PROJECT NOTE 1: MIDDLE RIVER COMPLEX SEDIMENT REMEDIATION

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SUBJECT: Monitored Natural Recovery at Contaminated Sediment Sites

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This Project Note provides additional information on processes included in the recommend alternative for sediments adjacent to Lockheed Martin's Middle River Complex (MRC).

The feasibility study project team recommends application of monitored natural recovery (MNR) technology as part of the remedial alternative to manage contaminated sediments in Dark Head Cove. Recommended remedial actions in Dark Head Cove include removal of higher concentration areas adjacent to the bulkhead, *in situ* treatment, and MNR after the application of *in situ* treatment.

Under the recommended alternative, site-wide human health remedial objectives will be met after the remedial construction is completed. Benthic remedial objectives will be achieved at 93% of the areas of concern at the end of the construction. Based on the preliminary calculations and conservative assumptions for the effectiveness of *in situ* treatment (assumed 50% effectiveness when most of the research studies show *in situ* treatment is 75-95% effective for PCBs/PAHs), approximately 3.7 acres of the 8.5 acres of the *in situ* treatment area is predicted to require MNR to achieve point-based benthic remedial goals after the application of *in situ* treatment. The time-frame to reach the benthic remedial goals is estimated to average 6 years after the construction; ranging from 1-13 years at specific locations. These estimates of the need for MNR and the associated time-frames will be refined during the design phase based on the results of bench scale treatability testing of *in situ* treatment effectiveness using MRC sediments. This treatability testing is scheduled for spring 2013.

This Project Note outlines general description of MNR, a partial list of sediment remediation projects where MNR has been applied, and a few case study reviews of projects that utilized MNR as a component of the remedy.

Monitored Natural Recovery:

MNR is recommended as part of the risk reduction approach for contaminated sediment that uses ongoing, naturally occurring processes to contain, destroy, or reduce the

bioavailability or toxicity of contaminants in sediment. MNR usually requires assessment, modeling, and monitoring to demonstrate that risk is actually being reduced. Natural processes that can reduce risk include the following:

- Processes that convert contaminants to less toxic forms (e.g., biodegradation)
- Processes that bind contaminants more tightly to the sediment (e.g., sorption)
- Processes that bury contaminated sediment beneath clean sediment (e.g., sedimentation)

General site conditions that are conducive to MNR include:

- Risk is low to moderate
- Anticipated land uses or new structures are not incompatible with natural recovery
- Natural recovery processes have a reasonable degree of certainty to continue at rates that will contain, destroy, or reduce the bioavailability or toxicity of contaminants within an acceptable time frame
- Expected human exposure is low and/or reasonably controlled by institutional controls
- Site includes sensitive, unique environments that could be irreversibly damaged by capping or dredging
- Sediment bed is reasonably stable and likely to remain so
- Sediment is resistant to resuspension, e.g., cohesive or well-armored sediment
- Contaminant concentrations in biota and in the biologically active zone of sediment are moving towards risk-based goals
- Contaminants readily biodegrade or transform to lower toxicity forms
- Contaminant concentrations are low and cover diffuse areas
- Contaminants have low ability to bioaccumulate

USEPA. 2004. Presenter's Manual for: Remediation of Contaminated Sediments. Office of Solid Waste and Emergency Response, 58 pp. http://www.clu-in.org/download/contaminantfocus/sediments/Presenters-Manual-Dredging.pdf

USEPA. 2005. Contaminated Sediment Remediation Guidance for Hazardous Waste Sites. OSWER 9355.0-85. EPA-540-R-05-012. http://www.epa.gov/superfund/health/conmedia/sediment/guidance.htm

MNR Case Studies:

An MNR Technical Guidance was developed by Environmental Security Technology Certification Program (ESTCP) through collaboration with DOD, EPA, and the Navy (ESCTP 2009). This MNR Guidance document presents more than a dozen case study sites for which MNR was evaluated and selected as the approved remedy or as a remedy component. Natural recovery timelines usually ranged from 5–30 years, and costs associated with MNR usually were orders of magnitude lower than those associated with dredging and capping. A summary of remedy selection at selected case study sites, the status as of 2008 and the success of MNR is presented in below tables, which were extracted from the 2009 MNR guidance document:

TABLE A-4. Monitoring design, current (2008) status, and current view of MNR success at MNR sites (page 1 of 3).

Site Name, Operable Unit/Subarea	Monitoring Objective	Monitoring Element	Current (2008) Status	MNR Viewed as Success?
Bellingham Bay, Whatcom Waterway	Physical isolation	Bathymetric surveys Sediment cores Visual inspections of intertidal and shoreline areas	An engineering design report describing long-term monitoring plan details is expected in 2009 or 2010. Monitoring data since the early 1970s show that natural sedimentation has occurred at significant rates and that mercury levels in surface sediments have decreased.	Yes
	Risk reduction	Surface sediment chemistry Mercury bioaccumulation in Dungeness crabs		
Bremerton Naval Complex, OU B	Physical isolation	Bathymetric surveys and modeling	Results of 2005 monitoring event indicate PCB concentrations continue to exceed cleanup levels. Monitoring is expected to extend until 2017, with at least four more sampling events planned.	Not yet determined
	Risk reduction	Surface sediment chemistry		
Commencement Bay, Nearshore/ Tideflats	Physical isolation	Sediment coring and vertical profiling Radioisotope analysis and sediment age dating Surface sediment chemistry and grain size	Area B of Sitcum Waterway: cleanup levels have been achieved with natural recovery, and the long-term monitoring therefore was deemed complete in 2004. Information regarding MNR in Hylebos Waterway is not available. Baseline monitoring has been performed in Thea Foss and Wheeler-Osgood Waterways, with long-term monitoring planned to begin in 2008.	Yes, where sufficient monitoring data have been collected (Sitcum Waterway)
	Chemical transformation	PAH fingerprint analysis to assess vertical/lateral profiles and trends in chemical transformation		
	Risk reduction	Visual inspection of exposed tideflats to document benthic burrowing activity Biota tissue analysis		
Elizabeth Mine	Risk reduction	Surface sediment chemistry Sediment toxicity analysis	No long-term monitoring program has been developed as of January 2008. Monitoring is expected to occur after upland remediation has been completed.	Not yet determined

TABLE A-4. Monitoring design, current (2008) status, and current view of MNR success at MNR sites (page 2 of 3).

Site Name, Operable Unit/Subarea	Monitoring Objective	Monitoring Element	Current (2008) Status	MNR Viewed as Success?
Hackensack River, Study Area 7	Physical isolation	Tide gauge monitoring to model shear forces Bathymetric surveys Sediment profile imagery to assess erosion	Baseline monitoring scheduled	Not yet determined
	Risk reduction	Pore water chemistry		
James River	Risk reduction	Monitoring of Kepone in fish tissue	Continued low-level contamination in fish tissue, below action level. Fish consumption advisory remains in effect but is less stringent than for PCBs in the same area (from other sources).	Yes
Ketchikan Pulp Company, Ward Cove	Risk reduction	Surface sediment chemistry Sediment toxicity analysis Characterization of benthic communities	MNR is functioning as intended. Recovery is sufficient to suggest cessation of monitoring in some areas.	Yes
Koppers Co., Inc., Barge Canal	Physical isolation	Bathymetric surveys Aerial photography to document sedimentation and vegetation encroachment	PAH concentrations have been decreasing. Lateral encroachment of shoreline vegetation has been observed in analysis of aerial photographs, confirming sedimentation. Bathymetric surveys show net sediment accumulation within the Barge Canal (0.5-2 feet accumulation from 2000–2004). Second Five-Year Review Report (2008) recommends discontinuing further monitoring in the Barge Canal.	Yes
	Risk reduction	Surface sediment chemistry		
Lavaca Bay	Risk reduction	Monitoring of mercury in fish tissue Surface sediment chemistry	Concentrations of mercury in surface sediments are achieving cleanup levels. Tissue concentrations of mercury in fish and crab have exhibited annual fluctuations but remain elevated compared to concentrations in the reference area.	Not yet determined

TABLE A-4. Monitoring design, current (2008) status, and current view of MNR success at MNR sites (page 3 of 3).

Site Name, Operable Unit/Subarea	Monitoring Objective	Monitoring Element	Current (2008) Status	MNR Viewed as Success?
Lower Fox River/Green Bay, OU2 and OU5	Risk reduction	Surface water quality Fish and waterfowl tissue sampling for human receptor risks Fish, bird, and zebra mussel tissue sampling for ecological receptor risks Surface sediment chemistry Population studies of bald eagles and double-crested cormorants for reproductive viability	Baseline monitoring of PCB concentrations in MNR-designated areas was completed in August 2007. The MNR program will be finalized in June 2009.	Not yet determined
Mississippi River Pool 15	Physical isolation	Not yet developed	As of October 2008, the MNR program that will be implemented during the Remedial Action phase has not yet been developed. Fish studies conducted prior to remedy selection demonstrated a decreasing trend in PCB levels in fish collected along the Alcoa-Davenport Works facility shoreline.	Not yet determined
	Risk reduction	Monitoring of PCBs in fish tissue		
Sangamo Weston/ Twelve-Mile Creek/ Lake Hartwell, OU2	Risk reduction	Deployment and tissue analysis of caged clams Monitoring of PCBs in fish tissue Surface water and surface sediment chemistry	General trends indicate significant reductions of PCB concentrations in surface sediment. The majority of surficial sediments in the Twelve-Mile Creek Arm of Lake Hartwell will achieve the 1 milligram per kilogram (mg/kg) cleanup level between 2007 and 2011. However, edible fish from Lake Hartwell continue to exceed the FDA tolerance limit for PCBs (2 mg/kg). It is suspected that groundwater contaminated with PCBs is a continuing source.	Mixed results due to incomplete source control
Wyckoff/Eagle Harbor, West Harbor Intertidal	Risk reduction	Biota collection and body burden analysis Surface and deep sediment chemistry	The implemented MNR remedy is achieving remedial goals.	Yes

Environmental Security Technology Certification Program (ESTCP). 2009. Magar, V.S., D.B. Chadwick, T.S. Bridges, P.C. Fuchsman, J.M. Conder, T.J. Dekker, J.A. Steevens, K.E. Gustavson, and M.A. Mills., Project ER-0622, 276 pp, May 2009. http://www.epa.gov/superfund/health/conmedia/sediment/pdfs/MNR_Guidance.pdf

Since 2008, additional monitoring results were reported at some of these sites. Selected publications about these updates and information about other remediation projects where MNR is part of the remedy are presented below:

Lower Fox River, Green Bay:

At the Fox River site, the 2002 ROD for OU1 recognized that although active remediation would achieve a sediment remediation goal of approximately 0.25 ppm PCBs at construction completion, it was estimated that it would take another 14 years before reduced PCB levels in fish tissue would allow relatively safe consumption of walleye for high-intake consumers. The ROD also recognized that if the remedy did not achieve the surface weighted average concentration (SWAC) goal, longer natural recovery periods would be required to meet RAOs. Recent fish tissue sampling has indicated that fish tissue levels are declining at greater levels than predicted. These results are summarized in the attached OU1 Post Remediation Executive Summary: (note each of the inserted pdf files indicated by icons are attached to the end of this document)

OU1 Executive Summary 2011-03-29

Sangamo Weston, Inc./Twelve-Mile Creek/Lake Hartwell, South Carolina:

One of the sites that have been highlighted by EPA is the Lake Hartwell site in South Carolina. A link to the EPA Region 4 website for background information:

http://epa.gov/region4/superfund/sites/npl/southcarolina/sangsc.html

Following paper evaluates MNR as a remedy at this site:

Lake-Hartwell-sed-recovery-06.pdf

Lavaca Bay, Texas:

EPA signed the Record of Decision (ROD) for Alcoa/Lavaca Bay Superfund Site is located in Calhoun County, Texas on December 20, 2001. Several remedial options were implemented at Lavaca Bay including source control, institutional controls, dredging and backfilling, enhanced natural recovery and MNR. It is estimated that surficial sediment mercury levels in all areas are

expected to decline to levels in the current range of open areas of the Bay within a 5 to 10 year time frame. The Five Year Review completed in June 2011determined that the remedy implemented at the Site currently protects human health and the environment. Natural recovery rate at some areas is determined to be slower than predicted due to ongoing sources. The report Five Year Review report identified recommendations that need to be implemented for the remedy to remain protective in the long term.

http://www.epa.gov/region6/6sf/pdffiles/alcoa-bay-tx.pdf
http://www.epa.gov/region6/6sf/pdffiles/lavaca-bay-1st-5yr-review-june2011.pdf

Wyckoff/Eagle Harbor, Washington:

Year 17 monitoring results of implemented remedies at Wyckoff/Eagle Harbor Superfund Site has been recently presented. The monitored natural recovery has shown orders-of-magnitude decreases in PAH levels in the 10 years since the last monitoring:

Battelle 2013_Wyckoff MNR a

Alaska Pulp Mill, Sawmill Cove, Alaska:

At Alaska Pulp Mill site, the Alaska Department of Environmental Conservation (ADEC) determined that the RAO would best be obtained by natural recovery with long-term monitoring every 10 years. The ultimate goal is to have 75 percent of the AOC in an equilibrium community by the year 2040. Most recent monitoring results indicate that natural recovery in the benthic ecosystem is occurring faster than originally predicted:

Battelle 2013_Alaska Pulp Mill MNR abstract

Onondaga Lake, New York:

MNR is selected as part of the remedy at Onondaga Lake to achieve the remedial action goals. Most recent monitoring data support the predicted performance of MNR:

Battelle 2013_Onondaga Lake

Lake Allegan, Michigan:

Lake Allegan, a Kalamazoo River hydropower impoundment, covers 1,690 acres and contains approximately 75% percent of the PCBs in river sediment of the Kalamazoo River Superfund Site. Source control actions together with natural recovery processes have significantly reduced the PCB loading, leading to attenuation of PCB in surface water, fish, and surface sediments.

Battelle 2013_Lake Allegan MNR abstract

Long Island Sound:

In Long Island Sound, a comprehensive study was initiated in 1982 to evaluate the environmental consequences of dredged material placement under aquatic conditions. The results of MNR nearly thirty years after the experimental aquatic placement of uncapped dredged material in Long Island Sound were presented. Thirty years of MNR, through bioturbation and ambient sedimentation result that there is now little biological or chemical difference relative to reference area sediments:

Battelle 2013_Long Island MNR abstract.

REFERENCES IN THE ORDER APPEARED IN THE TEXT

Wisconsin Department of
Natural Resources
101 S. Webster Street
Madison, Wisconsin

March 29, 2011

Lower Fox River Operable Unit 1 Post-Remediation Executive Summary

By the Agencies/Oversight Team

BOLDT

FINDINGS

BACKGROUND

The Record of Decision (ROD) issued for Operable Unit 1 (OU1), also known as Little Lake Buttes des Morts, based its polychlorinated biphenyls (PCBs) remedy on attaining sediment concentrations that corresponded with expected risk reductions to human health and ecological factors. The ROD called for remediation of all sediment that was contaminated with PCB concentrations greater than 1.0 parts per million (ppm or mg/kg) on a dry weight basis. The remedy also specified that all targeted sediment be removed, covered, and/or capped.

The OU1 remedy was implemented from 2004 through 2009 and resulted in a reduction of PCB concentrations in 2010 for the three media of interest: fish, sediment, and water. Natural recovery was occurring in these media pre-remedy, i.e., the PCB concentrations in fish, sediment, and water were declining; however, the remedy has markedly accelerated the rate of decline for PCB concentrations in all three media.

The following comparative analyses were performed on natural recovery data collected prior to 2004 and Long-Term Monitoring (LTM) results collected in 2010. The required baseline monitoring program collected samples in 2006/2007 but was not used in this analysis since this data was collected in the middle of the remedial action. The baseline monitoring program results showed elevated concentrations, which may have been due to OU1's ongoing remedial action.

These elevated results were expected and have been documented at other large dredging and remedial projects. PCB concentrations for fish and water increased above background levels during active remediation for fish and water but declined rapidly to substantively lower than expected levels post-remedy. If the following analysis had used the baseline monitoring program results for comparison, the reduction percentages for fish and water would show greater improvements.

FISH

PCB concentrations in walleye fillets decreased an average of 73% as a result of the sediment remediation as shown in Figure 1. Walleye were selected as the primary indicator species for the long-term monitoring program.

The primary concern, regarding PCBs in the Lower Fox River, is human health risks directly associated with consumption of fish from OU1. The current fish consumption advisory for walleye states: "Eat no more than one (1) meal per month or no more than 12 meals per year." This consumption advisory is based on PCB concentrations in walleye over time. Consumption advisories were developed using several criteria with fish tissue concentrations as one of the key components.

For unlimited fish consumption, the Wisconsin Department of Natural Resources generally uses a PCBs' threshold concentration of less than 0.05 ppm. Prior to the remedy, 3 of 79 walleye (4%) collected from OU1 had concentrations less than 0.05 ppm. After the remedy (2010), 24 of 27 walleye (89%) from OU1 had concentrations less than 0.05 ppm. The average PCB concentration for these 27 walleye is 0.03 ppm. The 2010 walleye results are very encouraging and will be utilized for future analyses in the State of Wisconsin's fish consumption advisory process.

For walleye, the ROD remedy versus natural recovery reduced the PCB fish tissue concentration by 73%. That is, the natural recovery remedy for walleye would reach this same level of PCB fish tissue concentration in approximately 15 to 20 years. The accelerated reduction effected by the ROD remedy is based on full time data set records from 1990 through 2003.

Note: As has been observed at other large dredging and remedial projects, fish-tissue PCB concentrations increased above background levels during dredging but declined rapidly to substantively lower than expected levels post-remedy. OU1's fish tissue PCB concentrations responded to the ROD remedy in a way that is consistent with other large dredging sites. E.g., at Bryant Mill Pond, part of the Allied Paper/Kalamazoo River/Portage Creek Superfund Site, PCB concentrations increased in fish captured during the years in which dredging occurred, but subsequently declined within one year to levels lower than expected with natural recovery. Similarly, at the Hudson River PCBs superfund site, PCB concentrations increased during dredging, but also declined in the first year of monitoring post-dredging. PCB concentrations in fish and water at OU1 appear to have exhibited the same general behavior, showing small increases during dredging but then declining quickly thereafter illustrating the benefit of the remedy.

SEDIMENT

The PCB concentration in the sediment was reduced an average of 94%, from an average of 3.7 ppm pre-remedy to 0.23 ppm post-remedy. See Figure 2.

The surface weighted average concentration (SWAC) in the top four inches of sediment was 3.7 ppm pre-remedy. The ROD specified a post-remedy SWAC of 0.25 ppm. The SWAC, measured immediately post-remedy (2009), was 0.23 ppm. The PCB concentrations, from 63 samples collected in 2010 by the USEPA Region 5 FIELDS Group, was 0.26 ppm which is not statistically different from the goal of 0.25 ppm or the 0.23 ppm measured post-remedy. Note the cap areas were not included in the FIELDS data set due to a paucity of soft sediments covering the caps. The expected PCB concentrations for the cap areas are less than 0.25 ppm.

WATER

For OU1's water column PCB concentrations, the post-remedy (2010) results are significantly lower than pre-remedy (1998) results and even more of a reduction when compared to the baseline monitoring (2006/2007) results. Since the most recent pre-remedy monitoring period (1998) was 12 years prior to remedial activities, and because laboratory analysis and field sampling methods varied among studies over the last few decades; the percentage change effected by the remedy, relative to expectations, cannot be reliably estimated for water column PCB concentrations. See Figure 3.

Note: As has been observed at other large dredging and remedial projects, water-column PCB concentrations increased above background levels during dredging but declined rapidly to substantively lower than expected levels post-remedy.

CONCLUSION

The 2010 post-remedy results for fish, sediment, and water show substantive improvements over natural recovery; however, a full understanding of the effects of the ROD remedy will be accomplished through the observation of PCB concentrations in these media as more data is collected through the long-term monitoring program over a period of years.

Introduction:

The Lower Fox River (LFR) is one of the most industrialized rivers in Wisconsin. It has experienced water quality problems related to municipal, industrial and non-point sources of contaminants since the early 1900s. Thick algal blooms and fish kills, due to heavy loads of organics and nutrients, were common up until the implementation of the **Federal Water Pollution Control Amendments of 1972 (**Clean Water Act).

PCBs were discovered in the LFR in the 1970s. PCB discharges from manufacturing and recycling of carbonless copy paper began in 1954. The LFR Site was identified under the Comprehensive Environmental Response, Compensation, and Liability Act of 1980 (CERCLA) in June 1997. The site for management purposes was divided into five (5) operable units (OUs). The LFR's most southerly and upstream section (from the outlet of Lake Winnebago to the Upper Appleton Dam) consisted of Little Lake Buttes des Morts (LLBdM) and was identified as Operable Unit 1 (OU1).

Fish PCB consumption advisories were in place for LLBdM since 1976. Historically walleye have had concentrations greater than 3.5 ppm (3.5 mg/kg) while the concentration in similar sized walleye in Lake Winnebago, the lake immediately upstream of LLBdM, have had concentrations less than 0.05 ppm (0.05 mg/kg).

Following an extensive Risk Assessment, Remedial Investigation (RI), and Feasibility Study a Record of Decision (ROD) was issued in December 2002 for OU1. The ROD established a sediment remediation standard designed to reduce human health exposure risks to an acceptable level. The goal established in the ROD was to eliminate fish PCB consumption advisories for recreational anglers within 10 years and for high intake-consumers within 30 years. Since the PCBs' content in the sediment surface (top 4 inches) is the primary factor that controls the amount of PCBs in fish, a cleanup standard was established for OU1. The ROD projected that if remedial actions were designed to address all sediment with PCB concentrations greater than 1.0 ppm (1.0 mg/kg), then a SWAC of 0.25 ppm (0.25 mg/kg) would be achieved in OU1.

The OU1 ROD issued in December 2002 required that all sediment with PCB concentrations greater than 1.0 ppm (1.0 mg/kg) be removed. Due to known limitations with environmental dredging, the ROD allowed an alternative demonstration of compliance. If post-dredge sampling indicated that the 1.0 ppm (1.0 mg/kg) PCBs' Remedial Action Level (RAL) had not been achieved, compliance with the ROD could be confirmed if an OU-wide SWAC of 0.25 ppm (0.25 mg/kg) PCBs was demonstrated. If a SWAC of 0.25 ppm (0.25 mg/kg) PCBs was not achieved, then the ROD required additional dredging and/or the placement of sand covers over dredged areas that would satisfy the 0.25 ppm (0.25 mg/kg) SWAC standard.

The ROD further stated, with specified performance and feasibility criteria from the Agencies, the use of capping as a contingent remedy supplementing sediment removal in order to achieve the ROD requirements. Based on information collected and analyzed after the issuance of the 2002 ROD, a 2008 ROD Amendment provided that, while dredging remained the primary

remedial action for OU1, alternate approaches, including engineered caps, and remedy sand covers, could be used under certain specified conditions.

The ROD Amendment continued the two standards used to judge the completion of the OU1 Remedial Action while allowing the contingent remedy to be used in addition to dredging. Simply stated: the Amended ROD declared that the remedial action (RA) Performance Standard was satisfied if all sediment exceeding the 1.0 ppm (1.0 mg/kg) PCBs' RAL was removed and/or contained using the primary remedial action and/or the alternate remedial action. If the RAL Performance Standard was not satisfied throughout the OU, but all sediment exceeding the RAL had been addressed, using the primary remedial action and/or the alternate remedial actions, the RA will be deemed complete if the Agencies determine that the SWAC goal of 0.25 ppm (0.25 mg/kg) PCBs had been satisfied.

However, the primary measure for compliance with the ROD is to reduce risks due to PCBs' exposure to fish consumers - both human and ecological. In an effort to understand remedial effectiveness, PCB samples from fish and water have been collected under the Baseline Monitoring Plan (BMP) and subsequently under the Long-Term Monitoring Plan (LTMP). These monitoring plans were developed collaboratively between the Agencies/Oversight Team (A/OT) and the Responsible Parties (RPs). Members of both groups were composed of experts in a range of technical disciplines including environmental engineering, analytical chemistry, toxicology, fish and wildlife management and statistics.

In addition to the BMP and LTMP, ecological PCB data are also available from other programs providing additional useful insight into the remedial effectiveness. The State of Wisconsin has analyzed fish tissue samples for PCBs since 1973 under its fish contaminant monitoring program. Water samples have also been collected under several programs since 1989 including other remedial investigations such as Lake Michigan Mass Balance studies. Sediment PCBs' data documenting pre-remedy conditions are available from RI investigations conducted in the 1980s and 1990s as well as pre-remedy design sampling conducted in 2003.

Remedial actions at Little Lake Buttes des Morts on the Lower Fox River, Wisconsin (OU1) were completed in (2009) and sediment, fish, and water samples were collected in 2010 for comparison with historical and baseline/pre-remedy action samples to evaluate effectiveness of the remedy.

A full understanding of the effects of the remedy will be accomplished through the observation of PCB concentrations in sediment, fish, and water over a period of years. PCB concentrations in these three media are influenced by many factors that may vary through time, including lipid content and size/age of fish, organic carbon content in the sediment, river flow and temperatures, as well as the new hydro-dynamics that may develop as a result of the remedy. The analyses reported in this document were conducted to minimize the potential effects of these factors regarding interpretation of the sample results.

Specific results of monitoring each individual media (fish, sediment, and water) are discussed below.

Fish Tissue

PCBs in fish consumed from OU1 are the source of health risks to humans and wildlife. OU1 fish, sampled since the late 1970s, have shown elevated levels of PCBs compared to fish from upstream Lake Winnebago.

The concentration of PCBs in fish is dependent on the level of PCBs in the system and the length, weight, age, and fat or lipid content of the fish. To assure that scientifically valid comparisons are made, fish of similar weight, length, and species are sampled for laboratory analysis. With these factors considered, it is then possible to determine the effectiveness of the ROD's remedy.

The concentrations of PCBs in walleye fish tissue caught in 2010 were compared with walleye that were caught and analyzed from 1990 through 2003. The PCB concentrations through this 14 year time period showed a rate of decrease due to natural recovery. The PCB concentrations in fish caught in 2010 were compared against predicted concentrations from walleye caught and analyzed from 1990 through 2003. Based on this analysis, the walleye caught in 2010 had PCB concentrations that were 73 percent less than the concentration that would have been expected had the remedy not been done.

The reduction to the PCB fish tissue concentration could also be performed from full time data set records dating from 1976 through 2003. However, the analytical technology to measure PCB concentrations in fish tissues prior to 1990 is not as representative as the analytical technology starting in 1990 and therefore is not as comparable. In addition, the rate of reduction in the earlier years (prior to 1990) was greater than the rate observed after 1990 primarily due to removal of original PCB sources to the river. The Agencies believe the 1990 through 2003 data set is more representative regarding natural recovery.

Post-remedy (2010) PCB concentrations in 89 percent (24/27) of skin-on walleye fillets were below the 0.05 ppm (0.05 mg/kg) threshold concentration generally used for unlimited consumption in the State of Wisconsin. This data will be combined with the fish tissue samples collected by the state to evaluate the State of Wisconsin's Fish Consumption Advisory in the future.

While this data confirms the impact of the sediment remediation project, additional rounds of long-term monitoring samples will be required to confirm the safety of consuming fish from OU1. The Long-Term Monitoring Plan, developed to conduct this appraisal, was designed to conduct at least three rounds of samples over a ten year timeframe. Based on all of these sampling events, the overall effectiveness of the ROD remedy will then be determined.

Note: Fish barrier(s) do not exist between Lake Winnebago (LW) and Little Lake Buttes des Morts. It is possible some of the fish sampled in 2010 were not "long-term" residents of OU1, and may have migrated downstream from LW or upstream from OU2. However, this can also be true for fish sampled since 1976. Regardless, this must be a point of consideration when interpreting results and reinforces the decision to be conservative in making any final decisions regarding the effectiveness of the ROD's remedy over time.

Sediment

In order to demonstrate compliance with the ROD's remedial design, post-remedy sediment samples were collected and analyzed for sediment PCB concentrations under a quality assurance project plan (QAPP) and construction quality assurance project plan (CQAPP). These tasks were conducted by the RPs and overseen by the A/OT. This confirmation program, composed of over 2200 analytical PCB tests, provided documentation of surface PCB concentrations in sediment immediately upon completion of the remedy, however long-term permanence will need further confirmation.

The results of these 2200 analytical PCB tests have been used to calculate a post-remedy SWAC for OU1. The method, to estimate SWAC, is based on statistical procedures drawn from established statistical literature using a weighted averaging approach known as stratified sampling and analysis. This method uses an area-based weighted average to combine data collected from different populations: for this case, different remedial action types, such as dredging areas, sand covered areas and no-action areas. This analysis, conducted by Foth (2010), resulted in an estimated SWAC of 0.23 ppm (0.23 mg/kg) which is less than the ROD-targeted SWAC of 0.25 ppm (0.25 mg/kg) with 95 percent level of confidence.

In an effort to evaluate the longer-term permanence of the remedy, USEPA also implemented an independent post-remedy sediment sampling and analysis program. This program, conducted in 2010 by the USEPA Region 5 FIELDS Group (USEPA, 2010a) collected 63 composite surface sediment samples suitable to estimate SWACs within subareas of OU1, as well as being suitable to estimate the SWAC for the entire OU1. In the event that a significant change in OU-wide SWAC was measured, sediment PCB levels in smaller sub-areas could be useful in the identification of root causes of change(s).

Using the PCB-sample results collected by the FIELDS Group, a similar SWAC calculation method was used to calculate an OU-wide SWAC of 0.26 ppm (0.26 mg/kg) with lower and upper confidence limits ranging from 0.17 ppm (0.17 mg/kg) to 0.32 ppm (0.32 mg/kg). These results do not exactly match those obtained by Foth because both estimates are based on a sample from the population of sediments in the lake and therefore have inherent uncertainty which is bounded. The Foth estimates are more precise, i.e., narrower confidence limits because of the number of samples collected and analyzed (greater than 2200). However, the FIELDS' data supports an OU-wide SWAC consistent with the Foth estimate and the remedial goal of (0.25 ppm (0.25 mg/kg), as demonstrated by the fact that the FIELDS-based confidence limits include both values. The FIELDS independent samples confirm that the OU-wide average PCB concentrations are approximately 0.25 ppm (0.25 mg/kg) to within the margin of error of their study.

Water

OU1 is the most upstream reach of the Fox River site. LTM data (2010) shows the ROD's remedy has apparently reduced the level of PCBs in the water column when compared with the pre-remedy and BMP results.

Water PCB concentrations are thought to be dependent on PCB concentrations in sediment along with the river's flow rate and temperature. Given these variables, the comparison must be made by accounting for natural variations in river flow rates and temperatures through the years prior to the remedial activities. Figure 3 shows the remediation resulted in apparent lower concentrations.

However, the pre-remedy data results were irregularly (1989, 1990, 1998, 2005) collected, and laboratory analysis and field sampling methods varied among studies over this time period; therefore, the percentage change effected by the remedy cannot be estimated for water column PCB concentrations with an appropriate level of statistical confidence.

Note: As has been observed at other large dredging and remedial projects, water-column PCB concentrations increased above background levels during dredging but declined rapidly to substantively lower than expected levels post-remedy.

Figure 1

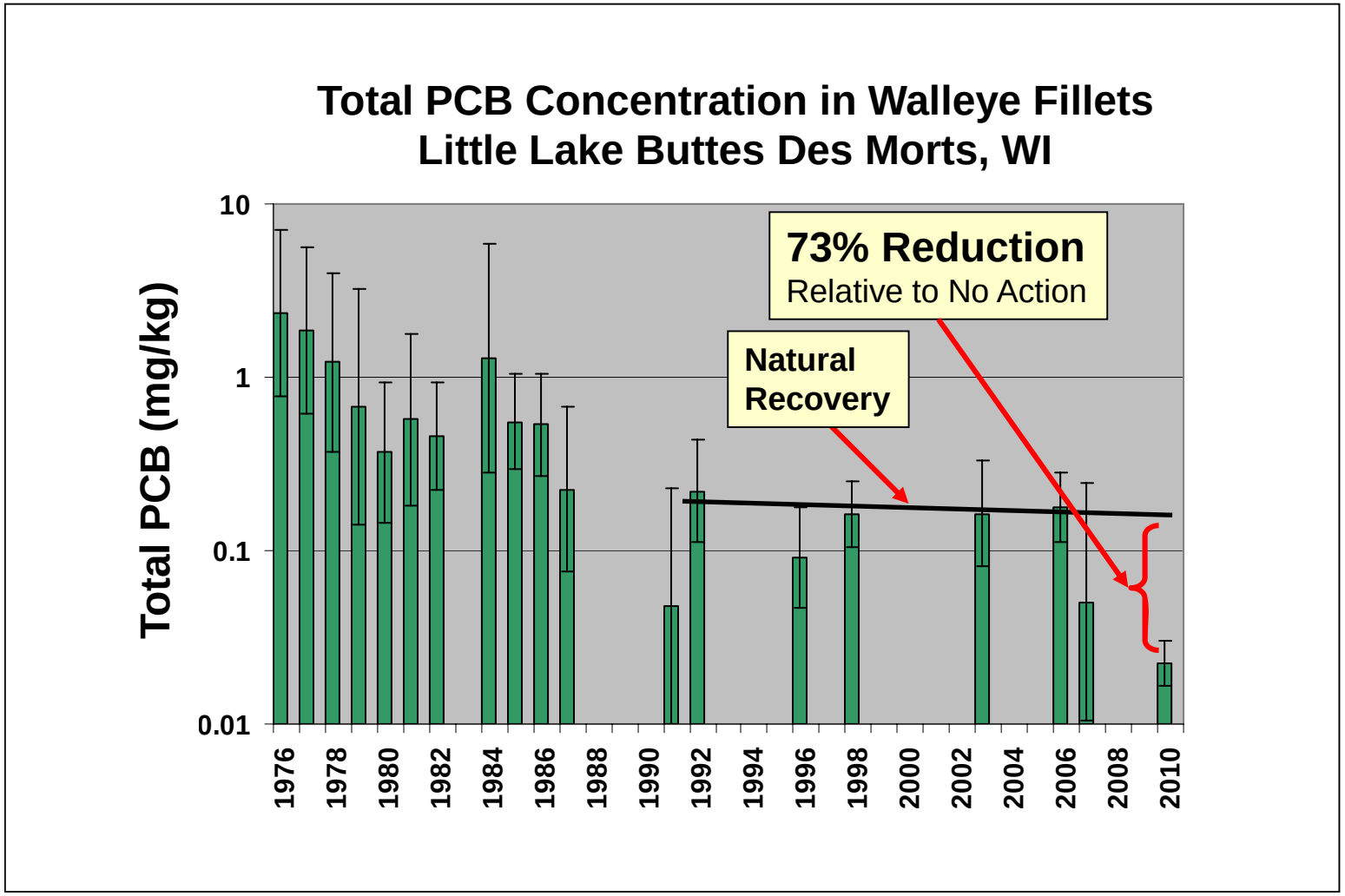

Total PCB Concentration in Walleye Fillets
Little Lake Buttes Des Morts, WI
Total PCB (mg/kg)
10
1
0.1
0.01
73% Reduction
Relative to No Action
Natural
Recovery
1976
1978
1980
1982
1984
1986
1988
1990
1992
1994
1996
1998
2000
2002
2004
2006
2008
2010

Figure 2

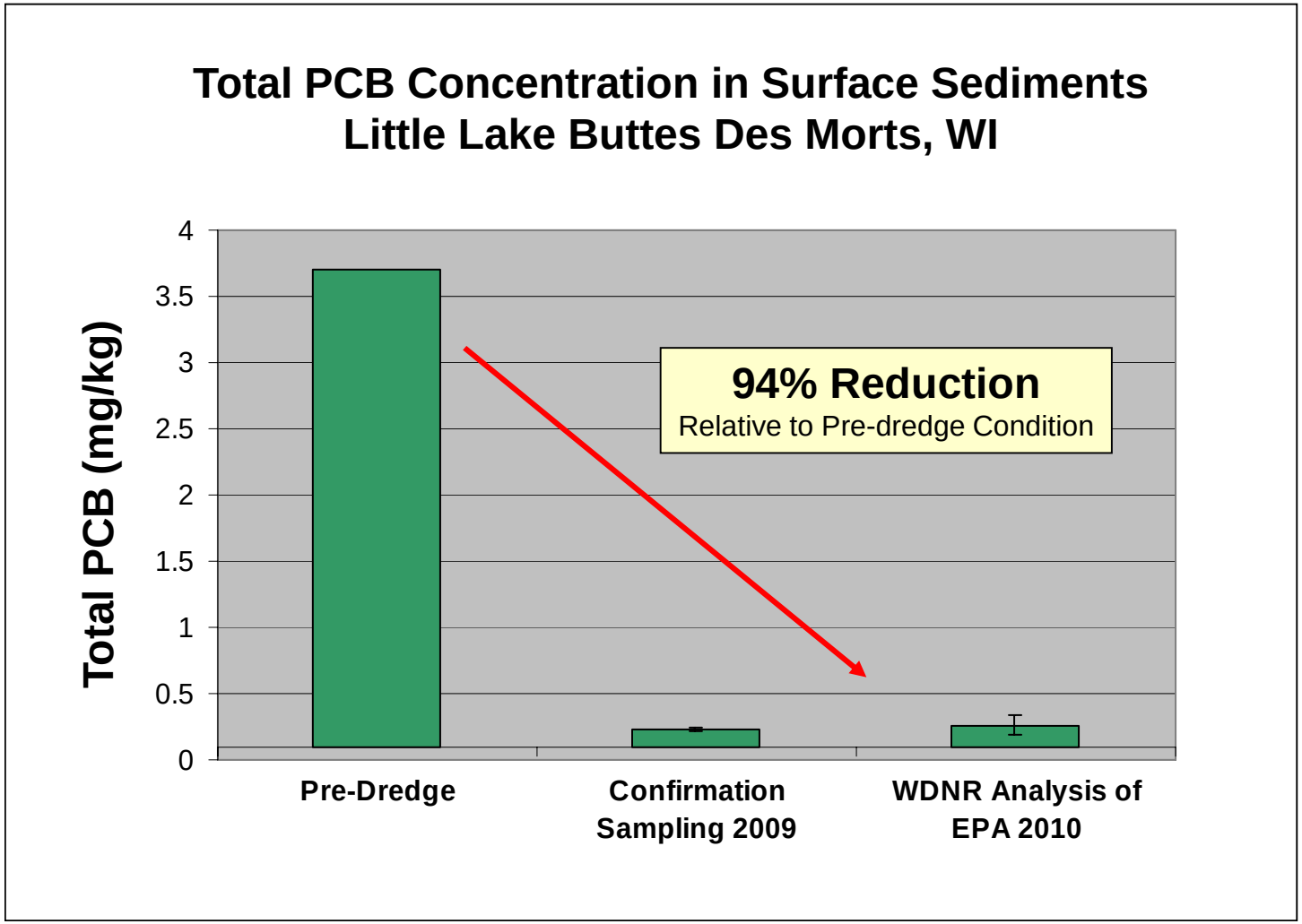
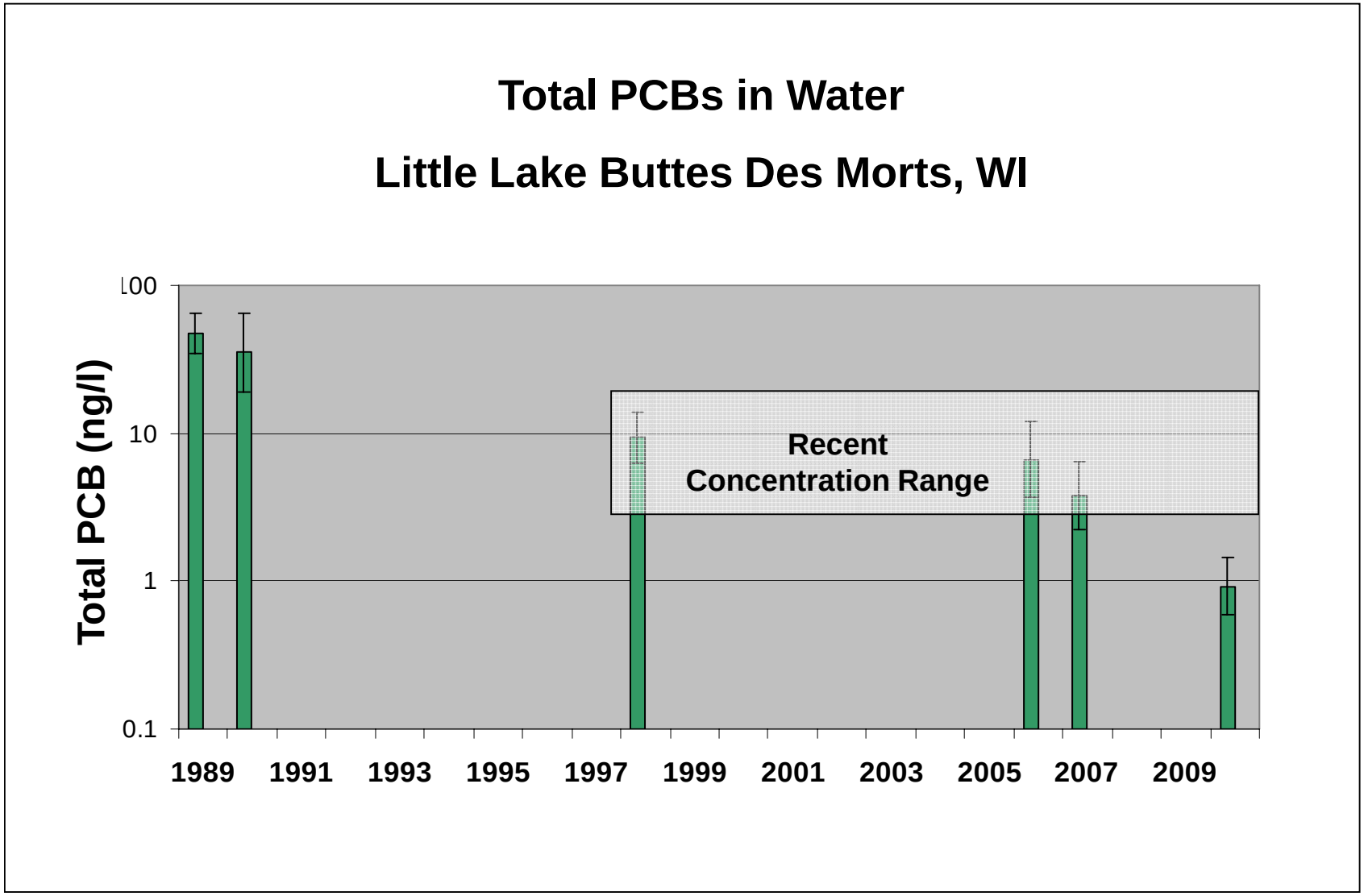

Total PCB Concentration in Surface Sediments
Little Lake Buttes Des Morts, WI
Total PCB (mg/kg)
4
3.5
3
2.5
2
1.5
1
0.5
0
94% Reduction
Relative to Pre-dredge Condition
Pre-Dredge
Confirmation Sampling 2009
WDNR Analysis of EPA 2010

Figure 3


Total PCBs in Water
Little Lake Buttes Des Morts, WI
Total PCB (ng/l)
100
10
1
0.1
Recent
Concentration Range
1989
1991
1993
1995
1997
1999
2001
2003
2005
2007
2009

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ScienceDirect
Chemosphere 66 (2007) 1821–1828
CHEMOSPHERE
www.elsevier.com/locate/chemosphere

Polychlorinated biphenyl contamination trends in Lake Hartwell, South Carolina (USA): Sediment recovery profiles spanning two decades

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Abstract

To assess the ca. 20-year polychlorinated biphenyl (PCB) contamination trends in Lake Hartwell, SC, sediment cores from the Twelve Mile Creek arm were collected in July 2004 at two sites (G30 and G33) first sampled in the mid-1980s. Congener-specific PCB data as a function of depth from the sediment–water interface for the 2004 sediment samples were compared to data obtained from 1987 and 1998 samples taken from the same locations. Despite modest decreases in total PCB levels near the G30 sediment–water interface, historical increases in average degrees of chlorination may elevate the overall toxic risk at this site. Unlike G30, the more rapid recovery in the near-surface sediment of G33 suggests that the effectiveness of the U.S. EPA natural attenuation record of decision is site-specific and is unlikely to result in uniform surface sediment recovery throughout the most contaminated regions of Lake Hartwell.

Keywords: PCBs; Natural attenuation; Congener-specific analysis; Reductive dechlorination; Aroclors

1. Introduction

Polychlorinated biphenyls (PCBs) are persistent organic pollutants with significant bioaccumulation potentials in environmental systems. Despite the cessation of domestic production in the late 1970s, sites contaminated with PCBs continue to pose a threat to human and ecosystem health. In aquatic systems, PCB partitioning and transformations are a function of molecular-level (i.e., congener-level) properties and structure (Erickson, 1986; Farley et al., 1994). The toxicological effects of PCBs also vary at the congener level (Safe, 1992). The variability observed in the physiochemical properties of PCBs suggests a differential partitioning (i.e., weathering) of congeners in multiphase environmental systems (Burkard et al., 1985; Dunnivant et al., 1988). Evidence of PCB physiochemical weathering in near-surface sediments in the Twelve Mile Creek arm of Lake Hartwell (the current study site) was reported by Farley et al. (1994). At this site, the degree of PCB chlorination in shallow sediments was observed to increase with increasing distance downstream of the source. During sediment resuspension and transport events, the preferential sorption of higher chlorinated congeners and volatilization of lower chlorinated congeners were proposed as the dominant PCB weathering mechanisms (Dunnivant et al., 1988).

Aerobic and anaerobic PCB degradation processes have been reported in laboratory investigations (Abraham et al., 2002; Master et al., 2002). Typically, PCBs with up to four chlorine atoms are susceptible to aerobic biotransformations. In anaerobic environments, reductive dechlorination is the most commonly reported PCB biotransformation mechanism (Bedard and Quensen, 1995; Pakdeesusuk et al., 2003). Reductive dechlorination of PCBs is a two-electron transfer mechanism in which chlorine atoms on the biphenyl moiety are replaced with hydrogen atoms (Nies and Vogel, 1991). Reductive dechlorination typically

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operates on the higher chlorinated PCB congeners (i.e., PCBs with four or more chlorines). Consequently, in closed systems, reductive dechlorination can reduce the total PCB level on a mass but not on a molar basis.

In sediment systems, an accumulation of lower chlorinated PCB congeners with a concurrent decrease in higher chlorinated congeners serves as compelling, albeit indirect, evidence of reductive dechlorination (Pakdeesusuk et al., 2005). When sediment PCB data are compared to known source composition data for the evaluation of *in situ* reductive dechlorination, the assumption is often made that source PCB compositions are identical to those deposited at sediment sampling sites. This assumption is not necessary, however, when congener-specific data exist for two or more sampling times. In these instances, the data from any sampling time (most logically the earliest) can serve as a datum from which PCB historical trends can be assessed. In all cases, it must be recognized that reductive dechlorination may not be the only process responsible for historical changes in PCB metrics. Dissolution, volatilization, aerobic biotransformations and sediment transport may be significant mechanisms in addition to reductive dechlorination (Farley et al., 1994; Bedard and Quensen, 1995).

Microcosm studies involving Lake Hartwell sediments have confirmed the presence of indigenous microorganisms capable of reductively dechlorinating PCB congeners in Aroclor 1254 (Pakdeesusuk et al., 2003). Congener-specific PCB analysis confirmed an overall decrease in the weight percent of the higher molecular weight species and a concurrent increase in the lower molecular weight congeners. The average number of total chlorines per biphenyl (Cl/biphenyl) decreased from ≈4.9 to 3.0 after 260 days of incubation. Analysis of dechlorination products indicated the preferential removal of *meta* and *para* chlorines; a decrease in *ortho* chlorines was not observed. As such, increases in the fraction of chlorine atoms occupying *ortho* positions (% *ortho* Cl) with concurrent decreases in total chlorines per biphenyl serve as compelling (indirect) evidence of reductive dechlorination.

Lake Hartwell, in northwestern South Carolina, is heavily contaminated with PCBs from the manufacturing and waste disposal practices of a capacitor manufacturer operating near Pickens, SC, from 1955 to 1987 (US EPA, 2004). The production of PCB-containing capacitors ended at the site in 1976, concurrent with the passage of the Toxic Substances Control Act, which subsequently banned PCB production in the US (LaGrega et al., 2001). PCB loading into the Lake Hartwell system via direct surface discharge and groundwater infiltration of PCB-containing wastes is estimated at 200 metric tons, ≈80% as Aroclor 1016 and 20% as Aroclor 1254 (Wong et al., 2001). In the mid-1980s, Germann (1988) conducted a broad survey of Lake Hartwell sediments to determine the geographic extent of PCB contamination downstream of the source. Sediment analyses from this survey suggested that PCB levels were the highest in the Twelve Mile Creek arm of Lake Hartwell. The entire Sangamo–Weston/Twelve Mile Creek/Lake Hartwell site was added to the EPA National Priority List in February 1990 (US EPA, 2004). The June 1994 record of decision (ROD) for this site called for natural capping of contaminated sediment with ongoing monitoring. A total-PCB cleanup requirement of 1.0 μg/g was stipulated. The rationale for this monitored natural attenuation strategy is to cover contaminated sediment with a sufficient amount of increasingly clean sediment such that PCB residues are prevented from entering the aquatic food chain (Brenner et al., 2004). Natural capping also reduces the potential for resuspension and subsequent relocation of contaminated near-surface sediments. Given sufficient time, the EPA hypothesizes that natural attenuation processes (e.g., biodegradation, burial and volatilization) will reduce the mass load of PCBs in Lake Hartwell (US EPA, 2004; Magar et al., 2005a).

A large historical dataset spanning two decades has been compiled for the most contaminated regions of Lake Hartwell. The data represent sediment core samples collected in 1987 (Germann, 1988), 1998 (Pakdeesusuk et al., 2005) and 2004 (the current work). Bulk PCB metrics (e.g., total PCBs, average chlorines per biphenyl, homolog distributions) and congener-specific concentrations were determined for all three sampling dates, which allow an analysis of historical PCB contamination trends. Examination of PCB depth profiles affords an assessment of the natural attenuation ROD assigned to the Lake Hartwell Superfund site. Historical congener-specific PCB data also permit an evaluation of *in situ* reductive dechlorination processes. The primary goal of the current work is to elucidate PCB contamination trends at two sediment-sampling locations (G30 and G33) in the Twelve Mile Creek arm of Lake Hartwell, SC. Data addressing the influence of (bio)chemical PCB weathering on congener profiles are discussed. An evaluation of the efficacy of the natural attenuation ROD invoked by the EPA at this Superfund site is also provided.

2. Experimental section

2.1. Sediment sampling

Sediment samples were obtained in July 2004 at sites G30 and G33 (one core per site) in the Twelve Mile Creek arm of Lake Hartwell. Sites G30 and G33 are ca. 36 and 38 km downstream of the original PCB discharge location (Farley et al., 1994) and represent heavily contaminated sites with historically similar sedimentation rates (Brenner et al., 2004). A map of the sampling locations is shown by Pakdeesusuk et al. (2005). Sediment cores were collected using a Wildco gravity corer fitted with a Lexan™ tube (5 cm diameter, 76 cm length). The cores were transported to the L.G. Rich Environmental Research Laboratory (Clemson University, Anderson, SC) and extruded within 24 h of the sampling event. The cores were sectioned every 5 cm, homogenized by manual mixing for 3 min, and stored in solvent-rinsed glass jars (0.5 l) at 4 °C prior to analysis. Prior to homogenization, the exterior portions

(~1 cm) of the 40–45 cm sediment fractions from both cores (G30 and G33) were isolated and stored separately from the interior portions not in contact with the Lexan™ tubes. Subsequent analyses were conducted for both the exterior and interior portions of the G30 and G33 (40–45 cm) fractions; sediment smearing was not observed.

2.2. PCB extraction

PCBs were extracted from sediment samples into acetone followed by solvent exchange into isooctane via a previously described sonication method (Dunnivant and Elzerman, 1987; Pakdeesusuk et al., 2005). Prior to sonication, all samples were spiked with octachloronaphthalene (3.4 μg in acetone) as a recovery standard. Extraction efficiencies averaged 81 ± 58%. Results were not corrected for extraction efficiencies.

2.3. PCB analysis

Sediment extracts were analyzed for PCBs on a Hewlett-Packard 6890 gas chromatograph (GC) equipped with a 30 m fused silica capillary column (ZB-5, Phenomenex, Torrance, CA; 0.25 mm diameter, 0.25 μm film thickness) and a ^{63}Ni electron capture detector (ECD). GC parameters are described by Pakdeesusuk et al. (2005). Blank GC runs (isooctane only) were conducted after approximately every five GC samples to check for analyte carry-over between injections. A 1:1 mixture of Aroclors 1016 and 1254 (AccuStandard; New Haven, CT) was used as a check standard after approximately every 12 field sample GC injections. Average GC–ECD response factors varied by less than 10% for all check standard analyses.

2.4. PCB quantification

Details of GC peak assignment, calibration and error propagation methods are described by Sivey (2005). Unless otherwise stated, all PCB error intervals reported below represent total analytical errors (Sivey, 2005). Total analytical error is a propagated uncertainty metric incorporating all significant error sources (e.g., sediment dry weight determinations, replicate GC–ECD analyses and calibrations). The current PCB quantification method, which quantifies 81 GC peaks, representing 128 PCB congeners, varies slightly from the method employed during the analyses of the 1987 and 1998 samples (Germann, 1988). Data from all sampling events were subjected to the current quantification method to facilitate congener-specific historical trend analyses.

3. Results and discussion

3.1. G30 PCB metrics and historical trends

Total PCB depth profiles for G30 samples collected in 1987 (Germann, 1988), 1998 (Pakdeesusuk et al., 2005) and 2004 (the current work) are shown in Fig. 1a. The 2004 total PCB concentration at the sediment–water interface (0–5 cm) is 3.0 ± 0.2 μg/g (9.5 ± 0.6 nmol/g). This value exceeds the EPA clean-up requirement of 1.0 μg/g and is indicative of a high potential for biota exposure and bioaccumulation in organisms residing or feeding in near-surface sediment. Significant PCB mass transport via resuspension of sediment at this site is also of concern. If recent near-surface recovery rates (0.19 ± 0.01 μg/g/yr; average annual decline in total PCB levels from 1998 to 2004) persist, G30 surface sediments will not be in compliance with the EPA clean-up requirement of 1.0 μg/g until 2015. Approximately uniform total PCB concentrations persist from 0 to 15 cm, indicating a well-mixed surface sediment layer. A sharp increase in total PCBs occurs at 15–30 cm. The maximum concentration captured by the 2004 core is 36 ± 1 μg/g (141 ± 8 nmol/g) at 30–35 cm. The entire vertical PCB profile was not captured in this core as evidenced by the concentrations failing to approach zero with depth.

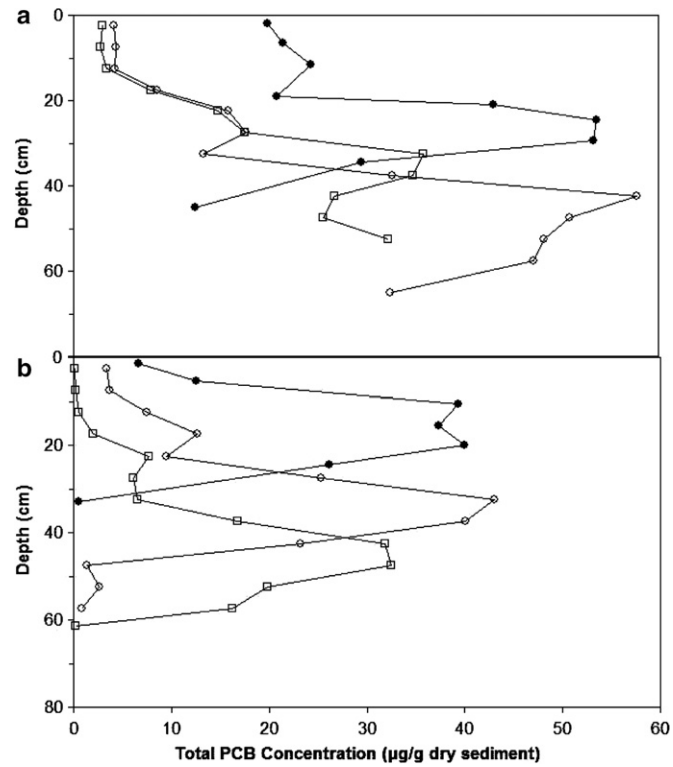

Fig. 1. Historical trends in (a) G30 and (b) G33 total PCB concentrations: (●) 1987; (○) 1998; (□) 2004.

Large decreases in total PCBs at the sediment–water interface are observed between 1987 and 1998 (Fig. 1a). Only modest decreases are observed between the 1998 and 2004 data for near-surface sediments. The 1998 and 2004 PCB profiles are very similar from 0 to 30 cm, which implies minimal net sedimentation between these two sampling dates. However, the maximum PCB concentration at this site is most likely deeper than that captured by the 2004 sampling event. Therefore, a reliable calculation of

recent sedimentation rates based solely on PCB profiles following the method described by Pakdeesusuk et al. (2005) is not possible. An examination of the historical data suggests that none of the cores captured the entire PCB profile at this site. An average sedimentation rate of 2.0 ± 1.8 g/cm^2/yr was reported by Brenner et al. (2004) at a transect of Lake Hartwell near G30 based on 2000 and 2001 sediment core radioisotope dating. Assuming a sediment bulk density of 2.6 g/cm^3 (Farley et al., 1994), this sedimentation rate translates into 0.8 ± 0.7 cm/yr. The G30 near-surface results of the current work suggest a six-year average (1998–2004) sedimentation rate near the lower end of the range calculated by Brenner et al. (2004).

Historical chlorine distribution profiles can be useful in assessing trends in PCB composition over time. Fig. 2a outlines the G30 Cl/biphenyl profiles for all three sampling years under consideration. Fig. 3a depicts % *ortho* Cl historical profiles. The approximate source composition (4:1 mixture of Aroclors 1016 and 1254) is demarcated in both figures by a dashed line (Cl/biphenyl = 3.39; % *ortho* Cl = 46). Dissimilarities in Cl/biphenyl levels at the sediment–water interface suggest a variable PCB composition of deposited sediment at G30 over time. The average Cl/biphenyl of PCBs in deposited sediment increased over time. This change reflects increasing degrees of source PCB weathering, as indicated by the increasing distances

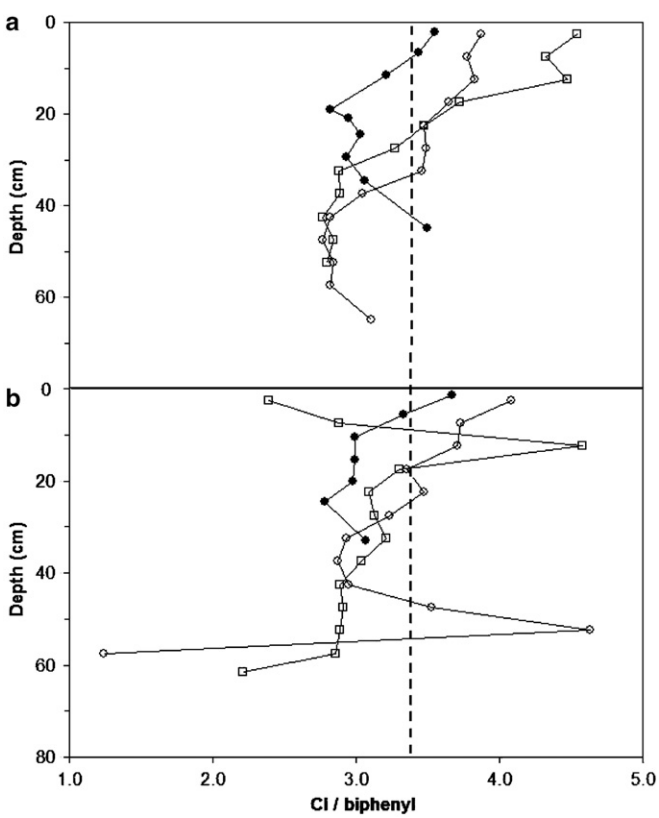

Fig. 2. Historical trends in (a) G30 and (b) G33 total chlorines per biphenyl: (●) 1987; (○) 1998; (□) 2004. The dashed line denotes the original PCB source composition (4:1 mixture of Aroclors 1016 and 1254).

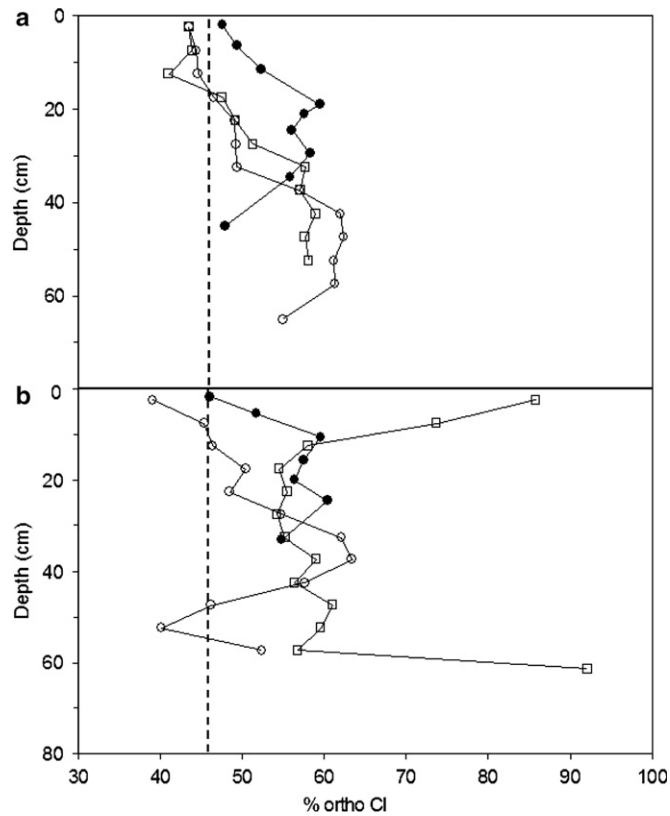

Fig. 3. Historical trends in (a) G30 and (b) G33 percent *ortho* chlorines: (●) 1987; (○) 1998; (□) 2004. The dashed line denotes the original PCB source composition (4:1 mixture of Aroclors 1016 and 1254).

from the dashed line in Fig. 2a for near-surface sediment. The most probable mechanisms acting here include the preferential volatilization and sorption of low and high molecular weight PCBs, respectively (Farley et al., 1994). The influence of sediment impoundment releases upstream of G30 may also impact the PCB composition of shallow sediments at this site. Aerobic biotransformations of low molecular weight congeners may be significant as well. Evidence of a variable deposition composition is also indicated at the congener-level. Fig. 4 displays the mole percent distributions of selected congeners in near-surface sediments for all three sampling dates. Increases in higher chlorinated congeners (245–25 + 236–35; 235–245) with concurrent decreases in lower chlorinated congeners (2–2 + 26; 4–4 + 24–2) over time are observed. Overall, PCB toxicity (Safe, 1992) and bioaccumulation (Morrison et al., 1996) potentials increase with the degree of chlorination. The bioaccumulation and toxicity potentials of individual congeners can vary by more than two orders of magnitude. As such, despite the continued trend of decreasing total PCB concentrations in G30 near-surface sediments, a concurrent decrease in the bioaccumulation and toxicity potentials of these sediments may not be occurring due to the shift toward higher chlorinated congeners over time. These trends, in conjunction with a total PCB concentration above the clean-up guideline, raise significant ecotoxicological concerns at G30.

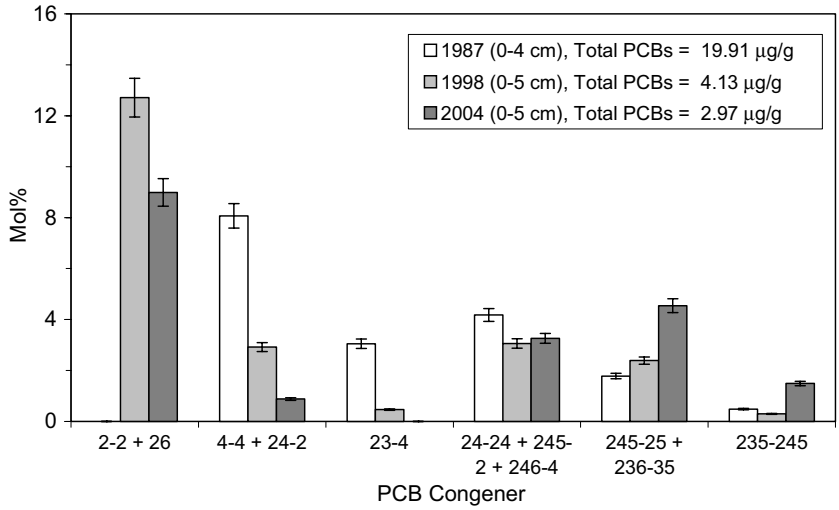

Fig. 4. Historical trends in the relative distributions of selected congeners at the G30 sediment–water interface. Error bars denote the error between the two analytical methods employed to obtain 2004 (Sivey, 2005) and pre-2004 (Pakdeesusuk et al., 2003) data.

As shown in Fig. 2a, the 1987 core depicts a rapid decline in Cl/biphenyl beginning near the sediment–water interface and continuing to a depth of 20 cm. For the 1998 and 2004 cores, a similar decrease in Cl/biphenyl levels occurs between 15 and 40 cm. Concomitant increases in % *ortho* Cl occur at the same depths (Fig. 3a). Taken together, these data are persuasive indicators of PCB biochemical weathering via reductive dechlorination (Pakdeesusuk et al., 2005) and that this process has likely been operating at this location for several decades. As sediment residence time increases with depth, Cl/biphenyl levels converge to ≈2.9 for all samples regardless of total PCB concentration (Fig. 2a). Similar convergence to approximately 60% *ortho* Cl with depth is also observed (Fig. 3a). At depths greater than 30 cm, changes in chlorine distributions are minimal (Figs. 2a and 3a), indicative of a plateau phase in reductive dechlorination at these depths (Pakdeesusuk et al., 2005). A plateau phase occurs when local reductive dechlorination rates approach zero. Historically, the plateau phase occurred at depths with total PCB levels exceeding those measured in the active reductive dechlorination zone (Fig. 1a). Therefore, a parameter other than (low) total PCB concentration must be responsible for the diminished reductive dechlorination rate. As observed by Pakdeesusuk et al. (2005), reductive dechlorination can be limited by a low availability of susceptible congeners (i.e., those with a spatial arrangement of chlorines favorable to biochemical reduction reactions) and by a population decrease in microbial communities capable of reductive dechlorination.

3.2. G30 Congener-specific analyses

Comprehensive congener distributions for the near-surface sediment (0–5 cm) and the fraction representing the highest captured total PCB level (30–35 cm) are depicted in Fig. 5. Of note is the predominance of higher chlorinated congeners in the shallow sediments. The dramatic shift to lower chlorinated congeners with depth is evidence of PCB weathering via reductive dechlorination at this site.

3.3. G33 PCB metrics and historical trends

Total PCB depth profiles for G33 samples collected in 1987 (Germann, 1988), 1998 (Pakdeesusuk et al., 2005) and 2004 (the current work) are shown in Fig. 1b. All three cores captured the entire PCB profile at this site. Consistent decreases in PCB levels at the sediment–water interface from 1987 to 2004 are observed. The 2004 total PCB concentration at the sediment–water interface is 0.133 ± 0.007 µg/g (0.56 ± 0.03 nmol/g). If the decrease in PCB concentration in near-surface sediments is assumed to be linear between 1998 and 2004, this site became compliant with the EPA clean-up requirement of 1.0 µg/g in 2002. The maximum concentration captured by this core is 33 ± 1 µg/g (128 ± 5 nmol/g) at 45–50 cm.

The Cl/biphenyl and % *ortho* Cl profiles for G33 are shown in Figs. 2b and 3b, respectively. It should be noted that all chlorine distribution metrics are susceptible to large errors at very low (i.e., <1 µg/g) total PCB concentrations. As individual congener concentrations approach their quantification limits, the concentrations of those congeners that are quantified can dominate chlorine distribution parameters. This results from variations in congener GC–ECD responses, which are magnified as concentrations approach the quantification limits of the analytical method. Therefore, the reliability of all chlorine distribution metrics may be low for G33 fractions at depths less than 15 cm and greater than 60 cm (Fig. 1b). In the 2004 core, a decrease in Cl/biphenyl from 3.21 to 2.86 is shown from 20–40 cm (Fig. 2b). The % *ortho* Cl values across these depths hover near 57% (Fig. 3b). In the sections of the cores with the highest PCB levels, Cl/biphenyl and % *ortho* Cl values converge near 2.9% and 60%, respectively.

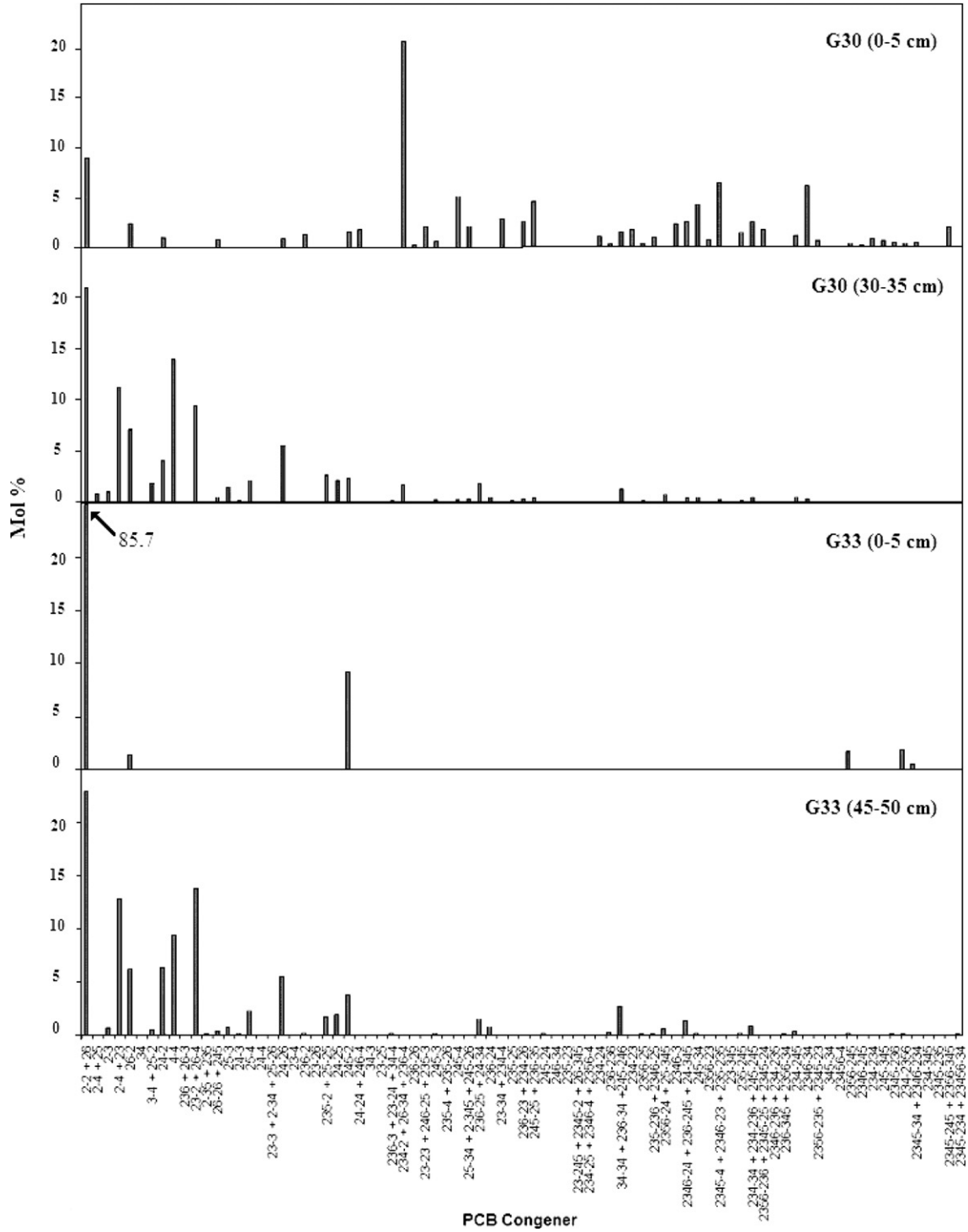

Fig. 5. Comprehensive congener profiles for the near-surface sediment and the highest recorded PCB contamination level at G30 and G33.

These data are considered modest evidence of reductive dechlorination at this site.

3.4. G33 congener-specific analysis

Comprehensive congener distributions for the near-surface sediment (0–5 cm) and the depth representing the highest captured total PCB level (45–50 cm) are shown in Fig. 5. Of note near the sediment–water interface (0–5 cm) is the predominance of three congeners, PCB 4 (2–2), 10 (26) and 48 (245–2). Each of these congeners has two ortho chlorines and are of low relative toxicity among all PCBs (Safe, 1992).

The PCB distribution from 45–50 cm is comprised largely of congeners with less than five chlorine atoms. Reductive dechlorination processes may be responsible for the

Table 1
G30 and G33 site comparisons based on the results of the 2004 sampling event

		G30	G33
Sediment–water interface	Depth (cm)	0–5	0–5
	Total PCBs (μg/g)	3.0 ± 0.2	0.133 ± 0.007
	Cl/biphenyl	4.6 ± 0.3	2.4 ± 0.1
	% *ortho* Cl	43 ± 3	86 ± 5
	Sediment recovery rate (μg/g/yr)[a]	0.19 ± 0.01	0.54 ± 0.04
Maximum PCB level	Depth (cm)	30–35	45–50
	Total PCBs (μg/g)	36 ± 2	33 ± 1
	Cl/biphenyl	2.9 ± 0.2	2.9 ± 0.1
	% *ortho* Cl	58 ± 3	61 ± 2

Uncertainty intervals denote total analytical errors (Sivey, 2005).
[a] Sediment recovery rates are based on the six-year average concentration differences between the 1998 and 2004 sampling events.

low levels of higher chlorinated congeners in this fraction. The congener-specific profile for 45–50 cm is representative of all samples from 35–63 cm (data not shown). These results are evidence of a reductive dechlorination plateau phase, as proposed by Pakdeesusuk et al. (2005). It may be argued that *in situ* biochemical weathering is not the only possible explanation for these congener-specific results. A potentially less probable explanation is the deposition of PCB-contaminated sediment over a span of several years with extremely similar congener signatures resulting from *upstream* physical and/or biochemical weathering processes.

3.5. Site comparisons

Representative PCB results from sites G30 and G33 are juxtaposed in Table 1. Significant differences exist between the two sites at the sediment–water interface. Whereas G30 sediments are well above the clean-up standard of 1.0 μg/g, G33 sediments are an order of magnitude below this level. The Cl/biphenyl level is also lower at G33. Taken together, these metrics implicate G30 as having a higher PCB toxicity and bioaccumulation potential relative to G33. Also of concern is the slower recovery rate at G30 (0.19 ± 0.01 μg/g/yr) relative to G33 (0.54 ± 0.04 μg/g/yr). This difference in recovery rates can be explained by a greater net sedimentation rate at G33, the deposition of less-contaminated sediment at G33, or both.

Unlike at the sediment–water interface, the PCB signatures at maximum contamination depths of G30 and G33 are very similar. In both cores, Cl/biphenyl and % *ortho* Cl values approach 2.9% and 60%, respectively, as total PCB concentrations reach their local maximum values. Similarities at these depths are also evident in the congener distributions (Fig. 5). These results are consistent with the most biochemically weathered end-member (EM 3) from the polytopic vector analysis for Lake Hartwell sediments reported by Magar et al. (2005b). These results indicate that PCBs comprising the maximum concentration levels in both cores were likely subjected to comparable weathering processes, including reductive dechlorination. The data listed in Table 1 suggest that PCB weathering processes occurred via similar mechanisms and to comparable extents at both sites.

In summary, the EPA remediation plan of natural sedimentation appears to be happening with a variable degree of success at sites G30 and G33. Local variations in sedimentation rates and PCB composition of deposited sediments appear to impact the efficacy of the natural attenuation ROD. The periodic release of contaminated sediment from impoundments upstream of the two sampling sites may also influence PCB profiles and recovery rates in a spatially heterogeneous fashion. These local disparities are responsible for the variable recovery rates calculated for the near-surface sediments at G30 and G33. To be sure, the uniform deposition of progressively less-contaminated sediments across the entire Twelve Mile Creek arm of Lake Hartwell is an unlikely phenomenon.

Congener profiles at G30 and G33 provide evidence of *in situ* reductive dechlorination as a probable PCB weathering mechanism at these sites. A shift to lower degrees of chlorination with concurrent increases in % *ortho* chlorines with increasing depth from the sediment–water interface is observed at both sites. Historical convergence to 2.9 average chlorines per biphenyl and 60% *ortho* chlorines with depth is also observed at both sites. Consistent with previous investigations (Pakdeesusuk et al., 2005), these trends are likely the result of *in situ* reductive dechlorination followed by a plateau phase with increasing distance from the sediment–water interface.

Acknowledgements

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East Eagle Harbor Bainbridge WA; 17 Years of Cap and Natural Recovery Monitoring

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Overview. The Wyckoff/Eagle Harbor Superfund Site, East Harbor Operable Unit (EHOU) is a former wood pole-treating facility located on Bainbridge Island, Washington. This paper presents the findings of the Year 17 monitoring of implemented remedies within the in-water operable unit. The remedies include over 60 acres of in-water subtidal isolation capping, two distinct intertidal isolation cap designs, and monitored natural recovery of intertidal areas that also contain ecologically critical eelgrass habitat. Overall the subtidal and intertidal caps are operating as intended; the caps are physically stable and the underlying PAH contaminants remain isolated. However, one area of the cap that is proximal to the Washington State Ferry Terminal has been eroded away, exposing the native NAPL-contaminated sediments. The monitored natural recovery has shown orders-of-magnitude decreases in PAH levels in the 10 years since the last monitoring.

Background/Objectives. The former Wyckoff treatment plant was heavily contaminated with principally with creosote and pentachlorophenol which permeated the soil, groundwater, and extensive areas of the subtidal and intertidal sediments. The site was added to the NPL in 1987 and the ROD signed in 1994. The subtidal cap was first placed in 1993-1994, with subsequent additional placements and remedial actions up through 2008. The objectives of this monitoring were to determine whether the caps are physically stable and effective at contaminant isolation, whether the monitoring natural recovery targets had been achieved, and as a secondary question whether the installed caps served as habitat for forage fish and if bivalves recovered at the site pose a human health consumption risk.

Approach/Activities. One of the oldest sediment capping projects in the U.S., the EHOU has been monitored since the original placement in 1993. The physical tools to evaluate site stability were primarily elevation surveys using bathymetry and photogrammetry. Through-cap coring was used to confirm cap thickness measures estimated by the elevation modeling. Changes in elevation indicated both erosion and accretion occurred over the site. In 2011 hydrodynamic modeling was added to assess the effects of wind and tidal-generated waves, and propeller wash from the Washington State Ferries that traverse the cap site. The tools used to assess chemical isolation included surface and subsurface sediment chemistry, and over a portion of the site *in-situ* sediment (SPME) samplers. Monitored natural recovery was assessed through surface and subsurface sediment sampling, as well checking for hydrocarbon seeps that were prevalent in the previous monitoring in 2002. Biological monitoring included collecting clams for tissue chemical analyses, habitat use surveys for birds, macroinvertebrates and algae, and a special study on use of the cap habitat by forage fish.

Results/Lessons Learned. The presentation will present the full results of Year 17 monitoring, and what additional activities are planned for operations and maintenance, and continued monitoring.

Monitored Natural Recovery at a Submarine Wood Waste Site: 10 Years after Baseline

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Background/Objectives. Sawmill Cove is located near the mouth of Silver Bay in southeast Alaska and was the receiving point for effluent and storm water discharges from the Alaska Pulp Mill, which produced pulp at the site from 1959 to 1993. Operations at the mill resulted in the accumulation of wood solids in some areas up to 20 ft or more in thickness on approximately 100 acres of the seafloor adjacent to the site. A remedial investigation and ecological risk assessment of the Bay Operable Unit (OU), encompassing Sawmill Cove, were conducted during 1996 and 1997. The results of these studies were used to delineate an Area of Concern (AOC) in Sawmill Cove. The Remedial Action Objective (RAO) for the AOC in Sawmill Cove, as defined in the Record of Decision (ROD), is to reduce to an acceptable level ecologically significant adverse effects to populations of bottom-dwelling life from hazardous substances, including wood waste degradation chemicals. The Alaska Department of Environmental Conservation (ADEC) determined that the RAO would best be obtained by natural recovery with long-term monitoring every 10 years. The ultimate goal is to have 75 percent of the AOC in an equilibrium community by the year 2040.

Approach/Activities. The long-term monitoring program was designed to measure the degree of natural recovery toward the natural resource management milestones outlined in the ROD. The baseline survey was carried out in the spring and fall of 2000 using sediment profile imaging (SPI), epifaunal video surveys, benthic community analyses, and sediment chemical analyses. Based on the positive results from the baseline monitoring, a cost-effective strategy was recommended for future monitoring that would save more than 50% of the projected future costs of the program. In 2011, the next round of monitoring based on baseline recommendations was carried out with sediment profile and plan view imaging, sediment chemical analyses, and bioaccumulation testing to address concerns about dioxin contamination.

Results/Lessons Learned. Natural recovery in the benthic ecosystem is occurring faster than originally predicted despite the slow decomposition rate of the wood waste, and bioaccumulation testing showed there were no risks posed by the low concentrations of dioxin detected in the sediment.

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Monitored Natural Recovery for Onondaga Lake: A Progress Report

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Background/Objectives. The Onondaga Lake (in Central New York State) Record of Decision specifies monitored natural recovery (MNR) as the remedy to achieve sediment performance criteria established for the profundal zone (deep-water areas that comprise 70 percent of the lake surface area). To evaluate the effectiveness of MNR, site-specific numerical modeling supported by several types of ongoing monitoring data was used to predict mercury concentrations into the future and estimate the time to recovery and to determine the likely future effectiveness of MNR. The remedy is predicted to successfully meet the sediment performance criteria prior to the end of the prescribed MNR period, which is the 10 years following the remediation of upland sources and the littoral zone (2027).

Approach/Activities. A site-specific model, developed in Microsoft® Excel using Visual Basic for Applications (VBA), is based on sediment processes that are well established in literature and incorporates site-specific monitoring data on sedimentation rates, sediment mixing depth, and surface sediment mercury concentrations. Mixing and rate of sedimentation are primary processes resulting in natural recovery in the profundal zone. Both typical and innovative monitoring data collected on these processes included sediment traps, high-resolution mercury cores, radioisotope cores, frozen cores, and most recently, deployment and monitoring of fluorescent microbead markers that marked the mudline elevation to quantify the thickness of newly deposited material. Supported by this monitoring data, the model was calibrated to mercury concentrations from surface sediment collected between 1992 and 2011 at almost 100 locations.

Results/Lessons Learned. Although previously available lines of evidence, such as macroinvertebrate sampling and current shear-stress evaluations, indicate that very little mixing is occurring in the profundal zone sediments, data from frozen cores and radioisotope cores were used to specify the site-specific mixing depth in the model. Visual inspection of frozen cores and the majority of radioisotope cores indicated that mixing of sediment in most areas of the profundal zone is taking place in the top few centimeters only. Sedimentation rates were also examined using the radioisotope cores, high-resolution mercury cores, and sediment trap data. Average sedimentation rates from these sampling programs ranged from 0.26 to 0.34 grams per square centimeter per year (g/cm2/yr). Cores collected from microbead marker plots in summer 2012 show the fluorescent microbead marker is clearly visible and at a depth below top of sediment consistent and is consistent with prior estimates of sedimentation.

Refined calibrated modeling based on these recent data indicated that observed mixing and sedimentation rates are expected to continue to support the attainment of the sediment performance criteria prior to the end of the 10-year MNR period. Model calibration and projection results will be presented.

Natural Recovery of PCB Sediments in Lake Allegan, Michigan

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Background/Objectives. Lake Allegan, a 1,650-acre impoundment of the Kalamazoo River is a significant repository of polychlorinated biphenyls (PCB) within the Kalamazoo River Superfund Site. Depositional processes have buried historic PCB accumulation under cleaner sediments since upstream sources have declined, leading to significant attenuation of PCBs in surface sediments and fish tissue. Residual PCB sources from eroding, exposed sediments in formerly impounded upstream areas could present limitations to ultimate recovery of the lake, unless controlled. This presentation describes an empirical record and mechanisms of natural recovery over the last two decades and examines the relationship between future recovery and incoming PCB loads.

Approach/Activities. PCB trends in sediment, surface water and fish data over the past 20 years are evaluated. Direct evidence of the burial and reduction in PCB surface concentrations are provided by three surface sediment sampling campaigns (1993, 2000, and 2009) and two series of radio-dated sediment core profiles (2000, 2009). Routine inlet/outlet surface water sampling by the State of Michigan and other sampling allow estimates of incoming PCB and sediment loads and retention in the lake. Mass balance analysis was conducted to examine recovery rate dependency on further source control.

Results/Lessons Learned. Sediment deposition in the lake averages 1.1 cm/year, based on Cesium-137 dated cores. Annual net retention of PCB (3.0 kg/yr) is 1.3 percent of the resident mass of PCB in the biologically active zone (BAZ, estimated to be the top 10 cm). The burial-driven annual loss of PCB mass from the BAZ (27 kg based on 2009 sediment data) is 11 percent of PCB mass in the BAZ. Sediment PCB profiles reflect burial-driven recovery with historic inventories and radionuclide profiles preserved at depth – consistent with long-term sedimentation. Lake-wide surface weighted average concentrations have declined from 2.9 mg/kg in 1994 to 0.99 mg/kg in 2009 - in agreement with core dating results, and observed rates of decline in fish tissue PCB levels. Radio-dated cores collected in 2009 following an extreme flow event produced by the remnants of Hurricane Ike in September 2008 indicate the event did not disrupt the continuing recovery of PCB levels in surface sediments. Inlet surface water PCB concentrations have also declined and will result in significant future reductions in lake bed PCB concentrations. With no further load reduction, surface sediment exposure concentrations in the BAZ are estimated to asymptote to approximately 0.11 mg/kg, approximately one tenth of concentrations measured in 2009. This analysis demonstrates the potential for natural recovery of Lake Allegan due to past upstream remedial actions and source control. A summary of important data for Lake Allegan and the mass balance analysis will be presented. The nature of the continuing PCB source and implications for recovery of PCB exposure concentrations in sediment and associated PCB levels in fish will be discussed.

The Long Haul: Results of Monitored Natural Recovery of the Field Verification Program Mound after Thirty Years

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Overview. Monitored Natural Recovery (MNR) is a sediment management technique that can require decades to evaluate. A comprehensive study was initiated in 1982 to evaluate the environmental consequences of dredged material placement under aquatic, wetland, and upland conditions. This study presents the results of MNR nearly thirty years after the experimental aquatic placement of uncapped dredged material in Long Island Sound.

Background/Objectives. An experimental mound was created in Long Island Sound as part of the joint USEPA/USACE Field Verification Program (FVP). The FVP objective was to field-verify existing methods for predicting the environmental consequences of dredged material placement. The FVP unconfined open-water disposal mound was created from the disposal of 55,000 m^3 of Black Rock Harbor (BRH) sediment in 1983. BRH sediment consisted of black, fine-grained silt and clay with high water content and elevated concentrations of metal and organic contaminants. Exposure to BRH sediment in laboratory tests resulted in both chronic and acute toxicity in several test species, as well as PCB and PAH bioaccumulation. The underlying assumption was that if adverse biological effects were observed in the laboratory, they should also be measurable in the natural environmental following placement of the unsuitable material in open water. Repeated monitoring of the mound has shown a wide range of benthic community responses, from an initial classic primary successional recovery following disturbance, to episodes of retrograde succession following Hurricane Gloria and hypoxic events in Long Island Sound. After 1988, the New England District continued to monitor the mound to observe and document the longer term environmental responses.

Approach/Activities. Benthic community conditions were surveyed with sediment profile imaging (SPI) in 1991, 1993, 1995, 1999, 2005 and 2011. Bulk sediment from grabs and cores has been collected and analyzed, most recently for PAHs, PCBs and metals in 2005 to assess distribution of contaminants within the mound. In 1997, 2000, 2005 and 2011 high resolution acoustic data were collected to assess small scale (< 10 cm vertical) changes in the surface of the mound and surrounding seafloor due to sediment transport processes.

Results/Lessons Learned. Multiple lines of evidence (SPI images, sediment chemistry, bathymetry, backscatter and side-scan sonar) demonstrated that the surface sediments at FVP support a robust, actively bioturbating benthic community with no evidence of sediment transport beyond >1 cm layers of fine sand. Thirty years of Monitored Natural Recovery of the FVP mound, through bioturbation and ambient sedimentation (~ 15 cm), has modified the seafloor to the point that there is now little biological or chemical difference relative to reference area sediments. BRH sediments are still present below the mound surface, but are not currently affecting the benthic community at the surface as a result of the long-term sediment deposition.