Distribution, source identification and potential ecological risk of heavy metals in surface sediments of the Mongla port area, Bangladesh

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ABSTRACT
This study comprehensively investigated the distribution, source identification, and potential ecological risk of heavy metals (As, Ni, Pb, Cd, Cr, Cu, and Zn) in surface sediments of the Mongla port area of Bangladesh. The sediment samples were collected from ten sampling points during the wet and dry season and heavy metals were analyzed by atomic absorption spectrophotometer. The average concentration of heavy metal in sediments was higher during the dry season (11.36 to 118.55 mg/kg) as compared with the wet season (8.70 to 88.58 mg/kg). Certain indices, including the enrichment factor, geoaccumulation index, and contamination factor demonstrated that Cd was highly responsible for sediment pollution. The range of pollution load index values was 1.12 to 1.55 during wet and 1.59 to 2.27 during the dry season, indicating progressive deterioration of sediments by studied heavy metals. Multivariate principal component analysis showed that the source of As, Cd, Cr, Ni, Cu and, Zn in the port area surface sediment mainly due to anthropogenic activities. Considering the potential ecological risk, surface sediments of the Mongla port area showed moderate to considerable ecological risk. Potential acute toxicity results in most of the sampling points during the dry season was greater than 4, shows moderate to the severe toxicity of heavy metals in sediment. Further studies to assess the widespread risk of heavy metals are recommended to protect the aquatic ecosystem from heavy metal contamination.

1. Introduction
Port area plays a significant role in global trade and the national economic progress of a country. It is considered as the midpoint of all types of transport networks and also provides vigorous opportunities for developing industrial, commercial, and recreation activities (Caglak et al. 2011, Birch 2017a). However, the port area creates environmental problems in their adjacent ecosystem by releasing wastewater, oil spilling, leaking of storing hazardous materials, waste dumping, ship painting, dredging of sediment, emitting of air pollutants, generating noise, etc. (UNCTAD 2015, Shen et al. 2017). Recently, heavy metal pollution in the port environment has been received more attention due to their sources availability, abundances, lower degradation rate, high accumulation rate, and potential eco-toxic character (Ali et al. 2016, Islam et al. 2015, Rahman et al. 2019). Larger quantities of heavy metals are entering the water bodies from port area activities (Nduka and Orisakwe 2011, Jahan and Strezov 2018) and is distributed throughout the water columns by the influencing of environmental and hydrodynamic factors (e.g. dissolution, sorption, precipitation, exchanges, complexation phenomena, and waves) (Abdel-Ghani and Elchaghaby 2007, Nouri et al. 2011). Primarily, heavy metals become fixed with the suspended particles and finally incorporated into sediments (Hosono et al. 2010, Islam et al. 2015), and the accumulation rate of heavy metals in sediment are influenced by diverse sediment constituents such as texture, mineralogical composition, and pH (Beveridge 1989), that’s why heavy metals concentrations are higher in sediments than water bodies. In the riverine environment, sediment is considered as most important and dynamic part due to its ecological significance for aquatic habitat- acts as feeding and breeding place (Haris and Aris 2015), so heavy metals pollution in
sediment is harmful to aquatic ecosystem (Bellas et al. 2008, Chen et al. 2016) because it hampers the growth and production of the aquatic organism by interrupting their physiological activities (Baker 1981). On the other hand, several studies indicate that elevated level of heavy metals such as Cd, Pb, Ni, Cr, As, Zn, Cu, Hg, Se, and Al in sediments have been proven as toxicants for aquatic life and in consequence of that pose a threat to maintaining ecosystem equilibrium (Wennberg 1994, Komoroske et al. 2011, Yi et al. 2011). Furthermore, heavy metals could be transferred from lower to higher-order trophic levels in a food chain through bioaccumulation and finally creating acute and chronic diseases in humans and animals (Sun et al. 2001, Al-Busaidi et al. 2011, Rahman et al. 2012, Fu et al. 2014). In sediment, heavy metals are existing in different chemical forms and their speciations go to numerous forms due to chemical weathering, adsorption, desorption, and diffusion during transportation throughout the aquatic environment, which influence their behavior and bioavailability (Reimann and de Caritat 2005, Marchand et al. 2006, Chen et al. 2018).

Due to numerous resuspension and deposition processes, surface sediment familiar as a sink of heavy metals starts releasing heavy metals and act as a source (Guevara et al. 2005, Chen et al. 2019). Therefore, regular monitoring and risk assessment of heavy metal pollution in the aquatic ecosystem is vital for taking defensive action before possibly irremediable harmful effects occur. Usually, water and biological samples are considered as favored media for the assessment of environmental and ecological health of aquatic ecosystem but water samples have some limitations due to high dynamic characteristics and sampling cost, lower concentration in sample create confusion about data quality and interpretation (Birch and Taylor 2000, Birch and Olmos 2008). Conversely, biological samples (e.g. flora and fauna) often fail to provide significant results because the concentration and effects of pollutant are varied from species to species due to distinct properties (e.g. nature of the chemical, age, gender, dose-time relationship, exposure route, metabolism rate, etc.) (Birch 2017b). Depending on heavy metals concentration, bioavailability and toxicity in surface sediment, different types of environmental risk assessment indexes such as enrichment factor (EF), contamination factor (CF), geoaccumulation index ($I_{\text{geo}}$), pollution load index (PLI), toxicological unit (TU) and potential ecological risk index (PERI), etc. have been developed (Yang et al. 2009, Zhang et al. 2013, Ali et al. 2016). In this aspect, sediment sampling is a quick, cost-effective, reliable, and larger information-carrying method to monitor the condition of aquatic ecosystems with respect to other methods (water and biological sample) because it’s a wide variety of habitats, storage bulk contaminants and substantially provides environmental and geochemical speciation (Maher et al. 1999, Birch et al. 2000, Uluturhan et al. 2011). Additionally, sediment analysis results not only provide the present status of ecosystem health but also projected the future changes of the riverine environment with time processes (Birch et al. 2000, Birch 2017b).

In this study, the Mongla port area was chosen as the study area due to its ecological significance of being surrounded by Sundarban mangrove forest and also for playing an important role as a commercial port, industrial center, fishing, tourism, and recreational area. Pasur and Mongla River are the important rivers in the port area and distributary of the Ganges tidal floodplain area (Ali et al. 2018). The port area adjacent rivers meet the Shibisa River within the Sundarban mangrove forests. Sundarban is the largest mangrove forest in the world and it has extensive ecological and economic importance (Kumar et al. 2016). Port activities gradually degrade their water and sediment quality. Consequently, there is a necessity to monitor the dispersal of heavy metals concentration in sediment, so that it could be compared with non-polluted background value as well as to assess the environmental quality and ecological health risk for this area. But unfortunately, the information on heavy metal pollution in the Mongla port area is inadequate. This study will help to fill the gap in knowledge as an effective scientific record on the aquatic ecosystem pollution by heavy metals in the Mongla port area sediment, Bangladesh. The objectives of this study were to determine the concentration of heavy metals (As, Ni, Pb, Cd, Cr, Cu, and Zn) in the surface sediments, as well as to the assess distribution, source, and potential ecological risk of the heavy metals in the Mongla port area.

2. Materials and methods

2.1. Description of the study area

This study was conducted in the Mongla port area (Figure 1), close to the Sundarbans and Bay of Bengal, Bangladesh. This is the second-largest seaport, located on the east bank of the Pasur River near its confluence with the Mongla Nulla, in the southwest part of Bangladesh. The average temperature is ranged from 19–31 °C and rainfall 1290.5 mm. The port also hosts the Mongla export processing zone (EPZ), mainly used for industrial, commercial, residential, and recreational purposes. The port area continuously received
untreated wastes and effluents from the EPZ area and port activities, which may have an extensive impact on the aquatic ecosystem in the study area.

2.2. Sediment sample collection, preparation and metal extraction

In this study, surface sediment samples of the Mongla port area in the Pasur and Mongla River were collected during the dry and wet season in 2019 from 10 sampling points (Figure 1). At every point, three composite sediment samples were collected by following standard procedure (USEPA 1999), where the river bed surface sediment samples were collected at a depth of 0 to 5 cm using an Ekman grab sampler (56 x 22 x 40 cm). Finally, the study samples were taken from the upper 2 cm of collecting composite sediment mixing pile with acid-washed (10% HNO₃) nonmetallic sampling bags to avoid any further metallic contamination. Then the collected samples were dried in an oven at 80°C for 24 h, grounded using mortar and pestle, and sieved (2 mm). Finally, the samples were stored in borosilicate airtight glass bottles for chemical analysis. All standard solutions for studied elements and other acids and chemicals for solution preparation were analytical grade received from Merck Germany, which was used throughout all the experiments. For the determination of heavy metals, the whole sediment digestion process was conducted by using modified USEPA Method 3050B (USEPA 1996); briefly, 2.0 g of each sediment sample was successively digested by using concentrated 10 ml HNO₃ (69%), 5 ml H₂SO₄ (98%), 5 ml HClO₄ (70%) and 3 ml H₂O₂ (30%). After completion of digestion, solutions were filtered by Whatman no. 41 filter paper (pre-washed with 0.1 M HNO₃) and made the solution final volume 100 ml with double-distilled water.

2.3. Analytical methods for physicochemical parameters

The physicochemical parameters (pH, electrical conductivity, % organic matter, and % organic carbon) of
sediment samples were determined. For pH measurement, sediment and double-distilled water were mixed at 1:2.5 ratios, kept overnight, and then pH was measured using a pH meter (MARTINI instruments, pH 56 pHWP, USA). In case of electrical conductivity (EC) determination, 5.0 g of dried sediment was taken in 50 ml beaker, then added 30 ml of double-distilled water and mixed it properly for 5 min, and EC was determined by using an EC meter (HACH Sension −156; multi-parameter, USA). Percentage of Organic matter (OM) and organic carbon (OC) were analyzed following Walkley and Black (1934) manual titration procedure.

2.4. Chemical analysis and quality assurance

The heavy metals in the samples were analyzed by atomic absorption spectrophotometer (AAS) (Model: AA-7000, Shimadzu, Japan.) using flame and hydride generator (HG) methods. The heavy metals measurements and analytical conditions for AAS, given in Table S1. The instrument calibration was done by using the standard solution (1,000 ppm), pursued from Sigma Aldrich, USA. All laboratory equipment and glassware were cleaned (i.e. used 20% HNO₃ acid for cleaning purposes and frequently washed with double distilled water and oven-dried before use). Before starting the analysis, a blank sample was run and validated; duplicate analyzed was performed of every sample and the average value was used. The relationship between the certified values (0.53–1.77%) and recovery values (93–100%) confirmed that the instrument analysis results are accurate and precise, presented in Table S2.

2.5. Evaluating the contamination level of heavy metals

In this study, contamination level is evaluated by sediment quality indices such as the enrichment factor (EF), geo-accumulation index (Igeo), contamination factor (CF), and pollution load index (PLI). EF is applied to assess the existence of heavy metals in sediments by anthropogenic influence or not. The EF method evaluates the degree of contaminants in the environment corresponding with a reference metal, such as Fe, Sc, or Al (Dickinson et al. 1996, Boës et al. 2011). In this study, Fe was chosen as the reference metal for its governing existence, which helps to know the variations of metals with respect to this metal. EF was calculated by using the following Equation (1).

\[ EF = \left( \frac{C_{xi}}{M_{xi}} \right)_{\text{sample}} / \left( \frac{C_{xi}}{M_{xi}} \right)_{\text{background}} \]  

where \( C_{xi} \) is the concentration of studied metal of \( i \)th parameter, \( M_{xi} \) is the reference metal of \( i \)th parameter, and \( \left( \frac{C_{xi}}{M_{xi}} \right)_{\text{sample}} \) and \( \left( \frac{C_{xi}}{M_{xi}} \right)_{\text{background}} \) are the ratios of the studied metal and the reference metal in the surface and background sediments, respectively. EF values are categories into five groups as proposed by Sutherland (2000); EF value <2.0, 2.0 to 5.0, 5.0 to 20.0, 20.0 to 40.0, and >40 represented minimal enrichments, moderate enrichment, significantly enriched, very highly enriched, and extremely enriched, respectively. Usually, an EF value less than 1.5 indicates crustal or natural origin, whereas an EF value higher than 1.5 indicates non-crustal or anthropogenic influences (Jahan and Strezov 2018).

\( I_{\text{geo}} \) is another indicator that measures the level of contamination in sediments (Muller 1969) and is expressed as the following Equation (2).

\[ I_{\text{geo}} = \log_2 \left( \frac{C_{xi}}{1.5 \times B_{xi}} \right) \]  

where \( C_{xi} \) is the studied metal concentration of \( i \)th parameter, \( B_{xi} \) is the geochemical background value of the heavy metal of \( i \)th parameter (i.e. representative of pre-industrial reference level of trace metals) and factor 1.5 is presented to reduce the probable differences in the background values due to lithogenic effects. Muller (1969) classified sediments according to the \( I_{\text{geo}} \) values, as extremely contaminated if \( I_{\text{geo}} >5 \), strongly to extremely contaminated if \( I_{\text{geo}} = 4–5 \), strongly contaminated if \( I_{\text{geo}} = 3–4 \), moderately to strongly contaminated if \( I_{\text{geo}} = 2–3 \), moderately contaminated if \( I_{\text{geo}} = 1–2 \) uncontaminated to moderately contaminated if \( I_{\text{geo}} = 0–1 \), and uncontaminated if \( I_{\text{geo}} \leq 0 \).

\( CF_i \) is the ratio of the individual metal concentration and its background value (Hakanson 1980), and it was calculated by using Equation (3).

\[ CF_i = \frac{C_i}{C_{xi}} \]  

\[ C_d = \sum_{i=1}^{n} CF_i \]  

where, \( C_i \) is the concentration of studied metal of \( i \)th parameter and \( C_{xi} \) is the background value of the studied metal of \( i \)th parameter. Background values of selected heavy metals for this study were taken from the preindustrial sediment samples of the study area in Bangladesh. The \( CF_i \) are categorized into four groups for observing the pollution of individual metal during a timeframe; \( (CF_i < 1) \), \( (1 \leq CF_i < 3) \), \( (3 \leq CF_i < 6) \)
and \((CF_i \geq 6)\); indicates a low, moderate, considerable and high degree of contamination, respectively (Hakanson 1980, Luo et al. 2007). However, \(C_d\) is the degree of contamination, i.e. combined effect of all individual metals in an individual point (Hakanson 1980), calculated by using Equation (4). It represents the quality of environment based on \(C_d\) values; \((C_d < 5)\) low; \((5 \leq C_d < 10)\) moderate; \((10 \leq C_d < 20)\) considerable; and \((C_d \geq 20)\) high degree of contamination (modified by Luo et al. 2007 and the original proposed by Hakanson 1980).

\(PLI\) is a combined approach for assessing sediment quality. The \(PLI\) is defined as the geometric mean of metals contamination factor (Tomlinson et al. 1980), presented in Equation (5).

\[
PLI = \sqrt[n]{CF_{i1} \times CF_{i2} \times CF_{i3} \ldots \ldots \times CF_{in}} \quad (5)
\]

where, \(CF_i\) is the individual metal pollution factor of \(i^{th}\) parameter and \(n\) is the number of studied heavy metals. Therefore, \(PLI\) values; \(<1\), \(1\), \(>1\), demonstrate no, baseline, and continuous degradation of the sediment quality, respectively (Tomlinson et al. 1980). This index has developed without accounting for the lithogenic and sedimentary inputs of metals (Jahan and Strezov 2018).

### 2.6. Assessment of potential ecological risk

In this study, sediment quality guidelines (SQGs), potential ecological risk index (\(PERI\)), and summation of toxic units (\(\sum TU\)) were used to measure heavy metal-associated ecological risk on the aquatic biota. For this purpose, two sets of SQGs: probable effect level (PEL) and threshold effect level (TEL); additionally, lowest effect level (LEL) and severe effect level (SEL), were used (NYSDEC 1999, MacDonald et al. 2000), detail description regarding these SQGs given in Table S3. The heavy metals concentrations were used for assessing the sediment grade as well as their risk on aquatic biota.

\(PERI\) is also presented to measure the level of heavy metal pollution in sediments. \(PERI\) was proposed by Hakanson (1980) and is calculated with the following Equation (6).

\[
E_i^j = T_i^j \times CF_i, \quad PERI = \sum_{i=1}^{n} E_i^j \quad (6)
\]

where, \(E_i^j\) is the single metal ecological risk index of \(i^{th}\) parameter. \(T_i^j\) is the biological toxic factor of \(i^{th}\) parameter. The study used the \(T_i^j\) value of As, Ni, Pb, Cd, Cr, Cu and Zn were 10, 6, 5, 30, 2, 5, and 1, respectively. (Hakanson 1980). \(PERI\) is the summation of \(E_i^j\), showing the overall ecological risk. \(E_i^j\) was classified into the following five groups: \(E_i^j < 40;\) \(40 \leq E_i^j < 80;\) \(80 \leq E_i^j < 160;\) \(160 \leq E_i^j < 320\) and \(E_i^j \geq 320\); indicates low, moderate, considerable, high and very high risk (Hakanson 1980), and \(PERI\) categories into following groups: \((PERI < 65)\) low; \((65 \leq PERI < 130)\) moderate; \((130 \leq PERI < 260)\) considerable and \((PERI \geq 260)\) very high (modified by Luo et al. 2007 and the original proposed by Hakanson 1980). \(PERI\) not only shows the sensitivity of aquatic organisms but also the level of risk with respect to the degree of contamination.

This study also used \(\sum TU\) for assessing the potential acute toxicity of studied heavy metals. \(TU\) is the ratio of the individual heavy metal concentration and its associated \(PEL_i\) value (Pedersen et al. 1998). The \(TU\) for all metals was calculated by using the following Equation (7).

\[
TU = \frac{C_i}{PEL_i} \quad (7)
\]

where \(C_i\) is the studied heavy metal concentration of \(i^{th}\) parameter in sediment and \(PEL_i\) is the studied heavy metal of \(i^{th}\) parameter associated value, presented as As = 17, Ni = 36, Pb = 91, Cd = 3.5, Cr = 90, Cu = 197, and Zn = 315 (MacDonald et al. 2000). The summation of the toxic unit (\(\sum TU\)) was calculated by using Equation (8).

\[
\sum TU = \sum_{i=1}^{n} \frac{C_i}{PEL_i} \quad (8)
\]

where \(\sum TU\) is the summation of the toxic unit of all heavy metal.

### 2.7. Statistical analysis

The multivariate statistical tools; Pearson’s correlation matrix (PCM), principal component analysis (PCA) and cluster analysis (CA) were applied to find the probable sources of heavy metals in the sediments. For PCA, the mean value of each variable was used, where eigenvalue > 1 and loading value > 0.5 for each principal component was taken from the analysis table for the explanation of the study results. CA was made on the standardized data sets (Z-cores) where the Euclidean distance and Ward method were applied, and the graphically represented dendogram shows the cluster interlink with the studied heavy metal each other as well as sampling points. The data was analyzed by Microsoft Excel-2010 and SPSS (V0.20) software. The Arc GIS (V.10.2) was applied for the spatial distribution of heavy metals in the study sites through the inverse distance weighting (IDW) method.
3. Results and discussion

3.1. Physicochemical characteristics of sediments

Due to differences of topographical and hydrogeological factors in the river-basin area, as well as the changing of local temperature and rainfall, physicochemical parameters and heavy metals may diverge significantly between a catchment area even within short distances (Costa et al. 2001, Wang and Qin 2006). The physicochemical characteristics of sediments are presented in Table S4. The study area sediment was moderately alkaline (pH ranged from 7.54 to 8.44, with an average value of 8.31). EC values in sediment were varied from 408 to 1060 μS/cm with an average value of 666 μS/cm. The organic matters in study site sediments were ranged from 1.01 to 5.4% with an average value of 2.86%. The organic carbon was varied from 0.58 to 3.13% with an average value of 1.57% due to discharging of waste and industrial sewage from the port area.

3.2. Distribution of heavy metals concentration in the sediments of the study area

The concentration ranges of studied heavy metals in the surface sediments of the study area were given as follows: Zn (137.5–87.0 mg/kg); Cr (116.08–73.17 mg/kg); Ni (52.79–31.52 mg/kg); Cu (46.8–35.63 mg/kg); Pb (36.11–22.75 mg/kg); As (13.45–8.60 mg/kg); Cd (1.7–0.95 mg/kg) during wet season and Zn (101.9–71.12 mg/kg); Cr (52.77–39.90 mg/kg); Ni (37.60–17.12 mg/kg); Cu (40.73–32.04 mg/kg); Pb (23.93–18.01); As (8.71–5.72 mg/kg); Cd (0.97–0.46 mg/kg) during dry season (Table 1). The variation of heavy metals may be attributed to changes in hydrodynamic factors and point and non-point sources in the study area. The mean concentration of heavy metals in sediment samples was in the decreasing order of Zn > Cr > Ni > Cu > Pb > As > Cd (Table 1). The dry and wet season scenarios revealed that the concentrations of studied heavy metals were comparatively lower in the wet season (Figure S1) than in the dry season (Figure S2), owing to lower river water flowing rates during the dry season, allowing for the storage of greater amounts of heavy metals in sediment. Heavy metal concentrations in study sites followed the descending order of site S7 > S1 > S6 > S9 > S8 > S4 > S3 > S10 > S5 > S2 (Table 1). Fascinatingly, the descending order of heavy metals in the study sites did not follow a downstream pattern, probably due to the influences of source variability, mineralogical composition and dominating of physicochemical processes such as the variation of organic matter, adsorption, absorption, precipitation and redox reaction, etc. With the exception of Pb, the mean concentration of all studied metals showed higher corresponds to the UCC and TRV as compared to those in other port area sediments around the world (Table S5), and the results showed that the concentrations of Cr, Cu, Ni, Cd, and Zn in this study are equivalent, if not higher, than those in other port area sediments.

3.3. Geochemical indices for sediment contamination assessment

3.3.1. Contamination factor (CF), enrichment factor (EF), geoaccumulation index (Igeo), and pollution load index (PLI)

The CFi and Cd analysis provide the existing level of contamination of the metals in the sediments of the

| Sites | As | Ni | Pb | Cd | Cr | Cu | Zn |
|-------|----|----|----|----|----|----|----|
| S1    | 7.99 | 13.2 | 27.97 | 45.46 | 18.01 | 23.75 | 0.97 | 1.05 | 47.64 | 116.08 | 35.13 | 46.80 | 85.56 | 121.00 |
| S2    | 7.13 | 8.60 | 20.71 | 31.52 | 20.20 | 24.28 | 0.77 | 1.30 | 39.90 | 98.69 | 36.12 | 41.68 | 71.12 | 87.00 |
| S3    | 7.22 | 10.85 | 20.81 | 42.39 | 22.68 | 33.19 | 0.65 | 1.15 | 44.49 | 73.17 | 34.80 | 39.52 | 76.32 | 113.50 |
| S4    | 6.77 | 11.05 | 23.09 | 41.09 | 19.92 | 24.01 | 0.48 | 1.35 | 44.10 | 84.85 | 37.45 | 44.65 | 86.11 | 126.50 |
| S5    | 6.26 | 11.65 | 17.12 | 40.74 | 21.03 | 34.83 | 0.54 | 1.45 | 42.22 | 79.15 | 35.75 | 39.52 | 76.32 | 113.50 |
| S6    | 7.64 | 12.05 | 21.33 | 52.79 | 22.21 | 33.74 | 0.67 | 1.70 | 41.86 | 88.31 | 36.21 | 44.25 | 85.97 | 137.50 |
| S7    | 8.13 | 13.45 | 37.66 | 51.19 | 20.90 | 31.01 | 0.67 | 1.30 | 47.58 | 84.95 | 40.73 | 44.65 | 101.9 | 132.00 |
| S8    | 7.83 | 10.80 | 23.39 | 39.08 | 23.93 | 36.11 | 0.53 | 1.20 | 52.76 | 91.30 | 34.49 | 35.63 | 91.68 | 114.50 |
| S9    | 8.71 | 11.55 | 25.67 | 43.69 | 22.27 | 29.02 | 0.63 | 0.95 | 45.27 | 85.23 | 34.26 | 41.59 | 98.21 | 119.00 |
| S10   | 5.72 | 10.40 | 24.88 | 42.86 | 19.92 | 34.28 | 0.47 | 1.10 | 42.32 | 79.99 | 32.04 | 39.32 | 94.33 | 119.00 |
| Maximum | 8.71 | 13.45 | 37.66 | 52.79 | 23.93 | 36.11 | 0.97 | 1.70 | 52.77 | 116.08 | 40.73 | 46.80 | 101.90 | 137.50 |
| Minimum | 5.72 | 8.60 | 17.12 | 31.52 | 18.01 | 22.75 | 0.46 | 0.95 | 39.90 | 73.17 | 32.04 | 35.63 | 71.12 | 87.00 |
| Mean  | 7.80 | 11.36 | 23.83 | 43.08 | 21.11 | 30.32 | 0.64 | 1.25 | 45.02 | 88.17 | 35.70 | 41.00 | 88.58 | 118.55 |
| SD    | 0.92 | 1.39 | 5.71 | 6.01 | 1.71 | 5.00 | 0.15 | 0.22 | 3.63 | 12.03 | 2.28 | 3.49 | 10.40 | 13.56 |

Abbreviation: SD: standard deviation.
study area. The \( CF_i \) indicated that the individual assessment of metal contamination, whereas \( C_d \) provides a complete evaluation of metal contamination for all localities. The \( CF_i \) values of all heavy metals in the study sites were \( 1 \leq CF_i < 3 \), indicating a moderate degree of contamination, while \( C_d \) (\( CF_i > 3 \)) displayed a considerable degree of contamination during the dry season (Figure 2(a)). However, the overall \( CF_i \) values for all heavy metals in study sites were found as decreasing order of \( Cd > As > Ni > Cr > Zn > Pb > Cu \) (Figure 2(a)).

The \( C_d \) values in the study sites were moderate contamination class (ranges from 8.01–10.59 with average value 9.11) during the wet season and considerable contamination class (ranges from 11.4–17.80 with average value 14.92) during the dry season for all sampling sites (Figure 2(b)), this can be ascribed to the discharging of industrial and urban wastewater, and dumping of waste materials over time.

The mean \( EF \) values of \( Cd \), \( As \), \( Ni \), \( Cr \), \( Zn \), \( Pb \), and \( Cu \) were 6.65, 4.85, 3.78, 3.76, 3.68, 3.58 and 3.40 during the wet season (Figure S3(a)) as well as 10.85, 7.03, 6.03, 5.91, 4.89, 4.24 and 3.66 during the dry season, respectively (Figure S3(b)), indicating that the sediment in the study area was moderately enriched with \( As \), \( Ni \), \( Cr \), \( Zn \), \( Pb \) and \( Cu \) (\( EF > 2–5 \)) and considerably enriched with \( Cd \) (\( EF > 5–20 \)) in the wet season, whereas considerably enriched with \( Cd \), \( As \), \( Ni \) and \( Cr \) (\( EF > 5–20 \)) and moderately enriched with \( Zn \), \( Pb \) and \( Cu \) (\( EF > 2–5 \)) in the dry season. The mean values of \( EF \) for all studied heavy metal was greater than 1.5, indicating that these metals perhaps originated from anthropogenic activities (e.g. human-induced pollution). However, \( Cd \) shows the highest \( EF \) values for both the dry and wet seasons in sediment samples (Figure S3(a,b)) might be due to higher accumulation by human perturbation.

![Figure 2.](image)

**Figure 2.** (a) \( CF_i \) values and (b) \( C_d \) values of heavy metals in surface sediments collected from the Mongla port area, Bangladesh.

| Sites | Season | As  | Ni  | Cd  | Pb  | Cr  | Cu  | Zn  | Risk index (PERI) | Risk level |
|-------|--------|-----|-----|-----|-----|-----|-----|-----|-----------------|-----------|
| S1    | Wet    | 16.30 | 8.39 | 97.45 | 4.50 | 2.32 | 5.01 | 1.20 | 135.17          | Considerable |
|       | Dry    | 26.93 | 13.63 | 105.00 | 5.68 | 5.66 | 6.68 | 1.70 | 165.28          | Considerable |
| S2    | Wet    | 14.55 | 6.21 | 77.11 | 5.05 | 2.09 | 5.16 | 1.00 | 111.17          | Moderate   |
|       | Dry    | 17.55 | 9.45 | 130.00 | 6.07 | 4.81 | 3.09 | 1.22 | 172.19          | Considerable |
| S3    | Wet    | 14.74 | 6.24 | 64.77 | 5.67 | 2.17 | 4.97 | 1.39 | 99.95           | Moderate   |
|       | Dry    | 22.14 | 12.71 | 115.00 | 8.29 | 3.56 | 5.27 | 1.62 | 168.59          | Considerable |
| S4    | Wet    | 13.78 | 6.92 | 47.57 | 4.98 | 2.15 | 5.35 | 1.24 | 81.99           | Moderate   |
|       | Dry    | 22.55 | 12.32 | 135.00 | 6.00 | 4.13 | 5.46 | 1.78 | 187.24          | Considerable |
| S5    | Wet    | 12.77 | 5.13 | 53.79 | 5.25 | 2.15 | 5.10 | 1.07 | 85.26           | Moderate   |
|       | Dry    | 23.77 | 12.22 | 145.00 | 8.70 | 3.86 | 5.64 | 1.59 | 200.78          | Considerable |
| S6    | Wet    | 13.75 | 6.40 | 66.90 | 5.55 | 2.04 | 5.17 | 1.11 | 100.92          | Moderate   |
|       | Dry    | 24.59 | 15.83 | 170.00 | 8.43 | 4.30 | 6.32 | 1.93 | 231.40          | Considerable |
| S7    | Wet    | 16.58 | 11.29 | 67.34 | 5.22 | 2.32 | 5.81 | 1.43 | 109.99          | Moderate   |
|       | Dry    | 27.44 | 15.35 | 130.00 | 7.75 | 4.14 | 6.37 | 1.85 | 192.90          | Considerable |
| S8    | Wet    | 15.98 | 7.01 | 52.52 | 5.98 | 2.57 | 4.92 | 1.29 | 90.27           | Moderate   |
|       | Dry    | 22.04 | 11.72 | 120.00 | 9.02 | 4.45 | 5.09 | 1.61 | 173.93          | Considerable |
| S9    | Wet    | 17.77 | 7.70 | 62.54 | 5.56 | 2.20 | 4.89 | 1.38 | 102.04          | Moderate   |
|       | Dry    | 23.57 | 13.10 | 95.00 | 7.25 | 4.15 | 5.94 | 1.67 | 150.68          | Considerable |
| S10   | Wet    | 11.67 | 6.14 | 46.51 | 4.98 | 2.06 | 4.57 | 1.32 | 77.25           | Moderate   |
|       | Dry    | 21.22 | 12.85 | 110.00 | 8.57 | 3.90 | 5.61 | 1.67 | 163.82          | Considerable |

**Table 2.** \( E'_f \) and PERI values of heavy metals in surface sediments collected from the Mongla port area, Bangladesh.
The mean $I_{\text{geo}}$ value for Cd in the wet season was 0.46 indicates uncontaminated to moderately contamination ($I_{\text{geo}} > 0$) and 1.46 in the dry season shows moderately contamination ($I_{\text{geo}} > 1$) of sediment due to a higher concentration of Cd in sediments than geochemical background values (Table S6). The major contributors for maximum cadmium values are atmospheric pollution, effluents, and waste materials from the port region. Conversely, $I_{\text{geo}}$ values for the rest of the other studies metals shows uncontamination ($I_{\text{geo}} < 0$) of sediments in the study sites during the wet season while As, Ni Cr and Zn exhibit uncontaminated to moderately contamination ($I_{\text{geo}} > 0$) during the dry season (Table S6).

$PLI$ was applied to evaluate the quality of the aquatic environment and ecosystem health with respect to sediment pollution status (Ali et al. 2016). $PLI$ values were ranged from 1.12 to 1.55 during the wet season with an average of 1.26; and 1.59 to 2.27 during the dry season with an average of 1.97, suggesting that the sediment of all sampling sites were noticeably polluted ($PLI > 1$), (Figure 3). Comparison between both seasons indicated that $PLI$ values of the dry season were higher than the wet season due to the higher storage of heavy metals in sediments. The overall assessment results indicated that the investigated area is being polluted and which have real threats to this susceptible aquatic ecosystem.

### 3.4. Potential ecological risk assessment

According to $PERI$ assessment results, the $E_i^j$ value for the single metal in all sites was found to be the following descending order of $\text{Cd} > \text{As} > \text{Ni} > \text{Pb} > \text{Cu} > \text{Cr} > \text{Zn}$ (Table 2). Among the analyzed metals, Cd showed moderate to considerable ecological risk with $E_i^j$ values ranging from 46.51–170.00 with a mean value of 94.57 (Table 2). The source of Cd in the sediments could be due to the discharging of oily effluents and wastes from the industries in the port area and land-based runoff into the river. The $E_i^j$ values for other studies of heavy metals (As, Ni, Pb, Cu, Cr, and Zn) show low ecological risk due to their lower toxic response factor ($T_i^j$). However, the $PERI$ values in the sampling sites were 77.25 to 231.40, suggesting that there was moderate to considerable potential ecological risk in all sampling sites (Table 2). Besides, SQGs assessment process and analysis results are present in Table S3 and Table S7, respectively. According to the TEL and PEL SQGs, any sampling site is considered contaminated if 10% values exceed TEL. During wet season As, Ni, Cd, Cr, and Cu exceed the 10% values of TEL. For As, 90%; Ni, 80%; Cd, 60%; Cr, 100%, and Cu, 40% of the sample have values between TEL and PEL; while only Ni shows 10% values exceed the PEL (Table S7). Whereas, based on LEL and SEL SQGs, heavy metals between LEL and SEL were following As (90%), Ni (90%), Cd (60%), Cr (100%), and Cu (100%) (Table S7). Additionally, With the exception of Pb, the concentration of all studied metals showed more than 10% values above TEL during the wet season. The concentration of As (100%), Ni (10%), Cd (100%), Pb (10%), Cr (70%), Cu (90%) and Zn (30%) has between the TEL and PEL values, while Ni (90%) and Cr (30%) shows values above PEL (Table S7). With respect to LEL and SEL SQGs, heavy metals between LEL and SEL were As (100%), Ni (80%), Cd (100%), Pb (60%), Cr (100%), Cu (100%) and Zn (40%), while only 20% of Ni values above the SEL (Table S7). Therefore, as per two sets of SQGs, the comparative results between two season indicated that As, Cd, Cr, Cu, and Ni were major concerning metals which might adversely affect the aquatic community.

Furthermore, this study tried to measure the potential acute toxicity of heavy metals on the aquatic organisms by calculating $TU$ and $\Sigma TU$, presented in Figure S4. Toxic units of heavy metals in the study sites followed the decreased order of $\text{Ni} > \text{Cr} > \text{As} > \text{Zn} > \text{Pb} > \text{Cd} > \text{Cu}$ during the wet season (Figure S4(a)) and $\text{Ni} > \text{Cr} > \text{As} > \text{Zn} > \text{Cd} > \text{Pb} > \text{Cu}$ during the dry season (Figure S4(b)). Figure S4 indicates two scenarios, where only one site (S7) in the wet season and majority of sites (S1, S4, S5, S6, S7, S8, and S9) in dry season exceeded the $\Sigma TU$ reference value ($\Sigma TU > 4$) due to higher accumulation and contamination, demonstrating that these areas sediment-dwelling fauna have moderate to the serious toxic effect of studied heavy metals.

### 3.5. Sources of heavy metals in the study area sediments

The multivariate analyses (PCA, CA, and PCM) are applied to know the source, distribution and
movement of studied heavy metal in the port area environment (Figures 4 and 5(a,b); Table S8). This study successively used PCA, CA, and PCM for precisely finding of studied heavy metal sources. PCA was executed based on the average concentrations of studied heavy metals with varimax rotation. The values of Kaiser-Meyer-Olkin and Bartlett were 0.291 and 45.658 (df = 21, p = 0.001), respectively (Table S9). The PCA results indicated that the variability of heavy metals could be represented by two principal components that cumulatively explained 74.769% of the total variance in the sediment (Figure 4). The first principal component (PC1) contributed 45.380% of the total variance and loading with As, Ni, and Zn (Figure 4), which also located in the same cluster (Figure 5(b)) and represent a significant positive correlation among them \( r^2 (\text{As–Ni} = 0.83), r^2 (\text{As–Zn} = 0.62) \) and \( r^2 (\text{Ni–Zn} = 0.79); \) Table S8. The sources of As, Ni and Zn in the study sites may be coming by direct discharging of untreated wastes from the residential area and several industries such as storage batteries, electroplating, varnish cosmetics and especially using of copper arsenate, arsenic sulfide for wood processing plant as well as surface runoff from agricultural land due to excess uses of As rich pesticides. The PC2 component accounted for 29.389% of the total variance
and showed positive loading for Cd, Cu and Cr \( \left( r^2 \text{ (Cd–Cu) = 0.62}, r^2 \text{ (Cr–Cu) = 0.38} \text{ and } r^2 \text{ (Cd–Cr) = 0.23}) \right) \) (Figure 4 and Table S8). The location of Cd–Cu–Cr in the same cluster shows their common source and existence (Figure 5(b)). The sources of Cd, Cu and Cr are possibly industrial activities such as sewage from dyeing industries, vehicle and coal combustion emissions from the traffic of port area, leachates from defused Cd batteries and Cd-plated items. On the other hand, the negative correlation of Pb with other studied heavy metals (Table S8) and individual position in the PCA and CA (Figures 4 and 5(b)) suggest that the Pb had a distinct geochemical or industrial source (e.g. Pb-gasoline, shipping paint, chemical or steelworks). Though some variations are found in multivariate analysis it represents a good agreement for studied heavy metal sources identification. Finally, the multivariate analysis result indicates that anthropogenic activities are dominating rather than geogenic sources, which match with previous findings of Ali et al. (2016) and Proshad et al. (2018).

Q-mode CA is applied to identify the spatial likenesses and sampling site grouping in the study area. This study represents two cluster, where cluster one consist of three sampling site (S1, S6-7) at low link distance, which is connected to cluster two seven sampling sites (S2-5, S8-10) at high link distance (Figure 5(a)), indicating that majority number of samples follow high link distance due to different parameters.

4. Conclusions

The surface sediment samples in the Mongla port area of Bangladesh have been analyzed for heavy metals (As, Ni, Pb, Cd, Cr, Cu, and Zn) to evaluate their distribution, source identification and potential ecological risk. The heavy metals concentrations showed that surface sediments were moderate to considerably polluted, and the metal pollution was higher in the dry season than wet season, and it also exceeded the TRV and UCC recommended values except for Pb. The study sites were found to be low to moderately polluted by As, Pb, Cr, Ni, Cu, and Zn, but especially by Cd, according to sediment quality indices. The pollution load index values indicated that the sediment qualities of the study sites were deteriorating. The multivariate analysis represented that the studied heavy metals originated from human activities. Ecological risk assessment (SQGs and PERI) showed that As, Ni, Cd, Cr, and Cu were considered as the concerning pollutants to the ecosystem of the study area. The potential acute toxicity results expressed that the study area sediments were moderate to severe toxicity for bottom-living organisms, especially in the dry season. Since the sources of heavy metals are linked to human activities, this study suggests developing and implementing wastewater treatment plants in the port region and upstream watersheds to minimize pollution and ecological risk.

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