

# Heavy Metals Concentration in Water, Sediments and their Bioaccumulations in Some Freshwater Fishes and Mussel in Dhaleshwari River, Bangladesh

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## ABSTRACT

The spatial and temporal distribution of heavy metals in water, sediment, fish and mussel of Dhaleshwari River, Bangladesh were determined by atomic absorption spectrophotometer. In water the concentrations of Ni, Pb, Cd, Cr and Cu varied seasonally and spatially from 5.47-9.74, 38.25-63.28, 5.29-8.20, 378.87-501.11 and 98.37-188.08 µg/L, respectively. The sediment also showed spatial and temporal variation of Ni, Pb, Cd, Cr and Cu ranges from 135.02-231.44, 58.19-70.26, 2.11-4.14, 95.76-141.27 and 31.53-76.52 mg/kg, respectively. These variations are likely to be due to different collection spots with point and non-point sources and seasons. The concentrations of Cd, Cr and Cu were higher, while Ni and Pb were lower in water than those of sediment. In *Trypauchen vagina*, a bottom living fish, the concentration of Ni, Pb, Cd, Cr and Cu varied seasonally from 6.35-9.56, 6.14-8.03, 0.51-0.73, 6.92-12.23 and 5.43-9.45 mg/kg, respectively. The concentrations of heavy metals in this fish were much lower than those of water and sediment. In *Glossogobius giuris*, also a bottom living fish, the concentration of Ni, Pb, Cd, Cr and Cu varied seasonally from 4.75-10.17, 4.25-8.17, 0.61-0.71, 7.15-11.92, and 5.17-7.48 mg/kg, respectively, which were slightly lower (except Cd) than those of *T. vagina*. In *Lamellidens marginalis*, a fresh water bivalve, the concentration of Ni, Pb, Cd, Cr and Cu varied seasonally from 6.07-11.32, 7.03-59.21, 0.56-7.23, 9.38-501.11 and 7.55-183.87 mg/kg, respectively. The concentrations of all the heavy metals studied were much higher in mussel than those of fish that indicate the greater rate of bioaccumulation in mollusc.

**Keywords:** aquatic pollution, *Glossogobius giuris*, *Lamellidens marginalis*, *Trypauchen vagina*

## INTRODUCTION

The Dhaleshwari River is a 160-Km-long tributary of the Jamuna River in central Bangladesh. It starts off the Jamuna near the northwestern tip of Tangail District. After that it divides into two branches: the north branch retains the name Dhaleshwari and merges with the other branch, the Kaliganga River at the southern part of Manikganj District. Finally the merged flow meets the Shitalakshya River near Narayanganj District. This combined flow goes southwards to merge into the Meghna River. Like most of the rivers of South Asian region, the water level of Dhaleshwari River remains very high during monsoon (July to October), whereas its level becomes low during pre- (April to June) and post-monsoon (November to March) due to reduced rain. Due to its geographical position, the Dhaleshwari River receives a large amount of domestic sewage, industrial waste, agricultural pesticides and pollutants from adjacent rivers, canals and direct run-off from land.

Indiscriminate use of potential toxic chemicals in agricultural fields ultimately drains into adjacent water bodies and are carried downstream through river waters. Their subsequent incorporation into the food chain, with biological magnification, at the highest tropic level, risks the stability of biota itself and also results in the disruption of biogeochemical cycles of the ecosystem. The problem may be exacerbated due to increased concentrations of toxicants during summer paddy cultivation when rivers have low discharge (Karim 1994).

Heavy metals that are introduced into the aquatic envi-

ronment are ultimately incorporated into aquatic sediments; organisms living in these sediments accumulate these heavy metals to varying degrees (Cross *et al.* 1970; Bryan and Hummerstone 1971). Contaminated sediments may be derived from inputs of suspended solids to which toxic substances are adsorbed, such as soil particles in surface water run-off from fields treated with pesticides. Alternatively, the natural suspended material in a watercourse as well as the river bed surface can adsorb chemicals from the water. When the suspended material settles out, the toxic material forms a sink or reservoir; the extent to which this can cause harm to aquatic life depends on the strength of the bond between the chemical and the particles. Substances with a very low solubility in water can be tightly bound and persist in the sediment for a long time. However, they may not be available for uptake by organisms in the sediment unless there is a direct route of entry through the skin surface or the gut. Although very low, the sediments are likely to continue for a long time at a slow steady rate.

Heavy metal contamination in an aquatic environment is of critical concern, due to toxicity of metals and their accumulation in aquatic habitats. Trace metals, in contrast to most pollutants, are not bio-degradable, and they undergo a global ecological cycle in which natural water are their main pathways. Of the chemical pollutants, heavy metals, being non-biodegradable, can be concentrated along the food chain, producing a toxic effect at points after far removed from the source of pollution (Tilzer and Khondker 1993). The toxic effect of such heavy metals, which are not utilized in the synthesis of new substances useful to orga-

nisms, lies in their ability to be stored up in enzymes and displace chemically similar elements. In this way vital biochemical reactions are blocked. Such an accumulation of heavy metals can harm the organism itself or can be transmitted to the trophic level of the food chain, where a similar toxic process can take place (Caspers 1975).

In West Bengal, India, due to rapid industrialisation and urbanisation of the city of Kolkata, Howrah and the newly developing Haldia complex, a negative impact has been exerted on the positive health of the ecosystem (Mitra and Choudhury 1993). Elevated concentration of heavy metals have been found in aquatic organism of Hooghly Estuary, India, where the sources of pollution were from natural as well as anthropogenic influences such as agricultural, industrial, urban input and sewage disposals (Bhattacharya *et al.* 1994). As a result, the numerous rivers and their tributaries that criss-cross Bangladesh may carry pollutants from the whole drainage area, including upstream areas in India, Nepal, Bhutan and China (Haque *et al.* 2006). Reduced flows in the dry season lessen the ability of the river Ganges to dilute and disperse pollutants from the whole drainage area. Growing industrialization on the bank of major river systems of Bangladesh has resulted in a considerable increase in water pollution. Different rivers have different trace metal concentrations. Usually during the monsoon trace metal concentrations are less and during pre- and post-monsoon trace metal concentrations are higher. The sediment load of Ganges-Brahmaputra-Meghna (GBM) river system consists exclusively of fine sand, silt and clay at their lower reaches within the Bengal basin, Bangladesh, and it deposited under uniformly fluctuating, unidirectional energy. The sediment has a close similarity in grain size with the sediments of the surrounding flood plain. The heavy mineral assemblage is dominated by unstable minerals which are mostly derived from high-rank metamorphic rocks. The mineral assemblage is dominated by quartz and feldspars. Illite and kaolinite are the major clay minerals and occur in almost equal proportion in bed sediments. Although the sediments of the GBM river system in the Bengal basin has the potential to trap contaminants because of their grain size and mineralogy, they are not heavily contaminated at the moment but considering the population density and fertilizer input they can be in future heavily contaminated (Haque *et al.* 2007). A quantitative analysis of sediments can reflect upon the current feature of a aquatic system whether the sediment behaves as a sink or sources of metals depends on the dynamics of the geo-chemical processes that occur at the sediment water interface under oxic/anoxic conditions (Santschi *et al.* 1990).

As a riverine country 230 rivers have created an intricate river system in Bangladesh. However, limited data are available on the impact of agriculture, domestic and industrial pollution, especially of heavy metals on aquatic environment. In Bangladesh the concentration of heavy metals in aquatic animals, water and sediment were studied by Sharif *et al.* (1991, 1992a, 1992b, 1993), Chowdhury (1994), Holmgren (1994), Hossain (1996), Khan *et al.* (1998), Biswas *et al.* (1998), Ahmed (2000), Ahmed *et al.* (2002, 2003, 2009a, 2009b), Haque *et al.* (2003, 2004, 2005, 2006, 2007). Their studies covered mostly coastal areas, parts of the GBM river system and some rivers of central parts. However, to date no systematic study yet has been carried out for heavy metal concentration in the Dhaleswari River.

The objectives of this study were to determine the concentration of heavy metals: nickel (Ni), copper (Cu), chromium (Cr), cadmium (Cd) and lead (Pb) from water, sediments, fish and mussels from Dhaleswari River, Bangladesh.

## MATERIALS AND METHODS

During pre-monsoon, monsoon and post-monsoon of 2001-2002, samples of water, sediment, two species of adult fish, *Trypauchen vagina* (Bloch and Schneider 1801) locally called cheua and *Glos-*

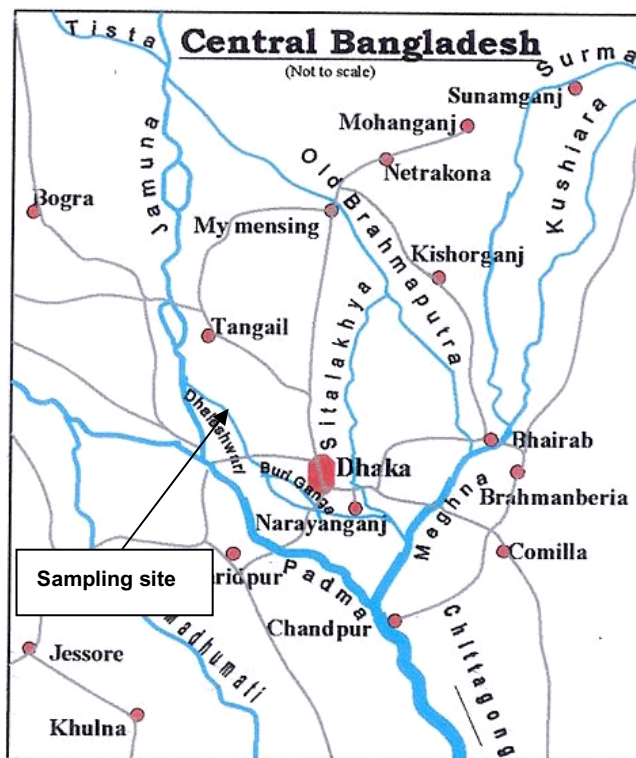


Fig. 1 Location map of the Dhaleswari River in central Bangladesh.

*sogobius giuris* (Hamilton 1822) locally called bila and one species of adult bivalve mollusc (freshwater pearl mussel; *Lamellidens marginalis*) were collected from three stations of Dhaleswari River, namely Station 1 (in the Buriganga-Dhaleswari confluence; locally called old launch ghat), Station 2 (in the Shitalakkha-Dhaleswari confluence; locally called launch ghat) and Station 3 (in the Dhaleswari-Meghna confluence; locally called Char-Kishorganj) (Fig. 1).

Both *T. vagina* and *G. giuris* are under the same family (Gobiidae) of order Perciformes. Cheua is demersal, occurs along the bottom in tidal rivers and estuaries and stays close to a self-dug burrow, while bila is benthopelagic, found in clear to turbid streams with rock, gravel or sand bottoms, feeds on small insects, crustaceans and small fish (Shafi and Quddus 1982).

The water samples were collected with the help of 1 L plastic bottles, which were previously soaked with a mixture of distilled water (975 ml), 2% HNO<sub>3</sub> (20 ml) and H<sub>2</sub>SO<sub>4</sub> (5 ml) for about 24 h. On the next day these bottles were scrupulously washed several times with distilled water, rinsed with double distilled water and then kept in an oven (60-70°C). The bottles were then ready for sample collection. The sediment samples were collected by a vertical corer and an Ekman grab sampler. The water and sediment samples were acidified immediately with 2 ml of HNO<sub>3</sub> per litre of water and preserved in refrigerator at 4°C for laboratory analysis. The fishes which were supplied by the local fishermen collected by netting, were identified according to Shafi and Quddus (1982). All samples were taken in different polythene bags. The mollusc samples were collected with an Ekman grab sampler, stake net (locally called Behundi net) and by hand. After collection mud and other fouling substances were removed and the molluscs were then cut open with the aid of a Teflon-coated stainless steel knife and then dried to room temperature and weighed. The soft flesh was dried in an oven at 90°C to an anhydrous state until a constant weight (dry weight) was obtained. After collection the sediment and fish samples were washed, weighed and dried in an oven at 105°C until gaining constant weight. After cooling in a dessicator, all the samples were ground by a carbide mortar and pestle to make powder and homogenised. The powdered sediment, fish and mussel samples were finally stored in a pre-cleaned dry plastic bottle and preserved in a dessicator for further analysis.

For the quantitative analyses of Ni, Cu, Cr, Cd and Pb, the samples were digested. Dried (0.5 g) powder was placed in a long test tube. Then 1 ml 70% HClO<sub>4</sub>, 4 ml concentrated HNO<sub>3</sub> and 1.5

**Table 1** Heavy metal concentrations with average ( $\pm$  standard deviation) in the water of Dhaleswari River.

Sampling season	Sampling station	Concentrations of heavy metals ( $\mu\text{g/L}$ )				
		Ni	Pb	Cd	Cr	Cu
Pre-monsoon (May)	Station-1	7.58	39.42	6.21	467.19	188.08
	Station-2	5.47	63.28	8.20	451.08	181.21
	Station-3	6.07	54.19	7.23	501.11	152.64
Monsoon (August)	Station-1	6.02	61.52	7.02	378.87	166.65
	Station-2	7.92	47.25	5.29	432.79	129.36
	Station-3	6.41	45.17	6.19	415.03	183.87
Post-monsoon (February)	Station-1	9.74	42.18	5.74	387.35	164.12
	Station-2	8.83	38.25	6.27	447.58	98.37
	Station-3	6.87	59.21	6.27	491.13	127.98
Average $\pm$ SD		7.21 $\pm$ 1.42	50.05 $\pm$ 19.28	6.49 $\pm$ 0.87	441.34 $\pm$ 42.48	154.69 $\pm$ 30.48
Range		5.47 - 9.74	38.25 - 63.28	5.29 - 8.20	378.87 - 501.11	98.37 - 188.08

ml concentrated  $\text{H}_2\text{SO}_4$  were added into the test tube. The samples were then heated gently in an oil bath ( $\sim 100^\circ\text{C}$ ) until the solid mass dissolved. If sample was not clear, 4 ml of  $\text{HNO}_3$  was added into the test tube and repeated until the solution was clear. Finally, the mixture was boiled at about  $210^\circ\text{C}$  in a paraffin oil bath in order to drive off the acids except  $\text{H}_2\text{SO}_4$  and then it was cooled down at room temperature. Blank digestion was also performed to quantify possible contamination during sample preparation and analysis. A standard solution of the elements Ni, Cu, Cr, Cd and Pb were prepared by pipetting the required amount of the solution from the stock solution, manufactured by Fisher Scientific Co., USA. The standard solution was prepared before every analysis of the current work. The samples were analyzed by using air acetylene flame with combination, as well as single element hollow cathode lamps into an atomic absorption spectrophotometer (Shimadzu, AAS-6800). The analytical quality of the work was checked by analysis of standard reference materials: NBS-SRM-1573, tomato leaves and NBS-SRM-1566, oyster tissue, prepared by the National Bureau of Standards, Washington DC, USA. The analytical procedures were also calibrated against the above standard reference materials. The average recovery ranged between 94 to 107%.

Statistical software SPSS ver. 12 was used to analyse the data using ANOVA followed by LSD *post-hoc* for multiple comparison at 5% level of significance to determine significant difference among mean values. In addition, the correlation matrix was carried out by Microsoft Excel.

## RESULTS AND DISCUSSION

Among all the five heavy metals studied, the concentrations of Cd, Cr and Cu were higher, while, Ni and Pb were lower in water than those of sediment. However, in comparison with heavy metals in fish and mussels, all metals were much higher in water except for Ni.

### Heavy metals in water

**Table 1** demonstrates the heavy metal concentrations with average ( $\pm$  standard deviation) in Dhaleswari River water. Cr was the most abundant ( $441.34 \pm 42.48 \mu\text{g/L}$ ), while Cd was the least abundant ( $6.49 \pm 0.87 \mu\text{g/L}$ ) heavy metal. The Cr concentration was higher ( $501.11 \mu\text{g/L}$ ) during pre-monsoon at station 3 but it was lower ( $378.87 \mu\text{g/L}$ ) during monsoon at station 1. These variations are likely to be due to different collection spots and seasons. However, the metals in water did not differ significantly ( $P > 0.05$ ) in pre-monsoon, monsoon and post-monsoon. A correlation matrix shows a very significant positive correlation between Cu and Pb, but very significant negative correlations between Ni and Pb as well as Cu and Ni (**Table 2**). Khan *et al.* (1998) found the concentrations of Cr ranged from 0.015 to  $0.491 \mu\text{g/ml}^{-1}$  in the water of the GBM estuary, values that are lower than the present study.

Cd concentration was higher ( $8.20 \mu\text{g/L}$ ) during pre-monsoon at station 2 and lower ( $5.29 \mu\text{g/L}$ ) during monsoon at the same station. These variations might be due to variation of collection time. Ahmed (1998) reported the

**Table 2** Correlation matrix of heavy metals in Dhaleswari River water.

	Pb	Cd	Ni	Cu	Cr
Pb	1				
Cd	0.6760*	1			
Ni	-0.9997*	-0.6935*	1		
Cu	0.9994*	0.7018*	-0.9999*	1	
Cr	0.1418	0.8253*	-0.1657	0.1769	1

\* Values  $>0.5$  or  $<-0.5$  are significantly correlated

concentration of Cd ranges between 0.018 and  $0.007 \text{ ppm}$  in water of the Sundarban Forest Reserve. Rao *et al.* (1985) found the range between 0.9 and  $1.3 \mu\text{g/g}$  in dissolved form and 6.6 and  $18 \mu\text{g/g}$  in particulate form. Values ranging between 0.2 and  $0.4 \mu\text{g/g}$  have been reported from Ostfjorden, Norway and Liverpool Bay, UK by Peterson *et al.* (1972) and Rojahn (1972), respectively. According to Environmental Quality Standard (EQS) the standard value of Cd for coastal water of Bangladesh is  $0.3 \mu\text{g/g}$  (EQS 1991). Khan *et al.* (1998) found the concentration of Cd ranges from 0.001 to  $0.107 \mu\text{g.ml}^{-1}$  in the water of the GBM estuary, which are much lower than the present study.

Ni was the second least abundant ( $7.21 \pm 1.42 \mu\text{g/L}$ ) heavy metal in the waters of the Dhaleswari River. A higher concentration ( $9.74 \mu\text{g/L}$ ) was found at station 1 during post-monsoon while a lower concentration ( $5.47 \mu\text{g/L}$ ) was found at station 2 during pre-monsoon.

The Pb concentration was maximum ( $63.28 \mu\text{g/L}$ ) during pre-monsoon at station 2, but it was minimum ( $38.25 \mu\text{g/L}$ ) during post-monsoon at the same station. However, variations of concentration at different stations in different seasons were high. Khan *et al.* (1998) found that the concentration of Pb ranged from 0.012 to  $0.431 \mu\text{g.ml}^{-1}$ , which are much lower than the current study.

Cu was the second most abundant ( $154.69 \pm 30.48 \mu\text{g/L}$ ) heavy metal in the water of the Dhaleswari River. However, variations of concentration at different stations in different seasons were significant ( $P < 0.05$ ). Ahmed (1998) reported that the concentration of Cu ranged between 0.11 and 0.021 ppm in water of the Sundarban Forest Reserve. Rao *et al.* (1985) found that the Cu concentration ranged between 6.8 and  $30.6 \mu\text{g/L}$  in dissolved and 822 and  $1801 \mu\text{g/g}$  in particulate form in Vishakhapatnam. The standard for Cu given by EQS is  $0.03 \mu\text{g/g}$ . The present findings differ greatly from above authors, which marks the state of Cu pollution in the Dhaleswari River.

Ahmed *et al.* (2009a) studied the heavy metal concentration in water of the Shitalakhya River, Bangladesh and found the concentration of Ni ranged from 4.31 to  $7.83 \mu\text{g/L}$ , Pb 41.24 to  $63.15 \mu\text{g/L}$ , Cd 7.12 to  $10.11 \mu\text{g/L}$ , Cr 192.18 to  $234.32 \mu\text{g/L}$ , and Cu 156.38 to  $254.07 \mu\text{g/L}$ , which are more or less similar with the present study and the findings of Islam (2002), who investigated the heavy metal concentration in the water of Buriganga River, Bangladesh.

**Table 3** Heavy metal concentrations (dry weight basis) with average ( $\pm$  Standard Deviation) in sediment of Dhaleswari River.

Sampling Seasons	Sampling Stations	Concentrations of heavy metals (mg/kg)				
		Ni	Pb	Cd	Cr	Cu
Pre-monsoon (May)	Station-1	182.13	58.19	4.14	96.85	36.53
	Station-2	195.08	64.51	3.71	95.76	76.52
	Station-3	141.29	62.90	2.92	110.25	33.65
Monsoon (August)	Station-1	135.02	63.56	2.11	113.54	32.54
	Station-2	183.51	68.93	3.18	141.27	31.53
	Station-3	231.44	60.47	3.95	136.71	55.02
Post-monsoon (February)	Station-1	153.52	70.26	2.95	121.74	40.41
	Station-2	221.45	63.18	3.05	132.24	36.82
	Station-3	186.10	66.05	3.13	109.76	53.46
Average $\pm$ SD		181.06 $\pm$ 33.27	64.22 $\pm$ 3.80	3.23 $\pm$ 0.61	117.56 $\pm$ 19.57	44.05 $\pm$ 14.93
Range		135.02 - 231.44	58.19 - 70.26	2.11 - 4.14	95.76 - 141.27	31.53 - 76.52

## Heavy metals in sediment

The heavy metal concentrations in the sediment of Dhaleswari River are given in **Table 3**. The most noticeable differences between the heavy metal concentrations in water and sediment are the much higher concentration of Ni and much lower concentration of Cr and Cu in sediment than those of water. Like water, Cd was also the least abundant heavy metal in sediment. Cu and Ni showed greater variation in concentrations in different seasons and at different sampling stations. However, Pb, Cd, Ni, and Cu in sediment did not differ significantly ( $P > 0.05$ ) in pre-monsoon, monsoon and post-monsoon. Only Cr of sediment in pre-monsoon was significantly ( $P < 0.05$ ) different than that of monsoon. The correlation matrix shows a very significant positive correlation between Ni and Pb, but very significant negative correlations between Cd and Pb, Cr and Cd as well as Cr and Cu (**Table 4**).

In the Dhaleswari River sediment average concentration of Ni was  $181.06 \pm 33.27$  mg/kg. The highest amount (231.44 mg/kg) was found at station 3 during monsoon, while the lowest (135.02 mg/kg) was found at station 1 during the same season. Although the average concentration of Pb was  $64.22 \pm 3.80$  mg/kg, the maximum (70.26 mg/kg) was noticed during post-monsoon at station 1, whereas, the minimum (58.19 mg/kg) was during pre-monsoon at the same station. Cd concentration was higher (4.14 mg/kg) at station 1 during pre-monsoon, on the other hand its concentration was lower (2.11 mg/kg) at the same station during monsoon, while average was  $3.23 \pm 0.61$  mg/kg. In the Dhaleswari River sediment average concentration of Cr was  $117.56 \pm 19.57$  mg/kg. The highest amount (141.27 mg/kg) was found at station 2 during monsoon, while the lowest (95.76 mg/kg) was found at the same station during pre-monsoon. Although the average concentration of Cu was found to be  $44.05 \pm 14.93$  mg/kg, the maximum (76.52 mg/kg) was noticed during pre-monsoon at station 2, whereas, the minimum (31.53 mg/kg) was during monsoon at the same station.

Sediments are the sources of organic and inorganic matter in the river, estuaries, and oceans. Bottom sediments have the capacity to exchange cations with the surrounding water medium. Trace elements, especially heavy metals, are concentrated in the surface micro-layer by transport of particle-associated bubbles. Despite injection of trace elements by streams, whether natural or artificial, since most of the inputs becomes trapped in the sediments and incorporated mainly into organic matter, most of the cationic forms have very little chance of leaving an estuarine and coastal area in solution (Haque *et al.* 2007). Any contaminant element is normally introduced into the sediments in a physico-chemical form, which permits it to enter the natural cycling of element in the discharge ecosystem (Segar and Pellenberg 1973).

The bio-availability of heavy metals may vary widely depending on sediment characteristics, water chemistry, hydrography, biological factors, etc. The deposits reflect the biological, chemical and physical condition of water body

**Table 4** Correlation matrix of heavy metals (dry weight basis) in Dhaleswari River sediment.

	Pb	Cd	Ni	Cu	Cr
Pb	1				
Cd	-0.9116*	1			
Ni	0.9725*	-0.9822*	1		
Cu	-0.6053*	0.8790*	-0.7739*	1	
Cr	0.6966*	-0.9299*	0.8444*	-0.9928*	1

\* Values  $> 0.5$  or  $< -0.5$  are significantly correlated

and become a signature to the state of heavy metal concentration in aqueous system (Ahmed *et al.* 2002). Many of these metals (for example, Co, Cu, Mn, Mo, Ni and Zn) are essential trace elements for aquatic organisms and are involved in biochemical processes such as enzyme activation. However, although essential in small amounts, many are toxic at only slightly elevated free ion concentrations (Furnas 1992). Others such as Cd, Pb and Hg have no known biological roles and are detrimental to essential life processes (Burden-Jones and Denton 1984).

Ahmed *et al.* (2009a) studied the heavy metal concentration in sediment of the Shitalakhya River, Bangladesh and found that the concentration of Ni ranged from 120.96 to 131.94 mg/kg, Pb 54.52 to 65.90 mg/kg, Cd 1.71 to 2.17 mg/kg, Cr 60.09 to 91.02 mg/kg, and Cu 56.07 to 91.51 mg/kg in dry weight basis, which are slightly lower than the present investigation, except for Cu.

Ahmed (1998) found the highest and lowest concentration of Pb as 61.66 and 10.96 mg/kg, Cd as 0.817 and 0.121  $\mu\text{g/g}$ , Cu as 50.5 and 6.89  $\mu\text{g/g}$  and Cr as 120.8 and 20.2  $\mu\text{g/g}$ , in sediment of the Sundarban Forest Reserve. Khan *et al.* (1998) reported the Pb concentration ranged from 2.355 to 26.086 mg/kg in sediment in the Ganges-Brahmaputra-Meghna Estuary.

The Cd concentration in the Sediments of the Baltic Sea ranged between 0.2 and 2.2  $\mu\text{g/g}$ , while the values for the North Sea are below 1  $\mu\text{g/g}$  for 80% of the samples (Nicholson and Moore 1981). Mehedi (1994) found that the concentration of Cd ranged from 0.8 to 0.88  $\mu\text{g/g}$  and that of Cu ranged from 3.8 to 60.88  $\mu\text{g/g}$  in the ship-breaking area of Chittagong. Chester and Stonar (1975) found as high as 2424  $\mu\text{g/g}$  of Cu and 624  $\mu\text{g/g}$  of Cr in sediments of Solfjord, West Norway. Bryan and Hummerstone (1977) studied the heavy metal concentration in the Looe estuary, Cornwall, UK and found the highest concentration of Cr as 5  $\mu\text{g/g}$ .

According to Legorburu and Canton (1992) the standard values from the analysis of marine sediment (SD-N-1/2), in the IAEA laboratory in Monaco, were 120 mg/kg for Pb, 11  $\mu\text{g/g}$  for Cd, 72  $\mu\text{g/g}$  for Cu and 149  $\mu\text{g/g}$  for Cr. The present investigations show lower concentration of the above heavy metals except for Cu. However, the certified value of Pb from the USSR standard reference material No. 2499 - 83, Soil SDPS-2 is  $7 \pm 5$   $\mu\text{g/g}$  and the present study did not find concentrations of Pb lower than the certified value and therefore harmful for soil and benthic community.

**Table 5** Heavy metal concentrations (dry weight basis) with average ( $\pm$  Standard Deviation) in cheua (*Trypauchen vagina*) of Dhaleswari River.

Sampling Seasons	Sampling Stations	Concentrations of heavy metals (mg/kg)				
		Ni	Pb	Cd	Cr	Cu
Pre-monsoon (May)	Station-1	6.35	7.08	0.58	7.11	5.58
	Station-2	9.56	6.14	0.73	9.26	8.27
	Station-3	7.08	6.17	0.51	8.17	7.61
Monsoon (August)	Station-1	7.13	6.98	0.51	11.42	5.91
	Station-2	8.45	7.43	0.56	9.37	5.43
	Station-3	7.19	6.46	0.65	9.13	6.25
Post-monsoon (February)	Station-1	6.53	8.03	0.51	10.15	9.45
	Station-2	7.82	7.14	0.51	6.92	5.87
	Station-3	7.06	6.49	0.55	12.23	8.52
Average $\pm$ SD		7.46 $\pm$ 1.01	6.88 $\pm$ 0.62	0.56 $\pm$ 0.07	9.30 $\pm$ 1.79	6.98 $\pm$ 1.49
Range		6.35 - 9.56	6.14 - 8.03	0.51 - 0.73	6.92 - 12.23	5.43 - 9.45

## Heavy metals in fish

**Table 5** depicts the heavy metals concentrations in cheua (*Trypauchen vagina*) of Dhaleswari River. The concentrations of heavy metals in cheua were much lower than those of water and sediment. However, among the five heavy metals studied in the whole body of cheua, Cr was the highest ( $9.30 \pm 1.79$  mg/kg), and Cd was the lowest ( $0.56 \pm 0.07$  mg/kg), while Ni, Pb and Cu were moderate in concentration compared to other heavy metals for this study. The Pb, Cd, Ni, Cu and Cr in cheua did not differ significantly ( $P > 0.05$ ) in pre-monsoon, monsoon and post-monsoon. The correlation matrix shows both positive and negative significant correlations among heavy metals (**Table 6**). According to Nauen (1983) the maximum consumption limit of Cd, Cr, Cu, Ni, Pb in aquatic animals are 1.2, 8, 120, 1 and 8 mg/kg, respectively. Therefore, some values of Cr, all the values of Ni, and one value of Pb exceeded the consumption limit thus not safe for human consumption. The much higher concentration of heavy metal in these fish are likely due to surrounding water and sediment which are polluted by industrial, agriculture and domestic waste originating from surrounding as well as upstream environment. Aquatic organisms living in the sediment accumulate heavy metals to varying degrees (Bryan and Hummerstone 1977).

An organism can uptake metals directly or with food particles from the water body and then these metals bind to the different parts of the body and may cause harmful effects. The spatial and temporal variations of heavy metals concentrations in the body of cheua were evident in the case of Cr and Cu.

The heavy metal concentrations in bila (*Glossogobius giuris*) of the Dhaleswari River are presented in **Table 7**. The heavy metal concentrations were slightly lower in bila (except for Cd) than those of cheua. Moreover, bila contained a much lower concentration of heavy metals than the water and sediment. However, like cheua, among the five heavy metals studied in the whole body of bila, Cr was the highest ( $8.84 \pm 1.74$  mg/kg), and Cd was the lowest ( $0.66 \pm 0.03$  mg/kg), while Ni, Pb and Cu were moderate in concentration. The Cd and Cr levels in bila did not differ significantly ( $P > 0.05$ ), whereas Pb, Ni and Cu differed signifi-

**Table 6** Correlation matrix of heavy metals (dry wt. basis) in cheua (*Trypauchen vagina*) of Dhaleswari River.

	Pb	Cd	Ni	Cu	Cr
Pb	1				
Cd	-0.9586*	1			
Ni	-0.8428*	0.9611*	1		
Cu	0.2115	-0.4809	-0.7044*	1	
Cr	0.8981*	-0.7357*	-0.5201*	-0.2399	1

\* Values  $>0.5$  or  $<-0.5$  are significantly correlated

cantly ( $P < 0.05$ ) in pre-monsoon, monsoon and post-monsoon. The correlation matrix shows both positive and negative correlations among heavy metals (**Table 8**). Cd levels usually increase with age (IPCS 1992). The acute toxicity of Cd to aquatic organisms is variable, even between closely related species, and is related to the free ionic concentration of the metal. Cd interacts with calcium (Ca) metabolism, and in fish it causes abnormally low Ca levels (hypocalcaemia), probably by inhibiting Ca uptake from the water. However, high Ca concentrations in the water protect fish from Cd uptake by competing at uptake sites. Effects of long-term exposure can include larval mortality and temporary reduction in growth (AMAP 1998).

Şirelil *et al.* (2006) measured the concentrations of Cd and Pb in vacuum-packaged smoked fish species (mackerel, *Salmo salar* and *Oncorhynchus mykiss*); these varied from 0.003 to 0.036 mg/kg with a mean of 0.01367 mg/kg for Cd, and 0.001 to 0.791 mg/kg with a mean of 0.17710 mg/kg for Pb. Ashraf (2006) studied 57 samples of canned tuna fish and found the concentration of Pb, Cr, Cd, Cu, Ni, and

**Table 8** Correlation matrix of heavy metals (dry wt. basis) in bila (*Glossogobius giuris*) of Dhaleswari River.

	Pb	Cd	Ni	Cu	Cr
Pb	1				
Cd	-0.6053*	1			
Ni	0.9187*	-0.2417	1		
Cu	0.5829*	0.2938	0.8565*	1	
Cr	0.9921*	-0.7003*	0.8619*	0.4766	1

\* Values  $>0.5$  or  $<-0.5$  are significantly correlated

**Table 7** Heavy metal concentrations (dry weight basis) with average ( $\pm$  Standard Deviation) in bila (*Glossogobius giuris*) of Dhaleswari River.

Sampling Seasons	Sampling Stations	Concentrations of heavy metals (mg/kg)				
		Ni	Pb	Cd	Cr	Cu
Pre-monsoon (May)	Station-1	8.27	8.17	0.68	7.15	6.36
	Station-2	7.45	6.09	0.71	9.07	7.28
	Station-3	10.17	6.43	0.61	11.19	7.48
Monsoon (August)	Station-1	5.19	5.60	0.69	8.17	5.52
	Station-2	4.75	4.25	0.62	7.23	5.25
	Station-3	4.80	4.85	0.69	7.92	6.03
Post-monsoon (February)	Station-1	9.46	8.16	0.64	9.46	6.36
	Station-2	6.03	6.30	0.67	11.92	6.28
	Station-3	7.23	7.18	0.67	7.46	5.17
Average $\pm$ SD		7.03 $\pm$ 2.00	6.33 $\pm$ 1.35	0.66 $\pm$ 0.03	8.84 $\pm$ 1.74	6.19 $\pm$ 0.82
Range		4.75 - 10.17	4.25 - 8.17	0.61 - 0.71	7.15 - 11.92	5.17 - 7.48



**Table 9** Heavy metal concentrations (dry weight basis) with average ( $\pm$  Standard Deviation) in mollusc (freshwater pearl mussel; *Lamellidens marginalis*) of Dhaleswari River.

Sampling Seasons	Sampling Stations	Concentrations of heavy metals (mg/kg)				
		Ni	Pb	Cd	Cr	Cu
Pre-monsoon (May)	Station-1	11.32	10.25	0.69	13.52	8.15
	Station-2	10.25	9.62	0.71	9.38	8.33
	Station-3	6.07	54.19	7.23	501.11	152.64
Monsoon (August)	Station-1	11.04	12.18	0.56	13.66	9.68
	Station-2	10.09	11.39	0.80	11.64	7.70
	Station-3	6.41	45.17	6.19	415.03	183.87
Post-monsoon (February)	Station-1	11.15	7.03	0.72	12.51	11.51
	Station-2	9.55	11.19	0.62	13.29	7.55
	Station-3	6.87	59.21	6.27	491.13	127.98
Average $\pm$ SD		9.19 $\pm$ 2.14	24.47 $\pm$ 21.63	2.64 $\pm$ 2.95	164.58 $\pm$ 229.59	57.49 $\pm$ 74.35
Range		6.07 - 11.32	7.03 - 59.21	0.56 - 7.23	9.38 - 501.11	7.55 - 183.87

Hg ranged between 0.14 and 0.82, 0.10 and 0.57, 0.08 and 0.66, 0.02 and 0.33, 0.09 and 0.48 and 0.18 and 0.86 mg/kg respectively, which are much lower than the present findings. Burgera and Gochfeld (2005) found As, Cd, Pb, Mn and Hg ranged from 0.23 to 3.3, 0.0001 to 0.01, 0.04 to 0.12, 0.1 to 1.0 and 0.05 to 0.6 ppm, respectively in the flounder, bluefish, yellow-fin tuna, Chilean sea bass, cod, croaker, porgie, red snapper, whiting, shrimp (large and small), and scallops of New Jersey, USA, which are also less than the current findings. They found interspecific differences in levels of metals for all metals. However, the same metals in fish did not have the highest values for more than two metals. They suggested that the differences were due to geography, trophic level, size, foraging method/location, and propensity of metals to undergo biomagnification in the food chain.

Ahmed *et al.* (2009a) studied the heavy metal concentration in *G. giuris* of the River Shitalakhya, Bangladesh and found the concentration of Ni ranged from 10.53 to 11.32 mg/kg, Pb - 7.17 to 9.17 mg/kg, Cd - 0.81 to 1.07 mg/kg, Cr - 3.13 to 3.37 mg/kg, and Cu - 5.19 to 6.03 mg/kg on a dry weight basis, which are close to the present study (except for Cr) and the findings of Islam (2002), who investigated the heavy metal concentration in *G. giuris* of Buriganga River, Bangladesh.

Sharif *et al.* (1992b) studied the heavy metal concentration in *T. vagina* and found the concentration of Ni, Cu, Pb and Cd as 2.4  $\pm$  0.00, 3.76  $\pm$  0.48, 2.42  $\pm$  0.27 and 0.11  $\pm$  0.00 mg/kg (dry weight basis), which are much lower than the present study. There may be several reasons for this; firstly, unlike the current study where fish were collected from one of the polluted rivers, in Sharif *et al.*'s study, fish bought from auction markets originated from variable habitats. Secondly, only the flesh of fish, which contains relatively less amount of heavy metal than other parts of the body (Wright 1976), was analysed, while on the other hand, the whole body was analysed in the current experiment. Moreover, the size was variable (100-500 g), whereas the present study was conducted with adult fish. Sharif *et al.* (1992a, 1993) found much lower concentrations of Pb, Cd and Cr in different marine fish such as *Lates calcarifer*, *Pangasius pangasius*, *Polynemus indicus*, *Ilisha megaloptera*, *Raconda russelliana*, *Leander styliferus*, *Rastrelliger kanagurta*, *Scomberomorus guttatum*, *Polynemus paradiseus* and *Usteogeniosus militaris* of the Bay of Bengal than those of our investigation and well below the permissible levels for human consumption. These variations are likely to be due to habitat differences, in addition to the reasons mentioned above for Sharif *et al.* (1992b).

In *Puntius sophore* from the Buriganga River, Haque *et al.* (2003) found seasonal variation of Pb, Cd, Cu and Ni ranged from 1.46 to 3.07, 0.24 to 0.41, 3.59 to 5.66 and 0.94 to 4.59 mg/kg, respectively in 1999. In the same river they also found seasonal variation of Pb, Cd, Cu and Ni ranged from 1.20 to 3.41, 0.24 to 0.58, 3.71 to 5.69 and 1.17 to 8.68 mg/kg, respectively in *Mystus vittatus* in 1999. These concentrations are much lower than the present study,

except for Cu. These variations may be due to the species and age of fish, temporal variation of sampling and use of different methods for sample analysis (spectrophotometer).

Haque *et al.* (2006) studied the seasonal variation of heavy metal concentrations in *Gudusia chapra* inhabiting the Sundarban mangrove forest and found the concentration of Cu, Zn, Fe, Pb, Cd, Cr and Ni seasonally varied from 0.527 to 3.99, 5.34 to 25.9, 0.038 to 0.221, 0 to 3.396 and 0.176 to 89.5  $\mu$ g/g dry weight basis, respectively. The concentrations of these heavy metals are well below those of the present findings.

Holden and Topping (1972) found that Pb concentrations ranged from 0.5 to 1.0 mg/kg on a wet weight basis of different fish species. Khan *et al.* (1987) found that Ni concentration varied between 2.70 to 15.2 mg/kg on a dry weight basis for five different species of fish. Zindge *et al.* (1979) found that the Cu concentration ranged between 2.80 and 12.40 ppm in different fish species. Khan *et al.* (1987) found that Cu varied between 0.65 and 58.1 mg/kg on a dry weight basis for five different species of fish in the Bay of Bengal region of Bangladesh coast. Roth and Hornung (1977) found that Cr varied between 2.8 to 4.9 mg/kg on a wet weight basis for different species of fish.

### Heavy metals in molluscs

**Table 9** shows the heavy metals concentrations in freshwater pearl mussel (*Lamellidens marginalis*) of Dhaleswari River. The Pb, Cd, Ni, Cu and Cr concentrations in mussel were not significantly different ( $P > 0.05$ ) in pre-monsoon, monsoon and post-monsoon. The correlation matrix shows both positive and negative significant correlations among heavy metals (**Table 10**). The most noticeable thing is that the concentrations of all the heavy metals studied are much higher in mussel than in fish. Moreover, the concentration of Ni in mussel was even higher than that of water and Cr concentration was also higher in mussel than in sediment. The maximum consumption limits of heavy metals in aquatic animals indicate that, except for Cu, almost all the values of other heavy metals exceeded the consumption limit (Nauen 1983) and harmful for human consumption. Molluscs are known to concentrate metals in their body shells to varying proportions depending upon the species, environmental condition, detoxification and inhibitory processes (Haque *et al.* 2007).

Ahmed *et al.* (2009a) studied the heavy metal concen-

**Table 10** Correlation matrix of heavy metals (dry wt. basis) in mollusc (freshwater pearl mussel; *Lamellidens marginalis*) of Dhaleswari River.

	Pb	Cd	Ni	Cu	Cr
Pb	1				
Cd	0.1773	1			
Ni	0.9935*	0.2879	1		
Cu	-0.9997*	-0.1554	-0.9908*	1	
Cr	0.8910*	0.6047*	0.9368*	-0.8808*	1

\* Values  $>0.5$  or  $<-0.5$  are significantly correlated

tration in *L. marginalis* of the River Shitalakhya, Bangladesh and found that the concentration of Ni ranged from 8.19 to 9.07 mg/kg, Pb - 9.16 to 13.09 mg/kg, Cd - 1.09 to 1.21 mg/kg, Cr - 8.12 to 9.07 mg/kg, and Cu - 5.47 to 8.19 mg/kg on a dry weight basis, which are close to the values detected in the present study, except for Cr and Cu, which are much higher than the values detected in the current investigation. Like the present study, Ahmed *et al.* (2009a) also mentioned that the variations in concentrations are likely to be due to spatial and temporal variation of sampling.

Ni concentration in mussel varied seasonally from 6.07 (in station 3 during pre-monsoon) to 11.32 mg/kg (in station 1 during the same season) with an average of  $9.19 \pm 2.14$  mg/kg. Sericano *et al.* (2001) reported the mean concentration of Ni in molluscs (bivalves) from Kara Sea and its adjacent Rivers (Yenisey and Ob rivers) of Russia as  $6.4 \pm 2.7 \mu\text{g.g}^{-1}$ . Bryan and Hummerstone (1977) studied heavy metal concentration in molluscs (*Littorina littorea*) from the Looe estuary, Cornwall, UK and found that the concentrations of Ni varied from 2.2 to 4.1  $\mu\text{g.g}^{-1}$ . Bustamante *et al.* (2000) reported that the mean concentration of Ni in molluscs from New Caledonia was  $6.7 \pm 3.0 \mu\text{g.g}^{-1}$ . Páez-Osuna *et al.* (1993) found the average concentration of Ni in molluscs from NW Coast Mexico to be  $3.8 \mu\text{g.g}^{-1}$ . Ahmed *et al.* (2009a) studied heavy metal concentrations in molluscs (bivalves) from the Shitalakhya River, Bangladesh and found that the concentrations of Ni varied seasonally from 8.19 to 9.07  $\mu\text{g.g}^{-1}$ . The close concentration of Ni of these two rivers are might be due to the fact that both receive domestic and industrial wastes from same sources mainly Dhaka city. Goldberg *et al.* (1983) found that the average concentration of Ni in molluscs from the Gulf Coast, USA was 1741  $\mu\text{g.g}^{-1}$ . Páez-Osuna and Marmolejo-Rivas (1990) found that the average concentration of Ni in molluscs, *Saccrostrea iridescens* from the Coast of Mazatlan, Sinaloa, Mexico was 424  $\mu\text{g.g}^{-1}$ .

The maximum concentration (59.21 mg/kg) of Pb in mussel was found during post-monsoon in station 3, while the minimum concentration (7.03 mg/kg) was detected in station 1 during the same season. Ahmed *et al.* (2009a) studied heavy metal concentration in molluscs (bivalves) from the Shitalakhya River, Bangladesh and found that the concentrations of Pb varied seasonally from 9.16 to 13.09  $\mu\text{g.g}^{-1}$ . Sericano *et al.* (2001) reported the mean concentration of Pb in molluscs (bivalves) from Kara Sea and its adjacent Rivers (the Yenisey and Ob rivers), Russia as  $1.4 \pm 0.1 \mu\text{g.g}^{-1}$ . Miramand and Bentley (1992) observed the highest concentrations of Pb as 3.75 and 2.0  $\mu\text{g/g}$  in *Petella* sp. and *Fucus serratus*, respectively, which are lower than the present findings. The concentration of Pb observed by Ahmed *et al.* (2009b) in mud crab (*Scylla serrata*) as 6.77  $\mu\text{g/g}$ , horseshoe crab (*Limulus* sp.) 5.28  $\mu\text{g/g}$ , mud skipper (*Gobus boddarti*) 4.99  $\mu\text{g/g}$ , hermit crab (*Eupagurus* sp.) 6.5  $\mu\text{g/g}$  and gastropod (*Assimineae vrevicula*) 4.66  $\mu\text{g/g}$ . Among the five samples, mud crab contained the highest value of Pb (6.77  $\mu\text{g/g}$ ) and hermit crab contained the lowest value of Pb (4.66  $\mu\text{g/g}$ ) in their body (Ahmed *et al.* 2009b). Zorba *et al.* (1992) observed the highest concentration of Pb in clam (*Circenita callipyga*) within a range between 1.23 and 2.91  $\mu\text{g/g}$  and stated that the variation in concentration was due to the seasonal changes and also with the influence of temperature and salinity.

Among the five heavy metals studied in mussel, Cd was found at lowest concentration ( $2.64 \pm 2.95$  mg/kg). Throughout the year, the Cd concentrations in station 3 were much higher than those of other stations. The highest concentration (7.23 mg/kg) was in station 3 during pre-monsoon, while, the lowest (0.56 mg/kg) was in station 1 during monsoon. Ahmed *et al.* (2009a) studied the heavy metal concentration in molluscs (bivalves) from the Shitalakhya River, Bangladesh and found that the concentrations of Cd varied seasonally from 1.09 to 1.21  $\mu\text{g.g}^{-1}$ . Sericano *et al.* (2001) reported the mean concentration of Cd in molluscs (bivalves) from Kara Sea and its adjacent Rivers, Russia as 2.7

$\pm 0.42 \mu\text{g.g}^{-1}$ . Bryan and Hummerstone (1977) studied the heavy metal concentration in molluscs (*Littorina littorea*) from the Looe estuary and found that the concentrations of Cd varied from 0.49 to 2.56  $\mu\text{g.g}^{-1}$ . Bustamante *et al.* (2000) reported the mean concentration of Cd in molluscs from New Caledonia to be  $16 \pm 6.5 \mu\text{g.g}^{-1}$ . Al-Madfa *et al.* (1998) found that the average concentration of Cd in molluscs (*Pinctada radiata*) from the Arabian Gulf as 0.49  $\mu\text{g.g}^{-1}$ . Páez-Osuna and Marmolejo-Rivas (1990) found the average concentration of Cd in molluscs (*Saccrostrea iridescens*) from NW Coast Mexico as 6.0  $\mu\text{g.g}^{-1}$ . Goldberg *et al.* (1983) found that the average concentrations of Cd in molluscs from Gulf Coast, USA to be 5.1  $\mu\text{g.g}^{-1}$ . Páez-Osuna *et al.* (1993) found that the average concentration of Cd in molluscs (*Crassostrea californiensis*, *C. subrugosa* and *Tellina* sp.) from the Coast of Mazatlan, Sinaloa, Mexico was 3.6  $\mu\text{g.g}^{-1}$ . Watling and Watling (1976) found that the average concentration of Cd in molluscs (*Choromytilus meridionali*) from Langebaan Lagoon, South Africa was 9  $\mu\text{g.g}^{-1}$ . Zorba *et al.* (1992) worked on clams (*Circenita callipyga*) as pollution bio-indicators in Kuwait's marine environment metal accumulation and depuration and observed highest concentration of Cd within range between 0.18 and 0.32  $\mu\text{g/g}$  and stated that accumulation of Cd in clam depends on the bio-availability of Cd in sea water and sediment. Miramand and Bentley (1992) observed the highest concentration of Cd within a range between 0.5 and 1.9  $\mu\text{g/g}$  in *Fucus serratus* and 2.7 and 7.5  $\mu\text{g/g}$  in *Patella vulgata*. Segar and Pellenberg (1971) worked on the distribution of the major and some minor elements in marine animals and observed the highest concentration of Cd as 31  $\mu\text{g/g}$ , which is much higher in comparison to the present findings. Zaroogian (1980) found that the uptake rate of heavy metals increased at lowest salinity and with an increase in temperature in *Crassostrea virginica*. The inhibition of Cd uptake by Ca channel blockers has been reported in excised gills of the bivalves *Anodonta anatina*. Increasing aqueous Ca concentrations reduced Cd accumulation in both the shore crab *Carcinus maenas* and the amphipod *Hyalella azteca* (Haque *et al.* 2007).

In mussel Cr was the most abundant ( $164.58 \pm 229.59$  mg/kg) compare to other heavy metals of this study. The Cr concentration was maximum (501.11 mg/kg) during pre-monsoon at station 3, whereas it was minimum (9.38 mg/kg) during the same season at station 2. Ahmed *et al.* (2009a) found that the concentrations of Cr varied seasonally from 8.12 to 9.07  $\mu\text{g.g}^{-1}$  in molluscs (bivalves) from the Shitalakhya River, Bangladesh. Sericano *et al.* (2001) reported the mean concentration of Cr in molluscs (bivalves) from the Kara Sea and its adjacent rivers in Russia was  $19 \pm 1.2 \mu\text{g.g}^{-1}$ . Bryan and Hummerstone (1977) studied the heavy metal concentration in molluscs (*Littorina littorea*) from the Looe estuary and found that the concentration of Cr varied from 0.13 to 0.98  $\mu\text{g.g}^{-1}$ . Bustamante *et al.* (2000) reported the mean concentration of Cr in molluscs from New Caledonia as  $2.6 \pm 0.3 \mu\text{g.g}^{-1}$ . Bryan (1973) studied seasonal variation of trace metal concentration in molluscs and found that the mean concentration of Cr was 1.3  $\mu\text{g.g}^{-1}$ . Al-Madfa *et al.* (1998) found that the average concentration of Cr in molluscs from Arabian Gulf was 2.79  $\mu\text{g.g}^{-1}$ . Páez-Osuna *et al.* (1993) found that the average concentration of Cr in molluscs from NW Coast Mexico was 2.7  $\mu\text{g.g}^{-1}$ .

Cu concentrations in station 3 were much higher than those of other stations all year round. Although the average concentration of Cu was  $57.49 \pm 74.35$  mg/kg, the highest concentration (183.87 mg/kg) was noticed in station 3 during the monsoon whereas the lowest (7.55 mg/kg) was found in station 2 during post-monsoon. Ahmed *et al.* (2009a) studied heavy metal concentration in molluscs (bivalves) from the Shitalakhya River, Bangladesh and found that the concentrations of Cu seasonally varied from 5.47 to 8.19  $\mu\text{g.g}^{-1}$ . Sericano *et al.* (2001) reported the mean concentration of Cu in molluscs (bivalves) from Kara Sea and its adjacent rivers in Russia to be  $51 \pm 32 \mu\text{g.g}^{-1}$ . Bryan and Hummerstone (1977) studied the heavy metal concentration

**Table 11** Maximum Cu, Zn, Cd and Pb concentrations (mg/kg) in reservoir sediments from the Lot River, other selected sites and UCC (upper continental crust).

Location	Cu	Cd	Pb	References
Cajarc site, Lot River, France	97.7	125	523	Audry <i>et al.</i> 2004
Temple site, Lot River, France	30.7	20.4	105	Audry <i>et al.</i> 2004
Marcenac site, Lot River, France	26.9	0.81	43.6	Audry <i>et al.</i> 2004
UCC, Western Europe	14.3	0.10	17	Wedepohl 1995
Malter Reservoir, Germany	196	37.5	465	Müller <i>et al.</i> 2000
Odiel River, Spain <sup>a</sup>	1282	8.5	649	Morillo <i>et al.</i> 2002
Tinto River, Spain	846	6.2	870	Galán <i>et al.</i> 2003
Guadamar River, Spain	165	3.9	127	Lacal <i>et al.</i> 2003
Lake Zurich, Switzerland	48.9	2.1	94.4	Von Gunten <i>et al.</i> 1997
Meuse River, The Netherlands	66	9.8	135	Van Den Berg <i>et al.</i> 1999

in molluscs (gastropods) from the Looe estuary and found that the concentrations of Cu varied from 10 to 194  $\mu\text{g.g}^{-1}$ . Bustamante *et al.* (2000) reported the mean concentration of Cu in molluscs from New Caledonia as  $73 \pm 20 \mu\text{g.g}^{-1}$ . Al-Madfa *et al.* (1998) found that the average concentration of Cu as  $7.08 \mu\text{g.g}^{-1}$  in molluscs from the Arabian Gulf. Páez-Osuna *et al.* (1993) found the average concentration of Cu in molluscs from the NW Coast Mexico to be  $119 \mu\text{g.g}^{-1}$ , Goldberg *et al.* (1983), in molluscs from Gulf Coast, USA, to be  $258 \mu\text{g.g}^{-1}$ , Páez-Osuna and Marmolejo-Rivas (1990), in molluscs from NW Coast of Mazatlan, Sinaloa, Mexico to be  $93 \mu\text{g.g}^{-1}$  and Watling and Watling (1976), in molluscs from Langebaan Lagoon, South Africa, to be  $12 \mu\text{g.g}^{-1}$ . Zorba *et al.* (1992) worked on clams as pollution bio-indicators in Kuwait's marine environment and observed highest concentration of Cu ( $12.5 \mu\text{g.g}^{-1}$ ) and stated that the concentration of Cu attained in clams by burrowing in sand as is their natural occurrence and by their enriched affected pollution from sediments. Miramand and Bentley (1992) worked on heavy metal concentrations in two biological indicators (*Patella vulgata* and *Fucus serratus*) and observed the highest concentration of Cu within the range of  $3.0$  to  $6.6 \mu\text{g.g}^{-1}$  in *Patella* sp. and  $0.8$  to  $2.0 \mu\text{g.g}^{-1}$  in *Fucus* sp. with seasonal fluctuation. He stated that Cu accumulated in both species due to the industrial activities of nuclear fuel processing plant which dumped nuclear fuel on the surrounding biological environment adjacent to the plant. The present findings substantiated the finding of Miramand and Bentley, although in the present investigation, the enrichment of Cu in macrobenthic fauna might be due to the industrial and agrochemical input of Cu in water and sediments. Cu is intimately related to the aerobic degradation of organic matter (Das and Nolting 1993).

Simkiss and Taylor (1989) discussed the pathways of metal accumulation by aquatic organisms, and identified six possible types. It is generally accepted that trace elements are taken up by aquatic biota in a passive process, down a concentration gradient into tissues. This can occur despite the presence of much higher concentrations of the elements in the tissues than in the external medium, as the metals in the tissues are bound to a wide range of biochemical sites (Manson *et al.* 1988). In a few instances, uptake may also occur through ion pumps, and in these cases, energy dependence exists (Rainbow 1995; Wright 1995). A review of the concentration of some heavy metals in reservoir from Lot River, other selected sites and UCC (upper continental crust) is given in **Table 11**.

The results of this study indicate that the concentrations of heavy metals in river environment of Bangladesh are higher than the permissible levels and the aquatic animals are not safe for human consumption. With the gradual development of industry, intensive use of pesticides and untreated domestic sewage as well as polluted discharge from neighbouring may further deteriorate the situation in future.

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## REFERENCES

- Ahmed F (1998) Heavy metals in the water and sediment of the Sundarbans Reserved Forest. Undergraduate thesis, Discipline of Fisheries and Marine Resource Technology, University of Khulna, Khulna, Bangladesh, 98 pp
- Ahmed MK (2000) An assessment of trace metal pollution in coastal areas of Bangladesh. *Proceedings of an International Symposium "Coastal Pollution and Nutrient Cycles"*, 3-10 December, 2000, Tokyo, United Nations University, 73 pp
- Ahmed MK, Bhowmik AC, Rahman S, Haque MR, Hasan MM (2009a) Heavy metal concentrations in water, sediments and their bio-accumulations in fishes and oyster in Shitalakhya River. *Wetlands Ecology and Management* (Submitted)
- Ahmed MK, Mehedi MY, Huque MR, Ahmed F (2002) Heavy metal concentration in water and sediment of Sundarbans Mangrove Forest, Bangladesh. *Asian Journal of Microbiology Biotechnology Environmental Sciences* **4** (2), 171-179
- Ahmed MK, Mehedi MY, Haque MR, Ghosh RK (2003) Concentration of heavy metals in two upstream rivers sediment of the Sundarban Mangrove Forest. *Asian Journal of Microbiology Biotechnology Environmental Sciences* **5** (1), 41-47
- Ahmed MK, Mehedi MY, Haque MR, Mondol P (2009b) Heavy metal concentrations in some selected macro-benthic fauna of the Sundarbans Mangrove Forest, Bangladesh. *International Journal of Environmental Research* in press
- Al-Madfa H, Abdel-Moati MAR, Al-Gimaly FH (1998) *Pinctada radiata* (pearl oyster): A bioindicator for metal pollution monitoring in the Qatari Waters (Arabian Gulf). *Bulletin of Environmental Contamination and Toxicology* **60**, 245-251
- AMAP (1998) *Assessment Report: Arctic Pollution Issues*, Arctic Monitoring and Assessment Programme, Oslo, 189 pp
- Ashraf W (2006) Levels of selected heavy metals in tuna fish. *Arabian Journal of Science and Engineering* **31** (1A), 89-92
- Audry S, Schäfer J, Blanc G, Jouanneau JM (2004) Fifty-year sedimentary record of heavy metal pollution (Cd, Zn, Cu, Pb) in the Lot River reservoirs (France). *Environmental Pollution* **132**(3), 413-426
- Biswas SK, Sharif AKM, Alamgir M, Choudhury DA (1998) Mercury level of some marine fishes from the Bay of Bengal. *The Journal of NOAMI* **15** (2), 1-5
- Bhattacharya B, Sarkar SK, Maji PK (1994) Bioaccumulation of heavy metals in flora and fauna of Hooghly Estuary, east coast of India. *Toxicological and Environmental Chemistry* **42**, 123-130
- Bryan GW (1973) The occurrence and seasonal variation of trace metals in the scallops *Pecten maximus* (L) and *Chlamys opercularis* (L). *Journal of the Marine Biological Association of the UK* **53**, 145-166
- Bryan GW, Hummerstone LG (1977) Indicators of heavy metal contamination in the Looe Estuary (Cornwall) with particular regard to Silver and Lead. *Journal of the Marine Biological Association of the UK* **57**, 75-92
- Burden-Jones C, Denton GW (1984) *Metals in Marine Organisms from the Great Barrier Reef Province* (Final Report, Part 1, Baseline Survey), Department of Marine Biology, James Cook University, Townsville, 155 pp
- Burgera J, Gochfeld M (2005) Heavy metals in commercial fish in New Jersey. *Environmental Research* **99**, 403-412
- Bustamante P, Grigioni S, Boucher-Rodoni R, Caurant F, Miramand P (2000) Bioaccumulation of 12 trace elements in the tissues of Nautilus (*Nautilus macromphalus*) from New Caledonia. *Marine Pollution Bulletin* **40**, 688-696
- Caspers H (1975) Pollution in coastal waters. An Interim Report on Results of a priority programme of the German Research Society (1966-1974), Germany, 142 pp
- Chester R, Stoner JH (1975) Trace elements in sediments from the Lower Severn Estuary and Bristol Channel. *Marine Pollution Bulletin* **6**, 92-96
- Cross FA, Duke TW, Willes JN (1970) Bio-geochemistry of trace elements in coastal plane estuary: Distribution of manganese, iron and zinc in sediments, water and polychaetous worms. *Chesapeake Science* **11**, 221-234
- Das JD, Nolting RF (1993) Distribution of trace metals in sediments and pore waters in the N. W. Mediterranean Sea. NIOZ, EROS-200 Project, 10 pp
- EQS (Environmental Quality Standards) (1991) Department of Environment,



- Government of the Peoples' Republic of Bangladesh, 44 pp
- Furnas MJ** (1992) The behaviour of nutrients in tropical aquatic ecosystems. In: Connell DW, Hawker DW (Eds) *Pollution in Tropical Aquatic Ecosystems*, CRC Press, Boca Raton, Florida, pp 29-65
- Galán E, Gómez-Ariza JL, González I, Fernández-Caliani JC, Morales E, Giráldez I** (2003) Heavy metal partitioning in river sediments severely polluted by acid mine drainage in the Iberian Pyrite Belt. *Applied Geochemistry* **18**, 409-421
- GESAMP** (1982) Joint Group of Experts on the Scientific Aspects of Marine Pollution, Review of potentially harmful substances – cadmium, lead and tin. *GESAMP Reports and Studies* **22**, 116 pp and *UNEP Reg. Seas. Reports and Studies* **56**, 85 pp
- Goldberg ED, Koide M, Holdge V, Flegal AR, Martin J** (1983) US mussel watch: 1977-1978 results on trace metals and radionuclides. *Estuarine, Coastal and Shelf Science* **16**, 69-93
- Haque MR, Ahmad JU, Chowdhury MDA, Ahmed MK, Rahman MS** (2004) Seasonal variation of heavy metals concentration in sediments of the rivers and estuaries of Sundarban mangrove forest. *Asian Journal of Microbiology Biotechnology Environmental Sciences* **6** (2), 175-185
- Haque MR, Ahmad JU, Chowdhury MDA, Ahmed MK, Rahman MS** (2005) Seasonal variation of heavy metals concentration in surface water of the rivers and estuaries of Sundarban mangrove forest. *Pollution Research* **24** (2), 463-472
- Haque MR, Ahmed MK, Ahmad JU, Chowdhury MDA** (2007) Seasonal variation of heavy metals in some molluscs of Sundarban mangrove forest, Bangladesh. *Pakistan Journal of Oceanography* **2** (2), 81-98
- Haque MR, Ahmed MK, Mannaf MA, Islam MM** (2006) Seasonal variation of heavy metals concentrations in *Gudusia chapra* inhabiting the Sundarban mangrove forest. *The Journal of NOAMI* **23** (1), 1-21
- Haque MR, Ullah SM, Ahmed MK, Azad MS** (2003) Seasonal variation in physico-chemical parameters of water and sediment in the rivers of Sundarban mangrove forest. *The Journal of NOAMI* **24** (2), 463-472
- Haque W, Ahmed ATA, Tarafdar SA, Akhter S, Quraishi** (2003) Trace elements in two small fishes, *Puntius sophore* (Hamilton) and *Mystus vittatus* (Bloch) from Buriganga River, Balu River and Ichamati Beel. Scientific Note, *Bangladesh Journal of Zoology* **31** (2), 247-251
- Holden AV, Topping G** (1972) Occurrence of specific pollutants in fish in the Forth and Tay estuaries. *Proceedings of the Royal Society of Edinburgh, Series, B* **71**, 189-194
- Holmgren S** (1994) An environmental assessment of the Bay of Bengal region. Bay of Bengal Programme, *BOBP/REP/67*, pp 79-112
- Hossain MS** (1996) Progress report on bio-accumulation and seasonal variation of trace metals in water, sediment and commercially important fishes and shellfishes of the north eastern Bay of Bengal. Institute of Marine Science, University of Chittagong, Bangladesh, 23 pp
- IPCS** (1992) Cadmium - environmental aspects, Environmental Health Criteria 135, International Programme on Chemical Safety (IPCS), a joint venture of the United Nations Environment Programme, the International Labour Organisation, and the World Health Organization, Geneva, Switzerland, pp59
- Islam MS** (2002) Heavy metals in water, sediment and some pelagic and benthic fishes of Buriganga River, Bangladesh. MSc thesis, Dhaka College, National University, Bangladesh, 121 pp
- Karim A** (1994) Environmental Impacts. In: Hussain Z, Acharya V (Eds) *Mangroves of the Sundarbans (Vol 2) Bangladesh*, IUCN, Bangkok, Thailand, pp 203-217
- Khan AH, Ali M, Biswas SK, Hadi DA** (1987) Trace elements in marine fish from the Bay of Bengal. *Science of the Total Environment* **61**, 121-130
- Khan YSA, Hossain MS, Hossain SMGA, Halimuzzaman AHM** (1998) An environment of trace metals in the GBM Estuary. *Journal of Remote Sensing and Environment* **2**, 103-113
- Lagorburu I, Canton L** (1992) Heavy metal concentration in littoral sediments from Guipuzcoa, Spain. *Marine Pollution Bulletin* **24**, 462-464
- Lacal J, Pilar da Silva M, García R, Sevilla TM, Procopio JR, Hernández L** (2003) Study of fractionation and potential mobility of metal in sludge from pyrite mining and affected river sediments: changes in mobility over time and use of artificial ageing as a tool in environmental impact assessment. *Environmental Pollution* **124**, 291-305
- Manson AZ, Jenkins KD, Sullivan PA** (1988) Mechanisms of trace metal accumulation in the polychaete *Neanthes arenaceodentata*. *Journal of the Marine Biological Association of the UK* **68**, 61-80
- Mehedi MY** (1994) A comparative pen-picture of pollution status of the coastal belt of Bangladesh with special reference to Halisahar and ship breaking area, Chittagong. MSc thesis, Institute of Marine Science, University of Chittagong, Bangladesh, 169 pp
- Miramand P, Bentley D** (1992) Heavy metal concentrations in two biological indicators (*Patella vulgata* and *Fucus serratus*) collected near the French nuclear fuel reprocessing plant of La Hague. *Science of the Total Environment* **3**, 135-149
- Mitra A, Choudhury A** (1993) Heavy metal concentrations in oyster *Crassostrea cucullata* of Sagar Island, India. *Indian Journal of Environmental Health, NEERI* **35** (2), 139-141
- Morillo J, Usero J, Gracia I** (2002) Partitioning of metals in sediments from the Odiel River (Spain). *Environment International* **28**, 263-271
- Müller J, Ruppert H, Muramatsu Y, Schneider J** (2000) Reservoir sediments – a witness of mining and industrial development (Malter Reservoir, eastern Erzgebirge, Germany). *Environmental Geology* **39**, 1341-1351
- Nauen CE** (1983) *Compilation of Legal Limits for Hazardous Substances in Fish and Fishery Products*, United Nations Food and Agriculture Organization, FAO Fisheries Circular No. 764, 102 pp
- Nicholson RA, Moore PJ** (1981) The distribution of heavy metals in the superficial sediments of the North Sea. *Rapp. P.-V. Reun. Conseil International Pour L'exploration De La Mer* **181**, 35-48
- Páez-Osuna F, Marmolejo-Rivas C** (1990) Occurrence and seasonal variation of heavy metals in the oysters *Saccostrea iridescens*. *Bulletin of Environmental Contamination and Toxicology* **44**, 129-134
- Páez-Osuna F, Osuna-López JI, Izaguirre-Fierro G, Zazueta-Padilla HM** (1993) Heavy metals in clams from a subtropical coastal lagoon associated with an agricultural drainage basin. *Bulletin of Environmental Contamination and Toxicology* **50**, 915-921
- Peterson A, Jefferies DF, Dutton JWR, Harvey BR, Steele AK** (1972) British Isles coastal waters: the concentration of selected heavy metals in seawater, suspended matter and biological indicators; a pilot survey. *Environmental Pollution* **3**, 69-82
- Rainbow PS** (1995) Physiology, physicochemistry and metal uptake – a crustacean perspective. *Marine Pollution Bulletin* **31**, 55-59
- Rao IM, Szxyarayana D, Reddy BRP** (1985) Chemical oceanography of harbour and coastal environment of Visakhapatnam (Bay of Bengal): Part 1- Trace metals in water and Particulate matter. *Indian Journal of Marine Sciences* **14**, 139-146
- Rojahn T** (1972) Determination of copper, lead, cadmium and zinc in estuarine waters by anodic-stripping-alternating-current voltammetry on the hanging mercury drop electrode. *Analytica et Chimica Acta* **62**, 438-441
- Roth I, Hornung H** (1977) Heavy metal concentrations in water, sediments and fish from the Mediterranean coastal area, Israel. *Environmental Science and Technology* **11**, 265-269
- Segar DA, Pellenberg RE** (1973) Trace metals in carbonate and organic rich sediments. *Marine Pollution Bulletin* **4**, 138-142
- Sericano J, Brooks JM, Champ MA, Kennicutt II MC, Makeyev VV** (2001) Trace contaminant concentrations in the Kara Sea and its Adjacent Rivers, Russia. *Marine Pollution Bulletin* **42**, 1017-1030
- Shafi M, Quddus MMA** (1982) *Bangladeshher Masho Shampad (Fisheries of Bangladesh)*, Kabir Publications, Dhaka, Bangladesh, 440 pp (in Bengali)
- Sharif AKM, Alamgir M, Mustafa AI, Hossain MA, Amin MN** (1992b) Trace element concentration in ten species of freshwater fish of Bangladesh. *Science of the Total Environment* **138**, 177-186
- Sharif AKM, Mustafa AI, Amin NM, Shafiullah S** (1991) Trace metals in tropical marine fish from the Bay of Bengal. *Science of the Total Environment* **107**, 135-142
- Sharif AKM, Mustafa AI, Hossain MA, Amin MN, Safiullah S** (1992a) Lead and cadmium contents in ten species of tropical marine fish from the Bay of Bengal. *Science of the Total Environment* **133**, 193-199
- Sharif AKM, Alamgir M, Krishnamoorthy KR, Mustafa AI** (1993) Determination of arsenic, chromium, mercury, selenium and zinc in tropical marine fish by neutron activation. *Journal of Radioanalytical and Nuclear Chemistry* **170**, 299-307
- Simkiss K, Taylor MG** (1989) Metal fluxes across the membranes of aquatic organisms. *Reviews in Aquatic Sciences* **1**, 173-188
- Şirelil UT, Göncüoğlu M, Yıldırım Y, Gücükoğlu A, Çakmak Ö** (2006) Assessment of heavy metals (cadmium and lead) in vacuum packaged smoked fish species (mackerel, *Salmo salar* and *Oncorhynchus mykiss*) marketed in Ankara (Turkey), E.U. *Journal of Fisheries and Aquatic Sciences* **23**, 353-356
- Tilzer MM, Khondker M** (1993) Hypertrophic and polluted freshwater ecosystems: Ecological basis for water resource management. Department of Botany, University of Dhaka, Bangladesh, pp 91-96
- Van Den Berg GA, Loch JPG, Van Der Heijdt LM, Zwolsman JGG** (1999) Mobilisation of heavy metals in contaminated sediments in the river Meuse, The Netherlands. *Water, Air, and Soil Pollution* **166**, 567-586
- Von Gunten HR, Sturm M, Moser RN** (1997) 200-year record of metals in lake sediments and natural background concentrations. *Environmental Science and Technology* **31**, 2193-2197
- Watling HR, Watling RJ** (1976) Trace metals in *Choromytilus meridionalis*. *Marine Pollution Bulletin* **7**, 91-94
- Wedepohl KH** (1995) The composition of the continental crust. *Geochimica et Cosmochimica Acta* **59**, 1217-1232
- Wright DA** (1976) Heavy metals in animals from the North East Coast. *Marine Pollution Bulletin* **7**, 36-38
- Wright DA** (1995) Trace metals and major ion interactions in aquatic animals. *Marine Pollution Bulletin* **31**, 8-18
- Zarogian GE** (1980) *Crassostrea virginica* as an indicator of cadmium pollution. *Marine Biology* **58**, 275-284
- Zingde MD, Singbal SYS, Moraes CF, Reddy CVG** (1979) Some observations on pollutants of Velsao Bay (Goa). *Mahasagar Bulletin of the National Institute of Oceanography* **12** (2), 69-74
- Zorba MA, Jacob PG, Al-Bloushi A, Al-Nafisi R** (1992) Clams as pollution bioindicators in Kuwait's marine environment: Metal accumulation and depuration. *Science of the Total Environment* **120**, 185-204