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HEAVY METALS IN WATER, SEDIMENT AND SOME FISHES OF KARNOFULY RIVER, BANGLADESH

F. ISLAM[1]*, M. RAHMAN[2], S.S.A. KHAN[3], B. AHMED[4], A. BAKAR[1] AND M. HALDER[1]

[1]*Bangladesh Council of Scientific and Industrial Research (BCSIR) Laboratories Chittagong, Chittagong-4220, Bangladesh*
[2]*School of Chemical Sciences and Food Technology, Faculty of Science and Technology, University Kebangsaan Malaysia (UKM), 43600-Bangi, Selangor, Malaysia*
[3]*Asian University for Women, Bangladesh*
[4]*TUV SUD Bangladesh (Pvt) Ltd, Dhaka, Bangladesh*

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ABSTRACT

The studies were carried out to estimate the Chemical and biological quality of water, sediment and fishes in various station of Karnofuly river, Bangladesh. In water, the heavy metal value of Lead, Cadmium, Chromium, Copper, Zinc, Iron, Manganese, Silver, BOD_5, COD, Nitrate, Nitrite, Phosphate were found at 0.107- 0.177 mg/L, 0038-.008 mg/L, 0.09-0.325 mg/L, 0.026-0.09 mg/L, 0.075-0.449 mg/L, 0.714-4.031 mg/L, 0.09-4.30 mg/L, 0.003-0.186 mg/L, 630 mg/L, 862 mg/L and 45 mg/L respectively. The highest value of BOD_5, COD and Phosphates were found in Fisheryghat station. All the sampling station of sediments showed moderate various. High concentration of Pb was found in Chapila Fish. The results also provided data to understand and quantify the threat of the impact of climate change on environment of this region. This study will convey a strong message for the government body, policy maker to establish rules and regulation in order to save our environment and will be helpful for public awareness. An immediate attention from the concerned authorities is required in order to protect the river from further pollution.

KEY WORDS : Water, Sediment, Fish, Heavy metal, Atomic Absorption Spectrophotometer

INTRODUCTION

Bangladesh is predominantly an agricultural based country within the south Asian region. Chittagong is the second largest city of Bangladesh. Chittagong is a developing and expanding city. Due to expedition of these city industrial activities, population growth, Agricultural practices, other manufactures, industrial effluents and oil and gas are discharged in a river named karnofuly and in early day's pollution was never even felt in this healthy city. Karnofuly the largest and most important river in Chittagong and the Chittagong Hill Tracts, is a 667 meters (2,188 ft)-wide river in the south-eastern part of Bangladesh. Originating from the Lushai hills in Mizoram, India, it flows 270 km (170 mi) southwest through Chittagong Hill Tracts and Chittagong into the Bay of Bengal. The mouth of the river hosts Chittagong's sea port, the main port of Bangladesh. Different kinds of industries are growing up beside the river Karnofuly.

The growing dearth of water has threatened profitable development, quality of the environment, sustainable of human livelihood, and a multitude of other public goals in many developing countries such as Asian and African regions. Unchecked growth of the urban population, particularly developing countries, places an enormous pressure on water and land resources. Wastewater is increasingly being used for agricultural irrigation in urban and peri-urban areas. Many industrial plants in developing countries operate in any or a normal wastewater treatment and routinely discharge their waste into drains that either contaminate rivers and

***Corresponding Author's email:** faridacct@yahoo.com (Faridul Islam, Scientific Officer, BCSIR Laboratories Chittagong, Chittagong-4220, Bangladesh. Phone: +8801717511576)

streams or add to the contaminant load of sewage sludge (Zeb *et al.* 2011). Contaminants from Industrial, urban and agriculture sources may enter the food chain in addition to the low water quality area (Khalid *et al.* 2011). The heavy metals contamination of agricultural soils and crops is causing concerns due to the probable effects on food production and human health in affected areas. Heavy metals are ubiquitous in the environment as a result of both natural and anthropogenic behavior, and humans are exposed to them their various pathways (Wilson and Pyatt, 2007).

Heavy metal contamination in water is an increasing worldwide environmental concern. Heavy metals are very harmful because of their non biodegradable nature, long biological half-lives and their potential to accumulate in different body parts. Most of the heavy metals are extremely toxic because of their solubility in water. Even low concentrations of heavy metals have damaging effects to man and animals because there is no good mechanism for their elimination from the body. Nowadays heavy metals are ubiquitous because of their excessive use in industrial applications. Wastewater contains substantial amounts of toxic heavy metals, which create problems (Chen *et al.* 2005; Singh *et al.* 2004). Dumping wastes from industries to rivers contribute to the larger problem of river pollution, which can seriously damage the marine environment and ecological imbalance is caused due to discharge of various industrial wastes into air and water bodies and ultimately affects on our food chain. The intensity of river pollution mainly depends on raw materials, location, their type, chemical effects, and production process. At present the industries such as such as 19 tanneries, 26 textile mills, 1 oil refinery, 1 TSP plant, 1 DDT plant, 2 chemical complexes, 5 fish processing units, 1 urea fertilizer factory, 1 asphalt bitumen plant, 1 steel mill, 1 paper mill (solid waste disposal hourly 1450 m^3), 1 rayon mill complex, 2 cement factories, 2 pesticide manufacturing plants, 4 paint and dye manufacturing plants, several soap and detergent factories and a number of light industrial units directly discharge untreated toxic effluent into Karnofuly river. From the survey of effluents from different industries, it has been found that the discharge is generally composed of organic and inorganic wastes. The organic wastes are the effluents from the tanneries, fish processing units, degradable wood chips, pulps and untreated municipal and sewage (about 40,000 kg BOD daily) etc. The inorganic wastes are chemicals used by the industries such as various acids, bleaching powder, lissapol, hydrogen peroxide, alkali, salts, lime, dyes, pigments, aluminium-sulphate and heavy metals etc. The DDT factory and fertilizer factory disposing of DDT, toxic chemicals and heavy metals to the Karnofuly River and ultimately to the Bay of Bengal.

Heavy metals that are introduced into the aquatic environment are ultimately incorporated into the aquatic sediments; organisms living in these sediments accumulate these heavy metals to varying degrees (Cross *et al.* 1970; Bryan and Hummerstone, 1977). It has been recognized that aquatic sediments absorb persistent and toxic chemicals to levels many times higher than the water column concentration (Casper *et al.* 2004; Vermeulen and Wepener, 1999; Linnik and Zubenko, 2000). Depending on the river morphology and hydrological conditions, suspended particles with associated contaminants can settle along the watercourse and become part of the bottom sediments, often for many kilometers downstream from the chemical sources (Ciszewski, 1997; Vigano, 2003; Wildi, 2004). So, the river water is seriously polluted by indiscriminate throwing of effluents and solid waste of the heavy industries. The heavy metal present in the waste is accumulated in soil of river bed. These metals get accumulated in fish through biomagnifications and directly entering in our food chain. Fish provide omega-3 (n-3) fatty acids that reduce cholesterol levels and the incidence of the heart disease, stroke, and preterm delivery (Daviglus *et al.* 2002; Patterson, 2002). Contaminant levels, particularly methylmercury (MeHg) and polychlorinated biphenyls (PCBs) are sufficiently high in some fishes to cause adverse human health effects in people consuming large quantities (Stem, 1993; Hightowe and Moore, 2003; Hites *et al.* 2004). Methylmercury is reported to counteract the cardioprotective effects (Guallar *et al.* 2002) Fish consumption is the only significant source of methylmercury for the public (Rice *et al.* 2000). Another is indirect way that is through irrigation of different crops by these heavy metal contaminated river water. Vegetables take up heavy metals and accumulate them in their edible (Bahemuka and Mubofu, 1991)and inedible parts in quantities high enough to cause clinical problems both to animals and human beings consuming these metal-rich plants (Alam, 2003). The present study was carried out to assess the levels of heavy metals in water, sediment and some fishes of karnofuly river, Bangladesh.

MATERIALS AND METHODS

The samples were collected in the heavy industrial zone of Chittagong city in Bangladesh from February to April during 2013.The samples of water and sediment were collected from five stations such as fishery ghat, Chakti khal, Mojjar tek, Kalur ghat east zone and Kalur ghat west zone of karnaphully river of Chittagong in Bangladesh and five types of fish were collected only Kalur ghat east zone because fishes are available under water (Table 1). The Fishery ghat and Chakti khal station is under the kotoali thana of Chittagong city where a lot of tanneries discharge their effluent. On the other hand Mojjer tek, Kalur ghat east and Kalur ghat west stations is industrial and heavily populated zone.

The water samples were collected in plastic bottles and acidified immediately with 2 mL of HNO_3 per litre of water and preserved in refrigerator at 4ºC for laboratory analysis. 100 mL of each sample was taken to 250 mL beaker and added 5 mL of concentrated nitric acid. Then evaporated the sample to near dryness on a hot plate. Then cooled the sample and added another 5 mL concentrated nitric acid. Samples were returned to hot plate and continued heating, adding additional acid as necessary until digestion was completed. This digestion decomposed organics. Then washed down the beaker walls with deionised water and filtered the sample into a 100 mL volumetric flask to remove silicate and other insoluble materials. Then each sample was made up to the mark with deionised water.

The sediment samples were collected by vertical corer and Ekman grab sampler. The sediments contain a lot of organic compounds. Prior to mineralization of samples, the organic compounds should be oxidized to increase the efficiency of acid digestion. At first, 500 g sediment samples were heated at 450 °C in a muffle furnace. Ash is treated with with 20% nitric acid by heating for 15 min. Then, the mixture of the sample and acid is filtered through glass filters. Then the solution was filtered and up to the mark with deionised water. These solutions were used for the determination of Pb, Cd, Cu, Mn, Zn, Ag, Cr and Fe.

Five different species of adult fish such as Chapila fish (Gonialosa manmina), Poua fish (Otolithoides pama), Rita fish (Rita rita), Chatka fish (Pellona ditchela) and Chiring fish (Apocrptes bato) were collected from river fishermen of fishery ghat station. The fish were washed, weighed and dried in

Table 1. Data of Soil, Fish and Water in the Karnofuly river of Bangladesh

Test		Soil (ppm)					Fish (ppm)					Water (ppm)			
Parameter	Mohora	Kal. east	Mojjar tek	Fishery	Chakti khal	Poua	Chring	Rita	Chapila	Chatka	Mojjar tek	Chakti khal	Kal. east	Fishery	Mohora
Pb	2.396	4.533	5.644	3.485	4.708	0.886	1.843	2.861	7.707	1.094	0.107	0.136	0.177	0.121	0.172
Cd	0.169	0.228	0.232	0.226	0.259	0.066	0.744	0.179	0.483	0.128	0.008	0.007	0.0038	0.006	0.004
Cu	0.432	1.227	1.999	1.135	5.263	0.842	0.77	1.508	2.806	1.093	0.036	0.069	0.032	0.09	0.026
Mn	32.845	55.853	85.372	71.562	77.614	1.733	6.123	13.301	4.784	1.078	0.173	1.336	0.09	4.3	0.11
Zn	4.229	9.785	15.708	11.265	23.394	8.535	11.45	72.701	33.46	15.794	0.449	0.25	0.115	0.317	0.075
Ag	0.172	0.613	1.012	0.358	0.723	0.278	0.237	0.992	1.737	0.012	0.186	0.085	0.012	0.077	0.003
Cr	0.152	0.849	2.563	0.812	0.678	0.569	1.077	0.064	0.099	0.429	0.325	0.204	0.09	0.315	0.149
Fe	558.72	647.375	928.434	752.31	921.396	7.465	145.131	73.732	63.082	8.703	1.534	4.031	0.714	3.121	0.887

an oven at 105°C for 3 days until gaining constant weight. After cooling in the desiccators, the samples were grinded by carbide mortar and pastel to make powder and homogenised. The powdered sample was finally stored in a pre-cleaned dry plastic bottle and preserved in desiccators for further analysis. For the quantitative analyses of Pb, Cd, Cu, Mn, Zn, Ag, Cr and Fe, the fish samples were digested. Dried (0.5 g) powder was taken in a long test tube. Then 1.0 mL 70% $HClO_4$, 4 mL concentrated HNO_3 and 1.5 mL concentrated H_2SO_4 were added into the test tube. The samples were then heated gently in an oil bath (~ 100°C) until the solid mass was dissolved. If sample was not clear 4 mL of HNO_3 was added into the test tube and repeated until the solution was clear. Finally, the mixture was boiled at about 210°C in order to drive off the acids except H_2SO_4 and then it was cooled down at room temperature. Blank digestion was also performed to quantify possible contamination during sample preparation and analysis.

The standard solution of the elements Pb, Cd, Cu, Mn, Zn, Ag, Cr and Fe were prepared by pouring the required amount of the solution from the stock solution, manufactured by Fisher Scientific Company, USA. Every metal standard solution was prepared for calibration the instrument for each element being determined on the same day as the analyses were performed due to possible deterioration of standard with time. Working standards were commonly prepared from stock solution at 1000 mg/L level using micropipettes. These solutions were prepared from their pure metal turnings and pure compound using nitric acid. Working standard and blanks were acidified to the same extent as samples. The atomic absorption instrument was set up and flame condition and absorbance were optimized for the analyte. Then blank, standards and samples were aspirated into the flame of AAS (Model- iCE 3000, Thermo Scientific). Only arsenic was estimated by hydride vapour generation technique. The analytical procedures were also calibrated against the above standard reference materials. The average recovery ranged between 92 to 104%.

BOD5 was determined by Winkler Method with slight modification (Rangana, 1986), COD was determined by the Dichromate Reflux Method (Rangana, 1986), Total Phosphorous, Nitrate and Nitrite was determined by the Colorimetric method using UV spectrophotometer at 405 nm, 420 nm and 520 nm (Rand, 1976).

RESULTS AND DISCUSSION

Soil

Different heavy metal concentrations at different places were determined in BCSIR laboratories in Chittagong. The average Pb concentration in sediments of karnofuly was 4.96±0.6ppm. Pb concentration was highest in sediments of Mojjar tek as 5.64ppm and lowest in place of Mohora as 2.4ppm. All the values were exceeded the guidelines of WHO. The average Pb concentration in Karnofuly was BDL-0.02ppm (Ahmed *et al*, 2010). So, all values of lead in different positions were highest than WHO standard and previous Pb concentration in karnofuli. Cd level in sediments of karnofuly was higher at Chakti khal as 0.26ppm in Karnofuli river but lower at Mohora as 0.17ppm. The average concentration of Cd is 0.24±0.017ppm. Previous average Cd levels were below detection level in Karnofuli river (Ahmed *et al.*, 2010). Cobult concentration was higher at Mojjar tek and lowest at Mohora in soil of Karnofuly River as 0.25ppm and BDL respectively. The average concentration of cobalt is 0.31±0.051ppm. Cobalt concentrations were highest in all value than WHO guidelines except Mohora. Concentration of cupper in sediments of karnofuly was 1.22±0.78ppm and range from 5.26ppm at Chakti khal to 0.43ppm at Mohora in Karnofuli. Among the heavy metals concentrations Fe reached at top position as average 832.40±160.28 ppm. Highest concentration of Fe was rerecorded at Mojjar tek as 928.43ppm and lowest in Mohora as 558.72ppm in Karnofuly sediments. Average Chromium concentration was 0.76±0.12ppm and range from BDL to 2.56ppm. In present Mn concentration was higher than 0.2 to 19.20ppm (Ahmed *et al,* 2010). At present Mn concentration range from 32.85ppm to 85.37ppm and average 72.95±15.30. Highest concentration of Mn was recorded at Mojjar tek and lowest concentration was recorded at Mohora in Karnofuly river sediments. Average Zn concentration was 16.30±6.82 and range from 4.23 to 33.40ppm in this river sediments and Ag concentration range from 0.17 to 1ppm and average 0.78±0.21ppm was recorded. Among the concentration of heavy metals Fe was highest (928.43ppm in Mojjar tek) and Chromium, Cobalt was lowest (BDL in Mohora) and descending series of these metals as Fe>Mn>Zn>Pb> Cu>Cr> Ag>Co>Cd. The sediments of Mojjar tek was mostly heavy metal contaminated among the places and Mohora was least heavy metal contaminated among the places. If we consider statistics there was a significant relationship between the heavy metal contents in soil and fish ($p<0.001$). The calculation was conducted by Minitab software. All the metal concentrations of soil, water, and fish (ppm) are given below Table 2.

Table 2. Heavy metal concentration (ppm) of soil, water and fish

Test Parameter	Soil (Metal Conc.±SD)	Water (Metal Conc.±SD)	Fish (Metal Conc.±SD)
Pb	4.96±0.60	0.14±0.031	1.67±0.89
Cd	0.24±0.02	0.01±0.002	0.40±0.25
Cu	1.22±0.78	0.05±0.028	1.40±0.84
Mn	15.30±72.95	0.12±0.043	3.77±2.23
Zn	16.30±6.82	0.28±0.139	20.79±12.42
Ag	0.78±0.21	0.06±0.040	0.60±0.39
Cr	0.76±0.12	0.25±0.068	0.45±0.25
Fe	832.40±160.28	2.06±1.456	69.95±5.96

Metal Conc. = Metal Concentration, SD=Standard Deviation

Fish

The heavy metals accumulate in the fish body through biomagnifications fro soil sediment of Karnofuly River. Average lead content in fish was 1.67 ±0.89ppm and range from 7.17 to 0.87 ppm. Highest lead concentration was obtained in Chapila fish (Gonialosa manmina) 7.17 ppm and lowest lead concentration was obtained in Poua fish (Otolithoides pama) 0.87 ppm. Highest cadmium concentration was found in Chring fish (Apocrptes bato) 0.74ppm and lowest in Poua (0.07ppm). The average concentration of the cadmium was 0.40 ±0.25ppm. The range of Cu in fish was 25.08 to 0.77ppm and average 1.4±0.84ppm. Highest Mn accumulation was occurred in Rita fish (Rita rita) (13.30ppm) and lowest in Poua (1.07ppm) but average concentration of Mn was 3.77±2.23ppm. Zinc was the second most abundant metal in Rita fish i.e. 72.70 ppm. Average concentration of Zn was 20.79 ±12.42ppm and range from 72.70 to 8.54ppm. Among the metals that were accumulated in fish, Ag concentration was very low. Average concentration of Ag was 0.60 ±0.39 ppm and range from 1.74 to 0.01ppm. Highest Cr concentration was found in Chring fish (1.08ppm) but lowest in Rita fish (0.06ppm) and average concentration was 0.45±0.25ppm. Among the heavy metals Fe were most abundant metals. Its average concentration was 69.95±5.96ppm but wider range from 145.13 to 7.47ppm. Highest concentration of Fe was obtained in Chring fish but lowest in Paua fish. From the above discussion it is clear that highly metal contaminated fish is Chring fish and lowest contaminated fish is Paua fish. All metal ranges are nearly same for Chapila fish and Rita fish. The Chatka fish (Pellona ditchela) contain average amount of metals. Heavy metal accumulation descending order in tested fish as Chring>Chapila= Rita>Chatka>Paua. Statistically there was a significant relationship between the heavy metal concentration in fish and water ($p<0.001$).

Water

The heavy metal concentration of the Karnofuly river varied in place to place as wide range from 4.3 to 0.003 ppm. The average lead concentration in water sample was 0.14±0.03ppm and range from 0.18 to 0.11ppm. The cadmium concentration was very low as average 0.0058±0.0018ppm with range from 0.008 (Mojjar tek) to 0.0038ppm (Kalur ghat East). The highest concentration of cupper was found in Fishery (0.09 ppm) and lowest in Mohora (0.026 ppm). The average concentration of cupper was 0.05±0.03ppm. The most dominant metal in water sample was Fe. The average concentration of Fe was 2.06 ±1.5ppm and range from 4.03 (Chakti khal) to 0.71(Kalur ghat East) ppm. The average Cr value was 0.25±0.09 ppm with highest and lowest concentration of Cr 0.33ppm and 0.09 ppm respectively. Highest Mn concentration (4.3ppm) was obtained in Fishery but lowest in Kalur ghat East (0.09ppm) with average 0.12±0.04 ppm. Average Zn concentration was found as 0.28±0.14ppm with range from 0.45 to 0.075ppm. Average concentration of Ag was 0.058±0.04ppm and range from 0.19 to 0.003 ppm. From the above

Table 3. The comparison data of water

Test Parameter	Karnofuly river water (Metal Conc.±SD) ppm	Agricultural use(NOM-001-ECOL-1996)	For Drinking water (ECR 1997)
Pb	0.14±0.031	1.0	0.05
Cd	0.01±0.002	0.4	0.005
Cu	0.05±0.028	6.0	1
Mn	0.12±0.043	-	0.1
Zn	0.28±0.139	20.0	5
Ag	0.06±0.040	-	0.02
Cr	0.25±0.068	1.5	0.05
Fe	2.06±1.456	-	0.3-1.0

Table 4. The value of BOD, COD, Nitrate, Nitrite and Phosphate in water.

Test Parameter	$BOD_{5\ ppm}$	COD_{ppm}	$NO_3^-{}_{ppm}$	$NO2^-{}_{ppm}$	$PO4^{-2}{}_{ppm}$
Chaktai khal	156	228.80	0.20	0.10	26
Kalurghat East side	42.50	83.2	0.55	0.30	0.5
Kalurghat west side	29.50	41.6	0.75	0.80	0.6
Mojjartek	15.50	20.80	1.50	0.25	0.65
Fisheryghat	630	832.0	0.51	0.42	45

discussion it was clear that highest contamination site was Mojjar tek and least contamination site was Khal.East. So the descending order of heavy metals concentration was Mojjar tek>Fishery>Chakti Khal> Khal East. All the metals except Cu and Zn were exceeded their permissible limit in water in respect of drinking purpose but not exceed their limit in respect of agriculture as shown in Table 3. Statistically there was a significant relationship between water and soil of heavy metal concentration ($p<0.001$). Comparison of heavy metal concentration (ppm) between Karnofuli river water and standard limit values of water for agriculture and drinking purpose.

Biochemical oxygen demand (BOD5) is one of the most important and useful parameters indicating the organic strength of a wastewater. BOD measurement permits an estimate of waste Strength in terms of the amount of dissolved oxygen required to break down the wastewater. The value of BOD, COD, Nitrate, Nitrite and Phosphate are shown in Table 4. In all cases BOD value is much higher than the value recommended by the "Environment Conservation Rules, 1997" which is a concern (Ministry of Environment and Forest, 1997). Highest level of BOD5 is 630 mg/L, found in fishery ghat. High level of BOD5 reveals that organic strength of water is very much high.

Chemical Oxygen Demand (COD) is a measure of the capacity of water to consume oxygen during the decomposition of organic matter and the oxidation of inorganic chemicals such as ammonia and nitrite. COD measurements are commonly made on samples of wastewaters or of natural waters contaminated by domestic or industrial waste. The highest value of COD is 832 mg/L in the fishery ghat. The abnormally high figures of BOD5 and COD content indicate the presence of pollutants other than usual domestic sewage.

The nitrate value is higher in the all area than the nitrite except kalurghat ghat west side region. The highest value of nitrate contain in the mojjar tek side (1.50 mg/L). Total phosphate value is much higher than the recommended value of The Environment Conservation Rules, 1997. The higher amount contain in the Fishery ghat area (45 mg/L). From the above value, the fishery ghat region is the most polluted area.

Overall, the Karnofuly River contains the low amount of water in dry season than rainy season. As a result only industries wastes are drop down the river. Fish is a living animal. The water and sediments are the supporting substances to survive of fish. We have analyzed five types of fish. These fishes are survived the all season. The selected location contains the huge amount of heavy industry than others area of Chittagong City.

CONCLUSION

Bangladesh is a riverine country. Most of the rivers water in here is salty. Few of them are fresh water. So the source of fresh water is very important. Chittagong is an industrial city in Bangladesh. Many industries are present near the side of the Karnofuly River. Others that are not present at side of the river but their waste and liquid effluents are reached at the river finally. Already the water of the karnofuly is exceeding the permissible limit of heavy metal for drinking purpose due to effluents from industrial use but yet not agriculture use. If in these condition prevails in future for a long time then it will exceed the limit of agriculture use. And ultimately through process of biomagnifications it will be reached and contaminate in food chain and entering in human body. So immediate step to reduce food chain contamination and further deeply more research are also required.

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