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Depth-related dynamics of physicochemical characteristics and heavy metal accumulation in mangrove sediment and plant: *Acanthus ilicifolius* as a potential phytoextractor

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ABSTRACT

The focus of this study was to determine the depth-wise variability of physicochemical properties (*i.e.*, pH, TOC, TN, and EC), and heavy metals (*i.e.*, Pb, Cu, Zn, As, and Cr) concentration, and the associated biological and ecological risks of the mangrove sediment. The accumulation of metal contents and the phytoremediation and phytoextraction were also investigated in a mangrove species, *Acanthus ilicifolius*. The mangrove sediment consists of a higher proportion of sand fraction (56.6–74.7%) followed by clay (10–28%) and silt (10.1–15. 7%) fractions. The concentrations (mg/kg) of Pb, Cu, Zn, As, and Cr were ranged from 22.05–34.3, 8.58–22.77, 85.07–114, 5.56–12.91, and 0.98–5.12 in all the sediment layers. The hierarchy of the mean metal concentration in sediment was Zn (102 mg/kg) > Pb (25.6 mg/kg) > Cu (14.8 mg/kg) > As (8.79 mg/kg) > Cr (2.74 mg/kg) respectively. The examined metal concentrations were below the respective average shale values (ASVs). The degree of environmental, ecological, and biological risks was minimal according to various pollution indices like geoaccumulation index (I_{geo}), contamination factor (*CF*), and pollution load index (*PLI*). According to sediment quality guidelines (SQGs), the adverse biological risk effect was not likely to occur. The result of the potential ecological risk index (*PERI*) demonstrated that the study area was in the low-risk condition as the corresponded *RI* value < 100. A combined influence of geogenic and anthropogenic factors was identified as the metal sources by multivariate analysis. The study found that the accumulation rate of the metal contents was higher in leaves than that of roots. The mean descending metal concentration values were Zn (107) > Pb (28. 7) > Cu (16.9) > As (11.2) > Cr (4.99) in leaves and Zn (104.32) > Pb (27.02) > Cu (15.29) > As (10.39) > Cr (3.80) in roots. The translocation and bioaccumulation factors of heavy metals suggested that the mangrove plant species, *A. ilicifolius* can be used for phytoremediation and phytoextraction since the bio-concentration factor and translocation factor > 1. The studied species exhibited the metal tolerance associated with two following strategies, metal exclusion, and metal accumulation. However, excess metal tolerance can impact the surrounding marine environment.

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1. Introduction

Mangroves are intertidal forest that occur in the tropical and subtropical coastal glove. The mangrove forest is one of the crucial habitats for various organisms, an intrinsic part of the ecosystem (Deng et al., 2019). Mangrove forests play an important function for the fisheries sector providing organic materials and detritus (Rahman et al., 2019a; Duke et al., 2014). It is a typical forest along with the unique geographical position and dynamic interface of land and sea. Mangrove makes up less than 1% of all tropical forests globally but signifies a valuable ecosystem delivering an array of crucial goods and services that contribute to the livelihoods and security of the coastal people (Duke et al., 2014). For instance, storm, flood, sediment erosion protection, coastal food items, nursery ground of numerous aquatic species are typical benefits from the mangrove forests. Moreover, it was estimated that 1 ha of healthy mangroves provides nearly 1.08 t of fish/year in the Philippines and 37,500 US dollar in the fisheris sector in the Gulf of California, Mexico (Schatz, 1991; Aburto-Oropeza et al., 2008).

The mangrove sediment is a sink of heavy metals received from tidal water, stormwater, surface water runoff and effluent from the coastal industries (Hossain et al., 2020). Globally, more than 50% mangroves are in danger due to climate change, population growth, and various industrial development near the coastal regions (Hamilton and Casey, 2016; ELTurk et al., 2018). The mangrove ecosystems are being gradually degraded through various human activities like industrialization, urbanization, tremendous unethical agricultural activities, chemical wastage from various untreated plants as well as terrigenous sources (Rahman et al., 2019b). Heavy metals contamination has become a global concern for its tremendous toxicity, non-biodegradable and persistent nature and higher abundance in sediment (Jingchun et al., 2010; Rahman et al., 2019b; Rahman et al., 2014). Heavy metals can be bio-concentrated and biomagnified adversely affect the biological health (Ahmed et al., 2019b; Saha et al., 2020). The physicochemical properties of sediment (*e.g.*, TOC, TN, pH, salinity, and EC) can affect the accumulation of heavy metals and trigger biological and ecological risks (Sundaramanickam et al., 2016). The physicochemical properties of the mangrove sediment are considered crucial abiotic factors that can support the growth of mangroves (Das et al., 2019).

The behavior and other potential characteristics of mangrove plants are influenced by the accumulated metal concentration in sediment (Rahman et al., 2019b). Similarly, the abundance of various plant species can influence the mangrove sediment properties (Weng et al., 2014; Rahman et al., 2019b). Mangrove or coastal plants play a crucial biological role and can uptake heavy metal due to their phytoremediation capability (Rahman et al., 2019b). For instance, *Sonneratia apetala* can uptake a considerable amount of metals from wetland (Zhang et al., 2010). Moreover, plants dispel the metal elements by cation exchange, filtration, and chemical structure changes using bioaccumulation mechanism (Rahman et al., 2019b). Metals are mostly accumulated in the plant roots and tissues and a small portion of them are translocated to the leaves and stems (Boularbah et al., 2006). Mangrove plants are adopted in the metal contaminated environment with appreciable amount of metal uptake capability (Windham et al., 2003). Therefore, native mangrove plants need to be investigated to determine heavy metals' phytoremediation and phytoextraction ability (Rahman et al., 2019b).

To the best of our knowledge, only a few studies have been reported on heavy metal contents in sediments and water in the south Sundarban mangrove forest of Bangladesh (Ahmed et al., 2002; Shil et al., 2017). A comprehensive analysis of physicochemical properties and heavy metals concentration in different sediment layers, and investigation of sediment quality in relation to environmental and ecological risks are very limited. Moreover, no study has been conducted to evaluate the phytoremediation ability of the mangrove plants in the present study area. It is noteworthy to mention that the present study area (Sundarban in Bangladesh) contains the largest mangrove forest in the world, which is unique in nature and provide supports and shelter to a lot of aquatic and terrestrial flora and fauna. However, a growing number of industrial establishments without proper effluent management control jeopardize the ecological health of the mangrove forests. Thus, it is an ideal study area for investigating the anthropogenic heavy metal accumulation in mangrove sediment and plants. Therefore, the objectives of the study were to (1) assess the physicochemical properties of sediment from three different depths, (2) monitor the heavy metal concentration in the mangrove sediments, (3) investigate the degree of the environmental, ecological and biological risks associated with the heavy metals in sediment, and (4) test the usefulness of a mangrove plant species, *A. ilicifolius* for phytoremediation and phytoextraction.

2. Materials and methods

2.1. Description of study area

The Sundarban is a natural mangrove forest located in the southern region in Bangladesh and a small part located in southeast India (Fig. 1). It is the largest delta in the world, which was formed by Ganges and Brahmaputra occupying the largest littoral mangrove ecosystem (Ahmed et al., 2002). The ecosystem covers 10,200 km^2 with the distribution of land of 60% in Bangladesh and 40% in India (Ranjan et al., 2018). The climate condition of Khulna, Satkhira and Bagerhat districts is typically warm ranging from 18 to 32 °C. Tides in these areas are so large that almost one-third area disappears and reappears every day. It is a tide-dominated mangrove with many water channels bringing huge loads of residue. The dominant mangrove species in the study area include *A. ilicifolius* and *A. alba*. Being one of the most diverse and productive environments, it is known a unique ecosystem in the world (Borrell et al., 2016). It provides extensive protection for the coastal community from cyclones, tidal flooding, erosion and other identical natural disasters (Das and Vincent, 2009). The ecosystem comprises 200 islands further dissected by 400 inter-connected tidal rivers and canals (Islam and Bhuiyan, 2018). Notable, more than 1.42 million people are directly and indirectly dependent on the mangrove forests. The rivers in the forest are a vast reservoir of natural resources. However, nowadays, several anthropogenic activities like burning of coal, crude and wood in brick kilning industries, ship breaking activities, oil spilling, use of excessive chemicals (*i.e.* fertilizers, pesticides, herbicides, detergent, induatrial waste) directly mix into waterbody, which are gradually deteriorating their physicochemical and geochemical features associated with the hydrological characters (Choudhury et al., 2021; Shil et al., 2017). Three districts *i.e.* Shatkhira, Bagherhat and Khulna are situated in the south-western costal region of Bangladesh and these areas are suitable for export quality shrimp farming. Industrial activities have been increasing in the adjunct area of the three districts. Therefore, it is an important study area for determining heavy metal accumulation in mangrove sediment and plant in relation to anthropogenic impact.

2.2. Sample collection and preparation for analysis

The sediment samples of three different depths were collected from ten different sampling sites in three districts: (i) Shatkhira, (ii) Bagherhat and (iii) Khulna (Fig 1). The sampling sites in three districts were selected where the mangrove forests were surrounded by developing industrial estates along with the residential dwellings. Moreover, in these sampling sites, a regular transition of agrochemical as well as industrial waste is occurred. Therefore, at each sampling spot, three sediment cores (0–90 cm) were recovered using acid-washed PVC pipes of length of 90 cm and internal diameter of 5 cm. All the collected cores were immediately sliced using a ceramic knife at an interval of 30 cm in the field. The sliced sediment samples were kept in the polytetrafluoroethylene bags and transported to laboratory of atomic energy centre Dhaka (AECD), Bangladesh to preserve at 4 °C cold room before freeze-drying to constant weight. The subsamples of freeze-dried

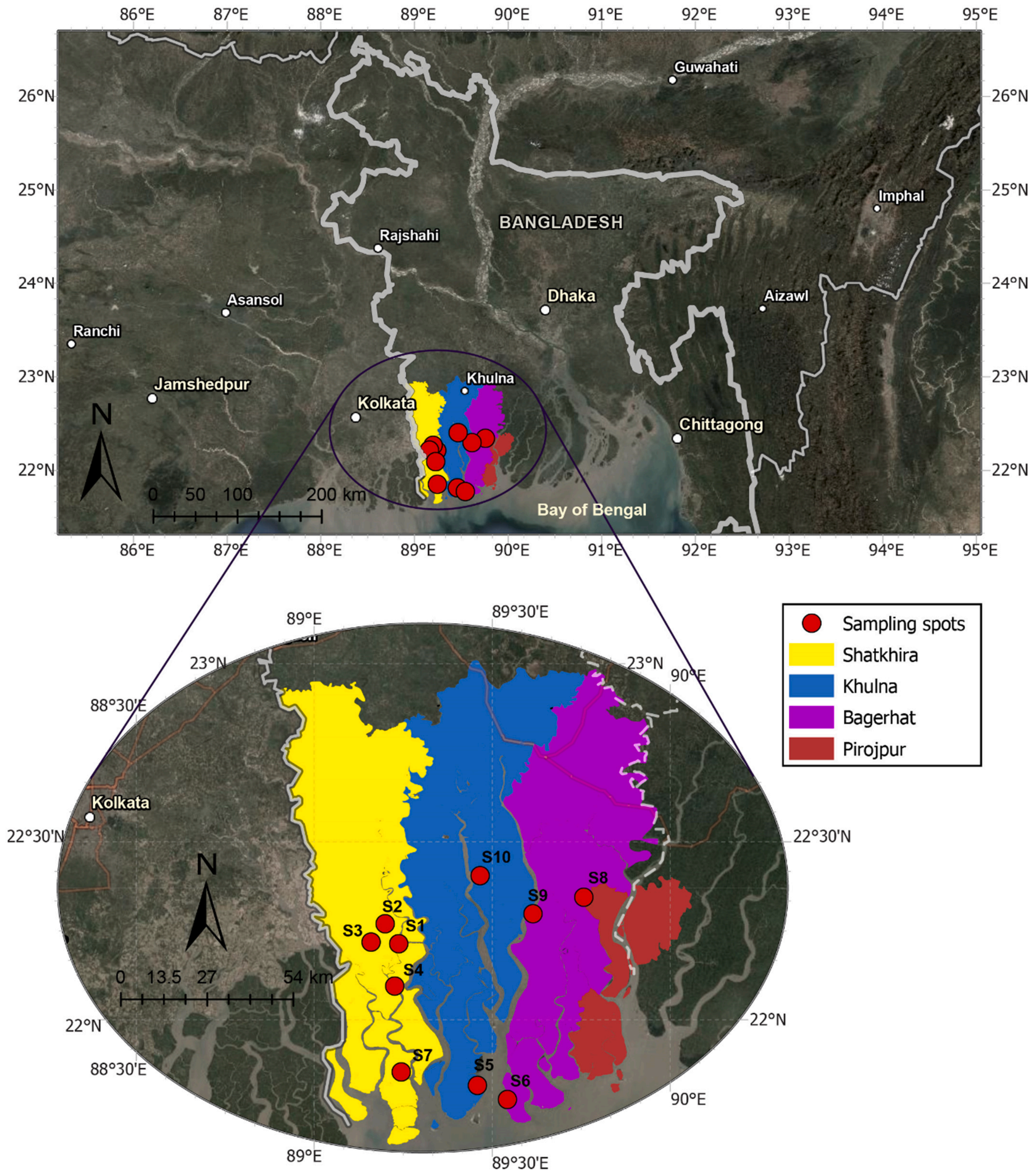


Fig. 1. Mangrove (Sundarban) sediment and plant sampling sites covering three districts, Bagherhat, Satkhira and Khulna.

sediment were then grounded using agate mortar and pestle and passed through 0.65 μm nylon screen. One portion of fine powdered samples were used for the elemental analysis and other portion for physico-chemical analysis.

Subsequently, mangrove plant species, *Acanthus ilicifolius* were collected from the same ten study sites following a standard procedure (Chowdhury et al., 2015; Rahman et al., 2019b). In brief, three subsequent plant were collected from each preselected sampling points and the samples were collected during low tide. Plants having a chest height > 18 cm with a height of 0.95 m were used for the current study. After air drying, plant samples were stored with proper labeling. After that, fouling substances were removed from the sampled plants through washing carefully. Then, leaf samples (young and green) were collected from strong plants using a sharp knife. The roots were collected from same trees in the study areas. Collected plant samples were washed carefully to remove surface bound particles.

2.3. Physicochemical analysis of sediments

The pH and electrical conductivity (EC) of the powdered sediment samples were measured in sediment water extracts of 1:5 ratio (*w/v*) using pH (EC-PH6-02K) and EC meters (HI, 2314). TOC and TN of sediment samples were measured using Vario EL Cube model CHNSO element analyzer manufactured by Elementar in Germany (Radhapriya et al., 2018; Sokolova and Vorozhtsov, 2014). The physical texture of the sediment was analyzed using particle size analyzer, PSA, Coulter L 230 (Beckman Coulter Inc., The U.S.A) (ELTurk et al., 2018).

2.4. Determination of metal concentration in sediment and plant

The leaf and root were cut into pieces with a sharp and clean stainless knife and dried at 70 °C in an oven to obtain a constant weight. The dried plant materials were then grounded to <0.65 μm fraction using agate mortar and pestle (Saha et al., 2016).Dried and finely grounded sediments and plant samples (∼0.65 μm) were subjected to 10 tons hydraulic pressure (Specac Ltd., UK) for 6 to 7 min to make 25 mm pellet for analyzing metal content (As, Pb, Cr, Cu, and Zn) using Energy Dispersive X-ray fluorescence (EDXRF, Epsilon 5, The Netherlands) following the method described elsewhere (Rahman et al., 2019b). A standard reference material from IAEA 433 (marine sediment) was used for the construction of calibration curves for quantitative analysis. The calibration curve for each element was constructed based on the K X-ray and L X-ray intensities calculated for the respective elements present in standard samples. The standard reference materials (marine sediment, IAEA 433) were prepared and examined following the identical method as followed for the investigational samples to check the quality assurance and control (QA/QC) (Rahman et al., 2019b). To check the precision of measurement, two randomly selected samples were measured three times and the calculated relative standard deviation (RSD%) was found at 3 to 5%. The accuracy of measurement was calculated by comparing the measured values of SRM IAEA 433 against the certified values and the percentage of recovery was within 94–106% (Table SI-1).

2.5. Sediment quality and pollution assessment

This study employed various assessment techniques for evaluating sediment quality and environmental risks; and such indices are useful for designing sediment quality goals and remediation strategies.

2.5.1. Geoaccumulation index (I_{geo})

I_{geo} was introduced and developed by Muller (1969) that provides anthropogenic influences based on the geochemical background concentration. The I_{geo} is calculated as follows:

$$I_{geo} = Log_2\left[\frac{Cm}{1.5Bdc}\right] \tag{1}$$

Where c_m is the metal concentration measured in the sample, B_{dc} is the background metal concentration and 1.5 is the background matrix correlation factor due to lithospheric effects (Yi et al., 2011). The B_{dc} values of Zn, Pb, Cu, As, and Cr were 95, 20, 45, 13, and 90, respectively (Turekian and Wedepohl, 1961). Pollution levels for I_{geo} were classified into the following seven classes (Muller, 1969). $I_{geo} < 0$ represents practically unpolluted sediment, while $0 < I_{geo} < 1$, $1 < I_{geo} < 2$, $2 < I_{geo} < 3$, $3 < I_{geo} < 4$, $4 < I_{geo} < 5$ and $I_{geo} \geq 5$ represent unpolluted to moderately, moderately, moderately to heavily, heavily, heavily to extremely and extremely polluted sediment respectively.

2.5.2. Contamination factor (CF) and pollution index (PLI)

The *CF*, proposed by Hakanson (1980) is calculated by normalizing the measured elemental concentrations against shale values taken from Turekian and Wedepohl (1961). The *CF* and *PLI* were calculated using the equations below (Hakanson, 1980):

$$CF = Cm_{(Sample)}/Bdc_{(Shale)} \tag{2}$$

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \ldots\ldots \times CF_n)^{1/n} \tag{3}$$

The sediment quality can be classified according to the *CF* values suggested by Hakanson (1980) and as follows: $CF < 1$: low contamination, $1 \leq CF < 3$: moderate contamination, $3 \leq CF < 6$: considerable contamination, and $CF \geq 6$: very high pollution and the classification of *PLI* is: $PLI < 1$: no pollution identification and $PLI > 1$: pollution confirmation. Additionally, *PLI* value higher than the threshold limit also denotes the progressive deterioration of the sediment quality (Hakanson, 1980).

2.5.3. Sediment quality guidelines (SQGs)

The SQGs are used to assess sediment pollution by comparing the measured elemental concentrations with guideline values (MacDonald et al., 2000). Moreover, to examine the potential environmental and biological risk associated with the abundance of metals in the mangrove sediments, the measured metals concentration were compared with the threshold effect level (TEL) and probable effect level (PEL) (Feng et al., 2011). Therefore, in the present investigation, two types of guidelines (TEL and PEL) were used to investigate the environmental risk of metals in the studied mangrove sediments. The metal concentration below TEL suggest the minimal effect with rarely observed biological effects and the concentration above PEL represents the probable occurrence of adverse effect on the ecosystem (MacDonald et al., 2000; Zhang et al., 2010).

2.5.4. Potential ecological risk index (PERI)

To determine the ecological risk, *PERI* was introduced by Hakanson (1980). The potential ecological risk from a single metal and total metals in sediment is defined as Eqs. (5) and (6).

$$E_r^i = T_r^i \times \left(C_n^i / C_o^i\right) \tag{5}$$

$$PERI = \sum_{i=1}^{n} E_r^i \tag{6}$$

Where E_r^i is the ecological risk factor, T_r^i is metal-specific toxic response factor (As: 10, Pb: 5, Cr: 2, Cu: 5, and Zn: 1) (Hakanson, 1980), C_n^i is individual element in the sediment and C_o^i is background value. C_o^i of As, Pb, Cr, Cu, and Zn was 13, 20, 30, 45, and 45 (Turekian and Wedepohl, 1961). The estimated risks can be classified as $E_r^i < 30$, $RI < 100$: low risk; $30 \leq E_r^i < 50$, $100 \leq RII < 150$: moderate risk; $50 \leq E_r^i < 100$, $150 \leq RI < 250$: considerable risk; $100 \leq E_r^i < 150$, $200 \leq RI < 350$: very high risk; $E_r^i > 150$, $RI > 350$: extreme risk (Hakanson, 1980).

2.6. Phytoremediation for the mangrove plant

In the present study, to evaluate phytoremediation ability, bioconcentration factor (*BCF*) and translocation factor (*TF*) were adopted (Rahman et al., 2019b). *BCF* and *TF* reflect phytoextraction and phytoremediation ability respectively (Phaenark et al., 2009; Rahman et al., 2019b) and the formulas are given below(Yoon et al., 2006; Rahman et al., 2019b):

$$BCF = Cm_r/Cm_s \tag{9}$$

$$TF = Cm_l/Cm_r \tag{10}$$

Where Cm_r is the metal concentration in root samples, Cm_s is the metal concentration in the sediment samples, and Cm_l is the metal concentration in leaves.

2.7. Statistical analysis

Multivariate analysis including correlation matrix (*CM*), principal component analysis (*PCA*) and hierarchical cluster analysis (*HCA*) was conducted to identify the associations of metals and their potential sources in sedimentary environment (Ahmed et al., 2019b). Multivariate analysis was performed using Origin Lab 9.4. ANOVA was conducted using SPSS (version 23) to investigate the significance of site-wise and depth-wise variability of physicochemical parameters and metal concentration in sediment.

3. Results and discussion

3.1. Vertical changes in physicochemical characteristics of mangrove sediments

The measured physicochemical parameters for sediment samples were displayed in Fig 2. The sediment of the mangrove forest was acidic to circumneutral with the pH ranges between 4.56 and 6.88. The finding is consistent with the previous studies (Zhou et al., 2010). This study revealed that the depth-wise variability of pH was significant at 95% confidence level ($p \leq 0.05$) (Fig. 2(a)). Interestingly, increased acidification of sediment was observed with increasing of depth (Fig. 2(a)). The acidic nature of the mangrove sediment may be attributed to release of low molecular weight organic acids through decomposition organic matters (such as root material and leaf litter), and/or oxidation of sulfide minerals (Tam and Wong, 2000). The circumneutral pH condition at the surface sediment layer is likely due to the presence of higher carbonate content with higher pH buffering capacity. The fluctuation of pH in sediment can be impacted by the availability of dissolved CO_2 in the pore water (Taylor et al., 2015). Also, marine calcifying organisms in the sediment m ay be considered one of the crucial factors in changes of pH (Murray et al., 2013; Hendriks et al., 2010). Although, all faunas may not have equal contribution in terms of changing pH, susceptible alteration of pH may occur based on the community structure as well as the excess carbon dioxide in the sediment (Murray et al., 2013).

A decreasing trend of TOC from top to bottom sediment layers was observed in this study. The average TOC concentrations at surface, middle and bottom layers were 2.57%, 1.92%, and1.61% respectively (Fig. 2(b)), and its depth distribution was significantly different ($p < 0.05$). The degree to which the organic matter exposed to heterotrophs may vary in different sediment layers and thus causes different rate of decomposition. Slow rate of microbial induced organic matter degradation under anaerobic condition relative to the oxic environment possibly resulted in gradual decrease in TOC from top to bottom sediment layers (Boyd, 1995). However, this hypothesis remained speculated due to the lack of redox potential data at three depths. Besides, roots have significant influence for carbon accumulation in mangrove forest. Differential roots turnover in different sediment layers may have influenced the vertical gradients of organic matter.

Nitrogen is one of the limiting nutrient factors for the coastal mangrove ecosystem (Liang et al., 2018). The abundance of N in sediment depends on terrigenous input, sediment fertility and texture, sediment redox status, salinity, and tidal flow occurrence (Lovelock et al., 2006). In the study area, the total nitrogen (TN) content was varied from 0.11 to 1.3 mg/g with the mean value of 0.64 mg/g for different deapth of sediment layeras (Fig. 2(c)). The site-specific differences in TN were not significant. This study revealed that the average TN content was decreased from the surface to bottom sediment (surface: 40.2%, middle: 32.1% and bottom: 27.0%) layers (Fig. 2(c)), which was

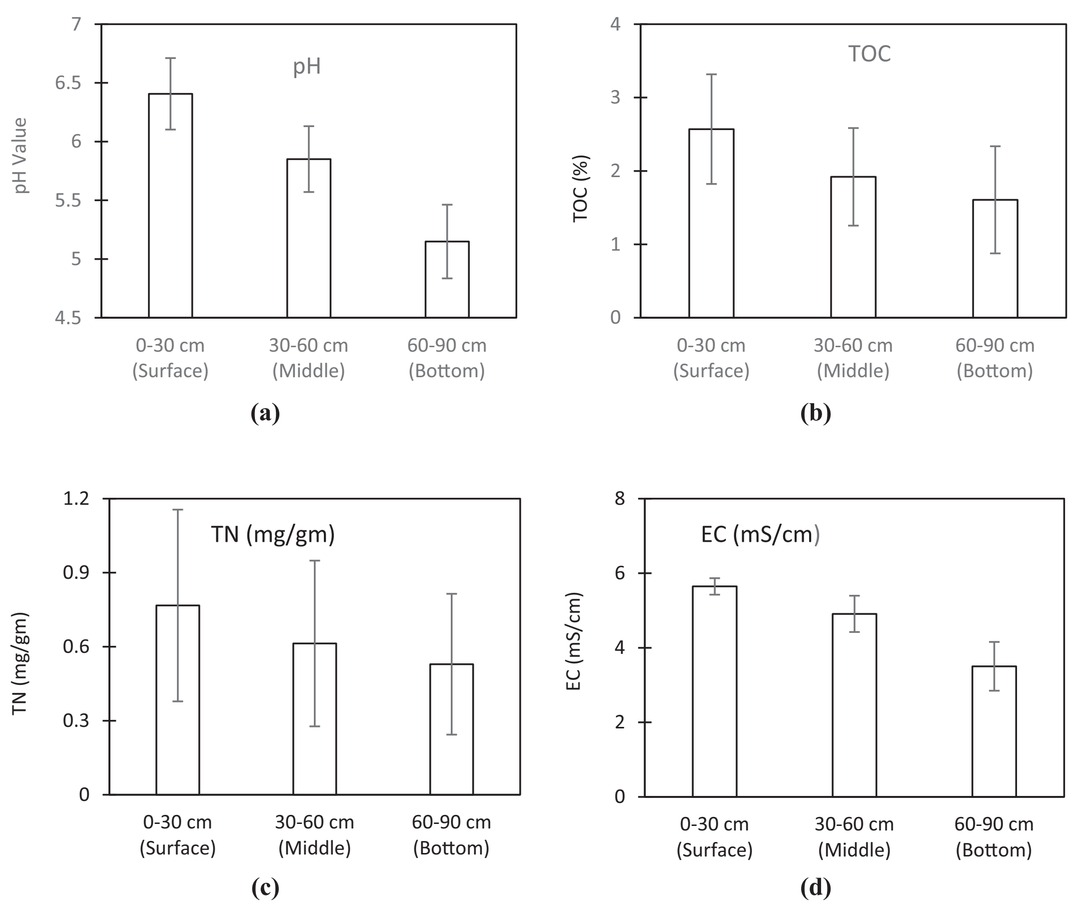


Fig. 2. The variance of layer-wise physico-chemical parameters of mangrove sediment (error bars represent the standard deviation).

consistent with the reported results in literature by several researchers (Cao et al., 2020; Meena et al., 2018; Sauer et al., 2012). Cao et al. (2020) reported that TN concentration can be decreased along with the increase of depth although the storage capacity of deep soil layer would enhance. On the other hand, some pneumatophores in the root system of *A. ilicifolius* species growing in the study area might enhance the retention and absorption of N at the surface rootzone layer (Lugomela and Bergman, 2002). However, similar findings were reported by Chai et al. (2019) wherein no significant variance of TN content was observed along the sediment depths.

Salinity (as represented by EC) is also an important factor in germination, growth of seedlings, and the reproduction of mangrove plants. The growth of mangrove plants is stimulated at moderate salinity, while significantly hampered by excess salinity (Patel et al., 2018). The EC values in sediment samples were ranged from 2.45 to 5.64 mS/cm with the mean value of 5.65 mS/cm (Fig. 2(d)), and the maximum EC value was observed at the sampling point S10 (Kalabogi). The EC were higher in surface sediment relative to middle and bottom layers, and the depth-wise distribution was significantly different ($p <$ 0.05). Our results are comparable with the findings of Srinivasalu et al. (2008), where the range of EC was reported 0.1 mS/cm to 6.1 mS/cm in the tsunamigenic sediment. However, the conductivity is significantly influenced by carbonate and sea salts, and thus the biologically valuable electrolytes (such as PO_4^{3-} and NO_3^-) may have minimal contribution to the measured EC values. In fact, the synchronous behavior EC with pH would suggest that the fluctuation of EC was predominantly influenced by the ions generated from carbonate dissolution.

3.2. Sediment texture

Sediment texture influences on the physicochemical properties, and the development and establishment of mangrove plant species (Tripathi and Misra, 2012; Nyangon et al., 2019). Sediment texture can vary according to sediment sources and their response to the wave and current, flora and benthic fauna's active involvement and their seasonal variability. The grain size of sediment is critical factor for heavy metals binding. The smaller sediment particles have higher metal sorption capacity due to higher surface area and associated higher reactive adsorption sites (Jain, 2005). Fig. 3 showed the variation of textural composition of sediment in terms of sand, silt and clay content. The mangrove sediment consisted of higher proportion of sand fraction (56.6–74.7% with the mean value of 64.9%) followed by clay (10–28% with a mean value of 21.6%) and silt (10.1–15.7% with the mean value of 13.6%) fractions respetively. According to our results, the sediment could be classified as medium loam. Our results are in line with the study conducted by Hossain et al. (2020). Also, Tue et al. (2018) carried out a study in the mangrove forests along the coast of northeast Vietnam, and reported that the sediment composition was mainly fine sand, silt, and clay.

3.3. Heavy metal in mangrove sediment

The elemental concentrations found in the mangrove sediments in three different depths were depicted in Fig. 4 (a), (b), (c). Metal concentrations (dry-weight) in the sediment samples at three different depths were decreased in the following order: Zn (range: 85–114 mg/kg; mean: 102.95 mg/kg) > Pb (range: 22.1–34.3 mg/kg; mean: 25.6 mg/kg) > Cu (range: 8.58–22.8 mg/kg; mean: 14.8 mg/kg) > As (range: 5.56–12.9 mg/kg; mean: 8.79 mg/kg) > Cr (range: 0.98–5.12 mg/kg; mean: 2.74 mg/kg) respectively. Zinc concentration was observed at the maximum level for all the depth concern. According to Zhang et al. (2010) when the coefficient of variation, CV ≤ 20%, there is a negligible variability, 21% < CV ≤ 50% indicates moderate variation, 51% < CV ≤ 100 indicates a high variability of the metal substances, and CV > 100% indicates very high variability in the metal accumulation in the sediment. Following this classification, we observed that 60% of the metal elements were moderately variated in the study area concerning all the layers as Cu, As, and Cr represented the moderate CV values in the surface (27.75, 21.98, and 40.76) and middle layer (27.35, 21.50, and 34.39). Rest of the metals corresponded to negligible CV values. In the bottom layer, only Cr exhibited the corresponded moderate CV value (47.61). Such observation indicated that only Cr was considered and moderately variated in the bottom sediment. The previous investigation illustrated that the vertical profile of the metal contents demonstrates a quick insight of the degree of pollution over time and geochemical composition and disposal of the contaminants (Qiao et al., 2013; Chai et al., 2019). The present study showed that the average metal concentration was relatively higher at the bottom sediment (35%) followed by surface (33%) and middle layers (31%). The downward migration of metals and their subsequent accumulation may have resulted in relative enrichment of metals in bottom layer. However, according to two-way ANOVA, the depth wise distribution of the metal concentration was statistically insignificant ($p > 0.05$). Although, the mean concentrations of Cu, Cr and As were below the respective average shale value (ASV), the concentration of Pb and Zn surpassed the ASV (Table 1). The sedimenatary Cu, As, and Cr were mainly originated from lithogenic sources as their depth-wise distribution was in line with the respective average shale values provided by Turekian and Wedepohl (1961). The higher concentration of Pb and Zn may have introduced to the study area by anthropogenic activities such as agricultural runoff, semi-urbanized mainland and untreated wastage from nearby industries and

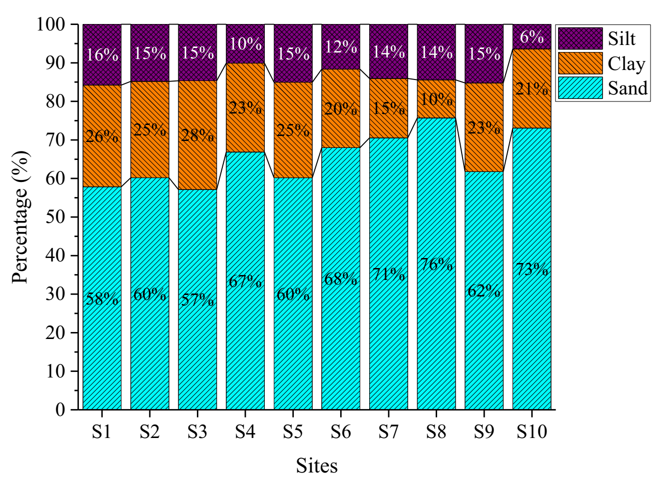


Fig. 3. Sediment texture properties from the study area (mangrove forest, Bangladesh).

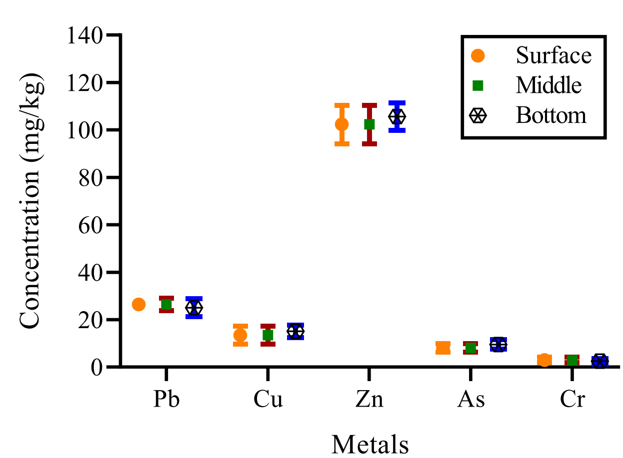


Fig. 4. Accumulation of heavy metal (dw) in the a) surface (0–30 cm), b) middle (30–60 cm) and c) bottom (60–90 cm) sediment in the study area.

Table 1
Average heavy metal concentration in mangrove sediment and their comparison with literature values.

Sites	Heavy metal (mg/kg) (dw)					
	Pb	Cu	Zn	As	Cr	References
S1	23.5 ± 1.5	15.9 ± 5.3	110.0 ± 1.7	8.8 ± 0.4	2.5 ± 0.2	This study
S2	24.7 ± 2.1	14.3 ± 0.3	101.3 ± 8.1	9.8 ± 0.9	2.9 ± 1.4	
S3	23.6 ± 2.2	16.6 ± 7.3	106.7 ± 2.5	9.1 ± 1.8	2.6 ± 1.4	
S4	27.2 ± 6.3	14.2 ± 2.3	104.0 ± 1.7	9.2 ± 2.4	2.4 ± 1.8	
S5	26.0 ± 4.7	12.5 ± 2.2	99.5 ± 14.5	6.1 ± 0.9	2.3 ± 0.2	
S6	27.4 ± 4.2	16.1 ± 3.0	103.6 ± 9.1	8.5 ± 1.0	3.2 ± 1.4	
S7	28.7 ± 2.5	15.5 ± 4.5	103.8 ± 6.3	9.7 ± 0.7	4.1 ± 0.9	
S8	23.4 ± 1.2	16.1 ± 3.8	98.8 ± 9.0	9.5 ± 2.5	2.2 ± 0.4	
S9	23.5 ± 1.1	15.8 ± 3.2	105.3 ± 4.6	10.0 ± 3.4	1.7 ± 0.3	
S10	28.5 ± 1.3	10.9 ± 0.6	96.6 ± 3.4	7.1 ± 1.1	3.8 ± 0.4	
Mean ± Std	25.6 ± 2.2	14.8 ± 1.8	102.9 ± 4.0	8.8 ± 1.3	2.7 ± 0.7	
ASV	20	45	95	13	90	(Turekian and Wedepohl, 1961)
Hong Kong, China	31.2	2.5	43	–	1.2	(Tam and Wong, 2000)
Pearl River Estuary, China	105	400	352	–	74.3	(Chai et al., 2019)
Galetea, Panama	32.5	4	10.9	–	12.8	(Guzmán and Jimenez, 1992)
Chuuk and Kosrae, Korea	8.5	46.9	148	15.6	642	(Ra et al., 2013)
Sungei Buloh, Singapore	12.3	7.06	51.2	–	16.6	(Cuong and Obbard, 2006)
Kuala Selangor estuary, Malaysia	76.6	3.55	28.8	20.7	–	(ELTurk et al., 2018)
Thi Vai Estuary, Vietnam	21	27	92	–	99	(Costa-Böddeker et al., 2017)

residences (Rahman et al., 2021). In comparison, the mean concentration of all the studied elements was much lower than the Pearl River Estuary (Chai et al., 2019). However, the mean concentration of Pb was 3 and 1.2 times higher than Chuuk and Kosrae (Ra et al., 2013) and Thi Vai Estuary (Costa-Böddeker et al., 2017) respectively. The Cu concentration was 3.7 and 5.3 times higher than Galatea (Tam and Wong, 2000) and Kuala Selangor estuaries (ELTurk et al., 2018) respectively (Table 1). Similarly, our Zn concentration was 9.4 and 3.6 times higher than that Galetea (Guzmán and Jimenez, 1992) and Kuala Selangor estuaries (ELTurk et al., 2018), respectively. As concentrations remained in line with the reported values for other international estuaries, but Cr was 2.3 folds higher than Hong Kong (Table 1).

3.4. Risk measurement of mangrove sediment

Geo-chemometrics were used to identify the metal enrichment status, sediment quality and risks for the mangrove ecosystem. The I_{geo} was calculated following the Eq. (1) and the results of I_{geo} are depicted in Fig. 5 (a). The mean I_{geo} in mangrove sediment follows the decreasing order: Pb (range: −0.58 to 0.30, mean: −0.23 ± 0.10) > Zn (range: −0.74 to −0.30, mean − 0.46 ± 0.07) > As (range: −1.81 to −0.59, mean: −1.23 ± 0.21) > Cu (range: −2.98 to −1.57, mean: −2.27 ± 0.24) > Cr (range: −7.11 to −4.72, mean: −5.74 ± 0.39) respectively. The grade of I_{geo} was slightly higher in surface sediment than the middle

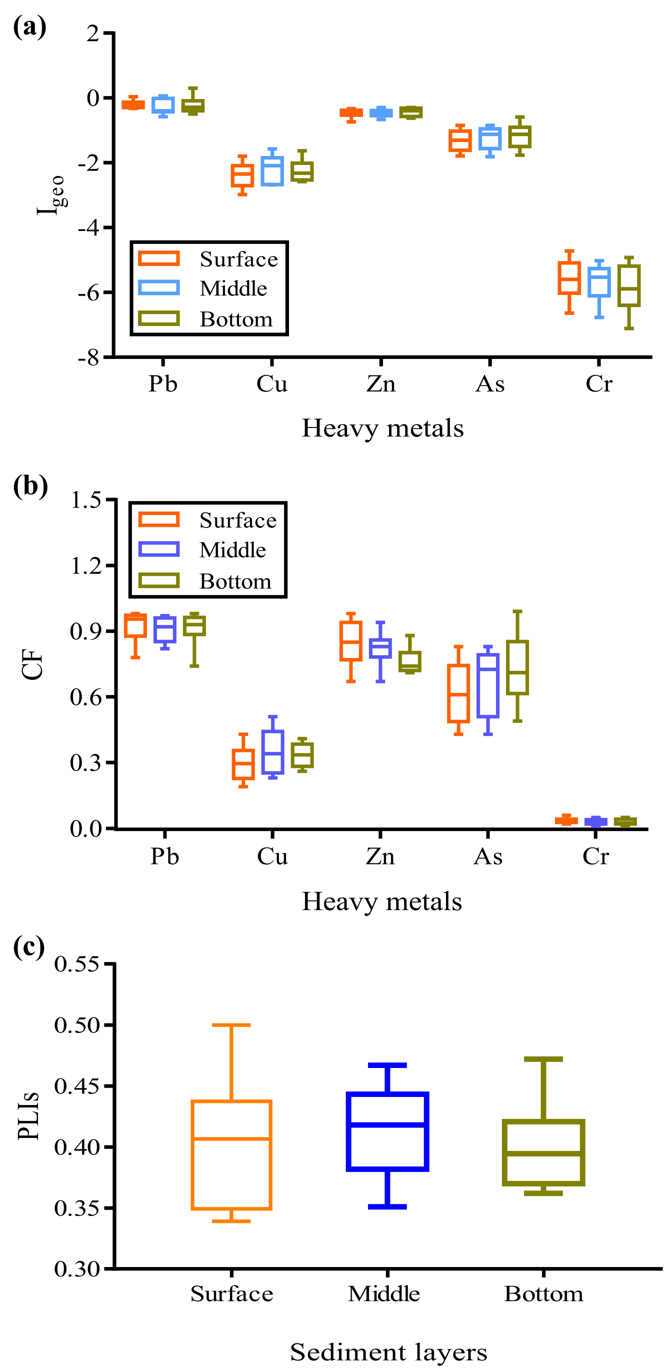


Fig. 5. (a) I_{geo} of the sediment samples replied that the sediment was at negligible pollution. (b) *CF* determined that the sediment samples were in low contamination degree (c) *PLI* of the sample sites denoted that the study area was below the considerable pollution line (<1).

and bottom. Based on the Muller (1969) contamination scale, the studied area was in unpolluted state.

To estimate the degree of contamination, contamination factor (*CF*) and pollution load index (*PLI*) were calculated following the Eqs. (2) and (3), and the findings are presented in Fig. 5 (b). The *CF* of the mangrove sediment decreases in the following order: Pb (range: 0.85 to 0.95; mean: 0.91 ± 0.03) > Zn (range: 0.72 to 0.92; mean: 0.81 ± 0.06) > As (range: 0.47 to 0.77; mean: 0.68 ± 0.1) > Cu (range: 0.24 to 0.37; mean: 0.33 ± 0.04) > Cr (range: 0.02 to 0.05; mean: 0.03 ± 0.01) respectively. The *CF* of all elements were < 1, suggesting low contamination. However, sewages and wastages from nearby sources might have a causative issue to rapid adsorption of metals in the mangrove sediment (Patel et al., 2018). Again, the results of *PLI* of the sediment samples in Fig. 5 (c) denoted that, *PLIs* from all sediment layers were < 1, hence, the study area was pollution-free for the current concentrations of the investigated metals.

The Sediment Quality Guidelines (SQGs) have been implemented by the numerous researchers for toxicological assessment of sediment-related metals for the management of aquatic environments, the safety of biota, and the execution of ecological arrangements and guidelines (MacDonald et al., 2000; Zhang et al., 2010; Souri et al., 2019). Accordingly, this study incorporates two SQGs indictors, probable effect level (PEL) and threshold effect level (TEL) to assess the quality of Sundarbans mangrove sediments. This study revealed that the mean metal concentrations were well within the TEL value, except for As. About 80% of As samples were less than the TEL limit, and 20% were between the TEL and PEL limit (Table 2). None of the samples surpassed the PEL range. This finding indicated no potential harmful effect on bottom-dwelling organisms. In general, the adverse biological effect rarely occurs when SQGs are greater than TEL but below the PEL limit (Costa-Böddeker et al., 2017).

The potential ecological risk index (*PERI*) provides a better understanding of heavy metal pollution by considering cumulative ecotoxicological effect to the sediment dwellers (Karydas et al., 2015). The estimated ecological risk factor (E_r^i) of the metals are presented in Fig 6, and the mean values of E_r^i follow the sequence of Pb (6.76 ± 1.47) > Cu (6.41 ± 0.84) > As (1.64 ± 0.40) > Cr (1.40 ± 0.08) > Zn (0.06 ± 0.02) respectively. The values are below the low-risk line (<30). Moreover, the evaluated *RIs* for top, middle, and bottom sediment layers were found to be <100 (Fig. 6 inset). The *PERI* value of bottom sediment was relatively higher than that of surface and middle sediment. Overall, the potential ecological risk associated with the examined metal concentrations in the mangrove sediment was low.

3.5. Potential sources of heavy metals

Pearson's correlation matrix was employed to identify the pairwise interrelationship among the heavy metals and physicochemical parameters (Fig. 7(a)). The strong correlation among the heavy metals suggests their common origin (Ahmed et al., 2019b). In our study, the significant positive correlation ($p < 0.05$) was observed in three pairs of heavy metals including Cr–Pb ($r = 0.78$), Zn–Cu ($r = 0.69$), and As–Cu ($r = 0.70$). A significant negative correlation was observed for Cu–Pb ($r = -0.53$). In terms of physicochemical parameters, TOC and pH were positively correlated ($r = 0.54$ at $p < 0.05$), while EC and pH were negatively correlated ($r = 0.75$ at $p < 0.05$). Furthermore, a negative correlation was found between EC and Zn ($r = -0.491$).

Table 2
Evaluation of consensus-based sediment quality of the mangrove sediment.

Properties	As	Pb	Cr	Cu	Zn
Mean conc. in mg/kg (dw)	8.8 ± 1.3	25.6 ± 2.2	2.7 ± 0.7	14.8 ± 1.8	103.0 ± 4.0
TEL[a]	9.79	35.80	43.40	31.6	121
PEL[a]	33	128	111	149	459
% of samples < TEL	80	100	100	100	100
% of samples between TEL-PEL	20	0	0	0	0
% of samples > PEL	0	0	0	0	0

[a] MacDonald et al., 2000.

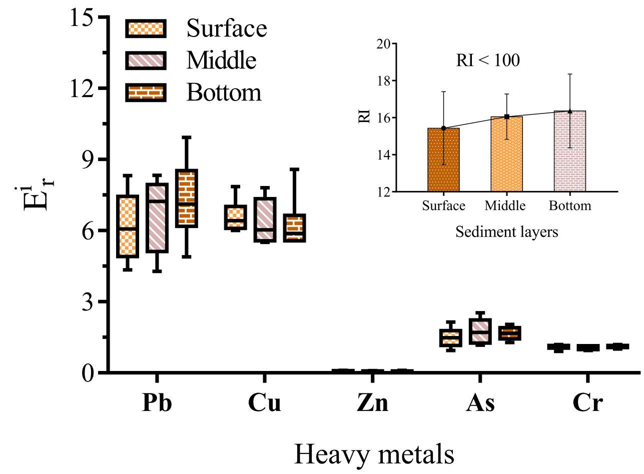


Fig. 6. The evaluation of E_r^i of the metals of the study area was <30 and *RI* < 100 (inside), both demonstrated that the study area was ecologically low risk.

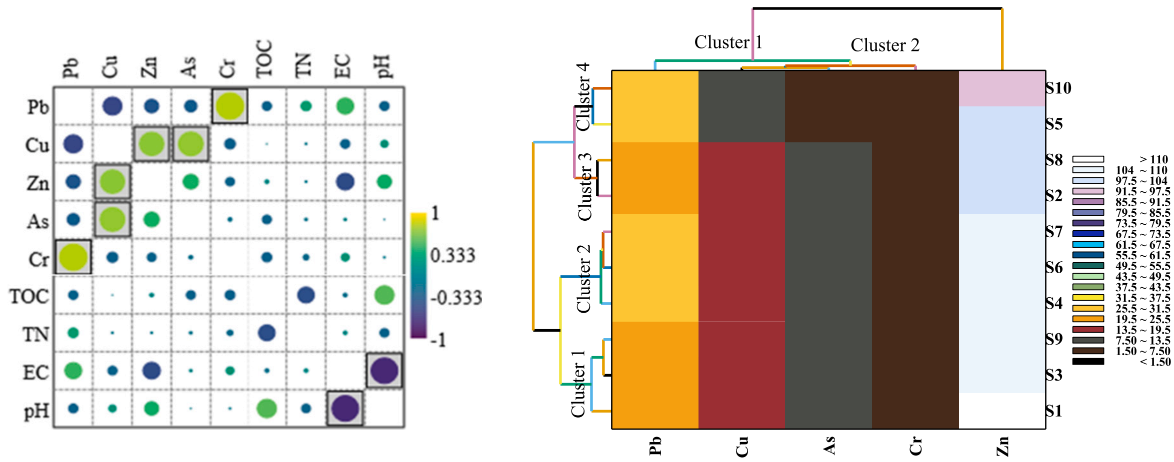
In this study, the datasets was subjected to principal component analysis (PCA) using the correlation matrix in order to standardize each variable and a varimax rotation was applied to aid interpretation of the results (Rahman et al., 2014; DelValls et al., 1998). The PCAs were then selected based on the Kaiser principle with eigenvalues >1 (Rahman et al., 2014; DelValls et al., 1998), which can be found in the supplemebtary section (Table SI-2 and Fig. SI-1). This study revealed that the PC1 and PC2 explained a cumulative variance of 81.36% of the total variance (Table SI-2). The PC1 represented 57.09% of the total variance, while PC2 contributed 24.28% to the total variance. Cu and Zn dominated the PC1 with the loadings of 51.84% and 44.26%, respectively. The PC2 was controlled by As with the loadings of 45%. The possible source of intrution of As in the study area was burning fossile fuels, unconscious use of fertilizers and different agrochemicals (Wu et al., 2015). Moreover, the establishment of some nearby textile industries and oil dropping from boats and ships, and antifouling paints are considered the common anthropogenic sources of metals in the study area (Rahman et al., 2019b; Islam et al., 2017; Islam and Bhuiyan, 2018).

HCA (heat map) was explored to better justify the relationship among the metal and their possible origins (DelValls et al., 1998). HCA differentiates the sampling sites based on similar chemical characteristic (Chung et al., 2011). Therefore, HCA was generated using Ward's method with Euclidean distance and presented in Fig 7(b). Three clusters among the sites were observed wherein S1, S3, and S9 (Table 1) belonged to cluster 1 covered 30% of the higher concentrated area and originated from anthropogenic/natural background. The sites S4, S6, and S7 formed cluster 2, which contributed the highest coverage of 31% concentrated zone. The cluster 3 (between sites S2 and S8) and cluster 4 (between sites S5 and S10) corresponds to 19% and 18% of concentrated site, respectively. The metals were divided into two clusters. Pb and Cr formed cluster 1, whereas Cu, As, and Zn were included in cluster 2. The identification of correlations among the studied metal contents through multivariate analysis rendered a similar characteristics of those metal elements (Ahmed et al., 2019a).

The measured metals in the mangrove sediment may have originated from a combined contribution of anthropogenic and geogenic sources.

3.6. Heavy metal bio-accumulation in the tissues of *A. ilicifolius*

The metal concentrations measured in leaves and roots of *A. ilicifolius*



(a) (b)

Fig. 7. a) Pearson correlation among the metal elements and physico-chemical properties of the sediment of mangrove forests. Data in the boxes exhibits the significant relationship ($p < 0.05$) among the contents. b) HCA (heat map) of the metal contents along with the sites based on the concentration similarity.

were represented in Fig. SI-2. The hierarchy of the mean metal concentration (mg/kg) in the leaves was as follows: Zn (107) > Pb (28. 7) > Cu (16.9) > As (11.2) > Cr (4.99) respectively. A similar elemental distribution pattern was observed for roots: Zn (104.32) > Pb (27.02) > Cu (15.29) > As (10.39) > Cr (3.80) respectively. Likewise sediment, we also tested the coefficient variability (CV) of the metal elements and observed that only Cr was variated in both leaves and roots (CV: 32.32 and 31.74 respectively). Also, the statistical result of one-way ANOVA demonstrated that the metal contents were significantly different ($p < 0.05$) in the plant tissues. The observed maximum accumulation of Zn in the plant tissues was due to its higher relative abundance in the sediments (Fig. 4). Actually, the uptake of Zn^{2+} does not respond to the metabolic inhibitor, therefore, this process is free from metabolic action (Kochian, 1993). Rather, the uptake kinetics of the metal element occurs as a fraction of the metal bound to the organic matter present in the xylem fluids, which results in high mobility of Zn in plants (Tsonev and Cebola Lidon, 2012; Kabata-Pendias and Pendias, 2001). The accumulation of Zn and Cu in the plant tissues revealed that these two metals are crucial trace elements for *A. ilicifolius*. The investigated metal accumulation in plant leaves was higher than plant roots (except for Zn), suggesting that leaves of the studied mangrove species are capable of uptake and store metals (Rahman et al., 2019b). In other words, the plant species' potentiality in extracting metal contents from the studied sites depends on the metal availability in the adjacent sediment.

Comparing with the other investigation around the world, the metal accumulation in the present plant tissues was higher than the other studies in most cases (Table 3). For instance, Chowdhury et al. (2015) conducted a relevant study in Indian Sundarban, phased of the Hugli (Ganges) River Estuary and reported that the mean concentrations of Pb, Cu, Zn, As, and Cr in mangrove tissues were 2.22, 14.37, 18.34, 0.08, and 1.66 mg/kg (dry-weight) respectively, and the values were lower than that of this study. Moreover, in terms of mean Pb (27.85 mg/kg)

Table 3
A comparison of heavy metal concentration in mangrove plants (mg/kg, dw) (leaves and roots) with other relevant studies.

Locations	Tissue types	Pb	Cu	Zn	As	Cr	References
Sundarban, Bangladesh	Leaves	28.67	16.94	107.09	11.19	4.99	Present study
	Roots	27.02	15.29	104.32	10.39	3.80	
	Mean	27.85	16.12	105.71	10.79	4.40	
Mangrove forest, India	Leaves	1.65	12.44	21.16	0.1	1.15	Chowdhury et al. (2015)
	Roots	2.78	16.30	15.51	0.06	2.17	
	Mean	2.22	14.37	18.34	0.08	1.66	
Red Sea, Saudi Arabia	Leaves	3.79	13.24	–	–	14.96	Alzahrani et al. (2018)
	Roots	3.67	9.82	–	–	17.46	
	Mean	3.73	11.53	–	–	16.21	
Futian Mangrove forests, South China	Leaves	1.6	17.01	40	–	3.00	Wang et al. (2013)
	Roots	18.3	106.5	108	–	13.2	
	Mean	9.95	61.75	74	–	8.10	
Farasan Island, Saudi Arabia	Leaves	–	356.6	29.5	–	9.30	Usman et al. (2013)
	Roots	–	270.5	34.9	–	14.90	
	Mean	–	313.55	32.2	–	12.10	
Futian Nature Reserve, China	Leaves	1.44	10.12	173.14	2.5	77.56	He et al. (2014)
	Roots	0.61	30.89	79.97	1.62	98.88	
	Mean	1.03	20.51	126.5	2.06	88.22	
Can Gio mangrove, Vietnam	Leaves	–	14.41	–	0.36	6.58	Thanh-Nho et al. (2019)
	Roots	–	18.97	–	8.53	11.85	
	Mean	–	16.69	–	4.44	9.22	
Pichavaram, India	Leaves	8.00	33.00	18	–	19.00	Kandasamy et al. (2014)
	Roots	9.00	58.00	18	–	35.00	
	Mean	8.50	45.5	18	–	27.00	
Southeast Sulawesi. Indonesia	Leaves	0.14	25.85	2.23	–	–	Analuddin et al. (2017)
	Roots	0.60	24.44	35.64	–	–	
	Mean	0.37	25.14	18.93	–	–	

accumulation, the concentration rate of the metal was much higher than the results from the Red Sea (Alzahrani et al., 2018), Futian mangrove forests (Wang et al., 2013) and Southeast Sulawesi (Analuddin et al., 2017). A similar trend was observed for As concentration (10.79 mg/kg) regarding to the comparable investigations. The mean of Cr (4.40 mg/kg) in our study area was only higher than the mean of the mangrove forest conducted by Chowdhury et al., (2015). The average accumulation rate of Cu (16.12 mg/kg) in the plant tissues was higher than in the mangrove forest and the Red Sea, however, the rest of the studies exhibited significantly exceeded values than ours. In consideration of Zn, the mean value (105.71 mg/kg) was only comparable with Futian Nature Reserve (126.56 mg/kg), whereas other considered studies did not surpass our respective mean value.

3.7. Phytoremediation potential of mangrove plant (A. ilicifolius)

Mangroves are capable of accumulating higher metal concentrations from the surrounding contaminated environment due to their specific tolerance to the metals (Zhang et al., 2010). In general, when a plant uptakes the contaminants from the surrounding environment and unable to metabolize rapidly, accumulation occurs at an early stage (Chandra et al., 2017). The bioaccumulation and phytoremediation potential of *A. ilicifolius* was tested using bioconcentration factor (*BCF*) and translocation factor (*TF*). According to Fitz and Wenzel (2002), plants with *TF*, especially $BCF > 1$ are capable of phytoremediation. However, some factors like pH of the environment, metal availability, and *rhizospheric* bacteria may control the metal uptake ability of the plants (Rahman et al., 2019b; Rosselli et al., 2003). The calculated *TF* and *BCF* of *A. ilicifolius* was greater than 1, suggesting that *A. ilicifolius* would show hyper-accumulation to the metals and suitable for phytoextraction and phytoremediation (Fig. SI-3). The results of *TF* also demonstrated the ability of the species to transport metals from roots to shoots due to the effective metal isolation and transport system in leaf vacuoles and apoplast (Lasat et al., 2000). Overall, by utilizing the higher metal accumulation capability of *A. ilicifolius*, the spread toxic metals in the coastal areas could be prevented to maintain a healthy mangrove ecosystem. In comparison, Marchand et al. (2016) investigated the uptake of the metal contents in some mangrove plants species located in New Caledonia and reported that the BCFs of the Co, Cu, Fe, Mn, Ni, and Zn were range from 0.06 to 0.13 and the associated TFs were varied from 0.03 to 0.15. Therefore, the investigated plant species there did not exhibit the phytoremediation and phytoextraction ability which was contrary to our investigation.

Metal tolerance in mangrove plants can be varied with the plant species and it is based on the two typical strategies named metal exclusion and its accumulation (Dahmani-Muller et al., 2000). The metal exclusion strategy consists of the abrogation of metal uptake together with the limitation of the metal transport to the shoot, which is generally adopted by pseudometallophytes (Wang et al., 2013). In the present study, we vividly observed that the investigated species were adopted both kinds of strategies. Previous studies reported that the limitation of upward transportation of the metal contents is a common metal tolerance strategy especially in wetland plants and such movement of the metal ions can induce significant stress to photosynthetic tissue (Liu et al., 2009). Sometimes, mangrove species can be used in wastewater treatment due to its low transportation rate of the metal elements enhance the facility to remove the contaminants from the marine environment (Yang et al., 2008). However, excess retention of metal contents, as well as the tolerance of heavy metals in plants, might destroy microbial symbiosis in fine roots (Wang et al., 2010). For example, arbuscular mycorrhizal fungi was recently observed to be ubiquitous in most of the studied mangrove plants which has a potential role in nutrient supplement security to the mangrove plants (Wang et al., 2011). Regarding this issue, it might pose an alarming threat to the, especially oligotrophic mangrove ecosystem. Moreover, some mangrove species like *mictyris brevidactylus*, *Crassostrea corteziensis*, *etc*. have the ability to accumulate a high extent of heavy metals in their tissues from the surrounding environment posing a risk to the associated food chain of the mangrove ecosystem (Yeh et al., 2009).

4. Conclusion

The present study describes the physicochemical features (pH, TOC, TN, and EC) and heavy metal concentrations (Pb, Cu, Zn, As, and Cr) in three different layers of mangrove sediment. Although the site-specific variability of the physicochemical properties was insignificant, the vertical fluctuations were statistically significant. The mangrove sediments were circumneutral to acidic with a gradual decline of pH from surface to bottom layer. A similar vertical trend was observed for TOC, TN, and EC. The sediment texture of the mangrove forest was mainly sandy and clay. The concentration of measured heavy metals was below the average shale values. Zn was the most accumulated metal content both in sediment and plant tissues. The adopted geochemical indices, I_{geo}, *CF*, and *PLI* implied that the sediment of the mangrove forest was not polluted. Even, SQGs in this area revealed that sediment-dwelling organisms were not be affected due to such limit of the metal attribution. The evaluated *PERI* indicated that the study area was not at risk level as the corresponded *PERI* was lower than 100. Multivariate statistical analysis (Pearson's correlation, PCA, and cluster analysis), suggested that the metals in mangrove sediment were sourced from the combined contribution of natural and anthropogenic activities. A significant site-specific variabiabilty of metal concentrations was observed in *A. ilicifolius* . The accumulation rate of the metal contents was comparable to other international investigations. The notable fact is that *A. ilicifolius* exhibited phytoremediation and phytoextraction ability as BCF and TF were greater than unity. Also, this specimen signified the metal tolerance as the species exhibited metal exclusion and metal accumulation strategies. The ability of the metal tolerance in the plant might facilitate the removal of the contaminants from the wastewater, however, excess metal tolerance might be detrimental to the adjacent environment.

CRediT authorship contribution statement

M. Safiur Rahman: Conceptualization, Methodology, Software, Validation, Formal analysis, Data Curation, Writing - Review & Editing, Supervision. **Narottam Saha:** Software, Validation, Formal analysis, Review & Editing. **A.S. Shafiuddin Ahmed, S.M. Omar Faruque Babu and Abu Reza Md Towfiqul Islam:** Visualization, Writing - Original Draft, Formal analysis. **Bilkis A. Begum:** Project management and Visualization. **Yeasmin N. Jolly and Shirin Akhter:** Investigation, Sample analysis using XRF. **Tasrina R. Choudhury:** Review & Editing and Visualization.

Declaration of competing interest

The authors wish to confirm that there are no known conflicts of interest associated with this publication and there has been no significant financial support for this work that could have influenced its outcome.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2021.113160.

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