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ORIGINAL ARTICLE

Monitoring and assessment of heavy metal contamination in surface water and sediment of the Old Brahmaputra River, Bangladesh

Md. Simul Bhuyan[1] · **Muhammad Abu Bakar**[2] · **Md. Rashed-Un-Nabi**[1] · **Venkatramanan Senapathi**[3,4] · **Sang Yong Chung**[5] · **Md. Shafiqul Islam**[1]

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Abstract

The present study was conducted to measure globally alarming of ten heavy metals (Pb, Cd, Cr, Cu, Hg, Al, Ni, Co, Zn and Mn) in surface water and sediment of the Old Brahmaputra River in Bangladesh. The observed order of heavy metal mean concentration in water and sediments is Al > Mn > Ni > Co > Cu > Pb > Zn > Cr > Cd > Hg in mg/l and Al > Mn > Zn > Ni > Pb > Cr > Cu > Co > Cd > Hg in mg/kg, respectively. The significant variations of Cr, Cu, Al and Ni were found in the water of all seasons ($p < 0.05$), while sediment showed Pb and Hg exhibited substantial changes in terms of seasons ($p < 0.05$). Principal component analysis and correlation matrix revealed that significant anthropogenic input of Pb, Cd, Cr, Cu, Hg, Al, Ni, Co, Zn and Mn in water and sediment. In case of water, very strong linear relationships exhibited in Ni versus Cu (0.911), Ni versus Al (0.910), Mn versus Co (0.882), Cr versus Al (0.877), Cu versus Cd (0.853), Ni versus Pb (0.850), Zn versus Cr (0.833), Ni versus Cd (0.828), Cu versus Cr (0.827), Al versus Cd (0.827) and Zn versus Co (0.804) at the significance level 0.05. In sediments, very strong linear relationships were noted in Zn versus Cr (0.889), Al versus Pb (0.848), Co versus Al (0.819) and Mn versus Co (0.806) at the significance level 0.05. The result discovered that water and sediment quality of the Old Brahmaputra River became contaminated due to the anthropogenic sources of industrial, domestic and irrigation discharges. This environmental monitoring and assessment research will be useful for the management and planning for the protection of this river.

Keywords Heavy metals · Contamination · Water · Sediment · Statistical analysis · Old Brahmaputra River

Introduction

Contamination of heavy metals in river water and sediments is the major environmental focus especially during the last decade (Ozmen et al. 2004; Fernandes et al. 2008) because of their profusion, persistence and toxicity (Islam et al. 2015a; Ahmed et al. 2015a, b). Heavy metal contamination of river water and surface sediments becomes the major quality problems in fast developing cities since water and sediment quality maintenance and hygiene structure do not grow along with population and urbanization (Ahmad et al. 2010). Both natural and anthropogenic activities are largely liable for the heavy metal abundance in the environment (Wilson and Pyatt 2007; Khan et al. 2008). Massive deposit of toxic heavy metals is discharged by anthropogenic activities (Gao et al. 2009; Nduka and Orisakwe 2011) as well as by natural process that also contributes to the metal contamination in aquatic environment (Tarra-Wahlberg et al. 2001; Jordao et al. 2002; Khan et al. 2008; Bai et al. 2011; Grigoratos et al. 2014; Martin et al. 2015). Geological weathering, industrial disposal of metals and metal components, leaching of metals from garbage, solid waste heaps, animal and human excreta are major sources of heavy metals (Forstner 1983). Moreover, metal contamination in aquatic

✉ Venkatramanan Senapathi
venkatramanan@tdtu.edu.vn

[1] Faculty of Marine Sciences and Fisheries, University of Chittagong, Chittagong, Bangladesh

[2] Bangladesh Council of Scientific and Industrial Research, Chittagong, Bangladesh

[3] Department for Management of Science and Technology Development, Ton Duc Thang University, Ho Chi Minh City, Vietnam

[4] Faculty of Applied Sciences, Ton Duc Thang University, Ho Chi Minh City, Vietnam

[5] Department of Earth Environmental Sciences, Pukyong National University, Busan, South Korea

ecosystems is due to the effects of unplanned urbanization and haphazard industrialization (Zhang et al. 2011; Bai et al. 2011; Grigoratos et al. 2014; Martin et al. 2015; Bhuyan and Islam 2017, Chung et al. 2018). These activities can generate heavy metals in sediment and water that pollute the aquatic environment (Sanchez-Chardi et al. 2007).

In an aquatic ecosystem, heavy metals scavenged by fine particles lead to their accumulation in sediments. The contamination of heavy metals in water and sediments has produced significant adverse ecologic effects (Yi et al. 2011; Islam et al. 2014; Martin et al. 2015; Islam et al. 2015b, c; Ahmed et al. 2015c). However, the level of the risks is very tough to measure accurately, due to the difficulty of biologic and chemical interactions that is hugely responsible for the alteration of the bioavailability of metals. It may undergo regular changes due to dissolution, precipitation and absorption which affect their performance and bioavailability (Nicolau et al. 2006; Nouri et al. 2011). Heavy metals are released from sediments by the processes of re-suspension of particulates and activities of micro-organisms within the sediments and at the sediment–water interface. These soluble forms of the heavy metals may be found in crustaceans, finfish and shellfish (NNPC and RIP 1986) and can be shifted to humans via the food chain pathways (Camusso et al. 1995; Sun et al. 2001; Zhou et al. 2004; Sharma et al. 2007; Yi et al. 2011; Alhashemi et al. 2012; Pan and Wang 2012; Rahman et al. 2013; Islam et al. 2015a; Ahmed et al. 2015a, b).

Sediments have been extensively regarded as environmental pointers for the evaluation of metal pollution in the watercourse (Islam et al. 2015d). Among the environmental pollutants, metals are of specific concern because of their possible toxic effects and capability to bioaccumulate in aquatic ecosystems (Censi et al. 2006). Sediment is an important part of the river basin, with the deviation of habitats and environments (Morillo et al. 2004). Thus, it is necessary to evaluate the concentrations of heavy metals in water and sediments.

Today, metal pollution has become the major problem in many fast-growing emerging countries like Bangladesh (Islam et al. 2015c; Kibria et al. 2016a, b; Bhuyan et al. 2016; Bhuyan and Islam 2017). The discharge of municipal wastes, unprocessed effluents from various industries and agricultural inputs in the open water bodies and rivers has created a frightening situation in Bangladesh (Venugopal et al. 2009; Islam et al. 2015a, c).

In Bangladesh, the Old Brahmaputra River is one of the most important rivers. Being the part of industrial developed zone, the pollution is increasing day by day, especially heavy metal pollution. This river is receiving a huge amount of untreated effluents from various industries such as textile crafts, dying industries, spinning mills, cotton, jute mills and others. High concentration of various heavy metals such as Al, Mn, Ni, Pb and Cu is discharged into the Old Brahmaputra River which contributes largely to pollution of the surface water and sediments. Unfortunately, no scientific research regarding heavy metal pollution in surface water and sediment of the Old Brahmaputra River has been conducted so far. Therefore, the foremost objectives of the present study were to determine the heavy metal concentrations in water and sediment. It is expected that this research can contribute to the identification of heavy metal contamination sources and origin and to the effective conservation and management of Old Brahmaputra River system.

Materials and methods

Sampling sites

The Old Brahmaputra River, close to the Narsingdi district (24°08′03.76″N & 90°47′09.62″E), is one of the most important ecosystems with more aquaculture farms (Fig. 1). It plays a very important role in minimizing rural poverty and supplying food to the poor fishing community as well as for local people. It is an active river that plays a significant role in morphological changes in the downstream area (Amacher et al. 1989). Continuous variations of the river's course constitute a significant factor in the hydrology of the Brahmaputra. Samples were collected at Drenerghat (24°08′54.42″N & 90°42′02.64″E) and Belanagor (23°56′19.12″N and 90°42′36.07″E) (Fig. 1). There are lot of textile craft industries and dying industries along the Old Brahmaputra River. Moreover, domestic sewage adds another pollution dimension of the river. In summer season, the river water layer gets down and water gets mixed with the pollutants discharged by these industries.

Geological information

The Brahmaputra River is a trans-boundary river which flows through China, India and Bangladesh. In Bangladesh, the Brahmaputra is linked by the Teesta River (one of its largest tributaries). The Brahmaputra River gets split into two distributary branches below the Teesta. The western branch contains the majority of the river's flow which continues as the Jamuna to merge with the lower Ganga, named the Padma River. The eastern branch is called the lower or Old Brahmaputra River. Brahmaputra River starts its 3000 km journey to the Bay of Bengal from the slopes of Kailash in western Tibet. The Tsangpo known as Tibet's great river crosses from east to the high-altitude Tibetan plateau, north of the Great Himalayan Range with various myriad channels and sandbanks on its way. It comes down from Sadiya in the south of Arunachal Pradesh and passes through the Dibrugarh, Neamati, Tezpur, Guwahati and

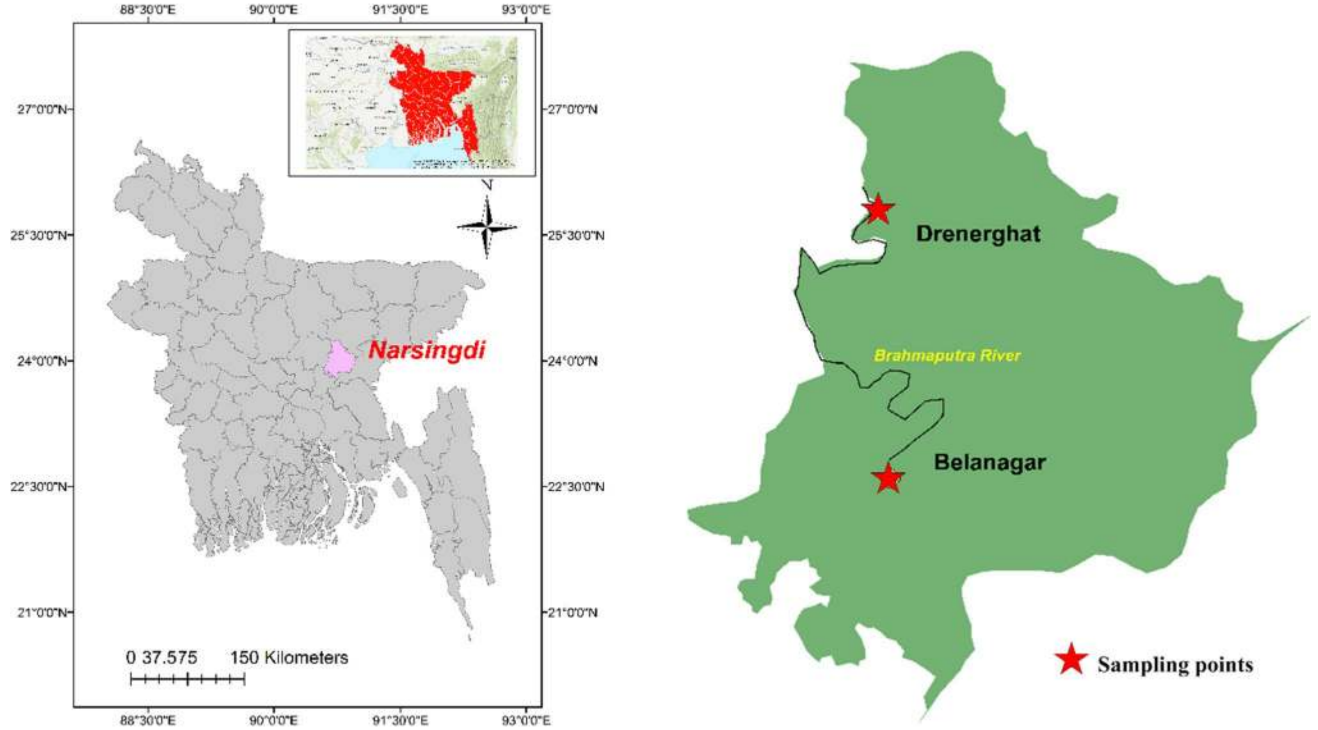


Fig. 1 Map showing sampling points of the Old Brahmaputra River

finally joins the Padma, the easternmost branch of the Ganges. This connected watercourse then flows into Bangladesh where it joins with the Meghna River. The Brahmaputra is navigable for favourable geological properties, especially for its length. The lower part of this river is revered to Hindus. Being one of the major rivers that are prone to tides in the world, the river is exposed to catastrophic flooding in spring when the Himalayan snows start to melt. Late Quaternary sediments of the Brahmaputra basin revealed a history of river swapping and climate change as exposed from sand- and clay-size mineralogy of shafts and up-to-date riverbed grabs. Epidote-to-crimson ratios in sand fraction sediments are diagnostic of the source with high E/G that designates Brahmaputra origin and low E/G indicates Ganges prominence. The Brahmaputra contains kaolinite (29%), illite (63%) and chlorite (3%) (Bhuyan and Islam, 2017).

The Ganges and Brahmaputra rivers have changed their position several times during the Holocene that is indicated by the analysis of mineralogical and stratigraphic data of Brahmaputra basin. Prolonged eras of mixed river inputs seem to be distant to the Early Holocene, representing rapid migrating and meandering channels during sea level low stand. During much of the Holocene, tectonic elevation of Sylhet Basin may have contributed to the larger course of this river. Varying degrees of physical and chemical weathering indicate the abundances of illite and chlorite contrast to smectite and kaolinite. In early post-glacial deposits, a dominance of physical weathering was suggested by High IC values in the Brahmaputra river course. Several geological changes in the basin were reported by improved chemical weathering under increasingly warmer and more humid conditions (Bhuyan et al. 2016).

Sample collection and preservation

A total of 12 surface water sample and 12 surface sediment samples were collected at three phases: September 2015 (rainy season), January 2016 (winter season) and March 2016 (pre-monsoon). Two litres of surface water sample was collected. Immediately after collection, water sample was acidified with nitric acid (pH = 2) and was transferred to the laboratory.

Heavy metal determination

The heavy metal contents of collected water and sediments were determined by atomic absorption spectroscopy (AAS, model iCE 3300, Thermo Scientific, UK) using standard analytical procedure (Table 1). Samples were carefully handled to avoid contamination. Glassware was properly cleaned by chromic acid and distilled water. Analytical-grade chemicals and reagents were used for blank determination and instrument readings. The techniques for sample

Table 1 Spectral lines used in emission measurements and the instrumental detection limit for the elements measured by using ICP–AES

Elements	Wavelength (nm)	Instrumental detection limit (mg/l)
Hg	253.7	
Pd	217.0	0.013
Cr	357.9	0.0054
Co	240.7	0.01
Cd	228.8	0.0028
Mn	279.5	0.0016
Ni	232.0	0.008
Zn	213.9	0.0033
Cu	324.8	0.0045
Al	309.3	0.028

and standard preparation and analysis of metal have been briefly described below.

Sample preparation (sediment)

This procedure was also used for the destruction of organic matter. Precaution was to be taken to avoid losses by volatilization of elements. The samples were weighed accurately, and a suitable quantity (10 to 20 g) was transferred to a silica crucible. After that, the samples were dried at 120 °C in a laboratory oven. These dishes were then placed in the muffle furnace at ambient temperature, and the temperature is raised slowly to 450 °C at a rate of no more than 50 °C/h. The samples were ignited in a muffle furnace at 450 °C for at least 8 h. After the samples are cooled, the dishes were removed from furnace. Then the sediment samples were digested in desired amount of 50% nitric acid on hot plate. Again samples were filtrated into a 100-ml volumetric flask using Whatman No. 44 filter paper and the residue was washed. Each sample solution was made up to the mark with distilled water (Chung et al. 2018).

Sample preparation (water)

A 100 ml water of each of the water samples was taken in a beaker. Then the samples were digested by adding 5 ml conc. HNO_3 on a hot plate. After that, the samples were filtrated into a 100-ml volumetric flask using Whatman No. 44 filter paper and made up to the mark with distilled water.

Standard preparation

All samples were prepared from chemicals of analytical grade with distilled water. 1 gm of metal cadmium, copper, lead and nickel was dissolved in HNO_3 solution; 1 g of cobalt, iron, manganese, zinc and aluminium was dissolved in HCl solution; 2.8289 g $K_2Cr_2O_7$ (= 1 g chromium) was dissolved in water and made up to 1 L in volumetric flask with distilled water; thus, stock solution of 1000 mg/l of Cd, Cu, Pb, Ni, Co, Fe, Mn, Zn, Al and Cr was prepared. Then 100 ml of 0.1, 0.25, 0.5, 0.75, 1.0 and 2.0 mg/l of working standards of each metal except iron was prepared from the stock using micropipettes in 5 ml of 2 N nitric acid. 100 ml of 2.0, 2.5, 5.0, 10.0 and 20.0 mg/l of working standards of iron metal was prepared from iron stock solution. Reagent blank was prepared in the same manner of sample preparation without sample to avoid reagent contamination (Thompson and Howarth 1976; APHA 1995).

Analysis of sample

The atomic absorption instrument was set up, and flame condition and absorbance were optimized. Then blanks (deionized water), standards, sample blank and samples were aspirated into the flame in AAS (model iCE 3300). The calibration curves were obtained for concentration versus absorbance. Data were statistically analysed using fitting of straight line by least square method. A blank reading was also taken, and necessary corrections were made during the calculation of concentration of various elements.

Statistical analysis

The research data sets were interpreted with various statistical analyses using SPSS (version 22). The one-way analysis of variance (ANOVA) was performed by the concentration of heavy metals in terms of seasons and sites. G-Graph was used for the graphical appearance of heavy metal in seasons and sites. Principal component analysis (PCA) was carried out on the source of heavy metal contamination. Pearson's product–moment correlation matrix was obtained to identify the relationship between the metals. Cluster analysis (CA) is an effective tool to find out the similarity and variation with the influencing factors on different data sets (Wang et al. 2014). Moreover, CA is an important tool for the characterization and simplification of data sets with the behaviour they possess. CA (dendrogram) was performed to show the similarity among variables and to identify their sources of origin using PRIMER (version 6).

Results and discussion

Seasonal variation of heavy metal levels in surface water samples

The concentration of heavy metals in surface water and its comparison with other rivers of Bangladesh are given in Table 2. The mean concentration of heavy metals is shown in Fig. 2 and Table 3. The observed concentration of heavy metals is in the following order: Al > Mn > Ni > Co > Cu > Pb > Zn Cr > Cd > Hg in mg/l. The concentrations of Al ranged between 1.5 and 11.5 mg/l. A higher concentration (11.5 mg/l) of Al was recorded at Belanagor in post-monsoon. The concentration exceeded the admissible limit (0.2 mg/l) set by ECR (1997). The lowest concentration of Al (1.15 mg/l) in monsoon season is at Belanagor. The highest concentration of Mn was recorded (2.5 mg/l) at Belanagor during post-monsoon that was above the permissible limit of standards as WHO (2004), ECR (1997) and EPA (1986). This concentration is also above the value recorded. This is due to the geogenic origin and also attributed to the minor input of anthropogenic sources (Balkis et al. 2010; Mokaddes et al. 2013). Ni concentration in water varied from 0.02 to 0.8 mg/l. A higher concentration of Ni was noted (0.8 mg/l) at Drenerghat in

Table 2 Comparison of the observed values of heavy metals in the water of the Old Brahmaputra River with other rivers of Bangladesh

River	Pb	Cd	Cr	Cu	Hg	Al	Ni	Co	Zn	Mn	References
Old Brahmaputra River	0.11	0.001	0.01	0.12	0.001	6.87	0.44	0.2	0.01	1.44	Present study
Buriganga	0.07	0.009	0.59	0.163	–	–	0.008	–	–	–	Ahmad et al. (2010)
Buriganga	0.112	0.059	0.114				0.15		0.332	0.157	Bhuiyan et al. (2015)
Balu	0.001	0.008	–	0.01	–	–	–	–	0.02	0.03	Mokaddes et al. (2013)
Dhaleshwari	0.05	0.006	0.44	0.15	–	–	0.007	–	–	–	Ahmed et al. (2009)
Dhaleshwari	0.20	0.00	0.13	0.00	–	–	–	–	–	–	Ahmed et al. (2012)
Khiru	0.02	0.13	–	0.004	–	–	–	–	0.006	0.17	Rashid et al. (2012)
Karatoa	Trace	–	0.005	Trace	–	–	0.005	–	Trace	0.101	Zakir et al. (2012)
Karnofuly	0.14	0.01	0.25	0.05	–	–	–	–	0.28	0.12	Islam et al. (2013)
Meghna	BDL	0.003	0.035		–	–	BDL	–	0.036	0.009	Hassan et al. (2015)
Shitalakhya	0.001	0.01	–	0.005	–	–	–	–	0.02	0.05	Mokaddes et al. (2013)
Shitalakhya	0.05	0.003	0.08	0.04			0.02		0.72		Islam et al. (2014)
Turag	0.002	0.01	–	0.004	–	–	–	–	0.02	0.06	Mokaddes et al. (2013)

Fig. 2 Graph showing mean concentrations (mg/l) of heavy metals in water during three seasons

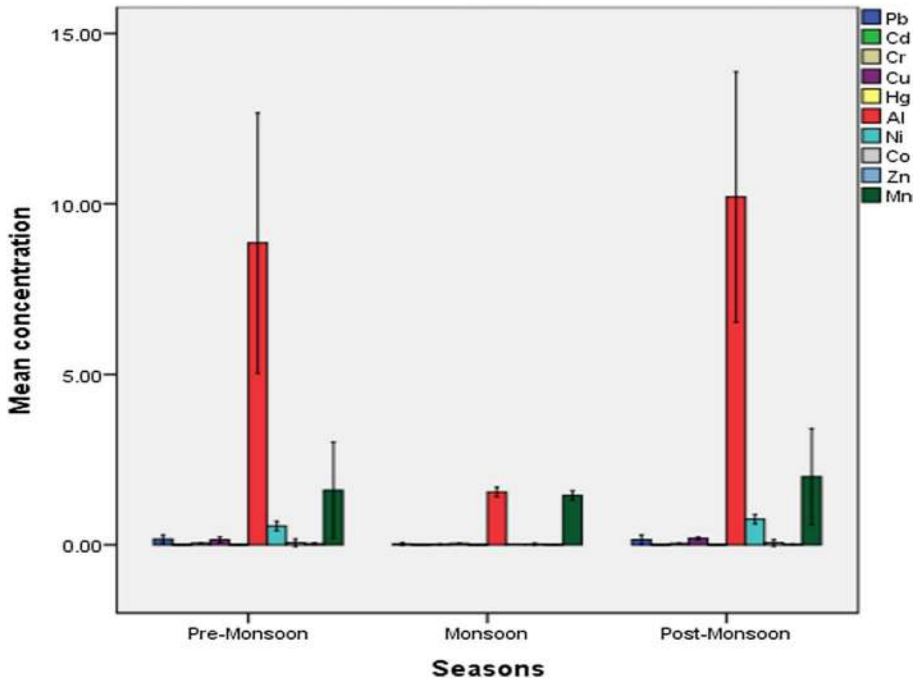


Table 3 Concentration of heavy metals in water and sediment at different sites in different seasons

Sites	Seasons	Pb	Cd	Cr	Cu	Hg	Al	Ni	Co	Zn	Mn
Heavy metals in water (mg/l)											
Belanagor	Pre-monsoon	0.12	BDL	BDL	0.18	BDL	10.2	0.5	BDL	BDL	2.1
Drenerghat		0.21	BDL	BDL	0.12	BDL	7.5	0.6	BDL	BDL	1.1
Belanagor	Monsoon	BDL	BDL	BDL	0.04	BDL	1.5	0.02	BDL	BDL	BDL
Drenerghat		0.04	BDL	BDL	0.03	BDL	1.6	0.02	0.03	BDL	1.4
Belanagor	Post-monsoon	0.10	BDL	BDL	0.2	BDL	11.5	0.7	BDL	BDL	2.5
Drenerghat		0.20	BDL	BDL	0.17	BDL	8.9	0.8	BDL	BDL	1.5
Heavy metals in sediment (mg/kg)											
Belanagor	Pre-monsoon	8.8	BDL	6.4	6.2	BDL	10,000	12.9	4.6	51.2	145
Drenerghat		8.1	BDL	5.2	5.7	BDL	8800	11.3	3.7	33.5	101
Belanagor	Monsoon	4.8	BDL	7.5	6.8	BDL	8200	11.5	3.85	81	105
Drenerghat		5.6	BDL	8	6.2	BDL	6900	13.5	3.9	65	145
Belanagor	Post-monsoon	9.8	BDL	6.5	6.4	BDL	11,200	14	4.8	52.5	155
Drenerghat		8.5	BDL	5.8	5.8	BDL	8900	13.5	3.9	32.78	106

a post-monsoon season that exceeded the allowable limit set by WHO (2004) and ECR (1997). A minimum concentration of Ni was observed (0.02 mg/l) at Belanagor and Drenerghat in monsoon season. The emission of contamination was caused by textile and dying industries (Ahmad et al. 2010; Faisal et al. 2004). Average Co concentration (0.20 mg/l) was measured at Drenerghat. Co is favourable to health, but the excess level of Co may pose lung and heart effects and dermatitis (ATSDR 2004). This is probably represented by geogenic and industrial inputs. A higher concentration of Cu was noted (0.2 mg/l) at Belanagor station in post-monsoon, and the average concentration of Cu is 0.12 mg/l, which is far below the allowable limit set of standards set by WHO (2004), EPA (2002) and ECR (1997). The concentration of Pb was recorded between 0.01 and 0.21 mg/l. The highest concentration was 0.21 mg/l at Drenerghat in pre-monsoon season. Pb exhibited the permissible limit set by standards of WHO (2004), USEPA (2006), EPA (2002) and ECR (1997). Moreover, Hassan et al. (2015) found the Pb concentration below detection limit from the Meghna River, Bangladesh. The lowest concentration 0.01 mg/l was recorded at Belanagor in monsoon season. Zn was found between BDL and 0.01 mg/l. This is significantly affected by paint and textile industries and associated with the discharge of sewage and irrigation runoff (Ahmad et al. 2010; Mokaddes et al. 2013). The concentration of Cr was observed between BDL and 0.01 mg/l, which was found below the limits of WHO (2004), USEPA (2006), ECR (1997). The concentration of Cd ranged from BDL to 0.001 mg/l. The maximum amount of Cd was recorded (0.001 mg/l) above the tolerable limits of USEPA 2006 and ECR (1997). In the case of Hg, it varied between BDL and 0.001 mg/l. This concentration was found within the limits of WHO (2004), ECR (1997), USEPA (2006). This is because of the effluents from industries and urban wastes (Ahmad et al. 2010).

ANOVA findings

ANOVA provided significant variations in the concentrations of Cr, Cu, Al and Ni that were found in terms of seasons ($p < 0.05$). No significant variations ($p > 0.05$) exhibited in metals Pb, Cd, Hg, Co, Zn and Mn. Moreover, predominant variations ($p > 0.05$) in the metal concentrations were recorded in terms of sites ($p > 0.05$).

Correlation matrix

Correlation matrix showed that a very strong linear relationship was found in Ni versus Cu (0.911), Ni versus Al (0.910), Mn versus Co (0.882), Cr versus Al (0.877), Cu versus Cd (0.853), Ni versus Pb (0.850), Zn versus Cr (0.833), Ni versus Cd (0.828), Cu versus Cr (0.827), Al versus Cd (0.827) and Zn versus Co (0.804) at the significance level of 0.05 (Table 4). Moreover, Al versus Cu (0.992) and Zn versus Hg (0.949) showed very strong linear relationship at the significance level 0.01. A strong relationship was observed in Cr versus Cd (0.760), Hg versus Cr (0.754), Cd versus Pb (0.742), Co versus Cr (0.742) and Ni versus Cr (0.719) at the level of 0.01. These results indicated that there was some original relationship between heavy metals, and revealed two different probable heavy metal sources such as anthropogenic and lithogenic (Islam et al. 2016a, b).

Seasonal variation of heavy metal levels in surface sediment samples

The concentration of heavy metals in surface sediments and its comparison with other rivers of Bangladesh are given

Table 4 Correlation matrix of heavy metals in water

	Pb	Cd	Cr	Cu	Hg	Al	Ni	Co	Zn	Mn
Pb	1									
Cd	0.742	1								
Cr	0.629	0.760	1							
Cu	0.620	0.853	0.827	1						
Hg	0.296	0.635	0.754	0.460	1					
Al	0.646	0.827	0.877	0.992	0.484	1				
Ni	0.850	0.828	0.719	0.911	0.253	0.910	1			
Co	−0.006	0.454	0.742	0.678	0.618	0.695	0.343	1		
Zn	0.217	0.574	0.833	0.567	0.949	0.603	0.302	0.804	1	
Mn	−0.194	0.304	0.453	0.641	0.259	0.619	0.312	0.882	0.492	1

Table 5 Comparison of the observed values of heavy metals in the surface sediment of the Old Brahmaputra River with other rivers of Bangladesh

River	Pb	Cd	Cr	Cu	Hg	Al	Ni	Co	Zn	Mn	References
Old Brahmaputra River	7.6	0.48	6.6	6.2	0.001	9000	12.8	4.1	52.7	126.2	Present study
Buriganga	69.75	3.33	177.5	27.85	–	–	200.5	–	–	–	Ahmad et al. (2010)
Buriganga	79.8	0.8	101.2	184.4	–	–	–	–	502.3	–	Saha and Hossain (2011)
Buriganga	31.4	1.5	173.4	344.2	–	–	153.3	–	481.8	4036	Mohiuddin et al. (2015)
Bangshi	59.99	0.61	98.1				25.67		117.15	483.4	Rahman et al. (2014)
Dhaleshwari	15.79	2.08	27.39	37.45	–	–	–	–	–	–	Ahmed et al. (2012)
Khiru	5.60	2.05	–	34.7	–	–	–	–	97.77	28.56	Rashid et al. (2012)
Karnofuly	4.96	0.24	0.76	1.22	–	–	–	–	16.30	15.30	Islam et al. (2013)
Karatoa	58	1.20	109	–	–	–	95	–	–	–	Islam et al. (2015c)
Meghna	9.47	0.23	31.74	–	–	–	76.1	–	79.02	442.6	Hassan et al. (2015)
Shitalakhya	28.36	5.01	63.22	–	–	–	39.22	–	75	–	Islam et al. (2014)
Shitalakhya	–	–	74.82	143.7	–	30,432.4	–	13.37	200.6	–	Islam et al. (2016a, b)
Turag	1.64	1.4	0.44	1.576	–	–	–	–	1.08	–	Banu et al. (2013)

in Table 5. The mean concentration of heavy metals was observed in the following order: Al > Mn > Zn > Ni > Pb > Cr > Cu > Co > Cd > Hg mg/kg (Fig. 3). Al concentration in sediments is ranged from 6900 to 11,200 mg/kg. The higher concentration of Al (11,200 mg/kg) was recorded at Belanagor in post-monsoon. The lowest quantity of Al (6900 mg/kg) was found at Drenerghat in monsoon season. Al concentration of sediments was above the permissible limit of FAO (1985).

The concentration of Mn was noted between 101 and 155 mg/kg. The highest level of Mn (155 mg/kg) was recorded at Belanagor, while the lowest concentration of Mn (101 mg/kg) was established at Drenerghat in pre-monsoon. The concentrations exceeded the permissible limit set by WHO (2008), USEPA (1999) and FAO (1985). It is suggested that the concentration Al and Mn originated from the lithogenic and is also associated with spinning miles and paint industries wastes (Rahman et al. 2014; Hassan et al. 2015). The concentration of Zn varied from 32.78 to 81 mg/kg. A higher concentration of Zn (81 mg/kg) was recorded at Belanagor, and lower concentration of Zn (32.78 mg/kg) was noted at Drenerghat in monsoon and post-monsoon seasons, respectively. This is above the limits of WHO (2008) and FAO (1985). Ni concentration was ranged from 11.3 to 14 mg/kg. A higher concentration of Ni (14 mg/kg) was observed at Belanagor in post-monsoon, while the lower level of Ni (11.3 mg/kg) was noted at Drenerghat in pre-monsoon. Ni was found below the allowable limits of WHO (2004) and USEPA (1999) but higher than the limits of FAO (1985). The concentration of Pb ranged between 4.8 and 9.8 mg/kg. The highest concentration of Pb (9.8 mg/kg) recorded at Belanagor in post-monsoon. This result is above the limit of FAO (1985). Effluents are from textile and paint industries and domestic waste runoff from the urban environment (Saha and Hossain 2011; Banu et al. 2013). The concentration of Cr varied between 5.2 and 7.5 mg/kg, whereas the higher concentration (7.5 mg/kg) of Cr was observed at Belanagor in monsoon season. The concentration of Cr was below the allowable limits of WHO (2004) and USEPA (1999). The concentration of Cu varied from

5.7 to 6.8 mg/kg. The maximum level of Cu (6.8 mg/kg) was noted at Belanagor in monsoon season. These results exceeded the permissible limits of FAO (1985). The concentrations of Co found ranged from 3.7 to 4.8 mg/kg. The highest concentration of Co (4.8 mg/kg) was recorded at Belanagor in post-monsoon. It is indicates that the concentrations exceeded the admissible limits (0.05 mg/kg) of WHO (2008) and FAO (1985). The concentrations of Cd were ranged between BDL and 0.54 mg/kg. The results are below the permissible limits of WHO (2004) and USEPA (1999). Hg concentration was recorded below the detection limit in all sediment samples in all seasons. A higher concentration of these metals was affected by discharges of textile and paint industries and domestic sewage waste (Datta and Subramanian 1998; Balkis et al. 2010; Ergul et al. 2008; Ahmad et al. 2010; Saha and Hossain 2011; Ahmed et al. 2012; Hassan et al. 2015).

ANOVA findings

Substantial changes in the concentrations of Pb and Hg were found in sediment in terms of seasons ($p < 0.05$). However, there are no significant variations ($p > 0.05$) in Cr, Cu, Al, Ni, Cd, Co, Zn and Mn. In the case of sites, no significant variations were found in the metal concentrations ($p > 0.05$).

Correlation matrix

Strong linear relationships were found in Zn versus Cr (0.889), Al versus Pb (0.848), Co versus Al (0.819) and Mn versus Co (0.806) at the significance level 0.05. Zn versus Cu (0.925) showed very strong linear relationship at the significance level 0.01. Strong relationships were observed in Cd versus Pb (0.788), Cu versus Cr (0.735) and Mn versus Ni (0.726) at the significance level of 0.05. In the case of grain size and metals, good linear relationships were observed in Ni versus % sand (0.953), Cu versus organic matter (0.925) and Cu versus organic carbon (0.924) at the significance level 0.01, and Zn versus % organic carbon (0.721), Zn versus % organic matter (0.737), Hg versus % sand (0.708) at the significance level of 0.05. Moderate relationships were observed in Mn versus % sand (0.598), Pb versus % sand (0.561) and Co versus % organic carbon (0.506) at the significance level 0.05 (Table 6). It suggested that grain size and organic matter and carbon play an important role in scavenging heavy metals in sediments (Venkatramanan et al. 2015a, b).

Table 6 Correlation matrix of heavy metals in sediment

	%Silt	%Sand	%Clay	%Organic carbon	%Organic matter
Pb	−0.515	0.561	−0.592	−0.143	−0.236
Cd	−0.265	0.331	−0.509	−0.345	−0.376
Cr	0.030	−0.019	−0.035	0.447	0.426
Cu	0.345	−0.264	−0.144	0.924	0.925
Hg	−0.666	0.708	−0.681	−0.112	−0.128
Al	−0.229	0.327	−0.635	0.391	0.310
Ni	−0.910	0.953	−0.859	−0.016	−0.141
Co	−0.380	0.488	−0.795	0.506	0.371
Zn	0.401	−0.364	0.109	0.721	0.737
Mn	−0.551	0.598	−0.620	0.297	0.136

Table 7 Component matrix of two-factor model with strong to moderate loadings in water

Water		
Eigenvalues (0.5)	Component	
	PC 1	PC 2
Pb	1.000	0.020
Cd	0.932	0.364
Cr	0.996	0.091
Cu	0.965	−0.263
Hg	0.670	0.742
Al	0.963	−0.268
Ni	0.904	−0.427
Co	0.999	−0.050
Zn	0.846	0.533
Mn	0.847	−0.531
Eigenvalue	8.416	1.584
%Total variance	84.157	15.843
Cumulative %	84.157	100.000

Principal component analysis (PCA)

The principal components analysis (PCA) are the uncorrelated variables, obtained by multiplying the original correlated variables with the eigenvalues. Surface water samples exhibited 100% in total sample variance (Table 7). Total variance of the PCs was 84.157% and 15.843% for PC 1 and PC 2, respectively. PC 1 is strongly correlated with Pb, Cd, Cr, Cu, Al, Ni, Co, Zn and Mn and PC 2 with Hg. The sources of PC 1 and PC 2 were derived from both lithogenic and anthropogenic inputs of paint and textile industries. Two PCs were extracted in sediment. The total variance of the PCs was 80.325% and 19.675% for PC 1 and PC 2, respectively (Table 8). PC 1 is strongly correlated with Pb, Al, Ni, Co, Mn and % sand and PC 2 with

Table 8 Component matrix of two-factor model with strong to moderate loadings in sediment

	Component	
Eigenvalues (0.6)	PC 1	PC 2
Pb	0.998	0.059
Cd	0.793	− 0.609
Cr	− 0.977	0.212
Cu	− 0.894	0.448
Hg	0.609	0.793
Al	0.962	0.274
Ni	0.948	0.317
Co	0.998	0.070
Zn	− 0.986	0.169
Mn	0.998	0.059
%Silt	− 0.892	− 0.452
%Sand	0.909	0.416
%Clay	− 0.978	− 0.210
%OC	− 0.580	0.815
%OM	− 0.772	0.635
Eigenvalue	12.049	2.95
%Total variance	80.325	19.675
Cumulative %	80.325	100.00

Hg and % OC. This is due to the sources of industrial and agricultural effluents.

Cluster analysis (CA)

CA was executed using square root and Bray–Curtis similarity to show the similarity among the parameters that contribute hugely to water pollution. From the output of the cluster analysis total, three clusters were found during different seasons. Cluster 1 includes Al and Mn; cluster 2: Co, Cr and Zn; and cluster 3: Ni, Pb and Cu (Fig. 4). Al and Mn represent strong linkage with minimum cluster distance that indicates those parameters have influencing power during seasonal variations. Parameters that are assembled together in less distance have a higher attraction with similar identical behaviour during temporal variations and also exert a possible effect on each other. Furthermore, Co, Cr and Zn have also strong linkage but lesser than cluster 1 but contribute largely to the environment. Ni, Pb and Cu are under the group of cluster 3 with minimum distance than cluster 1 and cluster 2 but have effects on the environment. This is revealed that these are affected by untreated industrial effluents from paint and textile and also agricultural inputs and domestic wastes (Chung et al. 2016). In the case of sediment, cluster 1 includes Cd and Hg; cluster 2: %OC and %OM; cluster 3: Co, Pb, Cr, Cu, Ni and %clay; and cluster 4: %silt, Mn, Zn and %sand (Fig. 5). Cd and Hg denote strong linkage with minimum cluster distance that designates those metals to have influencing power during seasonal variations. Heavy metals that are assembled together in less distance have a higher attraction with similar identical behaviour during temporal variations and have a possible effect on each other. %OC and %OM formed cluster 2 with minimum cluster distance. Moreover, Co, Pb, Cr, Cu, Ni and %clay have also strong linkage but lesser than cluster 1 and cluster 2 but contribute largely in the environment. %Silt, Mn, Zn and %sand are under the group of cluster 4 with minimum distance than clusters 1, 2 and 3 but have effects on the environment. This reflected the influence of effluents discharged from industries.

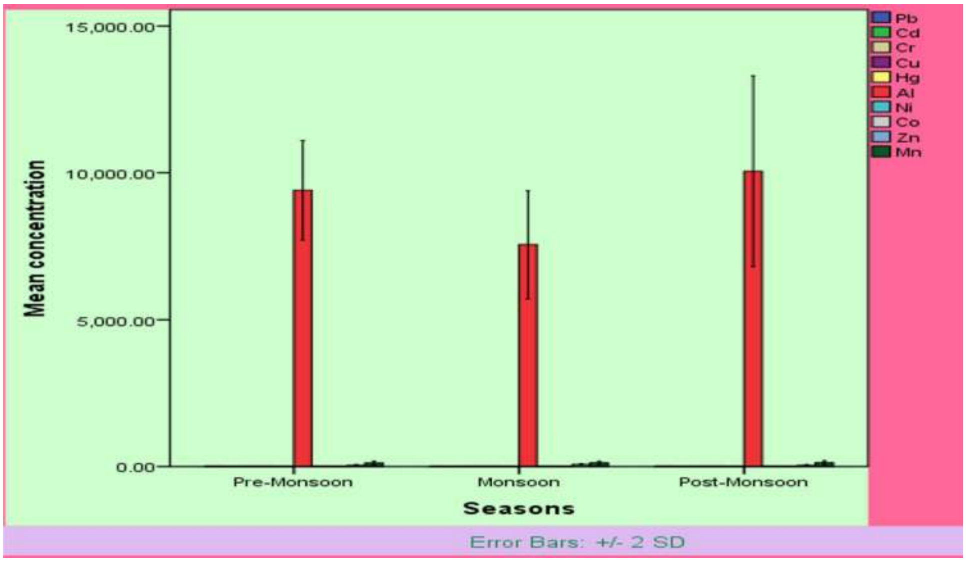


Fig. 3 Graph showing mean concentrations (mg/kg) of heavy metals in sediment during three seasons

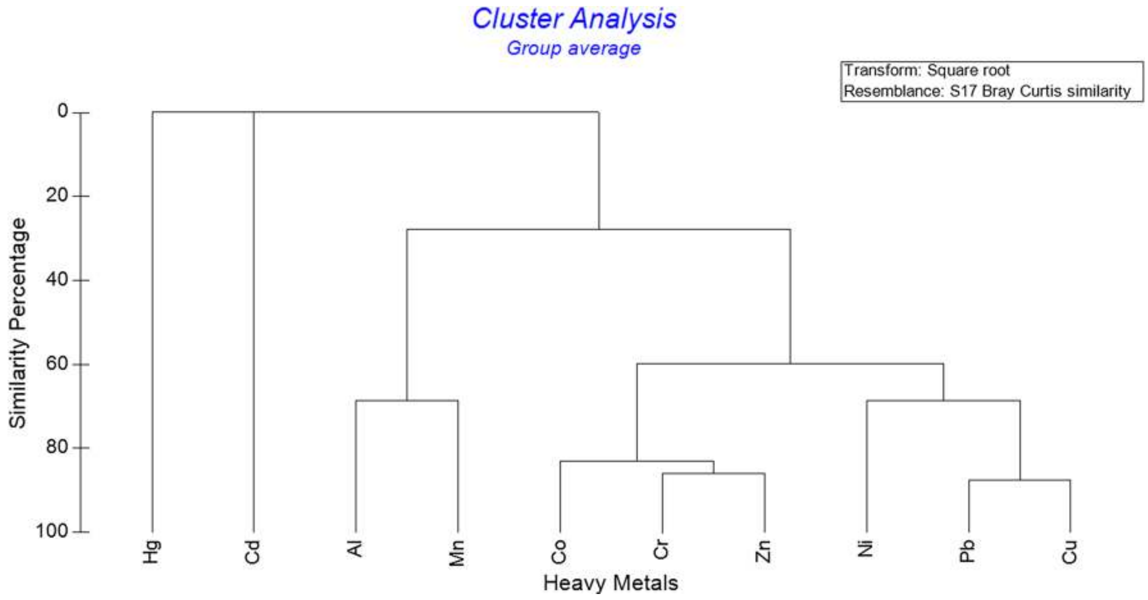
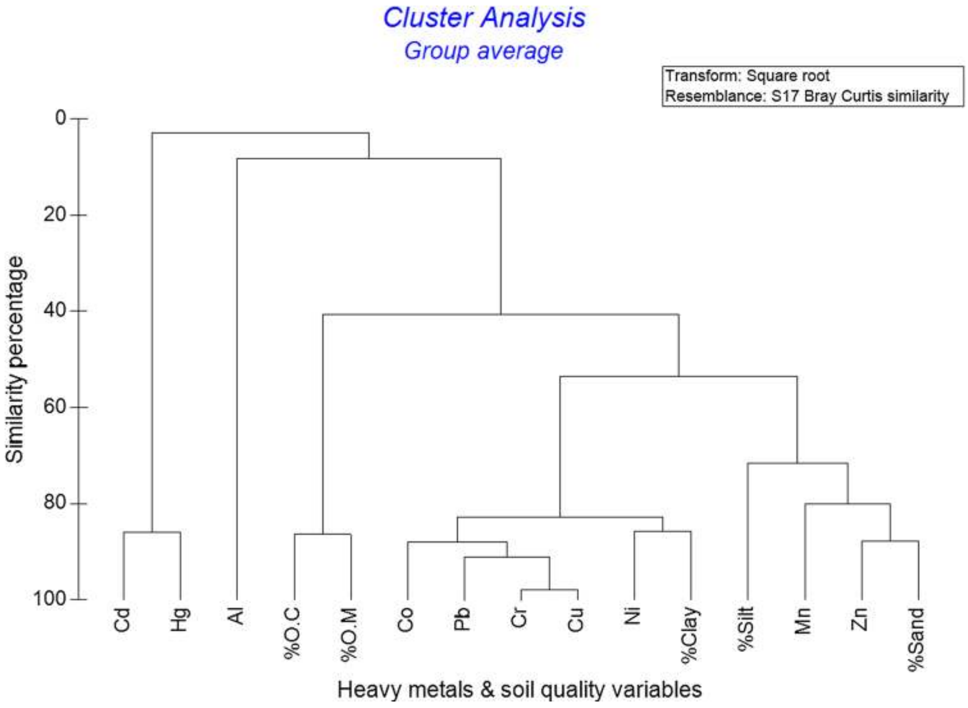


Fig. 4 Dendrogram showing the percentage of similarity among heavy metals during different seasons

Fig. 5 Dendrogram showing the percentage of similarity among heavy metals and soil quality variables in sediment during different seasons [OC = organic carbon; OM = organic matter]



Conclusions

Classical multivariate statistical analysis was used to evaluate the contamination sources of heavy metals at the Old Brahmaputra River. The observed order of heavy metal concentrations in surface water and sediments is as follows: Al > Mn > Ni > Co > Cu > Pb > Zn > Cr > Cd > Hg in mg/l and Al > Mn > Zn > Ni > Pb > Cr > Cu > Co > Cd > Hg in mg/kg, respectively. It suggested that effluents were discharged from paint and textile industries, irrigation and domestic wastes and also minor input from geogenic sources. Most of the metals exceeded the permissible limit set by the standards. PCA suggested that the contribution of metals in water and sediments was derived from the anthropogenic origin in addition to lithogenic sources. Moreover, grain size (size of sand, silt, clay) and organic matter acted as efficient scavengers for metals in sediments. Cluster analysis indicated that anthropogenic impact was accountable for monitoring the variability of metals in water and sediments and these metals leached from industrial wastewater. This research showed that the classical statistical analysis was a significant tool to identify contamination sources and origins. Further studies are needed on metal speciation and effects on metal uptake by human and organisms.

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Compliance with ethical standards

Conflict of interest The authors declare that there is no conflict of interest.

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