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Distribution and transport of sedimentary trace metals in the tidal portions of the Kennebec/Androscoggin River system, Maine, USA

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ABSTRACT

Previous investigations suggest that contaminant transport from the large Kennebec/Androscoggin watershed is an important large-scale process in mid-coast Maine. To investigate this phenomenon, we determined the concentrations of Cd, Cr, Cu, Ni, Pb, Sn and Zn in the surface sediments of 47 stations in the tidal Kennebec/Androscoggin system. Most stations exhibited elevated metal concentrations. Highest levels were found in the main stem of the system. Distribution patterns lead to the conclusion that metals enter the system from the watershed and are transported to the nearshore Gulf of Maine. The coarse-grained, ebb tide dominated flow prevents the accumulation of contaminants in the estuary. This supports the hypothesis of Larsen and Gaudette (1995) that the Kennebec and Androscoggin watersheds are sources for contaminants observed in the nearshore Gulf of Maine.

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1. Introduction

Several authors have documented elevated levels of toxic contaminants in the water, sediments and biota of the Gulf of Maine (Armstrong et al., 1976; Mayer and Fink, 1980; Lyons et al., 1978; Goldberg et al., 1983; Larsen et al., 1983a,b,1984; Ray and MacKnight, 1984; Gottholm and Turgeon, 1991, Larsen and Gaudette, 1995; Larsen et al., 1997; Getchell, 2002; others). A review of this material suggests that the patterns of contaminant distribution in the area between Cape Elizabeth and Boothbay, also known as mid-coast Maine, is particularly complex and interesting (Larsen, 1992). Larsen and Gaudette (1995) undertook, in 1991, a broad scale surficial sediment sampling and analysis program in this region. Their results suggested that an important large-scale process in the mid-coast region was the removal of contaminants from the large (27,700 km²), industrialized Kennebec/Androscoggin River watershed and their passage through the tidal reaches of the system, including the energetic and ecologically important Merrymeeting Bay, into the nearshore Gulf of Maine (Fig. 1). Merrymeeting Bay is significant because it represents the inland delta of the Kennebec and Androscoggin Rivers and is noted as the habitat of many threatened or endangered plants and animals. It is the largest tidal freshwater system north of Chesapeake Bay and it has an equally long history of human disturbance (Lichter et al., 2006).

The lower Kennebec River, the estuarine portion of the Kennebec/Androscoggin tidal system, connecting the predominantly freshwater Merrymeeting Bay (Larsen and Doggett, 1979) with the Gulf of Maine, does not fit the traditionally held model of estuaries as sediment traps (FitzGerald et al., 2005). This system is a narrow, shallow bedrock-lined conduit influenced by a strong freshwater flow and a mesoscale tidal range. The freshwater flow represents the second largest input of river water to the Gulf of Maine. During high flow periods the entire system is strongly ebb-current dominated (Fenster et al., 2001) but even at lower flows bottom ebb currents generally exceed bottom maximum flood currents (FitzGerald et al., 2005). The result is the net seaward transport of sediments as bedload that supply sand to the nearshore and the barrier-beach systems surrounding the estuary's mouth (Fenster et al., 2001). At times of low to moderate flows, a turbidity maximum with low suspended particle concentrations is formed in the upper estuary although all particulates are resuspended and exported to the Gulf of Maine during higher flows (Kistner and Pettigrew, 2001).

The present study is focussed on the tidal Kennebec/Androscoggin system with the aim of determining the distribution of sedimentary trace metals within the system and their passage through it.

2. Methods

Forty-seven stations (Fig. 1) were sampled in the summer of 1992 between Hallowell, ME (52 km inland) and the lower

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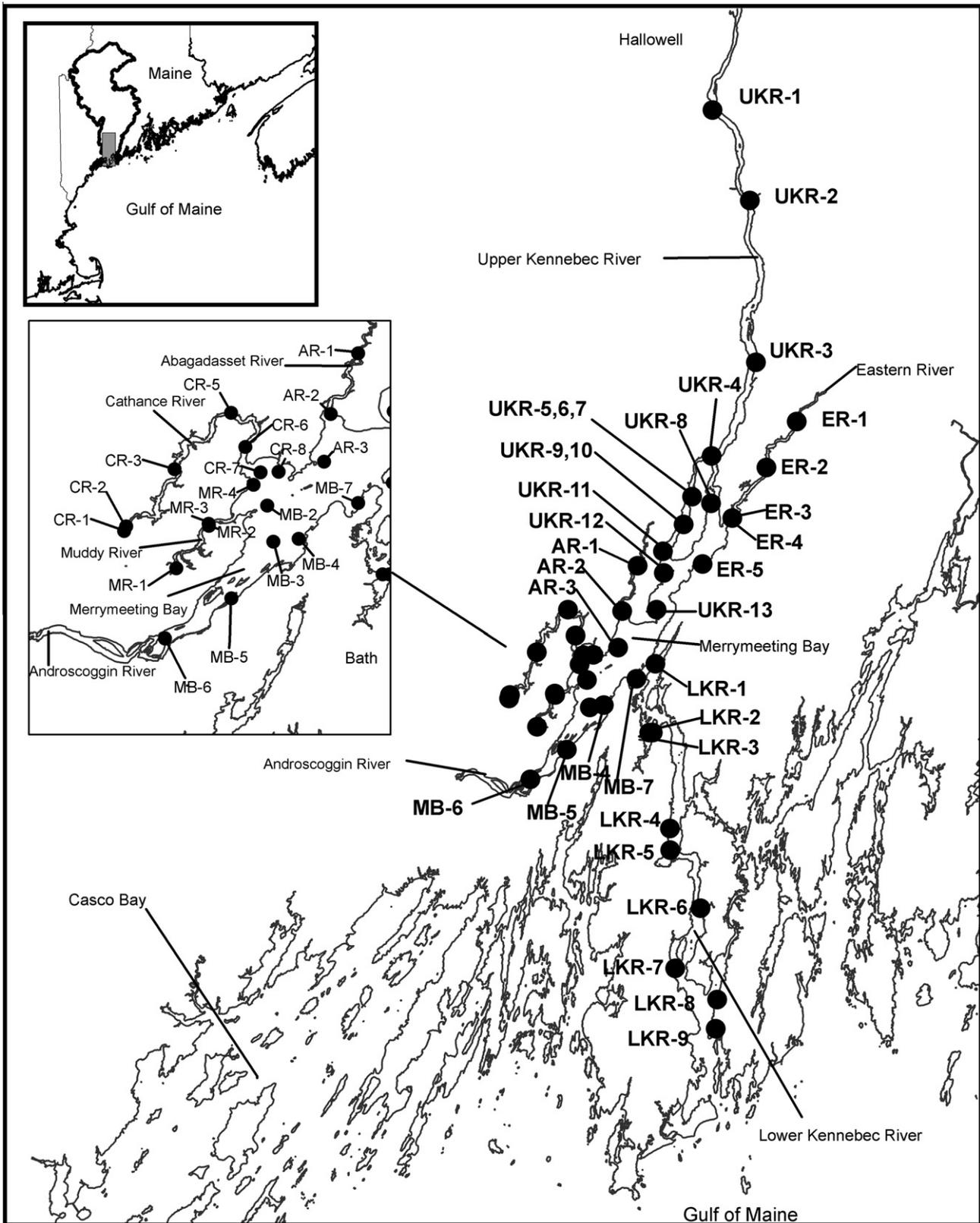


Fig. 1. Location of the Kennebec/Androscoquin River system and the stations occupied in the tidal reaches.

Kennebec River estuary. With the exception of the lower Kennebec estuary, this system may be characterized as tidal fresh water. Station abbreviations and distribution of stations within river segments is as follows: Lower Kennebec River (LKR)(9 sta-

tions), Merrymeeting Bay (includes lower Androscoquin River) (MB)(6), Upper Kennebec River (UKR)(13), Muddy River (MR)(4), Cathance River (CR)(7), Abagadasset River (AR)(3) and Eastern River (ER)(5). The lower Androscoquin River is included

Table 1Normalized concentrations of metals (ppm dry wt.) in surface sediments with percentage of sediment <63 μm and weight loss on ignition.

| River | Station # | Cd (%) | Cr (%) | Cu (%) | Pb (%) | Zn (%) | Sn (%) | Ni (%) | Fe (%) | Mn (%) | %<63 μm | % LOI |
|----------------------|----------------------|--------|--------|--------|--------|--------|--------|---------|---------|---------|--------------------|-------|
| Muddy River | MR-1 | 0.74 | 57.6 | 29.7 | 29.8 | 119.4 | 13.4 | 30.1 | 12150.9 | 125.7 | 46.7 | 10.1 |
| | MR-2 | 0.65 | 58.3 | 31.4 | 28.8 | 128.7 | 10.5 | 32.9 | 14514.5 | 143.9 | 49.4 | 5.8 |
| | MR-3 | 0.43 | 37.7 | 20.3 | 14.4 | 88.2 | 10.7 | 24.8 | 8864.0 | 78.8 | 72.8 | 7.0 |
| | MR-4 | 0.75 | 49.9 | 28.9 | 25.7 | 128.7 | 8.7 | 26.2 | 10637.3 | 122.8 | 77.2 | 8.9 |
| Cathance River | CR-1 | 0.20 | 60.2 | 31.9 | 26.3 | 144.6 | 16.0 | 41.2 | 22346.9 | 229.9 | 17.2 | 4.0 |
| | CR-2 | 0.20 | 42.6 | 23.6 | 16.4 | 100.7 | 14.0 | 33.6 | 13969.1 | 186.3 | 47.6 | 4.3 |
| | CR-3 | 0.51 | 47.5 | 22.4 | 20.1 | 96.4 | 11.1 | 24.2 | 8405.3 | 94.1 | 77.8 | 7.1 |
| | CR-5 | 0.79 | 46.5 | 28.6 | 26.7 | 121.9 | 14.9 | 26.8 | 11565.3 | 117.9 | 75.4 | 9.1 |
| | CR-6 | 0.37 | 45.3 | 24.7 | 22.3 | 101.6 | 7.5 | 31.1 | 11044.9 | 102.6 | 60.8 | 4.2 |
| | CR-7 | 0.86 | 50.7 | 29.1 | 24.6 | 144.0 | 6.7 | 29.8 | 16537.0 | 127.2 | 33.6 | 2.5 |
| | CR-8 | 0.33 | 25.6 | 13.5 | 9.5 | 64.0 | 6.1 | 18.8 | 6053.2 | 37.8 | 48.1 | 1.7 |
| | Abagadasset River | AR-1 | 0.58 | 72.6 | 29.6 | 21.0 | 127.6 | 15.3 | 39.9 | 16647.0 | 104.1 | 39.3 |
| AR-2 | 0.59 | 57.8 | 30.6 | 25.5 | 121.0 | 16.3 | 32.9 | 12839.7 | 127.6 | 66.8 | 7.8 | |
| AR-3 | 0.42 | 47.0 | 26.6 | 24.3 | 115.3 | 9.3 | 26.3 | 9584.5 | 101.3 | 79.9 | 6.5 | |
| Eastern River | ER-1 | 0.24 | 40.2 | 19.2 | 15.8 | 97.3 | 13.1 | 29.5 | 13813.2 | 135.4 | 49.8 | 3.4 |
| | ER-2 | 0.42 | 42.1 | 21.2 | 21.2 | 94.8 | 12.8 | 31.5 | 12166.3 | 144.4 | 71.3 | 5.9 |
| | ER-3 | 0.37 | 40.4 | 19.8 | 21.2 | 91.2 | 13.9 | 30.1 | 11956.2 | 137.3 | 71.6 | 4.9 |
| | ER-4 | 0.47 | 48.1 | 24.8 | 21.8 | 107.3 | 11.7 | 33.7 | 13158.4 | 157.5 | 66.2 | 6.6 |
| | ER-5 | 0.48 | 45.1 | 22.6 | 23.0 | 93.2 | 16.2 | 31.7 | 12418.4 | 116.3 | 54.1 | 4.0 |
| Upper Kennebec River | UKR-1 | 0.98 | 84.6 | 58.7 | 111.2 | 185.8 | 28.8 | 66.7 | 24110.3 | 257.7 | 33.7 | 3.4 |
| | UKR-2 | 0.53 | 175.1 | 78.3 | 80.5 | 400.5 | 34.6 | 145.0 | 48967.9 | 657.7 | 12.8 | 5.3 |
| | UKR-3 | 0.62 | 90.6 | 41.5 | 19.9 | 198.8 | 17.8 | 78.8 | 27029.0 | 366.2 | 22.5 | 4.9 |
| | UKR-4 | 0.96 | 102.5 | 49.9 | 284.7 | 248.5 | 36.4 | 79.4 | 37214.5 | 458.0 | 20.1 | 2.3 |
| | UKR-5 | 0.40 | 49.9 | 27.4 | 25.4 | 113.8 | 9.0 | 35.0 | 11597.9 | 118.1 | 48.6 | 4.8 |
| | UKR-6 | 0.65 | 50.5 | 27.5 | 24.6 | 39.8 | 11.6 | 31.8 | 11764.1 | 145.7 | 59.5 | 4.0 |
| | UKR-7 | 0.48 | 46.6 | 26.1 | 27.3 | 102.3 | 10.8 | 29.3 | 10023.0 | 105.4 | 59.5 | 4.4 |
| | UKR-8 | 1.82 | 218.5 | 98.4 | 94.3 | 474.6 | 92.1 | 184.2 | 72132.0 | 583.3 | 8.9 | 2.8 |
| | UKR-9 | 0.64 | 73.2 | 40.9 | 46.6 | 172.9 | 20.5 | 52.7 | 21052.9 | 244.8 | 41.2 | 3.4 |
| | UKR-10 | 0.62 | 66.6 | 35.1 | 38.8 | 154.4 | 21.2 | 45.7 | 20530.5 | 201.2 | 39.9 | 3.1 |
| | UKR-11 | 0.19 | 44.2 | 23.4 | 29.7 | 87.0 | 18.8 | 33.3 | 14248.6 | 134.5 | 59.9 | 2.5 |
| | UKR-12 | 0.21 | 30.8 | 15.7 | 10.0 | 56.1 | 7.6 | 23.3 | 10802.2 | 76.0 | 61.4 | 2.9 |
| | UKR-13 | 0.67 | 63.7 | 32.7 | 32.3 | 155.3 | 22.8 | 39.5 | 17387.0 | 153.3 | 37.8 | 3.9 |
| Merrymeeting Bay | MB-2 | 0.76 | 60.3 | 31.6 | 34.2 | 142.3 | 13.4 | 34.4 | 12811.6 | 136.8 | 33.6 | 3.4 |
| | MB-3 | 1.13 | 145.1 | 71.1 | 61.2 | 440.5 | 31.0 | 89.3 | 46427.9 | 406.1 | 11.5 | 2.5 |
| | MB-4 | 1.13 | 85.6 | 46.8 | 40.5 | 256.9 | 19.4 | 53.3 | 23039.4 | 224.6 | 21.1 | 3.5 |
| | MB-5 | 1.31 | 106.0 | 64.4 | 66.4 | 343.7 | 34.9 | 58.8 | 32214.5 | 159.7 | 17.8 | 3.4 |
| | MB-6 | 1.26 | 108.6 | 64.0 | 67.9 | 320.2 | 34.6 | 73.8 | 27869.9 | 344.9 | 13.3 | 1.8 |
| | MB-7 | 0.59 | 53.6 | 27.0 | 27.0 | 132.4 | 9.2 | 30.2 | 13454.7 | 81.8 | 38.0 | 3.0 |
| | Lower Kennebec River | LKR-1 | 1.04 | 97.4 | 51.3 | 40.9 | 236.9 | 34.5 | 69.1 | 32386.1 | 217.8 | 19.7 |
| LKR-2 | 0.99 | 74.6 | 45.1 | 39.6 | 209.5 | 27.3 | 50.9 | 23333.9 | 170.8 | 31.9 | 3.8 | |
| LKR-3 | 0.67 | 90.4 | 48.9 | 35.4 | 215.2 | 30.0 | 64.7 | 28630.4 | 242.2 | 22.5 | 2.9 | |
| LKR-4 | 1.24 | 121.1 | 69.8 | 46.2 | 276.8 | 41.3 | 95.4 | 39313.1 | 176.9 | 12.7 | 2.0 | |
| LKR-5 | 0.51 | 59.2 | 33.5 | 31.6 | 126.6 | 11.9 | 37.5 | 14437.1 | 88.4 | 40.6 | 5.0 | |
| LKR-6 | 0.89 | 88.4 | 55.7 | 57.2 | 179.5 | 26.9 | 60.1 | 26261.3 | 138.6 | 19.5 | 3.1 | |
| LKR-7 | 0.54 | 104.7 | 42.6 | 44.9 | 140.6 | 32.3 | 52.0 | 21963.6 | 115.8 | 30.7 | 4.9 | |
| LKR-8 | 0.66 | 56.0 | 29.4 | 18.3 | 116.6 | 20.4 | 33.7 | 14924.3 | 63.7 | 49.4 | 4.5 | |
| LKR-9 | 0.82 | 86.5 | 45.2 | 37.3 | 180.9 | 31.9 | 55.9 | 24452.2 | 118.8 | 29.5 | 4.2 | |
| | Mean | 0.68 | 73.2 | 38.4 | 41.7 | 170.4 | 21.3 | 50.3 | 20950.8 | 188.6 | 40.9 | 4.1 |
| | Min | 0.19 | 25.6 | 13.5 | 9.5 | 39.8 | 6.1 | 18.8 | 6053.2 | 37.8 | 8.9 | 1.7 |
| | Max | 1.82 | 218.5 | 98.4 | 284.7 | 474.6 | 92.1 | 184.2 | 72132.0 | 657.7 | 79.9 | 9.1 |
| | SD | 0.37 | 41.4 | 19.6 | 49.0 | 109.7 | 15.5 | 34.8 | 13783.8 | 144.0 | 21.4 | 1.7 |

as part of Merrymeeting Bay because no natural demarcation between them is evident. Fine sediments were sampled in the above areas and analyzed for seven trace metals (Cd, Cr, Cu, Pb, Zn, Sn and Ni) as well as major metals, grain size and organic carbon content.

Samples were obtained using a small, acid-cleaned stainless steel grab sampler of our own design (HEG). Undisturbed, surface sediment sub-samples (top 5 cm) for trace metal analysis were taken from the grab with acid-cleaned plastic scoops, transferred to clean polyethylene zip-lock bags and stored on ice for return to the laboratory. Separate sub-samples were taken for grain size analysis and organic matter determination. Emphasis was placed on finding fine-grained sediments that are uncommon in this hydrodynamically vigorous system.

Grain size distributions were determined by standard sieve and pipette methods (Folk, 1968). Organic matter in the sediments is expressed as percent weight loss on ignition obtained by heating a representative, dried subsample of the sediment to 540 °C for 24 h.

Trace metals were stripped off the sediment particle surfaces using the same strong acid leach process as Larsen et al. (1983a). In brief, approximately 3 g of dried sediment (60 °C, 18–24 h) were accurately weighed into a 100 ml glass beaker. Ten ml of concentrated reagent HNO₃ were added, and the samples evaporated to dryness. When cooled, each sample received 5 ml of 8% NH₄Cl (w/v), 5 ml of 0.02 M Ca(NO₃)₂ · 4H₂O, and 15 ml of an acid solution (80 ml concentrated HNO₃ plus 20 ml concentrated HCl diluted to 1 l with MilliQ water), and the volumes were reduced on a hot plate to 10–15 ml. Cooled samples

Table 2
Correlation matrix of Kennebec/Androscoggin data set.

| | Cd | Cr | Cu | Pb | Zn | Sn | Ni | Mn | Fe | % Fines | LOI |
|---------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----|
| Cd | XXX | | | | | | | | | | |
| Cr | 0.7456*** | XXX | | | | | | | | | |
| Cu | 0.8289*** | 0.9303*** | XXX | | | | | | | | |
| Pb | 0.3281* | 0.3746** | 0.4389*** | XXX | | | | | | | |
| Zn | 0.7373*** | 0.7844*** | 0.8297*** | 0.3562* | XXX | | | | | | |
| Sn | 0.4450** | 0.6611*** | 0.6266*** | 0.3970** | 0.4942*** | XXX | | | | | |
| Ni | 0.4917*** | 0.8374*** | 0.7982*** | 0.4386** | 0.6432*** | 0.7069*** | XXX | | | | |
| Mn | 0.3760** | 0.6045*** | 0.5921*** | 0.5019*** | 0.5490*** | 0.4576** | 0.8074*** | XXX | | | |
| Fe | 0.4874*** | 0.7629*** | 0.7367*** | 0.4738*** | 0.6473*** | 0.7904*** | 0.8843*** | 0.7512*** | XXX | | |
| % Fines | 0.6225*** | 0.7469*** | 0.7225*** | 0.4338** | 0.6036*** | 0.4241** | 0.6406*** | 0.5118*** | 0.5400*** | XXX | |
| LOI | 0.7491*** | 0.7923*** | 0.7740*** | 0.4766*** | 0.6868*** | 0.4564** | 0.5450*** | 0.4720*** | 0.5117*** | 0.7114*** | XXX |

Note: n = 47; Pb vs. fines and LOI correlations were not significant at n = 47, removal of one outlier resulted in significant correlations.

- * Significant.
- ** Very significant.
- *** Extremely significant.

were filtered using “Q” water; sediment trapped on the filter paper was washed several times with “Q” water, and the filtrate was brought to 50 ml total volume. These procedures have been shown to remove “environmentally available” metals without destruction of the mineral matrix (Tessler et al., 1979; Olsen et al., 1993).

The filtrates were analyzed by Atomic Absorption Spectrometry (AA) for Fe, Mn, Cd, Cr, Cu, Ni, Pb, Sn, and Zn, and concentrations as µg/gram dry weight sediment were calculated. Analytical variability could not be determined by replicate analysis of stan-

dard sediment samples (US Geological Survey standard MAG-1 (Marine Mud) and National Institute of Standards and Technology SRM 1646 (estuarine mud)) since our extraction procedure differed from the total dissolution procedures used to determine the certified values. Therefore, we have made within sample replicate analyses to estimate precision. These are: Cd 13.4%; Cr 4.4%; Cu 1.8%; Pb 4.8%; Zn 2.1%; Sn 20.9%; Ni 2.4%; Fe 5.9%; and Mn 1.3%. These uncertainty values are typical of AA analyses with the exception of Sn that was influenced by an outlier in the replicated samples.

Table 3
Summary statistics for metals in each of the seven subregions.

| Location | | Cd | Cr | Cu | Pb | Zn | Sn | Ni |
|----------------------|------|------|-------|------|-------|-------|------|-------|
| Muddy River | Mean | 0.64 | 50.9 | 27.6 | 24.7 | 116.2 | 10.8 | 28.5 |
| | Min | 0.43 | 37.7 | 20.3 | 14.4 | 88.2 | 8.7 | 24.8 |
| | Max | 0.75 | 58.3 | 31.4 | 29.8 | 128.7 | 13.4 | 32.9 |
| | SD | 0.15 | 9.6 | 4.9 | 7.1 | 19.2 | 1.9 | 3.7 |
| | SEM | 0.07 | 4.8 | 2.5 | 3.5 | 9.6 | 1.0 | 1.8 |
| Cathance River | Mean | 0.46 | 45.5 | 24.8 | 20.8 | 110.5 | 10.9 | 29.4 |
| | Min | 0.20 | 25.6 | 13.5 | 9.5 | 64.0 | 6.1 | 18.8 |
| | Max | 0.86 | 60.2 | 31.9 | 26.7 | 144.6 | 16.0 | 41.2 |
| | SD | 0.27 | 10.4 | 6.0 | 6.2 | 28.7 | 4.2 | 7.1 |
| | SEM | 0.10 | 3.9 | 2.3 | 2.3 | 10.9 | 1.6 | 2.7 |
| Abagadasset River | Mean | 0.53 | 59.2 | 28.9 | 23.6 | 121.3 | 13.6 | 33.0 |
| | Min | 0.42 | 47.0 | 26.2 | 21.0 | 115.3 | 9.3 | 26.3 |
| | Max | 0.59 | 72.6 | 30.6 | 25.5 | 127.6 | 16.3 | 39.9 |
| | SD | 0.09 | 12.9 | 2.1 | 2.3 | 6.2 | 3.8 | 6.8 |
| | SEM | 0.05 | 7.4 | 1.2 | 1.3 | 3.6 | 2.2 | 3.9 |
| Eastern River | Mean | 0.40 | 43.2 | 21.5 | 20.6 | 96.7 | 13.5 | 31.3 |
| | Min | 0.24 | 40.2 | 19.2 | 15.8 | 91.2 | 11.7 | 29.5 |
| | Max | 0.48 | 48.1 | 24.8 | 23.0 | 107.3 | 16.2 | 33.7 |
| | SD | 0.1 | 3.4 | 2.2 | 2.8 | 6.3 | 1.7 | 1.6 |
| | SEM | 0.04 | 1.5 | 1.0 | 1.3 | 2.8 | 0.7 | 0.7 |
| Upper Kennebec River | Mean | 0.67 | 84.4 | 42.7 | 63.5 | 183.8 | 25.5 | 65.0 |
| | Min | 0.19 | 30.8 | 15.7 | 10.0 | 39.8 | 7.60 | 23.3 |
| | Max | 1.82 | 218.5 | 98.4 | 284.7 | 474.6 | 92.1 | 184.2 |
| | SD | 0.42 | 54.6 | 23.6 | 73.3 | 127.7 | 22.0 | 48.5 |
| | SEM | 0.12 | 15.1 | 6.5 | 20.3 | 35.4 | 6.1 | 13.4 |
| Merrymeeting Bay | Mean | 1.03 | 93.2 | 50.8 | 49.5 | 272.7 | 23.7 | 56.6 |
| | Min | 0.59 | 53.6 | 27.0 | 27.0 | 132.4 | 9.2 | 30.2 |
| | Max | 1.31 | 145.1 | 71.1 | 67.9 | 440.5 | 34.9 | 89.3 |
| | SD | 0.29 | 34.1 | 18.5 | 17.8 | 120.3 | 11.3 | 22.7 |
| | SEM | 0.12 | 13.9 | 7.6 | 7.3 | 49.1 | 4.6 | 9.3 |
| Lower Kennebec River | Mean | 0.82 | 86.5 | 46.8 | 39.0 | 186.9 | 28.5 | 57.7 |
| | Min | 0.51 | 56.0 | 29.4 | 18.3 | 116.6 | 11.9 | 33.7 |
| | Max | 1.24 | 121.1 | 69.8 | 57.2 | 276.8 | 41.3 | 95.4 |
| | SD | 0.24 | 20.9 | 11.9 | 10.7 | 53.3 | 8.50 | 18.3 |
| | SEM | 0.08 | 7.0 | 4.0 | 3.6 | 17.8 | 2.8 | 6.1 |

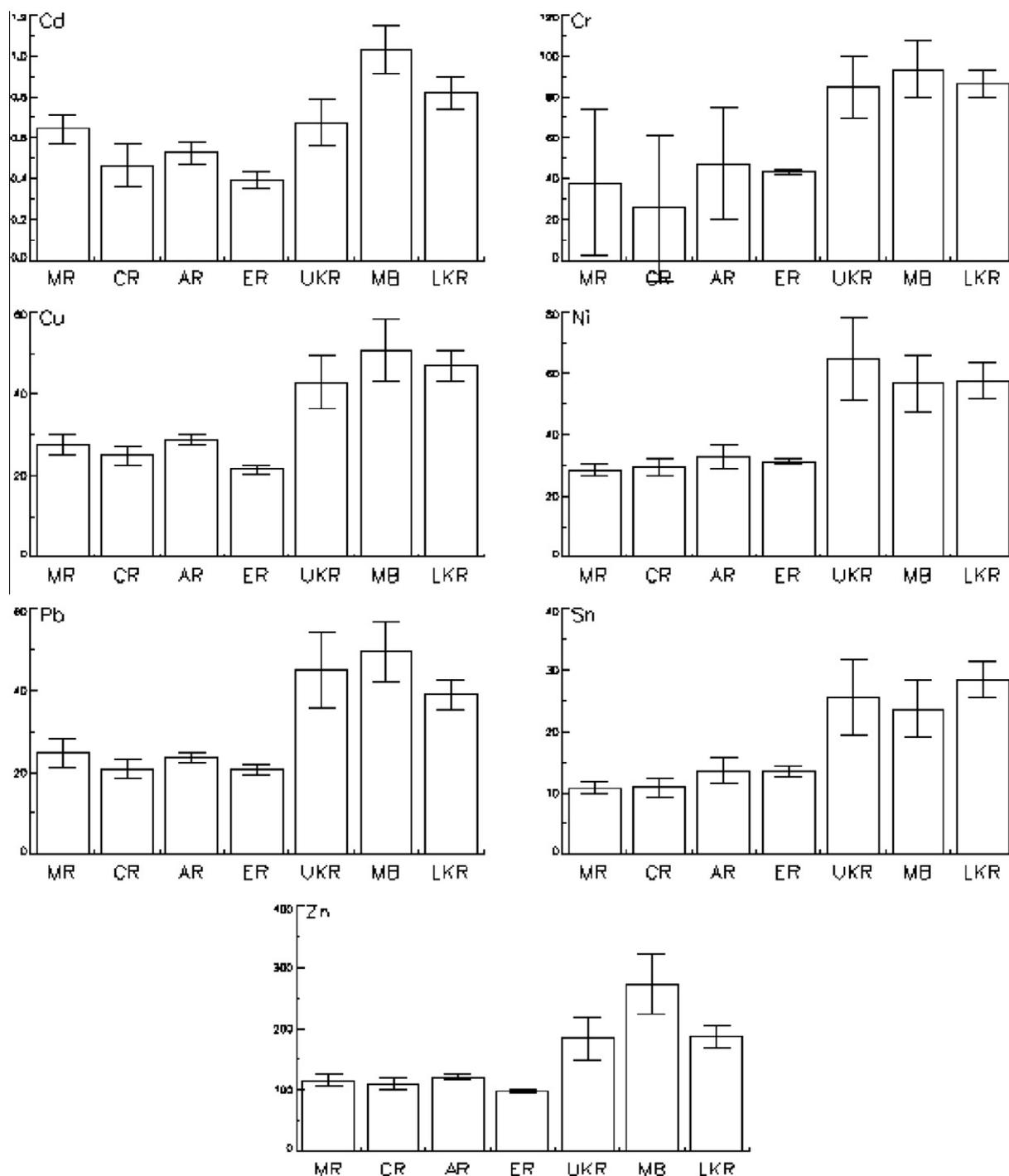


Fig. 2. Mean and SEM of trace metals in the seven defined subregions of the Kennebec/Androscoggin River system.

Except where noted, data were normalized to the fine sediment fraction by dividing the metal concentrations by the fraction of the sediment <63 μm (NOAA, 1988).

3. Results

Results of the sediment metal analyses with the percentages of fine sediments and loss on ignition are presented in Table 1. Examination of the summary statistics in Table 1 demonstrates that the individual metal concentrations were distributed widely

around the means. Nevertheless, only in the case of Pb does the standard deviation exceed the mean. Perusal of the Pb column reveals one very hardy outlier at Station UKR-4 located in the Kennebec River where it enters into Merrymeeting Bay.

A linear correlation matrix, using unnormalized data of trace metals, major metals and salient environmental variables was constructed to gain insight into the relationships among them (Table 2). Nearly all of the correlations between the trace metals, Mn, Fe, percent fines and LOI are extremely significant. Pb correlations are low and not significant with percent fines and LOI at $n = 47$. The

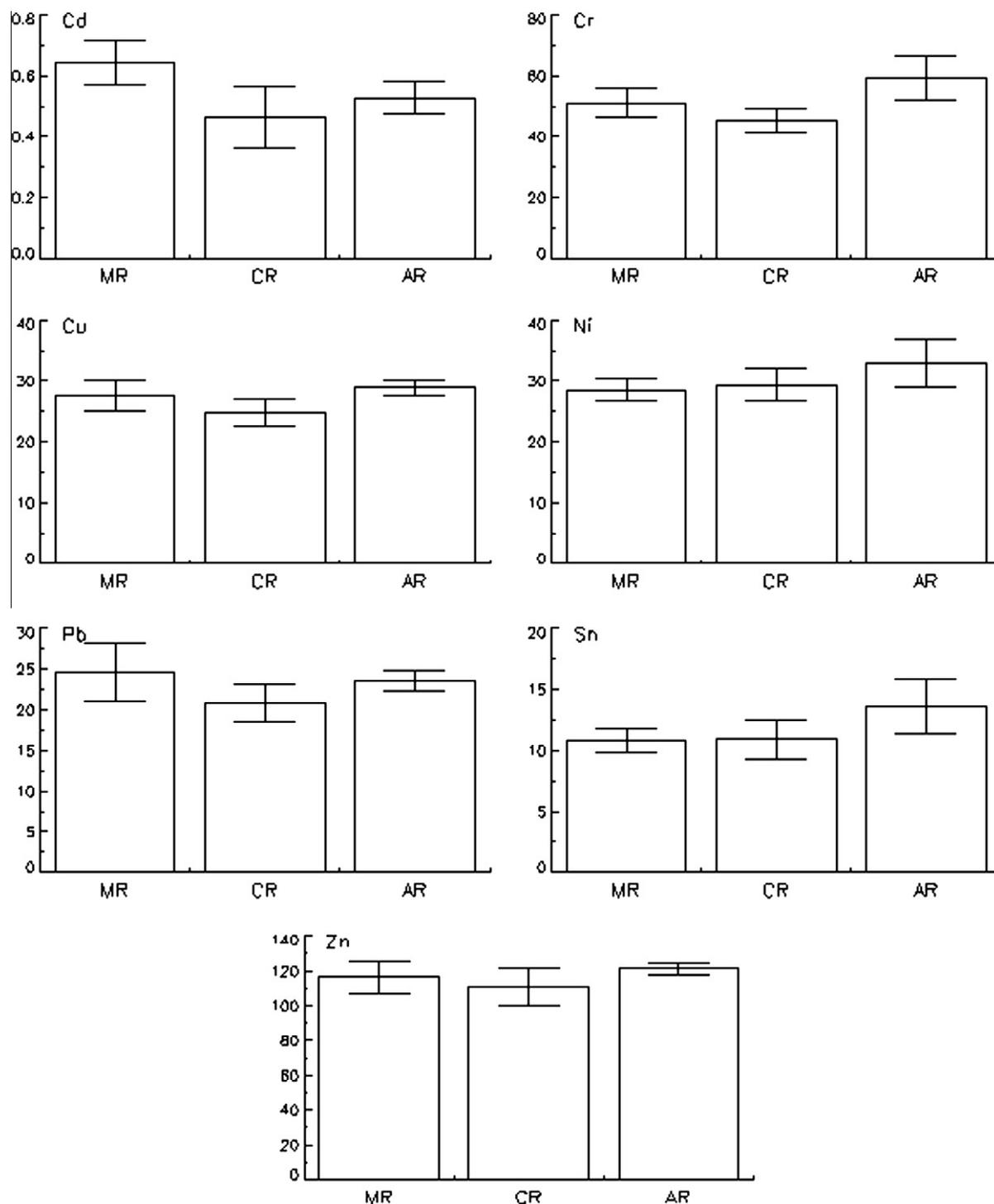


Fig. 3. Mean and standard error of metals in the three western tributaries of Merrymeeting Bay.

removal of the above-mentioned outlier at UKR-4, however, resulted in improved Pb correlations with every variable. With the noted exception of Pb, the correlation matrix indicates that the trace metals are normally distributed in association with the fine-grained and organic particles perhaps mediated by hydrous oxide coatings of Mn and Fe.

Grouping the stations by river segments and examining the summary statistics indicates that there is a clear and consistent geographic pattern exhibited by each of the seven trace metals (Table 3). Trace metal concentrations are higher in the Upper

Kennebec River (UKR), Merrymeeting Bay (MB) and Lower Kennebec River (LKR), the groupings that constitute the main stem of the system. Metal levels are uniformly lower in the four "local" Merrymeeting Bay tributaries, i.e. the Muddy (MR), Cathance (CR), Abagadasset (AR) and Eastern Rivers (ER) (Fig. 2).

An analysis to determine if the apparent differences in metal concentrations are statistically significant cannot be performed at the seven-group level because MR and AR are represented by too few stations. These two small tributaries, together with CR, are

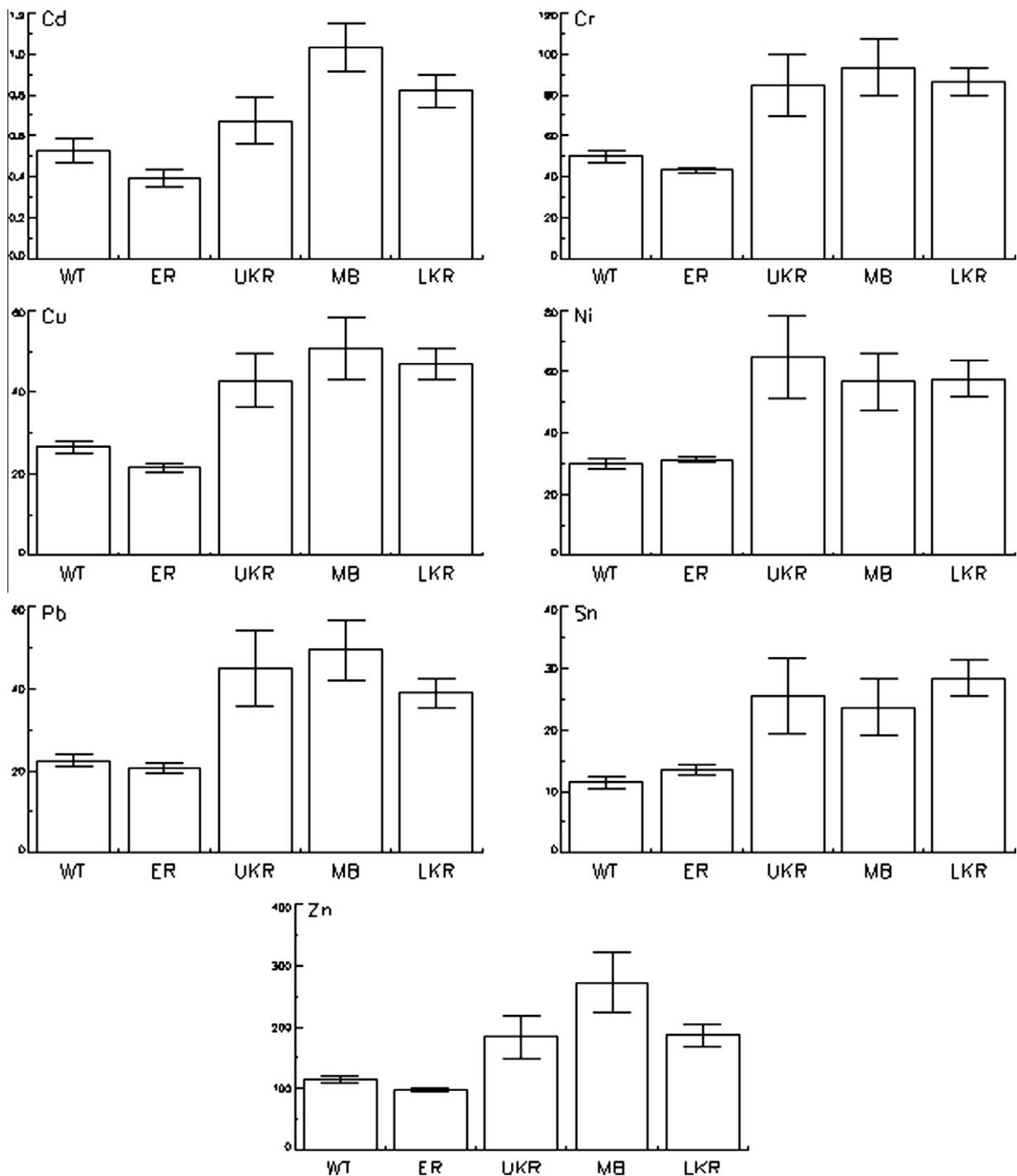


Fig. 4. Mean and standard error of metals in the five defined geographic groupings.

located on the western side of Merrymeeting Bay. They have contiguous watersheds and have especially uniform trace metal concentrations with the standard errors of the means overlapping in each case save one (Cr between CR and AR) (Fig. 3, Table 3). Data from these three tributaries, therefore, can be grouped together to increase the power of the statistical analysis. The new grouping is called Western Tributaries (WT). The means and standard errors of the resulting five groups are plotted in Fig. 4.

A Kruskal–Wallis test, a nonparametric analysis of variance, for each metal across the five geographic groupings of stations

Table 4

The level of significance of differences in levels of the seven metals over the five geographic groups.

| Metal | Significance level |
|-------|-----------------------|
| Cd | Very significant |
| Cr | Extremely significant |
| Cu | Extremely significant |
| Pb | Extremely significant |
| Zn | Extremely significant |
| Sn | Very significant |
| Ni | Extremely significant |

Table 5
Result's of Dunn's multiple comparisons test.

| Metal | Comparison | Significance level |
|-------|------------|--------------------|
| Cd | WT vs. MB | * |
| | ER vs. MB | ** |
| | ER vs. LKR | * |
| Cr | WT vs. MB | * |
| | WT vs. LKR | * |
| | ER vs. MB | ** |
| | ER vs. LKR | ** |
| Cu | WT vs. LKR | * |
| | ER vs. UKR | * |
| | ER vs. MB | ** |
| | ER vs. LKR | ** |
| Pb | WT vs. MB | * |
| | WT vs. LKR | * |
| | ER vs. MB | * |
| | ER vs. LKR | * |
| Zn | WT vs. MB | * |
| | ER vs. MB | ** |
| | ER vs. LKR | * |
| Sn | WT vs. LKR | ** |
| Ni | WT vs. UKR | * |
| | WT vs. MB | ** |
| | WT vs. LKR | ** |

* Significance at the < 0.05 level.

** At the < 0.01 level.

indicates that there are very significant or extremely significant statistical differences between the levels of metals in the groups (Table 4). The nonparametric test is used because parametric analysis of variance assumes identical standard deviations. Bartlett's test suggests that the differences between standard deviations are significant in each case.

The results of Dunn's Multiple Comparisons Tests are presented in Table 5. This test examines the results of the Kruskal–Wallis tests to determine which contrasts between geographic groupings are responsible for the statistically significant results. In each case, the significant differences are between one of the “local” tributaries, WT or ER, and one of the main stem groupings. To look at it another way, there is never a statistically significant difference detected between the “local” tributaries or between the main stem groupings.

A rank score analysis is applied to highlight the distributions of the metals over the entire study area. In this process, the stations are ranked for each metal from the highest concentration to the lowest (1–47) and then the scores are summed across the columns and the stations ranked from highest composite scores (low number) to the lowest (high score) (Table 6). Examination of the table indicates that there is considerable correspondence between the distribution of metals, i.e. a station with a high concentration of one metal is likely to have a high concentration of the other metals. In addition, the stations with the highest metal concentrations tend to be located along the main stem of the system, i.e. the Upper Kennebec River Channel, the western portion of Merrymeeting Bay, where Androscoggin River water enters, and in the Lower Kennebec River. With few exceptions, stations in the Western Tributaries and the Eastern River are in the third or fourth quartiles of stations. The geographic distribution of these rankings by quartile is presented in Fig. 5.

Several important insights are revealed by this summed rank score analysis. The 20 highest ranked stations are located in UKR, MB and LKR (Table 6). Furthermore, the most highly ranked stations among these are found in the UKR above Swans Island, in the confluence of the Androscoggin River and MB, and in the upper

reaches of the LKR between Bath and Merrymeeting Bay (Fig. 5). Stations in the minor tributaries are generally ranked in the third and fourth quartiles. In fact, four of the five ER stations and four of the seven CR stations are in the lowest quartile. Stations from UKR, MB and LKR ranked in the lower two quartiles are located at sheltered sites.

4. Discussion

The results of this study reveal a coherent explanation of the distribution and movement of trace metals into and through the Kennebec/Androscoggin River system. The major points are as follows. Metal levels are generally elevated above pre-industrial levels (Lyons et al., 1978; Larsen et al., 1983a) and above a Gulf of Maine baseline (Getchell, 2002) indicating that metals are presently entering the system. There are statistically significant differences in metal levels between our seven defined subregions that show that the greatest concentration elevations are limited to the main stem of the system, i.e. the Kennebec River and estuary and Merrymeeting Bay that, in our groupings, includes the lower Androscoggin River (Table 4). The four small tidal rivers that enter Merrymeeting Bay, the Muddy, Cathance, Abagadasset and Eastern Rivers, have watersheds limited to the Merrymeeting Bay vicinity and exhibit less elevated metal levels. In the case of Pb, sediment concentrations in these four rivers are actually below the Gulf of Maine baseline (Getchell, 2002). We, therefore, may conclude that the major portion of the observed trace metals is from outside of our immediate study area, i.e. from upstream sources in the Kennebec River and Androscoggin River watersheds.

The conclusion that the Kennebec and Androscoggin watersheds are the principal sources of metals in the system is reinforced by the distribution of the stations that ranked the highest in terms of metal concentrations (Table 6, Fig. 5). For instance, Stations MB -6, MB-5 and MB-3 are situated where the Androscoggin River broadens into Merrymeeting Bay. It is here where the currents would slow and the river would drop part of its suspended load during periods of low flow, i.e. summer. Likewise, highly ranked stations in the upper Kennebec are located where the river first meets the two-way tidal flow below the (former) Edwards Dam upstream of Hallowell (Stations UKR-1 and UKR-2) or where the river first broadens out into upper Merrymeeting Bay (Stations UKR-4 and UKR-8).

Four stations in the upper reach of the lower Kennebec River estuary, sometimes called the Sagadahoc estuary, also were highly ranked (Stations LKR-1, 2, 3 & 4). Whereas we cannot dismiss potential inputs from the population/industrial center of Bath, there is a hydrodynamic explanation why these stations would exhibit higher metal burdens than stations immediately upstream in Merrymeeting Bay. The Kennebec estuary, like other northern, rock-bound mesotidal estuaries, is strongly ebb flow dominated to the point that under high flow conditions bed load transport of sand-sized particles is seaward. Coarse-grained particles, and hence finer materials, are not confined to the estuary but are exported to the Gulf of Maine and are the principal source of the medium and coarse sands of the adjacent coastal barrier systems (FitzGerald et al., 2005). Nevertheless, periods of low or moderate flows, that occur about three-quarters of the time, allow for the formation of a turbidity maximum (Kistner and Pettigrew, 2001). The location of the Kennebec turbidity maximum, during our summer sampling period, is most often in the upper reach where we encountered metal levels higher than at stations both upstream and downstream.

The fact that metals are entering the Kennebec/Androscoggin system from upstream does not mean that they are accumulating

Table 6

Stations inversely ranked by their cumulative rank scores.

| Total rank | Station | Cd rank | Cr rank | Cu rank | Pb rank | Zn rank | Sn rank | Ni rank | Rank sum | Quartile |
|------------|---------|---------|---------|---------|---------|---------|---------|---------|----------|----------|
| 1 | UKR-8 | 1 | 1 | 1 | 3 | 1 | 1 | 1 | 9 | 1 |
| 2 | MB-6 | 3 | 5 | 6 | 5 | 5 | 6 | 3 | 33 | 1 |
| 3 | LKR-4 | 4 | 4 | 4 | 10 | 6 | 2 | 4 | 34 | 1 |
| 4 | MB-5 | 2 | 6 | 5 | 6 | 4 | 4 | 12 | 39 | 1 |
| 5 | MB-3 | 5 | 3 | 3 | 7 | 2 | 10 | 10 | 40 | 1 |
| 6 | UKR-4 | 10 | 8 | 10 | 1 | 8 | 3 | 2 | 42 | 1 |
| 7 | UKR-2 | 30 | 2 | 2 | 4 | 3 | 5 | 7 | 53 | 1 |
| 8 | UKR-1 | 9 | 15 | 7 | 2 | 13 | 12 | 6 | 64 | 1 |
| 9 | LKR-1 | 7 | 9 | 9 | 12 | 9 | 7 | 14 | 67 | 1 |
| 10 | LKR-6 | 11 | 12 | 8 | 8 | 15 | 14 | 13 | 81 | 1 |
| 11 | LKR-2 | 8 | 16 | 14 | 14 | 11 | 13 | 9 | 85 | 1 |
| 12 | LKR-9 | 13 | 13 | 13 | 16 | 14 | 9 | 8 | 86 | 1 |
| 13 | MB-4 | 6 | 14 | 12 | 13 | 7 | 19 | 19 | 90 | 2 |
| 14 | LKR-3 | 19 | 11 | 11 | 17 | 10 | 11 | 17 | 96 | 2 |
| 15 | LKR-7 | 29 | 7 | 15 | 11 | 22 | 8 | 5 | 97 | 2 |
| 16 | UKR-9 | 23 | 17 | 17 | 9 | 16 | 17 | 21 | 120 | 2 |
| 17 | UKR-13 | 18 | 20 | 20 | 19 | 17 | 15 | 11 | 120 | 2 |
| 18 | UKR-10 | 25 | 19 | 18 | 15 | 18 | 16 | 15 | 126 | 2 |
| 19 | UKR-3 | 24 | 10 | 16 | 41 | 12 | 21 | 24 | 148 | 2 |
| 20 | MB-2 | 15 | 21 | 22 | 18 | 21 | 30 | 36 | 163 | 2 |
| 21 | AR-2 | 27 | 25 | 24 | 29 | 29 | 22 | 16 | 172 | 2 |
| 22 | LKR-5 | 31 | 23 | 19 | 20 | 27 | 33 | 23 | 176 | 2 |
| 23 | MR-1 | 17 | 26 | 25 | 21 | 30 | 29 | 30 | 178 | 2 |
| 24 | CR-5 | 14 | 37 | 30 | 26 | 28 | 26 | 20 | 181 | 2 |
| 25 | LKR-8 | 20 | 27 | 27 | 42 | 31 | 18 | 18 | 183 | 3 |
| 26 | AR-1 | 28 | 18 | 26 | 39 | 26 | 25 | 29 | 191 | 3 |
| 27 | MR-2 | 22 | 24 | 23 | 23 | 25 | 39 | 45 | 201 | 3 |
| 28 | CR-1 | 46 | 22 | 21 | 27 | 19 | 24 | 43 | 202 | 3 |
| 29 | MB-7 | 26 | 28 | 33 | 25 | 23 | 41 | 32 | 208 | 3 |
| 30 | CR-7 | 12 | 29 | 28 | 31 | 20 | 46 | 46 | 212 | 3 |
| 31 | MR-4 | 16 | 31 | 29 | 28 | 24 | 43 | 44 | 215 | 3 |
| 32 | UKR-7 | 33 | 36 | 35 | 24 | 35 | 37 | 27 | 227 | 3 |
| 33 | ER-4 | 35 | 33 | 36 | 36 | 34 | 34 | 25 | 233 | 3 |
| 34 | UKR-6 | 21 | 30 | 31 | 32 | 47 | 35 | 40 | 236 | 3 |
| 35 | AR-3 | 38 | 35 | 34 | 33 | 32 | 40 | 33 | 245 | 3 |
| 36 | UKR-5 | 39 | 32 | 32 | 30 | 33 | 42 | 37 | 245 | 3 |
| 37 | ER-5 | 34 | 39 | 40 | 34 | 41 | 23 | 35 | 246 | 4 |
| 38 | UKR-11 | 47 | 40 | 39 | 22 | 44 | 20 | 38 | 250 | 4 |
| 39 | CR-2 | 45 | 41 | 38 | 43 | 37 | 27 | 22 | 253 | 4 |
| 40 | CR-3 | 32 | 34 | 41 | 40 | 39 | 36 | 34 | 256 | 4 |
| 41 | ER-1 | 43 | 44 | 45 | 44 | 38 | 31 | 26 | 271 | 4 |
| 42 | ER-2 | 37 | 42 | 42 | 37 | 40 | 32 | 42 | 272 | 4 |
| 43 | ER-3 | 40 | 43 | 44 | 38 | 42 | 28 | 41 | 276 | 4 |
| 44 | CR-6 | 41 | 38 | 37 | 35 | 36 | 45 | 47 | 279 | 4 |
| 45 | MR-3 | 36 | 45 | 43 | 45 | 43 | 38 | 39 | 289 | 4 |
| 46 | UKR-12 | 44 | 46 | 46 | 46 | 46 | 44 | 28 | 300 | 4 |
| 47 | CR-8 | 42 | 47 | 47 | 47 | 45 | 47 | 31 | 306 | 4 |

in the tidal portions of the system that we sampled. Indeed, the entire system exhibits ebb-dominated sediment transport (Fenster et al., 2001). The system cannot even be characterized as a sand sink since it is an active contributor of sand to the coastal environments (FitzGerald et al., 2005). Any deposition that occurs in sheltered areas or under the turbidity maximum during low flow periods is temporary due to resuspension and transport by surface to bottom ebb flow currents during periods of high flow.

The findings that the metals are being introduced into the lower Kennebec/Androscoggin system from upstream and are not accumulating in Merrymeeting Bay or the lower estuary supports the hypothesis of Larsen and Gaudette (1995) that the large Kennebec/Androscoggin watershed (27,700 km²) is the source for much of the contamination observed in the nearshore Gulf of Maine. Although we have emphasized trace metals in this research, the distribution of organic contaminants such as PAHs and dioxin should mirror the metal distribution because of similar affinities for fine-grained sediments and organic particles.

5. Summary and conclusions

Metal levels in the Kennebec/Androscoggin study area sediments are generally elevated relative to background and the highest metal levels are found in the main stem of the system. Principal sources of the metals are the watersheds of the Kennebec and Androscoggin Rivers. The smaller tributaries with watersheds in the immediate vicinity of Merrymeeting Bay have statistically significant lower metal concentrations. Higher metal levels in the upper reach of the lower Kennebec estuary may be explained by the location of the seasonal Kennebec turbidity maximum. The strong ebb-current dominated flow insures the transport of sand and smaller sized particles to the nearshore Gulf of Maine. Deposition of fine particles is temporary. Accumulation of metals and, by inference, other contaminants in the system is, therefore, negligible. These findings are further evidence that contaminants from the Kennebec/Androscoggin watershed are transported to the nearshore Gulf of Maine.

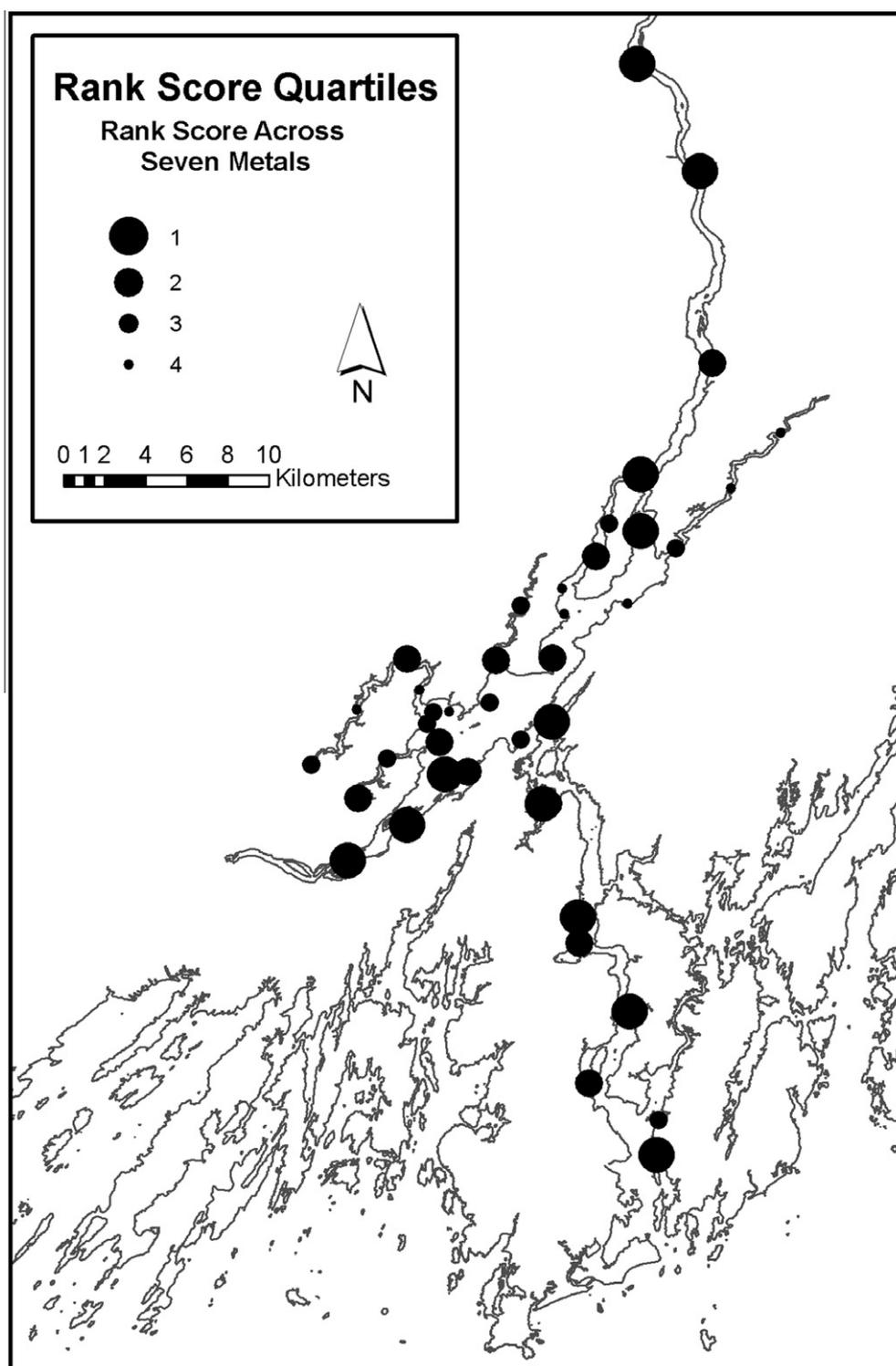


Fig. 5. Stations ranked by quartiles based on rank score sums across seven metals.

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