Heavy Metals in Salt Marsh Sediments of Porteresia Bed along the Karnafully River Coast, Chittagong

Mohammad Abdul Momin SIDDIQUE [1, 2] and Mahbuba AKTAR [2]

[1]Norwegian College of Fishery Science, University of Tromso, Tromso, Norway;
[2]Institute of Marine Science and Fisheries, University of Chittagong, Chittagong, Bangladesh

Abstract: Heavy metal (Cu, Zn, Cd, Pb and Fe) concentrations in salt marsh sediments of the Karnafully River coast were investigated in this study. Sediment samples of four different sites were analysed for selected heavy metals by AAS (atomic absorption spectrophotometry). The mean values of heavy metals were 45.79 µg/g for Cu, 105.0 µg/g for Zn, 0.43 µg/g for Cd, 26.70 µg/g for Pb and 3297.38 µg/g for Fe in this study. It is observed that Pb and Cu have positive correlations ($r = 0.370$, $P < 0.05$) and Fe and Zn have negative correlations ($r = -0.729$, $P < 0.05$) in the correlation matrix. Except Fe, all the metals were found at contamination levels which may indicate a recent and continuous contamination from domestic and industrial discharges. The results of sediment samples showed that the Karnafully River coast is facing heavy metal pollution.

Keywords: Bangladesh; heavy metals; Karnafully River coast; Porteresia bed

Karnafully is the most important tidal river of Chittagong district and Karnafully River estuary is the most tidal part of this river system that falls into the Bay of Bengal. The Karnafully River estuary is a typical estuary where both tidal oscillation and fresh water discharges are dominantly acting together in creating the high mixing mechanism of neritic and fluvial water for the estuarine system (Mahmood *et al.* 1976). This estuary is important from many aspects including navigation, transportation, fishing activities, docking yards, and the industrial utility of river water.

Thousands of industries and factories are situated on the bank of the Karnafully River or very close to the river system and they do not have any waste treatment facilities. They discharge the untreated waste into the nearest water bodies, which finally reach into the Karnafully River through different canal systems. Annually about 1216 ships and 45–60 oil tankers are handled at the Chittagong port (Ashraful 2003). As a result, various refuse and disposable materials are discharged and spills from ships, oil tankers and fishing boats get mixed with water and sediments.

Salt marshes are the most productive ecosystems and provide a buffer zone between terrestrial and aquatic ecosystems (Rajendran *et al*. 1993). Physical, chemical, and biological interactions between freshwater and saltwater systems can have significant influences on the transportation of trace and heavy metals in the estuarine environment (Ip *et al.* 2006). Therefore, salt marshes are excellent areas to study the pollution chronology of coastal and estuarine systems because of their fine sediments with high organic content (Ashraful *et al*. 2009). Salt marshes act as protective filters and repositories for runoff pollutants (Teal & Howes 2000), pathogens and different types of nutrients (Weis & Weis 2003).

In general, the salt marsh genesis is based on accretion due to sedimentation of suspended matter, supplied by tidal water or flood water of marine and riverine origin (Beeftink & Rozema 1988). As a consequence nutrients and contaminants like heavy metals, pesticides and halogenated hydrocarbons are transported into the salt marsh sediments (Delaune *et al.* 1990) since they are

partly bound to suspended particles. However, they can be available for uptake and accumulation by salt marsh plants (Otte *et al.* 1993) and animals (Banus *et al.* 1975). This might lead to a further transfer of heavy metals and pollutants into the food web via marine animals and birds.

The bioavailability of heavy metals in salt marsh sediments is low compared to terrestrial systems with oxidized soils. However, many pollutants are stabilized and immobilized, which can affect the availability of heavy metals in the salt marsh sediments (Stolz & Oremland 1999) and perform other ecosystem services (Harris 1999). Banus *et al.* (1975) demonstrated that heavy metals can partly be retained and accumulated (Gambrell 1994; Williams *et al.* 1994) in the salt marsh sediment.

In Porteresia bed, water-soluble metals and exchangeable metals are the most available and precipitated inorganic compounds, metal complexes with large molecular weight, humus materials and metals adsorbed to hydrous oxides are also possibly available (Gambrell 1994; Williams *et al.* 1994). But metals precipitated as insoluble sulphide forms and some metals bound to minerals are essentially unavailable in salt marsh sediments (Gambrell 1994).

The presence of heavy metals in the aquatic environment is of major concern because of their heavy toxicity, bio-accumulating tendency in the biota. Pollution by heavy metals is a threat to human life and the entire environment as well as the wetland ecosystem (Islam & Tanaka 2004; Igwe & Abia 2006). However, the structure and functioning of salt marshes are subjected to many human activities in coastal and estuarine areas.

Heavy metals are an important class of pollutants in the aquatic environment. Some heavy metals such as mercury, lead, cadmium, copper and zinc have been shown in some previous investigations to occur at a significant level in the salt marsh sediments. The accumulation of these heavy metals might affect the coastal ecosystems as well as the salt marsh biota. In view of the economic importance of the coastal regions and the adverse effects of metal pollution on living resources, the present study was carried out to investigate the concentration of heavy metals in salt marsh sediments of the Karnafully River coast. Besides heavy metals, the soil texture and organic matters of the salt marsh sediments are also assessed in this study for better understanding of the soil nature of Porteresia bed of the Karnafully River coast.

MATERIAL AND METHODS

Study locations. Karnafully is the most important river of Chittagong. The Karnafully river originates from the Lusai Hills of Assam in India at latitude 22°54'N and longitude 92°27'E (O'Mallery 1908) and then enters the district of Chittagong from the north-eastern side. The total length of the Karnafully River is about 170 miles and it empties into the Bay of Bengal (Mahmood *et al.* 1976). The study area presents a great interest because a lack of proper sanitation and waste collection facilities, sewage from all over the Chittagong city finally find their way into the Karnafully River through five major drainage systems. The study area receives the untreated wastage of many light and heavy industries. Four stations selected for the sampling were Station-1 (Rajukhali khal, location: 22°19.67'N and 91°51.13'E), Station-2 (Chaktai khal, location: 22°19.55'N and 91°50.49'E), Station-3 (Monowerkhali khal, location: 22°19.48'N and 91°49.98'E) and Station-4 (Chittagong Port, location: 22°18.48'N and 91°48.63'E) (see Figure 1).

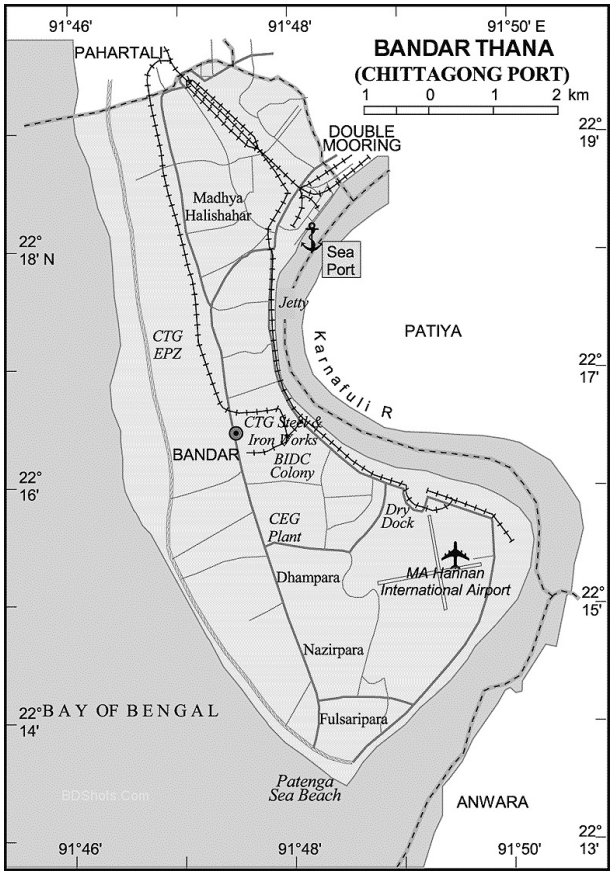

Figure 1. The study location on the Karnafully River coast

Sample collection. The four stations of study are exposed to different sources of pollution including domestic drainage, industrial effluent, oil refineries and ship's effluents, and coastal oil pollution. For each location samples were taken three times at a 5 m distance using an Ekman grab sampler from the Porteresia bed along the river. The position of sediment sampling locations was detected using GPS localization. The soils were collected from a depth of 0–10 cm at each sampling location and subsamples were taken from the central part of the grab to avoid contamination. The samples were kept in self-sealed acid pre-cleaned plastic bags, rinsed with metal-free water. The samples were deep-frozen until analysis.

Analysis of organic carbon. About 100 g of soil was dried at 80°C for 48 h to determine the moisture content of the soil, and for the analysis of organic matters. A portion of 30 g of soil was oven dried at 105°C for 24 h and burned in a furnace at 450°C for 4 h in order to estimate the organic matter content. Grain size distribution was determined by wet sieving of 100 to 150 g of sediment over a 63 pm mesh sieve. From the < 63 pm fraction, a subsample of 50 to 500 ml was filtered over a dried pre-weighed filter (0.45 pm, cellulose nitrate), dried for 24 h at 60°C and weighed. The residue (fraction > 63 pm) was dried for 24 h at 105°C and placed in the sieves of a mechanical sieve shaker with increasing mesh diameter (90 to 500 pm) for 15 min. The resultant fractions were weighed.

Analysis of sediment texture. To determine the soil texture about 25 g oven-dried soil (105°C) was taken in a 500 ml beaker and distilled water was added. The contents were stirred thoroughly with a glass rod for half an hour. Afterward, 10 ml of NaOH solution was added, stirred thoroughly and the contents were transferred quantitatively into a homogenizer cup with repeated washing. The material was thoroughly homogenized and transferred to 1000 ml sedimentation cylinder. The volume was made up to the mark with distilled water and stirred to prepare a uniform suspension. The suspension was allowed to settle and reading with a Bouyoucos soil hydrometer (model: 152H) was taken exactly after 4 min and 2 hours. A similar blank was also run without soil with the addition of 10 ml of NaOH solution to distilled water.

Extraction of heavy metals from sediments. 10.0 g of dried and homogenized sediment were weighed and placed into an acid washed PTFE digestion vessel. The digestion of total sediment (fraction < 63 μm) was performed with a mixture of HNO_3-HCl-HF (concentrated, Merck) at a hot plate and in a cooling system (Otte *et al.* 1993). The samples were filtered with GF/C filter paper. Metal analysis was carried out by flame atomic absorption spectrophotometer (Hitachi Z 8230 the model SSC 300) for Zn, Cu and Fe, and flameless atomic absorption spectrophotometer (Hitachi Z 9000) for Cd and Pb. The wavelengths of the most sensitive lines from the hollow cathode lamps (HCL) 228.8, 217.0, 213.9 and 324.7 nm were used for Cd, Pb, Zn and Cu, respectively.

Detection limits and accuracy. Table 1 shows detection limits for a number of common elements determined by flame atomic absorption and compares them with those obtained by other atomic absorption methods. Under usual conditions, the relative error of flame absorption analysis is in the order of 1% to 2%. With special precautions, this figure can be lowered to a few tenths of 1%. Note that flame AA detection limits are generally better than flame AE detection limits except for the easily excited alkali metals.

Data analysis. All calculations were based on dry weight of soil. Statistical software SPSS 16.0 was applied to determine the mean concentrations and standard deviation of heavy metals from the sampling sites. Relationships of heavy metal concentrations in sediments were tested by Pearson correlation analysis. Statistical significance was tested at 95% confidence level.

Table 1. Detection limits for some elements by atomic spectroscopy (absorption analysis)

Metals	Spectral line/nm	Flame	Detection limit (ppm)
Cd	229	C_2H_2-air	0.0005
Pb	283	C_2H_2-air	0.01
Zn	214	C_2H_2-air	0.008
Cu	325	C_2H_2-air	0.001
Fe	248	C_2H_2-air	0.01

RESULTS

Organic content and texture of sediment samples

Organic contents of different sites are not similar. The highest amount of organic carbon is found at Station 4 (2.18 ± 0.04%) and the lowest amount of organic carbon is obtained at Station 2 (1.03 ± 0.01%). On the other hand, a maximum amount of sand particles is found at Station 3 (50.08%) and maximum 30.8% of silt is found at Station 2. Organic content and the amount of sediment particles are shown in Table 2.

Concentration of heavy metals in salt marsh sediments

The concentrations of different heavy metals are not uniform at all the stations. The results of Fe, Cu, Zn, Cd, and Pb concentrations on the Karnafully River coast, expressed as μg/g, are shown in Table 3. The concentration of Cd and Pb were higher at all the stations compared to the other metals of this study. Maximum concentrations of Fe, Cu, Zn, Cd and Pb are found as 3 798.76 ± 67.19 (Station 2), 70.50 ± 3.53 (Station 3), 126.35 ± 15.66 (Station 1), 0.56 ± 0.49 (Station 3) and 42.02 ± 3.66 (Station 3), respectively. The lowest concentrations of Fe, Cu, Zn, Cd and Pb are obtained as 2723.38 ± 69.80 (Station 1), 34.71 ± 30.92 (Station 2), 91.71 ± 6.61 (Station 2), 0.27 ± 0.24 (Station 1) and 19.87 ± 34.41 (Station 4), respectively. It is also observed that the concentrations of Cu, Cd and Pb are higher at Station 3 compared with the other stations.

A comparison of the obtained values of Fe, Cu, Zn, Cd and Pb in the present study with the recommended values of unpolluted sediments is presented in Table 4. A significant correlation was observed between Pb and Cu (r = 0.370, P <0.05). A nega-

Table 2. Organic content and texture of salt marsh sediments of the Karnafully River coast (in %)

Study location	Organic matter	Organic carbon	Sand	Clay	Silt
Station 1	2.832 ± 0.03	1.49 ± 0.02	41.68	32.72	25.6
Station 2	1.951 ± 0.01	1.03 ± 0.01	38.08	31.12	30.8
Station 3	3.846 ± 0.02	2.02 ± 0.02	50.08	31.12	18.8
Station 4	4.147 ± 0.03	2.18 ± 0.04	38.08	39.12	22.8

Table 3. Heavy metal concentrations in salt marsh sediments of the Karnafully River coast (mean value ± standard deviation)

Metals	Station 1	Station 2	Station 3	Station 4
Fe	2723.38 ± 69.80	3798.76 ± 67.19	3633.26 ± 98.47	3034.11 ± 243.68
Cu	37.98 ± 7.20	34.71 ± 30.92	70.50 ± 3.53	39.98 ± 4.57
Zn	126.35 ± 15.66	91.71 ± 6.61	99.30 ± 8.73	103.19 ± 6.19
Cd	0.27 ± 0.24	0.42 ± 0.37	0.56 ± 0.49	0.49 ± 0.43
Pb	21.26 ± 2.07	23.67 ± 20.88	42.02 ± 3.66	19.87 ± 34.41

Table 4. Comparison of detected values of heavy metals with recommended values of unpolluted sediments (in μg/g)

Metals	Present study	Recommended value	References
Fe	3297.38	41 000.00	GESAMP 1982; Salomons & Forstner 1984
Cu	45.79	33.00	GESAMP 1982
Zn	105.00	95.00	Salomons & Forstner 1984
Cd	0.43	0.11	GESAMP 1982
Pb	26.70	19.00	Salomons & Forstner 1984

Table 5. Pearson correlation coefficients and their significance level for heavy metal concentrations in the sediments of the Karnafully River coast

Metals	Fe	Cu	Zn	Cd	Pb
Fe	–	0.228	–0.729*	0.268	0.370
Cu		–	–0.133	0.022	0.555*
Zn			–	–0.487	–0.206
Cd				–	0.102
Pb					–

*Correlation is significant at a 0.05 level

Table 6. Comparison of detected values of heavy metals with other salt marsh sediments of the world (in µg/g)

Sampling sites	Fe	Cu	Zn	Cd	Pb	Reference
Karnafully River coast	3297.38	45.79	105.0	0.43	26.7	present study
Tijuana Estuary	*	26.3	107.1	1.4	36.1	Weis *et al.* 2001
S. Joao da Talha (Tagus Estuary, Portugal)	*	89.1	427.4	4.7	126.9	Franca *et al.* 2005
Northern Europe	*	132.2	303.9	*	156.5	Callaway *et al.* 1998
Hortas (Tagus Estuary, Portugal)	*	27.6	167.6	1.7	65.2	Franca *et al.* 2005
Rosario (Tagus Estuary, Portugal)	*	62.7	320.6	5.9	199.3	Franca *et al.* 2005

*Data not available

tive correlation was found between Fe and Zn (r = –0.729, $P < 0.05$). The correlation matrixes of heavy metals in the sediments are shown in Table 5.

DISCUSSION

Organic content and texture of salt marsh sediment samples

In the present study, organic content has been found at a lower level. A higher amount of organic matter (4.15%) was found at Station 4. The mean concentration of organic matter on the Karnafully River coast is found to be 3.19%, which is less than the result obtained by Weis *et al.* (2001) in wetland sediments of the Tijuana Estuary, California. Weis *et al.* (2001) reported the organic content to range between 8.7% and 13.8% in their study. Leendertse *et al.* (1996) also found a higher level of organic content, 14.3% to 16.2% at Marsdiep and 9.6% to 14.8% at Delfzijl salt marsh sediments. Compared with the other salt marsh sediments, organic content of the Karnafully River coast is lower in concentration and does not exceed the contamination level.

The mean percentage of clay in Karnafully River coast sediments is 33.52%, which is less than the result (59.42%) obtained by Weis *et al.* (2001) from the Tijuana estuary. The mean percentage of clay was found to be 24.5%, which is very close to the result reported by Weis *et al.* (2001). According to the relative percentage of sand, silt and clay the sediments of the Karnafully River coast can be considered as clay-loam sediments (Siddique 2008).

Concentration of heavy metals in salt marsh sediment samples

A number of studies suggested that Fe was frequently used as an indication of natural changes in the heavy metal carrying capacity of the sediments (Rule 1986). In the present study, the highest value of Fe concentration was found to be 3798.76 µg/g at Station 2 (Chaktai khal). The mean concentration of Fe (3297.38 µg/g) does not exceed the recommended values of unpolluted sediments (GESAMP 1982; Salomons & Forstner 1984) in this study. This finding is very close to the findings of Banu (1995) and Ashraful (2003) in the sediments of the Karnafully river mouth and Chittagong coast of the Bay of Bengal, respectively.

Cu is an essential trace nutrient. The concentration of Cu was found lower at Station 1, 2 and 4 but

a higher value of Cu concentration of 70.5 µg/g was found at Station 3, which is twice more than the recommended value (33 µg/g) compared with the other stations. Similar results were obtained in S. Joao da Talha and Rosario salt marshes (Tagus Estuary, Portugal) by Franca *et al.* (2005). However, the mean concentration of Cu in sediments is slightly higher than the recommended values. This result of Cu concentration in sediments is higher than the results of Ashraful (2003) for the sediments of the Chittagong coast. It has been argued that Cu is closely related to the aerobic degradation of organic matter (Das & Nolting 1993) as most marine organisms have developed their mechanisms to regulate concentrations of Cu in their tissues in the presence of variable concentrations in the surrounding water, sediments and food (Neff 2002). However, the high level of Cu in sediments of the Karnafully coast indicates a higher input of deposited organic matter which comes from the industrial and domestic sewages.

Zn is one of the earliest known heavy metals and it has been stated that a large amount of Zn entering the oceans of the world is derived from aerial deposition (Neff 2002). The concentrations of Zn in estuarine sediments vary widely and its variation in concentration depends on the characteristics of sediments (Ashraful 2003). Zn concentrations in the sediments obtained from the study sites of the Karnafully River coast showed slightly higher values compared to the recommended values of unpolluted sediments. The mean concentration of Zn on the Karnafully coast was recorded to be 105.0 µg/g. This result is similar to the results of Mohanchandran (1988), who studied sediments of the Vetter River estuary, India and also very close to the findings of Weis *et al.* (2001) in wetland sediments of the Tijuana estuary. The extensive human activities of many light and heavy industries probably enhanced the concentration of Zn in the study area.

Cd is one of the most toxic metals measured in this study (Neff 2002). It can be toxic to aquatic biota as well as to humans. Previous studies showed that the amount of Cd in the estuarine sediments is related to the amount leached into the water (Neff 2002). Cd concentrations from sediments of the Karnafully coast indicate a significant environmental contamination by this metal (Cd > 0.11 µg/g) (GESAMP 1982). The mean value of Cd concentration was 0.433 µg/g, which is four times more than the recommended value (0.11 µg/g) of unpolluted marine sediments (GESAMP 1982; Solomons & Forstner 1984). But compared with Tagus and Tijuana estuary, the contamination level of Cd on the Karnafully River coast is lower than in these two salt marsh areas. A higher input of Cd to the estuary may be of anthropogenic origin. In the present study, the high level of Cd might be due to untreated sewage effluents (Ismail & Awad 1984) with recent deposit of organics and fine grain sediments (Mahmood *et al.* 1976) as a result of anthropogenic activities (Ashraful 2003).

Like the other metals discussed above, Pb is a natural component of marine ecosystems. It is present at low concentrations in clean seawater and sediments (Neff 2002). In this study, the concentration of Pd is twice higher (42.02 µg/g) at Station 3 than the recommended value (19.00 µg/g) of unpolluted sediments (Salomons & Forstner 1984). The mean value of Pb concentrations ranges from 19.87 to 42.02 µg/g in the sediments of the Karnafully River coast. This finding is consistent with the findings of Ashraful (2003) in the sediments of the Chittagong coast, Bay of Bengal. At all stations Pd concentrations exceeded the recommended values (19.0 µg/g) of unpolluted sediments but comparatively to a lesser extent than the other salt marshes like Tijuana and Tagus estuary. This could be attributed to the combined effect of oil spillage from the ships and fishing boats and the discharge of sewage effluents into waters along the Karnafully River coast (Ashraful 2003).

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Corresponding Author:

Mohammad Abdul Momin Siddique, M.Sc., University of Tromso, Norwegian College of Fishery Science, N-9037 Tromso, Norway
e-mail: tigermomin@yahoo.com