Environmental & Analytical
Toxicology

Bhattacharya et al., J Environ Anal Toxicol 2014, 4:6
http://dx.doi.org/10.4172/2161-0525.1000241

Research Article
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Assemblages of Total Mercury in the Tropical Macrotidal Bidyadhari Estuarine Stretches of Indian Sundarban Mangrove Eco-Region

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Abstract

The study was conducted to estimate total mercury in water and sediment of Bidyadhari river of Indian Sundarban delta in pre-monsoon, monsoon and post-monsoon period. Bidyadhari river presently serves as a sewage and excess rainwater outlet from the Kolkata metropolitan and adjacent area which ultimately empties at the Bay of Bengal in the course of the Indian Sundarban delta. Four different study sites situated around the course of the river were selected from the outfall of sewage canals at Kulti-Ghushighata (S1) where metropolitan sewages discharged and mixed up into water of Bidyadhari river which ultimately carried through this river via stations Malancha (S2), Kanmari (S3) to Dhamakhali (S4), just before the river confluences with the larger Raimangal river at northern Sundarban delta. Mean mercury concentration in collected water ranged BDL to 0.014 ± 0.001 μg ml^{-1} and sediment samples ranged BDL to 0.260 ± 0.014 μg g^{-1}. Highest mercury accumulations in river water both high tide and low tide was found at S4 followed by S3 with pronounced seasonal variation. Mercury present in the sediment (0-5 cm) showed a remarkable site and season specific differences with highest concentration in S4. Box whisker plot revealed that one extreme value was found at the S4 along with one outlier was at S3 and five outliers were at S4 during monsoon period. Regarding total mercury assemblages, PCA analysis showed all the sites except S4 are significantly associated. Based on Effective Range Low (ERL) value it is considered that sediment is still low mercury enrichment with less ecotoxicological risk while level often above the requirement desirable limit of drinking water recommended by WHO.

Keywords: Mercury; Bidyadhari river; Indian sundarban; Estuarine sediment

Introduction

Mercury (Hg) is a major toxic element and contaminant to the environment. It is produced by natural and anthropogenic activities and accumulating in the biotic systems. Mercury pollution in aquatic ecosystems has received great attention since the discovery of mercury as the cause of Minamata disease in Japan in the 1950s [1]. Mercury is a well-known neurotoxic compound [2] which is listed by the International Program of Chemical Safety (IPCS) as one of the six most dangerous chemicals in the world [3]. It was documented that Asia contributes 54% anthropogenic mercury emissions to global atmosphere [4]. In surface water, mercury can enter the food chain, or it can be released back to the nature by the process of volatilization [5,6]. Ecotoxicological behavior of mercury in aquatic systems has immense attention because it is the only heavy metal which bioaccumulates and biomagnifies through all levels of the aquatic food chain and converted into methylmercury [7]. Riverine drainage is the main source of metal contamination in coastal areas [8].

The sediments of mangrove ecosystems have a large capacity to retain heavy metals from tidal waters, freshwater rivers and storm water runoff. Mangrove wetland sediments possess many environmental factors that promote methylation of mercury and thus wetlands are recognized as "hot spots" for methylmercury (MeHg) production [9].

This study has been conducted at the north-eastern fringe area of the Sundarban delta around the river Bidyadhari. Bidyadhari river originates near Haringhata in Nadia district of West Bengal, India and then flows through areas of district North 24 Parganas before confluence with the Raimangal river in the Sundarban. At present the river has been used as the major drainage system and serves as sewage outlet of excess rainwater along with agriculture runoff from the city of Kolkata, North 24 Parganas and adjacent areas. The Bidyadhari and other such channels receiving brackish water under tidal effect and now carry little freshwater as they are mostly cut-off from the main stream of Ganges. However, a small branch of Ichamati river joining the Bidyadhari at Nazat near Dhamakhali which carries a portion of freshwater as well as pollution load. The pollution load contains industrial and municipal wastes along with agriculture runoff from the catchment areas at upstream of Ichamati river. Beside urban runoff, huge quantity of metropolitan waste from Kolkata city and North 24 Parganas district is carried through a long dry weather-storm water flow combined canal and discharged into the outfall on Bidyadhari river at Kulti-Ghushighata, situated at about 35 Km south-east of the eastern fringe of the Kolkata city. The pollutants discharged during low tides remains in the estuary for quite a long time and these were pushed up and downstream during high tides.

Many works were done with some considerable details in relation to mercury assemblages at Sundarban estuarine system focused on

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Received August 01, 2014; **Accepted** Septemebr 15, 2014; **Published** September 18, 2014

Citation: Bhattacharya S, Dubey SK, Dash JR, Patra PH, Das AK, et al. (2014) Assemblages of Total Mercury in the Tropical Macrotidal Bidyadhari Estuarine Stretches of Indian Sundarban Mangrove Eco-Region. J Environ Anal Toxicol 4: 241. doi: 10.4172/2161-0525.1000241

western, central and eastern part [10-15]. However, dearth information is available pertaining mercury load in northern part of Sundarban region focusing Bidyadhari estuarine stretches. Since substances like mercury in excessive amount can affect aquatic life and natural vegetation, this study has been designed to access the distribution and seasonal variations of the total mercury in the river water and sediments in different localities around the course of Bidyadhari river of Sundarban delta. This study will be useful for determining classical baselines for total mercury in Sundarban mangrove ecosystems and rivers quality too.

Materials and methods

Description of study area

The world's largest luxuriant mangrove chunk, the Sundarban is an archipelago of several hundred islands, spread across 9,630 sq km of which 4,266 sq km is in Indian side. This is a well-mixed estuary due to intense semidiurnal tide and wave action with a macrotidal setting. In the Indian Sundarban, approximately 2,069 sq km of the area is occupied by the seven tidal riverine system or estuaries, which finally end up in the Bay of Bengal. The flow of five major rivers including Bidyadhari river of the eastern and northern sector of the Sundarban has lost their previous upstream connections with another major river Ganges due to continuous heavy siltation and solid waste disposal from the adjacent cities and towns [16]. Various industries like paper, textiles, chemicals, pharmaceuticals, pesticides etc are situated in the bank of this basin, from where a considerable quantity of toxic and hazardous substance is being released into this important aquatic system along with huge organic load emanate from agricultural and aquaculture wastes as non-point sources [17].

To quantify the mercury (Hg) quantum in water and sediment samples of Bidyadhari river of northern Sundarban delta in different seasons of a year viz. pre-monsoon (March-June), monsoon (July-October) and post-monsoon (November- February) during the period from March, 2012 to February, 2013, four different stations situated around the course of the river at considerable distances have been selected from the outfall of sewage canals at Kulti-Ghushighata (S1) where metropolitan sewages discharged and mixed up into water of Bidyadhari river which ultimately flowed through this river via stations Malancha (S2), Kanmari (S3) to Dhamakhali (S4), just before the river confluences with the larger Raimangal river at Sundarban delta. The estuary gets semidiurnal tides with a maximum range of 5.5 m at spring and minimum 1.8 m at neap [18]. Hydrology of this estuary presents a cyclic pattern, characterized by large amount of precipitation and tidal interplay [19].

Kulti-Ghushighata (S1) and Malancha (S2) is located at the side of Bidyadhari river in its northwest. The site is fed by the huge discharge of the sewage sludge through the sewage disposal canal of the Kolkata mega polis. Brick kiln and aquaculture impoundments (*Bheries*) are situated either sides of the river. Mangroves like *Avicennia officinalis* and *Acanthus ilicifolius* is present elsewhere. Kanmari (S3) lies a considerable distance from the waste outfall point of the river and dominated by huge aquaculture activities. Dhamakhali (S4) is the north-easternmost station of Sundarban, lying just below the Dampier-Hodges line (the northern most boundary of the Indian Sundarban). In the downstream part, it meets the Raimangal river, the drainage channel demarcating the international boundary between India and Bangladesh. *Avicennia officinalis, Avicennia alba* and *Acanthus ilicifolius* is present in the tidal embankments. Sampling stations with coordinates and salient features were depicted in Table 1.

Name of station	Co-ordinates	Approximate aerial distance between	
Kulti-Ghushighata (S1)	22°31.368’N 88°41.537’E	Kolkata-S1	30.5 km
Malancha (S2)	22°30.688’N 88°46.157’E	S1-S2	7.0 km
Kanmari (S3)	22°26.464’N 88°48.246’E	S-2-S3	9.0 km
Dhamakhali (S4)	22°21.332’N 88°52.595’E	S-3-S4	12.5 km

Table 1: Co-ordination of four study sites with salient features.

Analytical procedure

Water from river collected during high-tides, ebb-tides as well as pooled of both tides separately. Water samples were collected at about 50 cm depth. Samples were stored in watertight neutral polyethylene containers previously soaked and washed with 10% nitric acid and double distilled water. The water samples were acidified in the field with concentrated HNO_3 at the rate of 5 ml/liter of water sample, to reduce the pH of the sample below 2.0 and stored at 4°C prior to analysis. Sediment samples were collected randomly from the top 0-5 cm of the surface using a metal pipe during ebb tides from the river. All samples collected in triplicate, pooled, thoroughly mixed and placed into pre-cleaned polyethylene containers. Sample were prepared through oven dried at 40°C, grind, sieved through 63 µm metallic sieve, visible marine organisms and coarse shell fragments along with grass leaves and roots when present were removed manually and stored in acid washed polythene containers until analysis.

Total mercury in water separately and sediment samples was quantified by wet ashing procedure in hot plate. Water samples were digested with 70% nitric acid while sediment samples were digested using tri-acid mixture of nitric acid, perchloric acid and sulphuric acid at 10:4:1 ratio. Successive steps of standard procedure of sample preparation were followed according to method of reading for estimation of Hg in Atomic Absorption Spectrometer (AAS) (Model: VARION AA 240) equipped with vapor generation accessories (VGA) for cold vapor mode (Model No. VGA77). Operating parameters and procedures viz. instrumental condition operation, preparation of working standard solution, instrument calibration and validation of total metal analysis methods were monitored accordingly.

Statistical analysis

One-way Analysis of Variance (ANOVA) was performed to assess whether mercury concentrations varied significantly between and within the sites with a Tukey's HSD means separation test to determine the differences among the means. Probabilities less than 0.01 ($P<0.01$) were considered statistically significant. The data were checked for homogeneity of variance before analysis. All statistical calculations were performed with SPSS 10.0 for Windows (SPSS Inc. Chicago, IL USA). Outlier detections for box whisker plot with D'Agostino- Pearson test for normal distribution and clustered multiple variable graphs were prepared using statistical software Medcalc® version 12.7.0. (MedCalc Software bvba, Ostend, Belgium). The box whisker plot shows the interquartile range (25 to 75 percentile) of the values. The line within the box represents the median value. White circles outside the box represent outliers and circles topmost represents extreme values. Using study sites as variable, principal component analysis (PCA) was used to characterize the metal composition in river. PCA is a multivariate statistical approach used for data reduction and for interpret patterns within large sets of data. With PCA, a large data matrix is reduced to two smaller ones that consist of principal component (PC) scores and loadings [20]. PCA was employed using PAST statistical software, version 3.0.1. Hierarchical cluster analysis characterizes similarities among samples by examining inter-point distances representing all possible sample pairs in high-dimensional space [21]. The Bray-Curtis Cluster Analysis (Single Link) was employed to estimate the distance and similarity matrix of metal distributions among the study sites (ordination method) and dendogram to show hierarchical clustering using Bio-Diversity pro software. All numerical data are represented as the mean ± SE.

Results

Mean mercury concentration in collected water (BDL to 0.014 ± 0.001 μg ml^{-1}) and sediment samples (BDL to 0.260 ± 0.014 μg g^{-1}) from four different localities were presented in Table 2. Mercury concentration in river water remains below detectable level in Kulti-Ghushighata (S1) and Malancha (S2) during all seasons. Highest mercury accumulations in river water both high tide and low tide was found at Dhamakhali (S4) followed by Kanmari (S3) and significant variation was observed between two sites (F ratio=31.27, $P<0.0001$; $R^2=0.30$). In S3, mercury concentration varied significantly within the seasons (F ratio=5.12, $P<0.01$; $R^2=0.23$). Highest mercury concentration was observed in pre-monsoon period but no significance differences was observed with monsoon period ($P=0.402$) in S3. In S4, significant seasonal variation in mercury accumulation was observed (F ratio=36.14, $P<0.0001$; $R^2=0.68$) with highest value in monsoon period. However, no significant differences were observed in river water mercury concentration in high tides and low tides within the sites (F ratio=0.37; $P=0.54$).

The mercury present in the sediment showed a remarkable site and season specific differences during the study period. A significant variation in yearly mercury accumulation in sediments (F ratio=7.86, $P<0.0001$; $R^2=0.25$) among the study sites were observed with highest concentration in S4 which were significantly different with S1 and S2 respectively ($P<0.0001$). Significant differences were also observed between S2 and S3 regarding total sediment mercury accumulation ($P<0.0001$). However, no significant differences were observed between S1 and S2 ($P=0.96$), S1 and S3 ($P=0.11$) and S3 and S4 ($P=0.44$). Site wise mercury assemblage can be demonstrates as S4>S3>S1>S2 (Figure 1). Significant seasonal variations of mercury concentration within the four sites were also observed during the study periods. In S1 (F ratio=24.10, $P<0.0001$; $R^2=0.76$), mercury in pre-monsoon was significantly different with monsoon and post-monsoon concentration ($P<0.0001$), however no differences of concentration was observed between monsoon and post-monsoon ($P=0.10$). In S2 (F ratio=18.05, $P<0.0001$; $R^2=0.70$), similar trend was observed like S1 where mercury in pre-monsoon was significantly different with monsoon and post-monsoon concentration ($P<0.0001$), and no differences of concentration was observed between monsoon and post-monsoon ($P=0.96$). In S3 (F ratio=30.81, $P<0.0001$; $R^2=0.80$) and S4 (F ratio=232.44, $P<0.0001$; $R^2=0.96$) mercury concentration in all seasons were significantly different with each other ($P<0.0001$) respectively. Season wise sequence of Hg concentration in river sediments at S1 and S2 was Pre-monsoon>Monsoon>Post-monsoon and at S3 and S4 in the sequence of Monsoon>Pre-monsoon>Post-monsoon. Two-dimensional scatter box plot of total Hg in sediments in the Bidyadhari tidal stretches are presented in Figure 2. An outlier value is defined as a value that is smaller than the lower quartile minus 1.5 times the interquartile range, or larger than the upper quartile plus 1.5 times the interquartile range. Extreme value is defined as a value that is smaller than the lower quartile minus 3 times the interquartile range, or larger than the upper quartile plus 3 times the interquartile range. These values are plotted using a different marker, drawn in the "warning" color (here is blue). Coefficient of Skewness was found 1.6983 ($P<0.0001$) and Coefficient of Kurtosis was 2.1941 ($P=0.0107$). The D'Agostino-Pearson test for normal distribution showed rejected normality ($P<0.0001$). In total, seven anomalous Hg values were found as follows. One extreme value was found at the Dhamakhali (S4) in monsoon period. One outlier was at Kanmari (S3) and five outliers were at the Dhamakhali (S4) in monsoon period.

Bray-Curtis Similarity Matrix of mercury distribution at four sites revealed that at 76% and 71% similarity between S1-S2 and S3-S4 respectively. However, S1 clustered with S3 and S4 forms clearly differentiated cluster showing nearly 50% and 75% dissimilarity while, station S2 clustered with S3 and S4 forms cluster with 73% and 83% dissimilarity respectively. Pearson Correlation (r) matrix revealed that S1 significantly associated with S2 and S3 significantly associated with S4 regarding total mercury enrichment in Bidyadhari river (Table 3).

Substrate	Station	Pre-monsoon	Monsoon	Post-monsoon	Overall year
River water-High tides	S1	BDL	BDL	BDL	BDL
	S2	BDL	BDL	BDL	BDL
	S3	0.002 ± 0.001^{b}	BDL	BDL	0.001 ± 0.001^{a}
	S4	0.004 ± 0.000^{aA}	0.004 ± 0.001^{aA}	0.011 ± 0.002^{aB}	0.006 ± 0.005^{b}
River water-Ebb tides	S1	BDL	BDL	BDL	BDL
	S2	BDL	BDL	BDL	BDL
	S3	0.001 ± 0.000^{aA}	0.002 ± 0.001^{aA}	BDL	0.001 ± 0.001^{a}
	S4	0.001 ± 0.000^{aA}	0.004 ± 0.001^{aB}	0.014 ± 0.001^{aC}	0.006 ± 0.006^{b}
River water (Pooled)	S1	BDL	BDL	BDL	BDL
	S2	BDL	BDL	BDL	BDL
	S3	0.002 ± 0.001^{aA}	0.001 ± 0.001^{aA}	BDL	0.001 ± 0.001^{a}
	S4	0.002 ± 0.001^{aA}	0.004 ± 0.003^{bA}	0.012 ± 0.004^{bB}	0.006 ± 0.005^{b}
River sediment	S1	0.057 ± 0.005^{bA}	0.026 ± 0.007^{cB}	0.011 ± 0.002^{aC}	0.031 ± 0.02^{ab}
	S2	0.040 ± 0.005^{bA}	0.011 ± 0.005^{cB}	0.010 ± 0.001^{abB}	0.02 ± 0.01^{a}
	S3	0.083 ± 0.010^{aA}	0.156 ± 0.021^{bB}	0.007 ± 0.001^{bC}	$0.08 \pm 0.06b^{c}$
	S4	0.085 ± 0.005^{aA}	0.260 ± 0.014^{aB}	0.002 ± 0.000^{cC}	0.11 ± 0.11^{c}

Table 2: Mean concentrations of total mercury (T_{Hg}) in river water (μg ml^{-1}) and river sediment (μg g^{-1}) at Bidyadhari estuarine stretches.
Kulti-Ghushighata (S1); Malancha (S2); Kanmari (S3); Dhamakhali (S4); BDL- Below Detection Level
Superscripts like a, b, c in same columns were significantly different ($P<0.01$) in Tukey's HSD mean separation test
Superscripts like A, B, C in same rows were significantly different ($P<0.01$) in Tukey's HSD mean separation test.

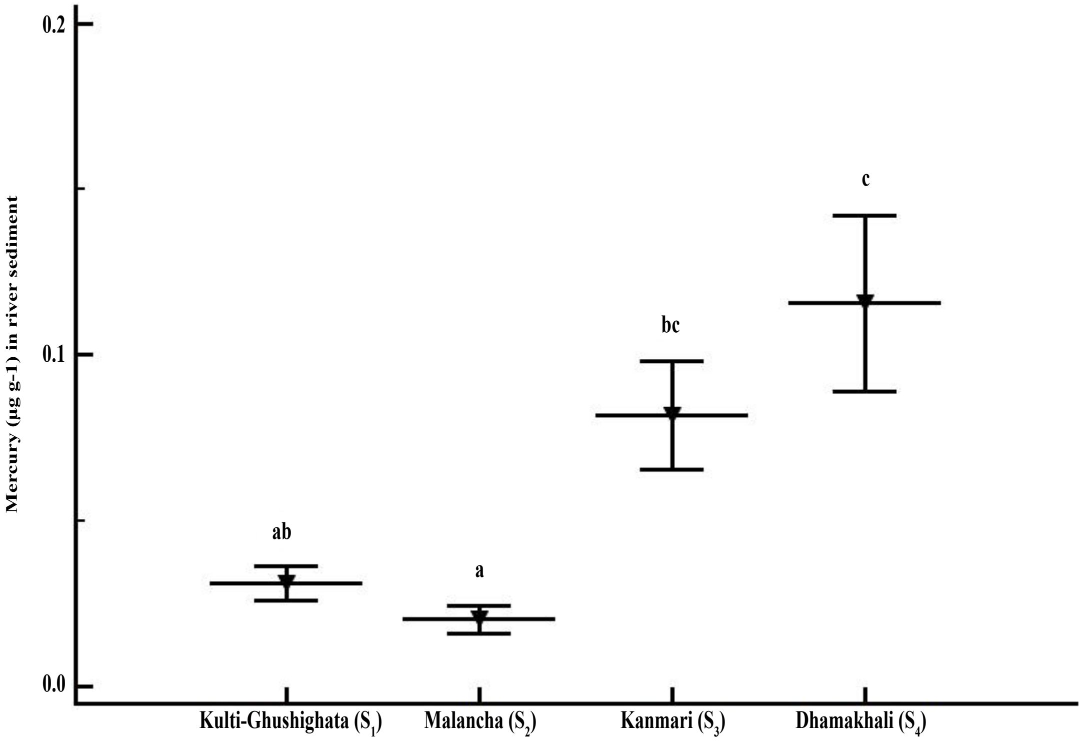

Figure 1: Distribution of mercury in sediments among the study sites. Different superscripts (a, b, c) were significantly different (*P*<0.01) in Tukey's HSD mean separation test.

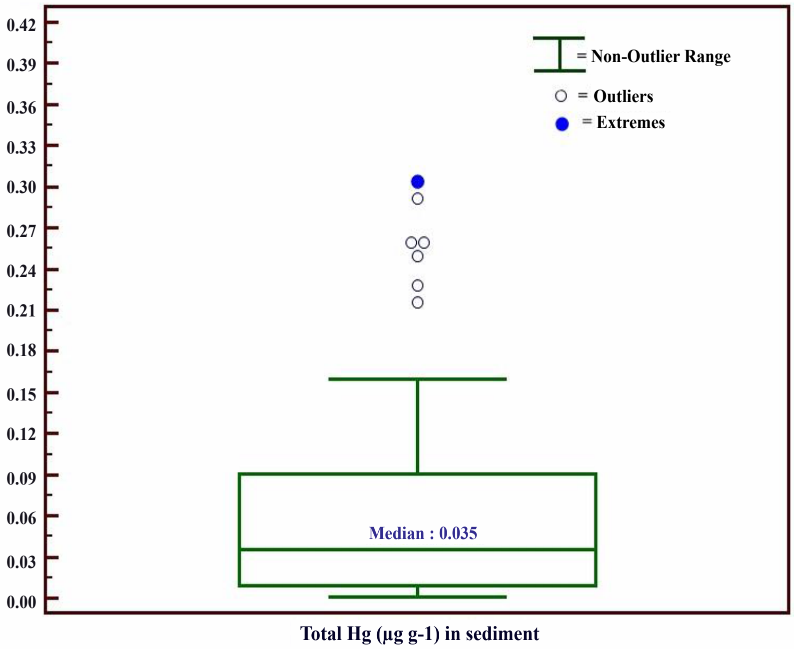

Figure 2: Two dimensional Box Whisker Plot of total mercury concentration in the sediments of Bidyadhari river. The box whisker plot shows the interquartile range of the values. The line within the box represents the median value. White circles outside the box represent outliers (values ranging from 0.20 to 0.28) and circles topmost represents extreme values (values>0.28).

Using study sites as variable, PCA was summarized in Table 4 and Figure 3. By studying the loadings of the variables on the components it can be seen that all the sites except Dhamakahli (S4) are significantly associated. PC 1 is responsible for 91.77% where S4 has strong positive loading. PC 2 shows a variance of about 5.39%, whereas S3 has positive loading. PC 3, which accounts 2.69% for total variance, has positive loading on S1 and S2.

Discussion

Though Hg was not detect in river water at S1 and S2, but Hg level in S3 and S4 were often above the requirement desirable limit (permissible limit) of drinking water (0.001 μg g^{-1}), recommended by WHO and IPCS [22,23]. Recent inventory on surface and core sediments in Indian Sundarban has exhibited contaminant values above the prescribed guidelines in few places indicating likely adverse impacts on adjacent biotic communities [10,12,13,15]. In a past study during 2005-2006, it was estimated that Hg contents in upper surface of sediment (0-4 cm) at Dhamakhali stretches of Bidyadhari river were 0.012 μg g^{-1} and 0.013 μg g^{-1} during post-monsoon and pre-monsoon season respectively [10] which are definitely intermediate values in between respective seasons at Dhamakhali under present study. In an another study, Kwokal et al. [15] showed that, the vertical distribution of total mercury in different sediment cores from the Hugli-Matla-Bidyadhari tidal complex within 0.09 μg g^{-1} (<93.3 ng/g), like 0.04 μg g^{-1} in Ghushighata and 0.02 μg g^{-1} in Dhamakhali, which is very seminal to our study. As compared to sediment quality benchmark criteria of Effects Range Low (ERL) value as reported by Long and Morgan [24], mercury enrichment in the present study are lower than the ERL values and thus it is likely that Hg levels in the sediment don't have lethal effects on benthic organisms [25].

Pearson Correlation (r)	**Kulti-Ghushighata (S1)**	**Malancha (S2)**	**Kanmari (S3)**	**Dhamakhali (S4)**
Kulti-Ghushighata (S1)	1			
Malancha (S2)	0.929	1		
Kanmari (S3)	0.627	0.476	1	
Dhamakhali (S4)	0.524	0.371	0.892	1

Table 3: Pearson Correlation (r) matrix of total mercury assemblages among the study sites (n=6). In bold, significant values at the level of significance alpha (α)=0.05 (two-tailed test).

Spatio-temporal heterogeneity in Hg enrichment and distribution pattern might be attributed to (a) location and distance of the four stations with the influence of rivers Ichamati and Raimangal with different tidal and geomorphic settings related to concerned river discharge and municipal outfall, (b) natural variability associated with physical mixing of the sediments, precipitation and run off (c) non-homogenous inputs from point and non-point sources of mercury as supported by Kwokal et al. [10].

Higher level of Hg in the river water at S3 and S4 is might be due to cumulative effect of sewage water contain Hg carried by the River Ichamati and discharged through a small tribulet to Bidyadhari river at Nazat in between Kanmari (S3) and Dhamakhali (S4), and due to close proximity of those stations with Bay of Bengal (north western part) from where sea water comes upwards in tidal effects which contains usually higher level Hg concentrations. Additional factors like bioturbational activities of macrozoobenthos refuge in Sundarban mudflat mainly Ocypodids, Thalassinids and other annelids like *Lumbrinereis spp.* and *Mastobranchus spp.* cause sedimentation turn over which bring physiochemical changes [26-28] that may contribute to the mobilization of Hg from the bottom to the superficial sediments and to the water column [29]. It is evident from the present set of data that among all stations, mercury content in water and sediment of river is usually less in Kulti-Ghushighata (S1) followed by Malancha (S2) suggesting the discharged through sewage canals in Bidyadhari river is not so high in mercury content for sediment contamination but alarming in respect of water quality which above the permissible limit of Hg for consumption in wide range of areas around the estuary. A large number of brackish water impoundments (*Bheries*) in either

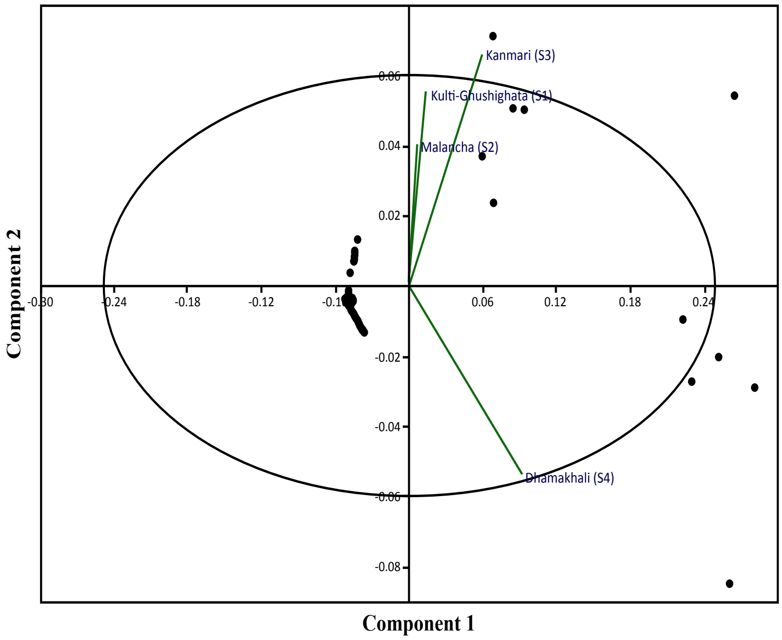

Figure 3: Principal Components Analysis (PCA): Variance-covariance plot with 95% ellipses.

Variable	PC 1	PC 2	PC 3	PC 4
Kulti-Ghushighata (S1)	0.11752	0.50275	0.59885	-0.61221
Malancha (S2)	0.060932	0.37142	0.4839	0.79005
Kanmari (S3)	0.53604	0.60832	-0.58452	0.030689
Dhamakhali (S4)	0.83375	-0.48911	0.25603	0.008821
Variance %	91.776	5.3929	2.6956	0.13548
Eigenvalue	0.00952	0.000559	0.00028	0.00001

Table 4: Results of Principal Components Analysis (loadings) after variance-covariance matrix. (The significant values are bold).

sides of the river exist where different fishes, prawns and crabs of commercial and export importance are cultivated using the waste water mixed tidal water through drainage systems from the river during high tides. Enhancement of Hg level in this river water may be dangerous for aquatic biota due to possibility of bio-accumulation of mercury as methyl mercury which is highly toxic to the living bodies including humans.

Conclusion

This study reveals that the Bidyadhari estuarine stretches area represents a safe environment in the context of sediment mercury contamination but frightening in respect to water. The data reported are useful baselines for total mercury in Sundarban mangrove, India and would be of help in future river quality studies. Further studies should account for all these parameters to understand the biogeochemical cycling of Hg in marine environment. With rapid development of electronic industries in West Bengal and large number of electronic wastes discarded into the environment might cause serious Hg pollution in Sundarban fluid mass in near future. To fully understand the fate of Total mercury (THg) and Methyl mercury (MeHg) in the Sundarban wetland ecosystem, these sources and transfer processes need to be identified, quantified and evaluated. Such a programme would facilitate us to develop sustainable remedial measures in future perspectives.

Acknowledgement

Authors are acknowledged and thankful to West Bengal University of Health Sciences for giving the necessary permission of the study which is a part of ongoing Ph. D works of the first author on *"Monitoring of xenobiotics in flora, fauna and human and their relationship at Sundarban Delta"*. Authors are grateful to Vice-Chancellor, West Bengal University of Animal and Fishery Sciences; Dean, Faculty of Veterinary &Animal Sciences, WBUAFS and Head, Department of Pharmacology and Toxicology, WBUAFS, Kolkata for providing necessary permission and laboratory facilities to do the research work. Authors are greatly indebted to anonymous reviewers for their constructive comments.

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Citation: Bhattacharya S, Dubey SK, Dash JR, Patra PH, Das AK, et al. (2014) Assemblages of Total Mercury in the Tropical Macrotidal Bidyadhari Estuarine Stretches of Indian Sundarban Mangrove Eco-Region. J Environ Anal Toxicol 4: 241. doi: 10.4172/2161-0525.1000241