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PROOF OF CONCEPT FOR THE USE OF MACROINVERTEBRATES AS INDICATORS OF POLYCHLORINATED BIPHENYLS (PCB) CONTAMINATION IN LAKE HARTWELL

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Abstract: The US Environmental Protection Agency (USEPA) develops methods and tools for evaluating risk management strategies for sediments contaminated with polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and other legacy pollutants. Monitored natural recovery is a risk management alternative that relies on existing physical, chemical, and biological processes to contain, destroy, and/or reduce the bioavailability or toxicity of in-place contaminants. These naturally occurring processes are monitored to ensure that management and recovery are progressing as expected. One approach frequently used to evaluate the recovery of contaminated sediments and associated biota is the assessment of contaminant tissue levels, or body burden concentrations, in top trophic level fish. In the present study, aquatic invertebrates were examined as an indicator of recent exposure to PCBs. The approach aimed to determine whether invertebrates collected using artificial substrates (i.e., Hester–Dendy samplers) could be used to discriminate among contaminated sites through the analyses of PCBs in whole homogenates of macroinvertebrates. Macroinvertebrates were sorted, preserved, and analyzed for total PCBs (t-PCBs), by summing 107 PCB congeners. Macroinvertebrate body burden concentrations showed similar trends to sediment t-PCB concentrations at the sites sampled. The results indicate that macroinvertebrates can be used to assess sediment contamination among sites that have different PCB contamination levels. *Environ Toxicol Chem* 2015;34:1277–1282. Published 2015 SETAC. This article is a US government work and, as such, is in the public domain in the United States of America.

Keywords: Monitored natural recovery Body burden Macroinvertebrates Polychlorinated biphenyls remediation Contaminated sediments

INTRODUCTION

Aquatic sediments are often the ultimate repository of contaminants in aquatic systems. The US Environmental Protection Agency (USEPA) estimates that approximately 10% (~917 million m^3) of the sediment underlying the country's surface water is sufficiently contaminated to pose potential risks to humans and wildlife [1]. To manage the risk of these sediments, it is critical to understand the fundamental mechanisms responsible for contaminant transport and fate; these mechanisms include chemical, biological, and physical processes. Metrics or approaches for assessing the impacts of sediment remediation are needed to provide quantitative measures of its success.

Monitored natural recovery is one approach toward managing the risk of contaminated sediments [2–7]. In terms of contaminated sediments, monitored natural recovery involves burial, by the deposition of increasingly clean sediments over time (i.e., natural capping). Natural capping reduces the risk of resuspension of contaminated surface sediments and reduces contaminant transport into the food chain by limiting bioturbation of contaminated surface or near-surface sediments [6]. The sediments are monitored to ensure that the management approach is effective. Long-term monitoring strategies are essential to demonstrate that these processes are occurring at a rate that adequately manages the risk of the contaminants remaining in place, and to verify the effectiveness of the approach. Exposures to contaminated sediment occur either in the water column during sediment transport or, more commonly, at the water–sediment interface once the sediment is deposited [3,6].

Monitored natural recovery generally relies on a few primary mechanisms for managing contaminant risk to human and ecological receptors. First and foremost is a reduction in the contaminant's availability to receptor organisms. This reduction in bioavailability can occur through physical processes that isolate the contaminated sediment or by biogeochemical processes that immobilize or degrade the contaminants. For monitored natural recovery, physical isolation is achieved primarily though the natural deposition of uncontaminated or less contaminated sediments on top of the contaminated sediments. This deposited sediment limits resuspension and direct contact of receptors with underlying bedded contaminated sediment [4,6].

Depending on the water body, contaminant exposure in the near surface sediment would be expected to be greatest in the biologically active zone of the sediment. This biologically active layer can vary from as little as 1 cm to 3 cm, to as deep as 1 m to 3 m, depending on the habitat and benthic organisms present [8]. Depending on the physical and hydrological nature of the site, net deposition of sediment in natural water bodies, defined as accretion, can be an on-going process. If the contaminant of concern source has been effectively managed (a term often referred to as source control) and the site is net

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depositional, the contaminated sediment will become physically isolated from the primary receptors in the aquatic system [8]. Biological metrics of exposure, such as tissue concentrations, can consist of biota impacted by the contaminant of concerns or other indicators used to demonstrate exposure or effects. These tissue concentration metrics can use organisms such as fish (which represent both an ecological and human health risk), benthic infauna, or surrogate physical samplers (passive samplers) [4,5]. Ideally, metrics should be chosen to quantify both the direct effects of the contaminant of concern and links to higher trophic levels [9,10]. One indicator adapted to evaluate the long-term recovery of contaminated sediments and associated biota following remediation is the collection of invertebrates using artificial substrate samplers such as Hester–Dendys [11].

In the present study, Hester–Dendy samplers were used to collect and quantify the accumulation of the contaminant of concern (i.e., polychlorinated biphenyls [PCBs]) in aquatic macroinvertebrates. The objective was to determine whether macroinvertebrates collected with Hester–Dendys after a 4-wk deployment and analyzed for PCBs could be used to discriminate among locations with different concentrations of PCBs in the sediment.

METHODS

Study site background

Operable Unit-2 of the Sangamo-Weston, Twelvemile Creek (T), and Lake Hartwell Superfund site in Pickens, South Carolina, USA (i.e., Lake Hartwell and Twelvemile Creek; Figure 1) was used as a field location to develop, test, and validate methods and tools that would support chemical, biological, and physical lines of evidence to evaluate the monitored natural recovery of PCB-contaminated sediments [2]. A capacitor manufacturing plant in Pickens was in operation from 1955 to 1987. The facility primarily used Aroclors 1016, Aroclors 1242, and Aroclors 1254 in its manufacturing activities [2]. The use of PCBs was terminated in 1977, before a USEPA ban on its use in January 1978 [4]. Waste, in the form of capacitors and wastewater treatment sludge, were buried on the plant site and at 6 satellite disposal areas. It has been estimated that the facility discharged more than 18 000 kg of PCBs into Lake Hartwell via Town Creek, a tributary to Twelvemile Creek and Lake Hartwell, during the period of its operation [2].

Lake Hartwell is a US Army Corps of Engineers-managed reservoir located in the northwest corner of South Carolina along the Georgia state line (Figure 1). It was created between 1955 and 1963, when the US Army Corps of Engineers constructed Hartwell Dam on the upper Savannah River, 11 km from the confluence of the Seneca and Tugaloo Rivers. Lake Hartwell extends 78 km and 72 km up the Tugaloo and Seneca Rivers, respectively. At full pool elevation (201 m mean sea level), the lake covers nearly 22 600 ha with a 1548-km shoreline [2].

Sediment and water sampling

Water samples for analysis of PCBs were collected first at each of the sampling sites in 2002 (background, T-M/N, and T-O [Figure 1]) at the middle of the water column during

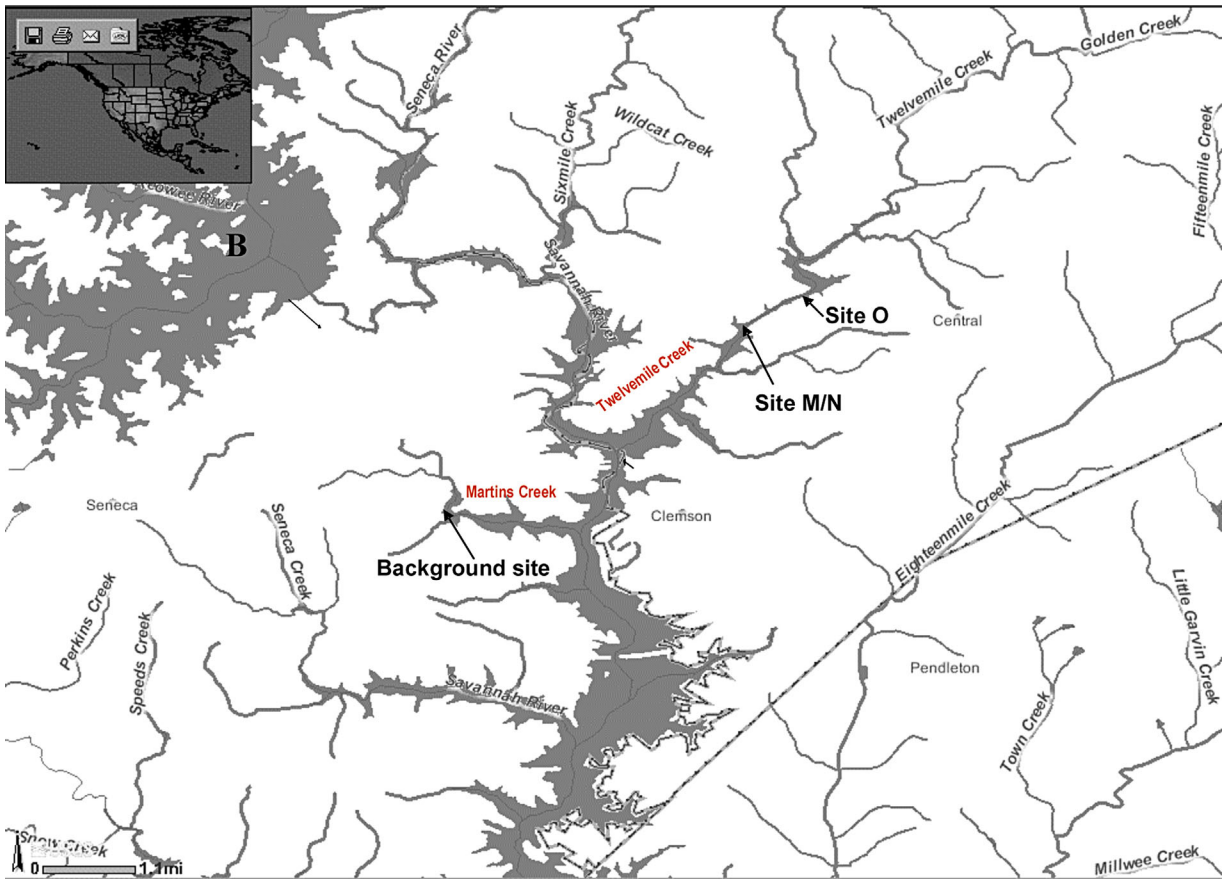

Figure 1. Lake Hartwell map with sample locations.

deployments of Hester–Dendys and again at retrieval of Hester–Dendys. Water was sampled using a peristaltic pump fitted with Teflon-lined polyethylene tubing to minimize sorption losses. A new sampling line was used for each sampling location. Before sampling, the sampling line was purged with approximately 5 L of lake water. Water samples were collected in 2 4-L amber bottles with Teflon-lined lids. Water was also collected before deployment and retrieval of Hester–Dendys in 2006.

Surface sediment was defined as the top 10 cm. This definition was developed during the remedial investigation and feasibility stage of the Superfund project and was followed throughout the present study [2]. Sediment samples were collected after water samples to avoid disturbing sediment fines that would contaminate the water samples. Surface sediment was collected with a Petite Ponar grab sampler using standard practices [2]. Three Ponar grab samples were transferred into a stainless steel mixing bowl and mixed to a consistent color and texture using a clean, stainless steel spoon. The homogenized sediment was transferred into 2 separate 1-dm^3 amber bottles with Teflon-lined lids.

Hester–Dendy methods

Macroinvertebrates were collected using Hester–Dendy multiple-plate artificial substrate samplers [11]. Each sampler unit consisted of 8 7.6-cm^2 plates constructed of 0.32-cm tempered hardboard, separated by 2.5-mm^2 spacers, and held together with an eyebolt and wing nut assembly. The plates and spacers were placed on a 0.635-cm eyebolt so that there were 3 single spaces, 3 double spaces, and 1 triple space between the plates. The components were secured with a wing nut (Figure 2A). The total surface area of each Hester–Dendy unit, excluding the eyebolt, was 924 cm^2 [11].

In 2002, the deployment system consisted of 4 rings wired onto a central chain, with each ring containing 5 individual Hester–Dendy samplers (Figure 2B). The spacing between the rings was approximately 0.6 m. Two deployments were made at each sampling site (Figure 1), yielding a total of 40 Hester–Dendy units at each location. In 2006, only the background and T-O sites were sampled because of increases in the number of Hester–Dendys deployed per site, to maximize the biomass collected. The number of Hester–Dendy samplers per ring was increased to 6, and the number of rings per deployment was increased to 5, for a total of 60 Hester–Dendy units at each site.

Each Hester–Dendy deployment was anchored above with a submerged float to prevent the deployment from coming into contact with the sediment (Figure 2B). The lowest ring was approximately 0.3 m above the sediment surface. After 28 d, the deployments were retrieved and the Hester–Dendy samplers were dismantled. The biofilm and associated macroinvertebrates from the sampler plates were scraped into a container filled with site-specific water. Macroinvertebrates were separated from the water and debris using a 10-μm stainless steel screen, and the samples were further processed by hand removing the macroinvertebrates completely from each plate. All macroinvertebrate specimens regardless of species were consolidated into a 50-cm^3 amber glass bottle with a Teflon-lined lid. The labeled bottles were shipped at 4 °C to the analytical laboratory (Battelle Memorial Institute, Duxbury, MA) for extraction and analysis of PCBs and lipids [3,4,12].

Chemistry methods

Macroinvertebrate and sediment samples were analyzed for 107 PCB congeners. The methods have been previously described [3–5]. Briefly, the PCB analysis was based on a modified version of SW-846 method 8270 [13]. Method 680 of the USEPA (primarily for level of chlorination analysis) [14] and method 1668 A of the USEPA (for individual PCB congener analysis) [12] were used for system calibration.

The analytical system was comprised of a gas chromatograph (Hewlett Packard 5890) equipped with an electronic pressure-controlled inlet and a mass selective detector (Hewlett Packard 5973) operating in the selected ion monitoring mode. A minimum of a 5-point response factor calibration was run with analyte concentrations in the standard solutions ranging from 0.001 ng/mm^3 to 0.005 ng/mm^3 in the low-level calibration standard to approximately 0.5 ng/mm^3 to 1.9 ng/mm^3 in the high-level standard. This method is based on the calibration approach of USEPA method 1668 A [12], which requires calibration of only 2 congeners for each level of the chlorination (i.e., 19 total congeners). The 2 PCB congeners used for each level of chlorination were the first and last eluting congeners within the level, so they also marked the level of chlorination integration window. The average response factor of the 2 congeners in each level of chlorination was used to quantify each of the individual congeners within that level of chlorination. The samples were bracketed by standard checks analyzed at least every 10 samples and at completion of the analysis sequence.

Quantification of individual compounds was obtained by the method of internal standards using internal standard compounds. Total PCBs (t-PCBs) were determined as the sum of the individual PCB congeners. The method detection limits for the PCB analytes were 0.01 ng/g to 0.04 ng/g dry weight in sediment, 0.2 ng/dm^3 to 0.8 ng/dm^3 in water, and 0.02 ng/g to 0.1 ng/g wet weight in tissue [3,4].

RESULTS AND DISCUSSION

In 2002, PCB body burden in macroinvertebrates showed the lowest t-PCBs (88.54 ng/g wet wt) at the background site and increased to 1995 ng/g wet weight t-PCBs at the T-M/N site, and to 2680 ng/g wet weight t-PCBs at the T-O site (Figure 3A). Percentages of lipids [13] were 0.98%, 1.47%, and 1.71% for background, T-M/N, and T-O sites, respectively. Midges were the dominant taxa found on the Hester–Dendys at sites T-M/N and T-O. More macroinvertebrate taxa were found at the background site; however, it too was dominated by midges.

In 2006, only 1 Hester–Dendy deployment was made per site; therefore, there were no replicates for comparison. Because of the increase in the number of Hester–Dendys per sampling location, site T-M/N was not sampled. In 2006, macroinvertebrate t-PCB body burden concentrations were not reflective of the sediment concentrations at site T-O but were much higher than at the background site (Figure 3B). Comparisons of total polychlorinated biphenyl (t-PCB) concentrations in water (μg/L), sediment (ng/g dry wt), and macroinvertebrate tissue (ng/g wet wt) at the background, T-M/N, and T-O sites are shown in Figure 3A. Mean percentage of lipids for the background site was 0.75% and 0.73% for the T-O site.

The t-PCB concentrations in water, sediment, and macroinvertebrates in 2002 are shown in Figure 3A. Very low to nondetectable concentrations in surface water samples were found in 2002 (0.21 ng/L–2.03 ng/L background, 59.7 ng/L–63.3 ng/L T-M/N, and 103.4 ng/L–135.81 ng/L T-O) and 2006 (0.14 ng/L–0 ng/L background and 10.84 ng/L–10.18 ng/L T-O). Surface sediment t-PCB concentrations were lowest at the

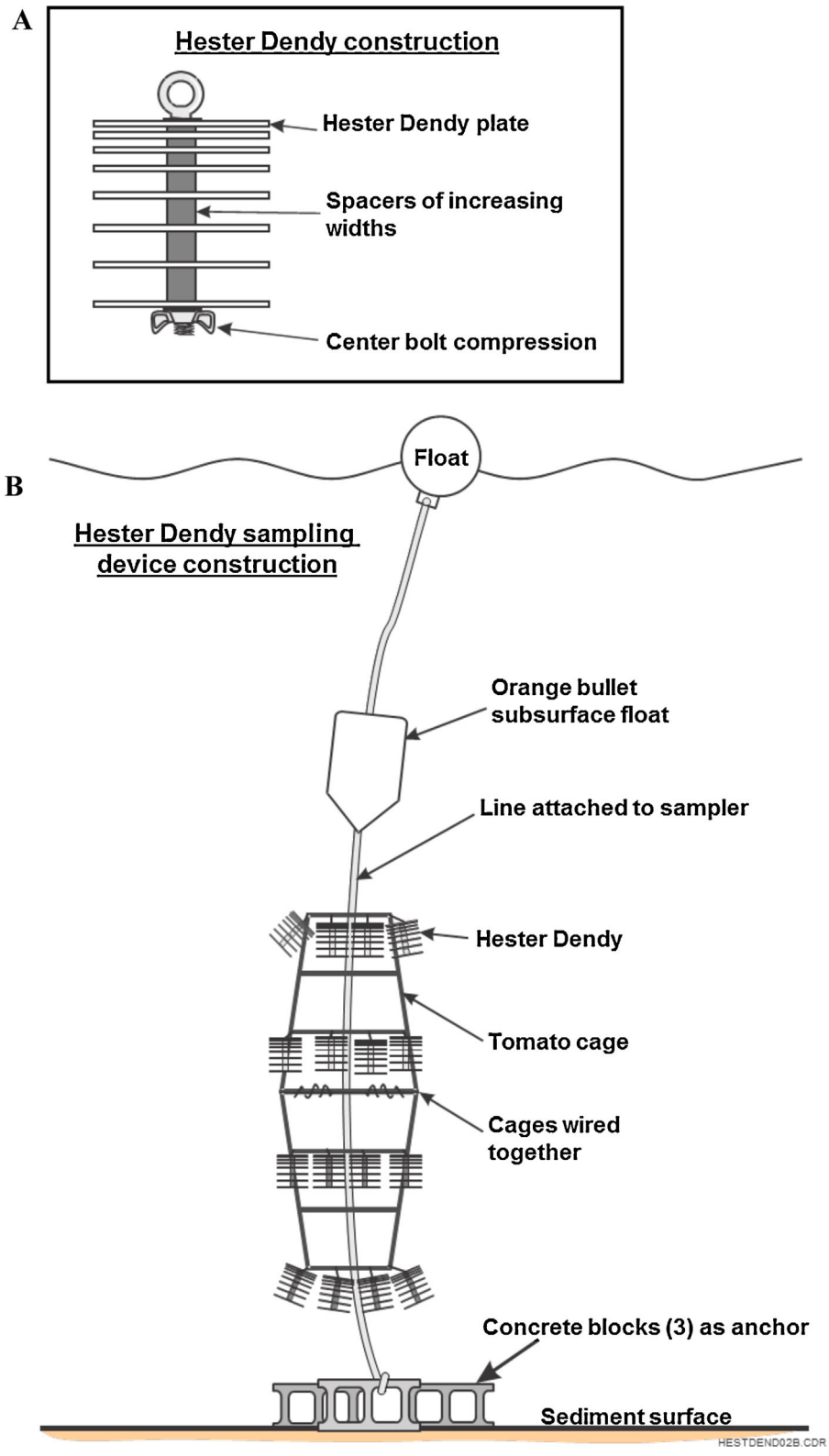

Figure 2. Hester Dendy deployment configuration.

background site and increased from 1154 ng/g at T-M/N to 2154 ng/g at T-O in 2002.

Figure 3B shows the t-PCB concentration results from 2006 water, sediment, and macroinvertebrate samples at the background and T-O sites (Figure 3B). In 2006, macroinvertebrate t-PCB concentrations were not reflective of the sediment concentrations at site T-O; however, it did show concentrations in macroinvertebrates to be much higher than at the background site, a variability that probably occurred as a result of the inherent heterogeneity associated with contaminant of concern concentrations in sediment. In addition, sample wet weights of the macroinvertebrates were much less in 2006 (1 g wet wt at site T-O $<$ 0.1 g background site) than the wet weights of the samples collected in 2002 (4.73 g and 1.22 g at T-O and

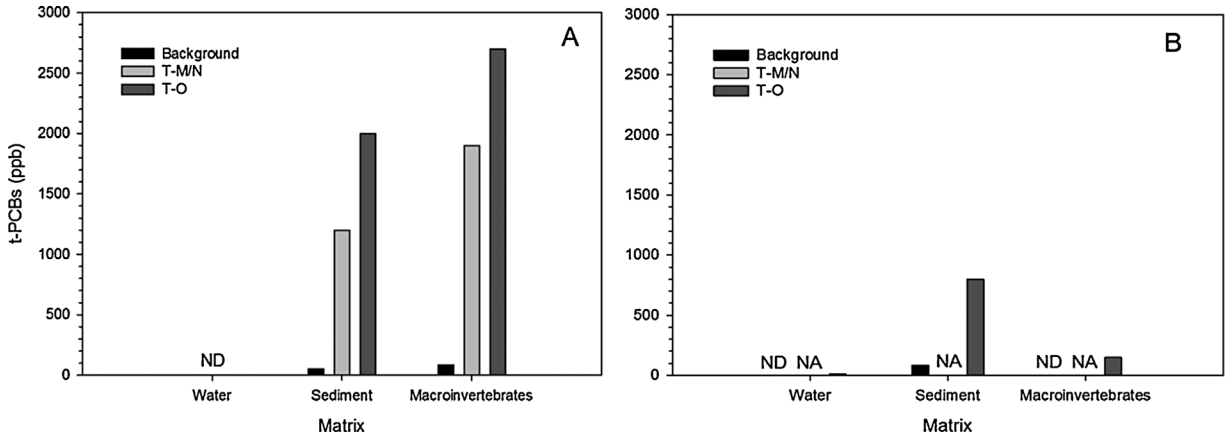

Figure 3. Total polychlorinated biphenyl (t-PCB) concentrations in water (μg/L), sediment (ng/g dry wt), and macroinvertebrate tissue (ng/g wet wt) at the background site, site T-M/N, and site T-O during (**A**) 2002 and (**B**) 2006 sampling events. Site T-M/N was not sampled in 2006. ND = not detected; NA = not analyzed.

background sites, respectively. The differences shown in 2002 versus 2006 could also be because of the collection periods (samples were collected in late August 2002 and May 2006).

Aquatic macroinvertebrates were tested as an indicator of recent exposure. Midge larvae were the dominant taxa found on the Hester–Dendys at sites T-M/N and T-O. More diverse taxa were found at the background site; however, it too was dominated by midges. A majority of the benthic invertebrates, such as midge larvae, annelids (aquatic worms), and mayfly larvae, have life cycles of 30 d to 90 d [15]. In comparison, the fish that are often used for contaminated sediment assessment, remediation evaluation, or fish advisories are usually 3 yr to 10 yr old and are usually more mobile, therefore representing a broader area of exposure. Contaminant tissue levels (i.e., body burden concentrations) in macroinvertebrates represent very recent contaminant exposure levels.

In the present study, our approach was to determine whether macroinvertebrates collected on artificial substrates (i.e., Hester–Dendy samplers) could be used for discrimination among various contaminated sites by comparing the body burden concentrations of macroinvertebrates with sediment concentrations. Macroinvertebrates did show similar trends to sediment t-PCB concentrations between sites in the 2002 sampling season, but some differences in the magnitude of PCBs were evident at site T-O in 2006. The use of Hester–Dendys to collect macroinvertebrates is a useful tool for assessing the biologically active zone; Hester–Dendy sampling also supports the evaluation of monitored natural recovery as a remedy for contaminated sediments. Although the Hester–Dendys were located above the sediment and were colonized by drift organisms, macroinvertebrates are not truly pelagic organisms, as they spend most of their time in the surface sediments. They release from such habitats under unfavorable conditions or in search of more favorable habitat [15]. Our hypothesis was that Hester–Dendys will collect nearby sediment-dwelling organisms and their tissue chemistry would reflect the nearby sediment chemistry. This was demonstrated in our 2002 results. Other factors may have affected our 2006 results, such as deployment location being too far above sediments during high flow or high lake levels and the time of year within the index period in which the Hester–Dendys were deployed. The hypothesis of the present study was further confirmed in a later study conducted on the Ashtabula River in Ohio (USA), in which some of the issues identified above were addressed, that is, location of Hester–Dendys closer to the sediment, surface increase in replication, and staying within the same 8-wk index period each year of deployment [16]. These factors may have also addressed the issue of sufficient biomass for tissue analyses, as there were no biomass issues in the Ashtabula study. However, additional validation is needed to demonstrate that a benthic body burden responds sooner to changes in sediment concentration than tissue concentrations in higher trophic level fish. This is being addressed in a current study on the Ottawa River in Ohio.

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Data availability—All data, associated metadata, and calculation tools are available on request from the corresponding author (lazorchak.jim@epa.gov).

REFERENCES

1. US Environmental Protection Agency. 1998. USEPA's contaminated sediment management strategy. EPA-823-R-98-001 Washington, DC.
2. US Environmental Protection Agency. 1994. USEPA Superfund Record of Decision: Sangamo Weston/Twelvemile Creek/Lake Hartwell Site, Pickens, GA. EPA/ROD/R04-94/178. Final Record of Decision. Washington, DC.
3. Magar VS, Wenning RJ. 2006. The role of monitored natural recovery in sediment remediation. *Integr Environ Assess Manag* 2:66–74.
4. Magar VS, Chadwick DB, Bridges TS, Fuchsman PC, Conder JM, Dekker TJ, Steevens JA, Gustavson KE, Mills MA. 2009. Technical guide. Monitored natural recovery at contaminated sediment sites. Environmental Security Technology Certification Program, Project ER-0622 (2009) 276. US Department of Defense, Washington, DC.
5. Brenner RC, Magar VS, Ickes JA, Foote EA, Abbott JE, Bingler LS, Crecelius EA. 2004. Long-term recovery of PCB-contaminated surface sediments at the Sangamo-Weston/Twelvemile Creek/Lake Hartwell Superfund site. *Environ Sci Technol* 38:2328–2337.
6. Foote E, Fimmen R Darlington R, Neff J, Durell G, Brenner R, Mills M. 2014. Technical resource document on monitored natural recovery. US Environmental Protection Agency, Washington, DC.
7. US Environmental Protection Agency. 2010. Methods and tools for the evaluation of monitored contaminated sediments: Lake Hartwell case study. EPA/600/S-10/006. Washington, DC.
8. Nogaro G, Mermillod-Blondin F, Valett MH, Francois-Carcaillet F, Gaudet JP, Lafont M, Gibert J. 2009. Ecosystem engineering at the

sediment–water interface: Bioturbation and consumer-substrate interaction. *Oecologia* 161:125–138.
9. MacDonald DD, Ingersoll CG, Berger TA. 2000. Development and evaluation of consensus-based sediment quality guidelines for freshwater ecosystems. *Arch Environ Contam Toxicol* 39:20–31.
10. Clarke DG, Palermo MR, Sturgis TC. 2001. Subaqueous cap design: Selection of bioturbation profiles, depths, and process rates. DOER Technical Notes Collection (ERDC TN-DOER-C21). US Army Engineers Research and Development Center, Vicksburg, MS, USA.
11. US Environmental Protection Agency. 1990. Macroinvertebrate field and laboratory methods for evaluating the biological integrity of surface waters. EPA/600/4-90/030. Washington, DC.
12. US Environmental Protection Agency. 1999. Method 1668, revision A: Chlorinated biphenyl congeners in water, soil, sediment, and tissue by HRGC/HRMS. EPA 821-R-00-002. Washington, DC
13. US Environmental Protection Agency. 2004. Semivolatile organic compounds by gas chromatography/mass spectrometry (GC/MS). Method 8270. In: *SW-846, Test Methods for Evaluating Solid Waste, Physical/Chemical Methods*. Washington, DC.
14. US Environmental Protection Agency. 1985 Method 680. Determination of pesticides and PCBs in water in water soil/sediment by GS/MS. Cincinnati, OH, USA.
15. Smith DG. 2001. *Pennak's Freshwater Invertebrates of the United States: Porifera to Crustacea*, 4th ed. John Wiley & Sons, Hoboken, NJ, USA.
16. Meier JR Lazorchak JM, Mills M, Wernsing P, Baumann P. 2015. Monitoring exposure of brown bullheads and benthic macroinvertebrates to sediment contaminants in the Ashtabula River before, during, and after remediation. *Environ Toxicol Chem* 34:1267–1276 (*this issue*).