

## Distribution of Mercury in different abiotic and biotic sectors of the Mandovi-Zuari estuary (Goa)

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This study highlights the behaviour of Mercury in the abiotic and biotic segments of the Mandovi-Zuari estuarine system. Spatial, seasonal (Premonsoon and Postmonsoon) and tidal, distribution of THg in dissolved/particulate fractions, sediment concentrations were mapped of the river mouths, Mormugao Port, inner and outer environs of this estuarine system. Observed seasonal partition studies provided an insight into the removal of the metal into the particulate fraction and subsequent deposition in sediments in the postmonsoon. Bioconcentration factor provided a link in the Hg cycle between the abiotic and biotic compartments of this ecosystem. Metal contaminations in different tissues of biota representing different trophic levels were evaluated by calculating the bioconcentration factor. Although biomagnification and bioaccumulation was observed in the representative biota of commercial importance, the observed total mercury levels are well within the safety limit 0.5 ppm set by WHO and 0.3 ppm set by UNEP for fish products for human consumption, thereby indicating no apparent mercury contamination.

**[Keywords:** Mandovi-Zuari, Dissolved, Particulate, Sediment, Biomagnification and bioaccumulation.]

### Introduction

Mercury (Hg) is one of the most toxic elements and its toxicity to humans has been well established<sup>1</sup>. It has been recognised as a severe environmental pollutant, not only because of its high toxicity, even at low concentrations, but also because of its ability to enter into biological systems<sup>2</sup>. The environmental behaviour of Mercury is interesting and differs from those of other toxic elements as it is ubiquitous and can exist as both a liquid and as a volatile form at ambient temperatures<sup>3</sup>. Mercury cycle is complex; the aquatic compartment represented by estuaries and coastal waters constitutes an important link between the terrestrial environment and the open ocean<sup>4</sup>. Estuaries serve as filters of river derived signatures of contamination. Accumulation of Mercury in biota depends on various physicochemical and biological factors. The ability of aquatic organisms to accumulate pollutants in their tissues makes them a useful index of pollution. The principal pathway for mercury exposure in humans is food consumption, particularly fish and its products, and exposure to Hg levels in multiple environmental compartments<sup>5,6</sup>.

Mandovi and Zuari estuaries are located on west coast of India in Goa. They are the lifelines of the state, used for navigation and transport of Iron and

Manganese ores from the hinterland to Mormugao harbour. Their physical, chemical and biological features are subjected to a seasonal rhythm induced by the annual cycle of the monsoon<sup>7</sup>.

The aim of the present investigation is to 1) Obtain an accurate Hg determinations in abiotic (water, sediment, particulate) and biotic (biota representing different trophic levels) compartments of this estuarine system, in order to assess their current contamination levels; 2) Explore the processes influencing the partitioning of mercury, between the dissolved and particulate fractions of seawater; 3) Assess the tissue wise distribution of THg from the food hygiene point of view, 4) Highlight the bio-magnification and bio-accumulation observed in the food web.

### Materials and Methods

The study area incorporated eighteen stations which spanned R. Mandovi & R. Zuari mouth, Mormugao port, and five transects covering the inner and outer estuarine region of Mandovi and Zuari. This tropical estuarine region located off Goa is regularly flushed with tides<sup>7</sup>. It extends from 15° 20' to 15° 30' N and 73° 40' to 73° 50' E. Although there was no apparent point source of Hg for both estuaries, they

are affected by non point inputs from sewage, mining and port activities. Seasonal sampling Premonsoon (March 2012) and Post monsoon (December 2012) was carried out in eighteen sampling stations (Fig. 1). These locations include five transects, three off Mandovi (March 2012 & December 2012) and two off Zuari (December 2012) each spanning three stations 0.5, 2 and 5 km from coast. Water depth varied from 4-20 m. Diurnal sampling was carried out at R. Mandovi and R. Zuari mouth at six hourly intervals for thirty six hours. Particulate samples were collected at nearshore stations of each transect (M1, M4, M7) during March 2012 at Low and High tide. In December 2012 the river mouth stations were sampled for partition studies at both tides.

#### Sampling and Storage

Water samples were collected using acid washed Niskin samplers at low and high tide, acidified and stored in glass bottles till analyses. Water was filtered through 0.45 microns to separate the dissolved and particulate fractions. Particulates on Millipore filters were stored in petriplate and dried in desiccators. The dissolved Hg fraction was determined from the difference between THg and particulate Hg. Surface sediments were sampled by clean Van veen grab from same locations and preserved at  $-20^{\circ}\text{C}$  till the analysis. Sediments were dried at  $35^{\circ}\text{C}$  and then ground to fine powder using an agate pestle mortar. Zooplanktons were collected by trawling the plankton net along five transects in Fig. 1. After collection, samples were stored at  $-20^{\circ}\text{C}$  till analysis was carried out at shore based laboratory. Ecologically and commercially important biota representing different

trophic levels (plankton, planktivores, filter feeders and predatory fish) of this estuarine system were assessed for THg. Planktivores fishes (*Tenulosa toli*, *Rastrelliger kanagartha*, *Sardinella longiceps*) filter feeding bivalves (*Paphia malabarica*, *Meretrix*, *Saccostrea cucullata*) predatory fish (*Arius maculatus*, *Johnius amblycephalus* and *Selaroides leptolepis*).

Fish samples were collected from the landing sites, dissected and tissues were separated (gill, liver, muscle) to evaluate THg in wet tissue. Filter feeders were collected from intertidal locations of R. Mandovi and after taxonomic identification of organisms, soft tissue was separated from the shells and analysed to determine the Mercury concentration by wet weight.

Mercury content in water, particulates, sediment and biota were analysed directly without sample preparation using the Direct Mercury Analyser, DMA-80 Tri-Cell (Milestone, Inc.) which follows EPA 7473 method<sup>8</sup>. This analytical technique is combination of thermal decomposition, amalgamation followed by atomic absorption detection<sup>8,9</sup>. Tri Cell model is state of the art, sensitive direct Mercury analyser with dynamic range of 0.0015 ng – 1200 ng. Calibration curves were generated, over a concentration range between 0.5 to 500 ng/g Hg, using multi-element standard solution (Merck, Germany). DMA Tri-cell is equipped with concentrate function which allowed the accurate measurements of low Hg in water samples. The precision and accuracy for THg estimation in different types of samples was verified against international reference materials, MESS-3 (Marine Sediment), NASS-5 (Seawater), LUTS-1 (Hepatopancreas of lobster) and DORM-4 (Dog fish tissue) from NRC, Canada. A close agreement with the certified values were obtained and presented in Table 1.

#### Results

Observed Mercury concentrations in water: THg, dissolved/particulate and sediment comprising the abiotic compartment of this ecosystem are presented in Table 2.

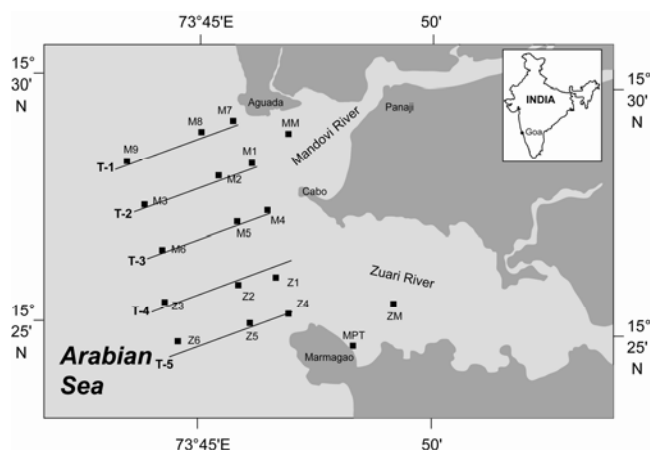


Fig. 1—Study Area- Map showing sampling locations

Table 1—Values obtained for certified reference materials

CRM	Reference value $\pm$ SD	Estimated value $\pm$ SD
NASS-5*	0.3 $\pm$ 0.1 ( $\mu\text{g/L}$ )	0.37 $\pm$ 0.08 ( $\mu\text{g/L}$ )
MESS-3	0.091 $\pm$ 0.009 ( $\mu\text{g/g}$ )	0.092 $\pm$ 0.0008 ( $\mu\text{g/g}$ )
LUTS-1**	16.7 $\pm$ 2.2 ( $\mu\text{g/Kg}$ )	17.35 $\pm$ 1.96 ( $\mu\text{g/Kg}$ )
DORM-4	410 $\pm$ 55 ( $\mu\text{g/Kg}$ )	407 $\pm$ 17.67 ( $\mu\text{g/Kg}$ )

\*Data obtained from Yu, L.Y *et al.* (2007)

\*\*Data obtained from Olson, L.M *et al.* (1991)

Table 2—Hg concentrations in water (THg, Dissolved/ Particulate Hg) and Surface sediments

Season			Mandovi estuary		Zuari estuary	
			Surface	Bottom	Surface	Bottom
Pre Monsoon	THg-Water ( $\mu\text{g/L}$ )	LT	0.54 (0.10-1.49)	0.29 (0.06 -0.64)	-	-
		HT	0.36 (0.05- 1.54)	0.32 (0.09 – 0.63)	-	-
	Dissolved Hg ( $\mu\text{g/L}$ )	LT	0.55 (0.38-0.80)	0.22 (0.01-0.37)	-	-
		HT	0.26 (0.14-0.37)	0.23 (0.12-0.32)	-	-
	Particulate Hg ( $\mu\text{g/g}$ )	LT	0.73 (0.40 -1.17)	0.43 (0.23-0.70)	-	-
		HT	0.30 (0.09-0.51)	0.39 (0.21-0.56)	-	-
Sediment ( $\mu\text{g/g}$ )		0.07 (0.04- 0.10)		-	-	
Post Monsoon	THg- Water( $\mu\text{g/L}$ )	LT	0.21 (0.11- 0.47)	0.17 (0.09-0.26)	0.09 (0.04-0.13)	0.08 (0.02-0.12 )
		HT	0.08 (0.03 - 0.12)	0.09 (0.05 -0.23 )	0.08 (0.04-0.18)	0.09 (0.05 -0.23)
	Dissolved Hg ( $\mu\text{g/L}$ )	LT	0.07	0.04	0.13	0.07
		HT	0.12	0.11	0.06	0.02
	Particulate Hg ( $\mu\text{g/g}$ )	LT	1.43	2.31	2.58	5.57
		HT	3.69	5.21	2.65	3.93
Sediment ( $\mu\text{g/g}$ )		0.08 (0.05 – 0.20)		0.08 (0.04-0.15)		

Table 3—Observed values of THg in Organisms

Organism	Concentration ( $\mu\text{g/Kg}$ wet weight)		
Planktons	2.29 $\pm$ 0.22		
<i>P. malabarica</i>	4.96 $\pm$ 0.91		
Filter feeding bivalves	14.99 $\pm$ 1.51		
<i>S. cucullata</i>	3.14 $\pm$ 0.82		
	Gill	Liver	Muscle
<i>R. kanagurta</i>	9.83 $\pm$ 1.46	3.07 $\pm$ 0.83	0.91 $\pm$ 0.2
Planktivores	1.09 $\pm$ 0.68	5.55 $\pm$ 0.1	4.32 $\pm$ 0.14
<i>S. longiceps</i>	1.09 $\pm$ 0.68	5.55 $\pm$ 0.1	4.32 $\pm$ 0.14
fishes	0.54 $\pm$ 0.07	4.15 $\pm$ 0.09	1.18 $\pm$ 0.04
<i>T. toli</i>	0.54 $\pm$ 0.07	4.15 $\pm$ 0.09	1.18 $\pm$ 0.04
<i>A. maculatus</i>	14.6 $\pm$ 1.9	134 $\pm$ 6.9	60.1 $\pm$ 4.3
Predatory fishes	21.3 $\pm$ 0.11	89.7 $\pm$ 1.6	94 $\pm$ 0.7
<i>S. leptolepis</i>	21.3 $\pm$ 0.11	89.7 $\pm$ 1.6	94 $\pm$ 0.7
<i>J. amblycephalus</i>	12.5 $\pm$ 0.01	47.2 $\pm$ 1.5	23.1 $\pm$ 0.5

In the biotic component observed trend was plankton < planktivores (*Tenulosa toli*, *Rastrelliger kanagurta*, *Sardinella longiceps*) < filter feeders (*Paphia malabarica*, *Meretrix meretrix*, *Saccostrea cucullata*) < predatores (*Arius maculatus*, *Johnius amblycephalus* and *Selaroides leptolepis*).

Observed tissue wise variation was Liver > Muscle > Gills (Table 3).

## Discussion

### Abiotic Component

A distinct seasonal, tidal and spatial variability of THg was observed in this estuarine system. Higher

concentrations observed in both seasons during low tide. Enhanced surface concentrations in Mandovi estuary were observed during both the seasons. (Fig. 2) (Table. 2). However in the Zuari estuary a uniform distribution of Hg was observed in the water column (Table 2) (Fig. 3). Spatial distribution of THg in the Mandovi and Zuari estuary indicated a nearshore > offshore distribution (Figs. 2 and 3). Enrichment factor EF calculated as Hg Sample/Hg Baseline<sup>12</sup>. A comparison of the values at the river mouths observed in this study with baseline concentrations (ICMAM) of this estuarine system indicated an enrichment (EF) of 15.2 in Mandovi during pre monsoon and 5.8 and 1.0 in Mandovi and Zuari during post monsoon respectively.

Partition studies of dissolved and particulate Mercury carried out at nearshore stations (M1, M4 & M7) and river mouths, exhibited a distinct seasonal variation. In premonsoon dissolved fraction was dominant 72% – 99% except a lower fraction (23%) observed at M7 LT B, in the post monsoon a reverse trend was observed, with the particulate fraction being significantly higher (31-82%) (Figs 4 and 5). The removal of Mercury from the dissolved phase may be attributed to the scavenging of the metal by Fe and Mn particulates which have been observed to be surfaces and possess a strong affinity for many trace metals including mercury is well documented<sup>3</sup> significant in this season<sup>13</sup>. Mercury in seawater occurs in electrically active cationic and complex

forms and is capable of adsorbing onto charged surface. The affinity of Fe and Mn hydroxides present large binding.

The equilibrium distribution of the metal was assessed as below.

$$K_D \text{ (ml/g)} = \frac{\text{[Total Concentration of a metal in SPM}(\mu\text{g/g)]}}{\text{[Concentration of a metal in water}(\mu\text{g/mL)]}}$$

Higher Partition co-efficient  $K_D$  was observed in Post-monsoon (4.46-4.75 mL/g) than in Pre-monsoon (2.70-4.09 mL/g). However no significant correlation of Log  $K_D$  with Log SPM was observed, which is in agreement with the findings in the northeastern and in Texas estuary by<sup>14,15</sup>.

Surface sediment showed marginally higher values in Postmonsoon. (Figs. 6 & 7). A similar seasonal variation in particulates was observed

1. R. Mandovi

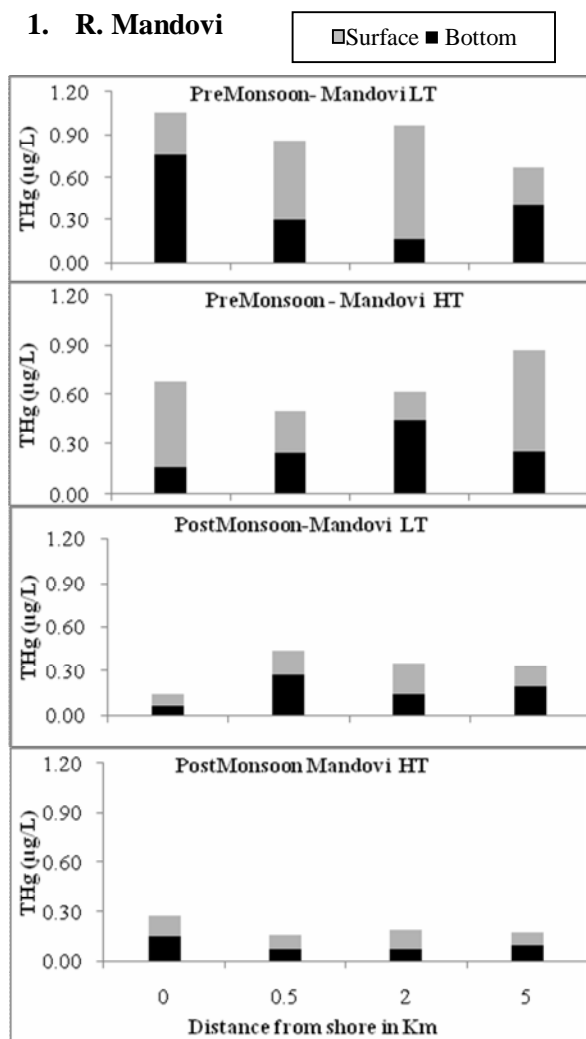


Fig..2—Spatial distribution in water at R. Mandovi

which is due to the strong affinity of the metal for solid phases in the receiving waters i.e., particulate rich in Fe and Mn hydroxides, making sediments the main repository in the aquatic environment<sup>16,17</sup>.

Biotic Component

Biomagnification of Mercury is well documented. A strong correlation between concentration of Total Mercury in aquatic organisms and their trophic levels in pelagic, estuarine and lake food webs has been seen by<sup>18-23</sup>. A significant stepwise biomagnification of Mercury was observed in this study with plankton < planktivores (*T. toli*, *R. kanagurta*, *S. longiceps*) < filter feeders (*P. malabarica*, *M. meretrix*, *S. cucullata*) < carnivores (*A. maculatus*, *J. amblycephalus* and *S. leptolepis*). Plankton comprising the first trophic level is the most sensitive marine organisms to inorganic mercury. The next trophic level is the planktivores which feed on plankton and in our study include: *T. toli*,

2. R. Zuari

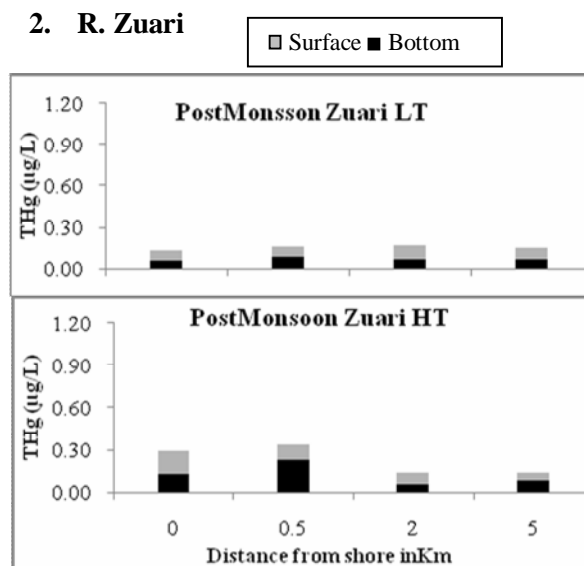


Fig. 3—Spatial distribution in water at R. Zuari

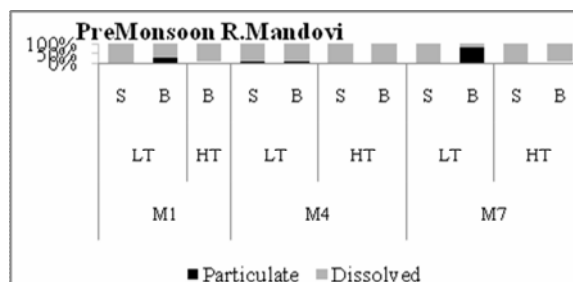


Fig.4—Partitioning between dissolved and particulate at R. Mandovi in Pre- Monsoon

*R. kanagurta*, *S. longiceps*, exhibited 5-10 times higher concentration than plankton. Predatory fish formed subsequent trophic level which included *A. maculatus*, *J. amblycephalus* and *S. leptolepis* which exhibited 10-20 times higher concentrations observed in plankton and planktivores (Fig. 8).

Different body tissues, concentrate different forms and levels of Mercury<sup>24-26</sup>. The Mercury distribution pattern observed in this study: Liver > Muscle > Gills, except for *R. Kanagurta* where highest concentration observed in gills and *S. Leptolepis* in Muscle tissue. The variation in the accumulation of metals in different organs of fish may be attributed to proximity of tissues to the availability of metals, i.e. the quantity present in the surrounding environment, age and type of the fish and role of the tissue in the detoxification process<sup>27</sup>. The high concentration in the digestive organ liver

concludes that major route of exposure of Mercury is food intake, as liver is the de-toxifying organ involved in food metabolism. Liver tissue has been observed to be the preferential site for Mercury accumulation<sup>3</sup>.

The interaction between abiotic and biotic components in Mercury distribution can be assessed from the Bioconcentration factor i.e. concentration of a particular metal in a biological tissue relative to the surrounding medium (Seawater- BCF<sub>mw</sub> or Sediment BCF<sub>ms</sub>). The ability of organisms to accumulate Mercury from the surrounding environment is calculated according to the ambient environment. Bio-concentration factor ranged from 6.37 in plankton to 1.43 – 26.10 in planktivores and 31.50 - 355.99 in predatory fish (Table 3). BCF (medium sediment) 0.04 - 0.22 was very low compared to BCF (medium water) (1.43-355.99). BCF criterion is an index for hazard classification for long – term environmental impacts. Above a certain threshold, a substance is deemed to be bio accumulative.

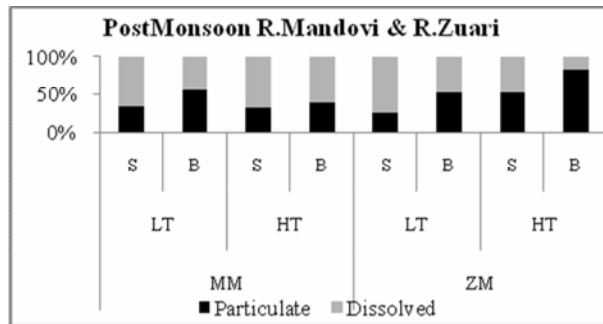


Fig.5. Partitioning between dissolved and particulate at R. Mandovi and R. Zuari in Post- Monsoon

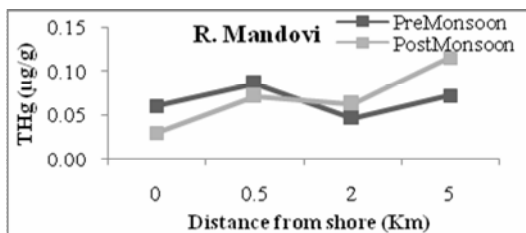


Fig..6. Spatial distribution of THg in Sediments at R Mandovi

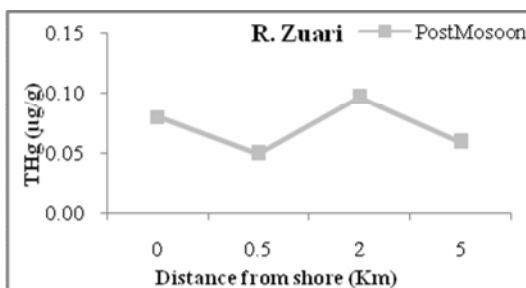


Fig..7. Spatial distribution of THg in Sediments at R. Zuari

Table 4—Observed values of Bio concentration Factor (BCF)

Organism	Bio-concentration Factor			
	BCF <sub>mw</sub>	BCF <sub>ms</sub>		
Planktons	6.37			
Filter feeding bivalves	<i>P.malabarica</i>	13.04	0.07	
	<i>M. meretrix</i>	39.44	0.22	
	<i>S. cucullata</i>	8.26	0.05	
Planktivores fishes		Gill	Liver	Muscle
	<i>R. kanagurta</i>	26.10	8.14	2.41
	<i>S. longiceps</i>	2.89	14.74	11.48
	<i>T. toli</i>	1.43	11.02	3.12
Predatory fishes	<i>A. maculatus</i>	32.13	355.99	159.50
	<i>S. leptolepis</i>	56.56	238.26	244.75
	<i>J. amblycephalus</i>	31.50	129.72	70.92

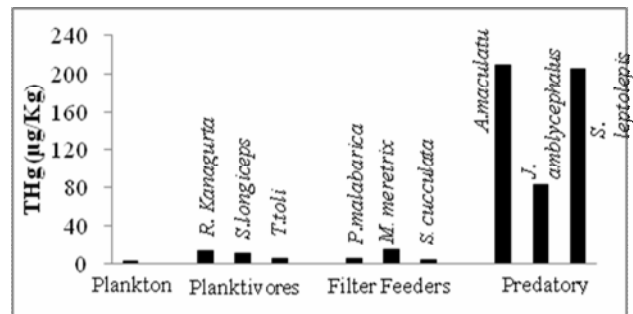


Fig.8. Biomagnification of Hg in marine Food web

The threshold BCF values used to classify substances range between 500 and 5,000, depending on the jurisdiction<sup>28</sup>. The higher BCF factor implies that Mercury is eliminated very slowly relative to uptake. The highest value observed being in this study, 355, is well below the acceptable threshold limit.

### Conclusions

The foregoing observations indicate that the behaviour of the metal in this estuarine system is impacted by seasonal variability. Higher THg concentrations are observed in the premonsoon with an enhancement of the metal in the particulate fraction in the post monsoon. The biotic compartment highlights the biomagnification and bioaccumulation of the metal. A marked correlation of the metal and their trophic levels was observed. It was observed that the liver tissue was the preferential site for Mercury bioaccumulation.

Although an enrichment of Mercury in water and a stepwise biomagnification of the metal observed in this food web, the Mercury level in representative biota is well below the acceptable safety limit 0.5 ppm for fish products set by WHO<sup>29</sup> and 0.3 ppm set by UNEP for human consumption, thereby indicating no apparent mercury contamination.

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