NATIONAL WATER-QUALITY ASSESSMENT PROGRAM: ENVIRONMENTAL DISTRIBUTION OF ORGANOCHLORINE COMPOUNDS IN THE APALACHICOLA-CHATTAHOOCHEE-FLINT RIVER BASIN

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ABSTRACT

The Apalachicola-Chattahoochee-Flint River basin, which drains about 20,000 square miles of Georgia, Florida, and Alabama, is one of 20 large hydrologic systems included in the first phase (1991-1996) of the U.S. Geological Survey's National Water-Quality Assessment program. One component of this program is the assessment of the occurrence and distribution of organochlorine compounds (pesticides and PCB's) in bed sediments and aquatic life in hydrologic settings representing ranges in drainage order, physiography, geology, climate, and land use. During August and November, 1992, and August and September, 1993, 37 stream sites and 7 reservoir sites in the ACF River basin were sampled for bed sediment (mineral and organic matter less than 2 mm) and either *Corbicula fluminea* (Asiatic clam) or *Gambusia affinis holbrooki* (mosquitofish). Bed sediments and tissues were analyzed for 38 organochlorine compounds. Results of these analyses are summarized and compared with historic data reported by Federal and State agencies. DDD, DDE, DDT, chlordane, dieldrin, and total PCB's were the most frequently detected organochlorine compounds in the ACF River basin, with detectable concentrations measured in more than 10 percent of samples. DDT and its homologs are widely distributed, but concentrations are much lower in recent (1992 and 1993) years. The widespread distribution and potential for continued bioaccumulation of chlordane and PCB's warrent their continued monitoring and assessment. Aldrin, heptachlor, toxaphene, endrin, endosulfan, methoxychlor, and hexachlorobenzene either were not detected or were detected in 2 percent or less of water samples.

INTRODUCTION

Background. Organochlorine insecticides, polychlorinated biphenyls (PCB's), polychlorinated dibenzo-*p*-dioxins (PCDD's), and polychlorinated dibenzofurans (PCDF's) comprise an environmentally significant group of chlorinated hydrocarbons because of their chemical properties. In the aquatic environment, these compounds are chemically stable, partition into both sediment and the lipid reservoirs of aquatic organisms, and are toxic or, in some other way, harmful to aquatic life (Delbeke and others, 1990; Day, 1990; Burton, Jr., 1992; Adams and others, 1992). The manufacture and use of organochlorine insecticides began in 1939 with DDT; followed by aldrin, dieldrin, chlordane, heptachlor, lindane, and toxaphene during the 1940's; and by endrin, endosulfan, methoxychlor, and mirex during the 1950's and 1960's (Smith and others, 1988). Though few historical data exist on the usage of organochlorine insecticides in the Apalachicola-Chattahoochee-Flint (ACF) River basin, the southeastern United States has accounted for the largest use of these compounds. Aldrin, chlordane, DDT, and toxaphene were the insecticides most used in this region, based on U.S. Department of Agriculture statistics for 1966. Control of cotton pests accounted for 70 percent of the DDT applied nationally through 1970 (U.S. Environmental Protection Agency [USEPA], 1972). Insecticide applications to cotton exceeded the combined total for all other crops. During the mid-1970's, production and use of many of the organochlorine insecticides dropped, and their current use is negligible. During 1990, about 48,000 pounds (active ingredient) of endosulfan were applied to 47 percent of the 66,000 acres of pecans grown in the ACF River basin, and about 5,400 pounds of lindane (active ingredient) were applied to 21 percent of the pecan acreage (Gianessi and Puffer, 1992). These are the only organochlorine insecticides currently used (1995) in the ACF River basin, the others having been replaced by more acutely toxic (Hudson and others, 1984; Smith, 1987) and more degradable (Morrill and others, 1982; Howard, 1991) organophosphates, such as chlorpyrifos and diazinon, or carbamates, such as carbaryl.

The dielectric and thermal properties of PCB's, first commercially produced in 1929, resulted in their widespread use in transformers, capacitors, and electromagnets, and as heat-transfer and hydraulic fluids (Smith and others, 1988). Before 1974, they also were used as plasticizers, lubricants, ink carriers, gasket seals, and additives in some pesticide formulations (USEPA, 1992). PCB use was restricted by the USEPA in 1977, and all manufacture and use of PCB's was prohibited in 1979 (Smith and others, 1988). PCDD's and PCDF's were not specifically manufactured, but occurred as contaminant byproducts of the manufacturing processes used for PCB's and organochlorine insecticides such as toxaphene (Smith and others, 1988).

Environmental Issues. The harmful properties of many of the organochlorine insecticides and PCB's prompted regulations that restricted or prohibited the use of these compounds (USEPA, 1990; table 1). Although the manufacture and use of many organochlorine compounds were discontinued during the 1970's and 1980's, these compounds are resistant to physical, chemical, and biological degradation; thus, they persist in the environment. Of the approximately 1.2 million tons of PCB's produced through 1980, it is estimated that 35 percent remains available in environmental compartments (Eisenreich, 1987). Organochlorine insecticides are neurotoxins. Although less acutely toxic than the organophosphates and carbamates in wide use today, they potentially are more chronically toxic because they are bioaccumulators (Schmitt and others, 1985; Eisler, 1990). These compounds have high k_{ow}'s (octanol-water partition coefficients) and high BCF's

Table 1: Uses, regulatory status, and environmental characteristics of some organochlorine compounds targeted for monitoring in sediments and aquatic tissues by Federal and State agencies

Organochlorine compound	Uses	Regulatory status[1]	Health and environmental concerns prompting regulation[1]
DDD	fruits, vegetables; degradation product of DDT	all uses except emergency public-health uses cancelled, January 1973 (applies to DDD, DDE, and DDT)	carcinogenic, bioaccumulator, wildlife hazard (applies to DDD, DDE, and DDT)
DDE	degradation product of DDD and DDT		
DDT	cotton, fruits, vegetables, mosquito control		
Aldrin	corn, termite control	agricultural uses cancelled, October 1974; termiticidal use cancelled, May 1987	carcinogenic, bioaccumulator, wildlife hazard, other chronic effects
Chlordane	corn, termite control; 'technical' chlordane is approximately 50 percent *cis*- and *trans*-chlordane with a variable mixture of other isomers	agricultural uses cancelled, December 1974; termiticide use, April 1988	oncogenic
Dieldrin	termite control; degradation product of aldrin	agricultural uses cancelled, October 1974; termiticide use, May 1987	carcinogenic, bioaccumulator, wildlife hazard, other chronic effects
Heptachlor	corn, termite control	agricultural uses cancelled, December 1974; termiticide use, April 1988	oncogenic
Lindane[1]	pecans, cucumbers, squash, livestock, seed treatment, structural treatment, residential use on ornamentals	restricted uses, February 1985. lindane must be 99.5 percent g- isomer. sale and use of 'technical' lindane prohibited	oncogenic, teratogenic, reproductive effects, other chronic effects, acute toxicity
Toxaphene	cotton, livestock	restricted uses, November 1982; all uses cancelled, 1989	oncogenic, population reduction in non-target species, acute toxicity to aquatic organisms, chronic effects on wildlife; contamination with PCDD's and PCDF's
Endrin	cotton, wheat	all uses cancelled, October 1984	oncogenic, teratogenic, population reduction in non-target and endangered species
Endosulfan[1]	pecans, peaches, apples, tomatoes, watermelons, squash, tobacco	in use	no restriction
Methoxychlor	fruit trees, vegetables, cattle[2]	in use	no restriction
Mirex	fire-ant control, degrades to kepone	all uses cancelled, December, 1976	carcinogenic, bioaccumulator, hazard to wildlife, other chronic effects
Hexachlorobenzene	seed protectant[2]	in use	no restriction
Total PCB's	dielectric fluids in transformers, capacitors, electromagnets; plasticizers, lubricants, ink carriers, gasket seals	restricted use, 1977; all manufacture and use cancelled, 1979	oncogenic; contamination with PCDD's and PCDF's

[1]U.S. Environmental Protection Agency (1990).
[2]1994 Farm Chemicals Handbook (1994).

(bioconcentration factors); and thus, readily partition into the sediment and tissue compartments of aquatic ecosystems (Jaffe, 1991). Aside from toxic effects on wildlife, most organochlorine insecticides are also carcinogenic (causing cancer), mutagenic (causing damage to cellular DNA), teratogenic (causing birth defects), and/or oncogenic (inducing tumors) (USEPA, 1990). PCB's are oncogenic and chronically toxic, but the primary concern is the PCDD and PCDF contaminants in PCB mixtures (E.T. Furlong, U.S. Geological Survey [USGS], written commun., 1994).

At present (1995), the only enforcable standards relating to organochlorine insecticides and PCB's are for surface- and ground-water drinking supplies, and edible and whole fish tissue and/or edible shellfish tissue used for consumption by humans. The USEPA has proposed sediment-quality criteria for hydrologic systems supporting aquatic life; however, these are to be guidelines rather than standards (Nowell and Resek, 1994), and thus, will not be enforcable.

Agencies Providing Data For This Assessment. Several Federal and State programs provide data on the occurrence and distribution of organochlorine compounds in bed sediments and in the tissues of aquatic biota in the ACF River basin. These data were obtained from the USEPA's Water Storage and Retrieval (STORET) data base (through April 1991) and from the USGS's National Water Information System (NWIS) data base (1992 and 1993). The USEPA, the U.S. Army Corps of Engineers (USACOE), the Georgia Department of Natural Resources (GADNR), and the Florida Department of Environmental Protection (FLDEP, formerly the Florida Department of Environmental Regulation) are contributors to the STORET data base. Bed-sediment and tissue data collected as part of the USGS National Water Quality Assessment (NAWQA) program reside in the NWIS data base. These data represent both long-term monitoring for status and trends and synoptic assessments of organochlorine contamination such as the USEPA's National Study of Chemical Residues in Fish (USEPA, 1992). This USEPA study, conducted during 1987, was a synoptic sampling of fish tissues at 388 stream sites nationwide. Seven of these sites are located in the ACF River basin. Stream, reservoir, and estuarine sites sampled by the above-mentioned agencies are shown in figure 1. Periods of record and a statistical summary of the data are given in table 2.

The USGS began its NAWQA program during 1991 to provide water-resource managers with relevant information about the major hydrologic systems in the United States. A key component of the NAWQA design is the assessment of the occurrence and distribution of organochlorine contaminants in streambed sediments and aquatic tissues. An important part of this assessment is the analysis of data collected as part of the NAWQA program, combined with historical data collected by other Federal and State agencies, to provide an integrated look at the water-resource issues of concern. The ACF River basin is one of 20 hydrologic systems nationwide where sampling began during 1991, with an additional 38 systems being phased in between 1991 and 1997. The ACF River basin drains about 20,000 square miles of northeastern and western Georgia, northwestern Florida, and southeastern Alabama into the Gulf of Mexico; and serves as a water-supply resource for Metropolitan Atlanta, Columbus, and Albany, Ga. (fig. 1). The southern half of the ACF River basin is in the Coastal Plain physiographic province and supports intensive silviculture, and both irrigated and non-irrigated row-crop agriculture.

ASSESSMENT APPROACH

Network Design. Much of the monitoring of sediments and tissues for organochlorine compounds by Federal and State agencies has been targeted to known sources and areas of contamination, an approach consistent with regulatory requirements focused on human health. The NAWQA approach is somewhat different with its emphasis on the entire hydrologic system. The NAWQA sampling was conducted to characterize the hydrogeographic distribution of organochlorine compounds in the ACF River basin across environmental gradients of stream order, physiography, geology, climate, and land use. Therefore, sampling sites were selected along these gradients to assess the occurrence and distribution of organochlorine compounds throughout the ACF River basin as contrasted with the typical focus on mainstem and reservoir locations (fig. 1). Organochlorine compounds with low water solubilities and high affinities for sediment organic matter or biogenic lipid material (Suntio and others, 1988) were selected for analysis. PCB's and most of the organochlorine insecticides have these characteristics. This paper presents the results of the NAWQA study, together with the results from historical data, to describe the hydrogeographic distribution of organochlorine compounds in bed sediments and in the tissues of aquatic biota within the ACF River basin.

During August and November 1992, and August and September 1993, 37 stream sites and 7 reservoir sites in the ACF River basin were sampled for organochlorine compounds in bed sediments and either *Corbicula fluminea* (Asiatic clam) or *Gambusia affinis holbrooki* (mosquitofish) tissue. Four of these sites were resampled during the August-September 1993 sampling period. Many previous investigations have sampled fish tissues because of health concerns related to human consumption of fish. However, the NAWQA focus is on environmental contaminant distributions in the hydrologic system. Thus, *Corbicula* was selected as the organism of choice because of its widespread distribution and limited mobility.

Methods. Bed sediments were collected and processed by methods described by Shelton and Capel (1994). Sediment samples were seived onsite to less than 2 mm, and this size fraction was submitted for chemical analysis. *Corbicula* were collected at stream and reservoir sites and processed according to methods described by Crawford and Luoma (1993). *Corbicula* were not obtainable in the floodplain system of the Apalachicola River; thus, *Gambusia* were collected using seines and dip nets. Whole fish, rather than fillets, were submitted for analysis. Bed-sediment and tissue samples were analyzed by gas chromatography at the USGS laboratory (W.T Foreman and T.J Leiker, USGS, written commun., 1994).

Data Analysis. Temporal and spatial distributions of data for specific organochlorine insecticide or PCB compounds typically are too low to support analysis of degradation or environmental processing pathways. This is true both historically and for the NAWQA sampling program. For this analysis, concentration data for specific compounds were aggregated to the compound groups presented in tables 1 and 2, where 'group' includes all isomers, homologs, metabolites, and degradation products. For multiple occurrences within a compound group (for example, cis- and trans-chlordane, cis- and trans-nonachlor, and oxychlordane are in the chlordane group) and within a specific medium (bed sediment or tissue), the values above detection

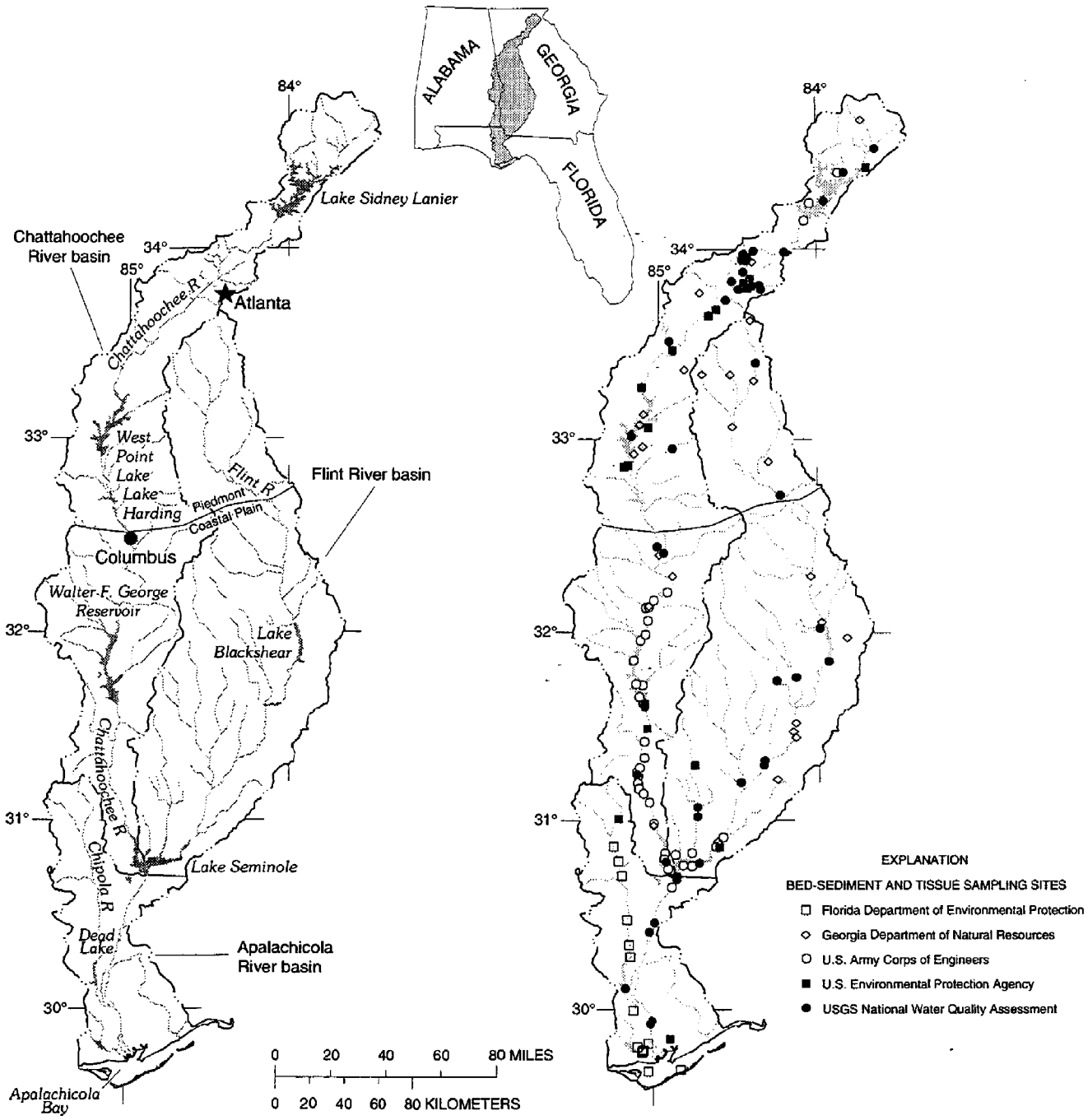

Figure 1. Location of Apalachicola-Chattahoochee-Flint River basin showing the stream, reservoir, and estuary sites sampled for organochlorine compounds in bed sediments and aquatic tissues, 1965-93.

to that for chlordane. However, in addition to widespread occurrence in the Piedmont reach of the Chattahoochee River, PCB's also were detected in the Chattahoochee River reach between Walter F. George Reservoir and Lake Seminole, in the Flint River downstream from Albany, Ga., and in Lake Seminole. PCB's, like chlordane, predominantly were detected in river mainstems and reservoirs.

DDT's widespread use in agriculture and mosquito control, and its environmental persistence, account for its ubiquitous occurrence in the ACF River basin (fig. 2). Data presented in this paper (table 2), and the findings of the U.S. Fish and Wildlife Service's National Contaminant Biomonitoring Program (NCBP; Jacknow and others; 1986, Schmitt and others, 1990) indicate that the DDE homologs (primarily *p,p'*-DDE) are the most prevalent form in both bed sediment and tissue. This indicates a low DDT influx into the environment and continued weathering of DDT residues. The NCBP (formerly part of the multiagency National Pesticide Monitoring Program) monitored organochlorine contamination in fish tissue at 112 stream sites nationwide from 1967 to 1984. Concentrations measured in sediment during the NAWQA sampling generally were lower than those reported in the historical data (table 2), indicating that environmental degradation is continuing.

Table 2. Organochlorine concentrations in bed-sediment and tissue samples collected as part of various Federal and State monitoring programs in the Apalachicola-Chattahoochee-Flint River basin, 1965-93

[Historical data were collected by the agencies shown in figure 1 (excluding the U.S. Geological Survey). Historical tissue data are derived predominantly from fish; NAWQA tissue data, from *Corbicula* (Asiatic clam). Sediment concentrations expressed as micrograms per kilogram dry weight; tissue concentrations, micrograms per kilogram wet weight; dl, detection limit; s, sediment; t, tissue; nd, no detection; nab, number of analyses above detection limit (actual values are reported when less than three samples had detectable concentrations of a compound, rather than the maximum and median values)]

	Historical data					U.S. Geological Survey NAWQA data (1992-93)			
		Sediment		Tissue		Sediment		Tissue	
Organochlorine compound	Period of record for historical data	Number of analyses (maximum and median conc. above dl)	Median dl (percent of analyses above dl)	Number of analyses (maximum and median conc. above dl)	Median dl (percent of analyses above dl)	Number of analyses (maximum and median conc. above dl)	Median dl (percent of analyses above dl)	Number of analyses (maximum and median conc. above dl)	Median dl (percent of analyses above dl)
DDD	s, 1973-1991	124	0.40	209	12.0	20	1.00	31	5.00
	t, 1965-1990	530-5.00	14	200-13.0	37	2.60-2.10	15	nd	0
DDE	s, 1973-1991	127	0.30	239	5.00	48	1.00	31	5.00
	t, 1965-1990	210-3.00	26	632,000-40.0	55	10.0-3.30	42	38.0-6.20	45
DDT	s, 1973-1991	172	3.00	214	10.00	20	2.00	31	5.00
	t, 1965-1990	364-13.2	12	60.0-13.0	12	3.60-2.80 (nab=2)	10	nd	0
Aldrin	s, 1973-1991	214	1.00	95	10.0	48	1.00	31	5.00
	t, 1978-1990	nd	0	nd	0	1.30 (nab=1)	2	nd	0
Chlordane	s, 1976-1991	192	3.00	126	80.0	48	1.00	31	5.00
	t, 1978-1990	3,300-106	15	857,000-375	43	9.50-4.80	19	83.7-34.3	10
Dieldrin	s, 1974-1991	206	2.00	129	10.0	48	1.00	30	5.00
	t, 1966-1990	10.0-2.60	1	94,100-21.0	17	3.80-1.10	10	7.90-6.45 (nab=2)	7
Heptachlor	s, 1973-1991	124	0.50	49	20.0	48	1.00	31	5.00
	t, 1978-1990	2,800 (nab=1)	1	nd	0	nd	0	11.0 (nab=1)	2
Lindane	s, 1973-1991	216	1.00	100	20.0	48	1.00	31	5.00
	t, 1978-1990	40.0-0.53	1	21,900-5,630	8	1.10 (nab=1)	2	6.00 (nab=1)	3
Toxaphene	s, 1973-1991	212	20.0	58	500	48	100	31	100
	t, 1978-1990	100 (nab=1)	<1	21.3 (nab=1)	2	nd	0	nd	0
Endrin	s, 1977-1991	178	2.00	81	30.0	48	2.00	31	5.00
	t, 1980-1990	nd	0	50.0 (nab=1)	1	nd	0	nd	0
Endosulfan	s, 1980-1991	32	8.00	45	30.0	48	1.00	na	na
	t, 1978-1990	nd	0	10.0 (nab=1)	2	nd	0		
Methoxychlor	s, 1977-1990	171	12.0	61	100	27	5.00	31	5.00
	t, 1978-1990	nd	0	17,900-380 (nab=2)	3	nd	0	nd	0
Mirex	s, 1973-1990	140	4.00	62	50.0	48	1.00	31	5.00
	t, 1978-1990	nd	0	35,500-855	19	1.10 (nab=1)	2	nd	0
Hexachlorobenzene	s, 1976-1991	124	0.50	58	20.0	48	1.00	31	5.00
	t, 1978-1990	nd	0	900-90.0 (nab=2)	3	nd	0	nd	0
Total PCB's	s, 1973-1991	246	6.00	141	200	48	100	31	50
	t, 1976-1990	753-68.0	31	6,060,000-490	70	79.0-76.0	6	180-63.0 (nab=2)	6

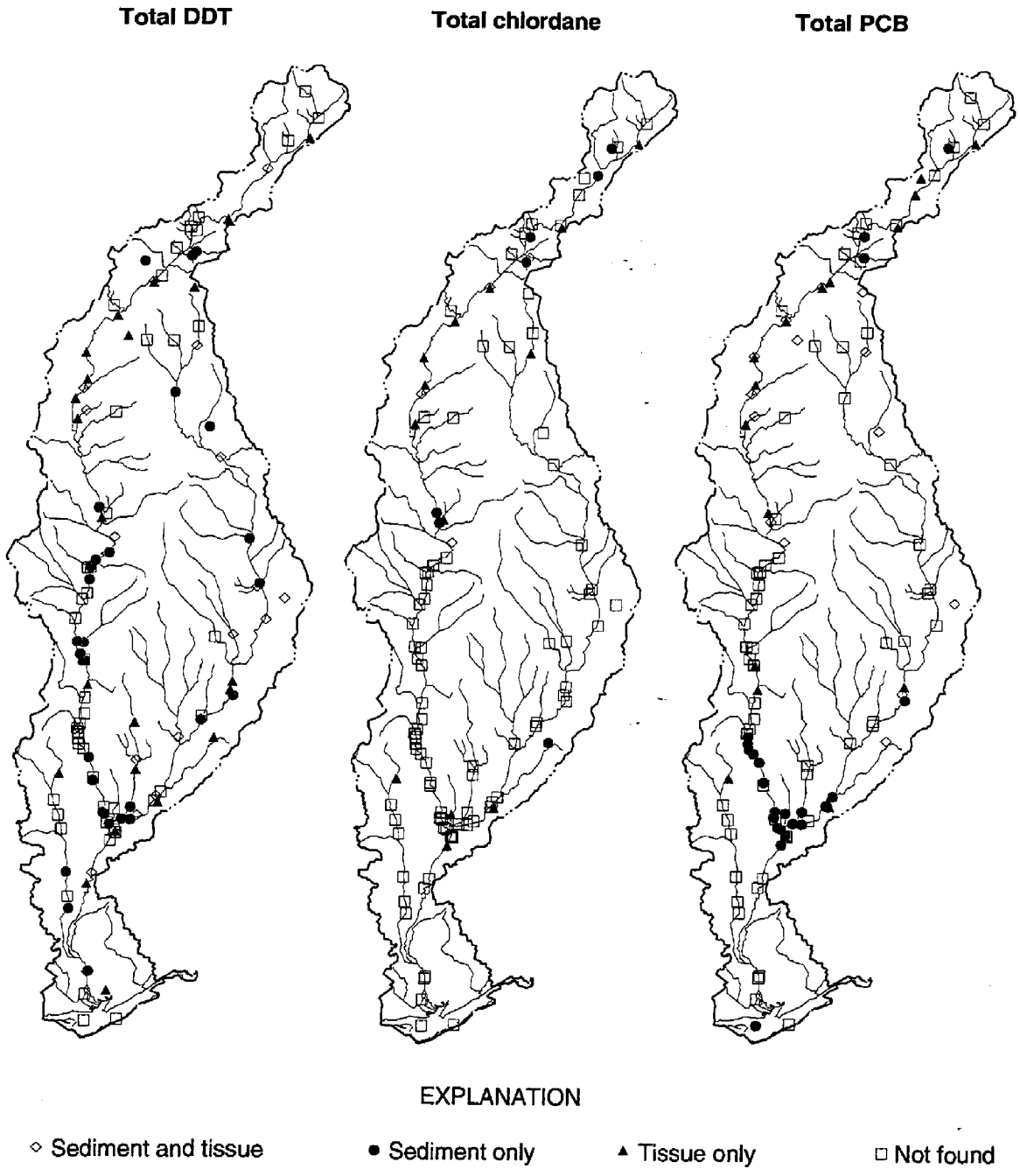

Figure 2. Geographic distributions of DDT and its homologs, chlordane, and total PCB's in bed sediments and aquatic tissues in the Apalachicola-Chattahoochee-Flint River basin, 1965-93.

Chlordane was primarily detected in mainstem reaches and reservoirs downstream from the urban areas of Atlanta, Columbus, and Albany, Ga. (fig. 2). Agricultural use of chlordane ceased in 1974, but termiticidal use continued through 1988, when chlordane use was banned by the USEPA (table 1), which partially may account for chlordane's presence in urban-affected areas of the ACF River basin and its relative absence in the agricultural areas of the Coastal Plain. The median chlordane concentration in bed-sediment samples collected during the NAWQA sampling was 4.80 µg/kg dry weight (maximum reported value, 9.50 µg/kg); whereas, the median concentration reported in the historical data (1976-91) was 106 µg/kg (maximum reported value, 3,300 µg/kg; table 2). These data indicate that chlordane is being removed from the environment by various degradation pathways. PCB occurrence in the ACF River basin indicates a similar urban or industrial effect but with a more widespread distribution than chlordane. Although PCB manufacture and use have ceased, utility companies are permitted to use transformers containing PCB's until failure, at which time the PCB's are recovered and disposed of. The relative absence of PCB's in the NAWQA bed-sediment and tissue samples seems anomalous, given their ubiquitous occurrence in the historical monitoring data (table 2). However, the median detection limit

limit in the list were summed. If all values were less than the detection limit, the average of all the values in the list was retained as the 'group' detection limit. This statistic presents a 'worst case' for a particular compound group and accounts for as much of the material as was analyzed for in its various forms.

RESULTS AND DISCUSSION

Frequency of Detection. DDD, DDE, DDT, chlordane, dieldrin, and total PCB's are the most frequently detected organochlorine compounds in bed sediment and tissue in the ACF River basin, with detectable concentrations measured in more than 10 percent of the samples (table 2). Results of the NAWQA sampling generally are comparable to those obtained from other Federal or State monitoring programs with a few exceptions. Historical data and the results of the NAWQA sampling indicate that DDE is the most common organochlorine contaminant in both bed sediment and tissue in the basin. Aldrin, heptachlor, toxaphene, endrin, endosulfan, methoxychlor, and hexachlorobenzene either were not detected in bed-sediment or tissue samples, or were detected in 2 percent or less of analyzed samples. Both spatial (fig. 1) and temporal (table 1) coverages by the various sampling programs indicate that these negative results are not anomalies of insufficient sampling. Toxaphene was extensively used throughout the southeastern United States (EPA, 1972) before its use was banned in 1989 (EPA, 1990), but was only detected in one sediment and one tissue sample.

Geographic Distribution. DDT and its homologs, chlordane, and PCB's were detected in wide distribution throughout much of the ACF River basin study area (fig. 2). Chlordane predominantly was detected in the Chattahoochee River reach from near its headwaters to Columbus, Ga., and, to a lesser extent, in the headwaters of the Chipola River, and in the lower Flint River and Lake Seminole. DDD, DDE, and DDT were the most uniformly distributed organochlorine contaminants. These compounds were detected in the Piedmont and Coastal Plain reaches of the Chattahoochee and Flint Rivers in Georgia and Alabama, and in the Chipola and Apalachicola Rivers in Alabama and Florida. Whereas chlordane predominantly was detected in river mainstems and reservoirs, DDT and its homologs also were detected in many tributaries. The geographic distribution of PCB occurrence was similar Although PCDD's and PCDF's were not sampled in the NAWQA program, fish collected for the USEPA's National Study of Chemical Residues in Fish (USEPA, 1992) at all seven sites within the ACF River basin had detectable quantities of these compounds in their tissues. These sites represent both mainstem (Chattahoochee River inflow to Lake Lanier, Chattahoochee River reach between Atlanta and West Point Reservoir, and Flint River inflow to Lake Seminole) and tributary (upper Chipola River and Lake Seminole) settings within the basin. Whole-body carp collected in the Chattahoochee River reach between Atlanta and West Point Reservoir ranked among the five sites nationally (of 388) with the highest concentrations of hexa- and heptachloro-dibenzo-*p*-dioxins and heptachloro-dibenzofurans.

SUMMARY

Organochlorine insecticides largely have disappeared from use in the United States having been replaced by organophosphates and carbamates. Virtually all of the PCB's in use today are in closed and recoverable systems such as transformers, and are being removed from circulation as the systems containing them fail. Endosulfan and lindane are the only two organochlorine insecticides presently used in the ACF River basin, and both are infrequently detected in sediment or aquatic life. Although DDT and its homologs are widely distributed, concentrations are much lower in recent (1992 and 1993) samples. Because of chlordane contamination, the states of Alabama and Georgia have issued consumption advisories for selected fish species in the Chattahoochee River reach between Atlanta and West Point Reservoir, and in West Point Reservoir (Alabama Department of Environmental Management, 1992; Georgia Department of Natural Resources, 1994). The widespread distribution and bioaccumulation of chlordane and PCB's warrant their continued monitoring and assessment.

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