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Level of Heavy Metals in Some Edible Marine Fishes of Mangrove Dominated Tropical Estuarine Areas of Hooghly River, North East Coast of Bay of Bengal, India

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Abstract The muscles of some important marine fishes collected in and around Hooghly estuarine coastal areas were analyzed for the heavy metals Cu, Zn, Ni, Cd, Cr and Pb. The concentration range of Cu (16.22–47.97 ppm), Pb (12.40–19.96 ppm) and Zn (12.13–44.74 ppm) were recorded comparatively higher and were similar to that found in contaminated areas. On the other hand the ranges of Ni (2.20–3.69 ppm), Cr (0–3.89 ppm) and Cd (0.62–1.20 ppm) were almost equal to those carried out over a wide range of geographical areas. The degree of bioaccumulations was metal-specific as well as species-specific in nature. The toxic groups of metals (Pb and Cd) showed higher variability than the essential metals (Cu, Zn and Ni). The calculated intake value of metals ($week^{-1}$ kg^{-1} body wt) varied from 14.88 to 27.60 of Pb, 0.87 to 1.68 of Cd, 0.0 to 5.45 of Cr, 22.70 to 137.16 of Cu, 3.08 to 5.17 of Ni and 16.98 to 62.60 of Zn through human consumption of these fishes and were compared with those of standard Provisional Tolerable Weekly Intake value (PTWI) per kg body weight as stipulated by WHO. The $PTWI_{Cal}$ values of Pb in some of the fishes recorded marginally excess values and may indicate a health risk through consumption of successive 7 days in a week.

Keywords Fish muscle · Heavy metal · Bioaccumulation · North east coast of India

Heavy metals are metals having a density of about 5 g/cc, which include elements such as Cu, Zn, Ni, Pb, Cd, Cr and others. Among them the metals like Cu, Ni and Zn are considered as essential micronutrients, but become toxic at concentration higher than the amount required for normal growth (Nies 1999). Other metals like Pb, Cd and Cr have so for unknown roles in living organisms and toxic even at low concentration (Wood 1974). They are not largely available in the living compartments of the nature and are brought into the ambient environment through human activities and become a source of pollution creating a decline in natural system, to human or to some living organisms in the environment through accumulation in soil, water and biota. Consumption of such aquatic food enriched with toxic metal may cause serious health hazards through food chain magnification (Miretzky et al. 2004).

Hooghly estuary and coastal environment (Fig. 1) in the north east coast of India (Lat. 21°53′N, Long. 88°15′E) are also not free from toxic metal pollution (Ghosh and Chowdhury 1989; Sarkar et al. 2002). The rapid human settlements, intensive boating and tourist activities, deforestation and ongoing agricultural and aqua-cultural practices due to high human population density and rapid economic growth of the countries surrounding the Bay of Bengal make the coastal environment vulnerable to a range of anthropogenic stress factors (Millennium ecosystem Assessment 2005; Ganguly et al. 2008). The coastal zone of this state is presently stressed with unplanned mushrooming of shrimp culture units, industries, and hotels, which in most cases release their respective wastes without any adequate treatment. These wastes containing appreciable amount of heavy metals thus find their way into the coastal water and adjacent estuaries. This estuarine complex is considered possibly the most polluted estuary in the world (Mukherjee and Kashen 2007) with large number of

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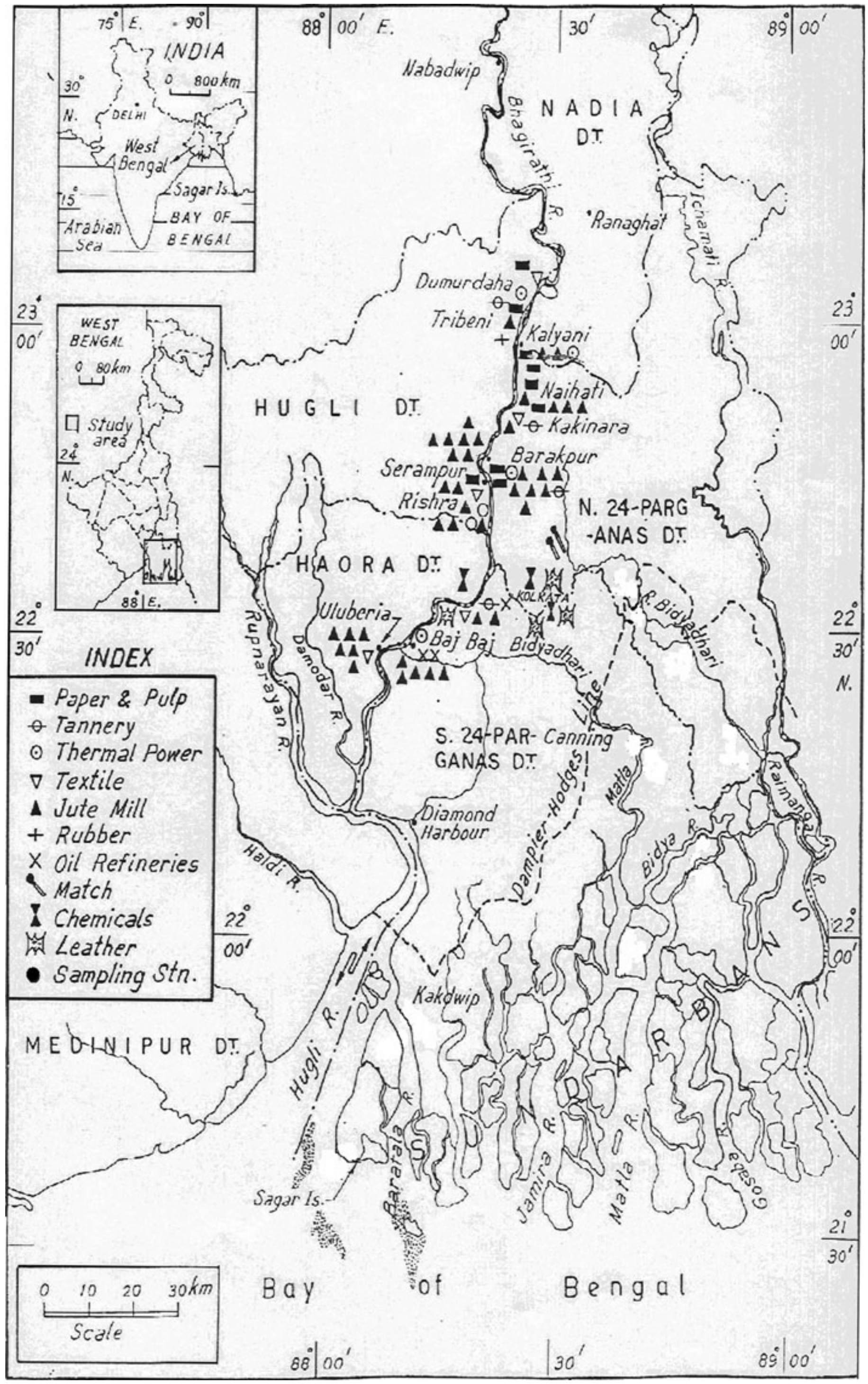

Fig. 1 Map of Hooghly estuary

factories located close to the mouth of the estuary discharging almost half a billion liters a day of untreated wastes including the effluent from pulp and paper mills, pesticide manufacturing plants, distilleries, thermal power plants, yeasts, rayon, cotton, vegetable oils, soap, fertilizers, leather manufacturing units and antibiotic plants. Being non-biodegradable, metals once get into soil or water, enjoy long residue time and spend for several years, before they are removed for other compartment of aquatic system (Walker et al. 1996). These elements usually remain in water as suspended colloid forming complex compound with inorganic and organic ligands and in course of time, they tend to accumulate in bottom sediments or being taken up by aquatic organisms, enjoying different trophic level of food chain. These heavy metals are taken up by aquatic animals directly through the epithelium of the skin, gills and elementary canals while some parts are accumulated in food organisms and are incorporated into the body of aquatic animals by nutrition (Chandra 1999). As a result the aquatic food produced in this territory may also contain significant level of toxic metals and also may pose health hazards on consumption. In the light of the above facts, the intent of the present paper is to study the degree of bioaccumulation of some heavy metals like lead (Pb), Cadmium (Cd), Chromium (Cr), Cupper (Cu), Nickel (Ni) and Zinc (Zn) in the flesh of some edible marine fishes and to compare the data obtained with that of other studies in order to investigate the degree of contamination of the area. Besides, the possible effects of human consumption through the uptake of these fishes are also highlighted.

Materials and Methods

Five species of fishes were collected directly from fisherman near Bakkhali region during winter season 2008. The species were *Setipinna phasa* (common Bengali name Phansa), *Pampus argentus* (Pomfret), *Glossogobius* sp. (Bele), *Liza parsia* (Parse) and *Cynoglossus* sp. (Sole fish). All these species have different food and feeding habit. Immediately after collection specimens were washed with clean seawater and finally with distilled water and stored in deep freezer. Samples of each species were taken to provide a consistent size range, as recommended by the guideline of the Joint Monitoring Group of Oslo & Paris convention. All samples consisted of a sufficient number of individuals to minimize errors due to individual variation (n = 10). Muscle tissues from the fishes were removed with plastic knife and kept in pre-cleaned plastic jar separately. Tissues were then oven dried at 105–110°C. All the dried samples were separately homogenized and 1 gm of each sample was digested with mixture of nitric acid and perchloric acid till a clear solution obtained, cooled at room temperature and filtered. The filtrate was then diluted with diluted HCl (1:1) and made to 50 mL with double distilled water. Blanks were also run at the same sequence of treatments. The solutions were analyzed for Pb, Cd, Cr, Cu, Ni and Zn by atomic absorption spectrophotometer (Model Varian Spectra AA55) using air-acetylene flame with digital read out system, deuterium lamp background corrector, and automatic zero to compensate the blank. Settings were those recommended by the manufacturer. A standard curve was run with each analysis. No detectable trace metals were found in the reagent blank. The results were expressed as mg/kg or ppm of dry weight. All chemicals and standard solutions used in the study were obtained from Merck and were of analytical grade. Double distilled water was used through out the study. All glassware and containers were thoroughly cleaned, finally rinsed with double distilled water for several times and air-dried prior to use. Accuracy and precision of the results were carried out by analysis of Bovine liver of known heavy metal content (SRM, Standard Reference Material, no 1577a, NIST, National Institute of Standard and technology, Former NBS). The recovery and mean relative standard deviations were within 97% and 10%, respectively. The detection limits of Pb, Cd, Cr, Cu, Zn and Ni are 0.05, 0.002, 0.02, 0.01, 0.005 and 0.02 mg/L, respectively. The obtained data were used for descriptive statistical analysis consisting of means, standard deviations and coefficient of variations. In addition, for comparing the different mean values, Duncan's multiple range tests were employed. Samples were considered significantly different at $p < 0.05$. All the analyses were performed by using SPSS—11 statistical packages for windows.

Results and Discussion

The average bioaccumulation level of the metals in different fishes along with their range of variation and standard deviation were presented in Table 1. It revealed that the mean values of the non toxic groups of metal like Cu and Zn (Table 2) showed comparatively higher level of occurrence in all fishes than other metals following the order of Zn > Cu > Pb > Ni > Cd > Cr, respectively. Among the fish, the average level of Zn varied from 12.13 to 44.74, Cu from 16.22 to 47.97, Pb from 12.40 to 19.96, Ni from 1.15 to 3.69, Cd from 0.62 to 1.20 and Cr from below detection limit to 3.89 mg kg^{-1} DW (dry weight). The metal chromium does not normally accumulate in fish and hence low concentrations were reported from everywhere (Moore and Ramamoorthy 1984). Our study also substantiates this fact showing negligible in amount in the fish muscle of Pomfret and Bele. The other fish contained Cr within the range between 1.0 and 3.89 mg kg^{-1} with higher values in Flat fish (3.89 mg kg^{-1}), followed by Phansa (1.02 mg kg^{-1}) and Parse (1.0 mg kg^{-1}). The variance analysis showed significant differences among the metal concentrations ($p < 0.05$). This vividly indicates that the degree of accumulation of the fish species is metal specific and the fishes must have the capability to differentiate the characteristics of the metals during their uptake from the available form. The study on the differences of metal accumulation among the fish also highlighted that only the species Pomfret and Bele registered comparatively lower level of bio-accumulation of almost all metals and showed statistical significant ($p < 0.05$) in their values only in cases of the metals Pb, Cd, Cr, and Zn, which may

Table 1 Descriptive statistics of metals in different fish species

Specimen	Pb	Cd	Cr	Cu	Ni	Zn
Phansa						
Mean level	18.33[a]	1.03[a]	1.02[b]	24.96[c]	3.17[a]	44.15[a]
Std Dev	12.2	0.17	0.8	6.9	1.23	15.66
Range	4.5–28.2	0.39–1.05	1.15–1.75	16.2–32.5	2.01–4.3	24.6–56.21
Pomfret						
Mean level	12.4[b]	0.99[a]	BDL	16.22[d]	3.41[a]	23.37[c]
Std Dev	8.7	0.4		4.65	2.15	9.9
Range	3.8–21.1	0.55–1.28		10.5–22.7	1.15–5.0	11.2–33.6
Bele						
Mean level	14.28[b]	0.62[b]	BDL	21.39[d]	2.2[b]	12.13[d]
Std Dev	7.65	0.5		8.21	0.99	5.98
Range	5.5–20.7	0.34–0.89		9.8–30.2	0.56–3.1	4.9–22.2
Parse						
Mean level	18.6[a]	1.2[a]	1[b]	32.02[b]	2.25[b]	30.04[b]
Std Dev	10.23	0.78	0.72	17.62	0.85	17.01
Range	5.9–31.6	1.0–3.01	0.25–1.65	14.3–45.9	1.65–3.98	12.4–36.7
Flat fish						
Mean level	19.96[a]	1[a]	3.89[a]	47.97[a]	3.69[a]	44.74[a]
Std Dev	6.85	0.55	1.67	15.22	1.67	15.01
Range	11.4–26.0	0.4–1.40	1.20–4.58	30.3–58.8	1.97–4.7	24.8–48.8

The result of the Groupings of Duncan's Multiple Range Test are also shown (a < b < c < d). Mean with different superscript data for each parameter is significantly different at $p < 0.05$. Concentrations are expressed in mg kg^{-1} dry wt

BDL below detection limit

Table 2 Comparison of metal accumulation in fishes from different regions versus present study

Area	Cd	Cr	Cu	Ni	Zn	Pb	References
Red Sea	0.5–2.0	1.0–10.3	0.5–2.0	1.0–5.0	1.9–35.0	1.5–8.3	Ahmed and Naim (2008)
Gulf of Cumbay	0.23 ± 0.03	0.77 ± 0.05	2.37 ± 0.45	ND	38.24 ± 1.64	1.09 ± 0.07	Reddy et al. (2007)
Southern California	0.60–1.0	ND	12.30–20.80	NA	27.8–54.8	1.6–13.3	Bruce et al. (1975)
Baluchistan coast Pakistan	0.04–0.12	NA	0.41–0.55	NA	3.90–4.32	0.25–0.44	Zehra et al. (2003)
Hooghly estuarine coastal areas	0.62–1.20	ND–3.89	16.22–47.97	2.20–3.69	12.13–44.74	12.40–19.96	Present study

All values are expressed in mg kg^{-1} dry wt

ND not detectable, *NA* not analyzed

be accounted for due to the major functional differences in their body as well as their position in the biological niche with different habits, their biology, physiology or feeding. As because, it is known that these two fish are found either in pelagic or mid-water level of coastal water and all are either plankto-phagous or predatory in habit and high accumulation of Cu and Zn in their mussel could be probably due to higher metabolism rate. On the other hand, the other fishes are benthic species, which live on the bottom, and their concentrations provide information about the degree of bioavailability of heavy metals from the sediments (Louma 1983). Thus, the present study also highlighted that metal accumulation in fish is also species-specific. The concentrations of the heavy metals detected in fishes were compared with the earlier reported values for other regions in an effort to determine the degree of contamination in the study area. The values of Cd in the present study (Table 2) were within the ranges as reported from the red sea area (Ahmed and Naim 2008) and Southern California (Bruce et al. 1975). However, a significant lower value was detected in fish muscles in gulf of Cambay (Reddy et al. 2007) and Baluchistan Coast of Pakistan (Zehra et al. 2003). According to Moore and Ramamoorthy (1984) the concentration of Cd < 1.5 mg kg^{-1} in muscle can be regarded as uncontaminated fish and the levels of Cd in the present study were recorded always less than this value indicating insignificant contamination of Cd in these fish muscles. Cr does not normally accumulate in fish and hence low concentrations were reported even from the industrialized part of the world (Moore and Ramamoorthy 1984). Our results also showed lower ranges (Nil to 3.89 mg kg^{-1}) and considerably lower than the muscles of the fish from the red sea (Ahmed and Naim 2008) indicating a less contamination of fishes in this area. Similarly, the lowest mean level of Ni was 2.5 mg kg^{-1} as examined and was lower than those in other fishes detected else where (Ahmed and Naim 2008). Our data show more pristine condition in the area and thus not necessarily indicative of Ni pollution problem. The mean levels of Pb and Cu were higher in the present study (Table 3) and are not consistent with what has been reported from other areas of Red Sea (Ahmed and Naim 2008), Gulf of Cumbay (Reddy et al. 2007) and Baluchistan Coast (Zehra et al. 2003), but to some extent comparable to the bioaccumulation levels of fish mussel in the polluted zone of Southern California (Bruce et al. 1975). Previous study (Mitra and Chowdhury 1993) on trace metal content in macro-benthic molluscs of this area highlighted elevated levels of both Cu and Zn ranging from 15.65 to 1,782 mg kg^{-1} and from 46.20 to 1,211 mg kg^{-1} of dry wt. The high levels of these metals in animal tissue may be partly due to contamination, caused by the presence of large numbers of vessel repairing facilities in the estuarine areas that use antifouling paints containing appreciable amount of these metals (Goldberg et al. 1983). In addition, the use of cupper sulphate in aqua culture situated in the upstream areas of Hooghly estuary as well as in the surrounding coastal areas as an alternative to antibiotic treatment could increase the levels of this metal and reduce water quality. Contrasting to Pb and Cu, however, the levels of occurrences of Zn in the present study were within the ranges of fish mussel studied in the region of Red sea and Southern California, but obviously less than other studies (Reddy et al. 2007; Zehra et al. 2003). Thus the present study highlights that anthropogenic loadings of the metals of Cu, Pb and Zn in these areas are relatively high due to which the fish muscle registered considerable amount of these heavy metals acting as bio-indicator of metal pollution. Moreover, since the selected species in the present study are of different feeding habits and habitats, the study of their metal contents may provide information about the main routs of heavy metal uptake and thus the above findings are also of interest because they indicate that these levels could be a reflection of bio-available of metal concentration in sediments (Louma 1983). Accumulation of these heavy metals in human beings due to consumption of these contaminated fishes has been assessed and presented in Table 3. According to Centre for Science and Environment (CSE), daily per capita fish consumption of Indian people is 12 gm per day in dry weight, which is about 1% of the total daily diet (CSE 2003) and calculation of weekly intake value of these metals per kg body weight has been done on the basis of this weight, taking 60 kg as an average weight of an individuals. Thereafter, these calculated values were compared with those of Provisional Tolerable Weekly Intake (PTWI) values stipulated by WHO as standard. It is clear that excepting with Pb, calculated PTWI values of other metals are always below the standard value. In respect to Pb, the calculated PTWI values of all fishes like flat fish, Parse and Phansa are marginally higher and thus, contribute significant amount of this metal through consumption. Because of the fact that the standard PTWI values are stipulated on considering the uptake of the toxic metals through all sources of intake like air, water and other various types of foods generally a person likes. Uptake of these fish alone contributes more than 100% of the tolerable enrichment of Pb in human through a single source. But it is to be noted that if a person consumes this amount of fish muscle for successive 7 days in a week, only then, the body accumulation level crosses the standard PTWI values. Moreover, these fishes are costly enough and also not always available in the respective market. Besides, the antagonistic effect caused by the presence of other metals may reduce the metal toxicity to a great extent. So, without having knowledge on the cumulative effect of all metals comprising of both essential and non-essential elements on the degree of metal toxicity, no concrete information could be made merely by the present studies.

Yet, the present study also highlights a grave concern that these areas are not at all free from heavy metal contamination and so the trace element emission into coastal waters also needs to be examined for its impact on the ability of marine organisms to survive and reproduce. A

Table 3 Accumulation level of different heavy metals in human being due to consumption of different fishes $week^{-1}$ kg^{-1} body wt

Specimen	Pb (μg kg^{-1})	Cd (μg kg^{-1})	Cr^a (μg kg^{-1})	Cu^a (μg kg^{-1})	Ni^a (μg kg^{-1})	Zn^a (μg kg^{-1})
Phansa	25.66	1.44	1.43	34.94	4.44	61.81
Pomfret	14.88	1.39	Nil	22.70	4.77	32.72
Bele	19.98	0.87	Nil	29.94	3.08	16.98
Parse	26.04	1.68	1.40	44.80	3.99	42.40
Flatfish	27.6	1.40	5.45	137.16	5.17	62.60
Std. PTWI Values	25.0	7.0	637	2,500	35	2,000
Ranges of $(PTWI_{cal}/PTWI_{std}) \times 100$	60–110	12–24	0.21–0.85	0.92–5.5	8.8–15.6	0.80–3.2

[a] Essential group of metals and their PTWI values (μg kg^{-1}) corresponds to minimum requirement of a person per kg body weight

proper evaluation of the most important and valuable coastal fisheries is necessary prior to the future development of industries or power plants near the marine environment.

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