

Assessment of Arsenic, Cadmium and Mercury Level in Commonly Consumed Coastal Fishes from Bay of Bengal, India

Mukherjee D. P.[1] and Kumar Bhupander[2*]

1. Central Pollution Control Board, 502, Rajdanga Road, Kolkata-700107, India
2. Central Pollution Control Board, East Arjun Nagar, Delhi-110032, India

*Corresponding Author: bhupander_kumar@yahoo.com

Abstract

Toxic heavy metal concentrations of Cadmium (Cd), Mercury (Hg) and Arsenic (As) were determined in muscle tissue of six marine fish species collected from north eastern Bay of Bengal, India. The concentrations of arsenic, cadmium and mercury were in range of 0.02-2.34 µg g^{-1}, 0.01-2.10 µg g^{-1} and 0.07-1.60 µg g^{-1} dry wt., respectively. Arsenic was the higher in average concentration followed by mercury and cadmium and their average concentrations were 0.66±0.09 µg g^{-1}, 0.62±0.05 µg g^{-1} and 0.47±0.07 µg g^{-1} dry wt., respectively. The concentration of heavy metals was species specific and metal specific significantly varied. The estimated intake values of these metals through human consumption were calculated (µg kg^{-1} body wt. day^{-1} and $weekly^{-1}$) and were compared with those of Provisional Tolerable Weekly Intake (PTWI) per kg body weight as stipulated by the Food and Agriculture Organization/World Health Organization (FAO/WHO) Joint Expert Committee on Food Additives (JECFA). The Estimated Tolerable daily intake $TDI_{(Estimated)}$ and Estimated Tolerable weekly intake $TWI_{(Estimated)}$ were lower than stipulated guidelines and therefore, were not considered to pose adverse effects to the humans. The Pearson product moment correlation was calculated and found that cadmium was positively correlated with mercury. Since there is a bioaccumulation of toxic heavy metals in fish tissues, therefore, a regular monitoring of heavy metals and other toxic pollutants such as polychlorinated biphenyls (PCBs) dioxins, and furans is proposed for fishes from northern Bay of Bengal.

Keywords: Heavy Metal, Arsenic, Cadmium, Mercury, Coastal Fish, PTDI, PTWI, Bay of Bengal, India

1. Introduction

Though preventive measures have been taken to reduce the input of trace metals into oceans, rivers and estuaries, accumulation in the different aquatic systems have been reported even today (Mortazavi and Sharifian, 2011; Kumar, *et al*., 2010; Meltem and Esra, 2010; Sajwan, *et al*., 2008; Senthil kumar, *et al*., 2007; Paller and Litterell, 2007). The anthropogenic activity generating cause for environmental concern and may have divesting effects on the ecological balance of the aquatic environment and diversity of biota (Forstner and Wittmann, 1983; Farombi, *et al.*, 2007). Trace elements that exist naturally at background levels in the environment include chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), manganese (Mn), and zinc (Zn), which are essential elements in living organisms. However, some trace elements or heavy metals such as mercury (Hg), cadmium (Cd), arsenic (As), chromium (Cr), and lead (Pb) are not required for metabolic activity and are toxic.

Heavy metals have the tendency to accumulate in the various aquatic animals, and the accumulation depends upon the intake and the elimination from the body (Karadede, *et al.*, 2004). Marine fishes exposed to these heavy metals have been consumed as sea foods and, hence are a connecting pathway for the transfer of toxic heavy metals in human beings. Fishes are major part of the human diet because, it has high protein content, low saturated fat and also contains omega fatty acids known to support good health (Dural, *et al*., 2007; Ikem and Egiebor, 2005). Therefore, various studies have been taken worldwide on the contamination of different fish species to determine their heavy metal concentration (Raja, *et al.*, 2009; Yilmaz, 2009; Ahmed and Nain, 2008; Nawal, 2008; Sivaperumal, *et al*., 2007; Carvalho, *et al*., 2005). Heavy metal studies of aquatic biota give an idea of how heavy metal concentrations in these organisms could become a more reliable water quality indicator compared to the chemical analysis of water and sediment.

Little information is available pertaining to the accumulation of heavy metals in coastal biota especially coastal fishes collected earlier from the North Eastern Bay of Bengal, India (Mitra, *et al.*, 2000; Bhattacharya, *et al*., 2001 & 2006; Sarkar, *et al.*, 2002). In this study we emphasized on measurement and distribution of heavy metals i.e. Cadmium (Cd), Mercury (Hg) and Arsenic (As) in the muscle tissues of six different but commonly consumable coastal fishes, from West Bengal coast, north east coast of India. Further, observed levels of heavy metal concentrations compared to the Provisional Tolerable Weekly Intake (PTWI) for toxic metals as set by the Food and Agriculture Organization/World Health Organization Joint Expert Committee on Food Additives (JECFA, 1978; 1989; 1983)

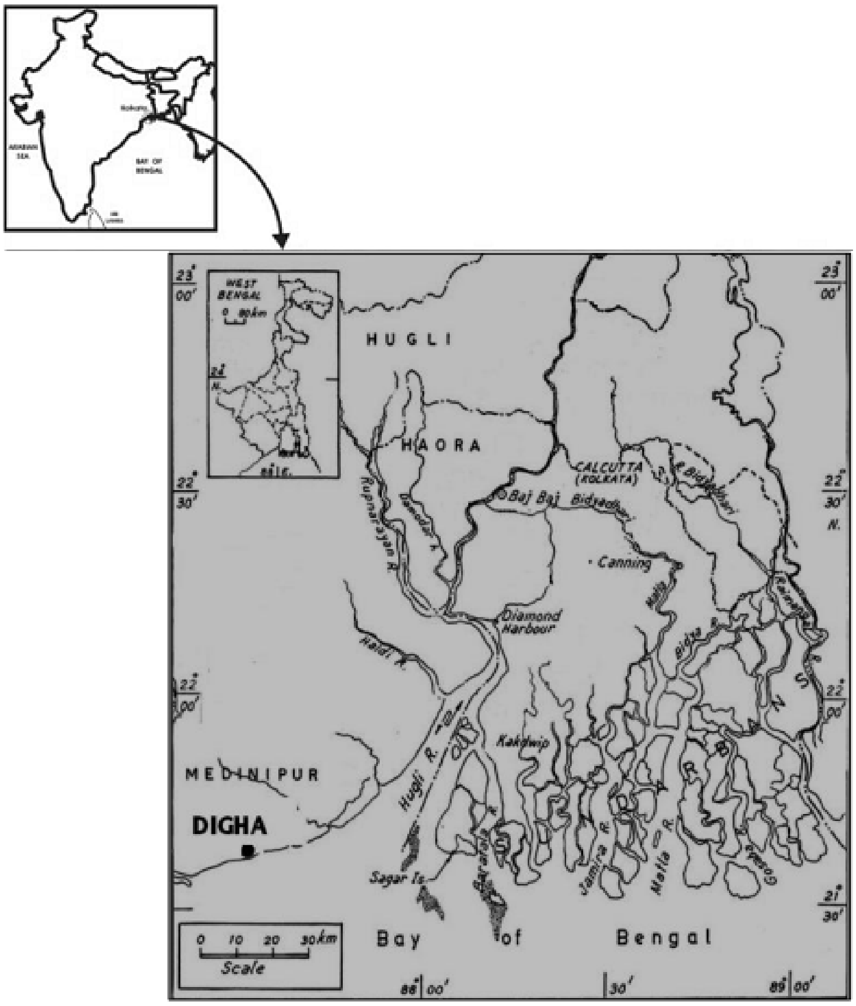

Figure 1: Map showing sampling collection location, North East Bay of Bengal, India

2. Materials and Methods

2.1 Sampling site description

Fish sampling was conducted in Digha a seaside resort and fish landing station at 24.68^{0}N, and 87.55^{0}E located in east Medinipur district of West Bengal and at the northern end of the Bay of Bengal (Figure 1). The geomorphology, hydrodynamics and ecology of the catchment area of the Digha is largely influenced by the estuarine and tidal network system of river Ganga (Hugli). The Ganga (Hugli) is the main river system in India, which covers a large area from north India and joins the Bay of Bengal to form deltaic Sunderbans ecosystem. The huge discharges through this reverine system, of municipal and industrial wastes was generated from industries include fertilizer, paints and pigments, dye manufacturing units, electroplating units and thermal power plants. A significant ecological change was pronounced in this area due to influence of Ganga (Hugli) river and wastes from Haldia ports, a major oil disembarkment terminal in eastern India (Chatterjee, *et al*., 2007).

2.2 Sampling

Six fish species, Bombay duck *(Harpadon nehereus)*, Bhola (*Daysciaena albida)*, white pomfret *(Pumpus argentius*), black pomfret *(Formio niger)*, Hilsa (*Hilsa ilisha)*, and mackerel (*Rastrelliger kanagurta)* were selected for this study. The selected fishes were the most abundant and commercially important species consumed by the people. Samples were collected from different counters of the fish landing station at Digha. Fish samples were labeled, they were preserved using ice and transported to main laboratory. All the samples were kept at -20^{0}C until further pre-treatment and analysis.

2.3 Sample pre-treatment

Samples were thoroughly washed with Mili-Q water after removing the scales, and muscle portion, which was taken for further processing. Muscle tissue was oven dried at 110^{0}C, powdered with pestle and mortar and stored until chemical analysis. Heavy metals were analyzed after digesting the homogenized samples in a mixture of nitric and perchloric (Honda, *et al*., 1982). Digestion was carried out after 0.5 gm homogenized powdered sample was placed in a Teflon beaker and digested with few drops of sodium chloride solution (30%) and a 10 ml mixture (1:5) of concentrate Nitric acid (65%) and concentrated Perchloric acid (70%). The free chlorine developed loosens the chemical bonds in organic compounds after gentle heating (at 70±5^{o}C) in a water bath for 12 hrs and destroys the organic matter in order to transfer the metals into the solution. The digested samples were centrifuged and the supernatant was analyzed. The results were expressed in $\mu g\ g^{-1}$ metal dry weight.

2.4 Instrumental analysis

Determinations of cadmium were carried out using graphite furnace Atomic Absorption Spectrometry (GF-AAS, Thermo, UK). Hydride generator (HG) coupled to atomic absorption spectrophotometer was used to analyzed total mercury (cold vapor mode) and arsenic (heating mode). Background corrections were applied whenever required during in the analysis and the method of standard additions was used to compensate for matrix effects. Performance of the instrument was checked by analyzing the standard reference material solutions (Merck NJ, USA) concurrently to check the precision of the instrument. After appropriate dilutions of stock standard solutions a five level calibration curve was prepared. Samples were analyzed in triplicate. The values obtained from the sample then corrected for final digestion volume and sample weight taken. Duplicate method blanks were also processes and analyzed alongside the samples to check any loss or cross contamination. The detection limit for Cd, Hg, and As was, 0.005, 0.001, and 0.002 ppm, respectively. The results were reported on dry weight basis.

2.5 Statistical analysis

Inter-heavy metal correlations in the fish muscle were investigated. The Pearson correlation coefficient was used to measure the strength of the association between heavy metal concentrations in muscle tissue and presented in correlation matrices (Pentecost, 1999). The *p*-values of less than 0.01 and 0.05 were considered to indicate statistical significance.

3. Result and Discussions

Concentration of cadmium, mercury and arsenic in fish muscle were presented in Table 1 & Table 2. The study of heavy metal concentrations in fishes was important with respect to human consumption of fish. Several studies shows heavy concentration in tissue of coastal fishes may vary considerably among the different species. This was possibly due to differences in metabolism and feeding patterns of the fishes.

3.1 Heavy Metal Concentrations in Fishes

The concentrations of heavy metals fishes in the present study were: As > Hg > Cd (Table 1). The average concentrations of cadmium, mercury and arsenic in muscle tissues were 0.47±0.07 µg g^{-1}, 0.62±0.05 µg g^{-1} and 0.66±0.09 µg g^{-1}, respectively. The ranges of concentration were 0.01-2.10 µg g^{-1}, 0.07-1.60 µg g^{-1} and 0.02-2.34 µg g^{-1} for Cd, Hg and As, respectively. These non-essential metals have no biological function or requirement and their concentrations in coastal fishes are generally low (Yilmaz, 2009; Ahmad, *et al.*, 2008).

Table 1: Concentrations of cadmium, mercury and arsenic in muscle tissue of fishes

	Heavy metal concentration (µg g^{-1} dry wt.)					
Heavy metals	min	max	mean	median	Std Err*	CV**
Cadmium	0.01	2.10	0.47	0.17	0.07	129.20
Mercury	0.07	1.60	0.62	0.40	0.05	73.51
Arsenic	0.02	2.34	0.66	0.41	0.09	108.87

*Std Err=standard deviation/√n, **CV=coefficient of variation

3.2 Heavy Metal Concentrations in Species

In our study the metal concentrations in muscle tissues varied significantly among the ten species of studied fishes (Figure 2 & Figure 3). Accumulations of metals were generally found to be species specific and may be related to their feeding habits and the bio-concentration capacity of each species (Fariba, *et al.*, 2009; Naim and Ahmed, 2008; Agoes and Hamami, 2007; Huang, 2003).

Cadmium values in this study ranged from 0.01 µg g^{-1} (*Rastreliger kanagurta*) to 2.10 µg g^{-1} (*Pampus argentius*) and the average concentration was 0.47±0.07 µg g^{-1} (Table 1 & 2). The observed values were similar to earlier studies from north east coast of bay of Bengal, India (De, *et al.*, 2010) and fishes from other geographical areas of gulf of Aquaba, Red Sea (Ahmed and Naim, 2008; Naim and Ahmed, 2008), Black, Marmara, Aegean and Mediterranean seas, Turkey (Mustafa, *et al*., 2008), and Iskenderun Bay, Turkey (Dural, *et al*., 2007). The observed values were lower than cadmium in fishes from Mangalore and Kochi, south est coast of India (Rejomon, *et al.*, 2010) but comparatively higher than fish species from Eastern Mediterranean coast, Turkey (Meltem and Esra, 2010), South east coast of India (Raja, *et al.*, 2009), Lagos lagoon, west Nigeria coast (Aderinola, *et al.*, 2009), gulf of Cambay, India (Reddy, *et al.*, 2007) and fishes from Gresik coastal waters of Indonesia (Agoes and Hamami, 2007). Cadmium has a high potential for bio-concentration in fish and is accumulated in multiple organs. The order of cadmium concentration in species was *Harpadon nehereus* > *Rastreliger kanagurta* > *Hilsa ilisha* > *Formio niger* > *Pampus argentius* > *Daysciaena albida.*

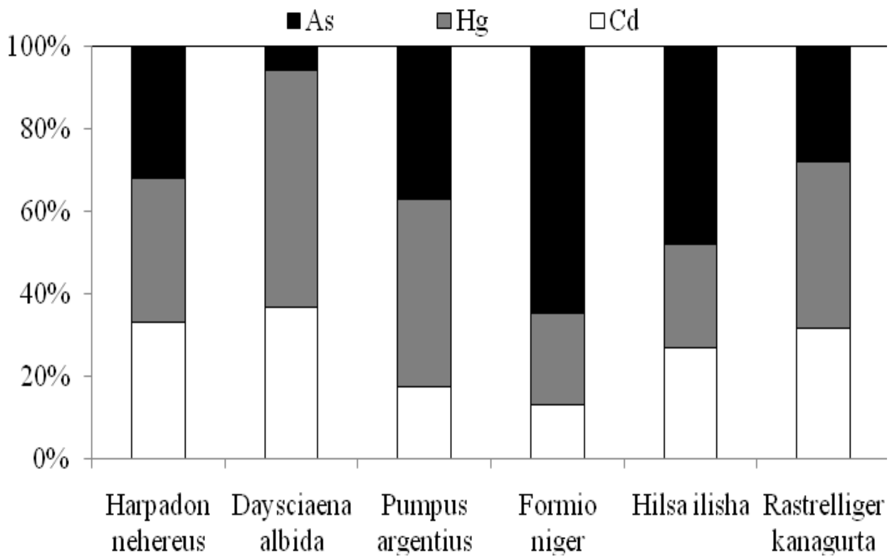

Figure 2: Percent distribution of cadmium, mercury and arsenic in muscle of fish species

Table 2: Concentrations of cadmium, mercury and arsenic in muscle tissue of fish species

Heavy metals ↓	**Statistics**	**Name of fish species**					
		Harpadon nehereus	***Daysciaena albida***	***Pumpus argentius***	***Formio niger***	***Hilsa ilisha***	***Rastrelliger kanagurta***
Cadmium	range	0.17-2.07	0.05-0.60	0.03-2.10	0.06-0.41	0.06-1.10	0.01-2.07
	mean	0.86	0.29	0.27	0.16	0.40	0.73
	median	0.37	0.07	0.08	0.11	0.15	0.40
	Std Err*	0.25	0.10	0.18	0.04	0.11	0.19
	CV**	94.75	99.08	138.29	85.80	108.73	95.81
Mercury	range	0.30-1.57	0.17-0.73	0.22-1.60	0.07-0.40	0.07-0.90	0.27-1.60
	mean	0.91	0.46	0.70	0.28	0.37	0.93
	median	0.95	0.48	0.34	0.34	0.34	1.00
	Std Err	0.14	0.08	0.15	0.04	0.06	0.13
	CV	49.67	50.34	77.29	47.52	61.99	54.08
Arsenic	range	0.07-1.96	0.02-0.17	0.02-1.74	0.02-2.34	0.02-2.34	0.07-1.63
	mean	0.83	0.05	0.57	0.80	0.71	0.65
	median	0.73	0.05	0.36	0.44	0.44	0.51
	Std Err	0.21	0.05	0.16	0.34	0.23	0.14
	CV	83.45	64.15	98.97	132.87	122.23	82.78

*Std Err=standard deviation/√n, **CV=coefficient of variation

The concentration of mercury in muscle tissues of different fish species from West Bengal coast varied from 0.07 to 1.60 $\mu g\ g^{-1}$ (mean 0.62±0.05 $\mu g\ g^{-1}$) (Table 1). The highest average concentration was observed in *Rastreliger kanagurta* (1.00 $\mu g\ g^{-1}$), and lowest was in *Formio niger* (0.04 $\mu g\ g^{-1}$) (Figure 2). The average concentration of Hg in fishes from Bay of Bengal were comparable with mercury in muscle tissue of marine fishes from Mosa Bay, Persian Gulf (Mortazavi and Sharifian, 2011), Malaysia (Hajeb, *et al.*, 2009), Bangladesh (Sharif, *et al.*, 2008) and Thailand (Agusa, *et al.*, 2007), however, concentrations were lower than those reported in fish from Gulf of Cambay (Reddy, *et al.*, 2007), but higher than Saudi

Arabia (Nawal, 2008; Waqar, 2004). Different species of fishes from West Bengal coast accumulated mercury in the order of: *Rastreliger kanagurta* > *Harpadon nehereus* > *Daysciaena albida* > *Hilsa ilisha* ≥ *Pampus argentius*, ≥ *Formio niger*.

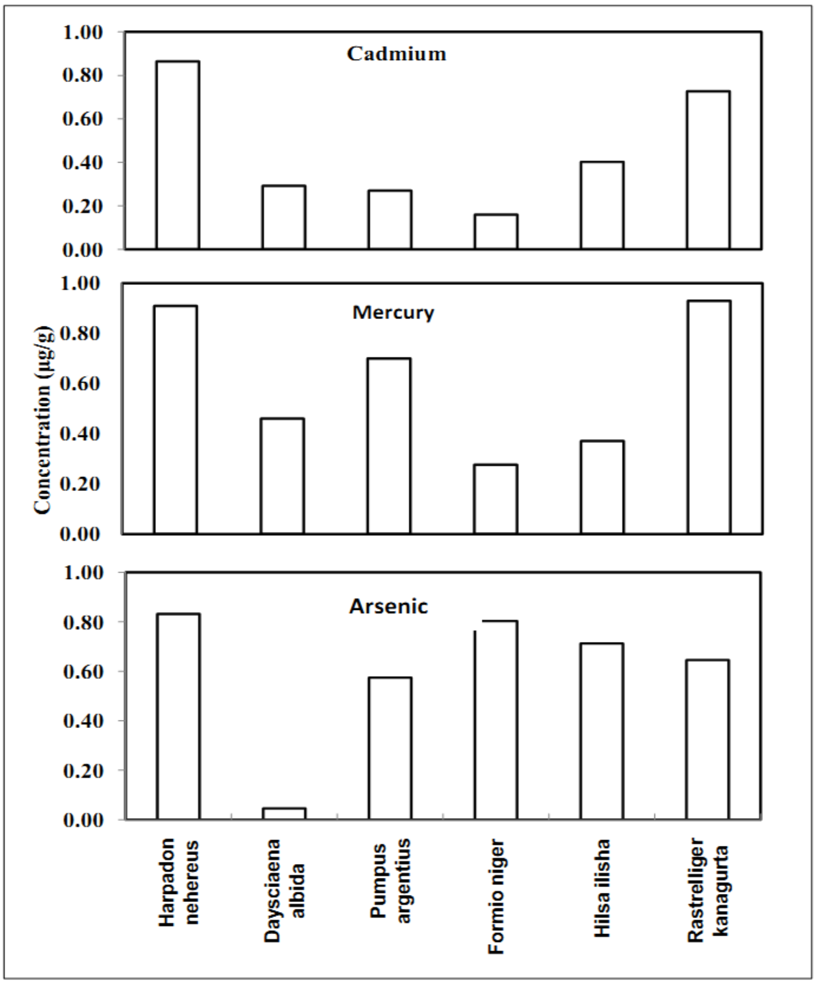

Figure 3: Heavy metal concentration profiles in different species of fishes

There was considerable variation of arsenic levels among the fish species from Bay of Bengal. The concentration of Arsenic was in range of 0.02-2.34 µg g^{-1} with the mean concentration of 0.66 µg g^{-1} (Table 1). Comparatively higher concentration range were observed in *Formio niger* and *Hilsa ilisha*. The observed concentrations of arsenic in muscle tissue were lower than those reported from Gulf of Cambay in North West coast of India (Reddy, *et al.*, 2007); Fangauta Lagoon, Tonga (Morrison and Brown, 2003); Pahang estuary, Thailand (Rattanachongkiat, *et al.*, 2004) and American Samoa, south pacific ocean (Peter, *et al*., 2007), but higher levels than our observations were observed in fishes from Gresik coastal waters of Indonesia (Agoes and Hamami, 2007). The observed average concentration order in different species was *Harpadon nehereus* > *Formio niger* > *Hilsa ilisha*, *Rastreliger kanagurta* > *Pampus argentius* > *Daysciaena albida*.

Pearson moment correlation coefficient analysis shows that cadmium is statistically (p=0.01 and p=0.05) correlated with mercury (r^2=0.703). Arsenic has not shown any positive correlation with mercury and cadmium.

3.3 Estimated Tolerable Intake of Heavy Metals

Foods having toxic metals could present a toxic hazard for the consumer which is dependent on the metal concentration in food and amount of food consumed (Hajeb, *et al*., 2009). Hazard consists of determining the toxicological properties related to a specific substance (Kuhnlein and Chan, 2000). The 'tolerable intake' is widely used to describe 'safe' levels of intake; and can be expressed on either a daily basis (TDI or tolerable daily intake) or a weekly basis (TWI or tolerable weekly intake). The tolerable intake of heavy metals as PTWI (Provisional Tolerable Weekly Intake), are set by the Food and Agriculture Organization/World Health Organization (FAO/WHO) Joint Expert Committee on Food Additives (JECFA). PTWI is the maximum amount of a contaminant to which a person can be exposed per week over a lifetime without an unacceptable risk of health effects.

The amounts daily and weekly consumption of fresh fish muscle In India, are 19.5 and 136.2 g, respectively (Speedy, 2003). The estimation of dietary intakes combines data on the levels of the heavy metal in fish muscle with the quantities of fish consumed daily or weekly by the population. The estimated tolerance daily intake (ETDI) and estimated tolerance weekly intake (ETWI) were calculated and presented in Table 3. Intake estimates were expressed as per unit body weight (µg/kg b.w. /day or weekly). The overall, ETDI for arsenic, cadmium and mercury was 0.18, 0.13, and 0.17 µg/kg b.w./day, respectively, and ETWI was 1.28, 0.91 and 1.21 µg/kg b.w./week, respectively.

Table 3: Estimated dietary intake (µg/kg b.w./day or week) of cadmium, mercury and arsenic of fish species from Bay of Bengal, India

Fish species	Cadmium		Mercury		Arsenic	
	EDI*	EWI**	EDI	EWI	EDI	EWI
Harpadon nehereus	0.24	1.68	0.25	1.77	0.23	1.62
Daysciaena albida	0.08	0.57	0.13	0.89	0.01	0.09
Pumpus argentius	0.08	0.53	0.19	1.36	0.16	1.12
Formio niger	0.04	0.31	0.08	0.54	0.22	1.56
Hilsa ilisha	0.11	0.78	0.10	0.72	0.20	1.39
Rastrelliger kanagurta	0.20	1.41	0.26	1.81	0.18	1.25
All samples	0.13	0.91	0.17	1.21	0.18	1.28
WHO/FAO	1	7	0.7	5	2.14	15

*EDI=Estimated daily intake, **EWI=Estimated weekly intake

3.3.1 Cadmium

Cadmium may accumulate in humans from food chain magnification and may induce kidney dysfunction, skeletal damage, and reproductive deficiencies (Commission of the European communities, 2001). The PTWI for cadmium has been set, by the JECFA, at 7 µg/kg b.w./week (equivalent to 1µg/kg b.w./day) (WHO, 1993). This value of the PTWI takes into consideration the fact that provides suitable time of renal exposure (WHO, 1989). This PTWI value corresponds to a daily tolerable intake level of 70 µg of Cd per day for the average 70 kg man and 60 µg of Cd per day for the average 60 kg woman. The ETDI and ETWI for cadmium in this study were 0.13 1µg/kg b.w. and 0.91µg/kg b.w. for daily and weekly consumption, respectively (Table 3, Figure 4). Clearly, indicates the Cd intake for the general population from fishes from this region is well below the guidelines.

3.3.2 Mercury

The toxicokinetics of mercury is associated with its chemical form: elemental, inorganic and organic. The organic form, usually methylmercury (MeHg) is more hazardous than both other forms. The liver and kidneys of stock animals, fish and shellfish tend to concentrate environmental mercury. Marine organisms possess a remarkable capacity to turn inorganic mercury into organic compounds (MeHg), thus rendering mercury more easily transferable throughout the aquatic food chain. As a result, marine organisms contain mercury in levels up to 5 mg/kg (Dudka and Miller, 1999).Earlier studies have shown that fish consumption may constitute an important source of mercury exposure for human (Fakour, *et al*., 2010; Díez, *et al*.,2008; Nasreddine and Parent-Massin, 2002). JECFA has established a PTWI of 3.3 µg MeHg/kg b.w./week for the general population (equivalent to 0.5 µg MeHg/kg b.w./day) but notified that pregnant and breast feeding women are likely to be at much greater risk due to the vulnerability of embryos and infants (WHO, 1989). JECFA has previously set a total mercury PTWI of 5 µg/kg b.w./week, which is equivalent to 0.7 µg/kg b.w./day (WHO, 1978). The observed ETDI and ETWI of Hg from this study were 0.17 and 1.21 µg/kg b.w. for daily and weekly consumption, respectively (Table 3, Figure 4), and therefore, were not considered to pose adverse effects to consumers.

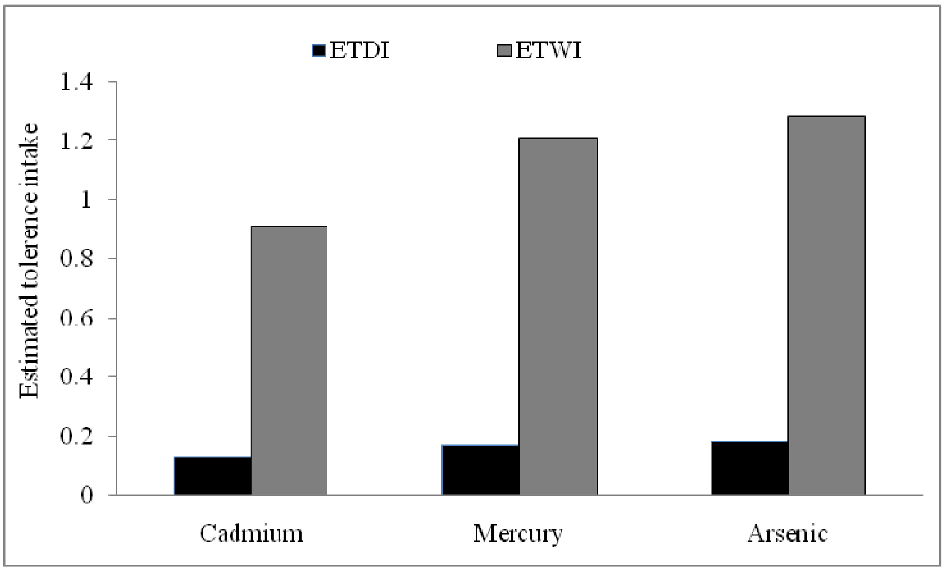

Figure 4: The estimated daily and weekly intakes of Cadmium, Mercury and Arsenic (µg/kg b.w./day or week) of the studied samples from the Bay of Bengal, India

3.3.3 Arsenic

The inorganic arsenic forms are more hazardous to humans than the organic ones, such as arsenobetaine, which are generally of low toxicity. In fact several days after its ingestion, around 80% of arsenobetaine get excreted unchanged from the human body. Both organic and inorganic forms of arsenic occur in food. It has been established that fish and seafood can accumulate sizeable quantities of organic arsenic from their environment. The JECFA has established a PTWI of 15 µg/kg b.w./week which is equivalent to 2.14 µg/kg b.w./day for inorganic arsenic (WHO, 1989). The daily intake of arsenic by humans reflects generally the quantities of seafood in the diet in which arsenic occurs mainly in the organic form. The ETDI and ETWI for arsenic was higher than Cd and Hg, with a range from 0.01 to 0.23 µg /kg b.w./day (mean, 0.18 µg /kg b.w./day) and 0.09 to 1.62 µg /kg b.w./week (mean, 1.28 µg /kg b.w./week), respectively (Table 3, Figure 4). Based on WHO data, the observed arsenic concentration in fish muscle tissues were much lower and pose no adverse effects.

4. Conclusion

When considering the heavy metals concentrations in fish species, suitable for human consumption, the most important aspect is their toxicity to humans. The results of this study revealed that consuming fish from north eastern Bay of Bengal may not have harmful effects because observed values of dietary intake of heavy metals were far below the permissible PTWI and PTDI limits for human consumption. More intensive study is needs to be conducted in order to determine the bioaccumulation of heavy metals in fishes from the study area. Further study on accumulation of organochlorines pesticides, PCBs, PAHs, and dioxins in fish tissues should be undertaken due to usage of these chemicals in India.

5. Acknowledgements

Authors are highly thankful to the Chairman and Member Secretary Central Pollution Control Board for support and encouragement to conduct this study. Mr. B. R. Naidu, then Incharge Zonal Office, Central Pollution Control Board, Kolkata is also acknowledged for providing the analytical facilities.

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