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Tidal Creek and Salt Marsh Sediments in South Carolina Coastal Estuaries: I. Distribution of Trace Metals

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Abstract. Twenty-eight tidal creeks were sampled along the South Carolina coast in the summer of 1995 to determine the levels of sediment trace metal contamination associated with different types and varying levels of human development in their watersheds. The particle size and total organic carbon (TOC) content of creek sediments in developed watersheds (*i.e.,* industrial, urban, and suburban) were similar to that in watersheds with little or no development (*i.e.,* forested or reference). Those trace metals commonly associated with urban and industrial sources, including Cu, Cr, Pb, Zn, Cd, and Hg, were in significantly higher concentrations in tidal creeks located in industrial/urban watersheds compared to the suburban and forested watersheds. Sediment trace metal concentrations were similar for creeks located in suburban and forested watersheds and 2 to 10 times lower than the creeks located in industrial/urban watersheds. Concentrations of trace metals primarily associated with the natural weathering of basement rock, including Al, Fe, As, Ni, and Mn, were not significantly different among watershed types. Four of the tidal creek–salt marsh systems were extensively sampled from the creek channel to the marsh-upland interface to characterize sediment trace metal spatial distributions within creek-marsh systems. Sediment particle size, TOC, and trace metal concentrations varied spatially within each creek-marsh system depending on the type of development in the watershed and the probable source of metals. The creek-marsh system selected to represent the industrial development had significantly higher ‘‘anthropogenic’’ trace metal concentrations compared to the other creek-marsh systems. This system also had trace metal distributional patterns that appeared to be associated with several localized sources of metals on the marsh surface. Both the ‘‘anthropogenic’’ and ‘‘natural’’ trace metal concentrations and spatial distributions were similar among and within the forested and suburban creek-marsh systems.

The human population of the United States is concentrated near coastal ecosystems with approximately 37% of the 1994 population living within 100 km of a coastline (Cohen *et al.* 1997). In addition, millions of tourists visit these coastal environments each year (Miller and Auyong 1991; Miller 1993). Over the next several decades, the coastal population of the southeastern United States is expected to increase by over 60% from the 1960 population levels (Culliton *et al.* 1990). This population growth will undoubtedly be accompanied by industrial expansion, suburbanization of forested areas, infrastructure development and improvement, and increased loadings of point and nonpoint source pollutants, including trace metals, polycyclic aromatic hydrocarbons, pesticides, and nutrients (US EPA 1993; Fortner *et al.* 1996; Kucklick *et al.* 1997). These changes will adversely affect the productivity, biodiversity, and ecological functioning of coastal ecosystems (Vitousek *et al.* 1997).

The watersheds that drain into the numerous meandering, shallow tidal creeks and salt marshes of the southeastern coastal zone are preferred sites for development. These creeks and their associated salt marshes serve as a refuge and nursery habitat for many fish and crustaceans (*e.g.,* Hackney *et al.* 1976; Weinstein 1979; Wenner and Beatty 1993) and provide feeding areas for wading birds (Dodd and Murphy 1996) and large predatory fish (Wenner 1992). Tidal creek and salt marsh sediments also have the potential to serve as repositories for contaminants that are transported into these systems from runoff and become adsorbed to sediments and organic materials (Weinstein 1996). The larval and juvenile life history stages of fish and shellfish that occur in tidal creek–salt marsh systems are frequently more sensitive to chemical contaminant exposure than adults (Baughman *et al.* 1989). Contaminants in these ecosystems may also be transferred through the food web to higher trophic levels

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(Barron 1995; DiPinto 1996; DiPinto and Coull 1997) including humans.

In 1994–1995, the South Carolina Department of Natural Resources conducted a study to define the linkages between tidal creek environmental quality and human uses of their watersheds. For this study, a population of 28 tidal creek systems was sampled using a comparative watershed approach. A suite of environmental quality indicators were examined for each creek including watershed characteristics (*e.g.*, land cover, human population density), environmental setting (*e.g.*, water quality, sediment characteristics), chemical contamination of the sediment, and ecological resources (*e.g.*, the kinds and abundances of living resources, health of individual resident biota).

The objectives of this particular study were to: (1) determine if the levels of sediment trace metal contamination in tidal creeks were associated with the type and degree of anthropogenic activity within the watershed; (2) determine the spatial distribution of sediment trace metal contamination across four creek-marsh systems associated with different types of land use; and (3) provide baseline information on trace metal distributions for tidal creek and salt marsh systems that could be used as a basis for the design of assessment, monitoring, and research programs. The potential biological effects associated with these patterns of trace metal contamination will be addressed in subsequent publications.

Materials and Methods

Study Area and Sampling Design

This study sampled trace metals from 28 tidal creeks located along the South Carolina coast from July to September 1995 (Figure 1). Twenty-four creeks were located in the Charleston Harbor Estuary. Two creeks were located in the Grand Strand area, approximately 120 km north of Charleston, and two creeks were located in the Hilton Head area, approximately 80 km south of Charleston. Each creek consisted of a relatively unbranched channel that drained a defined basin and formed the first-order connection between the drainage basin and estuarine habitats.

The 28 tidal creeks were classified into two populations (*i.e.,* upland and salt marsh) based on the predominant land cover of each watershed (Table 1). Upland creeks had from 15 to 97% of their watersheds as uplands. The watersheds of salt marsh creeks were entirely estuarine wetlands. Upland creeks were classified into the following five watershed classes that represented varying degrees of anthropogenic development: (1) forested or reference (*i.e.,* $<$15% of the watershed was developed); (2) suburban (*i.e.,* $>$45% urban/suburban land cover with a human population density $>$5 but $<$20 individuals/ha); (3) industrial (*i.e.,* $>$45% urban/suburban land cover which drained known industrial facilities); (4) urban (*i.e.,* $>$70% urban/suburban land cover with a human population density $>$20 individuals/ha); and (5) agriculture (*i.e.,* $>$40% agricultural land cover). Salt marsh creeks were classified into the following two classes: (1) unimpacted or reference, and (2) impacted. Salt marsh unimpacted creeks were located in relatively undeveloped regions of the harbor (*i.e.,* Fort Johnson), and impacted creeks were located in highly developed regions of the harbor (*i.e.,* James Island, West Ashley). The potential existed for contaminant loadings to enter salt marsh–impacted creeks from adjacent estuarine contamination and atmospheric deposition.

The upper boundary of each creek was defined as the point where water depth in the center of the channel was ~1 m deep on the average high tide. The lower boundary was defined as the point where the creek converged with another water body (secondary creek or tidal river) or the water depth in the center of the channel exceeded ~3 m on the average high tide. Creeks were stratified into 300 m reaches, and the creek lengths varied from one to five reaches.

Two sampling programs were conducted: (1) a tidal creek study (all 28 creeks), and (2) a creek-marsh transect study (four representative creeks). For the tidal creek study, a single, random sediment sample was collected at midtide elevation (intertidal) of the creek channel in both the upper- and lowermost reaches of each creek. The creek-marsh transect study examined the spatial distribution of sediment trace metals from the midtide level of the creek channel to the marsh-upland interface. Four creek-marsh systems were selected to represent the industrial (Diesel Creek), suburban (Shem Creek), and forested (Rathall Creek and Long Creek) upland watershed classes. For this study, three randomly located transects were defined perpendicular from the creek channel to the adjacent uplands in the upper- and lowermost reach of each creek (Figure 2). A single sediment sample was collected at four fixed locations along each transect: (1) the midtide level (intertidal) of the creek channel (creek bed), (2) the creek channel edge (berm), (3) a location 50% of the distance between the creek and the upland (midmarsh), and (4) a location 5 m from a vegetation shift from *Spartina alterniflora* to *Juncus roemerianus* or an upland vascular plant (high marsh). This vegetation shift indicates the mean to upper high-tide level. The distance from the creek channel to the adjacent uplands varied among creeks from 9 to 196 m.

Samples consisted of ~2 L of surface (top 2 cm) sediment placed in a solvent-clean stainless steel bowl using a stainless steel spoon. Sediments were homogenized, then divided into two aliquots. One aliquot was placed in an acid-washed plastic jar for trace metal analyses, and the other in a plastic bag for grain size and organic carbon analyses. The collecting utensils were rinsed in acetone and site water or deionized water between sites. All samples were placed on ice until reaching the National Oceanic and Atmospheric Administration, National Ocean Service (Charleston, SC) laboratory where they were stored at −60°C until being processed to determine trace metal content.

Physical and Chemical Analyses

Sediment grain size (*i.e.*, silt, clay, and sand) was determined for each sediment sample using protocols modified from Plumb (1981). Total organic carbon (TOC) was analyzed using a Perkin-Elmer 2400 Elemental Analyzer (Norwalk, CT) at a 950°C combustion temperature. Except for Hg, trace metal extraction was performed by closed-vessel, concentrated acid microwave digestion technique (Fortner *et al.* 1996). Weighed samples (~0.5 g) were placed in Teflon-lined digestion vessels to which concentrated HNO_3 and deionized water were added. Vessels were microwaved (CEM Model MDS-2000, Matthews, NC) for 2 h at 120 psi and allowed to cool before adding 30% H_2O_2. They were then microwaved for 10 min at 80 psi. Samples were filtered (#41 filter paper) and the remaining solution brought to a volume of 50 ml with deionized water.

Sediment trace metal concentrations of Al, Fe, Ni, Mn, Cu, Cr, Zn, and Sn were analyzed by inductively coupled plasma spectroscopy (ICP) on a Perkin-Elmer Plasma 1000 (Norwalk, CT) using an autosampler. Sediment concentrations of As, Pb, Cd, Ag, and Se were analyzed by graphite furnace atomic absorption (graphite furnace-AA) using a Perkin-Elmer 5100 Atomic Absorption Spectrometer (Norwalk, CT) with a Zeeman HGA 600 Graphite Furnace. The modifiers $Mg(NO_3)_2$ and/or palladium, and PO_4-$Mg(NO_3)_2$ were used to determine Pb, Ag, and As, Cd concentrations, respectively.

A separate extraction procedure was used for Hg. Dried sediment samples were weighed (~0.2–0.5 g) and transferred to a biological oxygen demand bottle to which deionized water, HNO_3, and HCl were added. The bottle was placed in a heated water bath (95 ± 2°C) for 2 min, cooled, and deionized water and $KMnO_4$ were added. The bottle was then heated in the water bath for 30 min and cooled. NaCl-

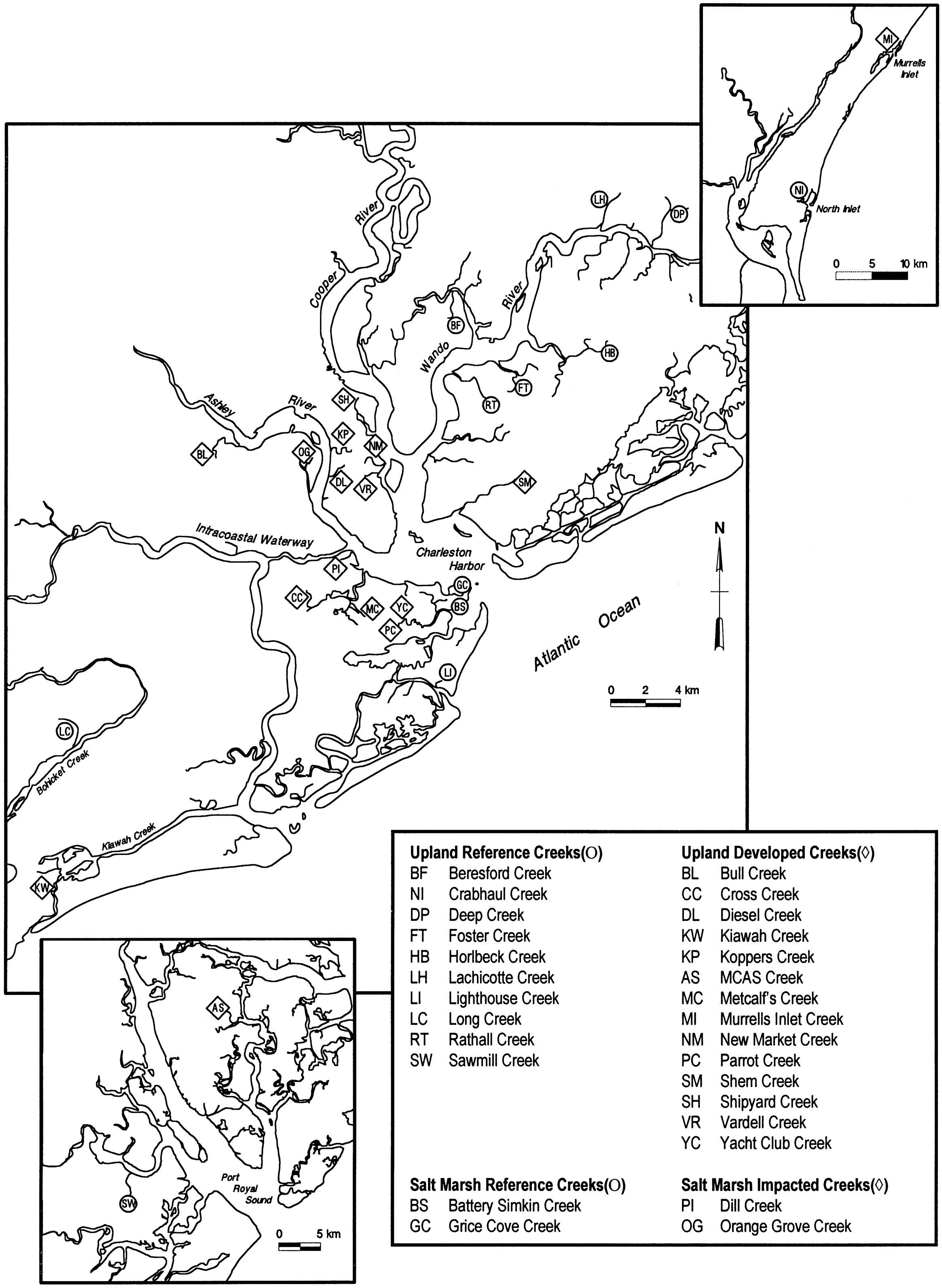

Fig. 1. Map showing location of tidal creeks sampled in South Carolina. The reference (○) and developed/impacted (◇) creeks are presented by their abbreviations. The inserts are the north (top) and south (bottom) sampling sites

hydroxyamine hydrochloride solution and deionized water were then added to complete the extraction process. Hg was analyzed by a cold-vapor atomic absorption procedure with a Leeman Labs PS200 mercury analyzer (Hudson, NH) at a wavelength of 253.7 nm. The Hg concentration was estimated from the slope of the calibration curve and the sample absorption. All sample extracts were analyzed in duplicate, and the results were averaged. Results are reported on a µg/g dry weight basis.

Table 1. Summary of the human population density, watershed size, and land cover for the 28 tidal creeks sampled. The creek name corresponding to the creek abbreviation can be found in Figure 1

Creek Abbreviation	Pop. Density (ind/ha)	Watershed Size (ha)	Forested (ha)	Salt Marsh (ha)	Urban/Suburban (ha)	Agriculture (ha)	Water (ha)	Barren (ha)
Forested or reference								
BF	0.0	25	5	20	0	0	0	0
NI	0.0	197	182	15	0	0	0	0
DP	0.0	65	40	12	0	13	0	0
FT	0.0	16	3	13	0	0	0	0
HB	0.1	238	185	9	0	22	4	18
LH	0.0	13	2	11	0	0	0	0
LI	0.0	37	3	32	0	0	0	3
LC	0.3	412	243	18	6	102	0	44
RT	0.6	72	30	35	0	0	0	7
SW	N/A	524	426	12	21	0	1	65
Suburban								
BL	12.7	434	84	21	320	0	16	4
CC	10.6	313	29	5	227	9	2	41
AS	N/A	434	101	7	266	0	0	61
MC	8.9	130	17	7	88	0	0	17
MI	N/A	83	34	10	39	0	0	1
PC	7.0	147	24	15	69	8	1	31
SM	15.7	428	62	9	286	0	4	67
YC	7.1	69	5	8	54	0	1	2
Industrial								
DL	6.9	105	0	33	64	0	0	9
KP	3.3	117	0	12	93	0	0	12
SH	8.2	280	14	10	180	0	0	75
Urban								
NM	21.1	198	3	27	140	0	0	18
VR	31.8	69	0	10	91	0	0	0
Agricultural								
KW	0.0	155	23	29	0	102	0	0
Salt marsh unimpacted								
BS	0.0	23	0	23	0	0	0	0
GC	0.0	21	0	21	0	0	0	0
Salt marsh impacted								
PI	0.0	20	0	20	0	0	0	0
OG	0.0	19	0	21	0	0	0	0

N/A: data not available

Calibration curves were developed for each metal and National Institute of Standards and Technology (NIST) Standard Reference Materials (SRM) were analyzed seven times during the processing of these samples. Recoveries ranged from 64% for Al to 175% for Cd and averaged 95% ± 25% (mean ± SD). All recoveries were within the acceptable confidence limits of the SRM material. Ag and Se concentrations were rarely found to exceed the level of detection, and Sn concentrations were consistently low. Consequently, these trace metals are not discussed further.

Data Analysis

Statistical analyses were performed using PC-SAS. Only two metals, Ni and Cd, had values below the level of quantification (LOQ) or the level of detection (LOD). Concentrations of Ni and Cd below the LOQ were set to the estimated value, whereas concentrations below the LOD were set to 0.00. Only 16% and 3% of the Cd concentrations were below the LOQ and LOD, respectively. Only 5% and 1% of the Ni concentrations were below the LOQ and LOD, respectively. Therefore, the level of skewing or biasing of the data from values below the LOD or LOQ was limited, and no mathematical alterations were performed. Linear regression analysis was used to determine the relationship between the trace metal concentrations and sediment clay or TOC content. Three data sets were established for the regressions: (1) the creek study and creek-marsh transect study sites ($n = 144$); (2) the reference sites from the creek study and creek-marsh transect study ($n = 69$); and (3) the developed and impacted sites from the creek study and creek-marsh transect study ($n = 75$).

The tidal creek study data ($n = 54$) was analyzed using a two-way analysis of variance (ANOVA) to determine differences in metal concentrations between watershed classes and reach. Data for all watershed classes were used in these analyses; however, least squares mean contrasts focused on comparing patterns for the upland watershed classes and the salt marsh watershed classes. The industrial and urban creeks were combined to increase the sample size for this class of creeks. Only one agricultural creek was sampled. The data for this creek are provided, but findings will not be discussed. Differences in sediment trace metal concentrations between the upper and lower reaches were examined within each watershed class using a one-way ANOVA.

The creek-marsh transect study was analyzed using two-way and three-way ANOVAs. The two-way ANOVAs (main effects = transect position and reach) were performed to evaluate the spatial distributions of trace metals within the individual creek-marsh systems sampled. The three-way ANOVAs (main effects = creek, transect position, and

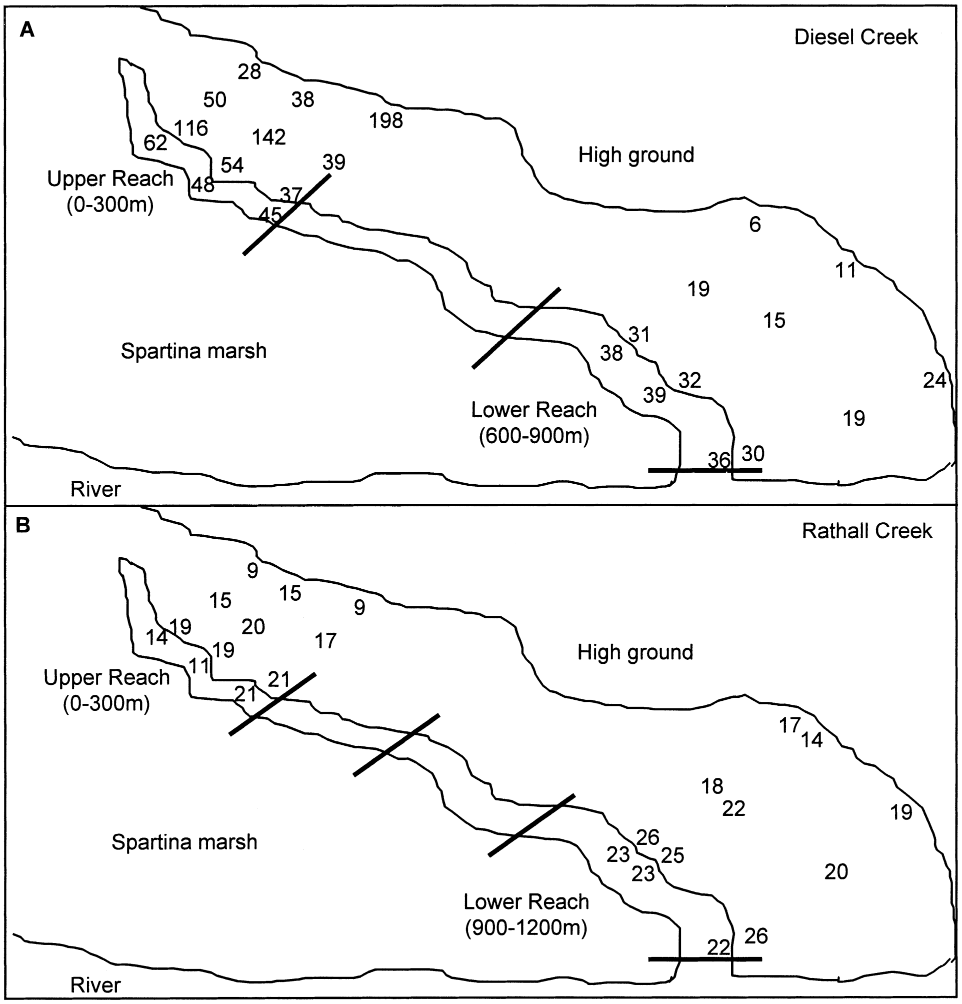

Fig. 2. The copper concentrations (µg/g dry weight) at each sampling site within the Diesel (A) and Rathall (B) tidal creek–salt marsh systems

reach) were performed to characterize the spatial distributions of trace metals within creeks and to evaluate differences among creeks.

The nonsignificant ($\alpha = 0.10$) interaction terms were removed from both the tidal creek study two-way ANOVAs and the creek-marsh transect study two-way and three-way ANOVAs and the analysis was reaccomplished. A significance level at $\alpha = 0.05$ was applied for all other statistical analyses. Contrasts were based on least squares means. The trace metal concentrations were $\log_{10}$ transformed (except Al and Fe), and the percentages of clay and TOC were arcsine-square-root transformed for the ANOVAs to help statisfy the homoscedasticity and normality assumptions.

Results

Overall, sediment particle size was heterogenous for the tidal creek study and the creek-marsh transect study. Sediments at sample sites ranged from muds (>50% clay) to sands (<15% clay); however, the majority of sample sites were characterized by mixed (15–50% clays) sediment types (Tables 2 and 3). The clay content of sediments was positively correlated with the silt-clay content ($r^2 = 0.87$, $p = 0.0001$). The silt content was about a third of the clay content.

Clay content of the creek channels in the tidal creek study ranged from 7–82% with a mean of 53% (Table 2). There was no consistent pattern in the distribution of clay with regard to watershed type or reach ($p = 0.69$). The clay content of creek channels for the upland watersheds, which included forested (50 ± 4, mean ± SE), suburban (53 ± 6), and industrial/urban (53 ± 5), were similar to each other (Figure 3A). The clay content of the salt marsh watersheds, which included unimpacted (57 ± 8) and impacted (74 ± 2) creeks, tended to be higher, but these differences were not statistically significant from the upland watershed classes (Figure 3A and Figure 4A).

The spatial distribution of sediment particle size was also heterogenous among and within the four intensively studied "representative" creeks (*i.e.*, creek-marsh transect study) (Table 3, Figure 5A). The clay content was significantly lower in Long Creek (38 ± 3) than in Rathall (58 ± 3), Diesel (52 ± 4), or Shem (60 ± 4) Creeks. In general, the berm and midmarsh sample sites had the highest clay content and the high marsh and creek bed had the highest sand content in these systems.

Total organic carbon of the sediment in the creek study ranged from 0.7 to 6.6% with a mean of 3.9% (Table 2). There was no consistent pattern in the distribution of TOC with regard to watershed class or reach ($p = 0.41$). The TOC content in the upland watershed classes, which included forested (3.9 ± 0.4), suburban (3.4 ± 0.4), and industrial/urban (4.5 ± 0.5), were similar to each other (Figure 3A) and to the salt marsh watershed classes, which included the unimpacted (4.0 ± 0.5) and impacted (5.6 ± 0.5) creeks (Figure 4A).

Table 2. Summary of the trace metal concentrations, clay, and TOC content collected in the tidal creek study

Site ID	Clay	TOC	Al	As	Cd	Cr	Cu	Fe	Pb	Mn	Hg	Ni	Zn
Forested or reference													
BF—U	77.2	5.8	5.76	14.5	0.20	94.3	29.0	3.11	28.6	173.2	0.12	23.5	99.2
BF—L	74.8	5.9	5.86	14.9	0.21	89.6	30.6	3.22	27.4	174.1	0.07	24.1	99.5
NI—U	11.4	0.9	1.05	3.5	0.00	12.7	2.2	0.56	4.2	38.2	0.01	4.4	15.0
NI—L	56.1	3.9	4.49	27.5	0.04*	48.8	11.4	2.42	15.0	122.6	0.08	15.2	55.1
DP—U	51.3	4.5	4.73	4.3	0.05*	56.9	12.6	2.28	17.0	162.0	0.04	14.0	52.9
DP—L	47.5	4.9	5.01	11.4	0.09	64.5	19.3	2.40	19.1	129.5	0.04	16.6	65.6
FT—U	67.2	5.9	7.01	10.2	0.13	83.7	26.4	3.62	18.9	162.6	0.10	23.1	89.1
FT—L	64.9	5.1	6.42	14.7	0.16	86.2	25.3	3.35	22.4	185.7	0.09	20.6	88.2
HB—U	32.1	3.3	3.64	5.4	0.12	42.4	12.0	1.95	15.6	123.5	0.06	10.5	48.7
HB—L	63.3	5.5	6.71	11.5	0.16	76.1	27.1	3.43	25.2	145.2	0.09	21.3	86.6
LH—U	66.1	5.7	7.29	8.7	0.16	82.9	25.2	3.48	24.7	163.8	0.12	21.4	91.5
LH—L	54.8	4.5	4.37	12.7	0.11	57.3	17.0	2.45	19.2	194.2	0.04	15.2	65.4
LI—U	53.5	3.4	6.60	18.1	0.08	66.5	16.2	3.79	20.1	247.1	0.05	21.0	68.9
LI—L	30.4	2.0	2.72	15.7	0.06*	38.0	7.7	1.88	10.0	221.3	0.03	8.7	40.5
LC—U	51.0	4.0	3.57	9.0	0.08	42.2	8.4	1.76	7.0	76.5	0.06	10.4	42.2
LC—L	19.8	0.9	1.65	3.5	0.04*	24.9	2.8	0.90	9.9	99.5	0.02	3.68*	20.4
RT—U	47.9	3.8	5.00	8.5	0.12	54.2	13.8	2.45	19.5	209.9	0.06	14.3	56.8
RT—L	64.6	4.1	6.72	12.1	0.14	77.1	23.3	3.59	22.2	256.9	0.10	21.7	85.6
SW—U	17.1	1.4	1.39	1.9	0.05*	16.2	1.6	0.52	5.1	37.9	0.02	3.51*	11.9
SW—L	40.2	2.9	4.60	13.5	0.07*	52.5	7.8	2.00	11.6	107.3	0.02	15.6	44.3
Suburban													
BL—U	73.7	6.0	6.72	9.5	0.25	92.2	26.4	3.19	41.0	260.9	0.15	19.4	112.9
BL—L	32.3	2.6	3.55	4.9	0.17	42.7	14.8	1.79	20.7	173.9	0.05	10.5	59.2
CC—U	25.3	2.4	2.52	7.5	0.13	33.4	10.5	1.44	21.8	111.4	0.10	8.5	53.0
CC—L	65.7	3.7	5.68	14.9	0.24	70.1	22.4	2.91	34.9	257.2	0.13	18.5	94.8
AS—U	7.4	0.7	0.84	1.9	0.39	14.6	3.0	0.42	10.5	32.8	0.03	3.23*	33.9
AS—L	15.0	1.1	1.37	4.3	0.15	16.9	3.1	0.74	7.1	68.3	0.01	2.80*	22.6
MC—U	27.6	1.7	2.65	9.8	0.09	34.0	10.0	1.49	19.4	115.0	0.06	9.9	50.1
MC—L	69.4	4.0	6.08	20.5	0.16	79.3	25.0	3.48	31.6	435.5	0.13	23.0	101.9
MI—U	69.7	4.2	5.90	25.0	0.08	69.7	16.1	3.27	22.2	169.5	0.06	19.1	70.0
MI—L	66.1	3.8	5.84	18.2	0.07*	71.4	16.0	3.18	21.4	157.0	0.05	19.0	74.1
PC—U	71.4	3.4	5.59	17.0	0.00	64.8	16.7	2.80	19.6	327.1	0.05	17.5	67.2
PC—L	81.7	4.6	6.38	11.2	0.06*	71.1	22.4	2.75	25.2	183.5	0.08	19.0	87.4
SM—U	26.1	2.7	2.26	3.7	0.11	36.9	16.4	1.45	17.3	96.0	0.05	6.7	58.6
SM—L	66.8	5.1	5.29	18.2	0.28	84.9	47.6	3.22	36.0	212.8	0.13	23.3	140.2
YC—U	79.4	4.8	7.31	16.1	0.20	79.3	25.2	3.92	25.0	268.4	0.09	23.4	97.6
YC—L	74.1	3.8	7.27	15.7	0.19	78.4	25.1	3.74	24.6	289.7	0.07	23.7	99.6
Industrial													
DL—U	42.2	2.6	2.98	11.0	0.28	104.1	44.7	1.78	101.3	166.7	0.07	13.2	194.5
DL—L	47.5	3.2	4.29	14.5	0.36	77.2	36.3	2.71	84.3	337.6	0.10	17.6	100.9
KP—U	76.5	6.0	6.74	17.4	0.78	130.8	70.5	3.27	112.4	174.9	0.22	22.3	235.3
KP—L	63.9	4.3	6.08	18.5	0.34	94.7	67.2	3.41	76.5	277.1	0.21	22.3	190.6
SH—U	35.7	5.0	3.41	8.3	1.03	2873	68.1	2.00	109.4	150.6	0.14	50.7	394.5
SH—L	29.4	2.5	2.34	7.2	0.48	2027	18.9	1.32	26.7	133.6	0.05	43.8	143.0
Urban													
NM—U	51.2	4.8	5.76	24.6	0.40	99.8	58.4	3.93	153.6	227.5	0.21	18.3	198.3
NM—L	73.0	5.3	8.01	16.2	0.44	106.8	59.4	3.91	124.9	212.2	0.54	26.3	225.3
VR—U	54.2	6.6	5.83	21.5	1.32	160.9	93.7	3.66	68.0	197.0	0.48	23.2	422.0
Agricultural													
KW—U	66.8	4.0	4.87	14.3	0.11	60.4	19.0	2.66	23.2	194.1	0.12	16.6	63.8
KW—L	42.9	2.4	3.47	14.6	0.06*	45.5	10.3	2.00	13.4	242.4	0.04	12.3	51.4
Salt marsh unimpacted													
BS—U	74.8	3.8	6.56	21.7	0.00	72.9	20.3	3.26	22.4	252.6	0.07	18.1	76.5
BS—L	52.5	5.3	5.13	18.8	0.04*	59.7	14.6	2.79	17.0	224.9	0.04	15.1	61.6
GC—U	63.0	4.2	6.35	11.5	0.12	74.0	23.1	3.18	23.1	251.9	0.05	21.8	83.2
GC—L	39.2	2.9	3.93	13.6	0.12	52.6	14.6	2.08	15.0	183.3	0.02	14.6	63.2
Salt marsh impacted													
PI—U	71.8	5.9	8.54	16.2	0.20	94.1	30.1	4.28	22.5	205.1	0.10	28.1	111.7
PI—L	70.7	4.6	6.88	15.5	0.20	74.1	24.6	3.65	28.2	247.7	0.06	20.7	98.2
OG—U	78.1	6.3	7.01	13.0	0.28	92.3	32.4	3.41	31.7	169.0	0.13	22.6	109.6

The first two letters of the Site ID indicate the creek (see Figure 1 for abbreviations and names) and the last letter indicates the reach (U = upper and L = lower)
Clay, TOC, Al, Fe are percentages and all other concentrations are ppm (µg/g dry weight)
0.00 indicates the concentration was below the level of detection
An * indicates the concentration was below the level of quantification

Table 3. Summary of the trace metal concentrations, clay, and TOC content [mean and standard error (Avg^{StErr})] collected in the creek-marsh transect study

Site ID	Clay	TOC	Al	As	Cd	Cr	Cu	Fe	Pb	Mn	Hg	Ni	Zn
Rathall Creek (forested watershed)													
U-CB	$53.8^{11.8}$	$4.3^{0.8}$	$5.2^{1.0}$	$9.5^{0.7}$	$0.12^{0.02}$	$65.6^{12.3}$	$15.1^{3.0}$	$2.6^{0.4}$	$21.7^{3.9}$	$209.6^{9.5}$	$0.07^{0.01}$	$15.4^{3.3}$	$61.5^{11.2}$
U-B	$58.3^{5.2}$	$7.2^{0.2}$	$5.1^{0.2}$	$14.2^{1.1}$	$0.16^{0.01}$	$80.7^{0.6}$	$19.6^{0.7}$	$2.8^{0.1}$	$24.1^{0.4}$	$157.8^{2.3}$	$0.09^{0.01}$	$19.0^{0.5}$	$79.4^{3.3}$
U-MM	$54.9^{3.7}$	$5.6^{0.4}$	$5.3^{0.4}$	$13.0^{1.6}$	$0.15^{0.01}$	$89.4^{1.4}$	$17.4^{1.4}$	$2.7^{0.2}$	$22.4^{1.7}$	$149.0^{13.5}$	$0.09^{0.02}$	$16.7^{1.5}$	$68.4^{4.5}$
U-HM	$36.2^{5.5}$	$5.3^{0.8}$	$2.8^{0.4}$	$5.8^{1.1}$	$0.09^{0.01}$	$50.4^{4.4}$	$11.0^{2.0}$	$1.4^{0.2}$	$12.7^{1.6}$	$82.5^{9.3}$	$0.04^{0.01}$	$11.0^{1.8}$	$43.0^{5.6}$
L-CB	$66.1^{1.2}$	$4.2^{0.1}$	$5.9^{0.4}$	$13.1^{1.2}$	$0.13^{0.02}$	$73.8^{1.8}$	$22.8^{0.3}$	$3.1^{0.3}$	$22.2^{0.8}$	$241.3^{11.8}$	$0.08^{0.01}$	$20.9^{0.5}$	$86.2^{1.7}$
L-B	$65.1^{11.5}$	$4.8^{0.2}$	$6.6^{0.3}$	$17.6^{1.3}$	$0.14^{0.01}$	$92.3^{6.1}$	$25.5^{0.2}$	$3.5^{0.2}$	$27.5^{1.7}$	$377.7^{11.2}$	$0.11^{0.01}$	$23.3^{0.6}$	$98.5^{1.2}$
L-MM	$69.5^{5.6}$	$6.9^{0.5}$	$5.3^{0.4}$	$10.3^{0.4}$	$0.14^{0.02}$	$84.7^{5.3}$	$19.8^{1.2}$	$2.3^{0.0}$	$22.2^{0.2}$	$110.0^{3.1}$	$0.07^{0.00}$	$18.3^{0.6}$	$72.3^{2.3}$
L-HM	$56.3^{8.1}$	$9.5^{0.7}$	$4.5^{0.2}$	$7.6^{0.8}$	$0.09^{0.01}$	$77.7^{1.3}$	$16.5^{1.4}$	$1.8^{0.1}$	$21.1^{1.0}$	$91.5^{1.4}$	$0.05^{0.01}$	$16.6^{1.8}$	$58.3^{4.5}$
Long Creek (forested watershed)													
U-CB	$27.9^{2.1}$	$2.4^{0.3}$	$2.4^{0.2}$	$3.3^{0.1}$	$0.09^{0.01}$	$30.2^{1.7}$	$6.6^{0.2}$	$1.3^{0.1}$	$9.9^{0.1}$	$96.3^{4.0}$	$0.04^{0.01}$	$7.9^{0.4}$	$29.9^{1.9}$
U-B	$28.7^{5.1}$	$2.6^{0.3}$	$2.2^{0.3}$	$3.0^{0.4}$	$0.00^{0.01}$	$27.6^{3.0}$	$5.6^{0.8}$	$1.1^{0.1}$	$10.4^{1.3}$	$91.8^{1.9}$	$0.04^{0.01}$	$5.6^{1.1}$	$24.5^{3.2}$
U-MM	$40.8^{3.8}$	$4.7^{0.3}$	$3.2^{0.1}$	$6.3^{0.5}$	$0.09^{0.00}$	$42.4^{1.7}$	$9.4^{0.3}$	$1.6^{0.0}$	$13.2^{1.1}$	$91.7^{4.7}$	$0.04^{0.01}$	$9.3^{0.4}$	$37.0^{1.1}$
U-HM	$28.8^{2.0}$	$4.7^{0.7}$	$2.0^{0.1}$	$3.9^{0.7}$	$0.10^{0.00}$	$26.2^{2.1}$	$6.3^{0.4}$	$1.0^{0.1}$	$9.8^{0.2}$	$78.5^{7.3}$	$0.04^{0.00}$	$6.0^{0.7}$	$24.3^{1.9}$
L-CB	$26.3^{11.6}$	$1.2^{0.6}$	$2.0^{1.0}$	$4.9^{2.4}$	$0.04^{0.02}$	$28.5^{12.0}$	$4.7^{2.7}$	$1.1^{0.5}$	$10.1^{3.9}$	$108.1^{34.5}$	$0.04^{0.02}$	$5.3^{3.7}$	$25.1^{10.8}$
L-B	$65.7^{9.4}$	$3.5^{0.3}$	$5.5^{1.0}$	$12.4^{2.0}$	$0.09^{0.01}$	$68.8^{12.8}$	$13.0^{2.4}$	$2.7^{0.5}$	$19.7^{2.4}$	$210.6^{26.5}$	$0.09^{0.01}$	$17.4^{2.4}$	$62.6^{9.8}$
L-MM	$52.9^{5.9}$	$3.9^{0.3}$	$4.6^{0.6}$	$9.4^{1.0}$	$0.08^{0.00}$	$60.4^{7.7}$	$11.5^{0.6}$	$2.4^{0.3}$	$17.7^{1.2}$	$146.1^{10.9}$	$0.04^{0.02}$	$15.3^{1.5}$	$52.9^{5.4}$
L-HM	$34.9^{8.8}$	$5.4^{1.5}$	$2.8^{0.8}$	$5.7^{1.2}$	$0.10^{0.02}$	$36.4^{9.5}$	$8.0^{2.3}$	$1.4^{0.4}$	$14.4^{3.5}$	$95.9^{11.3}$	$0.04^{0.01}$	$9.0^{3.0}$	$33.4^{9.8}$
Diesel Creek (industrial watershed)													
U-CB	$35.8^{3.5}$	$4.2^{1.9}$	$2.4^{0.3}$	$18.2^{3.9}$	$0.32^{0.02}$	$144.6^{28.4}$	$51.4^{5.3}$	$1.7^{0.1}$	$109.9^{45.3}$	$136.2^{17.3}$	$0.09^{0.01}$	$11.2^{2.0}$	$168.7^{25.8}$
U-B	$67.8^{0.3}$	$5.5^{0.2}$	$6.1^{0.4}$	$15.2^{1.1}$	$0.22^{0.06}$	$142.2^{18.9}$	$69.3^{24.0}$	$3.0^{0.2}$	$85.6^{30.1}$	$186.9^{25.2}$	$0.13^{0.03}$	$23.8^{1.3}$	$168.8^{14.9}$
U-MM	$67.7^{4.4}$	$14^{3.9}$	$3.9^{1.1}$	$16.7^{3.2}$	$0.74^{0.35}$	$270.2^{68.8}$	$77.0^{32.9}$	$2.3^{0.3}$	$93.6^{12.0}$	$133.9^{17.2}$	$0.24^{0.07}$	$28.7^{4.0}$	$211.0^{48.6}$
U-HM	$64.0^{3.7}$	$13.1^{2.3}$	$3.7^{1.1}$	$17.5^{3.8}$	$0.43^{0.11}$	$203.5^{41.2}$	$88.3^{55.1}$	$2.5^{0.2}$	$102.2^{30.0}$	$200.6^{100.4}$	$0.16^{0.04}$	$19.5^{3.3}$	$204.6^{44.3}$
L-CB	$62.6^{8.5}$	$4.1^{0.5}$	$5.4^{0.6}$	$17.5^{1.6}$	$0.34^{0.01}$	$91.8^{7.8}$	$37.6^{0.7}$	$3.1^{0.3}$	$60.7^{11.8}$	$318.8^{34.2}$	$0.11^{0.01}$	$22.1^{2.4}$	$127.4^{13.8}$
L-B	$70.7^{2.9}$	$5.1^{0.2}$	$5.9^{0.5}$	$15.7^{0.6}$	$0.19^{0.01}$	$114.0^{13.3}$	$31.1^{0.8}$	$3.0^{0.2}$	$37.8^{3.3}$	$210.9^{36.3}$	$0.14^{0.01}$	$22.3^{0.8}$	$107.3^{3.2}$
L-MM	$32.6^{4.5}$	$4.0^{0.6}$	$3.3^{0.2}$	$10.4^{1.0}$	$0.20^{0.03}$	$170.7^{14.7}$	$17.6^{1.2}$	$1.6^{0.1}$	$28.4^{1.8}$	$88.1^{14.0}$	$0.07^{0.00}$	$13.3^{0.1}$	$67.3^{1.3}$
L-HM	$12.9^{3.0}$	$2.9^{1.5}$	$2.3^{1.1}$	$7.3^{1.6}$	$0.17^{0.01}$	$139.9^{9.1}$	$13.8^{5.5}$	$1.1^{0.5}$	$17.9^{6.7}$	$75.0^{26.1}$	$0.06^{0.03}$	$12.2^{3.4}$	$51.7^{17.3}$
Shem Creek (suburban watershed)													
U-CB	$30.7^{2.5}$	$2.9^{0.3}$	$3.4^{0.8}$	$8.4^{0.6}$	$0.16^{0.02}$	$48.0^{7.1}$	$27.7^{3.5}$	$2.0^{0.4}$	$23.6^{0.6}$	$128.8^{11.8}$	$0.06^{0.01}$	$11.6^{2.2}$	$89.4^{13.6}$
U-B	$63.3^{1.9}$	$7.0^{0.4}$	$5.9^{0.5}$	$15.2^{0.6}$	$0.14^{0.01}$	$82.2^{5.9}$	$46.4^{3.7}$	$2.8^{0.2}$	$37.1^{1.1}$	$175.6^{20.8}$	$0.14^{0.01}$	$20.3^{0.8}$	$124.7^{1.0}$
U-MM	$72.6^{3.1}$	$8.9^{0.6}$	$5.7^{0.2}$	$14.9^{1.0}$	$0.17^{0.01}$	$71.5^{0.6}$	$36.6^{0.7}$	$2.9^{0.1}$	$34.3^{0.2}$	$120.0^{1.3}$	$0.12^{0.01}$	$18.8^{0.5}$	$110.5^{2.7}$
U-HM	$68.7^{2.8}$	$13.2^{1.6}$	$5.1^{0.5}$	$14.1^{0.8}$	$0.17^{0.06}$	$67.0^{4.9}$	$33.4^{1.1}$	$2.3^{0.1}$	$32.8^{2.0}$	$95.5^{1.4}$	$0.11^{0.01}$	$17.2^{1.3}$	$108.5^{5.9}$
L-CB	$57.1^{13.3}$	$4.6^{0.4}$	$4.6^{0.6}$	$18.6^{2.5}$	$0.20^{0.04}$	$68.9^{9.2}$	$36.4^{6.5}$	$2.8^{0.3}$	$29.6^{4.6}$	$180.4^{28.3}$	$0.11^{0.01}$	$17.7^{3.7}$	$112.0^{17.1}$
L-B	$69.5^{0.6}$	$5.8^{0.1}$	$6.4^{0.4}$	$16.9^{2.7}$	$0.17^{0.03}$	$94.6^{7.6}$	$45.7^{2.2}$	$3.2^{0.2}$	$32.9^{0.7}$	$199.1^{18.3}$	$0.14^{0.01}$	$22.0^{0.2}$	$132.9^{4.3}$
L-MM	$64.1^{1.5}$	$6.7^{0.3}$	$5.1^{0.4}$	$15.3^{1.1}$	$0.19^{0.01}$	$75.6^{3.8}$	$37.7^{2.4}$	$3.0^{0.1}$	$29.8^{0.9}$	$146.9^{10.0}$	$0.11^{0.01}$	$18.6^{1.6}$	$115.3^{6.6}$
L-HM	$51.7^{18.3}$	$5.8^{1.5}$	$4.1^{1.5}$	$12.2^{4.2}$	$0.14^{0.07}$	$61.0^{20.4}$	$30.3^{11.1}$	$2.2^{0.8}$	$25.7^{7.5}$	$121.1^{38.4}$	$0.10^{0.04}$	$15.6^{5.3}$	$90.9^{32.5}$

The first letter in the Site ID indicates the reach (U = upper and L = lower) and the second letter(s) indicates the transect position (CB = creek bed, B = berm, MM = midmarsh, HM = high marsh). Clay, TOC, Al, Fe are percentages and all other concentrations are ppm (µg/g dry weight)

The spatial distribution of TOC was also heterogenous among the four intensively studied representative creeks sampled in the creek-marsh transect study (Table 3). The TOC content was significantly lower in Long Creek (3.6 ± 0.3) than in Rathall (6.0 ± 0.4), Diesel (6.6 ± 1.0), or Shem (6.9 ± 0.7) Creeks. Different patterns of significance across the transect positions occurred within each creek. In general, TOC tended to be highest on the marsh surface (*i.e.,* midmarsh and high marsh).

The clay and TOC content of the sediments were significantly correlated with the trace metal concentrations for all sites sampled (*i.e.,* tidal creek study and creek-marsh transect study) (Table 4). Stronger correlations (*i.e.,* higher r^2s) were found between trace metal concentrations and the clay content than the TOC content of the sediment. The metals Al, Fe, As, and Mn had similar regression models with the clay content for the reference sites alone, the developed sites alone, and the combined data sets (Table 4). The metals Ni, Cu, Cr, Pb, Zn, Cd, and Hg had stronger correlations with the clay content for the reference sites alone than either the developed or combined data sets (Table 4). In addition, the regression models (*i.e.,* slopes, y-intercepts) for those metals were different between the reference and developed data sets. This finding suggests anthropogenic sources of these metals in the developed watersheds had altered the trace metal–particle size/TOC relationships.

The comparisons of trace metal concentrations among watershed classes, reaches, and across the creek-marsh system were evaluated without adjusting for sediment particle size or TOC content. We felt the data and results were more valuable to other investigators as absolute concentrations. In addition, the degree and type of development in the watershed altered the relationship between the sediment particle size and the trace metal concentrations suggesting that simple linear regressions would not be an adequate correction technique.

In the tidal creek study, the two-way ANOVA models were not significant for Al, Fe, As, Ni, and Mn, indicating there were no differences in concentrations of these trace metals among watershed classes (Figure 3B and Figure 4B). However, concentrations of the metals Cu, Cr, Pb, Zn, Cd, and Hg were significantly higher in the industrial/urban watershed class compared to the suburban and forested classes (Figure 3C). Trace metal concentrations in the suburban and forested

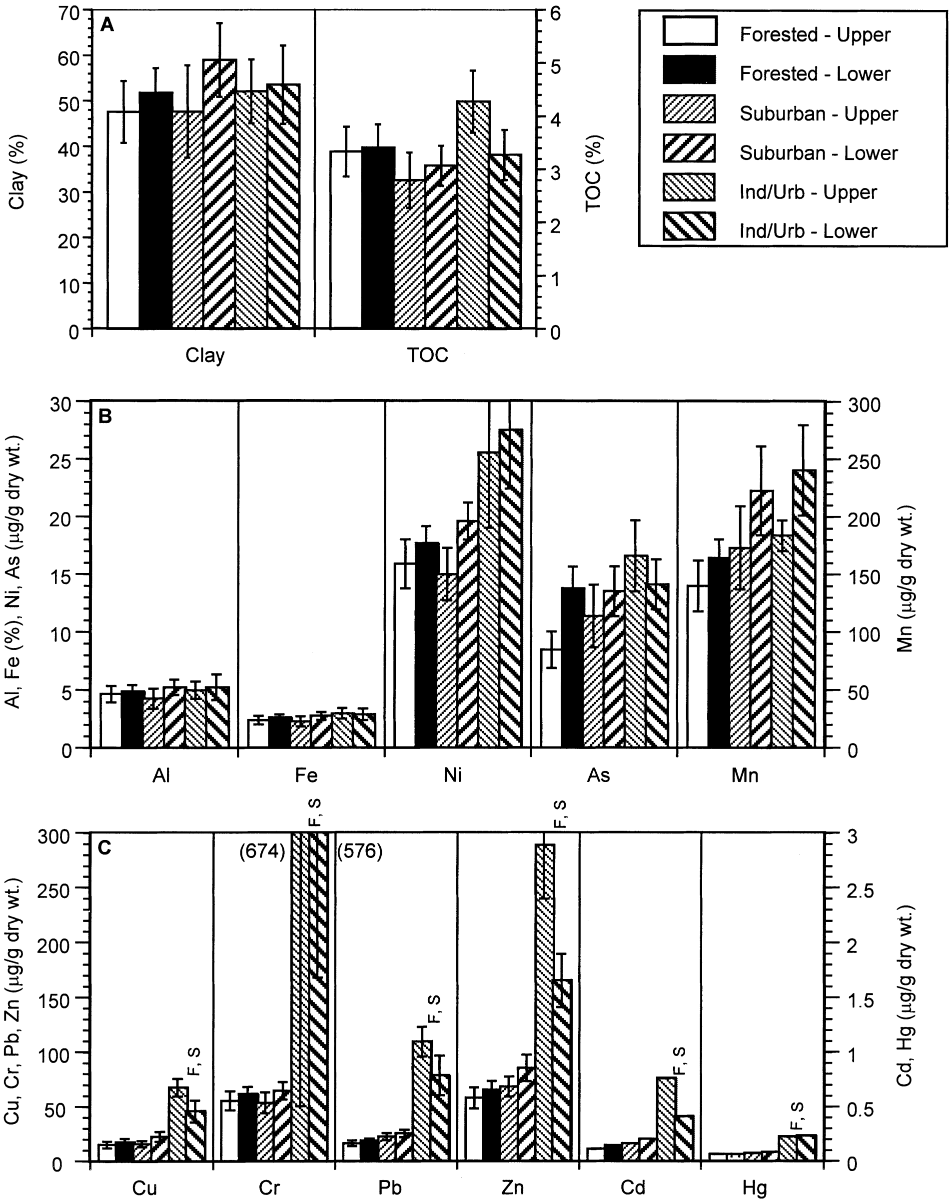

Fig. 3. The mean and standard error of the sediment characteristics (A) and trace metal concentrations (B and C) for the forested, suburban, and industrial/urban (ind/urb) watershed types by reach. F or S indicate the watershed class (combined upper and lower reach effect) was significantly higher than the forested or suburban classes, respectively

watershed classes were similar. Sediment trace metal concentrations (except Cd) were similar for impacted and unimpacted salt marsh creeks (Figure 4). The reach effect and the watershed by reach interaction were not significant for any of the metals evaluated. The upper reaches of the industrial/urban class generally had higher concentrations of the metals Cu, Cr, Pb, Zn, and Cd than the lower reaches (Figure 3C); however, these differences were not significant in a one-way ANOVA.

In the creek-marsh transect study, Long Creek (reference) had significantly lower concentrations for all metals than Rathall (reference), Diesel (industrial), and Shem (suburban) Creeks. Diesel and Shem Creeks had significantly higher concentrations of the metals As, Cu, Pb, Zn, and Hg than Rathall Creek (Table 5). In addition, the Pb concentrations were significantly higher in Diesel Creek than Shem Creek. Cr and Cd concentrations were significantly higher in Diesel Creek compared to Shem and Rathall Creeks, which had similar concentrations. Concentrations of Al, Fe, Ni, and Mn were either not different between the three creeks, or Rathall and Shem Creeks had higher concentrations compared to Diesel Creek (Table 5).

Within each of the four intensively studied creek systems, the clay content and the Al concentrations were concordant or in agreement from the creek channel to the marsh-upland interface (Figure 5A). The highest Al concentrations and clay content generally occurred in the berm and/or midmarsh transect positions, indicating fine sediment particles consistently accumulated at these sites. In Rathall, Long, and Shem Creeks, all of the trace metal concentrations were concordant with the Al and clay values (*i.e.*, highest values on the berm and/or midmarsh) for their respective transect position and reach (Figure 5).

Diesel Creek, the representative industrial watershed, had a number of trace metals (As, Cu, Cr, Pb, and Zn) whose spatial patterns were not concordant with sediment clay content or Al concentrations (Figure 5). In contrast, the metals Fe, Ni, and Mn had concordant patterns to the Al concentration. The

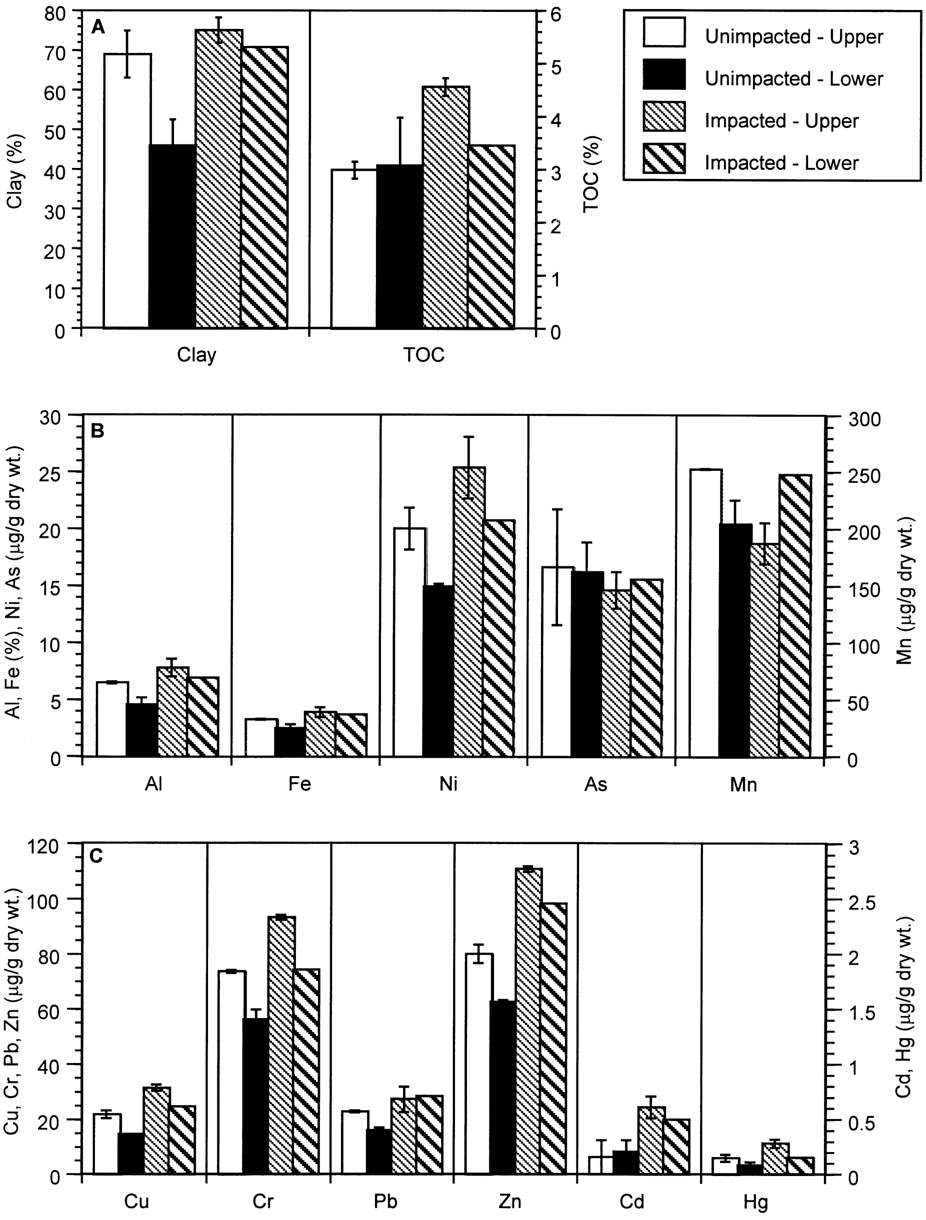

Fig. 4. The mean and standard error of the sediment characteristics (A) and trace metal concentrations (B and C) for the salt marsh unimpacted and impacted watershed types by reach. Note: There were no significant differences between the impacted and unimpacted classes of creeks

concentrations of the metals Cu, Cr, Pb, and Zn in the upper reach of Diesel Creek were two to three times higher than values found at similar locations in Rathall, Long, Shem Creeks and were significantly higher than values in the lower reach of Diesel Creek.

Variability in the trace metal concentrations within the tidal creek–salt marsh systems was dependent on the type of watershed development and the trace metal. For example, in the upper reach of Diesel Creek, the Cu concentrations in the creek channel were consistently high; however, the variability across the marsh platform was large (Figure 2A). The first transect had the highest concentration on the berm, the second transect had the highest concentration on the midmarsh, and the third transect had the highest concentration on the high marsh. This spatial heterogeneity was probably related to the pattern of historical contamination of the marsh platform of Diesel Creek. This high spatial variation of Cu was not found in the lower reach of Diesel Creek. The variability of Cu concentrations in Rathall, Long, and Shem Creeks was low in both the upper and lower reaches (Figure 2B). In general, the variability in metal concentrations within a system was similar in magnitude to the variability observed within their respective population of creeks (*i.e.*, forested, suburban, industrial/urban).

Discussion

The tidal creek–salt marsh complex is a dominant geographical feature of the South Carolina coast, comprising 50–70% of the estuarine area (Nummedal *et al.* 1977). These systems are important nursery and feeding grounds for fish and crustaceans (Hackney *et al.* 1976; Weinstein 1979; Wenner and Beatty 1993). The degree and type of development that occurs on the watersheds draining into these valuable natural resources influences trace metal distributions and concentrations. Levels of Cu, Cr, Pb, Zn, Cd, and Hg were significantly higher in the

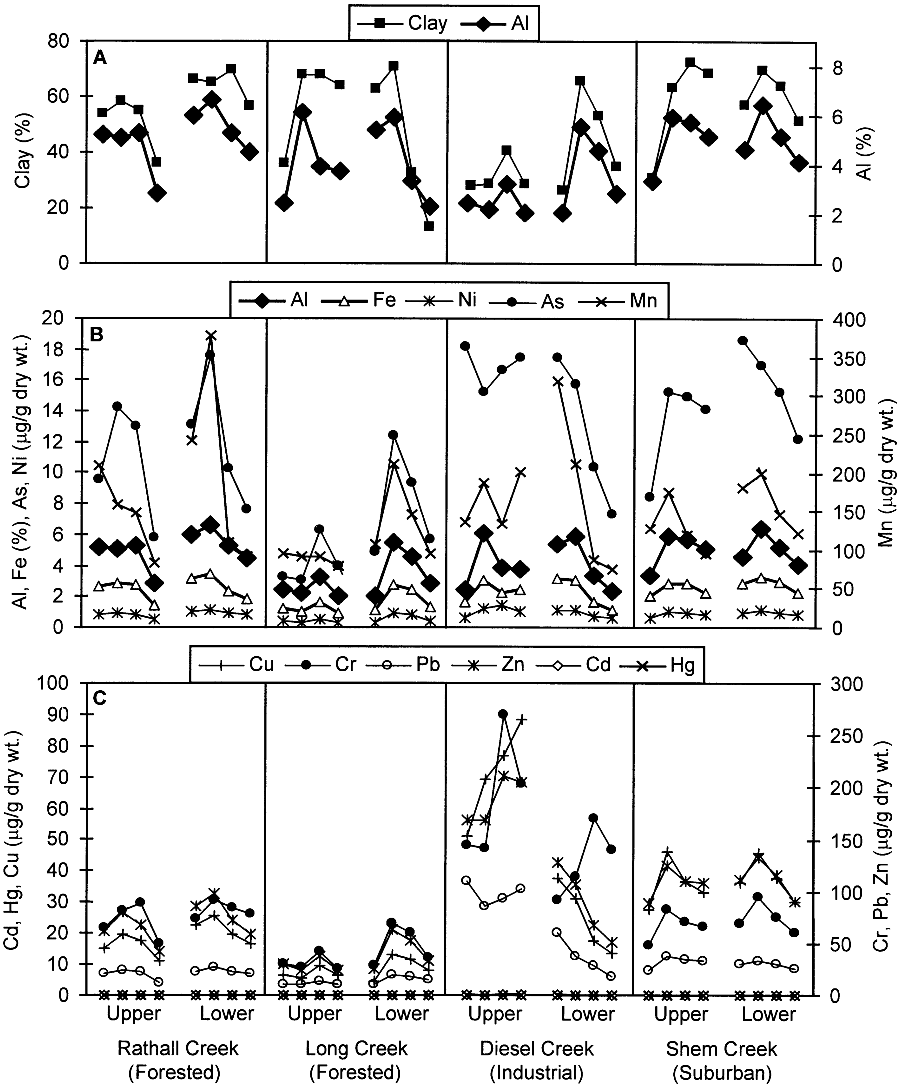

Fig. 5. Sediment grain size (A) and trace metal (B and C) distributions from the creek channel to the marsh-upland interface within each creek and reach. The four values for each group represent the average for the creek bed, berm, midmarsh, and high marsh proceeding from left to right

Table 4. Regression results of the trace metal concentrations and the clay content or total organic carbon (TOC) content

	Clay						TOC					
	Reference		Developed		Ref/Dev		Reference		Developed		Ref/Dev	
	r^2	p value	r^2	p value	r^2	p value	r^2	p value	r^2	p value	r^2	p value
Al	0.80	0.0001	0.65	0.0001	0.71	0.0001	0.37	0.0001	0.11	0.0039	0.19	0.0001
As	0.61	0.0001	0.53	0.0001	0.54	0.0001	0.19	0.0002	0.24	0.0001	0.22	0.0001
Cd	0.34	0.0001	**0.00**	**0.6924**	0.09	0.0003	0.36	0.0001	0.05	0.0452	0.15	0.0001
Cr	0.78	0.0001	**0.02**	**0.2874**	0.17	0.0001	0.50	0.0001	0.08	0.0164	0.19	0.0001
Cu	0.77	0.0001	0.29	0.0001	0.43	0.0001	0.51	0.0001	0.29	0.0001	0.35	0.0001
Fe	0.76	0.0001	0.66	0.0001	0.71	0.0001	0.28	0.0001	0.15	0.0005	0.21	0.0001
Pb	0.74	0.0001	0.18	0.0002	0.31	0.0001	0.49	0.0001	0.31	0.0001	0.34	0.0001
Mn	0.47	0.0001	0.38	0.0001	0.42	0.0001	**0.04**	**0.0831**	**0.01**	**0.3803**	**0.03**	**0.0513**
Hg	0.49	0.0001	0.36	0.0001	0.40	0.0001	0.26	0.0001	0.40	0.0001	0.34	0.0001
Ni	0.77	0.0001	0.49	0.0001	0.61	0.0001	0.42	0.0001	0.23	0.0001	0.31	0.0001
Zn	0.81	0.0001	0.28	0.0001	0.42	0.0001	0.39	0.0001	0.35	0.0001	0.34	0.0001

The bold values indicate the regression was not significant at $\alpha = 0.05$

urban and industrial watershed class than in the suburban and forested classes of creeks. This group of trace metals is commonly enriched in sediments due to anthropogenic sources (Bruland *et al.* 1974; Erlenkeuser *et al.* 1974; Goldberg *et al.* 1977; Kennish 1992). Levels of Al, Fe, Ni, As, and Mn were found to be similar among all watershed classes. This group of trace metals is primarily derived from natural sources (Bruland *et al.* 1974; Erlenkeuser *et al.* 1974; Goldberg *et al.* 1977).

Table 5. Significant differences from the three-way ANOVA least squares mean analyses comparing representative creek-marsh systems

	Shem vs. Rathall	Diesel vs. Rathall	Diesel vs. Shem
Clay	=	=	<
TOC	=	=	=
Al	=	<	<
Fe	=	=	<
Ni	=	=	=
As	>	>	=
Mn	=	=	=
Cu	>	>	=
Cr	=	>	>
Pb	>	>	>
Zn	>	>	=
Cd	=	>	>
Hg	>	>	=

Rathall Creek = reference; Diesel Creek = industrial/urban; Shem Creek = suburban

Long Creek is not shown because it was significantly lower than the other three creeks for all variables

An = indicates there were no significant differences, > indicates the first creek listed was significantly greater than the second creek, and a < indicates the first creek listed was significantly less than the second creek

The enrichment of specific trace metals in the tidal creek systems studied appears to be mainly related to past and present development activities. Industrial activities including metal fabrication facilities, historical power plants (*i.e.,* no longer in existence), fuel storage and distribution facilities, historical wood preservation facilities, phosphate mining, fertilizer plants, and ship building/overhauling facilities are all potential sources of trace metals to tidal creek and salt marsh systems for the industrial and urban watersheds sampled. A number of these activities occurred in the Diesel Creek watershed, the representative industrial/urban creek-marsh system examined. The anthropogenically enhanced trace metals in Diesel Creek were primarily found on the midmarsh and high marsh of the upper reaches, despite the deposition of fine particles and ‘‘natural’’ trace metals near the creek channel. This spatial pattern of trace metal enrichment suggests the upper reach of Diesel Creek was probably used as an undocumented disposal area for industrial wastes, particularly fly ash.

In contrast, suburbanized watersheds with medium to low density housing did not have elevated levels of metal contamination compared to the reference creeks. This is not surprising because suburban development does not generate large amounts of trace metals (Fortner *et al.* 1996). The representative suburban creek-marsh system, Shem Creek, had elevated levels of As, Cu, Pb, Zn, and Hg compared to Rathall Creek and Long Creek. This is not surprising since Shem Creek had a relatively high human population density (*i.e.,* 15.7 individuals per ha), which included commercial development (*e.g.,* shopping centers) and a major highway corridor (*i.e.,* US 17). In addition, the bottom portion (*i.e.,* ~3 km from our lowest sampling site) of Shem Creek is used as a dockage facility for ~50 commercial fishing vessels and dry stack boat storage yard. Increased Cu levels may have been associated with antifouling paints used by the many recreational and commercial vessels in this creek (US EPA 1985; Kennish 1992). Pb was associated with lead-based fuels, and Zn may have been associated with sacrificed anodes used on boats as well as from tire wear from roads (Cole *et al.* 1984; US EPA 1985; Maltby *et al.* 1995). Increased levels of As and Hg in Shem Creek are harder to explain. As is associated with phosphate mining and fertilizer production. These processes have historically occurred on the Ashley River. The location of Shem Creek in the bottom of the estuary may have influenced the As contamination of this creek. The slight increase in Hg levels may be the result of localized atmospheric fallout. The ‘‘anthropogenic’’ trace metal concentrations in Shem Creek and the two reference creeks were highest in areas with the highest levels of fine-grained sediments and ‘‘natural’’ trace metals. The overall low concentrations of trace metals observed in Long Creek were probably due to the low clay content and Al concentrations.

Overall, the association between trace metals and fine-grained sediments were found to be strongest in the reference systems. This association decreased in developed watershed systems suggesting that development activities enhanced the levels of trace metals. A reduction in the strength of metal-clay correlations was also reported for salt marsh sediments near human development in the Scheldt Estuary in The Netherlands (Beeftink *et al.* 1982). In the current study, tidal creeks and salt marshes were repositories of fine-grained sediment and trace metals, which is also in agreement with other studies (Beeftink *et al.* 1982; Fletcher *et al.* 1994; Williams *et al.* 1994). In addition, the hydrodynamic processes of these systems (*i.e.,* large tidal flushing, storm events) potentially resuspends the trace metals and fine-grained sediments causing the creeks, especially the industrial and urban watershed creeks, to have the potential to be a conduit of contamination to other regions of the estuary.

Trace metal deposition and resuspension in estuaries are dependent on a complex suite of physical, chemical, and biological factors (Olsen *et al.* 1982; Luoma 1989; Fletcher *et al.* 1994). Salinity is one example of a physical factor that can affect the association between trace metals and fine-grained sediments. The tidal creeks sampled in this study represented the broad salinity distributions (mesohaline to euhaline) that exist in the majority of South Carolina tidal creek systems. Within individual creeks studied, the salinity can fluctuate 5–15 ppt over a tidal cycle and from oligohaline (0.5–5 ppt) to euhaline (>25 ppt) during a single rain event. The largest fluctuations in salinity generally occurred in creeks located on developed watersheds (Holland *et al.* 1997).

The classification of creeks by land use type enabled the comparison of creeks with development in their watersheds to forested reference creeks that had similar predevelopment features rather than comparing them to salt marsh systems, which potentially have different groundwater flow, freshwater runoff, elevational differences, and sediment composition (*i.e.,* higher clay content). An understanding of the land cover in the surrounding watersheds proved valuable in interpreting the differences in watershed classes and the spatial patterns of trace metal contamination.

The tidal creek study sampling design enables statements to be made about classes of creeks with different types of land use in their watersheds, but does not allow for statements about individual creeks because of the lack of replication within a creek. However, the creek-marsh transect study was designed to extensively sample representative systems from each watershed class to investigate the within creek spatial variability. Uncertainties associated with this study involve the chemical analyses of sediments; however, a rigorous quality assurance/quality control program ensured reliable data.

This is the first study to link trace metal contamination in tidal creek–salt marsh habitats with the surrounding activities in the watershed for a large number of systems. The quality of the data generated in this study allows for comparisons with other monitoring programs (*e.g.,* NOAA National Status and Trends Program [NS&T] and the EPA Environmental Monitoring and Assessment Program [EMAP]). In general, the forested, suburban, and industrial/urban watershed categories had higher mean trace metal concentrations (*i.e.,* Ni, As, Cu, Cr, Pb, Zn) than the mean concentrations observed along the southeastern U.S. coast as determined in the 1995 EMAP-Carolinian Province (EMAP-CP) study (Hyland *et al.* 1998). In addition, the mean concentrations of Cu, Cr, Pb, and Zn for the industrial/urban class of creeks were higher than the maximum concentration observed in the EMAP-CP data set. One explanation for the higher values observed in this study is the silt-clay content of the sediment was lower in the EMAP-CP study compared to this study. Only the industrial and urban classes of creeks in this study were found to have trace metal concentrations which exceeded the NS&T ‘‘high’’ (mean of the NS&T sites plus one standard deviation) and ‘‘5xhigh’’ concentrations (NOAA 1991). The mean concentration of the metals As, Cu, Pb, Zn, Cd, and Hg would be classified as high and the mean concentration of Cr would be classified as 5xhigh. The high mean Cr concentration resulted from values recorded in Shipyard Creek. Therefore, the tidal creek trace metal concentrations were higher than the subtidal southeastern coastal concentrations, but only the industrial and urban creeks would be above the mean and standard deviation for the coastal United States.

One of the major objectives of the Tidal Creek Project was to develop monitoring recommendations to evaluate the status and trends of chemical contaminants for these important ecological systems. This study chose to concentrate its sampling efforts in the midtide level of the creek channel, where the organisms that use creeks as nurseries are concentrated for a large portion (50–70%) of their day (Holland unpublished data). Therefore, the potential exists for the transferring of trace metals deposited on this habitat to higher trophic levels. In addition, the physical-chemical conditions (*e.g.,* hydrodynamic disturbance, sediment resuspension and movement, and anoxic conditions) that increase the bioavailability of trace metals are also more pronounced in the channel habitats than on the salt marsh platform. Furthermore, the influence of environmental factors (*e.g.,* elevation in the intertidal zone, water depth, distance from uplands and potential sources) are easier to control in the channels than on the marsh platform. The sediment composition and trace metal concentration patterns occurred on small scales within the individual systems extensively examined. Data from the creek channel had the most consistent and interpretable concentrations. Therefore, future monitoring studies, which hope to examine the impacts of watershed development on the intertidal estuarine areas, should at a minimum sample a relatively constant elevation along the creek channel.

In addition to the type of habitat to sample, the location within the length of the creek is especially important. Despite the lack of statistically higher trace metal concentrations in the upper reaches of the industrial/urban watershed class, there were statistical differences within Diesel Creek. Therefore, we recommend that monitoring programs take independent samples of the upper and lower reaches of the creek if possible. If only one sample can be collected within a creek, then it should be taken in the upper reach. This is due to the stronger association of this reach with development activities, which may make the data more interpretable.

The scope of this study was limited to the characterization of sediment trace metal concentrations and distributions. We have not attempted to evaluate the bioavailability and the trace metal effects on living resources and ecosystems. Further studies by these authors and other researchers will investigate the biological effects associated with the trace metal concentrations observed in this study. These data will also serve as a baseline for trace metal sediment contamination in South Carolina tidal creeks and salt marshes. This is particularly important since several of the forested reference sites used in this study are being rapidly developed (*e.g.,* Horlbeck and Rathall Creeks).

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