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Research article

Heavy metals and metalloid contamination and risk evaluation in the surface sediment of the Bakkhali River estuary in Bangladesh

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ABSTRACT

Current state of contamination and subsequent risk of contaminated sediment of a tidal river of Bangladesh was evaluated in the present study. Sediment samples were collected from five locations in the tributary of Bakkhali River estuary during summer (April) and winter (December) season, 2020. Collected samples were processed using standard protocol and the content of heavy metals (Cd, Cr, Cu, Pb and Zn) and metalloid (As), were analyzed by the Flame Atomic Absorption Spectrometer. Sediment contamination was evaluated by pollution load index (PLI), contamination factor (CF), degree of contamination (*Cd*), potential ecological risk index (PERI), non-carcinogenic and carcinogenic risk (CR) due to the dermal contact of the sediment. Multivariate statistical analysis such as principal component analysis (PCA) and cluster analysis (CA) were also applied to find out the possible sources of the contaminant in the sediment. Results showed the average concentration of As, Cd, Cr, Pb, Cu and Zn was 9.74 ± 3.57, 2.00 ± 0.85, 48.75 ± 8.92, 29.78 ± 8.39, 5.44 ± 2.03 and 56.94 ± 8.57 mg/kg, respectively. Concentration of Cu, Pb and Zn were within the recommended level whereas the concentration of As, Cd and Cr were suppressed the recommended level of WHO and FAO/WHO standards. PLI, CF and *Cd* revealed considerably low degree of contamination of the sediment. Geo-accumulation index indicated uncontaminated to moderately contaminated condition of the sediment. Although the values of enrichment factor revealed no potential enrichment for most of the metals, Cd showed a minor enrichment during the winter season. Based on the ecological risk assessment, the sediment from all of the sample locations was found to be of moderate to low risk. PCA and CA analysis revealed the origin of contaminants mainly from anthropogenic sources. Although different metals showed non-carcinogenic risk to the inhabitants, cancer risk values for dermal contact (CR_{derm}) were much lower than 10^{-6} indicating no cancer risk for adult and child. However, the findings also revealed that children were more susceptible to CR_{derm} compared to adults. The present study concluded that long term dermal contact of the sediment of Bakkhali River estuary will be contagious to the people. Therefore, regular monitoring of the estuarine environment is necessary so that contamination does not get worse.

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1. Introduction

Riverine sediments are very vulnerable to pollution by heavy metals and metalloids [1]. Heavy metals and metalloid (HMs-M) in sediment exist in several chemical forms and undergo transformations in their speciation as they are transported through the river system. These changes are a result of dissolution, precipitation, absorption, and complication events. The amount of HMs-M in river sediments is determined by a number of factors, including the kind and amount of contaminant, the location, type and their sources, the geological and climatic circumstances, and the land use structure [2–4]. Key causes of contamination in riverine ecosystem comprise agriculture, industry, domestic and others [5–8]. Generally, the findings of various researches have revealed that HM concentration may be graded depending on the following land use features: industrial, urban, agricultural, and natural [9]. As a result of its location in an industrialized area, the river system in Bangladesh is becoming contaminated because of the influx of untreated wastewater, sewage, and industrial waste originated from towns and companies [10,11].

Sediments did not preserve the HMs-M perpetually and suspend into surface water with the changing of environmental conditions. Which are causing secondary pollution to aquatic organisms [12,13]. HMs-M present in the sediment has detrimental impacts on the benthic ecosystem and significantly harms other living things via their transmission throughout the food chain [14,15]. These toxic metals may transport via the gills, bile, kidneys, and skin of organisms, leading to alterations in organ function as well as modifications in their muscle taste and odour [16,17]. Furthermore, the high lipid solubility of HMs-M prevents their decomposition and transformation when entering the vital organs of aquatic organisms. This phenomenon will persistently be present in the surrounding environment and accumulated in aquatic species, hence leading to the relocation of hazardous materials to public health via the food chain [18,19]. Therefore, bioaccumulation and biomagnification of HMs-M in living organisms has been considered a major concern worldwide.

Estuarine rivers of Bangladesh are severally affected by HMs-M contamination. Current financial transition in Bangladesh has led to fast mechanization and unplanned urbanization, dumping huge amounts of toxic pollutants into the waterways [20,21]. These pollutants are ultimately discharged into the Bay of Bengal through numerous estuaries. Extensive research has been conducted on the presence and dispersion of high levels of metallic elements in the sediment of estuaries and coastal areas in several rivers of Bangladesh. For example, distribution of HMs-M in the sediment of Meghna River estuary [22], Karnaphuli River estuary [23], Feni River estuary [24] and Passur River [8] have been investigated. The research on HMs-M contamination in estuarine sand not only highlights environmental health concerns but also provides insights into human activities in the surrounding regions [25].

The Bakkhali River estuary is situated on the southeast coast of Bay of Bengal. This estuarine river is vital to local fisheries and livelihoods [26]. This estuarine zone is nutrient-rich [27]. Pollution, urban effluent, and sedimentation hinder this estuary's ecology [28]. Most of Cox's Bazar's home, municipal, and industrial effluents are released into the sea via uncontrolled canals along the river. Boat repair industry waste discharge and chemical spills pollute this region's water and sediments. HMs-M concentrations in sediments and water in the Bakkhali River estuary have been studied [29,30]. But no scientific investigation on HMs-M in sediment in the research region has been carried out up to now. Limited studies which have been carried out so far did not address the ecological and public health concern. Therefore, a systematic study is needed to investigate the sediment with the distribution, possible source,

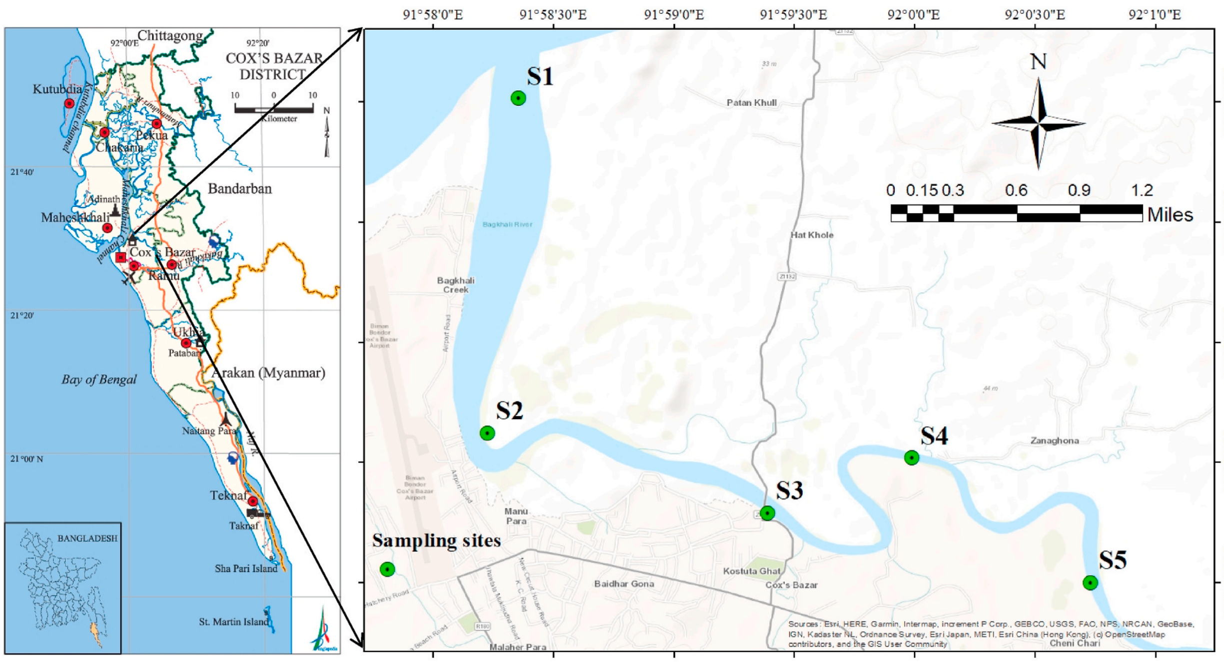

Fig. 1. Location of the sampling sites in the Bakkhali River estuary, Cox's bazar, Bangladesh.

contamination, risk assessment on ecology and public health. Thus, the present research examined the contamination status of the Bakkhali River estuary by evaluating the HMs-M concentrations and their subsequent ecological risks. Furthermore, the research was also examining the non-carcinogenic and carcinogenic risk of toxic HMs-M on adult and children population of the adjacent area. Our study will provide some theoretical framework and scientific aspects for ecological situation to evaluate the possible danger of HMs-M in this area. A systematic study is still lacking to associate the sediment with the distribution, characteristics, risk assessment, possible sources, and impact of HMs-M in the Bakkhali River estuary.

2. Materials and methods

2.1. Study area and sampling

The present study was conducted in the Bakkhali River estuary, Bangladesh during the summer (April) and winter (December) season of 2020. A total of five sampling stations were selected to collected sediment samples (Fig. 1). The sampling stations were as follows: S1 near mouth of Bay of Bengal (21°28′30.91ℙN, 91°58′21.32ℙE), S2 (21°27′05.84ℙN, 91°58′09.24ℙE), S3 (21°26′47.39ℙN, 91°59′23.46ℙE), S4 (21°27′01.12ℙN, 91°59′59.47ℙE) and S5 (21°26′29.97ℙN, 91^{0}00′43.31ℙE). Among the sampling locations, S1 and S2 are located near the estuary mouth and most of the industrial zones are located in these sites. Therefore, these two sites are more polluted with the effluents from different industries. Site S3 and S4 are located upstream of the estuary and are mostly influenced by the discharge of household garbage, septic tanks and market waste. Site S5 is located far from the estuary mouth and mostly influenced by the agricultural activities. Sediment samples (1 kg) were collected at depths ranging from 0 to 5 cm using a portable Ekman dredge sampler during the low tidal period. Triplicate samples were collected from each station and mixed thoroughly to prepare one sample. Therefore a total of 30 samples were collected during summer and winter season, respectively. Subsequently, the samples were carefully placed in a plastic bag and transported to the laboratory. In the laboratory, the samples were subjected to oven drying at 80 °C for duration of 24 h. Oven-dried samples were pulverized using a grinder and mortar, filtered through a 2 mm screen, and stored in an airtight, sanitary zip lock bag at a temperature of 8 °C while waiting for the chemical examination. A methodological flowchart of the present study is shown in Fig. 2.

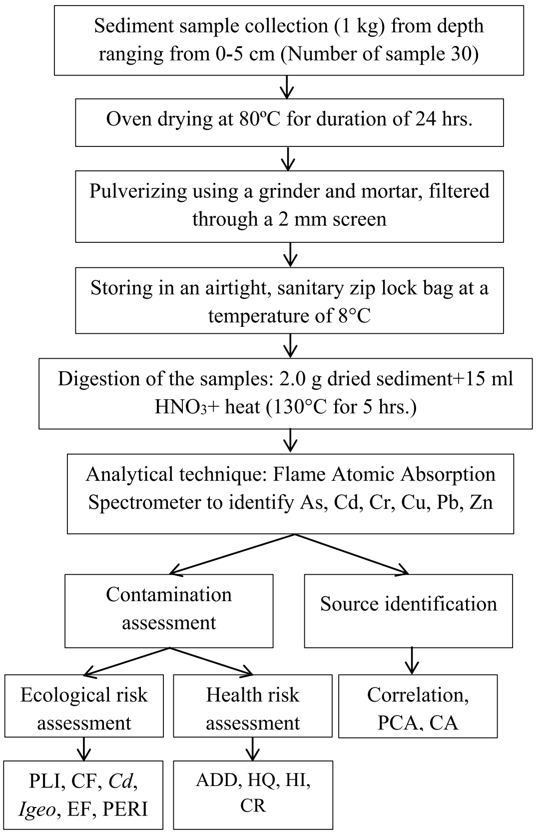

Fig. 2. Methodological flowchart of the study.

2.2. Digestion of the sample

A total of 2.0 g of dried sediment was collected for digestion. The sediment sample was combined with 15 ml of concentrated nitric acid (HNO_3) in a 100 ml beaker for digestion. The contents were subjected to a temperature of 130 °C for duration of 5 h. After being digested, the samples were diluted with deionized water to volume of 100 ml. Prior to the dilution, they were rinsed with 0.1 M HNO_3 and then passed over filter paper (Whatman no. 41).

2.3. Metal analytical technique

The analytical procedure was performed using deionized water. Every glass and container underwent a washing process using a solution of 20 % nitric acid, followed by thorough drying in an oven after being cleaned with deionized ultrapure water. The Shimadzu Flame Atomic Absorption Spectrometer (Model AA-6800) identified the elements arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), and zinc from the sediment samples.

2.4. Quality assurance and quality control

The correctness of the data for the research was validated by quality assurance (QA) and quality control (QC). Analytical blank (control sample) and spike samples were collected for every HMs-M to provide QA and QC. Samples without silt, samples with a multi-standard spike, and repeated samples were all analyzed simultaneously also to ensure the quality assurance. Tests were accomplished on three replicates to assure precision, and only the mean data was used. To avoid contamination, all laboratory equipment underwent a rigorous rinsing with distilled water, followed by immersion in a 10 % HNO_3 solution for at least 24 h. The Atomic Absorption Spectroscopy (AAS) was calibrated using conventional laboratory standards. The analytical approach used the certified reference material DORM-4 Fish protein, provided by the National Research Council of Canada, for the analysis of HMs-M. The proportion of the recovered amount ranged from 90 % to 99 %. The analytical parameters and technique for measuring HMs-M in a sample using AAS are presented in Supplementary Tables 1 and 2

2.5. Contamination assessment of HMs-M

2.5.1. Pollution load index

PLI is the nth root of metal CF multiplications calculated using the method of Tomlinson et al. [31]:

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \ldots \ldots \ldots. \times CF_n)^{1/n} \tag{1}$$

The PLI value of zero describes excellence, PLI = 1 specifies standard pollution level and PLI > 1 stipulates the deterioration of the waterbody.

2.5.2. Contamination factor (CF) and degree of contamination (C_d)

CF represents the ratio of the measured concentration to the baseline value of HMs-M. The determination of CF was conducted according to the method described by Tomlinson et al. [31]:

$$CF = C_{metal}/C_{background} \tag{2}$$

CF values classified the metal contamination as low-degree (CF < 1), moderate-degree ($1 \leq CF \leq 3$), considerable-degree ($3 \leq CF \leq 6$), and very high-degree ($CF \geq 6$) [10,23].

The value of *Cd* is calculated as the sum of the contamination factor (CF) as:

$$C_d = \sum_{i=1}^{n} CF \tag{3}$$

Cd is classified as low ($C_d < 8$), moderate ($8 \leq C_d < 16$), considerable ($16 \leq C_d < 32$) and very high degree of contamination ($C_d \geq 32$) accordingly [32].

2.5.3. The geo-accumulation index (Igeo)

The I_{geo} index was evaluated by Muller [33] as follows:

$$I_{geo} = \mathrm{Log}_2\ (C_n / 1.5 \times B_n) \tag{4}$$

where, Cn represents the concentration of the HMs-M (n), Bn represents background concentration of the metal, and factor 1.5 means the background matrix. I_{geo} values are classified as uncontaminated ($I_{geo} \leq 0$), uncontaminated to moderately contaminated ($0 \leq I_{geo} \leq 1$), moderately contaminated ($1 \leq I_{geo} \leq 2$), moderately to heavy contaminated ($2 \leq I_{geo} \leq 3$), heavy contaminated ($3 \leq I_{geo} \leq 4$), heavy to extremely contaminated ($4 \leq I_{geo} \leq 5$) and extremely contaminated ($I_{geo} > 5$).

2.5.4. The enrichment factor (EF)

The enrichment factor (EF) is determined using the formula:

$$EF = (C_M / C_{X\ Sample}) / (C_M / C_{X\ Earth\ crust}) \quad (5)$$

Where, C_M is the concentration of the HMs-M and C_X is their reference value of the HMs-M which was Mn [34]. Values of EF can be ranged between 1 and 25 whereas 1 indicates no enrichment, 1–3 minor enrichment, 3–5 moderate enrichment, 5–10 moderately severe enrichment, and 10–25 severe enrichment [35,36].

2.5.5. Potential ecological risk index

Potential ecological risk index (PERI) was determined after Hakanson [37] following the formula:

$$RI = \sum E_f^i \quad (6)$$

Where RI indicates risk index and E_f^i is the PERI for the contamination of individual HMs-M and calculated using the formula of -

$$E_f^i = C_f^i \times T_f^i \quad (7)$$

T_f^i represent the toxicity response coefficient of individual HMs-M. The pollution index is denoted as C_f^i and determined using the following formula:

$$C_f^i = C_s^i / C_n^i \quad (8)$$

Where, C_s^i and C_n^i represents the HMs-M content in the sediment controlled samples, respectively. E_r^i is characterized into the five groups as low (E_r^i <40), moderate ($40 \leq E_r^i$ <80), considerable ($80 \leq E_r^i$ <160), high ($160 \leq E_r^i$ <320) and very high risk ($E_r^i \geq 320$), respectively [34]. PERI was categorized as PERI low (<65), moderate (65 ≤ PERI <130), considerable (130 ≤ PERI <260) and very high (≥260) [37,38].

2.5.6. Non-carcinogenic and carcinogenic risk assessment

Humans may come into touch with sediment via activities such as bathing and washing. The Average Daily Dose (ADD) of the sediment for dermal contact was determined following Iqbal and Shah [39] as:

$$ADD_{dermal} = Cs \times SA \times Kp \times ET \times EF \times ED \times CF/BW \times AT \quad (9)$$

where, Cs denotes the average HMs-M content (mg/kg); SA measures the exposed area of skin (6600 cm^2 for the children and 18,000 cm^2 for the adults); Kp denotes the dermal penetrability coefficient. The Kp values are as follows: 0.0001 cm/h for Pb, 0.002 cm/h for Cr, 0.001 cm/h for Cd, As, and Cu, and 0.0006 cm/h for Zn; ET represents the exposure time (0.6 h/day); EF denotes the frequency of exposure (365 days/year); ED is the exposure duration (6 and 30 years for the children adult, respectively); AT is the ED × 365 for non-carcinogenic risk (2190 for the children and 10950 for the adult, respectively). CF denotes the unit conversion factor (0.001 L/cm^3).

Non-carcinogenic hazard quotient (HQ_{derm}) was determined using the equation as:

$$HQ_{dermal} = ADD_{dermal}/ RfD_{dermal} \quad (10)$$

where, RfD is the reference dose [40]. HQ > 1.0 defines an unacceptable risk of non-carcinogenic effects and HQ < 1.0 stipulates a tolerable level of risk for public health [41]. Hazard index (HI_{dermal}) was determined by Li et al. [42] as follows:

$$HI_{dermal} = \sum_{i=1}^{n} HQ_{derma}l \quad (11)$$

where, HI_{dermal} is the possible risk through dermal contact of HMs-M, i is the routes of contact; n is the type of HMs-M; HI > 1 unacceptable risk and HI < 1 tolerable value of non-carcinogenic risk on health.

The cancer slope factor (CSF) was employed to assess the carcinogenic risk. Cancer risk was assessed according to the formua

$$CR_{dermal} = ADD_{dermal} \times CSF \quad (12)$$

where, CR_{dermal} is the cancer risk through dermal contact of HMs-M. The allowable unit for lifetime CR exposure varies from 10^{-6} to 10^{-4} [43]. A CR score more than 10^{-4} suggests the likelihood of a carcinogenic risk [44].

2.6. Statistical analysis

All the relevant data was coded and presented as mean, and stander deviation. Assumptions of parametric verified and the normality of the data were tested by Shapiro-Wilk, and Kolmogorov-Smirnov test. HMs-M content of the sediment samples were tested for the significant differences among the stations and seasons using a one-way analysis of variance and *t*-test, respectively. Post-hoc analysis evaluated using Duncan multiple range test (DMRT), whereas P-value were considered significant at < 0.05. The relationship between all the HMs-M in the sediment samples was investigated using Pearson correlation coefficient. Multivariate statistical analyses (MVSA) such as principal component analysis (PCA) and cluster analysis (CA) were applied to identify the sources and distribution of HMs-M in the sediment. Cluster analysis was performed using two-way hierarchical cluster (heat map) with ward linkage method and Euclidean distance. ANOVA was performed in SPSS(Statistical Package for Social Sciences, version 25.0, IBM Corporation, Armonk, NY, USA) while MVSA were performed in Origin (Pro), Version 2024b, OriginLab Corporation, Northampton, MA, USA.

3. Results

3.1. Concentration of HMs-M in the sediments

River sediment contaminated with HMs and metalloids can pose a major threat to the aquatic ecosystem. Seasonal distribution of six HMs namely cadmium (Cd), chromium (Cr), lead (Pb), copper (Cu), zinc (Zn) and one metalloids namely arsenic (As) at five different locations in the sediment of Bakkhali River are given in Fig. 3. The mean concentration of As, Cd, Cr, Pb, Cu and Zn during the current study was recorded as 9.74 ± 3.57, 2.00 ± 0.85, 48.75 ± 8.92, 29.78 ± 8.39, 5.44 ± 2.03 and 56.94 ± 8.57 mg/kg, respectively. In both seasons, concentration of HMs-M in the sediment increased as Cd < Cu < As < Pb < Cr < Zn; additionally, investigated concentration of HMs-M in the sediment of Bakkhali River were significantly varied among the study locations and seasons (Supplementary Tables 3 and 4).

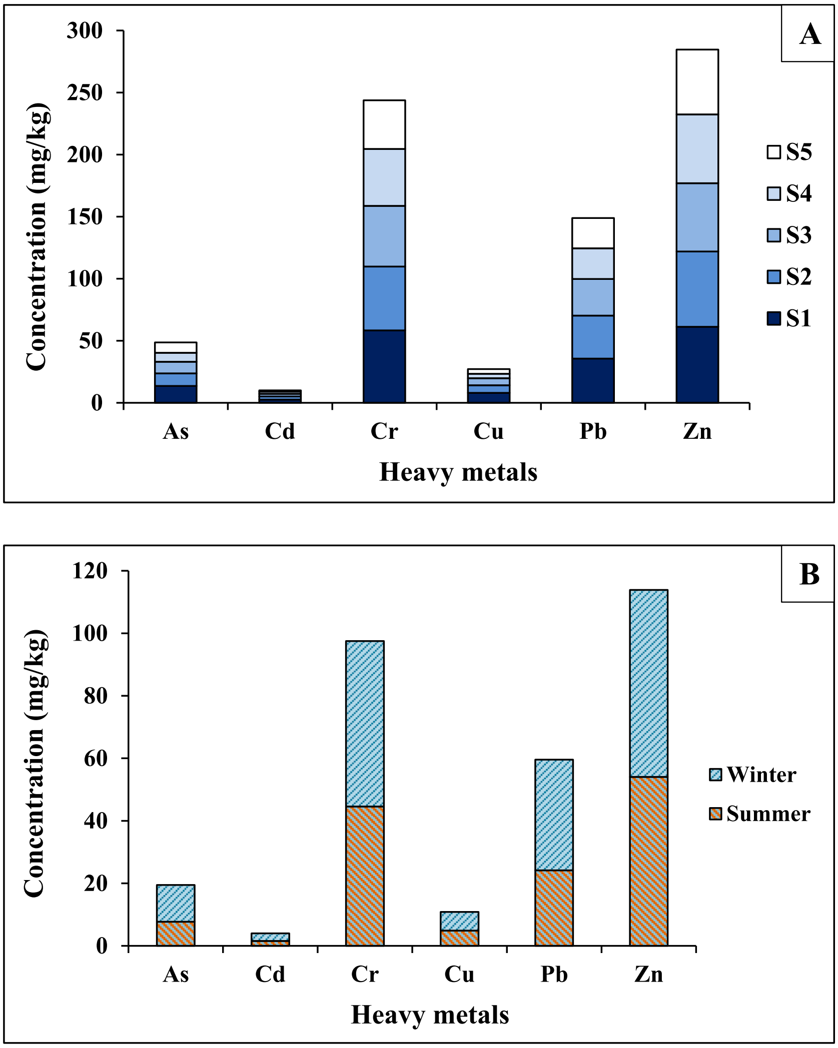

Fig. 3. HMs-M in the sediment of Bakkhali river estuary during among different stations (A) and seasons (B).

3.2. Correlation analysis and source appointment of HMs-M

The correlation matrix of the analyzed HMs and metalloids in the sediment sample under the study provides information on the inter-metal interaction (Table 1). Statistically significant correlations ($P < 0.05$) were found between the different HMs and metalloids suggesting that these contaminants have a common source of origin in the sediments of the Bakkhali River estuary.

The PCA was resulted in two principal factors, whereas PC 1 contributed 90.05 %, and PC 2 contributed 6.64 % (Fig. 4A). Meanwhile, these two PC contributed with the cumulative variance of 96.69 %. All the metals and metalloids were distributed in two quadrant of the biplot, whereas As, Cu were confined into the first quadrant and Cr, Cd, Pb and Zn were ran in the second quadrant. Arsenic and Cu is a common ingredient in phosphate fertilizers, fungicides and algaecides which are predominantly used in agricultural practices. Cr, Cd, Pb and Zn are used in electroplating, batteries, chemical plants, alloys and foils materials. Therefore, the primary factor responsible for the presence of HMs-M in the sediment of Bakkhali River was likely to be anthropogenic sources, such as pollution from river or sea water. A similar group of organization among the contaminants was also observed in cluster analysis (Fig. 4B). Dendogram developed by ward linkage method depicted two distinct clusters both vertically and horizontally. In vertical position, cluster 1 was consisted with As, Cd and Cu, while other metals were grouped together in cluster 2 and strongly confirmed their similar source of origin. In horizontal layout of the dendogram, sampling stations were divided into two distinct clusters, whereas S1, S2 formed cluster 1 and S3, S4 and S5 amalgamated in cluster 2.

3.3. Assessment of contamination

3.3.1. Pollution load index

The overall pollution load index (PLI) for the sediment sample were found higher in winter compared to summer and follows the descending order of S1 (0.98, 0.69) > S2 (0.84, 0.64) > S3 (0.75, 0.59) > S4 (0.66, 0.43) > S5 (0.54, 0.45). The calculated PLI values of all studied metals ranged from 0.43 (S4 in summer) to 0.98 (S1 in winter) with a mean value of 0.66 during the current study period (Fig. 5). The present findings revealed that, the PLI value of different seasons and stations along with the mean value were much lower compared to the threshold value of '1', which also confirmed that Bakkhali sediment are not polluted.

3.3.2. The contamination factor (CF) and degree of contamination (Cd)

The contamination factor (CF) and degree of contamination (Cd) are represented in Fig. 6. The mean CF value ranged from 0.14 to 2.00 and follows a descending order of Cd > Pb > As > Zn > Cr > Cu. In the present study, maximum CF value was recorded for Cd (3.18) whereas minimum value was found for Cu (0.07). However, the evaluated CF values for all the metals (except Cd and Pb) were less than threshold value of '1', indicating the studied sediments produce a low level of contamination (Fig. 6A). Degree of contamination (*Cd*), provide a complete assessment of metal contamination. The present findings reveal that, *Cd* of all stations and seasons showed a considerable low degree of contamination (except Site-1 in winter) and followed the descending order of S1 > S2 > S3 > S4 > S5 (Fig. 6B). The mean *Cd* was 5.41, indicating low contamination of the sediment.

3.3.3. The geo-accumulation index (Igeo)

The geo-accumulation index (I_{geo}) is an ecological index for determining the level of HMs-M contamination in the sediment samples (Fig. 6C). The estimated average I_{geo} for the metals in the Bakkhali River sediment was delineated in the following order: Cd (0.09) > Pb (−0.02) > As (−0.39) > Zn (−0.40) > Cr (−0.45) > Cu (−1.07). In the present study, Cd (0.33) had the maximum I_{geo} value while Cu (−1.07) had the minimum I_{geo} value. According to I_{geo} value proposed by Mullar (1979), mean I_{geo} value was uncontaminated to moderately contaminate at the sampling sites. On the contrary, the mean I_{geo} values for the rest of the other studied metals represent that all the sampling sites along the Bakkhali River are unpolluted ($0 > I_{geo}$) for As, Cr, Cu, Pb and Zn.

3.3.4. The enrichment factor (EF)

The enrichment factor of the HMs-M in the sediment samples is shown in Figrue 6D. The average EF value was less than 1 and arranged as Cu (0.05) > Cr (0.18) > Zn (0.20) > As (0.22) > Pb (0.50) > Cd (0.67). In the present study, maximum and minimum EF value was recorded for Cd (1.06) from S1 in winter and Cu (0.02) from S4 in summer, respectively. EF value of all studied metals indicated no enrichment (EF < 1) in all the sites along the Bakkhali River estuary whereas minor enrichment of Cd (1.06) was observed at S1 during winter.

Table 1
Correlation coefficients among the HMs of the sediment samples.

	As	Cd	Cr	Cu	Pb	Zn
As	1					
Cd	0.640[a]	1				
Cr	0.659[a]	0.726[a]	1			
Cu	0.718[a]	0.680[a]	0.680[a]	1		
Pb	0.692[a]	0.665[a]	0.668[a]	0.640[a]	1	
Zn	0.475[a]	0.518[a]	0.377[b]	0.339	0.451[b]	1

[a] Correlation is significant at the 0.01 level (2-tailed).
[b] Correlation is significant at the 0.05 level (2-tailed).

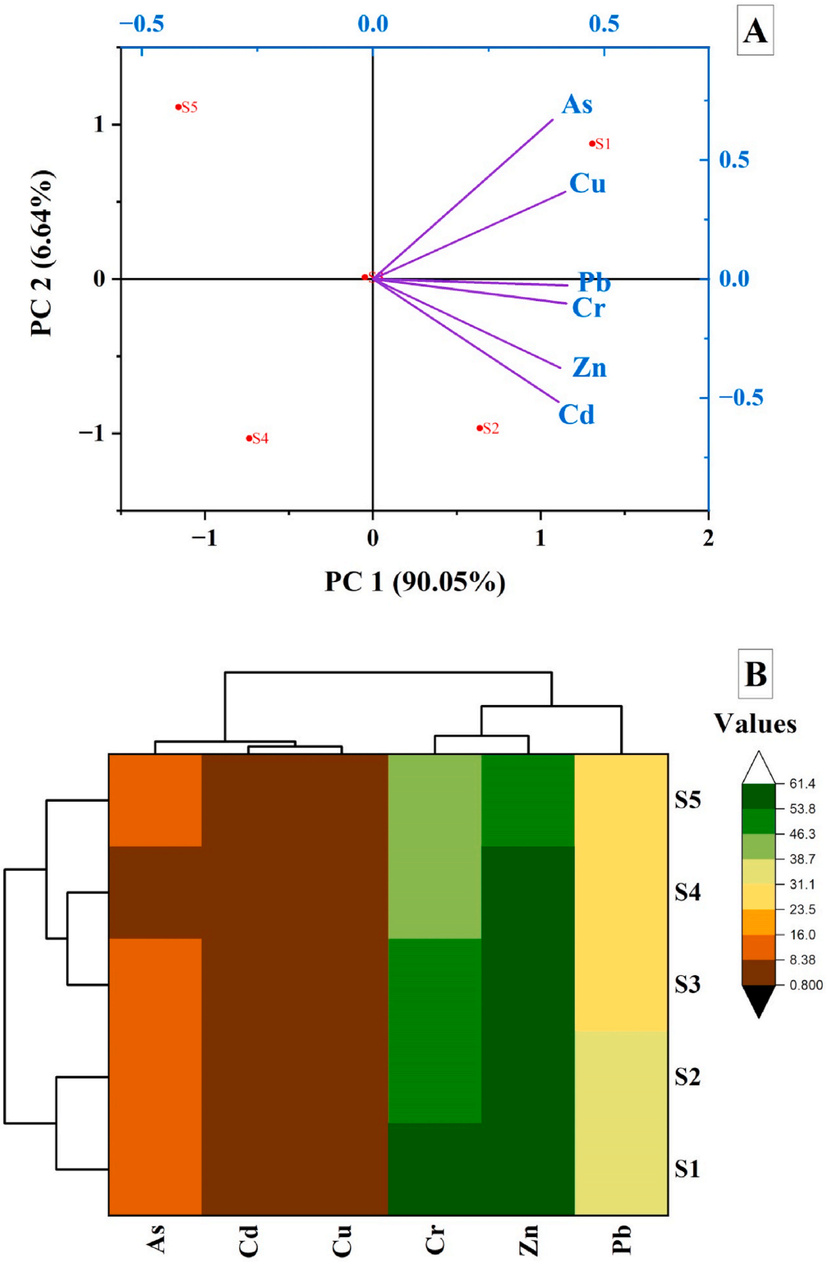

Fig. 4. PCA and cluster analysis among the HMs-M of the sediment of Bakkhali river estuary.

3.3.5. Potential ecological risk

The ecological risk factor (E_r^i) for individual metal and potential ecological risk index (PERI) are summarized in Fig. 7A. The ecological risk factors of HMs-M decreased as Cd (59.96) > Pb (7.45) > As (6.50) > Cr (1.08) > Cu (0.68) > Zn (0.60). Cd produced a moderate to considerable ecological hazard with E_r^i values in the range of 26.27–95.46 (with a mean value 59.96). PERI showed potential risk due to Cd exposure in all the sites except for S5 (Fig. 7B). Therefore, the average value of PERI for the studied metals was found 76.26 with a minimum and maximum PERI value of 38.04 (S5 in summer) and 120.21 (S1 in winter), respectively. The present findings revealed that all the stations exhibit moderate risk (65 ≤ PERI <130) whereas S3, S4 in summer and S5 in both seasons specify low risk (PERI <65) by the HMs-M in the Bakkhali River estuary.

3.3.6. Non-carcinogenic and carcinogenic risk assessment

The estimated ADD (dermal), HQ (dermal), CR (dermal) and HI values of the studied HMs-M for both adult and child are summarized in Table 2. In the current study, Zn exhibit highest ADD_{derm} value for both adult (2.15E-07) and child (1.21E-06) compared to

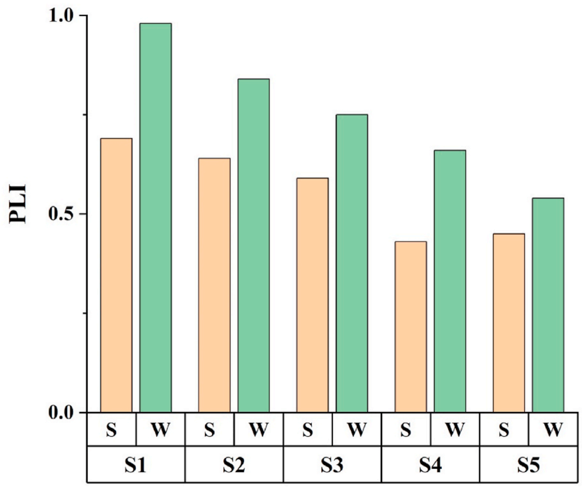

Fig. 5. Pollution load index (PLI) of the HMs-M of the sediment of Bakkhali river estuary. S1-S5 indicates sampling sites and S and W indicates summer and winter season, respectively.

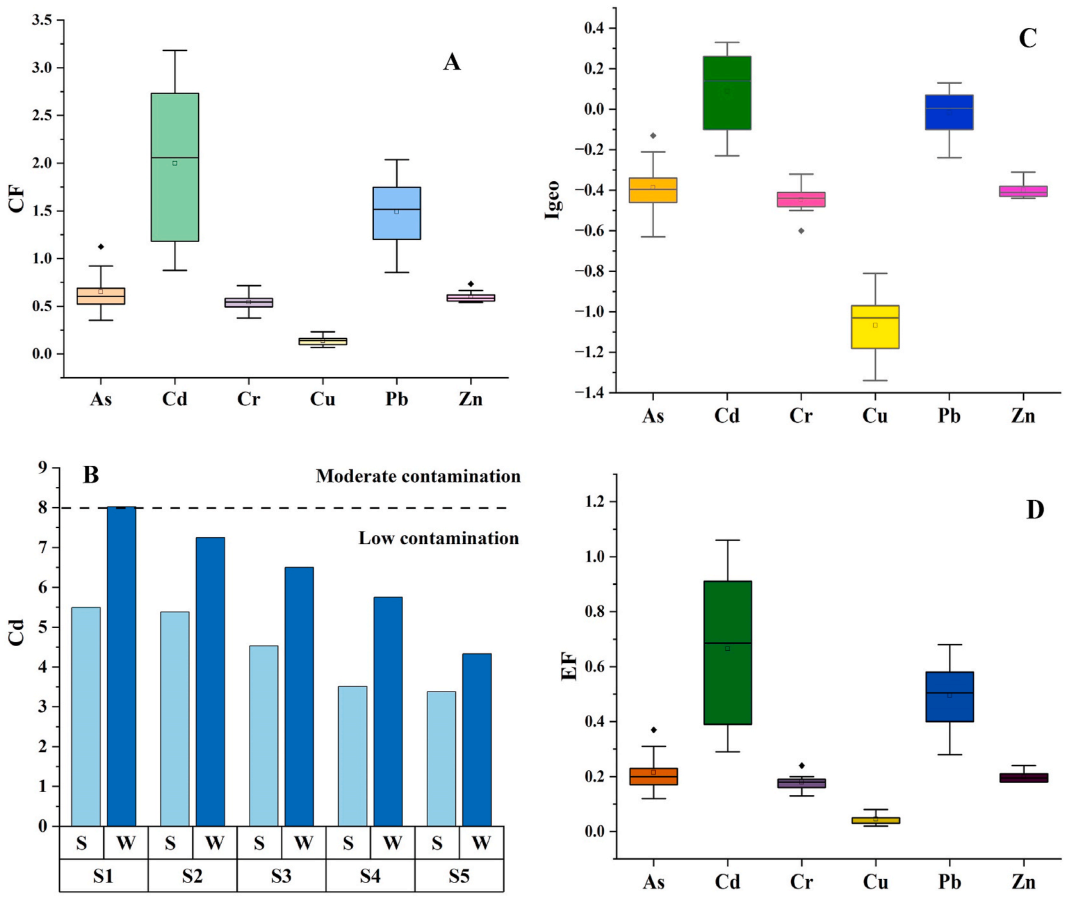

Fig. 6. Contamination factor (CF), degree of contamination (Cd), geo-accumulation index (Igeo) and enrichment factor (EF) of the HMs-M of the sediment of Bakkhali river estuary. A – CF, B – Cd, C – Igeo and D – EF. S1-S5 indicates sampling sites and S and W indicates summer and winter season, respectively.

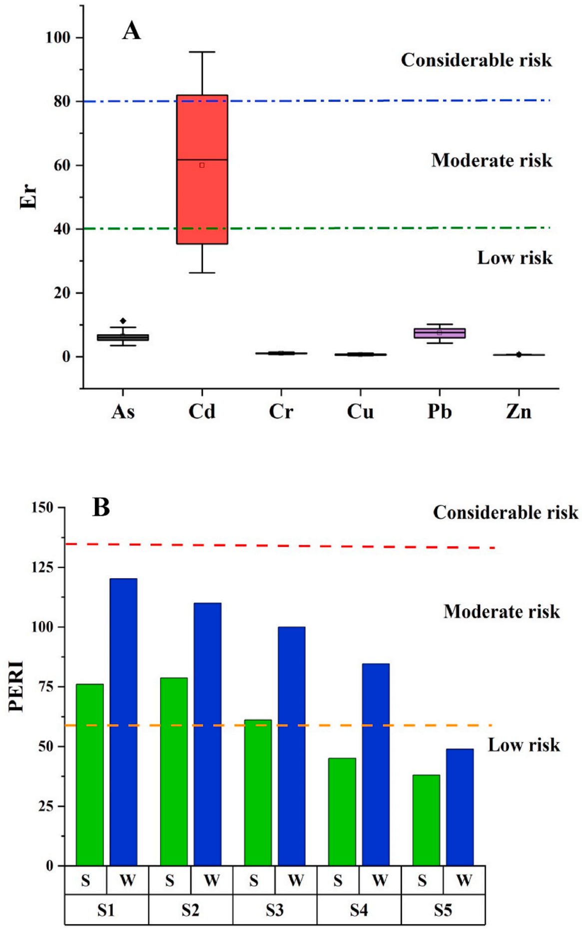

Fig. 7. Ecological risk factor (Er) and potential ecological risk index (PERI) of the HMs-M of the sediment of Bakkhali river estuary. A – Er, B – PERI. S1-S5 indicates sampling sites and S and W indicates summer and winter season, respectively.

Table 2
Health risk of HMs in the sediment of Bakkhali river estuary.

	Non-carcinogenic risk of adult		Non-carcinogenic risk of child		Carcinogenic risk of adult	Carcinogenic risk of child
	ADD_{derm}	HQ_{derm}	ADD_{derm}	HQ_{derm}	CR_{derm}	CR_{derm}
As	3.67E-08	2.16E-03	2.08E-07	1.22E-02	5.51E-08	3.12E-07
Cd	7.53E-09	1.51E-03	4.26E-08	8.53E-03	4.75E-08	2.69E-07
Cr	1.84E-07	2.45E-03	1.04E-06	1.39E-02	9.19E-08	5.20E-07
Cu	2.05E-08	1.71E-06	1.16E-07	9.66E-06	–	–
Pb	1.12E-07	2.67E-04	6.35E-07	1.51E-03	9.54E-10	5.40E-09
Zn	2.15E-07	3.58E-06	1.21E-06	2.02E-05	–	–
HI		6.39E-03		3.62E-02		

other HMs-M. In both adult and child HQ_{derm} value was found maximum for Cr (2.45E-03, 1.39E-02) and minimum for Cu (1.71E-06, 9.66E-06) respectively. HI value was found higher in child (3.62E-02) compared to adult (6.39E-03) during the study period. Furthermore, in both adult and child Cr displayed the highest CR_{derm} (9.19E-08, 5.20E-07) value whereas Pb represent lowest (9.54E-10, 5.40E-09) value respectively. No carcinogenic risk was indicated by the CR_{derm} value, which was lower than 10−6 for all HMs-M in both adults and children.

4. Discussion

Sediment is an important and constantly changing part of river ecosystems, and it is often used to evaluate pollution in the environment and in geochemical studies. During the present study, HMs-M was detected in all the sediment samples and the average concentration was directed as Zn (56.94 ± 8.57 mg/kg) > Cr (48.75 ± 8.92 mg/kg) > Pb (29.78 ± 8.39 mg/kg) > As (9.74 ± 3.57 mg/

kg) > Cu (5.44 ± 2.03 mg/kg) > Cd (2.00 ± 0.85 mg/kg). In the riverine ecosystem, higher and lower concentration of Cr and Cd were also previously reported by Kubra et al. [45] in the sediment of Rupsha River. Furthermore, in the present study, HMs-M was abundant in the winter months compared to summer. Seasonal influence on HMs-M content was also previously reported by Ali et al. [46] in Karnaphuli River and Islam et al. [47] in Halda River. It can be assumed that low water flow during winter months might concentrated higher amount of HMs-M in the sediment. Comparison of the present findings with other national, international and reference value (TRV) is shown in Table 3.

Zinc (Zn) was the most ubiquitous element in the sediment of Bakkhali River. Zn concentration (51.37–56.94 mg/kg) was below the TRV value reported by USEPA [53]. Furthermore, the recorded HMs-M concentration was also lower than the stated value (88.58–118.55 mg/kg) of Chakraborty et al. [54] in the Mongla port area, Bangladesh and higher than Siddique et al. [22] in the Meghna River (42.41 mg/kg). However, Kodat et al. [51] and He et al. [54] recorded a higher concentration Zn in the Black sea estuary (94.16 mg/kg) and Yangtze River estuary (82.92 mg/kg), respectively. Zn is a naturally occurring element that is often regarded as a pollutant in many sources such as agricultural and food waste, lubricating oil, waste water from the shrimp and fish industry, runoff from local markets, battery and pharmaceutical industries, and the manufacturing of insecticides and antifouling paints.

Mean Cr concentration was recorded as 48.75 ± 8.92 mg/kg where, winter exhibit the higher Cr concentration. In all the samples, Cr concentration was higher than TRV value of 26 mg/kg [53]. According to Hossain et al. [17], Cr is a powerful oxidizing agent with corrosive and oxidizing qualities that is very toxic and is widely employed in rust control, tanning, plating, and pigment manufacturing. Agricultural and industrial effluents, as well as untreated sewage and ship breaking operations, may contribute to the elevated Cr content in the research region.

Average lead (Pb) concentration (29.78 mg/kg) in the sediment of the Bakkhali River estuary remained within the TRV suggested by USEPA [53]. Concentration of HMs-M in the present study was lower compared to Pb (41.37 mg/kg) of Black sea estuarine sediment [51], while it was higher (3.37 mg/kg) than the sediment of the Terme River, Turkey [52]. Leaded petrol, crude oil pollution from cities, chemical compounds, lubricants, and ship maintenance, painting, and coating along the river may all contribute to greater Pb deposition in the sediment.

In the current study, maximum As concentration (13.59 ± 4.22 mg/kg) was recorded at S1 whereas minimum (7.22 ± 2.68 mg/kg) was recorded at S4 which exceed the TRV value of 6 mg/kg proposed by USEPA [53]. Chakraborty et al. [55] recorded As concentration ranged from 8.70 to 11.36 mg/kg at Mongla port area, Bangladesh which is somewhat similar to the present findings. Arsenic (As) is an inherent constituent in Bangladesh and even a little quantity may lead to chronic poisoning in animals [56]. The human body might potentially acquire cancer as a consequence of long-term repercussions, including genetic flaws, reproductive difficulties, and skin and vascular illnesses [46,57,58].

The average Copper (Cu) content in sediment sample was 5.44 mg/kg with higher value being 9.30 mg/kg during winter at S1. The minimum value of 2.77 mg/kg was detected at S4 throughout the summer. The observed value of Cu did not surpass the allowable concentration of 16 mg/kg [53]. These results were higher compared to the current findings. In the Meghna River, Bangladesh, Siddique et al. [22] found 6.22 mg/kg Cu which was within the range of the present findings.

The average Cd concentration (2.00 ± 0.85 mg/kg) exceed the USEPA [53] recommended TRV of 0.6 mg/kg indicating untreated pollutants from the local market and shipbreaking region cause this metal to precipitate in the Bakkhali River estuary. However, much higher concentration (3.78 mg/kg) in the estuarine Rupsha River, Bangladesh with Cd was reported by Proshad et al. [48].

The PLI index is often used to evaluate the quality of an environmental component and indicates the temporal and spatial trend. According to Manju et al. [59], PLI evaluates the extent of metal toxicity comparing experimental indices. The PLI of the HMs-M was ranged from 0.43 (S4 in summer) to 0.98 (S1 in winter), with the mean PLI value of 0.66. The present findings revealed the PLI values lower than 1 which demonstrated no pollution of the sediment. Khan et al. [60] and Kabir et al. [61] found higher PLI value in Brahmaputra River (1.11–1.28 with mean value of 1.20) and Shitalakhya River (0.33–1.21 with mean value of 0.75) than in the

Table 3
Comparison of HMs-M in sediment (mg/kg) with other national and international rivers, and reference value.

	As	Cd	Cr	Cu	Pb	Zn	
Bakkhali River, Bangladesh	9.74 (3.75–20.32)	2.00 (0.456–3.425)	48.75 (30.45–71.246)	5.44 (1.46–9.765)	29.78 (13.25–44.619)	56.94 (40.12–75.525)	This study
Pasur River, Bangladesh	10.28	1.59	49.15	6.28	26.58	61.04	[8]
Karnaphuli River, Bangladesh	81.09	2.01	20.30	–	43.69	–	[37]
Meghna River, Bangladesh	–	0.28	10.59	6.22	12.48	42.41	[23]
Rupsha River, Bangladesh	9.31	3.78	25.26	68.81	32.57	–	[48]
Sanmen county's tidal flat	14.3	0.112	84.8	40.7	30.5	116	[49]
Shantou Bay, China	–	1.1	47.5	39.7	50.3	205.9	[50]
Estuaries, Black Sea	7.36	0.20	60.64	45.66	41.37	94.16	[51]
Bahmanshir River, Iran	3.34	0.22	113	86.5	28.8	113	[6]
Terme River, Turkey	0.62	0.14	16.76	13.67	3.37	10.68	[52]
TRV	6	0.6	26	16	31	110	[53]

current study. The PLI documented in this study was also lower than Islam et al. [47] in Halda River, Bangladesh (PLI >1).

Contamination factor (CF) assesses HMs-M contamination in sediments whereas degree of contamination (Cd) estimates the proportion of potentially dangerous metals in sediments through comprehensive assessment [61,62]. The average CF values in the present study were in decreasing order of Cd > Pb > As > Zn > Cr > Cu. Haque et al. [63], observed CF values follows a descending trend of Cd > As > Cr > Pb in the sediment of the Buriganga River. With the exception of Cd and Pb, the CF values for all HMs-M were below 1, suggesting a low level of contamination. However, Cd (2.00) and Pb (1.49) surpassed the critical limit (1), suggesting moderate contamination by Pb and Cd. In the Shitalakhya River, Bangladesh, Kabir et al. [61] recorded the average CF values of Pb, Cu, Cr and Zn were below 1 (low contamination) whereas Cd (2.15) was responsible for moderate contamination of the sediment which is comparable to the present study. Islam et al. [64] conducted a research in the Old Brahmaputra River, Bangladesh, and found that sediments in the area had considerable contamination of Pb, whereas Cu exhibited minimal contamination. In the current study minimum and maximum degree of contamination (*Cd*) was recorded as 3.38 (S5) and 8.02 (S1) during summer and winter, respectively. The average *Cd* (5.41) indicated low contamination of the sediment. The mean *Cd* value observed in this study was alike to that reported by Kabir et al. [61] in the sediment of the Shitalakhya River, Bangladesh. Chakraborty et al. [54] documented the average *Cd* ranges from 9.11 to 14.92 in the sediments of the Pasur River during wet and dry season, respectively, indicating a moderate degree of contamination.

Geo accumulation index (I_{geo}) is a perilous environmental parameter for characteristic natural and anthropogenic sources of metal and evaluating single-metal contamination in sediments. Cd (0.33) had the maximum I_{geo} value and the estimated I_{geo} for the metals in the sediment of the Bakkhali River dropped gradually to the direction of Cd > Pb > As > Zn > Cr > Cu in both summer and winter, respectively. Kubra et al. [45] also recorded highest I_{geo} for Cd (1.89) in the sediment of the Rupsha River, Bangladesh, where the average I_{geo} value follows a downward order of Cd > Pb > Cr > As. In the present study, I_{geo} values of HMs-M (except Cd) were less than zero ($I_{geo} < 0$) demonstrating uncontaminated condition of the sediment by the studied HMs-M. However, I_{geo} value of Cd (0.09) indicated uncontaminated to moderately contaminated ($0 \leq I_{geo} \leq 1$) condition of the sediments. Unpolluted to moderately polluted ($0 \leq I_{geo} \geq 1$) state of the sediment of the Shitalakhya River was also reported by Kabir et al. [61]. However, they also reported the unpolluted ($I_{geo} \leq 0$) condition of the river sediment for other studied metals.

The enrichment factor (EF) evaluates the degree of ecological pollution and extensively used for assessing human activities to enrich the HMs-M in the environment. The average EF value recorded in the present study was decreased as Cd > Pb > As > Zn > Cr > Cu. The EF values for the HMs-M investigated were below 1, showing no enrichment (EF < 1) at any of the sites during the research period. The average EF ranged from 0.83 to 4.58 in the sediment of the Shitalakhya River, Bangladesh was higher compared to the present study [61]. However, maximum value was recorded as 1.06 at S1 in winter, indicating minor enrichment (EF = 1–3). Chakraborty et al. [54] revealed that the EF for HMs-M examined was over 1.5. Specifically, Cd exhibited the greatest EF values throughout both the dry and wet seasons, which corroborates the current results.

Ecological risk factor (E_r^i) and potential ecological risk index (PRRI) are widely used to assess the sediment contamination and the related biological risk [61]. The average E_r^i values of the Bakkhali River decreased in order of Cd > Pb > As > Cr > Cu > Zn. E_r^i of HMs-M (except Cd) in the sediment of the Bakkhali River indicated low ecological risk ($E_r^i < 40$). Haque et al. [63] reported that E_r^i of Pb, Cr, and As in the sediment of Buriganga River were below 40 (categorized as low ecological risk) in all sampling sites in the summer and winter. In the current study, Cd exhibits the higher E_r^i value (ranged from 26.27 to 95.46 with a mean value of 59.96) and indicated moderate to considerable risk at all stations (except S4, S5 in summer and S5 in winter). In Mongla port area, Bangladesh, Chakraborty et al. [54] found higher E_r^i value of Cd (range from 46.51 to 170.00 with a mean value of 94.57). However, PERI of the HMs-M ranged from 38.04 to 120.21 in summer and winter respectively, indicating low (PERI <65) to moderate risk (65 ≤ PERI <130) by the metals. Kubra et al. [45] found PERI score ranged from 160 to 320 in the Rupsha River, Bangladesh which is higher compared to the present findings.

Non-carcinogenic and carcinogenic risk assessment of HMs-M in sediments collected from the Bakkhali River estuary was studied for both adult and child who underwent dermal contact. The results provided essential evidence on ADD_{derm}, HQ_{derm}, CR_{derm} and HI. In both adult and child ADD_{derm} values were decreased as Zn > Cr > Pb > As > Cu > Cd where, Zn exhibit the highest ADD_{derm} value (2.15E-07 in adult and 1.21E-06 in child). In the Pasur River estuary, Jewel et al. [8] reported higher value for ADD_{derm} of Zn compared to other studied metals. In the current study, HQ_{derm} values in both adult and child were listed in ascending order of Cu < Zn < Pb < Cd < As < Cr and compared to other metals child (1.39E-02) exhibit the higher HQ_{derm} values for Cr than adult (2.45E-03). Furthermore, HQ_{derm}, and HI (6.39E-03, 3.62E-02) values of the studied HMs-M were below the threshold value (<1), specifying no potential danger to adult and child through dermal contact of sediment. Kubra et al. [45] and Topaldemir et al. [65] also reported non-carcinogenic risk of the inhabitants by the contamination of HMs-M as HQ and HI values of their studies did not exceeded the threshold value (<1). During the present study, CR_{derm} value for all the HMs-M were much lower than 10^{-6} indicating no cancer risk for the inhabitants due to dermal contact of sediments. However, the findings also revealed that children were more susceptible to CR_{derm} compared to adults. Similar finding was also reported by Kubra et al. [45], where they also showed higher risk of children due to HMs-M contamination of sediment.

5. Conclusion

This research comprehensively examines the contamination of HMs-M and metalloids in the Bakkhali River estuarine system utilizing several analytical methodologies and risk assessment indexes. Concentration of HMs-M in the sediment showed significant seasonal and spatial variations and the HMs-M and metalloids content were increased as Cd < Cu < As < Pb < Cr < Zn. MVSA revealed the possibility of common origin of the contaminants and they might be originated from anthropogenic sources. The PERI value for the metals examined indicating low (PERI <65) to moderate risk (65 ≤ PERI <130) by the studied contaminants. Furthermore, HI value was found higher in child (3.62E-02) compared to adult (6.39E-03) during the study period. In both adult and child Cr displayed the highest CR_{derm} (9.19E-08, 5.20E-07) value whereas Pb represent lowest (9.54E-10, 5.40E-09) value, respectively. During the study period, sediment properties were not evaluated which may have substantial effect on HMs-M distribution in bed sediment. Furthermore, vertical distribution of HMs-M in the sediment was also not evaluated. Therefore, the present study recommends through investigation on sediment properties and their influence on the distribution of metal and metalloids in the Bakkhali River estuary. However, the findings of the present study offer valuable information on the origins and behavior of HMs-M, which will be useful for policymakers and environmental professionals to protect river ecosystems and maintain environmental well-being.

CRediT authorship contribution statement

Sharmin Jahan: Investigation, Data curation. **Md Abu Sayed Jewel:** Supervision. **Bithy Khatun:** Writing – review & editing. **Arun Chandra Barman:** Writing – review & editing. **Sumaiya Akter:** Writing – review & editing. **Md Ayenuddin Haque:** Writing – review & editing, Writing – original draft, Software, Methodology, Formal analysis.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2024.e37496.

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