Marine Pollution Bulletin 203 (2024) 116429

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Heavy metals in afforested mangrove sediment from the world's largest delta: Distributional mapping, contamination status, risk assessment and source tracing

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ARTICLE INFO

Keywords:
Heavy metals
Distribution
Contamination
Mangrove sediments
Ecological risk
mHQ

ABSTRACT

This study aims to assess seasonal and spatial variations, contamination status, ecological risks, and metal sources (Ni, Pb, Cr, Cu, Mn, and Zn) in human-afforested mangrove sediments in a deltaic region. Five sampling locations were sampled during dry and wet seasons. Heavy metal concentrations followed the order: Mn $>$ Zn $>$ Ni $>$ Cr $>$ Cu $>$ Pb. Metal loads, except Cu and Pb, were higher during the dry season, aligning with national and international recommendations. Sediment quality guidelines, contamination factor, geoaccumulation index, enrichment factors, and pollution load index indicated uncontaminated sediment in both seasons. Potential ecological risk assessment showed low risk conditions in all sites. However, modified hazard quotient indicated moderate pollution risk from all metals except Pb. Analysis suggests anthropogenic sources, particularly evident near shipbreaking yards in Sitakunda. While initially uncontaminated, ongoing metal influx poses a potential risk to mangrove ecosystems.

1. Introduction

The mangrove ecosystem is renowned as among the most productive coastal wetlands globally (Rahman et al., 2019; Hossain et al., 2021a). These are widely dispersed in over 100 countries in the tropical and subtropical regions including Bangladesh, and offer wood and non-wood forest products for the local communities (Giri et al., 2011). These ecosystems are commonly recognized for their crucial role in coastal environments, protecting coastal communities from natural disasters, enhancing water quality, cycling carbon, mitigating soil erosion and promoting diverse biodiversity (Alongi, 2014; Uddin et al., 2022; Al Mahmud et al., 2024). Bangladesh boasts the world's largest natural mangrove forest area (Sundarbans), spanning approximately 6017 km^2 in the southern region (Al Mahmud et al., 2024). These forests have served as a biodiversity hotspot, a vital source of livelihood and have protected millions of people, acting as natural shields against climate-induced effects like cyclones, for decades (Hossain et al., 2021b; Al Mahmud et al., 2024). Recognizing these invaluable benefits, the Bangladesh government initiated mangrove afforestation in other open coastal areas during the late 1950s to safeguard communities from natural disasters and optimize the advantages (Hasan et al., 2013a).

Since 1966, approximately 280 km^2 of mangroves have been planted on the delta of the Ganges, Brahmaputra, and Meghna Rivers (Islam and Rahman, 2015). This area represents 7.5 % of the Sundarbans mangrove forest and 0.21 % of all mangroves worldwide (Uddin et al., 2022). Notable among these afforested mangrove zones are the lower Meghna and Feni River estuarine areas, including Nijhum Dwip, Sitakunda, and Swandwip. Situated along the northern coast of the Bay of Bengal, these afforested mangroves are currently susceptible to both anthropogenic and natural vulnerabilities such as storms, floods, rapid sediment

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https://doi.org/10.1016/j.marpolbul.2024.116429
Received 2 March 2024; Received in revised form 23 April 2024; Accepted 25 April 2024

erosion, industrial and agricultural pollution (Rahman et al., 2019; Ahmed et al., 2024). While large-scale natural mangrove ecosystems like the Sundarbans can somewhat mitigate the adverse effects of these natural and anthropogenic hazards, small-scale manmade mangroves are significantly vulnerable. Therefore, it is crucial to assess the impact of various anthropogenic activities, such as pollution, on afforested mangrove environments.

Mangroves experience direct pollution impacts largely due to their proximity to urban areas, which exposes them to contaminants from sewage runoff, farming practices, and industrial discharge. (Hossain et al., 2021a). In addition, other human activities such as clearing mangroves for urban expansion, aquaculture, or environmental restoration, can lead to the resurfacing of buried pollutants. This is because disturbing the sediment can release these pollutants back into the environment (Soper et al., 2019; Szafranski and Granek, 2023). In mangrove ecosystems, heavy metals (HMs), persistent organic pollutants (POPs), microplastics (MPs), polycyclic aromatic hydrocarbons (PAHs), and personal care products (PPCPs) are emerging contaminants (Hasan et al., 2013b). Of these, heavy metals have been identified as a significant concern and a priority pollutant for mangrove forests due to their bioavailability, persistence and potential toxicity to both aquatic organisms and surrounding ecosystems (Szafranski and Granek, 2023). Being the world's largest delta, the coastal area of Bangladesh receives substantial freshwater runoff from its river system leading to the introduction of large amounts of heavy metals (HMs). Additionally, the distinctive physical, chemical, and ecological characteristics of mangrove forests enable them to sequester heavy metals once they accumulate in sediment (Hossain et al., 2021b; Siddique et al., 2021). Consequently, toxic metals contaminate tidal water, infiltrate mangrove wetlands, mingle with pore waters, disperse throughout the water column, and ultimately accumulate in biota (Thanh-Nho et al., 2018).

Mangrove sediments are recognized as reservoirs for heavy metal pollutants, demonstrating rapid accumulation within their finely textured composition (Rahman et al., 2021). Furthermore, benthic organisms in mangrove sediments, including polychaetes, brachyuran crabs, gastropods, bivalves, and sipunculids, possess the capability to absorb metals from their surrounding environments (Enuneku et al., 2018; Kumar et al., 2019). These accumulated metals are then transferred to fish and shellfish, which play a role in bioaccumulating heavy metals (Hong et al., 2020). The subsequent bioaccumulation and biomagnification of toxic heavy metals through these aquatic food chains, sourced from water, food, and suspended sediment particles, pose persistent hazards to human health and other organisms (Rahman et al., 2021). Therefore, estimating the accumulation of these metals in mangrove sediments is a fundamental step towards further assessing their bioaccumulation in mangrove flora and fauna.

The distribution of metals in coastal sediment across Bangladesh is greatly influenced by seasonality and hydro-meteorological variables (Hossain et al., 2021a). During the rainy season, the increased precipitation and input of freshwater intensify the movement of sediment, leading to lower concentrations of metals due to dilution (Ahmed et al., 2020, 2021a, 2022, 2024). In contrast, in the dry season, decreased precipitation enables the settling of silt and the buildup of metals (Rahman et al., 2019). Changes in salt concentration, caused by the flow of rivers and the movement of tides, affect the chemical form and movement of metals in sediment (Ranjan et al., 2018). Temperature fluctuations impact biological activities, such as microbial activity, leading to changes in metal cycling and bioavailability. Modifications in the intake of pollutants and sediment load in rivers might additionally alter the composition and distribution of metals. The complex interaction between various factors influences the formation of metal patterns in terms of space and time, which in turn affects the environmental condition and the health of ecosystems in the coastal regions of Bangladesh.

We hypothesized that the accumulation of heavy metals in the afforested mangrove sediments is regulated by spatial and temporal variation. Moreover, the level of the HMs in the afforested mangrove sediments is high enough to show the potential ecological risk. Although several studies (Aktaruzzaman et al., 2014; Ali et al., 2016; Hossain et al., 2019; Islam et al., 2018; Rahman et al., 2019; Siddique et al., 2021) have investigated the prevalence and distribution of heavy metals in estuarine and coastal sediments, research specifically focusing on metal contamination in afforested mangrove sediment remains lacking. Hence, the study aimed to conduct a comprehensive assessment of heavy metal contamination in the sediment of artificially afforested mangrove forests. The objectives were to address the following inquiries: (i) What seasonal fluctuations occur in metal concentration levels within sediment of human-afforested mangrove forests? (ii) How does spatial variability affect metal concentrations in sediment? (iii) What is the contamination status and associated ecological risks linked with the presence of metals? and (iv) What are the sources of metals present in sediment in the study area? This study represents the first attempt to examine the presence and impacts of heavy metals in human-afforested mangrove sediment, contrasting with the natural mangrove forests of Sundarbans. This research will be crucial for establishing a baseline to gauge the impacts of forthcoming human activities and to furnish valuable insights for future management strategies and policy formulation in the region. Besides regional importance, understanding the extent and impacts of metal pollution in mangrove sediment can guide cross-border management approaches, promote worldwide collaboration, and uphold the indispensable ecological and socio-economic value of these ecosystems globally.

2. Martials and methods

2.1. Study area

We conducted our study in the afforested Mangrove sediments at Nijhum Dwip, Sitakunda, and Swandwip in the coastal delta of the lower Meghna river Estuary, Bangladesh. Nijhum Dwip, is an island and a tourist spot for the spotted deer in the forest, declared a national park by the Bangladesh government. Sitakunda is an Upazila of the Chittagong district is well-known for the Eco-Park, saltwater hot spring waterfall and other natural beauties, and the ship-scraping activities on the coast. Swandwip is an island which is also an upazila of the Chittagong district and situated at the coastal delta of the Feni River and Meghna River estuary. The Meghna River is the main outlet of the total (260) river system and 13 million tons of sediment is being transported in every year (Rahman, 2013). The sediment class is clay because of continues runoffs of river water. The study area has a monsoon environment and sunny, tropical weather. The seasonal monsoon winds have significantly impacted the region, like other regions of the coastal area. The average annual temperature is 26.24 °C, and the average annual rainfall is 3207 mm (Hossain et al., 2012). The entire area is tidal-influenced all year. Tides are semi-diurnal, with two high and two low waters during a lunar day. The tide ranges from 0.07 m during neap tide to 4.42 m during spring tide. Because of having tidal influence sediment's salinity approximately remains at 2–3 ppt in all seasons. The study area represents the broken coastal area by sediment erosion with heavy human activities such as construction, wrong disposal of sewerage and locomotives. The drainage system was linked nearby the sampling sites throw canal that carries total waste disposals of main town. Moreover, Sitakunda is one of the major coastal regions in Bangladesh next to the Bay of Bengal that has been subjected to pollution from ship breaking industry.

2.2. Selection of sampling sites, sample collection and preparation

The study area was divided into five sampling stations, encompassing nearly all crucial locations of artificially afforested mangrove forest sediments. The selection of sampling stations was based on the absence of river or canal bank-breaking and sediment settlement (Fig. 1). From

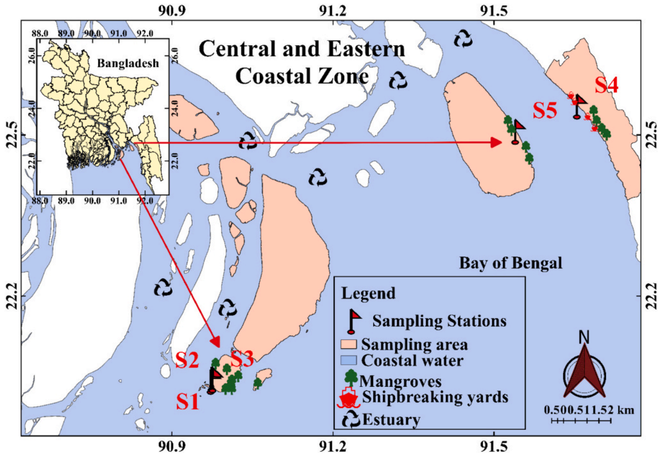


Fig. 1. Map of the area showing the sampling sites Nijhum Dwip (S1, S2, S3), Sitakunda (S4) and Swandwip (S5) of afforested Mangroves sediment of Bangladesh.

the five sampling stations (S1 to S3 for Nijhum Dwip, S4 for Sitakunda, and S5 for Swandwip), a total of four replicate samples with distinct ecological characteristics were chosen for seasonal sampling. Thus, we gathered a total of 40 surficial sediment samples during the dry season (December 2018 to January 2019) and wet season (June to July 2019). The distance between each station within the same site was approximately 2 km. Surface sediment samples from each site were taken at a 0–10 cm depth from the surface using a mud corer, which covers an area of (1 × 1) m. The top 2 cm of each sample were taken using an acid-washed plastic spatula from the mud corer's center to prevent contamination from the mud corer's metallic components (Ahmed et al., 2022). Plastic bags with the appropriate labelling were put to use for storing the samples after they had been previously sterilized with 10 % nitric acid and repeated for 20 min in an ultrasonic bath using distillate and ultrapure water (resistivity MΩ/cm) (Hossain et al., 2021b; Islam et al., 2018). After that, the samples were dried for 24 h at 50 °C (Hyun et al., 2007). Then, using a mortar and pestle, the dried samples were ground up, and the resulting lower particle-sized fractions were homogenized by grinding them in an agate mortar before being stored in glass bottles with labels (Ahmed et al., 2022; Islam et al., 2015). The labelled samples were then taken to the lab and underwent additional chemical analysis.

2.3. *Heavy metals analysis and quality assurance*

Samples were transported to the Soil and Environment Research Section, Bangladesh Council of Scientific and Industrial Research (BCSIR), and Bangladesh Atomic Energy Commission (BAEC) for chemical analysis. Samples were dried at 120 °C in a laboratory oven after being weighed in a tarred silica dish by a preferred amount of 10–20 g (Hossain et al., 2021a). To prevent the organic effluents, all measured samples were then put in a muffle furnace, and the temperature was gradually raised from 450 °C at a rate of 50 °C/h. Samples were digested correctly with 50 % nitric acid after 8 h of ignition, and they were then filtered using Whatman No. 44 filter paper in a 50-ml chemical flask. Following a standard analytical process, the residue was cleaned, and the metal contents were found using Atomic Absorption Spectroscopy (Model: AA-700, SHIMADZU) (Ahmed et al., 2022). The selection of heavy metals was based on their widespread occurrence in environmental matrices and their substantial influence on human health and ecosystems. Moreover, their regulatory significance and connection to industrial activity rendered them prime subjects for inquiry. Their established analytical methodologies and past study findings provided additional support for their selection, ensuring rigorous data collection and comparability with existing literature. We considered sample recovery, reagent blank analysis, and standard calibration to assess quality assurance and quality control (QA/QC). To ensure the validation, accuracy, and precision of the data analysis process, certified reference material from Merck KGaA (Germany) was utilized. Accuracy, indicated by recovery rates, ranged from 97 % to 110 % for all metals, while precision, measured by the Relative Standard Deviation (RSD), was below 5 % for all metals (Table S1). These findings closely mirrored the reference values, validating the results. The precision of the diagnostic strategy was maintained by reference sediment material (CRM320) ($N = 3$). The limit of detection (LOD) for trace elements was determined as follows: Ni and Zn were 0.001, Pb and Cr were 0.002, Mn was 0.1, and Cu was 0.003. Glassware was pre-washed for better precision by soaking in 10 % HNO_3 (w/w) for at least 48 h, and the reagents had the assurance to be an analytical grade or above.

2.4. *Sediment contamination level assessment*

For assessing metal contamination and risk calculation, scientists have developed models or indices that rely on background values to estimate contamination levels (Tomlinson et al., 1980; Muller, 1969). These models or indices have been utilized in pollution monitoring assessment and risk calculation studies for the past three decades (Birch, 2023). In this study Contamination factor (CF), Pollution load index (PLI), Geo-accumulation index (I_{geo}), Enrichment factor (EF), Potential ecological risk assessment (PERI) and Modified hazard quotient (mHQ) (Supplementary files A). However, there have been numerous critiques regarding the establishment of background concentrations that are used for risk calculations (Birch, 2023). This is because defining background data is crucial for accurately quantifying pollution levels in sediment and soil. The regional differences in sampling locations between contaminated and uncontaminated areas can lead to significant variations in sediment elemental compositions. Ranjan et al. (2008) utilized elemental abundances of the Earth's crust as baseline data, while Rubio et al. (2001) advocated for the use of local background values. Faganeli et al. (1991) suggested that the most representative background data could be extracted from deeper sediment layers, whereas Karaouzas et al. (2021) advocated for the establishment of regional values to assess sediment quality. In the current study, elemental abundances corresponding to the average crustal abundance data, as suggested by Turekian and Wedepohl, 1961, were utilized as background values, similar to many previous studies (Hossain et al., 2019; Islam et al., 2018; Singh et al., 2005).

2.5. *Sediment quality estimation by SQGs*

The sediment state was assessed for potential risks to the aquatic ecosystem using the consensus-based sediment quality guidelines (SQGs), a crucial technique for determining the degree of trace metal concentration in the sediment (MacDonald et al., 2000; Varol and Şen, 2012). The threshold effect concentration (TEC) and probable effect concentration (PEC) are two different kinds of identical limit values that make up the SQGs (Hossain et al., 2021b; Islam et al., 2018). The TEC is indicated by the effects data's lower 10th percentile, while the PEC is indicated by the effects of 50th percentile (MacDonald et al., 2000; Turekian and Wedepohl, 1961; Yeh et al., 2020). Values greater than PEC suggest severe risks to an aquatic habitat, whilst values below the TEC are not expected to have adverse biological effects (Ahmed et al., 2022; Hossain et al., 2021a).

2.6. *Statistical analyses*

Descriptive statistics were conducted using Microsoft Excel version 10. We conducted a parametric independent sample t-test ($n = 5 < 30$) for our data set to compare each metal concentration in Wet and Dry seasons. Before that, we tested the normality of our data set with the Shapiro-Wilk test and found they were normally distributed ($p > 0.05$). Multivariate and univariate analyses, including cluster analysis (CA), correlation matrix (CM), and two-way hierarchical cluster (heat map), were performed using Origin 9.5 (OriginLab Corporation, USA). Spatial distribution of sampling locations was illustrated using QGIS (v. 3.32 "LIMA"). Additionally, raster maps depicting spatial distribution were generated using inverse distance weighted (IDW) interpolation. The IDW technique, chosen for its popularity in interpolating scattered points, particularly favors local points over distant ones, aligning with the purpose of the study. For omitting biases, we selected three replication sites for each sampling point.

3. Results and discussion

3.1. Seasonal and spatial variation of metal levels in mangrove sediments

Considering both dry and wet seasons, the average metal concentrations in mangrove sediments were found in the decreasing order of Mn (464.7 ± 53.47 µg/g) > Zn (62.32 ± 22.45 µg/g) > Ni (38.77 ± 8.03 µg/g) > Cr (36.6 ± 6.67 µg/g) > Cu (35.74 ± 6.73 µg/g) > Pb (7.38 ± 7.14 µg/g) (Table 1). All the metals, excluding Cu and Pb, showed higher concentration loads during dry season and lower in wet season. The lower levels in the wet season compared to the dry season may be attributed to increased dilution effects caused by heavy rainfall, which disperses and transports metals downstream. Additionally, the higher water flow rates during wet season may facilitate the flushing out of metals from the coastal sediment, resulting in reduced concentrations. Because the grain size of the sediment and the size of the metals are the major controller in the spatial variability of the studied trace elements (Larrose et al., 2010). In dry season, highest concentration of Ni (47.88 µg/g), Cr (43.7 µg/g), Cu (46.9 µg/g), and Pb (2.79 µg/g) was observed in S4, whereas lowest concentration of Mn (433.08 µg/g), Ni (34.38 µg/g), Cr (30.84 µg/g), and Cu (22.91 µg/g) was in S2 (Fig. S 1). On the other hand, during the wet season, S4 had the highest concentration of Ni (47.14 µg/g) and Cu (42.77 µg/g), whereas S2 had a minimum load of Mn (364.95 µg/g), Ni (25.47 µg/g), Cr (23.58 µg/g), and Cu (33.31 µg/g). So, the findings indicated that S4 had a comparatively higher metal load and S2 had the minimum load in any season. Moreover, Independent sample t-test showed that the load of Pb (F = 53.92, p = 8.045E-05) and Zn (F = 387.4, p = 4.62E-08) highly significantly differed between dry and wet season. Nevertheless, during the two seasons under investigation, there was no significant change in the concentration of other metals.

As depicted in Fig. 2, heavy metals such as Cr, Cu, Pb, and Ni exhibited significantly higher levels in mangrove sediment near the shipbreaking-dominated site of Sitakunda compared to Swandwip and Nijhum Dwip. This suggests that the notable influx of metals into coastal sediments may originate from shipbreaking activities (Rahman et al., 2019). However, sources of Ni include electroplating, batteries, alloy manufacturing, spark plugs and other ignition devices (Kabata-Pendias and Mukherjee, 2007) though they contribute very little to be found in sediments. Various applications of Ni and Cu alloys, such as ship crankshafts, nozzles, steam valves, propellers, pistons, reduction gearboxes, and steam turbine blades, further contribute to their presence in

Table 1
Seasonal and spatial variation of heavy metal's concentrations (µg/g) in the mangrove sediment of coastal area in Bangladesh with comparison of other studies and standard values.

Descriptive statistics		Heavy metals (µg/g, dw)						Reference
		Ni	Pb	Cr	Cu	Mn	Zn	
Dry season	Mean	42.42	1.06	38.56	35.36	483.37	83.41	This study
	SD	5.80	1.25	5.43	9.31	41.65	3.39	
	CV (%)	12.22	105.60	12.59	23.55	7.71	3.63	
Wet season	Mean	35.12	13.71	34.63	36.12	446.04	41.24	
	SD	8.83	3.64	7.80	3.85	61.86	3.39	
	CV (%)	22.49	23.78	20.15	9.53	12.40	7.35	
Mangrove Sediment	Mean	38.77	7.38	36.60	35.74	464.70	62.32	
	SD	7.61	6.78	6.32	6.38	50.72	21.30	
	t-test (p-value)	0.161	0.0	0.382	0.869	0.295	0.0	
Sediment quality guidelines (SQGs)								
TEC		23	36	43	32	460	120	(Turekian and Wedepohl, 1961)
PEC		49	130	110	110	1100	460	(MacDonald et al., 2000)
% of samples < TEC		0	100	100	20	20	100	
% of samples between TEC-PEC		100	0	0	80	80	0	
% of samples > PEC		0	0	0	0	0	0	
TEL (Threshold effect level)		18	35	37.3	35.7	NG	123	
PEL (Probable effect level)		36	91.3	90	110	NG	315	
SEL (Severe effect level)		75	250	110	197	NG	820	
Literature (Metals concentration (µg/g dw) in mangrove sediment)								
Bangladesh	Nijhum Dwip	35.93	7.55	33.76	32.09	456.37	63.30	This study
	Sitakunda	47.51	8.43	39.92	44.84	474.09	61.69	
	Swandwip	38.58	5.84	41.79	37.61	480.32	60.03	
	Sundarbans	–	25.6	2.7	14.8	–	102.9	(Rahman et al., 2021)
China	Quanzhou	–	73.7	–	42.5	–	184	(Liu and Sun, 2013)
	Zhanjiang Bay	7.86	20.07	–	18.24	–	–	(Zhou et al., 2022)
	Shenzhen	–	105	74.3	400	–	352	(Chai et al., 2019)
India	Sundarbans	34.91	25.44	23.40	47.67	901	62.85	(Chowdhury et al., 2017)
	Gulf of Khambh	34.66	7.14	–	11.64	–	–	(Singh et al., 2020)
Saudi Arabia	Farasan Island	8.48	34.5	9.61	112	–	57.2	(Usman et al., 2013)
	Arabian Gulf	81.05	4.4	–	209.8	–	–	(Al-Kahtany et al., 2018)
Vietnam	Thi Vai Estuary	–	21	27	99	–	92	(Costa-Böddeker et al., 2017)
Average Shale value		68	20	90	45	850	95	(Turekian and Wedepohl, 1961)
Standard value		10-25[a]	19[b]	23[a]	33[c]	400[d]	95[e]	

NG = no guideline available.

[a] Canadian Council of Ministers of the Environment (CCME, 2002).

[b] Metals in the Hydro-Cycle. Berlin, Springer (Salomons and Forstner, 1984).

[c] Guidebook on applications of radiotracers in industry. Technical Report Series No. 316 (Donhoffer, 1991).

[d] NOAA screening quick reference tables (Buchman, 1999).

[e] The Review of the Health of the Oceans. Reports and Studies No. 15. GESAMP, Geneva, 108 (GESAMP, 1982).

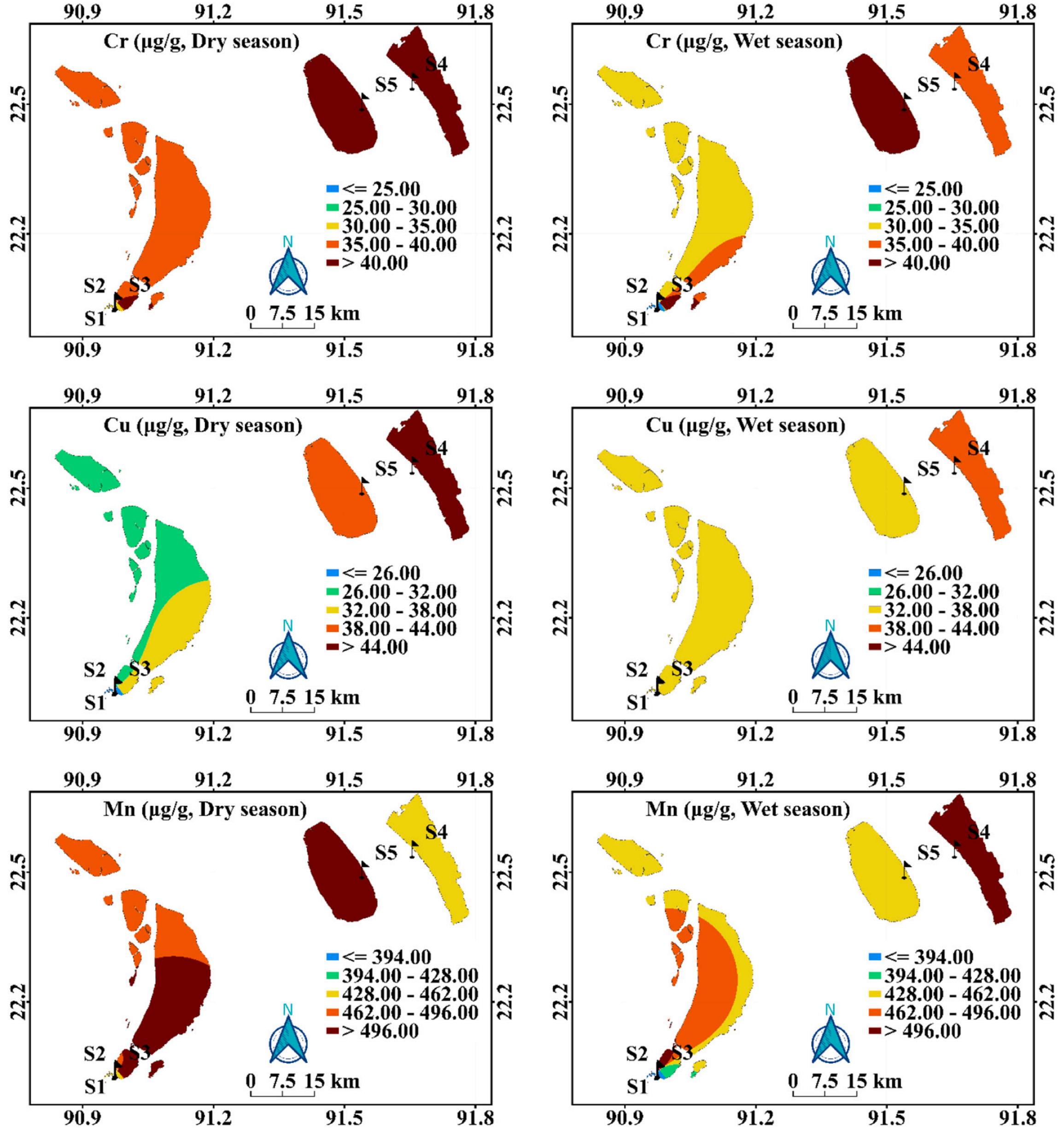


Fig. 2. Spatial variations of metals concentrations in the sediments during Dry and Wet season in the three mangrove sediment sites in Bangladesh.

the environment (Hasan et al., 2013a; Rahman et al., 2019). Besides, Cr and Pb are applied on the ship's deck fittings and rigging components to prevent corrosion (Kura and Mookoni, 1998). Moreover, batteries, paints and components of motors, generators, piping, and cables are also good sources of Pb in the aquatic environment (Rahman et al., 2017). Hence, when ships are recycled, these ecotoxic metals find their way into the aquatic ecosystem. So, both seasons' high metal concentrations of Cr, Cu, Pb, and Ni in S4 (Sitakunda, Chittagong) might be attributed mainly to the ship-breaking industrial activities and urban runoff (Aktaruzzaman et al., 2014; Hasan et al., 2013b; Rahman et al., 2019).

A comparative study of the heavy metal concentration of this experiment and the results reported worldwide, along with the average shale value and standard values, are also presented in Table 1. Metals load in the studied different areas of the world along with our study were lower than that in urban mangroves of Shenzhen, China (Al-Kahtany et al., 2018; Chai et al., 2019; Chowdhury et al., 2017; Costa-Böddeker et al., 2017; Liu and Sun, 2013; Singh et al., 2020; Zhou et al., 2022). However, this study recorded a higher concentration of Ni, Cr, and Zn compared to the sediment of Farasan Island, Saudi Arabia (Usman et al., 2013). Rahman et al. documented a minimum load of Cr (2.7 μg/g) and Cu (14.8 μg/g) in the sediment of the Sundarbans mangrove forest, Bangladesh, then the present study (Rahman et al., 2021). However, the high Zn in the mangrove sediments of the studied areas might be attributed to the industrial and domestic sewage discharge and application of fertilizers and pesticides in the agricultural fields (Banerjee et al., 2016). Further, a high load of Cr indicated the substantial release and accumulation of industrial effluent from dye and chrome plating (Benhaddya and Hadjel, 2014; Mathivanan and Rajaram, 2014; Sarkar et al., 2011). Moreover, the Swandwip and Nijhum Dwip islands are far from the urban runoff compared to Sitakunda, which had less metal concentration in the mangrove sediment. In addition, the mean values of the three sites, except Zn, were higher than the recommended values of different environmental quality standards (Buchman, 1999; CCME, 2002; Donhoffer, 1991; GESAMP, 1982; Salomons and Forstner, 1984). So, the condition of the sediment is vulnerable to the associated living organisms, and the aquatic ecology adjacent to the sediments is at

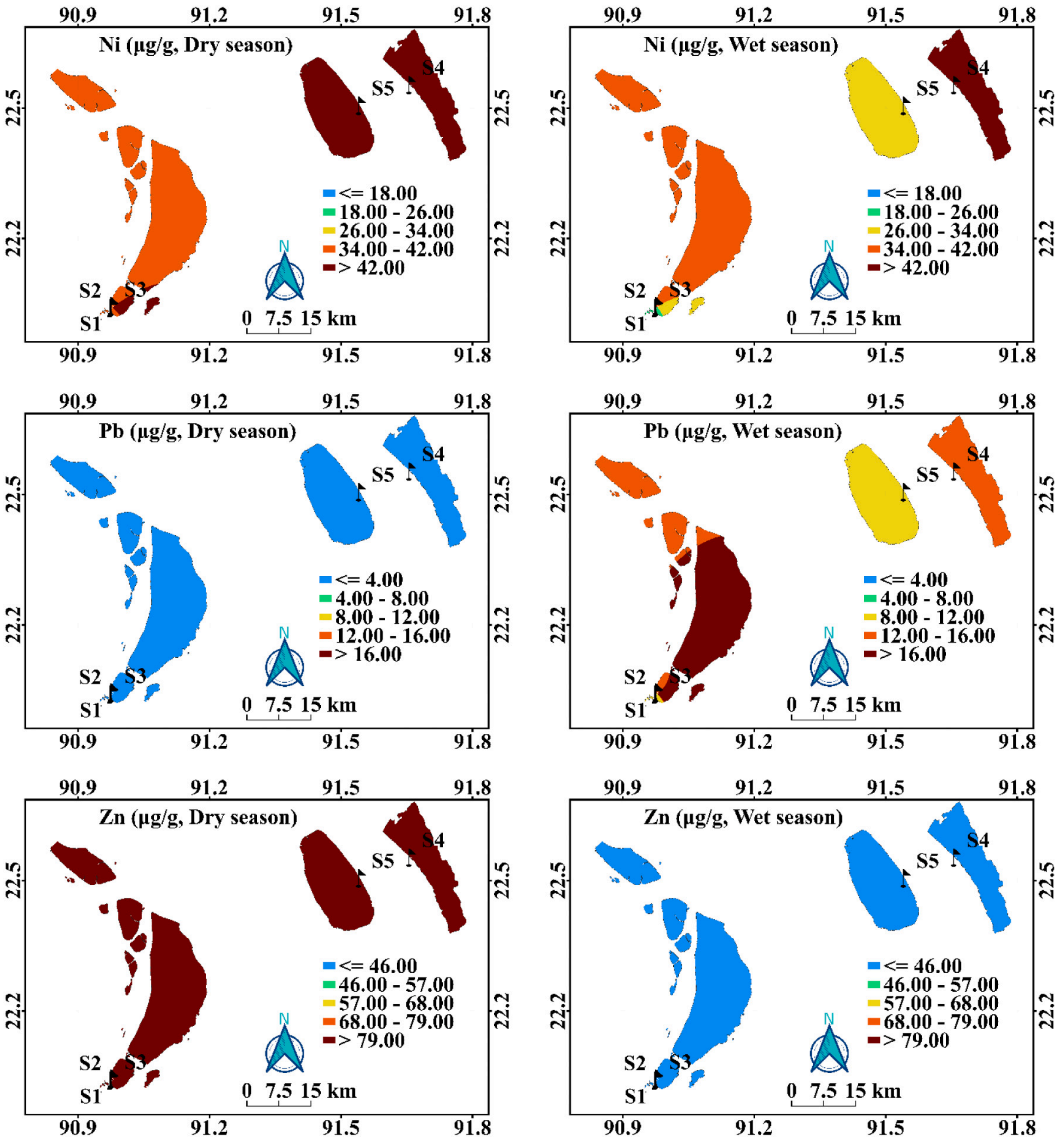


Fig. 2. (*continued*).

potential risk.

3.2. Ecological risk and metal contamination level assessment

3.2.1. Sediment quality guidelines (SQGs)

We assessed Sediment Quality Guidelines (SQGs) to discern the ecotoxicological impacts of contaminants (Islam et al., 2018). Sediment sample heavy metal concentrations were compared with consensus-based threshold effect concentration (TEC) and probable effect concentration (PEC) values. As per Table 1, Ni, Cu, and Mn samples largely fell within the TEC-PEC range, while Pb, Cr, and Zn concentrations remained below TEC values, suggesting minimal direct impact from unintentional human activities in the study area (Ahmed et al., 2022). Furthermore, SQG evaluation indicated that the biological risk posed by contaminants in mangrove sediment to resident species may not be substantial. However, comprehensive assessment of other geochemical indices is crucial for a more thorough understanding of current trace metal pollution levels (Siddique et al., 2021).

3.2.2. Pollution assessment indices

Fig. 3. displays the values for CF, PLI, Igeo, and EF. In general, CF values for all metals (Cu > Zn > Ni > Mn > Cr > Pb) in the study area were lower compared to the relatively higher values observed during the dry season, except for Pb and Cu. However, the CF value for Cu, particularly at S4 in Sitakunda, exhibited higher values (> 1), possibly influenced by external discrete sources such as anthropogenic inputs, industrial and agricultural runoff. Contaminated water from ship-breaking yards and runoff chemicals from nearby metal screaming shops may be the primary reasons for the elevated CF values for Cu in the study area (Rahman et al., 2019).

Mean EF values of studied metals showed no enrichment in both seasons (EF < 2), indicating insignificant metal concentrations for metal contamination (Fig. 3.). However, mean EF values for Ni and Zn were more significant in the dry season, but Pb (0.95–1.81) and Cu were higher in the Wet season. Lower values of metal EF in the dry season can be related to a decrease in anthropogenic activities. In contrast, higher concentrations in the Wet season indicate metal load from

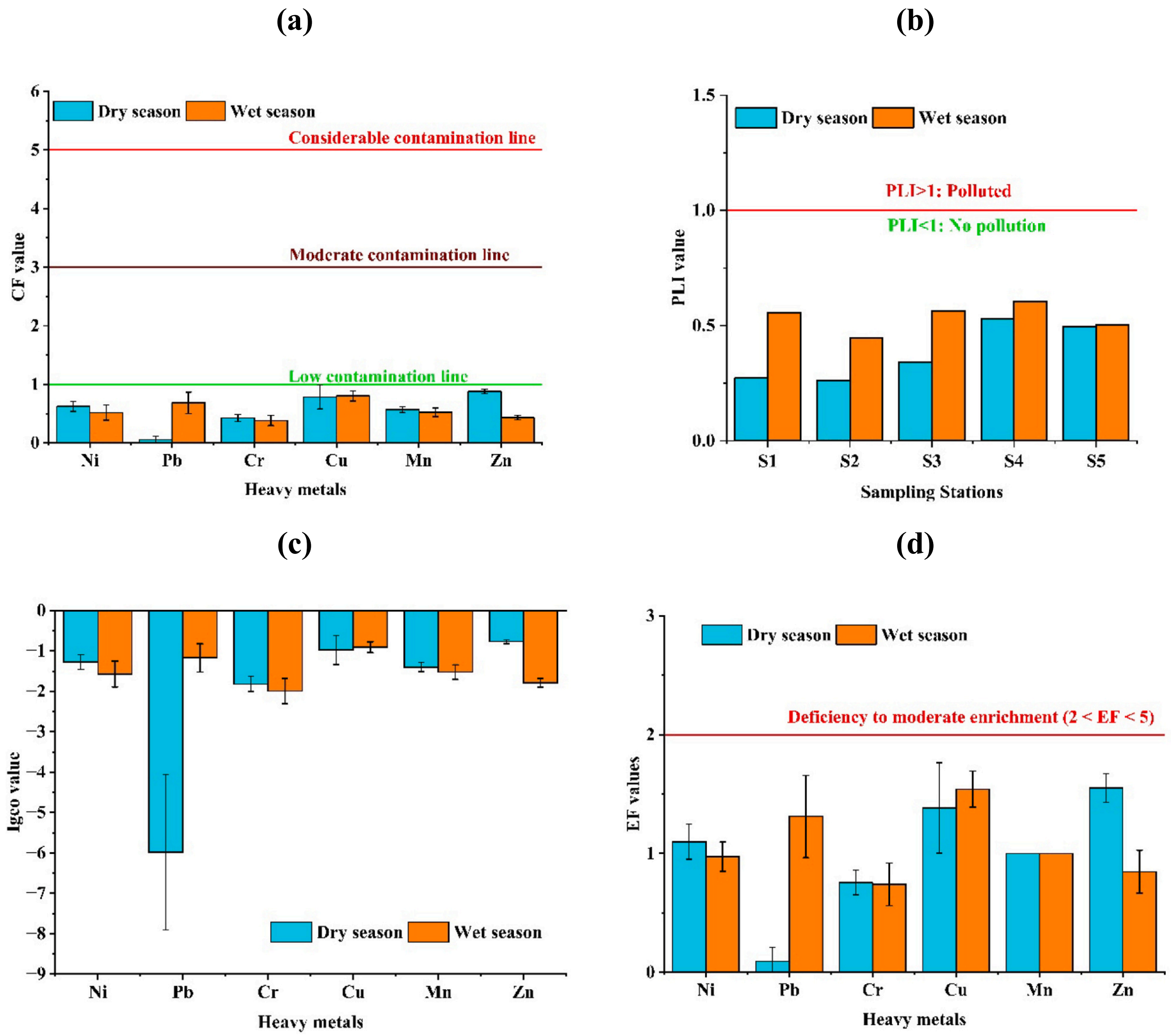


Fig. 3. Contamination factor (CF) (a), pollution load index (PLI) (b), geo-accumulation index (Igeo) (c) and Enrichment factor (EF) (d) for observed metals in mangrove sediment of coastal area in Bangladesh.

anthropogenic sources through rainwater. The total EF values exhibited the sequence of S4 > S3 > S2 > S5 > S1. The high EF values observed in S4 and S3 were attributed to runoff from urban sewage, agricultural land, and river bank erosion, as these sites are situated in close proximity to urban areas and estuaries. Pb and Cu are frequently discharged from shipbreaking and steel companies into the sediment of Sitakunda (Rahman et al., 2019). Consequently, the heavy metals in the mangrove sediments were not a result of enrichment but rather stem from the annual input of heavy metals via seasonal water runoff from anthropogenic sources.

The PLI values ranged from 0.26 to 0.53 during the dry season and 0.45 to 0.61 during the Wet season, confirming that the sediments of the studied sites were lower than 1, indicating no pollution (PLI<1) status (Fig. 3.). All the PLI values were higher at different stations in the Wet season with decreasing order as S4(0.605) > S3(0.564) S1(0.556) > S5 (0.504) > S2(0.447). Islam et al., 2017 and Ahmed et al., 2021a found PLI values >1 in mangrove Sundarbans' sediment and Meghna river upstream's sediments, respectively, indicating less pollution sources in our study area. However, higher PLI values in the Wet season illustrate the potential metallic discharge from shipbreaking operations and the improper use of water for irrigation in nearby agricultural fields fluxed by rainwater runoff (Rahman et al., 2019).

The Geo-accumulation index values (Igeo) showed shallow values (Igeo <0) for all the metals, indicating that sediments of the Cross River Estuary mangrove ecosystem are uncontaminated as a result of anthropogenic activities (Fig. 3.). However, the abrupt increase of Igeo values for Pb from Dry to Wet season indicates that Pb had not come from a regular source year-round but from any anthropogenic sources that flowed through rainwater. Rahman et al. (2019) demonstrated that mangrove sediments near ship-breaking yards exhibited moderate pollution levels (1 < Igeo <2) for Pb. Therefore, the elevated Pb concentration observed in our study area during the wet season could potentially originate from the nearby ship-breaking yards.

To assess heavy metal contamination in sediments based on their toxicity and environmental response, the potential ecological risk index (PERI) was introduced (Hakanson, 1980). Fig. 4(a). and Table 2. present the values of PERI for five heavy metals (Ni, Pb, Cr, Cu, and Zn) in mangrove surface sediment. The PERI value could not be computed for Mn due to unavailability of its biological toxicity factor (Tri). From the PERI calculation, heavy metal pollution by individual elements was determined, revealing no significant potential ecological risk from any metal (PERI<30). However, the ERI values decreased in the order: Cu > Ni > Zn > Cr > Pb (Dry season) and Cu > Pb > Ni > Cr > Zn (Wet season). Particularly, Cu displayed higher ERI values in both seasons in

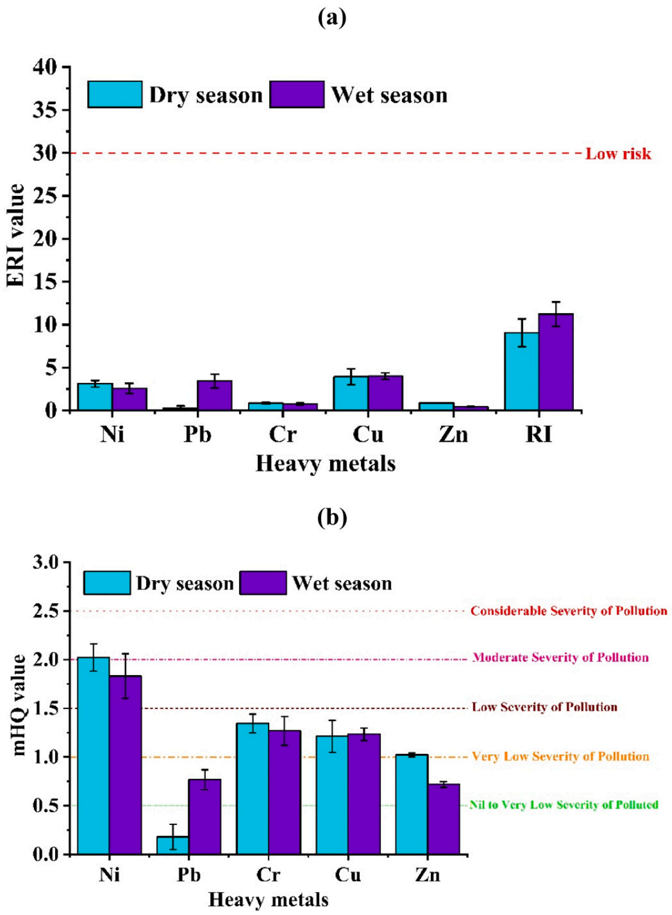


Fig. 4. Potential Ecological risk assessment (PREI) (a), Modified Hazard Quotient (mHQ) (b) for observed metals in the mangrove sediment of coastal area in Bangladesh.

sediment samples, indicating potential origin from anthropogenic activities. Spatially, PERI values for different sediment sites were also below 50, indicating negligible potential pollution. Despite S4 having the highest PERI values in both seasons, its ranking suggests that the increase may be significantly influenced by metals discharged from shipbreaking activities at sampling station S4, located near shipbreaking regions in Sitakunda. As the shipbreaking yards are one of the most considerable sources for Cu accumulation in the sediment and drain through the rainwater in the wet season to the estuarine water (Rahman et al., 2019). According to the study, hazardous metals do not threaten the ecology in the afforested mangrove area. However, sources of potentially hazardous metals, such as shipbreaking yards, are the source of chronic metal pollution. To preserve the afforested mangrove ecosystem, the report recommended implementing an emergency action plan.

Indices and reference values, such as the Pollution Load Index (PLI), Geoaccumulation Index (Igeo), and Potential Ecological Risk Index (PERI), are frequently used in assessing sediment contamination. Nevertheless, each of them has unique benefits and drawbacks in terms of reliability (Birch, 2023). For example, PLI may oversimplify complex environmental interactions, Igeo may disregard regional disparities in baseline metal levels and neglect temporal fluctuations, while PERI might not comprehensively capture the combined impacts of multiple metal pollutants and may not address the resilience and adaptive capacity of ecosystems. These indices frequently depend on simple models that may not comprehensively represent the intricacy of environmental systems and metal-ecosystem interactions (Benson et al., 2018). They mostly concentrate on individual metals and may unintentionally disregard the combined effects of several metal contaminants, whether they are synergistic or antagonistic. Moreover, discrepancies in the levels of metal present in the environment and the influence of natural phenomena might make it difficult to accurately understand the values assigned to certain indicators, thereby resulting in the incorrect assessment of environmental hazards. Moreover, these models may fail to sufficiently consider the fluctuations in metal levels and environmental factors over time and space, thereby restricting their usefulness in diverse geographical areas and time periods (Benson et al., 2018). The reliability and accuracy of these indices are contingent upon the quality of input data, such as metal concentrations and ecological characteristics, which may vary in terms of availability and precision (Birch, 2023). Thoroughly evaluating these constraints is crucial for accurately interpreting findings and formulating management strategies related to the risks of metal contamination.

3.3. *Modified hazard quotient (mHQ)*

The mHQ is employed to assess the harmful effects of an individual heavy metal on aquatic organisms (Benson et al., 2018). Fig. 4(b) depicts the average mHQ values representing the contributions of individual metals, while SF.2. showcases the spatial distribution of mHQ for metals across different sampling stations for ecological risk assessment. The pollution severity for Cr remained consistently very low to low, as indicated by all samples' mHQ values falling between 1.5 and 1 in both seasons, suggesting minimal ecological impact. Cu displayed similar trends, even though with seasonal fluctuations. Ni displayed moderate pollution severity ($2.5 > \text{mHQ} \geq 2$)) in the dry season and low severity ($2 > \text{mHQ} \geq 1.5$) in the wet season, with S4 consistently showing moderate pollution severity across both seasons. Although mHQ values of Pb remained below the unpolluted limit in the dry season, they indicated very low pollution severity ($1 > \text{mHQ} \geq 0.5$) in the wet season. Zn showed very low pollution severity ($1 > \text{mHQ} \geq 0.5$) in the dry season and low pollution severity ($1.5 > \text{mHQ} \geq 1$) in the wet season. Overall, these findings suggest a moderate ecological risk in the mangrove sediment areas, with the increasing concentration of these metals already beginning to significantly disrupt the community structure and environment of the study sites.

3.4. *Tracing metal sources*

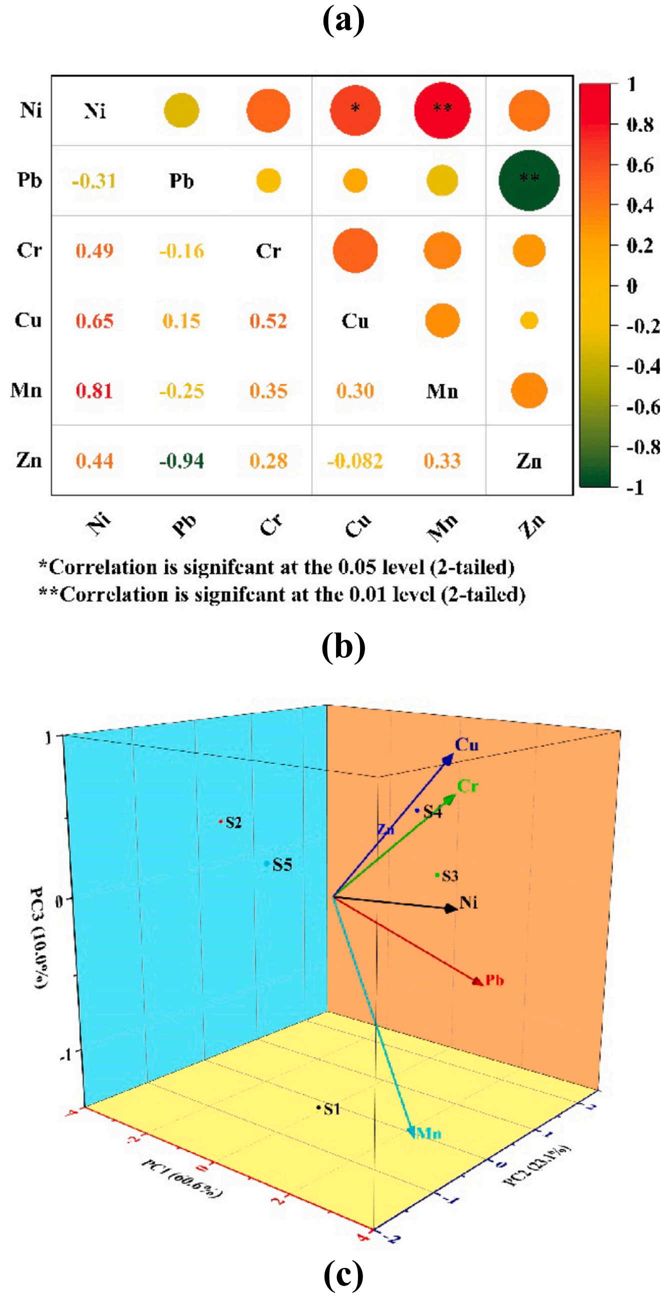
In our study, we observed both significant positive and negative correlations among the trace metals (Fig. 5a). For instance, there were

Table 2
Potential ecological risk index (PERI) of heavy metals in mangrove sediment of coastal area in Bangladesh.

Sampling stations	Ni (ERI)		Pb (ERI)		Cr (ERI)		Cu (ERI)		Zn (ERI)		RI	
	Dry season	Wet season	Dry season	Wet season	Dry season	Wet season	Dry season	Wet season	Dry season	Wet season	Dry season	Wet season
S1	2.82	3.04	0.03	3.92	0.78	0.67	3.29	3.97	0.91	0.39	7.83	11.98
S2	2.53	1.87	0.04	2.65	0.69	0.52	2.55	3.70	0.86	0.48	6.66	9.23
S3	3.29	2.30	0.06	4.63	0.91	0.94	4.15	3.74	0.93	0.44	9.33	12.04
S4	3.52	3.47	0.70	3.52	0.97	0.80	5.21	4.75	0.85	0.45	11.25	12.98
S5	3.43	2.24	0.50	2.42	0.94	0.92	4.45	3.91	0.85	0.42	10.17	9.90

positive and significant correlations between two pairs of the elements: Ni—Mn ($r = 0.81$, $p < 0.01$) and Ni—Cu ($r = 0.65$, $p < 0.05$). This relationship revealed a common source of origin for the positively correlated components (Ahmed et al., 2021a). On the other hand, significant negative correlations were identified in Pb—Zn ($r = -0.94$, $p < 0.01$). Their source may not be natural, for instance, geological rock. However, the mean concentrations of correlated metals (Ni, Cu, Mn, Zn, Pb) were not higher than their average shale values (Table 2), indicating that natural activities could have deposited those metals (Alam et al., 2019).

Analysis revealed that several of the examined metals, namely Cu, Zn, and Pb, exhibited comparable spatial distribution patterns (Fig. 2), with elevated concentrations notably observed in S4. This occurrence could be attributed to the substantial presence of major ship industries within the S4 region. Moreover, the prevalence of shipbreaking activities, alongside various anthropogenic factors such as the utilization of metal-containing pesticides, chemical fertilizers, herbicides, and fungicides in both agricultural and aquacultural practices, likely contribute to the increasing concentrations of Cu, Zn, and Pb (Alam et al., 2019; Hasan et al., 2013a; Rahman et al., 2019).



*Correlation is signifcant at the 0.05 level (2-tailed)
**Correlation is signifcant at the 0.01 level (2-tailed)

Fig. 5. Pearson correlation matrix (a), Principal component analysis (PCA) (b), Hierarchical cluster analysis (HCA) (c) of heavy metals in the mangrove sediment of coastal area in Bangladesh.

PCA (Principal Component Analysis) is employed to discern the variance within datasets by condensing a set of factors and conducting qualitative clustering analysis (Singh et al., 2005). This method visually represents positive and negative correlations among variables within intricate datasets, aiding in the classification of behavioral similarities among metals and their origins (Alam et al., 2019; Rahman et al., 2019; Ahmed et al., 2021b). The results, including loadings, eigenvalue variance (100 %), and cumulative variance for each factor, are summarized in ST.2. and Fig. 5(b). The principal component with the highest eigenvalue is considered the primary contributor to data dispersion. In this study, PCA reveals two primary grouping factors explaining a total variance of 83.67 %. PC 1, with the highest eigenvalue of 3.63, accounts for 60.56 % of the total variance, signifying its dominant and significant role. PC 2 explains 23.11 % of the total variance and exhibits moderate positive loadings (> 0.50) for Pb and Zn. These metals' potential sources may include urban and industrial waste, along with discharges from steel recycling industries such as shipbreaking activities (Mohiuddin et al., 2022). Consequently, the PCA results suggest that the distribution pattern of trace metals in soil fractions varies based on their sources (Rahman et al., 2019).

We further utilized cluster analysis to partition the distinct variability of the research area into groups based on similarities and differences, with each cluster representing sites with similar characteristics (Ahmed et al., 2021a; Hossain et al., 2019, 2021a; Rahman et al., 2019). Fig. 5(c). illustrates a dendrogram and a two-way hierarchical cluster heatmap generated using the Ward linkage method and Euclidean distance. The vertical metal section dendrogram identified two primary groups: Cluster 1 comprising Ni, Cu, Cr, and Mn, while Cluster 2 consisted of Pb and Zn. Both clusters, as indicated by PCA and Pearson's correlation results, suggested a shared origin for the metals. However, Cluster 2 attributed the emergence of elements primarily to metallic discharge, whereas Cluster 1 attributed them to natural weathering. Conversely, the horizontal dendrogram revealed three clusters: S1, S3, and S5 contributing to Cluster 1, S4 to Cluster 2, and S2 to Cluster 3. Our study suggests that elevated concentrations of Cu, Pb, and Zn in sediments from S4 may result from illegal disposal of industrial waste, sewage dumping, and aerial deposition of contaminants. Furthermore, the tidal movements of estuaries serve as a natural source of metal deposition into the mangrove sediment in the other two clusters.

4. Conclusion

Manganese and zinc exhibited the highest levels among the metals studied in afforested mangrove sediments. During the dry season, metal concentrations were generally elevated, attributed to reduced water flow and dilution effects, facilitating increased metal settling in sediments. Conversely, the wet season had lower metal contents, possibly due to the influx of “cleaner” sediments from freshwater sources. Higher freshwater flow during this period promoted sediment transport to the sea, effectively flushing metals from nearshore areas. Additionally, decreased salinity levels favored metal precipitation and binding with sediment particles, reducing their bioavailability. Spatially, Sitakunda demonstrated elevated metal loads in sediment, attributed to shipbreaking activities in the area. Nevertheless, these metal concentrations remained within the average shale value and fell within the recommended guidelines for sediment and uncontaminated in the study area. However, minimal ecological risk has been posed in the coastal mangrove sediment. Moreover, the Modified Hazard Quotient indicated low to moderate pollution severity from the metals, except for Pb. The findings highlighted a considerable risk associated with heavy metals if left uncontrolled in future. PCA, HCA, and Pearson correlation revealed Pb and Zn did not originate from natural sources. So, the primary site for those metal concentrations was identified as Sitakunda, known for its shipbreaking activities. In light of these results, the current study advocates for the implementation of a sustained monitoring program for the mangrove ecosystem of the Meghna estuary connected to the Bay of Bengal. Additionally, it emphasizes the necessity of maintaining heavy metal levels within globally recommended limits by halting shipbreaking activities and untreated industrial effluent discharge.

Funding

This study was supported by Researchers Supporting Project Number (RSP2024/144), King Saud University, Riyadh, Saudi Arabia.

CRediT authorship contribution statement

Md Saifur Rahman: Writing – original draft, Project administration, Methodology, Formal analysis, Data curation. **Moshiur Rahman:** Writing – original draft, Methodology, Investigation, Formal analysis. **Yeasmin N. Jolly:** Methodology, Investigation, Formal analysis, Data curation. **Md Kamal Hossain:** Resources, Methodology, Investigation, Formal analysis, Data curation. **Sanjida Semme:** Resources, Methodology, Investigation, Formal analysis. **Bilal Ahamad Paray:** Writing – review & editing. **Takaomi Arai:** Writing – review & editing. **Jimmy Yu:** Writing – review & editing. **M. Belal Hossain:** Writing – review & editing, Writing – original draft, Supervision, 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 provided in the article.

Acknowledgments

The authors would like to offer thanks to A.S. Safiuddin Ahmed for helping in lab and software support. The authors would like to extend their sincere appreciation to the Researchers Supporting Project Number (RSP2024/144), King Saud University, Riyadh, Saudi Arabia.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2024.116429.

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