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INTERNATIONAL JOURNAL OF CURRENT RESEARCH

International Journal of Current Research
Vol.3, Issue, 3, pp.219-229, March, 2011

ISSN: 0975-833X

RESEARCH ARTICLE

EVALUATION OF HEAVY METAL POLLUTION IN THE SURFACE SEDIMENTS OF ENNORE ESTUARY, TAMIL NADU, INDIA

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ARTICLE INFO

Article History:
Received 19th January, 2011
Received in revised form
1st February, 2011
Accepted 7th March, 2011
Published online 30th March, 2011

Key words:
Geoaccumulation index
Cadmium,
Copper, lead and zinc concentrations
Post-monsoon,
Summer,
Pre-monsoon
Monsoon

ABSTRACT

A study was conducted in Ennore estuary for the assessment of the surface sediments with respect to contamination factor and through geoaccumulation index. The data analysed shows that the cadmium, copper, lead and zinc concentrations were above the low interim sediment quality guidelines of ANZECC and below the high interim sediment quality guidelines of ANZECC during post-monsoon, summer, pre-monsoon and monsoon. The surface sediments of the Ennore estuary was highly contaminated with cadmium, copper, lead and zinc. The geoaccumulation index showed that the surface sediments were moderately to extremely polluted. The concentrations of trace metals were high in station 4 followed by station 3, station 2 and station 1 especially during monsoon. The correlation with the stations was highly significant. Zinc concentrations were high followed by copper, lead and cadmium. The major sources for the contamination of the heavy metals would had controlled by anthropogenic inputs from point and non-point sources.

INTRODUCTION

Trace metal contamination poses an environmental problem to aquatic biota (Esen, 2010). Contaminants such as trace metals, phosphorous, pesticides, PCBs and polycyclic aromatic hydrocarbons, get accumulated in sediments and suspended matter of aquatic systems (Jayaprakash *et al.,* 2007).

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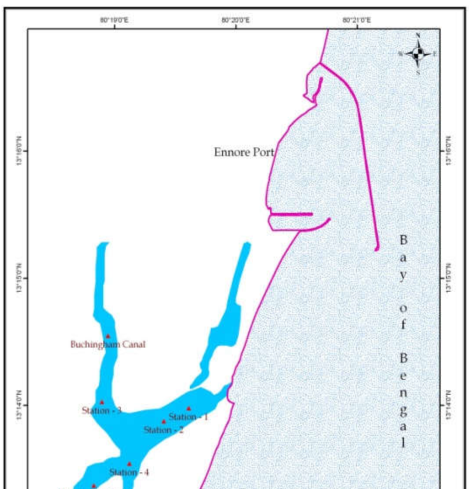
Among the innumerable contaminants, pollution by heavy metals in coastal environment has become a global phenomenon because of their toxicity, persistence for several decades in the environment (Laar *et al.,* 2011). Most of the contaminants leave their finger prints in sediments (Altun *et al.,* 2008). Estuarine sediments are the ultimate sinks for most of the pollutants (Ruzhong *et al.,* 2010; Zhihao *et al*., 2010). When dissolved metals from natural or anthropogenic sources come in contact with saline matter and are removed from water column to bottom deposits (Lee *et al.,* 2010). Heavy metal adsorption increases with decreasing grain size of the sediment. Thus, the metal concentrations are significantly enriched in the fine-grained sediment rich in clay minerals (Xu *et al.,* 2009). Metal assimilation aptitude is in the order of sand < silt < clay, due to increase in surface area, minerals and organic matter as particle size decreased from sand to clay (Rodriguez *et al.,* 2007). Fluxes of trace elements in estuaries and coastal waters are transported to the open ocean and the original composition of seawater is altered (Muthuraj and Jayaprakash, 2007). Sediment associated metals pose a direct risk to detritus and deposit feeding benthic organisms, and may also represent long-term source of contamination to higher trophic levels (Twining *et al.,* 2008).

Metals are used in contamination studies in marine systems due to their relationship with anthropogenic activities (Raju, 2010; Zhihao, 2010). The present study is aimed at evaluating the heavy metal measurements in surface sediments of Ennore creek in accordance with the numerical Interim Sediment Quality Guideline (ISQG) (ANZEEC and ARMCANZ, 2000) using Contamination Factor (CF) analyses (Muthuraj and Jayaprakash, 2007). Impacts of trace metals in sediments are assessed by means of the Geoaccumulation Index (I_{geo},) (Sekabira *et al.,* 2010; Ahiamadjie *et al.,* 2011). Study on distribution and enrichment of trace metals in the sediments is important to assess the probable influence of the heavy industrial activities in this region on marine environment (Mohiuddin *et al.,* 2010; Priadi *et al*., 2011). Ennore creek receives worldwide attention as it becomes an important point polluting source in the Bay of Bengal. Ennore Creek was the sole livelihood for thousands of fishermen families (Rajendran *et al.,* 2004). Ennore creek is an ecologically important but anthropogenic vulnerable site. Ennore creek once cherished for ecological richness is now heading towards a premature ecological death. Major source of metal pollution in the Ennore estuary are the surrounding key industries like Kothari Chemicals, Alkali Chemicals, Madras Refineries, Madras Fertilizers, Petrochemical Industries and such as Ennore Thermal Power Station (ETPS) (Shanthi and Gajendran, 2009). Recent studies have demonstrated that measurements of heavy metal concentrations could be an ideal tool for identifying sources and transport pathways of heavy metals in marine environments (Ahiamadjie *et al.,* 2011). Different metal assessment indices applied to estuarine environment have been developed (Nadia, 2009). The derived indices are used to evaluate the degree to which the sediment-associated chemical status might adversely affect aquatic organisms and are designed to assist sediment assessors responsible for the interpretation of sediment quality (Caeiro *et al.,* 2005). It is also to rank and prioritize the contaminated areas or the contaminants for the further investigation (Farkas *et al.,* 2007).

MATERIALS AND METHODS

Study area and sampling methods

Ennore creek (13°13'54.48" N, 80°19' 26.60" E) (Fig 1) is located in the northeast coast of metropolitan Chennai city, Tamil Nadu, India.



*Map was designed from ARCGIS 9.0

Ennore estuary consists of alluvial tracts and beach dunes, tidal flats and creek in the eastern part. Ennore comprises of lagoons, with salt marshes and backwaters, which are submerged under water during high tide and form an arm of the sea opening in to the Bay of Bengal. The total area of the creek is 2.25 sq km and is nearly 400 m wide. Its channels connect it to the Pulicat lake to the north and to the Kortalaiyar river in the south (Kannan *et al.,* 2007). Ennore coast receives untreated sewage from Royapuram sewage outfall, untreated / treated industrial effluents from Manali Industrial Belt, which houses many chemical industries. The dredging activities in Ennore area result in changes in the landscape, sediment transport, and dust pollution to the coast by quarrying process (Palanisamy *et al.,* 2006).

The surface sediment samples were collected through Peterson grab from the depth of 0.5 to 3.5 m. The sediment samples were immediately transferred to plastic bucket using plastic scoop and stored at 4°C in icebox with sealed labelled plastic bags and transferred to laboratory, stored at -20°C until analysis in deep freezer. Samples for water quality analysis were collected using 1 L Polyethylene-Tereftalate (PET) bottles. The samples were kept at 4°C in icebox, transferred to the laboratory and stored at -20°C until analysis in deep freezer. The surface sediments from the sampling locations of Ennore creek were dried at 50 to 60°C in a hot air oven and grinded in pestle and mortar. For a known quantity (Less than 1 g) of sediment was digested in a Teflon crucible with a solution of concentrated perchloric acid ($HClO_4$) (2 ml) and hydrofluoric acid (HF) (10 ml) to near dryness on the hot plate. Subsequently, a second addition of $HClO_4$ (1 ml) and HF (10 ml) was made and the mixture was evaporated to near dryness. Finally, $HClO_4$ alone was added and the sample was evaporated until white fumes appeared. The colourless residue (less than 8 ml) was dissolved in 5 ml of concentrated nitric acid and diluted with double distilled water to 25 ml in a glass standard flask and transferred to 50 ml Polyethylene-Tereftalate (PET) bottles (Tessier *et al.,* 1979). Trace metal concentrations (cadmium,

SpectraAA 220FS Atomic Absorption Spectrophotometer (AAS). Suitable internal chemical standards (Merck Chemicals, Germany) were used to calibrate the instrument. All the reagents used were analytical grade of high purity. The accuracy of the analytical procedures was assessed using the certified reference material from national research council of Canada for the present study (PACS-2, 2000).

Contamination Factor (CF)

The level of contamination is expressed by the Contamination Factor (CF) (Pekey *et al.,* 2004). CF was calculated as the ratio between the sediment metal content at a given station and the Normal Concentration Levels (NCLs), reflects the metal enrichment in the sediment.

$$CF = \frac{\text{Metal concentration in polluted sediment}}{\text{Background value of metal}}$$

The world crustal average contamination of the trace metals under consideration reported by Wedephol (1995) was used for background values of metals.
The CF was classified into four groups (Pekey *et al.,* 2004):-

1 ≤CF low contamination factor
1 ≤ CF<3 moderate contamination factor
3 ≤ CF<6 considerable contamination factor
6 ≥CF very high contamination factor

Geoaccumulation Index (I_{geo})

The geoaccumulation index was originally defined by Muller (1969) as:

$$I_{geo} = \text{Log2} \; \frac{C}{1.5 \times B}$$

Where, C is the measured sedimentary concentration for metal; B is the background value for the metal; factor of 1.5 was used because of possible variations in background values for a given metal in the environment, as well as very present study, I_{geo} has been calculated using background values for world crustal average metal concentrations as presented by Wedephol (1995). Muller (1969) proposed the descriptive classes for increasing I_{geo} values:

<0	Class 0 - Unpolluted
0–1	Class 1 - From unpolluted to moderately polluted
1–2	Class 2 - Moderately polluted
2–3	Class 3 - From moderately polluted to strongly polluted
3–4	Class 4 - Strongly polluted
4–5	Class 5 - From strongly to extremely polluted
>5	Class 6 - Extremely polluted

RESULTS

The percentage of recovery for trace elements using PACS-2, certified reference material (CRM), 96.66, 104.26, 105.31 and 97.64 per cent for cadmium, copper, lead and zinc respectively were observed (Table 1).

Table 1. Recovery of trace elements in certified reference material (PACS-2)

Element	Certified values (mg/kg)	Measured concentration (mg/kg)	Recovery (%)
Cd	2.11 ±0.15	299.66 ±2.05	96.66
Cu	310 ±12	2.20 ±0.31	104.26
Pb	183 ±8	192.72 ±2.43	105.31
Zn	364 ±23	355.39 ±1.96	97.64

* Measured concentration are the mean and standard deviation of n=24

The one way ANOVA results depict that the measured concentrations in all the stations were highly significant at $P<0.0001$. The concentrations of heavy metals were above the ISGQ low and were below the ISQG high in all the stations (Fig 2, 3, 4 and 5). The heavy metal concentration in the surface sediments of the station 4 was high in monsoon (Figure Fig 2, 3, 4 and 5). Maximum concentrations of cadmium were found in station 4 (9.13µg/g dry weight) during monsoon, 245.67, 94.89, 387.21 µg/g dry weights were the maximum concentrations of copper, lead and zinc in station 4 during monsoon. In station 1 minimum concentrations of 2.75, 108.33, 36.87 and 134.25 µg/g dry weight were measured for cadmium, copper, lead and zinc respectively in pre-monsoon, post-monsoon, summer and not in monsoon (Fig 2, 3, 4 and 5).

The source for heavy metal concentrations in the surface sediments was found in stations 3 and 4. The complete linkage drawn through the resemblance showed distribution of metals in stations 1, 2 and 3 was similar and station 4 contributed the highest concentrations of cadmium, copper, lead and zinc in the surface sediments (Fig 6). The draftsman sketch plotted for the correlation of stations in all the seasons showed strong correlation between stations 1 and 2 ($P<0.0001$) ($\alpha=0.05$), between stations 3 and 4, correlation was highly significant at $P<0.0001$($\alpha=0.05$) (Fig 7).

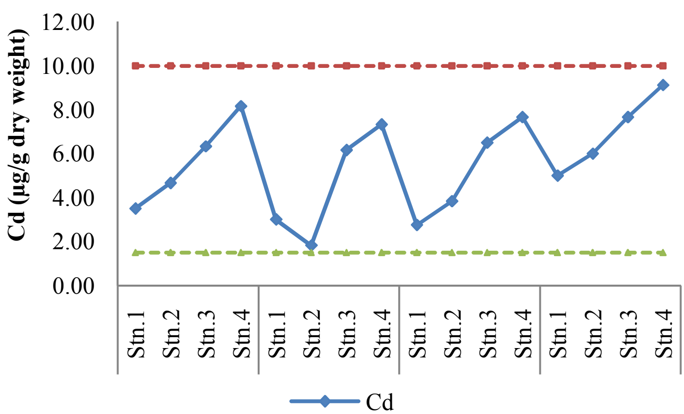


Fig. 2. Seasonal changes of cadmium in the surface sediments of the Ennore creek in stations, 1, 2, 3 and 4; (ISQG- Interim Sediment Quality Guideline (ANZEEC and ARMCANZ, 2000)

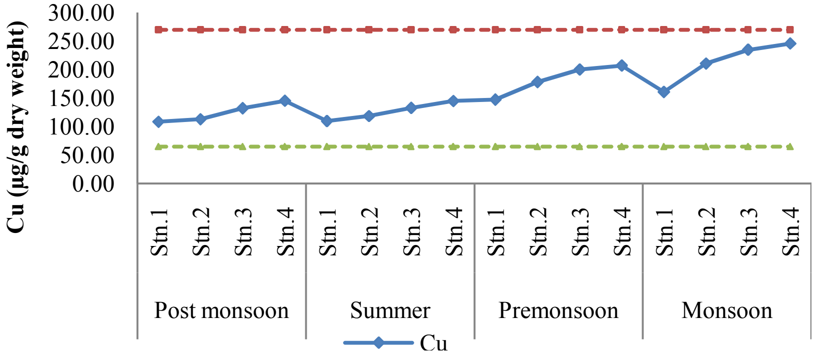


Fig. 3. Seasonal changes of copper in the surface sediments of the Ennore creek in stations, 1, 2, 3 and 4; (ISQG- Interim Sediment Quality Guideline (ANZEEC and ARMCANZ, 2000)

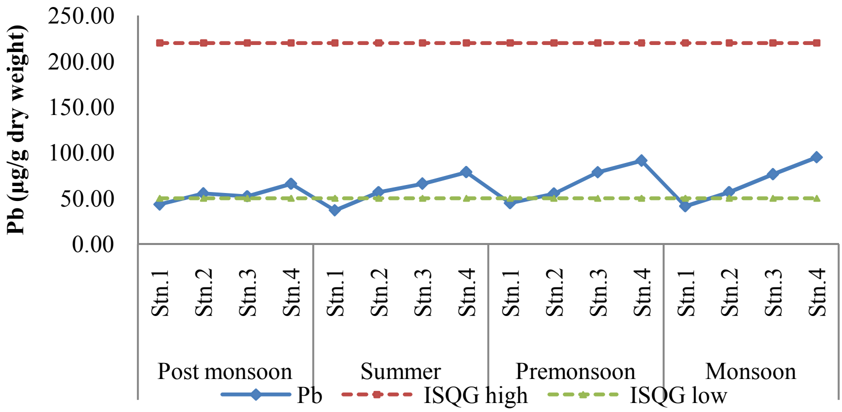


Fig. 4. Seasonal changes of lead in the surface sediments of the Ennore creek in stations, 1, 2, 3 and 4; (ISQG- Interim Sediment Quality Guideline (ANZEEC and ARMCANZ, 2000)

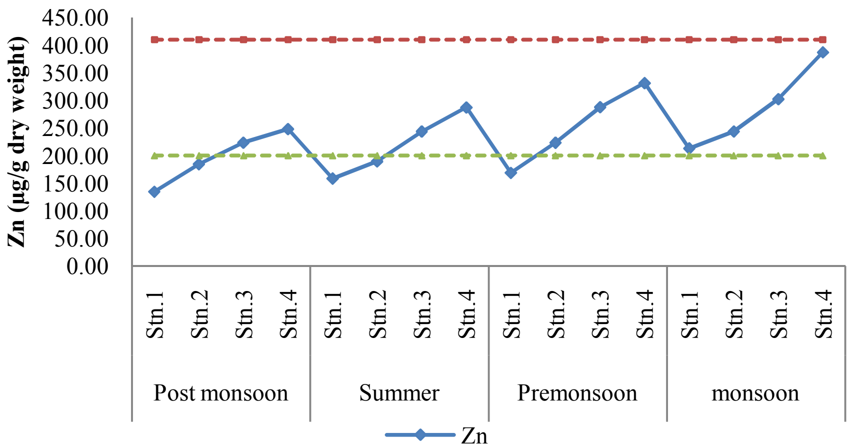


Fig. 5. Seasonal changes of zinc in the surface sediments of the Ennore creek in station, 1, 2, 3 and 4; (ISQG- Interim Sediment Quality Guideline (ANZEEC and ARMCANZ, 2000)

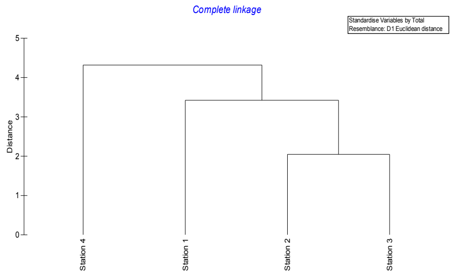


Fig. 6. Complete linkage of heavy metals in the surface sediments at stations (1, 2, 3 and 4)

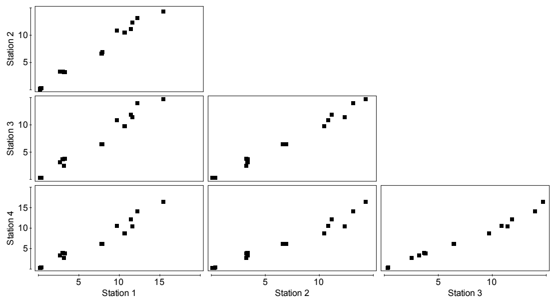


Fig. 7. Draftsman plot of the heavy metals in the surface sediments viewing the strong correlation between stations 1, 2, 3 and 4 (r^2= 0.98, 0.97, 0.95 and 0.96) (α=0.05) at *P*<0.0001 (2-tailed) in Ennore creek during post-monsoon, summer, pre-monsoon and monsoon

Seasons	Station	Cadmium	Copper	lead	Zinc
Postmonsoon	1	35.00	4.33	2.93	2.59
	2	46.67	4.51	3.73	3.54
	3	63.33	5.29	3.53	4.30
	4	81.67	5.81	4.45	4.77
Summer	1	30.00	4.38	2.49	3.04
	2	18.17	4.73	3.84	3.64
	3	61.67	5.31	4.46	4.68
	4	73.33	5.79	5.31	5.52
Premonsoon	1	27.50	5.89	3.03	3.24
	2	38.33	7.13	3.72	4.30
	3	65.00	8.01	5.30	5.53
	4	76.67	8.28	6.16	6.37
Monsoon	1	50.00	6.43	2.79	4.10
	2	60.00	8.42	3.84	4.68
	3	76.67	9.39	5.16	5.82
	4	91.30	9.83	6.41	7.45

Ennore creek

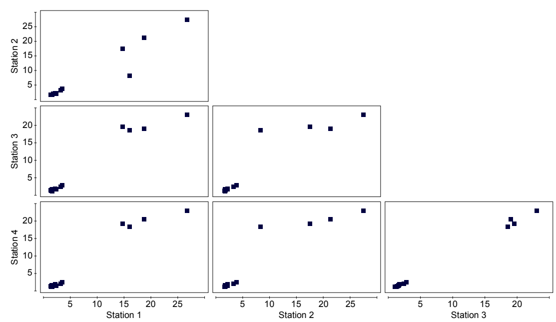


Fig. 8. Draftsman plot of the heavy metals in the surface sediments viewing the strong correlation between the stations 1, 2, 3 and 4 (r^2= 0.98, 0.97, 0.95 and 0.96) (α=0.05) at *P*<0.0001 (2-tailed) in Ennore creek during post-monsoon, summer, pre-monsoon and monsoon

Contamination Factor (CF)

The calculated contamination factor reveals that all stations in Ennore creek had a very high contamination factor of cadmium in the surface sediments during each season. The contamination factor of copper in post monsoon and summer for all stations was considerably contaminated. A very high contamination factor was observed of copper in all stations during premonsoon and monsoon. Stations during post-monsoon and summer were moderately and considerably contaminated with lead. In pre-monsoon and monsoon, stations 1 and 2 were moderately and considerably contaminated with lead and stations 3 and 4 were very highly contaminated (Table 2). Zinc also had the same pattern like lead in all the stations in post-monsoon and summer, moderately and considerably contaminated. Stations 1 and 2 were moderately and considerably contaminated during pre-monsoon and monsoon, stations 3 and 4 were very highly contaminated with zinc. The cadmium had the highest contamination factor of 91.30 in station 4 during monsoon, 9.83, 6.41 and 7.45 were the contamination factor for copper, lead and zinc in station 4 during monsoon. Strong correlation *P*<0.0001(α=0.05) was observed among the stations (1, 2, 3 and 4) (Fig 8).

Geoaccumulation Index (I_{geo})

Concentrations of cadmium in stations 1 and 2 of the surface sediments were strongly polluted to extremely polluted in post-monsoon, summer, pre-monsoon and monsoon. The surface sediments of the stations 3 and 4 were extremely polluted with cadmium in all the seasons. High geoaccumulation index for cadmium of 5.93 was found in station 4 stations in post-monsoon and summer was

Table 3. Geoaccumulation index (I_{geo}) for surface sediments of Ennore creek

Seasons	Station	Cd	Cu	Pb	Zn	Sediment quality
Post monsoon	1	4.54	1.53	0.97	0.79	Moderately polluted to extremely polluted
	2	4.96	1.59	1.31	1.24	
	3	5.40	1.82	1.23	1.52	
	4	5.77	1.95	1.57	1.67	
Summer	1	4.32	1.55	0.73	1.02	
	2	3.60	1.66	1.35	1.28	
	3	5.36	1.82	1.57	1.64	
	4	5.61	1.95	1.82	1.88	
Premonsoon	1	4.20	1.97	1.01	1.11	
	2	4.68	2.25	1.31	1.52	
	3	5.44	2.42	1.82	1.88	
	4	5.68	2.46	2.04	2.09	
Monsoon	1	5.06	2.10	0.90	1.45	
	2	5.32	2.49	1.35	1.64	
	3	5.68	2.65	1.78	1.96	
	4	5.93	2.71	2.10	2.31	

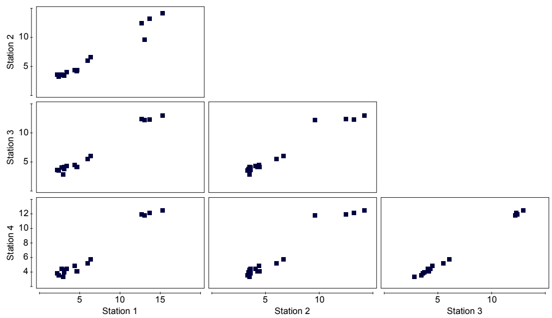


Fig. 9. Draftsman plot of geoaccumulation index (I_{geo}) of heavy metals in the surface sediments viewing the strong correlation between stations 1, 2, 3 and 4 (r^2= 0.96, 0.97, 0.96 and 0.94) (α=0.05) at P<0.0001 (2-tailed) in Ennore creek during post-monsoon, summer, pre-monsoon and monsoon

moderately polluted with copper. Surface sediments were moderately polluted to strongly polluted in all the stations except in station 1 during pre-monsoon. During monsoon all the stations were moderately polluted to strongly polluted with copper (Table 3). Station 1 surface sediments were unpolluted to moderately polluted during post-monsoon and summer. Stations 2, 3 and 4 were moderately polluted with lead. During the pre-monsoon and monsoon the stations 1, 2 and 3 were moderately polluted. In pre-monsoon and monsoon, station 4 was moderately polluted to strongly polluted with lead (Table 3). Surface sediments in station 1 were unpolluted to moderately polluted during post monsoon with zinc; stations 2, 3 and 4 were moderately polluted. and monsoon were moderately polluted. In summer all the stations was moderately polluted. In pre-monsoon and monsoon station 4 surface sediments was strongly polluted with zinc (Table 3). Geoaccumulation index was high for cadmium, copper, lead and zinc (5.93, 2.71, 2.10 and 2.31) was observed in monsoon in station 4. The low geoaccumulation index values for cadmium, copper, lead and zinc were (4.20, 1.53, 0.90 and 0.79) observed during post-monsoon in station 1, during monsoon in station 1 and post-monsoon in station 1 (Table 3). In all the seasons the geoaccumulation index was less in station 1. The correlation study revealed that all the stations in the creek was highly significant at P<0.0001(α=0.05) (2-tailed) (Fig 9). The heavy metal concentration in the surface sediments was much prevalent in all the stations which has made them entitled with moderately polluted to extremely polluted (Table 3).

DISCUSSION

The high concentrations of heavy metals in sediments may not necessarily indicate anthropogenic contamination, because of different background levels in parent materials and sediment properties (Esen, 2010). All studied metals were low in summer and high in monsoon. Al-Saadi *et al.* (2002) reported that the lead, copper and cadmium concentrations were high in spring in Habbaniya lake sediment. Zhihao (2010) reported an average concentration of cadmium 670 µg/g dry weight. Contamination level of cadmium in surface sediments of estuary is low and that of copper and zinc is high on the whole (Nadia *et al.,* 2009). Geoaccumulation index (I_{geo}) was originally devised for use with the global standard shale values as background metal levels, Rubio *et al.* (2000) have shown that the use of regional background values yields more appropriate results. In this study, I_{geo} has been calculated using background values for world crustal average metal concentrations as presented by Wedephol (1995). The index of geoaccumulation consists of seven grades or classes, with I_{geo} as 6 indicating almost a 100-fold enrichment above background values. High values of lead and zinc (435 and 1090 µg/g) sediments of Tamil Nadu Coast. Srinivasalu *et al.* (2007) reported copper in the northern, central sectors indicating a twofold increase in concentration (3320-6230 µg/g) at Kalpakkam (Tamil Nadu, India). Raju (2010) reported the average concentration of total trace elements of copper ranging from 729.33-625.2 µg/g; lead 26.64-24.68 µg/g; cadmium 6.0-4.45 µg/g; zinc 61.5-59.3 µg/g in the marine surface sediments of Bay of Bengal of Chennai, India. Lead is known as markers of paint industries and omnipresent in the study area (Lin *et al.,* 2002). The concentration of lead in the sediments of this region would have originated from the atmospheric deposition of automobile exhaust (Leopold *et al.,* 2008). The sources of pollution include industrial effluents such as food and beverage factories, and also domestic effluents from the population living along Korataliyar river (Jayaprakash *et al.,* 2010). Kehrig *et al.* (2003) reported that metal concentrations in sediment samples have significantly exceeded the natural concentration of heavy metals. Comparisons of sediment metal concentration along with various other coastal regions around the world indicate increase in cadmium and copper concentration in the study area which are well supported by the observations of Muthuraj and Jayaprakash (2007). Sediment samples from estuarine showed the order of metal levels as Zn> Cu> Pb> Cd in Ennore creek. Zulkifli (2009) reported that zinc (130.06 µg/g) was high followed by lead (53.16 µg/g), copper (36.98 µg/g) and cadmium (0.16 µg/g) in Malaysia, which can be compared with the present study. Hwang *et al.* (2008) has reported that heavy metal concentrations in the sediments of San Francisco bay USA of copper ranging from 101-541 µg/g dry weight, cadmium of 0.69- 7.75 µg/g dry weight, lead ranging from 218-750 µg/g dry weight and zinc varying from 280-1430 µg/g dry weight, which connects with the present study in Ennore creek.

Edinger *et al.* (2008) reported lead in the sediments of north Sulawesi, Indonesia from 2.5-12.0 µg/g dry weight; zinc concentration from 34-932.2 µg/g dry weight and copper from 10.0-62.9 µg/g dry weight. Copper concentrations in the sediments of mangrove area of Singapore varied concentrations varied from 11.27-134.13µg/g dry weight and cadmium 0.44µg/g dry weight (Cuong *et al.,* 2005). Lead ranged from 7.10-37.14 µg/g dry weight. Glasby *et al.* (2004) reported copper concentrations ranged from 20-103 µg/g dry weight, zinc varied from 256- 1310 µg/g dry weight, cadmium from 1.2-6.3 µg/g dry weight and lead ranged from 42-167 µg/g dry weight in the sediments of Szczecin lagoon, Singapore. Muthuraj and Jayaprakash (2007) have reported the maximum values of copper (651 µg/g), lead (38 µg/g), zinc (184 µg/g) and cadmium (7.5 µg/g) in the Ennore sediments associated with industrial contamination.

Sediments of Florida bay, USA contained copper ranging from 7-32µg/g dry weight, zinc ranging from 10-48 µg/g dry weight and lead ranging from 3-15.7 µg/g dry weight (Caccia *et al.,* 2003). Pekey (2006) reported copper concentration of 9.6-43.7 µg/g dry weight, 75-271 µg/g dry weight of zinc, 0.005-0.25 µg/g dry weight of cadmium and 22.3-89.4 mg/g dry weight of lead in sediments. Kucuksezgin *et al.* (2006) reported 3.3-8.6 µg/g dry weight of cadmium, 23.8-178 µg/g dry weight of lead, 60.6-139 µg/g dry weight of zinc. Lead concentration in sediments around Singapore ranged from 16 to 250 µg/g (Sin *et al.,* 1991). The concentration of lead in surface sediments of Pattani Bay (Thailand), which was contaminated, was determined to be 79.4-97 µg/g dry weight (Everaarts *et al.,* 1994). Samples of sediments from various locations along the coast of Malaysia analyzed by Ismail *et al.* (1995) contained 6.1-27.5 µg/g of lead. Concentrations of lead in the sediments of the present study are comparable to those reported elsewhere.

CONCLUSION

The sediments of Ennore estuary is extremely contaminated with cadmium, copper, leads and zinc due to the long term anthropogenic activities in and around the estuary through industrial and domestic sewage from point and non-point sources. The application of contamination factor and geoaccumulation index enabled us to find elevated contents of some toxic heavy metals in the sediments of Ennore estuary with cadmium, contaminated followed by station 3, station 2 and station 1. The concentrations of zinc were high followed by copper, lead and cadmium in monsoon. Elevated amounts may enter into the food chain and thus pose a hazard to human and animal health. The high content of toxic metals in the environment may also cause an increase in their content in ground waters as a result of leaching. The concentration of heavy metals in soils could be categorized as contaminated and extremely contaminated.

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