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Environmental controls on the speciation and distribution of mercury in surface sediments of a tropical estuary, India

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Abstract:

Distribution and speciation of mercury (Hg) in the sediments from a tropical estuary (Godavari estuary) was influenced by the changing physico-chemical parameters of the overlying water column. The sediments from the upstream and downstream of the estuary were uncontaminated but the sediments from the middle of the estuary were contaminated by Hg. The concentrations of Hg became considerably less during the monsoon and post monsoon period. Total Hg concentrations and its speciation (at the middle of the estuary) were dependent on the salinity of the overlying water column. However, salinity had little or no effect on Hg association with organic phases in the sediments at downstream. Increasing pH of the overlying water column corresponded with an increase in the total Hg content in the sediments. Total organic carbon in sediments played an important role in controlling Hg partitioning in the system. Uncomplexed Hg binding ligands were available in the sediments.

Key words: Hg distribution, Hg-sediment interactions, Salinity and Hg, River discharge and Hg, Intra-annual variation of Hg, Hg-TOC interactions

Introduction:

Mercury (Hg), a widely distributed toxic metal, has been considered as a high priority pollutant because of its persistence in environments and high toxicity to organisms (Merritt and Amirbahman, 2009; Wallace et al., 1982; Zahir et al., 2005).

Most anthropogenic Hg are released to air and water as by-products of various industrial processes (including coal combustion, fossil fuel combustion, mercury vapour lights and chloroalkali production) (Masekoameng et al., 2010; Pacyna et al., 2006; Pacyna et al., 2006; Streets et al., 2005). Strong affinity of Hg to natural particles helps this toxic metal to ultimately settle down and accumulate in sediments (Chakraborty et al., 2014a, 2014b, 2014c, 2014d, 2012a). Thus, sediments act as a major sink for Hg. However, it can be a potential source of Hg in aquatic environments (Randall and Chattopadhyay, 2013; Wang et al., 2012).

Changes in distribution and speciation of Hg in coastal/estuarine sediment can alter bioavailability of Hg in a system. Study of distributions and speciation of Hg in coastal/estuarine sediments has already received worldwide attention. However, not much study has been reported from the coastal/estuarine systems of India.

It is important to understand and identify different factors, which control the distribution and speciation of Hg in a tropical estuarine/coastal system. It has been reported that speciation of Hg in estuarine sediments changes significantly with salinity of the overlying water column (Bratkič et al., 2013; Du Laing et al., 2009; Noh et al., 2013). Hg (weakly associated with sediment) is likely to complex with chloride of the overlying water column and probably decreases total Hg content in sediments. Hg-chloro complexation increases the mobility of Hg in an estuarine system. Salinity intrusion is also often accompanied by formation of high turbidity zone. In a high turbidity zone, the quality and quantity of suspended particulate matter (SPM) also influence redistribution of Hg between water, SPM and sediment (Tseng et al., 2001). Increasing association of Hg with sediments has also been reported via “salt out” effects in estuary (Covelli et al., 2009; Guédron et al., 2012; Noh et al., 2013). pH of overlying water column has also been reported to have impact on the total Hg content in sediment (Hung et al., 2009; Schuhmacher et al., 1993).

Total organic carbon (TOC) (in sediment) and dissolved organic matter (DOM) (in water column) has been reported to play important roles in controlling the mercury sequestration process by marine sediments (Chakraborty et al. 2014a). Increasing concentrations of DOM in overlying water column increases complexation/reduction processes of Hg^{2+} within the water column and decrease the process of Hg sequestration by sediments. There have been other reports in the literature to

understand the impact of each biogeochemical parameter on Hg distribution and speciation under laboratory condition. This is the first attempt to understand the impact of varying physico-chemical parameters of the overlying watermass in regulating distribution, mobility and speciation of Hg in tropical estuarine sediments.

Study area and sampling

Study area

The Godavari estuarine system was chosen to understand the impact of different physicochemical parameters of overlying water column on Hg speciation in the bottom sediments. This estuarine system is located around 16°15'N and 82°5'E covering an area of 330 km². The Godavari River is one of the major rivers in India with a basin area of 3.1×10^5 km². It has been reported that sediment of this river are enriched with heavy metals. The high concentration of metals is probably because of the distribution of different types of rock in the main Godavari basin. The nature of the bed rock and the percentage of each rock type have been reported as: 20.1% of granites and hard rock; 19.2% of sedimentary rock; 55.7% of Deccan trap and 5.1% of recent alluvium. Tributaries of Godavari River which drain through Deccan traps have shown relatively higher concentrations of heavy metals compared to those tributaries which flow through granite rock. All these tributaries drain water from high rainfall areas. These tributaries contribute 80% flow of the total river. The river flow in the estuary is regulated by a dam at Dowleswaram. Monsoon derived freshwater is stored in this dam reservoir and conserved for utilization during the dry seasons by the Irrigation Department of Andhra Pradesh. With the onset of summer monsoon runoff, the dam reservoir is first filled and the surplus water is released downstream. Downstream of the dam, the Godavari River is naturally divided into Gautami and Vasishta distributaries that form respective estuarine systems (Figure 1) (Chakraborty et al 2012a). This study was conducted at Gautami Godavari estuary as it carries the major river discharge. The sampling was conducted from February 2010 to December 2010. Sediment samples were collected from three environmentally different locations to represent variety of environments (Figure 1). The sampling stations, Kotipalli represents upstream (dominated by freshwater), Yanam represents middle of the estuary and Bhairavapalem represents downstream (dominated by saline water) of the estuary.

Samples collection

A Van Veen stainless steel grab (with an area of 0.02 m²) was used to collect sediment. Without emptying the grab, sediment sample was collected from the center with a polyethylene spoon (acid washed) to avoid contamination by the metallic parts of the dredge. Multiple sampling was done at

each station. Details of sampling procedure has been mentioned in the literature (chakraborty et al., 2012b). The samples were stored at $-20\text{ }^{\circ}\text{C}$ and then dried at $30\text{ }^{\circ}\text{C}$ in a forced air oven (Kadavil Electro Mechanical Industry Pvt Ltd India, Model No. KOMS. 6FD). Sediments were subsequently ground and stored at $4\text{ }^{\circ}\text{C}$ until needed.

Cleaning Procedures for Containers

All containers used were made of Teflon. After cleaning with ultrapure water, all the containers and quartz boats were filled to the top with 10% (v/v) nitric acid and allowed to stand at the room temperature for 1 week. The containers and quartz boats were then rinsed five times with ultrapure water. The containers (acid cleaned) were refilled with ultrapure water every day and allowed to stand until they were used. Quartz boats were heated to 800°C prior to Hg analysis.

Sequential extraction protocol for Hg speciation

The sequential extraction protocols proposed by the European Community Bureau of Reference, usually called the BCR method (Ure et al., 1992) was used to understand the distribution of Hg in the different phases of the estuarine sediments. This protocol allows us to determine the sum of ion-exchangeable, water soluble and carbonate/bicarbonate form of Hg (Fr. 1); Hg bound to Fe–Mn oxyhydroxide i.e., reducible fraction of Hg (Fr. 2); concentrations of Hg associated with organic matter and sulfide, i.e., oxidizable fraction of Hg (Fr. 3) and residual Hg fraction (Fr. 4) in sediment. All the extraction processes were performed in Teflon containers. The reagents used in this study were of analytical grade or better (ultrapure). The protocol has been vividly described in the literature (Chakraborty et al., 2014a, 2014b, 2014c). The concentrations of Hg in each extracted solutions and in residual fractions were determined by direct mercury analyzer. All the extractions were done in triplicate. Two reagent blanks were analyzed for every extraction. In all cases blank results were below the detection limit of the analytical technique (0.0015 ng).

Determination of Hg in estuarine sediments by Direct Mercury Analyzer:

Mercury in the estuarine sediments was analyzed by Direct Mercury Analyzer (Tri cell DMA-80) from Milestone, Italy. The DMA-80 combines the techniques of thermal decomposition, catalytic conversion, amalgamation, and atomic absorption spectrophotometry. Controlled heating stages are implemented to first dry and then thermally decompose a sediment sample introduced into a quartz tube. A continuous flow of oxygen (99.9% pure) carries the decomposition products through a catalyst bed where interferences are trapped. All mercury species are reduced to elemental Hg and are then carried along to a gold amalgamator where the mercury is selectively trapped. The system is purged and the amalgamator is subsequently heated which releases all Hg vapours to the single

beam, fixed wavelength atomic absorption spectrophotometer. Here, absorbance measured at 253.7 nm is proportional to mercury content in the sample. The DMA-80 is fully compliant with US EPA method 7473.

Results and discussion:

Distribution of mercury at three locations in the Godavari estuary

Total Hg concentrations in the sediments and its variation at the three different locations (upstream, mid of the estuary and downstream) in the Godavari estuary are presented in Table 1. The highest concentration of Hg (ranging from ~ 44 to 276 $\mu\text{g.kg}^{-1}$) was found in the sediments collected at Yanam (middle of the estuary) followed by the sediments collected at Kotipalli (upstream). The concentrations of Hg in the sediments collected at Kotipalli were found to vary from 36.9 to 188.7 $\mu\text{g.kg}^{-1}$. The lowest concentration of Hg was found in the sediments collected at Bhairavapalem (downstream) (the concentrations of Hg in the sediment varied from 20.9 to 101.7 $\mu\text{g.kg}^{-1}$).

In estuarine system, complexation of Hg with dissolved organic matter, coupled with colloidal coagulation, has been reported to increase total Hg content in sediment at middle of the estuary (Chakraborty et al., 2012a; Foxa, 1983). The highest concentration of Hg in the sediments, collected at the middle of the Godavari estuary, was probably also due to coagulation of Hg associated suspended particulate matter (SPM) at the middle of the estuary (saline and fresh water mixing predominates at Yanam). The lowest concentration of Hg in the sediments at Bhairavapalem (downstream) depends on number of processes of the estuary.

- (1) Mixing of metal-rich fluvial sediments and metal-poor marine sediments was probably one reason for the decrease in the total Hg content in the sediments at Bhairavapalem.
- (2) It has also been reported that Cl^{-1} ion in overlying water column can form complex with Hg and can extract loosely bound Hg from sediments. Thus, this process may decrease the total concentrations of Hg in sediments. This was probably another reason for the low Hg content in the sediments at Bhairavapalem.
- (3) NaCl, likely through the action of Na^{+} , decreases inter-particle attraction that results in the release of particles and colloids from the sediment bed, along with particle-sorbed Hg (Chakraborty et al., 2012a). This was another possibility for the decreasing total mercury content in the studied sediments collected at Bhairavapalem.

The concentrations of Hg in the estuarine sediments were evaluated by following the sediment quality guideline (ERL/ERM) (Long and Morgan, 1990; Long et al., 1995; Long and Wilson, 1997;

Long and Macdonald 1998). The sediments collected at the downstream stations were not contaminated with Hg (throughout the year) and were in “good state” (in terms of Hg contamination). However, the sediments collected at the upstream (Kotipalli station) were in “intermediate state” (in terms of Hg concentration) during the pre-monsoon period. However, Hg loading became considerably less (and were in “good state”) during the monsoon and post monsoon period. The sediments collected at the middle of the estuary (Yanam) were found to have highest concentrations of Hg during the pre-monsoon period and were in “intermediate state” (in terms of Hg concentration). However, the concentrations of Hg gradually decreased and the quality of sediment became “good” during the post monsoon period.

The total Hg content in the estuarine sediments at all the three locations in the Godavari estuary is quite comparable with the other reported concentrations of Hg from unpolluted coastal sediments around the world (Conaway et al., 2003; Fran, 2002; Kannan et al., 1998; Kwokal et al., 2012; Leermakers et al., 2001; Magesh et al., 2013; Sankar et al., 2010; Yáñez et al., 2013; Yu et al., 2012).

It is very difficult to understand the impact of individual physico-chemical parameters (pH, Salinity, River discharge, and SPM) on Hg distribution in estuarine sediment samples. This study is based under the assumption that the sources of Hg to the estuary was constant and the amount of Hg that deposited in the estuarine sediments was only affected by the changing physico-chemical parameters of the sediments and the overlying water column.

The impacts of different parameters of overlying water column at three different stations on Hg distributions are discussed below.

Effect of salinity of the overlying water column on the total Hg content in the bottom sediments

Figure 3 shows the variation of total Hg content in the estuarine sediments collected at Yanam (the middle of the estuary) and Bhairavapalem (downstream) under varying salinity of the overlying water column. There was no variation of salinity in the water column at Kotipalli station (upstream) (mostly dominated by freshwater throughout the year). Thus, no impact of salinity in the water column on Hg content in the sediment was observed at the upstream of the Godavari estuary.

The concentrations of Hg in the sediments were found to increase with the increasing salinity of the overlying water column at Yanam and Bhairavapalem. It has been reported that flocculation of metal-humate complexes and desorption of metals from SPM are the two most important processes which affect the partition of metals in sediments and water column. The increasing salinity of the overlying water column (mixing of freshwater and brackish water) might have increased coagulation processes of Hg associated SPM and Hg-humate complexes and increased the total Hg content in the

sediments at Yanam (middle of the estuary) than Bhairavapalem (downstream). High concentration of SPM in water column at Bhairavapalem (downstream) in Godavari estuary (Sarma et al., 2012) might have enhanced metal exchange reaction between sediment and SPM. Mixing of metal-rich fluvial sediments and metal-poor marine sediments was probably another reason for the decrease of Hg content in sediment at Bhairavapalem. It is also possible that Na^+ (from NaCl from the overlying water column) decreased inter-particle attraction in sediments that results in the release of particles and colloids from the sediment bed, along with particle-sorbed Hg (Chakraborty et al., 2010).

Effects of river discharge on the total Hg content in the sediments

The impact of river discharge on the Hg content in the studied sediments at the three sampling locations is presented in Table 1. In general, there is virtually no river discharge between January and May due to drying of the river in the upstream. Usually the discharge starts from June, reached its maximal in August. Discharge is usually low during October-December (Chakraborty et al., 2012a). However, it depends upon the local rainfall. Table 1 shows that the concentrations of the total Hg in the sediments collected at Kotipalli (upstream station) suddenly decreased with the increasing river discharge. Presumably, the excessive inflow of freshwater from Dowleshwaram dam into the estuary would cause the dilution of Hg concentrations. It has been reported that quantity and quality of SPM influences redistribution of Hg between water, SPM and sediment (Tseng et al., 2001). Increase in concentrations of SPM by several orders of magnitude with the water discharge in this estuary probably facilitates exchange reactions between Hg (from the estuarine sediment at Kotipalli) with SPM of the overlying water column and decrease Hg concentration in the sediments. It must be noted that sediment sampling was not possible during the high river discharge during the monsoon time (June–September). The sharp decrease in concentrations of Hg in the sediments collected at Kotipalli and Yanam (from May to October) indicates that the river discharge into the estuary decreased the concentrations of Hg in the estuarine sediments.

Impact of pH of the overlying water column on the total Hg content in the sediments

The literature data shows that mobility of Hg at different pH in estuarine sediments is diverse. Some authors reported that low pH releases Hg from bottom sediments (Durate et al., 1991), while others claim that sorption of mercury on sediment increases at low pH (Schindler et al., 1980). It has been reported that the processes of sorption/desorption of Hg at different pH depends on the nature of Hg species present in the system. A distinct difference in absorption ability was observed among the sediments collected from the three different stations of the Godavari estuary. **Figure 4** shows the variation of total Hg in the estuarine sediments with the changing pH of the overlying water column.

It shows that the increasing pH of the water column increased the total concentrations of Hg in the studied sediments at all the three stations in the Godavari estuary.

This is probably the results of the combination of the following facts 1) formation of new binding sites in sediments at higher pH to binds Hg; 2) desorption of Hg at lower pH and binds with SPM or DOC. This study shows that the pH of the overlying water column in the Godavari estuary did influence Hg partitioning in the estuary.

Further geochemical fractionation study was performed to understand the impact of these physicochemical parameters of the overlying water column on Hg distribution in the different binding phases of the sediments.

Mercury speciation in estuarine sediments by using a modified BCR sequential extraction protocol

Figure 5 shows the distribution of Hg in the different binding phases of the sediments. The concentrations of water soluble, exchangeable and carbonate forms of Hg complexes (Fr. 1) were found to vary from 11.0-21.1% (at Kotipalli), 9.2-18.5% (at Yanam) and 2.1-10.9% (at Bhairavapalem) of the total Hg content in the studied sediments (Table 2). This fraction of Hg-complexes (Fr.1) is expected to release easily from sediments and can increase mobility and bioavailability of Hg in overlying water column. This change in concentrations of Hg as water soluble, exchangeable and carbonate complexes in the sediments is usually dependent on (i) Hg loading in the sediment (ii) concentrations of competing ligand in the water column (SPM and other ligands) (iii) availability of strong binding sites in the sediments.

In this study, relatively high Hg loading and low SPM concentrations (as reported in this location by Sarma et al., 2012) in the overlying water column at Kottipalli station increased the concentrations of Hg as water soluble, exchangeable and carbonate complexes in the sediments. Similar observation was found in the sediment collected at Yanam. Highest Hg loading in the sediments collected in the middle of estuary increased the relative concentrations of Hg in the fraction 1. However, lowest Hg loading, and more SPM concentrations (as reported by Sarma et al., 2012) and high salinity in the overlying water column at Bhairabvapalem probably responsible for the low concentrations Hg as fraction 1 in the sediments.

The concentrations of Hg associated with iron and manganese oxyhydroxide phases (Fr. 2) were found to vary from 8.6-18.2% (at Kotipalli), 11.1-14.3% (at Yanam) and 3.1-13.4% (at Bhairavapalem) of the total Hg content in the studied sediments. It was found that the major fraction of the total Hg was associated with the TOC in all the studied sediments. The concentrations of Hg

associated with organic phases were found to vary from 33–57% (at Kotipalli), 28-45.3% (at Yanam) and 21-36.4% (at Bhairavapalem) of the total Hg.

Figure 6 a, shows that the concentrations of Hg associated with organic phases (Fr. 3) in the sediments collected at Yanam gradually increased with the increasing salinity of the overlying water column. This is because of the fact that complexation of Hg with dissolved organic matter, coupled with colloidal coagulation, increased the total Hg content in the sediments collected at Yanam (middle of the estuary) and thus, the concentration of Hg associated with the organic phases (Fr. 3) in the sediment collected at Yanam gradually increased with the increasing salinity of the overlying water column.

However, the association of Hg with Fe/Mn oxyhydroxide phase gradually decreased in the sediments with increasing salinity of the overlying water column at Yanam. Enough uncomplexed Hg-binding sites were available in the organic phases of the estuarine sediments collected at Yanam and Hg prefers to form thermodynamically more stable complexes with organic phases than Fe/Mn-oxyhydroxide phases in the sediments.

Figure 6 b shows that the concentration of Hg associated with the organic phases (Fr. 3) in the sediment collected at Bhairavapalem (downstream) gradually decreased with the increasing salinity of the overlying water column. However, very low Hg to chloride ratio in this station, variation in salinity is not expected to play significant impact on Hg distribution. The high concentration of SPM (Sarma et al., 2012) in this region was probably responsible decreasing Hg association with organic phases in the sediments.

It has been reported that the processes of sorption/desorption of Hg at different pH depends on the nature of Hg species present in a system. **Figure 7** shows the variation of Hg association with organic (Fr 3) and Fe/Mn oxyhydroxide phases (Fr 2) in the sediment with varying pH. **Figure 7a** shows that the association of Hg gradually increased with the increasing pH of the overlying water column. However, the association of Hg with Fe/Mn oxide phase was found to decrease with pH at Yanam. This probably indicates that Hg prefers to form thermodynamically more stable complexes with organic phases than Fe/Mn-oxide phases in the sediments. However, completely opposite trend was observed in the sediments collected at Bhairavapalem (**Figure 7b**). High SPM in water column and settling of finer particles (mainly Fe/Mn oxyhydroxide) were responsible for the same.

The concentrations of Hg associated with organic phase gradually increased with the increasing Hg/TOC ratio in the estuarine sediments collected at Yanam and Kotipalli. This finding confirms that complexation of Hg with dissolved organic matter, coupled with colloidal coagulation and

becoming an integral component of the sediments, increased total Hg concentrations in the sediments collected at Yanam (middle of the estuary). Enough uncomplexed Hg-binding sites were also available in the organic phases of the estuarine sediments collected at mid of the estuary.

However, a steady decrease in Hg association with the organic phases of the sediments was observed with the increasing Hg/TOC ratio in the sediment collected at the downstream of the estuary. High SPM in the water column and settling of finer particles (mainly Fe/Mn oxide) might have helped Hg to undergo exchangeable reaction and thus, association of Hg with Fr 3 gradually decreased in the sediments collected at Bhairavapalem (downstream).

This study suggests the following:

1. The sediments collected at the Bhairavapalem (downstream) station were not contaminated with Hg (throughout the year) and were in “good state” (in terms of Hg concentration). The sediments collected at the Yanam (the middle of the estuary) were found to have highest concentrations of Hg during the pre-monsoon period and were in “intermediate state”. However, the concentrations of Hg gradually decreased and the condition of sediment became “good” during the post monsoon period at Yanam. However, the sediment collected at the Kotipalli station (upstream) was in “intermediate state” during the pre monsoon period. However, Hg loading become considerably less (and were in “good state”) during the monsoon and post monsoon period.
2. The concentrations of Hg associated with organic phases in the sediments were found to increase with the increasing salinity of the overlying water column at Yanam (at the middle of the estuary). However, the concentrations of Hg associated with the organic phases in the sediments were found to decrease (downstream) with the increasing salinity and SPM of the overlying water column at Bhairavapalem. Upstream (Kotipalli) of the estuary had very little influence of salinity of the overlying water column on Hg distribution in the sediments.
3. Although not statistically significant, an increase in the pH of the overlying water column corresponded with an increase in the content of Hg in the sediments. Increase in concentration of Hg associated with organic phases in the estuarine sediments was observed with the increasing pH of the overlying water column.
4. Taken as a whole, Hg speciation and partitioning in the Godavari estuary were strongly influenced by SPM loads, pH and salinity of the overlying water column and organic carbon in the sediments.

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CAPTION AND LEGENDS FOR FIGURES

Figure 1. Map of the sampling sites along the Godavari estuary (Andhra Pradesh region) of India. The filled circles are the locations of the three sampling sites.

Figure 2. Schematic diagram of the modified BCR protocol for sequential extraction study of metals in estuarine sediments.

Figure 3. Variation of total Hg content in the estuarine sediments with the varying salinity of the overlying water column at Yanam and Bhairavapalem

Figure 4. Variation of total Hg content in the estuarine sediments with the varying pH of the overlying water column at Kotipalli; Yanam and Bhairavapalem

Figure 5. Fractionation of Hg, associated with different solid-phases of the studied sediments (by modified BCR method) collected at three different location (a) Kotipalli (b) Yanam (c)

Bhairavapalem

Figure 6. Impacts of varying salinity on the Hg association with organic (▲) and Fe/Mn-oxyhydroxide binding (■) phases in the sediment collected at (a) Yanam and (b) Bhairavapalem

Figure 7. Impacts of varying pH on the Hg association with organic (▲) and Fe/Mn-oxyhydroxide binding (■) phases in the sediment collected at (a) Kottipalli (b) Yanam and (c) Bhairavapalem

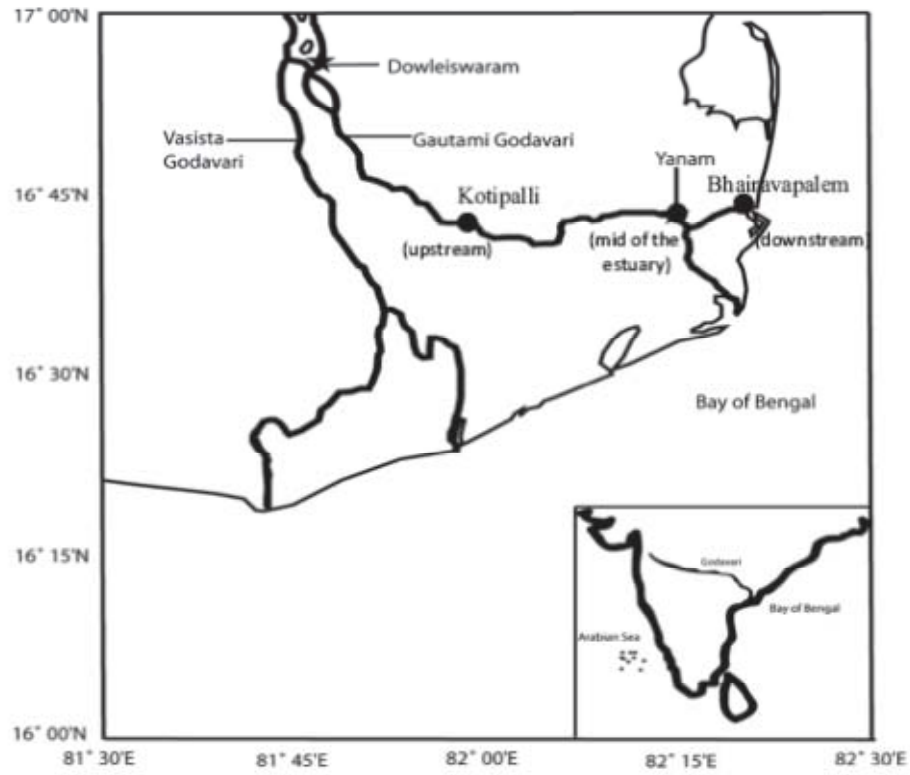


Figure 1

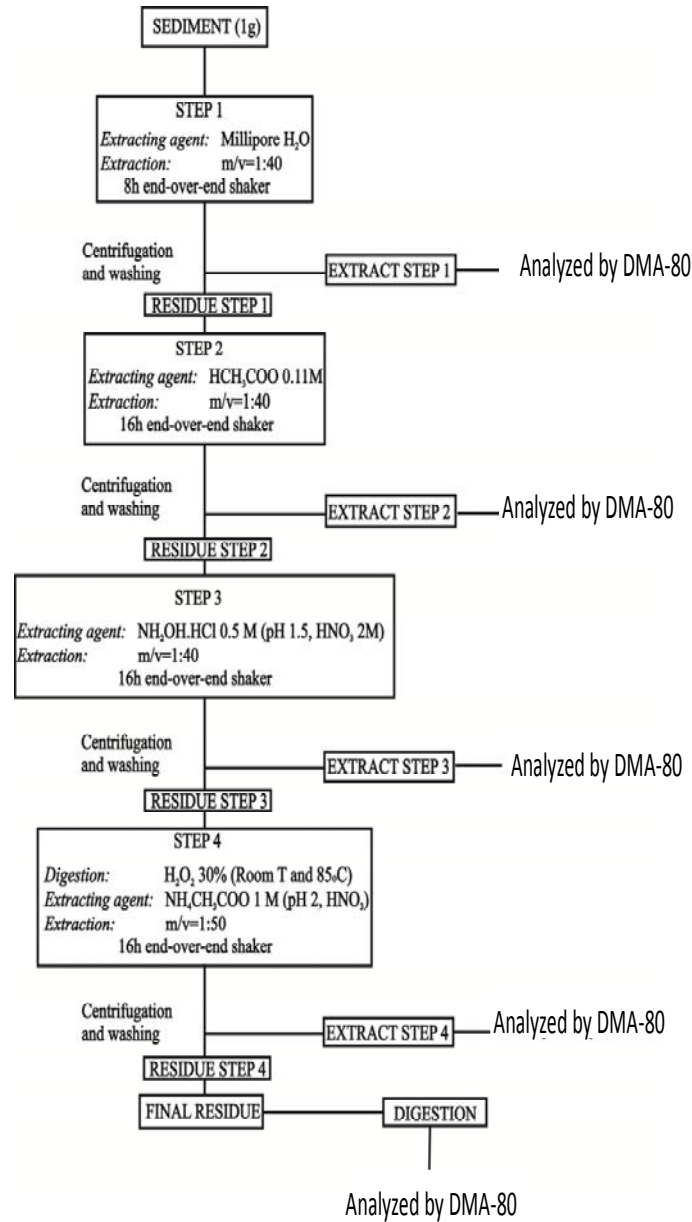


Figure 2

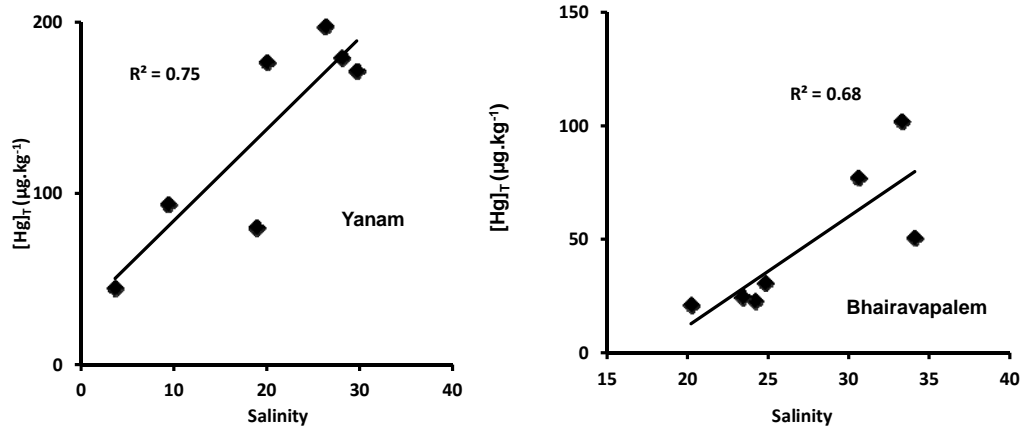


Figure 3

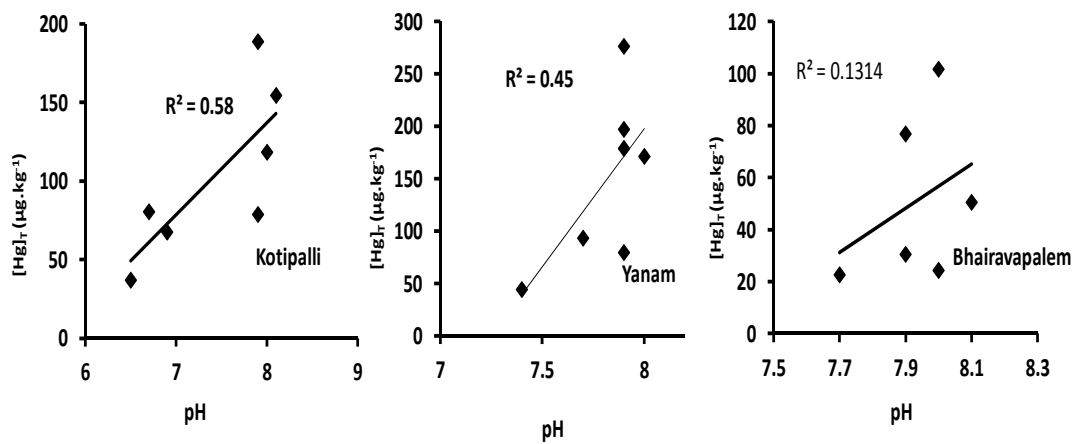


Figure 4

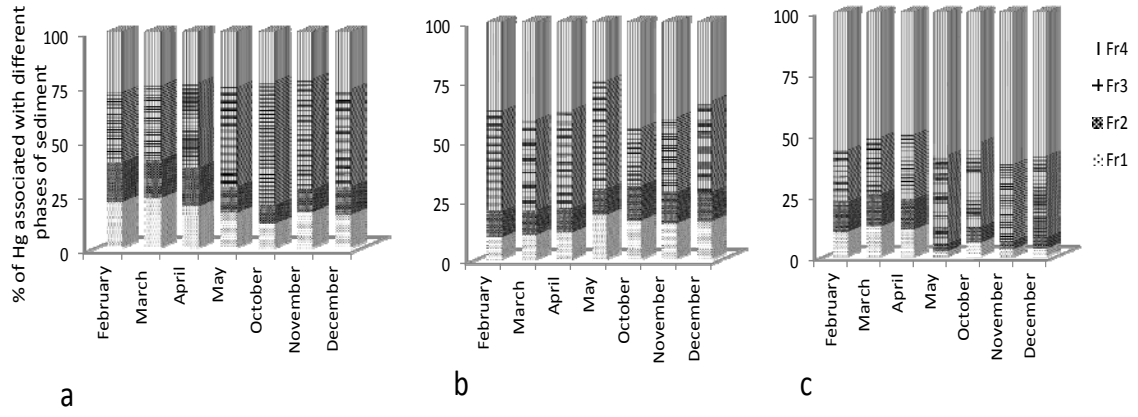


Figure 5

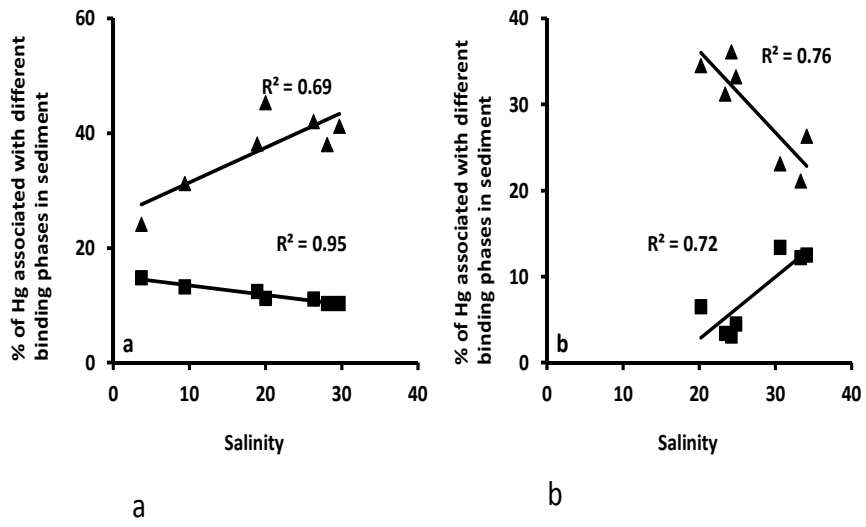


Figure 6

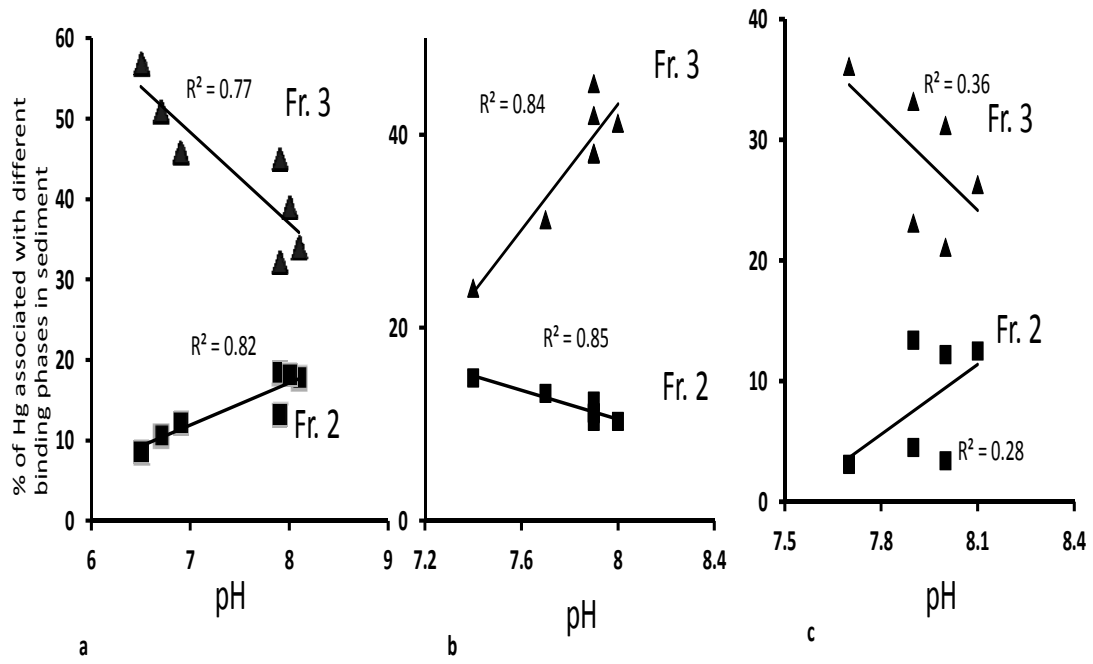


Figure 7

Table 1: Change in the concentrations of mercury ($\mu\text{g.kg}^{-1}$), total organic carbon (TOC) in the sediments and salinity, pH (of the overlying water column) at three different significant sites in Godavari estuary during Jan 10 to Dec 10.

	Date	Total Mercury ($\mu\text{g.kg}^{-1}$)	TOC (%)	Salinity (PSU)	pH	River discharge ($\text{m}^3.\text{s}^{-1}$)	ERL	ERM	State
Kotipalli	10-Feb	188.7	0.16	8.3	7.9	0.0			
	10-Mar	154.5	0.25	9.7	8.1	0.0			
	10-Apr	118.3	0.19	9.6	8.0	0.0			
	10-May	67.5	0.25	0.2	6.9	14.1	150	710	Good
	10-Oct	36.9	0.51	0.1	6.5	818.4			
	10-Nov	80.4	0.47	0.2	6.7	248.3			
	10-Dec	78.7	0.48	1.4	7.9	0.0			
Yanam	10-Feb	197.2	0.25	26.3	7.9	0.0			
	10-Mar	178.9	0.31	28.1	7.9	0.0			
	10-Apr	171.3	0.35	29.7	8.0	0.0			
	10-May	176.4	0.41	20.0	7.9	14.1	150	710	Good
	10-Oct	44.2	0.48	3.7	7.4	818.4			
	10-Nov	93.3	0.38	9.4	7.7	248.3			
	10-Dec	79.5	0.51	18.9	7.9	0.0			
Bhairavapalem	10-Feb	101.7	0.03	33.3	8.0	0.0			
	10-Mar	76.8	0.07	30.6	7.9	0.0			
	10-Apr	50.5	0.06	34.1	8.1	0.0			
	10-May	22.6	0.25	24.2	7.7	14.1	150	710	Good
	10-Oct	20.9	0.13	20.2	-	818.4			
	10-Nov	24.3	0.16	23.4	8.0	248.3			
	10-Dec	30.5	0.25	24.8	7.9	0.0			

Table 2. Sequentially extractable non-residual and residual Hg species in the estuarine sediments collected from three different locations of Godavari estuary.

Sampling location	Sampling time	Fr 1(%)	Fr 2(%)	(%)Fr 3	Fr 4 (%)
Kotipalli	February	21.1±0.5	18.5±1.5	32.2±1.5	28.2±1.2
	March	23.0±1.5	17.8±2.1	34.0±1.2	25.2±2.1
	April	19.0±0.6	18.2±1.1	39.0±3.5	23.8±1.4
	May	16.1±0.8	12.2±0.8	45.9±2.5	25.9±1.5
	October	11.2±0.7	8.6±0.5	57.0±3.2	23.4±1.8
	November	16.3±0.2	10.6±0.7	51.0±3.1	22.4±1.8
	December	15.1±1.0	13.2±0.9	45.0±2.7	26.8±1.6
Yanam	February	9.2±0.7	11.1±0.2	42.0±2.5	37.7±2.1
	March	10.2±0.9	10.3±0.3	38.0±1.5	41.5±1.5
	April	11.2±0.1	10.3±1.5	41.2±1.2	37.3±3.5
	May	18.5±0.8	11.2±1.5	45.3±1.3	25.2±1.2
	October	16.2±1.5	14.8±0.9	24.1±0.9	44.9±1.1
	November	15.1±0.8	13.2±0.5	31.2±0.8	40.5±2.1
	December	16.0±0.9	12.4±0.6	38.1±0.6	33.6±1.2
Bhairavapalem	February	10.1±0.1	12.2±0.1	21.1±0.7	56.7±3.1
	March	12.2±0.3	13.4±0.5	23.1±0.8	51.4±1.2
	April	10.9±0.4	12.5±0.4	26.3±1.5	50.3±0.5
	May	2.1±0.5	3.1±0.3	36.1±1.4	58.7±0.3
	October	5.6±0.7	6.5±0.9	34.5±1.7	53.4±1.4
	November	2.8±0.1	3.4±0.8	31.2±1.8	62.6±0.8
	December	3.2±0.2	4.5±0.1	33.2±2.4	59.1±0.5