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## **Changes in metal contamination levels in estuarine sediments around India-An assessment**

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### **Abstract**

This review is the first attempt to comprehend the changes in metal contamination levels in surface estuarine sediments with changing time around India. Contamination factor, geo accumulation index, pollution load index, effects range low and effects range median analysis were used to evaluate the quality of the estuarine sediments (by using the available literature data). This study suggests that estuarine sediments from the east coast of India were comparatively less contaminated by metals than the west coast. Sediments from those estuaries were found to be more contaminated by metals on which major cities are located. An improvement in estuarine sediment quality (in terms of metal contamination) over time around India was noticed. This study provides managers and decision-makers of environmental protection agency with a better scientific understanding for decision-making in controlling metal pollution in estuarine sediments around India.

**Key words:** Metal pollution; estuarine sediments; Pollution load index; Geoaccumulation index; contamination factor;

## **Introduction**

With rapid industrialization and economic development, metals are continuing to be introduced to estuarine sediments via several pathways and may potentially result in associated environmental and health problems. Heavy/trace metals are one of the serious pollutants in our natural environment due to their toxicity, persistence and bioaccumulation problems (Chakraborty et al., 2010). Trace/heavy metals in natural waters and their corresponding sediments have become a significant topic of concern for scientists and engineers in various fields associated with water quality, as well as a concern of the general public. It has been reported that estuarine sediments are contaminated mostly with heavy/trace metals. They serve as filter for many pollutants between land and sea (Chapman and Wang, 2001). Estuarine sediments appear to be major repositories for metals. The environmental problems of estuarine sediment pollution by heavy/trace metals have received increasing attention in the last few decades in both developing and developed countries throughout the world (Zhang et al., 2007).

It has been widely recognized that heavy/ trace metals content in sediments can be useful in assessing the quality of the sediments (Burton and Scott, 1992; Caccia et al., 2003). However total metal concentrations in sediment are not a good indicator of predicting their bioavailability and toxicity. Knowledge of distribution and speciation of metals in sediments is necessary for providing a better understanding of their potential impacts at elevated concentrations in environments. Several studies have been carried out to understand the distribution of total metals content in sediments of the Indian estuaries over the last few decades (Table 1& 2); nevertheless, the information is quite limited for metal distributions and their speciation in the estuarine sediments of India. The rapid population growth along with the sharp increase of industrial and agricultural developments may lead to an irrevocable metal pollution of Indian rivers. An effort was made (in this review) to provide a better understanding of metals contamination and their changes with the progress of industrialization and urbanization in estuarine sediments around India.

The degree of quantification of metal pollution in sediments has been determined with respect to the metal concentrations in average shale (Förstner and Wittmann, 1983; Muller, 1979). Some authors (Baruah et al., 1998) have considered the background value of their area of study to be the geometric mean of concentration at the different sample sites, which is the antilog of the arithmetic average of  $\log_{10}$  (log to the base 10) of the concentration values. The composition of upper continental crust

(UCC) (Taylor and McLennan, 1985) was used as the background concentration (provided as supporting document, Table 1SD) to theoretically predict the contamination factors (CF), pollution load index (PLI), geoaccumulation index ( $I_{geo}$ ) in estuarine sediments around India (by using the data available in the literature). The ERL (effects range low)/ERM (effects range median) analysis was also used to understand estuarine sediment quality. The objectives of this work were to (i) understand the distributions of metals in the estuarine sediments (from 8 major estuaries) (ii) understand the variation in the PLI, CF, and  $I_{geo}$  with respect to time and (iii) understand the contribution of human activities on metal loading in estuarine sediments around India. The summary of this review is expected to help: (1) environmental managers to identify and fixing problems in estuaries (in terms of metal contamination); (2) concerned citizens who are interested to know about the estuarine sediments around India; and (3) researchers who wish to know what type of data (metal distribution) are available (in the literature) in the estuarine sediments around India.

In this review, the complex geology of India has also been considered and an attempt was made to understand metal contamination levels and their change with respect to time in Indian estuaries.

### **Geology of India**

The diverse geology of India can be broadly classified into Deccan trap, Gondwana and Vindhyan. The Deccan trap covers a major part of Maharashtra, a part of Gujarat, Karnataka, Madhya Pradesh and Andhra Pradesh marginally. It is believed that the Deccan Trap was formed as result of sub-aerial volcanic activity associated with the continental deviation in this part of the India during the Mesozoic Era. The rocks found in this region are generally igneous type. Gondwana lands are spread widely across central India, and are a small amount almost everywhere; there is no unambiguous boundary to the region. However, the core region can be considered to be the eastern part of the Vidarbha region of Maharashtra, the parts of Madhya Pradesh immediately to the north of it, and parts of the west of Chhattisgarh. The wider region extends beyond these, also including parts of northern Andhra Pradesh, western Odisha and Uttar Pradesh. Geologically it is mostly Pre-Cambrian rock, with some areas dated to Permian and Triassic periods. Part of it is overlaid with alluvium, and in the west it is overlaid with the igneous rocks of the Deccan Traps. India's geographical land area is shown in Figure 1.

## **Estuaries in India**

In addition to the complex geology of the bed rock, estuaries in India are highly dynamic with complicated sedimentary environment. The morphological features play an important role in sediment distribution in Indian estuaries. Rapid urbanization and anthropogenic processes also influence sedimentation pattern in the Indian estuaries. In this review, eight major estuaries were selected around India to assess the quality of the sediments (available in the literature). Brief descriptions of these eight major estuaries are given below (Figure 1).

### **Major rivers and their estuaries from the east coast of India**

#### ***River Ganges and its Estuary:***

Ganges, the largest river of India, originates from the Gangotri glacier (at altitude of 13,800 ft) in the mountain range of Himalayas. The river ranks among the top 10 rivers in the world in terms of water ( $5.9 \times 10^{11} \text{m}^3 \text{year}^{-1}$ ) and Sediment ( $1,600 \times 10^{12} \text{g year}^{-1}$ ) discharge (Purushothaman and Chakrapani, 2007). The estuary passes through the Kolkata city (a major city of India). A major port is also located on its banks. Due to its shallow depth (average 6 m) and intense tidal mixing it is known as well-mixed estuary (Sadhuram et al., 2005). The Ganges River basin has been reported as one of the most densely populated basin in the world (Chakrapani et al., 1995). Port activities (Haldia and Kolkata port), semi-treated wastes from varied industries (tanneries, jute mills, pulp and paper mills, pesticide manufacturing plants, thermal power plants, Kilns, rubber, fertilizer, soap factories, antibiotic plants), and oil refinery (Haldia) (De et al., 2010; Gopal and Chauhan, 2006) are the potential sources of trace/heavy metals around this estuary. The sediments in the Ganges estuary are mainly illite with significant amount of detrital quartz and feldspar, particularly in coarse grain fraction.

#### ***River Godavari and its Estuary:***

The Godavari River (the third largest River in India) originates from the Western Ghats in Maharashtra state and flows through the states of Karnataka, Madhya Pradesh, Chattishgarh, Odhissa, and Andhra Pradesh, and empties into the Bay of Bengal. The river divides into two branches at the township of Dowaleshwram, which are named the Gautami (major branch) and the Vasista (minor branch). The basin of the Godavari River area includes some major geological formations (Krishnan, 1966) such as the Tertiary Deccan Traps (48%), Archean granites (39%), Precambrian and Gondwana

sedimentary rocks (11%) and recent alluvials (2%). Agriculture and aquaculture are the major sources of livelihood for local populations in these areas (the Gautami and the Vasistha Godavari estuary areas). The recent exploration of oil and natural gas reserves has increased the pace of the industrial development around the estuary. There are also few major fertilizer factories and gas based power plants and oil refineries operating around the estuary.

#### ***River Krishna and its Estuary:***

The Krishna River originates from Western Ghats (at an elevation of 1337 m above the sea level). Tungabatra and Bhima are the major tributaries of Krishna River. The Krishna basin has a drainage area of 258,945 km<sup>2</sup>, covering a distance of 1400 km before merging into the Bay of Bengal. The basin area composed of major geological formations as Archaean and younger crystalline rocks (78%), the Tertiary Deccan Traps (20%) and recent alluvial (2%) (Ramesh et al., 1989).

#### ***River Cauvery and its Estuary:***

The Cauvery River originates in the Brahmagiri range of the Western Ghats and flows through Karnataka and Tamil Nadu. Cauvery River flows through the areas which are dominated with Precambrian, Tertiary sedimentary rocks and Plio-Pleistocene sediments (Subramanian et al., 1985). The Cretaceous formations of the coastal tract of the Cauvery basin consist of faunal-rich marine sedimentary rocks, namely limestone, sandstones, clays and sandy beds etc. The mouth of the basin comprises alluvium deposits which are of clays and silts (Krishnan, 1966).

### **Major rivers and their estuaries from the west coast of India**

#### ***River Narmada and its Estuary:***

The Narmada River originates from Amarkantak in the Maikala Hills of Madhya Pradesh, India. It is the largest west flowing river and ranks seventh in terms of water discharge and drainage area in India. The river flows through Deccan traps, separated by the Vindhyan and Satpura ranges of hills. The Narmada River flows into the Arabian Sea at about 10 km north of Bharuch (via Gulf of Cambay).

The major lithologies of the basin are Deccan basalts, sedimentary rocks of Vindhyan and Gondwana super groups and Quaternary formations. Quaternary alluvium is restricted to the Narmada Valley. The river is polluted mostly by city sewage along with industrial effluents (Jain et al., 2008).

### ***River Tapti and its Estuary:***

The Tapti River originates from high range of Satpura Mountain at Multai in Madhya Pradesh. This river is 720 km long. The Tapti River flows through Madhya Pradesh, Maharashtra and Gujarat, and finally discharges into the Gulf of Kambhat (Cambay). A major part of the basin (62 %) is predominantly agricultural while 26.6 % of the area is under forest cover. The rest of the land is classified as non-arable and utilized for industrial activities and human settlement. There are 93 large and medium scale industrial units in the basin, plus innumerable small-scale units. Agricultural runs-off, urban sewage, and effluents from engineering industries, thermal power plants, food and beverages, textiles, chemical and metallurgical industries are the major sources of metal contamination in the Tapti estuary. (Krupadam et al., 2006).

### ***Ulhas Estuary***

The Ulhas River originates from the Westren Ghat (900m altitude above sea level). It reaches kalyan after travelling 80km mountain route through the western slope of the Ulhas plateau. Finally, it merges into the Arabian Sea at the Vasai Creek. The Ulhas basin is about 5000 sq.km and it is a monsoon fed river. The geology of the area is characterized by basaltic flows of Deccan traps having mineral assemblage of pyroxenes, plagioclase and opaque oxides with or without olivines. A large amount of effluents get discharged in the river from the industrial belts of Kalyan-Ambarnath and Ulhasnagar and the Waldhuni tributary which flows through these belts and is an effluent canal(Sahu and Mukherjee, 1983). This estuary passes through the Mumbai city.

### ***Cochin Estuary***

The Cochin estuary is one of the largest estuaries of India (256 km<sup>2</sup>). The mouth of the Cochin estuary is the widest (450 m) and considered as main entrance to the Arabian Sea from the west coast of India. This estuary generally has a width in the range of 0.8-1.5 km and depth of 4-13 m towards south and become shallow (0.5-3 m) in north with an area of 0.05-0.5 km.

The Cochin is the largest growing city after Mumbai in the west coast and is listed among the 17<sup>th</sup> major industrial cities of India according to the World Bank. It is also the 4<sup>th</sup> largest port in India. At the terminal of Willington Island (Cochin port) it handles export and import of container cargos. The principal Chemical industries of Kerala state (~70 %) are mainly located at the banks of Periyar and

Chitrapuzha. Apart from it approximately 250 industries are present around the estuary. Many of these industries are more than 50 years old. These industries take large amounts of fresh water from the river Periyar and in turn discharge~ 260 million liters of concentrated effluents daily with little treatment (Joy et al., 1990; Madhupratap, 1987; Martin et al., 2012; CPCB, 1996).

### ***Assessment of trace/heavy metal contamination in sediments***

Simple (which include the contamination factor, and Index of Geoaccumulation) and integrated indices (the Pollution Load Index) were used (in this study) to evaluate metal contamination level (based on the data available in the literature) in estuarine sediments around India.

### **Effects range low (ERL)/ Effects range median (ERM) Analysis**

The level of metal contaminants in estuarine sediments was evaluated by the ERL/ERM analysis.

This analysis was proposed by Long et al., 1995. Long et al., 1995 reviewed field and laboratory studies and identified nine metals (As, Cd, Cr, Cu, Pb, Hg, Ni, Ag, Zn) that were observed to have ecological or biological effects on organisms (in sediments). They defined ERL values as the lowest concentration of a metal that produced adverse effects in 10% of the data reviewed. Similarly, the ERM designates the level at which half of the studies reported harmful effects. Metal concentrations below the ERL value are not expected to elicit adverse effects, while levels above the ERM value are likely to be very toxic. ERL and ERM limits for metals in estuarine sediments are presented as supporting documents (Table 2SD).

A station is rated “good” if the concentrations of all the metals are below the ERL limit. An “intermediate” rating applies if any metal exceeds an ERL limit, and a “poor” rating for a metal indicates that its ERM limit has been exceeded.

### ***Contamination Factor***

The contamination factor (CF) was used to express the level of contamination by each metal in sediment. It is expressed as

$$\text{Contamination Factor (CF)} = \frac{\text{Metal Content in the sediment}}{\text{Background Value of Metal}}$$

“Background value of metal” indicates the concentration of metal (of interest) in the sediments when there was no anthropogenic input. Unavailable background value of metal in Indian estuarine systems forced us to consider the concentration of metal in upper continental crust (UCC) (Taylor and McLennan, 1985) as a representative of background value in the sediments.

$CF < 1$  – refers low contamination by a metal,

$1 \leq CF \leq 3$  – moderate contamination,

$3 \leq CF \leq 6$  – considerable contamination and

$CF > 6$  high contamination (Hakanson, 1980).

### ***Pollution Load Index***

Pollution Load Index (PLI) has been widely used to evaluate contamination level and pollution in coastal and estuarine sediments. A PLI value of zero indicates “no pollution”, a value of 1 indicates that only “baseline levels of pollutants” are present, and above 1 indicates “progressive deterioration of estuarine quality” (Tomlinson et al., 1980). The equation used here to calculate PLI was developed by Tomlinson et al., (1980).

$$CF = C_{\text{metal}} / C_{\text{background}} \quad (1)$$

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n} \quad (2)$$

$CF$  = Contamination factor

$n$  = Number of metals

$C_{\text{metal}}$  = concentration of metal in sediment sample

$C_{\text{background}}$  = background value of the metal.

### ***Geoaccumulation Index***

The geoaccumulation index ( $I_{\text{geo}}$ ) was calculated by using the following equation (Muller, 1969)

$$I_{\text{geo}} = \log_2 \frac{C_n}{1.5B_n} \quad (3)$$



Where  $C_n$  is the measured concentration of element 'n' in the sediment samples and  $B_n$  is the geochemical background value of the element 'n' in earth's crust (the concentration of the element 'n' in the UCC(Taylor and McLennan1985). The factor 1.5 is introduced to include differences in the background values due to lithological variations. Six classes of the geochemical index are given below.

*Index of geoaccumulation (Igeo) and contamination level*

Class	Value	Contamination Level
0	$I_{geo} < 0$	unpolluted
1	$0 < I_{geo} < 1$	unpolluted to moderately polluted
2	$1 < I_{geo} < 2$	moderately polluted
3	$2 < I_{geo} < 3$	moderately to heavily polluted
4	$3 < I_{geo} < 4$	heavily polluted
5	$4 < I_{geo} < 5$	heavily to extremely polluted
6	$I_{geo} > 5$	extremely polluted

### **Results and Discussion:**

Total metal concentrations in estuarine surface sediments around India at different times (as reported in the literature) are summarized in Tables 1 and 2. These data were used to calculate degree of metal contamination levels in the major estuarine sediments of India.

ERM/ERL analysis was performed on the basis of the reported concentrations of metals in the estuarine sediments. Figure 2 shows the rating of estuarine sediment quality (in terms of metal contaminations) with respect to time. Figure 2 indicates that the qualities of estuarine sediments from the Ganges and Cauvery estuaries (from the east coast of India) were in an "intermediate" and "good" state (recent status of estuary based on the recent publication). However, the quality of sediment from Godavari and Krishna estuary were in a "poor state. The sediments from the west bound estuaries (Narmada, Tapti, Ulhas and Cochin) were in a "poor" state. It is shown in the Fig 2 that the quality of estuarine sediments

(from the Godavari, Krishna estuaries from the east coast and the Tapti, Narmada, Ulhas and the Kochi estuaries from the west coast) deteriorated with time. However, the sediment quality guideline ERL may not be a threshold of any chemical concentrations in sediment at which the probability of toxicity shows an abrupt increase. Similarly, there is no basis for assuming that multiple concentrations above an ERL increase the probability of toxicity. Scarcity of systematic analysis and data from Indian estuary in the literature forced us to use ERL/ERM and other indices to estimate the quality of estuarine sediments around India.

The CF, PLI,  $I_{geo}$  were used for the environmental assessment of estuarine sediments (in this study). It is necessary to compare the level of metals (in estuarine sediments) with the pre-industrial reference level (from the same estuary). Unfortunately, no such background data are available for each individual estuary around India. In this study, the composition of upper continental crust (UCC) was used as a representative of pre-industrial reference level of trace/heavy metals.

Variations in the CFs of different metals in estuarine sediments around India at different times are shown in Fig. 3. The data are presented in Tables 3 (east coast) and 4 (west coast). The total Cr content in the estuarine sediments was found to be high in the west coast of India. High level of Cr contamination was found in the estuarine sediments from the Cochin and Ulhas estuaries (from west coast). This was probably because of the impact of high anthropogenic activities from the two major cities (Cochin and Mumbai) of India.

It is shown in Fig. 3 that the CF of Cr in the Ulhas estuary was changed from moderate (CF=1.20) to highly contaminated (CF= 14.18) with the progress of time from 1984 to 2000. The CF was highest in the Ulhas followed by the Tapti, Narmada and the Cochin estuary (from west coast). The reported concentrations of Cr in the estuarine sediments from the east coast of India indicate that the sediments from the Ganges, Godavari and the Cauvery estuaries were not contaminated with Cr.

The values of the CF for Ni were found to be less than 3 in most of the estuaries. However, the sediments from Tapti, Narmada, Ulhas (west coast) and Krishna estuaries (East coast) were reported to have very high Ni loading and the calculated CF value categorize them in considerable contaminated estuaries.

Most of the estuarine surface sediments (both in east and west coast) were fairly contaminated by Cu. The Ulhas estuary is in considerable contaminated state by Cu. Similarly, the CF of Cu in the estuarine sediments of the Narmada (7.5) and the Tapti River (13.05) were found to be high. The literature values showed that the estuarine sediments were not contaminated by Pb. A steady decrease in the CF of Pb was found in the major estuaries from the east coast of India. The sediments from the major estuaries were found to be contaminated by Cd. An abnormally high CF value for Cd has been reported in estuarine sediments from the Godavari estuary (by Chakraborty et al., 2012 in their recent study). Ray et al., 2006 have also reported a high CF (CF=92) for Cd in the Godavari estuary. The Cauvery estuary has also been reported to be highly contaminated with Cd. Sediment from the Ganges estuary has been found in the range of considerable to highly contaminated state for Cd. The sediments from the major estuaries from the west coast were found to be contaminated with Cd (except the Tapti estuary). Martin et al., 2012 has recently reported that the surface sediments from the Cochin estuary were also highly contaminated by Cd (92.5). The level of Cd loading on the surface sediment from the Ulhas estuary is not known. However, it has been reported (in 1984-86) that sediment from the Ulhas estuary was highly contaminated with Cd (with CF=34.83).

The pollution load index (PLI) of the estuarine sediments were calculated (based on the data available in the literature) to understand the change in overall contamination levels in the estuarine sediments around India. Lower values of the PLI (<1) imply no appreciable input from anthropogenic sources. However, the PLI value > 1 suggests pollution. The calculated PLI values showed that the sediments from the estuaries of west coast of India were more polluted compared to the estuarine sediments from east coast. The literature data suggest that the PLI values of most of the estuaries were within 2.

Due to wide variation in bed geology, sediment processes of rivers, variation in sediment input from rivers, mixing of fresh and salt waters under the influence of tides and the prevalent dry and wet seasons makes it extremely difficult to compare contamination level of estuarine sediments from east and west coast of India. It has been reported that sediments and suspended particle discharge from the rivers of east coast are much more than the rivers of west coast. Thus, this is probably one of the reasons for high metal loading in the estuarine sediments from the west coast of India. However, anthropogenic activities were also responsible for increasing the PLI in the estuarine sediments from the west coast of India (e.g., the Cochin and Ulhas estuaries).

*The changes in the PLI in estuarine sediments from the east coast of India are discussed below.*

Variations of the PLI in estuarine sediments from the east coast of India are presented in Table 5 and Figure 4. It is shown that the PLI in the estuarine sediments from Cauvery estuary gradually decreased from 3.47 (Seralathan and Seetaramaswamy, 1987; Seralathan, 1987) to 3.27 (Subramanian et al., 1989) to 1.78 (Ramanathan et al., 1988) and further decrease to 0.91 (Ramanathan et al., 1993) then to 1.18 (Ramesh et al., 1999) and finally 0.50 (Dhanakumar et al., 2013). This change in the PLI values in Cauvery estuary indicates a significant improvement in estuarine sediment quality (with respect to metal loading) over time.

The change in the PLI values of estuarine sediments from the Godavari estuary with respect to time also showed a steady decrease in metal pollution levels. However, the recent data reported by Chakraborty et al., 2012 showed very high PLI of 11.04, which may have occurred due to sampling at a particularly contaminated site. High loading of Cd, Zn, and Pb in the estuarine sediments was responsible for such high PLI value in Godavari estuary (Table 5). However, this very high value may not be a good representative of contamination level in the estuary.

The PLI of estuarine sediments from the Krishna River was very similar to the PLI of estuarine sediments in the Godavari estuary (Table 5). The PLI of estuarine sediments from the Ganges/Hooghly Rivers shows that the sediments were slightly contaminated with respect to UCC (Table 5). The PLI of the estuarine sediment were found to vary from 1.5 to 0.88. The recent study by Banerjee et al., 2012 has also indicated that the estuarine sediments (Ganges) are probably not being polluted due to anthropogenic activities in the Ganges estuary.

*The changes in the PLI in the estuarine sediments from the west coast of India are discussed below.*

The PLI in the estuarine sediment from the Cochin estuary was found to change from 0.11-8.17 with the exponential growth of human population and growing industrialization around the estuary over time. The PLI, calculated from the data reported by Venugopal et al.(1982) was 1.35. The PLI value further deteriorated to 8.17 (Balachandran et al., 2005) with the rapid growth of industrialization and urbanization (between 1982-2005). However, a decrease in the PLI was observed (PLI= 2.65) by Balachandran et al.(2006). and (PLI=3.53) Martin et al.(2012) (Table 5). These changes in the PLI in Cochin estuarine sediment suggest that the sediments are still polluted due to industrial activities around

the estuary (in the city Cochin). However, the decreasing trend in the PLI probably suggests that measures probably are being taken to control the input of metals to the estuary.

The PLI of estuarine sediment from the Ulhas estuary indicate that the estuary was highly polluted. The PLI in Ulhas estuary has been gradually increasing over time and due to the increase in anthropogenic activities as reported by Rokade (2009). The data obtained from earlier work (Sahu and Bhosale, 1991) show a PLI of 3.35 (Table 5). The PLI values of Narmada and Tapti river sediments showed a similar trend (Figure 4). Only two studies have been reported on these two rivers. Narmada River showed the PLI (1.06 & 3.25) and Tapti (1.74 & 3.55) indicating that the river is probably getting polluted with increasing anthropogenic activities.

The geoaccumulation index ( $I_{geo}$ ) for each element of all the estuaries around India is summarized in Table 6. The  $I_{geo}$  values of metals were lower in the estuarine sediments from the east coast compared to the west coast of India. This study indicates that estuarine sediments from the east coast were contaminated with Cd (the  $I_{geo}$  values ranging from 0.29-7.4). The sediments from Godavari estuary (7.4) was found to be extremely polluted by Cd and other three estuaries were also heavily polluted.

The  $I_{geo}$  values (Ramesh et al., 1999) indicate that Krishna estuary was reasonably pollute with Cd and moderately polluted with Cr, Ni and Co. The  $I_{geo}$  values also indicate heavy Cd pollution in Ganges/Hooghly estuary.

The  $I_{geo}$  values from the estuarine sediments from the West coast of India showed that the Cochin estuary was contaminated with Cd and Zn. The  $I_{geo}$  value of the Ulhas estuary revealed that the sediment in this estuary was heavily polluted with Cr. The estuarine sediments were moderately contaminated by Co, Ni and Cu in the Ulhas estuary.

## **Conclusion**

Comparison of the CF, PLI and  $I_{geo}$  and total trace metal content in the estuarine sediments from the east and west coast of India may be a convenient way of expressing some measure of pollution, but this method has its limitations. Sediment metal concentrations are influenced by a range of factors. They include physical and hydrological characteristics of the region and its benthos, atmospheric conditions, productivity, pH, sediment texture, redox potential and cation exchange capacity among others.

Internal geochemical processes can lead to remobilization of trace metals and this phenomenon should be taken into account while comparing the results. The quantity of metals retained in sediments is also affected by the characteristics of the sediment into which they are adsorbed. Grain size, partition coefficient ( $K_d$ ), cation exchange, organic matter content and mineral constituents all influence the uptake of heavy metals from the sediments by the aquatic organism.

However, elevated concentrations of metals do not necessarily pose a threat as they may never be released from the sediments and therefore may not be bioavailable. It is well known that sediments reflect the productivity of an area. The specific fate of heavy metals in the environment cannot be completely understood due to unexpected acts of nature. However, the best alternative is to make educated estimations using the available data.

The comparison of the contamination factor (CF) and geo accumulation index ( $I_{geo}$ ) and Pollution Load Index (PLI) of estuarine sediments from east and west coast of India (performed with the available data from the literature) may oversimplify the complex geochemical processes that are involved in each estuary. The intention of this study was to identify the estuaries with higher metal loading in the sediments. The ERL/ERM analysis suggests that the estuarine sediments around India are mainly in an “intermediate” to a “poor” state. Estuarine sediments of the Cochin and Ulhas were found to contain high metal loading. The sediment from the Ulhas estuary is contaminated with Cr. All the major estuarine sediments contain high concentrations of Cd. The PLI values and ERL/ERM analysis suggest that estuarine sediments from East coast are comparatively less contaminated with metals. It is also important to note that the decreasing trend in the PLI values indicate a significant improvement in some estuarine sediment (from Ganges, Cauvery and Cochin estuaries) quality with the progress of time. However, significant improvement is also required in some major estuaries (Ulhas, Cochin, Krishna estuaries) around India. Systematic investigation is recommended to monitor metal loading and change in estuarine sediment quality around India.

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**Figure Captions.**

Figure 1: Geological map with the major estuaries of India.

Figure 2: Status of the major estuaries of india with respect to the Effect range low (ERL) and Effect range median (ERM).

Figure 3: Changes in the contamination factor (CF) of heavy/ trace metals in the estuarine bed sediments over time.

Figure 4: Changes in the pollution load index (PLI) in the estuarine bed sediments over time.

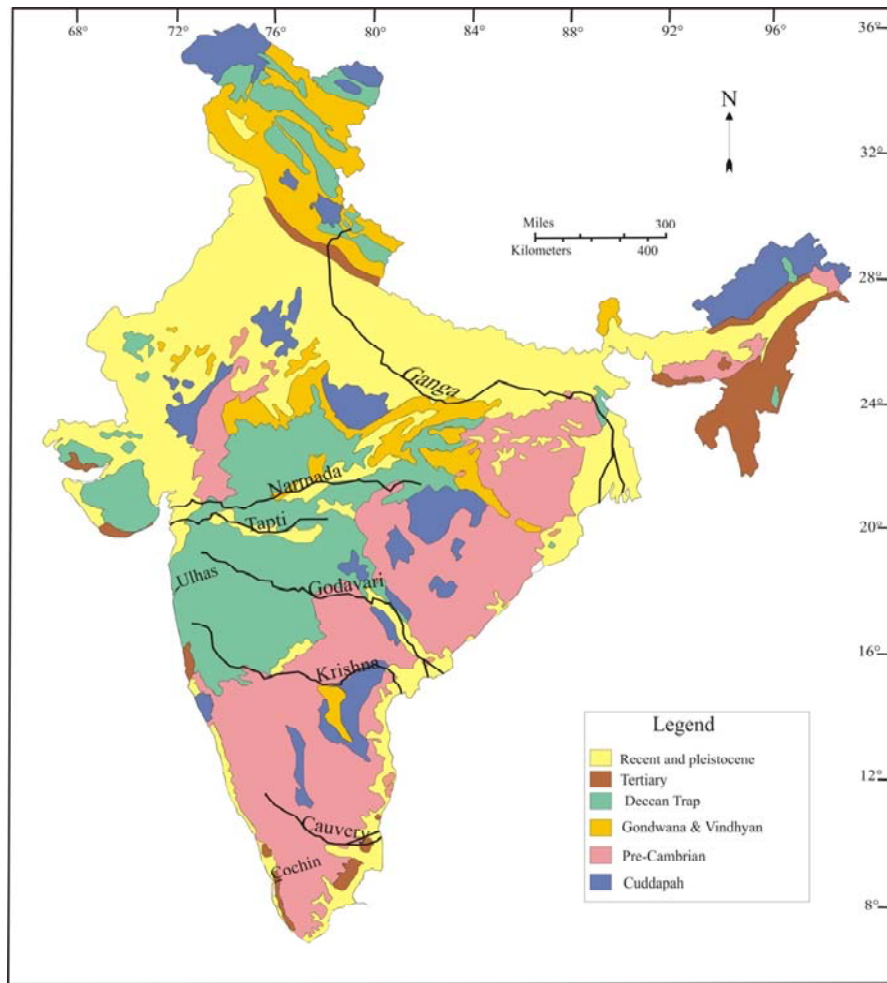


Figure 1

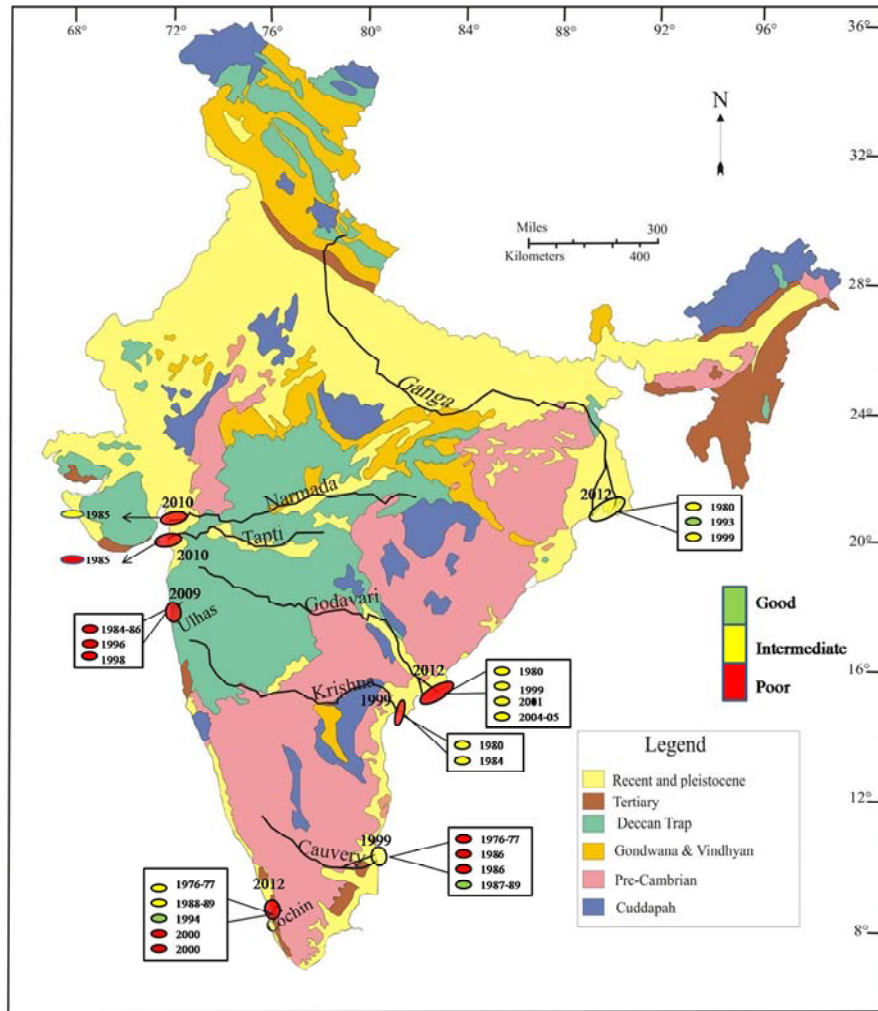


Figure 2

Contamination factor

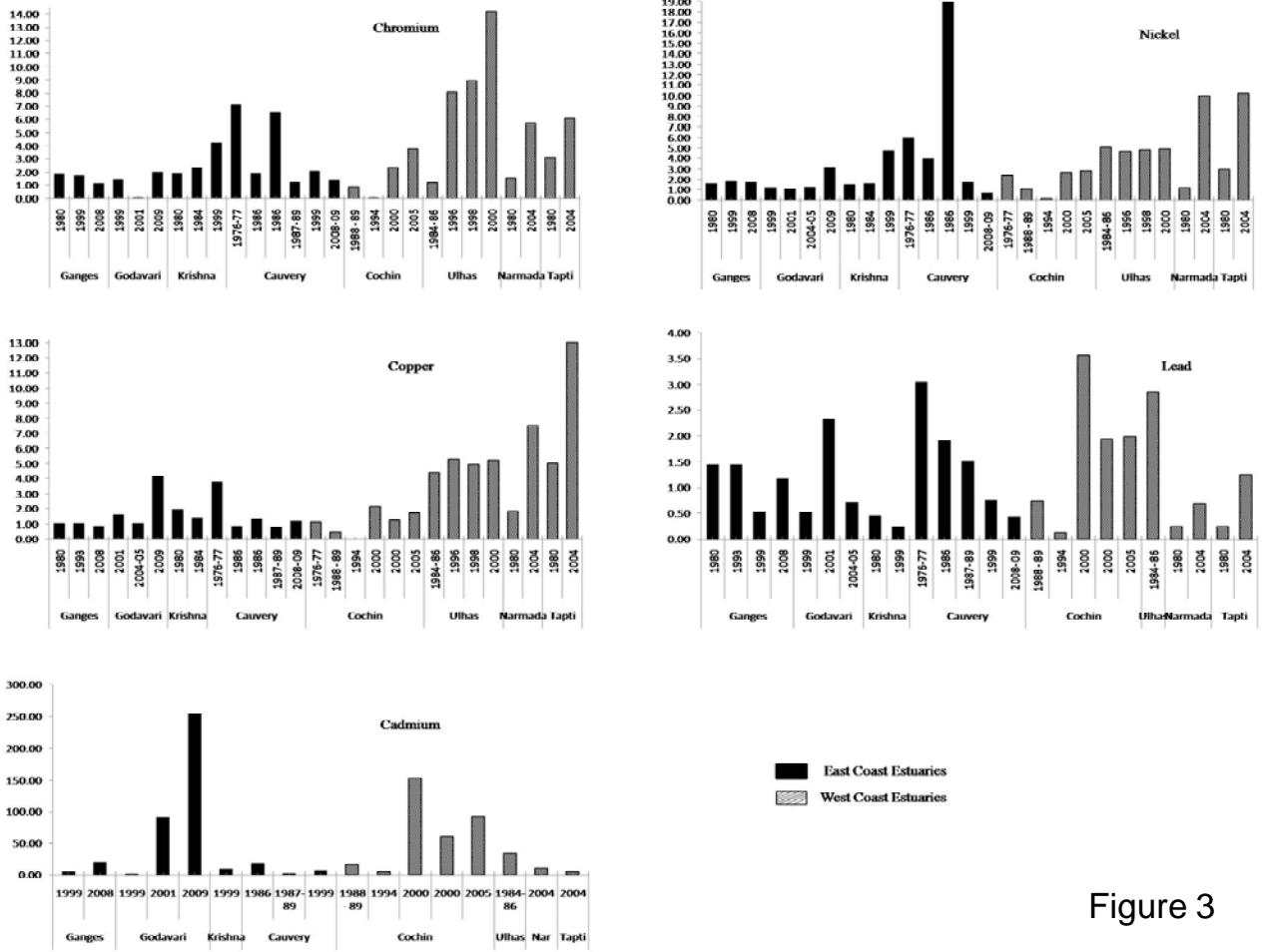


Figure 3

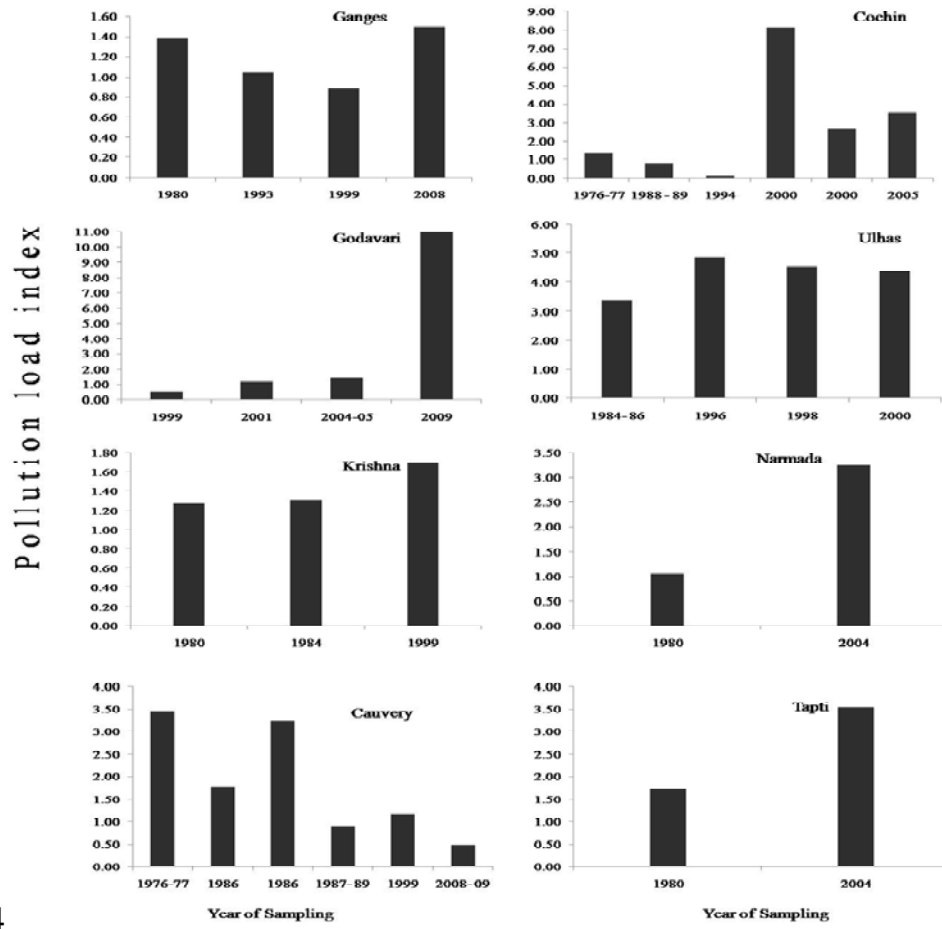


Figure 4

Table 1: Trace metal concentrations ( $\mu\text{g/g}$ ) in sediments from the major estuaries of the east coast of India.

Estuary	References	Cr	Ni	Cu	Zn	Pb	Cd	Hg	Mn	Fe	Co	As	No. of Samples	Year of sampling	Methods
Ganges	Subramanian et al 1988	67	32	26	71	29			553	31036	36		19	January 1980	EDXRF*
	Subramanian 1993			26	71	29			550	31000			19	Winter Season	AAS *
	Ramesh et al 1999	61.46	37.5		64.38	10.49	0.45				10.76	0.08	11		TIMS*
	Banerjee et al 2012	40.11	33.97	21.64	53.42	23.45	2.01		502.42	28600	18.23		17	June 2008	AAS*
	Ramesh et al 1999	50.88	23.55		27.28	10.15	0.18				6.04	0.05	6		TIMS*
Godavari	Ray et al 2006	1.73	20.7	40.2		46.7	9		429	4254	30.7		4	September 2001	AAS*
	Krupadam et al 2007		25.1	25.8	197.1	14.4					21.9		10	2004-2005	AAS*
	Chakraborty et al 2012	71.2	63.8	103.4	3876.7	424	24.8				25.5			2009	ICPMS*
Krishna	Subramanian et al 1985	68	30	49	31	9			1040	42280	29		19	October December 1980	EDXRF*
	Ramesh et al 1989	82	32	35	26				906	25100	32		14	Aug-84	XRF*
	Ramesh et al 1999	148.46	94.67		171.12	4.81	0.99				37.8	0.14	9		TIMS*
Cauvery	Srealathan 1987 & Srealathan and Seetaramaswamy	249.5	120.7	94.5	118.7	61			3100		14		6 & 15	1976-1977	colorimetrically and AAS*
	Ramanathan et al 1988	68.3	79.4	21.7					607.2	35552	46.1		18	July 1986	AAS*
	Subramanian et al 1989	229	379	33	75	38	1.85	0.118	1310	33500			18	July 1986	AAS* and Hydride system
	Ramanathan et al 1993	42.9		19.6	25.4	30.3	0.22		567.2	16285			16	June - 1987 & April-1989	AAS*
	Ramesh et al 1999	73.39	34.16		49.36	15.12	0.73				9.49	0.35	22		TIMS*
	Dhanakumar et al 2013	49.5	13.5	29.5	30	8.5			160	5228.5				10	May-2008 & Nov-2009

\*AAS-Atomic absorption spectrometry, TIMS- Thermal ionization mass spectrometer, XRF (EDXRF)- Energy dispersive x-ray fluorescence, ICPMS- Inductively coupled plasma mass spectrometry.

Table 2: Trace metal concentrations ( $\mu\text{g/g}$ ) in sediments from the major estuaries of the west coast of India.

Estuary	References	Cr	Ni	Cu	Zn	Pb	Cd	Hg	Mn	Fe	Co	As	No. of Samples	Year of Sampling	Methods
Cochin	Venugopal et al 1982		48.1	28	205				152.7		22.3			1976-1977	AAS *
	Nair 1992	29.99	21.15	12.2	70.4	14.86	1.66		66.03	8325	9.23		120	1988&89	AAS*
	Jayasree & Nair 1995	1	3	0.4	14	2.7	0.51		41	803	2.1		10	Jan- April 1994	AAS *
	Balachandran et al 2005			53.15	1266	71.28	14.94			61800				November 2000	AAS*
	Balachandran et al 2006	82.3	53.7	30.8	562.3	38.7	5.91		229.1	44700	18.3		17	November 2000	AAS*
Ulhas	Martin et al 2012	131.9	57.0	43.4	422.7	39.8	9.1		1147.8	29100.0	26.1		56	2005	AAS *
	Sahu & Bhosale 1991	42.1	101.2	109.7	143.6	57.0	3.4	0.7	731.0	66571.3	42.6		11	1984-86	AAS* and Hg analyzer
	Rokade 2009	282.6	93.2	132.6	862.3				1232.2	95000.0	47.4		10	1996	AAS*
Narmada	Rokade 2009	312.3	96.6	124.3	400.9				1301.8	84000.0	63.3		13	1998	AAS*
	Rokade 2009	496.3	98.0	130.9	217.5				1151.5	78000.0	64.1		11	2000	AAS*
	Subramanian et al 1985	55	23	46	50	5			514	31400	36		3	October-December 1980	EDXRF*
Tapti	Sanjay & Subramanian 2010	199.3	200.3	188.3	196.2	13.9	1.1		1214	89577	25.9	1.6	9	2004	WDXRF* and EDXRF*
	Subramnian et al 1985	108	60	126	118	5			1300	10900	64		3	October-December 1980	EDXRF*
	Sanjay & Subramanian 2010	212.6	205.5	326.2	216.7	25	0.5		1498	91128	27	1.7	5	2004	WDXRF* and EDXRF*

\*AAS-Atomic absorption spectrometry, EDXRF- Energy dispersive x-ray fluorescence,

WDXRF- Wavelength dispersive x-ray fluorescence



Table 3: The contamination factor (CF) of trace metals (East coast of India) calculated with respect to the Upper Continental Crust composition (Taylor and McLennan 1985).

Estuary	References	Cr	Ni	Cu	Zn	Pb	Cd	Mn	Fe	Co	As
Ganges	Subramanian et al 1988	1.91	1.60	1.04	1.00	1.45		0.92	0.89	3.60	
	Subramanian 1993			1.04	1.00	1.45		0.92	0.89		
	Ramesh et al 1999	1.76	1.88		0.91	0.52	4.59			1.08	0.05
	Banerjee et al 2012	1.15	1.70	0.87	0.75	1.17	20.51	0.84	0.82	1.82	
Godavari	Ramesh et al 1999	1.45	1.18		0.38	0.51	1.84			0.60	0.03
	Ray et al 2006	0.05	1.04	1.61		2.34	91.84	0.72	0.12	3.07	
	Krupadam et al 2007		1.26	1.03	2.78	0.72				2.19	
	Chakraborty et al 2012	2.03	3.19	4.14	54.60	21.20	253.06			2.55	
Krishna	Subramanian et al 1985	1.94	1.50	1.96	0.44	0.45		1.73	1.21	2.90	
	Ramesh et al 1989	2.34	1.60	1.40	0.37			1.51	0.72	3.20	
	Ramesh et al 1999	4.24	4.73		2.41	0.24	10.10			3.78	0.09
Cauvery	Srealathan , Srelathan & Seetaramaswamy 1987	7.13	6.04	3.78	1.67	3.05		5.17		1.40	
	Ramanathan et al 1988	1.95	3.97	0.87				1.01	1.02	4.61	
	Subramanian et al 1989	6.54	18.95	1.32	1.06	1.90	18.88	2.18	0.96	0.00	
	Ramanathan et al 1993	1.23		0.78	0.36	1.52	2.24	0.95	0.47		
	Ramesh et al 1999	2.10	1.71		0.70	0.76	7.45			0.95	0.23
	Dhanakumar et al 2013	1.41	0.68	1.18	0.42	0.43		0.27	0.15		

Table 4: The contamination factor (CF) of the trace metals (West coast of India) calculated with respect to the Upper Continental Crust composition (Taylor and McLennan 1985).

Estuary	References	Cr	Ni	Cu	Zn	Pb	Cd	Mn	Fe	Co	As
Cochin	Venugopal et al 1982		2.41	1.12	2.89			0.25		2.23	
	Nair 1992	0.86	1.06	0.49	0.99	0.74	16.94	0.11	0.24	0.92	
	Jayasree & Nair 1995	0.03	0.15	0.02	0.20	0.14	5.20	0.07	0.02	0.21	
	Balachandran et al 2005			2.13	17.83	3.56	152.45		1.77		
	Balachandran et al 2006	2.35	2.68	1.23	7.92	1.94	60.31	0.38	1.28	1.83	
	Martin et al 2012	3.77	2.85	1.73	5.95	1.99	92.86	1.91	0.83	2.61	
	Ulhas	Sahu & Bhosale 1991	1.20	5.06	4.39	2.02	2.85	34.83	1.22	1.90	4.26
Rokade 2009		8.07	4.66	5.30	12.15			2.05	2.71	4.74	
Rokade 2009		8.92	4.83	4.97	5.65			2.17	2.40	6.33	
Rokade 2009		14.18	4.90	5.24	3.06			1.92	2.23	6.41	
Narmada	Subramanian et al 1985	1.57	1.15	1.84	0.70	0.25		0.86	0.90	3.60	
	Sanjay & Subramanian 2010	5.69	10.02	7.53	2.76	0.70	11.22	2.02	2.56	2.59	1.07
Tapti	Subramanian et al 1985	3.09	3.00	5.04	1.66	0.25		2.17	0.31	6.40	
	Sanjay & Subramanian 2010	6.07	10.28	13.05	3.05	1.25	5.10	2.50	2.60	2.70	1.13

Table 5: The pollution load index (PLI) value of the estuarine sediment around India

<b>Estuary</b>	<b>References</b>	<b>PLI</b>
Ganges	Subramanian et al 1988	1.39
	Subramanian 1993	1.04
	Ramesh et al 1999	0.88
	Banerjee et al 2012	1.50
Godavari	Ramesh et al 1999	0.53
	Ray et al 2006	1.21
	Krupadam et al 2007	1.41
	Chakraborty et al 2012	11.04
Krishna	Subramanian et al 1985	1.27
	Ramesh et al 1989	1.31
	Ramesh et al 1999	1.70
Cauvery	Srealathan , Srelathan & Seetaramaswamy 1987	3.47
	Ramanathan et al 1988	1.78
	Subramanian et al 1989	3.27
	Ramanathan et al 1993	0.91
	Ramesh et al 1999	1.18
	Dhanakumar et al 2013	0.50
Cochin	Venugopal et al 1982	1.35
	Nair 1992	0.80
	Jayasree & Nair 1995	0.11
	Balachandran et al 2005	8.17
	Balachandran et al 2006	2.65
	Martin et al 2012	3.53
Ulhas	Sahu & Bhosale 1991	3.35
	Rokade 2009	4.86
	Rokade 2009	4.54
	Rokade 2009	4.37
Narmada	Subramanian et al 1985	1.06
	Sanjay & Subramanian 2010	3.25
Tapti	Subramanian et al 1985	1.74
	Sanjay & Subramanian 2010	3.55

Table 6: The geoaccumulation index (Igeo) values of estuarine bed sediments around India.

Estuary	References	Cr	Ni	Cu	Zn	Pb	Cd	Mn	Fe	Co	As
Ganges	Subramanian et al 1988	0.35	0.09	-0.53	-0.58	-0.05		-0.70	-0.76	1.26	
	Subramanian 1993			-0.53	-0.58	-0.05		-0.71	-0.76		
	Ramesh et al 1999	0.23	0.32		-0.73	-1.52	1.61			-0.48	-4.81
	Banerjee et al 2012	-0.39	0.18	-0.79	-1.00	-0.36	3.77	-0.84	-0.88	0.28	
Godavari	Ramesh et al 1999	-0.05	-0.35		-1.96	-1.56	0.29			-1.31	-5.49
	Ray et al 2006	-4.92	-0.54	0.10		0.64	5.94	-1.07	-3.63	1.03	
	Krupadam et al 2007		-0.26	-0.54	0.89	-1.06				0.55	
	Chakraborty et al 2012	0.44	1.09	1.46	5.19	3.82	7.40			0.77	
Krishna	Subramanian et al 1985	0.37	0.00	0.39	-1.78	-1.74		0.21	-0.31	0.95	
	Ramesh et al 1989	0.64	0.09	-0.10	-2.03			0.01	-1.06	1.09	
	Ramesh et al 1999	1.50	1.66		0.68	-2.64	2.75			1.33	-4.01
Cauvery	Seralathan 1987, Srelathan and Seetaramaswamy	2.25	2.01	1.33	0.16	1.02		1.78		-0.10	
	Ramanathan et al 1988	0.38	1.40	-0.79				-0.57	-0.56	1.62	
	Subramanian et al 1989	2.12	3.66	-0.18	-0.51	0.34	3.65	0.54	-0.65		
	Ramanathan et al 1993	-0.29		-0.94	-2.07	0.01	0.58	-0.67	-1.69		
	Ramesh et al 1999	0.48	0.19		-1.11	-0.99	2.31			-0.66	-2.68
	Dhanakumar et al 2013	-0.08	-1.15	-0.35	-1.83	-1.82		-2.49	-3.33		
Cochin	Venugopal et al 1982		0.68	-0.42	0.94			-2.56		0.57	
	Nair 1992	-0.81	-0.50	-1.62	-0.60	-1.01	3.50	-3.77	-2.66	-0.70	
	Jayasree & Nair 1995	-5.71	-3.32	-6.55	-2.93	-3.47	1.79	-4.46	-6.03	-2.84	
	Balachandran et al 2005			0.50	3.57	1.25	6.67		0.24		
	Balachandran et al 2006	0.65	0.84	-0.28	2.40	0.37	5.33	-1.97	-0.23	0.29	
	Martin et al 2012	1.33	0.92	0.21	1.99	0.41	5.95	0.35	-0.85	0.80	
Ulhas	Sahu & Bhosale 1991	-0.32	1.75	1.55	0.43	0.93	4.54	-0.30	0.34	1.51	
	Rokade 2009	2.43	1.64	1.82	3.02			0.45	0.86	1.66	

	Rokade 2009	2.57	1.69	1.73	1.91			0.53	0.68	2.08	
	Rokade 2009	3.24	1.71	1.80	1.03			0.36	0.57	2.10	
Narmada	Subramnian et al 1985	0.07	-0.38	0.29	-1.09	-2.58		-0.81	-0.74	1.26	
	Sanjay& Subramanian 2010	1.92	2.74	2.33	0.88	-1.11	2.90	0.43	0.77	0.79	-0.49
Tapti	Subramnian et al 1985	1.04	1.00	1.75	0.15	-2.58		0.53	-2.27	2.09	
	Sanjay& Subramanian 2010	2.02	2.78	3.12	1.02	-0.26	1.77	0.74	0.80	0.85	-0.40