

# Assessment of Total Mercury ( $Hg_T$ ) in Sediments and Biota of Indian Sundarban Wetland and Adjacent Coastal Regions

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## Abstract

The distribution of total mercury ( $Hg_T$ ) in surface sediments (0-5 cm; n = 12; particle size < 63  $\mu\text{m}$ ) and representative biota (benthic polychaetes, bivalve mollusks and finfish) were observed in the Sundarban mangrove wetland and adjacent regions nearby the Indian Ganges river estuary. Relatively low concentrations of  $Hg_T$  were measured in sediments ranging from 0.008  $\mu\text{g g}^{-1}$  to 0.056  $\mu\text{g g}^{-1}$ . There exist sharp differences in  $Hg_T$  accumulation in biota which revealed the following decreasing trend: polychaetes > fish > bivalve mollusks. These variations are related to a number of intrinsic (size, age and sex) and extrinsic (pH and salinity) factors together with the accumulation mechanisms intrinsic to each species for mercury. An organ-specific  $Hg_T$  accumulation in bivalve mollusks was evidenced with the following decreasing order: visceral mass > siphon > adductor muscle > mantle > gill, with a maximum value of 0.42  $\mu\text{g g}^{-1}$  in *Sanguinolaria acuminata*. Fishes showed wide efficiency in  $Hg_T$  accumulation in dorsal muscle, and of most concern, one species presented  $Hg_T$  above 0.05  $\mu\text{g g}^{-1}$  levels, the prescribed limit established by European Union. The benthic polychaetes showed extreme variations of  $Hg_T$  in their body tissues, with the maximum value of 0.603  $\mu\text{g g}^{-1}$  in *Dendronereis heteropoda* which is above the European Union threshold value. The authors strongly recommend further monitoring to investigate the source of toxic metals, including Hg which may originate from diverse potential sources such as industrial discharges, agricultural runoff and sewage sludge from upstream of the Ganges River Estuary.

**Keywords:** total mercury ( $Hg_T$ ), sediment, biota, bioaccumulation, Sundarban wetland

## 1. Introduction

Environmental mercury contamination is a global issue. In the past two decades, the concentration of Hg has been determined across the globe in numerous environmental matrices, including air, water, sediment, and biota (Driscoll et al., 2013; Taylor et al., 2012). Elemental mercury and mercury ions (mostly chloride, sulfide and organic acids bound and organic methyl mercury) are mainly formed in sediments and water (Ravichandran, 2004; Greenfield & Jahn, 2010). Recent works on mercury bioavailability (Hammerschmidt & Fitzgerald, 2004) and isotope studies confirmed the role of sediment as a potential source of mercury to living biota (Gehrke et al., 2011, Spada et al., 2012; Taylor et al., 2012). Tropical wetlands are typical in terms of environmental inputs favoring the biogeochemical processes that convert elemental mercury into organometallic mercury i.e., methylation and demethylation (Chatterjee et al., 2012; Liu & Ding, 2007). Primary consumers feeding directly from organic material in sediments receive mercury in lower levels (Choy et al., 2010). The organisms feeding on these primary consumers accumulate mercury in higher concentrations (DesGranges et al., 1998; Edmond et al., 2010; Wolfe et al., 1998). In this way the mercury concentrates through trophic transfer from lower to higher trophic levels in the aquatic food web. The highest body burden of mercury concentration could therefore be expected in the top predator fishes and human beings (NRC, 2000) accumulation factors of almost 5000 that of sedimentary mercury have been recorded (Sharma, 1993).

The Indian Sundarban (latitude 21°32' and 22°40' N; longitude 88°05' and 89°00' E) is the largest delta (area 9,630  $\text{km}^2$ ) in the estuarine part of the River Ganges. It is situated in the coastal region of Bay of Bengal is one

of the most dynamic, complex, bioclimatic zones; vulnerable to natural hazards such as tropical cyclones, floods, and man-made erosion, industrial and domestic effluents. The delta region is experiencing discharges from several upstream point sources such as coal fired power plants, cement industries, brick manufacturing units, chemical, paint and fertilizer manufacturing units and thermometer manufacturing units, small scale tanneries, (Figure 1) located along stretches of the Hugli River. Additionally, effluents from aquaculture ponds directly to the river could also act as individual point sources or diffuse sources as a group, of contaminants (Manna et al., 2010). In a recent study by Corsolini et al. (2012) the point sources in Indian Sundarban were identified as wastewater effluents whilst the diffuse source identified as surface runoff. These along with immense domestic and municipal discharges contribute input to the Sundarban of a number of pollutants including heavy metals and Persistent Organic Pollutants (POPs) (Binelli et al., 2007; Chatterjee et al., 2007, 2009, 2012; Guzzella et al., 2005; Sarkar et al., 2002, 2004, 2007; Watts et al., 2013). Recent research on surface and core sediments in Indian Sundarban has exhibited contaminant values above the prescribed guidelines indicating likely adverse impacts on adjacent biota (Kwokal et al., 2008, 2012; Chatterjee et al., 2009, 2012).

The objective of this study was firstly, to examine the pattern of enrichment of total mercury ( $Hg_T$ ) in the sediments collected from 14 sampling locations in the Indian Sundarban region. Secondly to measure the  $Hg_T$  in the tissues of five polychaete species (*Mastobranchus indicus*, *Glycera rouxii*, *Namalcastes fauveli*, *Dendronereis heteropoda* and *Pereneries cultifera*) and two bivalve mollusks (*Sanguinolaria acuminata*, *Macoma birmanica*) collected from the same study locations (see Table 1). And finally to measure the  $Hg_T$  in the tissues of 19 edible fish species (Table 1) collected from Gangasagar beach in the Indian Sundarban region, to obtain contamination status due to mercury sources and to assess probable health risks associated with the consumption of fish from this region

## 2. Method

### 2.1 Study Site, Sample Collection, and Preservation

Collection of sediments and biota was performed from twelve sampling sites using a PVC spatula acid cleaned between samples and sites. The sampling sites were, Barrackpur ( $S_1$ ), Nimtala ( $S_2$ ), Dakshineswar ( $S_3$ ), Babughat ( $S_4$ ), Nurpur ( $S_5$ ), Lot 8 ( $S_6$ ), Phuldubi ( $S_7$ ), Gangadharpur ( $S_8$ ), Gangasagar mudflat 1 ( $S_9$ ), Gangasagar mudflat 2 ( $S_{10}$ ), Gangasagar beach ( $S_{11}$ ) Mayagoyalinir Ghat ( $S_{12}$ ). Two more sites namely Chemagari ( $S_{13}$ ) and Gushighata ( $S_{14}$ ) were selected for collection of bivalve and polychaete samples. The sampling stations have been chosen considering the sediment dispersal patterns along the drainage network systems (Figure 1). Selection of sampling sites was based on difference in their tidal amplitude, distance from wave actions, and pattern of domestic and industrial impacts. In order to have biota samples representing different feeding guilds (see Table 1) we selected sediment dwellers (polychaetes and bivalves) with their corresponding sediment samples and fishes which are associated with different food preferences e.g. planktivorous/herbivorous, omnivorous or carnivorous and primarily carnivorous. Biota samples (polychaetes and bivalves) were collected from Gangasagar mudflat 2 ( $S_{10}$ ) and Gangasagar beach ( $S_{11}$ ) from the tidal mudflats during low-tide by hand picking methods wearing gloves, and where appropriate using a quadrat. The fish samples were collected from Gangasagar beach ( $S_{11}$ ) directly from fish trawlers catch landing. In our study, bivalve, polychaete and fish were considered as environmental indicators based on standard criteria (Wiener et al., 2003; Evers et al., 2008) which employ the selection of species that are abundant, widely distributed, easily distinguishable as well as easy to analyze. Moreover, these organisms were selected as they are tolerant of a wide range of salinity and pH, having a standard life span, dietary patterns and more importantly position in the food web.

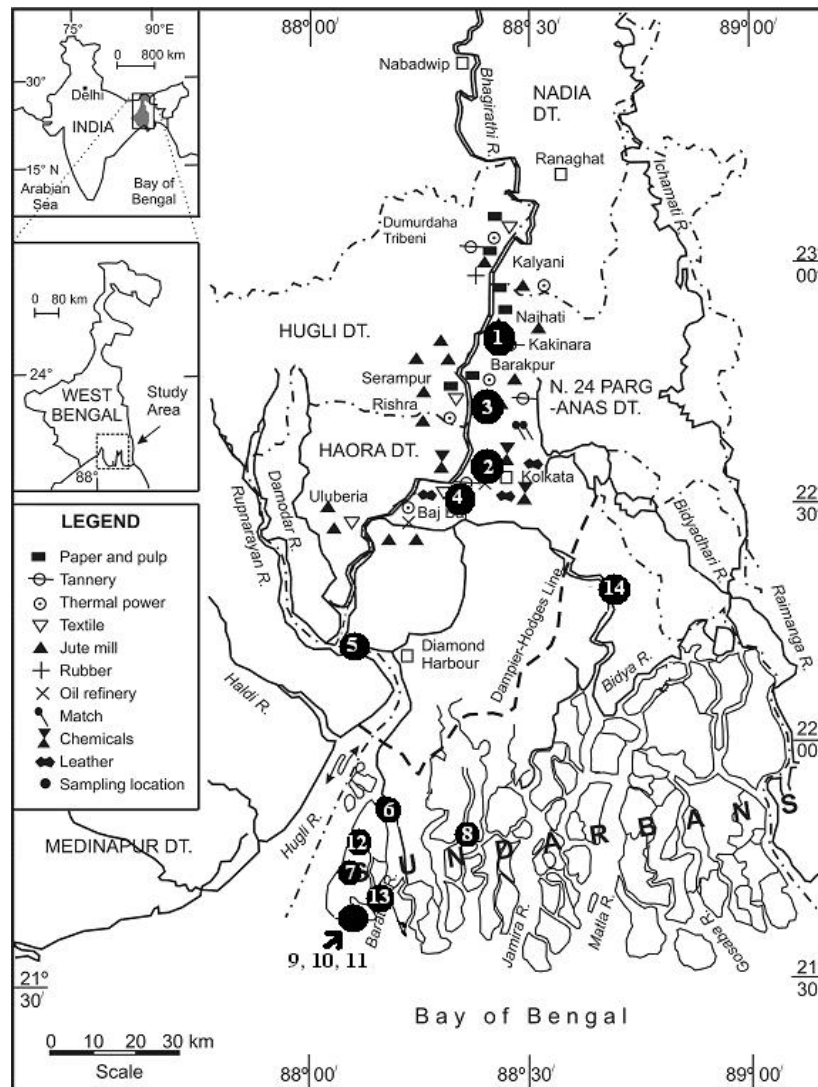


Figure 1. Map of the Sundarban wetland including lower stretch of Hugli River illustrating the locations of sampling sites

Sediments, bivalves and fish samples collected in the field were placed in sterile polyethylene bags, put on ice, transported to the laboratory where stored at  $-20^{\circ}\text{C}$ . Polychaetes after collection were taken to the laboratory and placed in aerated seawater collected from the same sampling sites for 48 hours for depuration. In the laboratory, biota samples were measured for biometry (e.g. length and weight) and processed for drying. All the biota samples were cut open and processed as a whole body. Bivalves were analyzed organ-wise (shell, visceral mass and siphon) in order to obtain an indication of bioavailable fractions of  $\text{Hg}_T$  (Zoluoga et al., 2009). For final preservation, all sediment and biotic samples were oven dried at  $60^{\circ}\text{C}$  for 48 h, homogenized with a mortar and pestle (biota only), sieved through  $63\ \mu\text{m}$  sieve (sediment samples only) and stored at room temperature in acid washed clear borosilicate vials. The drying temperature was maintained below  $80^{\circ}\text{C}$  in order to prevent volatilization of mercury.

Table 1. Common and scientific name of three different benthic biota: polychaetes (n = 5), bivalve mollusks (n = 2) and finfish (n = 19) collected from the Sundarban wetland, India. Individual habitat type and feeding behavior are indicated

Species	Common Name	Habitat Type	Feeding Habits
<b>Polychaetes</b>			
<i>Mastobranchus indicus</i>	Marine polychaetes	non-selective deposit feeders	Non-selective deposit feeders
<i>Glycera rouxii</i>		Demersal environment	Carnivorous
<i>Namalcastes fauveli</i>		Deposit feeder	Deposit feeder
<i>Dendronereis heteropoda</i>		benthic	Sub-surface deposit feeders
<i>Perinereis cultifera</i>		benthic	Herbivores (benthic microalgae)
<i>Mastobranchus indicus</i>		non-selective deposit feeders	Non-selective deposit feeders
<i>Dendronereis heteropoda</i>		Surface deposit feeding	Carnivorous
<i>Perinereis cultifera</i>		Surface deposit feeding	Deposit feeder
<i>Perinereis cultifera</i>		Surface deposit feeding	Sub-surface deposit feeders
<b>Bivalve mollusks</b>			
<i>S. acuminata</i>	Mollusk	benthic	Sedimentary filter feeder
<i>M. birmanica</i>	Mollusk	benthic	Sedimentary filter feeder
<b>Fish</b>			
<i>Chana punctatus</i>	Spotted snakehead/lata	Benthic-pelagic	Carnivorous
<i>Euthynnus affinis</i>	Mackerel	pelagic	Carnivorous/omnivorous
<i>Ompok pabo</i>	Pabo catfish	Muddy water/near bottom	Carnivorous/omnivorous
<i>Trichiurus sp</i>	Pompano	pelagic	Primarily carnivorous
<i>Coilia neglecta</i>	Anchovy/Amodi/Ruli	pelagic	Primarily carnivorous
<i>Nibea soldato</i>	Croaker/Bhola Bhetki	pelagic	Primarily carnivorous
<i>Trachinotus sp</i>	Pompano	pelagic	Primarily carnivorous
<i>Lates calcerifer</i>	Bhetki	pelagic	Primarily carnivorous
<i>Setipinna sp</i>	Phasa/hairpin anchovy	pelagic	Primarily carnivorous
<i>Pampus argenteus</i>	Pomfret	pelagic	Primarily carnivorous
<i>Pama pama</i>	croaker	pelagic	Primarily carnivorous
<i>Therapon jerbua</i>	Tiger bass	demersal	Primarily carnivorous
<i>Ailia coila</i>	Trout/Gangetic ailia	demersal	Primarily carnivorous
<i>Notopterus notopterus</i>	Bronze featherback	pelagic	Primarily carnivorous/omnivorous
<i>Harpodon nehereus</i>	Bombay duck	pelagic	Omnivorous
<i>Labeo bata</i>	bata	Benthic-pelagic	Omnivorous
<i>Oreochromis mossambicus</i>	Mozambique tilapia	demersal	Omnivorous
<i>Liza parsia</i>	Persia/mullet	demersal	Herbivorous/planktivorous
<i>Tilapia zilli</i>	Indian tilapia/grouper	demersal	Herbivorous/planktivorous

## 2.2 Analysis of Sediment pH, Organic Carbon and Textural Parameters

The sedimentary organic carbon (Corg) was analyzed using a rapid titration method (Walkey & Black, 1934) and the pH of the sediment samples were analyzed using Deluxe™ pH meter (model no. 101E). Textural parameters such as calculation of proportion of sand, silt and clay of sediment samples were done by sieving in a Ro-Tap shaker (Krumbein & Pettijohn, 1938). The textural parameters were calculated using statistical computation

using the formulae of Folk and Ward (1957).

### 2.3 Mercury Analysis of Sediments and Biota

The analyses were carried out using a Direct Mercury Analyzer, model DMA-80 from Milestone Srl, Italy. Samples were accurately weighed into pre-blanked 14 metal boats. The DMA-80 automatically transfers these to a furnace heated to 650 °C, to decompose and volatilize the Hg. After Hg pre-concentration on a gold amalgam and release, the AAS absorbance-time profile is recorded on a high and low concentration range detector. Sample concentrations were produced by comparison of their signal intensity against a calibration set of aqueous chemical standards (Romil Chemicals, UK) 0.01, 0.1 and 1  $\mu\text{g g}^{-1}$  Hg. Based on a sample weight of 0.1 g, the instrumental limit of detection was 0.0015  $\mu\text{g g}^{-1}$  Hg; 3SD of the blank ( $n = 10$ ). Data were validated against certified reference material ERM-BB422 (fish and biota) and NIST MESS-3 (sediments), which provided a recovery of  $99 \pm 4.5\%$  ( $n = 20$ ) and  $104 \pm 1.4\%$  ( $n = 5$ ), respectively (Table 2).

Table 2. Summary of Certified Reference Material data indicating their mean ppb, percentage relative standard deviation (%RSD) and percentage recovery.

Name	Number of analysis	Mean value (ppb)	%RSD	Recovery (%)
ERM-BB422	20	593	4.5	99
NRCC MESS-3	5	95	1.3	104

## 3. Results and Discussion

### 3.1 Sediment Quality Parameters

Table 3 summarises the sediment chemical parameters with pH ranging from slightly acidic (6.7 at Dakshineswar,  $S_3$ ) to basic (8.1 at Lot 8,  $S_6$ ). According to Liao (1990), the low pH in sediments could be partly resulting from oxidation of  $\text{FeS}_2$  and  $\text{FeS}$  to sulfate ( $\text{SO}_4^{2-}$ ) and partly from the mangrove litter decomposition and hydrolysis of tannin in mangrove plants releasing various organic acids. Lower pH values have also been reported in sediment samples from tropical wetland sites (Torres-Alvarado et al., 2013; Peck et al., 2002). The sediment samples exhibited low values of organic carbon (0.34 to 1.08%). Low organic carbon values in sediments from the intertidal zone of the Sundarban wetlands were also recorded in previous studies (Sarkar et al., 2004; Chatterjee et al., 2007). Previous works on sediments has shown that the predominance of negatively charged quartz grains in the sediments could be one of the reasons for less organics in this region. (Bhattacharya & Sarkar, 2003; Sarkar et al., 2004).

### 3.2 Sediment $\text{Hg}_T$ Concentration

Table 3 summarises  $\text{Hg}_T$  concentrations in surface sediments for 14 sampling sites; values ranged from 0.008  $\mu\text{g g}^{-1}$  (at Dakshineswar,  $S_3$ ) to 0.056  $\mu\text{g g}^{-1}$  (at Gangadharpur,  $S_8$ ) with mean and median values of 0.036 and 0.035  $\mu\text{g g}^{-1}$  respectively. The variation of  $\text{Hg}_T$  in the sites may be related with their locations and the fluvial characteristics of the river Hugli. The site Dakshineswar ( $S_3$ ) is an upstream site located 14 km from the centre of Calcutta having microtidal effects and organic effluents from the holy Kali temple. Gangadharpur ( $S_8$ ) by comparison is located about 100 km away from Calcutta, further downstream and having a meso-macrotidal environment. This site is part of the Indian Sundarban wetland and having the high organic carbon content and smaller grain size sediment characteristic. However weak positive correlation ( $r = 0.27$ ) was obtained for  $\text{Hg}_T$  and sedimentary organic carbon values. This could be due to the fact that sediment bound mercury is stimulating the microbial activities of the sediment and in situ geochemical conditions to act as a substrate for mineralization (Taylor et al., 2012) or result of some influences of episodic anoxia-hypoxia at sediment water interface (Sunderland et al., 2006). Also a positive correlation was obtained for sedimentary  $\text{Hg}_T$  with the smaller grain size (silt and clay) indicating binding mercury in the form of methylmercury with clay and mud fractions facilitating ingestion by aquatic organisms (Barandiaran, 2013; Zhang & Wang, 2008). The concentrations of  $\text{Hg}_T$  in sediments are nearly an order of magnitude lower than the previous study of  $\text{Hg}_T$  of Sundarban mangrove wetland which measured  $\text{Hg}_T$  and methylmercury in sediment cores, India (0.32-0.196  $\mu\text{g g}^{-1}$ , Chatterjee et al., 2012). Also the present result of  $\text{Hg}_T$  in sediments is much lower than that of Vembanad wetland system, west coast of India (0.016-4.22  $\mu\text{g g}^{-1}$ , Ramasamy et al., 2012). None of the study sites exceeded the prescribed ERL value (0.15  $\mu\text{g g}^{-1}$ ) of mercury proposed by Long et al. (1995, 1998) for adverse biological effects. Based on the results it could be stated that the sediments are uncontaminated with respect to mercury.

It has been suggested that mercury pollution in the Indian Sundarban region is largely caused by the burning of fossil fuel, wind-blown dust, erosion, aquacultural and agricultural wastes and untreated industrial discharges (Sarkar et al., 2006).

Table 3. Concentration of  $Hg_T$  (expressed in  $\mu g g^{-1}$  dry weight) measured in the surface sediments collected from 14 sampling sites in the Sundarban wetland area, India

Sampling Location	Sample sites	pH	Organic Carbon (%)	Texture property			Hg ( $\mu g g^{-1}$ )
				Sand (%)	Silt (%)	Clay (%)	
S1	Barrackpore	6.9	0.42	7.88	23.14	68.98	0.035
S2	Nimtala	6.9	0.56	9.43	17.47	73.1	0.024
S3	Dakshineswar	6.7	0.66	3.12	42.65	54.23	0.008
S4	Babughat	6.9	0.34	0.96	39.01	60.23	0.034
S5	Nurpur	6.94	0.96	8.76	35.25	55.99	0.041
S6	Lot 8	8.13	1.08	13.76	46.9	39.34	0.035
S7	Phuldubi	7.62	0.6	14.09	57.5	28.41	0.054
S8	Gangadharpur	7.22	0.87	0.4	39.6	60	0.056
S9	Gangasagar mudflat 1	7.91	0.59	26.14	15.1	58.76	0.032
S10	Gangasagar mudflat 2	8.04	0.56	19.6	20.2	60.2	0.025
S11	Gangasagar beach	7.66	0.39	98.1	0.71	1.2	0.053
S12	Mayagoalinir Ghat	7.11	0.48	3.5	41.1	55.7	0.031
S13	Chemaguri	7.41	0.43	61.66	37.14	1.2	0.041
S14	Gushighata	6.9	0.91	10.76	73.46	15.78	0.070

### 3.3 Concentration of $Hg_T$ in Biota

Benthic polychaetes exhibited a variable degree of accumulation strategies where minimum ( $0.083 \mu g g^{-1}$ ) and maximum ( $0.603 \mu g g^{-1}$ )  $Hg_T$  values were recorded in *M. indicus* from Chemaguri ( $S_{13}$ ) and *D. heteropoda* from Gushighata ( $S_{14}$ ) respectively (Table 4a). Considering uptake of  $Hg_T$  from dissolved phase of water and sediment ingestion are significant in this coastal environment it could be assumed that most of the  $Hg_T$  ( $> 70\%$ ) measured in polychaetes is derived by sediment ingestion (Wang et al., 1998). Representatives of family Nereidae like *D. heteropoda* prefer to inhabit mud burrows which have the unique biogeochemical properties e.g., oxygen and mucous promoting microbial growth to enrich  $Hg_T$  (Sizmur et al., 2013). This could explain why *D. heteropoda* have much higher  $Hg_T$  values compared to other polychaete species. The high  $Hg_T$  value reported for *D. heteropoda* is potentially a matter of concern because these species are used as a food source for shrimp ponds (Khan & Murugesan, 2005) indicating transfer and biomagnification of mercury in the form of methylmercury and probable adverse human health impacts from shrimp consumption. The differences in  $Hg_T$  patterns among the studied species of polychaetes may be attributed to minute differences in their feeding behaviors due to spatial distribution patterns and local inputs. We must also consider several other factors such as specimen size, sexual maturity, habit and habitats, trophic position, environmental parameters and pollution may also have impacts on the  $Hg_T$  contamination (Kehrig et al., 1998).

The bivalves exhibited lower  $Hg_T$  concentration values of  $Hg_T$  when compared to that of the polychaetes (Table 4a-4b), with individual tissue burden of  $Hg_T$  ranging from  $0.024 \mu g g^{-1}$  in adductor muscle of *S. acuminata* to  $0.24 \mu g g^{-1}$  in the visceral mass of *M. birmanica*. This variation in the concentrations of  $Hg_T$  reveals that in some way the organisms adopt specific preference for the accumulation of metals within their tissues. Bivalve mollusks are filter feeders and use their gills to capture food particles (e.g. plankton and organic matter) from the overlying water column. Therefore unlike polychaetes, bivalves receive  $Hg_T$  fractions from water rather than from direct sediment ingestion. Therefore, for bivalves there are two possible pathways in which both sediment and water could act as a source of  $Hg_T$ . The highest value of  $Hg_T$  in *S. acuminata* was observed in the siphon ( $0.11 \mu g g^{-1}$ ), with reduced  $Hg_T$  when measured in the visceral mass ( $0.04 \mu g g^{-1}$ ). This reduction may be the result of bivalve's unique detoxification mechanism by metallothionein in the visceral mass (Zhao et al., 2010;

Vasak, 2005), indicating the activity of the digestive glands to detoxify with response to higher  $Hg_T$  values. However, the mechanism used to detoxify the tissues in *S. acuminata* may not be as efficient in *M. birmanica* and may indicate this bivalve is not as adept at removing or indeed detoxifying its tissues with regards to higher concentrations of  $Hg_T$ . This may be of concern if this species of bivalve is then utilized as source of food for human consumption either directly or indirectly through use as a food source e.g. poultry feed (*pers. obs.*) which in turn is then consumed by humans. It is noteworthy that the organ in direct contact with the water i.e. the siphon is the organ containing the highest concentrations of  $Hg_T$  in the bivalve *S. acuminata* for this study.

Because mercury (in the form of methyl mercury) has been shown to bioaccumulate along the food chain (see Chen et al., 2008), it was expected that fishes which belong to a higher trophic position than that of polychaetes and bivalves would therefore contain higher concentration of  $Hg_T$  (see Table 4c). However, in this study  $Hg_T$  concentrations for finfish were found to be within a similar range observed among estuarine polychaetes, with  $Hg_T$  ranging from  $0.023 \mu\text{g g}^{-1}$  in muscle tissue of *Liza parsia* to a maximum of  $0.60 \mu\text{g g}^{-1}$  in the catfish *Ompok pabo* (Table 4c). Although  $Hg_T$  has been shown to vary according to the trophic position in fishes, it should be noted that trophic position is not always a key factor in determining  $Hg_T$  enrichment (see Sizmur et al., 2013). It could be suggested that deposit feeding bottom dwelling polychaete worms like *D. heteropoda* are less likely to be taken as prey items by pelagic fishes and are more likely to be ingested by demersal or benthopelagic species, where these fish species are more likely to come into contact with *D. heteropoda*. We may therefore assume that the largest amount of  $Hg_T$  accumulated in the muscle tissue would be observed for those fish species living a more demersal life style compared to those fish species found higher in the water column. However, this was not strictly the case for this study, where fish species found to have  $Hg_T$  greater than  $0.20 \mu\text{g g}^{-1}$  were found to be both pelagic and demersal (see Table 5) and may indicate other forms of  $Hg_T$  enrichment within the food chain.

According to Jackson (1990), mercury concentration in fish generally increases with age or size with respect to their exposure time to mercury. Jaworski and Ragnarsson (2006) observed the proportion of polychaetes and molluscs in the diet of predator demersal fish species comprised average  $> 5\%$  by weight of the total diet. This was quite in agreement with that of the finding of Ross (1982) for Gray tilefish from North and South Carolina waters where polychaetes and bivalves contribute little to the fish diet. However, Mojumder and Dan (1979) noted that the food of catfish consists of 26% polychaetes and sometimes these are preferred as a single food item. In our study highest concentration of  $Hg_T$  was found in a catfish *O. pabo* which could be due to predation on polychaetes. Additionally, fishes can accumulate mercury based on a number of other factors, including the availability of inorganic mercury in the sediment-water column, interaction between the trophic structures, rate of mercury methylation by microbes, accumulation strategies by different species and seasonal variations. The maximum  $Hg_T$  concentration was measured in the fish species *Ompok pabo* ( $Hg_T$ :  $0.6 \mu\text{g g}^{-1}$ ) (from Gangasagar beach,  $S_{11}$ ) which feeds on small fish, aquatic insects, insect larvae and earthworms followed by *Ailia coila* ( $Hg_T$ :  $0.37 \mu\text{g g}^{-1}$ ) which feeds on small gastropods and followed by *Trichiurus* sp (Ribbon fish) ( $Hg_T$ :  $0.27 \mu\text{g g}^{-1}$ ) the adults of which are carnivorous, whilst their juveniles and sub-adults are mainly herbivorous (Martins et al., 2005; Chiou et al., 2006; Bittar et al., 2008). The concentration of  $Hg_T$  relative to trophic level feeding behavior in fish is illustrated in Figure 3, with the higher trophic levels observed primarily for the more carnivorous fish species. However, two of the omnivorous species in Figure 3 (*Ompok pabo* and *Ailia coila*) have particularly high  $Hg_T$  perhaps suggesting a more carnivorous nature. The highest concentration in *O. pabo* (from Gangasagar beach  $S_{11}$ ) is likely due to the bioaccumulation of  $Hg_T$  from both high and low trophic levels through its omnivorous food chain (Beneditto et al., 2013) and were notably identical to the  $Hg_T$  observed within the whole body mass of the polychaete *D. heteropoda* collected from the same location of Gangasagar ( $S_9$ ) which is situated at the far south of Lower Long Sand Island at the mouth of the River Hugli. Although only one fish was measured for  $Hg_T$  from the 19 fish species represented for this study (with the exception of *Nibea soldato* where there were two fish measured), the high total mercury concentrations observed in the tissues of these fish species would indicate the urgency for further monitoring and scientific studies to be conducted around this fragile wetland ecosystem.

The measurement of methyl-Hg was beyond the scope of the current study. However, several authors have suggested that in a majority of cases the percentage methylmercury in fish is in the range 55-100% (Kruzikova et al., 2008; Agah et al., 2007; Storelli et al., 2003). We can therefore assume in a worst case risk assessment,  $Hg_T$  is still an appropriate measure.

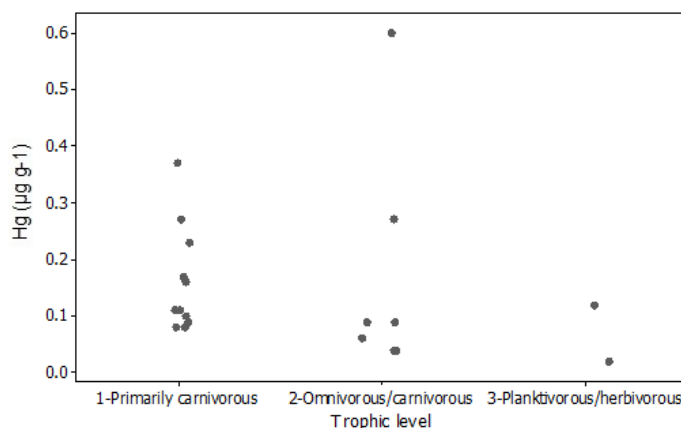


Figure 3. Range plot of  $Hg_T$  concentration in fish showing increase in higher trophic level

$Hg_T$  concentrations of biota collected from Sundarban wetland were comparable or lower than similar organisms in different estuarine systems of the United States. For example, studies on *Acartia* copepods ( $\sim 0.03$  to  $0.05 \mu\text{g g}^{-1}$ ), *Littorina littorea* ( $0.18 \pm 0.03 \mu\text{g g}^{-1}$ ), *Carcinus maenas* ( $0.11 \pm 0.02 \mu\text{g g}^{-1}$ ), and *Fundulus heteroclitus* ( $0.15 \pm 0.03 \mu\text{g g}^{-1}$ ) in Long Island Sound and Gulf of Maine, reported similar contamination patterns of  $Hg_T$  (Watras & Bloom, 1992; Hammerschmidt & Fitzgerald, 2006; Chen et al., 2009). Home et al. (1999) obtained very high levels of  $Hg_T$  in *L. littorea* collected from Georgia salt marshes, ranging between  $0.6$  and  $33.1 \mu\text{g g}^{-1}$ . Polychaetes (*Nereis occidentalis* and *N. succinea*), bivalves (*Ensis minor*), and decapods (*Callinectes sapidus*) from Lacava bay, United States also exhibited about 34 to 90 percent higher  $Hg_T$  values compared to the present study. The internal geochemical environment favoring mercury methylation as well as metabolism pattern along with the differential patterns of mercury input in the external environment is the key factor responsible for the observed variations in  $Hg_T$  in biota (Benoit et al., 2003). Also, variations in estuarine processes affecting ecological and physico-chemical parameters from site to site may affect the intake of methylmercury as food/prey in different trophic positions (Chen et al., 2008).

Table 4a. Total mercury  $Hg_T$  (expressed in  $\mu\text{g g}^{-1}$  dry weight) in ascending order of concentration observed in whole body mass of five polychaete species collected from five sites in Sundarban wetland, India

Species	Site	Tissue type	$Hg_T$ ( $\mu\text{g g}^{-1}$ )
<b>Polychaetes</b>			
<i>Mastobranthus indicus</i>	Chemagari ( $S_{13}$ )	Whole body	0.08
<i>Glycera rouxii</i>	Gangasagar ( $S_9$ )	“	0.09
<i>Mastobranthus indicus</i>	Gangasagar ( $S_9$ )	“	0.12
<i>Namalcastes fauveli</i>	Gangasagar ( $S_9$ )	“	0.12
<i>Perinereis cultifera</i>	Lower Long Sand ( $S_{15}$ )	“	0.12
<i>Perinereis cultifera</i>	Gangasagar ( $S_9$ )	“	0.18
<i>Perinereis cultifera</i>	Maya Goyalinir Ghat ( $S_{12}$ )	“	0.20
<i>Dendronereis heteropoda</i>	Gushighata ( $S_{14}$ )	“	0.37
<i>Dendronereis heteropoda</i>	Gangasagar ( $S_9$ )	“	0.60



Table 4b. Total mercury  $Hg_T$  (expressed in  $\mu g g^{-1}$  dry weight) in ascending order of concentration observed in three different organs of two bivalve mollusks collected from two sites in Sundarban wetland, India

Species	Site	Tissue type	$Hg_T$ ( $\mu g g^{-1}$ )
<b><i>Bivalve mollusks</i></b>			
<i>S. acuminata</i>	Gangasagar ( $S_{10}$ )	Adductor muscle	0.02
		Visceral mass	0.04
		Siphon	0.11
<i>M. birmanica</i>	Chemagari ( $S_{13}$ )	Visceral mass	0.03
		Adductor muscle	0.24
		Siphon	0.42

Table 4c. Total mercury  $Hg_T$  (expressed in  $\mu g g^{-1}$  dry weight) measured in the muscle tissue of twenty fish species collected in Gangasagar from the Sundarban wetland, India in ascending order of concentration observed

Species	$Hg_T$ ( $\mu g g^{-1}$ )
<b><i>Fish</i></b>	
<i>Liza parsia</i>	0.02
<i>Euthynnus affinis</i>	0.04
<i>Pampus argenteus</i>	0.04
<i>Harpodon nehereus</i>	0.06
<i>Nibea soldado</i>	0.08
<i>Chana punctatus</i>	0.08
<i>Nibea soldato</i>	0.09
<i>Notopterus notopterus</i>	0.09
<i>Labeo bata</i>	0.09
<i>Therapon jerbua</i>	0.10
<i>Trachinitus sp.</i>	0.11
<i>Lates calcerifer</i>	0.11
<i>Tilapia zilli</i>	0.12
<i>Setipinna sp.</i>	0.16
<i>Pama pama</i>	0.17
<i>Coilia neglecta</i>	0.23
<i>Trichiurus sp.</i>	0.27
<i>Oreochromis mossambicus</i>	0.27
<i>Ailia coila</i>	0.37
<i>Ompok pabo</i>	0.60

Table 5. Fish species showing trophic level and which indicate concentrations of  $Hg_T$  greater than  $0.20 \mu g g^{-1}$  captured from Gangasagar ( $S_9$ ) in the Sundarban wetland, India

Species	Common Name	Habitat	Feeding Habits	$Hg_T$ ( $\mu g g^{-1}$ )
<i>Coilia neglecta</i>	Anchovy/Amodi/Ruli	Pelagic/Neteric	Primarily carnivorous	0.23
<i>Trichiurus sp.</i>	Ribbon fish	Demersal	Primarily carnivorous	0.27
<i>Oreochromis mossambicus</i>	Mozambique tilapia	Benthic-pelagic	Omnivorous	0.27
<i>Ailia coila</i>	Gangetic ailia/Kajuli	Pelagic	Primarily carnivorous	0.37
<i>Ompok pabo</i>	Pabo catfish	Demersal	Carnivorous/omnivorous	0.60

#### 4. Conclusion

The results obtained in the current study from the lower stretch of Hugli (Ganges) River along with Indian Sundarban wetlands reveal that  $Hg_T$  concentration in sediments are currently low due to a number of factors such as geomorphological positions, tidal and riverine inputs despite increasing anthropogenic factors associated with the growth of population and industry around Calcutta. Although the  $Hg_T$  values in surface sediments are well below the prescribed ERL value ( $0.15 \mu g g^{-1}$ ), sediment dwelling biota are high in terms of  $Hg_T$  in their body tissues. The maximum concentration of  $Hg_T$  was obtained in benthic polychaete *Dendronereis heteropoda* ( $0.603 \mu g g^{-1}$ ) from Gushighata ( $S_{14}$ ) which is greater than the maximum permissible range proposed by European Union ( $0.5 \mu g g^{-1}$ ). Although this species of polychaete is less likely consumed by carnivorous fishes but realizing their extensive used as a feed for shrimp and possible health risks to human beings and birds it should be taken into care for future monitoring and management of this region. Also there is a trend of  $Hg_T$  increasing from sediment < bivalve < fish < polychaete (Figure 2). There was a trend of  $Hg_T$  accumulation in carnivorous fish species than that of the herbivorous/planktivorous fish species reflecting the role of trophic position in  $Hg_T$  accumulation however it was not proven true for surface dwelling deposit feeders. The highest  $Hg_T$  concentration in fish was  $0.6 \mu g g^{-1}$  in *Ompok pabo* from Gangasagar beach ( $S_{11}$ ) is also higher than the European Union maximum permissible ( $0.5 \mu g g^{-1}$ ) and USEPA 'fish tissue residue criterion' ( $0.3 \mu g g^{-1}$ ) which are the maximum tolerable mercury intake levels in different edible fishes. Therefore fish, especially carnivorous fish species may require further consideration for human consumption, particularly high risk groups such as children or pregnant women. Future extensive studies are required to comment on the probable human health hazards from fish consumption in this study region.

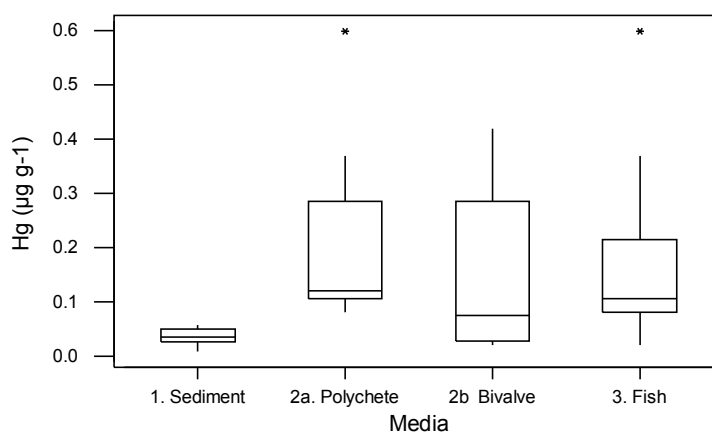


Figure 2. Box-plot showing ranges of  $Hg_T$  concentration in different sample media

This area is a zone of high environmental and socio-economic significance and the current study took a broad approach to examining a variety of environmental compartments for evidence of the accumulation of harmful concentrations of total mercury ( $Hg_T$ ). It demonstrated that the sediments sampled are not currently a sink for  $Hg_T$  based on a comparison with other sediments across the world, despite concentrations in biota being at

concentrations of concern, based on ERLs. This disparity, coupled with the increase in urbanization and industrialization around Calcutta suggests two types of detailed study are required in the future: (i) a longer term, time based monitoring programme of all compartments, at key diagnostic sites to highlight if environmental conditions change to a critical state; (ii) a detailed investigation of individual food chains linking back from commercial fish, through prey-lower life forms to representative sediments via suspended particulates to identify the key stages of bio-magnification and any potential remediation strategies. Mercury intake in the form of methylmercury from fish diet and other food sources is considered the most serious in terms of general impacts on human health. Several countries and international organizations have calculated and established the recommended dose or levels of mercury for health and safety (UNEP, 2010). The maximum value of  $Hg_T$  in our study ( $0.603 \mu g g^{-1}$ ) exceeded the prescribed values for India ( $0.5 \mu g g^{-1}$ ), Canada ( $0.5 \mu g g^{-1}$ ), China ( $0.3 \mu g g^{-1}$ ), Georgia ( $0.3 \mu g g^{-1}$ ), United Kingdom ( $0.3 \mu g g^{-1}$ ) and Japan ( $0.3 - 0.4 \mu g g^{-1}$ ). Although, this value well below the range of Safety Guidelines of WHO/FAO for predatory fishes ( $1 mg/kg$ ), USA ( $0.5 - 1 \mu g g^{-1}$ ) and Croatia ( $0.5 - 1.5 \mu g g^{-1}$ ). The intake of mercury depends not only on the amount of  $Hg_T$  in fish, but the proportion of fish consumed. Therefore, in order to have a comprehensive health risk assessment of  $Hg_T$  from fish consumption, a detailed survey regarding age, sex and fish consumption frequencies of a studied population has to be considered. Furthermore, the safety guidelines with regards to the maximum allowable concentrations of total mercury ( $Hg_T$ ) for consumption has been shown to vary between countries and international organizations (i.e.  $0.3 - 1.5 \mu g g^{-1}$ ). This issue would (and may) become more contentious if fish captured in the Indian Sundarban wetland were then found to be sold in fish markets further afield (outside of India) and in doing so would therefore contravene safety guidelines stipulated for  $Hg_T$  in these countries.

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