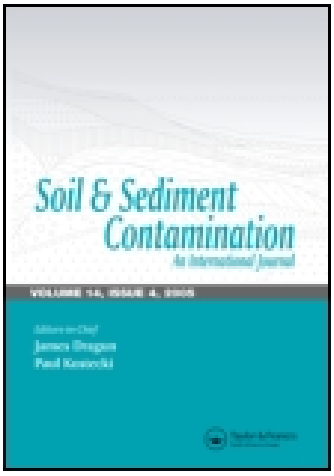

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Mercury Concentration in Sediment Cores from Sundarban Mangrove Wetland, India

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The work presents an extended database ($n = 123$) of total mercury (T_{Hg}) in fine-grained sediment fraction (<63 μm) of core samples in 10 sampling stations of the Sundarban mangrove wetland, India, formed at the estuarine realm of the Hugli (Ganga) River. Results revealed a wide range of spatial, seasonal, and intertidal flat variations of T_{Hg} (7.3 to 93.3 ng/g) with a definite enhancement level at the lower stretch of the estuary, which has extreme mangrove vegetation. An overall enrichment of T_{Hg} in surface/subsurface layers of the core is tentatively explained by remobilization and resuspension of the metal from deeper sediments (36–40 cm). A strong positive correlation was observed between the Hg and clay fraction content of the sediments, while correlations of Hg with organic carbon was poor. Based on the index of geoaccumulation (I_{geo}), enrichment factor (EF), and anthropogenic factor (AF) values, it is suggested that the sediments of Sundarban were found to be less polluted with respect to total mercury. The data reported are a useful baseline for T_{Hg} in Sundarban and would be of importance in future sediment quality studies.

Keywords Mercury, sediment, geoaccumulation index, Sundarban mangrove wetland (India)

1. Introduction

Mercury is one of the most toxic metals for marine fauna with a complex biogeochemical cycle in air, soil, water, and biota. It is listed as a priority pollutant by international agencies in charge of marine environmental protection (Beiras et al., 2002; Ikem and Egiebor, 2005; McAloon and Mason, 2003). Most of the sources of mercury in aquatic ecosystems are the atmosphere, primarily associated with rainfall; however, there are significant sources from mining and industrial processes. Atmospheric deposition contains the three main forms of mercury (Hg^{2+}, Hg^0, or $MeHg^+$), although the majority is inorganic mercury (Beiras et al., 2002; Carreón-Martínez et al., 2002). It was reported that 54% of all anthropogenic emissions of mercury to the atmosphere are from Asia (Jaffe and Strode, 2008). Once

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in surface water, mercury can enter the food chain, or it can be released back to the atmosphere by volatilization (McAloon and Mason, 2003; Marins et al., 1998; During et al., 2009; Rinklebe et al., 2009; 2010). Mercury and other heavy metals are not only transformed or cycled in the aquatic environment, but may be concentrated by factors of millions by aquatic organisms along the food chain (Barnabé and Barnabé-Quet, 2000).

The input of mercury in the littoral sediments and waters is variable in different parts of the world. For example, Minamata disease, the worst outbreak of mercury pollution from an industrial accident, caused the death of many people who had eaten fish with a high concentration of mercury. Remediation work for contaminated Minamata Bay, Japan, was described by Hosokawa (1993). Cossa (1995) estimated that 5% of Hg discharged into the Mediterranean Sea turns up again in the sea fish species, rendering them unfit for human consumption. Of particular importance is the North Adriatic Sea, where the effects of mercury mining regions on quality of stream sediments in Idrija, Slovenia, were studied (Frančišković-Bilinski et al., 2005). The contamination of mercury in the Middle Adriatic Sea was described by Kwokal et al. (2002) and Mikac et al. (2009). According to Barnabé and Barnabé-Quet (2002), the input of mercury to littoral areas of the French River is 2.6-5.6 tons/year and concentration of mercury in mollusks varies from <0.2 tons to >0.4 mg/kg of dry weight in relation to rearing zone. There are also examples of low mercury concentration in estuaries, like Öre River (Sweden) and Krka River (Croatia), described by Kwokal et al. (2002), or of high ones in floodplain soils (Devai et al., 2005).

The Indian Sundarban (Lat. $21^0 32'$ and $22^0 40'$ N; Long $88^0 11'$ and $89^0\ 05'$E), as part of the vast delta (area 9,630 sq.km) formed at the mouth of the Ganges-Brahmaputra system, is a unique bioclimatic zone in a tropical geographical situation in the coastal regions of the Bay of Bengal. It has the potential for being a global biodiversity "hotspot" as it is a reservoir of very rich and diverse faunal and floral communities (Bhattacharya and Sarkar, 2003). The estuarine stretch is the recipient of wastes from urban settlements and a variety of industries including paper and pulp, electrical and pharmaceutical, etc. (as shown in Figure 1). Hydrology of this estuarine system presents a cyclic pattern, characterized by a large amount of monsoonal precipitation and tidal interplay (Bhattacharya, 1988; Sarkar et al., 1985). To date there are only limited data pertaining to T_{Hg} composition of sediment profiles of this estuarine complex (Kwokal et al., 2008; Chatterjee et al., 2009). A considerable number of small-scale industries such as dental amalgam, batteries, thermometer, barometer, electrode, and bulb manufacturing units are present by the sides of the Ganga River upstream, and contribute to a significant amount of mercury in the Sundarban wetland. Hence the present collaborative study has been initiated to extend the mercury database, and to evaluate factors that affect the spatial distribution of T_{Hg} in this estuarine wetland complex (consisting of Hugli-Matla-Bidyadhari tidal river systems) of northeast India.

2. Materials and Methods

2.1. Study Area

Ten sampling stations, designated as Light House (S_1), Kachuberia (S_2), Lot 8 (S_3), Kakdwip (S_4), Gangadharpur (S_5), Jharkhali (S_6), Gosaba (S_7), Canning (S_8), Dhamakhali (S_9), and Gushighata (S_{10}), have been chosen along the lower stretch of the Hugli estuary covering a part of the Indian Sundarban mangrove wetland (Figure 1). Sampling was done covering all three seasons of the year (pre-monsoon, monsoon, and post-monsoon). The

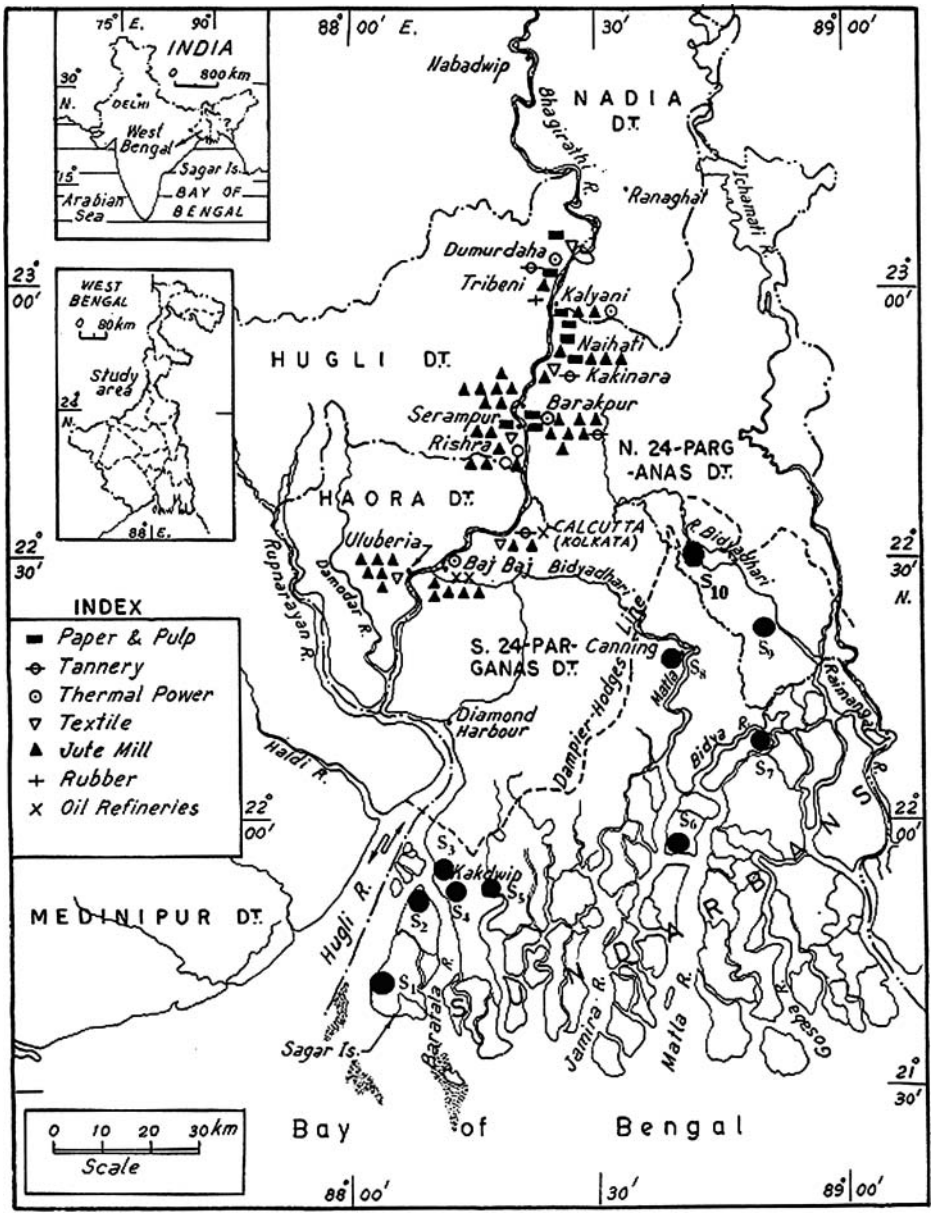


Figure 1. Map of Sundarban showing location of 10 sampling stations.

sampling locations included the upper, middle, and lower intertidal flats having widths ranging from 200-300 m, and showed a wide range of sediment dispersals under a complex drainage network of a tropical meso-macrotidal setting. The studied stations lie at different distances from the sea and thus experience different wave-tide-riverine energy fluxes, various geomorphic situations, and diverse human interference with a variable degree of exposures to heavy metal contamination.

The first station near the Light House (S_1) is situated at the southwestern vertex of Sagar Island and faces the eastern margin of the funnel-mouth Hugli estuary. This station experiences moderate to high waves from the Bay of Bengal and macrotidal amplitude almost throughout the year. As a result, this station is prone to erosion, especially during the episodic tropical cyclones, with a recurrence interval of 3.28 years within 100 km of the island (Bandyopadhyay, 1998). The sandy beach of this station often shows an admixture of coarse and fine particles together with material derived from the breaking down of the brick embankments. The upper intertidal materials are generally more homogeneous in terms of composition and texture compared with that of the lower intertidal zone. The lower intertidal sediments are subject to constant reworking by waves and tidal currents and these show a wide dispersal of sediments. Because of the high-energy situation prevailing in this area, resuspension of sediments' biogenic activity is rather small.

The second station, Kachuberia (S_2), lies at the northernmost tip of Sagar Island at the mouth of the Hugli estuary. The western bank of the tapering end of this island faces the macrotidal Hugli estuary at a point 35 km upstream from the Bay of Bengal. The eastern bank of the island faces the meso-macrotidal Muriganga River, the bifurcating right arm of the Hugli River. Both of the banks near the northern vertex of the island are erosion prone and have been protected by brick paving. Recent tidal sedimentation takes place on the sloping hard substrates, which are exposed during low tides. This station is characterized by a flow separation of the Hugli River against the northern tip of Sagar Island and is naturally protected from direct wave action. The lower and upper intertidal zones of this station have different amounts of sediment accumulation capacity. The accumulated slimy mud of the intertidal zone often slumps downward to form a greater thickness of the bottom mud. Bioturbational activities causing re-suspension of sediments are mainly caused by mud crabs and gobid fishes.

The third station, Lot No. 8 (S_3), is situated at the eastern bank of the Muriganga River about 40 km upstream from its mouth. The flood and ebb tidal currents have distinctive channels with occasional mixing of the two flows at points of major subaqueous shoals. The sediments are dominantly clay and are supposed to be deposited from a "turbidity maxima" (Dyer, 1986) occurring at this stretch of the Hugli estuary. This station experience moderate waves caused by the south summer wind from the seaward side and thus has been given protection by brick pavements. The lower intertidal zone of this locale experiences severe anthropogenic stresses from multifarious commercial activities. The accumulated mud of the upper intertidal zone is apt to be transported downslope by gravitational movements to be accumulated in the lower intertidal region. Re-suspension of sediments by biogenic activities is rather insignificant.

Sediment samples from the fourth station, Kakdwip (S_4), have been collected from the mudflat of the east-west trending Moynapara creek about 1.5 km east of the Muriganga River. The mud flat, in its lower part, contains marshy vegetation with a few saplings of mangroves, and is bioturbated by the burrows of *Sesarma* sp. (crab), *Nereis* sp., and *Dendronereis* sp. (tube-forming polychaete). Deep burrows (3–4 m) with surface mounds at and around the burrow openings are produced by *Thalassina* sp (ghost shrimp) in the upper intertidal to supratidal mudflats.

The mudflat of Gangadharpur (S_5) is situated on the western bank of the wide Saptamukhi River, a major tidal inlet in the Hugli-Matla delta complex. Sediment recycling and water circulation are mostly restricted as the associated creeks have very low flow. Resuspension of sediments by biogenic activities is rather poor as the station belongs to a low energy domain.

The mudflat of Jharkhali (S_6) area occurs in a cove at the junction of the eastern bank of Matla River and northern bank of the Bidya River, a major distributory of the former river system. Vertical sediment movement by bioturbating macro infauna is limited and hence lateral transport of sediment by wave and tides is inhibited to a great extent.

The mudflat of Gosaba (S_7) occurs in the meander bend of the Bidya River, a major distributory of the Matla River. The mudflat is cresentic in outline and forms a point bar on the southern flank of the Bidya River. The diurnal tidal flushing is responsible for the erosion-accretion processes in the point bar. Bioturbational activities of macrobenthic organisms, particularly by crabs, enhance sediment dynamics together with the influence of physical processes.

The sampling locality Canning (S_8) is situated by the side of the upper stretch of the Matla River and belongs to the tidal channel network of the Hugli-Matla system. This station, situated at the lower deltaic plain, experiences semi-diurnal tides and moderate to low wave action. Hydrology of this station represents a cyclic pattern, characterized by flood and ebb flows. The sediments are generally silty clay with subordinate sand fraction. Mud crabs and fiddler crabs generally cause bioturbation and enhance resuspension of sediments.

The Dhamakhali (S_9) mudflat is situated by the side of the Bidyadhari River, a distributory system of the Sundarban mangrove wetland. In the downstream part, it meets the Raimangal River, the drainage channel demarcating the international boundary between India and Bangladesh. It is the north-easternmost station of Sundarban, lying just below the Dampier-Hodges line (the northern most boundary of the Indian Sundarban). Geomorpologically, this station is situated in the inner concave meander bend of Bidyadhari and is fed by the drainage carrying industrial effluents from Kolkata (former Calcutta) metropolitan city. Tidal amplitude and current velocities are rather low. The sediments are mostly silty clay to clayey silty. The substrates are sparsely bioturbated by polychaetes and fiddler crabs like *Uca* sp.

The mudflat of the tenth station Gushighata (S_{10}) occurs by the side of the north-south trending Karati River, a tidal channel upstream of Matla River in its south and Bidyadhari River in its northwest. Fed by the huge discharge of the sewage sludge through the sewage disposal canal of the Kolkata metropolitan area, the sediments of the tidal mudflats of this station are highly affected by pollutants like heavy metals. Some selective polychaetous annelids like *Mastobranchus* sp. and *Dendronereis* sp. are the most common species as they can inhabit the pollution-inflected substrates.

2.2. Sampling and Analyses

Core sediments were collected twice (October-November 2005 and April-June 2006), covering two successive sampling seasons postmonsoon and premonsoon. Cores were collected with the help of a steel corer (40 cm length and 5 cm diameter) by gently pushing into the sediments. The cores were capped and frozen on return to the laboratory. At each station, cores were taken from two transects of the littoral zones (upper and lower) and the upper 40 cm of each core was sliced into 4-cm fractions (subsamples) with the help of a PVC spatula. Prior to sample collection, all the glassware for the collection and storage of sediment samples was thoroughly cleaned with acid (10% HNO_3), and then rinsed in double-distilled (Milli-Q) water before each use.

The core-length size differs between the stations due to variations in the nature of the substratum. Core fractions were stored in labeled polyethylene bags 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 and divided into two aliquots. One aliquot was sieved through a 63μm metallic sieve and stored in hermetically sealed plastic bags until elemental analysis. All visible marine organisms and coarse shell fragments, sea grass leaves and roots when present were removed manually. The other unsieved aliquot was used for sediment quality parameters (organic carbon, pH, percentage of sand, silt, and clay). Organic carbon (C_{org}) content of the soil was determined following a rapid titration method (Walkey and Black, 1934) and pH was determined with the help of a deluxe pH meter (model no. 101E) using a 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 completed using a formulae of Folk and Ward (1957). Total determination of Al was performed by digestion of the sediments with a mixture of acids (HF, HNO_3 and HCl), according to the method described by Rantala and Loring (1975). Aluminum concentrations were determined by Flame-AAS (Perkin-Elmer Analyst 100) using direct aspiration into a N_2O-acetylene flame. For total mercury measurements, sediment samples (about 0.2 g) were decomposed in a closed quartz vessel (pressure 3 bars) with a mixture of 10 ml of nitric and perchloric acids at 140°C. The Cold Vapor Atomic Absorption Spectrometry (CVAAS) method was used for total mercury measurements. The protocol was described by Fitzgerald and Gill (1979) and has been used ever since by numerous workers (see 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 vapor by Elemental Mercury Detector (mercury Monitor TM 3200 by a Thermo Separation Products).

The detection limit of the method for sediment samples is 0.01 ng/g. The reproducibility is 2% for concentrations >200 ng/g and 10% for concentrations >1 ng/g. 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 (ng/g respectively)). Program Statistica (StatSoft, 2001) has been used for all statistical calculations in this work: cluster analysis of R-modality, cluster analysis of Q-modality, correlation analysis, determination of anomalies by boxplot method.

3. Results and Discussion

3.1. Sediment Characteristics

Sediment characteristics such as pH, organic carbon (%), and textural properties (percentages of sand, silt and clay) obtained in different profiles of the 10 sampling stations are shown in Table 1, in which average values are presented.

Sediments characterized by pH varied from slightly acidic to basic (6.6 to 8.8). The acidic nature is mainly pronounced in S_1 and S_2 in the surface and subsurface layer of the lower littoral zone (LL) during the post-monsoon season; in S_5, S_6, S_7, S_8 and S_{10}, the acidic nature was observed only in surface layer. 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). When pyrite is exposed to air and water, it also involves several reactions, as

Table 1

Mean concentrations and standard deviations (mean ± SD) of total mercury (T_{Hg}), Al, pH, organic carbon, and sediment quality parameters, determined at sampling stations S_1–S_{10} (UL = Upper Littoral zone, ML = Mid Littoral zone, LL = Lower Littoral zone)

	T_{Hg} (ng/g)	Al (mg/kg)	pH	OC%	Sand%	Silt%	Clay%
S1/UL/POM	25.8 ± 16.18	119410 ± 26886.2	7.01 ± 0.35	0.44 ± 0.22	19.39 ± 22.35	47.86 ± 22.73	32.75 ± 25.17
S1/LL/POM	12.94 ± 15.74	63490 ± 23079.11	6.83 ± 0.26	0.18 ± 0.15	94.51 ± 3.33	5.48 ± 3.33	0
S2/UL/POM	24.44 ± 3.02	128000 ± 11956.03	7.3 ± 0.26	0.44 ± 0.06	11.80 ± 6.66	40.49 ± 14.96	48.74 ± 11.78
S2/LL/POM	19.17 ± 7.77	80840 ± 26377.23	6.96 ± 0.36	0.35 ± 0.15	54.21 ± 27.76	24.49 ± 16.80	21.28 ± 19.45
S3/UL/POM	27.2 ± 10.85	122400 ± 11979.43	8.17 ± 0.63	0.24 ± 0.05	29.02 ± 19.64	27.92 ± 10.05	43.06 ± 13.42
S3/LL/POM	39.06 ± 4.42	152780 ± 22448.99	8.3 ± 0.15	0.66 ± 0.02	1.20 ± 1.56	36.36 ± 32.51	61.39 ± 31.42
S3/UL/PRM	38.1 ± 6.62	63380 ± 10541.47	8.7 ± 0.10	0.75 ± 0.04	13.24 ± 12.70	45.43 ± 7.70	41.31 ± 6.87
S3/ML/PRM	34.86 ± 9.28	78358 ± 4998.95	8.46 ± 0.21	0.55 ± 0.41	9.25 ± 6.18	56.03 ± 24.99	34.71 ± 22.39
S3/LL/PRM	27.48 ± 8.19	58398 ± 8286.15	8.55 ± 0.13	0.95 ± 0.18	41.04 ± 14.87	41.40 ± 6.96	17.55 ± 9.89
S4/LL/PM	29.67 ± 12.94	32775 ± 4026.06	8.22 ± 0.27	0.58 ± 0.01	9.46 ± 3.46	36.08 ± 24.79	54.46 ± 27.89
S5/LL/M	28.77 ± 4.95	71950 ± 6903.62	6.97 ± 0.33	0.84 ± 0.33	0.40 ± 0.33	44.11 ± 17.40	55.48 ± 17.35
S6/LL/PM	22.56 ± 3.13	55842.86 ± 8796.56	7.27 ± 0.53	0.78 ± 0.06	11.54 ± 2.91	39.08 ± 20.65	49.37 ± 22.23
S7/LL/PM	28.78 ± 7.75	79900 ± 8306.14	7.61 ± 0.56	0.69 ± 0.08	0.93 ± 0.79	49.86 ± 12.51	49.20 ± 12.92
S8/UL/PM	44.62 ± 18.36	77616.5 ± 2294.09	7.45 ± 0.47	0.26 ± 0.18	2.70 ± 1.16	48.10 ± 16.30	49.19 ± 16.20
S8/ML/PM	39.45 ± 5.67	63758.75 ± 7092.63	7.7 ± 0.14	0.82 ± 0.04	7.31 ± 2.33	47.24 ± 13.82	45.44 ± 15.99
S8/LL/PM	47.3 ± 40.05	35951.33 ± 1732.29	6.66 ± 0.05	0.56 ± 0.11	46.98 ± 15.73	38.23 ± 20.79	14.78 ± 7.85
S9/UL/PM	28.9 ± 10.76	69161.4 ± 4784.09	7.88 ± 0.31	0.60 ± 0.11	1.74 ± 0.68	53.16 ± 8.81	45.09 ± 8.57
S9/ML/PM	23.7 ± 4.27	70372.25 ± 3392.35	8.07 ± 0.20	0.51 ± 0.05	3.14 ± 1.44	65.82 ± 17.87	31.04 ± 18.18
S9/LL/PM	21.42 ± 4.04	66943.4 ± 9011.68	7.46 ± 0.27	0.45 ± 0.12	15.28 ± 12.75	38.42 ± 8.27	46.33 ± 8.61
S10/LL/PM	41.6 ± 23.12	66200 ± 5167.39	13.8 ± 0.18	1.82 ± 0.18	21.52 ± 1.83	146.92 ± 8.22	31.56 ± 9.65

described by Stumm and Morgan (1996). More significant is oxidation of sulfide of pyrite to sulfate, which releases dissolved ferrous iron and acidity into the water (Chatterjee et al., 2007). The basic nature was mainly observed at station Lot 8 (S_3) during the post-monsoon and pre-monsoon time, which was also recently recorded by Zuloaga et al. (2009). The sediment core samples show a variable admixture of sand, silt, and clay with an overall size range from coarse loamy to clayey very fine. The absolute dominance of sand in the core sediments at S_1, lower littoral zone (LL), may be referred to its relatively high-energy beach setting influenced by wave and long shore currents. A similar setting of dominance was observed by Chatterjee et al. (2009) at one site of Sundarban, namely Jambu Island. The low C_{org} values (0.46–1.26%) 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).

3.2. Total Mercury (T_{Hg}) in Sediments

The vertical distribution of total mercury in different sediment cores from the Hugli-Matla-Bidyadhari tidal complex at 10 different stations reveals very low values (<93.3 ng/g), as shown in the appendix of Table 1. Comparable concentrations of total mercury (<4–93 ng/g) were recorded by Chatterjee et al. (2009), using a different experimental technique. The prevalent spatial variations in mercury concentration in sediments are very much pronounced, which might be governed by (i) wide variations of geographic-geomorphic situations; (ii) natural variability associated with the physical mixing of the sediments; (iii) differences in hydrodynamic regimes related to river discharge and tidal influx; (iv) changes in sediment particle size characteristics (coarse loamy to clayey very fine); and (v) non-homogenous inputs from point and non-point sources of mercury.

The highest concentration of 535 ng/g was recorded earlier by Kwokal et al. (2008), at the station Canning (S_8) at a depth of 32–36 cm during pre-monsoon. In the present work, the highest concentration of 93.3 ng/g was recorded at the subsurface layer (4–8 cm) at the same site during pre-monsoon. The majority of the core samples reveal enrichment of mercury at the surface/subsurface layers. This may be the result of post-depositional diagenetic processes that remobilize the metal from deeper sediments and cause upward migration in the sediment column (Rasmussen, 1994). Canario et al. (2003; 2005) also found high mercury concentration in the surface sediments of the Tagus estuary, Portugal, and interpreted that Hg may be mobilized under suboxic conditions and retained in the upper sediment layers mainly associated with Fe/Mn oxy-hydroxides. The binding between Hg and sedimentary phases also plays an important role (Silva et al., 2003). Sulphides are also potential binding constituents involved in the cycling of Hg in sediments (Benoit et al., 2003). Like Fe and Mn, the distribution of solid sulphides (such as acid volatile sulphur, AVS) in sediments is influenced by diagenetic processes. Strong correlations of elemental S and sulphides (AVS) with Hg in aquatic sediments suggest the importance of these constituents in influencing the diagenesis of Hg (Fabbri et al., 1991). We did not measure AVS in our sediments and therefore cannot quantitatively examine the significance of sulphides as a controlling factor on Hg variability. Other factors, like the bioturbational activities of macrozoobenthos (*Sesarma* sp; mud-crab, *Ocypode* sp.; ghost crab, *Lumbrinereis notocirata* and *Mastobranchus indicus*; polychaetes) present in the mudflat of Sundarban, which produce physicochemical changes of the substrates (Bhattacharya, 2002), may contribute to the remobilization of Hg from the bottom to the superficial sediments and to the water column (Birkett et al., 2002). The lowest concentrations of T_{Hg} observed in coastal sediments

at Light House (S_1) can be attributed to minimal anthropogenic stresses and the longest distance from the industrial belts of the cities of Kolkata and Howrah located upstream of the Hugli estuary (Figure 1). 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 elaborate knowledge of the environment of deposition is urgent and the problem specific to each site must be taken into consideration.

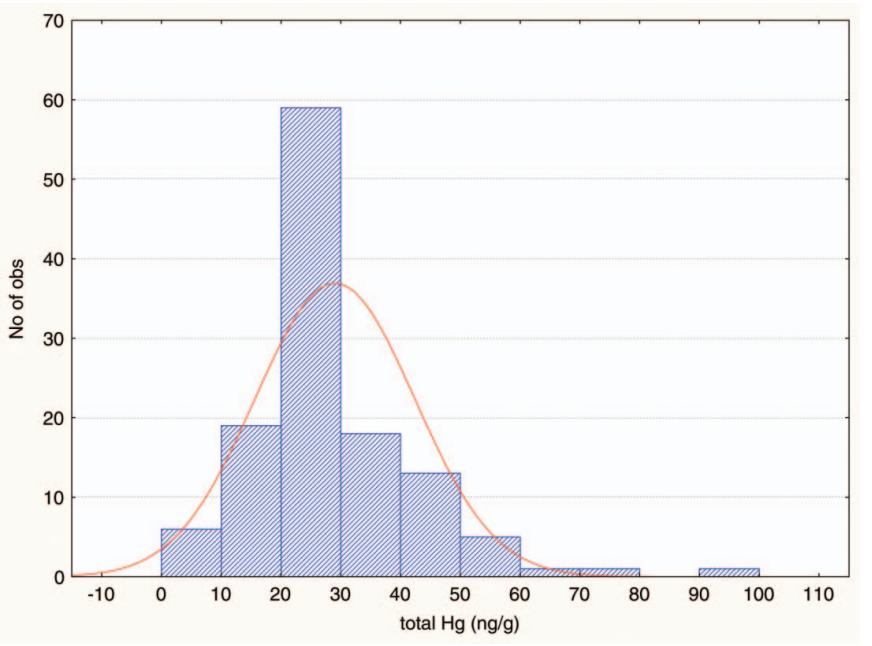
The aluminum (Al) content of sediment that is considered as a good indicator of the amount of fine material present in the samples (Loring, 1991; Din, 1992) varies randomly from 28,900 to 171,900 μg/g (Table 1). The concentration varies with depth and the lowest concentration is observed at the 12–16 cm depth of the core at Kakdwip (S_4) in the lower littoral zone during post-monsoon, while the highest concentration of Al was observed at Lot 8 (S_3) in the lower littoral zone at 4–8 cm depth during post-monsoon, which indicates that the sediment becomes finer at station S_3.

When mercury concentrations of the present study were compared to sediment quality benchmark criteria (Effects Range Low (ERL) and sediment toxic value (0.15 μg^{-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. These data, however, do not support the model of Jaffe and Strode (2008) about the fate and transport of the Asian Hg emissions. It is the GEOS-Chem model, a chemical transport model, which showed that the emissions are associated with high deposition of Hg within south and southeast Asia, including India.

3.3. Statistical Interpretations

Frequency histogram distribution of total Hg in sediments of the Sundarban region and a two-dimensional scatter box diagram obtained by program Statistica are presented in Figures 2a and 2b, respectively. The boxplot gives the interquartile range of the values. The line within the box represents the median value. Circles outside the box indicate two outliers. The asterisk represents an extreme value. In total, three anomalous Hg values were found as follows. One extreme value was found at the Canning (S_8) subsurface layer (4–8 cm), lower littoral (LL) in a post-monsoon period. Two outliers were at Canning (S_8) in the subsurface layer (5–10 cm), upper littoral (UL) in the post-monsoon period, and the other one at Gushighata (S_{10}) at 12–16 cm. Frequency histogram distribution and two-dimensional scatter box diagram for Al in sediments are presented in Figures 3a and 3b, respectively. The histogram shows bimodal distribution. The boxplot method shows three outliers for aluminum at station Lot 8 (S_3) at depths of 0–4, 4–8, and 12–16 cm, in the lower littoral (LL) zone in the post-monsoon period. This finding is in support of the presence of a "turbidity maxima" in this station characterized by dominant clay (Dyer, 1986). The highest concentration of organic carbon was found at S_{10} station (16–20 cm) in the post-monsoon period. Sampling locations and anomalies determined from the total dataset (in the appendix of Table 1) are presented in Table 2. Basic statistical parameters are presented in Table 3. R-modality cluster analysis, which shows the relation between variables (T_{Hg}. Al, pH, OC, sand, silt, and clay), shows linkage only between Al and silt. The correlation between total mercury and other studied components does not show significant correlation coefficients, which indicates some other influence on mercury. Obtained graphs were only described and not presented.

To establish the variation of the total mercury in 20 cores, analysis of variance (ANOVA) was undertaken using mercury concentration as variables, and stations and depth profiles



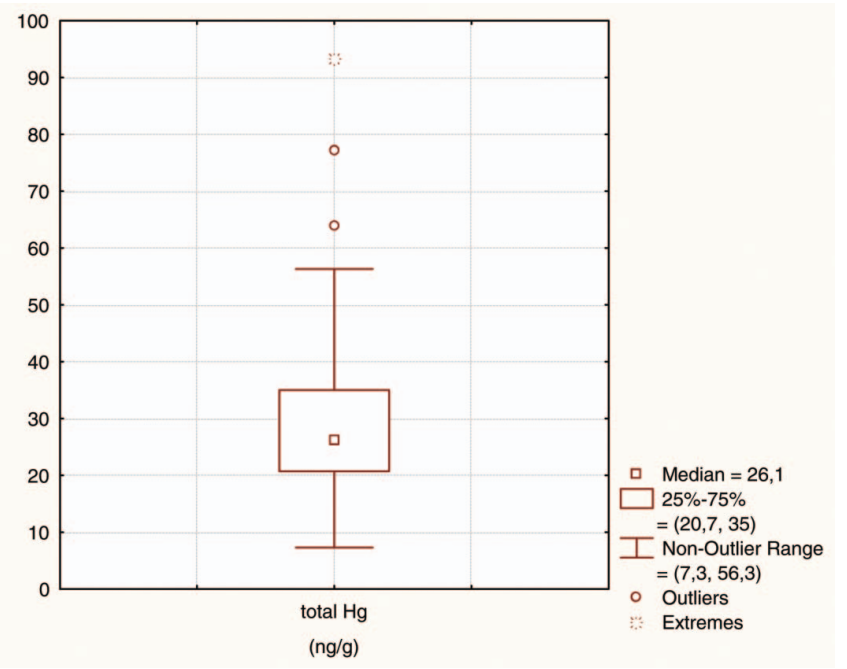


Figure 2. (a) Frequency histogram distribution of total Hg in Sundarban mangrove wetland. (b) Two-dimensional scatter box diagram of total Hg in Sundarban mangrove wetland (Color figure available online).

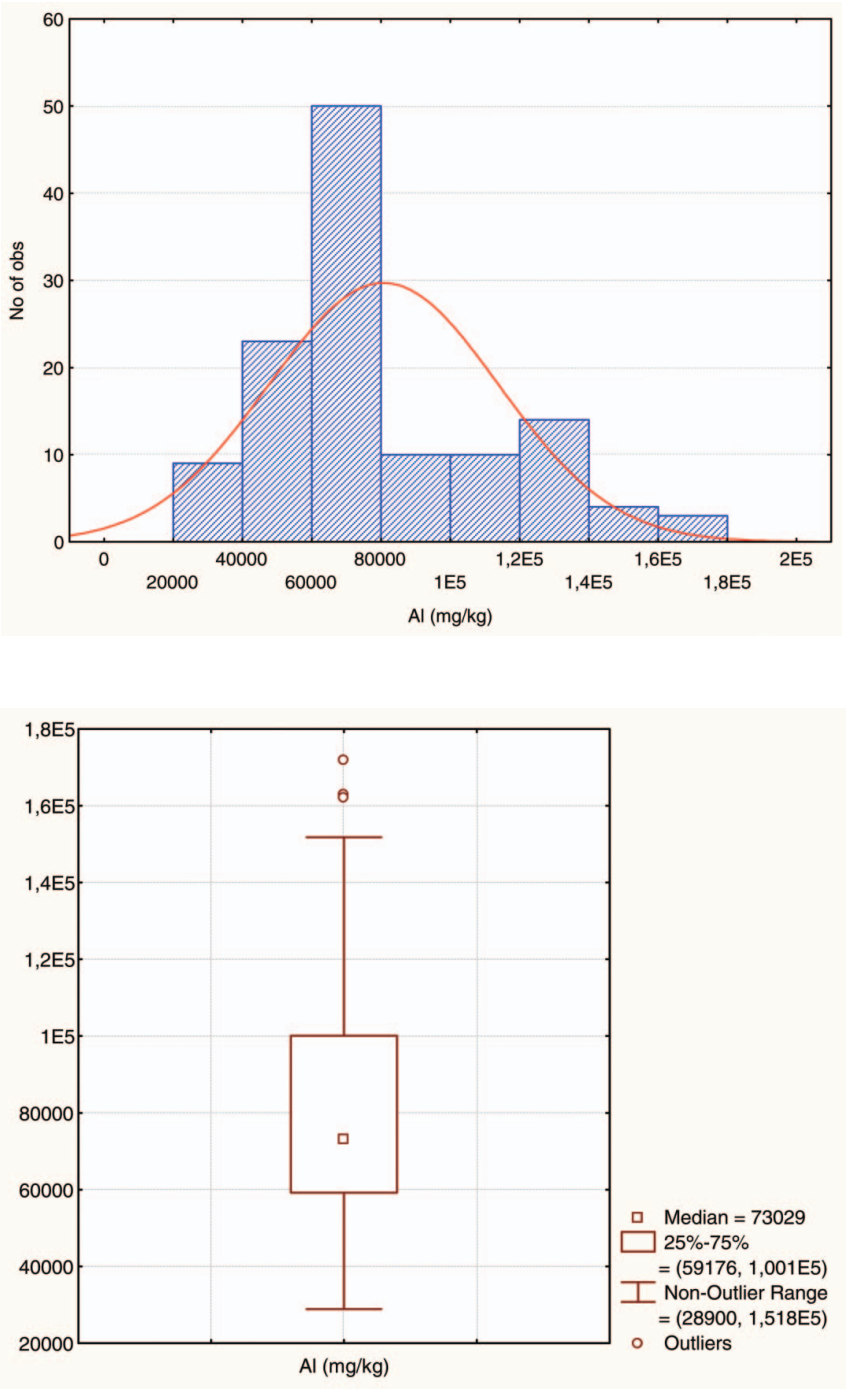


Figure 3. (a) Frequency histogram distribution of Al in the Sundarban mangrove wetland. (b) Two-dimensional scatter box diagram of Al in Sundarban mangrove wetland (Color figure available online).

Table 2
Sampling locations and anomalies determined from the total dataset (PM = Post-monsoon, LL = Lower Littoral zone, UL = Upper Littoral zone)

					Positive anomaly (box plot)	
Location	Sample No.	Depth (cm)	Season	Littoral zone	Extreme	Outlier
S_8	S8/PM	4–8	PM	LL	Hg	
S_8	S8/PM	5–10	PM	UL		Hg
S_{10}	S10/PM	12–16	PM	LL		Hg
S_{10}	S10/PM	16–20	PM	LL		OC
S_3	S3/PM	0–4	PM	LL		Al
S_3	S3/PM	4–8	PM	LL		Al
S_3	S3/PM	12–16	PM	LL		Al
S_1	S1/PM	0–4	PM	LL		Sand
S_1	S1/PM	4–8	PM	LL		Sand
S_1	S1/PM	8–12	PM	LL		Sand
S_1	S1/PM	12–16	PM	LL		Sand
S_1	S1/PM	16–20	PM	LL		Sand
S_1	S1/PM	20–24	PM	LL		Sand
S_1	S1/PM	24–28	PM	LL		Sand
S_1	S1/PM	28–32	PM	LL		Sand
S_1	S1/PM	32–36	PM	LL		Sand
S_1	S1/PM	36–40	PM	LL		Sand

Note: pH, silt, and clay do not show anomalies.

as different factors. Significant value was obtained only for stations as variables ($F = 1.83$; $p < 0.02$) and insignificant for depth profiles ($F = 0.32$; $p < 0.97$) and seasons ($F = 1.39$; $p < 0.5$). Correlation values (r) between T_{Hg}, organic carbon, pH, percentage of sand, silt, and clay reveal an overall insignificant picture in the majority of the cases. Birkett et al. (2002) indicated that organic matter is not a major factor to control mercury distribution; instead, other factors like macroinvertebrates might be important. In aquatic systems, the macroinvertebrates, such as mussels, cockles, polychaetes, and sea anemones present in the mudflat of the estuarine environment, may produce physicochemical changes that may contribute to the mobilization of mercury from the bottom to the superficial sediment and to the water column (Birkett et al., 2002). The lack of correlation between T_{Hg} and organic carbon suggests the role of other associated factors such as bioturbation, tidal flushing, and temperature. A moderate to strong positive correlation between T_{Hg} and clay fraction of sediments was found in station Lot 8 in the lower littoral zone, ($r = 0.86$, $p < 0.07$), which was observed in other estuarine environments (Panda et al., 1999; Sahu et al., 1998). It can be explained because a greater proportion of finer-sized particles such as clay will increase the ability of sediments to retain Hg through adsorption and ion exchange mechanisms because of the high cation exchange capacity, large surface area, and surface charge in these grain-size fractions (Anderson, 1979; Williams et al., 1994; Cundy et al., 1997). Particle size indirectly reflects the hydrodynamic environment; therefore, the observed relationship between particle size (here clay) and T_{Hg} levels is a key in understanding Hg

Table 3
Basic statistical parameters for Grain size, Total mercury and Aluminum

	Mean	Geometric	Median	Minimum	Maximum	Variance	Std. Dev.	Skewness	Kurtosis
T_{Hg} (ng/g)	28.80	26.20	26.10	7.30	93.3	$1.767188E+02$	13.29	1.703063	4.94532
Al (mg/kg)	80307.19	74037.39	73029	28900.00	171900.0	$1.091661E+09$	33040.29	0.827579	−0.04646
pH	7.54	7.51	7.50	6.40	8.8	$4.694655E-01$	0.69	0.215572	−1.07686
OC%	0.58	0.48	0.59	0.03	1.4	$8.272399E-02$	0.29	0.238799	0.03794
Sand%	22.45	7.39	9.36	0.16	99.4	$8.469778E+02$	29.10	1.527923	1.17256
Silt%	40.63	32.53	39.12	0.56	94.4	$4.430004E+02$	21.05	0.257965	−0.22305
Clay%	36.96	—	38.92	0.00	77.8	$5.158899E+02$	22.71	−0.165182	−1.10977

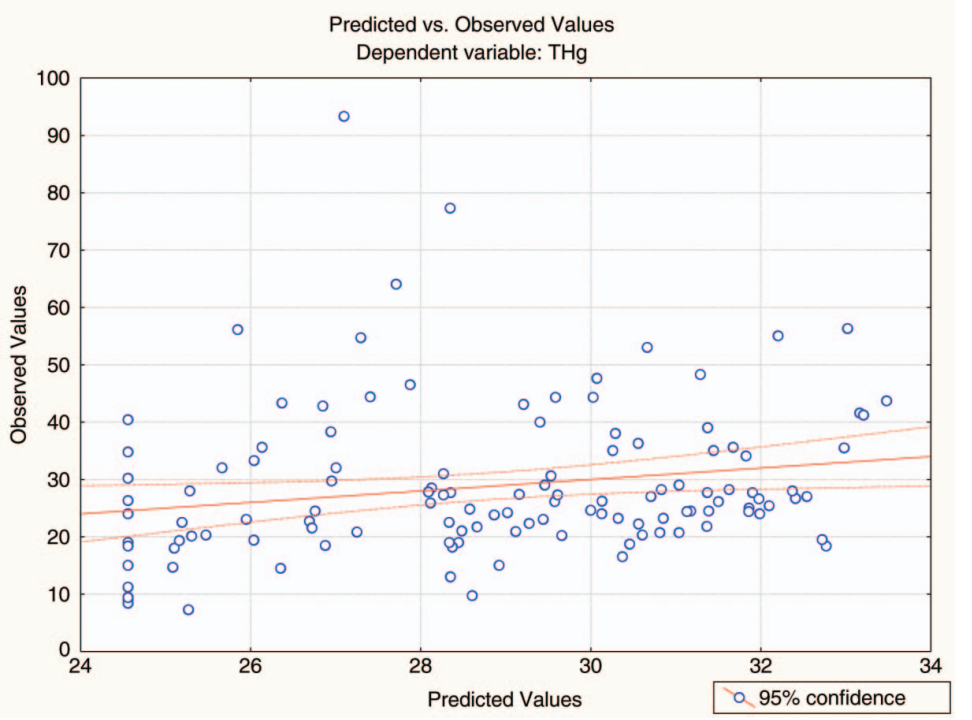


Figure 4. Regression line for correlation of Total mercury (T_{Hg}) with clay%. The broken lines represent the 95% confidence interval for all the data (Color figure available online).

distribution in sediments. Regression line (predicted vs. observed) of T_{Hg} with clay% is shown in Figure 4. The broken lines represent the 95% confidence interval for all the data. There is a good fit between T_{Hg} and clay%; i.e., a good linear relationship between the two parameters. The graph shows that the individual points lie within 90% confidence level, except for a few cases.

3.4. Sediment Quality Tests

Index of Geoaccumulation (I_{geo}), Enrichment Factor (EF) and Anthropogenic Factor (AF). Possible sediment enrichment of metals was evaluated in terms of the I_{geo} presented by Muller (1979). The formula used for the calculation of I_{geo} 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" (Turekain and Wedepohl, 1961). The geoaccumulation Index (I_{geo}) in the present work showed very low values (−3.674 to −4.94), indicating that sediments are uncontaminated (Muller, 1979), endorsing the previous observation reported from the same area by Kwokal et al. (2008) and by Chatterjee et al. (2009) from other sediment samples of the Sundarban region. To distinguish elements originating from human activities between those from natural weathering is an essential part of geochemical studies. One such technique largely applied is "normalization", i.e., metal concentrations were normalized to a textural or compositional characteristic of sediments. Normalizing elements in relation to Al is widely used to compensate for variations in both grain size and composition, since it represents

the bulk quantity of aluminosilicates, which is the predominant carrier phase for adsorbed elements in coastal sediments (Chatterjee et al., 2007). According to Nolting et al. (1999), this method is also a powerful tool for the regional comparison of trace metal content in sediments and can be applied to determine enrichment factors (EF). The values of EFs can be obtained using the equation $EF = (metal/Aluminum_{sediment})/(metal/Aluminum_{shale})$. EFs close to 1 indicate crustal origin, whereas those greater than 10 are considered to be non-crustal sources (Nolting et al., 1999). The minimum EFs obtained for many elements are lower than 1, implying that these elements are depleted in some of the phases relative to crustal abundance in the study area. However, it is evident that elements with a mean EF value greater than 1 reveal sediment contamination. As evident in Table 4, overall lower EF values for most of Hg in most of the sampling areas and sampling in different seasons suggest that mercury is crustal in origin.

Table 4 also shows the values of anthropogenic factors (AF = Cs/Cd, where Cs and Cd refer to the concentrations of the elements in the surface sediments and at the deepest part in the sediment column, respectively) calculated according to Szefer et al. (1998). These values show irregular distribution within the sediment column. Anomalous concentrations of Hg (Table 2) are up to three times higher than the mean Hg value presented in Table 3. It can be hypothesized that they present anthropogenic contamination.

Table 4
Values of Anthropogenic Factors (AF) and Enrichment Factors (EF) in the studied sediment cores (PrM = Premonsoon, PM = Postmonsoon, LL = Lower Littoral Zone, ML = Mid Littoral Zone, UL = Upper Littoral Zone)

Core Names	AF	EF
S_1/UL PM	2.01	0.33
S_1/LL PM	0.31	0.40
S_2/UL PM	1.24	0.47
S_2/LL PM	0.38	0.67
S_3/UL Pr M	0.95	1.22
S_3/ML Pr M	1	1.09
S_3/LL Pr M	1.08	1.03
S_3/UL PM	0.92	0.57
S_3/LL PM	1.84	0.62
S_4/LL PM	0.75	2.03
S_5 /LL PM	0.78	1.03
S_6 LL PM	1.16	1.08
S_7 LL PM	1.18	0.94
S_8/UL PM	1.56	1.23
S_8/ML PM	1.27	1.86
S_8/LL PM	6.43	6.92
S_9/UL PM	1.10	1.06
S_9/ML PM	1.53	0.84
S_9/LL PM	0.45	0.86
S_{10} LL PM	0.79	2.75

Table 5
Comparison of literature concentration of mercury with the present study

Locations	Range of Hg (ng/g)	Reference
Aveiro lagoon (Portugal)	320-343,000	Pereira et al., 1998
Santa Gilla lagoon (Italy)	60-828,000	Degetto et al., 1997
Rivers and estuaries in Guanabara Bay (Brazil)	210-19,450	Mosca, 1980
Numerous rivers and sediments of the Guanabara Bay (Brazil)	50-12,070	FEEMA, 1986
Sao Joao de Meritti River and estuary (Brazil)	320-3380	Barracas and Wasserman, 1998
Sediment core from Guapimirim (Brazil)	125-219	Wasserman et al., 2000
Average shale of Earth's crust	400	Turkian and Wedepohl, 1961
Sediment cores from Sundarban	7.3-93.3	present study

Table 5 is a summary comparison between ranges of concentration reported in the literature and in the present work. It can be observed that the present study reports the narrowest range of concentration in comparison to other studies in the world.

4. Conclusions

The large mercury database in the sediment profiles of the Sundarban mud-flat demonstrates low levels of mercury in the sediments. The observed anomalous and complex vertical distribution of T_{Hg} is related to anthropogenic activities (industrial wastes, combustion of fossil fuel, uses of antifouling paints, dredging activities, etc.) as well as physical and biogenic processes (like natural erosion, estuarine mixing and re-suspension, bioturbation, etc.) in this estuarine system. Enrichment in surface and subsurface sediments indicates remobilization of mercury from deeper sediments. The vertical distribution of the metal in the mudflats of the Sundarban wetland is relatively erratic and does not provide a time-integrated record of historical pollutant inputs, hence it is unsuitable in studying pollution trends. With rapid development of the electronic industry in West Bengal, a large number of outdated electronic products have been discarded into the environment and these "e-wastes" might cause serious mercury pollution in the Sundarban wetland in the near future. Quantifying storage of Hg in wetland sediments provides an opportunity to assign ecological value to the ability of a wetland to act as a sink for this metal; i.e., "ecosystem service" (Costanza et al., 1997). By valuing a wetland in economic terms, fair weight can be given to these systems in decision-making processes, such as wetland restoration, as an important service provided for the human population.

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