, and PCB were all studied, in no case was a significant correlation found in any of the six species (Table 4). In two of the six species, the correlations between length and lipid and also between weight and lipid were significant. In both species, red snapper and king mackerel, the  $P$  values for lipid versus chlorinated hydrocarbon, were below 0.01. Giam et al. (1974) noted a relationship between size and concentration of pollutants in groupers from the Gulf of Mexico, but only in the area with the highest contamination, i.e., up to 0.1 ppm.

$\Sigma$ DDT and PCB levels within single species were compared at the various sites. Fish from the

TABLE 4.—Sample correlation and coefficients (R) and their significance (P) between body dimensions, lipids, and organochlorine residues in fishes from the northwest Atlantic Ocean and Gulf of Mexico.

Species	No. <sup>1</sup>	Length vs. lipid		Weight vs. lipid		Length vs. $\Sigma$ DDT		Weight vs. $\Sigma$ DDT		Lipid vs. $\Sigma$ DDT		Length vs. PCB		Weight vs. PCB		Lipid vs. PCB	
		R	P	R	P	R	P	R	P	R	P	R	P	R	P	R	P
Gag	8	-0.369	0.368	-0.317	0.444	0.189	0.653	0.232	0.580	0.552	0.156	0.085	0.842	0.117	0.782	0.755	0.030
Black grouper	6	0.102	0.847	0.056	0.917	0.426	0.400	0.356	0.489	0.896	0.016	( <sup>2</sup> )	( <sup>2</sup> )	( <sup>2</sup> )	( <sup>2</sup> )	( <sup>2</sup> )	( <sup>2</sup> )
Red grouper	10	0.536	0.110	0.595	0.070	0.576	0.082	0.511	0.132	0.256	0.476	( <sup>2</sup> )	( <sup>2</sup> )	( <sup>2</sup> )	( <sup>2</sup> )	( <sup>2</sup> )	( <sup>2</sup> )
Red snapper	18	0.571	0.013	0.663	0.003	0.323	0.191	0.438	0.069	0.668	0.002	0.296	0.233	0.374	0.126	0.718	0.001
King mackerel	18	0.575	0.013	0.598	0.009	0.357	0.146	0.333	0.176	0.722	0.001	0.344	0.162	0.333	0.177	0.694	0.001
Spanish mackerel	10	-0.035	0.923	-0.117	0.747	0.468	0.173	0.417	0.231	0.159	0.661	0.214	0.552	0.209	0.562	0.306	0.390

<sup>1</sup>Number of composites, each consisting of 10 fish.  
<sup>2</sup>DDE only, because IDE and DDT not always quantifiable.  
<sup>3</sup>PCB not quantifiable.

Florida Keys contained the greatest amounts of both substances, indicating that contamination from agricultural and domestic effluents was greatest in south Florida, an area of intense agriculture and relatively dense population as well. Fish caught in the vicinity of Beaufort and Mobile contained slightly less  $\Sigma$ DDT and PCB. Fish from the other sites contained lower levels of chlorinated hydrocarbons, which were not distinguishable by site, except that only DDE was quantifiable in the one sample from Pascagoula, Miss. The Mississippi River, which receives the runoff from 40% of the land mass of the conterminous United States, including the corn belt and the cotton belt, did not seem to be the main source of either  $\Sigma$ DDT or PCB.

Low levels of dieldrin and endrin, two highly toxic organochlorine pesticides, were found in the fish included in this study. The highest concentration of either compound was 0.026 ppm. The dieldrin and endrin content of only the three species with the highest levels of  $\Sigma$ DDT and PCB were determined. Nonetheless, dieldrin and endrin were quantifiable only in about one-third of the samples. Two of 18 red snapper, 7 of 18 king mackerel, and 6 of 10 Spanish mackerel samples contained quantifiable amounts of both compounds. The mean levels for Spanish mackerel, the species with the highest mean concentrations, were 0.007 ppm dieldrin and 0.008 ppm endrin (Table 5). King mackerel from Aransas Pass contained the highest concentration of dieldrin, 0.026 ppm; Spanish mackerel from Panama City, Fla., the highest concentration of endrin, also 0.026 ppm. The dieldrin and endrin concentrations in the three species followed the same distribution patterns with relation to species and lipid content as was observed for  $\Sigma$ DDT and PCB. Butler (see footnote 4) found no dieldrin in the muscle of small Spanish mackerel from the Savannah River estuary.

### CONCLUSIONS

Residues of  $\Sigma$ DDT, PCB, dieldrin, and endrin, although generally low, were found in all species and all locations except Pascagoula, Miss., where only DDE was quantifiable. The highest levels of  $\Sigma$ DDT and PCB, and the only ones to reach 1 ppm, were in one composite sample of king mackerel from the Florida Keys, 0.996 ppm  $\Sigma$ DDT and 1.8 ppm PCB. The highest level of dieldrin, 0.026 ppm, was in king mackerel from Aransas Pass, Tex., and of endrin, also 0.026 ppm, in Spanish mack-

TABLE 5.—Dieldrin and endrin in fishes from the northwest Atlantic Ocean and Gulf of Mexico. nd = not detected; nq = not quantifiable.

Species	No. <sup>1</sup>	Dieldrin (ppm)		Endrin (ppm)	
		Mean±SD	Range	Mean±SD	Range
Red snapper	18	nd	nd-nq	nd	nd-0.003
King mackerel	18	0.005±0.006	nd-0.026	0.004±0.004	nd-0.014
Spanish mackerel	10	0.007±0.004	nd-0.014	0.008±0.010	nd-0.026

<sup>1</sup>Number of composites, each consisting of 10 fish.

erel from Panama City, Fla. The species with the highest lipid contents contained the highest concentrations of chlorinated hydrocarbons. Significant correlations ( $P < 0.05$ ) were found between lipid and size and between lipid and chlorinated hydrocarbon content in two of the six species. In two other species the correlation between lipid and either  $\Sigma$ DDT or PCB was significant. In all cases but one, the chlorinated hydrocarbon levels were substantially below the U.S. Food and Drug Administration guidelines: 5 ppm  $\Sigma$ DDT, 2 ppm PCB, and 0.3 ppm dieldrin and endrin.

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## LITERATURE CITED

- ANDERSON, D. W., J. R. JEHL, JR., R. W. RISEBROUGH, L. A. WOODS, JR., L. R. DEWEESE, AND W. G. EDGEComb.  
1975. Brown pelicans: Improved reproduction off the southern California coast. *Science* (Wash., D.C.) 190:806-808.
- ANONYMOUS.  
1966. Report of a new chemical hazard. *New Sci.* 32:612.
- 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.
- BOWES, G. W., AND C. J. JONKEL.  
1975. Presence and distribution of polychlorinated biphenyls (PCB) in arctic and subarctic marine food chains. *J. Fish. Res. Board Can.* 32:2111-2123.
- 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.
- BURKE, J. A., AND W. HOLSWADE.  
1966. A gas chromatographic column for pesticide residue analysis: Retention times and response data. *J. Assoc. Off. Anal. Chem.* 49:374-388.
- BURNETT, R.  
1971. DDT residues: Distribution of concentrations in *Emerita analoga* (Stimpson) along coastal California. *Science* (Wash., D.C.) 174:606-608.
- BUTLER, P. A.  
1973. Organochlorine residues in estuarine mollusks, 1965-1972—National pesticide monitoring program. *Pestic. Monit. J.* 6:238-362.
- BUTLER, P. A., AND R. L. SCHUTZMANN.  
1978. Residues of pesticides and PCBs in estuarine fish, 1972-1976—National pesticide monitoring program. *Pestic. Monit. J.* 12:51-59.
- CARSON, R.  
1962. *Silent spring*. Houghton-Mifflin Co., Boston, Mass., 368 p.
- GIAM, C. S., A. R. HANKS, R. L. RICHARDSON, W. M. SACKETT, AND M. K. WONG.  
1972. DDT, DDE, and polychlorinated biphenyls in biota from the Gulf of Mexico and Caribbean Sea—1971. *Pestic. Monit. J.* 6:139-143.
- GIAM, C. S., R. L. RICHARDSON, D. TAYLOR, AND M. K. WONG.  
1974. DDT, DDE and PCBs in the tissues of reef dwelling groupers (Serranidae) in the Gulf of Mexico and the Grand Bahamas. *Bull. Environ. Contam. Toxicol.* 11:189-192.
- GOLDBERG, E. D., V. T. BOWEN, J. W. FARRINGTON, G. HARVEY, J. H. MARTIN, P. L. PARKER, R. W. RISEBROUGH, W. ROBERTSON, E. SCHNEIDER, AND E. GAMBLE.  
1978. The mussel watch. *Environ. Conserv.* 5:101-125.
- HENDERSON, C., A. INGLIS, AND W. L. JOHNSON.  
1971. Organochlorine insecticide residues in fish—Fall 1969 National pesticide monitoring program. *Pestic. Monit. J.* 5:1-11.
- HENDERSON, C., W. L. JOHNSON, AND A. INGLIS.  
1969. Organochlorine insecticide residues in fish (National pesticide monitoring program). *Pestic. Monit. J.* 3:145-171.
- JENSEN, S., A. G. JOHNELS, M. OLSSON, AND G. OTTERLIND.  
1969. DDT and PCB in marine animals from Swedish waters. *Nature* (Lond.) 224:247-250.
- LEBOEUF, B. J., AND M. L. BONNELL.  
1971. DDT in California sea lions. *Nature* (Lond.) 234:108-110.
- LIEB, A. J., D. D. BILLS, AND R. O. SINNHUBER.  
1974. Accumulation of dietary polychlorinated biphenyls (Aroclor 1254) by rainbow trout (*Salmo gairdneri*). *J. Agric. Food Chem.* 22:638-642.
- MARKIN, G. P., J. C. HAWTHORNE, H. L. COLLINS, AND J. H. FORD.  
1974. Levels of mirex and some other organochlorine residues in seafood from Atlantic and Gulf coastal states. *Pestic. Monit. J.* 7:139-143.
- MCDERMOTT-EHRlich, D., D. R. YOUNG, AND T. C. HEESSEN.  
1978. DDT and PCB in flatfish around southern California municipal outfalls. *Chemosphere* 6, p. 453-461.
- MEARNS, A. J., AND M. J. SHERWOOD.  
1977. Distribution of neoplasms and other diseases in marine fishes relative to the discharge of waste water. *Ann. N.Y. Acad. Sci.* 298:210-224.
- REINERT, R. E.  
1970. Pesticide concentrations in Great Lakes fish. *Pestic. Monit. J.* 3:233-240.
- RISEBROUGH, R. W., W. WALKER II, T. T. SCHMIDT, B. W. DE LAPPE, AND C. W. CONNORS.  
1976. Transfer of chlorinated biphenyls to Antarctica. *Nature* (Lond.) 264:738-739.

- SIMS, G. G., J. R. CAMPBELL, F. ZEMLYAK, AND J. M. GRAHAM.  
1977. Organochlorine residues in fish and fishery products from the Northwest Atlantic. *Bull. Environ. Contam. Toxicol.* 18:697-705.
- SLADEN, W. J. L., C. M. MENZIE, AND W. L. REICHEL.  
1966. DDT residues in Adelie penguins and a crabeater seal from Antarctica. *Nature (Lond.)* 210:670-673.
- SNYDER, D., AND R. REINERT.  
1971. Rapid separation of polychlorinated biphenyls from DDT and its analogues on silica gel. *Bull. Environ. Contam. Toxicol.* 6:385-390.
- STOUT, V. F., 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.
- VEITH, G. D.  
1975. Baseline concentrations of polychlorinated biphenyls and DDT in Lake Michigan fish, 1971. *Pestic. Monit. J.* 9:21-29.
- YOUNG, D. R., D. J. MCDERMOTT, AND T. C. HEESEN.  
1976. DDT in sediments and organisms around southern California outfalls. *J. Water Pollut. Control Fed.* 48:1919-1928.
- ZITKO, V.  
1978. The interference of aromatic hydrocarbons in the determination of PCB's. *Proceedings, 4th Joint Conference on Sensing of Environmental Pollutants, American Chemical Society, Pap. 201, p. 757-760.*
- ZITKO, V., O. HUTZINGER, AND P. M. K. CHOI.  
1972. Contamination of the Bay of Fundy—Gulf of Maine area with polychlorinated biphenyls, polychlorinated terphenyls, chlorinated dibenzodioxins, and dibenzofurans. *Environ. Health Perspect.* 1:47-50.