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Contamination status and associated ecological risk assessment of heavy metals in different wetland sediments from an urbanized estuarine ecosystem

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ABSTRACT

Sediment samples of different wetland types (saltmarsh, mangrove, tidal pool, mudflat and sandflat) from an urbanized estuary were analyzed to evaluate the contamination level and ecological risks of five heavy metals (Pb, Fe, Zn, Ni and Cr). The findings showed that the mean concentration (mg/kg) of heavy metals followed the order of Fe > Zn > Ni > Pb > Cr, while Pb and Fe concentrations exceeded the recommended guidelines. Heavy metals levels were highest in saltmarsh and mudflats. Contamination assessment indices e.g., contamination factor (CF), degree of contamination (CD), enrichment factor (EF), and geo-accumulation index (Igeo) revealed that the studied wetlands had low to moderate levels of pollution, meaning these sites receive medium levels of anthropogenic contamination compared with background values. For some of the studied metals, such as Pb, Zn, Fe, and Ni, the EF value was >1 in certain types of wetland, indicating anthropogenic sources, while Cr was <1 indicating natural sources. The pollution load index (PLI) value was determined to be <1, indicating perfection of soil, and was in the following order: mudflat> saltmarsh> tidal pool> mangrove > sandflat. The ecological risk (RI) value was the highest for saltmarsh and the lowest for sandflats. However, the RI value for Cr, Zn, Ni, and Pb was <30 suggesting that these metals pose a low risk in the local ecosystem. Cluster analysis (CA), principal component analysis (PCA), and Pearson's correlation specified that anthropogenic sources of metals.

1. Introduction

Estuarine wetlands such as salt marsh, mangrove, tidal pool, mud flat and sand flat are special types of habitats or ecosystems situated where the marine and terrestrial environments converge. They might be covered or exposed by the daily tidewater intrusion, or they might be accessible during only low tide. It is known that estuarine environment of Bangladesh is incredibly productive in terms of nutrient absorption from a variety of sources (Abu Hena et al., 2013). They provide shelter, feeding and breeding ground for countless resident and migratory species, including numerous critically endangered and commercially valuable species (Abu Hena et al., 2013; Barbier, 2019; Batzer and Sharitz, 2014; Mehvar et al., 2019; Piersma and Van Gils, 2011). In Bangladesh, estuarine wetlands have great importance for the country's socioeconomic, cultural, and ecological aspects (Hena et al., 2007; Abu Hena and Khan, 2009; Mehvar et al., 2019). Increasing anthropogenic activity and development pressure expose wetland habitats to high levels of contamination, and are emerging as the biggest threats to wetland loss (Billah et al., 2014; Islam et al., 2018). Heavy metal contamination, among these difficulties, is a particular concern for wetland ecosystems due to its severe toxicity and considerable species enrichment via bioaccumulation (Mohiuddin et al., 2022; Mehvar et al., 2019).

Heavy metals are present in the environment naturally, but human activities like fast industrialization, urbanization, significant changes in

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land use, and associated increases in terrestrial runoff can make them more abundant (Rahman et al., 2012). Furthermore, natural processes like weathering and erosion of constituent materials may exacerbate heavy metal loading in the aquatic environment. As a result, throughout their transmission, heavy metals released into a river system by natural or man-made causes are distributed between the aquifer and bed sediments (Gibbs, 1973). A portion of metals which transfer to sediment through being adsorbed onto suspended matter and subsequently undergoing a sedimentation process (Zhu et al., 2016). The most important factors determining metal absorption in sediments are sediment matrix type, total organic carbon (TOC) content, grain-size distribution, fractionation and dissolved organic matter (DOM) (Dendievel et al., 2022). Estuarine and coastal sediments are recognized to be significant sinks for heavy metals when environmental circumstances change (Tian et al., 2020).

Around the world, many towns and cities have grown up close to estuaries to take use of the advantages for food, transportation, and water supply. In order to enhance these advantages, people have made changes to estuaries over the centuries. Examples include creating channels for navigation, filling in shallow areas for construction, and managing river outflows. The estuaries that have continued to see growth and industrialization have transformed into urbanized estuaries (Newton and McLellan, 2015). In a similar vein, the Karnaphuli River Estuary is a living example of an urbanized estuary. The Karnaphuli is one of the major estuaries in Bangladesh flows through the commercial capital of Chattagram city, which also serves as the country's principal seaport and nerve centre for the economy. It provides an excellent ecosystem goods and services to the local inhabitants and dwelling organisms possessing some unique ecohabitats like mudflats, tidal pools, sandfalts, saltmarsh and mangroves. Some of the local economically important fish species e.g., *Macrobrachium rosenbergii*, *Lates calcarifer* and *Mughil cephalus* uses these habitats as their breeding ground (Mohiuddin et al., 2022). However, the water resources of Chattagram specifically Karnaphuli River estuary are under severe environmental disturbance as a result of effluent discharge from cement and ship recycling industries, oil refineries and tanneries, paper and rayon factories, iron and steel industries, naval and merchant ships, fertilizer and other chemical industries, as well as direct disposal of sewage and solid waste from the city into the neighboring Karnaphuli River and coastal regions of the Bay of Bengal (Ali et al., 2016; Hossain and Khan, 2002; Mohiuddin et al., 2022). The incidence of toxic and hazardous materials in river, estuary and marine ecosystems not only threaten ecological functions (Rahman et al., 2014) but also presents potential risks to public health and livelihood in Bangladesh (Islam et al., 2014). Estuarine wetlands along the Karnaphuli River are influenced by the chemical and mineral composition of suspended materials, anthropogenic influences, deposition, enrichment, and pollution from the river have an influence on the environment (Ahmed et al., 2019; Jain et al., 2007).

Over the past three decades, research on heavy metal contamination in coastal ecosystems has developed quickly throughout the world. In Bangladesh, previous studies had also reported the distribution, and contamination of heavy metals in different coastal rivers and estuaries such as the Sangu River (Hossain et al., 2019), Meghna river (Bhuyan et al., 2017), Feni river estuary (Islam et al., 2018), Karnaphuli river (Ali et al., 2016) and Halda river (Bhuyan and Bakar, 2017). However, none of the earlier research focused on the level of metal contamination and associated ecological risks in the special habitats (such as saltmarsh, mangrove, tidal pool, mudflats, and sandflats), despite these types of wetlands were inherently a part of the tropical rivers and estuaries. To implement the proper protective measures for these habitats, it is dire important to assess the concentration of metal contaminants, possible risks and their sources in this scenario. Therefore, in this study we aimed to i) assess the extent of metal pollution in different wetland sediments (saltmarsh, mangrove, tidal pool, mudflats, and sandflats) of the Karnaphuli river estuary, Bangladesh, ii) evaluate the ecological risk due to sediment contamination, and iii) identify the possible sources of heavy metals using chemometrics and multivariate analyses. The findings will be helpful for coastal managers and policy makers who are adopting laws to safeguard the priceless variety of wetlands in this estuary from long-term buildup of heavy metals as well as for maintaining food safety obtained from there, ensuring human welfare.

2. Materials and methods

2.1. Study area

The study was conducted along the Karnaphuli River estuary (22°12′33″ to 91°38′54″), which runs through Chattagram City and is near to the Bay of Bengal in Bangladesh (Fig. 1 and Table S1). The study locations were chosen after a preliminary survey was conducted to identify and pinpoint low, medium, and highly impacted sites that are frequently known to be contaminated by domestic, agricultural, and industrial wastes, including oil refineries, iron and steel industries, sewage, farming, landfills, fertilizer factories and paper mills. Geographically, the bed of the river is made up of tertiary rocks covered in alluvial deposits, with the underlying accumulations being constituted of mud and sand in progressively finer stages (Rizbi, 1971). Additionally, the Karnaphuli River exhibits semidiurnal tides with a range of 2–4 m. Its mean depth in the external zone is 8 to 10 m (m) (Lara et al., 2009), and the ecosystem's environmental parameters change periodically as a result of the strong influence of the Indian monsoon (Alam and Zafar, 2012).

2.2. Sample collection

For the assessment of the levels of heavy metals, a total of 30 samples of sediments were taken using a mud corer from 10 stations (designated as S1, S2, S3, S4, S5, S6, S7, S8, S9, and S10) in various types of wetlands, such as mud flats, sand flats, salt marshes, tidal pools, and mangrove areas of the Karnaphuli River estuary (Fig. 1). Samples were appropriately labelled and placed in polythene bags after collection. Following that, the samples were kept in an ice box at 4 °C for further examination.

2.3. Sample preparation and metal extraction

In the laboratory, a portion of the representative sediment samples was kept in a hot air oven on a petri dish at a temperature of 110 °C for 8 h. After cooling, the samples were pulverised in a mortar and pestle. The crushed materials were sieved through a 2 mm mesh strainer before being homogenised. 15 mL of aqua regia, which is composed of HNO3 and HCl in a 1:3 M ratio, was added to 1 g of dried and powdered sediment sample in a pre-acid-washed 100-mL beaker. The beaker was then covered with a watch glass and heated on a hot plate set to 100 °C until brown vapours were visible. Heating the solution caused it to evaporate without boiling. After the digesting procedure was finished, the most sediment sample was dissolved in acid to lower the overall material volume. The mixture was then allowed to cool before being transferred to a volumetric flask with deionized water and filtered using a 125-mm membrane filter paper (Whatman® Schleicher & Schuell, Darmstadt, Germany). All of the materials, along with sample blanks (made of deionized water) and standards, were breathed into an atomic absorption spectrophotometer (AAS) furnace (Model-AA-7000 Atomic Absorption Spectrophotometer, Shimadzu, built in Japan). The statistical analysis of the data employed both the least squares method and straight line fitting. When calculating the concentration of different components, the required adjustments were applied. Replicate samples, blank samples, and certified reference materials (CRM) were employed to ensure the accuracy of the experiment. The recovery findings showed good accuracy and precision because they were within 10 % of the authorized values.

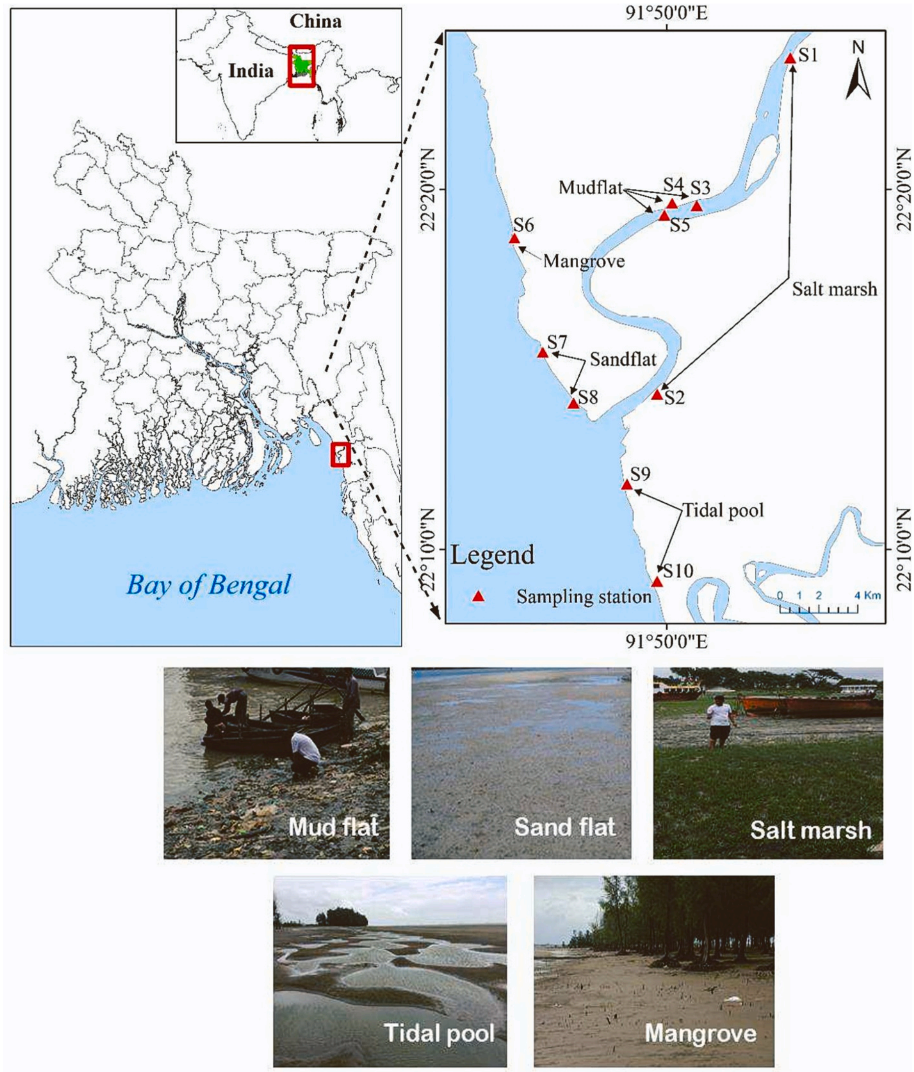

Fig. 1. Location of the sampling stations with wetland types around the Karnaphuli River at Chittagong, Bangladesh.

2.4. Quality control (QC) and quality assurance (QA)

The current study adopted a thorough QA/QC strategy. Beginning with standard sediment reference materials (GBW07314 and GBW07333) recommended by the Second Institute of Oceanography (SOA), China, the correctness of procedure and precision were evaluated. In addition, procedure blanks and duplicates were employed in this investigation for quality control. The relative standard deviations (RSDs) for repeated samples were below 10 %, while the analytical results for the reference materials were within 10 % volatility. Each analysis began with the calibration of the equipment. The blanks in every set had been tested in duplicates according to the identical methodology. The percentages of recovery were found between 93 % - 106 %. The limit of detection was 0.001 for Pb and Fe; 0.002 for Cu and Cd and 0.1 μg/g for Ni and Cr. The reported results are the mean of repeated analysis. Samples for repeated tests were chosen at random. All glasses, plastics, and quartz were washed in 10 % HNO_3, and then rinsed with deionized water (18.25 M ohm cm^{-1}).

2.5. Risk assessment in sediments of different estuarine ecosystem

The level of metal pollution status in diverse wetland sediments from urbanized estuarine habitats was assessed in this study using a number of metal assessment indices (Hakanson, 1980; Tomlinson et al., 1980; Varol, 2011) e.g., Contamination Factor (CF), Enrichment factor (EF), Degree of Contamination (CD), Pollution Load Index (PLI), Potential Ecological Risk Assessment (PERI) (Table 1).

2.6. Statistical analysis

The most popular multivariate statistical techniques, correlation and principal component analysis, were applied to explore meaningful correlations between the heavy metals in the sediment samples. The statistical procedures were carried out with a 95 % confidence interval (significance $p < 0.05$). To identify contamination sources, a factor approach that relies on principal component analysis (PCA) was used (natural and anthropogenic). The spatial diversity among the locations was identified using cluster analysis. The Euclidean distance was used as the dissimilarity matrix, whereas the coupling method was the Ward's method. PAST, a free statistical program was used for all of the statistical studies (Hammer et al., 2001).

3. Results and discussion

3.1. Heavy metal concentrations in sediments of different wetlands

In this study, the mean concentrations of the studied metals in the experimented areas followed the decreasing order of Fe (17,402.06 ±

Table 1
Pollution indices used in the present study.

Eq. No	Index	Depiction and objectives	Principle	Explanation	Pollution degree criteria
1.	Contamination factor (CF)	CF estimates the heavy metal contamination.	$CF = C_{metal}/C_{background}$	Here, C_{metal} = the concentration of each metal and $C_{background}$ = background concentration of that metal.	CF > 6: Very high pollution 3 < CF < 6: Considerable contamination 1 < CF < 3: Moderate contamination CF < 1: Low contamination (Hakanson, 1980)
2.	Degree of contamination (CD)	CD evaluates metal contamination status.	$CD = \sum CF$	Here, CF = the contamination factor.	CD < 5: Low 5 < CD < 10: Moderate 10 < CD < 20: Considerable 20 > CD: Very high (Hakanson, 1980)
3.	Pollution load index (PLI)	PLI is used for comparison of contamination status among sites.	$PLI = n\sqrt{(CF_1 \times CF_2 \times CF_3 \times .... CFn)}$	Here, CF = the contamination factor and n = the total number of studied metals.	PLI < 1: No pollution; PLI > 1: Polluted (Tomlinson et al., 1980).
4.	Geo-accumulation index (I_{geo})	I_{geo} evaluates the contamination comparing with background values (Muller, 1969)	$I_{geo} = log_2\left(Cn/_{1.5Bn}\right)$	Here, C_n = the concentration of the metals in samples. B_n = the background value of the metal (n), Constant 1.5 = the variations in the background values (Ke et al., 2015).	>4: Extremely polluted, 3–4: Heavily polluted; 2–3: Moderately to heavily polluted; 1–2: Moderately polluted; 0–1: Unpolluted to moderately polluted; <0: Practically unpolluted; (Chowdhury et al., 2015; Varol, 2011, Islam et al., 2018).
5.	Enrichment factor (EF)	EF measures influence of anthropogenic (Sayadi et al., 2010).	$EF = \frac{\left(C_n/_{C_{Mn}}\right) sample}{\left(C_n/_{C_{Mn}}\right) background}$	Here, C_n = the concentration of the metals in samples, C_{Mn} = the concentration of Mn in that area.	2–5: Deficiency to moderate enrichment 5–10: Moderately severe enrichment 10–25: Severe enrichment 25–50: Very severe enrichment > 50: Extremely high enrichment (Birch and Olmos, 2008)
6.	Potential ecological risk index (PERI)	PERI was proposed by Hakanson (1980) and is applied to appraise the potential ecological risk of studied metals in sediment.	$E_r^i = T_r^i C_f^i$ $C_f^i = C_n^i / C_o^i$ $ER_I = \sum E_r^i$.	Here, E_r^i = the potential ecological risk factor and T_r^i = the toxic response factor of metals. Here, Cu = Pb = Ni = Co = 5, Mn = Zn = 1, As = 10, and Cr = 2 were used (Suresh et al., 2011).	Er < 40: Low ecological risk Er ≤80: Moderate ecological risk; 80 < Er ≤ 160: Appreciable ecological risk; 160 < Er ≤320: High ecological risk; Er > 320: Serious ecological risk And RI <150: Low pollution; 150 < RI < 300: Considerable pollution; 300 < RI < 600: High pollution; RI ≥600: Very high pollution (Hakanson, 1980; Ke et al., 2015).

8220.77 mg/kg) > Zn (40.47 ± 23.49 mg/kg) > Ni (18.34 ± 16.26 mg/kg) > Pb (12.54 ± 6.53 mg/kg) > Cr (0.85 ± 0.68 mg/kg). One-way ANOVA results showed that the mean concentration of essential metals (Fe, and Zn) and non-essential metals (Ni, Pb, and Cr) differed highly significantly ($F = 21.27$, $p < 0.01$) at 99 % confidence interval. However, higher load of Fe in the sediments from the urbanized estuarine ecosystem could be attributed to terrigenous sources such as sedimentary and industrial material transported by seasonal flashfloods (Boyko et al., 2019). In addition, there are ship recycling companies, iron and steel industries, as well as mining and mineral processing, all of which have a significant impact on the higher Fe concentrations. Besides, previous studies suggested that Zn, Ni, Pb, and Cr might be originated through anthropogenic activities (Islam et al., 2018; Rahman et al., 2019), and the load of these metals in the surficial sediment could have been escalated through runoff from agricultural land, residential area, and domestic sewage (Islam et al., 2018).

The concentration of Fe and Zn in different wetlands followed the decreasing order of saltmarsh > mudflat > tidal pool > sandflat >

mangrove, whereas Pb, Ni, and Cr load were recorded decreasing in order of saltmarsh > mudflat > tidal pool > mangrove > sandflat (Fig. 2). The metal concentrations in saltmarsh, mudflat, and tidal pool wetlands were the highest indicating that these wetlands received metals from residential sewage, farm lands and industrial discharge through Karnaphuli River. Again, saltmarsh and mudflat had higher load of metals as these wetlands were formed mostly with clay. In contrast, the metal loads in sandflat and mangrove wetlands were relatively lower than the other studied wetlands because of sediment properties (grain size) and also these sites receive comparatively small number of pollutants from the industrial and residential areas. An expansion of undistributed soil that is primarily found in the lower water column is known as a sandflat. Sandflat has the lowest content of clay (Li et al., 2007). It is a fragile area because tidal surges and ebb flux periodically resuspend sediment. Neap tides cause the finer muddy sediments to build up, while spring tides cause the rougher granular sediments to build up which generally act as a source of lower metal accumulation (McLachlan and Brown, 2006).

This study was a pioneer to scrutinize the load of heavy metals in sediments at different types of wetlands along with the Karnaphuli River. However, the present findings were assessed by comparing with other similar works (Table 2) as there was no existing data on metal loads at different types of wetlands in this river. It was observed that the load of Pb, Zn, and Ni in the saltmarsh of this study was higher than previous studies (Rakib et al., 2021; Idaszkin et al., 2020). In contrast, Pb, Zn, Ni, and Cr load in mudflat of this experiment was lower than that of Ulhas Estuary, India (Fernandes and Nayak, 2012). This is due to mudflats being frequently surrounded by low-elevating sandflats and at high levels of neap tide, a marsh vegetation region (Dyer, 1998). That vegetation has been efficient in absorbing metal pollutants from soils such as lead, cadmium, chromium, arsenic, and different radionuclides (Tangahu et al., 2011). Further, all the metals assessed in this study was found to be lower in the mangrove sediment than other mangroves worldwide (Rahman et al., 2021; Chowdhury et al., 2017; Wu et al., 2019; Shi et al., 2019). Similarly, sandflat and tidal pool of this study were found to be less polluted with Zn and Cr.

3.2. Contamination level and ecological risk assessment

3.2.1. Contamination factor (CF), degree of contamination (CD) and pollution load index (PLI)

The degree of heavy metal pollution is estimated using CF (Hakanson, 1980). The estimated CF value of <1 denotes no contamination, 1–3 moderate, and > 3 substantial or high contamination. The highest values of CF for Pb (0.92), Fe (0.59), Zn (0.72), Ni (0.53), and Cr (0.019) were estimated in the saltmarsh. In contrast, Pb (0.17), Ni (0.08), and Cr (0.0038) were found the lowest in sandflat whereas Fe (0.19) and Zn (0.17) were the lowest in mangrove. However, the values of CF for all metals were below 1 (Fig. 3) indicating that the studied wetlands had low metal contamination. Further, CD was calculated to estimate the degree of contamination of each wetland. According to the value of CD, studied wetlands followed the decreasing order of saltmarsh (2.78) > mudflat (2.22) > tidal pool (1.52) > mangrove (0.98) > sandflat (0.74) (Fig. 4). The CD values found in this study indicated that the studied wetlands had low degree of contamination. However, the PLI provides an understanding of the overall toxicity level of the sample. The PLI will give the occupants a certain understanding of the nature of the environment. This also offers valuable information for decision-makers on the state of the area's pollution rate (Suresh et al., 2011). In this study, the PLI values followed the order of: saltmarsh (0.33) > mudflat (0.25) > tidal pool (0.16) > mangrove (0.09) > sandflat (0.08). All the values of PLI were found to be below 1, indicating that the wetlands were not polluted with the metals (Fig. 4).

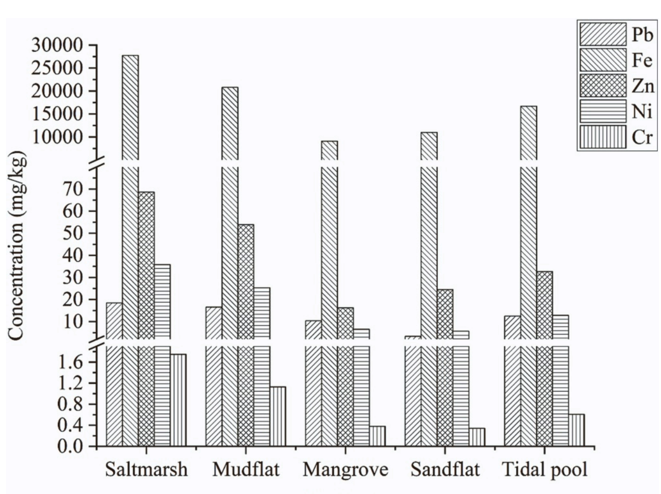

Fig. 2. Concentrations of heavy metals in sediments at different wetlands in the Karnaphuli River Estuary (mg/kg).

3.2.2. Enrichment factor (EF)

EF measures influence of anthropogenic activities (Sayadi et al., 2010). EF value of <2 indicates no enrichment; 2–5 moderate enrichment and > 5 considerable enrichment. In this method, heavy metal enrichment in sediments is evaluated through the values found in comparison with metal load in sediments with geochemical background values (Tuna et al., 2007). The mean EF values of Pb, Fe, Zn, Ni, and Cr followed the order of Pb (1.73) > Zn (1.1) > Fe (1) > Ni (0.63) > Cr (0.02) (Fig. 3). The values of EF revealed that all the heavy metals were reported to be <2 at all wetlands, except Pb in mangrove, suggesting the minimal enrichment in the area. The EF value for Pb in mangrove (2.70) showed moderate enrichment. Heavy metal influx through surface runoff and industrial activities may cause moderate increase in Pb, which can be severe in the near future.

3.2.3. Geo-accumulation index (I_{geo})

I_{geo} evaluates the contamination levels of metals comparing with its background values (Muller, 1969). To determine sediment quality, the values of I_{geo} have been used (Karbassi et al., 2008). The values of I_{geo} for five heavy metals in this experiment ranged from −8.63 (for Cr in sandflat) to −0.7 (for Pb in saltmarsh). The average values of I_{geo} for each metal decreased as per the following order: Pb (−1.51) > Zn (−2.05) > Fe (−2.17) > Ni (−2.93) > Cr (−7.62) (Fig. 3). However, all the quantified heavy metals in every wetland had values of <1.0 indicating that the sampling sites were in uncontaminated to a moderately contaminated degree.

3.2.4. Potential ecological risk factor (Er) and risk index (RI)

The ecological risk index indicates not only the single ecological risk of individual metal but also indicates the integrated ecological and toxicological effects of the increased pollution by diving the ecological risk levels of soil contamination (Karydas et al., 2015). The average monomial risk factor E_r^i of metals in sediments samples from different wetlands of around the Karnaphuli River were ranked in the order: Pb (3.06) > Ni (1.27) > Zn (0.41) > Cr (0.02) (Fig. 3). All the values of ecological risk for Pb, Ni, Zn, and Cr were found below 40 which indicates that this metals are posed to low risk in the surrounding ecosystem.

The calculated potential ecological risk index (RI) for metals in this study is shown in Fig. 4. Samples from different wetlands followed the order of: saltmarsh (8.0) > mudflat (6.59) > tidal pool (4.41) > mangrove (3.25) > sandflat (1.51) (Fig. 4). The RI values revealed that the studied wetlands posed the lowest ecological risk (RI < 150).

Table 2
Comparison of the findings (mg/kg) of present study with the experiments conducted worldwide and some standard values.

Sampling location	Pb	Fe	Zn	Ni	Cr	References
Saltmarsh						
Chittagong, Bangladesh	18.46	27,719.83	68.61	35.76	1.75	This study
Chittagong, Bangladesh	5.48	31,658	41.72	NA	NA	Rakib et al., 2021
Pozo Salt Marsh, Patagonia, Argentina	10.28	NA	19.89	23.29	62.64	Idaszkin et al., 2020
			Mudflat			
Chittagong, Bangladesh	16.57	20,779.32	53.84	25.30	1.13	This study
Ulhas Estuary, India	82	9.02	180	190	239	Fernandes and Nayak, 2012
			Mangrove			
Chittagong, Bangladesh	10.38	9086.99	16.21	6.51	0.38	This study
Sundarban, Bangladesh	25.6	NA	102.9	NA	2.7	Rahman et al., 2021
Sundarban, India	25.44	NA	62.85	34.91	23.40	Chowdhury et al., 2017
Futian Mangrove, Shenzhen, China	NA	NA	351.15	117.92	49.76	Wu et al., 2019
Xixiang Mangrove, Shenzhen, China	NA	NA	429.41	168.12	103.09	Wu et al., 2019
Shajing Mangrove, Shenzhen, China	NA	NA	649.19	397.87	311.16	Wu et al., 2019
Dongzhaigang Mangrove, Hainan, China	19.32	NA	48.57	18.97	65.77	Shi et al., 2019
			Sandflat			
Chittagong, Bangladesh	3.33	10,992.03	24.485	5.64	0.34	This study
Niger Delta, Nigeria	0.024	NA	74.51	2.278	7.392	Benson et al., 2016
			Tidal pool			
Chittagong, Bangladesh	12.46	16,685.50	32.67	12.82	0.61	This study
Jangsong tidal flat, Korea	17.9	NA	68.3	15.5	15.2	Kim et al., 2021
			Standard vales			
Shale value	20	47,200	95	68	90	Turekian and Wedepohl, 1961
Lowest effect level (LEL) for sediment	31	2 %	120	16	26	MacDonald et al., 2000
Threshold Effect Level (TEL) for sediment	30.2	–	124	22.7	37.3	MacDonald et al., 2000
Probable Effect Level (PEL) for sediment	112	–	271	48.6	90	MacDonald et al., 2000
Severe Effect Level (SEL) for sediments	250	4 %	820	75	110	MacDonald et al., 2000

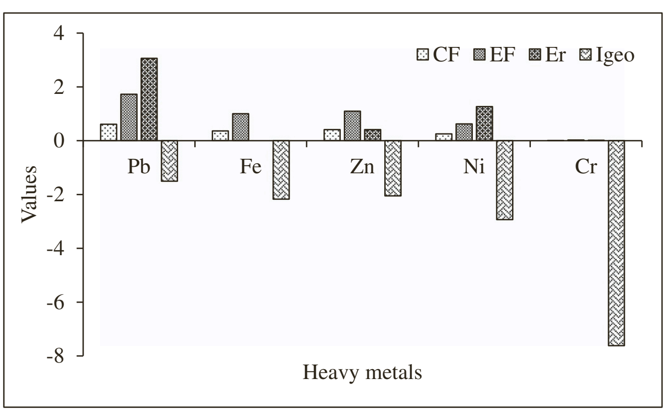

Fig. 3. CF, EF, Er and Igeo of the studied metals in different wetlands.

3.3. Potential source identification

Enrichment factor (EF) <1 indicates natural sources and enrichment factor (EF) > 1 indicates anthropogenic source of heavy metals (Conrad et al., 2017). The sediments were normalized with respect to the reference elements, including Fe (Amin et al., 2009; Karbassi et al., 2008). In the current study, EF value for some of the metals such as Pb, Zn, Fe and Ni is >1 in some stations indicates the anthropogenic sources. The EF value of Cr is <1 indicates that Cr is coming from only natural sources.

Correlation between heavy metals can indicate the origin and movement of these metals. When there is no correlation between the elements, therefore not one single factor regulates the metals. The correlation matrix (Table 3) revealed that the Fe, Zn, and Ni were highly significantly ($p < 0.01$) and positively correlated with each other indicating their similar types of origin. Further, Pb and Ni were found to be significantly correlated ($p < 0.05$) in this study highlighting their identical sources.

The chemometric technique PCA was utilized to comprehend the intricate relationship between sediment samples and heavy metals. This multivariate technique is founded on the correlation matrix and eigenvalue analysis (Dendievel et al., 2022). Each variable has a loading that indicates how well the model components are taking that variable into account. In our investigation, the PCA analysis produced two major PC components that together accounted for 99 % of the data variability (Fig. 5). Ni, Cr, Fe, and Zn all had very strong positive loads in the first principal component (PC1), which made up 94.56 % of the total variation. Our investigation corroborated with a correlation coefficient between them of $r = 0.97$. Aside from the anthropogenic inputs from nearby fertilizer industry, they can be originated from natural limestone (Mohiuddin et al., 2022; Dendievel et al., 2022). Once again, it is known that Cr and Ni are mutually related with a variety of rocks (Jain et al., 2007). High levels of Cr can be present due to direct release of effluents from paint and ink industries. A highly positive load of Pb was seen in the PC2, which explained 4.59 % of the observed variation, but only a medium load of Zn. According to the current results, these two elements in natural soils exhibit the same close geochemical dependence as the iron family, with a correlation coefficient of $r = 0.82$. Agricultural runoff, disposed car batteries and commercial phosphate fertilizers and its raw materials, is shown to be a significant supplier of such Zn and Pb (Tian et al., 2020; Mohiuddin et al., 2022).

CA was performed to group the similar sampling stations (spatial variability) and recognize specific areas of contamination (Simeonov et al., 2000). Heavy metal origins in the studied areas are primarily affected by natural factors as well as human (Liu et al., 2022). Spatial CA rendered a dendrogram (Fig. 6) where all five wetlands around the Karnaphuli River were grouped into two statistically significant groups

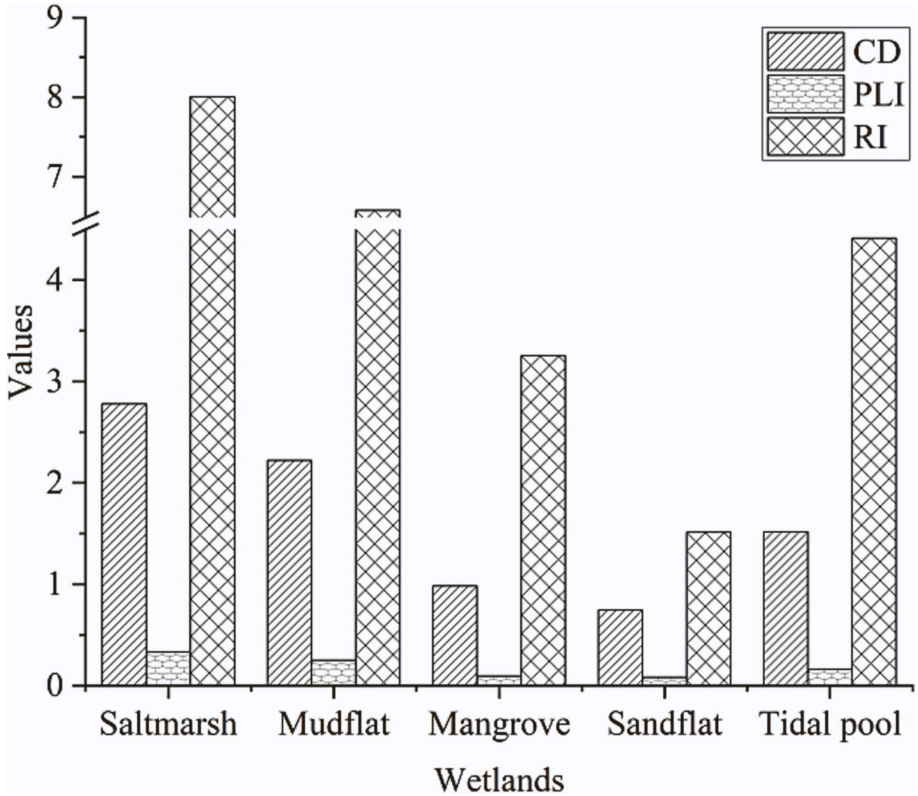

Fig. 4. CD, PLI and RI in different wetlands around the Karnaphuli River estuary, Chittagong.

Table 3
Correlation of metals in different wetlands sediments of Karnaphuli estuary.

	Pb	Fe	Zn	Ni	Cr
Pb					
Fe	0.84544				
Zn	0.81818	0.98554**			
Ni	0.8838*	0.97977**	0.98621**		
Cr	0.86036	0.97346**	0.97749**	0.9956**	

* $p < 0.05$.
** $p < 0.01$.

at (Dlink/Dmax) × 100 < 10,000. Group 1 is consisted of two wetlands (mangrove and sandflat) which is mostly dominated by naturally occurring metals. Group 2 is consisted of three wetlands (saltmarsh, mudflat, and tidal pool) which are mostly dominated by both natural and anthropogenic activities due to the presence of textiles mills, fertilizer company, power plant and oil refinery. Anthropogenic activities e.g., wastewater and sewage discharge from domestic and industries, urbanization, dumping of solid waste, automobile washing and auto workshops near the study sites put a great impact on the heavy metal enrichment.

4. Conclusion

This is the pioneer study to assess the contamination levels and potential ecological risks of heavy metals in the different types of wetland sediments (saltmarsh, mangrove, tidal pool, mudflat and sandflat) within the Karnaphuli river estuarine system. The mean concentration (mg/kg) of heavy metals in all wetland types followed the decreasing sequence of Fe > Zn > Ni > Pb > Cr. Pb and Fe concentrations was found to be exceeded the safe recommended limit. Pb levels was found in order of saltmarsh > mangrove > tidal pool > mudflat > sandflat, and Fe in order of mudflat > saltmarsh > tidal pool > sandflat > mangrove.

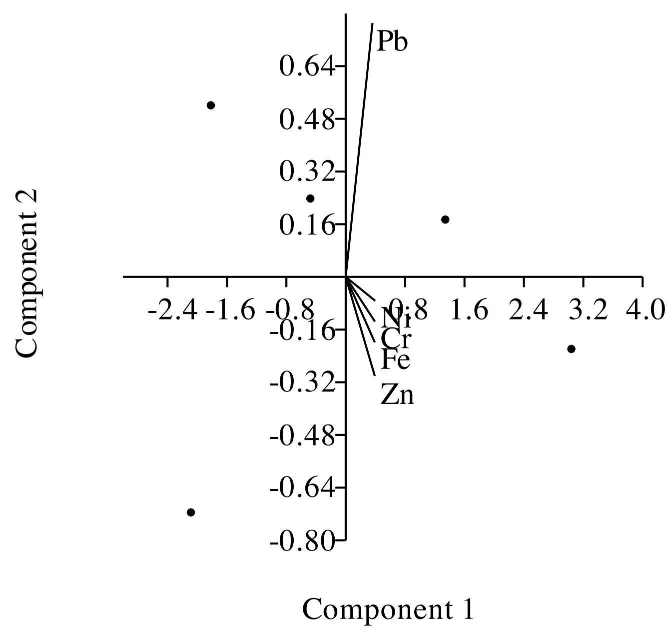

Fig. 5. PCA analyses of heavy metals in different wetlands of Karnaphuli Estuary.

Saltmarsh and mudflat possessed a considerably larger metal load as they primarily comprised of clay. In contrast, sandflats have the lowest concentration of clay due to the buildup of rougher granular sands during spring tides, which prevent higher metal accumulation in them. The CF, CD, EF, and I_{geo} values showed that studied wetlands had low to moderate levels of pollution. Enrichment Factor (EF) value of Pb, Zn, Fe and Ni was >1 for some wetland types indicating the anthropogenic

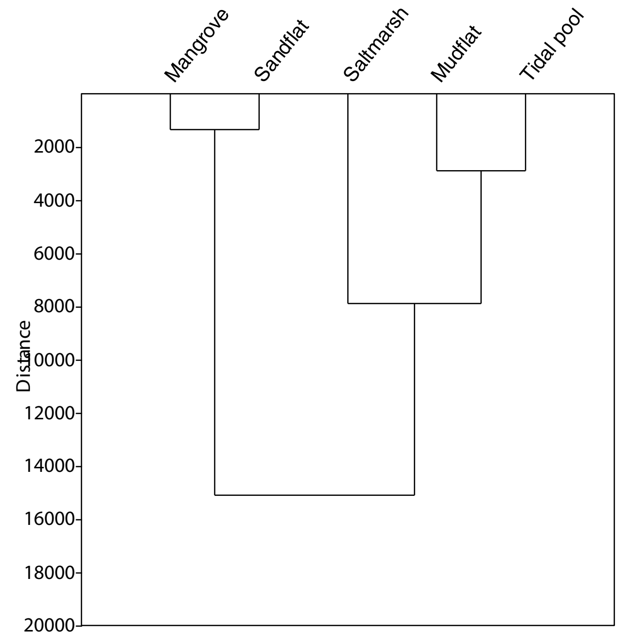

Fig. 6. Cluster analysis of heavy metals in different wetland types in the Karnaphuli estuary.

sources, and for Cr was <1 indicates originated from natural sources. Ecological risk assessment value (PERI) for Cr, Zn, Ni, and Pb was found below 30 suggesting that these metals posed a low risk in the local ecosystem. Multivariate analyses identified anthropogenic sources of metals in the study area. The data provided in the present experiment could be useful for environmental agencies or governments in the monitoring and management of the vast coastal ecosystem of the Karnaphuli river estuary. In order to protect the wetland environment, wastewater and sewage management should be strictly enforced in industries and cities. To reduce heavy metal concentrations from polluted soils, ex situ remediation techniques should be applied. Traditional remediation methods include scouring the soil, land filling, electrokinetic ablation, and soil sequestration.

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CRediT authorship contribution statement

Conceptualization, M.B.H.; methodology, M.A.R and M.B.H; formal analysis, M.A.R and M.B.H; investigation, M.K.H, M.A.R. S.S. and M.B.H; resources, M.B.H. and M.K.H; data curation, M.K.H, S.S. and M.B.H; writing—original draft preparation, M.A.R., A.A.U.N, S.S; writing—review and editing, M.B.H., M.F.A; J.Y. and T. A.; visualization, M.F.A.; supervision, M.B.H. and M.K.H.; project administration, M.B.H.; funding acquisition, M.B.H., M.F.A. and T.A.

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Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Mohammad Belal Hossain reports financial support was provided by King Saud University. Mohmmad Belal Hossain reports a relationship with King Saud University that includes: funding grants.

Data availability

Data will be made available on request.

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