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Heavy Metal Accumulation in Sediments of a Tropical Estuary: A Case from the Southwest Coast of India

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Abstract

Levels of heavy metals (Cr, Cu, Pb, Zn, Fe, Mn), organic carbon content and textural characteristics in the surficial sediments of Akkulam-Veli estuary (SW coast of India) are presented. Spatial variations were in accordance with textural charaterstics and organic matter content. Anthropogenic inputs (sewage, effluent discharge, municipal solid waste, eutrophication and tourism influx) have given rise to a gradient in concentration of metals in estuarine sediments. Extent of heavy metal accumulation in estuarine sediments has been evaluated using Contamination Factor (CF), Index of Geoaccumulation (Igeo) and Pollution Load Index (PLI). High CF and PLI values in the sediments of estuary can be attributed to high inflow of these metals through anthropogenic inputs and the role played by the finer fraction and TOC in the distribution and retention of trace elements in the sediments. Subsequently the accumulation of metals in this estuarine system is aggravated by weak flushing. However Igeo of Fe, Mn, Cr, Cu, Mn remains zero class probably due to the predominance of lithogenic flux.

Keywords: estuary; texture; organic carbon; heavy metals; pollution load index; contamination factor, Index of geoaccumulation; Akkulam-Veli estuary

INTRODUCTION

Estuarine sediments constitute a fundamental step in the pathway of contaminants to the ocean as estuaries filter the fluvially fluxed metals derived from both natural and anthropogenic sources (Larrose et al 2010). Sediment, a complex and dynamic matrix derived from weathering of crustal rocks, can be considered as a data-source of environmental information, because chemical constituents of sediments reflect time integrated information of local respectability (Hackanson and Jansson, 1983; Fostner and Wittman, 1983; Demirak et al. 2006; Cevik et al. 2009; Shirani 2020)

Trace metals are significantly hazardous pollutants in aquatic environments, even at very low concentrations (Salomons and Forstner, 1984; Nriagu and Pacyna, 1988; Salmons and Stigliani, 1995; Birch and Olmos, 2008). In this sense, study of the occurrences, fate and distribution of trace metals in estuarine environments has received major attention, mainly because of the persistent toxic effects of heavy metals (Lacerda *et al.,* 1988; Martin *et al*., 2012) as well as their ability to be accumulated within compartments of these environments (Lacerda and Abrao, 1984, Paneer Selvam *et al.,* 2012).

Heavy metal concentration of the sediments is potentially good indicators of the state of the environment (Forstner and Wittman, 1983). Information on the level of heavy metal pollution in the marine and fresh water environment is important as they cause serious health hazards. These elements are magnified in the food chain and reach human beings causing deleterious effects (Nitta, 1992; Buccolieri *et al*., 2006; Ip *et al*., 2007).

The present paper deals with the analysis of several heavy metals viz., zinc, copper, lead, chromium, iron and manganese, their distribution in Akkulam-Veli estuary. The source of trace elements, their transport path-ways and the various physical and chemical factors that control their distribution in the estuarine are discussed, further the extent of metal concentration in the estuary is assessed using various statistical tools like Contamination factor (CF), Index of geoaccumulation (Igeo) and Pollution load index (PLI). Metal levels were correlated with organic matter contents within sediments, keeping in mind that is a significant scavenger of metals in both coastal and marine environments (Salmons and Stigliani, 1995). Thus, the knowledge on the metal concentrations in sediment is always considered to be a vital face of aquatic environmental assessment studies (Jain *et al*., 2005; Karbassi *et al*., 2008), because it provides data for planners and decision makers for its proper management

STUDY AREA

The Akkulam–Veli (AV) estuary (area = ~1.0 km^2; N. Lat. 8^0 25’ to 8^0 35’ and E. Long. 76^0 50’ to 76^0 58’), a shore perpendicular estuary (Fig. 1.1), located at the NW edge of Thiruvananthapuram city, along the south west coast of India. Rocks and/or sediments exposed at the estuarine shore are progressively older along the northern shoreline, where as, the southern shore is characteristically made of modern beach alluvium, ancient kayal deposits and laterite bluffs. The eastern side is surrounded by lateritic hillocks. The coastal zone enjoys a humid tropical climate with an average annual rainfall of 2048.86 mm, relative humidity of 77.2% and temperature of 27.40 ^{0}C.

Severe environmental degradation is experiencing in the Akkulam-Veli estuary due to municipal waste disposal, effluent discharge, eutrophication, excessive tourism load, developmental activities etc (Nair *et al.,* 1998, Sheela *et al*., 2012, Swarnalatha *et al*., 2013). Lack of proper flushing results in the piling up of pollutants in the estuary. For most of the year, this estuary remains separated from the sea by the development of sand bars. The estuary is partially separated into two by the existence of a bund across it. The western part with a length of 1.25 km and width of 100 m forms the Veli estuary. The eastern part starting from the bund forms the Akkulam estuary. The Veli side is having few important species of mangroves and the Akkulam side is congested with floating aquatic weeds.

The important canals which drain into this estuary include Kannammoola canal, Kulathur canal and Parvathy Puthen ar. Among these, the Kulathur canal joins the estuary at its northern shore. Kannammoola Thodu joins along its eastern shore. The Parvathy Puthen Ar (waterway) connects this estuary with Kadinamkulam estuary in the north and Poonthura estuary in the south. Sewage from Thiruvananthapuram city and drainage from the suburbs flow into the estuary through the Kannammoola Thodu. Seepage from the waste water of Muttathara sewage farm makes the canal water extremely polluted. Two factories, viz., The English India Clays Ltd. and Travancore Titanium Products (TTP) Ltd, are situated in the vicinity of the estuary. The Clay Factory discharges its treated effluents directly to the estuary on the southern shore.

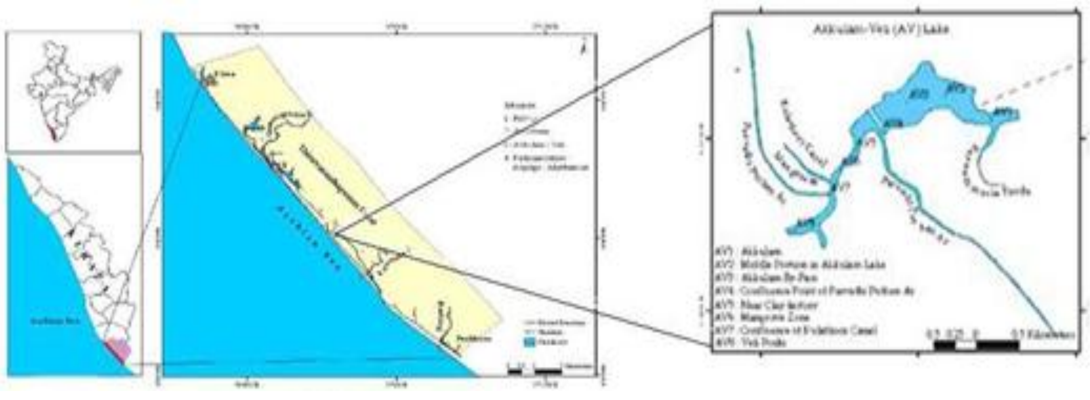

Figure 1. Study area and sampling locations

MATERIALS AND METHODS

Oceanographic surveys were carried out within the Akkulam-Veli estuary, using a transect which included eight (8) sampling stations (Figure 1) during premonsoon. A Van Veen Grab was used to collect surficial sediment samples. Relatively mostundisturbed-core portion of the grab sample was carefully transferred to wide mouth polythene bottle using a plastic spatula and capped air-tightly.

A known quantity of the well-powdered, bulk sample was digested with HF-HNO_3-$HClO_4$ acid mixture using standard procedures for the preparation of solution 'B' (APHA, 1985). From this, the content of heavy metals (Fe, Mn, Zn, Pb, Cu and Cr) was determined using Atomic Absorption Spectrophotometer (AAS) GBC 932 AA model (Rantala and Loring (1975).

The total organic carbon (TOC) in sediment was determined by wet oxidation method of El Wakeel and Riley (1957) technique and particle size analysis was carried out using wet sieving and pipette analysis technique (Lewis, 1984). In order to obtain the geochemical mapping, ArcGIS 8.5 was applied. According to the obtained data the spatial distribution of heavy metals was used to predict the un-sampled sites in the study area in which the inverse distance weighting method was used.

Results and Discussion

Analytical results of texture, TOC and heavy metals in the sediments of the estuary are given in table 1 and 2. Correlation matrices of textural and chemical parameters in the estuary are given in Table 3. Spatial maps are generated using Arc GIS 8.5.

Table.1. Granulometric composition (in wt %) and sediment type (after Pickard, 1971) of Akkulam-Veli estuary

Stations	Sand	Silt	Clay	Mud*	Sediment type
1.	2.30	28.40	69.30	97.70	Silty clay
2.	3.20	28.40	68.40	96.80	Silty clay
3.	4.10	24.50	71.40	95.90	Silty clay
4.	26.20	45.20	28.40	73.60	Clayey silt
5.	42.30	45.20	12.30	57.50	Sandy silt
6.	43.10	38.20	18.10	56.30	Sandy silt
7.	41.50	28.10	30.40	58.50	Silty mud
8.	79.80	3.40	16.80	20.20	Silty sand
Average	30.31	30.17	39.38	69.56	

Table. 2. Heavy metal concentration (Fe in %) and (Cu, Zn, Pb, Cr, Mn in ppm) in the surficial sediments of Akkulam-Veli estuary

Stations	**TOC**	**Cu**	**Mn**	**Pb**	**Zn**	**Cr**	**Fe**
1.	11.79	39.80	192.80	133.40	124.30	129.60	4.23
2.	8.97	43.40	142.30	144.30	139.80	36.40	6.89
3.	8.66	53.60	504.60	142.40	84.60	137.80	7.34
4.	8.66	46.40	138.80	194.20	43.30	108.80	9.86
5.	4.17	88.60	338.60	190.30	16.40	177.60	5.57
6.	7.31	46.30	291.70	94.60	94.60	137.40	4.26
7.	10.27	31.20	175.80	101.30	54.80	96.30	6.86
8.	0.73	37.80	198.60	50.40	88.30	59.40	4.54
Average	7.57	48.38	247.90	131.36	80.76	110.41	4.95

Texture

The spatial distribution of sand, silt and clay content in the estuarine sediments are illustrated in Figure 2 and Table 1. In the estuary, considerable mixing of textural grades is observed spatially. The content of sand varied from 2.30 to 79.80% (avg. 30.30%), and silt from 3.40 to 45.20 % (avg. 30.17%) and clay from 12.30 to 71.40% (avg. 39.38%). Station 8 recorded the highest percentage of sand (79.80%) and st. 1 the lowest (2.30%). The st. 3 showed the maximum clay content (71.40%) which is towards the Akkulam part and with a minimum (20.20%) at estuarine mouth (St. 8). The sand fraction has an inverse relationship with silt (r=-0.81) and clay (r=-0.73) fraction (Table 3). The sediment nomenclature, according to Pickard (1971) is shown in Table 1.

The wide spatial variation in the sediment distribution in Akkulam-Veli indicates additional detrital input from various sources like the Kannammoola Thodu, Parvathy Puthen Ar and Kulathur Thodu. The Akkulam part (i.e., eastern part) registers dominance of clay and silt (mud) over sand owing to the proximity to the Kannammoola Thodu and bank erosion. These sediments fall in the silty clay category. However, the Veli side (i.e., western side) exhibits high content of sand indicating the source mainly from the sand bar.

In general, three dominant categories of sediment types, viz., silty clay with 37% followed by 25% sany silty and 12% each for sandy silt, Silty mud and clayey silt each are dispersed in the estuary. The landward side of the estuary consists of silty clay only. Yet, clayey silt and silty mud sediments are widespread in the remaining part of the estuary. However, the downstream of the estuary (near pozhi) is sand dominated. According to Arunkumar and Sabu Joseph (2007), poor sediment sorting is related to the superposition of different sediment sources and low energy of tidal circulation.

In the estuarine mouth and in the main trunk due to various *insitu* sorting processes (like scouring by tides), the sediments get re-suspended and flushed out to the open sea during the ebb flow, and such physical processes in the waterbody primarily influence the grain size variation of sediments.

Total organic carbon (TOC)

The spatial distribution of TOC in the sediment substratum of the estuary is given in Figure 2 and Table 2. The TOC ranges vary from 0.73 to 11.79% (avg. 7.57%). The maximum TOC content (11.79%) was recorded at the Akkulam part of (st.1) and minimum at bar mouth (st.8). In the present study, OC in all the stations (except st.5 & st.8) exceeds the proposed limit of 5% (Alagarsamy, 1991) and these areas are shown in Figure 2. In this estuary, suspended organic matter received by the lake through land run-off, sewage etc. settles and forms part of bottom sediments. Dead aquatic plants and organisms also contribute much to the organic content of sediments. Enriched with organic matter, the sediments act as good reservoir of nutrients.

Spatial distribution of TOC shows higher concentration in Akkulam side of the estuary (Fig. 2). This indicates the textural control over TOC as evidenced by high silt and clay sediments towards Akkulam part. The negative correlation of TOC with sand (r= -0.77), and positive correlation with silt (r = 0.89) and clay (r = 0.74) support this observation (Table 3). The positive relationship of TOC with clay and silt indicates size dependent scavenging. It was experimentally proved that various clay minerals absorb substantial amount of organic matter formed by decomposition of phytoplankton (Muller & Suess, 1979; Rodríguez-Barroso et al. 2010). Added to this co-sedimentation of organic matter with fine particles due to similarity in settling velocity help for the sympathetic relationship of organic matter content to fine particles of the sediments (Valdes et al. 2005). Increase in TOC with decrease in particle size is also attributed to the increase in surface area of fine particles.

Contribution from Kannammoola thodu and debris of aquatic weeds are also more at Akkulam part and these play a major role in controlling TOC content. The confluence of Parvathy Puthen ar and Kulathur Thodu in Veli part also recorded high content of TOC.

However, low OC is recorded in the Veli part (estuarine mouth) due to the sandy nature of bottom sediments. From the study it is evident that low level of tidal mixing and seasonal monsoonal dilution are not adequate to enable the self-purifying mechanism of the estuary, and further the carrying capacity of estuary is vehemently reduced due to the enormous load contributed by sewage drain and autochthonous source.

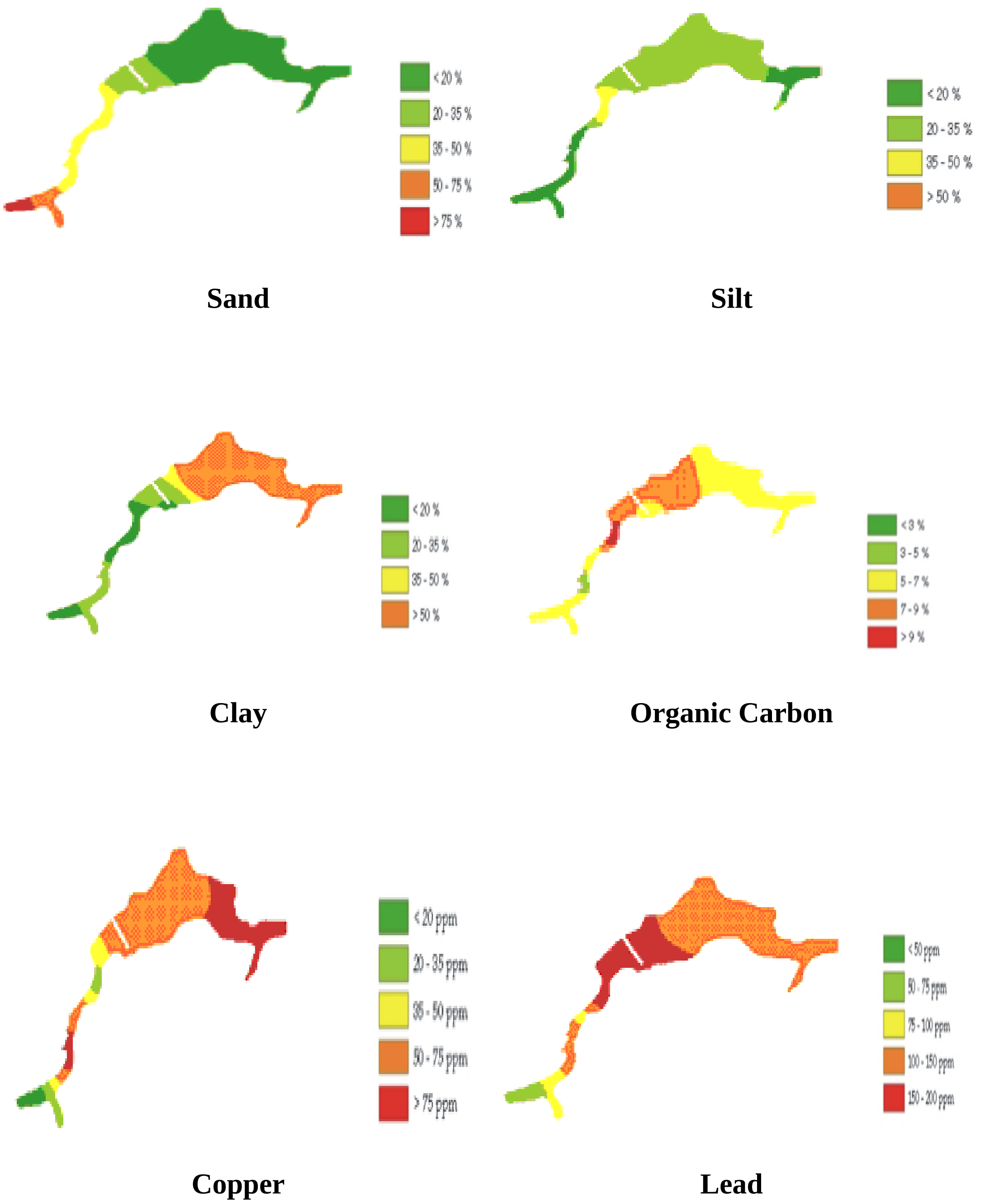

Figure 2. Spatial variation sand, silt, clay, OC, Cu and Pb of Akkulam-Veli estuary

Heavy metals

The distribution of Cu, Cr, Zn, Pb, Fe and Mn in estuarine sediments are Showed in Table 2. Cu fluctuates considerably from 31.2 to 88.6 ppm (avg.= 48.38) . In the estuary, the Akkulam part of the estuarine sediments shows comparatively higher Cu (Fig. 2) due to the fine texture and high TOC, as evidenced by the positive correlation of Cu with clay (r= 0.75) and silt (r = 0.60), and negative correlation with sand (r = -0.51).

Table. 3. Pearson correlation matrix for the geochemical attributes of sediments of the estuary.

Parameters	Clay	Cr	Cu	Fe	Mn	TOC	Pb	Sand	Silt	Zn
Clay	1.00									
Cr	0.47	1.00								
Cu	0.75	0.24	1.00							
Fe	0.88	0.50	0.49	1.00						
Mn	0.71	0.62	0.27	0.60	1.00					
TOC	0.74	0.60	0.59	0.63	0.58	1.00				
Pb	0.70	0.43	0.43	0.55	0.11	0.52	1.00			
Sand	-0.73	-0.06	-0.51	-0.46	-0.09	-0.77	-0.52	1.00		
Silt	0.06	0.24	0.60	0.29	-0.14	0.89	0.71	-0.81	1.00	
Zn	0.44	-0.56	0.07	-0.20	-0.20	0.58	-0.40	-0.33	-0.08	1.00

The ranges of Zn in the sediments are between 16.4 to 139.4 ppm (avg. = 80.76 ppm), However, Akkulam part of the estuarine sediments showed a comparatively high content of Zn than downstream sediments. This could be due to the fine nature of sediment and enrichment of OC at Akkulam sediments. Zinc showed positive correlation with OC (r= 0.54), which indicates that the enrichment of zinc in the present sediments takes place through biological process or reaching through organic matter.

The ranges and averages of Pb are between 50.4 to 194.2 ppm (avg. = 131.36 ppm) and the spatial distribution shown in Figure 2. However, Cr content is high, and it ranges between 31.2 and 88.6 ppm (avg. =48.38). High content of Cr is due to the input of sewage through Parvathy Puthen ar and Kannammoola thodu.Fe fluctuates considerably from 4.23 to 9.86% (avg.=4.95%), whereas the Mn ranges between 142.30 and 194.20 ppm (avg.=247.90ppm).

In order to consider the environmental assessment of Akkulam-Veli estuary, a range of pollution indicators, such as Contamination Factor (CF), geoaccumulation index (Igeo) and pollution load index (PLI) were estimated. All indices (CF, Igeo and PLI) used in the present study specify various levels of contaminatio,n in the studied areas although their outcomes are mostly uniform. The reason behind varying level of contamination is that each index assesses different pollutants of importance for their evaluation.

Contamination Factor, CF

The degree of contamination of aquatic sediments was quantified earlier by comparing the elemental concentration with uncontaminated background levels (Forstner and Muller, 1973; Nikofarova and Smirnova, 1975).

In the present study, the method suggested by Forstner and Muller (1973) was used for the calculation of contamination factor (CF) using the following equation.

$$CF = \frac{\text{Metal concentration in polluted sediments}}{\text{Background value of the metal or average shale}}$$

The CF < 1 refers to low contamination, 1 ≤ CF ≤ 3 means moderate contamination, 3 ≤ CF ≤ 6 indicates considerable contamination, and CF > 6 indicates very high contamination (Hakanson, 1980). A CF, calculated as the ratio between the sediment metal content at a given station and the normal concentration levels, reflects the metal enrichment in the sediment when CF > 1 for a particular metal, it means that the sediment is contaminated by the element, and if CF < 1, then there is no metal enrichment by natural or anthropogenic inputs. While calculating the CF of the sediments in the study area, we have taken the world crustal average contamination of the trace metals under consideration reported by of background values.

The CF for Fe in the estuary ranges from 0.40 to 2.14 ppm; Mn 0.16 to 0.59; Cu from 0.69 to 1.97; Pb 2.52 to 9.71; Cr 0.40 to 1.97 and Zn 0.17 to 1.47 ppm (Table 5).

Concentration of metals like Fe, Pb, Cr, Zn, Cu in Akkulam-Veli estuary is much higher (>1) than those in average Shale (Turekain and Wedephol, 1961) indicating contamination.

From the CF values it can be noted that all the metals which were studied, showed contamination and of these very high values were observed for Pb and Cu, indicating high build up of these metals in the sediments. This highest value of CF can be attributed to their incorporation by anthropogenic inputs mainly from urban domestic sewage and land run-off.

Index of Geoaccumulation, Igeo

In order to compare present day heavy metal concentration in the sediments of rivers and estuaries with pre-civilization background values, and for a quantitative measurement of possible contamination in these sediments, an "Index of geoaccumulation" (Igeo) proposed by Muller (1979) was used for quantification of metal accumulation in polluted sediments by following equation,

$$I_{geo}=\log_2 (C_n/1.5B_n)$$

Where, C_n is the measured concentration of heavy metal 'n' of the sediments and B_n is the geochemical background concentration of element (n)- either measured directly in pre-civilization sediments or taken from the literature (average shale value – Turekian and Wedepohl, 1961). The factor 1.5 is introduced to include possible variation of the background values that are due to lithogenic variations (Chakravarty and Patgiri, 2009), as well as very small anthropogenic influences (Qingjie and Jun, 2008). Muller (1981) proposed seven grades or classes of the geo-accumulation index. Different geo-accumulation index classes are given in (Table 4); the I_{geo} class 0 indicates the absence of contamination while the I_{geo} class 6 represents the upper limit of the contamination. The highest class 6 (very strong contamination reflects) 100-fold enrichment of the metals relative to their background values (Harikumar and Jisha, 2010). On the other hand, Karbassi et al. (2008) mentioned that I_{geo} failed to various degrees to designate the intensity of pollution.

The calculated Igeo of sediments of the estuary is given in Table 5. The Igeo class of the heavy metals (Fe, Mn, Cr, Cu, Mn) in the estuary remains in the uncontaminated category, class zero at all the station probably due to the predominance of lithogenic flux, whereas (Igeo) values of Pb greater than two indicating the moderately contaminated category. Higher values of Pb may suggest the anthropogenic source.

Table.4. Seven Igeo-classes based on the numerical value of the index (Muller, 1979).

Igeo	Igeo-Class	Designation of sediment quality
>5	6	Extremely contaminated
4-5	5	Strongly/extremely contaminated
3-4	4	Strong contaminated
2-3	3	Moderately/strong contaminated
1-2	2	Moderately contaminated
0-1	1	Uncontaminated/moderately contaminated
0	0	Uncontaminated

Pollution Load Index, PLI

Tomlinson et al., (1980) employed Pollution Load Index (PLI), to assess the extent of pollution by heavy metals in the environment. The range and class are same as Igeo.

Pollution Load Index, PLI

$$\text{PLI} = \text{N}\sqrt{\text{Product of N number of CF values}}$$

The pollution load index provides simple comparative means for assessing a site or area quality (0.0 indicates perfection, 1.0 indicates only baseline levels of pollutants present and >1.0 indicates progressive deterioration of the site, Tomlinson et al., 1980, Cabrera et al., 1999). The PLI of the estuary show contamination, and the values ranging from 0.74 to1.56 (avg. = 1.8) for AV (Table 5). The PLI of the zone or the whole investigated area of the estuary however, was 1.8 which confirmed that the lake sediments are polluted. Once heavy metals are discharged into the estuarine and coastal waters, they rapidly become associated with particulates and are incorporated in bottom sediments (Muraleedharan and Ramachandran, 2002). The PLI can provide some understanding to the public of the area about the quality of a component of their environment. It also indicates the trend spatially and temporarily. In addition, it also provides valuable information and advice to the policy and decision makers on the pollution level of the area (Mohiuddin et al., 2010, Harikumar and Jisha, 2010).

Table. 5. Contamination Factor and PLI of the estuary

Stations	**Contamination Factor**						**Pollution Load Index**
	Fe	**Cu**	**Pb**	**Zn**	**Cr**	**Mn**	
1.	0.99	0.88	6.67	1.31	1.44	0.23	1.23
2.	0.70	0.96	7.22	1.47	0.40	0.17	0.98
3.	2.14	1.19	7.12	0.89	1.53	0.59	1.56
4.	0.94	1.03	9.71	0.46	1.21	0.16	0.97
5.	1.50	1.97	9.52	0.17	1.97	0.40	1.25
6.	0.93	1.03	4.73	1.00	1.53	0.34	1.15
7.	0.40	0.69	5.07	0.58	1.07	0.21	0.81
8.	0.55	0.84	2.52	0.93	0.66	0.23	0.74
Igeo.	0.51	0.39	2.69	-0.03	0.03	-1.34	

CONCLUSION

The heavy metal concentration in the sediments from the Akkulam-Veli estuary shows a combined influence of mineralogy, TOC content and texture. The estuary is polluted by heavy metals, and very high values are observed by Pb and Cu indicating high build up of these metals in the sediments. The highest value of CF can be attributed to their incorporation by anthropogenic inputs mainly from urban domestic sewage and land run-off. Based on the value of CF, PLI and Igeo, Akkulam-Veli is identified as a potential 'hot spot' in terms of heavy metals. The sediment heavy metal contamination of this estuary is a cause for concern as these metals may undergo bioaccumulation and affect benthic organisms. The complex nature and flow restriction favouring accumulation of pollutants have transformed this estuary into a delicately poised system. It is important to determine the source of these heavy metals and to manage their input into the estuary so that their concentration in the sediment does not reach toxic levels.

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