



Metal accumulation in salt marsh soils along the East Coast of the United States

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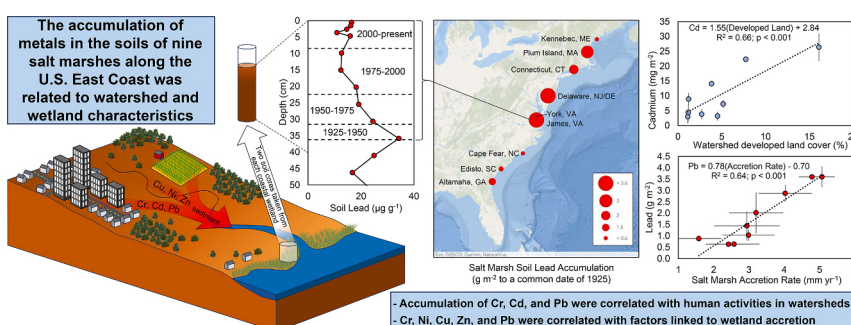
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HIGHLIGHTS

- The chronology of metal accumulation in nine salt marshes along U.S. East Coast was investigated.
- A laboratory experiment indicated that Cr, Ni, Cu, Zn, Cd, Pb, and U were immobile in salt marsh soils.
- Accumulation of Cr, Cd, and Pb in salt marsh soils was linked with human activities in the landscape.
- Cr, Ni, Cu, Zn, and Pb accumulation was correlated with wetland accretion.
- We observed declines in metal concentrations over recent decades at several sites.

GRAPHICAL ABSTRACT



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ABSTRACT

Coastal salt marshes are depositional environments that can accumulate pollutants introduced to the environment from human activities. Metals are a contaminant of concern in coastal environments due to their longevity and toxicity. We assessed metal concentrations and accumulation rates in nine salt marsh sites along the U.S. East Coast from Maine to Georgia. Following a metal mobility assay in organic-rich and mineral dominated salt marsh soils under aerobic/anaerobic and freshwater/saltwater conditions, we focused on profiles of chromium, nickel, copper, zinc, cadmium, lead, and uranium in two soil cores from each of the nine marshes that had previously been dated using lead-210 radioisotope techniques. We examined how land cover and the spatial distribution of land cover, marsh vertical accretion, and other watershed characteristics correlated with metal concentrations and depth/time-integrated accumulation of metals. We found statistically significant differences in metal concentrations and/or inventories between sites, with accumulation of metals positively correlated with both developed land cover in the watershed and rates of vertical accretion in the tidal marsh. The accumulation of chromium, cadmium, and lead were significantly correlated with developed land cover while the accumulation of chromium, nickel, copper, zinc, and lead were correlated with factors that determine sediment delivery from the landscape (e.g., riverine suspended sediment, soil erodibility in the watershed, and agricultural land cover skewed towards the coast) and measured wetland accretion rates. We observed declines in the concentration of

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many metals since 1925 at sites along the U.S. East Coast, indicating pollution mitigation strategies have succeeded in reducing metal pollution and delivery to the coastal zone. However, increasing rates of salt marsh vertical accretion over recent decades largely offset reductions in metal concentrations, resulting in rates of metal accumulation in coastal salt marsh soils that have not changed or, in some instances, increased over time.

1. Introduction

Coastal tidal salt marshes are productive ecosystems that exist at elevations close to local mean sea level (Morris et al., 2002). Global sea levels have risen by an average of about 3 mm yr⁻¹ over the past century with an acceleration in the rate of sea-level rise in recent decades in response to increased warming of the climate system (Church and White, 2006; Nerem et al., 2018). Salt marshes rely on the formation of new soils to build vertical elevation and keep pace with sea-level rise. Salt marsh soils are formed by the sequestration of organic matter derived largely from salt marsh plant production and from the trapping of sediments delivered to the marsh surface during tidal flooding (Morris et al., 2016). Sediments supporting marsh accretion can be supplied from oceanic sources and by rivers and streams delivering sediments from the terrestrial landscape (Baranes et al., 2022). Fluvial sediments can be an important source of material allowing tidal marshes to keep pace with sea-level rise, and depositional coastal environments that trap fluvial sediments can store the signals of changing watershed land uses within the vertical profile of salt marsh soils that have accumulated over time (Cochran et al., 1998).

The quality and quantity of materials delivered from the landscape to the coast can be influenced by human activities. Globally, soil erosion has accelerated as a result of deforestation, increased agricultural land use and urban development, and other human activities (Syvitski et al., 2005; Walling, 2006). Simultaneously, sediment availability to some coastal tidal wetlands has declined due to reductions in agricultural land use, soil conservation policies, and trapping of sediments in reservoirs (Weston et al., 2023). Urban development, agriculture, and industrial activities have altered water-quality and resulted in increased levels of pollutant delivery to coastal systems (Gedan et al., 2009; Kennish, 2002).

Toxic metals such as lead (Pb), cadmium (Cd), and zinc (Zn) are typically defined as trace metals that naturally occur at relatively low concentrations in the environment and can be toxic to living organisms (Smith and Huyck, 1997). Once metals are introduced into an environment, they are difficult to remove and can therefore become hazardous to organisms within the ecosystem (Lambers et al., 2009). The mobility and bioavailability of metals is influenced by a number of factors, including the concentration and speciation of the metal and the physicochemical status of the surrounding environment (Smith and Huyck, 1997; Ure, 1996; Williams et al., 1994). High organic matter availability in salt marsh soils fuels microbial respiration which, in saturated soils with limited oxygen diffusion, results in anaerobic, reducing conditions. Sulfate reduction is the dominant anaerobic microbial respiration pathway in most salt marsh soils due to the lack of oxygen and prevalence of sulfate (SO₄²⁻) in seawater (Capone and Kiene, 1988), and this metabolic process produces sulfide that can precipitate metals as metal sulfide compounds (Morse and Luther, 1999). Most metals are thought to be highly immobile under these conditions (Smith and Huyck, 1997), though there is some evidence of mobility for some metals in marsh soils (Wiese et al., 1997; Zwolsman et al., 1993).

There are numerous human activities that result in metal contamination of environmental systems, including agriculture (He et al., 2005; Herawati et al., 2000), industry (Lü et al., 2018; Shallari et al., 1998), boating (Turner, 2010), and urban development (Davis et al., 2001; Ward, 1990). The delivery of metal pollution from the landscape to coastal marsh ecosystems can be achieved through both river flow from the landscape (Rozan and Benoit, 1999) and atmospheric deposition (Cochran et al., 1998). Metals are among the more concerning pollutants

in coastal ecosystems (Gedan et al., 2009; Kennish, 2002). Prior studies have evaluated metals in salt marshes (e.g., Chen and Torres, 2012; Otero and Macias, 2002; Williams et al., 1994) as well as the impact on salt marsh plants (Duarte et al., 2010), animals (Goto and Wallace, 2010; Vinagre et al., 2008) and the microbial community (Mucha et al., 2013).

The purpose of this study was to assess metal mobility in salt marsh soils and to evaluate the record of metal pollution as stored in salt marsh soils along the U.S. East Coast. We focus on accumulation of seven metals [chromium (Cr), nickel (Ni), copper (Cu), Zn, Cd, Pb, and uranium (U)] in salt marsh soils based on mobility findings and concern about the toxicity and contamination of these elements in the environment (Smith and Huyck, 1997). We establish a chronology of metal accumulation in salt marsh soils from 1925 to present at nine sites from Maine to Georgia by pairing marsh soil dating data (from Weston et al., 2023) with soil metal content. We examine how rates of metal accumulation have changed over the past century and we relate metals in salt marsh soils to wetland and watershed characteristics to assess influences on metal accumulation in salt marshes over a broad geographic range.

2. Methods

2.1. Study sites, soil collection, and dating

This study was conducted in nine salt marsh sites along the U.S. East Coast ranging from Maine to Georgia. Salt marsh soils were collected from the Kennebec River (KEN), Maine, Plum Island Sound (PIE), Massachusetts, Delaware River (DEL), New Jersey/Delaware, York River (YRK), Virginia, James River (JMS), Virginia, Cape Fear River (CPF), North Carolina, Edisto River (EDS), South Carolina, and Altamaha River (ALT), Georgia (Fig. S1). The average annual temperature at the salt marsh sites, the average suspended sediment concentration of the river draining to the coastal zone, the average watershed slope, and soil erodibility are given in Weston et al. (2023). The vegetation was dominated by *Spartina patens* in the KEN, PIE, and DEL sites and by *S. alterniflora* (= *Sporobolus alterniflorus*) in the remaining sites. In each system, two soil cores were collected on the vegetated marsh platform in the mesohaline region of the salt marsh. Additional surficial soil (0–5 cm depth) was collected at the PIE and DEL sites for the metal mobility experiment (Fig. S2). Soil cores were sectioned into 1 cm (to a depth of 50 cm) or 2 cm (beyond 50 cm) depth increments. The soil bulk density was determined by weighing soil sections of known volume after drying (at 60 °C until no further change in weight was observed), and soil organic matter content was determined by measuring the weight change of a subsample of dried soil samples following combustion at 500 °C for four hours (Fig. S2; Craft et al., 1991). The soil sections were visually inspected for the presence of salt marsh plant material. The age of each soil section was determined by ²¹⁰Pb radiometric dating, as described in detail for these cores in Weston et al. (2023), wherein additional details on soil core location and collection may be found. To determine the chronology of metal accumulation in these salt marshes, a subset of the core sections from two cores at each site were analyzed for metal content. Each section from 0 to 5 cm was analyzed, and then sections approximately every 5–6 cm were selected to characterize the entire depth of each soil core.

2.2. Metal mobility experiment

Soils from the organic-rich PIE site and the more mineral-dominated DEL site, as defined by soil organic matter content, were used in a metal

mobility experiment to test how mobility of metals differed between soils with different organic content under freshwater/saltwater and aerobic/anaerobic conditions in a factorial manner (eight treatments). Wet soils from each site were thoroughly homogenized and subsampled (in triplicate) for analysis of initial metal content (Fig. S2). Homogenization disrupts biogeochemical and sedimentological zonation that may be important in determining metal mobility in salt marsh soils, but this approach allowed us to investigate how oxygen and salinity influenced mobility. A portion of each sediment sample were dried at 60 °C until no further weight change was detected, and the organic content was determined by weight change after loss on ignition (LOI) at 500 °C for four hours (Craft et al., 1991). A portion of the homogenized and dried soil from each site was analyzed for metal content as described below to determine the total amount of each metal in the mobility treatments (Fig. S2).

For the mobility experiment, approximately five grams of homogenized wet soil was placed in a 50-mL tube along with 25 mL of either deionized water or saltwater (with a salinity of 34 made with Instant Ocean® diluted with deionized water). Aerobic treatments were assembled with aerated dilution solutions and with air in the sample tube headspace (25 mL). Anaerobic treatments were prepared in a glovebox under a nitrogen atmosphere with dilution solutions purged with nitrogen gas to remove oxygen. Three replicates were prepared for each of the eight treatments ($n = 24$ total). The treatment tubes were placed on an orbital shaker table at room temperature (~ 18 °C) for a one-week incubation period. Following the incubation, the sample tubes were centrifuged and the porewater was filtered (0.45 μm PTFE) and analyzed for metal content. The metal content of both the freshwater and saltwater amendment solutions were measured. The amount of metal in solution for each treatment was compared with the total metal in the initial soil, yielding a measurement of the amount of each metal mobilized in each treatment and soil combination.

2.3. Elemental analysis

All soil samples for metal analysis (Fig. S2) were dried (60 °C) and ground with a metal-free ceramic mortar and pestle. A 0.5 g sample of soil was placed in a Teflon digestion tube along with 10 mL of trace metal grade nitric acid. The samples were digested in a Mars™ 6 One Touch microwave digestion system with a 20-min ramp and a 15-min hold at 175 °C. At least one acid blank containing 10 mL of nitric acid was included with each digestion. The digested sample was transferred to a volumetric flask and brought to a volume of 25 mL with deionized water. These samples were filtered (0.45 μm PTFE) and further diluted (1:10 by mass) with deionized water prior to analysis. Filtered water samples from the metal mobility experiment were diluted and analyzed without digestion. All samples were diluted to approximately 4 % nitric acid before analysis.

Standards of known elemental concentration (ranging from approximately 0.1 to 1000 $\mu\text{g L}^{-1}$) in 4 % nitric acid were prepared using an Agilent 8500–6940 multi-element stock standard that included silver (Ag), aluminum (Al), As, barium (Ba), beryllium (Be), calcium (Ca), Cd, cobalt (Co), Cr, cesium (Cs), Cu, iron (Fe), gallium (Ga), potassium (K), lithium (Li), magnesium (Mg), manganese (Mn), sodium (Na), Ni, Pb, rubidium (Rb), selenium (Se), strontium (Sr), thallium (Tl), U, vanadium (V), and Zn and a Ricca PSB10KW antimony (Sb) stock standard. Check standards, instrument blanks, and digest blanks were analyzed every approximately 10 samples.

2.4. Land cover analysis

Land cover was evaluated using Geographic Information Systems (GIS) for each of the watersheds draining to the nine study sites (Figs. S1 and S2). We assessed developed, agricultural (crop), and forested (combined deciduous, evergreen, and mixed forest) land cover from 1938 through 2016 back-casted by Sohl et al. (2016). The number of

dams in each watershed was determined from the National Inventory of Dams (<https://nid.sec.usace.army.mil/>), which includes dams above 15 m in height, and was supplemented by data for smaller dams acquired from state agencies in each of the watersheds (Rodriguez, 2018).

The potential impact of the spatial distribution of land cover types within each watershed on metals in coastal soils was evaluated by determining the skewness index (SI) value for developed, agricultural, and forested land covers as well as the SI for dams. The SI, developed by Mineau et al. (2015), is calculated as:

$$SI = \frac{FP_{LC}}{FP}$$

Where FP_{LC} is the weighted mean flow path distance from each pixel of the specific land cover to the coast in each watershed, and FP is the unweighted mean flow path distance for the entire hydrologic network in each watershed. An SI value >1.0 represents skewness away from the watershed outlet, while a value <1.0 represents skewness towards the watershed outlet (Mineau et al., 2015). We calculated the time-averaged land cover and SI for each land cover and for dams between 1938 and the year the soil core was collected (2014–2017). We further assessed potential point-sources of metal pollution within each of the watersheds using data from the United States Environmental Protection Agency (USEPA) Toxics Release Inventory (TRI), National Pollutant Discharge Elimination System (NPDES), and brownfield databases for 2017 (Jezyski, 2019). There was not enough information to assess historical changes in TRI, NPDES, and brownfield point-source pollution pathways, and thus only the data for 2017 were used in our analysis. The number of these potential point-sources of pollution in each watershed were normalized to the watershed area.

2.5. Data analysis

The total amount of each element in solution in the mobility experiment was compared to the total amount of metal in the initial soil subsamples, and the amount of the element mobilized, on a percentage basis, was calculated for each treatment. A general linear model of mobilization relative to soil type (organic/mineral), salinity (fresh/salt), and oxygen availability (aerobic/anaerobic) for each element was used to determine significant treatment/soil type effects ($\alpha = 0.05$). A Pearson correlation of relative mobilization factors across all elements elucidated elements with similar patterns of mobility from marsh soils.

Chronologies of soil Cr, Ni, Cu, Zn, Cd, Pb, and U content were determined by pairing the soil dating from Weston et al. (2023) with the metal analysis described above. Due to varying vertical accretion rates measured across sites (~ 2 – 5 mm yr^{-1} ; Weston et al., 2023), the same depth does not correspond to the same age at each site. Further, radiometric dating based on Pb-210 becomes error-prone for dates in excess of approximately 100 years (Appleby and Oldfield, 1992). To compare metal concentrations and accumulation rates of metals over a common and reliable timeframe, we determined the depth of a common date, 1925, across all cores. The depth-weighted average concentration ($\mu\text{g g}^{-1}$) of each metal above the 1925 time-horizon was calculated. The total inventory (mg m^{-2}) of each metal above this time horizon was calculated using the metal concentration ($\mu\text{g g}^{-1}$) and the soil dry bulk density (g cm^{-3} ; from Weston et al., 2023). Metal accumulation rates ($\text{mg m}^{-2} \text{yr}^{-1}$) were also determined for four discrete time periods (1925–1950, 1950–1975, 1975–2000, and 2000-core collection date).

Differences in depth-weighted average metal concentrations between sites (to a common date of 1925) were assessed with analysis of variance (ANOVA), followed by Tukey's post-hoc test ($\alpha = 0.05$). Linear regressions of metal concentration over time (from 1925) were used to determine significant change in metal concentrations. Pearson correlations were performed on the depth-weighted average metal concentrations across all sites and the soil inventories of each metal across sites to determine patterns in the relationships between metals.

To determine correlations between watershed characteristics and metals, we conducted a principal components (PC) analysis of the watershed characteristics with Varimax rotation and included PCs with Eigenvalues >1.0. Time-averaged developed, forested, and agricultural land cover and skewness, USEPA NPDES and TRI permits, brownfields, dams and dam skewness, temperature, slope, soil erodibility, riverine suspended sediment, and salt marsh accretion rates were used in the PC analysis. We then performed Pearson correlation between the PC scores and the depth-weighted average sediment concentrations and total metal inventories to a common date of 1925. Statistical analyses were conducted in SPSS® (IBM® Corporation).

3. Results

3.1. Metal mobility

The organic matter content as determined by loss on ignition of the organic (PIE) soils was 34 %, and the organic matter content of the mineral (DEL) soils was 19 %. Results from the mobility assay indicated a range of metal mobility potentials from salt marsh soils, and the metals demonstrated a variety of responses to organic/inorganic, salt/fresh, and aerobic/anaerobic conditions (Tables 1 and S1). Eight elements (V, Cr, U, Rb, Mn, As, Ga, and Ba) demonstrated significantly higher mobility from organic soils, and four of these same elements (V, Cr, Mn, and As) also exhibited greater mobilization under anaerobic conditions ($p < 0.05$; Group A in Table 1). In contrast, five elements (Pb, Ni, Co, Zn, and Tl) were mobilized more readily from mineral soils, with all but Tl also demonstrating significantly higher mobilization in aerobic conditions ($p < 0.05$; Group C in Table 1). There were an additional seven elements (Group B in Table 1; Cu, Sb, Cd, Ag, Se, Sr, and Cs) that did not clearly fall into either of these two categories.

Eight elements demonstrated a significant ($p < 0.05$) salinity effect on mobility, with six (V, Rb, Mn, Ba, Se, and Sr) more mobile under saline conditions with two (Ga and Cu) more mobile in freshwater treatments (Table 1). There were only two metals, Sb and Cs, that demonstrated no significant treatment effect ($p > 0.05$; Table 1). A correlation between mobility values across treatments for all elements (Table S2) indicated three groups of elements based on their mobility characteristics. The mobility patterns of V, Cr, U, Rb, Mn, As, Ga, Ba (Group A metals) were similar and highest from organic soils under anaerobic conditions (Tables 1 and S2). In contrast, the mobility patterns for Pb, Ni, Co, Zn, and Tl (Group C metals) tended to be correlated with each other and negatively correlated with Group A elements

(Table S2), and these elements demonstrated greater mobility from mineral soils under aerobic conditions (Table 1). Group B metals (Cu, Sb, Cd, Ag, Se, Sr, Cs) had more variable or no significant treatment response (Table 1) and their mobility patterns tended not to be well correlated with other elements (Table S2).

The metals V, Cr, Cu, Ag, Cd, Cs, Tl, Pb, and U exhibited low mobility ($< 1\%$ mobility) across all treatments (Table S1), while Co, Ni, Zn, and Ga also demonstrated low mobility under salt marsh conditions (saline and anaerobic; Fig. 1). Elements with low mobility in anaerobic and saline conditions would serve as potential candidates for assessing accumulation of pollutants in salt marsh soils. In contrast, Mn, As, Se, Rb, Sr, Sb, and Ba demonstrated higher ($> 1\%$) maximum (Table S1) and salt marsh treatment mobilities (Fig. 1).

3.2. Metals in salt marsh soils

We examined the chronologies (Fig. 2), depth-weighted average concentrations and total inventories (Fig. 3), and accumulation rates (Fig. 4) of Cr, Ni, Cu, Zn, Cd, Pb, and U in the nine sites along the U.S. East Coast (the full dataset is given in Table S4). Our analysis focused on the top 10–40 cm of soil in each core for which we were able to confidently date the soil sections using Pb-210 techniques and confirm the soils were salt marsh deposits as indicated by the soil bulk density, organic content (Fig. S3), and visual observations of salt marsh plant material. We observed a decline in concentration for many of the metals over recent decades in the top portion of the soil cores (Fig. 2). We evaluated significant change in metal concentrations since 1925 (the dashed line in Fig. 2) by pairing the dating information from Weston et al. (2023) with the measured metal concentrations. We observed significant declines over time in Zn at two sites, Cr and Cd at three sites, Cu at four sites, and Pb at five sites (Table 2; $p < 0.05$). Nickel and U increased significantly at the KEN site, with declines at the JMS (both Ni and U) and YRK (U only) sites. The concentrations of all metals declined at the JMS site, while there was no significant change in any metal with time in the three southeast coast sites (CPF, EDS, ALT; Table 2).

The depth-weighted average metal concentrations indicated significant differences between sites for Ni, Cu, Zn, and Pb ($p < 0.05$; Fig. 3). The DEL site had the highest concentrations of nickel and had higher zinc than all sites except for JMS ($p < 0.05$; Fig. 3). Integrating concentrations with depth (to a common date of 1925 and correcting for soil bulk density) yielded total soil inventories of each metal (mg m^{-2} ; Fig. 3) and accumulation rates for specific time periods ($\text{mg m}^{-2} \text{yr}^{-1}$; Fig. 4). There were significant site differences in the inventories of all

Table 1
Significant treatment effects from a general linear model of element mobilization from organic (Plum Island Sound) or mineral (Delaware River) soils under freshwater/saltwater and aerobic/anaerobic conditions. When there is a significant treatment effect, the treatment that resulted in higher element mobility is listed.

	Element	Site		Salinity		Oxygen	
Group A	V	Organic	<0.001	Salt	0.002	Anaerobic	<0.001
	Cr	Organic	<0.001		NS	Anaerobic	0.030
	U	Organic	0.001		NS		NS
	Rb	Organic	<0.001	Salt	<0.001		NS
	Mn	Organic	<0.001	Salt	0.029	Anaerobic	0.007
	As	Organic	0.010		NS	Anaerobic	0.044
	Ga	Organic	0.020	Fresh	0.010		NS
	Ba	Organic	0.041	Salt	0.004		NS
				Fresh	<0.001		NS
Group B	Cu		NS		NS		NS
	Sb		NS		NS		NS
	Cd	Organic	0.010		NS	Aerobic	0.002
	Ag		NS		NS		NS
	Se	Mineral	0.055	Salt	<0.001	Anaerobic	<0.001
	Sr	Mineral	0.002	Salt	<0.001		NS
	Cs		NS		NS	Aerobic	<0.001
Group C	Pb	Mineral	0.011		NS	Aerobic	0.045
	Ni	Mineral	0.004		NS	Aerobic	0.026
	Co	Mineral	0.001		NS	Aerobic	0.004
	Zn	Mineral	<0.001		NS	Aerobic	0.001
	Tl	Mineral	<0.001		NS		NS

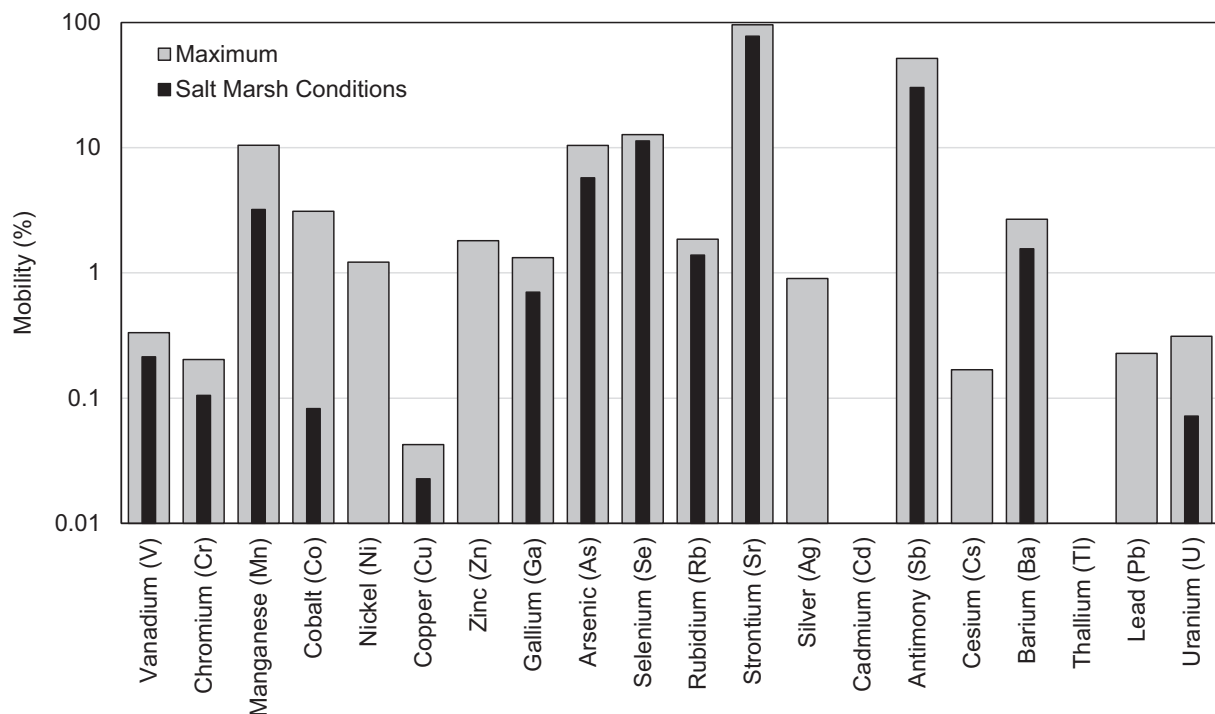


Fig. 1. Maximum mobilization measured for each element across all treatments, and the mobilization of each element in salt marsh (anaerobic and saline) conditions.

metals ($p < 0.005$) except for U ($p > 0.05$; Fig. 3). The PIE and DEL sites contained more Cr than the KEN, CON, YRK, CPF, or EDS sites. DEL had higher Ni inventories than all but the JMS site, and the DEL and JMS sites had higher Zn content than other sites ($p < 0.05$; Fig. 3).

A regression of time-specific metal accumulation rates (1925–1950, 1950–1975, 1975–2000, and 2000-core collection; Fig. 4) over time resulted in a different pattern than changes in concentration over time (Table 2). We observed significant declines in Cr, Zn, Cd, and Pb accumulation at PIE ($p < 0.05$; note that the 1925–1950 period for Pb was excluded), a decline in Pb at KEN, and a decline in Cd at the JMS site (Table 2). We also observed increased metal accumulation over time at several sites. Accumulation rates of Ni and Cu increased over time at the DEL and JMS sites, and Cr increased at the DEL site while Zn increased at the JMS site (Table 2). We also observed an increase in U accumulation at the CON site.

Depth-weighted average Ni, Cu, Zn, Cd, and Pb concentrations tended to be significantly correlated with each other ($p < 0.05$), while there was little correlation between Cr and U concentrations and the other metals (Table S3). Correlations between soil inventories of the metals tended to be stronger, with significant correlations between all metals ($p < 0.01$) except for U which was correlated with Cr, Cu, Cd, and Pb ($p < 0.05$), but not Ni or Zn ($p > 0.05$; Table S3).

3.3. Land cover

The general pattern of land cover change in all nine watersheds was of increasing development and declining agricultural land use (Fig. S4). Forest land cover increased throughout the 1900s in most watersheds (except for PIE and DEL which exhibited little change in forest cover) before starting to decline around the turn of the century in all watersheds (Fig. S4). The spatial pattern of land cover was more variable between watersheds, as demonstrated by the skewness index (SI, Fig. 5). Developed land cover was skewed towards the coast in the KEN, CON, DEL, and JMS watersheds but away from the coast in the CPF and ALT (Figs. 5 and S1). Agricultural land cover was skewed towards the coast in the KEN, DEL, YRK, JMS, and ALT watersheds, and away from the coast

in the PIE and EDS watersheds. Forest cover was skewed away from the coast in all watersheds, though more markedly in the DEL, YRK, and JMS (Fig. 5).

3.4. Relationship between salt marsh soil metals and watershed/wetland characteristics

The principal components (PC) analysis indicated that there were three components that explained about 84 % of the variability in watershed characteristics (Table 3). PC1 was linked to development, including higher developed land cover, NPDES and TRI permits, and more brownfields and dams (Fig. S4). PC2 was linked to agricultural and forest land cover patterns and can be characterized by higher agricultural and lower forested land cover, lower slopes, urban land cover skewed away from the coast, and higher annual average temperature in the coastal zone (Table 3). PC3 was characterized by factors that influence sediment delivery from the watershed and tidal marsh accretion rates and included higher agricultural and lower forested land cover towards the coast, higher riverine suspended sediment concentrations, higher watershed soil erodibility, and higher salt marsh accretion rates (Table 3; Fig. S5).

Depth-weighted average soil Cd and Pb concentrations were significantly positively correlated with PC1, and Ni, Zn, and Pb concentrations were correlated with PC3 ($p < 0.05$; Table 3). Soil inventories of Cr, Cd, and Pb were correlated with PC1, and inventories of Cr, Ni, Cu, Zn, and Pb were significantly correlated with PC3 ($p < 0.05$; Table 3). None of the metal concentrations or inventories were significantly correlated with PC2, and none of the PCs were significantly correlated with either concentrations or inventories of U ($p > 0.05$; Table 3).

4. Discussion

4.1. Metal mobility

The pattern of element mobility in salt marsh soils was characterized by metals that were mobilized from organic soils under anaerobic

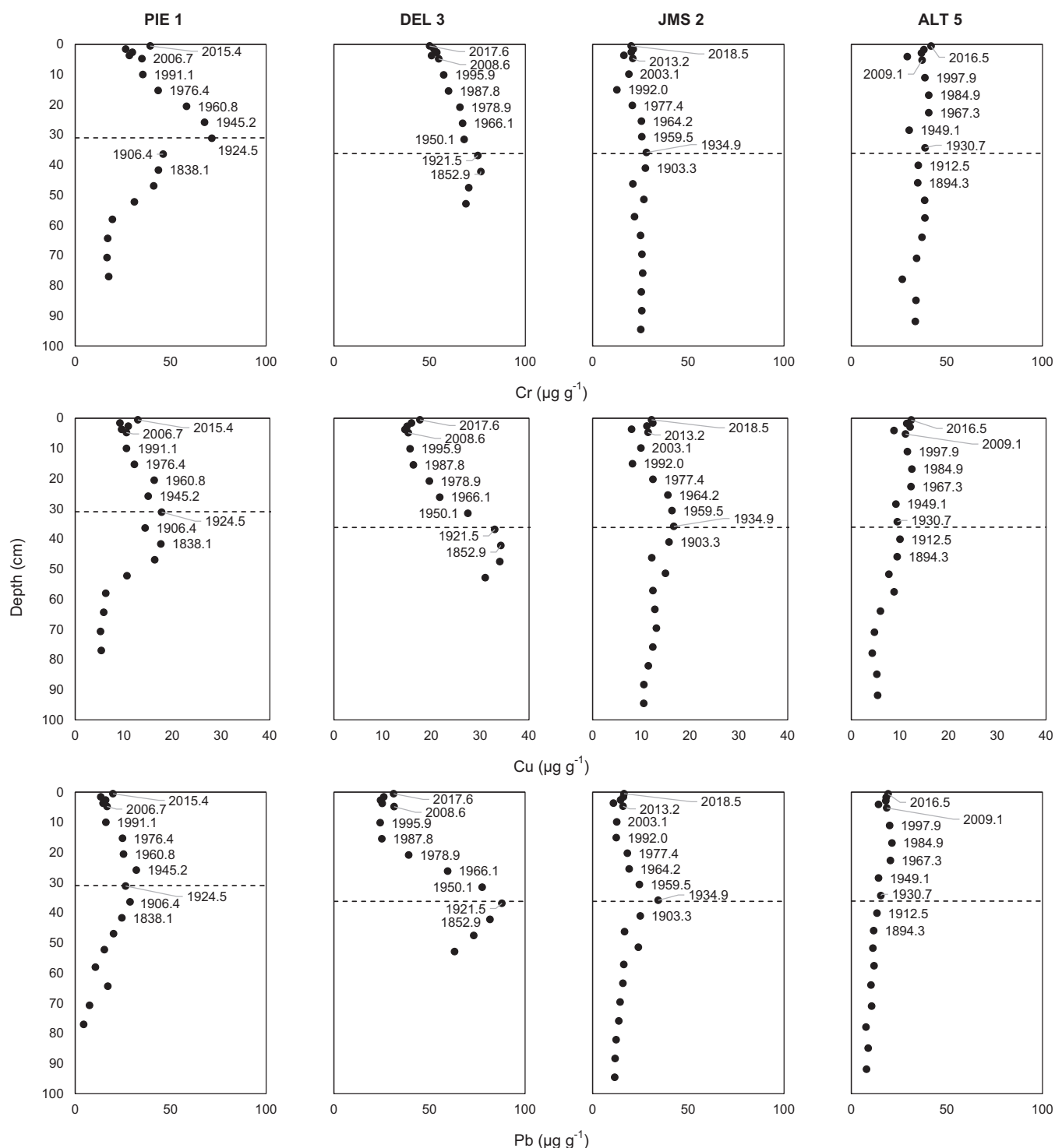


Fig. 2. Example of chromium (top), copper (middle), and lead (bottom) profiles in salt marsh cores collected from (left to right) the Plum Island, Delaware, James, and Altamaha estuaries. The dashed line indicates the 1925 date, above which depth-weighted average concentrations and soil inventories were calculated. Core dating is from [Weston et al. \(2023\)](#).

conditions (Group A) and elements that were mobilized from mineral soils under aerobic conditions (Group C; [Tables 1](#) and [S2](#)). These results likely reflect the form that the metals take in the soil environment and the mechanism(s) of mobilization from soils. The release of metals into solution may be as a result of processes including ion exchange, dissolution reactions, and organic matter mineralization. Metals can be found in soils in many forms and binding states, including oxides and other oxygen complexes, cationic species (often forming complexes with

inorganic soil particles), metal sulfides, and metals bound with organic matter ([Caporale and Violante, 2016](#); [Violante et al., 2010](#)). The metals in Group C ([Table 1](#)), for instance, tend to be cationic and form complexes or precipitates that are often bound to negatively charged clay particles, while several of the metals in Group A tend to form oxides/hydroxides that may be more susceptible to release under anaerobic conditions.

Given that salt marsh soils are largely anaerobic and salty,

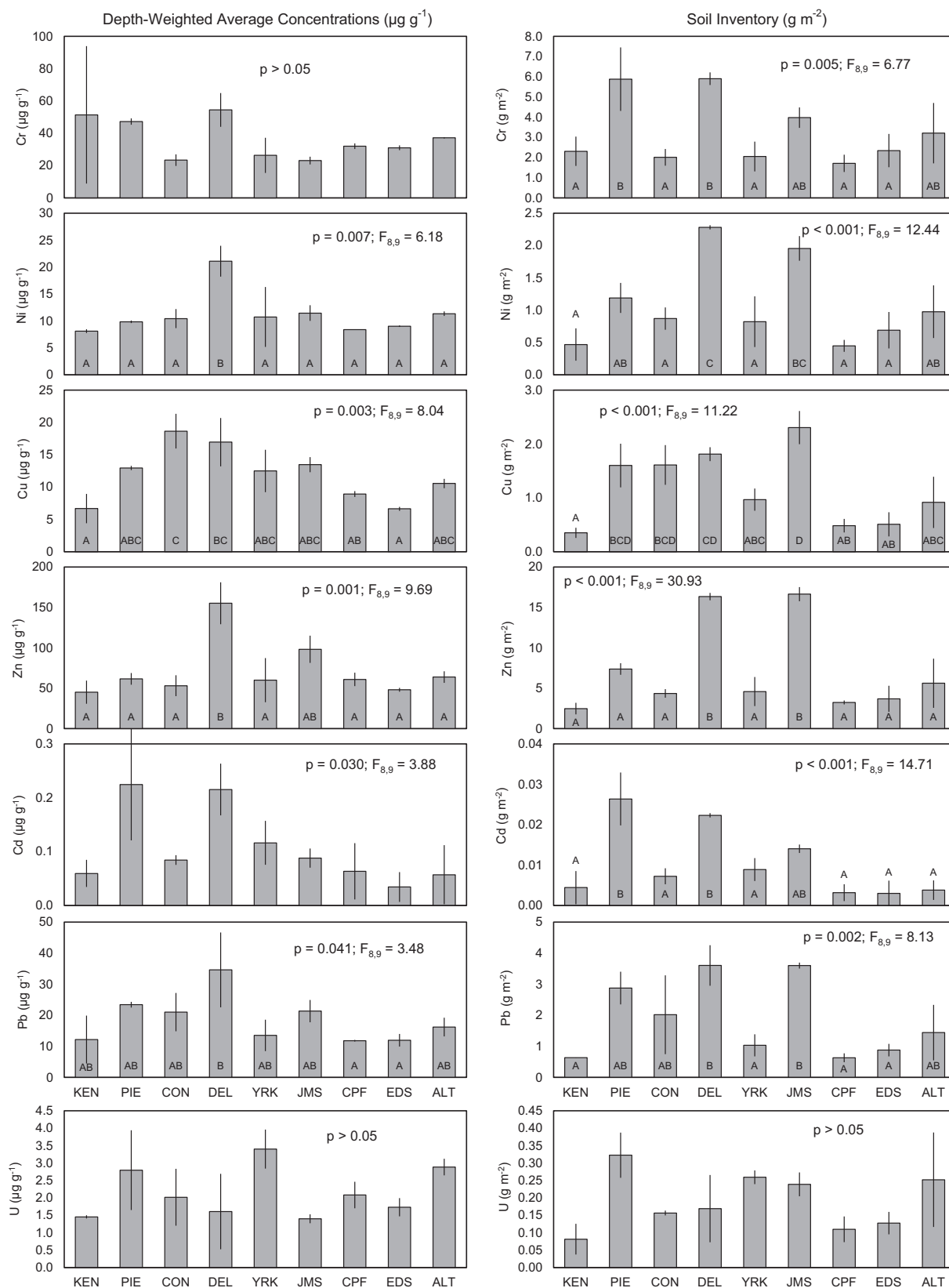


Fig. 3. Post-1925 depth-weighted average metal concentrations in salt marsh soils along the U.S. East Coast (left) and inventories of these metal accumulated in soils (right). Error bars denote standard deviation ($n = 2$). Significant site effect is indicated by analysis of variance (ANOVA) statistics, and sites that do not share a letter were found to have significantly different metal concentrations or inventories (Tukey post-hoc $p < 0.05$).

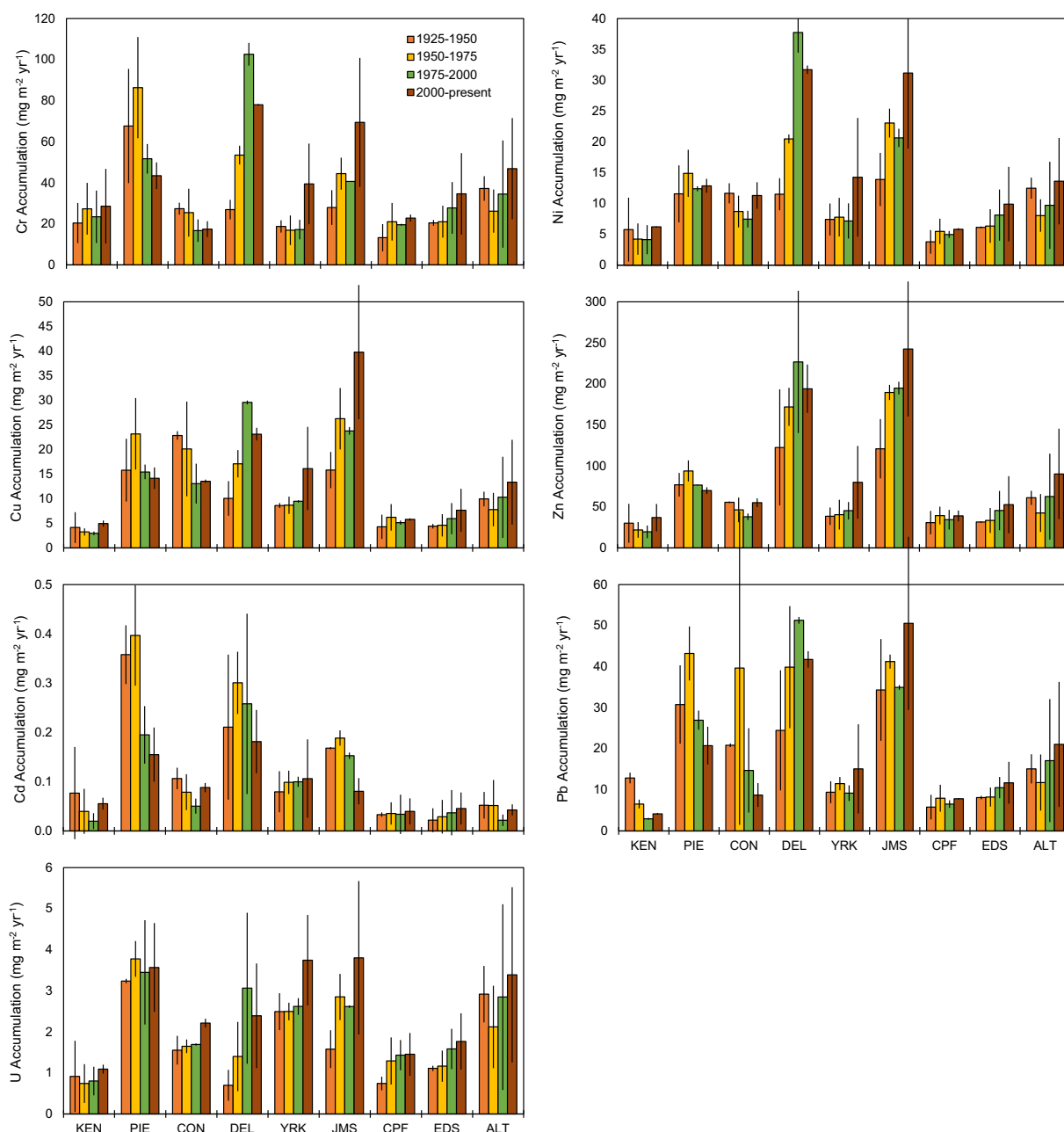


Fig. 4. Average ($\pm$ SD) accumulation rates of chromium, nickel, copper, zinc, cadmium, lead, and uranium in nine salt marsh soils during discrete time periods (1925–1950, 1950–1975, 1975–2000, and 2000-core collection).

mobilization in response to these conditions would likely correspond to greater mobilization in the salt marsh environment. Salinity increases the ionic strength of the soil porewater and the concentration of cations (Na^+ , K^+ , Ca^{2+} , Mg^{2+}) and anions (Cl^- , SO_4^{2-}) found in seawater. The cations in seawater compete with cationic metals for negative sorption sites in the soil, increasing metal availability (Tam and Wong, 1999). We found that Mn, for instance, was mobilized more strongly under salt-water conditions (Table 1) and had high maximum mobilization ($\sim 10\%$; Fig. 1), which would suggest Mn would not be strongly retained in salt marsh soils.

The introduction of oxygen and other oxidants can oxidize metal sulfides and promote mobilization of metals (Otero and Macias, 2002). It is likely that the oxidation of reduced metal sulfide and similar compounds resulted in the significant oxygen effect we observed for the Group C metals (Table S2). In contrast, reducing conditions can promote

the biotic and/or abiotic dissolution of metals that form oxides/hydroxides (Caporale and Violante, 2016). We found, for instance, that As was mobilized more readily under anaerobic conditions (Table 1) and had relatively high maximum mobilization ($\sim 10\%$; Fig. 1). Similarly, Ahmann et al. (1997) found that reducing conditions resulted in mobilization of As from soils, which they attribute to direct microbial reduction of arsenate minerals. Our results indicate that As would not be expected to accumulate in salt marsh soils in a manner relative to its delivery to the coastal environment and would thus not be a strong candidate for assessing the chronology of pollutant delivery to salt marsh systems.

The design of the metal mobilization experiment, in which soil slurries with a high water to sediment ratio were used, encouraged mobilization of metals from the soil matrix. The addition of artificial saltwater with very low metal concentrations increased the

Table 2

Significant linear regression coefficients for the within-site analysis of metal concentrations (i.e., Fig. 2) over time since 1925 ($\mu\text{g g}^{-1} \text{yr}^{-1}$) and accumulation rates for specific periods (1925–1950, 1950–1975, 1975–2000, and 2000-core collection; i.e., Fig. 4) over time since 1925 ($\text{mg m}^{-2} \text{yr}^{-2}$; $p < 0.05$).

	Site	Cr	Ni	Cu	Zn	Cd	Pb	U
Concentrations ($\mu\text{g g}^{-1} \text{yr}^{-1}$)	KEN		0.077				−0.237	0.015
	PIE	−0.414		−0.063	−0.344	−0.004	−0.241	
	CON	−0.158		−0.137			−0.414	
	DEL			−0.065		−0.003	−0.254	
	YRK							−0.023
	JMS	−0.089	−0.057	−0.040	−0.575	−0.002	−0.217	−0.007
	CPF							
	EDS							
	ALT							
Accumulation Rates ($\text{mg m}^{-2} \text{yr}^{-2}$)	KEN						−0.135	
	PIE	−1.072			−0.599	−0.003	−0.561 ^a	
	CON							0.008
	DEL	0.899	0.343	0.230				
	YRK							
	JMS		0.216	0.299	1.632	−0.001		
	CPF							
	EDS							
	ALT							

^a Regression significant from 1950-present, excluding the 1925–1950 period.

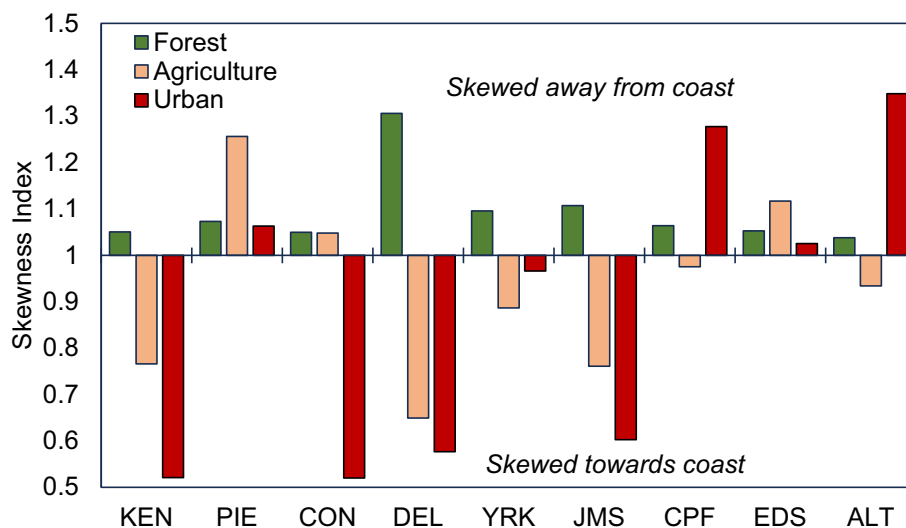


Fig. 5. Skewness index for forest, agricultural, and developed land cover in each watershed. Values above 1.0 indicate the land cover is skewed away from the coast, while values <1.0 indicate land cover is skewed towards the coast.

concentration gradient between the soil/metal matrix and the pore-water, which would promote desorption and mobilization of metals. We therefore suggest the mobilization factors we observed are higher than the release of metals from the soil matrix in salt marsh environments. In our analysis of metals in salt marsh soil cores representing the chronology of metal accumulation over the previous century, we focused on seven elements (Cr, Ni, Cu, Zn, Cd, Pb, and U) that were found to be relatively immobile in salt marsh soils (< 0.1 %; Fig. 1) and have widespread industrial use and/or have been recognized as toxic. Elements with significant mobility in salt marsh soils may not be preserved in soil cores in a manner that reflects the chronology of their delivery to the coast over decades. As an example, As would be of potential interest but, with a mobility >1 % in salt marsh soil conditions (Fig. 1), would not be preserved with fidelity in salt marsh soils.

4.2. Metal accumulation in salt marsh soils

The concentrations of metals we observed in salt marshes along the U.S. East Coast were generally comparable with prior observations in salt marsh systems. For instance, in a review of salt marsh metal

concentrations (Williams et al., 1994 and references therein) found average concentrations of 2.3, 185.1, 67.6, 91.2, 100.7, and 349.3 $\mu\text{g g}^{-1}$ of Cd, Cr, Cu, Ni, Pb, and Zn, respectively, in surficial salt marsh soils across North America, Europe, and Australia. The concentrations we observed at nine sites (Fig. 3) are similar, though at the lower end of the ranges found in Williams et al. (1994). Concentrations of Cd, Zn, Pb, and Cu were higher in a *Spartina alterniflora* marsh in Bohai Bay, China (Chai et al., 2014), but similar to concentrations found in a *S. alterniflora* marsh in the Yangtze River estuary (Quan et al., 2010). Spencer et al. (2003) found declining Cu, Zn, and Pb concentrations in sediments deposited in recent decades in the Medway Estuary, UK, comparable with our findings of declines in metals over time at many U.S. East Coast marsh sites (Fig. 2; Table 2). The depth-weighted concentration of Cr at the KEN, PIE, and DEL sites and the Zn concentration at the DEL site exceeded the threshold effect concentrations (TEC) for these metals (43.4 $\mu\text{g g}^{-1}$ for Cd and 121 $\mu\text{g g}^{-1}$ for Zn) proposed by MacDonald et al. (2000) as consensus-based sediment quality guidelines. While depth-weighted average concentrations of Pb did not exceed the consensus-based TEC for Pb (35.8 $\mu\text{g g}^{-1}$) at any site, concentrations of Pb at specific depths exceeded the TEC at the KEN, PIE, CON, DEL, and JMS

Table 3

Principal component (PC) matrix for watershed land use land cover and physical characteristics including toxic release inventory (TRI) and National Pollutant Discharge Elimination System (NPDES) densities in each watershed and the skewness index (SI) for various land use types. The dominant partial correlations for each PC with eigenvalues >1.0 are noted in bold. The eigenvalues and percent of variance (84.1 % cumulative variance) for each component are given. The lower half of the table gives Pearson correlation coefficients (r) between the salt marsh soil depth-weighted average concentrations ($\mu\text{g g}^{-1}$) and inventories (mg m^{-2} ; both to a common date of 1925) and the principal components with significant correlations indicated by asterisks.

	PC1	PC2	PC3
Developed Land Cover	0.973	−0.058	0.100
NPDES	0.960	−0.144	−0.109
TRI	0.945	0.164	0.029
Brownfield	0.867	0.001	0.431
Dam	0.752	−0.264	0.029
Forested Land Cover	−0.273	−0.916	−0.065
Dam SI	−0.036	0.880	0.025
Temperature	−0.402	0.834	−0.047
Agricultural Land Cover	−0.381	0.823	−0.257
Urban SI	0.079	0.813	−0.287
Slope	0.557	−0.763	0.085
Forest SI	0.200	−0.009	0.897
Agricultural SI	0.553	0.268	−0.753
Suspended Sediment	0.133	0.583	0.750
Accretion Rate	0.368	−0.286	0.705
Soil Erodibility	−0.161	−0.452	0.542
Eigenvalues	6.281	4.338	2.830
% Variance	39.253	27.110	17.690
Depth-Weighted Averages ($\mu\text{g g}^{-1}$)			
Cr	0.221	−0.025	0.146
Ni	0.189	0.027	0.773**
Cu	0.422	−0.387	0.425
Zn	0.175	0.035	0.878**
Cd	0.681**	−0.136	0.427
Pb	0.496*	−0.167	0.602**
U	0.162	0.189	−0.181
Inventories (mg m^{-2})			
Cr	0.644**	0.009	0.483*
Ni	0.288	−0.079	0.792**
Cu	0.429	−0.318	0.526*
Zn	0.208	−0.076	0.815**
Cd	0.754*	−0.181	0.462
Pb	0.497*	−0.206	0.604**
U	0.438	0.054	0.113

** Correlation is significant at the 0.01 level.

* Correlation is significant at the 0.05 level.

sites (Table S4).

Our examination of the concentrations and accumulation rates of metals in salt marsh soils along the U.S. East Coast indicates that there are two main factors that influence both the concentration and accumulation of metals: 1) human activities in the watershed that impact metal abundance and 2) sediment delivery from the watershed and accumulation of that sediment in the coastal zone (Table 3; Fig. S5). The average concentration of Cd and Pb, and the accumulation of Cr, Cd, and Pb were found to be significantly correlated with watershed development and the point sources of pollution accompanying development (Fig. 6; Table 3). We also found that average soil concentrations of Ni, Cu, Zn, Cd, and Pb were significantly correlated across the nine salt marsh study sites (Table S3), indicating that the mechanisms of delivery and retention of these elements in salt marsh soils are similar across broad geographic scales. More concentrated human activities in the landscape directly impacts the amount of metal pollution delivered from that landscape to the coastal zone. Uranium is not typically attributed to development and human activities other than the mining of U for use as a source for nuclear energy (Lu and Liu, 2018). The lack of any significant signal in U concentrations or inventories (Table 3) strengthens the conclusion that human activities are responsible for the signals of the

other metals we observed. It is possible that there was a significant contribution of metals to these salt marsh sites from atmospheric deposition derived from sources beyond the watershed boundaries examined here (Cochran et al., 1998). There may also be a significant lag in the timing of metal pollution in the watershed and the delivery and accumulation of metals in the coastal system (Van Meter and Basu, 2017).

Despite clear links between greater proportions of human development in the watershed resulting in higher metals in coastal salt marshes (Fig. 6) and the continuing increase in development along the U.S. East Coast (Fig. S4), our study also indicates that concentrations of metals in Northeast and Mid-Atlantic salt marsh soils have generally declined in recent decades (Table 2). This demonstrates that policy and management decisions have had a significant impact on metal delivery and accumulation in salt marsh ecosystems. The Clean Water Act of 1972 created regulations for the point source discharge of pollutants into the nation's waterways and established the NPDES permit system. The Clean Air Act of 1970 increased enforcement and requirements for motor vehicle emissions. For instance, the use of Pb as an additive in paint and gasoline was phased out beginning in the 1940s and 1970s, respectively (Filippelli et al., 2015), and atmospheric deposition of Pb has decreased (Eisenreich et al., 1986). We observed convincing declines (> 25 %) in Cr, Cu, Zn, Cd, and Pb concentrations in at least two of the salt marsh sites over time, with the strongest record of reductions in Pb concentration (5 sites; Table 2). The concentration of Pb in marsh soils declined by over 50 % from peak concentrations at the PIE, CON, and JMS sites (Table S4). These are meaningful reductions in soil metal concentrations in several of the Northeast and Mid-Atlantic sites.

In addition to human sources of metal pollution, we observed that factors associated with the delivery of sediment from the landscape to the coast and the subsequent accumulation of those sediments in salt marsh soils are important in determining the concentration and accumulation of metals in coastal systems. We observed that concentrations of Ni, Zn, and Pb along with inventories of Cr, Ni, Cu, Zn, and Pb were higher when rates of salt marsh soil accretion were greater (Fig. 6), which was associated with systems with higher soil erodibility and riverine suspended sediment (Table 3). These findings indicate that sources of metals in the watershed together with the delivery and accumulation of those sediments in depositional coastal environments drive the accumulation of metals in salt marsh soils.

The general pattern of declining soil metal concentrations over time (Fig. 2; Table 2) was not reflected in the record of change in accumulation rates over time (Fig. 4; Table 2). Instead, we found that accumulation rates of several metals accelerated over time, most notably in the DEL and JMS sites (Table 2). Weston et al. (2023) found that rates of salt marsh soil vertical accretion accelerated at all nine of the sites for which we have examined metals in response to accelerating rates of relative sea-level rise, with particularly large accelerations at the DEL and JMS sites (about double the average). It therefore appears that declines in the concentrations of metals in the soil have been largely offset by increases in soil accumulation rates. Metal accumulation rates have not changed significantly or even increased (particularly in the DEL and JMS) in recent decades, despite reductions in soil metal concentrations (Table 2).

5. Conclusion

We have documented links between human activities in the landscape that influence the accumulation of toxic metals in coastal salt marsh soils along the U.S. East Coast. The accumulation of metals in salt marsh soils is also influenced by the factors that determine rates of salt marsh vertical accretion. Successful environmental policy implementation has resulted in declines over time in some metal concentrations in the Northeast and Mid-Atlantic salt marsh soils. However, the legacy of pollution over the past century from human activities in the watersheds draining to the coast has resulted in the accumulation of metals to

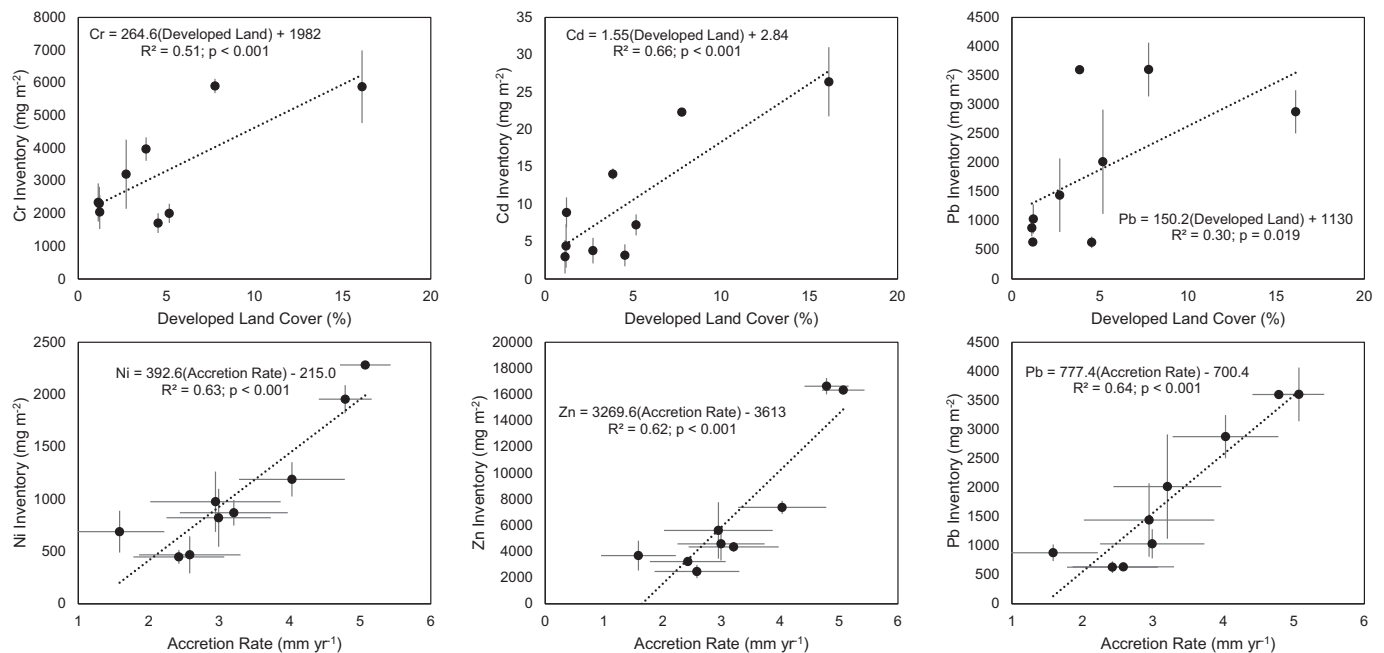


Fig. 6. The relationship between inventories (from 1925 through core collection) of salt marsh soil chromium (Cr), cadmium (Cd), and lead (Pb) and developed land use in the watershed (top row) and soil inventories of nickel (Ni), zinc (Zn), and lead (Pb) with measured rates of vertical accretion (bottom row; accretion from (Weston et al., 2023)). Linear regression statistics are shown, and the error bars denote standard error.

potentially toxic levels in salt marsh soils along much of the U.S. East Coast. Accelerating salt marsh soil accretion rates largely offset reductions in metal concentrations, resulting in little change in metal accumulation rates in recent decades.

CRediT authorship contribution statement

Kristen E. Jezycki: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Elise Rodriguez:** Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Christopher B. Craft:** Writing – review & editing, Investigation, Funding acquisition, Conceptualization. **Scott C. Neubauer:** Writing – review & editing, Investigation, Funding acquisition, Conceptualization. **James T. Morris:** Writing – review & editing, Funding acquisition, Conceptualization. **Steven T. Goldsmith:** Writing – review & editing, Methodology, Funding acquisition, Conceptualization. **Peleg Kremer:** Writing – review & editing, Methodology, Conceptualization. **Nathaniel B. Weston:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data are included in the supplementary material.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2024.171025>.

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