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

PETER C. VAN METRE*
U.S. Geological Survey, 8011 Cameron Road, Austin, Texas 78754

EDWARD CALLENDER
U.S. Geological Survey, Reston, Virginia 22092

CHRISTOPHER C. FULLER
U.S. Geological Survey, Menlo Park, California 94025

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 reconstruing 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 km$^2$, 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 km$^2$ 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 km$^2$ 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 km$^2$ 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 km$^2$ 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 km$^2$ 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 $14 \times 14 \times 20$ 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...
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 137Cs 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 137Cs 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 137Cs peaks with exponential decreases in 137Cs to the sediment surface in each reservoir core indicate relatively little mixing (Figure 2). The ratios of peak 137Cs to surface 137Cs 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 137Cs 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 137Cs in 1952, the peak 137Cs 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 137Cs activities, ϕc, were calculated by multiplying 137Cs 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 ϕc, in picocuries per square centimeter (Table 1).
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, $\phi_f$ (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 $\phi_c$ by $\phi_f$, 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 $\mu$g/kg in a sample deposited in about 1969 and then decrease to 3 $\mu$g/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 $\mu$g/kg (Figure 3). Concentrations in the two most recent samples are 42 and 46 $\mu$g/kg, 88% below the maximum concentration. The largest PCB concentration in Lake Walter F. George is 220 $\mu$g/kg in a sample that was deposited in about 1968. Concentrations decrease very rapidly at shallower depths and are less than 14 $\mu$g/kg in five samples deposited since the late 1970s, a 94% decrease.

**FIGURE 2.** Cesium-137 concentrations in reservoir sediment cores and the atmosphere.

**TABLE 1. Sediment Focusing Data**

<table>
<thead>
<tr>
<th>reservoir</th>
<th>max depth of $^{137}$Cs (cm)</th>
<th>integrated $^{137}$Cs activity, pCi/cm² ($\phi_c$, $\mu$)</th>
<th>$^{137}$Cs fallout activity, pCi/cm² ($\phi_f$)</th>
<th>location of precipitation station used for $\phi_f$</th>
<th>focusing factor (FF) ($\phi_c/\phi_f$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>White Rock Lake</td>
<td>63</td>
<td>11.6 (0.4)</td>
<td>5.1</td>
<td>Dallas, TX</td>
<td>2.3</td>
</tr>
<tr>
<td>Coralville Reservoir*</td>
<td>&gt;155</td>
<td>&gt;49.2 (1.2)</td>
<td>8.8</td>
<td>Iowa City, IA</td>
<td>&gt;5.6</td>
</tr>
<tr>
<td>Lake Harding</td>
<td>125</td>
<td>39.4 (1.4)</td>
<td>6.2</td>
<td>Columbus, GA</td>
<td>6.3</td>
</tr>
<tr>
<td>Lake Walter F. George†</td>
<td>&gt;156</td>
<td>&gt;36.0 (1.5)</td>
<td>6.2</td>
<td>Columbus, GA</td>
<td>&gt;5.8</td>
</tr>
<tr>
<td>Lake Blackshear</td>
<td>58</td>
<td>17.6 (0.8)</td>
<td>5.7</td>
<td>Albany, GA</td>
<td>3.1</td>
</tr>
<tr>
<td>Lake Seminole</td>
<td>100</td>
<td>42.3 (1.5)</td>
<td>6.0</td>
<td>av of Albany and Columbus, GA</td>
<td>7.1</td>
</tr>
</tbody>
</table>


**TABLE 2. Sediment Core Burdens of PCBs and Total DDT**

<table>
<thead>
<tr>
<th>compd</th>
<th>reservoir core burdens (ng/cm²)</th>
<th>published core burdens attributed to atmospheric fallout (ng/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>White Rock Lake</td>
<td>Lake Superior (21)</td>
</tr>
<tr>
<td></td>
<td>Lake Harding</td>
<td>Lake Walter F. George</td>
</tr>
<tr>
<td>PCB</td>
<td>250</td>
<td>28 000</td>
</tr>
<tr>
<td>total DDT</td>
<td>250</td>
<td>2 600</td>
</tr>
<tr>
<td>Normalized for Sediment Focusing*</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PCB</td>
<td>110</td>
<td>4 400</td>
</tr>
<tr>
<td>total DDT</td>
<td>110</td>
<td></td>
</tr>
</tbody>
</table>

| 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, $\phi_f$ (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 $\phi_c$ by $\phi_f$, 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 $\mu$g/kg in a sample deposited in about 1969 and then decrease to 3 $\mu$g/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 $\mu$g/kg (Figure 3). Concentrations in the two most recent samples are 42 and 46 $\mu$g/kg, 88% below the maximum concentration. The largest PCB concentration in Lake Walter F. George is 220 $\mu$g/kg in a sample that was deposited in about 1968. Concentrations decrease very rapidly at shallower depths and are less than 14 $\mu$g/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
be comparable to concentrations in Lake Harding after 1974. If, however, the decreasing trend is caused by West Point Lake intercepting much of the 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...
Sediments at the coring site could, therefore, be a mixture of the Flint River arm of Lake Seminole about 1 km from where the coring site was located. The coring site is on the east side of Lake Seminole. The temporal trend in DDT concentrations in Lake Walter F. George peak at 74 µg/kg in about 1965 by 93% to 2 µg/kg in 1971; its use was banned nationwide in 1972. DDE was detected in all core samples deposited since about 1965 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 67, 74, 79, and 78% of the total DDT measured in samples from Lakes Blackshear, 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 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 maximum years 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 affected 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 has less than 1.0 µg/kg of o,p′-DDE, p,p′-DDT, o,p′-DDD, and o,p′-DDE. The drainage area for Coralville Reservoir is about 50% 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 1972 (26); however, significant urban use as a termiticide and for the control of fire ants continued until at least 1990 (27).

Chlordane was 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 fluxes from the watershed are the most important sources.

Reservoir core burdens normalized by sediment FF are also listed in Table 2. 137Cs 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/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban 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/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² 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/cm², is comparable to the maximum estimated fallout rates. In the three reservoirs most impacted by urban sources. In the three reservoirs most impacted by urban areas, normalized PCB burdens, which range from 110 ng/cm² in White Rock Lake to 4400 ng/cm² in Lake Harding, are still much greater than fall