

Investigating toxicological effects of short-term resuspension of metal-contaminated freshwater
and marine sediments

By

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Abstract

Sediments in navigation-dominated waterways are frequently contaminated with a wide range of chemicals and are subject to resuspension events. There is little information documenting whether adverse ecological effects result from these resuspension of contaminated sediments. I studied the mobilization of metals (Zn, Cu, Cd, Pb, Ni and Cr) during resuspensions of freshwater and marine sediments and whether or not this resulted in toxicity to organisms. Sediment resuspension events were simulated using sediment flux exposure chambers (SeFEC) to resuspend metal-contaminated sediments from Lake DePue (IL) and two Portsmouth (ME) Naval Shipyard sites. Various resuspension events of environmentally relevant suspended particulate matter concentrations (200-1,000 mg/L) were performed. Short-term resuspension events (4 h) resulted in metal- and sediment-specific mobilization. Toxic effects on organisms were limited (*Hyalella azteca* and *Daphnia magna* survival, *Neanthes arenaceodentata* survival, growth and body burden, *Pyrocystis lunula* bioluminescence) over both the time period of the exposure and during a 4 to 10-d post-recovery period. I conclude that sediment resuspension events at relevant suspended particulate matter concentrations do not release large amounts of dissolved metals, most likely due to the oxidation of reduced iron, manganese and amorphous sulfide species and subsequent readsorption to iron and manganese oxides and organic matter, therefore limiting organisms' exposure and effects. The redeposition of suspended sediments and associated metal speciation changes, from sulfides to oxides, has the potential to increase metal bioavailability and toxicity to sensitive benthic organisms. While the release of dissolved metals are low, resuspension events still pose the risk of causing toxic responses due to mobilization of contaminated sediments.

Table of Contents

1.0 Introduction.....	1
2.0 Material and Methods	6
2.1 Site Descriptions	6
2.2 System Setup	7
2.3 Sampling Protocol	8
2.4 Sample Analysis	12
2.5 Quality Assurance and Data Analysis	14
3.0 Results.....	16
3.1 Sediment Characterization	16
3.2 Overlying Water Comparisons (bedded v. resuspension)	18
3.3 Sediment Comparisons (bedded v. redeposited)	25
4.0 Discussion	30
5.0 Conclusion	37
Supporting information.....	39
Appendix.....	48
Literature Cited.....	54

1. Introduction

In aquatic systems, sediments play an important ecological role for aquatic organisms; however, sediments are also sinks for many contaminants. One such class of contaminants are metals (e.g., Zn, Cu, Cd, Cr, Ni, Pb), and in aquatic sediments, metals can be complexed to a number of ligands, including reduced sulfur species, organic matter, carbonates, iron and manganese oxides (Luoma and Bryan, 1981, Calmano et al 1993, Cantwell et al. 2002). When complexed, metals are considered non-bioavailable and therefore, not toxic to resident biota (Calmano et al. 1993, Chapman et al. 1998, Eggleton and Thomas 2004). Many metal complexes are reversible and environmental conditions (e.g., pH, redox) can alter the partitioning of metals. Sediment disturbances, such as resuspension due to propeller wash events, expose anoxic sediments to oxygen. The chemical changes that occur when reduced metal species are oxidized creates a complex scenario where there is potential for solubilization of, which can potentially cause toxicity to aquatic organisms (Fig.1, Zhuang et al. 1994, Simpson et al. 1998, Caetano et al. 2002, Hill et al. 2009, Kalnejais et al. 2010, Hong et al. 2011a).

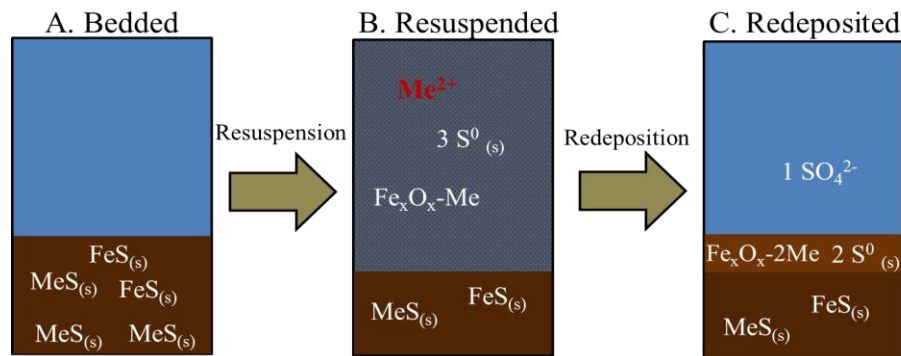


Figure 1. Conceptual model of important metal (Me) speciation in various sediment redox states. In bedded sediment (A) metals are typically bound as insoluble sulfides. When resuspended, (B) sulfide species are oxidized, mobilizing metals into the overlying water. As particles redeposit (C), mobilized metals are scavenged by Fe_xO_x and returned to the benthic environment, with further oxidation of sulfur to sulfate. Note that not all potential chemical reactions and species are represented in this schematic, and sulfur stoichiometry is balanced for clarity but speciation may differ depending on environmental conditions.

Assessments of environmental risk are usually based on assumptions of equilibrium conditions existing in sediments and effect thresholds are derived from steady-state laboratory exposures in non-turbid systems (Burton et al. 2005, Burton and Johnston 2010). In human-dominated watersheds, harbors and navigation channels continuously experience sediment resuspension events due to propeller wash, tidal actions, currents, and storm events (Kalnejais et al. 2007), which results in systems not under equilibrium conditions. The use of steady-state models to describe these ecosystems results in substantial uncertainties in any determinations of toxicological risk, because steady-state exposures do not reflect environmentally relevant conditions. The environmental significance of resuspension events is ill-defined, and given the magnitude of occurrence there needs to be a better quantified approach to facilitate more accurate risk assessments and management decisions.

Managing the risks associated with the resuspension of metal contaminated sediments requires knowledge of the dominant physical and chemical processes; particularly those environmental conditions (pH, Eh, O₂) that are important for metal speciation (Calmano et al. 1993, Saulnier and Mucci 2000, Cantwell et al. 2002). It has been observed that overlying water pH and dissolved O₂ are the main factors controlling iron, manganese and sulfur speciation, which in turn influence metal mobilization during sediment resuspension events (Atkinson et al. 2007, Jones-Lee and Lee 2005, Hong et al. 2011a). Readily oxidized sulfide minerals (FeS and MnS) are rapidly oxidized (Simpson et al. 1998), and such oxidation reactions can hydrolyze water and lower pH (Calmano et al. 1993). These processes lead to sediment particles being suspended in oxidizing, acidic conditions that can potentially cause dissolution of particulate-bound metals (Saulnier and Mucci 2000). In marine sediments, pyrite (FeS₂) is the main sulfur species, and once metals coprecipitate with pyrite they become more resistant to oxidation during

short-term resuspension events compared to more amorphous, reactive sulfide phases, such as mackinawite (Morse and Luther, 1999, Caetano et al. 2002, Rickard and Morse, 2005). Other metal sulfides, such as CdS, CuS, PbS and ZnS, can also take more than 8 h to be oxidized and dissolved (Simpson et al. 1998). Particulate and dissolved organic matter can limit the mobilization of metals, because of metals' affinity for organic ligands (Chapman et al. 1998, Cantwell et al. 2008). Complexation of metals to dissolved ligands (i.e., DOC and inorganic anions) can increase metal solubility, but decrease bioavailability (Chapman et al. 1998, Hong et al. 2011a). Sediments with a greater fraction of fine particles (i.e., silt and clay) have greater particle surface area and cation exchange capacity which can lead to greater metal sorption capacity limiting metal bioavailability, and increased scavenging of metals (Cantwell et al. 2002, Cantwell et al. 2008, Kalnejais et al. 2010).

While dissolved metals are considered the most important fraction when discussing bioavailability, the resuspension of particulate bound metals can also negatively impact ecosystems and resident biota through ingestion and transformation (i.e., remobilization) of particulate bound metals in the gut (Calmano et al. 1993, Allen et al. 1995, Schlegel et al. 2000, Weltens et al. 2000, Hill et al. 2009). Suspended particles are enriched in metals compared to the bedded sediments (Cantwell et al. 2002, Kalnejais et al. 2007), thus creating scenarios where the presence of suspended particles may lead to increased exposure to metals. In combination with aqueous exposure to metals, contaminated suspended particles have been shown to increase lethal and sublethal toxicity when compared to aqueous-only exposures (Weltens et al. 2000, Hill et al. 2009, Cloran et al. 2010). However, the toxicity of suspended particles depends on the primary metal contaminant (Fichet et al. 1998). The bioavailability of particulate bound metals is

controlled by the metal partitioning behavior, binding strength, and also potential liberation after organism digestion (Bryan and Langston 1992, Calmano et al. 1993).

Exposure to either contaminated particles or dissolved metals during resuspension is not the only concern for biota because these sediments are redeposited, often under different physical and chemical conditions. Redox conditions in sediment are vertically stratified due to differences in dissolved oxygen concentrations, with oxic conditions at the surface and anoxia at depth. Resuspension of sediment may temporarily disrupt vertical stratification; as stratification reestablishes, redox sensitive molecules (e.g., sulfide) in redeposited sediments can be oxidized, which can potentially increase metal bioavailability to benthic organisms (Fig. 1). Bonnet et al. (2000) investigated this modification of redox stratification due to resuspension and found no significant differences in toxicological effects to *Chironomus tentans* and *Hyaella azteca*. Changes in redox stratification during non-equilibrium episodes are not well studied so further investigation is necessary.

While toxicological studies are becoming more environmentally realistic by introducing non-equilibrium conditions, many resuspension studies have been done either as slurry experiments or with unrealistic levels of suspended particulate matter (SPM), ranging from 10 – 3,000 g sediment/L (Fichet et al. 1998, Simpson et al. 1998, Bonnet et al. 2000, Caetano et al. 2003, Atkinson et al. 2007, Maddock et al. 2007, Cantwell et al. 2008, Hong et al. 2011a, Duran et al. 2012). In natural systems, SPM concentrations rarely exceed 0.8-1 g/L during resuspension events (Ludwig and Probst 1998, Wilber and Clarke 2001, Reine et al. 2007). While these studies are important for elucidating mechanisms of release and establishing worst-case resuspension events, the release of metals into the overlying water and associated toxicological effects may over exaggerate what is realistic under environmentally relevant conditions.

The studies listed above have begun to investigate environmental risk under non-equilibrium conditions, thus improving our understanding of contaminant flux in human-dominated ecosystems, but lack realism in at least one dimension (i.e., SPM concentrations, resuspension duration, use of laboratory formulated sediments). The objective of this study is to examine the changes in metal bioavailability in a variety of naturally-contaminated sediments during more realistic laboratory resuspension events. Further, concurrent toxicity assays with key indicator organisms were used to link exposure to effects. I examined changes in environmental conditions during bedded and resuspension events and investigated how these changes affected fluxes of metals in freshwater and marine sediments. Effects on biota from exposures to 4-h resuspension and bedded scenarios were investigated with relevant biological endpoints (i.e., survival, growth, bioluminescence, metal-tissue content). Changes in toxicity due to shifts in vertical redox stratifications were examined with bedded and redeposited sediment toxicity tests. Results from this study further contribute to understanding the environmental risks of sediment resuspension events, linking organism exposure to effects, and provide a more holistic characterization to better assess and manage these associated risks.

2. Methods

2.1 Site descriptions

Lake DePue, IL, USA (41° 19' N, 89° 18' W) is a shallow lake connected to the Illinois River by a small channel (Gough et al. 2008). The lake is contaminated from historic industrial activities, including ~80 years of Zn-smelting, along with sulfuric acid and diammonium phosphate fertilizer production (Webb et al. 2000, Cahill and Bogner 2002). The lake has a gradient of Zn concentrations in sediment, ranging from 300 to 42,300 mg/kg, from the western side of the lake towards the decommissioned smelting facility in the east (Cahill and Bogner, 2002). The site is currently listed on the United States Environmental Protection Agency (USEPA) National Priority List (NPL, Site ID# ILD062340641)). Sediment samples were collected in September 2011 and stored under anoxic conditions at 4 °C in the laboratory.

Portsmouth Naval Shipyard (PNS, 43° 5' N, 70° 44' W) in Kittery, ME, USA is located at the mouth of the Great Bay Estuary and Piscataqua River. The shipyard was established as a government facility in the early 1800's, and has built and repaired ships and submarines to date (USEPA, 2007a). The site is currently listed on the USEPA NPL (Site ID# ME7170022019). Sediments were collected from two sites, MS04 location 1 and MS03 location 2 (USEPA, 2007a). The sediments at these sites are contaminated with a mixture of metals and PAHs from foundry slag, historic industrial point sources and groundwater/surface water runoff (USEPA, 2007a). Sediment samples were collected in September 2012 and stored similarly to those from Lake DePue.

2.2. System setup

Sediment flux exposure chambers (SeFEC) were modified from designs of Hammerschmidt and Fitzgerald (2008) and Eggleston (2011). The SeFEC chambers were constructed out of polycarbonate core tubes (7.1 cm ID, 16.5 cm height, ~580 ml volume) with polycarbonate end caps. The chambers were modified into a recirculating-system, with two chambers per experimental unit: a resuspension chamber and an organism exposure chamber (Fig. 2). Sediment resuspension was driven by an inverted Wheaton quad stir plate (Wheaton, Millville, NJ) and Teflon coated stir bar (1.5 inch starburst stirring head, Cole Parmer). Homogenized sediment was placed into the resuspension chamber to create a 2.5 to 3.5 cm core depending on sediment type. A sampling port at 1.5 cm from the bottom was included to take porewater samples via Rhizon CSS soil samplers (Rhizosphere Research Products, The Netherlands).

To initiate resuspension, the inverted stir plate was turned on and the speed was adjusted (370-400 rpm) to achieve approximately 200 - 1,000 mg/L of suspended particulate matter (SPM). Overlying water containing suspended particles was recirculated between the resuspension and exposure chambers by a Gilson Minipuls peristaltic pump (Gilson, Inc. Middleton, WI) at 12.5 mL/min python airline tubing (Python Products, Inc. Milwaukee, WI). Initial experiments demonstrated it took about 1 to 1.25 h for complete mixing of the water column between the two chambers. The SPM in the exposure chamber was kept in suspension by an identical quad stir plate and stir bar as above (Fig. 2).

The exposure chamber contained an inner chamber to contain test organisms. The inner chamber was a polycarbonate tube, 9 cm height x 2 cm ID with a 26.4 cm² window covered with 243 µm Nitex bolting cloth (Wildlife Supply Company, Yulee, FL). The 243 µm mesh size

allowed for mixing of suspended particles between the exposure and inner chambers (Burton et al. 2005). All resuspension and bedded (non-resuspension) sediment experiments were four hours in duration with four replicates and 10 organisms per replicate.

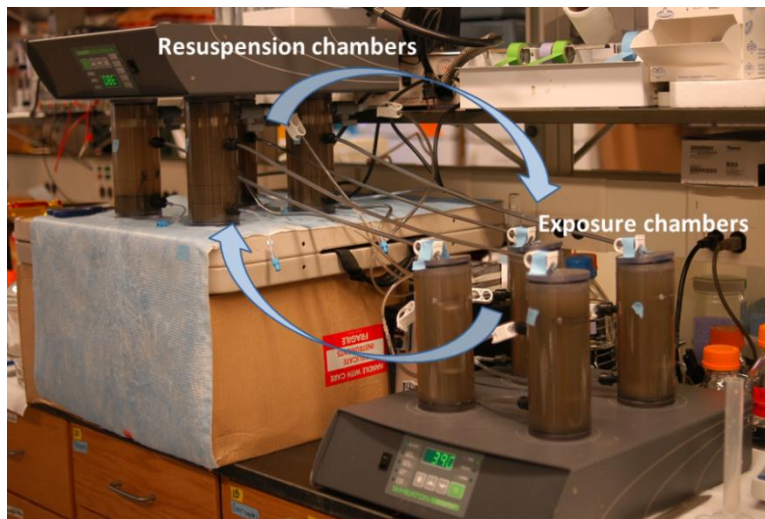


Figure 2. Sediment flux exposure chambers (SeFEC) recirculating resuspension system with separate resuspension and exposure chambers.

2.3 Sampling protocol

2.3a. Overview

For each sediment type, we chemically characterized and measured toxicity of three sediment integrity conditions: bedded, resuspended, and redeposited sediment (Fig. 1). Direct comparisons in geochemistry and toxicity were made between: 1) the overlying water during 4-h bedded and resuspension experiments (Fig. 1. panels A and B) and 2) the benthic environment during bedded and redeposited sediment experiments (Fig. 1. panels A and C). In the above scenarios, toxicity was assessed with either an epibenthic/benthic (either *Hyalella azteca* or *Neanthes arenaceodentata*) and pelagic (either *Daphnia magna* or *Pyrocystis lunula*) organism for each of the sediments. Exposure time for all organisms was 4 h, and followed by either: 1)

depuration and tissue metal analysis, or 2) held for 4 to 10 d (depending on organism) to monitor post-exposure survival and growth. Freshwater experiments with Lake DePue sediment were conducted with moderately hard reconstituted water (101 mg/L CaCO₃) (Smith et al. 1997) and marine experiments with PNS sediments were conducted with artificial seawater prepared from commercial sea salt to a salinity of 31 (Tropic Marin, Wartenberg, Germany).

2.3b. Overlying water comparisons (bedded v. resuspension)

Geochemistry

Bedded and resuspension experiments were completed identically in the SeFEC recirculating system, either with or without induced shear stress. Physicochemical parameters (dissolved O₂, pH, temperature and conductivity) were measured hourly in the exposure chamber. Porewater for metal analysis was sampled from the resuspension chamber before experiment initiation and overlying water in the exposure chamber was monitored over the experiment duration (0, 1 and 4 h). SPM was collected during resuspension experiments for determination of total suspended solids, particulate metal concentrations (1 and 4 h) and particulate organic carbon (POC, 4 h). Turbidity (NTU) was measured with Hach 2100N turbidimeter (Loveland, CO.). Porewater and overlying water was filtered (0.2 µm) and acidified to 2% with trace metal grade Nitric acid (Fisher Scientific).

Toxicity

For freshwater exposures, lab-reared *H. azteca* (10 organisms, 7-14 d old) and *D. magna* (10 neonates <24 h old) were placed within the inner chambers and exposed to recirculating water for 4 h. Following the exposure, five of the *H. azteca* were depurated and digested for determination of metal body burden (Appendix A3) and the remaining five organisms were

placed in 150 ml of clean reconstituted water with a strip of unbleached paper towel and three pre-weighed, preconditioned 1-cm diameter disks of red maple leaf (*Acer rubrum*). Organisms were held for an additional 6 d (7-d total exposure) to monitor survival and leaf disk processing (Wilding and Maltby 2006). Following either bedded or resuspension exposures, *D. magna* were placed in 25 ml of clean reconstituted water and monitored for an additional 3 d (4-d total exposure) to determine survival. Each beaker of 10 organisms was fed 0.25 ml of Sel-cero (*Selenastrum capricornutum* and wheat grass powder suspension) at initiation and on 3 d.

For marine exposures, ten 2 to 3-week post-emergent *N. arenaceodentata* (Aquatic Toxicology Support, Bremerton, WA) were placed in the inner chambers. Following the 4-h exposure, five organisms were placed into clean seawater for depuration and analysis of subsequent metal body burden. The other five organisms were placed in 150 ml of clean seawater and held for an additional 9 d (10-d total exposure) to monitor survival and growth rates. Growth rates were determined by comparing body mass after 10 d to those at test initiation (ASTM 2007). *Neanthes arenaceodentata* were fed (10 mg rabbit chow) at initiation and every third day; following water changes of two-thirds volume (modified from ASTM 2007).

Pyrocystis lunula (Lapota et al. 2007, Qwiklite biosensor, Assurance Controls, Vista, CA.) were exposed to filtered and unfiltered samples taken after the 4-h experimental durations. After dosing, the sample-organism mixture was incubated for 24-h in a 12:12 light - dark cycle. Following this exposure, the light intensity of the bioluminescent organisms was measured and compared to unexposed controls, with results being expressed as a percent bioluminescence inhibition. MS04 bedded experiment control and treatments had low output intensities (4 to 5 times lower than other experiments) and were excluded from statistical analyses.

Controls for *H. azteca*, *D. magna* and *N. arenaceodentata* exposures consisted of four additional replicates that were not exposed to either contaminated water or sediment. Organisms were held in 25 ml of clean water without food during 4-h test durations, but otherwise treated identically through either the depuration and digestion or post-exposure hold procedures. Overlying water quality (dissolved O₂ and pH) was monitored and remained satisfactory over the duration of the experiment.

2.3c. Sediment comparisons (bedded v. redeposited)

Geochemistry

After resuspension experiments, bedded sediments were subsampled with cores (10-mm deep, sectioned at 5-mm resolution) to investigate effects of resuspension on acid volatile sulfides (AVS) and simultaneously extracted metals (SEM) depth profiles. Un-manipulated sediment was used to determine pre-resuspension (bedded) conditions. Additionally, water for analysis of filter-passing metals was collected in the resuspension chambers after overnight settling (additional ~20 h) to determine bedded and redeposited metal fluxes. Filtered water samples were also taken before various water changes (see below) to monitor metal concentrations in the test chambers.

Toxicity tests

Sediment cores resulting from bedded and resuspension experiments were used for a static-renewal sediment toxicity test. An additional resuspension experiment was completed to obtain redeposited cores, due to the destructive nature of sampling for AVS/SEM analyses. Following the 4-h experiment, the chambers sat overnight (~20 h), received a 100% water change, and three leaf disks and organisms were added, which allowed for direct contact with the

bedded or redeposited sediments (modified USEPA, 2000; ASTM, 2007). For freshwater experiments, 10 *D. magna* and 10 *H. azteca* were included in the test for 4 and 7 d, respectively. For marine experiments, five *N. arenaceodentata* were included for 10 d and fed (10 mg rabbit chow in each chamber) every third day (ASTM, 2007). Additionally, *P. lunula* were exposed to filtered marine sediment elutriates (1:4 sediment to water ratio) for determination of bedded and redeposited sediment toxicity. Control chambers were setup with clean water and either unbleached paper towel and leaf disks (*D. magna* and *H. azteca*) or clean Ottawa sand (*N. arenaceodentata*). Treatments and controls for freshwater and marine experiments received one volume (~275 ml) water change per day with intermittent physicochemical sampling to insure adequate water quality. Following the various exposure durations, surviving organisms were counted, and *H. azteca* (Appendix A4) and *N. arenaceodentata* were depurated prior to metal analysis in their tissue. Relative growth rates were determined for *N. arenaceodentata*.

2.4 Sample analysis

2.4a. Sediment characterization

Water content of sediments was determined gravimetrically after drying a subsample of homogenized sediment (~10 g) for 24 h at 100°C. The dried sediment was homogenized with a mortar and pestle for analysis of total carbon (TC), total organic carbon (TOC), and total recoverable metals. TOC was determined by pretreatment of ~20 mg sediment in silver capsules with 50 µL 1% HCl, followed by 8 h fumigation with 12 M HCl (Walthert et al. 2010). TOC and TC were determined with a CHN Elemental Analyzer (Costech Elemental Combustion System).

Total recoverable metals were determined by microwave assisted (MARS 5, CEM, Matthews, NC) acid digestions with a 3:1 ratio of HNO₃ and HCl (USEPA 2007b). The digestate was filtered (0.2 µm polycarbonate membrane), diluted, and analyzed by inductively coupled

plasma-optical emission spectrometry (ICP-OES). PNS digestates and wet PNS sediment samples were sent to Calscience Environmental Laboratories (Garden Grove, CA) for determination of total metals (EPA method 6010B) and PAHs (EPA method 3545 followed by method 8270C).

Acid volatile sulfides (AVS) and simultaneously extracted metals (SEM) were determined by procedures developed by USEPA (1991). Briefly, AVS was determined by acidification of sediment-water slurry to 1M HCl, trapping the generated H₂S in NaOH, and colorimetric analysis (USEPA 1991). The sediment-water slurry was filtered (0.2 μm), quantitatively diluted, and analyzed for SEM by ICP-OES. Total reduced inorganic sulfur (TRIS) was extracted with 5.65M HCl and chromium powder (Sullivan et al. 2000); TRIS was trapped and determined colorimetrically, as described above for AVS. Sediment grain size distribution was determined by wet-sieving the sediment slurry for 30 min through a series of sieves, which was dependent on sediment type (A.I. Packman, Northwestern University, Evanston, IL, USA, personal communication). The resulting sediment fractions were dried at 70° C, and weighed to calculate the mass fraction in each size class. Permeability was measured by imposing a constant head difference of 1.6 m on a filtration column (ID 2.5 cm) (Omnifit, Danbury, CT, USA) packed with sediment.

2.4b. Dissolved and particulate metals and organic carbon

Water samples were syringe filtered with a 25 mm 0.22 μm Isopore™ polycarbonate membrane filter and housing (EMD Millipore Corporation, Billerica, MA). Water samples were acidified to 2% with trace metal grade Nitric acid (Fisher Scientific). Metals in freshwater were analyzed by ICP-OES, while was sent to Calscience Environmental Laboratories (Garden Grove, CA) and analyzed via EPA method 6010B. Dried filters were weighed and microwave digested

as detailed for sediments (USEPA 2007b). Digestates were filtered (0.2 µm), diluted, and analyzed via ICP-OES. Additional aliquots of filtered water (0.2 µm) were acidified to a pH of 1-2 for determination of dissolved organic carbon on a carbon analyzer (OI Analytical TOC Analyzer). Samples for analysis of particulate organic carbon (POC) were collected by filtering water through 25 mm GF/F glass fiber filters (Whatman Corp). POC was determined by pretreatment of filters with 50 µL 1% HCl, followed by 8 h fumigation with 12 M HCl (Walther et al. 2010). The filters were dried and rolled in 25 mm tin discs. These samples were analyzed on a Costech Elemental Analyzer.

2.4d. Tissue metal analysis

Metal body burden was determined on surviving *H. azteca* and *N. arenaceodentata*. *Hyaella azteca* were depurated for 24 h in reconstituted water containing ~40 µM of EDTA and food (ground tetramin). *Neanthes arenaceodentata* were removed from test chambers and depurated for 24 h in clean seawater containing 10 mg rabbit chow. Organisms were dried (60°C), weighed (to nearest 0.001 mg) and digested for 6 d in nitric acid (Trace Metal Grade, Fisher Scientific), followed by the addition hydrogen peroxide (ACS grade, Fisher Scientific) for an additional 24 h (Norwood et al. 2006, Rosen et al. 2008a). The digested organisms were diluted and analyzed by inductively coupled plasma mass spectrometry (Elan 9000 ICP-MS).

2.5 Quality Assurance and Data Analysis

All appropriate quality control analyses were done and within acceptable ranges (Table SI.1.). Statistical analyses were conducted using R 2.14.0 (R Development Core Team). Two way ANOVAs were used to investigate differences in endpoints (i.e., survival, growth and tissue concentrations) for overlying-water and sediment exposures. A significant interaction between

sediment integrity (e.g., bedded v. redeposited) and exposure conditions (i.e., control v. treatment) indicates an effect of resuspension on the endpoint of interest. For experiments with incomplete treatment combinations (i.e., *H. azteca* feeding was not measured from bedded exposures) we conducted two-sample t-tests. A nonlinear least squares regression with a Michaelis-Menten kinetics equation was fit to the filtered metal release data for Lake DePue sediments (Bolker 2008). One-way ANOVAs and t-tests were used to determine differences in Qwiklite results. Two sample t-tests were used to investigate differences in AVS, SEM and $\Sigma(\text{SEM-AVS})/\text{foc}$ between bedded and resuspended samples. Simple linear regressions were used to determine relationships between pH and dissolved O₂ with turbidity. When necessary, data was normalized with appropriate transformations (natural log and arcsine square root) to insure assumptions were met (equal variance and normality). Non-parametric tests (i.e., Kruskal-Wallis test) were completed when normality could not be achieved with transformations.

3. Results

3.1 Sediment Characterization

Sediments from the three study locations were anoxic (porewater $O_2 < 1$ mg/L), circumneutral (porewater pH $\sim 7.0 - 7.3$), with high concentrations of sulfide, and moderate organic carbon content (Table 1 and SI.2). The sediments differed in texture with the freshwater sediment containing more fine particles (<45 μm , Lake DePue = 81%) than the marine sediments (PNS MS04 = 39% and PNS MS03 = 23%) (SI.3). Despite differences in particle size distributions the permeability of all sediment types was very low (10^{-15} to 10^{-14} m^2). Porewater DOC concentrations varied by sediment type, with Lake DePue (15 ± 1 mg/L) having lower DOC than both MS03 (103 ± 6 mg/L), and MS04 (98 ± 4 mg/L). MS03 and MS04 sediments contained appreciable amounts of sulfide in excess of AVS, (97 and 91 $\mu\text{mol/g dw}$, respectively) which is an approximation of pyrite-S (Table 1, Cantwell et al. 2002). In porewaters, Zn was detected in all three sediment types (28-146 $\mu\text{g/L}$), Ni was present at low levels in both PNS sediments (18 and 22 $\mu\text{g/L}$), and other metals (Cu, Cd, and Cr) were not detected (Table SI.2). All three sediments were potentially toxic; total recoverable metals in Lake DePue and PNS sediments exceeded effects thresholds based on probable effect levels (MacDonald et al. 1996 and Smith et al. 1996) and Lake DePue sediments also exceeded bioavailability thresholds ($(\sum\text{SEM-AVS})/f_{\text{oc}} > 130 - 3000$ $\mu\text{mol/g OC}$, USEPA 2005). The majority ($>85\%$) of total recoverable Zn and Mn in Lake DePue was 1M HCl-extractable (SEM) while other metals (Cu, Ni, and Fe) were less reactive (Table 2). In PNS sediments, Zn was also labile ($\sim 60\%$ of total recoverable as SEM), whereas Fe, Mn, Cu, Ni, Pb, and Cr had relatively little metal in the SEM fraction ($<30\%$) indicating the recalcitrance of these metals (Table 2). PAHs are present in PNS sediments but are at concentrations that are unlikely to cause toxicity (Long et al. 1995).

Table 1. Select mean geochemical parameters of three sediment types investigated in this study

	Total Metals (mg/kg dw) ^a								TOC (%) ^a	AVS (μmol/g dw) ^a	TRIS (μmol/g dw) ^b	Σ(SEM-AVS)/foc (μmol/g OC) ^b	ΣPAHs (μg/kg) ^c
	Fe	Mn	Cd	Cr	Cu	Pb	Ni	Zn					
PNS MSO3	39,600	332	ND ^d	96	575^e	181	69	533	1.6	20	117	-921	3178
PNS MS04	24,900	289	ND	114	364	83	48	223	2.0	9	98	-267	2052
Lake DePue	36,600	1,129	109	84	403	NA ^f	49	13,889	2.7	60	NA	4,973	NA

^a n = 3 ^b n = 4 ^c n = 1

^d ND = not detected

^e Bolded values exceed recommended probable effect levels (PELs, MacDonald et al. 1996 and Smith et al. 1996)

^f NA = Not available

Table 2. Fraction of total recoverable metals as 1M HCl extractable (SEM) in bedded sediments

	[SEM]/ [total recoverable metals] (%)		
	Lake DePue	MS03	MS04
Fe	36	14	22
Mn	86	12	11
Zn	95	55	65
Cu	25	9	15
Ni	45	29	26
Pb	NA ^a	6	7
Cr	NA	30	27
[AVS]/[TRS]	NA	17	9

^a NA = not available

3.2 Overlying water comparisons (bedded v. resuspension)

3.2a. Geochemistry

Physicochemical:

During resuspension events, suspended particulate matter in the exposure chambers were held between 250 – 1,000 mg/L in most replicates (SI.4 and 5). Resuspension of Lake DePue sediments caused a consistent decline in pH (~0.4 units, Fig. 3 and SI.4) of overlying water; pH declines were greatest after the first hour of resuspension, and then leveled off (Fig. 3A). Without resuspension, declines in pH were minimal (< ~0.05 units, SI.5). Turbidity (NTU) was unrelated to pH ($p = 0.17$, $R^2 = 0.28$). During resuspension experiments, dissolved O₂ in overlying water decreased but the overlying water remained oxidic (≥ 6.2 mg/L, Fig 1B). There was a strong inverse linear relationship between 4 h turbidity and O₂ concentrations ($p < 0.001$, $R^2 = 0.89$).

Resuspension of PNS sediments caused declines in pH (~0.2 units, Fig. 1 and SI.4) and unlike Lake DePue, pH in PNS declined steadily over the 4 h (Fig. 3A). Without resuspension,

declines in pH were minimal ($< \sim 0.05$ units, SI.5). For resuspension of MS03, turbidity was inversely related to pH ($p = 0.02$, $R^2 = 0.64$), but not for MS04 ($p = 0.84$, $R^2 = 0.01$). During the PNS resuspension experiments, dissolved O_2 in overlying water declined but the overlying water remained oxic (≥ 4.7 mg/L, Fig 3B). There was a strong inverse linear relationship between 4 h turbidity and O_2 concentrations ($p < 0.001$, $R^2 > 0.86$).

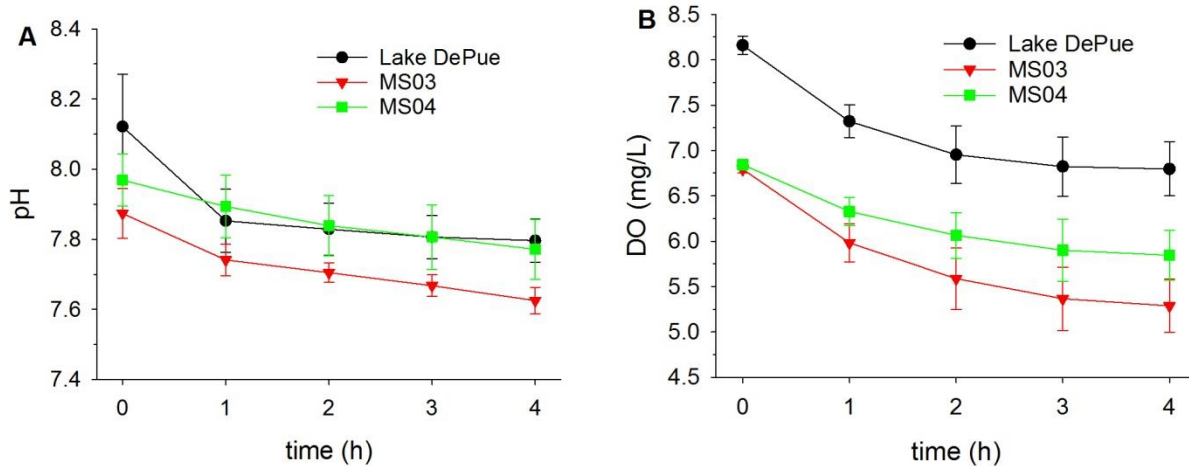


Figure 3. Mean (± 1 SD, $n = 8$) pH (A) and dissolved oxygen (B, DO) in overlying water during 4-h resuspension experiments. pH and DO remained stable through time for all bedded experiments (data not shown).

Dissolved metals

Resuspension of Lake DePue increased the mobilization of filtered Zn and Mn compared to bedded experiments (Fig. 4). Zn increased during the first 2 h of resuspension reaching a maximum of 83 ± 11 $\mu\text{g/L}$ at 3 h, and then leveled off, following a Michaelis-Menten kinetic function ($F_{1,30} = 156$, $R^2 = 0.73$, $p < 0.0001$, Fig. 4). During the bedded experiment, Zn diffused into the overlying water, reaching a maximum of 20 ± 3 $\mu\text{g/L}$ after 4 h. Mn showed similar trends as Zn, increasing the first two hours and then leveled off at ~ 120 $\mu\text{g/L}$, following

Michaelis-Menten kinetics ($F_{1,30} = 290$, $R^2 = 0.90$, $p < 0.0001$, Fig. 4). During the bedded experiment, Mn diffused into overlying water, reaching a maximum concentration of 5 ± 0.3 $\mu\text{g/L}$ at 4 h. Other dissolved metals (Fe, Cu, Cd, Cr and Ni) were near or below detection limits (SI 6a.). Even after overnight settling (~ 20 h) of resuspended particles, dissolved Zn remained significantly elevated compared to the bedded exposures (144 ± 3 v. 59 ± 8 $\mu\text{g/L}$, Mann Whitney U, $p = 0.029$). Mn followed the same trend, with dissolved Mn remaining in the overlying water after settling of resuspended particles (126 ± 7 v. 86 ± 9 $\mu\text{g/L}$, $p < 0.001$, $t = 7.15$).

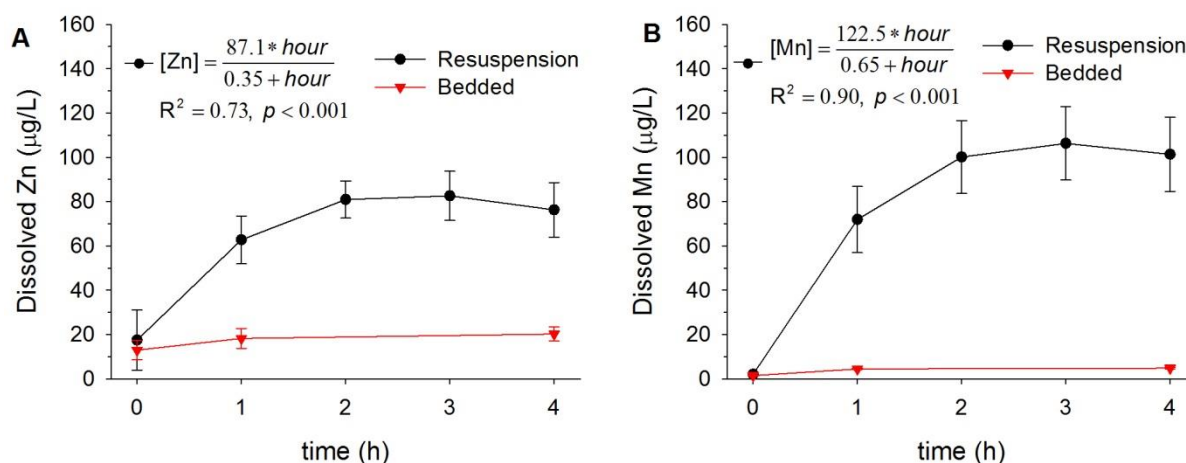


Figure 4. Mean (± 1 SD, $n=4$ to 8) filtered Zn (A) and Mn (B) during 4-h resuspension and bedded experiments for Lake DePue. Equation represents nonlinear least squares regression fit model for dissolved metal release during sediment resuspensions.

Resuspension of both MS03 and MS04 resulted in minimal release of metals to overlying water, with concentrations in filtered water below method detection limits (SI 6b and 6c). In MS03, Ni reached a maximum of 6 ± 1 $\mu\text{g/L}$ and Mn 16 ± 2 $\mu\text{g/L}$ after 4 h of resuspension (bedded Ni = not detected, Mn = 9 ± 0 $\mu\text{g/L}$). During MS04 resuspensions, Mn was the only metal observed in the filtered fraction in the overlying water (max 10 ± 1 $\mu\text{g/L}$, bedded Mn = 8 ± 0.1 $\mu\text{g/L}$). In both MS03 and MS04 bedded exposures, Zn increased marginally through time (SI

Table 6b and 6c) After settling overnight, there was no additional solubilization of metals into the overlying water for either sediment (SI 6b and 6c).

Particulate metals

Table 3. Mean (\pm 1 SD, n = 4 to 8) 4-h calculated metal enrichment factors (EF^a)

	Enrichment factor							
	Zn	Cu	Ni	Cr	Pb	Fe	Mn	OC ^b
Lake DePue	0.75 (0.02)	1.08 (0.04)	1.54 (0.35)	1.51 (0.10)	NA ^c	1.13 (0.05)	0.65 (0.03)	1.31 (0.02)
MS03	1.45 (0.06)	1.67 (0.09)	1.14 (0.09)	2.24 (0.14)	1.72 (0.02)	1.14 (0.07)	1.18 (0.08)	2.62 (0.78)
MS04	1.93 (0.16)	1.06 (0.10)	1.38 (0.11)	2.14 (0.15)	1.98 (0.20)	1.74 (0.13)	1.41 (0.08)	2.38 (0.42)

^aEF = [Me] SPM/[Me] bulk; EF > 1 indicate enrichment of metals in SPM compared to bulk sediment

^bOC = organic carbon

^cNA = not available

Generally, SPM enrichment factors (EF) were similar between 1-h and 4-h samples across all sediment types (SI.7). For Lake DePue resuspensions, Zn and Mn were the only metals for which EF was less than one (Table 3) and Ni, Cr and OC had the largest EF (1.54, 1.51, and 1.31, respectively). For MS03 and MS04 resuspensions, all EF were greater than 1; SPM was most enriched with Cr, Cu (MS03 only), and Pb (Table 3). In both PNS resuspension experiments, SPM was highly enriched in organic carbon, although the enrichment values were highly variable (Table 3).

3.2b. Toxicity

Lake DePue exposures

For the DePue sediment resuspension exposures, we observed no differences between our treatments for any of our toxicological endpoints. *Daphnia magna* survival was not affected by

exposure to overlying water from Lake DePue bedded (100%) or resuspension ($96 \pm 11\%$) trials (Fig. SI.1). Survival was lower (70%) in one of the replicate chambers that had the greatest concentration of SPM (1,012 mg/L), but dissolved metals were not elevated above other replicates, which suggests physical stress as a potential cause. Little additional mortality was observed when *D. magna* were held for 3 additional days for either exposure scenario (Fig. SI.1). *Hyalella azteca* survival was not affected by exposure to overlying water from bedded ($98 \pm 5\%$) or resuspended sediment ($98 \pm 5\%$) even after a 6-d post-experiment hold. *Hyalella azteca* feeding rates were not different between resuspension and unexposed controls ($p = 0.25$, $t = 1.35$, Fig. 5).

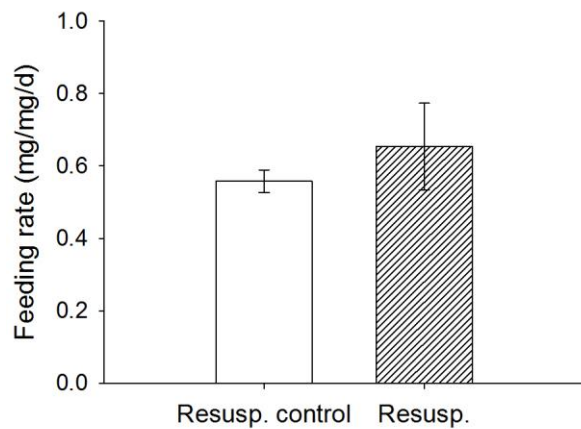


Figure 5. Mean (± 1 SD, $n = 3$) *Hyalella azteca* post-resuspension feeding rates.

PNS exposures:

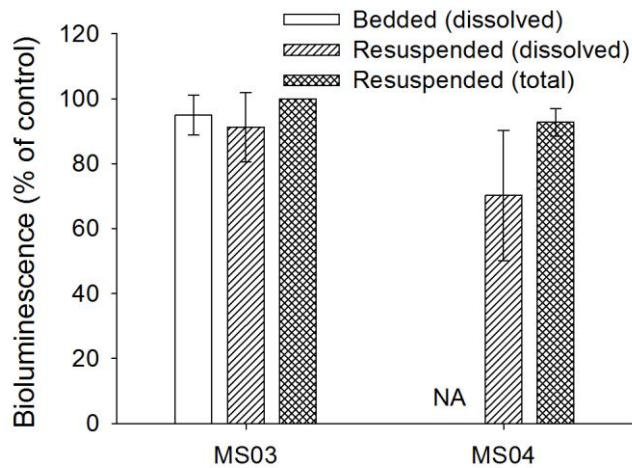


Figure 6. Mean (± 1 SD, $n = 4$) response of *Pyrocystis lunula* exposed to filtered and unfiltered overlying water during resuspension and bedded experiments for MS03 and MS04, expressed as % of control. Percentages > 100 were rounded down to 100% to allow for arcsine square root transformations. NA = not available.

Pyrocystis lunula exposed to overlying water from resuspension and bedded exposures showed minimal bioluminescence inhibition (Fig. 6). For MS03 exposures, there were no differences in the bioluminescence between organisms exposed to bedded, dissolved resuspension, or total resuspension 4-h water samples ($F_{2,9} = 2.8$, $p = 0.114$). For MS04 exposures, although treatments were not significantly different ($p = 0.07$, $t = 0.27$), filtered resuspension exposures exhibited the greatest light inhibition ($70 \pm 20\%$ of control), while unfiltered samples were less affected ($93 \pm 4\%$ of control, Fig. 6). *Neanthes arenaceodentata* survival was 100% following exposure to overlying water from MS03 and MS04 resuspension and bedded treatments and $> 95\%$ after the 9-d post exposure hold (Fig. SI.1). For MS03 and MS04 experiments, there was no difference in *N. arenaceodentata* relative growth rates ($\mu\text{g}/\text{ind}/\text{d}$) between any of our exposure treatments (all $p > 0.29$, Fig. 7). Organisms exposed to SPM had slightly reduced growth rates compared to control and bedded exposures, but results

were not significant (Fig. 7). Also, *N. arenaceodentata* exposed to SPM had slightly elevated concentrations of Zn, Cu, and Cr compared to bedded and control exposures; however, these differences were not statistically different due to large variability and low power (all $p > 0.05$, Table 4). Exposure to MS03 and MS04 resuspensions increased Zn body burdens 20 and 37% over controls, respectively. Ni, Pb, and Cr were very low in all exposure scenarios, typically < 5 mg/kg dw.

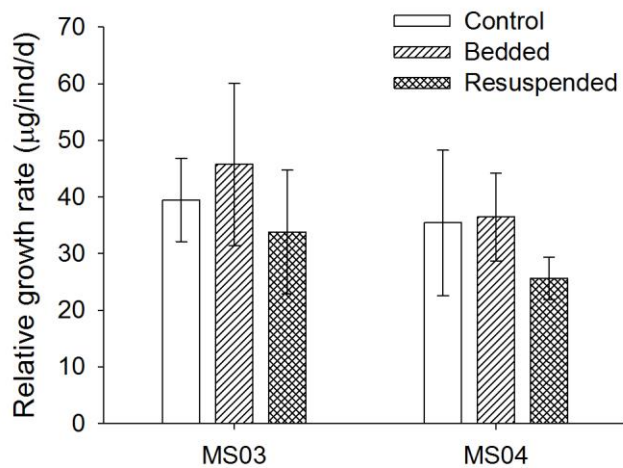


Figure 7. Mean (± 1 SD, $n = 4$ for bedded and resuspended, 8 for controls) *Neanthes arenaceodentata* relative growth rates between control, bedded and resuspended exposures after 9-d post-exposure hold. Bedded and resuspended controls for each sediment type are depicted as a single bar but were analyzed separately.

Table 4. Mean (± 1 SD, $n = 4$) *Neanthes arenaceodentata* body burden following 4-h resuspended and bedded experiments.

Sediment	Exposure	Body Burden (mg/kg dw)				
		Zn	Cu	Ni	Pb	Cr
MS04	Bedded control	137 (8)	24 (2)	3.1 (0.2)	0.8 (0.2)	2.5 (0.3)
	Bedded	159 (23)	24 (2)	3.5 (0.5)	0.8 (0.1)	2.7 (0.5)
	Resusp. control	130 (32)	26 (4)	3.9 (1.2)	0.8 (0.2)	3.9 (0.2)
	Resuspended	156 (23)	29 (4)	3.6 (1.0)	1.1 (0.4)	5.2 (1.9)
MS03	Bedded control	180 (36)	23 (2)	3.3 (0.7)	0.6 (0.1)	2.4 (0.4)
	Bedded	185 (21)	24 (2)	3.8 (0.6)	0.7 (0.1)	2.4 (0.4)
	Resusp. control	124 (20)	26 (1)	3.7 (0.3)	0.8 (0.2)	2.8 (0.9)
	Resuspended	170 (50)	25 (2)	4.0 (0.9)	0.9 (0.2)	3.9 (1.2)

3.3 Sediment comparisons (bedded v. redeposited)

3.3a. Geochemistry

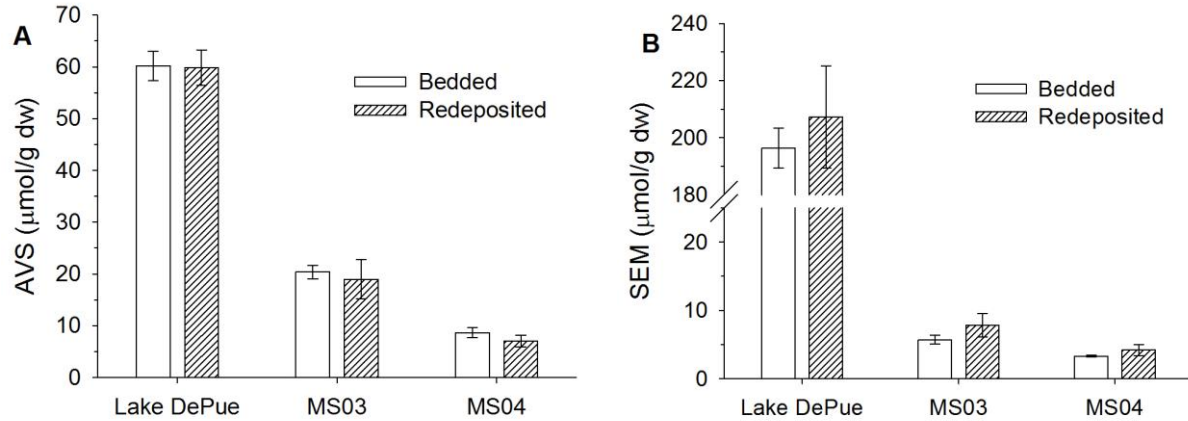


Figure 8. Mean (± 1 SD, $n = 4$) AVS (A) and SEM (B) for bedded and redeposited sediments. Graphs have different y-axes.

No differences in AVS or SEM (all $p > 0.05$) were observed between bedded and redeposited sediments (Fig. 8). We visually observed color changes (from dark to lighter) of surficial sediments and resuspended particles, indicating oxidation (Fig. SI.2), but this visual change was not reflected in a geochemical change, possibly due to coarse core sectioning (0.5 cm depth). Bioavailable metals (i.e., $\sum(\text{SEM}-\text{AVS})/\text{foc}$) were slightly higher in MS04 exposures ($p = 0.041$, $t = 2.59$) but not Lake DePue or MS03 (both $p > 0.19$, Table 5). Although we did observe slight changes in bioavailable metals, the changes were small and did not result in changes in predicted toxicity from bioavailability thresholds (USEPA 2005).

Table 5. Mean (± 1 SD, $n = 4$) $\sum(\text{SEM}-\text{AVS})/\text{foc}$ ($\mu\text{mol/g OC}$) bioavailability for bedded and redeposited sediments.

	$\sum(\text{SEM}-\text{AVS})/\text{foc}$ ($\mu\text{mol/g OC}$)		
	Lake DePue	MS03	MS04
Bedded	4,973 (251)	-921 (74)	-267 (49)
Redeposited	5,384 (631)	-701 (288)	-140 (84)

On day 7 of Lake DePue exposures, filtered Zn was elevated above USEPA acute aquatic life criteria (120 µg/L) in both the bedded (451 ± 51 µg/L) and redeposited treatments (493 ± 72 µg/L) despite daily water changes. Ni, Fe, Cu, Cd, Cr, and Pb were below detection limits in all samples throughout the experiments (SI.9a). For PNS exposures, 10-d overlying-water concentrations were typically below detection limits (SI.9b). Cu was near the MDL (2.67 µg/L), but did not exceed acute aquatic life criteria (4.8 µg/L), and Zn was typically less than 10 µg/L but highly variable (SI.9b). Mn was elevated in the PNS controls (maximum 200 µg/L), but < ~25 µg/L in chambers containing sediment.

3.3b. Toxicity

Lake DePue exposures

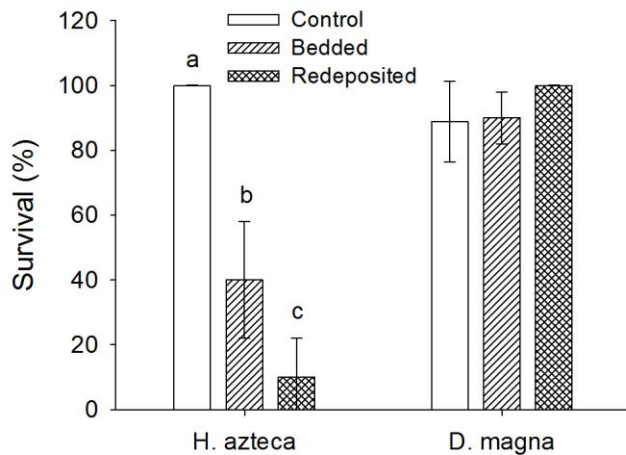


Figure 9. Mean (± 1 SD, n = 4 for bedded and redeposited, 8 for control) *Hyalella azteca* (7 d test) and *D. magna* (4 d test) survival after bedded and redeposited sediment toxicity exposures. Survival of *H. azteca* differed among treatments (ANOVA $p < 0.001$) and different letters above bars indicate treatments that were significantly different (Tukey HSD). Survival of *D. magna* did not differ among treatments (Kruskal-Wallis $p = 0.11$)

Daphnia magna exhibited no differences in toxicity between control, bedded and redeposited sediment exposures (Kruskal-Wallis $p = 0.11$, $H = 4.48$, Fig. 9). *Daphnia magna* are

primarily planktonic organisms, but they were observed feeding at the sediment surface in these experiments, which suggests some exposure to sediment. Survival of *H. azteca* was lower than controls in both bedded ($40 \pm 18\%$) and redeposited ($10 \pm 12\%$) exposures (Tukey HSD, $p < 0.001$, Fig. 9). Amphipods exposed to redeposited sediments had significantly decreased survival compared to bedded exposures (Tukey HSD, $p < 0.01$).

PNS exposures

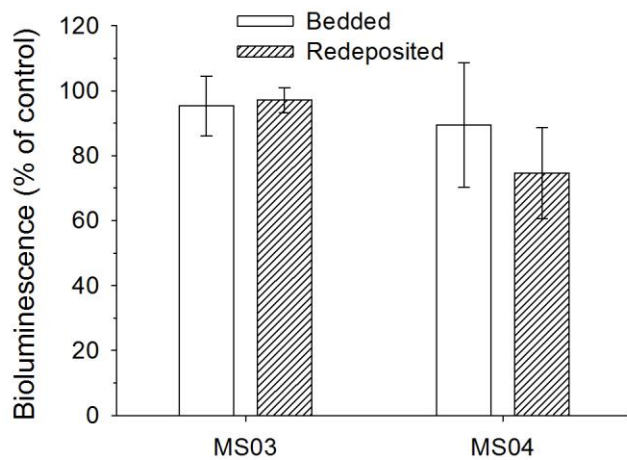


Figure 10. Mean ($n = 4$, ± 1 SD) response of *Pyrocystis lunula* exposed to filtered sediment elutriates (1:4 sediment to water) of MS03 and MS04, expressed as % of control. Percentages > 100 were rounded down to 100% to allow for arcsine square root transformations.

Pyrocystis lunula exposed to sediment elutriates of bedded and redeposited sediment showed minimal light inhibition (Fig. 10). In MS03, there were no differences between bedded and redeposited exposures (Kruskal-Wallis $p = 0.74$, $H = 0.11$), with both treatments near 100% of controls. In MS04, the redeposited response was lower ($75 \pm 14\%$), but not significantly different from the bedded treatment response ($89 \pm 19\%$, $p = 0.14$, $t = 1.70$).

Minimal mortality of *N. arenaceodentata* was observed following 10-d bedded and redeposited sediment toxicity tests for both PNS sediments (Fig. 11). For MS03 sediments, there was no difference in relative growth rates due to sediment exposure ($F_{1,12} = 0.51$, $p = 0.49$) and

no differences between bedded and redeposited exposures ($F_{1,12} = 0.77, p = 0.40$, Fig. 12). For MS04 sediments, exposure to bedded and redeposited sediments reduced relative growth rates compared to controls ($F_{1,12} = 7.71, p = 0.017$), but there was no difference between bedded and redeposited experiments ($F_{1,12} = 0.95, p = 0.35$) (Fig. 12).

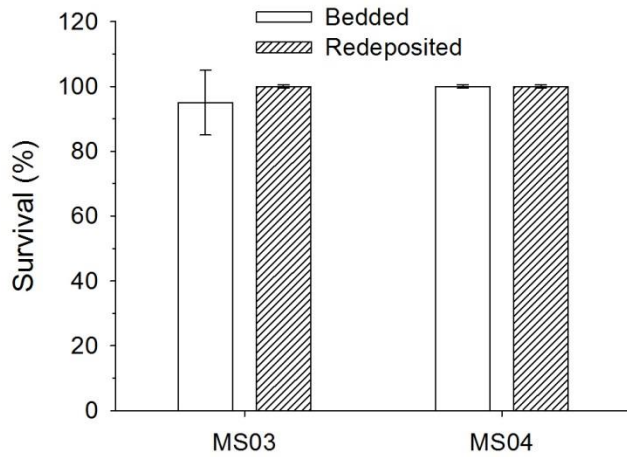


Figure 11. Mean (± 1 SD, $n = 4$) *Neanthes arenaceodentata* survival following 10-d bedded and redeposited sediment toxicity tests for MS03 and MS04 sediments. Controls for bedded and redeposited exposures for both sediment types were 100% (data not shown).

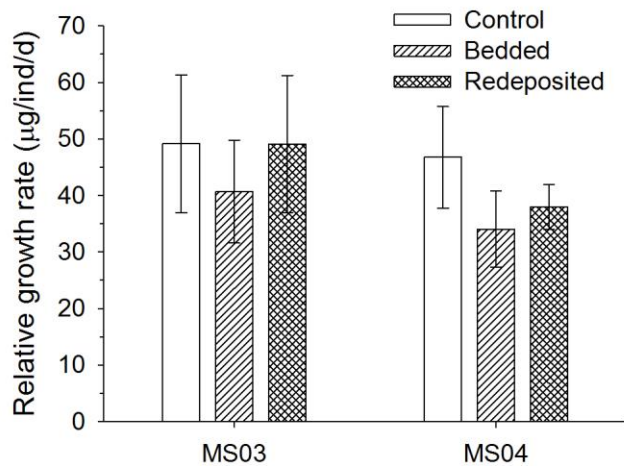


Figure 12. Mean (± 1 SD, $n = 4$ and 8) relative growth rates between 10-d control, bedded and redeposited sediment toxicity exposures for MS03 and MS04. Bedded and redeposited controls for each sediment type are depicted as a single bar but were analyzed separately.

Exposure of *N. arenaceodentata* to MS04 sediment resulted in elevated Cu body burdens compared to controls ($F_{1,12} = 13.20, p < 0.01$), while no differences were observed for Zn, Cr, Ni and Pb (all $p > 0.05$, Table 6). No differences between sediment exposure and controls were observed for MS03 (all $p > 0.05$), with the exception of Ni, which was elevated in controls ($F_{1,12} = 84.57, p < 0.001$) and Cu, which was marginally greater in sediment exposures compared to controls ($F_{1,12} = 4.60, p = 0.053$) (Table 6). No differences in *N. arenaceodentata* body burdens were found in both sediment types between bedded and redeposited sediment exposures (all $p > 0.05$), with the exception of MS03 Pb (Tukey HSD $p < 0.001$), which was slightly elevated in redeposited compared to bedded exposures, but overall concentrations were low (< 5 mg/kg, Table 6). Correlations between *N. arenaceodentata* body burdens and relative growth rates showed no relationships with most metals (all $R^2 < 0.2$).

Table 6. Mean (± 1 SD, $n = 4$) *Neanthes arenaceodentata* body burden following 10-d bedded and redeposited sediment toxicity tests

Sediment	Exposure	Body Burden (mg/kg dw)				
		Zn	Cu	Ni	Pb	Cr
MS03	Bedded control	159 (10)	44 (16)	14 (2)	2.6 (0.3)	4.0 (1.0)
	Bedded	146 (18)	82 (54)	5 (2)	1.6 (0.2)	3.3 (0.7)
	Redeposited control	196 (25)	36 (1)	12 (2)	2.2 (0.3)	4.5 (1.3)
	Redeposited	189 (29)	62 (20)	6 (1)	2.8 (0.2)	3.8 (0.9)
MS04	Bedded control	161 (20)	28 (1)	11 (1)	1.9 (0.1)	3.2 (0.9)
	Bedded	128 (42)	54 (30)	8 (3)	1.5 (0.5)	2.3 (0.8)
	Redeposited control	176 (29)	31 (8)	12 (3)	1.6 (0.5)	2.5 (0.5)
	Redeposited	157 (21)	51 (9)	10 (2)	1.6 (0.5)	3.5 (0.9)

4. Discussion

Environmentally-relevant SPM concentrations used in our experiments resulted in modest pH and O₂ declines in water. Other resuspension studies report varying physicochemical changes during short-term simulated resuspensions, with pH changes similar to what was observed here (typically < ± 1.0 pH unit) and wildly varying O₂ declines (no change to anoxia) (Zhuang et al. 1994, Cantwell et al. 2002, Maddock et al. 2007, Hong et al. 2011a, Hwang et al. 2011). pH declines during the first few hours of resuspension are mediated by the oxidation of Fe²⁺, but buffered by carbonate and other inorganic species, with large declines observed over longer term resuspensions (5 h to days) due to oxidation of S⁰_(s) (product of sulfide oxidation) to SO₄⁻², which has implications for metal mobilization (Calmano et al. 1993 Callie et al. 2003, Burton et al. 2006, Atkinson et al. 2007, Maddock et al. 2007, Hong et al. 2011a). If resuspension leads to hypoxic conditions, Fe²⁺ oxidative precipitation and metal scavenging by iron oxides will be slower creating a potential for increased bioavailability of metals (Caetano et al. 2002; Atkinson et al. 2007, Ye et al. 2013). Scavenging by iron oxides could explain the small fraction of metals mobilized in all resuspensions.

In tests with Lake DePue sediment, we observed Zn and Mn dissolution but it might not have been bioavailable because of colloidal complexation. Concentrations of filtered Zn and Mn were elevated during resuspensions of Lake DePue sediment, while other metals (Cu, Ni, Cd, Cr, Pb and Fe) were not affected by resuspension. The differences between bedded and resuspension exposures show that Zn and Mn mobilization is greater than what would be expected by diffusion alone. Cu, Cd and Pb were likely not observed in filtered water because of either their relatively greater affinities for organic matter (Coquery and Welbourn 1995 and Lin and Chen 1998) or slower oxidation kinetics of their sulfide minerals over the course of short-term

resuspensions (Slotton and Reuter 1995, Simpson et al. 1998, Caetano et al. 2002). These metals, along with Cr were not detected in porewaters, indicating their probable association with solid phases, therefore lack of bioavailability (Chapman et al. 1998). The released Zn may have been from non-sulfide origin, given that metal sulfides can take more than 8 h to oxidize (Simpson et al. 1998) and ~50% of Zn in the sediment was associated with carbonates and other inorganic species (Gough et al. 2008). Other processes must be occurring due to resuspension, such as ion exchange reactions between SPM and overlying water and increased porewater flushing, which can overload the binding capacity of newly formed iron oxides (Chapman et al. 1998, Kalnejais et al. 2010). In a separate study with Lake DePue sediment, no ZnS was found in SPM, but this experiment was conducted over 14 d experiments with *in situ* sediment cores, allowing for the preservation of *in situ* redox gradients so the resuspended surficial sediment was already partially oxidized, therefore a lack of ZnS is not indicative of complete oxidation during my short-term resuspensions (C. Cadoux, Northwestern University, Evanston, IL, USA, personal communication). The observed Zn in overlying water during my resuspensions was likely not free metal given the elevated turbidity (50 ± 19 NTU), but rather associated with colloids and dissolved ligands in the overlying water as found in other studies for both Zn (Wen et al. 1999) and other metals (Ag, Cu and Pb; Kalnejais et al. 2010). Given that Zn and Mn had similar release kinetics, it is possible that the filtered Zn was associated with either dissolved or colloidal Mn, as previously observed for Cu and Mn (Stauber et al. 2000). EF for Zn and Mn were less than 1, indicating some degree of release from SPM. This could be due to Zn having a lower affinity for sediment solid phases compared to other metals (Cu, Pb, Hg; Caille et al. 2003) and Mn being slower to form oxides (Stumm and Morgan 1996). Other studies have observed SPM enrichment and depletion, and can be explained by a number of physical, chemical and

diagenetic processes (Cantwell et al. 2002, Kalnejais et al. 2007). EF for other metals were greater than bedded sediment in my study, indicating the possibility of SPM acting as a reservoir for remobilized metal due to adsorption onto iron oxides, clay materials or POC (Chapman et al. 1998, Caetano et al. 2002, Cantwell et al. 2002). The majority of enrichment is likely due to the preferential resuspension of fine-grained sediments, which have a greater surface area for metal sorption (Kalnejais et al. 2007). Enriched SPM metals have the potential to be many times greater than bulk sediment, indicating that resuspension events could significantly increase exposure and effects to resident biota.

Despite exposure to elevated filtered Zn and metals associated with SPM, a lack of remobilized, bioavailable metals resulted in few observed adverse effects to *D. magna* and *H. azteca*. The non-selective, filter-feeding *D. magna* is sensitive to metals and physical stress from suspended particles (Weltens et al. 2000, Robinson et al. 2010, Cloran et al. 2010), but combining these two stressors (physical and chemical) did not result in increased mortality during resuspension events or latent effects following exposures. Particulate Cd and Zn has been shown to become more bioavailable in the organisms' gut track, causing toxicity that could not be explained by the dissolved fraction alone, but *D. magna* are also believed to reduce ingestion rates upon exposure to contaminated sediments, which could manifest into chronic, population and community level effects (Allen et al. 1995, Weltens et al. 2000). *Hyalella azteca* exposed to resuspended sediment did not reduce feeding rates during post-exposure holds, as seen in Wilding and Maltby (2006), who showed that exposure to aqueous Zn resulted in irreversible or slowly recovering feeding inhibition, which is an ecologically relevant endpoint, affecting the organisms' energy budget and therefore survival, reproduction and population growth. A lack of Zn toxicity could be explained because colloiddally bound or organically complexed Zn is less

bioavailable compared to Zn^{2+} (Carvalho et al. 1999), but short exposure durations and Zn concentrations below EPA aquatic life criteria (120 $\mu\text{g/L}$) could also be responsible for minimal effects observed.

While metal solubilization was observed with Lake DePue, it was very limited with both PNS sediments. Cu was the primary metal of concern in MS03 and MS04, but was not above method detection limits throughout the tests. The observed low solubility of Cu is likely due to recalcitrance of CuS (Chapman et al. 1998, Caetano et al. 2002) and a strong affinity for organic matter and iron oxides (Lors et al. 2005). Pyrite is the dominant form of sulfur present in marine systems, is more resistant to oxidization during short-term resuspensions and trace metals can coprecipitate with pyrite to varying degrees (moderate coprecipitation: Cu, Mn, Ni, low: Cr, Pb, Zn Cd; Huerta-Diaz and Morse 1992, Caetano et al. 2002, Cantwell et al. 2002, Rickard and Morse 2005). Our [SEM]/ [total metals] percentages go along with these coprecipitation trends, with the exception of Pb. In PNS sediments, AVS is in excess of SEM, indicating these sediments have free binding sites for metal scavenging during resuspension, preventing mobilization (Cantwell et al. 2008). In PNS experiments, the release of Fe^{2+} and formation of iron oxides, with subsequent aggregation into flocs, could have been responsible for scavenging dissolved metals and SPM enrichment observed (generally > Lake DePue), returning the metals to the benthic environment more rapidly than individual particles after termination of resuspension (Jones-Lee and Jones 2005, Plach et al. 2011). While diffusion presumably dominated the slight Zn mobilization during PNS bedded exposures, this diffusion was countered by readsorption to particles/floc in resuspension exposures (Chapman et al. 1998). Different complexing behavior of seawater and a greater pH can also explain the quicker floc

aggregation/settling and lack of bioavailable metals in the overlying water (Bruland 1989, Wen et al. 1999, Atkinson et al. 2007, Burton and Johnston 2010 and Hong et al. 2011b).

Pyrocystis lunula bioluminescence (Qwiklite) was not inhibited from exposure to overlying water from bedded and resuspended exposures. These organisms were exposed to discrete samples representing one time period (4h), so it is possible that we missed the quick release and peak of dissolved metals that is thought to occur within an hour of resuspension (Caetano et al. 2002). This potential quick metal release would have only represented short exposure durations for organisms, therefore not likely important toxicologically. *Pyrocystis* spp. have been found to be more sensitive to metals than other rapid toxicity bioassays (e.g., Microtox; Rosen et al. 2008b), with moderately low 24-h EC₅₀'s for Cu ($185 \pm 101 \mu\text{g/L}$) and Zn ($345 \pm 16 \mu\text{g/L}$), so a lack of responses in the filtered samples was not surprising. Organisms exposed to unfiltered water samples generally showed less light output inhibition compared to filtered samples, indicating that the metals associated with SPM were also not bioavailable to *P. lunula*. No acute mortality or latent growth effects were observed with *N. arenaceodentata*. Growth is considered a more sensitive endpoint (Lee and Lee 2005), but was only slightly reduced with exposure to contaminated SPM. *Neanthes arenaceodentata* body burdens were not elevated due to SPM exposure, despite observations of sediment excretion during the depuration period, indicating the organisms were ingesting SPM, but the metals were not liberated and absorbed in the gut. Uptake from the dissolved phase is most important for Zn and Cd bioaccumulation (Lee and Lee 2005), but the particulate phase was more important for Ni bioaccumulation, while other studies have shown synergistic effects between particulate and dissolved metals, with bioaccumulation being metal and species specific (Fichet et al. 1998, Weltens et al. 2000, Hill et al. 2009, Cloran et al. 2010). Despite high levels of total metals

resuspended into the water column on SPM, these metals must not have been present in bioavailable fractions explaining a lack of effects.

A lack of quantifiable AVS, SEM and \sum SEM-AVS/foc changes were observed due to resuspension and redeposition of Lake DePue sediment (despite visual color changes) likely due to: 1) low sediment permeability, 2) small layer of surficial sediment resuspended and 3) coarse core sectioning (0.5 cm depth). Bioavailability predictions were correct for Lake DePue sediment, as model values for bedded and redeposited sediments were $> 3000 \mu\text{mol/g OC}$ and toxicity to *H. azteca* was observed, but *D. magna* were not negatively affected by sediment or elevated water column Zn. *Hyalella azteca* exposed to redeposited sediments exhibited greater toxicity than sediment without disturbances, suggesting the proposed changes in metal speciation of SPM, from sulfides to oxides, appear to slightly increase the toxicity of solid phase bound metals. These results contrast Bonnet et al. (2000), who found no differences in toxicity between perturbed and non-perturbed sediments. In a separate study with Lake DePue sediment, it was found that immediately following resuspension the top 1 mm of sediment was void of ZnS species, but 2 d following such events, the sulfide was present again, indicating that resuspension can change metal speciation (C. Cadoux, Northwestern University, Evanston, IL, USA, personal communication), but the changes may be temporary depending on the redox conditions of the sediment.

Minimal AVS, SEM and \sum SEM-AVS/foc changes were observed for PNS sediments, although bioavailable metals (SEM) were slightly greater in both redeposited compared to bedded exposures, possibly due to the settling of metal-enriched flocs (Plach et al. 2011). The increase in MS04 bioavailability prediction after redeposition corresponds to slightly reduced *P. lunula* bioluminescence of sediment elutriates. Compared to Miller et al. (2000), our sediment

elutriates had higher suspended solids concentrations (250 g sediment/L), but large effects were still not observed, with a more sensitive organism (Lapota et al. 2007). Exposure to MS03 and MS04 bedded and redeposited sediments did not cause *N. arenaceodentata* mortality but MS04 sediments (bedded + redeposited) experienced significantly lower growth rates compared to controls. An absence of growth depression in organisms exposed to MS03 sediment indicates the importance of AVS in mitigating metal toxicity. MS03 had greater metal (and PAH) concentrations compared to MS04, but also more AVS, therefore smaller $\sum\text{SEM-AVS}/\text{foc}$ model predictions, and a lack of growth effects were observed. Similar to what Pesch et al. (1995) observed for Cd and Ni, *N. arenaceodentata* in our exposures bioaccumulated significant amounts of Cu despite AVS being in excess of SEM, with no observed mortality. This could be explained by a number of scenarios, including 1) oxidation of AVS and release of Cu in the burrow microenvironment and 2) Cu could have been released from digested sediment particles (Pesch et al. 1995). In MS03, Pb was also elevated in redeposited compared to bedded exposures, but concentrations were < 3 mg/kg dw, indicating Pb may be slightly more bioavailable in redeposited sediments. *Neanthes arenaceodentata* body burdens could have been underestimated in this study, by as much as 40-60%, due to selective feeding on uncontaminated, supplemental food (Lee et al. 2001). Some of the metal body burdens in these exposures could also be attributed to the rabbit chow slurry, which contained various forms of essential trace elements: Cu, Zn, Co, Fe, and Mn. Elevated concentrations of Ni in control organisms can be attributed to the substrate (Ottawa sand) and contamination in the food source. In the overlying water exposures, the beakers contained no sand substrate and had low Ni body burdens (< 4 mg/kg dw). A lack of endpoint differences between bedded and redeposited *N. arenaceodentata*, but differences for the epibenthic *H. azteca* implicate the importance of life histories and feeding

strategies when trying to assess risks to ecosystems. *Neanthes arenaceodentata* burrows were observed 2-3 cm below the sediment water interface, to depths at which sediment resuspension did not chemically or physically alter, while *H. azteca* forage at the sediment surface, where most of the chemical oxidation occurs. Decreased growth rates and elevated metal body burdens have the potential to manifest into population and community-level effects (Roberts 2012).

5. Conclusions

This research investigated the changes in metal bioavailability due to resuspension of contaminated sediments under more realistic laboratory exposures. A host of physical and chemical factors were responsible for controlling potential metal mobilization and subsequent impacts to biota. We observed site and metal specific mobilization, with heavily-contaminated (SEM > AVS), freshwater sediments representing the greatest potential for release of bioavailable metals. Metal mobilization from two marine sediments seems to be controlled by sulfide chemistry and the high concentrations of inorganic ligands in seawater. The exposure of organisms to metal-contaminated SPM did not result in reduced survival, growth, feeding or increased body burdens, indicating that single, short-term resuspension events may not be ecologically relevant. These results support the conclusions of Eggleton's (2004) literature review that says metal mobilization is not significant enough to be acutely toxic, although the author notes chronic effects have been observed. While single, short-term events investigated do not present risks to resident biota, it is possible with increased frequency, duration and intensity there is potential for negative impacts. At sites similar to PNS, metals are present, but are not in a bioavailable form, even after resuspension into oxic waters. PNS sediments completely settle within a day, so prolonged exposure to oxic waters will typically not occur, therefore limiting the amount of recalcitrant sulfide oxidation with subsequent acidification and metal release. In high traffic waterways, the

frequency, duration and intensities of these events could result in greater mobilization of metals. The organisms present in these systems are exposed to multiple events, potentially compounding the toxic responses. In most cases, we were not able to differentiate the toxicity between bedded and redeposited sediments, implying that there were not large shifts in metal bioavailability, or if these shifts did occur, redox gradients were quickly re-established. In cases where toxicological differences were observed, redeposited sediments seemed to be related to greater toxicological impacts. The conservative nature of our system (small size and no water replacement) should have detected any large physicochemical changes, but a lack of such changes show that realistic concentrations of SPM will not pose great risks during 4-h resuspensions.

This research highlights the importance of fully characterizing initial sediment and water geochemistry and understanding disturbance regimes, which are paramount in making more accurate predictions of potential metal mobilization and assessing ecological risks due to resuspension events. Resuspension of historically contaminated sediments are important because they can negate improvements made by restoration, remediation and management activities and also act as a significant source of bioavailable metals in these systems (Kalnejais et al. 2010 and Roberts 2012). Further research is needed to examine ecologically relevant endpoints, including; community level responses, predator-prey interactions, recruitment and reproduction under resuspensions of varying frequencies, durations and intensities.

Supporting Information

Table SI.1. QA/QC for analytical procedures performed, with number of replicates in parenthesis.

Parameter	Procedural Blank	Procedural Spike Recovery (%)
AVS	ND ^a (2)	89 ± 3 (3)
SEM _{Fe} ^b	71 ± 19 µg/L (2)	NA ^c
SEM _{Zn}	13 ± 2 µg/L (2)	NA
TOC ^d	0.05 ± 0.01% (9)	97 ± 4 (6)
Sediment Zn ^e	5 ± 8 µg/L (10)	93 ± 5 (9)
Sediment Cu	ND (10)	90 ± 4 (9)
Sediment Cr	ND (9)	82 ± 6 (8)
Sediment Pb	ND (9)	99 ± 4 (5)
Sediment Ni	ND (10)	101 ± 14 (9)
Sediment Fe	9 ± 7 µg/L (10)	92 ± 3 (9)
Sediment Mn	ND (10)	94 ± 2 (5)
Organism Cu ^f	3 ± 3 µg/L (13)	91 ± 10 (13)
Organism Cd	ND (5)	109 ± 5 (6)
Organism Ni	1 ± 1 µg/L (13)	92 ± 20 (12)
Organism Zn	194 ± 195 µg/L (7)	117 ± 41 (7)
Organism Cr	51 ± 10 µg/L (8)	NA

^aND: below detection limit

^bOther SEM metals (Mn, Cu, Ni, Pb and Cd) were below detection limits.

^cNA: parameter not available

^dTOC recoveries includes Caffeine, E-horizon and NIST 2702

^eSediment Me represents both sediment and SPM filter digestions, averaged and combined.

These include NIST 2702 and NIST 2781. Cd and Pb did not work for LD sediment and SPM digestions.

^fOrganism recoveries from both *H. azteca* and *N. arenaceodentata* are with NIST 1570a. Pb certified values were too low to detect and no listed Cr value. Laboratory dilution blanks for organism digestions are all < 1 µg/L. *H. azteca* Zn body burden are not reported in this study due to pre-analysis dilution errors.

Table SI2 Initial mean (n = 3 to 12 ± 1 SD) porewater physicochemistry and metal concentrations (ug/L)

	pH ^a	DO ^a (mg/L)	DOC ^a (mg/L)	Dissolved metals (ug/L) ^b						
				Zn	Cu	Ni	Cd	Cr	Fe	Mn
Lake DePue	7.32 (0.03)	1.01 (0.13)	15 (1)	146 (37)	ND ^c	ND	ND	ND	1006 (771)	913 (178)
MS03	7.05 (0.02)	1.04 (1.00)	103(6)	28 (10)	ND	22 (19)	NA ^d	ND	14,063 (3,809)	351 (93)
MS04	7.08 (0.04)	0.81 (0.52)	98(4)	40 (18)	ND	18 (18)	NA	ND	13,973 (5,763)	103 (34)

^a n = 2 for Lake DePue and 3 for PNS sediments

^b n = 12 for Lake DePue and 8 for PNS sediments. Data represents means between experiments (resuspension and bedded) analyzed on separate ICP-OES runs, resulting in large standard deviations for certain metals.

^c ND = below MDL

^d NA = not measured

Table SI3. Particle size distribution (%) of sediment types.

	Particle size distribution (%)					
	< 45 μm	45 - 106 μm	106 - 150 μm	150 - 250 μm	250 - 1000 μm	>1000 μm
Lake DePue	81	6	13 ^a	NA	NA	NA
MS03	23	17	12	6	16	26
MS04	39	37	11	12 ^b	NA	NA

^a > 106 μm largest fraction analyzed for Lake DePue

^b > 150 μm largest fraction analyzed for MS04

Table SI.4 Mean ($n=8 \pm 1$ SD) physicochemistry during resuspension and redeposited experiments.

	Hour	pH	Temp (°C)	DO (mg/L)	Turb (ntu)	Specific Conductance ($\mu\text{S}/\text{cm}$)	TSS (mg/L)
Lake DePue	Initial	8.16 (0.04)	22.7 (0.1)	8.12 (0.11)	0.1 (NA)	361 (7)	
	0	8.12 (0.15)	22.5 (0.1)	8.16 (0.10)	0.1 (0.07)	351 (1)	
	1	7.85 (0.09)	23.0 (0.1)	7.32 (0.18)	383 (150)	366 (3)	439 (159)
	2	7.83 (0.07)	23.6 (0.1)	6.95 (0.32)	529 (275)	372 (3)	599 (223)
	3	7.81 (0.06)	24.0 (0.1)	6.82 (0.33)	598 (322)	375 (3)	626 (222)
	4	7.80 (0.06)	24.2 (0.2)	6.80 (0.30)	658 (330)	378 (4)	643 (284)
	24				50 (19)		
MS03	Initial	7.92 (0.03)	23.0 (0.1)	6.75 (0.07)	0.6 (0)	NA	
	0	7.87 (0.07)	23.1 (0.3)	6.80 (0.04)	0.6 (0)		
	1	7.74 (0.05)	23.8 (0.4)	5.98 (0.21)	171 (68)		387 (171)
	2	7.71 (0.03)	24.4 (0.3)	5.59 (0.34)	240 (111)		
	3	7.67 (0.03)	24.7 (0.2)	5.37 (0.35)	259 (117)		
	4	7.63 (0.04)	24.8 (0.2)	5.29 (0.29)	265 (126)		576 (281)
	24						
MS04	Initial	8.02 (0.10)	22.7 (0.2)	6.85 (NA)	0.5 (0)	NA	
	0	7.97 (0.07)	23.0 (0.2)	6.84 (0.04)	0.5 (0)		
	1	7.89 (0.09)	23.3 (0.2)	6.33 (0.16)	150 (70)		337 (166)
	2	7.84 (0.09)	23.8 (0.1)	6.07 (0.25)	246 (143)		
	3	7.81 (0.09)	24.0 (0.1)	5.90 (0.34)	266 (197)		
	4	7.77 (0.09)	24.0 (0.2)	5.85 (0.28)	294 (235)		686 (438)
	24	7.66 (0.07)	23.3 (0.1)				

Table SI.5 Mean ($n = 4 \pm 1$ SD) physicochemistry during bedded experiments.

	Hour	pH	Temp (°C)	DO (mg/L)	Turb (ntu)	Specific Conductance (μ S/cm)
Lake DePue	Initial	7.98	22.9	8.40	0.1	347
	0	8.06 (0.02)	22.7 (0.2)	8.38 (0.08)	0.1 (0)	349 (1.4)
	1	8.02 (0.02)	23.2 (0.2)	8.37 (0.05)	2 (1)	353 (1)
	2	8.04 (0.01)	23.8 (0.2)	8.38 (0.02)	2 (1)	354 (1)
	3	8.02 (0.01)	24.1 (0.1)	8.34 (0.05)	2 (1)	354 (1)
	4	8.03 (0.01)	24.3 (0.2)	8.35 (0.04)	2 (1)	355 (1)
	24					
MS03	Initial	8.17	22.5	7.08	1.0	NA
	0	8.11 (0.02)	23.4 (0.1)	7.13 (0.03)	1.0 (0)	
	1	8.10 (0.01)	23.9 (0.1)	7.05 (0.02)	1.0 (0.2)	
	2	8.09 (0.02)	24.5 (0.1)	6.98 (0.03)	0.9 (0.1)	
	3	8.08 (0.01)	24.8 (0.1)	6.94 (0.02)	0.8 (0.1)	
	4	8.07 (0.01)	24.8 (0.1)	6.87 (0.04)	0.7 (0.1)	
	24	7.96 (0.00)	23.4 (0.1)	6.50 (0.07)		
MS04	Initial	8.03	22.7	7.19	0.8	NA
	0	8.00 (0.02)	23.3 (0.1)	7.18 (0.03)	0.8 (0.0)	
	1	8.00 (0.01)	23.6 (0.1)	6.95 (0.04)	0.8 (0.1)	
	2	7.99 (0.01)	24.2 (0.1)	6.89 (0.03)	0.7 (0.0)	
	3	7.99 (0.01)	24.5 (0.2)	6.86 (0.04)	0.6 (0.0)	
	4	7.98 (0.01)	24.6 (0.1)	6.84 (0.05)	0.6 (0.0)	
	24	7.96 (0.01)	23.3 (0.1)	6.60 (0.10)		

Table SI.6a Mean ($n = 4 \pm 1$ SD) dissolved metals ($\mu\text{g/L}$) during Lake DePue resuspension, redeposited and bedded experiments.

	Hour	Zn	Cu	Ni	Cd	Cr	Pb	Fe	Mn
	$\sim\text{PQL}^a$ ($\mu\text{g/L}$)	4	8	10	2	4	20	2	2
Lake DePue resuspension	Initial	ND ^b	ND	ND	ND	ND	ND	ND	2
	0	17 (14)	ND	ND	ND	ND	ND	ND	2 (0)
	1	63 (11)	ND	ND	ND	ND	ND	6 (3)	72 (15)
	2	81 (8)	ND	ND	ND	ND	ND	6 (1)	100 (16)
	3	83 (11)	ND	ND	ND	ND	ND	8 (4)	106 (17)
	4	76 (12)	ND	ND	ND	ND	ND	6 (1)	101 (17)
	24	144 (3)	ND	ND	ND	ND	ND	ND	126 (7)
Lake DePue bedded	Initial	ND	ND	ND	ND	ND	ND	ND	ND
	0	13 (4)	ND	ND	ND	ND	ND	3.5 (1)	ND
	1	18 (4)	ND	ND	ND	ND	ND	2 (1)	4 (0)
	4	20 (3)	ND	ND	ND	ND	ND	ND	5 (0)
	24	59 (8)	ND	ND	ND	ND	ND	ND	86 (9)

Note: resuspension and redeposited concentrations combined

^a PQL = practical quantification limit based on Perkin Elmer detection limits * 20

^b ND = less than PQL

Table SI.6b Mean ($n = 4 \pm 1$ SD) dissolved metals ($\mu\text{g/L}$) during MS03 resuspension and bedded experiments.

	Hour	Zn	Cu	Ni	Cr	Pb	Fe	Mn
	MDL ^a ($\mu\text{g/L}$)	3.52	2.67	2.98	2.71	4.06	10.1	2.7
MS03 resuspension	Initial	ND ^b	ND	ND	ND	ND	ND	7
	0	ND	4 (1)	ND	ND	ND	ND	7 (0)
	1	6 (NA)	ND	4 (1)	ND	ND	ND	12 (1)
	4	ND	ND	6 (1)	ND	ND	ND	16 (2)
	24	5 (0)	3 (0)	7 (1)	ND	ND	ND	20 (2)
MS03 bedded	Initial	ND	ND	ND	ND	ND	ND	7
	0	13 (5)	3 (NA)	ND	ND	ND	ND	8 (0)
	4	32 (7)	ND	ND	ND	ND	ND	8 (0)
	24	36 (7)	ND	ND	ND	ND	ND	9 (0)

^a MDL = method detection limit based on Calsciences analytical capabilities

^b ND = less than MDL

Table SI.6c Mean ($n = 4 \pm 1$ SD) dissolved metals ($\mu\text{g/L}$) during MS04 resuspension and bedded experiments.

	Hour	Zn	Cu	Ni	Cr	Pb	Fe	Mn
	MDL ^a ($\mu\text{g/L}$)	3.52	2.67	2.98	2.71	4.06	10.1	2.7
MS04 resuspension	Initial	ND ^b	4	ND	ND	ND	ND	7
	0	20 (16)	ND	ND	ND	ND	ND	7 (0)
	1	7 (NA)	ND	ND	ND	ND	ND	9 (1)
	4	ND	ND	3 (NA)	ND	ND	ND	10 (1)
	24	31 (43)	ND	3 (0)	ND	ND	ND	10 (1)
MS04 bedded	Initial	ND	ND	ND	ND	ND	ND	7
	0	5 (1)	ND	ND	ND	ND	ND	8 (0)
	4	17 (5)	ND	ND	ND	ND	ND	8 (0)
	24	22 (12)	ND	ND	ND	ND	ND	8 (0)

^a MDL = method detection limit based on Calsciences analytical capabilities

^b ND = less than MDL

Table SI.7. Table of mean (n = 4 to 8 ± 1 SD) enrichment factors though time

Sediment	Hour	Enrichment Factor							
		Zn	Cu	Fe	Mn	Ni	Pb	Cr	OC ^a
Lake DePue	1	0.72 (0.02)	1.07 (0.05)	1.11 (0.05)	0.60 (0.03)	1.64 (0.39)	NA ^b	1.68 (0.08)	NA
	2	0.74 (0.03)	1.09 (0.03)	1.15 (0.02)	0.61 (0.03)	1.25 (0.13)	NA	NA	NA
	3	0.76 (0.02)	1.09 (0.04)	1.16 (0.04)	0.63 (0.02)	1.37 (0.16)	NA	NA	NA
	4	0.75 (0.02)	1.08 (0.04)	1.13 (0.05)	0.65 (0.03)	1.54 (0.35)	NA	1.51 (0.10)	1.31 (0.02)
MS03	1	1.35 (0.06)	1.58 (0.07)	1.06 (0.03)	1.12 (0.05)	1.18 (0.11)	1.65 (0.13)	2.09 (0.05)	NA
	4	1.45 (0.06)	1.67 (0.09)	1.14 (0.07)	1.18 (0.08)	1.14 (0.09)	1.72 (0.02)	2.24 (0.14)	2.62 (0.78)
MS04	1	1.65 (0.20)	0.89 (0.03)	1.45 (0.20)	1.21 (0.22)	1.36 (0.16)	2.02 (0.23)	1.91 (0.14)	NA
	4	1.93 (0.16)	1.06 (0.10)	1.74 (0.13)	1.41 (0.08)	1.38 (0.11)	1.98 (0.20)	2.14 (0.15)	2.38 (0.42)

^a Organic Carbon

^b NA = not available

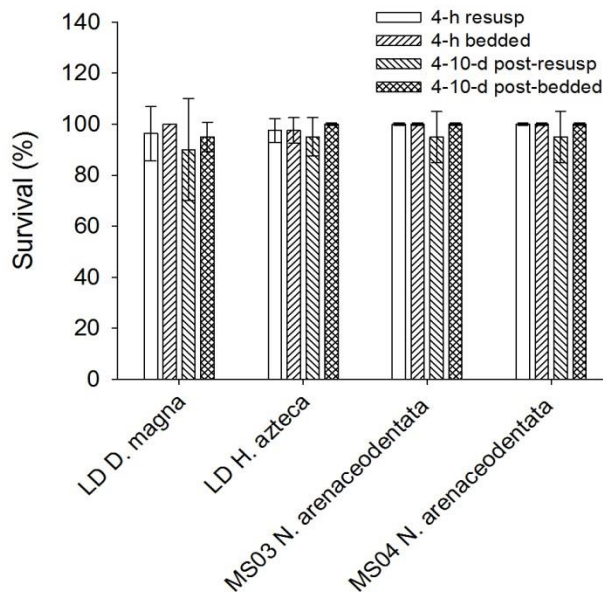


Figure SI.1. Mean (± SD, n = 4 - 8) organism survival following resuspension and bedded experiments, and subsequent post-exposure hold. Note: *Hyalella azteca* held for 7 d, *Daphnia magna* 4 d and *Neanthes arenaceodentata* 10 d.

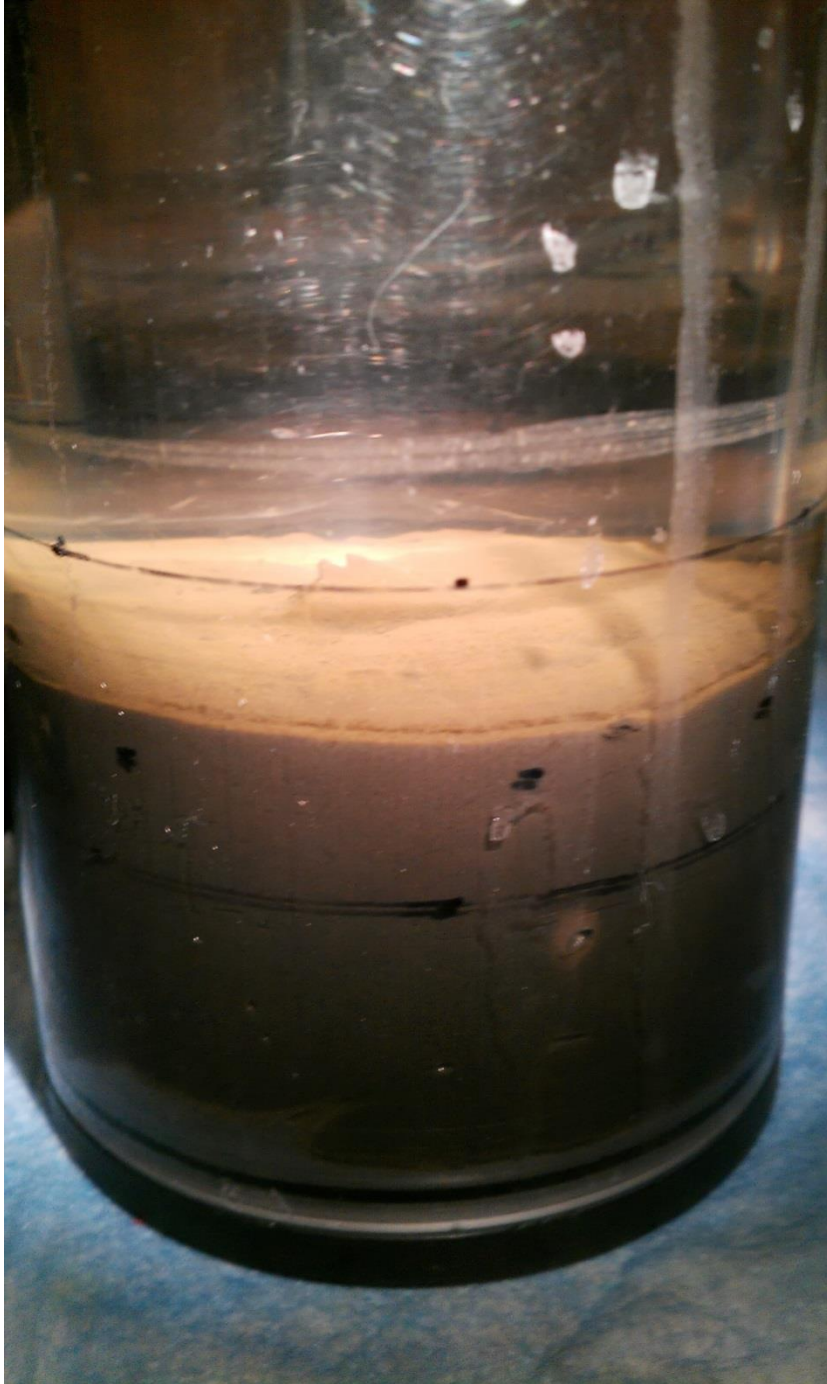


Figure SI.2. MS03 surficial sediment oxidation and color change following resuspension and settling.

Table SI.9a. Mean (± 1 SD, $n = 4$) dissolved metal concentrations ($\mu\text{g/L}$) of Lake DePue overlying water after 7 d redeposited and bedded sediment toxicity tests.

		Dissolved metals ($\mu\text{g/L}$)							
		Cr	Cu	Pb	Ni	Fe	Mn	Zn	Cd
PQL ^a		4	8	20	10	2	2	4	2
Lake DePue redeposited	Control	ND ^b	ND	ND	ND	ND	ND	ND	ND
	Treatment	ND	ND	ND	ND	ND	108 (12)	493 (72)	ND
Lake DePue bedded	Control	ND	ND	ND	ND	ND	ND	ND	ND
	Treatment	ND	ND	ND	ND	ND	144 (11)	451 (51)	ND

^a PQL = practical quantification limit based on Perkin Elmer detection limits * 20

^b ND: below PQL

Table SI.9b. Mean ($n = 4$, ± 1 SD) dissolved metal concentrations ($\mu\text{g/L}$) of PNS overlying water after 10 d redeposited and bedded sediment toxicity tests.

		Dissolved metals ($\mu\text{g/L}$)						
		Cr	Cu	Pb	Ni	Fe	Mn	Zn
MDL ^a		2.71	2.67	4.06	2.98	10.1	2.7	3.52
RL ^b		10	10	10	10	100	5	10
MS03 redeposited	Control	ND ^c	ND	ND	ND	32 (1)	120 (13)	24 (11)
	Treatment	ND	ND	ND	ND	35 (3)	23 (1)	20 (18)
MS03 bedded	Control	ND	4 (1)	ND	ND	37 (3)	182 (19)	9 (1)
	Treatment	ND	4 (1)	ND	ND	48 (11)	26 (2)	7 (2)
MS04 redeposited	Control	ND	4 (1)	ND	ND	ND	111 (32)	ND
	Treatment	ND	5 (NA ^d)	ND	ND	40 (2)	16 (1)	6 (1)
MS04 bedded	Control	ND	ND	ND	ND	35 (1)	135 (46)	8 (1)
	Treatment	ND	3 (0)	ND	ND	39 (1)	15 (1)	8 (3)

^a Calsciences method detection limit

^b Calsciences reporting limit

^c ND = below MDL

^d NA: No SD because only 1 rep had Cu above MDL.

Note: Concentrations of Fe more than likely resulting from interaction of saltwater with instrument detector, given the relatively high reporting limit.

Appendix

Table A1. Mean (n = 4 to 8 ± 1 SD) particulate metal concentrations through time.

Sediment	Hour	Metal concentration (µg/g dw)							
		Zn	Cu	Fe	Mn	Ni	Pb	Cr	OC (%)
Lake DePue	1	10,047 (345)	432 (19)	40,502 (1,839)	682 (35)	79 (19)	NA ^a	142 (7)	NA
	2	10,262 (446)	439 (13)	41,971 (767)	689 (36)	60 (6)	NA	NA	NA
	3	10,516 (324)	440 (18)	42,528 (1,354)	710 (24)	66 (8)	NA	NA	NA
	4	10,485 (335)	437 (17)	41,564 (1,829)	731 (38)	75 (17)	NA	127 (8)	3.6 (0.1)
MS03	1	720 (30)	911 (43)	42,000 (1,083)	372 (18)	82 (8)	299 (24)	200 (5)	NA
	4	771 (31)	961 (51)	45,075 (2,729)	394 (27)	79 (6)	312 (3)	215 (14)	4.2 (1.2)
MS04	1	367 (44)	325 (10)	36,145 (4,869)	349 (63)	65 (8)	168 (19)	218 (16)	NA
	4	430 (35)	384 (36)	43,270 (3,137)	407 (24)	66 (5)	139 (52)	245 (17)	4.8 (0.8)

^a NA = not available

Table A2. Mean (n = 4 ± 1 SD) AVS, SEM and $\sum(\text{SEM-AVS})/foc$ concentrations from bedded and resuspended (0.25 and 0.75 cm depths) sediments

	Exposure	AVS	\sum SEM	$\sum(\text{SEM-AVS})/foc$
		µmol/g dw	µmol/g dw	µmol/g OC
Lake DePue	Bedded	60 (3)	196 (7)	4,973 (251)
	Redeposited (0.25cm)	60 (3)	207 (18)	5,384 (631)
	Redeposited (0.75cm)	57 (4)	214 (8)	5,737 (318)
MS03	Bedded	20 (1)	6 (1)	-921 (74)
	Redeposited (0.25cm)	19 (4)	8 (2)	-701 (288)
	Redeposited (0.75cm)	18 (2)	7 (0)	-648 (117)
MS04	Bedded	9 (1)	3 (0)	-267 (49)
	Redeposited (0.25cm)	7 (1)	4 (1)	-140 (84)
	Redeposited (0.75cm)	7 (0)	4 (1)	-133 (51)

Table A3. Mean (± 1 SD, $n = 4$) *Hyaletta azteca* Cu, Ni and Cd body burdens following exposure to overlying water from 4-h bedded and resuspended events

	Body burden ($\mu\text{g/g dw}$)		
	Cu	Ni	Cd
Bedded control	38.4 (8.8)	2.7 (0.5)	0.9 (0.2)
Bedded	33.5 (3.9)	4.4 (3.2)	0.9 (0.2)
Resuspended control	47.1 (13.3)	3.8 (2.1)	1.2 (0.2)
Resuspended	43.5 (3.9)	4.7 (3.7)	0.8 (0.2)

Table A4. Mean (± 1 SD, $n = 4$) *Hyaletta azteca* Cu and Cd body burdens following exposure to 7-d bedded and redeposited sediment toxicity exposures

	Body burden ($\mu\text{g/g dw}$)	
	Cu	Cd
Bedded control	73.3 (7.7)	1.2 (0.1)
Bedded	107.7 (18.7)	15.0 (4.9)
Redeposited control	92.7 (10.2)	1.3 (0.1)
Redeposited ^a	58.4 (43.0)	15.4 (0.4)

^a $n = 2$ because of poor survival ($10 \pm 12\%$)

Table A5. Summary of simple linear regression statistics comparing 4-h DO and pH to 4-h turbidity

	Comparison	R^2	p -value	Shapiro-Wilk p -value
Lake DePue	DO v. turbidity	0.8921	0.000409	0.2286
	pH v. turbidity	0.2848	0.173	0.02539
MS03	DO v. turbidity	0.8571	0.000965	0.4917
	pH v. turbidity	0.6394	0.01721	0.2322
MS04	DO v. turbidity	0.96	0.0000203	0.9533
	pH v. turbidity	0.007026	0.844	0.06118

Table A6. Summary of t-test statistics comparing bedded to redeposited metal bioavailability measurements

	Parameter	t-statistic	<i>p</i> -value	Shapiro-Wilk <i>p</i> -value
Lake DePue	AVS	0.1449	0.888	0.1211
	SEM	1.1327	0.301	0.7802
	Bioavailability	1.2095	0.272	0.7825
MS03	AVS	0.6899	0.516	0.1719
	SEM	2.3407	0.0578	0.2130
	Bioavailability	1.4802	0.189	0.0628
MS04	AVS	2.1817	0.0719	0.2922
	SEM	2.1939	0.0707	0.0223
	Bioavailability	2.5869	0.0414	0.4937

Table A7. Summary of 2-way ANOVA statistics (df, F and *p*-values) for *Neanthes arenaceodentata* metal body burdens Following exposure to MS03 and MS04 bedded and resuspended overlying water

Factor	df ^a	Zn		Cu		Ni		Pb		Cr ^b		
	n	F	<i>p</i>	F	<i>p</i>	F	<i>p</i>	F	<i>p</i>	F	<i>p</i>	
MS03	Controls v. treatments ^c	1	4.1015	0.06781	0.4585	0.51117	0.0485	0.8294	1.8665	0.1969	2.3035	0.1550
	Bedded v. resuspended ^d	1	0.2215	0.64711	4.2182	0.06245	1.4733	0.2482	0.8895	0.3642	26.8257	0.00023
	Interaction	1	0.0317	0.86189	1.4870	0.24611	0.7230	0.4118	1.8300	0.2011	0.7445	0.40514
	Shapiro-Wilk (<i>p</i>)			0.4727		0.7045		0.2411		0.0128		0.3707
MS04	Controls v. treatments	1	2.4782	0.1414	0.0081	0.9300	1.1436	0.3059	2.6973	0.1264	2.4330	0.1448
	Bedded v. resuspended	1	3.9558	0.0700	5.1406	0.0427	0.6389	0.4396	8.1921	0.0143	5.8772	0.0321
	Interaction	1	1.2017	0.2945	1.3749	0.2637	0.0536	0.8208	0.0178	0.8961	1.9322	0.1898
	Shapiro-Wilk (<i>p</i>)			0.3270		0.2802		0.5168		0.9164		0.3923

^a degrees of freedom

^b MS03 Cr was natural log transformed prior to analysis

^c Controls v. treatments = (bedded control + resuspended control) v. (bedded treatment + resuspended treatment)

^d Bedded v. resuspended = (bedded control + bedded treatment) v. (resuspended control + resuspended treatment)

Table A8. Summary of 2-way ANOVA statistics (df, F and *p*-values) for *Neanthes arenaceodentata* metal body burdens following MS03 and MS04 bedded and redeposited sediment toxicity test

Factor	df ¹	Zn		Cu ²		Ni		Pb		Cr	
	n	F	<i>p</i>	F	<i>p</i>	F	<i>p</i>	F	<i>p</i>	F	<i>p</i>
MS03 Controls v. treatments	1	0.8754	0.3679	4.5966	0.0532	84.5726	<0.001	1.6608	0.2218	2.2369	0.1606
MS03 Bedded v. redeposited	1	13.7660	0.0030	0.9158	0.3575	0.9303	0.3538	9.2637	0.0102	1.0464	0.3265
MS03 Interaction	1	0.1020	0.7549	0.1582	0.6978	4.3265	0.0596	41.8103	< 0.001	0.0025	0.9609
MS03 Shapiro-Wilk (<i>p</i>)			0.9618		0.0761		0.7935		0.2231		0.9590
MS04 Controls v. treatments	1	3.0943	0.1040	13.2042	0.0034	4.1958	0.0631	1.2251	0.2901	0.0942	0.7646
MS04 Bedded v. redeposited	1	2.2451	0.1599	0.2108	0.6544	2.4312	0.1449	0.2579	0.6208	0.0612	0.8091
MS04 Interaction	1	0.1937	0.6677	0.0296	0.8663	0.5032	0.4917	0.6559	0.4338	5.3813	0.0406
MS04 Shapiro-Wilk (<i>p</i>)			0.9950		0.3110		0.5034		0.7736		0.1226

¹ degrees of freedom

² MS04 Cu was ln transformed prior to analyses

Table A9. Tukey post hoc HSD *p*-values for *Neanthes arenaceodentata* body burdens following MS03 and MS04 bedded and redeposited sediment toxicity test.

Comparison	MS03 Pb <i>p</i> -value	MS04 Cr <i>p</i> -value
Treatment:Bedded – Control:Bedded	0.0006974	0.4662267
Control:Redeposited – Control:Bedded	0.1258423	0.5277204
Treatment:Redeposited – Control:Bedded	0.6144620	0.9783382
Control:Redeposited- Treatment:Bedded	0.0424527	0.9962581
Treatment:Redeposited – Treatment:Bedded	0.0001083	0.2942750
Treatment:Redeposited – Control:Redeposited	0.0149289	0.3289475

Table A10. Summary of 2-way ANOVA statistics (df, F and *p*-values) for *Neanthes arenaceodentata* growth rates following MS03 and MS04 bedded and resuspended exposures

		df ¹	Growth rate	
Factor		n	F	<i>p</i>
MS03	Controls v. treatments	1	0.0042	0.9496
	Bedded v. resuspended	1	4.1235	0.0651
	Interaction	1	0.1214	0.7335
	Shapiro-Wilk (<i>p</i>)			0.7212
MS04	Controls v. treatments	1	0.6652	0.4306
	Bedded v. resuspended	1	0.8521	0.3742
	Interaction	1	1.2115	0.2926
	Shapiro-Wilk (<i>p</i>)			0.9217

¹ degrees of freedom

Table A11. Summary of 2-way ANOVA statistics (df, F and *p*-values) for *Neanthes arenaceodentata* growth rates following bedded and redeposited sediment toxicity test

		df ¹	Growth rate	
Factor		n	F	<i>p</i>
MS03	Controls v. treatments	1	0.5139	0.4872
	Bedded v. redeposited	1	0.2839	0.6039
	Interaction	1	0.7664	0.3985
	Shapiro-Wilk (<i>p</i>)			0.4057
MS04	Controls v. treatments	1	7.7087	0.0168
	Bedded v. redeposited	1	0.0012	0.9735
	Interaction	1	0.9525	0.3484
	Shapiro-Wilk (<i>p</i>)			0.4258

¹ degrees of freedom

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