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Spatial and Temporal Aspects of Sedimentary Contaminant Concentrations in the Tidal Portions of the Kennebec/ Androscoggin River System : A Report to the Surface Water Ambient Toxics Monitoring Program, Maine Department of Environmental Protection

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SPATIAL AND TEMPORAL ASPECTS OF SEDIMENTARY CONTAMINANT
CONCENTRATIONS IN THE TIDAL PORTIONS OF THE
KENNEBEC/ANDROSCOGGIN RIVER SYSTEM

A Report to the
Surface Water Ambient Toxics Monitoring Program
Maine Department of Environmental Protection

by

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ABSTRACT

The concentrations of Cd, Cr, Cu, Ni, Pb, Sn and Zn in the surface sediments of 47 stations in the tidal Kennebec/Androscoggin system of the Gulf of Maine were determined. For data analysis purposes the region was divided into seven subregions consisting of five tributaries of Merrymeeting Bay, i.e. the Upper Kennebec, Muddy, Cathance, Abagadasset and Eastern Rivers, Merrymeeting Bay proper and the Lower Kennebec River connecting Merrymeeting Bay and the Gulf of Maine. Special emphasis was given to locating fine-grained depositional areas in this generally energetic, coarse grained system.

Most stations exhibited elevated metal concentrations. Statistically significant differences existed between the four small "local" tributaries and one or more of the three station groupings representing the main stem of the system. The distribution of metals indicated that the sources were the upstream Kennebec and Androscoggin watersheds. Metal levels in the upper reach of the lower Kennebec estuary were higher than found immediately upstream and downstream. This distribution can be explained by the existence of a turbidity maximum.

It is believed that the system is in a dynamic equilibrium with regard to particle and contaminant deposition and that further accumulation is negligible. 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.

INTRODUCTION

Elevated levels of toxic contaminants in the water, sediments and biota of several estuaries and embayments of the Gulf of Maine have been documented over the last three decades (Armstrong, *et al.*, 1976; Mayer and Fink, 1980; Lyons, *et al.*, 1978; Goldberg, *et al.*, 1983; Larsen, *et al.*, 1983a, 1983b, 1984; Ray and MacKnight, 1984; Gottholm and Turgeon; 1991, Larsen and Gaudette, 1995; Larsen, *et al.*, 1997; others). Taken together, these studies suggest considerable variability in the degree of enrichment as a function of source and transport mechanisms. A review of the environmental quality of the Gulf of Maine region (Larsen, 1992) suggests that the area between Cape Elizabeth and Boothbay is particularly complex and interesting. For instance, in the first comprehensive baseline survey of Casco Bay proper, Larsen, *et al.* (1983a) found all measured metals but cadmium to

be elevated well above pre-industrial levels, as defined by Lyons, *et al.*, (1978). Geographic distributions suggested anthropogenic inputs associated with activities in and around the commercially important Portland Harbor. Subsequently, the NOAA National Status and Trends Program (NS&T) reported that Casco Bay sediments were moderately enriched with metals and other toxics and that metal levels in livers of non-migratory fish collected near Cape Small, not far from the mouth of the Kennebec estuary, ranked high on both a Gulf of Maine and national scale (Gottholm and Turgeon, 1991; Larsen 1992). Data from the EPA Mussel Watch Program indicated that mussels from the isolated and undeveloped Cape Newagen ranked surprisingly high in lead and zinc content (Goldberg, *et al.*, 1983; Larsen, 1992).

These patterns of toxics in both sediments and biota over a relatively large area demonstrate that the study area is affected by numerous sources and complex, dynamic processes. Surveys of limited geographic scope, while important for local management concerns, are inadequate for determining and evaluating larger scale processes which may dominate regional fluxes of contaminants. One such larger scale process that may be important in Maine's mid-coast region is 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. Evidence from the distribution of heavy minerals (Ross, 1967), hydrographic modeling (D.A. Brooks, personal communication) and anecdotal accounts of pulpwood drift support this possibility. Most recently, Stumpf and Goldschmidt (1992) used satellite imagery to show the development and dispersion of a sedimentary plume from the Kennebec River estuary into the Gulf of Maine as a result of a major (100 year) storm. This one event could have transported over 500,000 metric tons of sediments and associated toxics through the estuary (R. Stumpf, personal communication), and the dispersion of the plume in the days following the initial event could explain many of the contaminant distributions noted in the above site-specific studies. Clearly, baseline surveys were needed on appropriate scales to evaluate suspected operative mechanisms.

Prompted by the above reports of contaminant concentrations in sediments and biota from mid-coast Maine, Larsen and Gaudette (1995) undertook, in 1991, a broad scale surficial sediment sampling and analysis program. Their goals were to document geographic distributions of contaminants on a regional level and to gain insight into possible sources and transport mechanisms. Trace metals were used as surrogates for the suite of toxics

moving through the region. Results reaffirmed the suspicion that the Kennebec/Androscoggin system may play a key role in regional contaminant dynamics. They concluded that more information was needed for both scientists and managers to understand the distribution and movements of contaminants in both space and time.

As an initial step in building a detailed understanding of the sources, movements and deposition of contaminants in the tidal Kennebec/Androscoggin system, Dr. Henri Gaudette of the University of New Hampshire and a graduate student undertook a focussed survey of the system. Sampling design and fieldwork was supervised by Dr. Peter Larsen as part of the Kennebec Area Research Endowment program. Once again, trace metals were used as surrogates for all contaminants that are associated with fine sediments and organic matter. Considerable effort was expended to locate stations with sufficiently fine sediments to provide a valid characterization of metal levels and distributions.

The resulting 1992 data set consisted of 47 stations between Hallowell, ME (52 km inland) and the lower Kennebec River estuary. With the exception of the lower Kennebec estuary, this system may be characterized as tidal fresh water. The distribution of stations within river segments is as follows: Lower Kennebec River(9), Merrymeeting Bay (includes lower Androscoggin River) (6), Upper Kennebec River (13), Muddy River (4), Cathance River (7), Abagadasset River (3) and Eastern River (5). The lower Androscoggin River is included as part of Merrymeeting Bay because no natural demarcation between them is evident. On the other hand, whereas it is commonly accepted that the northern limit of Merrymeeting Bay on the Kennebec River is the Richmond Bridge, we followed the convention of nautical charts and topographic maps and called everything north of Abagadasset Point the upper Kennebec River. Abagadasset Point is such a strong constriction that we assumed that the water above it is Kennebec water with only a slight dilution from the Eastern River. 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.

Specific goals of the investigation included:

- Documentation of geographic distribution of metals in the dynamic Kennebec/Androscoggin system. 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.
- To gain insights into locations of possible sources.
- To gain insights into the generic activities which may produce the contamination.

- To gain insights into temporal trends in sediment metal concentrations.

METHODS

Forty-seven stations (Fig. 1, Appendix 1) were sampled in the summer of 1992 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.

Grain size distributions were determined by standard sieving 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 hours.

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 grams of dried sediment (60°C, 18-24 hours) 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 liter with MilliQ water), and the volumes were reduced on a hot plate to 10-15 ml. Cooled samples 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 ug/gram dry weight sediment were calculated.

Analytical variability could not be determined by replicate analysis of standard sediment samples (U.S. 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 analytical error. 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 which was influenced by an outlier in the replicated samples.

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

RESULTS

Results of the sediment metal analyses with the percentages of fine sediments and loss on ignition are presented in Table 1. Background material on concentrations of major metals, pre-normalized trace metal concentrations, grain size calculations and data and loss on ignition calculations are presented in Appendices 2-5, respectively. Examination of the summary statistics at the bottom of 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 just upstream of Swans Island.

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 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; Fig. 2). 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).

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 located on the western side of Merrymeeting Bay. They have contiguous watersheds and have especially uniform trace metal loads 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 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 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 there are the differences between standard deviations are significant in each case.

Table 4. The level of significance of differences in levels of each 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

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 (Tables 6-12). The results are presented in a geographical context in Figs. 5-11. Examination of the tables and figures 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, that 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 quartile of stations.

The data can be further reduced by summing the rankings across the seven metals (Table 13). For instance, Station UKR-8 in the Kennebec River just north of Swans Island is ranked number 1 for six of the seven metals and number 3 for the seventh. Summing these rankings results in a score of 9. Hence, we can conclude that station UKR-8 has the highest trace metal burden of the 47 stations. Station MB-6 with a sum rank score of 33 is second, LKR-4 with a total score of 34 is third, and so on through the 47 stations. The geographic distribution of these rankings by quartile is presented in Fig.12.

Several important insights are revealed by this summed rank score analysis. The 20 highest ranked stations are located in UKR, MB and LKR (Table 13). 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 (Fig. 12). Stations in the minor tributaries are generally ranked in the third and fourth quartile. 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.

Table 5. Results of Dunn's Multiple Comparisons Tests. * indicates significance at the <0.05 level; ** at the <0.01 level.

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	**

Table 6. Stations ranked by the concentration of Cd.

Rank	Station	Cd Conc.	Quartile
1	UKR-8	1.820	1
2	MB-5	1.309	1
3	MB-6	1.263	1
4	LKR-4	1.236	1
5	MB-3	1.130	1
6	MB-4	1.128	1
7	LKR-1	1.036	1
8	LKR-2	0.991	1
9	UKR-1	0.976	1
10	UKR-4	0.955	1
11	LKR-6	0.892	1
12	CR-7	0.863	1
13	LKR-9	0.824	2
14	CR-5	0.789	2
15	MB-2	0.756	2
16	MR-4	0.751	2
17	MR-1	0.739	2
18	UKR-13	0.675	2
19	LKR-3	0.671	2
20	LKR-8	0.658	2
21	UKR-6	0.652	2
22	MR-2	0.648	2
23	UKR-9	0.636	2
24	UKR-3	0.622	2
25	UKR-10	0.622	2
26	MB-7	0.589	3
27	AR-2	0.588	3
28	AR-1	0.575	3
29	LKR-7	0.544	3
30	UKR-2	0.531	3
31	LKR-5	0.507	3
32	CR-3	0.505	3
33	UKR-7	0.484	3
34	ER-5	0.481	3
35	ER-4	0.465	3
36	MR-3	0.433	3
37	ER-2	0.421	4
38	AR-3	0.418	4
39	UKR-5	0.395	4
40	ER-3	0.369	4
41	CR-6	0.367	4
42	CR-8	0.328	4
43	ER-1	0.241	4
44	UKR-12	0.205	4
45	CR-2	0.200	4
46	CR-1	0.198	4
47	UKR-11	0.189	4

Table 7. Stations ranked by the concentration of Cr.

Rank	Station	Cr Conc	Quartile
1	UKR-8	218.54	1
2	UKR-2	175.08	1
3	MB-3	145.13	1
4	LKR-4	121.10	1
5	MB-6	108.57	1
6	MB-5	106.01	1
7	LKR-7	104.66	1
8	UKR-4	102.54	1
9	LKR-1	97.36	1
10	UKR-3	90.58	1
11	LKR-3	90.40	1
12	LKR-6	88.36	1
13	LKR-9	86.54	2
14	MB-4	85.64	2
15	UKR-1	84.57	2
16	LKR-2	74.64	2
17	UKR-9	73.20	2
18	AR-1	72.65	2
19	UKR-10	66.59	2
20	UKR-13	63.68	2
21	MB-2	60.33	2
22	CR-1	60.23	2
23	LKR-5	59.16	2
24	MR-2	58.30	2
25	AR-2	57.80	3
26	MR-1	57.58	3
27	LKR-8	55.95	3
28	MB-7	53.58	3
29	CR-7	50.71	3
30	UKR-6	50.45	3
31	MR-4	49.94	3
32	UKR-5	49.90	3
33	ER-4	48.13	3
34	CR-3	47.48	3
35	AR-3	47.05	3
36	UKR-7	46.61	3
37	CR-5	46.53	4
38	CR-6	45.33	4
39	ER-5	45.12	4
40	UKR-11	44.19	4
41	CR-2	42.56	4
42	ER-2	42.13	4
43	ER-3	40.41	4
44	ER-1	40.22	4
45	MR-3	37.71	4
46	UKR-12	30.80	4
47	CR-8	25.61	4

Table 8. Stations ranked by the concentration of Cu.

Rank	Station	Cu Conc	Quartile
1	UKR-8	98.43	1
2	UKR-2	78.28	1
3	MB-3	71.13	1
4	LKR-4	69.76	1
5	MB-5	64.38	1
6	MB-6	63.98	1
7	UKR-1	58.69	1
8	LKR-6	55.74	1
9	LKR-1	51.32	1
10	UKR-4	49.85	1
11	LKR-3	48.89	1
12	MB-4	46.78	1
13	LKR-9	45.19	2
14	LKR-2	45.14	2
15	LKR-7	42.64	2
16	UKR-3	41.51	2
17	UKR-9	40.95	2
18	UKR-10	35.14	2
19	LKR-5	33.52	2
20	UKR-13	32.67	2
21	CR-1	31.92	2
22	MB-2	31.64	2
23	MR-2	31.36	2
24	AR-2	30.61	2
25	MR-1	29.66	3
26	AR-1	29.59	3
27	LKR-8	29.39	3
28	CR-7	29.11	3
29	MR-4	28.91	3
30	CR-5	28.59	3
31	UKR-6	27.50	3
32	UKR-5	27.41	3
33	MB-7	27.03	3
34	AR-3	26.56	3
35	UKR-7	26.13	3
36	ER-4	24.80	3
37	CR-6	24.65	4
38	CR-2	23.63	4
39	UKR-11	23.37	4
40	ER-5	22.55	4
41	CR-3	22.44	4
42	ER-2	21.16	4
43	MR-3	20.34	4
44	ER-3	19.78	4
45	ER-1	19.24	4
46	UKR-12	15.70	4
47	CR-8	13.45	4

Table 9. Stations ranked by the concentration of Pb.

Rank	Station	Pb conc.	Quartile
1	UKR-4	284.68	1
2	UKR-1	111.25	1
3	UKR-8	94.27	1
4	UKR-2	80.47	1
5	MB-6	67.89	1
6	MB-5	66.40	1
7	MB-3	61.22	1
8	LKR-6	57.23	1
9	UKR-9	46.60	1
10	LKR-4	46.22	1
11	LKR-7	44.89	1
12	LKR-1	40.91	1
13	MB-4	40.52	2
14	LKR-2	39.59	2
15	UKR-10	38.85	2
16	LKR-9	37.29	2
17	LKR-3	35.38	2
18	MB-2	34.17	2
19	UKR-13	32.30	2
20	LKR-5	31.55	2
21	MR-1	29.81	2
22	UKR-11	29.72	2
23	MR-2	28.83	2
24	UKR-7	27.28	2
25	MB-7	27.03	3
26	CR-5	26.67	3
27	CR-1	26.34	3
28	MR-4	25.73	3
29	AR-2	25.46	3
30	UKR-5	25.39	3
31	CR-7	24.58	3
32	UKR-6	24.57	3
33	AR-3	24.29	3
34	ER-5	23.03	3
35	CR-6	22.27	3
36	ER-4	21.77	3
37	ER-2	21.19	4
38	ER-3	21.19	4
39	AR-1	20.97	4
40	CR-3	20.05	4
41	UKR-3	19.87	4
42	LKR-8	18.28	4
43	CR-2	16.45	4
44	ER-1	15.76	4
45	MR-3	14.40	4
46	UKR-12	10.05	4
47	CR-8	9.54	4

Table 10. Stations ranked by concentration of Zn.

Rank	Station	Zn conc	Quartile
1	UKR-8	474.61	1
2	MB-3	440.52	1
3	UKR-2	400.47	1
4	MB-5	343.71	1
5	MB-6	320.15	1
6	LKR-4	276.77	1
7	MB-4	256.92	1
8	UKR-4	248.46	1
9	LKR-1	236.85	1
10	LKR-3	215.24	1
11	LKR-2	209.47	1
12	UKR-3	198.84	1
13	UKR-1	185.79	2
14	LKR-9	180.88	2
15	LKR-6	179.49	2
16	UKR-9	172.91	2
17	UKR-13	155.29	2
18	UKR-10	154.41	2
19	CR-1	144.59	2
20	CR-7	143.99	2
21	MB-2	142.32	2
22	LKR-7	140.55	2
23	MB-7	132.39	2
24	MR-4	128.70	3
25	MR-2	128.70	3
26	AR-1	127.58	3
27	LKR-5	126.63	3
28	CR-5	121.86	3
29	AR-2	121.03	3
30	MR-1	119.36	3
31	LKR-8	116.56	3
32	AR-3	115.28	3
33	UKR-5	113.77	3
34	ER-4	107.28	3
35	UKR-7	102.25	3
36	CR-6	101.64	3
37	CR-2	100.71	4
38	ER-1	97.29	4
39	CR-3	96.41	4
40	ER-2	94.78	4
41	ER-5	93.16	4
42	ER-3	91.23	4
43	MR-3	88.21	4
44	UKR-11	86.96	4
45	CR-8	63.97	4
46	UKR-12	56.12	4
47	UKR-6	39.76	4

Table 11. Stations ranked by concentration of Sn.

Rank	Station	Sn Conc	Quartile
1	UKR-8	92.13	1
2	LKR-4	41.34	1
3	UKR-4	36.37	1
4	MB-5	34.89	1
5	UKR-2	34.61	1
6	MB-6	34.59	1
7	LKR-1	34.52	1
8	LKR-7	32.28	1
9	LKR-9	31.90	1
10	MB-3	31.04	1
11	LKR-3	30.04	1
12	UKR-1	28.84	1
13	LKR-2	27.30	2
14	LKR-6	26.87	2
15	UKR-13	22.78	2
16	UKR-10	21.23	2
17	UKR-9	20.46	2
18	LKR-8	20.38	2
19	MB-4	19.38	2
20	UKR-11	18.83	2
21	UKR-3	17.78	2
22	AR-2	16.26	2
23	ER-5	16.17	2
24	CR-1	15.99	2
25	AR-1	15.32	3
26	CR-5	14.91	3
27	CR-2	14.03	3
28	ER-3	13.90	3
29	MR-1	13.43	3
30	MB-2	13.36	3
31	ER-1	13.07	3
32	ER-2	12.82	3
33	LKR-5	11.95	3
34	ER-4	11.69	3
35	UKR-6	11.61	3
36	CR-3	11.14	3
37	UKR-7	10.76	4
38	MR-3	10.69	4
39	MR-2	10.55	4
40	AR-3	9.29	4
41	MB-7	9.16	4
42	UKR-5	9.03	4
43	MR-4	8.73	4
44	UKR-12	7.62	4
45	CR-6	7.52	4
46	CR-7	6.73	4
47	CR-8	6.11	4

Table 12. Stations ranked by the concentration of Ni.

Rank	Station	Ni Conc	Quartile
1	UKR-8	184.16	1
2	UKR-4	145.00	1
3	MB-6	95.35	1
4	LKR-4	89.30	1
5	LKR-7	79.35	1
6	UKR-1	78.80	1
7	UKR-2	73.76	1
8	LKR-9	69.14	1
9	LKR-2	66.68	1
10	MB-3	64.71	1
11	UKR-13	60.05	1
12	MB-5	58.82	1
13	LKR-6	55.90	2
14	LKR-1	53.27	2
15	UKR-10	52.72	2
16	AR-2	51.99	2
17	LKR-3	50.88	2
18	LKR-8	45.66	2
19	MB-4	41.16	2
20	CR-5	39.92	2
21	UKR-9	39.47	2
22	CR-2	37.54	2
23	LKR-5	35.04	2
24	UKR-3	34.43	2
25	ER-4	33.73	3
26	ER-1	33.68	3
27	UKR-7	33.59	3
28	UKR-12	33.29	3
29	AR-1	32.89	3
30	MR-1	32.87	3
31	CR-8	31.76	3
32	MB-7	31.66	3
33	AR-3	31.51	3
34	CR-3	31.12	3
35	ER-5	30.18	3
36	MB-2	30.06	3
37	UKR-5	30.06	3
38	UKR-11	29.79	4
39	MR-3	29.52	4
40	UKR-6	29.34	4
41	ER-3	26.82	4
42	ER-2	26.27	4
43	CR-1	26.19	4
44	MR-4	24.85	4
45	MR-2	24.22	4
46	CR-7	23.34	4
47	CR-6	18.79	4

COMPARISON WITH OTHER STUDIES

Comparisons between studies are often difficult due to differences in sampling techniques, analytical methodology and documentation. Nevertheless, even with the limitations, valuable insights can be discovered and the effort is usually rewarding. In the present case, there are a small number of recent studies that can be utilized. An initial observation is that, since the studies are all relatively recent, temporal comparisons would have little meaning.

The results, or selected results, of five studies are summarized in Table 14. Most of the included numbers represent means. The reader is reminded that there are variances around these mean values. The first three studies listed employ very comparable methodologies.

The first data set presented in Table 14 includes the mean concentrations of seven metals in the seven subregions of the present study. The previously noted concentration differences between the four smaller tributaries and the main stem regions are obvious. The results of Getchell (2002) from the nearby Boothbay region are included as a baseline. Her Gulf of Maine stations were taken 2-8 kilometers off Cape Newagen. Although no sites downwind of a continent are unimpacted by contaminants, these sites are isolated from direct inputs and may be considered to represent regional background contaminant levels. Her Boothbay and Inner Boothbay Harbor stations represent sites along a gradient of presumed increasing contaminant input. Comparison of the present results with Getchell's reveals that, with one exception, samples for the Kennebec/Androscoggin system contains elevated levels of metals. Zn appears to be especially elevated. The one exception is Pb that exhibits concentrations in the four small Merrymeeting Bay tributaries that are below our chosen Gulf of Maine background level.

There is good correspondence between the present results and those of Larsen and Gaudette (1995). Stations 23-25 of Larsen and Gaudette (1995) are located in the lower Kennebec River and in each case the range of values reported for these stations bracket the mean values reported for the LKR grouping in the present study. These authors had reported that metal levels in the region, especially in the main stem of the Kennebec estuary, were elevated above pre-industrial levels.

Results from the FOMB/DEP study are in general agreement with the present study for the two metals that were analyzed in common. Pb levels are near or below the Gulf of Maine baseline and Zn levels are in agreement for similar areas; for instance, in the Muddy River 127.9 vs. 116.2 and in the Abagadasset River

114.4 vs. 121.3. The FOMB/DEP study is still in production. Once it is complete with detailed methodology and specific sampling sites, it would be productive to do more thorough comparisons of these and other parameters.

Chilcote and Waterfield (1995) sampled 14 stations in the Merrymeeting Bay area. Because of the extremely sandy nature of their samples, and basic differences in methodology, we are not able to compare results.

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 (Table 14). 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 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 13, Fig.12). 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. Likewise, highly ranked stations in the upper Kennebec are located where the river first meets the two-way tidal flow below the (former) dam in Augusta (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, 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. When fresh, river water collides with seawater to form an estuary, unique physical and chemical processes result. Seawater is denser than fresh water. As a result, in a constricted tidal estuary, it sinks and produces a bottom current with a net upstream movement. Conversely, the fresh water floats upon seawater and produces a surface current with a net downstream movement. Hence, as sediment particles carried by the downstream flowing river water sink, as they tend to do, they become entrained in the upstream moving bottom current. Further up estuary, the particles will be mixed back into the downstream surface current to sink again into the bottom current. Many particles become captured in this cyclic estuarine circulation. At the same time, when the fine river borne sediment and organic particles, with which the contaminants are associated, come into contact with the salts in the seawater, chemical and electrostatic changes occur. This causes changes in the solubility of many contaminant complexes and, very dramatically, it causes the small contaminant laden particles to flocculate, i.e. bind together, and become less buoyant. The result of these processes is that the upper reaches of estuaries are often characterized by a region of increased suspended loads and underlain by muddy deposits. This region is called the turbidity maximum and it is here where higher levels of contaminants would be expected. Hydrographic conditions in the Kennebec estuary allow for the formation of a turbidity maximum during periods of low or moderate flows which occur about three-quarters of the time (Kistner and Pettigrew, 2001). The location of the Kennebec turbidity maximum 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 in the tidal portions of the system that we sampled. Olsen, *et al.* (1993) investigated a range of US east coast estuaries in an effort to explain patterns observed in estuarine particle retention or export. The Kennebec/Androscoggin system fits into their Type I where "sediment and contaminant accumulation are negligible". Like our study area, Type I areas have noncohesive sediments strongly influenced by physical or biological mixing. They are in "a state of dynamic equilibrium with respect to sea level, river discharge, tidal currents and wave activity" and have "apparently obtained an equilibrium depth above which net particle and contaminant deposition is negligible, despite an excess of both." They say further that the entire suspended

sediment and contaminant load bypasses these areas. Any deposition that occurs is temporary due to resuspension by currents and waves.

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.

SUMMARY AND CONCLUSIONS

Metal levels in the Kennebec/Androscoggin study area sediments are generally elevated relative to background

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 Merrymeeting Bay area have statistically significant lower metal levels

Higher metal levels in the upper reach of the lower Kennebec estuary may be explained by the location of the Kennebec turbidity maximum

The system is in dynamic equilibrium in regards to particle and contaminant deposition. Accumulation of metals and, by inference, other contaminants in the system is negligible

These findings are further evidence that contaminants from the Kennebec/Androscoggin watershed are transported to the nearshore Gulf of Maine

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1.5

ESTUARINE SEDIMENT CHARACTERIZATION
(from 1999)

ESTUARINE SEDIMENT CHARACTERIZATION

Recent hazardous waste site assessments in lower river systems and estuaries have demonstrated the need for a better understanding of toxic contaminant levels in estuarine sediments. These areas, neither river nor marine, and a transition zone between erosional and depositional areas are not well characterized. Waste discharge license limits are based on ambient concentrations of a toxicant after mixing. Due to stoichiometric changes between fresh and salt water, many contaminants settle shortly after reaching saline conditions. The amount of contaminants deposited in these areas is a reflection of the actual load delivered from the river (and treatment plants) and is largely independent of ambient concentrations. Concern has been raised that although concentrations may be decreasing, loading may be actually increasing due to increased discharge flows.

Some estuarine sediment chemistry has been conducted, but most work has been in euryhaline areas. In the 1999-2003 five year plan, we intend to characterize sediments in the major estuarine areas at a rate of one estuary area each year. The Friends of Merrymeeting Bay helped collect samples from Merrymeeting Bay in 1999 and results were reported in the 1999 SWAT report. Samples for dioxins and furans, however, were not analyzed. New samples were collected for dioxin and furan analysis in 2000. Results are as follows.

STATIONS

AB Abagadasset River near Bald Head N43:59.787, W69:51.073.
AR Androscoggin River near Bayshore Road N43:57.446 W69:51.591
KR Kennebec River near Abagadasset Point N43:59.915 W69:49.826
MR Muddy River near Pleasant Point N43:58.205, W69:52.871
SI Swan's Island south end N43: 59.787 W69:51.073
WC Whiskeag Creek mouth N43:56.169 W69:49.827

TABLE 1.5.1 DIOXIN IN 2000 MERRYMEETING BAY SEDIMENT SAMPLES

Sed ID		AR-1	SI-2	AR-2	KR-3	AR-3	WC-2
Congener	DL (ng/Kg, dry weight)						
2,3,7,8-TCDF	0.11	6.88	1.17	2.22	5.68	2.55	1.10
1,2,3,7,8-PeCDF	0.25	1.99	0.21	0.64	1.79	0.81	<DL
2,3,4,7,8-PeCDF	0.25	2.14	0.26	0.78	1.73	1.10	<DL
1,2,3,4,7,8-HxCDF	0.25	10.1	0.77	1.82	2.52	3.62	0.18
1,2,3,6,7,8-HxCDF	0.25	8.50	0.58	1.31	2.03	3.34	0.24
2,3,4,6,7,8-HxCDF	0.25	3.34	0.23	0.79	1.19	2.44	<DL
1,2,3,7,8,9-HxCDF	0.25	1.29	<DL	0.110	0.71	0.30	<DL
1,2,3,4,6,7,8-HpCDF	0.50	173	14.9	140.4	27.5	101	7.68
1,2,3,4,7,8,9-HpCDF	0.50	12.9	0.715	2.77	2.25	32.3	<DL
OCDF	0.50	282	8.77	156	85.4	227	20.7
2,3,7,8-TCDD	0.10	0.55	0.07	0.37	0.59	0.19	1.30
1,2,3,7,8-PeCDD	0.25	1.69	<DL	0.36	0.83	0.43	<DL
1,2,3,4,7,8-HxCDD	0.25	3.55	0.40	0.82	1.07	1.17	<DL
1,2,3,6,7,8-HxCDD	0.25	27.1	1.61	5.27	3.20	9.11	1.38
1,2,3,7,8,9-HxCDD	0.25	20.4	1.56	2.68	2.60	6.62	1.36
1,2,3,4,6,7,8-HpCDD	0.50	365	19.9	78.8	54.3	177	43.6
OCDD	0.50	4279	141	1183	1329	3166	450
TEQ ND=0		17.491	1.216	5.002	5.259	7.573	2.281
TEQ ND=DL		17.491	1.491	5.002	5.259	7.573	2.749
sample weight (g wet wt)		100	100	94	125	91	143
% solids		50	50	53	40	55	35
Sediment amounts are based on the % solids to give a 50 g sample weight of dry material.							

Sed ID		WC-3	AB-c3	MR-c2	AB-c3	MR-c3
Congener	DL (ng/Kg, dry weight)					
2,3,7,8-TCDF	0.11	1.62	<DL	<DL	<DL	<DL
1,2,3,7,8-PeCDF	0.25	0.81	<DL	<DL	<DL	<DL
2,3,4,7,8-PeCDF	0.25	0.81	<DL	<DL	<DL	<DL
1,2,3,4,7,8-HxCDF	0.25	1.01	<DL	<DL	<DL	<DL
1,2,3,6,7,8-HxCDF	0.25	1.02	<DL	<DL	<DL	<DL
2,3,4,6,7,8-HxCDF	0.25	0.45	1.05	3.02	1.05	3.02
1,2,3,7,8,9-HxCDF	0.25	<DL	2.26	4.98	2.26	4.98
1,2,3,4,6,7,8-HpCDF	0.50	18.6	66.5	112.0	66.5	112.0
1,2,3,4,7,8,9-HpCDF	0.50	0.98	<DL	6.95	<DL	6.95
OCDF	0.50	51.5	117	156	117	156
2,3,7,8-TCDD	0.10	0.33	<DL	<DL	<DL	<DL
1,2,3,7,8-PeCDD	0.25	0.69	<DL	<DL	<DL	<DL
1,2,3,4,7,8-HxCDD	0.25	0.67	<DL	<DL	<DL	<DL
1,2,3,6,7,8-HxCDD	0.25	2.96	<DL	<DL	<DL	<DL
1,2,3,7,8,9-HxCDD	0.25	2.20	<DL	<DL	<DL	<DL
1,2,3,4,6,7,8-HpCDD	0.50	65.9	106	332	106	332
OCDD	0.50	826	1776	2550	1776	2550
TEQ ND=0		3.397	2.25	5.58	2.25	5.58
TEQ ND=DL		3.422	2.87	6.20	2.87	6.20
sample weight (g wet wt)		125	51.0	51.0	51.0	51.0
% solids		40				

Sediment amounts are based on the % solids to give a 50 g sample weight of dry material.

