Role of Sediment Resuspension in the Remobilization of Particulate-Phase Metals from Coastal Sediments

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The release of particulate-phase trace metals due to
sediment resuspension has been investigated by combining
erosion chamber experiments that apply a range of
shear stresses typically encountered in coastal environments
with a shear stress record simulated by a hydrodynamic
model. Two sites with contrasting sediment chemistry were
investigated. Sediment particles enriched in silver,
copper, and lead, 4–50 times greater than the bulk surface-
sediment content, were the first particles to be eroded.
As the shear-stress level was increased in the chamber,
the total mass eroded increased, but the enrichment of these
trace metals fell, approaching the bulk-sediment content.
From the temporal distribution of shear stress generated by
the hydrodynamic model for a site in Boston Harbor,
resuspension fluxes were estimated. The erosion threshold
of this site is exceeded during spring tides, releasing the
particles enriched in trace metals into the water column. Due
to the higher trace metal content and the regularity of
resuspension, low-energy resuspension events (up to a
shear stress of 0.2 N/m²) contribute up to 60% of the
resuspension metal flux in an average year. The estimated
annual quantity of copper and lead resuspended into the
water column is higher than estimates of the total riverine
flux for these metals. These results indicate that sediment
resuspension is a very important mechanism for releasing
metals into the water column and provide new insight into
the chemical and physical processes controlling the long-
term fate of trace metals in contaminated sediments.

Introduction

The sediments of many coastal areas have become large
repositories of contaminant metals. Due to shallow water
depths and bottom stress induced by waves and currents,
the sediments in coastal areas are often subject to regular
episodes of sediment resuspension, so that the particulate-
phase metals may not be permanently sequestered in the
bottom sediments. The impact of resuspension on coastal
water quality and long-term contaminated sediment distri-
bution will depend on both the sediment geochemistry and
the coastal hydrodynamics.

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The focus of this paper is on the release of metals from
fine-grained coastal sediments by resuspension. Apart from
occasional events, such as dredging, resuspension is the only
mechanism that releases solid-phase metals into the water
column. All other release mechanisms, such as diffusion and
advection, require the metal to be in the dissolved phase.
The largest proportion of metals reside in the solid phase,
so determining the quantity and speciation of metals that
can be eroded during resuspension events is important for
understanding metal release from contaminated sediments.
Several field and laboratory studies have investigated the
chemical composition and changes that occur when sedi-
mentary metal phases are suspended in oxygenated water
(e.g. refs *1–4*). However, none of these investigations have
assessed how the release of metals varies over the range of
shear stresses that occur in the natural environment.

Sediment suspension occurs once critical flow conditions
within the water column have been exceeded, so that particles
are eroded and maintained in suspension by the exchange
of momentum from the fluid to the particles (*5*). In coastal
environments, tidal currents, wind waves, storm events, and
wave-current interactions drive resuspension (*6*). Multiple
factors including grain size, mineralogy, and the activity of
benthic organisms (*7*) determine the sediment's cohesive
strength and thus its ability to resist the shear stress imposed
by the overlying fluid. Due to the wide variety of parameters
that can influence the erodibility of cohesive sediments,
resuspension is difficult to predict. Studies investigating the
erodibility of cohesive sediments have used erosion chambers
and flumes to measure the magnitude and nature of
suspension with increasing bottom stress (e.g. refs *8* and *9*).

The release of metals due to resuspension is affected not
only by the composition of the sediment but also by the
diagenetic processes in the sediments. In sediments with an
oxic or suboxic layer, cycling of Fe and Mn can concentrate
trace metals near the sediment–water interface. Iron oxy-
hydroxides and manganese oxides present in this zone can
scavenge trace metals (*10*), so that erosion can resuspend
enriched particles. In organic-rich coastal-marine sediments,
sulfate reduction is important, and trace metals can be
coprecipitated or adsorbed to sulfide minerals (*11*). If these
particles are resuspended, they can be oxidized, releasing
dissolved metals to the water column.

The purpose of this study is to investigate metal release
over the range of shear stresses that are typically expected
in the coastal environment, from quiescent to annual storm
conditions. We coupled measurements of three contaminant
metals, Ag, Cu, and Pb as well as Fe and Mn, with
meteorological observations and a hydrodynamic model to
quantify the role of resuspension as a source of contaminants
to Boston Harbor, U.S.A. This combination of geochemistry,
erosion experiments, and hydrodynamics has enabled a more
complete understanding of benthic metal fluxes.

Materials and Methods

Erosion Chamber. A modified EROMES laboratory erosion
device (*12*) was used to determine the sediment response
over the range of shear stresses that occur annually in Boston
Harbor. Turbulence is generated by a propeller mounted 3
cm above the sediment, and baffles mounted in the 10.7 cm
core barrel suppress full-body rotation of the water column
(Supporting Information Figure S-1). All metal parts in the
original design were replaced with plastic to enable trace
metal sampling. Water and suspended sediment samples
were collected through a port in the top cap. The imposed
bottom shear stress is a function of the propeller rotation

TABLE 1. Site Locations and Characteristics of Surface Sediments

location	water depth	collection method	sand/silt/clay, %	surface solid-phase content[a] (mmol/kg dry weight)						
				Fe	Mn	Ag	Pb	Cu	AVS[b]	%OC
Hingham Bay 42.29083N 70.92778W	5 m	diver collected	6/62/32	760 ± 30	12.0 ± 0.4	0.024 ± 0.003	0.37 ± 0.02	1.1 ± 0.04	1.8	3.1 ± 0.2
Massachusetts Bay 42.38981N 70.8305W	30 m	damped gravity corer (14)	18/57/25	570 ± 70	10 ± 0.9	0.006 ± 0.001	0.30 ± 0.04	0.57 ± 0.08	nd[c]	2.0 ± 0.2

[a] Average and standard deviations. HB: 6 cores collected between 2002 and June 2004; MB: 6 cores collected between 2002 and Sept. 2003. For samples collected simultaneously with the erosion experiment cores see Table S-3. Data for surface samples with depths of either 0−0.5 cm or 0−0.3 cm depending on sampling date (see Supporting Information for sampling resolution and dates). [b] AVS data from June 2004 for HB, Sept 2003 for MB. [c] nd - not detected.
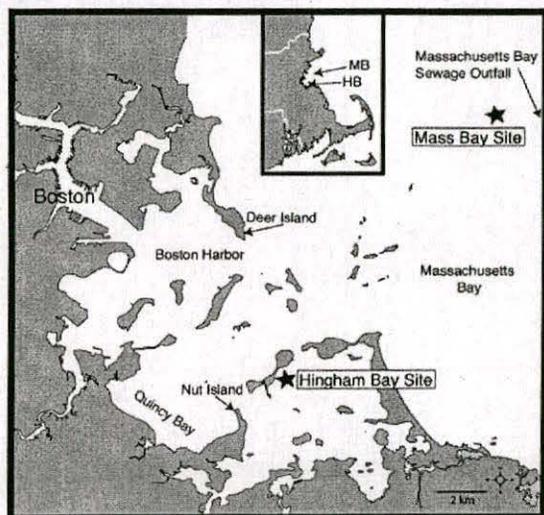
FIGURE 1. Map of coastal Massachusetts showing sampling site locations: MB, Massachusetts Bay site; HB, Hingham Bay site.

rate. Sieved sand and Shield's erosion threshold curve was used to calibrate the chamber. The relationship between the propeller rotation rate and shear stress at the sediment−water interface was linear. More details of the chamber calibration procedure are provided in the Supporting Information.

Field Sites and Sampling. Erosion studies were undertaken at two sites in coastal Massachusetts, northeastern U.S.A., characterized by fine-grained sediment, one in Massachusetts Bay (MB) and the other in Boston Harbor (Table 1 and Figure 1). The sites span the range of geochemical environments expected in coastal regions. The MB site is 11 km offshore of Boston Harbor and located 2 km west of the sewage outfall for Boston which opened in 2000. The chemical composition of this site has been monitored since 1989 (13). The Boston Harbor site is a more sheltered site, in Hingham Bay (HB). This site has high concentrations of metals and other contaminants in sediments resulting from more than 130 years of sewage discharge to the southern embayments of the harbor (14). This site has been sampled periodically since 1977 (14) and has been more intensively sampled since 2001 (15, 16). Sedimentation rates are not well-known at either site, but an upper limit for the HB site is 0.6 cm/y (15) and an upper limit for MB is 0.3 cm/y (17).

Sediment samples from both sites were collected in ways that minimized the disturbance of the sediment−water interface (Table 1). At each site, cores for porewater and solid-phase analysis were held vertically on ice and sectioned under a nitrogen atmosphere within 6 h of collection for MB cores

and 8 h for HB cores. Cores for the erosion experiments were collected at the same time. More details on the sampling, sectioning, and experimental procedures are provided in ref 16, and a list of all the cores collected and the analyses performed on each are provided in the Supporting Information.

Erosion Experiments and Sample Analysis. Cores for the erosion experiment were transported by car to the laboratory, 2 h away. The cores had at least 15 cm of overlying water and no headspace, which minimized turbulence and sediment disturbance during transport. For HB cores, experiments were started within 6 h of core collection. For the MB sample, the core was refrigerated overnight, and the experiment was started 18 h after core collection. Although there is some risk that the erosional characteristics of the sediment were altered during sampling and transport, Tolhurst et al. (12) found with careful transportation, even with a delay of up to 20 h between collection and erosion analysis, the erosion thresholds of cores brought back to the laboratory were not statistically different from in situ determinations.

For the erosion experiment, the erosion core barrel was mounted rigidly in a stand with the experimental geometry shown in Figure S-1. All items used during the experiment were cleaned with trace-metal clean protocols (16). An initial water sample was collected, and then the rotation rate of the propeller was incrementally increased, until a maximum shear stress of 0.33−0.4 N/m^2 was reached, depending on the experiment. Ten to twelve stress increments were imposed for each experiment. Each rotation rate was held for 20 min.

After 20 min at each rotation rate, water samples were withdrawn. Fifty milliliters of sample was removed from the chamber and replaced with bottom water collected from the same site. The sample was filtered through a 47 mm acid-cleaned, polysulfone 0.45 μm filter membrane (Pall-Gelman) using a Savillex Teflon filtering tower. Each filter membrane was dried under an infrared lamp. The quantity of material on each filter membrane was determined gravimetrically.

The solid phases of the sectioned sediments and eroded particles were analyzed with a total digestion (18) involving additions of concentrated HCl, HNO$_3$, HClO$_4$, and HF. The concentrations of Fe, Mn, Ag, Pb, and Cu in the digests were quantified with ICP−MS. Acid volatile sulfide (AVS) was determined on frozen samples by a method adapted from ref 19 with a detection limit of 0.05 mmol/kg dry sediment. More details on the analytical methods are provided in the Supporting Information.

Hydrodynamic Modeling of Shear Stresses. To determine the temporal variation of bottom shear stresses at the Hingham Bay site a modeling and statistical approach was used. The method used is described briefly here, and more details are provided in the Supporting Information. The depth-averaged tidal model TRIM (20) run with 100 m
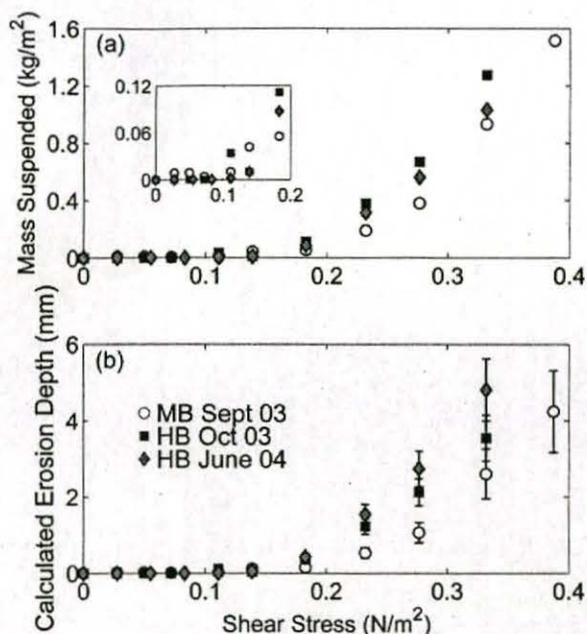
FIGURE 2. (a) Mass of particles resuspended at each shear stress increment. The inset is an expanded scale to show the response at low shear stresses (b) The calculated depth to which sediment was eroded at each shear stress. Error bars are calculated from the standard deviation in porosity.

resolution, wind data from NOAA buoy 44013, 20.6 km northeast of HB (hourly data from August 1984 to December 2003), and the NOAA tidal constituents for Boston Harbor were used to calculate the tidal and wind- and wave-driven currents at the site for the 19.4 year time period covered by the wind data. The Grant and Madsen (21) wave-current interaction formula was used to calculate combined shear stress. The depth-averaged currents (tide + wind-driven) were converted to 1 m above bottom using the model drag coefficient ($c_d = 0.003$). A constant physical bottom roughness of $k_b = 0.0001$ m was assumed, and a 19.4 year record of shear stress was generated.

Results and Discussion

Site Characteristics. While the Hingham Bay site has only slightly higher organic carbon and slightly finer sediments compared to the Massachusetts Bay site (Table 1), the two sites have very different geochemical compositions. HB is a strongly reducing site with acid volatile sulfide (AVS) and high levels of contaminant metals in surface sediments (Table 1). Porewater sulfide is detected below 6 cm (Figure S-3 and ref 15). The MB site is less reducing, with no AVS detected in the upper 1 cm of sediments and no porewater sulfide detected down to a depth of at least 30 cm. Ag, Cu, and Pb are all enriched at HB relative to MB, with the order of enrichment Ag ≫ Cu > Pb.

Erosional Characteristics. Erosion experiments were undertaken to provide a seasonal comparison (HB in June and October) and an intersite comparison (MB in October). The sediment surface at both sites in fall was largely free from benthic colonization except for 20−25 polychaete worm tubes protruding through the surface of the core. A red algal mat covered the entire sediment surface at HB in June.

No material is eroded into the chamber with the first shear stress steps (Figure 2). Only at a shear stress of 0.11 N/m² for the fall experiments and at 0.14 N/m² for the June HB experiment is there a significant release of particles. These points are the critical shear stress τ_c, where the imposed shear stress first exceeds the cohesive forces within the

sediments. Beyond the τ_c, the mass eroded rises steadily with each shear stress increase. The values of τ_c, measured here are similar to but higher than the average $\tau_c = 0.10 \pm 0.04$ N/m² determined by Ravens and Gschwend (9) using an in situ linear flume at nearby Quincy Harbor. These authors also found a decrease in erodibility when algal mats were present, and this may be responsible for the higher erosion threshold observed here for June.

Even at the highest shear stresses imposed in each experiment, the erosion depths did not exceed 6 mm (Figure 2). The depths calculated are average erosion depths as the sediments were assumed to have eroded uniformly over the core surface area. At all shear levels, the HB site was eroded to a deeper level than the MB site, and the HB site eroded to essentially the same depth in each experiment. These calculated erosion depths compare with the work of Ravens (22) who found that two storms exerting a shear stress of 0.5 N/m² and 0.35 N/m² eroded 4.4 and 3.1 mm of sediment, respectively, from the Quincy Bay site.

Particle Release. The total amount of particulate trace metal eroded at each stress level is shown in Figure 3. Once the critical shear stress for erosion was exceeded, increasing the shear stress further led to increasing amounts of particulate-phase trace metals being eroded. Fe, Mn, Pb, Ag, and Cu all show a smoothly increasing release as the shear stress increases beyond the threshold of erosion for both HB experiments. The total amount of metal eroded in October 2003 is consistently higher than in June. Lead shows the same monotonic increase as the other metals up to the shear stress of 0.28 N/m², but becomes constant in June 2004, and is variable at high shear stresses in October 2003. The MB data also show a smooth increase in solid-phase metals after the erosion threshold has been exceeded, and the total amounts of each metal eroded become constant at higher shear stresses.

Although the total amount of solid-phase metal resuspended generally increased with increasing shear stress, the metal content in the eroded particles (in μmol(metal)/g(eroded sediment)) changed with each shear stress step (Figure 4). There is considerable variation in the metal content over the range of shear stresses for each metal, indicating that as erosion progresses, sediment particles with different levels of metal enrichment are eroded.

Both HB experiments show some release of Fe and Mn prior to the erosion threshold, likely associated with the few particles that were visually evident in the water column before the onset of bulk erosion. There are peaks in Ag, Pb, and Cu that coincide with these prethreshold Fe and Mn peaks, suggesting that the trace metals may be adsorbed to the readily erodible Fe and Mn phases.

For Ag, Cu, and Pb the particles with the highest metal content are eroded at or just beyond the critical erosion threshold. Beyond 0.2 N/m² the metal contents fall with continuing erosion. The only exception is the behavior of lead in October 2003, where peak content occurs at 0.23 N/m², well beyond the erosion threshold. The variation in Fe content is different, with the content rising gradually after the erosion threshold and then reaching a steady, maximum level beyond 0.28 N/m² for HB and falling for MB. The manganese data show a similar trend to the iron data, except for the one highly enriched sample collected at a shear stress of 0.14 N/m² in HB in June 2004.

The metal content of eroded particles is compared to that of bulk sediments, averaged over the upper 3 mm of the sediment column, in Figure 5. The metal contents of the initially eroded particles are higher than the bulk contents, and as erosion increases the level of enrichment decreases. Calvo et al. (23) and Cantwell et al. (3) also reported enrichments above the bulk content in resuspended sediments. As erosion progresses and reaches the depth over
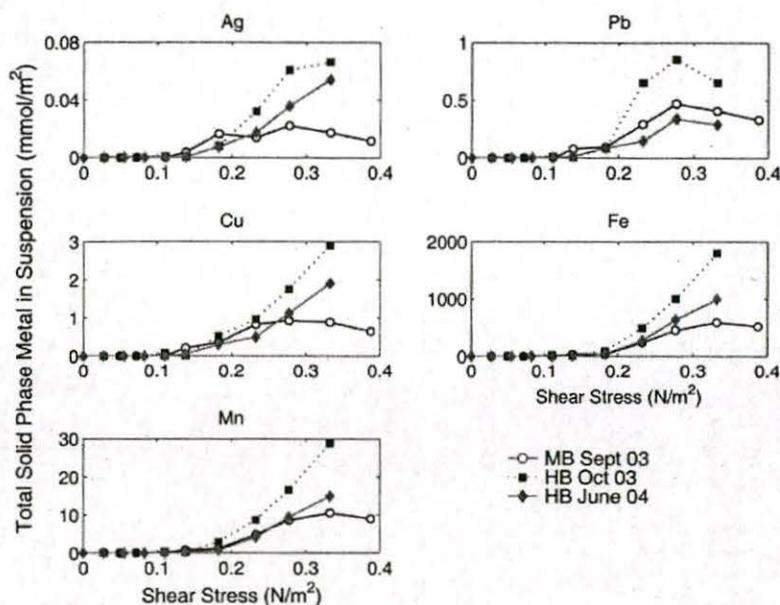
FIGURE 3. The total amount per sediment surface area of particulate trace metals resuspended in the erosion chamber for each experiment.
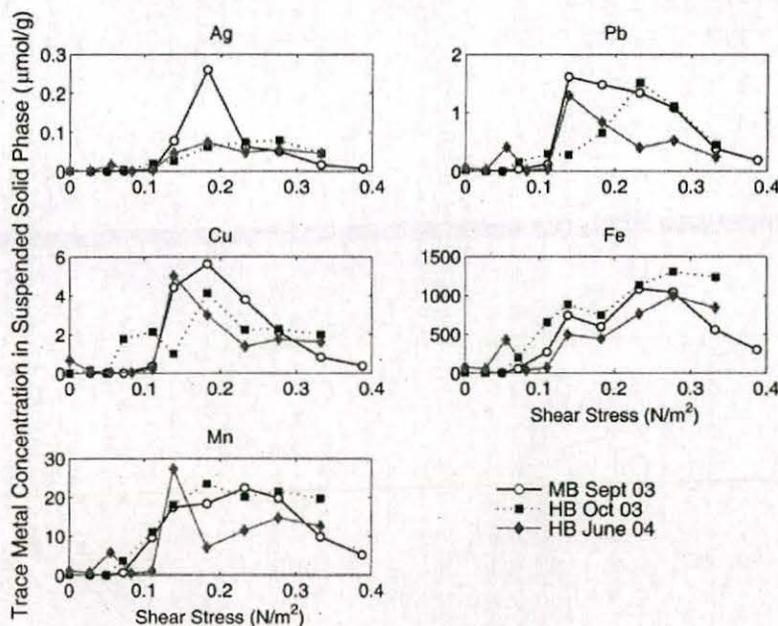

**FIGURE 4.** Trace metal content in the eroded solid phase as a function of shear stress.

which the sectioned sediments were sampled, the content of the metals, normalized by the average bulk surface sediment content (Table 1), tend toward one (Figure 5). This suggests that, at the highest shear stress, the erosion chamber is resuspending bulk sediments. The metal enrichments at low shear stress could indicate resuspension of a much thinner layer than the 3 mm sediment layer sampled or selective resuspension of enriched particles.

The level of enrichment for the MB samples is greater than for HB. In MB, the most readily eroded particles have a silver content 45 times greater than bulk sediment. The order of metal enrichment is Ag > Cu > Pb > Mn, Fe. In the HB experiments, Cu ~ Ag > Pb > Mn, Fe. This is the same order (without Ag) of enrichment as found by Cantwell et al. (3).

Potential processes responsible for the enrichment observed in the most readily erodible sediments include both physical and chemical mechanisms. The physical mechanisms could be due to preferential association of metals with finer, more readily erodible particles within the sediments (14). The higher levels of enrichment in MB may be due to a higher fraction of sand and therefore a disproportionate amount of the fine-grained, metal-enriched particles eroding (3). If this were the sole mechanism leading to the differences, Fe would also be enriched in the initially eroded particles of MB, which is not observed.

The chemical mechanism leading to the enrichment may be a diagenetic process that focuses metals at the sediment–water interface. Porewater analysis of MB sediments indicates that there is always dissolved iron below 1 cm, but no measurable iron in the top 3 mm throughout the year (Figure S-3). This suggests that close to the sediment–water interface, iron oxyhydroxides are being precipitated, thereby removing dissolved iron from the porewaters. Trace metals are often
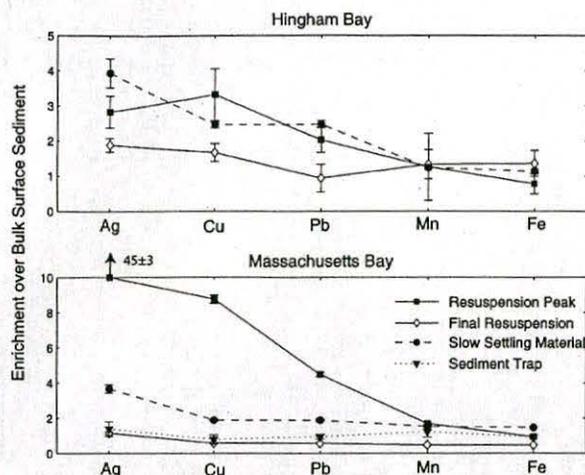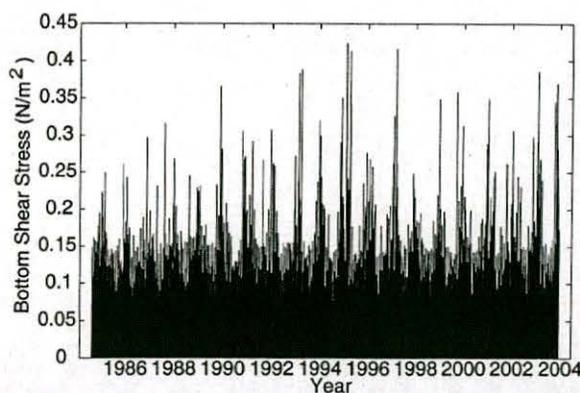
FIGURE 5. Particle metal contents normalized by the average bulk surface sediment content (Table 1) for both Hingham Bay and Massachusetts Bay. The particles shown are as follows: (1) the resuspension peak - particles resuspended at or one stress step beyond the erosion threshold, (2) final resuspension - the resuspended particles sampled at the maximum shear stress, (3) slow settling material - eroded particles in suspension after 8 h, and (4) sediment trap - particles collected in USGS sediment traps moored 16 m above the seafloor, 4 km from the MB sampling site, 2001–2003 data.

FIGURE 6. Simulated bottom shear stress distribution at the Hingham Bay site.

TABLE 2. Number of Shear Stress Events in Hingham Bay Simulated Record

shear stress (N/m^2)	no. of events[a] in simulated record	average no. of events (/year)
0.11	1436	74
0.14	729	37
0.18	191	9.8
0.23	81	4.2
0.28	31	1.6
0.33	13	0.7
0.35	10	0.5

[a] If two or more events occurred within 24 h they were defined as only a single event.

strongly scavenged by iron and manganese oxide phases (24, 10), so that precipitation of these oxides close to the sediment–water interface can lead to an enriched layer of trace metals close to the sediment–water interface. At MB the Fe, Mn, Ag, Cu, and Pb contents in the upper 1 cm are 1.2–1.4 times higher than the contents below 10 cm (16). This diagenetically enriched surface layer can then be eroded. HB is more reducing, and dissolved iron in the 0–3 mm porewater samples is detected in every season except winter (Figure S-3 and ref 16). As reducing conditions within the sediments intensify over summer the surface iron oxyhydroxide layer is reduced, such that the trace-metal-trapping oxide layer is destroyed or restricted to a very thin surface layer that is not detected by bulk sampling techniques. The erosion chamber may be detecting this thin oxidized layer as the enriched, initially eroded particles, that represent an erosion depth of less than 1 mm. Because the surface oxide layer is restricted or does not persist throughout the year at HB, the levels of trace metal enrichment at the sediment surface cannot reach those observed at the MB site.

Alternatively, the enrichments in the most readily erodible material may reflect newly arrived matter that has reached the sediment–water interface but has not been incorporated into the sedimentary matrix. However a comparison of the eroded material composition with that of the USGS sediment trap data (25) indicate that the two materials are quite different at the MB site (Figure 5). The peak contents of Ag, Pb, and Cu are all higher than the contents measured in the sediment traps, while the Fe and Mn contents are similar. Material collected in sediment traps does not exactly capture material reaching the seafloor, due to processes such as hydrodynamic sorting and resuspension, but the data strongly suggest that Ag, Cu, and Pb are enriched in the most readily erodible sediments relative to settling material, while Fe and Mn are not.

A combination of winnowing of the fine-grained particles as well as erosion of the diagenetically formed surface enriched layer in the sediments is likely to be responsible for the high contents of trace metals that are initially eroded at both sites. Whatever the mechanism, the erosion of enriched particles at low shear stresses increases the impact of

sediment resuspension on remobilizing metals from coastal sediments. Low shear stresses will occur more frequently, and so regular erosion of enriched particles is a significant mechanism of remobilization.

The metal content of material that remained in suspension 8 h after the erosion experiments had ended is also shown in Figure 5. At both sites the trace metal contents of this fine material are up to a factor of 4 times higher than the bulk surface sediment. An important result is that the particles remaining in suspension over time scales of hours are enriched in metals relative to the bulk sediments from which they were derived. These particles have a long residence time in the water column. Sediment resuspension events can thus result in a dispersal of metal-enriched fine particles.

Temporal Variation of Shear Stress in Hingham Bay. To put the shear stresses imposed in the erosion chamber into context in the marine environment, the modeled shear stress distribution for the HB site is shown in Figure 6. The number of times each imposed shear stress level is reached (an event) in the record is shown in Table 2. The erosion threshold at the Hingham Bay site is exceeded up to 74 times per year, several times during each spring tide. More intense erosion events are due to strong winds associated with the passage of storms. However, the impact of a wind event is strongly dependent on both wind direction and the tidal stage when the wind event occurs.

As an estimate of the importance of contaminant release, we combine data from erosion chamber experiments with data from our hydrodynamic model and assume the Hingham Bay site is representative of the total 41 km^2 of fine-grained sediment regions within Boston Harbor (calculated from ref 26). Below we consider how representative the chemistry, erosion characteristics, and shear stress exposure of the HB site are to those Harbor-wide. Data from other fine-grained sites within the Harbor (27, 28) (Figure S-4) show that the metal contents found at HB are typical. The porewater profiles

of sites in the northern Harbor (27) are also similar to those of HB, suggesting that the sediment geochemistry of sites at opposite ends of the Harbor are comparable. The water depth at HB is close to average for the Harbor (4.9 m (29)), but the site is more sheltered, particularly from northeast storms, than many other fine-grained areas. Therefore the shear stress frequencies given in Table 2 are likely to be underestimates for the Harbor. Although erosion characteristics are difficult to predict, and therefore extrapolate, Ravens et al. (9) found similar, slightly lower, erosion thresholds and depths for Quincy Bay, the largest region of fine-grained sediments in the Harbor. The erosion thresholds for the main regions of fine-grained sediments are thus comparable, but slightly lower, than those found at HB. Use of the HB site to represent the Harbor is likely to yield a lower bound of the Harbor-wide release as other sites have approximately the same metal content but will experience more intense shear stresses than the HB site.

Assuming that characteristics of the Hingham Bay site are representative of the fine-grained sediment within Boston Harbor and using the average erosion release from the two HB erosion experiments, we calculate that sediment resuspension events up to a shear stress of 0.33 N/m² mobilize 20 000, 20 000, and 800 kg/yr of solid-phase Pb, Cu, and Ag from Boston Harbor as well as 7000 and 100 tons/yr of Fe and Mn. While there is significant uncertainty in this estimate, it far exceeds the estimated riverine input to the Harbor of lead and copper of 2200 and 2700 kg/yr (30) and represents a large quantity of contaminated sediments that are in motion. Much of this mobilized load will settle back to the seafloor; however, the small fraction of particles that remain in suspension are enriched in trace metals and can be transported away from the source. The Ag, Pb, and Cu contents in the surface sediments of Boston Harbor have been decreasing since 1977 (14). Resuspension is likely to be a factor contributing to this decrease. Further work investigating particle settling rates and dynamics is needed to quantify the metal loss from the sediments and to fully understand the long-range transport and redeposition of these contaminated particles. Erosion experiments undertaken at additional sites within the Harbor are also needed to improve the preliminary estimates given here.

At the Hingham Bay site, erosion events ≤0.14 N/m² suspend 1% of the mass of particles that a 0.33 N/m² event suspends. However these lower shear stresses suspend 14%, 18%, and 30% of the calculated annual resuspension load of particulate Ag, Pb, and Cu, respectively. This disproportionate contribution is due largely to the regularity at which these low shear stresses occur as well as the trace metal enrichment in the most readily erodible material. Sites with readily erodible particles and/or geochemical processes that trap trace metals in the surface oxide layer will have an even greater contribution from the low-energy resuspension events. Resuspension of contaminated particles from coastal sediments is thus not restricted to extreme episodic events but occurs throughout the year. This should be considered when assessing the long-term redistribution of contaminated sediments.

These experiments attempt to quantify resuspension release under the typical range of environmental shear stress conditions. Field investigations during resuspension events need to be conducted to both verify and improve the flux estimates determined using the erosion chamber. As the erosion chamber investigations have indicated, resuspension is a very important mechanism releasing contaminated particles into the water column; additional studies are warranted to further our understanding of the fate of metals in contaminated sediments.

Supporting Information Available

Detailed information on the modified EROMES erosion chamber, trace metal, and AVS analysis and the method used for simulating the temporal record of shear stress for the HB site, description of cores collected and analyses undertaken for each cruise, iron and sulfide porewater profiles for each site, solid-phase metal data for the dates of the erosion experiments, porosity data and data on the overlying water used in the erosion experiment, and a plot comparing the HB site with other data for Boston Harbor. This material is available free of charge via the Internet at http://pubs.acs.org.

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