Marine Pollution Bulletin 140 (2019) 255–261

Contents lists available at ScienceDirect

Marine Pollution Bulletin

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

Baseline

Baseline study of heavy metal contamination in the Sangu River estuary, Chattogram, Bangladesh

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ARTICLE INFO

Keywords:
Heavy metals
Contamination
Sangu Estuary
Reference site
Bangladesh

ABSTRACT

Sediment samples were collected from twelve selected sites of the Sangu River estuary and seven metals (As, Cr, Cu, Cd, Pb, Ni, Zn) were analyzed with Inductively Coupled Plasma-mass Spectrometry (ICP-MS) to assess the contamination degree of heavy metals in the Sangu River estuary and to represent it as a reference site. This study revealed the descending order of studied metals (mg/kg) observed in sediment as Zn (88.97 ± 58.97) > Ni (32.75 ± 16.09) > Cu (29.2 ± 10.78) > Cr (25.14 ± 5.20) > Pb (19.57 ± 7.01) > As (2.58 ± 2.55). Cadmium was observed below the detection level. Various indices like geo-accumulation index (I_{geo}), contamination factor (CF), pollution load index (PLI) suggested that the Sangu River estuary is not contaminated by studied metals excepting Pb. PCA and correlation matrix analysis indicates that Pb and Zn may have anthropogenic sources and As, Ni, Zn, Cu, Cr may come from natural sources.

Estuaries are known to have an intricate, highly fertile and dynamic nature. Various anthropogenic pollutants are dispensed into these transition zones from adjacent industrial and urban areas (Monteiro et al., 2016). Among all types of pollutants some properties like non-biodegradability, toxicity, ecological risk, bio-accumulation, persistence, ubiquity, biogeochemical recycling and abundance make the heavy metals an interesting research topic (Malvandi, 2017; Liu et al., 2016; Varol, 2011).

Metals can make the water and bottom sediment unsuitable for aquatic lives, play role in reducing species abundance and depleting the native species, adversely affect the estuarine habitats (Kibria et al., 2016; Wu et al., 2007; Kibria et al., 2012), accumulate in organism's body through food chain and gradually increase health risk (Malvandi, 2017). Heavy metals can occur in the estuarine environment through both natural and anthropogenic activities. Natural processes include physical and chemical weathering of rocks, erosion and atmospheric deposition, and conversely, sewage discharge, industrial wastewater, agricultural and coastal land runoff, change in lacustrine environments are regarded as anthropogenic activities (Mondal et al., 2018). A large portion of free metal ions are more easily deposited into sediments than water (Bahloul et al., 2018). So sediments function as major storage of heavy metals as well as are considered as an effective tool to evaluate the contamination level of an aquatic environment (Santos et al., 2004; Ke et al., 2017).

A few studies have been conducted to assess the heavy metal contamination in estuaries of Bangladesh like in the Feni River estuary (Islam et al., 2018), the Karnafuli River (Ali et al., 2016), the Bakkhali River estuary (Siddique et al., 2012). However, there is no baseline information about the heavy metal contamination in non-polluted or less polluted estuaries in Bangladesh.

There is an assumption that the Sangu River estuary, located in the southeast region of Bangladesh, can be used as a reference site to determine the contamination level as it is relatively far away from the industrial areas. A study was found on finfish and shellfish fauna and fishery of the Sangu River estuary (Uddin, 2014). However, there was no single study to observe the pollution status in the Sangu River estuary. Here, this research attempts to represent the metal concentration in the surface sediments and to appraise the contamination level in the study area. The major objectives of this research work are 1) to assess the degree of contamination level, 2) to use this ecosystem as a reference site, and 3) to determine the possible sources of pollutants.

The Sangu River originates from the North Arakan Hills of Myanmar and enters Bangladesh through Bandarban district. It drains the water from Patiya, Satkania, Banskhali district and finally falls into the Bay of Bengal near Chattogram about 16.09 km at 22°6′N 91°51′E. Nationally it is an important estuary containing thirty three species of finfish and

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https://doi.org/10.1016/j.marpolbul.2019.01.058
Received 11 August 2018; Received in revised form 15 January 2019; Accepted 28 January 2019

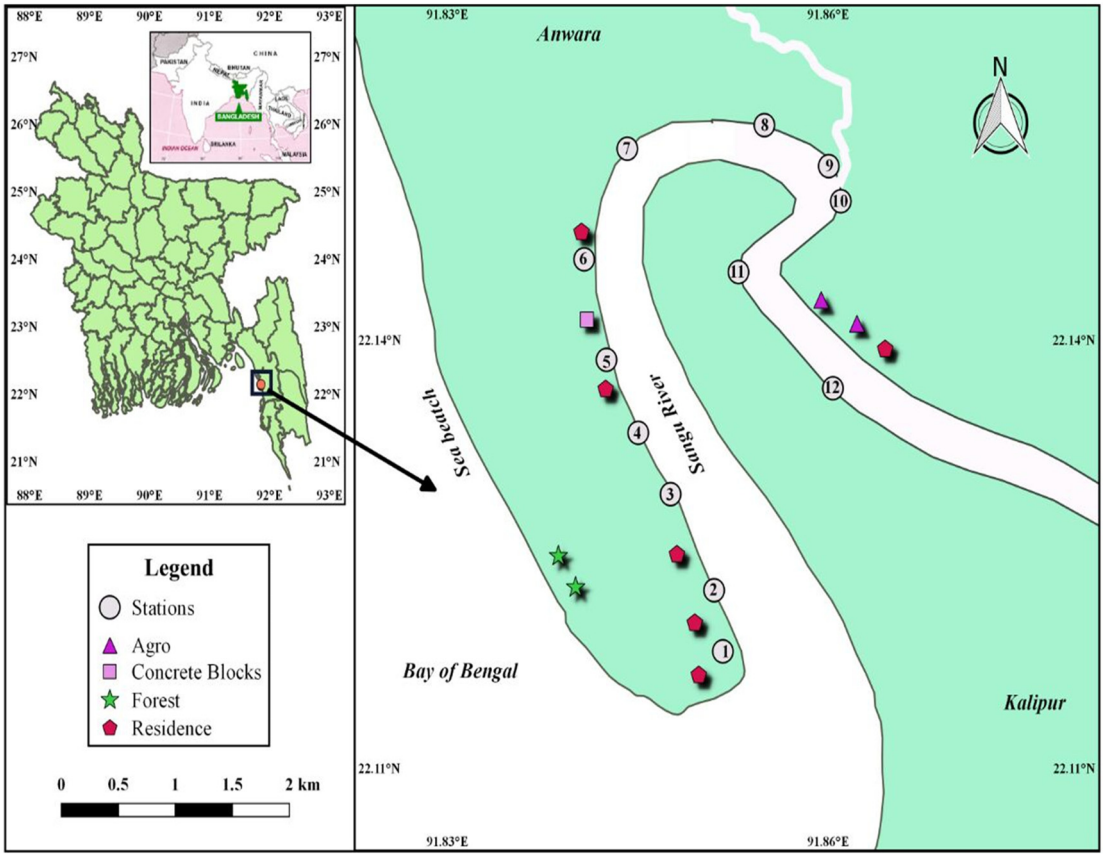

Fig. 1. The location of the study area and sampling sites in the Sangu River estuary.

five species of shellfish (Uddin, 2014). Besides, it provides the livelihood of people living in the adjacent areas. The nature of sediment is sandy at the mouth of the estuary and muddy landwards.

Sediment samples were taken at a depth of 0–10 cm of the riverbed from 12 selected sites of the Sangu River estuary on 21st May 2017 (Fig. 1). 12 sediment samples have been collected from the study area. After collection, samples were sealed up using polythene bags with proper labeling. Then the sediment samples were kept in an ice box at 4° centigrade for further analytical process.

Sediment samples were air dried and crushed well then sieved with 2 ml strainer. 1.8 g of each sediments was processed in 10 ml of ultrapure HNO_3 utilizing Micro Wave Digestion System (WX-6000, origin from China), filtered and diluted for 2 h. Then the solution is kept into a tezaron tube (50 ml). Level of metal concentration of As, Cr, Ni, Pb, Cu, Cd and Zn was resolved in triplicate in air/acetylene fire utilizing Inductively Coupled Plasma-mass Spectrometry (ICP-MS) (ELAN9000, Perkin-Elmer, Germany). Sediment reference material CRM320 was used ($N = 3$) to confirm the approval of information and the exactness and accuracy of diagnostic strategy. All arrangements were done utilizing ultrapure water and all plastic, quartz and crystal were absorbed HNO_3 (10%) for no < 24 h and washed over and again with ultrapure water.

Mathematical calculations were performed by using Microsoft Excel version 10 while Qgis @2.17 was used for map plotting. Pearson coefficients were applied to assess the relationships among the considered variables ($P < 0.05$). Multivariate and univariate statistical analysis like correlation matrix (CM) and principal component analysis (PCA) were carried out by using both origin9 @2015 and Graphpad Prisom7@2015.

Recently, various indices have been developed to assess the heavy metal status in sediments which can be categorized into four types: background enrichment indices, pollution indices, toxicity indices and ecological risk indices (Caeiro et al., 2005; Xiao et al., 2013). In this study, quite a few metal assessment indices were used to evaluate the contamination level of heavy metals in sediments of the Sangu River estuary (Table 1).

Table 2 represents the concentrations of heavy metals at twelve different sites of the Sangu River estuary sediments. In sediments, the studied metals ranged from 12–33.42 for Pb, 9.52–31.4 for Cr, 2.23–39.8 for Cu, 15.6–79.7 for Ni, 22.8–80.83 for Zn and 2.1–2.9 mg/kg for As. Cadmium was observed below the detection level. The concentration level of studied metals followed the order as: Zn > Ni > Cu > Cr > Pb > As.

Zn showed the highest concentration at S6. Maximum level of Ni and Pb was observed at S9 and S10 respectively. Sediments collected from site-5 showed highest concentration of Cr and Cu and the maximum concentration of As was found at S7 (Fig. 2).

This is the first attempt to scrutinize the level of heavy metals in sediments of the Sangu River estuary. The present data were evaluated by comparing with other works (Table 3) as there was no available information on metal concentrations in that estuary. The Pearl River estuary has considerably higher concentration profile of metals than the Sangu River estuary (Zhou et al., 2004). Metal concentration in sediment samples of the Feni River estuary was comparatively greater than the present study, while As (0.85) and Pb (6.47) are lower in the Feni River estuary than the Sangu River estuary (Islam et al., 2018). Values of metals in sediments of the Bakkhali River estuary were higher than present study area (Siddique et al., 2012). Similarly, metal concentration level in the Karnafuli River was higher than present study (Ali et al., 2016). Concentration levels in sediment samples from the Sangu River estuary were lower than Ganges River estuary. However, concentration of Ni (8–57 mg/kg) (Subramanian et al., 1988) was lower in the Ganges River estuary than present study. The Tapti River estuary (Shah et al., 2013) has considerably higher level of metals than Sangu

Table 1
Pollution indices used in the study.

Index	Depiction and objectives	Principle	Explanation	Pollution degree criteria
Geoaccumulation index (I_{geo})	The geoaccumulation index (I_{geo}) provides a means to evaluate the contamination status of the environment by comparing with geochemical background concentrations (Muller, 1969)	$I_{geo} = Log_2\left[\frac{Cn}{1.5Bn}\right]$	C_n is the concentration of the metals observed in sediment samples. B_n is the geochemical background concentration of the metal (n), the factor 1.5 means the "background matrix correlation value" responsible for lithospheric differences (Ke et al., 2017).	I_{geo} index includes seven classes which represent the degree of contamination according to the index values: < 0 – Practically unpolluted; 0–1 – Unpolluted to moderately polluted; 1–2 – Moderately polluted; 2–3 – Moderately to heavily polluted; 3–4 – Heavily polluted; 4–5 – Heavily to extremely polluted; > 5 – Extremely polluted. (Chowdhury et al., 2015; Varol, 2011; Islam et al., 2018).
Contamination factor (CF) and Pollution Index (PLI)	Contamination factor (CF) is used to estimate the heavy metal contamination in the study area. Pollution Load Index (PLI) can be used for assessment of comparison of contamination status among the study sites.	$CF = Cn_{(Sample)}/Bn_{(Shale)}$ $PLI = (CF1 \times CF2 \times CF3 \times \ldots \times CFn)^n$	The CF value can be acquired by dividing the concentration of each metal in sediments by background level (Hakanson, 1980).	CF < 1: Low contamination 1 < CF > 3: Moderate contamination 3 < CF > 6: Considerable contamination CF > 6: Very high pollution (Hakanson, 1980) And PLI < 1: No pollution; PLI > 1: Polluted (Maanan et al., 2015; Tomlinson et al., 1980).
The Potential ecological risk index (PERI)	The Potential ecological risk index (PERI) was originally proposed by Hakanson (1980) and it is applied to appraise the potential ecological risk of studied metals in sediment. This index provides a better	$RI = \sum_{i=1}^{n} E_r^i$ $E_r^i = T_r^i \times CF$	E_r^i is the potential ecological risk factor and T_r^i is the toxic response factor of studied metals. T_r^i was determined for Cu = Pb = Ni = 5, Zn = 1, As = 10, Cr = 2 and Cd = 30 (Suresh et al., 2011).	RI < 150: Low pollution; RI: 150–300: Considerable pollution; RI: 300–600: High pollution; RI ≥600: Very high pollution (Hakanson, 1980; Ke et al., 2017).

(*continued on next page*)

Table 1 (*continued*)

Index	Depiction and objectives	Principle	Explanation	Pollution degree criteria
	evaluation of the potential ecological risk factor of heavy metal contamination by combining ecological and environment effects with toxicology (Ke et al., 2017).			

Table 2
Measured concentrations (mg/kg) of studied metals from all sampling sites of the Sangu River estuary.

Stations	Metal Concentration						
	Pb	Cd	Cr	Cu	Ni	Zn	As
Station 1	19.6	BDL	25.36	36.4	20.4	67.08	2.6
Station 2	12.41	BDL	27	2.23	23.32	22.8	2.5
Station 3	20.53	BDL	25.36	34	30	80.8	2.8
Station 4	21.01	BDL	27	36.05	32.1	75.41	2.9
Station 5	23.4	BDL	31.4	39.8	29.3	93.5	2.3
Station 6	29.6	BDL	27.15	30.5	27.9	261.8	2.5
Station 7	12.9	BDL	26.2	34.31	28.6	112.5	2.9
Station 8	12.41	BDL	9.52	15.4	15.6	42.22	2.8
Station 9	22.44	BDL	25.24	22.26	79.7	73.47	2.7
Station 10	33.42	BDL	24.86	33.12	39.5	80.83	2.6
Station 11	12	BDL	26.2	33.81	32	77	2.1
Station 12	15.2	BDL	26.5	32.94	34.6	80.28	2.3
Mean ± Std	19.58 ± 7.017	–	25.15 ± 5.21	29.24 ± 10.78	32.75 ± 16.09	88.97 ± 58.97	2.58 ± 0.25

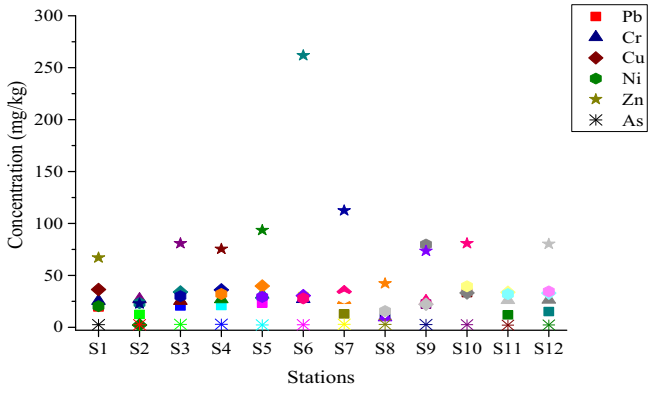

Fig. 2. Metal concentration in all sampling sites.

River estuary.

The I_{geo} values for the study area are presented in Table 4. The I_{geo} values of As, Cr, Cu, Ni in sediment of the Sangu River estuary were < 0, representing the category 'Practically unpolluted' at all sites. Consequently, all selected sites are not polluted by these metals. In contrast, site S10 and site S6 are considered as unpolluted to moderately polluted as these sites showed the highest I_{geo} value of Pb (0.16) and Zn (0.877) respectively (Fig. 3). I_{geo} values followed the order as: Pb > Zn > Cu > Ni > Cr > As. However, the average I_{geo} values for all studied metals were < 0 (Table 1), suggesting that the study area is unpolluted by all studied metals.

CF values of As, Cr and Cu for all selected sites were detected < 1 which is a sign of low contamination. Site S9 was marked as moderately contaminated by Ni (1.22) and Pb (1.17). Site S10 was also experienced with moderate contamination due to high CF value of Pd (1.67). Similarly, Zn showed high CF values at S6 (2.75) and site S7 (1.18), both are > 1 and < 3.Consequently, these sites were tagged as moderately contaminated by Zn (Fig. 4).

Table 4
I_{geo} values for metals in the sediment samples.

Stations	Igeo values					
	As	Pb	Cr	Cu	Ni	Zn
Station 1	−2.91	−0.61	−2.41	−0.89	−2.3219	−1.08701
Station 2	−2.96	−1.27	−2.32	−4.92	−2.1289	−2.64386
Station 3	−2.80	−0.55	−2.41	−0.99	−1.7655	−0.81853
Station 4	−2.75	−0.51	−2.32	−0.90	−1.6679	−0.91813
Station 5	−3.08	−0.36	−2.10	−0.76	−1.7996	−0.60792
Station 6	−2.96	−0.02	−2.31	−1.15	−1.8702	0.877503
Station 7	−2.75	−1.22	−2.37	−0.98	−1.8344	−0.34104
Station 8	−2.80	−1.27	−3.83	−2.13	−2.7089	−1.75496
Station 9	−2.85	−0.42	−2.42	−1.60	−0.3559	−0.95573
Station 10	−2.91	0.16	−2.44	−1.03	−1.3686	−0.818
Station 11	−3.2150	−1.3219	−2.3653	−0.9974	−1.6724	−0.88803
Station 12	−3.0837	−0.9808	−2.3489	−1.0350	−1.5597	−0.82785
Average	−2.92	−0.70	−2.47	−1.45	−1.75	−0.89863
Max	−2.75	0.16	−2.10	−0.76	−0.36	0.88
Min	−3.22	−1.32	−3.83	−4.92	−2.71	−2.64

The PLI values for all sampling sites were ranged from 0.22 to 0.63 and mean PLI value was 0.47. In all sites, PLI values were < 1 (Fig. 5) which indicates that the Sangu River estuary is not polluted. As per PLI values, the sampling sites can be arranged as S6 > S10 > S9 > S5 > S4 > S3 > S7 > S12 > S1 > S11 > S8 > S2.

According to the potential ecological risk index the Sangu River estuary is in low ecological risk by As, Cr, Cu, Ni, Zn as the E_r^i values were < 40 and in moderate ecological risk by Pb for having E_r^i value > 40 (Fig. 6). Moreover, all study sites had RI values < 150 which leads to a decision that the Sangu River estuary has no ecological risk. As per the E_r^i values, studied metals were arranged as: Pb > Cu > Ni > As > Zn > Cr. However, in the sediment samples of the Feni River estuary, E_r^i values of Ni pointed to pollution (Islam et al., 2018).

To determine the relationship among the studied heavy metals a

Table 3
Metal concentrations (mg/kg) in sediment samples from Sangu River estuary and other selected estuaries around the world.

River estuary, location	AS	Cr	Cu	Ni	Pb	Zn	References
Luanhe River estuary, China	3.4–13.6	11.6–76.2	9.6–35.6	3.5–35.8	22.6–43.7	12.9–94.7	(Liu et al., 2016)
Pearl River estuary, China	–	89	46.2	41.7	59.3	150.1	(Zhou et al., 2004)
Feni River estuary, Bangladesh	0.85	35.28	–	33.27	6.47	–	(Islam et al., 2018)
Karnafuli estuary, Bangladesh	37.23–160.32	11.56–35.48	–	–	21.98–73.42	–	(Ali et al., 2016)
Bakkhali River estuary, Bangladesh	–	–	34.93	–	27.14	100.85	(Siddique et al., 2012)
Ganges estuary, India	–	21–100	4–53	8–57	12–115	12–611	(Subramanian et al., 1988)
Tapti river estuary, India	N/A	55.20	148.32	80.92	56.69	143.55	(Shah et al., 2013)
Sangu River estuary, Bangladesh	2.58	25.149	29.235	32.751	19.576	261.8	Present study

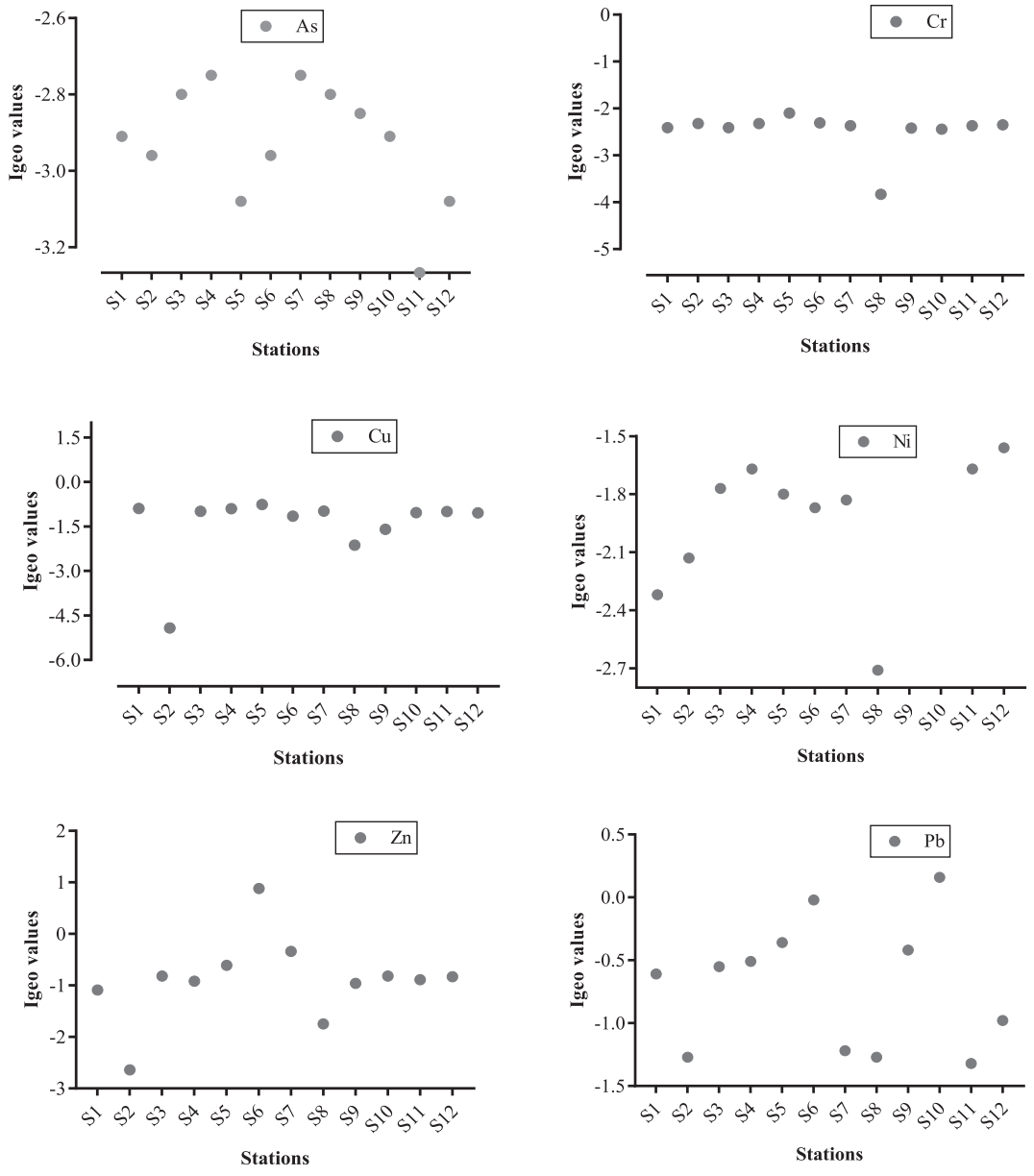

Fig. 3. I_{geo} values for all studied metals at all sampling stations.

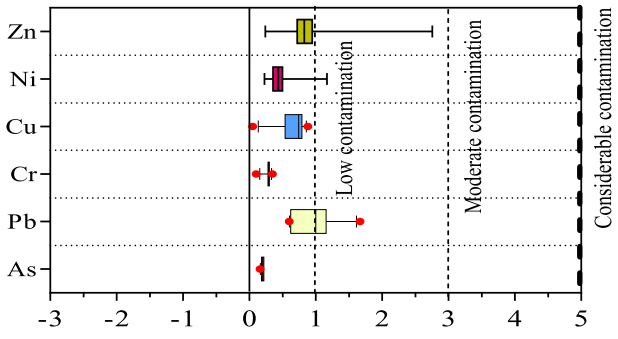

Fig. 4. CFs for sediment samples in the study area.

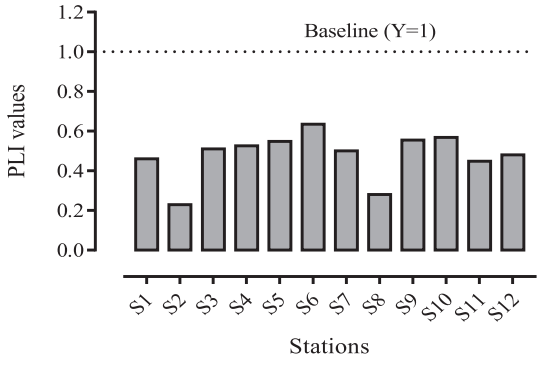

Fig. 5. PLIs for all sediment samples in the study area.

correlation matrix is used. By correlation matrix, it can be possible to find a common source of metal in the study area (Malvandi, 2017). The results of the Pearson correlation analysis are shown in Table 5. There were no highly significant correlations among metals in the Sangu River estuary. Only Pb and Zn showed a significant relation which suggested that Pb and Zn might have common anthropogenic sources.

Table 6 summarizes the results of PCA with eigenvalue, variance of 100% and cumulative variance for each factor. Three principal components (PCs) with eigenvalues > 1 were taken out as a consideration.

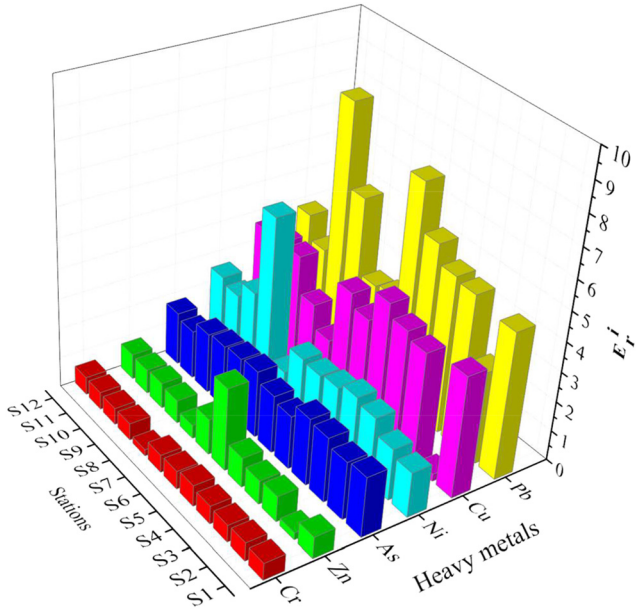

Fig. 6. Values of E_r^i at all sampling sites.

Table 5
Pearson correlation matrix of studied metals of the Sangu River estuary.

	Pb	Cr	Cu	Ni	Zn	As
Pb	1					
Cr	0.301	1				
Cu	0.372	0.426	1			
Ni	0.304	0.233	0.013	1		
Zn	0.515*	0.282	0.360	−0.001	1	
As	0.058	−0.369	−0.097	0.034	−0.078	1

* Significant at 5% level.

Table 6
Principal component analysis of collected sediments of the Sangu River estuary.

Variable	PC1	PC2	PC3
Pb	0.498*	0.410	−0.019
Cr	0.485	−0.344	0.261
Cu	0.470	−0.072	−0.278
Ni	0.213	0.334	0.808**
Zn	0.467	0.136	−0.410
As	−0.182	0.760**	−0.181
Eigenvalue	2.235	1.209	1.057
Variance (%)	37.25%	20.14%	17.61%
Cumulative of variance (%)	37.25%	57.39%	75.00%

** Highly significant at 5% level.

PC1 contributed 37.25% variance with the highest eigenvalue 2.235 having a high loading on Pd (R = 0.498). PC2 elucidated 20.14% of the total variance with a high loading of As (R = 0.76) and revealed an eigenvalue of 1.209. PC3 had a high loading on Ni (R = 0.808) and accounted for 17.61% of total variance with an eigenvalue of 1.057 (Fig. 7).

In the present study, the metal contamination status in the Sangu River estuary, Chattogram, Bangladesh has been revealed for the first time. This study explained that this estuary is not highly concentrated by As, Pb, Cr, Cu, Ni and Zn. Results from various indices (I_{geo}, CF and PLI) disclosed that the Sangu River estuary is not polluted by studied metals. According to I_{geo} and CF values two study sites were unpolluted to moderately polluted by Zn, however the entire study area is not polluted by Zn. The pollution ecological risk index indicates that the estuary is moderately contaminated by Pb (E_r^i > 40). The possible

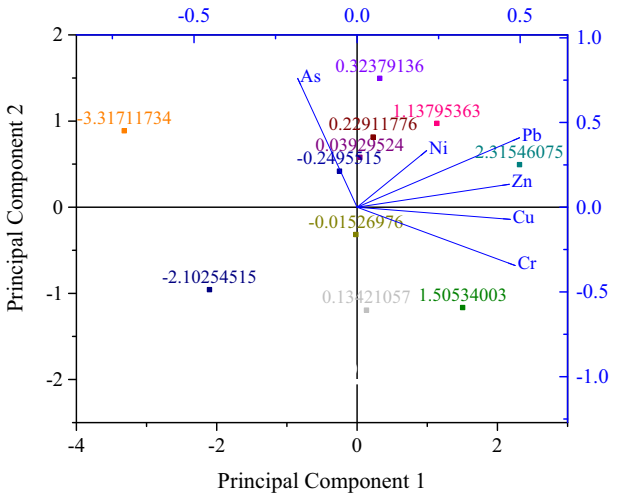

Fig. 7. PCA of studied metals of the Sangu River estuary.

source of Pb may be the gasoline and diesel fuel from engine boats. Zn might come from municipal and domestic wastes passed through the Parki canal. Therefore, Pb and Zn may come from similar anthropogenic sources. The other metals may originate from natural sources. This study also revealed that the Sangu River estuary cannot be considered as a reference site in any case for Pb. In general, this is a baseline study which first presents the current contamination status of the Sangu River estuary for future researchers.

Acknowledgements

The author would like to thank Bangladesh Council of Scientific and Industrial Research (BCSIR) for providing necessary fund for metal analysis and instrument facilities. This work was done under the partial on-going R & D projects.

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