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Persistence and Distribution of PCBs in the Sediments of a Reservoir (Lake Hartwell, South Carolina)

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PCBs are one of the most persistent and ubiquitous pollutants occurring in the environment today. Since their release to the environment, PCBs have been found in water, air, soil, sediment, and human samples (Erickson 1986). Low levels are now identifiable even in remote sites (Eisenreich 1987; Muir et al. 1988). Once in aquatic systems, PCBs tend to associate with particulate matter which may subsequently settle to the bottom. Thus, lake and river sediments have been found to be a major PCB sink in aquatic systems (Karickhoff 1984; Steen et al. 1978; Eisenreich et al. 1981), as observed for the reservoir system studied here.

Lake Hartwell is a PCB-contaminated reservoir located in the upper Savannah River basin in South Carolina and Georgia. It has received the majority of the PCB contamination from Twelve-Mile River, a major tributary at the top of the reservoir located in the northwestern portion of South Carolina in Pickens County (Billings 1976; DHEC 1977). The purpose of this investigation was to provide information on the lake-wide fate and distribution of PCBs and effects of natural and anthropogenic redistribution processes. Although congener-specific data were generated, space for only total PCB concentrations is available here. Congener-specific distributions will be the subject of a later report.

MATERIALS AND METHODS

Thirteen sediment cores were collected from locations shown in Figure 1 during 1983 and 1984. Cores were taken with a benthos sediment corer (Wildco, Inc) containing 75 x 5 cm (i.d.) polycarbonate tubes. Sample locations were selected by using bathymetry data which located the deepest region in each area. In addition, sediment cores were selectively chosen which contained fine sediment particles (high clay content) with little sand and detrital material. Sampling was conducted in this manner to provide a more uniform composition between sediment samples and to maximize the likelihood of sampling the higher contaminated sediments in each region of the lake (Steen et al. 1978; Choi and

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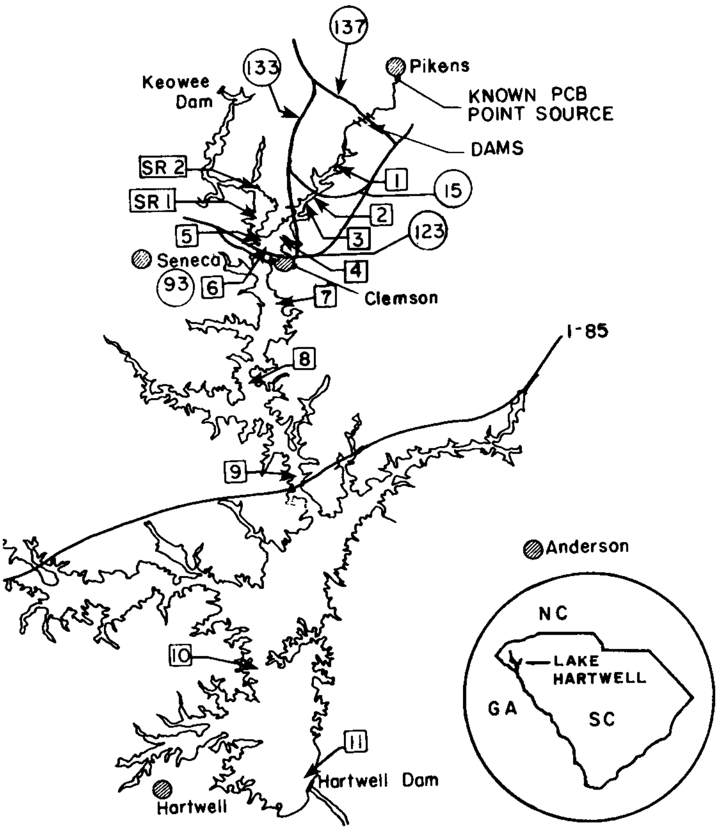

Figure 1. Location of sample sites on Lake Hartwell.

Chen 1976). Core samples were returned to the laboratory, separated into 4-5 cm sections, discarding sediment near the walls of the tube, and stored field-wet in solvent-rinsed glass jars at 4 C until extraction.

Two extraction techniques were used to recover PCBs from sediments. An initial investigation (Polansky 1984) used a steam distillation technique in extraction of sediment samples from Twelve-Mile Creek (sediment cores 1, 2, 3, 5, 6, and SR2 in Figure 1 and Table 1). The steam distillation technique was a modified version of the Nielson-Kryger steam distillation apparatus described by Veith and Kiwus (1977). For a detailed description of the extraction method and sample cleanup used here refer to Swackhammer (1981) or Polansky (1984). A later study (Dunnivant 1985) used a sonication technique for extraction of sediment cores 4, 7, 8, 9, 10, 11, and SR1 (refer to Figure 1 and Table 1). Recoveries for the two extraction procedures were found to be similar with the steam distillation technique

Table 1. Total PCB concentrations for individual core section

Sample	Distance[b] (km)	Total PCB (dry basis) in core section[a] (μg/g) 1	2	3	4	5	6	7	8	9	10
1	29.4	18.1	9.5	9.1	12.0	13.2	13.6	13.6	11.4	24.6	10.0
2	32.2	18.4	19.4	10.3	11.5	55.6	88.5	78.8	61.0	21.3	-
3	33.9	2.6	2.4	11.8	16.8	3.6	2.3	13.1	M	32.9	-
4	35.4	19.4	40.7	55.6	47.5	26.6	31.8	4.6	1.7	0.28	0.27
5	36.8	8.4	15.0	26.2	9.3	4.1	-	-	-	-	-
6	38.0	18.3	14.5	10.7	15.8	6.9	M	-	-	-	-
7	43.0	4.3	5.1	23.4	10.5	1.8	0.15	0.06	0.06	0.03	-
8	51.8	3.9	2.5	2.7	3.3	7.0	9.3	11.7	8.9	2.5	-
9	63.6	1.0	2.1	4.3	3.8	2.2	0.66	0.35	0.28	0.35	-
10	78.2	1.1	0.01	0.03	BDL	-	-	-	-	-	-
11	91.1	0.71	1.1	2.7	6.4	15.4	11.5	8.6	8.8	9.8	-
SR2	NA	0.03	0.03	0.06	0.11	0.26	0.25	0.18	-	-	-
SR1	NA	0.29	3.6	2.0	2.9	5.3	15.1	4.8	-	-	-

M Designates lost sample; - Designates a distance greater than the maximum sample (no sample); BDL Below detection limit (0.03 μg/g;dry basis); [a] = Distance measured downstream from input in Pickens, S.C.; [b] = Increasing core number means increasing depth in sediment, but average core section varied between the two independent sampling trips. Refer to Figure 2 for the average depth of a given section.

recovering approximately 10 percent higher amounts. For a detailed description of the sonication technique and comparisons between the sonication and steam techniques refer to Dunnivant and Elzerman (1988).

Extracted PCBs were separated and quantified on a capillary column gas chromatograph. The chromatography system was a Hewlett-Packard Model 5880A equipped with a ^{63}Ni electron-capture detector (ECD) and a 30-meter DB-5 capillary column with 0.25 mm column i.d. and 0.25 μm film thickness. Peak and/or congener-specific quantification of 64 predetermined peaks was achieved using internal (Aldrin) and external standard calibration. External standards consisted of a mixture of Aroclors 1016, 1242, and 1254 (1:1:1 respectively) which were known to be the primary Aroclors present in Lake Hartwell sediments (Billings 1976). Pure Aroclors (>99%) were obtained from the Quality Assurance Branch of EPA (Cincinnati, OH). For a more detailed description of the quantitation scheme refer to Dunnivant (1985) or Dunnivant and Elzerman (1988).

Due to space considerations and the large volume of data generated by peak-specific capillary-column analysis, only total PCB concentrations will be given here. Total PCB concentrations were calculated by summing approximately 64 peaks present in measurable quantities. Because congeners are subject to differential environmental weathering when released to the environment, calculations of this type should provide a more accurate total concentration than ones based on total area or on only a few peaks. The above quantification scheme allows peak (and generally, congener) specific analyses without the use of mass spectrometer detection, which would greatly increase the cost and complexity of analysis. For this initial investigation, the capillary column GC/ECD technique was determined to be sufficient.

RESULTS AND DISCUSSION

As shown in Figure 1, eleven locations were sampled from potentially contaminated regions of Lake Hartwell (sites 1-11), and two locations from the Seneca River (SR1 and SR2) which at the time of sampling had no known point sources of PCBs. Table 1 is a summary of total PCB concentrations for each sample and core section. All values reported here are dry weight basis.
In order to compare PCB concentrations versus core section depth and site distance from the point source, data from Table 1 are shown in Figure 2. As seen from these data, PCB sediment concentrations from site SR1, initially thought to be from an uncontaminated tributary (Seneca River), contained fairly high levels of PCBs (>1 μg/g dry weight basis). Preliminary investigations indicated the congener composition of PCBs at site SR1 were significantly different from Lake Hartwell sediments downstream of the Twelve-Mile River influent, probably indicating different sources (see below). Possible PCB sources to the Seneca River are abandoned landfills, known to contain waste drums and capacitors, which are located adjacent to the Seneca River and tributaries.

Sample SR2 appears more representative of local "background" concentration. Concentrations of total PCBs ranged from 0.03 to 0.26 μg/g. It was taken below the effluent of a "pristine system", Issaqueena Lake, which has no known PCB point source. As seen from these data, sediment from this site show a gradual increase in PCB concentration with increasing sediment depth. This increase may reflect changes in levels of atmospheric inputs peaking during years of active PCB use.

Data for the Twelve-Mile River sediments, sites 1-11, are also presented in Figure 2 as a function of depth in the sediment. A combination of input history and post-depositional processes and transport has apparently resulted in a complex pattern of PCB concentrations with depth in the sediment and distance from the source. However, rational explanations for these observed distributions can be hypothesized.

Figure 3 shows locations 1 through 11 arranged by increasing distance from the point source (located in Pickens, S.C.) and plots average concentration per core, highest concentration per core, and total PCB in the top 30 cm. In general, PCB concentrations decreased with increasing distance downstream, after site 2, showing an exponentially decreasing trend. Sediment from site 2 contained the highest concentration found to date, with

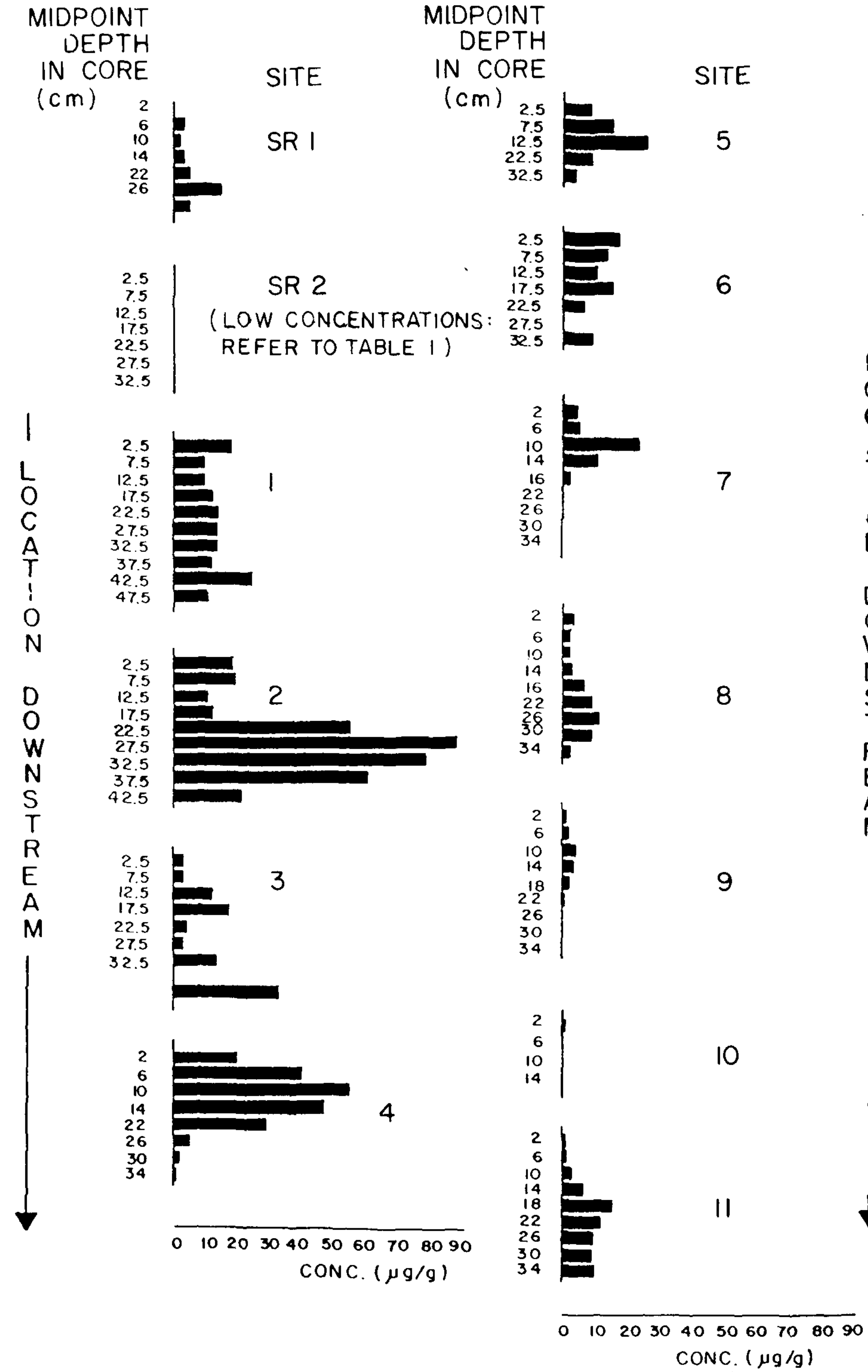

Figure 2. PCB concentration versus depth in core and distance from point source.

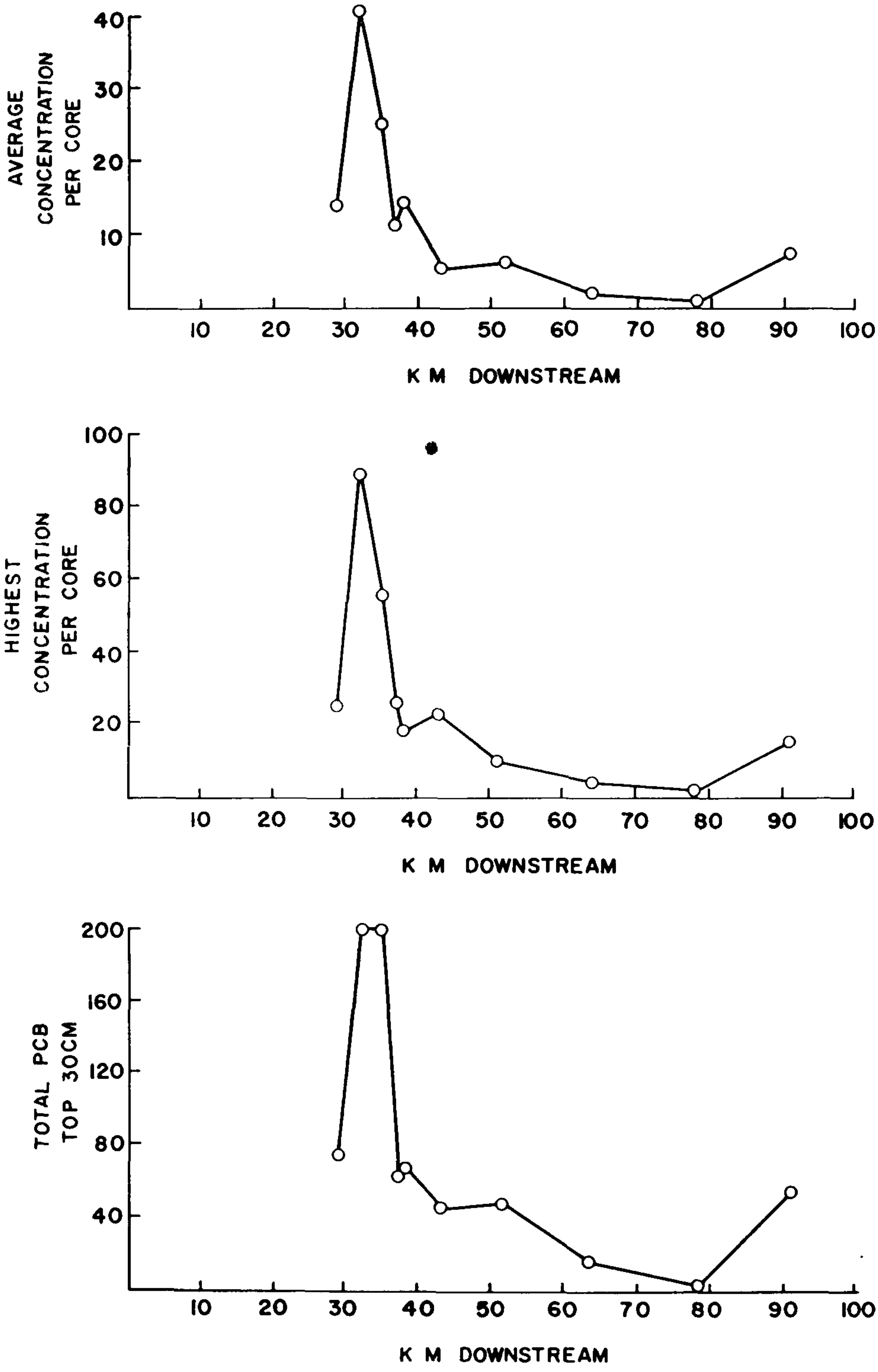

Figure 3. PCB contamination versus distance downstream.

the highest concentration, 88.5 μg/g, occurring at a depth of approximately 27.5 cm in the sediment. Bopp et al. (1981) reported similar decreases with distance from multiple point source emissions in the Hudson River system. Sediments from site 1, the closest to the point source, would normally be expected to contain the highest PCB concentrations. However, due to the physical features of the lake, sedimentation of fine clays known to concentrate PCBs (Steen et al. 1978; Choi and Chen 1976) does not occur until reaching the area surrounding site 2. Around site 2 in the Twelve-Mile River is the first region which acts as a natural sedimentation basin.

Two other sample concentrations which do not ideally fit the exponential decrease characterized in Figure 3 are sites 3 and 11. Site 3 was located in a small cove, offset from the main channel, where it might receive less contaminated sediment and also be influenced by inputs of "clean sediment" resulting from storm drainage of the local watershed. Thus, it can be seen that PCB concentrations in Lake Hartwell sediments are not only determined by proximity to the point source and upstream contamination, but are also dependent on physical features of the lake such as sediment mixing or tributary inputs and local deposition patterns. Site 11 was located just above the Lake Hartwell Dam. Bathymetric and topographic map data suggest this sample was taken from the old river bed that was deposited prior to the filling of Lake Hartwell and during the period of highest PCB inputs. Lower concentrations in the surficial layers at site 11 may reflect more recent transport of sediment from the upper reservoir while greater concentrations at depth may reflect the former river bed. Alternatively, additional unknown sources may affect site 11 or in-lake distribution processes may concentrate fine particles which adsorb relatively higher PCB concentrations at this site.

Conclusive statements concerning the overall distribution of PCBs in Lake Hartwell require more data, however, some additional useful observations can be made. In general, sediment cores showed a gradual increase in PCB concentrations with depth in the sediment, reaching a maximum and then slowly falling off as the underlying pre-reservoir soil material was approached. This is the expected profile for undisturbed sediments resulting from initiation and then reduction of inputs of a refractory compound such as PCBs. Exceptions such as sites 1, 2, and 6 show an increase in PCB concentrations in the surficial sediments. The increases for sites 1 and 2 can probably be explained by activities related to three small power generation dams located upstream of the sample sites in Twelve-Mile River (refer to Figure 1). Sediment collected behind these dams has recently been flushed at irregular intervals, releasing large amounts of sediment which had been accumulating for many years. Data on the level of PCB contamination in these sediments are not available, but the proximity to the point source and probable sedimentation of the smaller, more contaminated particles, behind the dams indicates potentially high levels. Resuspension of contaminated sediment can result in both PCB desorption (Coates and Elzerman 1986) and deposition of contaminated sediments downstream, in this case primarily at sites 1 and 2. Another factor which hampers interpretation of the profile data is sand dredging operations located downstream of the PCB input, but upstream of all sample locations presented here. As mentioned, redistribution of suspended sediments, by dredging or any other mechanism, could also result in irregular sediment patterns downstream.

In summary, high levels of PCBs are still present in Lake Hartwell sediments. The highest level of PCBs were found, in general, closer to the point source and decreased with distance downstream. Data from sediment core profiles indicate that the highest PCB concentrations have been buried, however, surficial sediments still contain elevated levels of PCBs.

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