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Assessment of Toxic Heavy Metals in Surface Water of the Meghna River Estuary: An Integrated Statistical Approach

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ABSTRACT

Rapid industrial development and negligible waste disposal management are considered the driving factors for the major environmental crisis in developing countries like Bangladesh. In the present study, the surface water of Meghna River estuary was evaluated for the concentrations of five toxic heavy metals, namely chromium (Cr), lead (Pb), cadmium (Cd), arsenic (As), and mercury (Hg). All the heavy metals in question were found to be higher than the background value of natural surface water. The toxic heavy metals were in the descending order of Pb>Cr>Hg>Cd>As in the collected water samples. Pb and Hg were found to exceed the lowest biological chronic safety concentration limits recommended by the National Oceanographic and Atmospheric Administration (NOAA), whereas Cr, Cd, and As were under the admissible limits. Principal Component Analysis (PCA), Hierarchical Cluster Analysis (HCA), and Pearson's correlation analysis suggested that the sources and distribution of those toxic heavy metals are similar. Anthropogenic activities such as industrial waste, agricultural runoff, and municipal solid waste disposal may be responsible for the increased concentration of those metals. Compared to China's secondary seawater quality standard, the concentration of heavy metals in the Meghna estuary is considerably higher, except arsenic. When compared with previous studies, the heavy metals concentration in the Meghna estuary is significantly higher, which is alarming. From this integrated study, the Meghna estuary is under threatening conditions from ecological and human health perspectives. Further studies are required for temporal monitoring and assessment of the Meghna River estuary.

Introduction

The continuous development of industry, agriculture, and urban areas contribute to a considerable amount of chemical hazards in the aquatic environment (Ali et al., 2016; Islam et al., 2015). Both natural and anthropogenic activities are responsible for this situation. However, anthropogenic activities outweigh the natural causes in terms of heavy metal release in the runoff. As the estuary and continental shelf are active sinks for the suspended materials, higher concentrations of chemical components and heavy metals accumulate in the sediments and water. The rate of accumulation of heavy metals in the estuarine area has elevated to such an extent that the local ecosystem is in threatening conditions (Chapman et al., 2013; Liu et al., 2020; Pandey et al., 2019). As a part of the global ecological cycle, the humans cannot avoid the toxic effects of heavy metals because of dependence on aquatic foods for the source of complete protein, vitamins, nutrients, and necessary amino-acids (Ahmed et al., 2019; Fakhri et al., 2018; Fuentes et al., 2009; Longo et al., 2013; Miri et al., 2017; Özden et al., 2010).

Apart from that, seafood consumption is getting popular globally, but chemical pollution in the marine environment makes it hazardous to human health. Chemical contamination has been putting the biodiversity in a very vulnerable condition, and in turn, the unsafe biodiversity is posing different types of risk to humans. The highly chemically polluted countries are suffering much where proper filtration and management of industrial waste are not in place; most

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of the third world countries like Bangladesh are facing that condition (Ahmed et al., 2019; Ahmed et al., 2016; Ali et al., 2016; Gani et al., 2017; Kibria et al., 2016b; Liang et al., 2018).

Bangladesh is a land of rivers. Its locality, urbanization, transportation, agriculture, and industrialization solely depend upon the drainage system. As a result, a large amount of discharging hazardous chemical wastes into the water system,which makes the rivers and estuaries vulnerable. Many industries, particularly oil refineries, textiles and dying factories, tanneries, fertilizer industries, battery recycling factories, along with runoff from agricultural land and domestic solid waste, are releasing a huge amount of untreated effluent into the river, which is ultimately transported to the Bay of Bengal through the Meghna estuary.

Besides, the rivers of Bangladesh are a route that carries the world's most sediment load with their discharge. The sediment loads are the naturally occurring heavy mineral sources in the study area. Both point and non-point sources are responsible for the contamination load of the estuary (Barceló, 2004). Thus, estuarine water is highly vulnerable to contamination from numerous sources of pollutants.

From the previous studies, pollution has appeared to have reached a level that needs immediate attention from the regulatory bodies. Considering this, pollutants and hazardous chemical components have already raised alarming symptoms, which are indicative of supervision and consideration of the estuarine water body; otherwise, pollution could be uncontrollable shortly. The research work previously carried out has mostly been confined to the assessment of heavy metal pollution in the adjacent rivers of the industrially significant cities in Bangladesh, such as Dhaka (Ahmad et al., 2010; Mohiuddin et al., 2011; Rahman et al., 2014; Islam et al., 2015) Chittagong (Ali et al., 2016; Hossain and Islam, 2006; Kibria et al., 2016b), Khulna (Proshad et al., 2019; Islam et al., 2015). The number of studies on heavy metal pollution in the estuary area is scant. Several studies performed have taken a deterministic approach to identifying heavy metal sources, taking into account the uptake in flora and fauna, and the spatial and temporal distribution of heavy metals (Hossain and Islam, 2006; Hossain and Rahman, 2011; Islam et al., 2015; Islam and Habibullah, 2017; Khan et al., 1998; Kibria et al., 2016a, 2016b; Proshad et al., 2019).

The current study aimed to investigate the spatial distribution of some toxic heavy metals in surface water collected from the Meghna River estuary. Moreover, environmental risk will be assessed by making a comparison with several estuarine conditions as well as by identifying the possible sources and controlling factors of those toxic heavy metals.

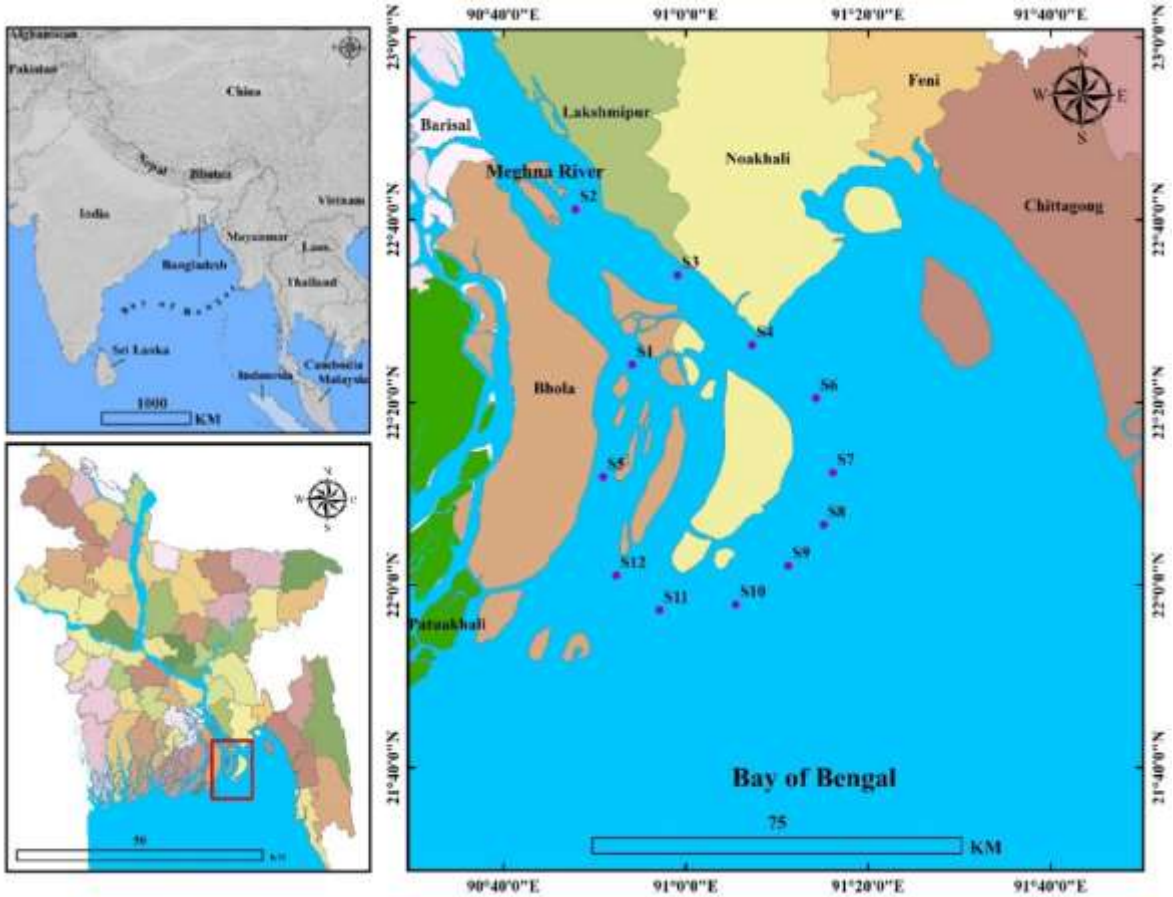

Figure 1: Map showing the study area with sample locations

Study Area

The Ganges, the prime source of the river system of the study area as well as for Bangladesh, enters the country through the north-western side, namely the Padma River which is afterward joined by the Brahmaputra River and flows southward as the Padma River. Finally, the Padma-Brahmaputra River joins the Meghna River near Chandpur and is discharged into the Bay of Bengal as Meghna River which forms a huge Meghna River estuary system. Almost all the smaller rivers in the country contribute to that joint discharge of the Ganges-Brahmaputra-Meghna River system. This study area is bounded by Barisal and Bhola districts in the west and Noakhali, Lakshmipur, and Chittagong in the east. This long and extensive river system carries a huge amount of sediment from upstream (Himalayan region). Moreover, accretional and erosional processes in the study area create several smaller islands(Biswas, 1978; Curray and Moore, 1971; West, 1973) namely Hatiya, Sandwip, Manpura, Nijhum Dwip, Domar Char, Sona Char, Rabipur etc. are the major ones (Figure 1). Consequently, the bathymetry, tides, and outflow from the Meghna River contribute to the driving forces of the estuary which makes the estuary indispensable both for economic and ecological diversity (Khan et al., 1998).

From the economic point of view, the study area is important for its location as it is regarded as the major commercial hub for shipping and water-based transportation. Thus, all kinds of river-going motorized and non-motorized vessels, including the passenger, the lighter cargo, and the oil tankers, etc. pass through the study area both from Chittagong port and the newly built Payra port to the upstream areas and even to the capital city and the other way around (Ahmed et al., 2019; Khan et al., 1998). Hence, the estuary under study serves as a potential shipping route for transportation; oil leakage, oil tanker accidents, waste dumping from lighters, and other ships. In addition, industrial, domestic, municipal, and agricultural untreated effluents get mixed with rivers, and the pollutants find their way to the Bay of Bengal through the Meghna estuary. Therefore, it is imperative to investigate the estuary for heavy metals and other important parameters.

Materials and Methods

Surface Water Sampling and Analysis

A total of 12 surface water samples were collected randomly from upstream to downstream of the Meghna River estuary during December 2018. Samples were collected in high-grade polythene bottles. The bottles were treated with 5% conc. HNO_3 acid and washed with deionized water before sample collection to avoid contamination. All the samples were then filtered with a 0.45µm syringe-head membrane filter and the filtered samples were acidified with ultrapure concentrated nitric acid to make a pH of <2 (Cenci and Martin, 2004; Islam et al., 2015). The collected water samples were sealed and stored at 4˚C and were brought to the laboratory for further analysis. During collecting the samples, the physiochemical properties, mainly pH, electrical conductivity (EC), and Total Dissolved Solids (TDS) of surface water samples were also measured and recorded in situ with calibrated portable instruments. Hanna's combo EC-TDS digital meter (Hanna HI98129) was used for the measurement of EC and TDS, and the pH of the samples was recorded by a portable digital pH meter (Hanna HI98312).

In the laboratory, heavy metals (Cr, Pb, Cd, As, and Hg) concentrations were estimated by using Inductively Coupled Mass Spectrometry (ICP-MS) (Bruker Aurora M90, Bremen, Germany) and using the method as described by Kimura and Iwano (2000). Analytical grade chemicals were used for preparing standards and reagents for analytical works. The mixture of HNO_3 and HCl (1:3) acid was used for digestion since it gives the maximum analyte recovery for heavy metal analysis (Uddin et al., 2016). From 100 ppm stock standards of 100 µg/L concentration of transition metals (Merck, Germany), intermediate standard solutions of 50 µg/L, 20 µg/L, 10 µg/L, 5 µg/L were prepared. Deionized water was used throughout the experiment to prepare all the standards. Procedural blanks were also run with the samples. All the elements were analyzed using calibration curves after the parameters were optimized for maximum signal intensity and sensitivity of the instrument. To locate the sampling points, GPS device was employed. The obtained data was subjected to further analysis.

Statistical Analysis

Multivariate statistical techniques are a quantitative and independent technique to simplify, concise and understand variations to provide meaningful insights like modelling and interpretation of large data set (Laaksoharju et al., 1999; Wu et al., 2005). In this study, multivariate statistical methods such as Principal Component Analysis (PCA), Hierarchical Cluster Analysis (HCA), and Correlation analysis were performed with the help of SPSS software (Version 20). These statistical methods are performed to evaluate the heavy metal distribution and their probabilistic sources in the surface water of the study area. Multivariate statistical analysis methods were applied to a dataset that is comprised of the concentrations of five (5) heavy metals collected from twelve (12) different locations. The heavy metals included Cr, Pb, Cd, Hg, and metalloid As. To check the suitability of dataset for PCA, Kaiser-Meyer-Olkin (KMO) and Bartlett's sphericity tests were carried out. KMO is a sampling acceptability that indicates the common variance which might be induced by underlying factors. PCA might be more functional if the values are close to 1. Basic statistics such as range,

mean, and standard deviation were computed in Microsoft Excel.

Principal Component Analysis (PCA)

Principal Component Analysis (PCA) is a statistical technique for identifying and expressing interrelationships among various sets of surface water heavy metals concentration data. It is used to extract the most significant variables for deciphering patterns with similar characteristics in a most concise way (Mustapha and Aris, 2012; Sârbu and Pop, 2005; Stetzenbach et al., 1999; Vasanthavigar et al., 2010; Wold et al., 1987). Its function is to convert the original variables into new set of variables (Axes) termed as principal components (PCs), which describe a linear combination of the original variables (Helena et al., 2000; Mellinger, 1987; Sârbu and Pop, 2005). The factor loadings are responsible for the correlation between PCs and variables, while the greatest positive and negative loadings make the largest contribution and provide subsequent information (Liu et al., 2003).

Hierarchical Cluster Analysis (HCA)

Hierarchical Cluster Analysis, which comprises a group of multivariate techniques, are executed to classify the variables (Massart, 1983) into a cluster that possesses a high internal uniformity within the clusters and a high external variability between the clusters based on the selected variables (Bhuiyan et al., 2016; Islam et al., 2018; Lattin et al., 2003). HCA can be interpreted by a 2D illustration commonly known as Dendrogram (Lokhande et al., 2008; McKenna, 2003; Rogerson, 2001). The linkage of branches of a dendrogram that are in proximity to each other indicates a stronger relationship between samples or clusters of sampling sites (Usman et al., 2014). In this study, HCA was carried out using Ward's linkage method that provides an extended proportion of accurately classified observations (Willett, 1987) utilizing squared Euclidean distances (Massart, 1983; Ward Jr, 1963). It also enables the verification of the output found from PCA (Ahmed et al., 2019).

Results and Discussions

Variation in Electrical Conductivity (EC), pH and Total Dissolved Solids (TDS) and concentration of heavy metals (Cr, Pb, Cd, As, and Hg) of the surface water of the estuary are the prime indicators for nature, quality, and aquatic life. Different physical parameters and toxic heavy metals concentration with their statistical summary as maximum, minimum, mean and standard deviation (SD) are shown in Table 1. The pH level of the collected samples varied from 7.40 to 8.32 with an average value of 7.86 that signifies surface water were slightly alkaline in nature. The pH values exhibit comparatively higher value from sample S1 to S6 than S7 to S12 (Table 1). EC and TDS values gradually increased from S1 to S12, where the EC values ranged from 11.61 to 28.35 mS/cm and the TDS values ranged from 7.63 to 18.14 g/L. Samples S1 to S5 were located in the upstream area, whereas S6 to S12 were located close to the saline ocean water (Figure 1).

The variation in heavy metal concentrations in the estuary is caused by highly loaded heavy metals input from upstream that make their way to the sea, which contains a comparatively lower concentration of heavy metals (Khan et al., 1998). Seasonal variation, amount of river discharge, residence time and hydrogeologic cycle govern the heavy metal concentration and distribution in the estuary area. The suspended materials and soluble heavy metals carried by the river water may deposit in the sediment and water column at the estuary which subsequently act as secondary sources of pollution (Khan et al. 1998). From the collected samples the concentrations of Cr, Pb, Cd, As, and Hg were found to range from 0.5-46.17 µg/L, 0.9-164.39 µg/L, 0.23-5.06 µg/L, 0.11-2.55 µg/L, and 0.05-5.07 µg/L with a mean concentration of 17.88 µg/L, 66.80 µg/L, 2.36 µg/L, 1.17 µg/L, and 2.60 µg/L respectively (Table 1). The mean concentrations of the heavy metals in the study area were obtained in the descending order of Pb>Cr>Hg>Cd>As.

Naturally occurring causes are not responsible for higher concentration of chromium in water body. Anthropogenic activities are accounted for higher concentration of chromium. A significant spatial heterogeneity is observed in the collected samples. The highest Cr concentration was observed at sample S2 (46.17 µg/L) followed by S3 (40.25 µg/L) and S4 (35.84 µg/L) which were located in the upstream area of the estuary and the lowest value was obtained at S11 (0.50 µg/L), which is located near open sea (Figure 2A). Cr concentrations were within the lowest biological chronic safety concentration recommended by the United States National Oceanic and Atmospheric Administration (NOAA) (Table 2). It also beyond the natural surface water background value. High Cr concentration increased the probability of toxicity in marine organisms due to uptake and accumulation in the living cells. The concentration of Cr might have increased due to direct disposal of industrial waste from textile, steel, petroleum, tanneries, fertilizer industries along with refused materials from ships and oil tankers (Ali et al., 2016; Facetti et al., 1998; Islam et al., 2015; Khan et al., 1998; Mohiuddin et al., 2012; Proshad et al., 2021).

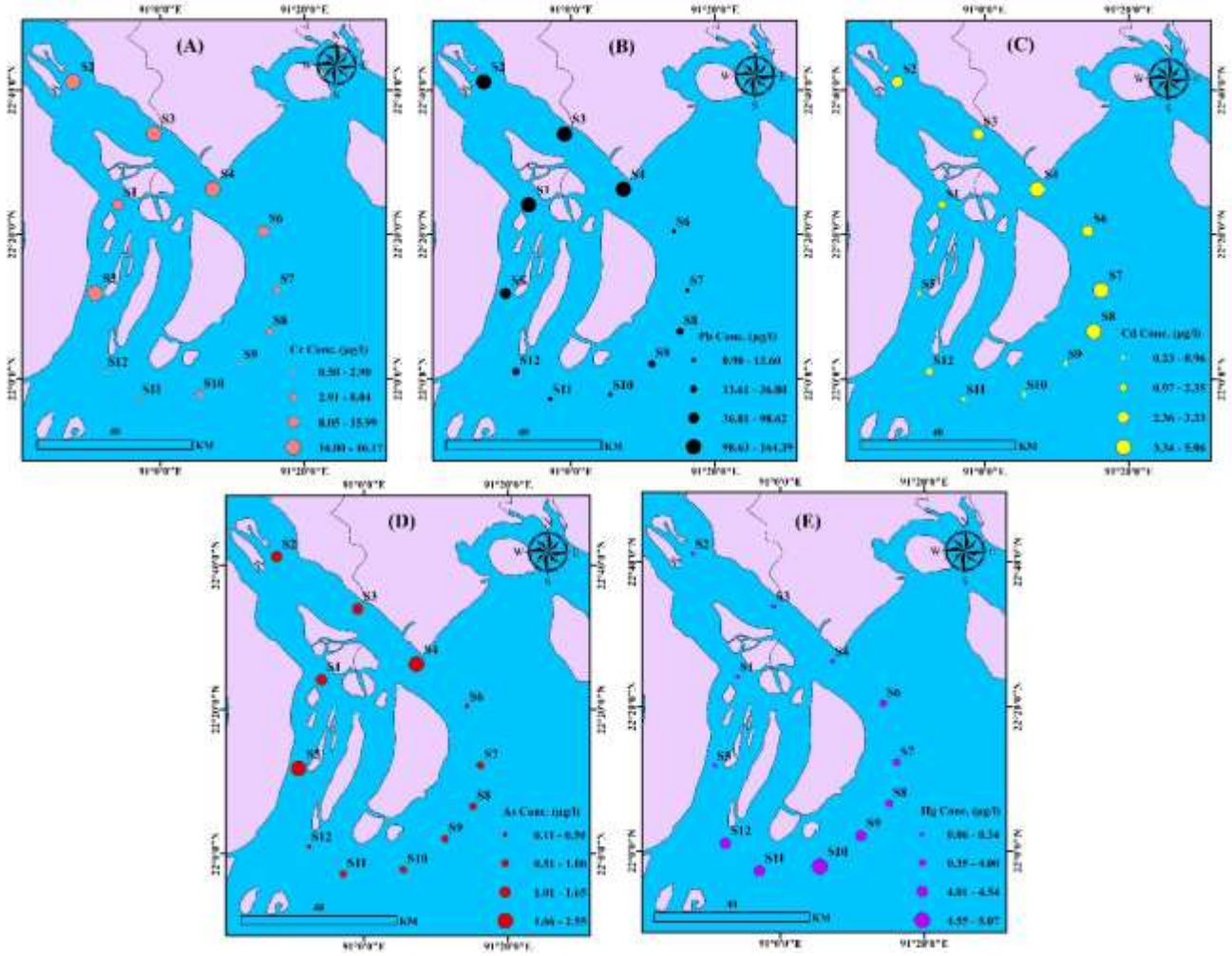

Figure 2: Spatial distribution of five toxic heavy metals in the Meghna estuary

Seawater contains some trace amount of lead and under normal conditions lead does not react with water (Kar et al., 2008). Lead and lead compounds are generally toxic pollutants that have adverse effects on aquatic biodiversity. The concentrations of Pb were found to fluctuate throughout the study area. The highest Pb concentration was found at sample S2 (164.39 µg/L) which was the northern most sampling point, and the lowest value was obtained at S6 (0.90 µg/L) (Figure 2B). The concentrations of Pb were much higher than the lowest biological chronic safety concentration recommended by NOAA (Table 2). It also exceeded the natural surface water background value (Table 2). The probable sources of lead in the study area might be battery recycling factories, ship breaking industries, municipal solid waste, electronic manufacturing, cement factories, and tires and steel works (Ali et al., 2016; Mohiuddin et al., 2012; Shikazono et al., 2012).

Cadmium is considered to be one of the most hazardous pollutants because of its toxicity to the organisms in the aquatic environment. It was found at all the sampling sites and showed an irregular trend in change among the sites (Figure 2C). The highest concentration of Cd was observed at sample S7 (5.06 µg/L) followed by S8 (3.75 µg/L) and S4 (3.73 µg/L), which were located in the southern part of the estuary close to the open sea. The lowest Cd concentration wasidentified at S5, located close to the land area towards south-western part of the study area. Cadmium concentration exceeded the background value of natural surface water but was much lower than the lowest biological chronic safety concentration set by NOAA (Table 2). The high concentration level of Cd in the estuary water might be due to the disposal of Cd polluted industrial waste in the river and upstream agricultural fields where cadmium-containing phosphate fertilizers have been used (Khan et al., 1998). Moreover, steel processing (smelting and electroplating) and untreated municipal sewage are regarded as potential sources for an elevated concentration of Cd in the aquatic environment (Ahmed et al., 2019; Mohanta et al., 2019).

Table 1: Physical and chemical parameters of the surface water of Meghna River estuary

Sample No.	pH	EC (mS/cm)	TDS (g/L)	Cr (µg/L)	Pb (µg/L)	Cd (µg/L)	As (µg/L)	Hg (µg/L)
S1	8.20	15.39	10.31	15.99	134.58	2.35	1.40	0.05
S2	8.10	12.88	8.24	46.17	164.39	3.33	1.40	0.34
S3	8.32	11.61	7.63	40.25	158.20	2.86	1.65	0.26
S4	8.30	19.80	12.85	35.84	131.31	3.73	2.10	0.09
S5	8.21	12.87	8.43	34.99	98.62	0.23	2.55	0.33
S6	8.00	25.10	16.06	15.73	0.90	2.88	0.50	3.80
S7	7.71	23.40	14.97	6.72	5.99	5.06	0.75	3.99
S8	7.53	26.65	17.05	5.35	30.24	3.75	0.70	4.00
S9	7.41	28.35	18.14	2.90	21.09	0.70	0.90	4.37
S10	7.58	27.30	17.47	8.04	5.89	0.96	1.00	5.07
S11	7.55	27.05	17.39	0.50	13.60	0.63	0.95	4.39
S12	7.40	25.75	16.56	2.09	36.80	1.89	0.11	4.54
Maximum	8.32	28.35	18.14	46.17	164.39	5.06	2.55	5.07
Minimum	7.40	11.61	7.63	0.50	0.90	0.23	0.11	0.05
Mean	7.86	21.35	13.76	17.88	66.80	2.36	1.17	2.60
SD	0.35	6.18	3.88	16.02	62.28	1.44	0.66	2.04

SD = Standard Deviation

Arsenic is one of the major concerns for humans due to its lethal effects on the human body (Mukherjee and Bhattacharya, 2001). As was detected at all the sampling sites of the study area. The concentration of As was higher in the upstream of the estuary than the southern part, which was in close proximity to the open sea (Figure 2D). The highest value was detected at sampling site S5 (2.55 µg/L) and the lowest value at S12 (0.11 µg/L). Arsenic concentrations were insignificant compared to the biological chronic safety concentration set by NOAA (Table 2). Both geogenic and anthropogenic sources are probably responsible for the present level of arsenic in the study area (Liu et al., 2020; Mukherjee and Bhattacharya, 2001). Arsenic rich parent rock from Himalayan catchment area is a significant source of arsenic in the study area (Ali et al., 2016; Chowdhury et al., 1999; Mitamura et al., 2008). The anthropogenic sources of arsenic could be attributed primarily to arsenic-based pesticides and fertilizers. Furthermore, some chemicals, such as copper arsenate, which is used to treat wood, and arsenic sulfide, which is used for tanning may increase the concentration of arsenic in the environment (Ahmed et al., 2016; Ali et al., 2016; Baeyens et al., 2007; Bhuiyan et al., 2011; Fu et al., 2014; Singare et al., 2012).

Mercury is a major concern because of its ubiquitous state that can coexist in soil, water, and air. The distribution of Hg in the study area was unique from the other studied metals as higher concentrations were observed in the lower part of the estuary, whereas the concentrations of other metals were low (Figure 2E). The highest concentration of Hg was detected in the seaward location at S10 (5.07 µg/L) and the lowest value at S1 (0.05 µg/L). The concentration of Hg is comparatively higher than the admissible limit of lowest biological chronic safety concentration recommended by NOAA (Table 2). Higher concentration of Hg in the Meghna estuary may have originated from steel industries, transportation of coal and oil, cement manufacturing industry and effluent discharge from textile mills and factories near the estuary (Bhuyan et al., 2019; Hassan et al., 2015; Saha and Hossain, 2011; Tang et al., 2010).

Principal Component Analysis

In PCA, numbers of PCs equal to the number of original variables and each PCs comprise the entire measured surface water quality variables. PCs with eigen values of≥1 are more significant; as a result, PCs having eigenvalues ≥1 were taken in order to interpret the relevant data structure (Jackson, 2005). The plot of eigenvalues and PCs (Scree plot) are shown in Figure 3 to determine the number of PCs retained. Thus, the first 2 PCs (Eigen value >1) were extracted with a total eigenvalue sum of 4.51, which appears to have a significant effect on the surface water quality and explained about 90.06% of the total variance in data. The factor loading, eigenvalues, percentage of variance, and cumulative percentage of variance elucidated by each factor for PCs are tabulated in Table 3.

Table 2: Comparison of surface water heavy metal concentration between Meghna estuary and several water quality standards (all are in µg/L unit)

Areas	Cr	Pb	Cd	As	Hg	References
Meghna Estuary	17.88	66.80	2.36	1.17	2.60	This study
The background value of natural surface water	0.2	0.005-0.015	0.01	ND	ND	Wu and Zeng (1983)
The lowest biology chronic safety concentration limit of the NOAA	>50	>8.1	>8.8	>36	>0.94	Long et al. (1995)

ND= Not determined/No data

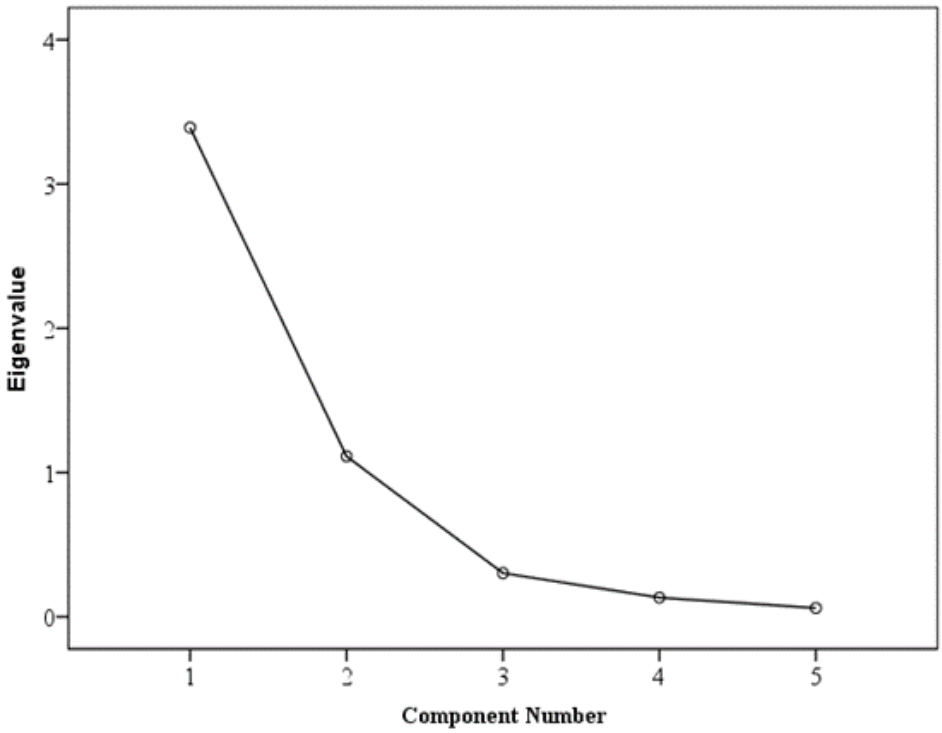

Figure 3: Scree Plot of eigenvalues of PCA

PC1 accounts for about 67.81% of the total variance, which signifies the heavy metals that are most influential. It includes the most significant toxic metals, which are positively loaded by Cr (0.95), Pb (0.90) and As (0.83) and negatively contributed by Hg (- 0.97). Higher loading of Cr, Pb, As, and Hg suggest the possible similar anthropogenic sources which may be attributed to the discharged water carried by the Meghna River. The industrial zone and big cities established on the bank of the river discharge toxic effluents which afterward make their way to the Bay of Bengal through the Meghna estuary. Those toxic effluents may be responsible for the elevated concentration of heavy metals in the Meghna estuary. Besides, agricultural, urban solid waste, and inland river transportation from Chittagong and Payra ports play a crucial role in elevating the concentration of toxic heavy metals in the Meghna estuary. Moreover, As is noteworthy because it has both active natural and anthropogenic sources. The Himalayan parent rock is the prime natural source of As in Bangladesh water (Mukherjee and Bhattacharya, 2001). Considering the anthropogenic source of As, it was reported that the imported coal transportation through that waterway contributes to a higher concentration of arsenic (Liu et al., 2020; Yu et al., 2007; Yudovich and Ketris, 2005).

The second component, PC2 contributes about 22.25% of the total variance with an eigenvalue of 1.12. PC2 shows a strong positive loading for Cd (0.95) and weak negative loading for As (-0.40) which is indicative of anthropogenic sources for both Cd and As. Cd polluted industrial waste (Steel plant), municipal solid waste, along with phosphate fertilizer are vital driving factors for the higher concentration of Cd accumulation in the study area (Mohanta et al., 2019).

Table 3: Factor loading of each toxic heavy metals of surface water with their variance and eigenvalues

Variables	PC1	PC2
Cr	**0.950**	0.168
Pb	**0.904**	0.164
Cd	0.189	**0.958**
As	**0.834**	−0.409
Hg	**−0.970**	0.120
Eigenvalue	3.39	1.12
% Of Variance	67.81	22.25
Cumulative %	67.81	90.06

Hierarchical Cluster Analysis (HCA)

The results of the HCA are presented through a dendrogram in Figure 4 which grouped all the 12 sampling sites into two statistically significant clusters under the similarity of surface water heavy metal content. Cluster-1, comprises about 42.6% (5 sites) of the total samples, whereas cluster 2 has incorporated about 58.4% (7 sites) of the total samples (Figure 4). The observation of the dendrogram shows an analogy between two clusters due to differences in the level of toxic heavy metals content. Mean values of surface water quality parameters resulting from the cluster analysis are presented in Table 4.

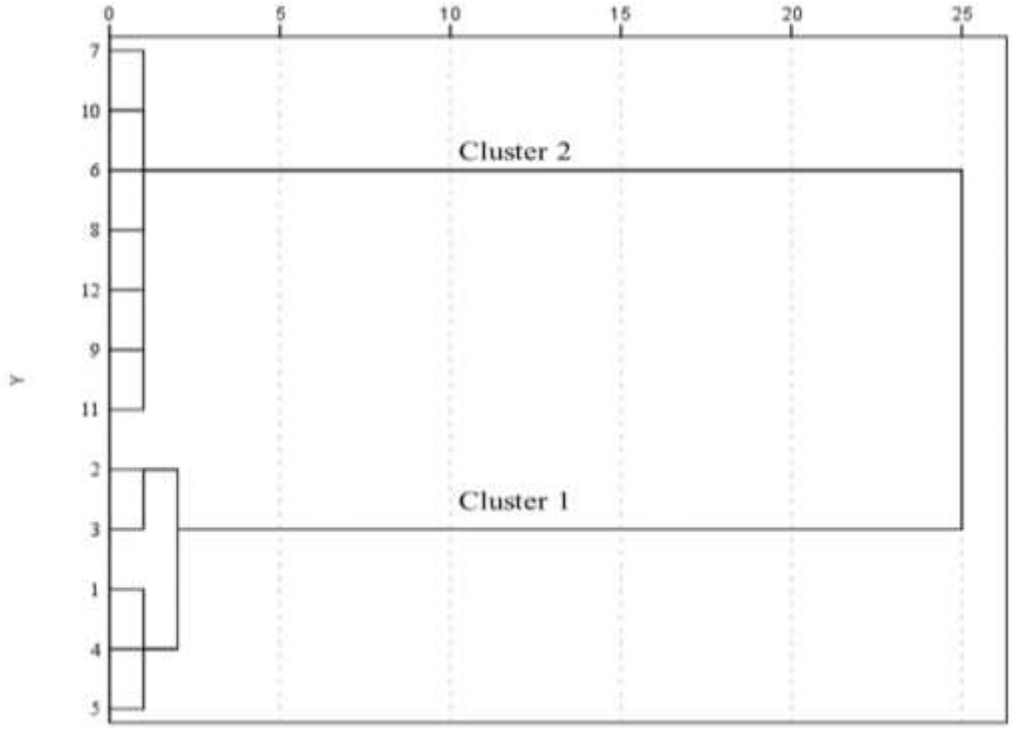

Figure 4: Dendrogram showing the clustering of the surface water sampling sites

Cluster 1 is generally characterized by the highest average concentration of Cr, Pb, Cd and As than cluster 2 (Table 4). This cluster is Cr, Pb, Cd, and As dominated. Samples in cluster 1 are mostly distributed in the upper part of the estuary (Figure 2) due to active anthropogenic sources are located at the upstream district or industrial areas to be more precise. The sampling site within cluster 2 is generally characterized by the lowest mean content of studied toxic metals (Table 4). All the samples in cluster 2 were located at the lower part of the estuary. It is the transitional zone where discharge from the upstream flushes out into open sea and may get diluted. Therefore, the concentration of metals in cluster 2 lower than cluster 1 except for Hg. Comparatively higher concentrations of Hg may be due to the impact of non-point anthropogenic sources found within the external portion of the estuary. The sampling sites in cluster 2 were located close to two important seaports, i.e., Chittagong and Payra along with highly polluted Karnaphuli River discharge in the south-eastern part, which may have elevated the Hg concentrations (Ali et al., 2016; Liu et al., 2020).

Table 4: Mean values of toxic heavy metals resulting from cluster analysis in the study area (all are in μg/L unit)

Parameters	Cluster 1 (n=5)	Cluster 2 (n=7)
Cr	34.65	5.90
Pb	137.42	16.36
Cd	2.50	2.27
As	1.82	0.70
Hg	0.21	4.31

N = Number of surface water samples in each cluster

Although there are a few dissimilarities between the outcomes of CA and PCA, however, a good agreement inferred from the dataset that was analyzed. In cluster 1, significantly higher values of Cr, Pb, Cd, and As are shown irrespective of their location, where as those variables exhibit higher loading in PC1 and PC2 (Table 3).

Correlation Matrix Analysis

The correlation coefficient matrix is a simplified statistical method that summarizes data for advanced analysis and determination of relationship and degree of reliance between a pair of variables (Bahar and Reza, 2010) within a range from -1 to +1. Where +1 stand for total positive linear correlation, -1 for total negative correlation and 0 for no correlation. The Pearson's correlation coefficients of 5 toxic heavy metals were encountered and displayed in Table 5. From the study it is found that Cr showed a strong positive correlation with Pb (0.838), As (0.752), and Hg (0.877). Lead showed a significant positive correlation with Hg (0.860), Cd (0.763), and As (0.577). Arsenic possessed a strong negative correlation with Hg (-0.811). The strong correlation among Cr, Pb, Cd, As, and Hg may signify the same sources (anthropogenic), their degree of pollution and independence during transportation in the river system (Ali et al., 2016; Chen et al., 2012; Håkanson and Jansson, 1983; Islam et al., 2015; Jiang et al., 2014; Li et al., 2009; Suresh et al., 2012). Besides, the high correlation among toxic heavy metals demonstrated that they would sustain in nature by forming strong

bonding with themselves and water (Proshad et al., 2021; Yang et al., 2015). The strong negative correlations of As with Hg suggested that they have different origins (Liu et al., 2020). Cluster Analysis and PCA mentioned that As and Hg may have dissimilar sources which may be the result of the joint influence of the Karnaphuli River and Chittagong seaport (Liu et al., 2020).

Table 5: Results of the correlation analysis of the toxic heavy metals of the study area

	Cr	Pb	Cd	As	Hg
Cr	1.0				
Pb	**0.838**	1.0			
Cd	0.196	**0.763**	1.0		
As	**0.752**	**0.577**	-0.155	1.0	
Hg	**0.877**	**0.860**	-0.171	**-0.811**	1.0

Comparison with Other Studies

The comparison of the toxic heavy metal concentration of the surface water of Meghna estuary with some rivers, estuary, and sea surface water along with international standards are given in Table 6. Since Bangladesh has no seawater quality standard. Therefore, the China's secondary seawater quality standard was used in this study. From the comparison, it was found that the mean value of lead, chromium, and mercury surpass China's secondary seawater quality standard whereas cadmium and arsenic were within the limit (Table 6). The mean concentrations of Cr, Pb, Cd, and Hg were higher than the Rupsha River, Bangladesh; Jinzhou Bay, China; Meiliang Bay of Taihu Lake, China; Yellow River estuary, China; Changjiang River estuary and Hangzhou Bay, China and Bohai Sea, China (Table 6). Cr, Pb, and Cd concentrations were significantly lower than the Karnaphuli River, Bangladesh, and Okumeshi River, Nigeria. Moreover, arsenic concentration was remarkably lower than the Karnaphuli River, Bangladesh; Rupsha River, Bangladesh; Jinhou Bay, China; Changjiang River estuary and Hangzhou Bay, China but higher than the Yellow River estuary, China. Rapidly growing industry and poorly managed solid waste putting the river system under threat in Bangladesh. Meghna River and estuary are in the most threatening situation as the overall data set suggested. Subsequently, those heavy metals gradually accumulated in the biodiversity and micro-organisms of the studied area through the food chain and disturbing the ecological balance afterward may affect consumers; humans to be more precise (Ahmed et al., 2019).

Table 6: Comparison of toxic heavy metals in surface water (μg/L) with international guideline and other studies in the world and Bangladesh

Areas	Cr	Pb	Cd	As	Hg	References
Meghna Estuary	17.88	66.80	2.36	1.17	2.60	This study
The Secondary Standard of Seawater quality standard of China	≤10.0	≤5.0	≤5.0	≤30	≤0.2	Wang et al. (2011)
Karnaphuli River, Bangladesh	78.25	13.34	8.55	28.91	ND	Ali et al. (2016)
Rupsa River, Bangladesh	8.03	7.21	1.18	5.75	ND	Proshad et al. (2021)
Jinzhou Bay, China	ND	0.61	0.92	2.19	0.03	Wang et al. (2012)
Ganges River, India	13.0	42.0	9.0	ND	ND	Gupta et al. (2009)
Meiliang Bay of Taihu Lake in China	2.84	5.06	0.74	ND	ND	Rajeshkumar et al. (2018)
Yellow River Estuary, China	ND	0.51	0.68	0.92	0.013	Tang et al. (2010)
Changjiang River Estuary and Hangzhou Bay, China	ND	0.9	0.387	3.6	0.172	Sun et al. (2009)
Bohai Sea, China	ND	1.05	0.45	1.66	0.04	Zhan and Wang (2005)
Okumeshi River (Nigeria)	870	450	1320	ND	ND	Ekeanyanwu et al. (2010)

ND = Not determined/No data

Conclusions

In this study, an approach was undertaken to assess the overall toxic heavy metals distribution within the Meghna estuary as it has the foremost diverse aquatic biodiversity and economically noteworthy region for Bangladesh. To portray an extensive overview of distribution and understanding probabilistic source of the heavy metals, the study confronts a multivariate statistical approach, i.e., PCA, HCA, and correlation analysis. PCA has identified active sources of heavy metals, most of which are anthropogenically driven to the estuary from upstream districts. Additionally, PCA analysis suggested that arsenic may have both geogenic and anthropogenic sources. Since arsenic (As) has anthropogenic sources, arsenic-rich Himalayan parent rock might be the dynamic natural source within the Meghna estuary. Concurrently, correlation analysis provided similar sources of the heavy metals inferred from PCA. HCA also showed that the spatial distribution of heavy metals is mainly from the upper part of the estuary to the open sea. Finally, correlation analysis verified the sources and enumerated the degree of pollution during toxic metal transportation through the studied estuary. From the study, it was also found that the concentration of toxic heavy metals exceeded the natural surface water background value of China. In some cases, concentration also outweighs the lowest biological chronic safety concentration limits recommended by NOAA. From the comparison of the

heavy metal concentrations of several rivers, bays, and estuaries with the heavy metal concentrations of Meghna estuary, it is suggested that Pb, Cr concentrations are higher. But As concentration is comparatively lower. Industrial, municipal, irrigational and sediment transport through the river is the active source of heavy metals in the study area. Overall, the results of the study suggest that the Meghna estuary has fallen under immense risk due to the higher accumulation of toxic heavy metals. Thus, extensive and persistent monitoring and reconnaissance are essential to upholding vigilant structures by incorporating appropriate environmental legislation. Besides, strictly maintained Environmental Impact assessment (EIA), Effective Effluent Treatment Plant (ETP), and restrained municipal solid waste discharge to the river should be undertaken to conserve the aquatic environment and biodiversity from toxic heavy metal pollution.

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