



Monitoring Trace Metal Contamination in the Sediment of Meghna River Estuary, Bangladesh: A Case Study with Environmental Risk Assessment

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ABSTRACT

Trace metal contamination of water and sediment may have a serious ecological risk to aquatic environmental health. The present study was organized to determine the five globally alarming heavy metals (Cu, Zn, Pb, Cd and Cr) levels in the sediment and assess their potential biological danger. Five different stations were chosen to collect the sediment samples of Meghna River Estuary during two different seasons; monsoon and post monsoon. Results demonstrated the descending order of the observed metals in sediment as follows: $Zn < Cr < Cu < Pb < Cd$. Geo-accumulation index (Igeo), CF (contamination factor), PLI (Pollution Load Index) and PERI (Potential ecological risk index) were calculated to evaluate contamination level which suggested that Meghna River Estuary is not contaminated with those metals and there is no risk of ecological degradation. Furthermore, the multivariate analysis, such as PCA and Pearson correlation matrix analysis disclosed that Zn and Pb may have originated from anthropogenic sources, and other metals may come primarily from a natural source.

INTRODUCTION

The global environment has been experiencing a possessive appearance of heavy metals in the last few decades through the contamination in aquatic biome. Bioaccumulation, biomagnification and longtime persistence nature made heavy metals a prime global concern (Duman *et al.* 2007). Heavy metals hamper the aquatic ecology through making the niche unstable for aquatic lives, dropping species richness and reducing the native species (Wu *et al.*, 2007; Kibria *et al.*, 2012). The mobility of heavy metals in aquatic habitat can be natural or human induced. These two causes are responsible for heavy metal abundance in the environment (Wilson & Pyatt, 2007; Khan *et al.*, 2008). Sediment is the ultimate sink of these metals (Zhang *et al.*, 2017; Bahloul *et al.*, 2018). Hence, this storage of heavy metals is widely regarded as an ecological indicator for assessing the level of metal contamination in the aquatic environment (Ke *et al.*, 2017).

Studies showed that metals are deposited in sediment through absorption, precipitation and hydrolysis, and their gradual accumulation in the sediment has detrimental effects on benthic organisms and many other organisms through the food web and thus endanger the wellbeing of the aquatic ecosystem (Singh *et al.*, 2005; Suresh *et al.*, 2015). A heavy metal can reach the human body through finfish, shellfish or crustaceans, where heavy metals may accumulate in a soluble form (Sharma *et al.*, 2007; Yi *et al.*, 2011; Alhashemi *et al.*, 2012; Rahman *et al.*, 2013; Islam *et al.*, 2015).

The Meghna River Estuary is a key river directly connected to the bay of Bengal in the southern coastal belt of Bangladesh. This estuary supports millions of livelihoods as well as providing service to both industry and agriculture. Besides, local and national economies are highly dependent on this estuary since it supplies the country with a huge amount of fish. However, limited studies have been conducted on some parts of Meghna River (Hassan *et al.*, 2015; Bhuyan *et al.*, 2017); while, the southern part of the estuarine area remains unassessed, except for the study of Sarker *et al.* (2020) who addressed the health risk through heavy metal contamination in fish. Therefore, a methodical study on the sediment distribution, ecological risk assessment, possible sources, and the impact of heavy metals is necessary. Hence, the present study aimed to observe the seasonal distribution of heavy metals in sediment, assess the ecological risk in sediment and find out the probable sources of metal using statistical technique.

MATERIALS AND METHODS

2.1 Sampling Site

The Meghna River Estuary, adjacent to the Noakhali region, is one of the important ecosystems for fisheries. This estuary is heavily used as fish landing area, irrigation purpose, fishing, dumping waste, sewage disposal, water-based transport etc. Numerous human individuals use the Chairman ghat (Site 1) area on daily basis for various purposes, viz. fishing, river crossing, livelihood etc. This area is a junction of various wood made engine boats and fishing trawlers. As a result, this area receives huge amounts of pollutants.

2.2 Sample collection and preservation

The primary goal of this study was to determine Cd, Cr, Cu, Pb and Zn concentrations in surface sediments which act as contamination indicators. The surface layer of sediment was chosen because it controls the exchange of metals between sediments and water. The samples were collected from 5 stations during April to September 2016 (Chairman ghat, Satla1, Satla2 Char kering and Vangon nadi) along the Meghna River, at a depth of 0-12 cm with a Ekman dredge. The collected sediment samples were sealed up with proper labelling using plastic bag. Then, the sediment samples were transferred to the laboratory of the Department of Fisheries and Marine Science, NSTU for further analysis using an ice box.

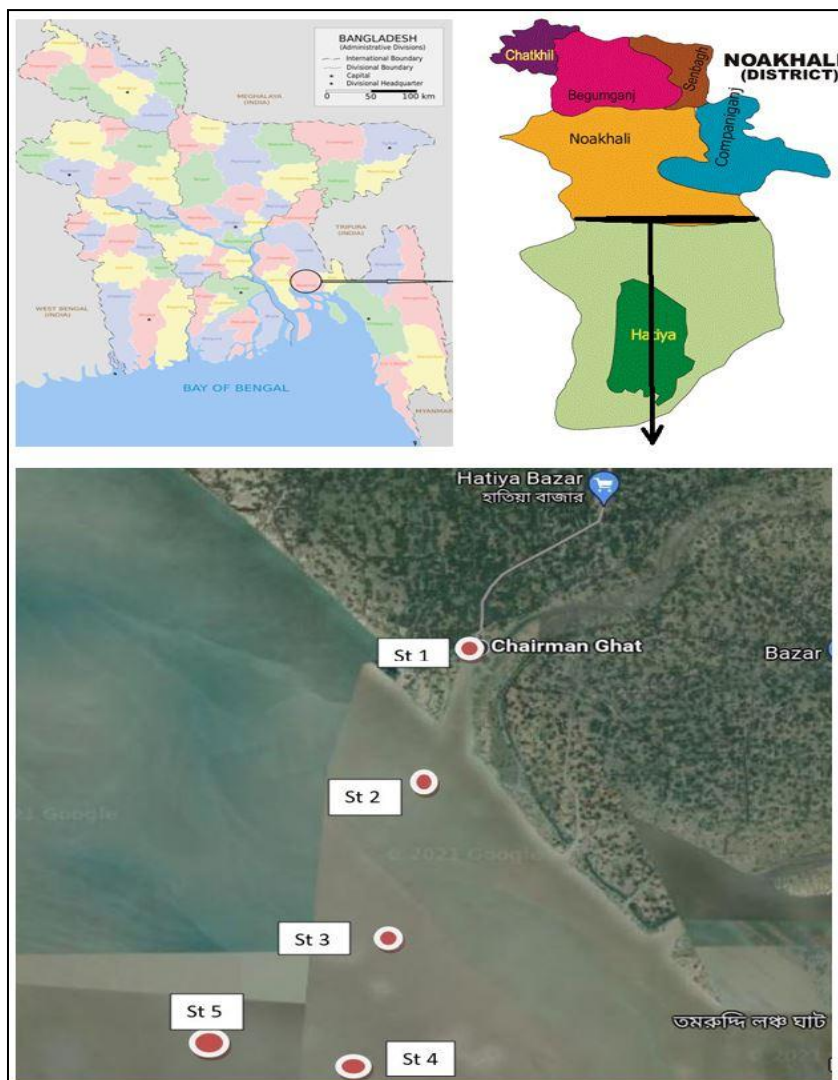


Fig. 1. Five sampling stations of Meghna River Estuary showing the location of the study area

2.2 Sample preparation and analysis

The sediment samples were kept in a dryer at 70°C for 24 hours. Counter drying makes the sample ready to heavy metal analysis. A percaline mortar cleaned with acid water were used to grind the dried sample into powder using a pestle. These powdered samples were then sieved with a strainer (2ml). Digestion procedure was carried out in a 50ml beaker containing 1g of dried sample, followed by the assimilation of sample in 50% HNO₃. A temperature of 190°C for 1.5 h on a hot plate was maintained to perform the procedure. After cooling the sample, a final volume (30 ml) with distilled water was made in a volumetric flask (Chung *et al.*, 2018). Metal contents of Cu, Zn, Pb, Cd and Cr were then determined by atomic absorption spectrometry (AAS), a VARIAN model (AA2407) in the laboratory of Bangladesh Agricultural Research Institute by complying standard procedure (APHA, 1995).

2.3 Sediment pollution load assessment

Several indices were used to assess the heavy metal pollution status in sediments of the Meghna River Estuary. These indices can be categorized as background enrichment indices, pollution indices, toxicity indices and ecological risk indices (Xiao *et al.*, 2013). All the indices and their pollution degree assessment are presented in Table (1). In the current study, geo-accumulation Index (Igeo), contamination Factor (CF), pollution index (PLI) and potential ecological risk index (PERI) were estimated to assess the contamination load.

Table 1. Indices used in the studied area and their pollution degree criteria

Index	Value	Description	Assessment of Pollution degree
Geo-accumulation index (Igeo)	$I_{geo} = \log_2 [C_n / (1.5 \times B_n)]$	C_n = metal concentration in samples. B_n = geochemical background concentration, factor 1.5 = lithospheric changes (Ke <i>et al.</i> , 2017).	< 0 : Practically unpolluted 0–2 = not polluted to moderately polluted 2–3 = Moderate to heavy pollution 3–4 = Heavy pollution 4–5 = Heavy to extreme pollution > 5 : Extreme pollution (Varol, 2011; Chowdhury <i>et al.</i> , 2015; Islam <i>et al.</i> , 2018).
Contamination factor (CF)	$CF = \frac{C_n(\text{Sample})}{B_n(\text{Shale})}$		< 1 : Low contamination 1–3: Moderate contamination 3–6: Considerable contamination $CF > 6$: Very high pollution (Hakanson, 1980)
Pollution Index (PLI)	$PLI = (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{1/n}$		$PLI < 1$: No pollution; $PLI > 1$: Polluted (Tomlinson <i>et al.</i> , 1980; Maanan <i>et al.</i> , 2015).
Potential ecological risk index (PERI)	$RI = \sum_{i=1}^n E_r^i$ $E_r^i = T_r^i \times Cf$	E_r^i = potential ecological risk factor T_r^i = toxic response factor T_r^i = Cu and Pb: 5, Zn: 1, Cr: 2 and Cd: 30 (Suresh <i>et al.</i> , 2011).	$E_r^i < 40$: Low pollution 40–80 : Moderate 80–160 : Considerable $E_r^i > 160$: High pollution RI < 150: Low pollution; 150–300: Considerable pollution; 300–600: High pollution; RI \geq 600: Very high pollution (Hakanson, 1980; Ke <i>et al.</i> , 2017).

2.5 Statistical analysis

SPSS (version 25) was used to perform statistical research data analysis. Seasonal variation in heavy metals concentration was determined by ANOVA. Graphical presentation of heavy metals against seasons and sites (SPSS v.25) and mathematical equation were carried out using Microsoft office excel 10 and SPSS v 25. Besides, for the source identification of metal, the PCA was performed. Pearson's correlation matrix was used to detect the relationships between the metals.

RESULTS AND DISCUSSION

3.1 Heavy metal concentration in sediment

Table (2) depicts the concentration of heavy metals at the five study sites in two seasons along with the background values and sediment quality guidelines (SQGs). In both the seasons, metal ranked in the order of $Zn < Cr < Cu < Pb < Cd$. In particular, highest amount of metal concentration found in site 1 in wet season. Here Zn and Cr were particularly higher in concentration. Also in St 4 these two are in high concentration during the dry season. Both spatially and temporally there were no significant differences of heavy metals found in sediment (significance level $p > 0.05$). Concentration of all metal were higher in dry season than the wet season in sediment sample (Fig. 1). However, due to the increased water dilution, sediment runoff and comparatively pure in wet season lower concentrations of metals could be attributed. Geological features, hydrological effects, lithological inputs and vegetation types may also influence the metal concentration (Jain *et al.*, 2007).

Table 2. Heavy metal concentration in sediment of Meghna River estuary in monsoon and post monsoon (n= 10 for each station).

Stations	Season	Cu	Zn	Pb	Cd	Cr
St-1	Monsoon	24.80	83.12	0.091	0.009	75.72
	Post Monsoon	20.36	73.24	0.024	0.007	54.60
St-2	Monsoon	16.24	61.00	0.013	0.008	48.20
	Post Monsoon	18.20	69.60	0.021	0.008	51.76
St-3	Monsoon	14.44	69.84	0.031	0.003	65.28
	Post Monsoon	20.84	74.40	0.022	0.005	57.92
St-4	Monsoon	19.12	68.76	0.034	0.002	55.16
	Post Monsoon	23.24	80.24	0.048	0.001	63.92
St-5	Monsoon	18.60	66.68	0.029	0.002	56.12
	Post Monsoon	23.00	80.32	0.064	0.003	61.80
Mean	Monsoon	18.64	69.88	0.0396	0.0048	60.096
	Post Monsoon	21.128	75.56	0.0358	0.0048	58
TEC		31.60	121.00	35.80	0.99	43.40
PEC		149	459	128	4.98	111
Background value		45	95	20	0.30	90

Cu concentration varied between 14.44-24.80mg/kg where average concentration was 19.89mg/kg (Table 2). This concentration was much higher than the permissible limit of FAO 1985. **Mohiuddin *et al.* (2015)** recorded 344.5mg/kg from Buriganga River in 2015 which is much greater than our studied area (Table 4). Moreover study from Karnafully, Halda and Turag, **Islam *et al.* (2013)**, **Banu *et al.* (2013)** and **Bhuyan *et al.* (2017)** found less concentration than the present study.

Zn concentration was higher in every station in all the season. It was ranged between 61.00 and 83.12 mg/kg (Table 2). Concentrations were higher than the tolerable limit set by **FAO (1985)** and **WHO (2008)** but under permissible limit set by **USEPA 1999** and **WHO 2004** (Table 3). **Islam *et al.* (2013)** found almost same result from Halda River water. The present study is far below the result of **Mohiuddin *et al.* (2015)** (481.8mg/kg) from Buriganga.

In present study Pb and Cd concentration were below the guideline value (Table 3). Pb concentration were far below the result of **Ahmed *et al.* 2012**, **Banu *et al.* 2013**, **Islam *et al.* 2013**, **Mohiuddin *et al.* 2015** and **Bhuiyan *et al.* 2017**. Their findings also higher for Cd than present study.

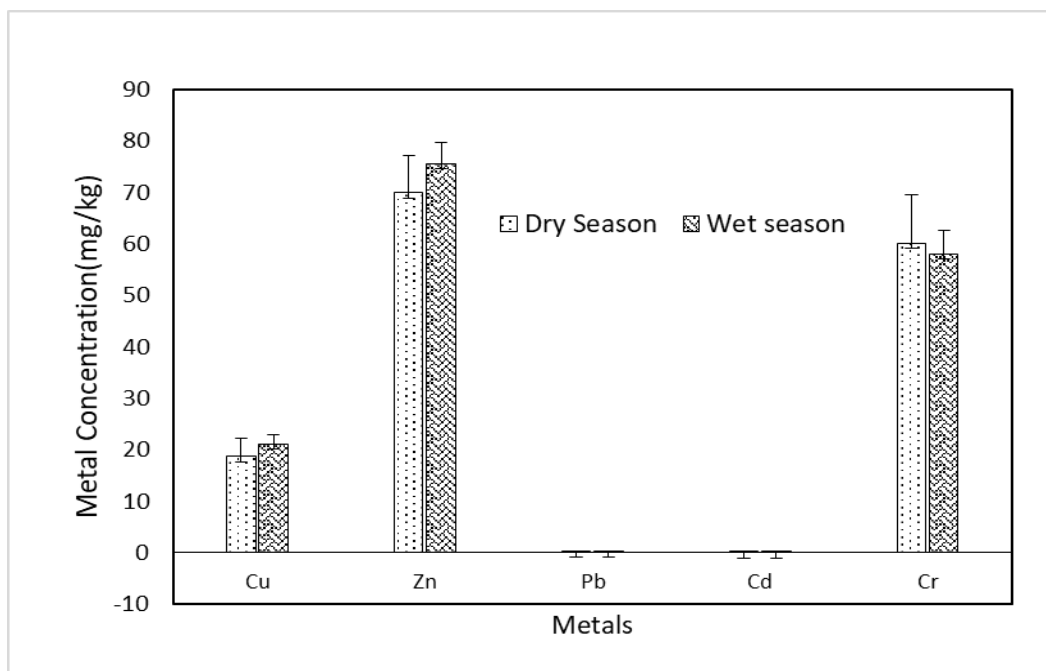


Figure 2. A Graph showing mean metal concentration in two seasons

Highest concentration of Cr recorded from station 1 during the monsoon. Islam *et al.* 2016 found almost same Sitalakhya River (Table 4). Cr concentration varied between 48.20 to 75.72mg/kg. These concentrations exceed the permissible limit set by **FAO 1985**, **USEPA 1999**, **WHO 2004** and **WHO 2008** (Table 3). Besides, metal concentrations in sediment, the average shale values and sediment quality guidelines (SQGs) used in this study are also presented in Table 2. When average values of heavy metals compared with the average shale values, it is found that all the

values were below the shale value in both seasons which indicates that the manmade activities had no direct effect on the concentration of these metals in sediment (Chai *et al.*, 2014). Moreover, Comparison have been also made between heavy metals concentration as well as the consensus-based threshold effect concentration (TEC) and probable effect concentration (PEC) values (Table 2). Here, Cr was between TEC and PEC indicating that the concentration of Cr exhibit adverse effects on the ecosystem.

Table 3. Metal concentration in present study compared with different international standard.

Heavy metals	Present study	WHO 2008	WHO 2004	USEPA 1999	FAO 1985
Cu	19.89	–	–	–	0.2
Zn	72.72	5.0	123	110	2
Pb	0.038	–	–	40	5
Cd	0.005	–	6	0.6	–
Cr	59.05	0.05	25	25	0.1

Table 4. Comparison of heavy metals concentration in the sediment of the Meghna estuary with different rivers

River	Cu	Zn	Pb	Cd	Cr	Reference
Karnofuly	1.22	16.30	4.96	0.24	0.76	Islam <i>et al.</i> 2013
Halda	5.90	79.58	8.80	0.04	8.84	Bhuyan <i>et al.</i> 2017
Dhaleshwari	37.45	-	15.79	2.08	27.39	Ahmed <i>et al.</i> 2012
Shitalakhya	143.7	200.6	-	-	74.82	Islam <i>et al.</i> 2016
Feni River	–	–	6.47	-	35.28	Islam <i>et al.</i> 2018
Buriganga	344.2	481.8	31.4	1.5	173.4	Mohiuddin <i>et al.</i> 2015
Turag	1.576	1.08	1.64	1.4	0.44	Banu <i>et al.</i> 2013
Meghna estuary	19.89	72.72	0.038	0.005	59.05	Present study

3.2 Pollution risk assessment of heavy metal

3.2.1 Geo-accumulation index (Igeo)

The Geo-accumulation index (Igeo) introduced by Muller (1969) values have been used to explain sediment quality (Karbassi *et al.*, 2008) which are presented in Table 5. Based on classification (Table 1), the calculated values of Igeo for all the metal in the sediments were < 0,

indicating the category ‘Practically unpolluted’ at all sites. However, Igeo values followed the order as $Pb < Cr < Cu < Cd < Zn$.

Table 5. Igeo values for metals in the examined sediment samples with mean, maximum, minimum and pollution level

Stations	Igeo values				
	Cu	Zn	Pb	Cd	Cr
St 1	-0.48	-0.26	-2.71	-1.75	-0.32
St 2	-0.59	-0.34	-3.25	-1.75	-0.43
St 3	-0.58	-0.295	-3.05	-2.05	-0.34
St 4	-0.503	-0.281	-2.86	-2.47	-0.35
St 5	-0.511	-0.287	-2.809	-2.25	-0.36
Mean	-0.5328	-0.2926	-2.9358	-2.054	-0.36
Max	-0.48	-0.26	-3.25	-1.75	-0.32
Min	-0.59	-0.34	-2.71	-2.47	-0.43
Pollution level	0	0	0	0	0

3.2.2 Contamination factor (CF) and Pollution load index (PLI)

In all the station CF values of all metals were identified <1 which is a sign of low contamination. In station 1, Zn showed the highest CF values (0.82) (Fig 2). On the other hand Pb showed tiny CF values which indicate that there is no chance of contamination by this metal.

The PLI values for all sediment sample ranged from 1.85-2.08 (Fig 3). PLI values were >1 in all stations indicating that the entire metals Meghna River estuary is polluted. Highest load was recorded from station 1(2.078). According to PLI values sampling stations follows the arrangement as $St\ 1 > St\ 4 > St\ 5 > St\ 3 > St\ 2$.

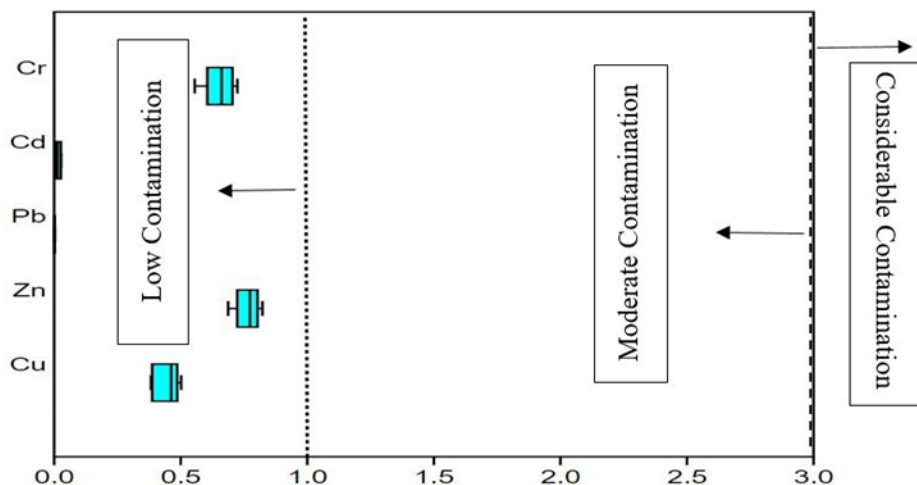


Figure 3. Contamination Factor (CF) for sediment sample where dot line indicates range of contamination level

3.2.3 Potential ecological risk index (PERI)

Based on potential ecological risk index, all five metals (Cu, Zn, Pb, Cr and Cd) were less than 40 indicating that there is no severe ecological risk in the studied area of Meghna River estuary by these metals (Fig. 4). Furthermore, RI values were < 150 in all the stations which point to a decision that the Meghna River estuary has no ecological risk. E_r^i Values arranged the studied metals as: Cu > Cr > Cd > Zn > Pb.

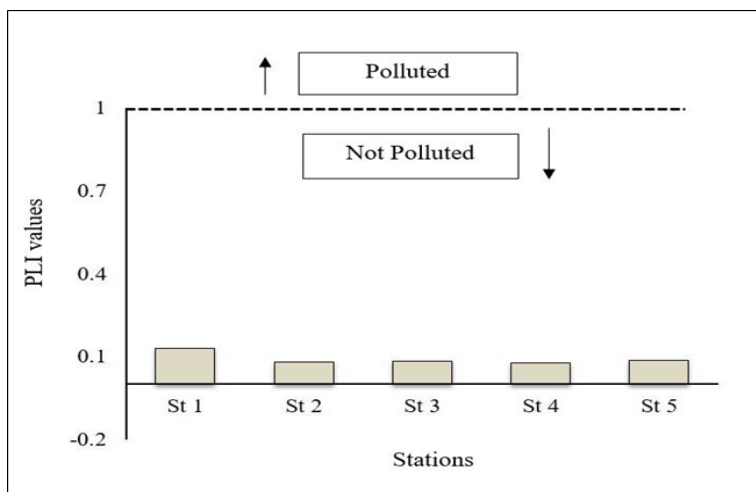


Figure 4. Pollution Load Index (PLI) values for all five stations in Meghna River Estuary

3.3 Identification of metal source

In aquatic atmosphere, study of Metal's interrelation can provide important information about the origin and pathway of metal (Suresh *et al.*, 2011; Wang *et al.*, 2012). Industrial runoffs, civic wastes, fertilizer or other agricultural inputs are possible sources which could be indicated through correlation parameters such as very strong, strong, and moderate correlation. According to K  krer *et al.* (2014), metals are not controlled by a single factor if there is no correlation among the metals. Table 6 shows the Pearson correlation analysis of the present study

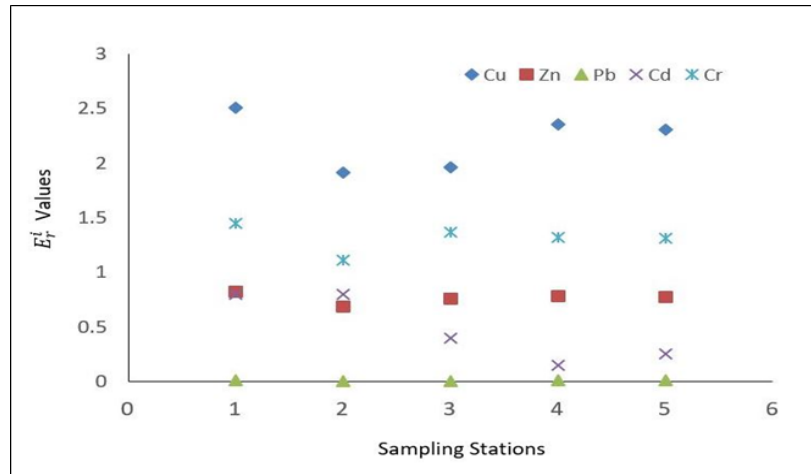


Figure 5. E_r^i Values for all the sampling stations

Not only origin, correlations among heavy metals may also reflect the path of these elements. Strong positive correlation was found between the following pairs of metal: Cu-Zn (.880) and Pb-Zn (.927) at the significance level 0.05. At 0.01 significance level a pair, Cu-Pb exhibit strong positive correlation. . On the other hand, Cu-Cd, Zn-Cd, Cd-Pb and Cd-Cr showed negative weak relationship. This finding indicates that Cu, Zn and Pb might originate from same source.

Table 6. Pearson correlation matrix of the studied metal of Meghna River Estuary

Correlations					
	Cu	Zn	Pb	Cd	Cr
Cu	1				
Zn	.880*	1			
Pb	.970**	.927*	1		
Cd	-.157	-.234	-.128	1	
Cr	.674	.942*	.777	-.188	1
*. Significant at 0.05 level					
**. Significant at 0.01 level					

The objective of PCA is to reduce the data size by extracting a small number of concealed factors by analyzing the relationship among the observed variables (Loska and Wiechula, 2003; Ma *et al.*, 2016).

Table 7 shows the results of PCA for heavy metals of sediment samples. With 87.09% of the total variance two rotated principal components (PCs) were extracted with eigenvalues > 1 . PC1 explained 66.90% of total variance with the eigenvalue 3.345 which was highly loaded with Zn (.96) and Pb (0.95). PC 2 contributed 20.19% of total variance with a high load of Cd (.998) and exhibited an eigenvalue 1.01. Zn and Cu exhibit a positive correlation; thus indicating that, Zn and Cu may exist from a common source (Wang *et al.*, 2015). The source of PC 1 and PC 2 can be considered as mixed source from anthropogenic inputs particularly from water transport discharge and agricultural activities in the study area.

Table 7. Principal Component Analysis of sediment sample of Meghna River Estuary (Component Matrix of two factor model showing different degree of relation)

Variables	Components	
	PC 1	PC 2
Cu	.871	.074
Zn	.956	-.035
Pb	.946	.057
Cd	-.029	.998
Cr	.882	-.064
Eigenvalue	3.345	1.010
% total variance	66.900	20.192
Cumulative %	66.900	87.092

CONCLUSION

Concentration of heavy metals in Meghna estuary does not exceed the permissible USEPA standard for any of the sites. This indicate regarding the readily toxicity pollution by heavy metal, Meghna river estuary sediments are not in severe state. PCA recommended that the metals source in sediments was more or less anthropogenic origin rather than lithogeny. However, effluents and fuel from engine boats may be the possible source of metal such as Pb. Domestic and municipal wastes through the adjacent canal might play role for Zn deposition in water as well as sediment. These indicate that Pb and Zn may come from similar anthropogenic sources. Other metals may be results of lithogenic activities. The results evidently indicate the sediment quality of the Meghna River estuary to be unpolluted to low polluted. Constant monitoring and

assessment will be beneficial for checking pollution status of the river water and sediment. Furthermore, this study will help in understanding the amount of toxic compounds (heavy metals) being received in the river and its biological magnification in animals, particularly those at the lower level of food chain. As well as it will also help to make aware those local people or adjacent farmers for proper management of waste disposal and also to minimize use of synthetic inputs.

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