

Declining metal levels at Foundry Cove (Hudson River, New York): Response to localized dredging of contaminated sediments

Joshua A. Mackie^{a,*}, Susan M. Natali^b, Jeffrey S. Levinton^b, Sergio A. Sañudo-Wilhelmy^{b,c}

^a Moss Landing Marine Laboratories, Invertebrate Zoology, 8272 Moss Landing Road, CA 95039-9647, USA

^b Stony Brook University, Department of Ecology and Evolution, Stony Brook, NY 11794-5245, USA

^c University of Southern California, Marine and Environmental Biology, Los Angeles, CA 90089-0371, USA

Received 31 August 2006; received in revised form 11 January 2007; accepted 11 January 2007

Dredging of a hotspot of metal-contaminated sediment is associated with a recognizable local and river-wide decline in cadmium in the Hudson River, New York.

Abstract

This study examines the effectiveness of remediating a well-recognized case of heavy metal pollution at Foundry Cove (FC), Hudson River, New York. This tidal freshwater marsh was polluted with battery-factory wastes (1953–1979) and dredged in 1994–1995. Eight years after remediation, dissolved and particulate metals (Cd, Co, Cu, Pb, Ni, and Ag) were found to be lower than levels in the lower Hudson near New York City. Levels of metals (Co, Ni, Cd) on suspended particles were comparatively high. Concentrations of surface sediment Cd throughout the marsh system remain high, but have decreased both in the dredged and undredged areas: Cd was 2.4–230 mg/kg dw of sediment in 2005 vs. 109–1500 mg/kg in the same area in 1983. The rate of tidal export of Cd from FC has decreased by >300-fold, suggesting that dredging successfully stemmed a major source of Cd to the Hudson River.

© 2007 Elsevier Ltd. All rights reserved.

Keywords: River pollution; Cadmium; Metal removal; Superfund project; Hudson River

1. Introduction

The potential hazards of metal contaminants to humans and ecological systems are well-recognized (Gochfeld and Burger, 1998; Jarup, 2003; US EPA, 2004) and have promoted large-scale remediation efforts in a range of ecosystems. For example, in the United States, the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA), commonly known as the “Superfund Act” of 1980, has focused attention and resources on identifying contaminants, their associated risks, and on developing technologies for cleaning and containing hazardous waste. However, while 25 years have passed since the inception of CERCLA, there is still

a small amount of objective data on the efficacy of remediation projects. These data are essential to guiding future projects, and for increasing our understanding of how chemical parameters affect biotic systems.

Remediation of benthic systems is primarily accomplished by dredging-removal of contaminated sediments with or without sediment replacement. The determined recovery status of a remediated site reflects both the particular system dynamics and response variables measured (Kelaher et al., 2003; Kemble et al., 2000; Schuwerack et al., 2001; Voie et al., 2002; Zajac and Whitlatch, 2001). Dredging itself is considered a major ecological disturbance to the benthic community and may affect sediment structure, and species diversity and richness (Kelaher et al., 2003). In many cases, this immediate ecological effect will be outweighed by the primary goal of removing pollutants from a locale where they pose a threat to human health.

* Corresponding author. Tel.: +1 831 771 4403; fax: +1 831 632 4403.

E-mail address: jmackie@mml.calstate.edu (J.A. Mackie).

This study examines the effect of dredging on metal concentrations in Foundry Cove (FC), in the Hudson River, just south of Cold Spring, NY. Waste effluents containing large amounts of cadmium (Cd), cobalt (Co) and nickel (Ni) were released into the Cove by a battery factory from 1953 to 1979. Prior to remediation FC was the most Cd-polluted riverine site known (Förstner, 1984; Moore and Ramamoorthy, 1984). Large areas of the cove had sediment Cd concentrations of 10,000 mg/kg (Knutson et al., 1987). Contaminated sediments were removed without replacement from FC during two clean-up campaigns. In 1972–1973, 90,000 m³ of sediments were removed by dredging, however, Cd concentrations were not significantly altered (Knutson et al., 1987). In 1994 under the auspices of the 1980 Superfund Act, the U.S. Environmental Protection Agency (EPA) began a larger-scale clean-up. The upper 30 cm of sediment was removed from across the entire main body of FC (an area referred to as ‘Eastern Foundry Cove’ in reports); the highly contaminated factory outfall was dredged and capped with gravel (US EPA, 1989, 1998).

Considering both the direct ecological impacts and considerable financial costs of dredging operations, further studies are required to document the effectiveness of this strategy in removing toxicants, as well as resulting ecosystem effects. The unusual history of FC has made it a useful site for studying the ecological and genetic effects of Cd toxicity (Klerks and Levinton, 1989; Levinton et al., 2003; Martinez and Levinton, 1996) and the ecological and evolutionary effects of removing contaminated sediments (Kelaher et al., 2003; Levinton et al., 2003).

The objectives of the present study were to determine the concentrations of metals in the water column at FC, 10 years after environmental remediation, and to compare these values to other locations within the Hudson and other rivers. Total dissolved and suspended particulate concentrations of a suite of metals (Cd, Co, Cu, Pb, Ag, and Ni) were measured in the water column at FC. Concentrations of Cd in surficial sediments at FC were determined, including samples within the dredged location and neighboring non-dredged areas. If dredging has resulted in comprehensive removal of heavy metals, we would expect low metal levels in FC, relative to other areas in the Hudson River and to pre-dredging values. Alternatively, elevated metal levels may persist in FC due to incomplete remediation or transport from other sources. If high concentrations of metals other than those discharged by the battery plant are detected in the Cove, this may indicate the presence of additional sources of metals into FC and the Hudson River.

2. Methods

2.1. Study area

Foundry Cove is situated on the eastern side of the Hudson River south of the village of Cold Spring, and 90 km north of The Battery, New York City, NY. The main body of water of Eastern FC is 12 ha and is bordered to the north, east, and south by cattail marsh (Fig. 1). Localized exchange of water between FC and the Hudson River occurs through a ca. 20-m wide opening, beneath a railroad trestle (Knutson et al., 1987). Although FC is tidally

influenced, salinity only reaches 3–6 psu during periods of low freshwater flow (Bower et al., 1978).

2.2. Water samples

Surface water samples were collected from FC ($n = 11$) in August 2002. Sampling was conducted over two half ebb tides and a complete flood tide (Appendix 1). Unfiltered and filtered water samples were collected 1 m below the surface through acid-washed Teflon tubing into acid-cleaned polyethylene (LDPE) bottles. Filtered samples were passed through an acid-cleaned polyethylene filter cartridge (0.45 μm) attached to the pump system. The unfiltered and filtered samples were acidified to $\text{pH} < 2$ using quartz-distilled hydrochloric acid and stored for 3 months prior to analysis. Most of the metals attached to suspended particles as well as those bound to dissolved organic ligands are released by this passive acid-digestion protocol (Sañudo-Wilhelmy et al., 1996). The accuracy of this digestion protocol is suggested by the fact that distribution coefficients (K_d) between the particulate and dissolved metal pools calculated using this digestion protocol are indistinguishable from those calculated using a strong acid-digestion (Sañudo-Wilhelmy et al., 1996). Therefore, the particulate fraction of the samples ($>0.45 \mu\text{m}$) was calculated as the difference between the unfiltered and the filtered ($<0.45 \mu\text{m}$) dissolved fractions. Water samples were preconcentrated with an APDC/DDDC organic extraction (Bruland et al., 1985). Samples were analyzed for Cd, Co, Cu, Pb, Ni, and Ag using a Thermo-Finnegan Element2 Inductively Coupled Plasma Mass Spectrometer (ICP-MS). Analytical procedures yielded full recoveries of all trace metals measured within the Canadian National Research Council standard reference material for trace metals (CASS-3) and relative standard deviations of $<10\%$ for replicated analyses of samples. Analytical detection limits were 15 pM for Pb, 0.60 pM for Ag, 0.15 nM for Cu, 0.20 pM for Cd, 0.60 nM for Ni and 0.10 nM for Co.

The opening beneath the railway trestle provides the single major water connection between FC and the main Hudson River. Metal fluxes (the product of metal concentration, water velocity, and area of water movement between FC and the Hudson River) were estimated assuming a 1 m water column depth and 20 m width, approximately matching the dimensions of the opening. The export term for each metal, based on sampling of one tide, was determined by calculating the difference in areas of flux curves of the incoming and outgoing tide. Areas were determined by summing trapezoids of flux plots. Water velocity (cm/s) was measured with the aid of a stopwatch (0.1 s accuracy) at the surface using a free-moving float moving past a fixed 2 m floating rod, at 11 time points (with 10 replicates during each period) throughout the tidal cycle. Estimated velocities at metal sampling times were not made simultaneously as water samples were collected, but inferred via graphical interpolation. The Mann–Whitney–Wilcoxon (MWW) test, assuming independence of data points, was used to test whether flux of an element was consistently higher on either the incoming or outgoing tide. Significance was estimated from a two-tailed distribution of the T statistic. Probability values of <0.05 were considered significant.

Pre-dredging tidal transport data are available for Cd only (Hazen and Kneip, 1980; J. Levinton, unpublished data). We estimated water column-based metal transport between FC and the Hudson River for Cd following a previously published method: the difference between the mean influx and efflux concentrations of Cd in one tidal cycle was multiplied by 125,000 m³, the estimated volume of water exchanged per tidal cycle (Hazen and Kneip, 1980).

2.3. External reference site for measurement of trace metals

Water samples from one location in the Lower Hudson River were collected and analyzed for comparison with levels measured at FC. This site (referred to as ‘LHR’), is 19 miles north of The Battery, in a highly urbanized drainage system receiving a range of anthropogenic-source toxicants. Wastewater emissions from the nearby North River Wastewater Treatment Plant (DEP NY, 2006) appear to contribute to the elevation of some metals (Ag, Ni, Cu) (Sañudo-Wilhelmy and Flegal, 1992). Water samples ($n = 5$) were collected at LHR during one complete ebb tide and two half flood tides, and corresponded to the maximum and minimum current speeds during the

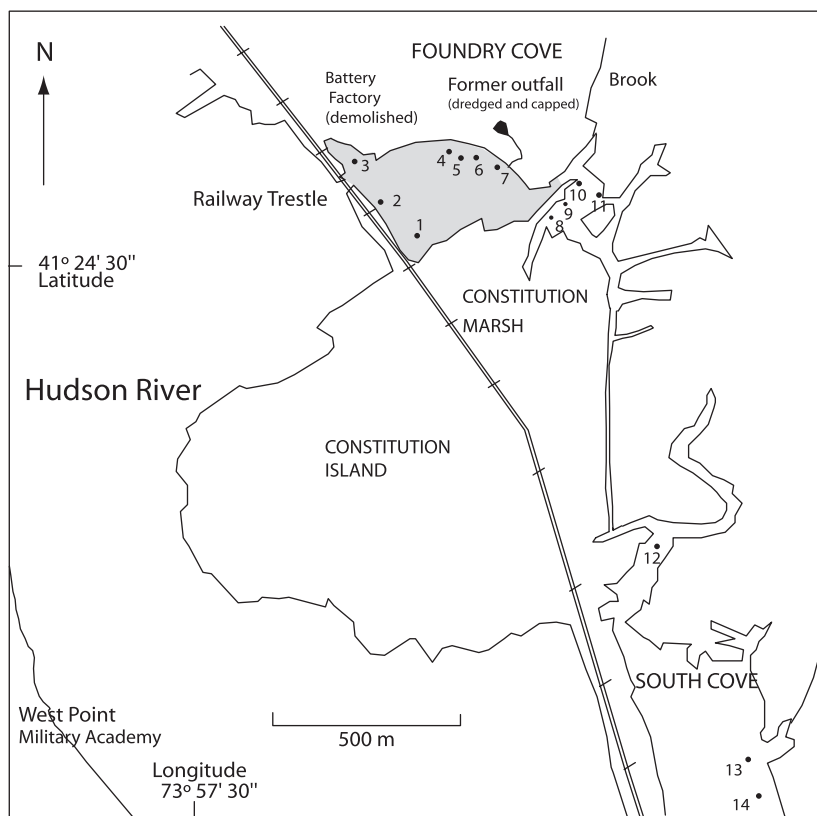


Fig. 1. Map of Foundry Cove and surrounding area. Thirty centimeters of sediment was dredged from the shaded area in the 1994–1995 Superfund remediation campaign. Numbers correspond to sampling stations at which surface sediment cores were collected and analyzed for cadmium in 2005 (Table 2).

tidal cycle (Appendix 1). Differences in surface water metal concentrations between FC and LHR were tested by multivariate analysis of variance (MANOVA; JMP 5.0, SAS Institute, 2002). When the MANOVA was significant, analysis of variance (ANOVA) was used to detect among site differences in individual dissolved and suspended particulate metals. Data were transformed when necessary to meet the assumptions of normality.

2.4. Measurement of surficial sediment cadmium

Surficial (top 5 cm) sediment samples were collected in 2005 from the dredged area within FC, Constitution Marsh, and neighboring South Cove ($n = 32$). Sediment was dried for 72 h at 60 °C in a Fisher Isotemp Oven, homogenized, and digested using repeated additions of nitric acid and H_2O_2 (EPA Method 3050B) and analyzed for Cd by flame atomic absorption spectrometry. Data were compared with measurements recorded prior to the clean-up by Hazen and Kneip (1980) and Knutson et al. (1987).

3. Results

3.1. Metals in the water column

Concentrations of dissolved metals were higher overall at LHR compared to FC (MANOVA: $p < 0.0001$; Table 1). Dissolved Cd was two times greater at LHR (1.01 ± 0.03 nM) than FC (0.46 ± 0.05 nM). Dissolved Ni was 1.5 times greater at LHR (17.25 ± 0.63 nM) than FC (11.22 ± 0.27 nM). Concentrations of Co, which along with Cd and Ni was released into FC by the battery plant operation, did not differ

significantly at the two sites (LHR: 0.74 ± 0.11 nM; FC: 0.64 ± 0.04 nM).

In contrast, particulate metal levels were higher overall at FC compared to LHR (MANOVA: $p < 0.002$; Table 1). Silver was the only metal found at higher concentration in suspended sediments at LHR. Levels of Cd, Co, and Ni in the suspended particulate fraction (Cd, 139 ± 27 nmol/g; Co, 1026 ± 149 nmol/g; Ni, 2039 ± 315 nmol/g) were at least twice as abundant at FC. Particulate Cd at LHR was undetectable by our method of analysis as the levels of this trace element in the dissolved and unfiltered samples were similar.

3.2. Flux of metals between Foundry Cove and the Hudson River

Maximum water velocities recorded during flood and ebb tides at FC were 74.6 and 66.7 cm/s, respectively. Standard errors of velocity measurements were < 0.6 cm/s. Based on the relative areas of the incoming and outgoing rates-of-change curves (Fig. 2), it is estimated that greater amounts of Ag, Cd and Co were ejected from FC on the outgoing tide relative to the ingoing tide (MWW test: $p < 0.05$). Suspended particulate Co, Cu, Pb, Ni, and Ag showed nominally greater levels of influx into the Hudson River from FC; however, MWW tests were non-significant, indicating that outgoing flux estimates did not consistently outrank the incoming flux estimates in size. In contrast, Cd showed a net efflux (MWW: $p < 0.05$)

Table 1
Range, mean, and standard error of dissolved and suspended particulate metals in the water column of Foundry Cove (FC), and the Lower Hudson River (LHR)

	FC			LHR		
	Range	Mean	SE	Range	Mean	SE
Dissolved						
Ag (pM)	2.55–7.28	4.91	0.51	60.6–83.3	68.4	4.37**
Cd (nM)	0.26–0.72	0.46	0.05	0.92–1.11	1.01	0.03**
Co (nM)	0.32–1.44	0.74	0.11	0.50–0.75	0.64	0.04
Cu (nM)	15.4–24.4	20.52	0.98	15.4–24.4	29.9	0.93**
Pb (nM)	0.09–0.20	0.17	0.01	0.28–0.31	0.30	0.004**
Ni (nM)	9.8–12.6	11.2	0.27	15.4–18.7	17.2	0.63**
Particulate						
Ag (nmol/g)	11–46	25	3.6	12.7–80.7	43	12.9
Cd (nmol/g)	28–317	139	26.5	Not detected		
Co (nmol/g)	470–1832	1026	149.1	216–777	406	101.8*
Cu (nmol/g)	1337–5297	2908	379.5	1420–3580	2330	370.1
Pb (nmol/g)	814–2939	1737	234.6	624–1620	1101	180.8
Ni (nmol/g)	868–4029	2039	314.8	268–898	587	104.5**

ANOVA: *significantly different between FC and LHR at $p < 0.005$; **significantly different at $p < 0.008$.

from FC. A total of 38.5 mmol Cd was estimated to be ejected over the tidal cycle.

3.3. Distribution of cadmium in FC surficial sediments

Sediment-surface-layer (SSL) Cd was assayed in 2005 within the dredged area of FC, and the adjacent undredged areas to the east (in Constitution Marsh), and 2 km to the south

(South Cove) (Fig. 1). At the northern margin of FC, an area with high Cd contamination prior to the clean-up, Cd levels in 2005 ranged from 2.4 to 230 mg/kg (mean: 59.7 ± 16.8) (Table 2). Cadmium measured in three cores from the western margin of FC, an area of lower contamination prior to the clean-up, ranged from 9.4 to 21.1 mg/kg (mean: 14.3 ± 4.9 mg/kg). Four samples collected directly to the east of the dredged area within Constitution Marsh had Cd

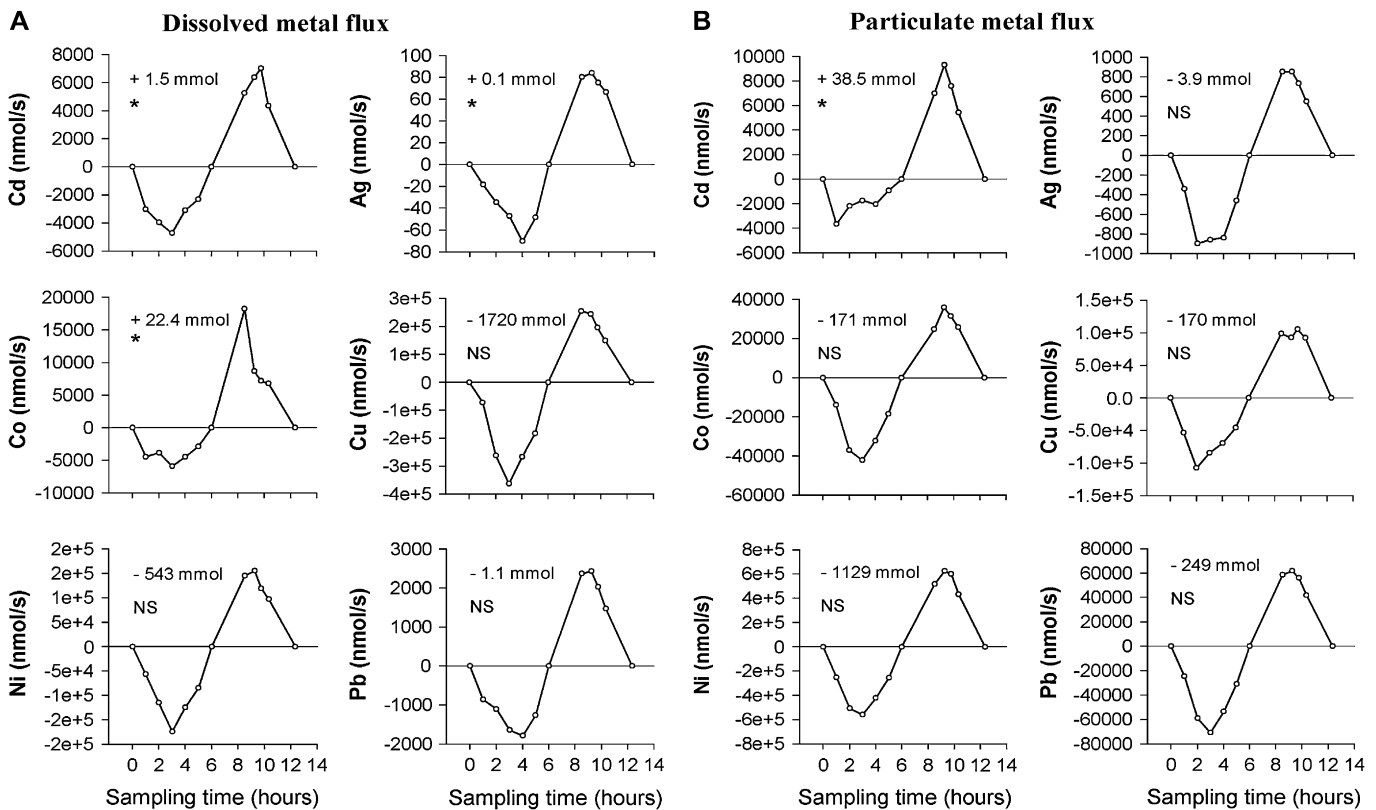


Fig. 2. Flux of (A) total dissolved metals and (B) suspended particulate metals in the water column between FC and the Hudson River calculated for one tidal cycle in 2002. Negative values on the Y-axis represent inflow into FC and positive values represent outflow to the Hudson River. The net direction of transport and magnitude of export terms are estimated as the sum of the area of inflow and outflow curves. The Mann–Whitney–Wilcoxon test was used to compare the median values of (non-zero) measures of flux in each tide phase (* $p < 0.05$; NS = non-significant).

in the range of 29–209 mg/kg (mean: 101.5 ± 41.5 mg/kg). Cores sampled at South Cove ranged from 1.1 to 8.0 mg/kg, with one core (no. 24), collected at the southern part of Constitution Marsh reporting 12.4 mg/kg Cd.

The SSL Cd data were compared with measurements from the same areas recorded prior to the clean-up during two periods: 1971–1975 (Hazen and Kneip, 1980), and 1983 (Knutson et al., 1987) (Table 3). These data are shown graphically in the online supporting material. Scatter plots of the Log concentrations of SSL Cd, excluding concentrations measured directly within the former outfall are shown in Fig. 3. Analyses of covariance (ANCOVAs), treating distance from outfall as a covariate, and analyzing measures from dredged and undredged zones separately, indicated significant declines in SSL Cd occurring in both zones (dredged: $p < 0.000$; non-dredged: $p = 0.036$).

4. Discussion

In 1983, 4 years after battery-factory waste dumping was stopped, SSL Cd levels at FC averaged 4005 ± 1431 mg/kg (range of 12–39,500 mg/kg) (Knutson et al., 1987). The Cd

was unevenly distributed. Extreme concentrations (range: 3200–120,000 mg/kg, mean: $25,026 \pm 7485$ mg/kg) were reported within the waste pond and natural inlet that constituted the outfall that fed into FC (Hazen and Kneip, 1980; Knutson et al., 1987). A hotspot of slightly lower concentration occurred within the main part of FC in proximity to the outfall. The majority of Cd occurred in the first 30 cm of sediment; however, a few cores indicated Cd levels of >100 mg/kg extending below this depth (Knutson et al., 1987); FC was dredged to a depth of 30 cm in 1994–1995.

Our measurements indicate that elevated SSL Cd remains at FC. An average of 59.7 mg/kg was found in cores sampled at the northern margin near the former outfall, suggesting that Cd extended significantly below 30 cm of depth at the time of dredging, or that dredging resulted in incomplete removal of contaminated sediment from across the hotspot area. Cores from an undredged region of Constitution Marsh, directly adjacent to the FC, had the highest levels of Cd detected in our sampling (range: 28.7–209, mean = 101.5 ± 41.5 mg/kg).

At South Cove, which is connected to FC by a series of shallow channels, mean SSL Cd in the 1970s was estimated to be 35.5 ± 14.6 mg/kg (range: 14–77 mg/kg) (Hazen and Kneip, 1980). In 2005, the SSL Cd level at South Cove was 5.4 ± 1.3 mg/kg (range: 1.1–12.4 mg/kg). This level in fact remains high, both globally and relative to other areas of the Hudson River. Cadmium concentrations found in river sediments that are impacted by urbanization and industry (excluding the Hudson River Basin) were recently summarized as being between 0.1 and 2.9 mg/kg (Acevedo-Figueroa et al., 2006). The Hudson River Basin has received an unparalleled load of metal contamination from industrial and urban sources. Cd levels in recently deposited sediments are generally between 0.5 and 3.0 mg/kg, and have decreased slightly at a river-wide scale since the 1960s (Chillrud et al., 2003).

Cadmium was significantly increased in concentration in dissolved and particulate fractions in water leaving FC. In contrast to Cd, other metals Co, Cu, Pb, Ni, and Ag, measured as suspended particulate load, displayed a nominal, but non-significant, influx into FC from the Hudson River.

Estimates of the mass of Cd ejected from FC per tidal cycle made before the clean-up are: 1600 g in one cycle in 1974, 350 g in one cycle in 1976 (Hazen and Kneip, 1980), and 1000 g in one cycle in 1994 (J. Levinton, unpublished data). Sources of variation in these flux estimates are not known. Annual variations in the flow rate of the Hudson River may alter the rate and pattern of surface sediment exchange occurring at the mouth of FC. Secondly, pulses of saltwater which occur sporadically at FC in summer months, may mobilize sediment-bound Cd into solution, contributing temporal variability (Hazen and Kneip, 1980).

Dredging of FC in 1994 reduced flux of Cd to the Hudson River substantially. The difference in mean total (particulate + dissolved) Cd concentrations recorded throughout one tidal cycle at FC in 2002 was 0.013 $\mu\text{g/l}$, or equivalent to only 1.6 g of Cd being exported over the tidal cycle. A strong reduction in suspended Cd flux from FC may have occurred shortly after dredging, as a similarly low level of 4 g of Cd

Table 2
Cadmium in surface layer cores collected at Foundry Cove in 2005

Area	Core	Station	Cd (mg/kg)	Mean Cd \pm SE (mg/kg)
FC western margin	1	1	9.5	14.3 ± 4.9
	2	2	9.4	
	3	3	24.1	
FC northern margin	4	4	10.4	59.7 ± 16.8
	5		54.3	
	6		25.3	
	7		44.7	
	8	5	50.7	
	9		230.4	
	10		98.1	
	11		159.6	
	12	6	62	
	13		47.8	
	14		93.6	
Constitution Marsh	15		43.4	101.5 ± 41.5
	16	7	11.1	
	17		3.9	
	18		2.4	
	19		10.9	
	20	8	28.7	
	21	9	208.7	
South Cove	22	10	43.8	5.4 ± 1.3
	23	11	124.9	
	24	12	12.4	
	25	13	2.4	
	26		1.1	
	27		1.1	
	28		2.4	
	29	14	6.6	
	30		7.9	
	31		8.0	
	32		4.8	

Sample stations are shown in Fig. 1. Four cores were collected from within a 1-m² quadrat at stations 4, 5, 6, 7, 13, and 14. Other stations are represented by single cores.

Table 3
Historical measurements of surficial Cd (mg/kg dw of sediment) at Foundry Cove

Zone	Location (meters from outfall) ^a	Sample year						
		1971 ^b	1973 ^b	1974 ^b	1975 ^b	1976 ^b	1983 ^c	2005
Within the former waste outfall	Outfall 05 m						5700	
	Outfall 10 m	30,500	9630	42,000	120,000	11,500		
	Outfall 25 m						39,500	
	Outfall 50 m						12,800	
	Outfall 60 m	18,500	10,680					
	Outfall 160 m						6980	
	Outfall 175 m	10,400	25,700	28,300		3200		
FC (w. margin)	–605						38	
	–603						188	
	–601							9
	–600							24
	–600		33	790				
	–560						725	
	–550							10
	–535						554	
FC (n. margin)	–480		550					
	–420			760	430	3		
	–390	3						
	–300		450	1680	290	1600		
	–295	160						
	–220							35
	–200						109	
	–150						816	
	–140							124
	–100						1020	
	–60							55
	–50						1500	
	–40						671	
	–10							7
	0	38,200						
5						273		
15						6470		
Const. Marsh	480		150					
	485							125
	570							44
	580					780		
	600							209
	660							29
	660		430					
	690		870					
760		760		430	3			
South Cove	1530							12
	1800		35					
	2100		14					
	2120							2
	2180							7
	2330		77					
	2510		16					

^a Measurements from within the outfall channel run north-south, according to Hazen and Kneip (1980). Other measurements are positioned to the west (–) or east (+) of the outfall in FC.

^b Reported in Hazen and Kneip (1980).

^c Reported in Knutson et al. (1987).

was estimated to be ejected over one tide in September 1995, shortly after the clean-up (J. Levinton, unpublished data).

Dissolved concentrations of Cd and Ni, which were historically discharged into FC (Bower et al., 1978), were found to be significantly higher at the LHR site, suggesting the effective removal of sources of these metals to water at FC. A high

volume of sewage is discharged from the North River Sewage Treatment Plant, located near the LHR sampling site, and accordingly may elevate dissolved metals. Ag, a sensitive tracer of sewage (Sañudo-Wilhelmy and Flegal, 1992), was almost 14 times greater in the LHR (68.4 ± 4.4 pM Ag) than in FC (4.9 ± 0.5 pM Ag). Copper was slightly elevated in the

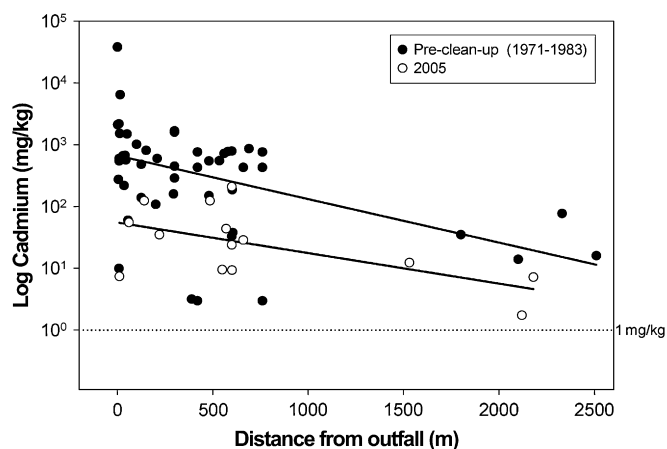


Fig. 3. Scatter plots of sediment-surface-layer measurements of Cd (Log scale) and distance from the former outfall for measurements made before and after the clean-up (Table 3). Lines of best fit for each distribution were fitted by linear regression.

LHR (29.9 ± 0.9 nM) compared to FC (20.5 ± 0.10 nM). Elevation of Cu in the LHR may be the result of wastewater effluent (Sañudo-Wilhelmy and Gill, 1999), or possibly benthic remobilization of contaminated estuarine sediments (Hall et al., 1988). Dissolved Pb, which has not previously been associated with wastewater runoff in the LHR, was also found at a higher level at LHR (0.30 ± 0.004 nM) compared to FC (0.17 ± 0.01 nM).

The ability to resolve significant anthropogenic differences in the sediment-bound fractions of metals is compromised by physical difference in the FC and LHR sites. The LHR site is closer to fully marine seawater, and therefore experiences higher and more heterogeneous saline concentrations. During sample collection, salinity at FC remained at 0 psu. The salinities that were recorded in the LHR (6–10 psu) are sufficient to cause particulate Cd to desorb and form complexes with dissolved organic and inorganic ligands (Libes, 1992; Rainbow, 1997; Sañudo-Wilhelmy and Gill, 1999). Therefore an absence of detectable particulate Cd in the LHR may be attributable to desorption (Sañudo-Wilhelmy and Gill, 1999).

To put the suspended particulate metal data recorded at FC into a wider context, concentrations of Cd, Ni, and Pb in FC were contrasted with levels in the San Francisco river system (SFRS) as reported by Flegal et al. (1996), using only SFRS sites with a salinity of 0 psu at sample collection. Mean suspended particulate Ni in FC was 1.5 times greater than in the SFRS, and suspended particulate Cd was 41 times greater. Suspended particulate Pb was 62 times more abundant at FC than the SFRS. Lead, which is not known to have been released by battery plant, is relatively common in Hudson River sediments (Bower et al., 1978). Further work is, however, required to distinguish geological and anthropogenic sources of Pb recorded at FC.

The reduction in Cd at FC has had a positive biological effect locally. Prior to clean-up, the population of a widespread freshwater oligochaete, *Limnodrilus hoffmeisteri*, that occurred within the highly contaminated portion of FC had a genetic mutation for resistance to Cd (Klerks and Levinton, 1989).

Genetic resistance, and the portion of protein-bound Cd found in the cytosol of worms decreased following the clean-up (Levinton et al., 2003). Prior to the clean-up, blue crab *Callinectes sapidus* had high tissue Cd concentrations throughout the Hudson River; subsequently levels have dropped to 20% of their previous values (Levinton et al., 2006). It remains uncertain as to whether this reduction in tissue-accumulated Cd can be explained simply as a consequence of stemming the FC Cd source. A reduction was observed in crabs sampled in proximity to FC and over the entire tidal Hudson River. Secondly, multiple sources of Cd to the Hudson River have recently declined (Bopp et al., 2006).

A range of general water-usage measures (Sañudo-Wilhelmy and Gill, 1999) and efforts to remove point-sources of contamination are being applied in the Hudson River. Notably, a large-scale dredging operation is planned with the aim of removing organic contaminants including sediment-associated polychlorinated biphenyls (PCBs) released by the General Electric Company's (GE) plant in Hudson Falls, New York. Remediation is planned to reduce organic contamination currently affecting 200 miles of the river (Chillrud et al., 2003). Consequently, ongoing studies that trace the origins and fate of the conglomerate of bioavailable pollutants in the Hudson provide a valuable opportunity for analyzing large scale effects of remediation strategies.

5. Conclusions

Our field results suggest that Superfund dredging has effectively but not completely removed a point source of metal contamination at FC. The present level of SSL Cd within the marsh system has decreased to <10% of pre-clean up levels. Cd contamination remaining at the former hotspot of contamination (60 mg/kg dw Cd) and proximal marsh (102 mg/kg dw Cd) is high in a global context, and exceeds the consensus-based threshold at which effects on test organisms are likely to be observed when exposed to freshwater sediments (5.0 mg/kg dw Cd; Crane et al., 2002). The volume of Cd transported per tide from FC to the main Hudson River was estimated to have decreased by over 300-fold, indicating a broad-ranging effect of remediative dredging.

Acknowledgements

We thank A. Grabman, B. Allen, R. Anderson, R. Young, D.E. Martinez, D. Wang, and A. Tovar-Sanchez for assistance with sample collection and analyses. Research was partially supported by a grant from the Hudson River Foundation to J. Levinton and a National Science Foundation Predoctoral Fellowship to S. Natali. Comments by an anonymous reviewer improved the manuscript.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.envpol.2007.01.010.

References

- Acevedo-Figueroa, D., Jiménez, B.D., Rodríguez-Sierra, C.J., 2006. Trace metals in sediments of two estuarine lagoons from Puerto Rico. *Environmental Pollution* 141, 336–342.
- Bopp, R.F., Chillrud, S.N., Shuster, E., Simpson, H.J., 2006. Contaminant chronologies from Hudson River sedimentary records. In: Levinton, J.S., Waldman, J.R. (Eds.), *The Hudson River Estuary*. Cambridge University Press, New York, pp. 383–397.
- Bower, P.M., Simpson, H.J., Williams, S.C., Li, Y.H., 1978. Heavy-metals in sediments of Foundry Cove, Cold Spring, New York. *Environmental Science and Technology* 12, 683–687.
- Bruland, K.W., Coale, K.H., Mart, L., 1985. Analysis of seawater for dissolved cadmium, copper and lead – an intercomparison of voltammetric and atomic-absorption methods. *Marine Chemistry* 17, 285–300.
- Chillrud, S.N., Bopp, R.F., Ross, J.M., Pederson, D.C., Chaky, D.A., Tolley, L.R., Estabrooks, F., 2003. Stable lead isotopes, contaminant metals and radionuclides in upper Hudson River sediment cores: implications for improved time stratigraphy and transport processes. *Chemical Geology* 199, 53–70.
- Crane, J.L., MacDonald, D.D., Ingersoll, C.G., Smorong, D.E., Lindskoog, R.A., Severn, C.G., Berger, T.A., Field, L.J., 2002. Evaluation of numerical sediment quality targets for the St. Louis River Area of Concern. *Archives of Environmental Contamination and Toxicology* 43, 1–10. Department of Environmental Protection (DEP), New York City, 2006. New York City Drainage Areas and Water Pollution Control Plants. Available from: <<http://www.nyc.gov/html/dep/html/drainage.html>>.
- Flegal, A.R., Rivera-Duarte, I., Ritson, P.I., Scelfo, G.M., Smith, G.J., Gordon, M.R., Sañudo-Wilhelmy, S.A., 1996. Metal contamination in San Francisco Bay waters: historic perturbations, contemporary concentrations, and future considerations. In: Hollibaugh, J.T. (Ed.), *San Francisco Bay: The Ecosystem*. American Academy for the Advancement of Science, pp. 173–188.
- Förstner, U., 1984. Cadmium in sediments. *Experientia* 40, 23–29.
- Gochfeld, M., Burger, J., 1998. Temporal trends in metal levels in the eggs of the endangered Roseate Tern (*Sterna dougallii*) in New York. *Environmental Research (Section A)* 77, 36–42.
- Hall, W.S., Bushong, S.J., Lenwood, W., Hall, J., Lenkevich, M.J., Pinkney, A.E., 1988. Monitoring dissolved copper concentrations in Chesapeake Bay, U.S.A. *Environmental Monitoring and Assessment* 11, 33–42.
- Hazen, R.E., Kneip, T.J., 1980. Biogeochemical cycling of cadmium in a marsh ecosystem. In: Nriagu, J.O. (Ed.), *Cadmium in the Environment*. John Wiley and Sons, New York, pp. 399–424.
- Jarup, L., 2003. Hazards of heavy metal contamination. *British Medical Bulletin* 68, 167–182.
- Kelaker, B.P., Levinton, J.S., Oomen, J., Allen, B.J., Wong, W.H., 2003. Changes in benthos following the clean-up of a severely metal-polluted cove in the Hudson River estuary: environmental restoration or ecological disturbance? *Estuaries* 26, 1505–1516.
- Kemble, N.E., Hardesty, D.G., Ingersoll, C.G., Johnson, B.T., Dwyer, F.T., MacDonald, D.D., 2000. An evaluation of the toxicity of contaminated sediments from Waukegan Harbor, Illinois, following remediation. *Archives of Environmental Contamination and Toxicology* 39, 452–461.
- Klerks, P., Levinton, J.S., 1989. Rapid evolution of metal resistance in a benthic oligochaete inhabiting a metal-polluted site. *Biological Bulletin* 176, 135–141.
- Knutson, A.B., Klerks, P.L., Levinton, J.S., 1987. The Fate of Metal Contaminated Sediments in Foundry Cove, New York. *Environmental Pollution* 45, 291–304.
- Levinton, J.S., Pochron, S.T., Kane, M.W., 2006. Superfund dredging restoration results in widespread regional reduction in cadmium in blue crabs. *Environmental Science and Technology* 40, 7597–7601.
- Levinton, J.S., Suatoni, E., Wallace, W., Junkins, R., Kelaker, B., Allen, B.J., 2003. Rapid loss of genetically based resistance to metals after the cleanup of a Superfund site. *Proceedings of the National Academy of Sciences of the United States of America* 100, 9889–9891.
- Libes, S.M., 1992. *An Introduction to Marine Biogeochemistry*. John Wiley and Sons, Inc., New York.
- Martinez, D.E., Levinton, J., 1996. Adaptation to heavy metals in the aquatic oligochaete *Limnodrilus hoffmeisteri*: evidence for control by one gene. *Evolution* 50, 1339–1343.
- Moore, J.W., Ramamoorthy, S., 1984. *Heavy Metals in Natural Waters: Applied Monitoring and Impact Assessment*. Springer-Verlag, New York.
- Rainbow, P.S., 1997. Ecophysiology of trace metal uptake in crustaceans. *Estuarine, Coastal and Shelf Science* 44, 169–175.
- Sañudo-Wilhelmy, S.A., Flegal, A.R., 1992. Anthropogenic silver in the Southern California Bight – a new tracer of sewage in coastal waters. *Environmental Science and Technology* 26, 2147–2151.
- Sañudo-Wilhelmy, S.A., Rivera-Duarte, I., Flegal, A.R., 1996. Distribution of colloidal trace metals in the San Francisco Bay estuary. *Geochimica et Cosmochimica Acta* 60, 4933–4944.
- Sañudo-Wilhelmy, S.A., Gill, G.A., 1999. Impact of the Clean Water Act on the levels of toxic metals in urban estuaries: the Hudson River estuary revisited. *Environmental Science and Technology* 33, 3477–3481.
- SAS Institute, 2002. *JMP Start Statistics, A Guide to Statistics and Data Analysis using JMP and JMP IN Software*, second ed. Thomson Learning, Duxbury.
- Schuerer, P.M.M., Lewis, J.W., Jones, P., 2001. The potential use of the South African river crab, *Potamonautes warreni*, as a bioindicator species for heavy metal contamination. *Ecotoxicology* 10, 159–166.
- US EPA, 1989. EPA Superfund Record of Decision, EPA/ROD/R02-89. Available from: <<http://www.epa.gov/superfund/sites/rods/fulltext/r0289097.pdf>>.
- US EPA, 1998. Five-year Review Report: Marathon Battery Company Superfund Site, Village of Coldspring, Putnam County. Available from: <<http://www.epa.gov/superfund/sites/fiveyear/f98-02005.pdf>>.
- US EPA, 2004. Region 3 Risk Based Concentration Table. Available from: <<http://www.epa.gov/reg3hwm/risk/human/index.htm>>.
- Voie, O.A., Johnsen, A., Rosslund, H.K., 2002. Why biota still accumulate high levels of PCB after removal of PCB contaminated sediments in a Norwegian fjord. *Chemosphere* 46, 1367–1372.
- Zajac, R.N., Whitlatch, R.B., 2001. Response of macrobenthic communities to restoration efforts in a New England estuary. *Estuaries* 24, 167–183.