CHARACTERIZATION OF FATE AND TRANSPORT PROCESSES: COMPARING CONTAMINANT RECOVERY WITH BIOLOGICAL ENDPOINT TRENDS

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ABSTRACT

The Remediation Technologies Development Forum (RTDF) Sediment Subgroup is developing a framework for assessing Monitored Natural Recovery (MNR) as a sediment risk-management practice. The RTDF Framework relies on a weight-of-evidence approach, focusing on 1) historical contaminant sources; 2) contaminant fate and transport; 3) temporal contaminant trends in sediment and biological endpoints; and 5) defensible predictive tools to allow prediction of future sediment recovery.

This paper is part of a series of papers co-authored by the RTDF sediment subcommittee, focusing on different elements of the RTDF MNR Framework. It is hoped that his framework will lead to more cost-effective and standardized evaluations of MNR at contaminated sediment sites, which could lead to increased acceptance of MNR by scientists, engineers, site owners, regulators, and the public. Results from the Sangamo-Weston/Twelvemile Creek/Lake Hartwell Superfund Site (Lake Hartwell) are presented. EPA Region 4 has been collecting fish and invertebrates for PCB analyses in Lake Hartwell and Twelvemile Creek since the 1970s. These results are compared with measurements of surface sediment recovery, conducted by EPA National Risk Management Research Laboratory (NRMRL) and Battelle.

Despite the relatively low PCB levels in surface sediments, fish collected from Twelvemile Creek and Lake Hartwell continued to have t-PCB concentrations above the FDA tolerance level of 2 mg/kg; results showed no apparent concentration trends since 1992. However, continued decreases in sediment concentrations could lead to measurable decreases in fish PCB concentrations. Whether the forage fish are impacted solely, or primarily, from surface sediment concentrations, upward advection or diffusion from buried contaminated sediments, or influx of PCBs from an upgradient source, cannot yet be deciphered from the data. Further research is being conducted to elucidate these parameters and their potential impact on forage fish PCB concentrations. The Corbicula clam t-PCB concentrations appeared to correspond closely to surface sediment t-PCB concentrations; lowest t-PCB levels were observed at background stations and the highest concentrations were observed immediately downstream of the former Sangamo-Weston plant.

INTRODUCTION

The Remediation Technologies Development Forum (RTDF) Sediment Subgroup is interested in developing a better understanding of the natural mechanisms that contribute to the recovery of contaminated sediments, and to develop a framework for assessing Monitored Natural Recovery (MNR) as a sediment management practice. The *RTDF Framework* relies on five primary lines of evidence, namely: 1) characterization of historical contaminant sources and controls; 2) characterization of fate and transport processes; 3) compilation of a sufficient historical record for chemicals of interest to evaluate temporal trends; 4) compilation of historical trends in biological endpoint data to corroborate chemical data; and 5) development of acceptable and defensible predictive tools to allow prediction of future sediment recovery.

Natural recovery of contaminated sediments relies on two primary mechanisms (Brenner et al., 2004; Magar, 2001; Stout et al., 2001): (1) natural sediment deposition (burial of contaminated sediments by clean sediments, and (2) weathering of contaminants to less toxic products or less bioavailable forms. Sediment deposition results in capping of contaminated sediments, which works to protect the water column (and resident fish) from the vertical diffusion and advection of contaminants from surface sediments. Natural capping also reduces the risk of resuspension of contaminated surface sediments during high flow events or storms, and reduces the possibility of contaminant transport into the food chain by limiting bioturbation and bioaccumulation in surface or near-surface sediments (Cardenas and Lick, 1996).

Sediment burial occurs in net depositional environments, where the rate of sediment deposition exceeds the rate of sediment scouring and resuspension. Most depositional environments require relatively slow surface water velocities and commonly occur in deltas, lakes, and the slow-moving portions of rivers. Ironically, these depositional environments are likely repositories for contaminated sediment particles; in other words, the same natural sediment transport mechanisms that promise to recover contaminated sediment environments through natural burial were probably the cause of the initial deposition and accumulation of contaminated particles. It is therefore clear that natural depositional environments will continue to accumulate contaminated particles until contaminant sources are terminated, and that source removal must be an integral component of natural recovery. Evaluation of sediment burial requires an assessment of the following processes that may impact sediment dynamics:

- Source attenuation and potential ongoing primary or secondary sources
- Surface sediment recovery via deposition of increasingly clean sediments
- Sediment stability and potential sediment resuspension and migration
- Sediment and water column contaminant fate and transport processes

Evaluation of sediment weathering requires an understanding processes that effect contaminant fate and that may result in contaminant transformation (partial or complete). For buried sediments, the primary weathering mechanisms include biotransformation, sequestration, and benthic mixing.

The RTDF MNR Framework for contaminated sediment sites includes the following five interrelated elements:

I. Characterize Contamination Sources and Controls
II. Characterize Fate and Transport Processes (both sediment and contaminant)
III. Establish Historical Record for Sediment Contaminants (including bed stability)
IV. Corroborate MNR Based on Biological Endpoint(s) Trends
V. Develop Acceptable and Defensible Predictive Tools

Previous reports by Brenner et al. (2004), Magar et al. (2002), and Battelle (2002) demonstrated the contributions of natural capping to the natural recovery of surface sediments at the Sangamo-Weston/Lake Hartwell/Twelvemile Creek Superfund Site. Natural sedimentation of progressively cleaner sediments over time resulted in the gradual recovery of surface sediments from peak concentrations of approximately 40 mg/kg to concentrations below the 1 mg/kg total PCB (t-PCB) cleanup goal (Figure 1). Corresponding reductive dechlorination activity resulted in the transformation of higher-chlorinated PCB congeners (congeners with 4 or more chlorines) to *ortho*-saturated mono-, di-, and trichlorobiphenyl congeners with sediment depth and age (Figure 2).

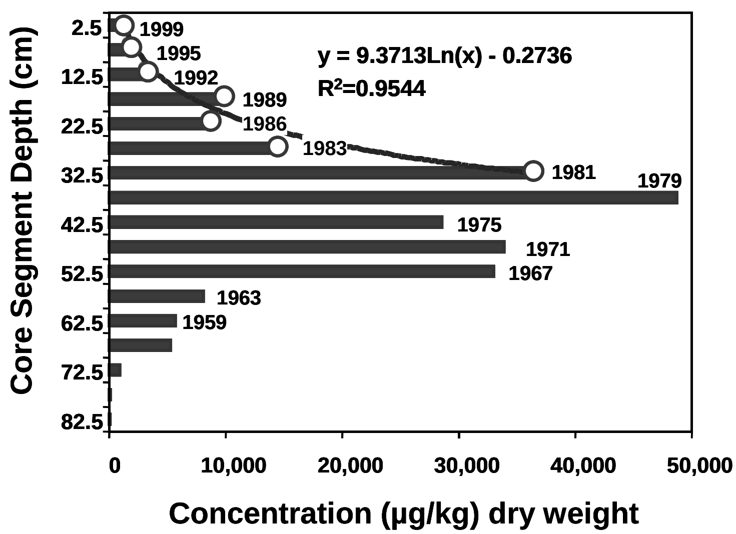

Figure 1. Total PCB concentration and age dating profile at Transect L, Lake Hartwell

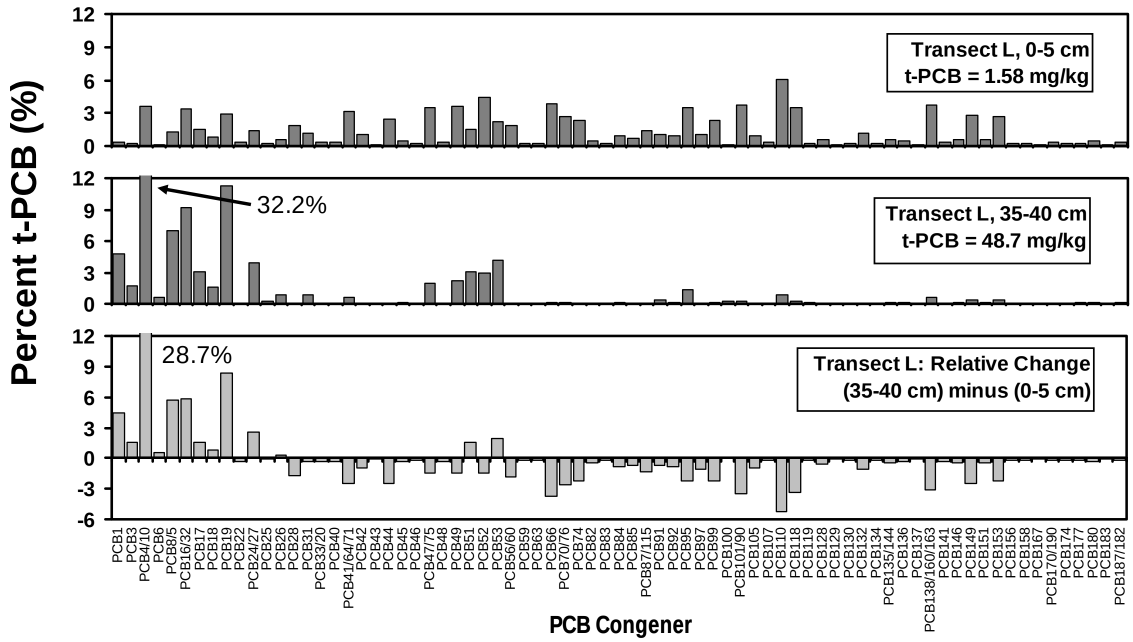

Figure 2. PCB congener distributions in surface and buried Transect L sediments. Bottom figure shows the percent difference between surface and buried sediments, identifying net congener increases and decreases with depth.

This paper combines information on natural recovery processes responsible for the recovery of PCB-contaminated sediments with data from fish and invertebrate studies at Lake Hartwell. We compare the results of the Lake Hartwell sediment Natural Recovery studies (Brenner et al., 2004; Magar et al., 2002) with fish and invertebrate data collected by EPA Region 4 under their annual monitoring program, to examine the extent to which surface sediment recovery has measurably impacted the fish recovery. The results shown in Figures 1 and 2 were reported previously, and involve Categories II and III of the RTDF MNR Framework; the data reported below falls into Category IV.

SITE DESCRIPTION

The 730-acre Sangamo Weston/Twelvemile Creek/Lake Hartwell site is located in Pickens County, SC (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 were released into Town Creek, a tributary of Twelvemile Creek, which is a major tributary of Lake Hartwell, between 1955 and 1977 (EPA, 1994).

MATERIALS AND METHODS

Surface sediment and water samples were collected at six locations. Sampling locations corresponded with former *Corbicula* Clam sample locations previously deployed by the EPA Region 4 (RMT Inc., 1999). Two surface sediment samples were collected from each of the C-0, C-1, C-2, C-3, C-4, C-5, and C-6 locations and labeled with the sample location and A or B for consecutive samples.

Fish and *Corbicula* Clam sampling were conducted by EPA Region 4 between 1995 and 2002, according to procedures described in the EPA Final Record of Decision (ROD) (EPA, 1994). Fish samples were collected at six locations; only two locations, SV-106 and SV-107, are discussed in this paper, due to their proximity to sediment coring locations. SV-107 is located just downstream from EPA Region 4's Transect L, discussed in previous studies (Battelle, 2002). SV-106 is further downstream from Battelle's previous sampling locations, but is discussed in this paper as a comparison to location SV-107. Game fish (i.e., hybrid bass and largemouth bass) were prepared for analyses in accordance with the standard FDA fillet method and were analyzed for PCB Aroclors (EPA SW-846 8082). (Channel Catfish also are monitored by EPA Region 4, but were not included in this paper.) *Corbicula* Clams were placed in baskets for 28 days, then were removed and analyzed for PCB Aroclors (EPA SW-846 8082).

For comparison with the fish and clam results, Battelle and EPA analyzed surface sediment samples for PCBs at each of the clam monitoring locations (C-0 through C-6; Figure 3). PCBs were measured using a congener method (EPA SW-846 8270); t-PCB concentrations were determined by the sum of 107 PCB congeners, for each sample.

RESULTS AND DISCUSION

Figure 3 shows water concentrations, surface sediment sample results, and clam concentrations for Stations C-0 through C-6. Average Hybrid Bass and Largemouth Bass concentrations at Stations SV-106 and SV-107 also are shown. Figure 4 shows Hybrid Bass and Largemouth Bass whole filet concentrations at Stations SV 106 and SV-107.

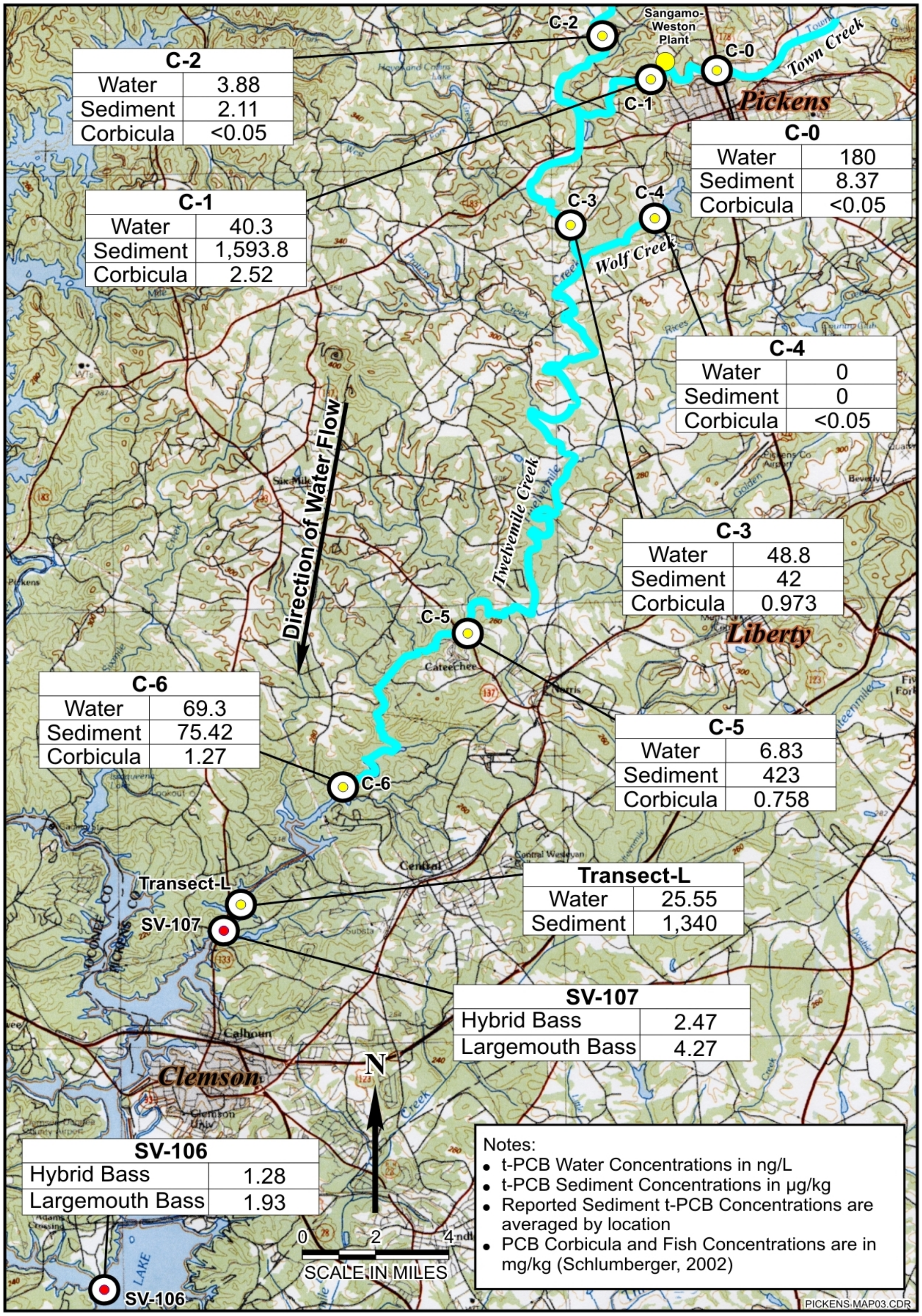

Figure 3. Sangamo-Weston/Lake Hartwell/Twelvemile Creek superfund site map showing EPA Region 4 Corbicula Clam and average fish results for 2002, and EPA and Battelle water and sediment concentrations.

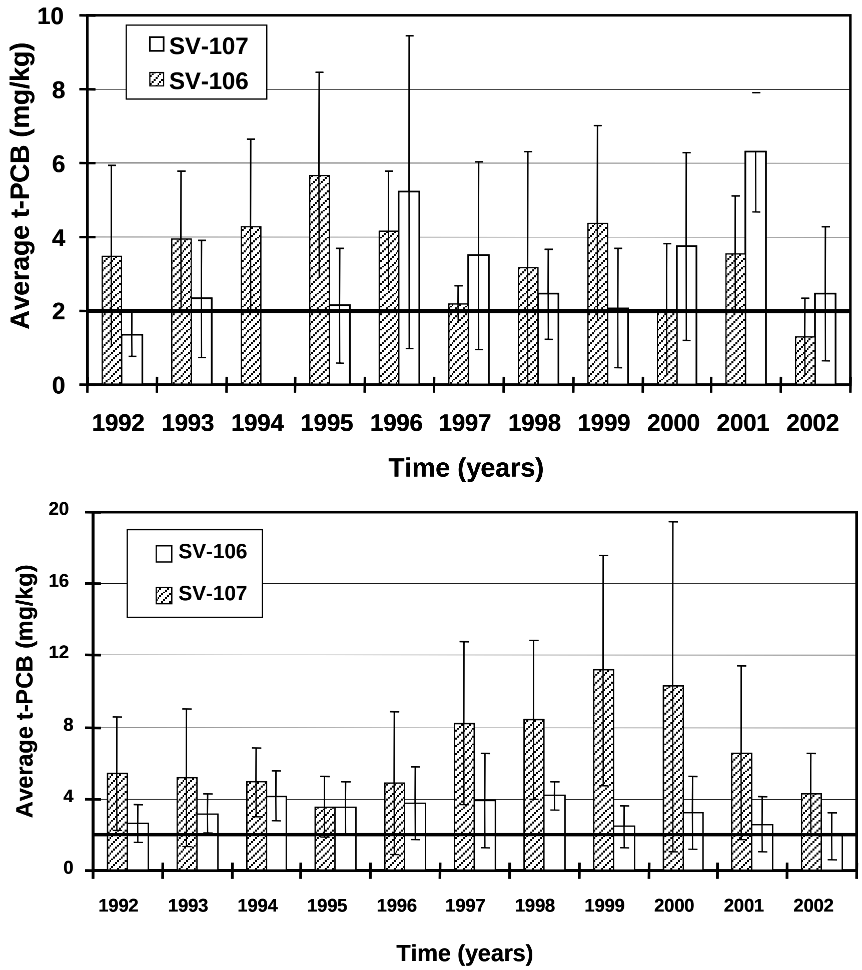

Figure 4. EPA Region 4 Hybrid Bass (top) and Largemouth Bass (bottom) concentrations over time, monitored by EPA Region 4; whole fish filet concentrations are reported for two monitoring stations in Lake Hartwell. (Hybrid Bass were not sampled at SV-107 in 1994.)

Three of the surface sediment sample locations (C-0, C-2, and C-4) were collected upstream from the Sangamo-Weston Plant and represent background samples. All of the samples from these three locations had low t-PCB concentrations ranging from below detection limits to 12.5 μg/kg. All three of these locations also had correspondingly low *Corbicula* PCB concentrations of <0.05 mg/kg (Schlumberger, 2002).

Samples collected at C-1, located directly downstream from the former Sangamo-Weston plant, had the highest PCB concentrations of the surface sediment samples. These were the only two surface sediment samples with concentrations exceeding the 1 mg/kg target clean-up concentration. The C-1 *Corbicula* clam samples also had the highest t-PCB concentration (2.52 mg/kg) of the seven caged *Corbicula* deployments (Schlumberger, 2002). Moving downstream from the Sangamo-Weston plant, the t-PCB concentration decreased at C-3, then increased again at C-5. Core samples collected at C-3 also had low t-PCB concentrations at deeper depths up to 30 cm (data not shown). The surface sediment samples collected furthest downstream, before Lake Hartwell, were collected in Twelvemile Creek at Highway 337 (Maw Bridge). Two surface sediment samples were collected at this location and had t-PCB concentrations of 3.84 and 147 μg/kg, respectively, showing a wide range of concentrations from a single station. The *Corbicula* samples collected at C-3, C-5, and C-6 had PCB concentrations of 0.973, 0.758, and 1.27 mg/kg, respectively (Schlumberger, 2002).

Average Hybrid Bass and Largemouth Bass whole filet concentrations at Stations SV-106 and SV-107 are shown in Figure 4. The historical data show varying concentrations over time (note the relatively large error bars). In general, average t-PCB concentrations remained above the FDA tolerance level of 2 mg/kg, and showed no apparent concentration decrease since 1992. This was a surprising result, because sediment core samples collected by Battelle and EPA in 2000 and 2001 showed substantial decreased surface sediment concentrations; current surface sediment concentrations are approaching the 1 mg/kg surface sediment goal and generally are less than 3 mg/kg in the surface 5 cm (Battelle, 2002). Hybrid bass t-PCB concentrations at both locations SV-107 and SV-106 generally exhibited more variability than the largemouth bass t-PCB concentrations.

CONCLUSIONS

EPA Region 4 has been collecting fish and invertebrates for PCB analyses in Lake Hartwell and Twelvemile Creek since the 1970s. A fish advisory has been in place for Lake Hartwell since 1976 and fish continue to exceed the U.S. Food and Drug Administration (FDA) tolerance level for PCBs in fish of 2.0 mg/kg, despite the substantial historical decrease in PCB sediment concentrations. Continued decreases in sediment concentrations could lead to measurable decreases in fish PCB concentrations. Whether the forage fish are impacted solely, or primarily, from surface sediment concentrations, upward advection or diffusion from buried contaminated sediments, or influx of PCBs from an upgradient source, cannot yet be deciphered from the data. Further research is being conducted to elucidate these parameters and their potential impact on forage fish PCB concentrations.

Surface sediment samples taken from Twelvemile Creek generally have low t-PCB concentrations. The *Corbicula* clam t-PCB concentrations corresponded to surface sediment t-PCB concentrations, with the lowest t-PCB levels at background locations (Stations C-0, C-2, and C-4) and the highest concentrations observed immediately downgradient of the former Sangamo-Weston Plant (Station C-1).

These results were evaluated in accordance with the RTDF MNR Framework. The framework uses a line-of-evidence approach to evaluate the efficacy of MNR as a risk-management remedial alternative. It is hoped that his framework will lead to more cost-effective and standardized evaluations of MNR at contaminated sediment sites, which should lead to increased acceptance of MNR by scientists, engineers, site owners, regulators, and the public.

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