



Chapter 6

Metal Distributions in the Surface Sediments of Long Island Sound

By

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INTRODUCTION

The distribution of metal contaminants in surface sediments has been measured and mapped as part of a U.S. Geological Survey study of the sediment quality and dynamics of Long Island Sound. Surface samples from 219 stations were analyzed and mapped for trace (Ag, Ba, Cd, Cr, Cu, Hg, Ni, Pb, V, Zn and Zr) and major (Al, Fe, Mn, Ca, and Ti) elements, grain size, and *Clostridium perfringens* spores.

Knowledge of the magnitude of contaminant concentration in sediments is important for regulation of contaminant discharges as well as management of the natural resources throughout the Long Island Sound watershed. A geochemical study of Long Island Sound was undertaken to understand the regional concentrations, distribution, and fates of contaminants in sediments as a component of a larger study by the U.S. Geological Survey. In this report, we present tables containing the analytical data for each of the analyses in the surface sediments of the Sound. Maps of the concentration distributions are presented to show patterns of regional contaminant distributions and metal co-variances that imply common sources or behaviors. This work offers a regional view of metal contaminants and the processes responsible for their observed distributions. This publication compliments and supplements the interpretive regional analysis of the U.S. Geological Survey's efforts in Long Island Sound that was initially presented in Poppe and Polloni (1998) and recently published in the *Journal of Coastal Research* (Buchholtz ten Brink and others, in press; Mecray and Buchholtz ten Brink, in press; Varekamp and others, in press).

METHODS AND ANALYSIS

Field Operations

Surface sediments were collected using a Van Veen grab sampler equipped with a video and still camera system (Blackwood and Parolski, 2000). The *R/V Seaward Explorer* was equipped on two cruises in 1996 to collect 191 samples from 130 stations within Long Island Sound (Fig. 1). The first cruise (SEAX96017) visited sample sites that were chosen to verify the sidescan sonographs used to interpret the sedimentary environments (Knebel and Poppe, in press). Sample locations on

the later cruise in 1996 (SEAX96024) were selected to measure the spatial distribution of contaminants across the Sound and were taken in a series of north-south transects from the Connecticut shoreline towards Long Island. On a third cruise (JD97001), aboard the *R/V John Dempsey* in 1997 (Poppe and Polloni, 1998), surface sediment samples were collected from the westernmost end of Long Island Sound. The combined sample set from all of the cruises provided an even sample distribution throughout the Sound. Samples were obtained only from those grabs that had visible features that appeared minimally disturbed and identified the sediment-water interface. The upper 2-cm of sediment were removed from the Teflon-coated grab using acid-washed, methanol-rinsed, Teflon-coated, titanium tools to prevent sample contamination. Sediment was placed in acid-washed, polyethylene containers, sealed, and those samples destined for chemical analysis were stored frozen. Salinity in the bottom water was also measured at selected stations during each cruise.

Laboratory Sample Analysis

Aliquots of the wet sediment were sampled in the laboratory using acid-washed, titanium tools. Samples for chemical analysis were freeze-dried to constant weight and the water content was calculated from mass lost. A total acid digestion was performed on 0.1200g (+/- 0.0005g) of the dried sample using hydrofluoric, nitric, perchloric, and hydrochloric acids under microwave-induced pressure (Murray and Leinen, 1996). Analysis of major and trace metals in the resulting solution followed standard procedures of inductively coupled plasma atomic emission spectroscopy (ICP- AES) and graphite furnace atomic absorption spectroscopy (GFAAS). Concentrations of 16 elements were reported in $\mu\text{g/g}$ dry sediment for the trace metals (silver (Ag), barium (Ba), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), manganese (Mn), nickel (Ni), vanadium (V), zinc (Zn), and zirconium (Zr)) and in weight percent (dry sediment) for the major elements (aluminum (Al), calcium (Ca), iron (Fe), and titanium (Ti)). Mercury (Hg) concentrations were reported in $\mu\text{g/kg}$ dry sediment after strong acid leach and elemental analysis (see Varekamp and others, in press). *Clostridium perfringens* concentrations were reported in spores/g dry sediment (see Buchholtz ten Brink and others, in press).

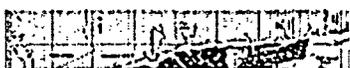
Analytical and procedural variability from the ICP-AES and the GFAAS were determined by simultaneously extracting and analyzing Standard Reference Materials (SRMs) along with a high-concentration sediment sample in triplicate. The SRMs and the method blanks were also analyzed in triplicate; results for each SRM analysis were within 10% of the accepted value. Sample replicates were within 5% of the mean value. Maximum salt content was calculated by measuring the salinity and the mass of water lost from representative samples. The salt calculation was performed to assess the effects on mass and metal content from the salts. The mass of salt remaining was determined to be less than 5% of the total mass of the sample. The mass difference and the resulting metal concentrations from the salts were within the error of analysis for all samples. The grain-size distributions were measured on wet sample aliquots from each surface sample site using the method reported in Poppe and others (1998).

Statistical means, medians, and ranges were calculated for the entire Sound (Table 2) and to elucidate regional patterns (Table 3). Natural background concentrations (Tables 2 and 3) were determined from cores taken in depositional environments within Long Island Sound. Background values would most likely be reached in areas of mostly fine-grained sediment than in areas dominated by coarse-grained sediment.

The maps were created in MapInfo (MapInfo Corp., 1998). The locations where the surface samples were taken were georeferenced from within the program. The point concentrations of the contaminants were displayed by creating thematic maps of the contaminant concentration attribute. The ranges were classified based on the sediment quality criteria of Long and others (1998). When those criteria were not available, ranges were created by dividing the number points equally. The contour maps were interpolated from the point concentrations using Vertical Mapper (Northwood Geoscience, 1999). The method of interpolation used was Triangulation with Smoothing where the data points are connected by a series of triangles. The points for the triangles are chosen by the Delaunay criterion, which causes the triangles to be drawn from points that are nearest neighbors. The resulting three-dimensional surface is smoothed by using a bivariate fifth-order polynomial expression in the X and Y direction for each triangle face to fit a smooth grid surface to the triangular faces. The color gradation is adjusted to match the point ranges manually.

RESULTS

Sample locations and the suite of metal concentrations measured in the surface sediments of Long Island Sound are documented in Buchholtz ten Brink and Mecray (1998). In this paper, we present the total metal concentration distributions for the



trace (Ag, Ba, Cd, Cr, Cu, Hg, Ni, Pb, V, Zn and Zr) and major (Al, Fe, Mn, Ca, and Ti) elements in Figures 2-17. Figure 1 shows the locations of all of the surface samples

taken over 3 separate field seasons. These sample locations are superimposed over the sedimentary environments maps for comparison purposes (Knebel and others, 1999). Table 1 contains all of the data for the 16 elements measured along with the latitude and longitude location information. Tables 2 and 3 present the results of the statistical analyses for the entire data set.

To open georeferenced displays of the metal distribution-theme in ESRI's ArcView program make sure the application is loaded on your computer. Users should go to the *lisound* directory located on the top level of this CD-ROM and double click on the *lisound.apr* project file. The individual ArcView shapefiles may also be opened directly with any Arc application (e.g. ArcInfo, ArcExplorer) and can also be found on the data page. Further detailed information can be found on the ArcView Project File page.

DISCUSSION

Metal Concentration Distributions

Maps of anthropogenic metal concentrations show regional distribution patterns of high concentrations in the western Sound with low concentrations measured in the eastern Sound and on the shoal complexes that divide the basins within the Sound. These patterns are similar to those of sedimentary environments (see Knebel and Poppe, in press) and current-generated bottom-stress (Signell and others, in press). This similarity reflects the common observation that fine-grained sediments dominate in areas that have high metal concentrations in the sediments and in areas in depositional sedimentary environments. To aid in the evaluation of the metal distributions in the region, samples were categorized by the sedimentary environment in which they are located (Fig. 1; Knebel and others, 1999; in press). The anthropogenic metals, Ag (Fig. 2), Cd (Fig. 3), Cr (Fig. 4), Cu (Fig. 5), Hg (Fig. 6), Pb (Fig. 7), and Zn (Fig. 8), all have concentration values that increase as the sedimentary environment changes erosional to depositional. Al (Fig. 9), Ba (Fig. 10), Fe (Fig. 11), Ni (Fig. 12), Ti (Fig. 13), V (Fig. 14), and Zr (Fig. 15), i.e. those elements that are lithogenic or involved in diagenesis, show patterns similar to one another, but do not show any significant spatial or statistical trend in relation to the sedimentary environments.

Spatial Variability of Anthropogenic Elements

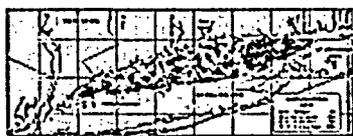


Figure 7. Pb concentrations

Spatial distributions of the anthropogenic elements, Pb (Fig. 7), Cu (Fig. 5), Cr (Fig. 4), Cd (Fig. 3), Ag (Fig. 2), Zn (Fig. 8), and Hg (Fig. 6; Varekamp and others, in press) are very similar in the surface sediments of Long Island Sound. High concentrations of these elements were measured in sediments from the central and western basins of the Sound. The concentrations measured in the eastern Sound were low and generally indistinguishable from natural background. Low concentrations were also measured in the sediments collected from the shoal complexes and the axial depression.

The distribution maps of Pb (Fig. 7) and Cu (Fig. 5) concentrations are grouped into ranges that are defined by the natural background values, which were measured in regional sediment cores (see Tables 2 and 3), and by the potential to induce toxic effects in macrobenthos (Long and others (1996, 1998). Long and others (1996, 1998) use two sediment quality criteria, the Effects Range-Low (ER-L) and the Effects Range-Medium (ER-M), to define the potential for the metal concentrations in sediments to induce toxicity. The concentration ranges outlined in the methods were also used to calculate the degree of metal enrichment (Tables 2 and 3). Pb and Cu are enriched in the whole Sound and have mean concentrations that exceed their ER-Ls. The high anthropogenic metal concentrations in the western and central basins of the Sound generally exceed these sediment criteria, which provide a warning of potential toxic effects in macrobenthos exposed to these sediments.

Throughout the Sound, higher values of the anthropogenic elements were found in samples with a high percentage of fine-grained sediment than in samples in sandier regions (Salomons and Forstner, 1984; Buckley and Cranston, 1991). The high concentrations of metals in the central and western basins, and the absence of enrichment in the sandy eastern Sound and the axial depression, indicate that fine-grained sediments, and the contaminants associated with them, are focused in the basins by current activity. The concentration distributions of these elements follow patterns that correlate with the sedimentary environments (Knebel and Poppe, in press and this CD-ROM), the sediment texture and TOC (Poppe and others, in press and this CD-ROM), and the *Clostridium perfringens* spore concentrations (Buchholtz ten Brink and Mecray, 1998; Buchholtz ten Brink and others, in press). The distributions of Pb (Fig. 7), Cu (Fig. 5) and the other anthropogenic elements (Figures 2-8) are also similar to the patterns of bottom stress generated by the hydrodynamic regime in that areas of high concentration are found in basins where winnowing by current and tidal activity is minimal (Signell and others, in press).

Concentrations are low in the eastern Sound, the axial depression, and the shoal areas despite their proximity to nearshore contaminant sources. These low values correspond with the occurrence of predominantly coarse-grained sediment (Poppe and others, in press and this CD-ROM), non-depositional environments (Knebel and Poppe, in press), and high bottom stress generated by the local hydrodynamic regime (Signell and others, 1998, in press). Low metal concentrations are measured on the shoal complexes and in the axial depression where, when winds blow parallel to the axis of the Sound, the currents are stronger which winnows the fine-grained, metal-bearing particles and deposits them in the relatively quiescent basins (Signell and others, in press).

In addition to the sedimentary environment and the hydrodynamic regime, the distribution of anthropogenic metal concentrations in Long Island Sound surface sediments is also affected by the proximity to contaminant sources. Within the Sound, particulates and associated contaminants exist as suspended material which becomes homogenized and dispersed (Kim and Bokuniewicz, 1991). After solids and associated metals sink towards the bottom, they are transported by the net westerly bottom flow (Signell and others, in press), which traps the sediments in the western end. Anthropogenic sources of metals are greater in the central and western regions than elsewhere in the Sound (Grieg and others, 1977; Strategic Assessments Branch, 1990; Breault and Harris, 1997; Buchholtz ten Brink and others, in press and this CD-ROM). Metal loadings in the western Sound originate from direct discharge and industrial effluent from the largest metropolitan region of the United States (Farrow and others, 1986; Benoit and others, 1999). The combined effects of population density in the western Sound, point and non-point sources in the region, the general transport of contaminant-bearing fine-grained material, and the location of depositional areas yield the contaminant concentration patterns that are observed in the Sound.

Spatial Variability of Major Elements

The elements, Al (Fig. 9), Ba (Fig. 10), Ca (Fig. 16), Fe (Fig. 11), Mn (Fig. 17), Ti (Fig. 13), and V (Fig. 14) are associated with source rocks, fine-grained mineral structure, and biogenic sediments. High concentrations of Mn (Fig. 17) and Fe (Fig. 11) were found in the western basin of the Sound; however, elevated levels were also measured east of the Connecticut River mouth where strong bottom currents dominate and the sediments are predominantly sandy. The spatial distributions observed for these elements in Long Island Sound are more variable than the anthropogenic metal distributions with regard to sedimentary environments, hydrodynamic regime, and source proximity.

Four samples from the scoured, eastern Sound had concentrations of Mn, Fe, V, Ti, and Zn, that were more than ten times greater than elsewhere in the Sound. An explanation for the elevated concentrations of these elements in the easternmost Sound may be the presence of local sources or lag deposits containing these elements. Major elements, particularly Fe and Mn, are found in steel manufacturing (Farrow and others, 1986). Zn is used in the metal industry and also as a component of rubber tires (Nriagu, 1980). Both point and non-point sources are likely to affect the distribution of these elements over a broad area of the Sound. Diagenetic metals, such as Fe, Mn, Ti, and V, may accumulate in the sediments as oxide crusts formed in the presence of high water volumes flowing across the surface sediments in the eastern Sound. Such oxide crusts may be a recent addition to the Sound, or they may be the result of a relict lag deposit (Mecray and Buchholtz ten Brink, in press).

Regional Management Issues

This study has found that establishing a representative suite of samples in a study area, assessing the level of metal enrichment, and grouping the elements by chemical behaviors may serve as a useful tool for those involved in management of our natural resources. The geochemical data from Long Island Sound reveal patchy contaminant distributions that range from background values to potentially toxic levels (Long and others, 1998). The distribution of the elements in the Sound demonstrates that the variance in the metal concentrations is low within each of the sedimentary environments. It is possible, therefore, to obtain representative samples at strategic locations to assess the spatial geochemistry of the Sound using knowledge of the sedimentary and physical oceanographic regimes. As such, a regional assessment of sediment contamination can be made by: (1) taking fewer samples in each sedimentary regime; (2) determining elevations in metal concentrations relative to the potential for toxicity; and (3) identifying elements that co-vary and using them as proxy elements. The benefits of such a regional, and multi-disciplinary, assessment would be: (1) reducing the total cost of the study without losing scientific value; (2) improving the accuracy of system predictions used for environmental management decisions; and (3) understanding the scales of contaminant variability and sediment processes to target critical management factors.

SUMMARY

This report presents data, maps, and tables of metals in Long Island Sound surface samples collected in 1996-1997. This information is used to: (1) establish a regional baseline for temporal studies of contamination in Long Island Sound; (2) identify areas of high contamination in the Sound; (3) elucidate transport and deposition processes affecting contaminant distributions; and (4) generate predictive and assessment tools for regional management in Long Island Sound and elsewhere (Mecray and Buchholtz ten Brink, in press).

The regional patterns of metal concentrations in Long Island Sound result primarily from variations in bottom energy that result in differing grain sizes and particle lithology for the sedimentary environments. The anthropogenic metals are widely dispersed in Long Island Sound and are subject to redistribution in some regions. Their distributions co-vary with the texture, the sedimentary environments, and the lithology of the sequestering material (Fe, Al, or TOC) measured in the sediments.

ACKNOWLEDGMENTS

A great number of people from the U.S. Geological Survey in Woods Hole as well as our collaborators at Boston University, Wesleyan University, and the Woods Hole Oceanographic Institution have made this work possible. The authors wish to thank the field team from the 1996 sampling season, especially Peter Gill, Rick Rendigs, Dann Blackwood, Michael Casso, Bart Krueken, and the crew of the *R/V Seaward Explorer*. The samples were processed through the lab very efficiently thanks to Michael Casso, Emily Banks, Katie Meigs, Shannon McDaniel, Erin Galvin, and Brian Flynn. Metals analyses were conducted by Adila Jamil, Eric Grunwald, and Lauren Vigliotti, under the guidance of Rick Murray at Boston University. Brenton Taylor performed the texture analysis in the sediment laboratory of the USGS Woods Hole Field Center. We thank Erin Galvin for her attention to detail and her constant help in the final phases of this work. The authors greatly benefited from discussions with Larry Poppe, Rich Signell, and Harley Knebel on Long Island Sound topics. Funding for this project came from the U.S. Geological Survey's Coastal and Marine Geology Program in cooperation with the State of Connecticut Department of Environmental Protection.

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FIGURE CAPTIONS

Figure 1. Locations of surface sediment samples taken on three cruises to Long Island Sound aboard the *R/V Seaward Explorer*, the *R/V John Dempsey*, and the *R/V Connecticut*. Stations were selected to representatively sample the different sedimentary environments, sediment types, and proximity to potential metals sources in the Sound. Sedimentary environments from Knebel and others (1999) have been superimposed.

Figure 2. Silver (Ag) concentrations ($\mu\text{g/g}$ dry sediment) in the surface sediments (0-2 cm) of Long Island Sound. The upper divisions on the legend are defined by the Effects Range-Low (ER-L) and Effects Range-Medium (ER-M) sediment quality guidelines of Long and others (1996, 1998). The low range is defined by the natural background values observed in sediment cores from western Long Island Sound. Purple dots represent concentrations measured above the ER-M for Ag in sediments. Red dots represent samples above the ER-L, but below the ER-M. Yellow dots are concentrations below the ER-L, but above the natural background concentrations. The green circles are concentrations that are below naturally-occurring values for Ag, as measured in fine-grained, regional sediment cores (see Table 2 and 3).

Figure 3. Cadmium (Cd) concentrations ($\mu\text{g/g}$ dry sediment) in the surface sediments (0-2 cm) of Long Island Sound. Concentration ranges used on the map are determined by the sediment quality guidelines of Long and others (1996, 1998) for Cd. Natural background values for Cd are presented in Table 2. The distributions of silver (Ag; Fig. 2), chromium (Cr; Fig. 4), copper (Cu; Fig. 5), and mercury (Hg; Fig. 6) are similar to this Cd distribution.

Figure 4. Chromium (Cr) concentrations ($\mu\text{g/g}$ dry sediment) in the surface sediments (0-2 cm) of Long Island Sound. Concentration ranges used on the map are determined by the sediment quality guidelines of Long and others (1996, 1998) for Cr. Natural background values for Cr are presented in Table 2.

Figure 5. Copper (Cu) concentrations ($\mu\text{g/g}$ dry sediment) in the surface sediments (0-2 cm) of Long Island Sound. Concentration ranges used on the map are determined by the sediment quality guidelines of Long and others (1996, 1998) for Cu. Natural background values for Cu are presented in Table 2.

Figure 6. Mercury (Hg) concentrations (ppb dry sediment) in the surface sediments (0-2 cm) of Long Island Sound. Concentration ranges used on the map are determined by the sediment quality guidelines of Long and others (1996, 1998) for Hg. Natural background values for Hg are presented in Table 2.

Figure 7. Lead (Pb) concentrations ($\mu\text{g/g}$ dry sediment) in the surface sediments (0-2 cm) of Long Island Sound. Concentration ranges used on the map are determined by the sediment quality guidelines of Long and others (1996, 1998) for Pb. Natural background values for Pb are presented in Table 2.

Figure 8. Zinc (Zn) concentrations ($\mu\text{g/g}$ dry sediment) in the surface sediments (0-2 cm) of Long Island Sound. Concentration ranges used on the map are determined by the sediment quality guidelines of Long and others (1996, 1998) for Zn. Natural background values for Zn are presented in Table 2.

Figure 9. Aluminum (Al) concentrations (weight %) in the surface sediments (0-2 cm) of Long Island Sound. Blue dots represent samples below the natural background value measured in sediment cores taken from Long Island Sound. All other concentration ranges were determined by an equal spread in the values.

Figure 10. Barium (Ba) concentrations (weight %) in the surface sediments (0-2 cm) of Long Island Sound. Concentration ranges used on the map are determined by the same criteria as in Figure 9.

Figure 11. Iron (Fe) concentrations (weight %) in the surface sediments (0-2 cm) of Long Island Sound. Concentration ranges used on the map are determined by the same criteria as in Figure 9. The elevated concentrations in the eastern Sound are samples that contain oxide coatings.

Figure 12. Nickel (Ni) concentrations ($\mu\text{g/g}$ dry sediment) in the surface sediments (0-2 cm) of Long Island Sound. The upper concentration ranges used on the map are determined by the sediment quality guidelines of Long and others (1996, 1998) for Ni. The lower concentration ranges are determined by an equal spread in the values since the ER-L exceeded the natural background values found in Long Island Sound.

Figure 13. Titanium (Ti) concentrations (weight %) in the surface sediments (0-2 cm) of Long Island Sound. Concentration ranges used on the map are determined by the same criteria as in Figure 9.

Figure 14. Vanadium (V) concentrations ($\mu\text{g/g}$ dry sediment) in the surface sediments (0-2 cm) of Long Island Sound. Concentration ranges used on the map are determined by the same criteria as in Figure 9.

Figure 15. Zirconium (Zr) concentrations ($\mu\text{g/g}$ dry sediment) in the surface sediments (0-2 cm) of Long Island Sound. Concentration ranges used on the map are determined by the same criteria as in Figure 9.

Figure 16. Calcium (Ca) concentrations (weight %) in the surface sediments (0-2 cm) of Long Island Sound. Concentration ranges used on the map are determined by the same criteria as in Figure 9.

Figure 17. Manganese (Mn) concentrations ($\mu\text{g/g}$ dry sediment) in the surface sediments (0-2 cm) of Long Island Sound. Concentration ranges used on the map are determined by the same criteria as in Figure 9.

TABLE CAPTIONS

Table 1. A summary of the surface sample locations, metal concentrations, texture analyses, and *Clostridium perfringens* spores counted for 219 surface samples from Long Island Sound. A copy of this data table (chemtab1.xls) may also be downloaded in Microsoft Excel format. The data dictionary is also available in HTML, text file, and Microsoft WORD document.

Table 2. Summary statistics for the surface samples collected around Long Island Sound from 1996-1999. A copy of this table (chemtab2.xls) may also be downloaded in Microsoft Excel format.

Table 3. Summary statistics grouped by sedimentary environment (Knebel and Poppe, in press) for the surface samples collected by the U.S. Geological Survey in Long Island Sound. A copy of this table (chemtab3.xls) may also be downloaded in Microsoft Excel format.

¹ U.S. Geological Survey, Coastal and Marine Geology Team, Woods Hole, MA 02543