Seasonal and spatial distribution of heavy metals in an industrially affected river sediment and evaluation of ecological risk, health risk & pollution source

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Research Article

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Abstract

Present study sketched to quantify heavy metals (Cr, Mn, Ni, Cu, Zn, Pb, Cd, As) in sediment samples (Dry and Rainy season) of an industrially affected river namely Shitalakshya by Energy Dispersive X-ray fluorescence (EDXRF) technique. Different geochemical indices and multivariate statistical analysis were applied to define the accumulation, source and level of metal pollution in the sediment samples and probabilistic health risk implications due to dermal contact of sediment was also evaluated. Study revealed mean heavy metal (HM) concentrations in a sequence of Mn > Zn > Cu > Cr > Ni > Pb > As > Cd for both the seasons and almost all the elements were found within the suggestive standard value by various agencies with an exception of Cd and As. Enrichment factor (EF), geo-accumulation index (I_{geo}), contamination factor (CF) and pollution load index (PLI) indicated a high level of contamination by HM and a moderate level of Ecological risk was assessed for both the season. Hazard Index (HI), known as non-carcinogenic health risk for all the elements studied found bellow 1, indicating no harm for health and total carcinogenic risk also revealed the safe range. Results of multivariate statistical analysis indicates, the possible sources are mostly anthropogenic which may be owing to discharge of untreated wastes from various industries, metal and waste dumping sites, oil and refinery industries, glass and ceramic industries as they are located closely to the sampling sites of the Shitalakshya river.

Introduction

Heavy metal (HM) Pollution of rivers and other water bodies has become a major concern worldwide over the years, which is more acute in a river oriented country like Bangladesh. Basically, rivers and other water bodies are the one natural resource that people of Bangladesh has exploited the most In recent years, the reverine systems of Bangladesh have become more polluted as a consequence of rapid population growth, uncontrolled development on the riverbanks, urbanization, unplanned industrialization, and agricultural operations. Industries are considered the prime polluters because they utilize a huge amount of water and release untreated wastewater throughout the production cycle in the water bodies. Most of the industries in Bangladesh are river oriented and discharged their waste to the river prior to any treatment. Thus Aquatic ecosystem is the ultimate recipient of almost every waste including heavy metals. Due to non-degradable characteristics, accumulation of heavy metal in water bodies causing serious water and sediment pollution, moreover, HMs are carcinogenic, teratogenic, and mutagenic and thus it poses serious human health implications as well. The pollution level of the rivers in Bangladesh has increased rapidly, resulting a potential source of huge amount of toxic heavy metal compounds in the riverbed sediments (Ali et al.,2019;Bhuyan & Bakar, 2017). Depending on the types of pollutants it can be extremely hazardous to the flora and fauna present in or near the riverbed which by virtue of their nature can easily contaminate the environment as well as affect human health for a longer time exposure. The interaction mechanism depends on their toxicity, persistency, hard degradation capability, bio-accumulation capacity by organisms and bio-magnification mechanism through the food chain (Huang et al. 2020, Wei et al. 2016; Zahra et al. 2014; Zheng et al. 2013) etc.

Heavy metals have mostly originated from anthropogenic and geological /natural sources. Natural sources include weathering of metal containing rocks, erosion, forest fires, and volcanic eruptions while anthropogenic sources are mining and smelting, domestic discharges, industrial effluents, metallo–pesticides and combustion of fossil fuel(Gautam et al.,2016; Mirza et al., 2019; Karbassi et al., 2008; Malik et al., 2010; Martin JAR et al., 2015; Förstner et al.,1973;Reza et al., 2010) etc. Thus heavy metals discharged into a river system by natural or anthropogenic sources during their transportation are dispersed between the aqueous phase/water and bed sediments by leaching, diffusion and infiltration. The next phase is the accumulation of these heavy metals in sediment and biota (Proshad et al. 2019). Once they enter the water body may retain in the sediment for long time and usually affect the ecosystem and subsequently the human health. Retention of heavy metals in sediment depends on different physicochemical processes like adsorption/desorption, precipitation and complex processes during their transportation through the reverine system (Caporale and Violante, 2016). Usually sediment provides useful information for environmental and geochemical pollution status (Uluturhan et al., 2011; El-Said et al., 2014), as it is considered as an adsorptive sink for metals. A wide variety of habitats are living in sediments, therefore, determination of heavy metal contamination status in river sediments is important to ensure a safe, secured environment for them.

The Shitalakshya River flows through the east of Narayanganj District of Bangladesh, which is originated from one of the distributaries of the old Brahmaputra River and falls into the Dhaleshwari River near Kalagachhiya, in Barguna district, Barishal division, Bangladesh. The river is extremely significant to the peoples living nearby but due to excessive pollution caused by the disposal of various wastes into the river has made the future of the river itself to be uncertain and thus an urgent need for research on the potential adverse effect of heavy metals to assess the ecological risk and health risk due to dermal contact and their source identification in this terrestrial ecosystem is required. Thus the study was conducted to determine the concentration of heavy metals (Cu, Pb, Cr, Zn, Ni, Cd, Mn, As) in dry and rainy season, collected from the Shitalakshya river , which was eventually used to calculate different pollution indices such as Enrichment Factor (EF), Geo-accumulation Index (I_{geo}), Contamination Factor (CF)and Pollution Load Index (PLI). Identification of possible source applying Multivariate statistical analysis and ecological & health risk assessment of the heavy metal contaminated sediment of Shitalakshya river was done as well.

Materials And Methods

Sample collection and preparation

Ten (10) sediment samples were collected from Shitalakshya River at Narayanganj area which was apart from each other by 400 meters approximately (Fig.1). From each point, samples of around 2 gm were collected, kept in fresh zipper poly bag and tagged with sample location, number and date of collection. Sampling was started from Sultana Kamal Bridge Demra (23° 43' 18.462" N-Latitude and 90° 30' 2.1348" E-Longitude) and ended at Kanchpur Landing Station (23° 42' 3.96" N° N-Latitude and 23° 42' 3.96" NE-

Longitude) (Fig. 1) and collected in two different seasons; March (Dry season) and October (Rainy season) from ten (10) different locations (namely S-1, S-2, S-3, S-4, S-5, S-6, S-7, S-8, S-9, S-10).

Analytical method

Preparation of sample for elemental analysis by EDXRF

Visible roots and plant fragments were removed from the sediment samples manually and discarded. Small portions of each sample was placed in a cleaned and acid treated porcelain dish, dried at 60^{o}C for 48 hours in an oven till constant weight, grounded initially in a ball mill and finally with a mortar-pestle (carbide) to get fine particle of homogeneous mixture for the formation of pellets using the hydraulic press pellet maker machine (Specac, UK) applying 10 ton pressure (Tamim et al. 2016). To avoid any contamination, the grinder and pellet maker were cleaned properly before grinding and preparing the next sample.

Sample Irradiation and Method Validation

Sediment samples were analyzed for heavy metal concentration using a nuclear analytical technique called Energy Dispersive X-ray Fluorescence Spectroscopy (EDXRF) (Epsilon 5, Panalytical, the Netherlands).A Gadolium radioactive source which excite the sample, causes the atoms of the samples to absorb the radiation and emit radiations of a specific frequency corresponding to the character of the elements present in the sample (Jolly et al., 2013). As such, comparing the obtained spectral lines or frequency to the reference values of various heavy metals give the constituent elements present in the sample (Gilfrich, 1994).However for the validation of the method quality assurance and quality control test was performed using the standard reference material (Marine Sediment IAEA 433). The precision and accuracy were found within the acceptable limit (10%) as described in detailed by Jolly et al.,(2018).

Pollution load assessment of sediment

Evaluation of sediment contamination status was conducted by calculation of different indices and detailed of those indices with references are presented in the Table 1.

Evaluation of Health risk associated with the contaminated sediment

Ingestion, dermal contact and respiration are the three major routes usually considered in human health risk issue regarding soil/dust but in case of sediment dermal contact is only the pathway as sediment may come to contact with human by different household works like washing, cleaning, bathing or other recreational activities. This study thus focused only on health risk assessment owing to dermal contamination of sediment with human. The exposure through dermal contact can be calculated for non-carcinogenic effect using the equation   (USEPA 1989, 2004, Rovira et al. 2011, Iqbal et al. 2013, Jewel et al. 2020) as follows:

$$EXP_{dermal} = \frac{Cm \times CF \times SAXAF \times ABS \times EF \times ED}{BWXAT}$$

Where, EXP_{dermal} is the exposure via dermal intake mg/kg/day; C_m is the concentration of heavy metal in sediment mg/kg; CF is the unit conversion factor (10^{-6} kg/mg); SA is the exposed skin surface area (5700 cm2); AF is the adherence factors from sediment to skin (0.07 mg.cm-2); ABS is the dermal absorption from sediment (0.001); EF is the exposure frequency (350 days/years); ED represents the exposure duration (30 years); BW is the body weight (70 kg) and AT represents the average days (10,950 days).

Health risk assessment guideline (USEPA 2004) described Hazard quotient (HQ) as non-carcinogenic health effect due to heavy metal exposure of contaminated sediment, which can be calculated by the equation:

$$\mathbf{HQ_{Dermal}} = \frac{\mathbf{EXP_{Dermal}}}{\boldsymbol{Rf_d}}$$

Where, Rf_d is the oral reference dose of the respective contaminants and hence it is hypothetical that the reference dose via dermal contact is identical to the reference dose via dermal contact as reported by Iqbal et al. (2013).

HI is the Hazard Index via dermal contact of the contaminated sediment and calculated by the summation of the corresponding HQ_{Dermal} value as:

$$\mathbf{HI} = \sum_{i=1}^{n} \mathbf{HQ_{Dermal}}$$

When $HI<1$, it represents highly unlikely significant toxic interaction and $HI>1$ represents potential non-cancer health effect (Enuneku et al. 2018). In the present study, Non-carcinogenic Health risk assessment for the element Cr, Ni, Cu, Zn, Cd, Pb, As for the dry and rainy season has been calculated.

Whereas, Carcinogenic risk assessment was carried out by multiplying exposure dose (dermal) and the dose response relationship of particular chemical component as:

$$CR = EXP_{Dermal} \times SF$$

Where, SF is the carcinogenicity slope factor (per mg/kg-day).In case of multiple carcinogenic contaminants the cancer risk can be calculated by summation of all chemicals and routes and can be calculated as:

$$TCR = \sum CR_i$$

It is notable that risks lying between 1.0E−04 and 1.0E−06 are considered as an acceptable level (USEPA 1989), while risk exceeding 1.0E−04 are considered as a lifetime carcinogenic risk to the human body. Among studied elements only Cr and As have dermal effect in respect to carcinogenic view and the Slope factor (SF) for Cr and As is 2.00E+01 and 3.66 E+00 respectively (Wang et al., 2021).

Statistical analysis

The complex ecotoxicological processes can be well defined by the interrelationship and dependency among the variables and their relative weights (Bartolomeo et al., 2004) and Multivariate statistical method is the most suitable tools to find out the relationships among the variables and parameters. In the present study, Cluster analysis (CA), Principal Component analysis (PCA), and the Pearson correlation analysis were applied (Li and Zhang 2010; Varol et al., 2011), where PCA was applied to the dimensionless standardized data set of heavy metals for dry and rainy season and Cluster analysis (CA) was used to identify spatial variability among the sites (Mao et al., 2013) for both the season. All the statistical analyses were done using free statistical software R and IBM SPSS Statistics 25.

Results And Discussion

Seasonal and Spatial variation of heavy metals in the surface sediment samples

Mean concentrations (mg/kg) of measured heavy metals and their range are summarized in Table 2 for both the seasons (dry & rainy season). In dry season the mean heavy metal concentrations can be ordered as: Mn (611.90 ± 4.90) > Zn (162.70 ± 1.30) > Cu (147.03 ± 1.18) > Cr (69.42 ± 0.56) > Ni (39.45 ± 0.32) >Pb (20.01 ± 0.16) > As (13.94 ±0.11) > Cd (5.03 ± 0.04) and in rainy season the same trend was observed as: Mn (555.90 ± 4.45) > Zn (153.00 ± 1.22) > Cu (140.71 ± 1.13) > Cr (66.85 ± 0.53) > Ni (35.09 ±0.28) >Pb (19.60 ± 0.16) > As (13.37 ± 0.11) > Cd (4.74 ±0.04). In dry season the concentrations of all metals were slightly higher than the rainy season which can be attributed due to the fact that pollutants in river may be diluted by rain water thus lowering the value. To evaluate the sediment contamination level and its possible adverse impact in aquatic ecosystem various sediment quality guidelines (SQGs) have been developed over the years (Burton 2002,Li et al., 2013, MacDonald et al., 2000) and hence the metal concentrations were compared with the NOAA Marine Sediment Quality Guideline (ERL and ERM), the Canadian Interim Marine Sediment Quality Guideline (TEL and PEL), the United States Environmental Protection Agency sediment quality guidelines (USEPA,1997). When measured value of metal found below the threshold effect level (TEL), adverse biological effects are expected to occur rarely,whereas metal concentrations higher than Probable effect level (PEL) indicates possibility of frequent adverse effects(MacDonald et al., 2000). Long et al. (1995), identified the 10th and 50th percentile of the effects data as Effect range low (ERL) and effect range median (ERM) respectively were also used to compare with the present data (Table 2).The sediment samples from the Shitalakshya River having arsenic (As) concentration ranged from 12.50-15.22 mg/kg in dry season and in rainy season it was 12.04-14.70 mg/kg with an average of 13.94and 13.37 mg/kg in dry and rainy season respectively. According to USEPA (1997) the maximum limit of arsenic (As) in sediment is 7.24 mg/kg. The ERL and ERM limit of arsenic in sediment is 4.2 and 70 mg/kg respectively and the concentration of As in Shitalakshya river sediments was almost double and higher than maximum permissible limit according to USEPA (1997).Concentration of Cu ranged from 130.00-158.00 and 122.00-157.60 mg/kg in dry and rainy season with a mean value of 147.03 and 140.71 mg/kg respectively and hence the values are higher than USEPA (1997), TEL, PEL, ERL suggested value but lower than ERM suggested limit and 8-11 fold higher than USEPA, SQG suggested limit. Concentration of Cr ranged from 61.00 to 73.71 mg/kg in and dry season with a mean of 69.42 mg/kg and 61.00 to 72.36 mg/kg in rainy season with a mean of 66.85 mg/kg. From a previous study, the mean Cr concentration in Shitalakshya river sediment was found to be 63.22 mg/kg. From Table 2, it is observed that Cr concentration in Shitalakshya River was lower than the safety limit of USEPA, ERL, ERM and PEL recommended values but higher than the TEL suggested value. Level of Mn in sediments ranged in between 563-658 mg/kg in dry season and in rainy season it was 502-613 mg/kg with an averageof 611.90 and 555.90 mg/kg in dry and rainy season respectively. The level of Ni ranged from 33.30-46.28 with mean of 39.45 mg/kg in dry season and in rainy season it was 29.99-42.99 mg/kg, with a mean of 35.09 mg/kg but in a previous study meanof Ni wasfound 39.22 mg/kg, which agrees the present value. In comparison to the other river, Shitalakshya river sediment contains less amount of Ni and the concentration of Ni is higher than the ERL, TEL,and USEPA limit but lower than the standard values set by ERM and PEL. The sediment samples from the Shitalakshya river having zinc (Zn) ranged from 132-195 mg/kg in dry season and in rainy season it was 131-192 mg/kg with an average of 162.70 and 153.00 mg/kg in dry and rainy season respectively.Islam et al. (2015), reported the concentration of Zn in Shitalakshya river sediments was 75 mg/kg, however, Zn in Shitalakshya river sediments lies within ERM and PEL limits but exceeded USEPA, TEL, ERL SQG and slightly above the ERL limit in all points but far below the ERM limit. Concentration of Cadmium (Cd) ranged from 4.55-6.32 mg/kg in dry season and in rainy season it was 4.18-6.00 mg/kg with an average of 5.03 and 4.74 mg/kg respectively and hence was much lower when it is compared to other rivers sediment in Bangladesh. However, Cd concentration lies within the safety limit of ERM but exceeded all other recommended SQG limits (Table 2). The concentration of lead (Pb) in the sediment samples ranged from 18.31-22.65 mg/kg in dry season and in rainy season it was 18.25-21.91 mg/kg with an average of 20.01 and 19.60 mg/kg in dry and rainy season respectively and lied within the safety limit of ERL, ERM, TEL, PEL values but exceeded the SQG limit of USEPA (0.6 mg/kg). Previous study showed a mean concentration of Pb of 28.36 mg/kg in Shitalakshya river sediment. In rainy season the most polluted sampling sites can be ordered as S-6>S-9>S-10>S-1> S-4>S-8>S-3>S-2>S-7>S-5 whereas in dry season it was ordered as S-9>S-6>S-4>S-2>S-1>S-10>S-7>S-3>S-8>S-5.

Assessment of sediment pollution degree

Enrichment factor (EF)

EF is measured to identify the level of anthropogenic impact on sediment that is contribution of each element for the enrichment of sediment of an individual site and can be calculated by the equation shown in Table 1.The EF values of heavy metals in the Shitalakshya river sediments for both dry and rainy season are shown in Fig. 2 (a). EF values of As and Cu in the dry and rainy season at every point were significantly higher compared to other metals. The maximum enrichment of As (14.41 and 12.70 for rainy and dry season respectively) was found in the S-7 sampling site in both the season and EF values of Cu in dry season ranged from 6.57 to 4.91 and in rainy season it was 7.21 to 4.80 which lied in EF> 5 in almost every point, showing significant enrichment .The range of calculated EF values for the other studied elements were Cr:1.28 to 1.06 and 1.35 to 1.10 in dry and rainy season, Ni:1.25 to 0.86 and 1.24 to 0.82 in dry and rainy, Zn:1.93 to 1.23 and 2.25 to 1.30 in dry and rainy, Pb:1.68 to 1.30 and 1.85 to 1.39 in dry and rainy, Cd :0.03 to 0.02 and 0.03 to 0.02 in dry and rainy season respectively indicating deficiency to minimal enrichment (EF< 2). By considering the sampling point based on the enrichment value the most significant sites can be ordered as S-7>S-3>S-2>S-4>S-1 >S-8>S-5>S-9>S-10.

Contamination Factor (CF) and Pollution load index (PLI)

To measure the pollution level of the environment by each studied elements the parameter contamination factor (CF) usually calculated and the equation used for the estimation is expressed in detail in Table 1. Present study, computed the contamination factor for Cr, Mn, Ni, Cu, Zn, Cd, Pb As that is presented in Fig.2 (b), showing the variations of the calculated CF values for various heavy metals analyzed at different points in both dry and rainy seasons. The mean contamination factor for all the studied heavy metals of Shitalakshya river sediment showed similar trends for both the season. In the dry season the mean CF values showed the following trends As (9.29) > Cu (4.59)>Zn(1.28) >Pb(1.25) > Cr(0.97)>Mn(0.81) > Ni(0.80) > Cd(0.05) and in rainy season the trends was As(8.91)> Cu(4.40)>Pb(1.22) > Zn(1.20)> Cr(0.94)>Mn(0.74)> Ni(0.71)> Cd(0.04). Among all the studied heavy metals As had very high contamination factor both in dry (9.29) and rainy season (8.91). In case of Cu the range of CF values in both seasons is 4.93 to 4.06 in dry season and 4.925 to 3.81 in rainy season thus indicating considerable pollution in both seasons. Zn and Pb had a CF value greater than > 1 in both the season for all the sampling points and hence sediments of Shitalakshya river are moderately polluted by Zn and Pb (Table 1), whereas the mean CF values of Cr, Ni, Mn, and Cd are less than 1 and hence indicating low pollution.

PLI for sediment generally calculated to estimate the level of pollution associated by more than one contaminant in a particular site and calculated using the equation as illustrated in Table 1.Estimated value of PLI for different sampling points to evaluate the quality of the aquatic environment and the pollution status with Seasonal variation is shown in Fig. 2(c). In both season the calculated PLI values for all the points are above or near one (1) indicating, polluted by heavy metals. Comparing the seasonal variation of PLI values it can be concluded that the sediment samples are more polluted in dry season than in the rainy season which was consistent with the measured metal concentrations. From the Fig.2(c) the spatial variation of PLI values can also be predicted and can be apprised that site S-6, S-3, S-1, S-4, S-9 and S-2 are more polluted compared to other sites in both the season.

Geo-accumulation index (I_{geo})

I_{geo} is calculated to measure the degree of pollution by each heavy metal in the sediment sample, which is calculated by the equation shown in Table 1. In the present study I_{geo} for the element Cr, Ni, Cu, Zn, Cd, Pb, As is measured considering the mean value of the elements for the ten different site and presented in Fig. 3. Calculated I_{geo} values for both the season revealed that As and Cu have the maximum geo-accumulation index and showed positive I_{geo} values in all points. The mean I_{geo} values of As was 2.62 for dry and 2.57 for rainy season whereas for Cu it was 1.62 for dry and 0.69 for rainy season, indicating moderate to strong pollution by As, whereas Cu showed variation in I_{geo} values appraising moderately polluted by Cu in dry season and unpolluted to moderately polluted in rainy season.. However, all other heavy metals showed negative I_{geo} and their mean I_{geo} for both in dry and rainy season can be attributed as Cr; - 0.61 and -0.67, Ni; - 0.90 and -1.07, Zn; -0.23 and -0.32, Cd; -6.48 and -6.57 ,Pb;-0.26 and -0.29 in dry and rainy season respectively.

Assessment Potential Ecological Risk index (PERI)

Table 3 represents the potential Ecological risk index of the studied heavy metals. Potential ecological risk factor (E^i_r)of the studied heavy metals follows the order As>Cu>Pb>Ni>Cr>Cd>Zn. From the PERI calculation it has been observed that As has the highest single pollution load index and site S-7 showed maximum value. According to the classification of Potential ecological risk factor (E^i_r), arsenic (As) contamination can be categorized in the considerable ecological risk group, on the other hand all other studied heavy metals have (E^i_r) values less than 40, comprising low ecological risk at the studied area. Overall impression is that the studied sampling sites have PERI value greater than 110 and thus indicating moderate ecological risk by the toxic elements.

Associated Human Health Risk Assessment

Mean value of estimated dermal exposure (EXP_{dermal}) by the heavy metals Cr, Ni, Cu, Zn, Cd, Pb, and As in the sediment samples of Shitalakshya river have been computed along with Hazard Quotient (HQ) , Hazard Index (HI) and carcinogenic risk during dry and rainy season in the Table 4**.**The HQ value was found in the order as As>Ni>Cr>Pb>Cd>Cu>Zn in both the season and the values for dry season are higher than the rainy season. Hazard Index (HI) value for Dry season was 6.78 10^{-4} and for rainy season it was 6.36 10^{-4}. The calculated HI value for both the season was found below 1 (HI<1), indicating an acceptable non-carcinogenic risk by the heavy metal contaminated sediment collected from the Shitalakshya river.Carcinogenic Risk (CR) value for Cr and As was found 7.59 E-06 & 2.79 E-07 in Dry season and 7.31 E-06 & 2.67 E-07 in rainy season respectively and in both cases Cr showed a higher value than As but however all the CR value lied in the acceptable range (1.0E−04 and 1.0E−06) , indicating no risk posed.TCR value for both the season also found in the acceptable range of 10^{-7} –10^{-6} , indicating that the health risk is low or negligible, and will not cause obvious carcinogenic health effect to the population.

Source identification of metals based on Pearson correlation, Principal Component analysis and Cluster analysis

Pearson correlation

Table 3 shows the Pearson correlation coefficient of the studied toxic elements. From the Table 5 it is evident that Cr-Pb pairs has a strong positive correlation (0.695) at the significance level of $p<0.01$. Cr also shows a strong positive correlation with Mn (0.602) at the significance level of $p<0.01$, which indicates Cr, Pb and Mn may have common source of origins. It is also seen that Ni-Mn pairs has a moderate positive correlation of (0.457) at the significance level of $p<0.05$ and Ni-As shows moderate positive correlation of (0.344) whereas Ni-Cd pairs shows a strong positive correlation of (0.557) at the significance level of $p<0.05$. Cu-Cd pairs also has a moderate positive correlation of (0.437) and Cu-As has a correlation coefficient of (0.362) .This indicates that Ni, Cu, Cd and As may have originated from the same sources.

Principal component analysis

The Principal component analysis (PCA) results of the standardized dataset of the toxic elements have been shown in Table 5. showing that the PCA reduced the number of variables to three principal components (PCs) with Eigen values >1. From the Table 5, it is clear that PC1 has an eigenvalue 2.37 and it explains 29.62% of the data variance and highly loaded with Ni (0.636), Cu (0.759), Cd (0.762) and As (0.666). PC2 has eigenvalue 2.17 and is accounted for 27.21% of the data variance and has high positive loadings of Cr (0.896),Mn (0.863) and Pb (0.596). PC3 has eigenvalue 1.22 and it can explain 15.28% of the data variance with higher loading of Ni (0.625) and Zn (0.712). Fig.4a represents the scree plot and Fig.4b shows the PCA correlation circle for metals based on Pearson correlation matrix. This figure is a graphical representation of Table 5. For the first PC (DIM-1, Horizontal axis) accounting for 29.6% of the data variation and the second PC (DIM-2, vertical axis) it describes 27.2 % of variation.

Cluster Analysis

To find out the group of analyzed variables Cluster analysis was performed using Wards method with square Euclidean distance metric . From the cluster dendrogram (Fig.5), it can be elucidated that the studied elements have been divided into three statistically significant groups. Group -1 consists of Cr, Pb, Mn, group-2 consists of Ni, Cd, Cu, As and group-3 consists of only Zn which builds up a cluster with the second group with a long distance. Narayanganj and Demra asindustrially prone area, situated on the bank of Shitalakshya River discharge a huge quantity of wastes both solid and liquid, thus contributing heavy metal/toxic elements into the river. Similarly, Shiddirganj area was also highly polluted by heavy metals as power station, textile industries are located there. From field observation three main sources of pollution are identified in all the four reaches, which are: Dhaka city domestic wastewater, Industrial waste and Local waste. Oil industry and refinery are the main source of Pb and Cd. Metal and waste dumping place contribute to the high concentration of Zn, Pb, Mn . Boat and ship dock yard is probably the main source of Mn ,Pb, Zn whereas Soap factory is the contributory source of Pb. Dye factory, textile and tannery contribute mainly Pb, Cr, Co, Zn, Cd. Pb, Fe, Cr, Zn can be originated from Electroplating and High way & rail station respectively. Thus in group 1 the contributory source for Mn, Pb was boat and ship dock yard, metal and waste dumping station, whereas for Pb Cr the possible source may be dye, textile and tannery factory.In group 2 the contributory source for Cu, As was Glass and ferrous metal industries that are located near the bank of Shitalakshya river and Cd, Ni coming from Oil and Refinery industry, untreated industrial effluents from paint and textile industries. In group 3, Zn can have derived from multiple sources dye factory, textile and tannery, Electroplating and High way & rail station respectively.

Conclusion

The overall findings can be summarized as:

- The Shitalakshya river sediments is highly contaminated by arsenic (As),Cu shows considerable pollution whereas Pb and Zn are moderate contributor for polluting the river sediment. All other elements (Cr, Mn, Ni, Zn, Pb, Cd) have lower pollution status with comparison to As and Cu.
- Enrichment factor, geo-accumulation index, contamination factor and pollution load index indicate a high level of sediment contamination by heavy metals and also pollution levels in the dry season was slightly higher than the rainy season.
- In both the season, arsenic (As) has the highest contribution to the single factor pollution in all the sampling sites regarding ecological risk assessment and the ecological risks of other elements are low.
- Calculated value of HQ, HI, CR and TCR revealed that there is no non-carcinogenic and carcinogenic risk by the heavy metals studied in the sediment sample right now but a continuous monitoring is suggested to assess more precisely the non-cancer and cancer risk to the population.
- From Pearson correlation, PCA and CA analysis, the identified possible sources of the toxic elements Cr, Pb, and Mn may be from iron and steel industries, Cr originated from vegetable oil industries, Mn, Pb may have origin from Boat and ship dock yard and from metal & waste dumping sites. Pb can also come from the dust of lead acid battery factory, river bank erosion and atmosphere. On the other hand, Ni, Cu, Cd and As may have originated from the untreated industrial effluents of paint and textile industries, non-ferrous metal industries and also agricultural inputs. Finally Zn can have derived from multiple sources like dye factory, textile and tannery, Electroplating and High way & rail station respectively.

Declarations

Author contributions: Y.N. Jolly: Conception and design, writing and final approval of the manuscript; Atahar Rabby: sample collection and analysis; Mehedi Hasan: Data calculation and curation; S. Akter, K.M. Mamun and J. Kabir. M.S. Rahman: sample irradiation and data aquisation; Taseen Jubair Bhuiyan: Data calculation; A.M. Sarwaruddin Chowdhury: Visualization, Writing.

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Tables

Due to technical limitations the Tables are available as a download in the Supplementary Files.

Figures

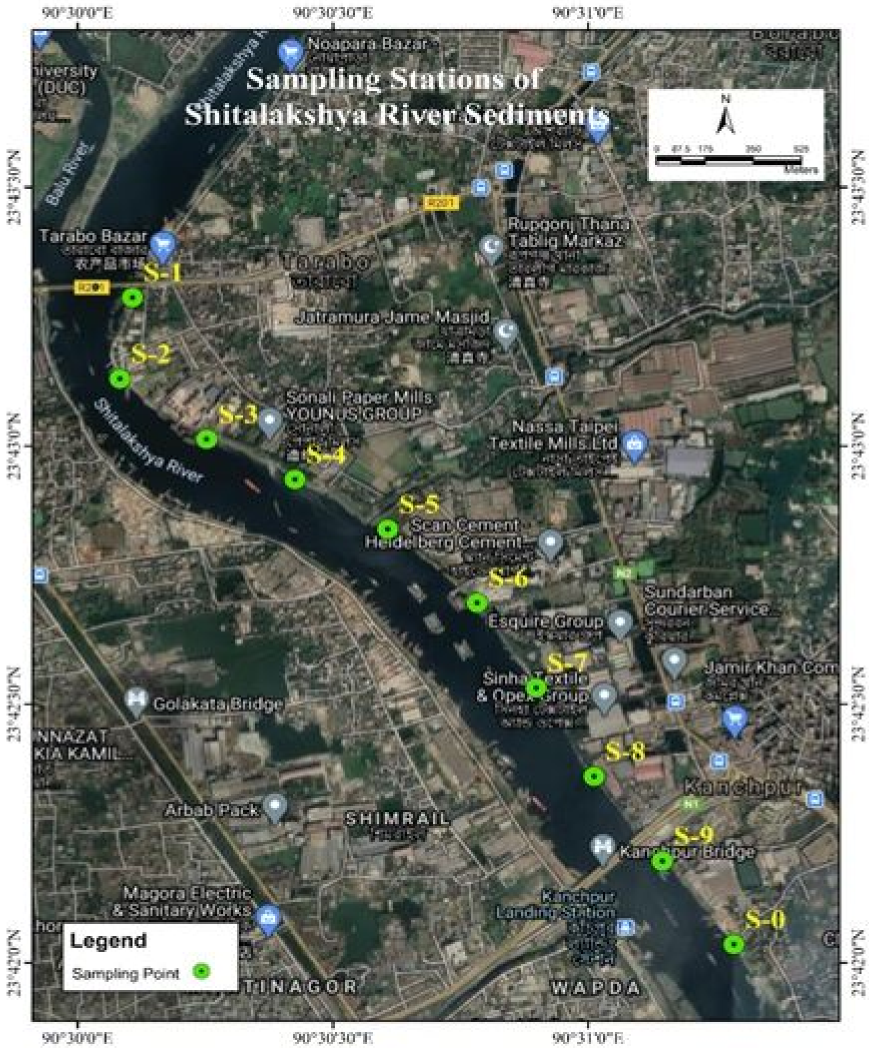
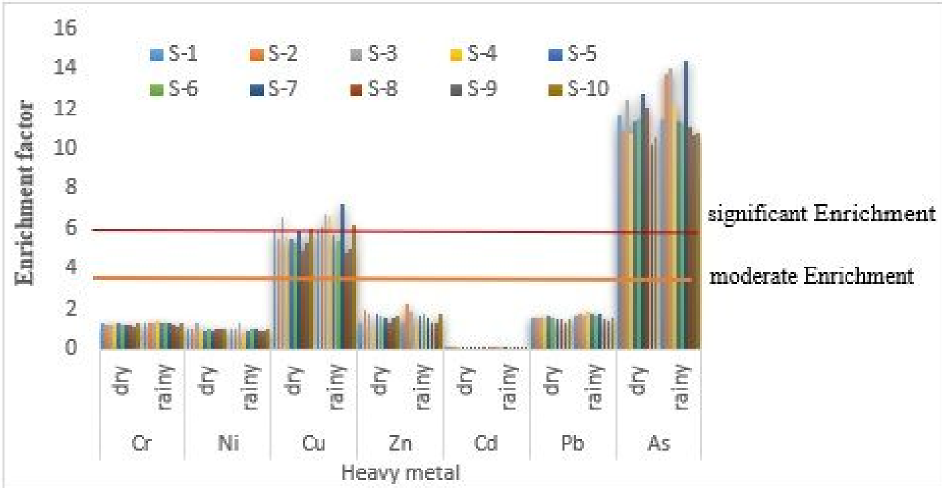
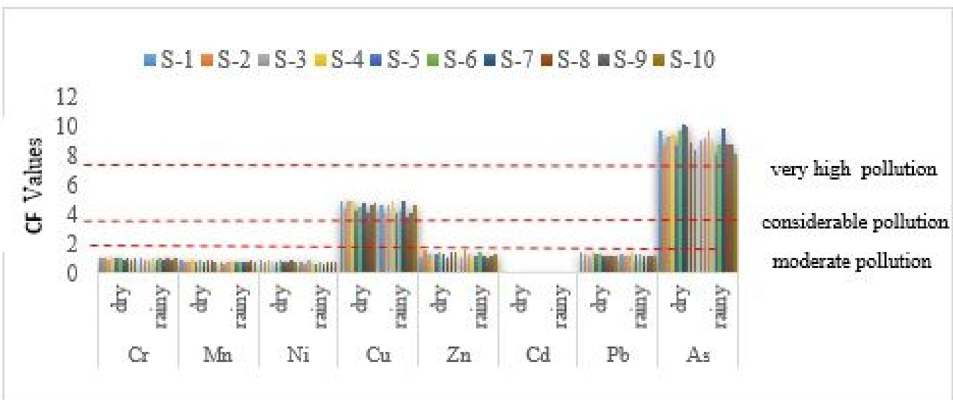


Figure 1

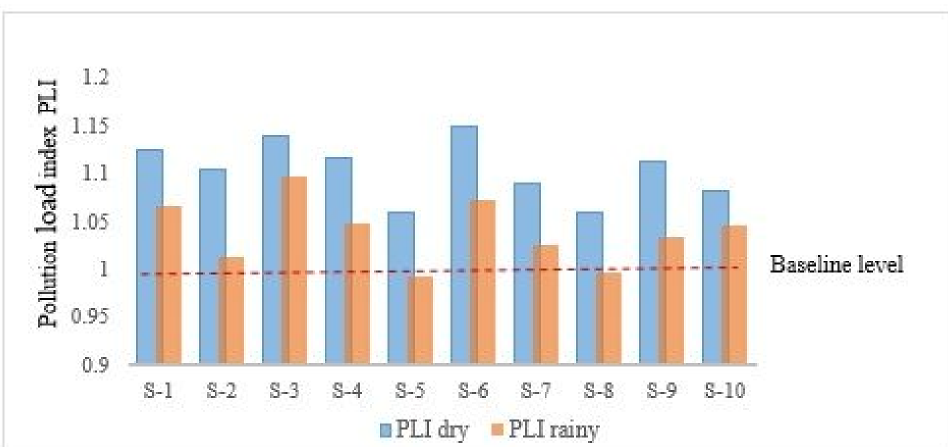
Sampling location Ma



2(a) Enrichment Factor



2(b) Contamination Factor



2(c) Pollution load Index

Figure 2

2(a), 2(b) and 2(c) shows variation of Enrichment factor (EF) values, Contamination factor (CF) and Pollution load index (PLI) of studied .heavy metals in both seasons.

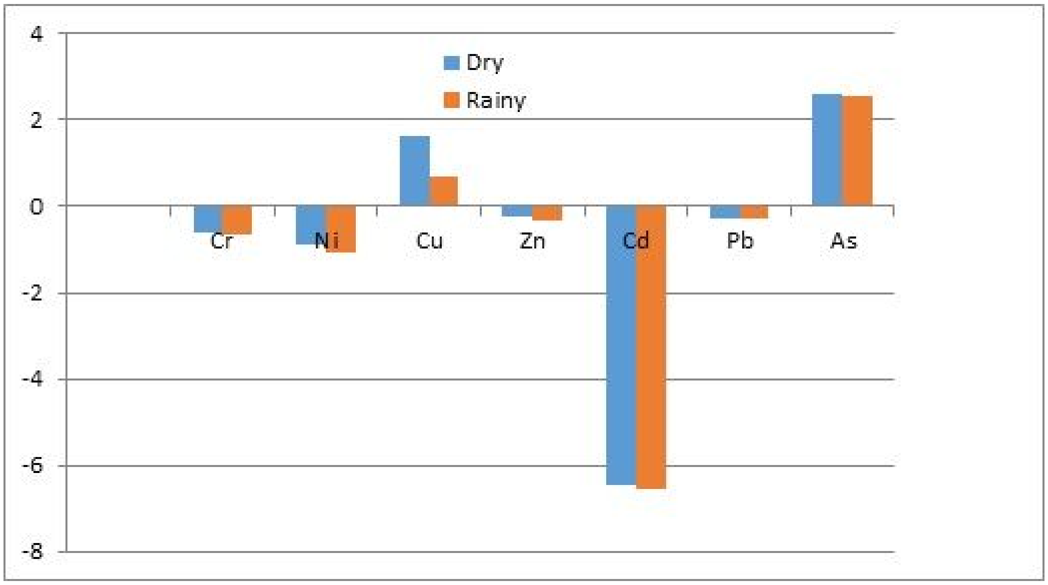


Figure 3

Geoaccumulation Index (I geo) for different elements for different sampling site of Shitalakshya river sediment

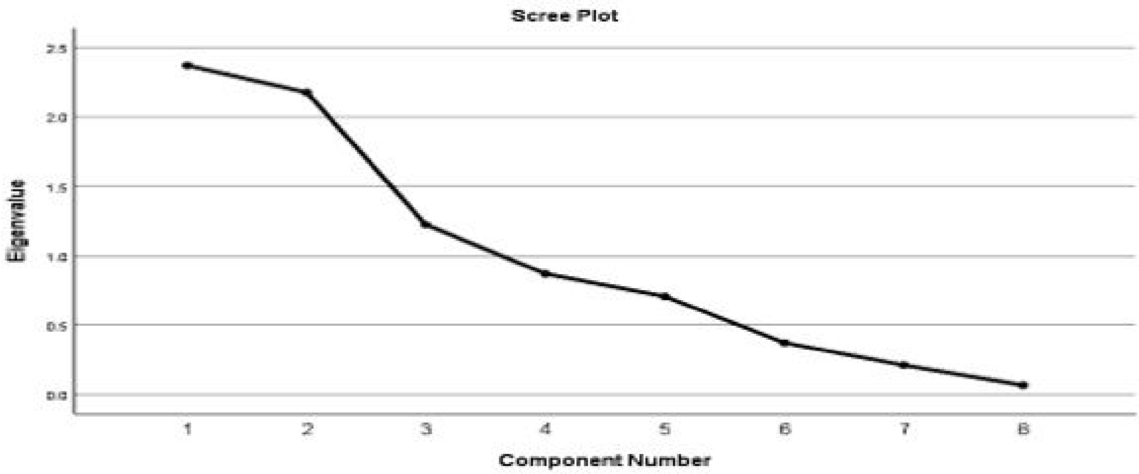


Fig. 4a

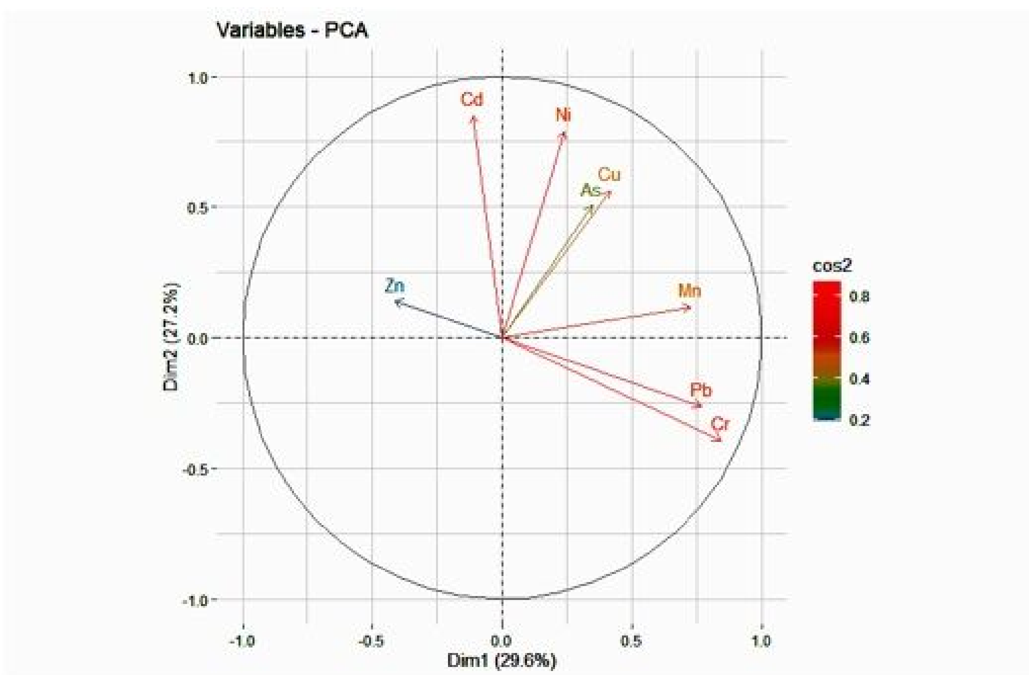


Fig. 4b

Figure 4

Scree plot (a) and variable PCA plot (b) for the heavy metals data set

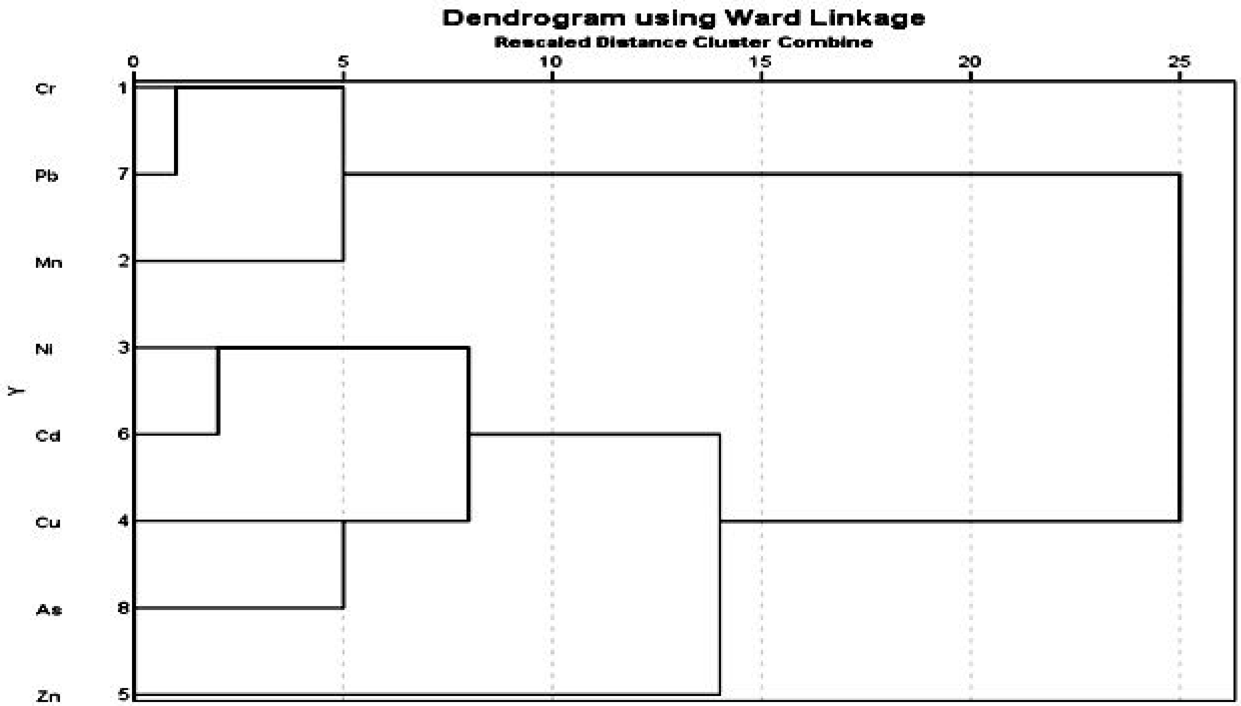


Figure 5

Dendrogram showing clustering elements in the studied sediment of Shitalakshya river

Supplementary Files

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- tables.pdf