Historical Trends in Organochlorine Compounds in River Basins Identified Using Sediment Cores from Reservoirs

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This study used chemical analyses of dated sediment cores from reservoirs to define historical trends in water quality in the influent river basins. This work applies techniques from paleolimnology to reservoirs, and in the process, highlights differences between sediment-core interpretations for reservoirs and natural lakes. Sediment cores were collected from six reservoirs in the central and southeastern United States, sectioned, and analyzed for ^{137}Cs and organochlorine compounds. ^{137}Cs analyses were used to demonstrate limited post-depositional mixing, to indicate sediment deposition dates, and to estimate sediment focusing factors. Relative lack of mixing, high sedimentation rates, and high focusing factors distinguish reservoir sediment cores from cores collected in natural lakes. Temporal trends in concentrations of PCBs, total DDT (DDT + DDD + DDE), and chlordane reflect historical use and regulation of these compounds and differences in land use between reservoir drainages. PCB and total DDT core burdens, normalized for sediment focusing, greatly exceed reported cumulative regional atmospheric fallout of PCBs and total DDT estimated using cores from peat bogs and natural lakes, indicating the dominance of fluvial inputs of both groups of compounds to the reservoirs.

Introduction

The U.S. Geological Survey (USGS) National Water-Quality Assessment (NAWQA) Program has three objectives, one of which is to "define trends (or lack of trends) in our nation's water quality" (*1*). This study, a part of the NAWQA Program, used chemical analyses of dated sediment cores from reservoirs to define historical trends in water quality in the influent streams. The sediment-core/paleo-environmental approach for documenting changes in water quality of natural lakes has a long history (*2*−*6*); however, there are important differences between natural lakes and reservoirs that affect the sampling and interpretation of sediment cores. Sedimentation rates for most natural lakes vary from 0.05 to 10 mm/yr (*3, 4*). In contrast, reservoirs exhibit sedimentation rates that range from 10 to 200 mm/yr (*7−9*). Greater sedimentation rates convey several advantages to recon-

structing trends, including allowing a finer temporal discretization and limiting the effects of post-depositional mixing and diagenesis (*7, 10*).

Settings. The reservoirs sampled are Coralville Reservoir in Iowa, White Rock Lake in Texas, and Lakes Walter F. George, Harding, Blackshear, and Seminole in Georgia. Coralville Reservoir is located on the Iowa River in east central Iowa (Figure 1). The reservoir was created in 1959, has a surface area of 101 km², and has a drainage area to surface area ratio of 80. The watershed is about 90% farmland, principally corn and soybeans.

White Rock Lake is located on White Rock Creek within the city limits of Dallas, TX. The reservoir, filled in August 1912, has a surface area of 4.4 km² and a drainage area to surface area ratio of 59. Agriculture dominated land use when White Rock Lake was constructed. Intensive urban development of the White Rock Creek drainage began in the 1950s, and the drainage was 72% urban in 1990 (Samuel Brush, North Central Texas Council of Governments, written communication, 1992).

Lake Harding is located on the Chattahoochee River 184 km downstream of Atlanta and 29 km upstream of Columbus, GA. Storage began in 1926. Lake Harding has a surface area of 24 km² and a drainage area to surface area ratio of 452. Lake Harding receives drainage and effluent from the Atlanta metropolitan area, which was reduced in 1974 with the completion of West Point Lake 25 km upstream.

Lake Walter F. George is located 290 km downstream from Atlanta on the Chattahoochee River and is separated from the Atlanta metropolitan area by Lakes Harding and West Point. Storage began in May 1962. Lake Walter F. George has a surface area of 188 km² and a drainage area to surface area ratio of 103.

Lake Blackshear is located 314 km downstream from Atlanta on the Flint River. Storage began in 1930. Lake Blackshear has a surface area of 35 km² and a drainage area to surface area ratio of 276. Land use is agricultural, and approximately half the drainage is forested.

Lake Seminole is located at the confluence of the Chattahoochee and Flint Rivers where they form the Apalachicola River. Storage began in May 1954. Lake Seminole has a surface area of 163 km² and a drainage area to surface area ratio of 272. The Chattahoochee arm of the reservoir is 83 km downstream of Lake Walter F. George. The Flint River arm of Lake Seminole is 160 km downstream of Lake Blackshear and 110 km downstream of Albany, GA. Land use in the lower parts of both the Chattahoochee River and Flint River drainages is largely agricultural.

Field and Laboratory Methods

Sampling. Cores were collected using a Benthos gravity corer with a 3.05 m long, 6.3 cm diameter, plastic-lined barrel and a Wildco box corer with a 14 × 14 × 20 cm box. (Any use of trade, product, or firm names is for descriptive purposes only and does not constitute endorsement by the U.S. Government.) One site was sampled in the middle or lower part of each reservoir. The site was selected based on reconnaissance sampling of each reservoir and bathymetric surveys with the objective of obtaining as thick a sequence of undisturbed lacustrine sediment as possible, yet still penetrating the pre-reservoir land surface. The pre-reservoir surface provided a date marker in the lower part of each core and was identified based on lithologic and chemical changes.

Analysis. Cores were subsampled for measurement of porosity, organochlorine compounds, organic carbon, and ^{137}Cs. Porosity was calculated based on measured wet and dry weights of samples and an assumed density of solids of

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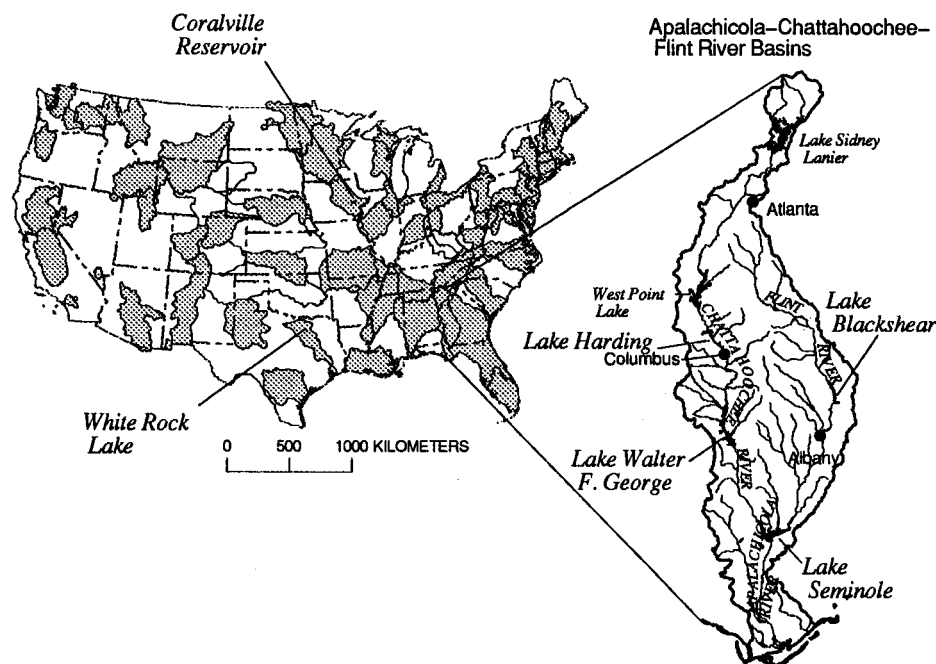
FIGURE 1. USGS National Water-Quality Assessment study units and sediment coring sites.

2.5 g/cm³. Organic carbon concentrations were measured using the method described in ref 11. Organochlorine compounds in samples from five of the six reservoirs were determined in organic solvent extracts using a dual capillary-column gas chromatograph with dual electron capture detectors following the method of Wershaw et al. (*11*). Compounds in samples from the other reservoir, Coralville Reservoir, were also determined in organic solvent extracts using a dual capillary-column gas chromatograph with dual electron capture detectors; however, the extraction and cleanup procedures recommended by Foreman et al. (*12*) were used. The Wershaw method uses a hand-shaken extraction first with acetone and then hexane, followed by an alumina/silica adsorption chromatographic cleanup. The Foreman method uses a Soxhlet extraction with dichloromethane and methanol followed by gel permeation and adsorption chromatographic fractionation. Because of these differences in methods, comparisons of PCB and total DDT (DDT + DDD + DDE) burdens among reservoirs did not include Coralville Reservoir.

Reporting levels for the Wershaw method are 0.1 μg/kg for DDT metabolites and 1.0 for technical chlordane and PCBs. Reporting levels for the Foreman method are 1.0 μg/kg for DDT metabolites and chlordane compounds (including *trans*-chlordane) and 50 μg/kg for PCBs. Accuracy and precision were determined by analyzing spiked samples and monitoring recovery of surrogates in the laboratory and by analyzing duplicate samples from the field. The median difference for 25 duplicate analyses of organochlorine compounds (five compounds in each of five samples) was 22%.

Sediment ^{137}Cs activity profiles were measured by counting freeze-dried sediments (20−75 g) in fixed geometry with a high-resolution (1.2 KeV fwhm at 661.6 KeV), intrinsic germanium detector γ-spectrometer. The method is similar to that used by Callender and Robbins (*7*). Replicate counts of samples agreed within ±15%. Detection limits ranged from 0.02 to 0.1 pCi/g, depending on sample mass analyzed.

Results and Discussion

The lacustrine sediments in cores from all six reservoirs are characterized by uniform, fine-textured sediments and no visible evidence of bioturbation. Median percent silt- and clay-sized particles (<0.062 mm) for the six reservoirs range

from 99.1 to 99.9%, and median percent clay-sized particles (<0.004 mm) range from 72.6 to 87.1%. Median organic carbon concentrations range from 2.0 to 2.4 wt % for five of the reservoirs; the median for the sixth, Lake Seminole, is 3.3 wt %. Because lacustrine sediments are relatively homogeneous with respect to grain size and organic carbon content, concentrations of ^{137}Cs and organochlorine compounds were not normalized prior to interpretation of trends. In no case would normalization have altered the interpretation of trends.

Post-Depositional Sediment Mixing. Post-depositional mixing can significantly alter profiles of particle-bound constituents recorded in bottom sediments (*13, 14*). Processes that cause sediment mixing include bioturbation and physical reworking by current and wave action. The pronounced ^{137}Cs peaks with exponential decreases in ^{137}Cs to the sediment surface in each reservoir core indicate relatively little mixing (Figure 2). Ratios of peak ^{137}Cs to surface ^{137}Cs activity are 5.9 ± 1.6, indicating limited mixing at these sites. By comparison, ratios of peaks to surface activity of about 3 and 8 were observed in cores from Lake Erie with "restricted mixing" (*15*). Relatively high sedimentation rates, the presence of laminations in sediment cores, and the low abundance of burrowing benthic organisms also suggest limited mixing. Therefore, post-depositional mixing probably has not significantly altered profiles of ^{137}Cs or other particle-bound constituents in these reservoirs.

Age Dating. Four dates can be identified in cores from the reservoirs constructed prior to 1952: the reservoir impoundment date (matched to the pre-reservoir surface), the first occurrence of detectable ^{137}Cs in 1952, the peak ^{137}Cs concentration in 1963, and the date of core collection. Based on these four dates, mean sedimentation rates were calculated for intervening time intervals, which were then used to assign estimated deposition dates to samples from each core. Sedimentation rates ranged from 0.52 to 3.4 g/cm² yr (0.9−4.7 cm/yr).

Sediment Focusing. Integrated ^{137}Cs activities, ϕ_c, were calculated by multiplying ^{137}Cs activity by the effective density to yield activity per volume of wet sediment. The activities per volume were then multiplied by the interval thickness and summed over the length of the core to give ϕ_c in picocuries per square centimeter (Table 1).
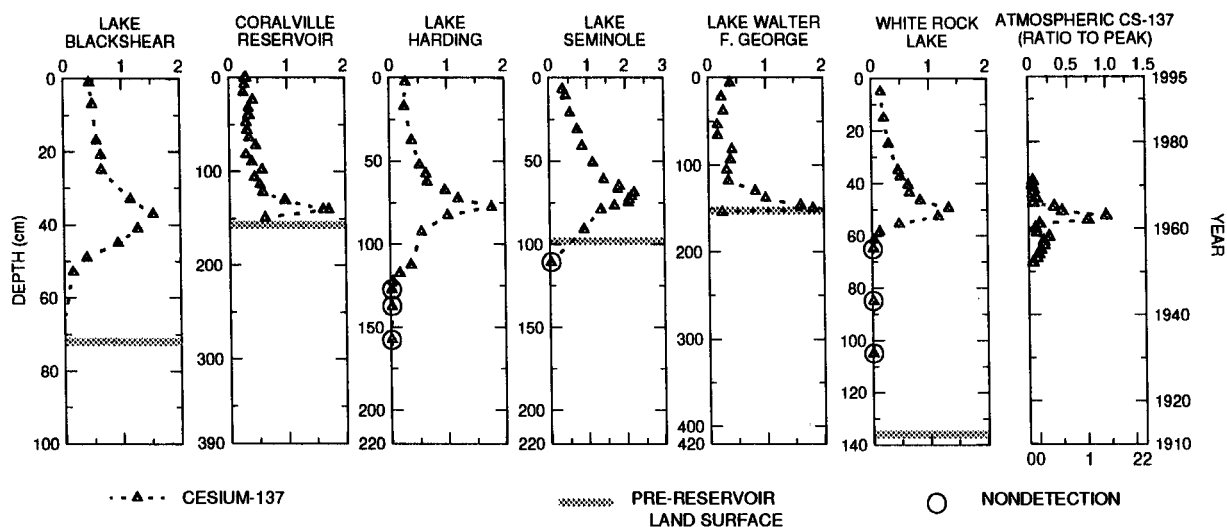
FIGURE 2. Cesium-137 concentrations in reservoir sediment cores and the atmosphere.

TABLE 1. Sediment Focusing Data

reservoir	max depth of ^{137}Cs (cm)	integrated ^{137}Cs activity, pCi/cm^2 (ϕ_c) ($\pm\sigma_c$)	^{137}Cs fallout activity, pCi/cm^2 (ϕ_f)	location of precipitation station used for ϕ_f	focusing factor (FF) (ϕ_c/ϕ_f)
White Rock Lake	63	11.6 (0.4)	5.1	Dallas, TX	2.3
Coralville Reservoir[a]	>155	>49.2 (1.2)	8.8	Iowa City, IA	>5.6
Lake Harding	125	39.4 (1.4)	6.2	Columbus, GA	6.3
Lake Walter F. George[b]	>156	>36.0 (1.5)	6.2	Columbus, GA	>5.8
Lake Blackshear	58	17.6 (0.8)	5.7	Albany, GA	3.1
Lake Seminole	100	42.3 (1.5)	6.0	av of Albany and Columbus, GA	7.1

[a] Coralville Reservoir lacustrine sediment record starts in 1959, but total ϕ_f extends from 1954 to 1974. If 1959−1974 flux used, FF = 6.6. [bb] Lake Walter F. George lacustrine sediment record starts in 1962, but total ϕ_f extends from 1954 to 1974. If 1962−1974 flux used, FF = 9.4.

TABLE 2. Sediment Core Burdens of PCBs and Total DDT

compd	reservoir core burdens (ng/cm^2)					published core burdens attributed to atmospheric fallout (ng/cm^2)		
	White Rock Lake	Lake Harding	Lake Walter F. George	Lake Blackshear	Lake Seminole	Lake Superior (21)	rural Wisconsin lakes (28)	peat bogs (29)
PCB	250	28 000	2 300	54	270	0.1−8	1.3−8.8	2.9−15.9
total DDT	250	2 600	970	370	540	10.4−27		2.6−21.6
	Normalized for Sediment Focusing[a]							
PCB	110	4 400	400	17	38			
total DDT	110	410	170	120	76			

[a] Core burden divided by sediment FF determined using ^{137}Cs.

The ^{137}Cs atmospheric fallout flux for each reservoir was derived from the fallout prediction model of Sarmiento and Gwinn (16). Monthly ^{137}Cs fallout delivery was decay corrected and summed to yield the total fallout deposition, ϕ_f (Table 1). Atmospheric deposition of ^{137}Cs after 1974 is not included and is estimated by Robbins (17) to account for only about 3% of the total flux. Sediment FFs, calculated for each core by dividing ϕ_c by ϕ_f, range from 2.3 to more than 7.1 (Table 2). FF provides an estimate of the degree of focusing of particle associated contaminants from both the drainage basin and within the reservoir (16, 18). Generally smaller, but occasionally comparable FFs have been reported for lakes. FFs ranging from less than 1 to more than 5 were reported for Lake Huron (15). FFs of 1.2 and 1.7 were reported for Lake Ontario (5).

Trends in PCBs. PCBs were not detected in White Rock Lake sediments deposited before about 1950 (Figure 3). Concentrations increase sharply in White Rock Lake sediments deposited from about 1950 to a peak of 21 μg/kg in a sample deposited in about 1969 and then decrease to 3 μg/kg

in the two most recent samples; a decrease of 86% since the peak. Changes in PCB concentrations in White Rock Lake sediments compare well with U.S. production and regulation of PCBs (19, 20).

Larger concentrations are found in the Georgia reservoirs downstream of Atlanta (Lakes Harding and Walter F. George). PCB concentrations in four Lake Harding samples deposited from about 1950 to 1966 range from 280 to 380 μg/kg (Figure 3). Concentrations in the two most recent samples are 42 and 46 μg/kg, 88% below the maximum concentration. The largest PCB concentration in Lake Walter F. George is 220 μg/kg in a sample that was deposited in about 1968. Concentrations decrease very rapidly at shallower depths and are less than 14 μg/kg in five samples deposited since the late 1970s, a 94% decrease.

West Point Lake was constructed on the Chattahoochee River 25 km upstream from Lake Harding and downstream from Atlanta in 1974 (Figure 1). If decreasing trends in PCBs in Lake Harding are the result of the elimination of PCB use, then PCB concentrations in West Point Lake sediments should
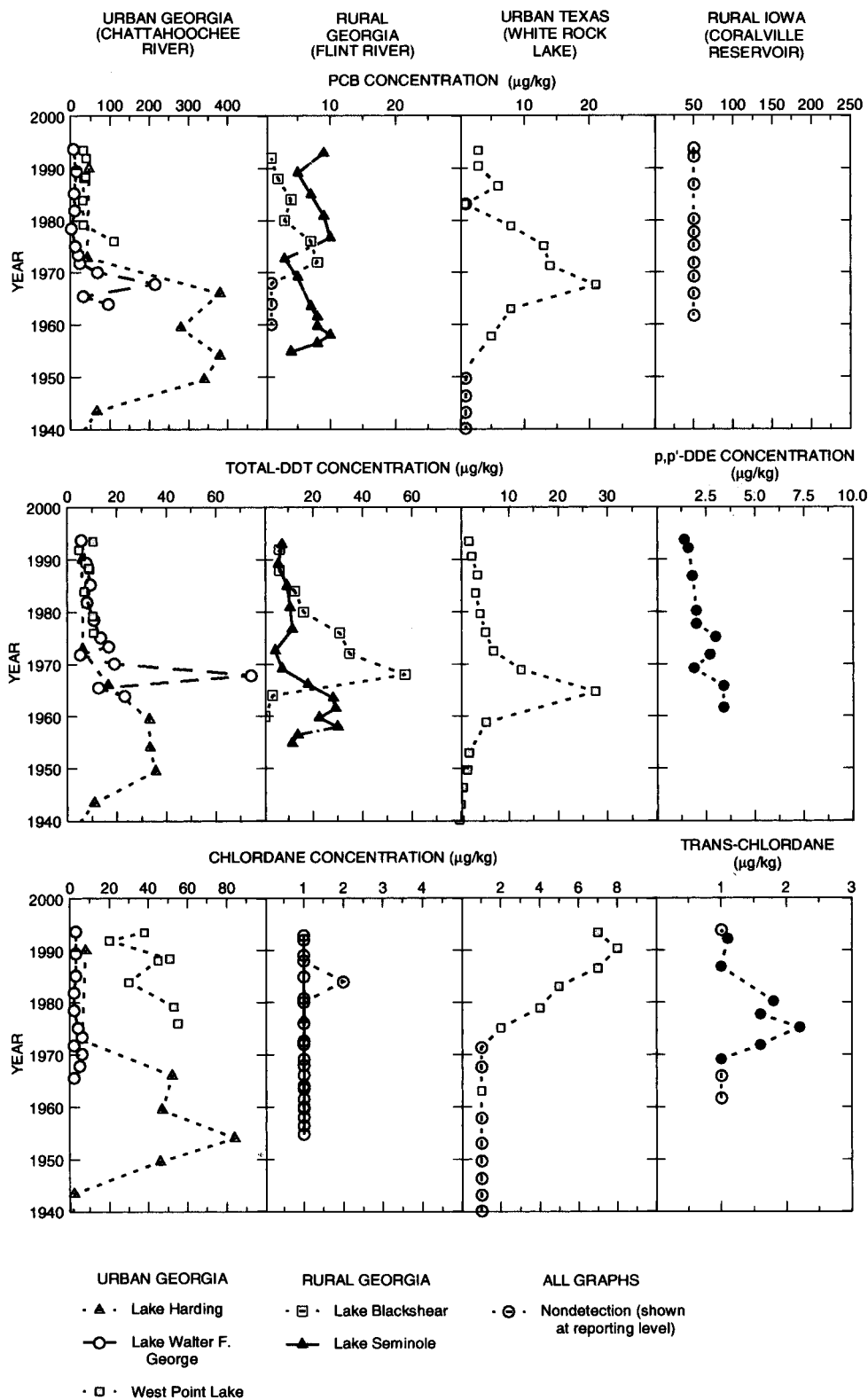
URBAN GEORGIA
(CHATTAHOOCHEE RIVER)

RURAL GEORGIA
(FLINT RIVER)

URBAN TEXAS
(WHITE ROCK LAKE)

RURAL IOWA
(CORALVILLE RESERVOIR)

PCB CONCENTRATION (µg/kg)

TOTAL-DDT CONCENTRATION (µg/kg)

p,p'-DDE CONCENTRATION (µg/kg)

CHLORDANE CONCENTRATION (µg/kg)

TRANS-CHLORDANE (µg/kg)

URBAN GEORGIA

· ▲ · Lake Harding

─○─ Lake Walter F. George

· □ · West Point Lake

RURAL GEORGIA

· ⊟ · Lake Blackshear

─▲─ Lake Seminole

ALL GRAPHS

· ⊖ · Nondetection (shown at reporting level)

FIGURE 3. Concentrations of organochlorine compounds in reservoir sediment cores.

be comparable to concentrations in Lake Harding after 1974. If, however, the decreasing trend is caused by West Point Lake intercepting much of the of PCB load to Lake Harding from an upstream source (e.g., Atlanta), then larger concentrations are expected in West Point Lake. PCB concentrations in seven sediment-core samples from West Point Lake, analyzed using the same methods as those used in Lake Harding, range from 110 µg/kg in the oldest sample (~1974) to 32 µg/kg in the most recent sample (1990s) (Figure 3; Van Metre, unpublished data), similar to Lake Harding. These

data suggest that decreasing PCB trends in Lake Harding are not caused by construction of West Point Lake.

Smaller concentrations and less-pronounced trends in PCBs occur in the two more rural Georgia reservoirs (Lakes Blackshear and Seminole) as compared to the three more urbanized reservoirs (Lake Harding, Lake Walter F. George, and White Rock Lake in Texas) (Figure 3). The largest concentration in Lake Blackshear, 8 µg/kg, occurred during the late 1960s, followed by a decrease to 1 µg/kg in the most recent sample. PCB concentrations in Lake Seminole range

from 3 to 10 μg/kg and, unlike in the other four reservoirs, did not have a distinct temporal trend. The coring site is on the Flint River arm of Lake Seminole about 1 km from where the Flint River and Chattahoochee River arms converge. Sediments at the coring site could, therefore, be a mixture from two different sources, causing some variability.

No PCBs were detected in 10 samples from Coralville Reservoir; however, the method used to analyze samples from Coralville is not as sensitive for PCBs (see Field and Laboratory Methods).

Historical trends in PCBs have been identified using sediment cores in other hydrologic settings (5, 21, 22). Eisenreich et al. (5) found temporal trends in PCBs in Lake Ontario cores similar to reservoir cores of this study. PCB concentrations and accumulation rates in the Lake Ontario cores had large increases from the 1940s to the late 1960s and then decreased to about one-third of maximum by about 1980 (5).

Trends in DDT, DDD, and DDE. DDT was first synthesized in 1874 and was used as an insecticide starting in 1939. DDT usage peaked in the United States in the early 1960s (23). DDT was widely used in the United States until about 1970; its use was banned nationwide in 1972. DDE was detected in all core samples deposited since about 1950 in all six reservoirs. DDE, on average, accounts for 58% of the total DDT (DDT + DDD + DDE) measured in samples from White Rock Lake and for 47, 74, 79, and 78% of the total DDT measured in samples from Lakes Harding, Walter F. George, Blackshear, and Seminole, respectively. DDD accounts for most of the remainder of total DDT in all of the reservoirs. DDT was detected in only 1 of 13 samples in White Rock Lake in which DDE was detected; however, DDT was detected in 8 of 10 samples from Lake Harding, in all 14 samples from Lake Walter F. George, in 7 of 9 samples from Lake Blackshear, and in all 13 samples from Lake Seminole.

Total DDT concentrations in White Rock Lake increased from the mid-1940s to a maximum of 27 μg/kg in about 1965 (Figure 3), about the same time that DDT usage peaked in the United States (23). Concentrations have decreased since 1965 by 93% to 2 μg/kg in the most recent sample. Total DDT concentrations in Lake Walter F. George peak at 74 μg/kg in sediments deposited in about 1968. By the early 1970s, concentrations were about 20% of the peak, and the total DDT concentration of the most recent sample, 5.5 μg/kg, is 93% less than the maximum concentration. A similar temporal pattern occurs at Lake Blackshear. The maximum total DDT concentration in the core from Lake Blackshear is 57 μg/kg as compared with 6.5 μg/kg in the most recent sample, an 89% decrease. Total DDT trends in the other two Georgia reservoirs are similar, with maximums of 30 and 35 μg/kg in Lakes Seminole and Harding, respectively. In both, the maximum occurs in samples deposited in the 1950s. The concentration in the most recent sample from Lake Harding is 5.9 μg/kg, an 83% decrease from the maximum. The concentration in the most recent sample from Lake Seminole is 7.5 μg/kg, a 75% decrease. The unusual temporal pattern of PCBs in Lake Seminole, with a concentration low in the 1970s, is repeated for total DDT but with a less pronounced rise in the 1980s. As suggested for PCBs, West Point Lake does not appear to have effected trends in total DDT in Lake Harding. Concentrations of total DDT in West Point sediments are similar to concentrations in the two downstream lakes after the early 1970s (Figure 3).

Concentrations of p,p'-DDE in Coralville Reservoir also decrease from the 1960s to the present but are smaller than DDE concentrations in the other five reservoirs (Figure 3). Each of the 10 Coralville samples have less than 1.0 μg/kg of o,p'-DDE, p,p'-DDT, o,p'-DDT, p,p'-DDD, and o,p'-DDD. The drainage area for Coralville Reservoir is about 90% agriculture, with corn and soybeans accounting for most of that.

Trends in Chlordane. Technical chlordane, a mixture of more than 140 compounds, is a pesticide with both agricultural and residential uses (24). Agricultural use of chlordane was principally on corn, and nationwide agricultural use in 1971 was an estimated 857 000 kg of active ingredient (25). Agricultural uses were cancelled in 1974 (23); however, significant urban use as a termiticide and for the control of fire ants continued until at least 1990 (26, 27).

Chlordane was not detected in samples from Lake Blackshear and was detected in only 1 of the 14 samples from Lake Seminole at a reporting level of 1.0 μg/kg. *trans*-Chlordane, shown for Coralville Reservoir (Figure 3), is one of the two most abundant components of technical chlordane (24) and, along with *trans*-nanochlor and *cis*-chlordane, is used to quantify technical chlordane using the Wershaw method (11). Trends in *trans*-chlordane in Coralville Reservoir reflect historical agricultural use with the peak of 2.2 μg/kg in sediments deposited in the mid-1970s followed by a decreasing trend to the top of the core.

Chlordane was first detected in the White Rock Lake core in a sample deposited in 1965; however, unlike Coralville, chlordane concentrations in White Rock Lake continued increasing from the 1960s to about 1990 (Figure 3). The largest concentration is 8 μg/kg in about 1990, followed by a small decline in concentration in the most recently deposited sample, a pattern indicative of continued urban use during the 1970s and 1980s.

Chlordane was detected in all 11 samples from Lake Walter F. George, and the maximum concentration is 6.0 μg/kg in a sample deposited in about 1973. Relatively large chlordane concentrations were found in Lake Harding core samples deposited from about 1950 to 1970, ranging from 46 to 84 μg/kg (Figure 3). Concentrations in the two Lake Harding samples deposited since the early 1970s are 6.0 and 8.0 μg/kg. Unlike PCBs and total DDT, chlordane concentrations in West Point Lake are larger than post-1974 concentrations in Lake Harding, ranging from 20 to 55 μg/kg (Figure 3; Van Metre, unpublished data). Recent urban use of chlordane, the presence of Atlanta upstream, and higher concentrations in the upstream reservoir coincident with a decrease in concentrations in the downstream reservoir suggest that much of the decreasing trend in Lake Harding can be attributed to construction of West Point Lake.

Sources of PCBs and Total DDT. Burdens of PCBs and total DDT, ϕ_{pcb} and ϕ_{ddt}, were calculated for five reservoirs by multiplying concentration for each measured interval by the effective density for that interval to yield mass of contaminant per volume of wet sediment. The mass of contaminant for unanalyzed intervals was estimated by linear interpolation of adjacent measured intervals. Mass per volume was then multiplied by the interval thickness and summed over the length of the core to give ϕ_{pcb} and ϕ_{ddt} in nanograms per square centimeter (Table 2). For comparison, Table 2 lists burdens reported for cores from Lake Superior, rural Wisconsin lakes, and peat bogs, all of which are attributed primarily or solely to atmospheric fallout (21, 28, 29).

Core burdens of PCBs are much greater in the reservoirs than in the natural lakes and bogs (Table 2). PCB burdens in the latter settings range from 0.1 to about 16 ng/cm². In contrast, PCB burdens in the reservoirs range from 54 ng/cm² in Lake Blackshear to 28 000 ng/cm² in Lake Harding. This comparison suggests that only a small fraction of the PCBs in these reservoir cores can be attributed to direct (regional) atmospheric fallout of PCBs on the reservoir surface and indicates that fluvial inputs are predominant.

Reservoir core burdens normalized by sediment FF are also listed in Table 2. ^{137}Cs is supplied to the reservoirs only by atmospheric fallout on the reservoir and its drainage. Normalized burdens, therefore, represent the estimated cumulative atmospheric loading of PCBs and total DDT over the watershed that would be necessary to supply the measured

reservoir core burdens, assuming no local point and non-point sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm^2 in White Rock Lake to 4400 ng/cm^2 in Lake Harding, are still much greater than fallout rates estimated from natural lakes and peat bogs. Lake Seminole, with a mixed land use, has a normalized burden approximately 3 times greater than maximum estimated fallout rates. Only Lake Blackshear, with a normalized burden of 17 ng/cm^2, is comparable to the maximum burdens reported for peat bogs, rural Wisconsin lakes, and Lake Superior. Therefore, we conclude that (1) Lake Blackshear provides an upper estimate of regional atmospheric inputs of PCBs in Georgia; (2) regional atmospheric sources cannot account for the relatively large burdens observed in the other four reservoirs, even when sediment focusing is accounted for; and (3) local point and non-point sources of PCBs contribute significantly to the PCB burdens of cores, especially in the more urban drainages. A similar conclusion was reached by Gschwend et al. (30) for polycyclic aromatic hydrocarbons (PAHs) in cores from remote and urban sites in the northeastern United States. They concluded that the delivery of PAH to urban sites via runoff completely overwhelms the background atmospheric depostion rates indicated for remote sites.

Total DDT burdens in the five reservoirs are 1−2 orders of magnitude larger than burdens in Lake Superior and in peat bogs (Table 2). Unlike Lake Blackshear with PCBs, none of these five reservoirs can be considered relatively unimpacted by local sources of DDT. The smallest normalized burden of the five is for Lake Seminole, and it is about 3 times greater than the maximum reported for Lake Superior and peat bogs. Lakes Blackshear and White Rock are about 4 times greater, Lake Walter F. George is about 6 times greater, and Lake Harding is about 15 times greater. As with PCBs, total DDT reservoir core burdens indicate that fluvial inputs from the drainage basins greatly exceed regional atmospheric fallout directly to the reservoir surfaces.

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