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An Assessment of Mercury Loading in Core Sediments of Sunderban Mangrove Wetland, India (A Preliminary Report)

Ž. Kwokal · S. K. Sarkar · M. Chatterjee · S. Franciskovis-Bilinski · H. Bilinski · A. Bhattacharya · B. D. Bhattacharya · Md. Aftab Alam

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Abstract This is a preliminary report on total mercury (T_{Hg}) in core sediments (<63 μm particle size) of Sunderban mangrove wetland, northeastern part of the Bay of Bengal, India. Cold vapor atomic absorption spectrometry (CVAAS) was used for T_{Hg} determination. The concentration varies from 9.8 to 535.1 ppb (ngg^{-1}). Results revealed variations over premonsoon and postmonsoon month at different core depth, as well as in studied three sampling stations, located at the site of three rivers: Hugli River (S_1), Matla River (S_2) and Bidyadhari River (S_3). Elevated concentration of T_{Hg} in subsurface layer (4–8 cm) of the core at S_2 is attributed to remobilization of mercury from deeper sediment (32–36 cm). Positive correlation is present between total Hg and clay content. Based on index of geoaccumulation (I_{geo}) and Effects-Range Low (ER-L) value (150 ppb) it is considered that the sediments are till now unpolluted. As a consequence, there is less chance of ecotoxicological risk to organisms living in studied sediments. Two statistical methods were applied to determine T_{Hg} anomalies. Box plot method showed one extreme and three outliers in S_1 at postmonsoon season. Two extremes were found at S_2 at 4–8 and at 32–36 cm in premonsoon period. In S_3 there was no anomaly by box plot method. MAD method was more sensitive than box plot method and T_{Hg} anomaly was detected at 12–16 cm in S_3 during postmonsoon season. The data reported are useful baselines for T_{Hg} in Sunderban mangrove wetland, India and would be of help in future sediment quality studies.

Keywords Marine sediments · Estuarine sediments · Texture class · Total mercury · Mercury anomalies · Statistical analysis

Ž. Kwokal
Centre for Marine and Environmental Research, Rudjer Boskovic Institute, P.O. Box 180, 10001 Zagreb, Croatia

S. K. Sarkar (✉) · M. Chatterjee · A. Bhattacharya · B. D. Bhattacharya · Md. A. Alam
Department of Marine Science, University of Calcutta, 35 Ballygunge Circular Road, Calcutta 700019, India
e-mail: sarkar.santosh@gmail.com

S. Franciskovis-Bilinski · H. Bilinski
Department of Physical Chemistry, Rudjer Boskovic Institute, P.O. Box 180, 10001 Zagreb, Croatia

Mercury is a global pollutant that, cycles between air, water, sediments, soil and organisms in various forms (Moreno et al. 2005). With well known toxic effects (Boening 2000; Pilgrim et al. 2000) the amounts of Hg, which are mobilized and released into the environment, have increased considerably since the onset of the industrial age. Although anthropogenic Hg emissions have been reduced by half in the last decades (Pacyna et al. 2001), ongoing contamination is still a worldwide phenomenon. In recent years depletion of atmospheric Hg during spring time in the Arctic has been discovered which may act as a sink for the global Hg cycle (Schroeder et al. 1998; AMAP 2005). Depending on the Hg discharge and the ecosystem dynamics, Hg in sediments may be present in concentrated hotspots as dispersed over large areas in the world (Hines et al. 2000; Hinton and Veiga 2001; Kwokal et al. 2002; Frančišković-Bilinski et al. 2005; Moreno et al. 2005).

The presence and behavior of mercury in aquatic systems is of great interest and importance since it is the only heavy metal which bioaccumulates and biomagnifies through all levels of the aquatic food chain (Lindqvist et al. 1991). A number of coastal and marine environmental studies are based on mercury concentrations in sediment profiles (Andren and Harriss 1973; Marins et al. 1997;

Pereira et al. 1998; Quevauviller et al. 1992). Some of these works have shown that sediment profiles can be a reliable record of the history of natural (Marins et al. 1997) and anthropogenic stresses (Pereira et al. 1998). The world's largest magnificent mangrove block, the Sunderban is a typical and unique bioclimatic zone at the land-ocean boundary of the Bay of Bengal (20° 32′–20°40′ N and 88° 05′–89° E). It is formed at the estuarine and tidal network systems of River Ganges (Hugli) and Bay of Bengal covering an area of 9647 sq km. The macrotidal Hugli estuary together with the luxuriant Sunderban mangrove complex forms the genetically diverse ecosystems in the Bay of Bengal coast of northeast India. The region has been experiencing massive changes in forms of growth of industry and shifts in population and social structure. A significant ecological change is pronounced in this estuarine and coastal wetland environment due to huge discharges of domestic and industrial wastes along with the rapid emergence of Haldia port complex, a major oil disembarkment terminal in eastern India. The estuary has become vulnerable to chemical pollutants such as heavy metals, organochlorine pesticides, petroleum-derived hydrocarbons, chlorinated hydrocarbons etc., which have changed the biogeochemistry of the area and affected the quality of the local coastal environment. (Chatterjee et al. 2007; Guzzella et al. 2005; Saha et al. 2006).

Since substances like mercury in excessive amount can affect both aquatic life and natural vegetation, and enhances

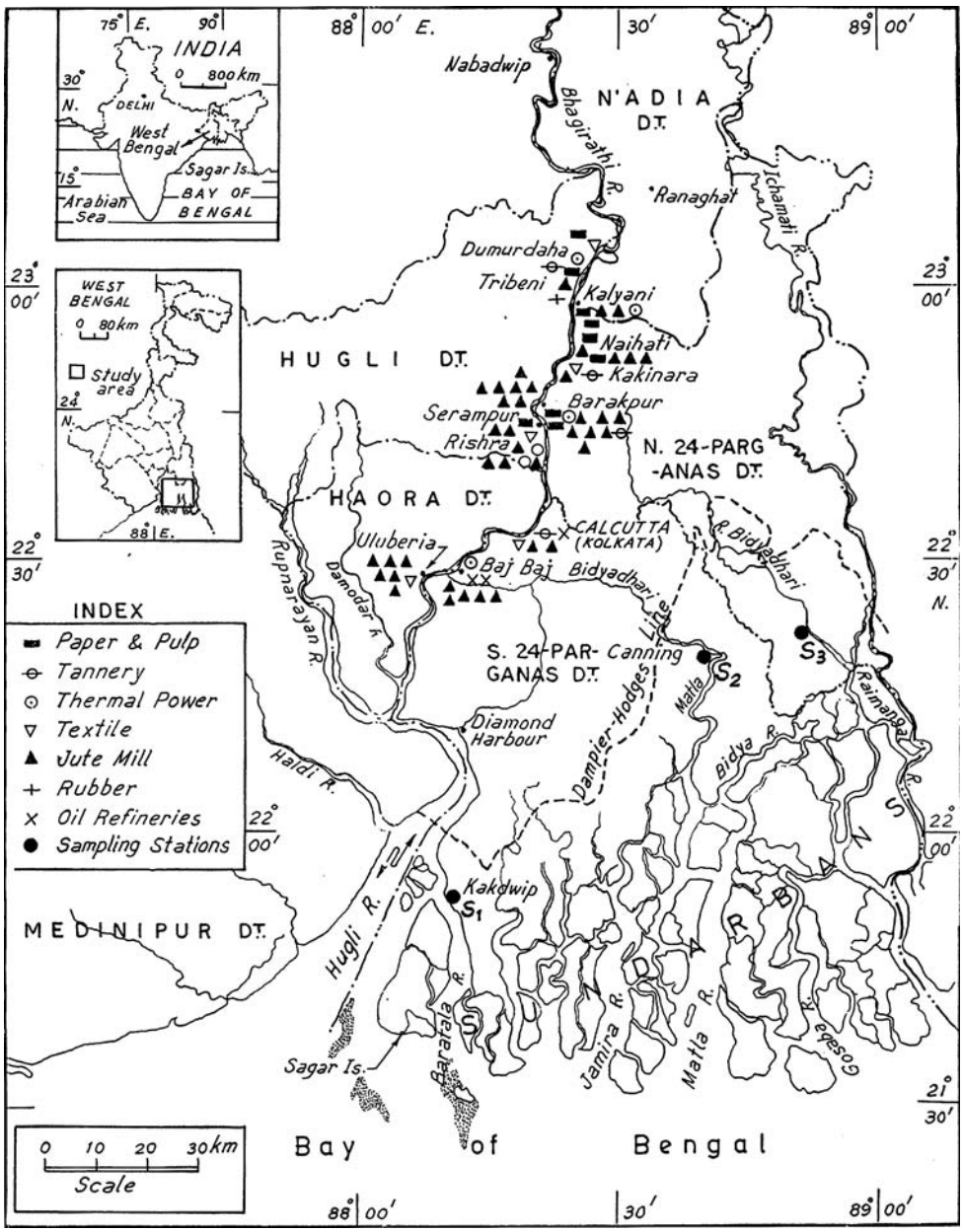

Fig. 1 Map of Sunderban showing the location of three sampling stations

the loss of ground cover leading to erosion, a study on the distribution and seasonal variations of the total mercury in the sediment core of this complex wetland has been attempted to assess the probable influence of both natural and anthropogenic factors. The distribution of total mercury in sediment profiles of three sampling sites (Fig. 1) in the Sundarban mangrove wetland has been examined in the present paper for ascertaining the state of T_{Hg} concentration in this economically important coastal region.

Materials and Methods

Core sediments were collected twice (October–December, 2005) and (April–June, 2006) during the postmonsoon and premonsoon seasons of two successive years, from three sampling stations located along east-west flank of Sunderban mangrove wetland covering a distance of 80 km in a northeast to southwest direction. Three sampling sites namely, Lot 8 (S_1, Canning (S_2) and Dhamakhali (S_3) by the Hugli, Matla and Bidyadhari rivers respectively, were chosen in Sunderban wetland because they belong to different tidal environments and they have different wave energy fluxes and distances from the sea (Bay of Bengal) (Fig. 1). Being located by the side of three rivers they maintain a difference with reference to geomorphic and tidal settings. Cores were collected with the help of a steel corer (40 cm length and 5 cm diameter) by gently pushing it into the sediments and dug out, capped and frozen on return to the laboratory. At each station every core was sliced into 4 cm fractions (sub samples) with the help of PVC spatula. Prior to sample collection, all the glasswares for the collection and storage of sediment samples were thoroughly cleaned with acid (10% HNO_3), and then rinsed in double-distilled (Milli-Q) water before each use.

Core fractions were stored in labeled polyethylene bags stored in iceboxes and transported to the laboratory where they were frozen to −20°C. Within two days, a portion of each sample was placed in a ventilated oven at a very mild temperature (max. 40°C). Dried samples were then disaggregated using an agate mortar and pestle, sieved through 63 μm metallic sieve and stored in hermetic plastic bags until analysis. All visible marine organisms and coarse shell fragments, sea grass leaves and roots when present were removed manually. These were divided into two aliquots, unseived one for sediment quality parameters (organic carbon, pH, % of silt, clay and sand) and the second (sieved) for mercury analysis. Organic carbon (C_{org}) content of the soil was determined following a rapid titration method (Walkey and Black 1934) and pH with the help of a deluxe pH meter (model no. 101E) using combination glass electrode manufactured by M.S. Electronics (India) Pvt. Ltd. Mechanical analyses of sediment were done by sieving in a Ro-Tap Shaker (Krumbein and Pettijohn 1938) manufactured by W.S. Tyler Company, Cleveland, Ohio, and statistical computation of textural parameters was done by using formulae of Folk and Ward (1957).

Sediment samples (about 0.2 g) were decomposed in a closed quartz vessel (pressure 3 bars) with mixture of 10 mL of nitric and perchloric acids at 140°C. Cold Vapour Atomic Absorption Spectrometry (CVAAS) method was used for total mercury measurements. The protocol was described by Fitzgerald and Gill (1979) and was used ever since by numerous workers (see in Kwokal et al. 2002). This method requires conversion of all forms of mercury to Hg^{2+} (acid, oxidative digestion) and its reduction to Hg^0 by Sn^{2+} solution, adsorption/desorption on Au—wire and detection of Hg^0 vapour by Elemental Mercury Detector (a mercury Monitor^{TM}3200 by a Thermo Separation Products). The detection limit of the method for sediment samples is 0.01 ppb (ngg^{-1}). The reproducibility is 2% for concentrations >200 ngg^{-1} and 10% for concentrations >1 ng g^{-1}.The efficiency is >90% for solid matrices. Quality assurance for determination of Hg was linked to Standard Reference Material for total mercury (marine sediment SRM 2702: designated concentration versus observed concentrations 0.4474 ± 0.0069 and 0.4414 ± 0.0098 ppb (μgg^{-1} respectively).

Factor analysis, correlation (r) values and analysis of variance (ANOVA) were performed in order to determine relationships between variables. All statistical analyses were performed using the computer software STATISTICA (STATSOFT, 1998; 2000; Minitab).

Two different statistical procedures to identify anomalous geochemical data were used. First was the box plot method. Dependent on the empirical cumulative distribution plots, normal or lognormal box plots are constructed. The box length is the interquartile range. Outliers are values between 1.5 and 3 box lengths from the upper or lower edge of the box. Far outliers (extremes) are values more than 3 box lengths from the edge of the box (Tukey 1977; see also Reimann et al. 2005).

The second complementary statistical method was the median absolute deviation (MAD). It is defined as the median of the absolute deviations from the median of all data (Tukey 1977). The median value ±2 MAD defines a fence, which separates outlier and extremes from a population.

Results and Discussion

Sediment characteristics such as pH, organic carbon (%), and percentages of sand, silt and clay obtained in different profiles of the three sampling stations have been shown in Table 1. Sediments are characterized by pH varying from slightly acidic to basic (6.6 to 8.7). The acidic nature is mainly pronounced in Canning (S_2) during postmonsoon

Table 1 Values of T_{Hg} (Total mercury), pH, organic carbon and textural properties in the three studied stations covering two ecological seasons

Season	Station	Depth (cm)	T_{Hg} (ng g^{-1})	pH	Organic carbon%	Sand%	Silt%	Clay%	Texture
Postmonsoon	Lot 8	0–4	46.3	8.2	0.63	0.6	25.95	73.45	Clayey very fine
		4–8	42	8.1	0.69	4.0	18.2	77.8	Clayey very fine
		8–12	43	8.5	0.67	0.6	24.35	75.05	Clayey very fine
		12–16	42	8.4	0.65	0.4	24.175	75.425	Clayey very fine
Premonsoon		0–4	22	8.3	0.93	61.6	32.86	5.54	Sandy loam
		4–8	16.3	8.6	0.89	32.88	47.14	19.98	Coarse loamy
		8–12	21.7	8.6	0.75	18.64	50.56	30.8	Clay loam
		12–16	13.5	8.6	1.26	49.88	43.78	6.34	Coarse loamy
		16–20	16.8	8.7	0.81	45.88	34.94	19.18	Loamy
		20–24	12.9	8.5	1.08	37.4	39.12	23.48	Fine loamy
Postmonsoon	Canning	0–4	14.8	6.7	0.68	20.88	30.54	48.58	Clayey fine
		4–8	9.9	6.7	0.46	1.16	51.58	47.46	Silty clay
		8–12	20.4	6.6	0.56	1.88	39.42	58.7	Clayey fine
Premonsoon		0–4	9.8	8.3	0.79	0.6	29.04	70.36	Clayey very fine
		4–8	61	8.2	0.71	0.2	38.62	61.18	Clayey very fine
		8–12	15.9	7.7	0.67	0.6	50.04	49.36	Silty clay
		12–16	15.1	7.9	0.65	0.6	48.14	51.26	Silty clay
		16–20	16	8.3	0.71	0.64	53.06	46.3	Silty clay
		20–24	15.4	8.3	0.62	0.56	66.5	32.94	Silty clay loam
		24–28	16.6	8.4	0.65	1.48	57.52	41.0	Silty clay loam
		28–32	16	8.3	0.72	2.48	66.58	30.94	Fine silty
		32–36	535.1	8.2	0.81	0.72	30.92	68.36	Clayey very fine
		36–40	18.8	8.2	0.76	0.28	44.44	55.28	Silty clay
Postmonsoon	Dhamakhali	0–4	12.7	7.2	0.48	25.92	38.28	35.8	Clayey fine
		4–8	10.2	7.4	0.25	26.56	32.3	41.14	Clayey fine
		8–12	14.5	7.2	0.46	20.88	30.54	48.58	Clayey fine
		12–16	30	7.8	0.47	1.16	51.58	47.46	Silty clay
Premonsoon		0–4	13.2	8.1	0.78	15.12	73.0	11.88	Coarse loamy
		4–8	11.4	7.8	0.76	15.72	47.86	36.42	Clay loam
		8–12	9.9	8.0	0.82	6.64	66.64	26.72	Fine silty
		12–16	12.6	7.7	0.82	24.04	64.38	11.58	Coarse loamy
		16–20	11.3	7.8	0.93	12.76	34.8	52.44	Clayey fine
		20–24	13	8.1	0.83	9.16	26.92	63.92	Clayey very fine

season. This is partly due to the oxidation of FeS_2 and FeS to SO_4^{2-} and partly results from the decomposition of mangrove litter and hydrolysis of tannin in mangrove plants releasing various kinds of organic acids (Liao 1990). The sediment core samples show a variable admixture of sand, silt and clay with an overall size range from sandy loamy to clayey very fine. The low organic carbon values (0.25–1.26%) obtained might be the result of marine sedimentation and mixing processes at the sediment water interface where the rate of delivery as well as rates of degradation by microbial-mediated processes can be high (Canuel and Martens 1993). This is also related with the poor absorbability of organics on negatively charged quartz grains, which predominate in sediments in this estuarine environment (Sarkar et al. 2004).

The vertical distribution of T_{Hg} in 33 core samples at three stations covering post and premonsoon season from Hugli-Matla-Bidyadhari estuarine complex reveals very low to moderate values as shown in Table 1. T_{Hg} values of 33 samples ranged from 9.8 ng g^{-1} to 535.1 ppb (ng g^{-1}). Spatial heterogeneity in T_{Hg} distribution might be attributed to (i) location of the three stations by the side of three rivers of different tidal and geomorphic settings and differences in hydrodynamic regimes related to concerned river discharge, (ii) natural variability associated with physical mixing of the sediments, (iii) variations in sediment particle size (sandy loam to clayey very fine), and (iv) non-homogenous inputs from point and non-point sources of mercury.

An overall consistent pattern of T_{Hg} distribution was observed in sediment core at S_1 taken in postmonsoon season. An increasing trend of T_{Hg} enrichment at the surface/near subsurface layer was observed in the core at S_2 taken in premonsoon season. The highest concentration of T_{Hg} was observed in the same station at the depth of 32–36 cm. This value (535.1 ppb) can be compared with the value 654 ppb recently determined in surface sediment at Canning station, as reported by Sarkar et al. (2004). It has been revealed that mercury forms strong aqueous complexes with dissolved sulphides (e.g., sulphides, polysulfphide and thiols) which can be an important mechanism forming mobilization of mercury from deeper sediments to the overlying water column (Bothner et al. 1980; Wang and Driscoll 1995; Carreon-Martinez et al. 2002). Recently, Canario et al. (2008) interpreted that variation of T_{Hg} concentrations in surface sediments in Upper St. Lawrence River, Canada were strongly influenced by the formation/deposition/retention of organic sulphur compounds in the sediment water interface. Enrichment of T_{Hg} in surface sediments was also observed by Canario et al. (2003, 2005) from Tagus estuary, Portugal. They interpreted that mercury may be mobilized under suboxic conditions and retained in the upper sediment layers mainly associated with Fe/Mn oxyhydroxides, where interactions strength of the binding between mercury and sedimentary phases (silt and clay) also plays an important role (Silva et al. 2003). In addition, bioturbational activities of the benthic macrozoobenthos present in Sunderban mudflat (namely *Ocypode* sp (crab), *Sesarma* sp (crab), *Lumbrinereis notocirrata* and *Mastobranchus indicus* (polychaetes)) produce physiochemical changes (Bhattacharya 2002) that may contribute to the mobilization of Hg from the bottom to the superficial sediments and to the water column (Birkett et al. 2002).

A comparatively higher value of T_{Hg} in all the subsamples of the core was recorded during postmonsoon sampling at S_1 than in the rest two stations. This might be related to the location of the station beside the main stream of the dominant Hugli estuary. Potential sources of Hg may be related to industrial discharges (paper factories, electrical industries etc.), agricultural run off (Hg–containing fungicides) and sewage sludge from the upper stretch of Ganga river (location of multifarious industries is shown in Fig. 1) (Sarkar et al. 1999). In addition, fouling of sediments by frequent dredging activities near the station S_1 occurs to maintain the navigation channel. Regular operation of dredging activities cause high turbidity in this region resulting considerable retention of mercury within the estuarine system as ascertained by Elliott and Griffiths 1986. Other potential factors for T_{Hg} enrichment are natural erosion from a dynamic system, combustion of coal used as energy source in a local scale (Wang et al. 2000; Pacyna and Pacyna 2001) together with extensive use of antifouling paints to safeguard the warves and fishing boats. Similar pattern of mercury enrichment due to natural and anthropogenic sources was also endorsed by Beldowski and Pempkowiak (2007) in marine coastal sediments of Southern Baltic Sea.

The variations of T_{Hg} can also be the result of post depositional diagenetic processes that remobilize the metal from deeper sediments and cause upward migration in the sediment column (Rasumussen 1994). Additionally, sediment resuspension and mixing of higher concentration bottom waters might also act as potential sources of Hg to be brought to the surface (Mason et al. 1999). Transport, burial and diagenesis play a key role in the preservation of historical records for metal contamination (Valette-Silver 1993). Hence for reliable results an excellent knowledge on the environment of deposition is urgently needed and the problem specific to each site is to be taken into consideration. Moreover, changing conditions can change a repository to a source and vice versa.

When mercury concentrations of the present study were compared to the Effects Range Low (ERL) sediment toxic value (150 ppb ($\mu g\ g^{-1}$)) reported by Long and Morgan (1991), it is revealed that the T_{Hg} levels in all core samples are lower than the ERL values and thus have no toxic effects on the biota inhabiting in the sediments, except the value 535.1 at S_2 (32–36 cm).

Possible sediment enrichment of metals was evaluated in terms of the Igeo of Muller (1979). The formula used for the calculation of Igeo is: $\log_2$ (Cn/1.5 Bn), where Cn is the measured content of element "n", and Bn the element's content in "average shale" (Turkian and Wedepohl 1961). The geoaccumulation Index (I_{geo}) in the present work showed very low values (−3.06 to −5.64) indicating that sediments are uncontaminated (Muller 1979), endorsing the previous observation reported from the same area by Chatterjee et al. (2007) from the core sediment samples in Sunderban region.

The interrelationship among the trace metals with silt content of the sediment is observed in many estuarine environments (Panda et al. 2006; Sahu et al. 1998). In the present study a coincidence of elevated mercury in the sediment profiles with the fine-grained clay minerals have been recorded for all the stations. This might be due to greater active surface area of the clay minerals on which metals may absorb. However, relatively high correlation value between T_{Hg} and clay was recorded only at S_1 ($r = 0.95$, $p = <0.00001$, $n = 10$). While the other two stations (S_2 and S_3), the r-values present positive but insignificant ($r = 0.46$, $p = <0.116$ and $r = 0.21$, $p = <0.555$ for S_2 and S_3 respectively). The relationship of T_{Hg} between pH and organic carbon reveals a very complex and anomalous pattern. The insignificant as well as negative relationship between T_{Hg} and organic carbon, as observed in the present study, might be attributed to their intrusion in the estuarine system through diverse sources at different locations. As a result the areas influenced by

industrial mercury may have relatively low organic matter and areas receiving domestic wastewater have high organic matter as emphasized by Zingde and Desai (1981) while studying the mercury contamination in Bombay harbor, western part of India.

To establish the variation of T_{Hg} among the samples analyzed, a one-way analysis of variance (ANOVA) was undertaken using T_{Hg}, stations, depths and seasons as different factors. It has been revealed that the variations of T_{Hg} concentration within the three sampling stations and depth profiles are highly significant ($F = 4.37$; $p < 0.01$ and $F = 462.35$; $p < 0.00001$ respectively) whereas, variations between the two seasons were not significant ($F = 3.87$; $p > 0.05$).

For better illustration of elevated T_{Hg} concentrations in some samples, statistical analysis was performed using box plot and MAD methods. Results are presented in Table 3. Box plot method showed one extreme and three outliers at S_1 during postmonsoon season. Two extremes were found at S_2 at 4–8 cm and 32–36 cm in premonsoon period. At S_3 there was no anomaly by box plot method. MAD method was more sensitive than box plot method and T_{Hg} anomaly was detected at 12–16 cm at S_3 during postmonsoon season besides six positive anomalies detected previously by box plot method.

The results of sorted factor loading scores along with percentage variances and cumulative percentage variances are shown in Tables 2 and 3. It is observed that the four factors derived are showing a total cumulative variance of 50.48% only. Out of which Factor-1 is responsible for 16.13%, which indicate poor response of individual factor towards defining the geochemical responses. However Factor 1 is found to constitute sand and clay as major components, which show strong negative correlation towards each other. This is a common phenomenon in the fluvial deposits. Considering the above observations, Factor 1 may be termed as "sand factor". Factor 2 shows a variance of about 13.84%, where silt has a strong positive loading. It is difficult to predict association of silt and T_{Hg} with the limited analytical result in the present preliminary work. However, association of T_{Hg} may be related to granulometry (Loring 1978). So, this factor can be termed as "silt Factor". Factor 3, which explains 10.39% of total variance, has strong positive loading on pH and organic carbon. This shows a common physicochemical source, may be related with adsorption of T_{Hg} in the sediments. Factor 4 shows a variance of about 10.11% with major positive influence from T_{Hg} and moderate positive influence by clay.

Table 2 Results of factor analysis after Varimax rotation

Variable	Factor1	Factor2	Factor3	Factor4
THg	−0.114	−0.112	0.051	0.985
pH	0.048	0.019	0.963	0.053
oc%	0.278	0.055	0.324	0.052
Sand%	0.970	−0.086	0.049	−0.088
Silt%	0.005	0.995	0.012	−0.096
Clay%	−0.762	−0.609	−0.047	0.134
Variance%	16.133	13.845	10.393	10.107
Cumulative Var%	16.133	29.978	40.371	50.478

The range of T_{Hg} levels in sediments recorded (9.8–535.1 ppb (ng g^{-1})) in the present study, can be compared with reported levels of mercury in the sediments of other Indian rivers, coastal regions, estuaries, etc. (Selli et al. 1973, Renzoni et al. 1973; Baldi et al. 1979; Baldi and Bargagli 1984; Trivedi and Dubey 1978; Zingde and Desai 1981; Subramanian et al. 2003; Shaw et al. 1988) and with some rivers and estuaries from Europe (Kwokal et al. 2002; Frančišković-Bilinski et al. 2003; Frančišković-Bilinski et al. 2005; Frančišković-Bilinski et al. 2006; Frančišković-Bilinski 2007; Frančišković-Bilinski 2008). Concentrations of T_{Hg} determined in the present work resemble to total T_{Hg} concentrations determined in clean estuary of Öre River (Sweden) and of Krka Estuary (Croatia) by Kwokal et al. 2002 and to stream sediments of Kupa River drainage basin (Frančišković-Bilinski et al. 2005).

During Premonsoon a solitary case of abrupt elevated value of 535.1 ng/g in 32–36 cm depth at S_2 was recorded which might be associated to digenetic migration (Spencer 2002).

The preliminary study assessed the T_{Hg} in sediment cores in Sunderban mangrove wetland covering Premonsoon and postmonsoon seasons. Results indicate relatively

Table 3 Sampling locations, season, core depths and Hg anomalies determined by boxplot and MAD methods from 33 samples

Sampling location	Season	Depth (cm)	Positive anomaly (boxplot)		Postive anomaly (MAD)
			Extreme	Outlier	
S1	Postmonsoon	0–4	+		+
		4–8		+	+
		8–12		+	+
		12–16		+	+
S2	Premonsoon	4–8	+		+
		32–36	+		+
S3	Postmonsoon	12–16			+

low levels of T_{Hg} in the sediments and its complex vertical distribution is related to anthropogenic (industrial wastes, combustion of fossil fuel, antifouling paints, dredging, etc) and physical processes (natural erosion, estuarine mixing and resuspension, tidal dynamics and bioturbation processes) in this estuarine system. Enrichment in surface and near surface sediments indicates remobilization of Hg from deeper sediments. A good correlation is seen between T_{Hg} concentration and grain size distribution. T_{Hg} is always high in sediments having greater mud (silt and clay) contents. The vertical distribution of heavy metals in these mudflats is relatively erratic and does not provide a time-integrated record of historical pollutant inputs hence unsuitable in studying pollution trends.

The authors recommend a continuous monitoring of heavy metals considering both biotic and abiotic compartments as this coastal region is considerably under stress due to implementation of dredging, drilling, impact of chemical industries and development of ecotourism resorts. Moreover there is always a change in the positions of the repository and source of pollutants in such dynamic coastal environment.

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