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Research Article

SPATIO-TEMPORAL VARIATION OF MERCURY IN BIDYADHARI RIVER OF SUNDARBAN DELTA, INDIA

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ABSTRACT: Bidyadhari river originates in Nadia district of West Bengal, India and then flows through North 24 Parganas district and now serves as a sewage and excess rainwater outlet from the city of Kolkata and adjacent area, which ultimately empties at the Bay of Bengal through the Indian Sundarban delta. 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 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. This study was conducted to estimate total mercury (Hg) concentration in waters (during high tides and ebb tides) and sediments of Bidyadhari river in pre-monsoon, monsoon and post-monsoon seasons during the period from March, 2012 to February, 2013 at those stations. It is revealed from the estimated data that agricultural runoff, sewage, effluents from various industries and Kolkata metropolitan, Salt Lake City and adjacent areas of North 24 Parganas district carried and discharged in Bidyadhari river through sewage canals are not so high in mercury content for sediment contamination but alarming in respect of water quality, which crosses the permissible limit of Hg for consumption (0.001 ppm) in wide range of areas at Kanmari and Dhamakhali around the estuary. Enhancement of Hg level in this river water and transportation of the metal through tidal effects to and fro mangrove land of Sundarban may be dangerous for aquatic lives and supposed to be grave concern for the ecology of the Sundarban delta including humans.

Keywords: Bidhyadhari river, Sundarban delta, Mercury, High tide, Ebb tide, Sediment.

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INTRODUCTION

Mercury (Hg) is a global pollutant which cycles between air, water, sediments, soil and organisms in various forms (Moreno *et al.*, 2005). With well known toxic effects the amounts of Hg, which are mobilized and released into the environment, have increased considerably since the onset of the industrial age (Boening 2000).The presence and behavior of Hg in aquatic systems is of great interest and importance since it bioaccumulates and biomagnifies through all levels of the aquatic food chain (Lindqvist *et al.*,1991). Riverine drainage is the main source of metal contamination in coastal areas (Mitra 1998, Bastidas *et al.*,1999). This study has been conducted at the north-western fringe area of the Sundarban delta around the river Bidyadhari, which originates near Haringhata in Nadia district of West Bengal, India and then flows through North 24 Parganas district before confluence with the Raimangal river in the Sundarban. Now this river has been used as a major drainage system for carrying sewage and excess rainwater from the city of Kolkata and North 24 Parganas (Chatterjee 2006, Mitra *et al.*, 2009). The Sundarban mangrove estuarine chunk is considered as most diversified and productive eco-region of the tropics which has a dense network of intersecting creeks, estuaries, inlets and tidal water ways. 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 the Ganges, the major source of freshwater (Guhathakurta and Kaviraj 2000).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 contains industrial and municipal wastes along with agriculture runoff from the catchment areas at upstream of Ichamati river. Beside runoff from a wide catchment areas comprise dual cities Kolkata and Salt Lake along with North 24 Parganas district, a huge quantity of metropolitan waste arising both from industrial and domestic sources is carried through a long dry weather- storm water flow combined canal and discharged into the outfall on Bidyadhari river at Kulti-Ghushighata (Haroa block adjacent to the border of Minakhan block), situated about 35 Km south-east of the eastern fringe of the city. The pollutants discharged during low tides remains in the estuary for quite a long time and these were pushed above during high tides. Systematic information pertaining to the biology of the estuary and impact of sewage on ecosystem are meager as very few workers have attempted to study this aspect (Khan 1995).The wastes from these industries and municipal discharges can be considered as potential sources of metals (both toxic and non-toxic) as observed by various workers (Shamberger 1981, Lemly 1985, Sarkar *et al.*, 1999). Although, the sediments of mangrove ecosystems have a large capacity to retain heavy metals from tidal waters, fresh water rivers and storm water runoff, and they often act as sinks for heavy metals (Yim and Tam1999, Tam and Wong 1993, 1995, 2000).

Many scientists conducted some studies in considerable details in relation to sewage disposal on either side of outfall for a considerable distance. Their studies also covered on nature of sewage and its use in pisciculture, fish seed resources of the estuary, physico-chemical, biological and bacteriological condition of the estuary including its contamination by heavy metals and

aquaculture in sewage fed ponds etc.(Saha *et al.*, 1958, David 1959, Thakur 1975, Ray *et al.*, 1984, Ray and Bandhopadhyaya 1984 and Khan 1995). A few studies have indicated that heavy metals (relative density over 5) are generally increasing in the water, sediment and biota of Sundarban (Guhathakurta and Kaviraj 2000). In respect of the occurrence of heavy metals in river water and sediment is due to the discharge of industrial effluents from various sources including untreated sewage, municipal waste and agrochemical runoff from nearby cities and villages directly into the river (Singh *et al.*, 2005),the study of the distribution of metals in water and especially of sediments of estuaries is very important from the point of view of environmental pollution because sediment concentrates metal from aquatic systems, and represents an appropriate medium to monitor contamination (Sarkar *et al.*, 2004).

However, dearth information was available pertaining Hg load in northern part of Sundarban region focusing Bidhyadhari estuarine stretches. In continuation to the fact, this study is design to access the total Hg concentration and to monitor the spatio-temporal variation as well as xenobiotic quantum of mercury in the river water and sediments in different localities around the course of Bidyadhari river of Sundarban delta.

MATERIAL AND METHODS

Study seasons and sites: The study was undertaken to quantify the recent xenobiotic quantum of Hg in water and sediment samples of Bidyadhari river of Sundarban delta due to disposal of huge quantity of metropolitan wastes disposal 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 (Fig. 1). Stations were

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

Name of station	Name of the Community Development Block	Co-ordinates	Approximate aerial distance between	
Kulti-Ghushighata (S1)	Haroa	22°31.368'N, 88°41.537'E	KSC - S1	30.5 km
			Bantala - S1	26.5 km
			KLC - S1	18.0 km
Malancha (S2)	Minakhan	22°30.688'N,88°46.157'E	S1 - S2	7.0 km
Kanmari (S3)	Sandeshkhali-I	22°26.464'N, 88°48.246'E	S-2 - S3	9.0 km
Dhamakhali (S4)	Sandeshkhali-II	22°21.332'N, 88°52.595'E	S-3 - S4	12.5 km

KSC: Kolkata Science City, KLC: Kolkata Leather Complex.

Table 2: Inter seasonal concentration of mercury (ppm) in river sediment and waters during high tides and ebb tides at different stations (Mean ±SE of six replicates).

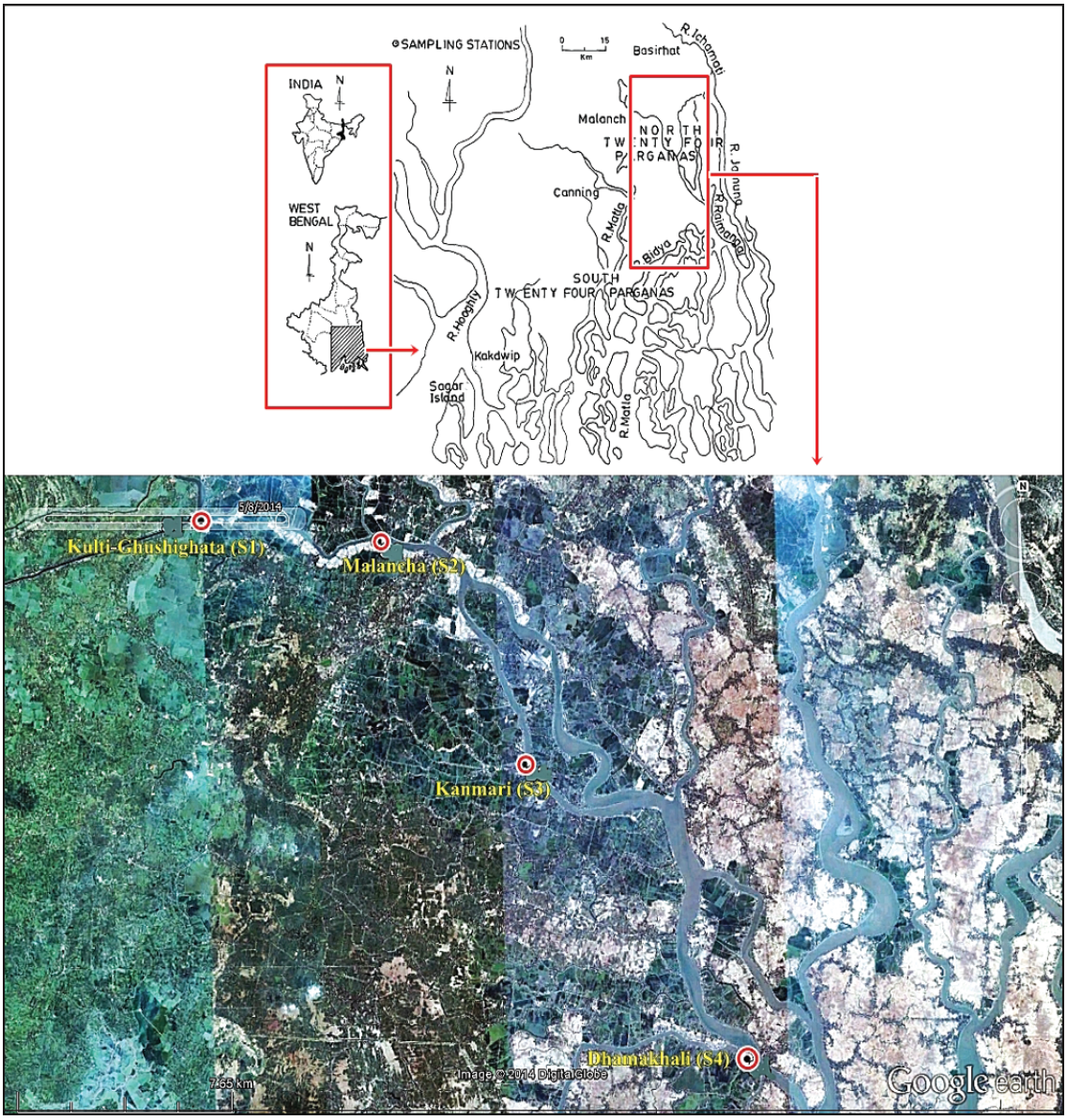
Substrate	Station	Pre-monsoon	Monsoon	Post-monsoon
River water -	Kulti-Ghushighata (S1)	BDL^{c}	BDL^{b}	BDL^{b}
High tides	Malancha (S2)	BDL^{c}	BDL^{b}	BDL^{b}
	Kanmari (S3)	0.002 ± 0.001^{b}	BDL^{b}	BDL^{b}
	Dhamakhali (S4)	0.004 ± 0.000^{a}	0.004 ± 0.001^{a}	0.011 ± 0.002^{a}
River water -	Kulti-Ghushighata (S1)	BDL^{b}	BDL^{b}	BDL^{b}
Ebb tides	Malancha (S2)	BDL^{b}	BDL^{b}	BDL^{b}
	Kanmari (S3)	0.001 ± 0.000^{a}	0.002 ± 0.001^{ab}	BDL^{b}
	Dhamakhali (S4)	0.001 ± 0.000^{a}	0.004 ± 0.001^{a}	0.014 ± 0.001^{a}
River sediment	Kulti-Ghushighata (S1)	0.057 ± 0.005^{b}	0.026 ± 0.007^{c}	0.011 ± 0.002^{a}
	Malancha (S2)	0.040 ± 0.005^{b}	0.011 ± 0.005^{c}	0.010 ± 0.001^{ab}
	Kanmari (S3)	0.083 ± 0.010^{a}	0.156 ± 0.021^{b}	0.007 ± 0.001^{b}
	Dhamakhali (S4)	0.085 ± 0.005^{a}	0.260 ± 0.014^{a}	0.002 ± 0.000^{c}

Different superscripts (a, b, c) in same columns were significantly different (P< 0.01) in Tukey's HSD mean separation test.
BDL = Below detectable Limit.

selected considering the sediment dispersal patterns along with the drainage network systems and their position were fixed by a global positioning system (GPS) which is supported by Corsolini *et al.*, (2012). Co-ordinate description of four study stations with aerial distances from each other was presented in Table-1.

Sample collection and storage: Surface sediments and water samples (n=6) were collected during each study period from four key stations along the stretch of Bidhyadhari river namely, Kulti-Ghusighata (S1), Malancha (S2), Kanmari (S3) and Dhamakhali (S4). Water from river collected during high-tides as well as ebb-tides separately. Water samples were collected from surface level of the river. 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 5ml/liter of water sample, to reduce the pH of the sample below 2.0 and stored at 4^0C prior to analysis (Singh *et al.*, 2005).

Fig. 1: Photograph showing the different study area along the stretches of Bidyadhari river of Sundarban delta.



Sediment samples were collected randomly from the top 0-5 cm of the surface using a grab sampler 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 40^0C, lightly ground, sieved through 63 µm metallic sieve, visible

Table 3: Inter-seasonal variation of pH and salinity (ppt) in high tides and ebb tides at different stations (Mean ±SE of six replicates)

Station	Substrate	Pre-monsoon		Monsoon		Post-monsoon	
		pH	Salinity (ppt)	pH	Salinity (ppt)	pH	Salinity (ppt)
S1	High tide	7.13±0.04	9.00±0.74	6.96±0.06	1.00±0.36	7.48±0.12	0.75±0.17
	Ebb tide	7.11±0.07	5.50±0.36	6.28±0.04	0.33±0.21	7.53±0.13	0.66±0.16
S2	High tide	7.16±0.04	9.91±0.55	6.58±0.03	2.08±0.32	6.98±0.07	3.75±0.89
	Ebb tide	7.26±0.13	7.16±0.527	6.43±0.09	1.0±0.36	7.40±0.11	1.91±0.27
S3	High tide	7.03±0.08	11.08±0.32	7.35±0.14	4.91±0.52	7.41±0.07	6.41±0.97
	Ebb tide	7.55±0.10	7.91±0.82	7.18±0.12	2.83±0.45	7.58±0.03	3.75±0.72
S4	High tide	8.21±0.13	15.0±0.88	7.21±0.08	12.08±0.82	7.10±0.06	5.41±0.20
	Ebb tide	8.20±0.08	17.16±1.13	7.18±0.10	9.0±0.36	7.35±0.02	6.50±0.22

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.

Estimation of pH and salinity of water: pH and salinity of the water sample were determined directly in the field measured by using Hanna multi-parameter water analysis kit (Model: HI9828, Hanna instruments, USA).

Estimation of Hg: Total Hg in water and sediment samples was quantified by wet ashing procedure in hot plate. Water samples were digested with 70% nitric acid as per method of Carbrey *et al.* (2009) while sediment samples were digested using tri-acid mixture of nitric acid, perchloric acid and sulphuric acid at 10:4:1 ratio following some modifications of the method of Welsch *et al.*(1990) and Datta *et al*. (2010). Successive steps of sample preparation as per standard procedure were followed according to method of reading for estimation of Hg in Atomic Absorption Spectrometer (AAS) (Model: VARION AA 240). The AAS was equipped with vapor generation accessories (VGA) for cold vapor mode (Model No.VGA77) using Argon. 0.3 % Sodium Borohydride + 0.5 % Sodium hydroxide were used as reducing agent. In 10 ml sample 2.5 ml HCL and 2.5 ml HNO_3 was added and the volume was made to 25 ml with Millipore water. 0, 5, 10, and 20 ppb standards were prepared accordingly from 1 ppm standard Hg solution.

Every time before taking the reading the instrument was calibrated and standard curve was prepared with 0, 5, 10 and 20 ppb freshly prepared standards. Absorbance was recorded at wave length 253.7 nm. 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.

Reagents: Millipore water was used and all

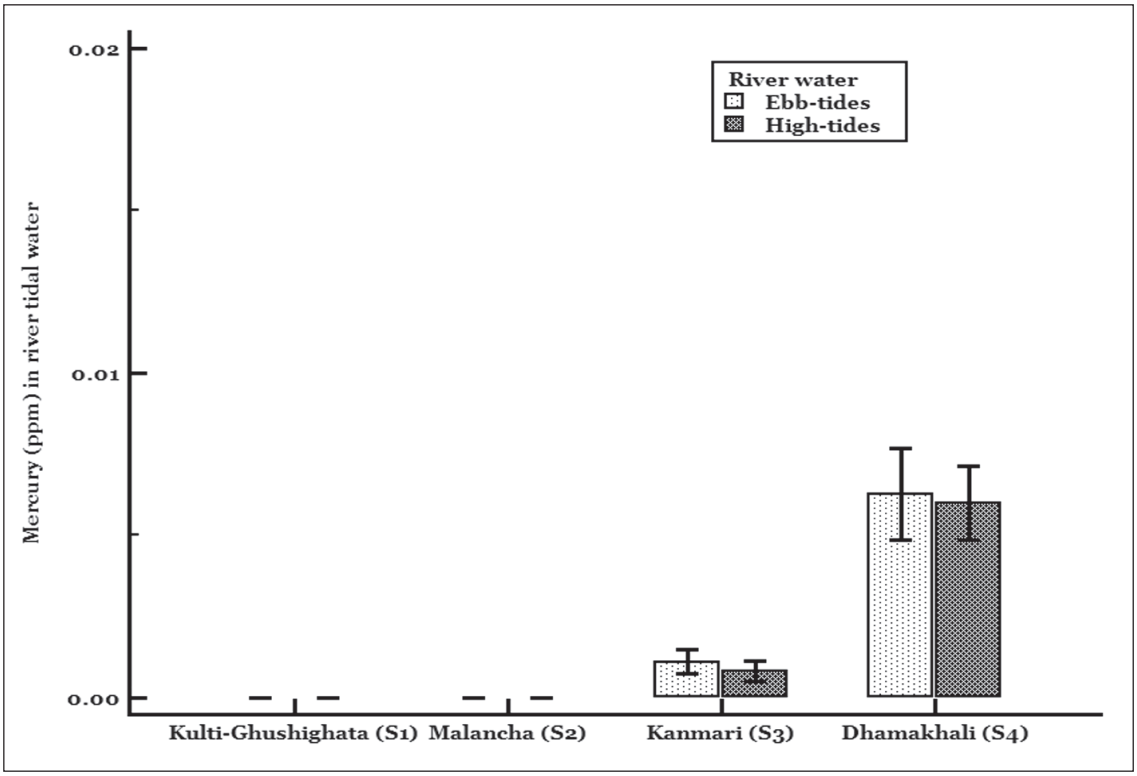


Fig. 2: Differences of Hg concentrations (ppm) in high-tidal and ebb-tidal water of river among the study sites throughout the year (Mean ±SE of six replicates)

chemicals of analytical grade were purchased from Rankem Pvt. Ltd., E-Merk (India) and Sigma Aldrich (USA).

Statistical analysis: 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. Possibilities less than 0.01 ($P< 0.01$) were considered statistically significant. All statistical calculations were performed with SPSS 10.0 for Windows (SPSS Inc. Chicago, IL USA). Clustered multiple variable graphs were prepared to establish variation within the sites using statistical software Medcalc® version 12.7.0. (MedCalc Software bvba, Ostend, Belgium). 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 (Mc Aleece *et al.,* 1997).All numerical data are represented as the mean ± SE.

RESULT AND DISCUSSION

Mercury concentration in river water remains below the detectable level at S1, S2 during all

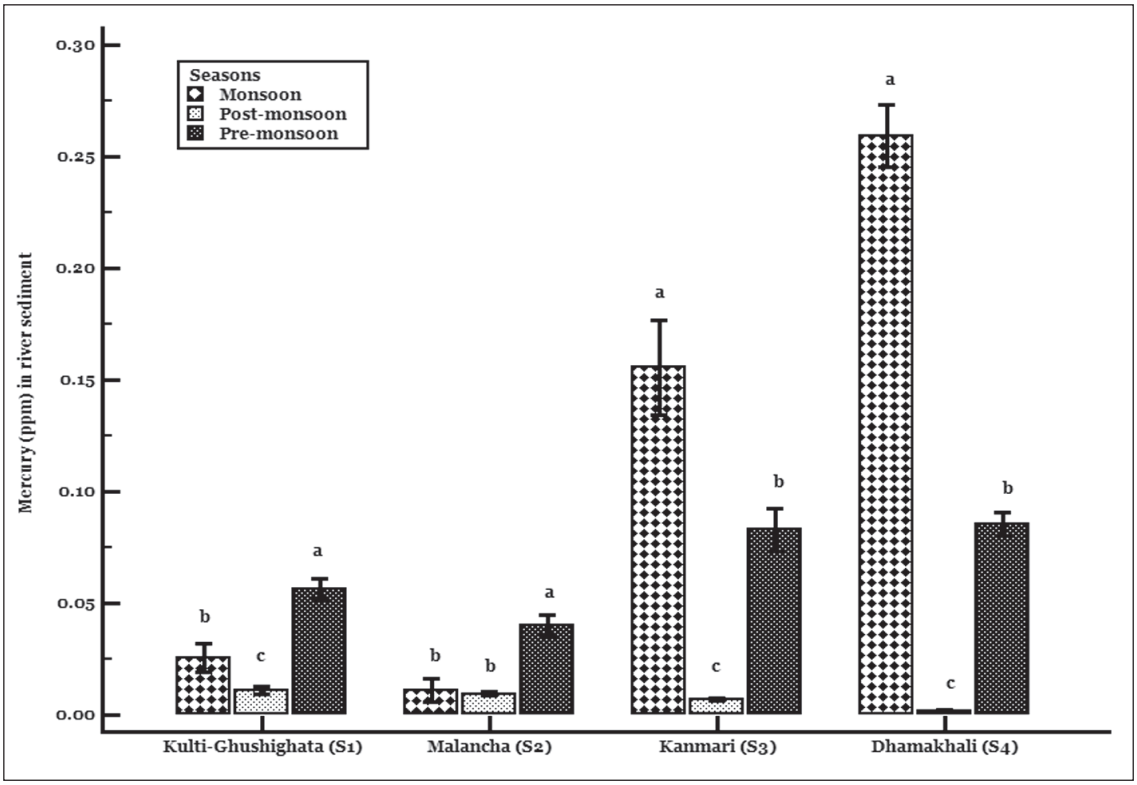


Fig. 3: Inter- seasonal variation in total Hg concentrations (ppm) in sediment of river among the study stations (Mean ±SE of six replicates). Different superscripts (a, b, c) in bar were significantly different ($P< 0.01$) in Tukey's HSD mean separation test.

seasons. But concentration of Hg (0.001-0.002 ± 0.001ppm) during pre-monsoon and monsoon seasons is observed in river waters at S3 whereas river water at S4 contains always some amount of Hg (0.001-0.014± 0.001ppm) through over the seasons.

In all seasons, S4 is having significantly higher Hg content in river water, usually in high tides, than remaining stations ($P< 0.01$). Mean Hg content of river water in ebb tides at S3 and S4 during pre-monsoon and monsoon are homogeneous and these values are significantly higher than that of S1 and S2($P< 0.01$). Graphical representations regarding differences of mercury concentrations (ppm) in high-tidal and ebb-tidal water of river among the study sites (S1 to S4) throughout the year have been presented in Fig. 2. From the Fig. 2 it is evident that prevalence of Hg is higher at S3 and S4 during ebb tides. River sediments at S1 and S2 contained metal concentration of 0.010 ± 0.001 to 0.057±0.005 ppm in spite of non-detectable level of Hg in river water at those stations. Mean concentration of Hg in sediments present in the station S4 (0.260±0.014 ppm) followed by S3 (0.156±0.021 ppm) is quietly higher in monsoon

Table 4: Mercury (ppm) distribution among the study sites: Bray-Curtis Similarity Matrix

Stations (S4)	Kulti-Ghushighata (S1)	Malancha (S2)	Kanmari (S3)	Dhamakhali
Kulti-Ghushighata (S1)	*	76.4031	50.5505	35.3011
Malancha (S2)	*	*	37.1526	23.8327
Kanmari (S3)	*	*	*	71.9806
Dhamakhali (S4)	*	*	*	*

than that of S1 and S2. The sequence of Hg concentration in river sediments at S1 and S2 is Pre-monsoon> Monsoon> Post-monsoon. Hg concentration in river sediments remains higher at S3 and S4 in the sequence as Monsoon> Pre-monsoon> Post-monsoon. In pre-monsoon, mean Hg contents in river sediments at S3 and S4 are homogeneous and significantly higher than that of S1 and S2 whilst the fact is become reverse at post-monsoon and at that season mean Hg contents at S1 and S2 are homogeneous and significantly higher than that of S3 and S4 ($P< 0.01$). However, mean Hg contents at S4 during monsoon is significantly higher than that of remaining stations. In relation to inter-seasonal variation in mercury concentration in river sediments at different stations under study, it has been observed that mercury concentration in river sediments during pre-monsoon at S1 and S2 is significantly higher than that of monsoon and post-monsoon ($P< 0.01$), whereas mercury concentration in river sediments during monsoon at S1 is also significantly higher than that of post-monsoon but Hg content during monsoon and post-monsoon at S2 remain homogeneous. On the contrary, mercury concentration in river sediments during monsoon at S3 and S4 is significantly higher than that of pre-monsoon and post-monsoon seasons. During monsoon in relation to other seasons, this phenomenon happens may be due to cumulative effect of more runoff, municipal sewage and effluents from catchment areas as well as more tidal influx of sea water which contains usually higher level Hg concentrations from Bay of Bengal at closer proximity of the stations S4 and S3. Mercury concentration in river sediments at those stations during pre-monsoon is also significantly higher than that of post-monsoon ($P< 0.01$) (Fig. 3).

It has been observed that cluster showing nearly 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. Bray-Curtis Similarity Matrix (%) of mercury (ppm) distribution and hierarchical clustering dendogram among the study sites were presented in Table-4 and Fig. 4 respectively.

Mean values of the physico-chemical parameters of water like pH and salinity of different stations during high and ebb tides were

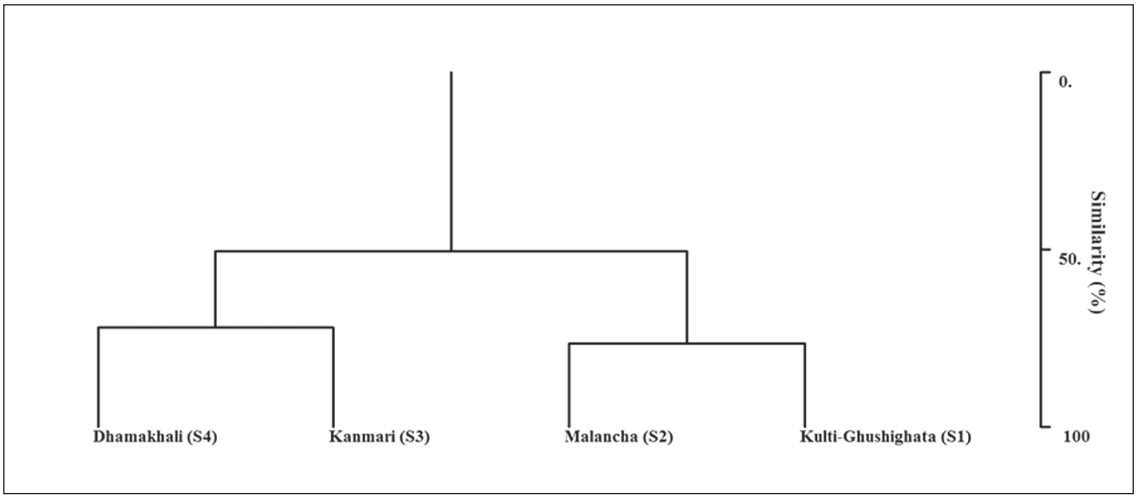


Fig. 4: Dendogram showing hierarchical clustering of study sites based on the Hg abundance indicating sites of similarity.

shown Table 3. In all seasons, Hg is significantly (P<0.01) and positively correlated with pH (r = 0.58 to 0.68) and salinity (r =0.52to0.79) of the respective waters.

Though Hg is non-detectable in river water at S1 and S2, but Hg level often crosses the requirement desirable limit (permissible limit) of drinking water *i. e.* 0.001 ppm recommended by WHO, EU (Lenntech 2013) and IS: 10500 (1992) in case of river water at S3 and S4 (Table 2).

As per reports of the WBPCB (2010 and 2011) (West Bengal Pollution Control Board, India), Hg concentration in ground waters at Dhapa (dumping grounds of industrial and domestic wastes of city Kolkata) and Kolkata Leather Complex (both are situated around the sewage canal *i.e.* dry weather- storm water flow combined canal which meets with Bidyadhari river at Kulti-Ghushighata and discharged the wastes) was above the permissible limit (0.001 ppm) at pre-monsoon season (April) in most of the recent years past. Similarly, river water at Malancha and Haroa adjacent area (a few kilometer away from Kulti –Ghushighata on upstream of Bidyadhari river) shows Hg concentration has been enhanced recent years from not traceable to 4.605 ppb (0.005 ppm) and 1.99 ppb (0.002 ppm) respectively on April, 2012 as estimated by WBPCB, India. In another study during 2005 and 2006, it was estimated that Hg contents in upper surface of sediment of Bidyadhari river at Dhamakhali were 12.7 ppb (0.013ppm) and 13.2 ppb (0.013 ppm) during post-monsoon and pre-monsoon season respectively (Kwokal *et al.,* 2008), which are definitely intermediate values in-between respective seasons at Dhamakhali under present study (*i.e*. 0.002 ppm at post-monsoon to 0.085±0.005 ppm at pre-monsoon).

Grain size distribution of the sediments has a role in accumulation of mercury in sediments (Kwokal *et al.,* 2008). Nature of river water is mostly neutral to alkaline and saline (Table 3) at all the study stations and the concentration of Hg in sediments is found usually considerably higher than those obtained in

surface water of river. Due to the neutral to alkaline nature of the river water, most of the heavy metals have precipitated and settled as carbonates, oxides, and hydroxides bearing sediments and elevated levels indicate higher exposure risks to the benthic biota of the river and ponds (Singh *et al.* 2005). However, Hg concentrations in river sediments at all stations under study are definitely remain below the permissible limit 1.0 ppm and 0.3 ppm formulated by USEPA and OME respectively (Table 2). Mitra *et al.* (1999) stated that fluctuation of salinity is a prime factor in the coastal areas, which influences partitioning and bioavailability of metals.

Higher level of Hg in the river water at S3 and S4 stations is might be due to cumulative effect of sewage water contain Hg carried by the Ichamati river and discharged through a small branch to Bidyadhari river at Nazat in between Kanmari (S3) and Dhamakhali (S4), and due to close proximity of those stations with Bay of Bengal (western part) from where sea water comes upwards in tidal effects which contains usually higher level Hg concentrations. Literatures suggest Hg eventually accumulates more in ocean and sea (Griesbauer 2007 and Ullrich *et al.,* 2001). Non-detectable level of Hg in river water at station 1 and 2 indicated that the sewages /effluents under question may not be contaminated much with Hg. It is found that Hg levels in all sediment samples from all stations are lower than Effects Range Low (ERL) value and thus having no toxic effects on the biota in habiting in the sediments (Kwokal *et al.,* 2008).

It is clear that among all stations, Hg content in water and sediment of river is usually less in Kulti-Ghushighata (S1) followed by Malancha (S2) suggesting agricultural runoff, sewage outfall and contaminated effluents from various industries along with anthropological activities as point sources and Kolkata metropolitan, Salt Lake City and adjacent areas. Though 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 crosses the permissible limit of Hg for consumption (0.001 ppm) in wide range of areas around the estuary. A large number of brackish water fish-ponds (*Bheries*) in either sides of the river exist where different fishes, prawns and crabs are cultivated usingthe 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 flora and fauna due to possibility of bio-accumulation of mercury in those substrates like fish, prawns and crabs as methyl mercury which is highly toxic to the living bodies including humans.

CONCLUSION

Agricultural runoff, sewage and effluents from various industries at Kolkata metropolitan and adjacent areas of North 24 Parganas district carried and discharged in Bidyadhari river through sewage canals is not so high in Hg content for sediment contamination but alarming in respect of water quality which crosses the permissible limit of Hg for consumption (0.001 ppm) in wide range of areas around the estuary. Enhancement of Hg level in this river water and transportation of the metal through tidal effects to and fro mangrove land of Sundarban may be dangerous for aquatic flora and fauna and supposed to be grave concern for the ecology of the Sundarban delta including humans. 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 in the Sundarban wetland ecosystem, these sources and transfer processes need to be identified, quantified and evaluated. Such a programme would facilitate to develop sustainable remedial measures in future perspectives.

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