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Natural Recovery of PCB-Contaminated Sediments at the Sangamo-Weston/Lake Hartwell Superfund Site

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ABSTRACT: The goal of this study was to investigate and identify natural recovery processes responsible for the recovery of polychlorinated biphenyl (PCB)-contaminated sediments at Lake Hartwell, SC. Ten sediment cores were collected from predetermined transect locations. Cores were analyzed by depth for ^{210}Pb , ^{137}Cs , and PCBs. ^{210}Pb and ^{137}Cs were used to date sediments in order to determine sediment accumulation rates and sedimentation rates. Upgradient transect cores were impacted significantly by sediment built up in the headwaters of Lake Hartwell, due to historical sediment releases from upstream impoundments. The highest PCB concentrations were associated with buried silt/clay layers. Downgradient transect cores consisted primarily of silt and were not noticeably impacted by the impoundments. Surface sediments showed decreasing t-PCB concentrations, which approached the target t-PCB cleanup goal of 1.0 mg/kg. The PCB congener compositions became increasingly dominated by lower-chlorinated congeners with sediment depth and corresponding age, resulting in a relative accumulation of *ortho* chlorines and loss of *meta* and *para* chlorines. End member (EM) patterns had characteristics of Aroclors 1248 and 1254, Aroclor 1248 dechlorination via *Process C* (Bedard and Quensen, 1995), and *Process H'* dechlorination (Bedard and Quensen, 1995).

INTRODUCTION

The U.S. EPA's NRMRL is interested in developing effective, inexpensive remediation technologies for contaminated sediments. It has been determined that anaerobic and aerobic biotransformation/biodegradation of PCBs reduces the overall contaminant mass (Quensen and Tiedje, 1997), sorption onto organic particles reduces bioavailability (McGroddy et al., 1996), and sediment containment through natural capping can act as a natural barrier to protect the aquatic environment from contaminated sediments (Cardenas and Lick, 1996). The goal of this study is to better understand the natural mechanisms that contribute to the recovery of PCB-contaminated sediments. The study focuses on the natural recovery of PCB-contaminated sediments at the Sangamo-Weston/Twelve-Mile Creek/Lake Hartwell Superfund site in South Carolina (i.e., the Lake Hartwell site).

In general, natural attenuation of contaminated sediments relies on two primary mechanisms:

- Burial of contaminated sediments with clean sediments, and
- Contaminant weathering.

Sediment burial (capping) acts both to protect the water column from the vertical diffusion and advection of contaminants from near surface sediments, and to reduce contaminant transport into the food chain that can occur through bioturbation and bio-accumulation in surface or near-surface sediments. PCB weathering, which includes such mechanisms as dilution, volatilization, biodegradation, and sequestration, can provide a permanent reduction in levels of PCB contamination and of PCB chlorination. Biological reductive dechlorination under anaerobic conditions preferentially dechlorinates higher-chlorinated PCB congeners, transforming them to lower-chlorinated congeners. Other weathering processes such as dilution and volatilization preferentially remove lower-chlorinated PCBs that are more mobile.

This study focused on evaluating natural sediment capping and contaminant weathering at the Lake Hartwell site. The contributions of the various potential weathering mechanisms were not examined separately, except by careful analysis of PCB congener profiles in aged sediments.

Site Description. The 730-acre (295-hectare) Sangamo Weston/Twelve-Mile Creek/Lake Hartwell site is in Pickens County, SC (U.S. EPA, 1994). The Sangamo-Weston plant was used for capacitor manufacturing from approximately 1955 to 1978. The plant used a variety of dielectric fluids in its manufacturing processes, including ones containing PCBs. Waste disposal practices included land burial of off-specification capacitors and wastewater treatment sludge on the plant site and at six satellite disposal areas. PCBs also were discharged with effluent directly into Town Creek, a tributary of Twelve-Mile Creek, a major tributary of Lake Hartwell. The site was divided into two operable units (OUs). OU 1 addressed the land-based areas (including the Sangamo-Weston land) and six satellite disposal areas. The source of PCB contamination was cleaned as part of the remediation of OU 1. This study pertains to OU 2, which consists of PCB-contaminated sediments at the bottom of Lake Hartwell (Figure 1).

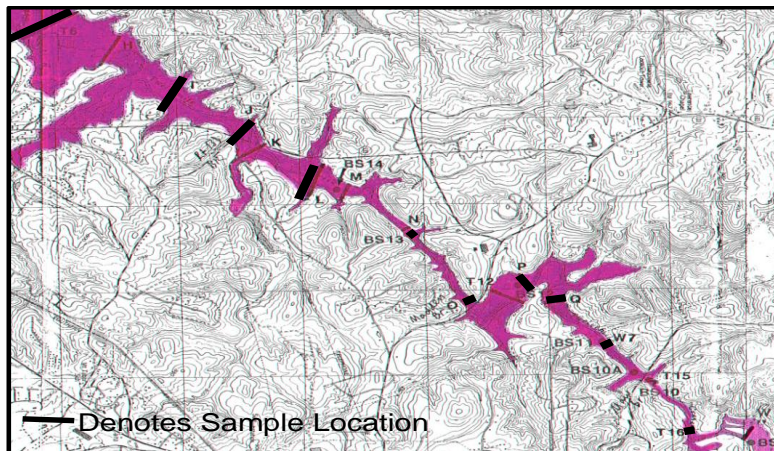


FIGURE 1. Sangamo-Weston/Lake Hartwell/Twelve Mile Creek Superfund site map showing sample transect locations (dark lines transecting the river).

MATERIALS AND METHODS

The field studies included the collection of sediment cores and subsequent chemical analyses at 10 sample locations at the Lake Hartwell site. Cores were collected at pre-determined river transects already established by the U.S. EPA Region 4 and United States Army Corps of Engineers (USACE) under the site's ongoing annual monitoring program (Figure 1). Sediment cores were collected using either 5-cm by 183-cm LexanTM tubes or 5-cm by 244-cm transparent polyvinyl chloride (PVC) pipe. Triplicate sediment

cores were taken at each of the sample locations. Each core was segmented vertically into 5-cm samples and sealed in glass containers. The three cores were used as follows. One core was used for contaminant analyses (Battelle Ocean Sciences Laboratory, Duxbury, Massachusetts), which included PCB concentrations with congener distributions for the Lake Hartwell samples. The second core was used for lead-210 (^{210}Pb) and cesium-137 (^{137}Cs) isotope concentrations (Battelle Marine Sciences Laboratory, Sequim, Washington) for sediment age-dating analyses. The final core was used for PCB partitioning experiments (Battelle Columbus, Ohio). Additional analyses from each sample location included total organic carbon (TOC), grain-size/particle-size distribution, and moisture content (Soil Technology Inc., Bainbridge, Washington).

PCB analyses were conducted using high-resolution gas chromatography with mass spectrometry (GC/MS). The PCB method included the separation and analysis of 107 PCB congeners. ^{210}Pb was analyzed using the alpha counting technique described by Koide et al. (1973); ^{137}Cs concentrations were determined by gamma counting a known amount of sediment (~100 g dry) on a Ge-diode detector.

RESULTS AND DISCUSSION

Sediment cores were obtained from 10 transect locations in Lake Hartwell. To the extent possible, sample cores were taken from the deepest portion of the river. PCB congener analyses were conducted; total PCB concentrations were determined by the sum of individual congeners. Vertical PCB profiles were established at each transect location. Typical PCB profiles are shown for Transect Q (Figure 2) and T6 (Figure 3), from upgradient and downgradient locations in Lake Hartwell.

The most upgradient transects (T16, W7, Q, and P) were impacted significantly by historical releases of sediment built up in the headwaters of a dam located upgradient of the lake. As expected, the highest PCB concentrations were associated with silt layers while sand layers contain very low PCB concentrations. In Transect Q (Figure 2), the top 25 cm of sediment contains less than 93 $\mu\text{g}/\text{kg}$ total PCBs, well below the cleanup goal of 1 mg/kg for surface sediments (U.S. EPA, 1994). Transect T6 (Figure 3), consisting primarily of silt, was not noticeably impacted by the release of silt and sand from the dam, and has a more typical PCB profile, where the highest PCB concentrations in the middle of the core are associated with the period of maximum PCB release into the lake. The top 5, 10, and 15 cm of the core contained 0.93, 2.6, and 8.9 mg/kg. While surface sediment concentrations appeared to be decreasing, at the time of sampling they continued to exceed the 1 mg/kg PCB goal.

PCB Composition Data. PCB composition (i.e., the relative concentrations of PCB congeners) was examined based on PCB homologues (i.e., level of chlorination) and congener data. The PCB homologues and congener compositions became increasingly dominated by lower-chlorinated congeners with sediment depth and corresponding age of the deposited sediments, which is consistent with changes observed in the homologue composition. A significant loss (~45%) of tetra-, penta-, and hexachlorobiphenyl congeners and accumulation of mono-, di-, and trichlorobiphenyls dominated at greater depth. In surface sediment samples, tetrachlorobiphenyls were most abundant, followed by the tri- and pentachlorobiphenyls, then by di- and hexachlorobiphenyls, and mono- and heptachlorobiphenyls.

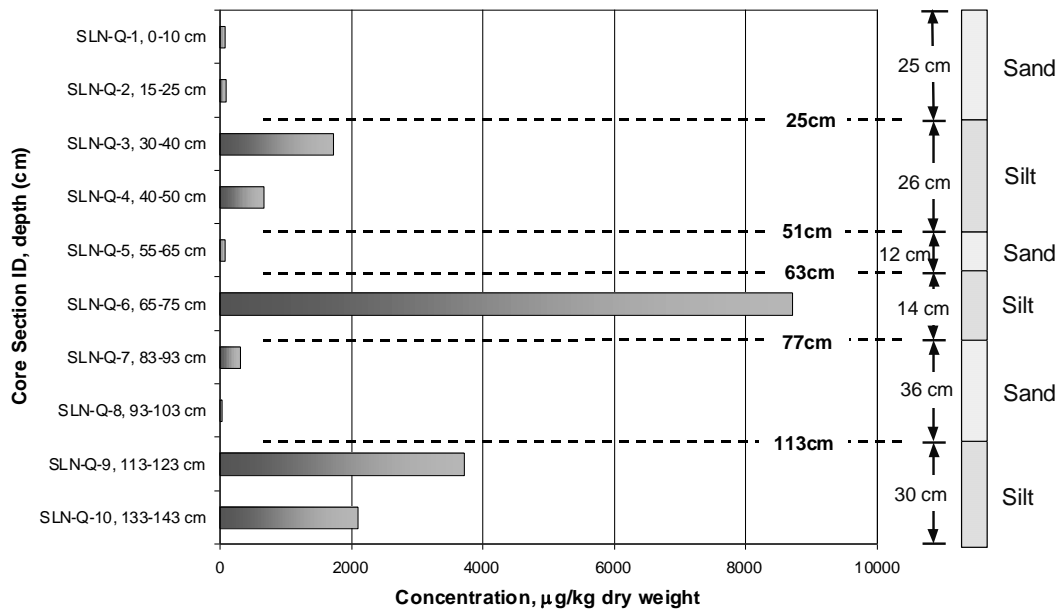


FIGURE 2. Vertical PCB profile from upgradient transect Q. Silt/sand layers are formed in the upgradient portion of the lake due to planned releases of sediment built up behind a dam located upstream of the lake.

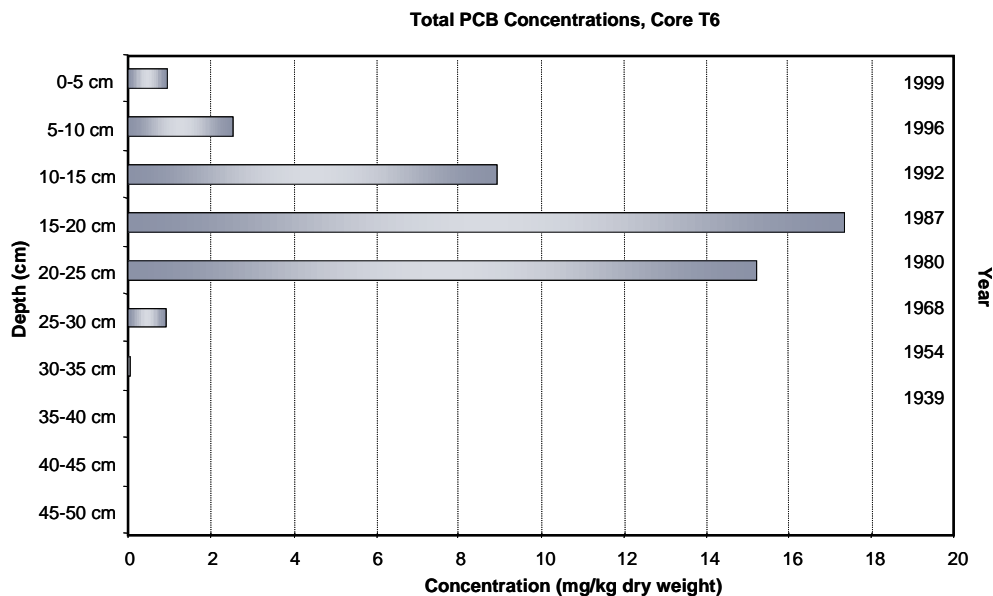


FIGURE 3. t-PCB vertical concentration profile in downgradient Core T6.

Polytopic Vector Analysis. The PCB data generated for this study were modeled using the multivariate statistical method known as Polytopic Vector Analysis (PVA) to identify fingerprint (also known as end-member) compositions from the data generated for Lake Hartwell. End-member (EM) patterns then were compared to source patterns reported in the literature (e.g., Aroclor compositions and known PCB dechlorination or weathering patterns).

Initial results suggest that PVA appeared to resolve three EM patterns. The composition and distribution of EM-1 had characteristics of a mixture of Aroclors 1248 and 1254. EM-2 was dominated by low-chlorinated congeners, including mono-, di-, and trichlorobiphenyls. The congeners that made up EM-2 preferentially exhibited chlorines in the 2 (*ortho*), 4 (*para*) and 6 (*ortho*) positions. The dominance of *ortho*-chlorines suggested that EM-2 was a result of a microbial dechlorination process. EM-3 (End-Member 3) was characterized by di-, tri-, and tetrachlorobiphenyl congeners, but also showed an increase in *ortho*-chlorinated congeners relative to EM-1. EM-3 appears to be an intermediate between EM-1 and EM-2. EM-1 was most abundant in surface sediments, and EM-2 and EM-3 dominated deeper sediment segments.

CONCLUSIONS

Vertical PCB profiles indicate that the sediments are recovering by the slow deposition of increasingly less-contaminated sediment, but that the downgradient reaches of the lake have not yet recovered to the target concentration of 1 mg/kg. The presence of *ortho*-halogenated compounds found in high concentrations provides indications of reductive dechlorination of higher-chlorinated PCBs in deeper sediments. PVA provided an excellent tool to evaluate end-member PCB patterns.

The coring approach demonstrated in this research provides a uniquely effective approach to understanding the history of contaminated sediment deposition, weathering processes, and contaminant sources in the interest of studying natural recovery of PCB-contaminated sediments. This research summarizes just one method of studying recovery of contaminated sediments at Lake Hartwell. In order to fully study PCB-contaminated sediments it is necessary to study other mechanisms that may play a role in sediment recovery and the fate and transport of sediment contaminants, such as bioaccumulation, bioturbation, and volatilization to name a few. In addition, a better understanding is needed of how major events such as 100- or 500-year storms, sea level rise, and human activity impact sediment erosion and deposition.

ACKNOWLEDGMENTS

Funding for this study was provided by the U.S. EPA National Risk Management Research Laboratory under Contract No. 68-C5-0075, Work Assignment 4-30. This technical note summarizes recent research and does not represent U.S. EPA policy.

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