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Superfund Dredging Restoration Results in Widespread Regional Reduction in Cadmium in Blue Crabs

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A nickel−cadmium battery factory released about 53 tons of mostly cadmium and nickel hydroxide suspended solid waste between 1953 and 1979 into Foundry Cove, which is tidally connected to the Hudson River estuary. A major Superfund dredging cleanup in 1994−1995 removed most of the cadmium from the sediment from within Foundry Cove. Here, we demonstrate that the cleanup reduced cadmium tissue concentrations (hepatopancreas and leg muscle) in an important fishery species, the blue crab *Callinectes sapidus* near Foundry Cove, but also across a broad reach of the Hudson River. Before the cleanup, cadmium concentrations in crabs were 4−5 times higher on average than after the cleanup and geographic variation in crab cadmium concentration along the Hudson River estuary was strongly reduced after the cleanup. The factor of reduction in crab tissue concentrations was far less than the factor of reduction of export of cadmium from Foundry Cove into the Hudson or the factor of reduction of cadmium sediment concentrations within the cove following the cleanup. This unique study demonstrates the efficacy of a major dredging cleanup and quantifies the spatial and temporal impact of the cleanup. It demonstrates that cleanup of a point source can have dramatic effects over large spatial scales.

Introduction

The widespread presence of metals in coastal and aquatic environments presents a major risk to human health (*1*, *2*). The most worrisome exposures result from consumption of fish, crustacea, and molluscs, which commonly contain metal concentrations above acceptable standards (*3*). For example, the Mussel Watch program reports high concentrations of metals such as mercury, cadmium, and copper near sources of pollution along much of our coastline (*4*). Understanding the impact of restoration on polluted areas is crucial, especially given the controversy concerning major dredging projects, which are costly and may remobilize and spread toxic substances over broad spatial scales (*5*). To what degree can such cleanups result in reductions of human exposure to toxic substances? What is the spatial impact of such cleanups of point sources?

Foundry Cove is a tidal freshwater water body on the east side of the Hudson River estuary, about 90 km north of The Battery in New York City. During the period 1953−1979 a battery factory released about 53 T of nickel-cadmium waste into Foundry Cove, resulting in very high sediment cadmium concentrations (*6*). Nearly all exchange of water and cadmium-laden particles between Foundry Cove and the adjacent Hudson River occurred through a narrow opening of about 26 m wide. The cove was dredged in 1994−1995, following a 1989 decision (*7*) under the auspices of the "Superfund" Act (Comprehensive Environmental Response, Compensation, and Liability Act; CERCLA). Before cleanup, cadmium concentrations in the sediment were in the range of 5−40 000 $\mu g\ g^{-1}$ (*8*) and as much as 250 000 $\mu g\ g^{-1}$ in the area near the outfall (J. Levinton, unpublished data). The entire cattail marsh area near the outfall was dredged and replanted. The rest of Foundry Cove was dredged to a depth of 30 cm. After the cleanup concentrations in the sediment and in the dominant oligochaete worm *Limnodrilus hoffmeisteri* were strongly reduced (*7*).

Previously, strong evolutionary (*7*, *9*) and ecological (*10*) impacts were demonstrated *within* Foundry Cove following the cleanup. Before the cleanup, cadmium within sediments was available to invertebrate feeders and cadmium found in the cytosol of invertebrate prey was efficiently transferable to predators (*11*). But Foundry Cove had also exported large amounts of cadmium to the adjacent Hudson River estuary (*12*), where tidal exchange and estuarine circulation could have strong impacts throughout a large portion of the estuary. Before the cleanup the edible blue crab *Callinectes sapidus* had high cadmium concentrations throughout much of the tidal freshwater Hudson River (*13*). Because the blue crab comprises an important sports and commercial fishery, uptake of cadmium by crabs is deemed a risk to human health in the Hudson River (*14*, *15*).

We here compare cadmium concentrations before (1981, 1984) and after the cleanup (2000, 2004) in the tissues of the blue crab from the Hudson River near Foundry Cove and from a series of localities, up- and down-river. This gives us the unique opportunity to assess the following: (i) if the cadmium concentrations in highly mobile crabs collected in the Hudson River near Foundry Cove and throughout the river were significantly reduced after the cleanup; (ii) if a geographic structure in crab tissue cadmium concentration existed before and after the cleanup for a stretch of the estuary of over 100 km; and (iii) if the change in tissue concentrations of cadmium in crabs corresponded to reductions in the export of cadmium from Foundry Cove to the Hudson following the cleanup or to the reduction of concentration of cadmium in Foundry Cove sediments before and after the cleanup. Therefore, our data allow us to uniquely assess the spatial impact of a point source cleanup on an adjacent water body with active water motion and long-distance dispersing organisms that readily absorb cadmium.

Estimates before the cleanup in 1974, 1976, and 1992 suggested a release from Foundry Cove to the open Hudson River of 1600 (*16*), 350 (*16*), and 1000 g (Levinton, unpublished data) of cadmium adsorbed to sedimentary particles per tidal cycle, whereas estimates of release after the cleanup in 1995 and 2002 were 4.0 (*17*) and 1.6 g (*18*) cadmium per tidal cycle, respectively. The differing estimates before and after the cleanup are broadly consistent; the rate of cadmium release into the main part of the Hudson was reduced by an

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average factor of over 300, following the 1994−1995 remediation. Before the cleanup Foundry Cove sediment cadmium concentrations (excluding measurements within the outfall marsh creek) averaged 3532 μg g^{-1} ± 1662 SE (*8*) (range of 12−39 500 μg g^{-1}) but averaged 10.9 μg g^{-1} afterward, from one estimate (*19*), ranging from 0.7 to 81.2 μg g^{-1}. The sediment concentration within Foundry Cove was therefore reduced on average also by a factor of about 324, which is close to the estimate of reduction of export on suspended particles per tidal cycle from Foundry Cove to the Hudson River estuary. Another more recent estimate in 2005 yielded an average sediment concentration of 52.5 ± 14.6 μg g^{-1} cadmium (*16*). This yields a lower reduction ratio of 67. Before the cleanup, blue crabs in the Hudson River estuary had high cadmium concentrations, which was related to export of cadmium from Foundry Cove (*13*).

Experimental Section

Data Collection and Analysis. All data on cadmium in leg muscle and hepatopancreas tissues of crabs were obtained from the New York State Department of Environmental Conservation toxic materials database. Cadmium analyses for 1981 collections, before the cleanup, were reported by Sloan and Karcher (*13*) following methods of Feinberg and Ducauze (*20*), and these data were entered later into the New York State database, our data source for this study. The database also contained some data for collections in 1984. Leg muscle and hepatopancreas tissues of blue crabs collected in the summer of 2004 were analyzed by Pace Analytical (Madison, WI). The method was similar to that used in the 1981 study and involved extraction using repeated additions of nitric acid and H_2O_2 (U.S. EPA method 3050b) and the analytical method followed U.S.EPA method 6020, using a Hewlett-Packard model 4500 ICP-MS analyzer. Results were also entered into the New York State database. No percent recovery data were reported for the analyses before the cleanup. For post-cleanup analyses, recovery was measured on selected samples in 11 batches. Pre-digest spike percent recovery was 87.9 ± 1.3 (SE) and post-digest spike percent recovery was 104.4 ± 1.2 (SE). We report cadmium concentrations within tissues of Hudson River male crabs only since females were underrepresented in the field collections of 2004.

There was no significant regression between hepatopancreas cadmium and body mass, either in the pre-cleanup (ANOVA on regression: $n = 65$, $F = 0.418$, $p = 0.520$) or post-cleanup (ANOVA: $n = 55$, $F = 0.385$, $p = 0.385$) samples. Similarly there was no significant regression between muscle cadmium in the post-cleanup samples (ANOVA: $n = 55$, $F = 2.2524$, $p = 0.139$). There was a significant negative regression between cadmium and body mass for the pre-cleanup muscle samples (ANOVA: $n = 65$, $F = 5.2378$, $p = 0.026$) but the regression only explained 0.07 of the total variance, which is very weak. Average body mass in the pre-cleanup samples was 195.6 ± 8.1 (SE) g, and for the post-cleanup crabs average body mass was 134.4 ± 6.2 g. Given that the regression of pre-cleanup muscle cadmium on body mass was negative, and the body size change was in a direction opposite than would be expected for body mass to explain the change in cadmium concentrations in leg muscle, we can conclude that size was not an explanatory factor in the cadmium reduction we found post-cleanup.

Localities are described by river kilometer, the number of kilometers north of The Battery (southernmost tip of Manhattan Island, New York City). Data from before the cleanup were obtained from localities sampled by net (once at each station) in the summers of 1981 and 1984. Data after the cleanup were obtained in the same manner in the summer of 2004. Owing to uneven sampling success, the same river kilometer locations were not sampled in the before- and after-cleanup samples, with the exception of river kilometer 90, which is near the Foundry Cove opening to the Hudson. Localities had replicate crabs, so it was possible to estimate heterogeneity among localities by means of ANOVA.

Change in Cadmium Concentration

Changes near Foundry Cove. At Foundry Cove (River Mile 54, about River Kilometer 90), pre-cleanup mean cadmium concentrations in male crab hepatopancreas tissue declined from 11.86 μg g^{-1} ± 2.69 SE, ($n = 12$) to 2.89 μg g^{-1} ± 0.08 ($n = 15$) following the cleanup (Figure 1). Cadmium concentrations within crab leg muscle tissue declined from 0.30 μg g^{-1} ± 0.08 ($n = 12$) to 0.06 μg g^{-1} ± 0.03 ($n = 15$). This drop was statistically significant (Welch ANOVA comparison allowing for unequal variances: $p < 0.007$ for hepatopancreas and $p = 0.0142$ for leg muscle). The drop was by a factor of 4.1 for hepatopancreas and 5.0 for muscle cadmium. Even the higher hepatopancreas cadmium levels today are, on average, below the Food and Drug Administration's standard of concern for crustacea (3.0 μg g^{-1}) for safe consumption (*21*) and average leg muscle tissue concentrations are below the U.S. Environmental Protection Agency's risk factor (*22*) of 0.68 μg g^{-1}.

Changes throughout the Estuary. The cleanup of Foundry Cove was not a localized phenomenon. Averaged over all sampled localities within the Hudson River (Figure 1), pre-cleanup cadmium concentrations in crab hepatopancreas tissue declined from 8.13 μg g^{-1} ± 5.67 SE ($n = 65$) to 2.39 μg g^{-1} ± 2.01 ($n = 58$) post-cleanup. Cadmium within crab leg muscle tissue declined from 0.19 μg g^{-1} ± 0.20 ($n = 65$) to 0.04 μg g^{-1} ± 0.09 ($n = 58$). The reductions of a factor of 3.4 and 4.8, respectively, were substantial and statistically significant (Welch ANOVA: $p < 0.0001$ for cadmium concentration within both hepatopancreas and leg muscle tissue). However, this factor of reduction and the reduction near Foundry Cove was much less than the factor of reduction of sediment concentration or export of cadmium from Foundry Cove into the Hudson River.

Changes in Geographic Heterogeneity. Before cleanup, substantial and significant geographic variation existed in cadmium concentration in Hudson River crabs with maxima for hepatopancreas near Foundry Cove and the nearest down-river location and another significant peak near Croton Point for hepatopancreas but not for muscle (Figure 2). For both hepatopancreas and muscle, among-locality variation in cadmium was significant (for hepatopancreas, Welch ANOVA: $p < 0.0064$; for muscle, Welch ANOVA: $p < 0.0001$). Tissue concentrations of cadmium before the cleanup declined from Foundry Cove toward the north, indicating that Foundry Cove was the main source of cadmium involved in uptake by blue crabs. The peak near Croton Point could have derived from another cadmium source, dispersal of cadmium-rich crabs from upriver, or both. A sanitary landfill near Croton Point, however, showed no detectable source of cadmium for possible export into the Hudson River before it was remediated (*23*). After the Foundry Cove cleanup, overall concentrations of cadmium in tissues were reduced substantially over the entire region, and geographic variation in cadmium was reduced. No significant geographic variation existed in blue crab hepatopancreas after cleanup (Welch ANOVA: $p = 0.1893$). After cleanup, no significant geographic variation existed in muscle cadmium concentrations (Welch ANOVA: $p = 0.2650$), but there was a local (non-significant) maximum near Beacon (Figure 2).

Discussion

The availability of pre- and post-remediation data for blue crabs, with a series of localities spread up- and down-river from the Superfund site has given us an opportunity to

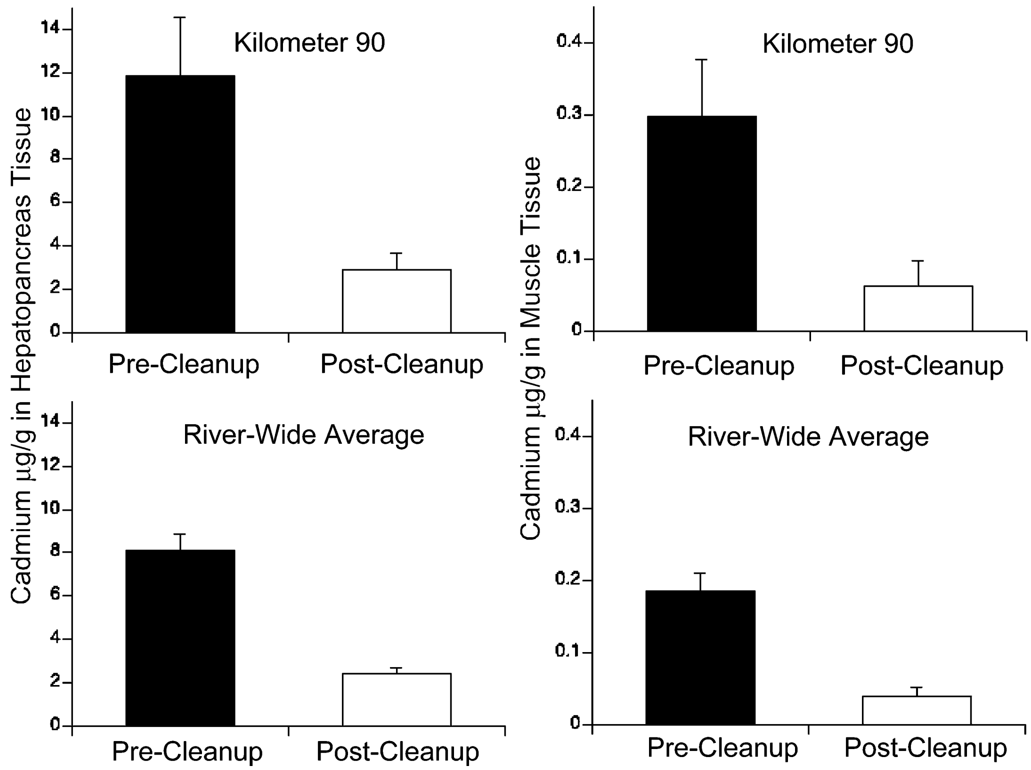

FIGURE 1. Mean changes over all Hudson River localities (± SE) in cadmium concentrations within hepatopancreas and leg muscle tissue of blue crabs from 1981 and 1984 (before cleanup) to 2000 and 2004 (after cleanup). Foundry Cove, the source of cadmium, lies at river kilometer 90.

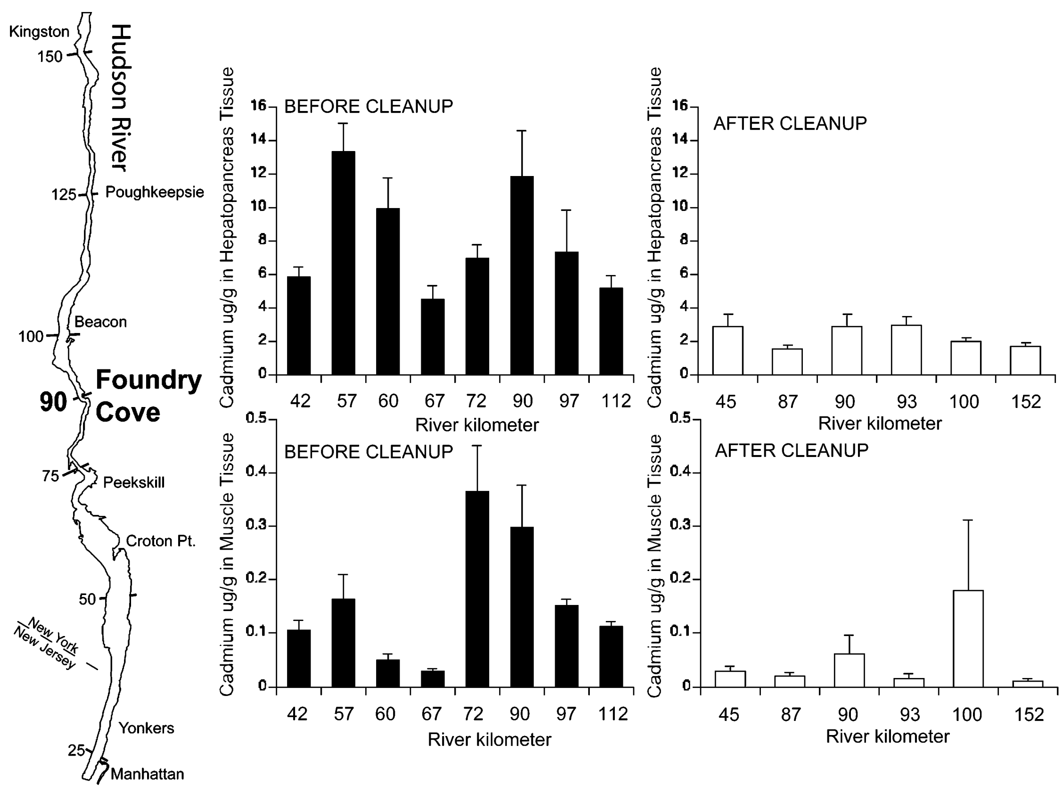

FIGURE 2. Changes in geographic distribution of cadmium concentrations (± SE) within hepatopancreas and leg muscle cadmium of blue crabs before and after the cleanup. Localities could not be matched with successful crab catches in both years. Foundry Cove, the source of cadmium, lies at river kilometer 90. Map data at left provided by the Hudson River Foundation.

characterize the success of the cleanup and the change in geographic pattern of cadmium in crabs. Blue crabs readily take up cadmium from the environment and uptake is mediated in the cytosol by a series of isoforms of low-molecular-weight metal-binding proteins (*24*). Influx and efflux of cadmium may be regulated across crab gills (*25*). Cadmium is more bioavailable to blue crabs at low salinities, and cadmium uptake increases (*26*). Salinities in the vicinity of Foundry Cove and localities covered in this study range from 0 to 5 psu (*27*). Before cleanup, Hudson River blue crabs had high concentrations of cadmium, and crabs collected near the point of tidal exchange with Foundry Cove had maximum concentrations. Following cleanup, concentrations of cadmium throughout the Hudson River declined, and geographic variation was greatly reduced. Although post cleanup crabs occasionally demonstrate cadmium concentrations that exceed health standards, our overall temporal and geographic findings demonstrate that the cleanup was very successful, despite a broad geographic impact of Foundry Cove on the Hudson River estuary before the cleanup. Dredging within the cove resulted in reduced cadmium concentrations of blue crabs throughout the Hudson.

Before cleanup, tidal mixing and crab mobility resulted in widespread exposure of cadmium to human consumers along the tidal freshwater Hudson, with local hotspots of cadmium concentration in crabs, especially near Foundry Cove itself. Following cleanup, both geographic variation and total concentration were reduced. Because large amounts of cadmium were exported to the open tidal freshwater Hudson, it is not clear that blue crabs obtained cadmium only near Foundry Cove. The salinity over much of the sampling area is usually less than 5 psu, which suggests that cadmium would be available for uptake from dissolved sources(26). Feeding within Foundry Cove, however, would have resulted in high degrees of trophic transfer of cadmium from benthic prey to crustacean predators (*11*).

While the case for the Foundry Cove cleanup as the direct cause for the reduction of cadmium in blue crabs is compelling, we must remember that other sources of metals have been cleaned up in our coastal waters during the same period. In the Hudson River basin, cadmium concentrations in sediments have declined since the 1960s (*28*). On the other hand, Croton Point, as mentioned above, was not likely a major source, nor was a pigment plant in the upper Hudson likely a source because cadmium concentrations declined strongly with increasing distance north of Foundry Cove in crab samples that were taken before the cleanup (*13*). In the decade that preceded the cleanup, cadmium concentrations in lower Hudson River freshwater sediments in the geographic range of this study were $<10\ \mu g\ g^{-1}$ (*28*). The export of cadmium from Foundry Cove was at a truly extraordinary level and it is therefore unlikely that more than a small proportion of the reduction of cadmium in blue crabs over time may be explained by a more regional recovery of contaminants.

Our overall conclusions of reduced overall cadmium concentrations in blue crabs and a reduction of geographic heterogeneity in concentrations is robust and well supported by the statistical analysis. Nevertheless, the structure of the data is far from ideal and was not the result of a pre-designed statistical model. It would have been preferable to have direct comparisons before and after the cleanup, with preferably multiple control localities, as conceived in BACI and related designs (*29*). We did not design this study and were only able to analyze the data available to us.

Large-scale cleanups of toxic sediment are costly and may remobilize sediment that might result in the transport of toxic substances to downstream sites (*5*) or even upstream in the case of the tidal lower Hudson River estuary. Having information on the degree to which such cleanups affect areas distant from the Superfund site and the overall degree of transfer of toxics to consumable species following cleanup is therefore critically important. Blue crabs in the tidal freshwater Hudson can move passively at least 20 km in a tidal rise or ebb (*30*), and this movement can rapidly spread cadmium-laden crabs to other areas. Females are known to use tidal streams to enhance migration distances (*31*).

In the Hudson, a 2002 decision to dredge the bulk of sediments containing high concentrations of PCBs (*32*) can benefit from comparisons with the present study, which supports the assertion that a large-scale dredging can result in salutary effects on exposure of toxins to humans and other species over a broad spatial scale.

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Supporting Information Available

MS Word file with all individual blue crab cadmium analyses for hepatopancreas and leg muscle, river km location, before-after cleanup, and body mass (g). This material is available free of charge via the Internet at http://pubs.acs.org.

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