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Assessment of trace metal in macroalgae and sediment of the Sundarban mangrove estuary

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Abstract

This study was carried out to assess the levels of trace metals (Fe, Zn, Mn and Cu) in the macroalgae and sediment of the Sundarban mangrove estuary in Bangladesh. In this connection, macroalgae, water and sediment samples were collected seasonally (pre-monsoon, monsoon and post-monsoon) for a year from the study area. The surface water of the estuary was found to be quite rich in NO₃, PO₄ and NH₄, which is more than that of the sediment pore water. Trace elements were determined using an atomic absorption spectrophotometer. The amount of trace metals in the samples were found in the order of Fe > Mn > Zn > Cu. Enrichment factor was done for trace metals in sediment and showed non-crustal origin of Zn and Cu in the monsoon period. Analysis of variance revealed the seasonal variation of trace metal concentration in sediment (except Mn; p > 0.05) and macroalgae (except Zn; p > 0.05). Iron in sediment showed significant positive correlation both with Fe (r=0.84, p < 0.05) and Mn (r=0.87, p < 0.01) in macroalgae. Similarly, Cu in sediment demonstrated a significant positive correlation of metals in sediment and macroalgae.

Keywords Trace metals · Macroalgae · Sediment · Water nutrient · Sundarban mangrove · Estuarine ecosystem

Introduction

Estuaries are considered to be significant reservoir of trace metals that have originated from both anthropogenic and natural sources. As a favourable site for human settlement and industrial activities, estuaries become polluted with a wide variety of organic and inorganic contaminants derived from municipal and industrial sources, agricultural runoff, recreational boating, commercial shipping and other points (Tam and Wong 1995; Townend 2002; Melville and Pulkownik 2006). The coastal metropolitan areas discharge a huge amount of industrial and domestic wastes that led the pollution in coastal estuaries and affect marine communities (Amado Filho and Pfeiffer 1998). Trace elements those discharged with municipal and industrial wastes are significantly accumulated by many marine and estuarine species. The accumulation of pollutants gives rise to concerns of the possible detrimental effects of toxic substances on coastal resources and their potential impacts on human health (Hatje et al. 2009; Brito et al. 2012). Trace metals like Cu, Zn, Fe, and Mn are essential for aquatic biota to a specific concentration, but they can become toxic if their concentration level in the environment is high (Yap et al. 2003). Thus, it is essential in detecting the sources of such pollutants and measuring trace elements concentration and distribution in the marine environment for better understanding of their behaviour and fate in the aquatic environment (Kumar et al. 2010). Macroalgae and seagrasses are used as biomonitors of metal pollution in many continents, although it is more difficult to assess the amounts of trace elements accumulated in the latter because of their capacity to incorporate elements from both the sediments and the water column (Phillips 1990; Villares et al. 2005). Biomonitoring of metals using macroalgae has been reported in many geographic regions including the Sarawak Malaysia (Billah et al. 2017),



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Egyptian Mediterranean coast (El-Din et al. 2014), Aegean coast (Akcali and Kucuksezgin 2011; Sawidis 2001), Indian coast (Chakraborty et al. 2014), Australia (Melville & Pulkownik 2007) Tyrrhenian coastal areas (Conti and Cecchetti 2003), Aughinish Bay, Ireland (Reis et al. 2016), Polish coast (Rajfur and Kłos 2014; Ozyigit et al. 2017), Malaysian coast (Mashitah et al. 2012), and Sudanese Red Sea coast (Ali et al. 2017).

The Sundarban mangrove forest in Bangladesh coast is the harbour of large diverse macroalgae. Various species of macroalgae are abundant in water, pneumatophores and prop root of mangroves which can play a vital role in detecting trace metal concentration. Moreover, tidal activities induced erosion and physical mixing of the sediments, runoff, oil spillage, industrial activities like discharges from ports and urban area are common in this region. Trace metal concentration has been determined by many researchers (Chatterjee et al., 2009; Filho et al. 2011; Islam and Wahab 2005) in water and sediment of Sundarban mangrove estuary in both part of Bangladesh and India, but no such research has been conducted to determine the concentration of trace element in macroalgae in the Sundarban yet. It is significant to investigate and reveal the potential of macroalgae as a natural bioindicator for trace metal contamination in the Sundarban estuarine ecosystem. To fulfil the gaps, the present research has been conducted with the objectives of estimation of the trace metal concentration in mangrove macroalgae associated with pneumatophore and sediment and to evaluate the potentiality of macroalgae as natural bioindicator test species for metal pollution. The findings of this study would be relevant in coming up with strategies on how to address metallic pollution in estuaries.

Materials and methods

Study area

The Sundarban mangrove forest is the single largest continuous mangrove forest in the world and intersected by the number of river estuaries. The forest has already been recognized as an international Ramsar Wetland Site and declared as a World Heritage Site (WHS) by the UNE-SCO in 1997. The Sundarban forests tract covered an area of 10,000 km² of which 66% are land and the remaining is water (Rashid et al. 2008). Samples were collected from five sampling stations which encompassed Dhanghmari (22°43'72.74" N and 89°58'83.2" E), and Koromjol (22.43°N and 89.592°E) of Passur River estuary (Fig. 1). Sampling stations were selected on the basis of topography and the availability of macroalgae adhered to pneumatophores.



Sample collection

Macroalgae in pneumatophore

Macroalgae associated with pneumatophores were collected seasonally (pre-monsoon, monsoon, and post-monsoon) for the metal analysis. Pneumatophores were cut randomly at mud line with clipper from each transect in the sub-littoral zone (0.5–3.0 m) of the selected sites during low tide when the mangrove foreshore is exposed. Collected pneumatophores were kept in a plastic bag and transported to the laboratory under cool conditions.

Sediment

Intertidal surface sediment (5–10 cm) samples were collected in triplicate from each station with the plastic trowel. After cleaning the surface litter, samples were sealed in plastic bags and kept cooler box during transportation to the laboratory for analysis. Sediment samples were air-dried for 3 weeks prior to the analysis.

Water

The estuarine surface water quality parameters such as temperature, pH and dissolved oxygen (DO) were measured in situ using water pH meter (Model AD1000) and DO meter (PDO 519). The surface water and sediment pore water samples were collected in preconditioned (pre-washed in 10% nitric acid) 500-mL polyethylene bottles from each sampling stations in triplicate for nutrient analysis. The collected water samples were filtered using a Millipore filtering system (MFS). Filtered samples were acidified by adding 2 mL of concentrated nitric acid in each 100 mL sample.

Determination of water nutrients

Dissolved inorganic nitrate (NO₃), phosphate (PO₄), and ammonium (NH₄) in both the surface and pore water samples were determined following the methods described by Kitamura et al. (1982), Weatherburn (1967), and Parsons (1984), respectively.

Trace metal analysis for macroalgae and sediment

For the determination of metal (iron, zinc, manganese, and copper), the macroalgae samples were scrapped off from each segment of pneumatophore taking care of not removing the pneumatophore tissue. Algal samples were then washed thoroughly to remove any adhering sediment particles and separated onto small glass Petri dishes. The samples were



placed in the oven and dried at 105 °C overnight (12 h); then dried material was ground to fine powder according to the method described by Gopinath et al. (2011). While sediment samples were placed in the oven and dried at 105 °C overnight (12 h), dried sediments were mortared to the fine powder and sieved through a 300 BSS mesh sieve. Acid digestion of both macroalgae and sediment samples was performed by using high-pressure laboratory microwave oven following the EPA-3050B method. The concentration of the common contaminant metals, e.g., iron, zinc, manganese and copper, was determined from the digested macroalgae filtrates as dry weight basis (ppm or $\mu g g^{-1}$ dry weight) using a

Shimadzu AA-7000 Atomic Absorption Spectrophotometer in Designated Reference Institute for Chemical Measurements (DRiCM) following the procedure of American Water Works Association (AWWA 3113B).

Enrichment factor (EF)

Enrichment factor is an effective method to estimate the anthropogenic impact on sediments (Huang et al. 2014). Enrichment factor (EF) was estimated using normalization of metal concentrations above uncontaminated background levels (Kumar et al. 2012). The enrichment factor is based on



the standardization of a tested element against a reference. A reference element is the one characterized by low occurrence variability. The most common reference elements are Sc, Mn, Ti, Al, and Fe (Sutherland 2000). In the present study, Fe was used as the reference ($C_{\rm ref}$) element for EF calculations using the following equation

$$EF = \frac{\left(\frac{c_x}{c_{ref}}\right) \text{ sample}}{\left(\frac{c_x}{c_{ref}}\right) \text{ background}}$$
(1)

where C_x = concentration of the element of interest and, C_{ref} = concentration of the reference element for normalization.

In the present study, the background levels (i.e., the average contents of elements in the Earth's crust) given by Rudnick and Gao (2004) were employed for each of the trace metals investigated. The EF values close to unity indicate crustal origin and EF < 1.0 suggests a possible mobilization or depletion of elements (Zsefer et al. 1996). According to EF classification, the EF values 1.5–3.0, 3.0–5.0, 5.0–10 and > 10 are the prominent evidence of minor, moderate, severe and very severe contamination of the elements in the sediments, respectively (Brich and Olmos 2008).

Statistical analyses

Pearson correlation coefficient was used for checking any significant relationship among the parameters tested. Oneway ANOVA was likewise carried out to determine the seasonal variations of different factors. For both cases of analyses, the Statistical Package for Social Science (IBM SPSS Statistics 20.0) was used. The results of the statistical analyses are further elaborated in the succeeding section, and findings are the bases for coming up with inferences relevant in providing recommendations.

Results and discussion

Environmental parameters

In the pre-monsoon season, surface water temperature ranged from 27.4 °C to 28.8 °C and 29.3 °C to 30 °C in the post-monsoon season (Table 1). The maximum temperature (33.2 °C) was recorded in the early monsoon (Table 1). Significant seasonal variation of surface water temperature (df=2, F=166.4, p < 0.05) was observed (Table 4). Mostly similar observation to the present study was reported by Rahman et al. (2013) in the Passur River, Sundarban; Billah et al. (2016) in Miri estuary, and Saifullah et al. (2014) in



 Table 1
 Mean values of environmental parameters in different season at Sundarban

Season	Station	SWT (°C)	pН	DO (mg/L)
Pre-monsoon	St 01	27.4±0.51	8.10±0.07	7.50 ± 0.36
	St 02	28.8 ± 0.50	8.03 ± 0.02	7.33 ± 0.70
	St 03	28.4 ± 0.35	8.08 ± 0.04	8.26 ± 1.73
	St 04	28.2 ± 0.20	8.08 ± 0.04	7.80 ± 1.44
	St 05	27.7 ± 0.30	8.15 ± 0.03	8.73 ± 0.55
Monsoon	St 01	33.2 ± 0.05	7.19 ± 0.03	7.50 ± 0.17
	St 02	33.03 ± 0.20	7.19 ± 0.07	7.16 ± 0.05
	St 03	32.6 ± 0.4	7.26 ± 0.01	6.73 ± 0.49
	St 04	33.2 ± 0.17	7.22 ± 0.02	6.70 ± 0.17
	St 05	32.1 ± 0.52	7.28 ± 0.02	6.66 ± 0.40
Post-monsoon	St 01	29.5 ± 0.5	7.38 ± 0.04	2.83 ± 0.47
	St 02	29.8 ± 0.76	7.46 ± 0.07	2.56 ± 0.47
	St 03	29.4 ± 0.52	7.44 ± 0.03	2.96 ± 0.20
	St 04	29.3 ± 0.65	7.55 ± 0.04	5.26 ± 0.15
	St 05	30.0 ± 0.26	7.48 ± 0.05	5.43 ± 0.37

SWT surface water temperature, DO dissolved oxygen

Sibuti mangrove estuary in Malaysia. The maximum water pH was observed in the pre-monsoon (8.15) and minimum in the post-monsoon season (7.19) (Table 1). A negative correlation (r = -0.80, p < 0.01) between the surface water temperature and the pH was revealed (Table 3). The water pH reported by Rahman et al. (2003) mostly coincided with the present study. The present findings indicated that the water of the Passur River is alkaline (pH > 7), and there were insignificant variations between the sampling stations. Dissolved oxygen was maximum (8.73 mg/L) in the premonsoon (Table 1). Chakraborty et al. (2015) also found considerably higher DO during pre-monsoon season, and the most similar pattern was observed in the study of Hoque et al. (2015).

Water nutrients in surface and sediment pore water

The concentration of nitrate (NO₃) in surface water was found to be higher in the monsoon (0.11 mg/L) and lower in the pre-monsoon season (0.02 mg/L) (Table 2). While, in sediment pore water, the maximum concentration of NO₃ was found in the pre-monsoon season (0.28 mg/L) and minimum in the post-monsoon season. (0.02 mg/L) (Table 2). Generally, the concentration of nitrate in surface water remains higher during the monsoon season which could be due to the freshwater influx (Kumar et al. 2009). Land drainage and precipitation could also increase the concentration of nitrate during the monsoon season (Sarkar and Bhattacharya 2010). The nitrate content

Season	Station	SW NO ₃ (mg/L)	SW PO ₄ (mg/L)	SW NH ₄ (mg/L)	SPW NO ₃ (mg/L)	SPW PO ₄ (mg/L)	SPW NH ₄ (mg/L)
Pre-monsoon	St 01	0.06 ± 0.01	2.13 ± 0.08	0.04 ± 0.02	0.25 ± 0.13	0.07 ± 0.005	0.03 ± 0.010
	St 02	0.04 ± 0.01	1.99 ± 0.07	0.03 ± 0.008	0.28 ± 0.08	0.09 ± 0.004	0.02 ± 0.01
	St 03	0.05 ± 0.009	2.14 ± 0.17	0.07 ± 0.05	0.06 ± 0.001	0.09 ± 0.02	0.04 ± 0.007
	St 04	0.05 ± 0.008	2.10 ± 0.01	0.03 ± 0.01	0.22 ± 0.15	0.04 ± 0.047	0.08 ± 0.07
	St 05	0.06 ± 0.01	2.18 ± 0.09	0.06 ± 0.01	0.11 ± 0.09	0.01 ± 0.001	0.03 ± 0.006
Monsoon	St 01	0.11 ± 0.05	0.60 ± 0.02	0.05 ± 0.03	0.06 ± 0.02	0.05 ± 0.01	0.05 ± 0.008
	St 02	0.07 ± 0.03	0.57 ± 0.01	0.03 ± 0.01	0.06 ± 0.01	0.03 ± 0.02	0.05 ± 0.01
	St 03	0.03 ± 0.008	0.56 ± 0.04	0.03 ± 0.007	0.04 ± 0.007	0.06 ± 0.01	0.05 ± 0.02
	St 04	0.07 ± 0.05	0.55 ± 0.07	0.03 ± 0.005	0.04 ± 0.009	0.07 ± 0.02	0.04 ± 0.02
	St 05	0.03 ± 0.01	0.49 ± 0.01	0.05 ± 0.03	0.09 ± 0.10	0.05 ± 0.02	0.06 ± 0.02
Post-monsoon	St 01	0.02 ± 0.004	1.84 ± 0.15	0.02 ± 0.006	0.03 ± 0.004	1.47 ± 0.07	0.02 ± 0.006
	St 02	0.02 ± 0.003	2.07 ± 0.25	0.12 ± 0.09	0.02 ± 0.007	1.51 ± 0.181	0.07 ± 0.024
	St 03	0.10 ± 0.14	1.83 ± 0.078	0.22 ± 0.24	0.04 ± 0.007	1.62 ± 0.103	0.14 ± 0.14
	St 04	0.02 ± 0.003	1.81 ± 0.127	0.13 ± 0.103	0.04 ± 0.002	1.72 ± 0.103	0.18 ± 0.06
	St 05	0.02 ± 0.001	1.91 ± 0.17	0.28 ± 0.015	0.02 ± 0.007	1.82 ± 0.034	0.14 ± 0.12

Table 2 Water nutrients in surface and sediment pore water of Passur River estuary, Sundarban

SW surface water, SPW sediment pore water

found in the present study is guite similar to that reported by Wahid et al. (2007). The concentration of phosphate (PO_4) in surface water was maximum in the pre-monsoon season (2.18 mg/L). During the monsoon and post-monsoon season, phosphate concentration ranged between 0.49-0.60 mg/L and 1.81-2.07 mg/L, respectively, while phosphate content in sediment pore water was maximum in the post-monsoon season (1.82 mg/L). Almost similar seasonal distribution pattern of phosphate in the water of Passur River was reported by Rahman et al. (2013). Ammonium (NH₄) concentration in the surface water was found to be higher (0.28 mg/L) in the pre-monsoon and lower (0.02 mg/L) in the post-monsoon season (Table 2). In the monsoon season, it ranged from 0.03 mg/L to 0.05 mg/L for surface water. For sediment pore water, ammonium concentration was higher in the post-monsoon season (0.18 mg/L) and lower in the pre-monsoon season (0.02 mg/L) (Fig. 2). Rahman et al. (2015) reported the higher concentration of ammonium in the surface water of the Passur River in the post-monsoon season. Another research by Hoq (2007) also reported the similar pattern of ammonium distribution which mostly coincided with the present study.

Trace metal concentration in sediment

The concentration of Iron (Fe) was found higher than that of other trace metals detected. The maximum content of





Table 3 Tł	he Pearson c	orrelation c	oefficient (r)) among the	e physical pa	rameters, n	utrients and	trace meta	l concentrat	ions in est	uarine eco	systems					
	SWT	DO	Hq	NO ₃ (s)	PO ₄ (s)	NH ₃ (s)	NO ₃ (p)	$PO_4(p)$	NH ₃ (p)	Fe (s)	Zn (s)	Cu (s)	Mn (s)	Fe (a)	Zn (a)	Cu (a)	Mn (a)
STW	1																
DO	0.14	1															
Hd	-0.80^{**}	0.41	1														
$NO_3-(s)$	- 0.003	0.24	0.05	1													
$PO_4 - (s)$	- 0.94**	-0.20	0.73^{**}	-0.01	1												
$NH_3+(s)$	018	-0.51	-0.17	- 0.16	0.24	1											
$NO_3^{-}(p)$	- 0.42	-0.55*	0.76^{**}	0.04	0.36	- 0.36	1										
$PO_4-(p)$	- 0.31	-0.84^{**}	- 0.23	- 0.32	0.34	0.77^{**}	-0.50	1									
$NH_3+(p)$	- 0.33	0.28	0.04	-0.11	0.31	0.77^{**}	- 0.22	0.66^{*}	1								
Fe (s)	-0.51^{*}	- 0.69**	0.03	- 0.39	0.48	0.57*	- 0.32	0.90^{**}	0.55	1							
Zn(s)	- 0.86**	0.15	0.65^{**}	0.08	-0.80^{**}	-0.14	-0.31	-0.23	- 0.25	- 0.33	1						
Cu (s)	0.46	-0.66^{**}	-0.80^{**}	- 0.34	- 0.39	0.59^{*}	-0.78^{**}	0.66^{**}	0.34	0.43	0.47	1					
Mn (s)	- 0.45	- 0.26	0.33	- 0.05	0.40	-0.21	0.31	0.11	- 0.32	0.25	- 0.53	- 0.25	1				
Fe (a)	- 0.35	-0.62*	- 0.07	-0.18	0.32	0.52^{*}	0.36	0.78^{**}	0.52^{*}	0.84^{*}	-0.17	0.46	0.08	1			
Zn (a)	- 0.39	0.34	0.55*	0.58*	0.22	- 0.28	0.46	-0.31	-0.18	- 0.09	- 0.22	-0.60*	0.30	0.04	1		
Cu (a)	0.78^{**}	- 0.36	- 0.90**	- 0.24	-0.71^{**}	0.12	- 0.66**	0.22	-0.003	0.007	0.71^{**}	0.816^{*}	- 0.33	0.22	- 0.54*	1	
Mn (a)	- 0.74**	- 0.64**	0.31	- 0.21	0.74^{**}	0.59*	- 0.06	0.81^{**}	0.53*	0.87^{**}	- 0.59*	0.175	0.43	0.71^{**}	0.10	- 0.29	1
sw surface	water, spw s	sediment po	re water, s su	ediment, a	algae												
*Correlatio	n is Signific	ant at the 0.	.05 level (2-i	tailed)	1												

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Parameter	Pre-monsoon (Mean \pm SD)	Monsoon (Mean \pm SD)	Post-monsoon (Mean \pm SD)	ANOVA
SWT (°C)	28.1 ± 0.37	32.8 ± 0.26	29.6 ± 0.54	df = 2, F = 166.4, p < 0.05
pН	8.08 ± 0.04	7.22 ± 0.03	7.46 ± 0.04	df = 2, F = 81.43, p < 0.05
DO (mg/L)	7.9 ± 0.95	6.95 ± 0.25	3.8 ± 0.03	df = 2, F = 27.31, p < 0.05
SW NO3 (mg/L)	0.04 ± 0.01	0.06 ± 0.01	0.04 ± 0.003	df = 2, F = 0.789, p > 0.05
SW PO ₄ (mg/L)	2.10 ± 0.07	0.55 ± 0.03	1.89 ± 0.15	df = 2, F = 44.27, p < 0.05
SW NH ₄ (mg/L)	0.04 ± 0.02	0.04 ± 0.01	0.15 ± 0.03	df = 2, F = 6.65, p < 0.05
SPW NO3 (mg/L)	0.18 ± 0.03	0.06 ± 0.01	0.03 ± 0.005	df = 2, F = 10.46, p < 0.05
SPW PO ₄ (mg/L)	0.06 ± 0.006	0.05 ± 0.01	1.62 ± 0.07	df = 2, F = 547.67, p < 0.05
SPW NH ₄ (mg/L)	0.04 ± 0.02	0.05 ± 0.02	0.11 ± 0.03	df = 2, F = 3.709, p > 0.05
Fe (Sediment)	$31,870.09 \pm 5042.7$	$27,378.9 \pm 40,433.9$	$43,461.4 \pm 6093.2$	df = 2, F = 42.06, p < 0.05
Mn (Sediment)	359.8±114.8	298.2 ± 56.3	347.6 ± 42.2	df = 2, F = 1.600, p > 0.05
Zn (Sediment)	74.7 ± 12.2	113.3 ± 15.8	84.05 ± 13.4	df = 2, F = 13.83, p < 0.05
Cu (Sediment)	7.7 ± 3.9	33.06 ± 6.9	40.96 ± 15.9	df = 2, F = 75.34, p < 0.05
Fe (Algae)	$14,876.8 \pm 2687.2$	$13,243.5 \pm 1186.04$	$26,887.35 \pm 1819.7$	df = 2, F = 9.77, p < 0.05
Mn (Algae)	280.07 ± 3.8	49.06 ± 2.7	465.8 ± 8.08	df = 2, F = 112.83, p < 0.05
Zn (Algae)	66.9 ± 4.1	48.03 ± 2.3	48.3 ± 2.1	df = 2, F = 4.034, p > 0.05
Cu (Algae)	5.06 ± 1.2	38.2 ± 2.7	28.8 ± 1.4	df = 2, F = 85.32, p < 0.05

Table 4 Analysis of variance (ANOVA) in physicochemical characteristics of water and sediments among the seasons

df degree of freedom, F test of model fitting, p significance level, SWT surface water temperature, SW surface water

Fe was observed in the post-monsoon (46,771.67 ppm) and the lowest in the monsoon (24,443.33 ppm). The present finding coincided with the concentration of Fe $(42,172.17 \ \mu g/g)$ detected by Kumar et al. (2016) in the sediment of Sundarban. Significant seasonal variation was observed in sediment iron (df = 2, F = 42.06, p < 0.05; Table 4). The pore water phosphate demonstrated a strong positive relationship (r = 0.90, p < 0.01) with Fe (Table 3). Billah et al. (2014) also reported the higher concentration of Fe in the sediment of an estuary in Malaysia. The amount of Zn was found to be higher (142.70 ppm) in the

estuary

monsoon followed by the post-monsoon (90.8 ppm) and pre-monsoon (89.25 ppm). Analysis of variance revealed seasonal variation of Zn concentration in sediment Zn (df=2, F=13.83, p < 0.05). Zn showed the negative correlation (r = -0.80, p < 0.01) with surface water phosphate (Table 3). Melville and Pulkownik (2006) and Billah et al. (2014) found a higher concentration of Zn than that of present findings (Fig. 3). The maximum content of Mn (481.80 ppm) was found in the post-monsoon (Fig. 4). Manganese did not show any significant relationship with any other parameters (Table 3). Kumar et al. (2016) found



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Fig. 4 Sediment manganese concentration $(\pm SD)$ variation at different seasons in different sampling stations of Sundarban mangrove estuary



Fig. 5 Sediment copper concentration (\pm SD) variation at different seasons in different sampling stations of Sundarban mangrove estuary

the higher content of Mn (740.95 µg/g) in the sediment of the Sundarban mangrove estuary than that of the present study. The higher concentration Cu was found in the postmonsoon season (49.11 ppm) followed by the monsoon (37.95 ppm) (Fig. 5). Significant seasonal variation of Cu was observed in sediment (df=2, F=75.34, p < 0.05) (Table 4).

Enrichment factor

The enrichment factor (EF) analysis has been commonly used to assess human-made contamination in sediment. It is based on the standardization of a tested element against a reference. In the present study, Fe was used as the reference element for EF calculations. The EF values of 0.5–1.5 reflect regional rock compositions, whereas EF values that are > 1.5 indicate non-crustal contributions and/or non-natural weathering processes (e.g., anthropogenic influences) (Zhang and Liu 2002; Zhang and Shan 2008). In monsoon values of EF for Zn were found greater than 1.5, which indicated anthropogenic impact on the Zn levels in sediment (Fig. 6). However, EF values of Mn were found lower than 1.5 indicating natural or crustal origin (Fig. 7). Copper also showed EF values greater than 1.5 in the monsoon season (Fig. 8), which suggested non-crustal origin of Cu.

Trace metal concentration in macroalgae

The maximum concentration (28,960 ppm) of Fe in macroalgae was found in the post-monsoon and the minimum



Fig. 6 Enrichment factor for Zinc



Fig. 7 Enrichment factor for Manganese



Fig. 8 Enrichment factor for copper









Fig. 10 Algal zinc concentration $(\pm SD)$ variation at different seasons in different stations of Sundarban mangrove estuary

in the pre-monsoon (4901.07 ppm) (Fig. 9). Significant seasonal variation for Fe in macroalgae was confirmed through ANOVA (df=2, F=9.77, p < 0.05). It showed significant positive correlation (r=0.84, p < 0.05) with Fe in sediment (Table 3). The content of Zn was found to be higher in the pre-monsoon (81 ppm) followed by the monsoon (66.82 ppm) and the post-monsoon (52.71 ppm) (Fig. 10). However, Zn concentration in macroalgae did not differ significantly (df=2, F=4.034, p > 0.05) among the sampling seasons. The maximum concentration of Mn (477.50 ppm) in macroalgae was found in the post-monsoon and the minimum in the monsoon (27.30 ppm) (Fig. 11). Profound seasonal variation of Mn in macroalgae was observed (df=2,



F = 112.83, p < 0.05). Mn also demonstrated significant positive correlation (r = 0.876, p < 0.01) with Fe in sediment and Fe in macroalgae (r = 0.71, p < 0.01). Chernova (2012) recorded the content of Mn (12–455 µg/g) in the macroalgae which mostly coincided with the findings of the present study. The concentration of Cu was found to be higher in the monsoon (42.72 ppm) and lower (1.45 ppm) in the pre-monsoon (Fig. 12). There was significant seasonal variation of Cu in macroalgae (df = 2, F = 85.32, p < 0.05). The Cu concentration showed significant negative correlation (r = -0.90, p < 0.01) with pH and positive correlation (r = 0.81, p < 0.01) with Cu in sediment (Table 3).





Fig. 12 Algal copper concentration $(\pm$ SD) variation at different seasons in different stations of Sundarban mangrove estuary

Conclusion

There has been very few information about the trace metal distribution in surface water and sediment and none for the macroalgae of the Sundarban. This might be the first initiative to assess trace metal accumulation in macroalgae in the Sundarban mangrove estuary in Bangladesh. The Sundarban mangrove estuary was (Dhangmari-Koromjol) found with more trace metals concentration than that of findings for other estuaries. This location is the entry point to the forest and possesses more anthropogenic activities. There revealed a succession of trace elements from sediment to macroalgae. Seasonal variation of trace element (Mn, Cu) was observed. The study has assessed the distribution of

trace metals in macroalgae and sediments, which possess the ability to impact the estuarine ecosystem of the Sundarban. The heterogeneous distribution of these elements in this region might be ascribed to anthropogenic inputs coupled with erosion and physical mixing of the sediments, oil spillage, the fast development of industries, discharges from ports and urban area etc. Findings of this study also suggest that as a potential bioindicator, mangrove macroalgae can be used to assess the status of the estuarine ecosystem.

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Compliance with ethical standards

Conflict of interest There is no conflict of interest regarding this manuscript.

Proper permission from authority Prior permission was taken from Divisional forest officer, Sundarban West Division, Khulna, Bangladesh.

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