

## Organochlorine Residues in Animals from Three Louisiana Watersheds in 1978 and 1979

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Environmental contamination by various chemicals is a problem of constant occurrence. In order to determine the levels of harmful chemicals in the environment, monitoring programs have been set up throughout the United States. In 1964, the National Pesticide Monitoring Program was set up to include sites in Louisiana (such as watersheds) to assess the presence of organochlorine compounds including insecticides and their metabolites, and polychlorinated biphenyls (PCBs). The presence of organochlorines has been reported previously for Louisiana in preliminary residue sampling studies of the 1960's (Epps et al. 1967), in river drainage areas (Schmitt et al. 1981), and in fish after extensive sampling efforts (Butler and Schutzmann 1978). This study, in cooperation with Patuxent Wildlife Research Center (USDI) in Laurel, Maryland, was designed to continue monitoring of organochlorine residues in Louisiana wildlife and fish.

Characteristically, organochlorines may concentrate in certain animals due to their feeding habits or physiological makeup (such as lipid content) (see Bulkley et al. (1981) for discussion); hence these animals are often sampled as indicators of organochlorine contamination. However, animals monitored as indicator species are often not used for human consumption. Therefore, the animals selected for this study were chosen both to reflect immediate risks to humans, and to be useful indicators of environmental contamination through biological magnification. Collection sites for the different specimens reflected different insecticide use areas, and therefore different contamination For example, the East Franklin site is located in an area of formerly heavy organochlorine insecticide use and has been previously reported to have relatively high levels of contamination of fish. The Bayou Plaquemine-Brule site was an area of intermediate insecticide use, and the Lake Verret site was an area of low insecticide use (Graves et al. 1981).

## MATERIALS AND METHODS

Specimens were collected throughout 1978 and 1979 at times when the particular species were sufficiently abundant. The herons (yellow crowned night heron, <a href="Nyctanassa">Nyctanassa</a> violacea</a>), snakes (water

snakes, Natrix spp.), and raccoons (Procyon lotor) were collected by shooting or trapping. The fish (shad, Dorosoma spp.; channel catfish, Ictalurus punctatus; crappie, Pomoxis spp.), frogs (Rana spp.), and crawfish (Procambarus clarkii) were collected through use of an electrical shocking device, hook and line, or nets. Whole body samples were used for all animals except raccoon, where only legs muscles were retained. Samples of catfish also included fillets alone, while frog samples also included legs alone. Samples of individual specimens from each watershed and year were pooled in groups of five (five groups total) for residue analysis except for crawfish, which were pooled in groups of ten, and heron and raccoon, which were analyzed individually (ten individuals total). Immediately after collection, specimens were packed in dry ice, and then homogenized in Waring® blenders or Hobart® choppers. Samples were repacked in dry ice and sent to Raltech Scientific Services, Inc. (Madison, Wisconsin) for analysis. The organochlorines of interest included alpha- and beta-BHC, alpha- and gamma-chlordane, compound E, DDD, DDE, DDT, dieldrin, endrin, heptachlor, PCBs, trans- and cis-nonachlor, and toxaphene.

Fish, frog, and snake samples were initially homogenized in a Hobart food grinder, extracted with petroleum ether, and concentrated by evaporation. Samples were cleaned up on a Florisil column by eluting with ethyl ether and then concentrated. Further cleanup was done on a Silicar CC-4 column by elution with petroleum ether, followed by a mix of 1% acetonitrile, 19% hexane, and 80% methylene chloride, and then concentration to 2 ml. Samples were brought to 10 ml with petroleum ether, and quantitated by gas-liquid chromatography. Bird and raccoon samples were placed initially in a Whatman® extraction thimble, desiccated overnight, and then extracted on a Soxhlet extractor for 8 hours with 50:50 petroleum ether:ethyl ether. The samples were then concentrated over a steam bath to 5-10 ml, and brought to 25 ml with petroleum ether. Cleanup again involved Florisil (elution with 5% and 20% ethyl ether in petroleum ether) and Silicar columns (same method as for fish, frogs and snakes). The second elution from the Silicar column was used for the dieldrin and endrin determinations.

The lipid content was determined by drying at 40°C for 24 hrs, followed by desiccation and reweighing. Total moisture was determined by drying at 40°C for 2 weeks, followed by desiccation and reweighing. Gas-liquid chromatographic determinations were done on an HP5710A instrument with an electron capture (Ni<sup>63</sup>) detector. The packing for non-chlordane isomers consisted of 1.5/1.95% OV-17/QF-1 on 80/100 supelcoport, while for chlordane isomer detection the packing was 3% OV-1 on 80/100 supelcoport. Both types of samples were injected with a gas flow of 33 m/min of 95% argon and 5% methane. Detection limits for most organochlorines were 0.05 ppm, except for PCBs and toxaphene (0.10 ppm).

## RESULTS AND DISCUSSION

Little or no residues of any organochlorine compound were detected in any of the catfish fillet, crawfish, frog leg and whole body, raccoon, or snake samples taken from the Lake Verret site (Tables 1 and 2). The compounds most frequently found as residues in catfish whole body, crappie, shad and heron were DDE, dieldrin, and PCBs. There was a general decline in the levels of DDE and dieldrin from 1978 to 1979. However, levels of PCBs that were detected remained approximately the same or increased over the same period (up to a 3% increase in herons).

Samples of catfish fillet, frog, and raccoon from the Plaquemine-Brule site were generally free of organochlorine residues (Tables 3 and 4). The compounds that were most frequently encountered as residues in samples from catfish whole body, heron, shad, crappie, crawfish, and snake species were chlordane isomers, DDE, and PCBs. Declines in residue levels of DDE and chlordane were noted from 1978 to 1979, although PCB levels remained the same or increased. An increase in the level of toxaphene residues was also noted for crappie over this same period.

Samples of catfish fillet, crawfish, frog, raccoon, and snake from the East Franklin site were relatively free of organochlorine residues (Tables 5 and 6). Residues from catfish whole body, crappie, heron, and shad primarily consisted of DDE and PCBs, except for shad, which had residues of several compounds in 1978. Declines in levels of DDE were noted from 1978 to 1979 for all samples except shad. PCB levels remained fairly constant over the 2 year period.

The data presented herein indicate that heron and shad were the species most likely to accumulate organochlorine residues, regardless of the contamination level of the site from which they were collected. Thus, these species can serve as indicators of organochlorine contamination in the state of Louisiana, as has been indicated previously for shad (Graves et al. 1981). Levels of organochlorine compounds were greatly reduced for raccoon, shad, and catfish when compared to the levels reported during the years of widespread organochlorine insecticide use (1962) (Epps et al. 1967), and to residue levels just after the discontinuation of DDT use (1973) (Graves et al. 1981). DDT was present in only one instance, and the levels of DDT metabolites present were often barely detectable. Toxaphene residues reported in this study probably reflected its use as a herbicide in soybean fields, since it is seldom used as an insecticide anymore (Graves et al. 1981).

All average levels were below maximum tolerance levels for the organochlorine compounds. Although values reported are averages, some individual specimens or groups of specimens may have contained levels at least 10X the mean.

Many parts of the U.S. have already reported to be contaminated

Table 1. Organochlorine residues in animals from Lake Verret site (1978)<sup>a</sup>.

BHC-A BHC-B BHC-B Chlordane-A Chlordane-G Compound E DDD NR NR NR DDD NR NR NR DDE NR NR NR DDE NR NR NR DDE NR NR NR DDE NR NR NR D12-1	NR NR NR NR					
NR N	NR NR NR NR					
NR N	NR NR NR	NR			NR	NR
NR NR NR NR NR	NR NR NR	NR			NR	NR
NR NR NR NR	NR NR	NR	NR NR	R NR	NR	.072 <sub>h</sub>
NR NR NR	NR	NR			NR	.050
NR NR NR		NR			NR	NR
NR NR	NR,	NR			NR	.084
NR NR	.012 <sup>b</sup>	NR		_	NR	.126
	NR	NR			NR	NR
MB	NR	NR			NR	060.
IND	NR	NR			NR	NR,
NR,	NR,	NR			NR	.010
.052 <sup>b</sup> .	9090°	NR			NR	.228°
NR	NR	NR			NR	.010
	NR	NR			NR	NR
NR	NR	NR			NR	NR

A=alpha, B=beta, G=gamma, C=cis, T=trans, FIL=fillets, WB=whole body, LEG=legs. NR=no residue detected at levels indicated in the text, PCB=polychlorinated, biphenyls (as Arochlor 1260), BHG=benzene hexachloride. b Otpusmyre was exceeded by the standard error.

Organochlorine residues in animals from Lake Verret site (1979)ª. Table 2.

Compound	Ca	Catfish IL WB	Crappie	Crawfish	Frog LEG W	og WB	Heron	Raccoon	Shad	Snake
	1						ع. ا			
BHC-A	NR	NR	NR	NR	NR	NR	.025	NR	NR	NR
BHC-B	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR
Chlordane-A	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR
Chlordane-G	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR
Compound E	NR	NR,	NR	NR	NR	NR	NR	NR	NR	NR
ወወወ	NR	.010	NR	NR	NR	NR	NR	NR	NR	NR.
DDE	NR	.042	NR	NR	NR	NR	$125_{L}^{D}$	NR	NR	.020 <sup>b</sup>
DDT	NR	NR	NR	NR	NR	NR	.014 <sup>D</sup>	NR	NR	NR
Dieldrin	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR
Endrin	NR	.022	NR	NR	NR	NR	NR	NR	NR	NR
Heptachlor	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR.
PCBs	NR	.136	.124	NR	NR	NR	.235 <sup>D</sup>	NR	.422	.110 <sup>b</sup>
Nonachlor-T	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR
Nonachlor-C	NR	NR	NR	NR	NR	NR	NR,	NR,	NR	NR
Toxaphene	NR	NR	.082	NR	NR	NR	.247 <sup>D</sup>	.095 <sup>b</sup>	.218 <sup>b</sup>	NR

a A=alpha, B=beta, G=gamma, C=cis, T=trans, FIL=fillets, WB=whole body, LEC=legs.
NR=no residue detected at levels indicated in the text, PCB=polychlorinated
biphenyls (as Arochlor 1260), BHC=benzene hexachloride.

Means are exceeded by the standard error.

Organochlorine residues in animals from Plaquemine-Brule site (1978). $^{\rm a}$ Table 3.

Compound	Ca FIL	Catfish FIL WB	Crappie	Crawfish	Frog LEG W	og WB	Heron	Raccoon	Shad
		l							
BHC-A	NR		NR	NR	K	NR	NR	NR	NR
BHC-B	NR		NR	$NR_{\lambda}$	NR	NR	NR	NR	NR
Chlordane-A	NR	•	NR	.020°	NR	NR.	NR	NR	.076 <sup>5</sup>
Chlordane-G	NR	•	NR	.026 <sub>1</sub>	NR	NR	NR	NR	.082
Compound E	NR		NR	.010	NR	NR	NR	NR	NR
DDD	NR		NR	NR	NR	NR	$NR_{ m c}$	NR	.106
DDE	NR	•	NR	NR	NR	NR	,690	NR	.122
DDT	NR		NR	NR	NR	NR	NR	NR	NR
Dieldrin	NR	NR	NR	NR	NR	NR	NR	NR	.092
Endrin	NR		NR	NR	NR	NR	NR	NR	$NR_{\lambda}$
Heptachlor	NR		NR	NR	NR	NR	$NR_{\rm r}$	NR	.012
PCBs	060.	•	990.	$NR_{\rm r}$	NR	NR	.083	NR	.318
Nonachlor-T	NR	•	NR	.020	NR	NR	NR	NR	NR
Nonachlor-C	NR		NR	NR	NR	NR	NR	NR	.026
Toxaphene	NR		NR	NR	NR	NR	NR	NR	NR

A=alpha, B=beta, G=gamma, C=cis, T=trans, FIL=fillets, WB=whole body, LEG=legs. NR=no residue detected at levels indicated in the text, PCB=polychlorinated biphenyls (as Arochlor 1260), BHC=benzene hexachloride. b Means are exceeded by the standard error.

Table 4. Organochlorine residues in animals from Plaquemine-Brule site (1979)<sup>a</sup>.

Compound	Cat	Catfish FIL WB	Crappie	Crappie Crawfish	Frog LEG WE	og WB	Heron	Raccoon	Shad	Snake
BHC-A	NR	2	NB	MR	NR	NR	NR	NR	NR	MR
BHC-B	NR.	NR	NR	NR	NR	NR	NR	NR	NR	NR NR
Chlordane-A	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR
Chlordane-G	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR
Compound E	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR
מממ	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR
DDE	NR	.022	NR	NR	NR	NR	.307 <sup>D</sup>	NR	.238	NR
DDT	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR
Dieldrin	NR	NR	NR	NR	NR	NR	.011 <sup>D</sup>	NR	MR	NR
Endrin	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR
Heptachlor	NR	NR	NR	NR	NR	NR	NR	NR	NR.	NR
PCBs	NR	.018	.154	NR	NR	NR	.057	NR	1.126	NR
Nonachlor-T	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR
Nonachlor-C	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR
Toxaphene	NR	NR	.704	NR	NR	NR	NR	NR	NR	NR

<sup>a</sup> A=alpha, B=beta, G=gamma, C=cis, T=trans, FIL=fillets, WB=whole body, LEG=legs. NR=no residue detected at levels indicated in the text, PCB=polychlorinated, biphenyls (as Arochlor 1260), BHC=benzene hexachloride. b Means are exceeded by the standard error.

Table 5. Organochlorine residues in animals from East Franklin site (1978).

compound	Cat	Catrish (	Crappie	Crawiish	FF	26 26	Heron	Raccoon	Shad
	FIL	WB	i I		LEG WE	WB			
BHC-A	NR	NR	NR	NR	NR	NR	NR	NR	NR
BHC-B	NR	NR	NR	NR	NR	NR	NR	NR	NR
Chlordane-A	NR	NR	NR	NR	NR	NR	NR	.017 <sup>b</sup>	$.012^{\rm D}_{ m L}$
Chlordane-G	NR	NR	NR	NR	NR	NR	NR	.017 <sup>D</sup>	.010
Compound E	NR	NR	NR	NR	NR	NR	NR	NR	NR
DDD	NR	NR	NR,	NR	NR	NR	NR	NR	.084,
DDE	NR	.010 <sup>b</sup>	.010 <sup>b</sup>	NR	NR	NR	2.00	NR	.128 <sup>D</sup>
DDT	NR	NR	NR	NR	NR	NR	NR	NR	NR
Dieldrin	NR	NR	NR	NR	NR	NR	.029 <sup>b</sup>	NR	920.
Endrin	NR	NR	NR	NR	NR	NR	NR	NR	NR
Heptachlor	NR	NR	NR	NR	NR	NR	NR	NR	.016 <sup>D</sup>
PCBs	NR	.020°	NR	NR	NR	NR	.071 <sup>D</sup>	NR	$NR_{\mu}$
Nonachlor-T	NR	NR	NR	NR	NR	NR	NR	NR	.012
Nonachlor-C	NR	NR	NR	NR	NR	NR.	NR	NR	NR
Toxaphene	NR	NR	NR	NR	NR	NR	.082	NR	NR

A=alpha, B=beta, G=gamma, C=cis, T=trans, FIL=fillets, WB=whole body, LEG=legs. NR=no residue detected at levels indicated in the text, PCB=polychlorinated biphenyls (as Arochlor 1260), BHC=benzene hexachloride.

b orpnengis (as attended), uncluence Means are exceeded by the standard error.

Organochlorine residues in animals from East Franklin site (1979). Table 6.

Compound	Cat	Catfish IL WB	Crappie	Crawfish	Frog LEG W	WB WB	Heron	Raccoon	Shad	Snake
BHC-A BHC-B Chlordane-A Chlordane-G Compound E DDD DDE DDT Dieldrin Endrin Heptachlor	NR NR NR NR NR NR	NR NR NR NR NR NR NR	NR N	NN N N N N N N N N N N N N N N N N N N	NR NR NR NR O24 NR NR	NR NR NR NR NR NR	NR NR NR NR NR NR OS6 NR OS6	NN	NR	AN A
PCBs Nonachlor-T Nonachlor-C Toxaphene	NR NR NR	. 026 NR NR NR	.052 NR NR .084	NR NR NR	NR NR NR	NR NR NR	• 675- NR NR NR	NR NR NR	NR NR NR .456	NR NR NR

a A=alpha, B=beta, G=gamma, C=cis, T=trans, FIL=fillets, WB=whole body, LEG=legs. NR=no residue detected at levels indicated in the text, PCB=polychlorinated biphenyls (as Arochlor 1260), BHC=benzene hexachloride. Means are exceeded by the standard error.

with PCBs, such as the Great Lakes region (Veith et al. 1981), and the present study indicates that PCBs are also present in Louisiana watersheds. Levels of organochlorine compounds such as DDE and dieldrin generally decreased both in this study, and in previous studies of Louisiana wildlife as well (Schmitt et al. 1981. Butler and Schutzmann 1978). However, some previous studies in Louisiana (Schmitt et al. 1981) have indicated that PCB levels have increased, and this study demonstrates that levels of PCBs either remained the same or increased. were found at some sites in species used for human consumption (crappie, catfish), continued monitoring is advisable in order to prevent harmful levels of PCBs appearing in animals used in human In addition, since PCBs are known to be associated with sediment movement (Frink et al. 1982), monitoring appears necessary to detect the potential spread of PCBs to drainage areas of watersheds or into groundwater.

Acknowledgements. Support of this research furnished by the U.S. Department of the Interior through the Patuxent Wildlife Research Center Contract 14-16-0009-77-950.

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Received June 25, 1984; accepted July 10, 1984.