

**ORIGINAL ARTICLE****RISK ASSESSMENT ON HEAVY METAL ENRICHMENT IN THE SURFACE SEDIMENTS  
OF CUDDALORE, SOUTH EAST COAST OF INDIA****<sup>1</sup>K. Karikalan, <sup>1</sup>S.P. Mohan, and <sup>2</sup>S. Srinivasalu**<sup>1</sup>Department of Geology, University of Madras, Guindy Campus, Chennai – 600025, India.<sup>2</sup>Department of Geology, Anna University, Chennai – 600025.

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**ABSTRACT**

The surface sediments were analyzed on grain size, carbonates (CaCO<sub>3</sub>), organic matter (OM) to ascertain the environmental conditions of off Cuddalore along the south east coastal tract, India. Studies on the accumulation of heavy metals (Mn, Cr, Cu, Ni, Pb, Zn) in the sediments of Cuddalore, East coast of India were carried out using Atomic Absorption Spectrophotometer. In surface sediments collected, the order was Ni > Mn > Cr = Zn > Pb > Cu > Cd. The high concentration of sand in the surface sediments is due to high tidal influence in the study area. The tidal currents significantly play a role in separating the fines from the river mouth. The content of organic matter (OM) does not reveal a significant difference in amount.

**Keywords:** Trace elements, CaCO<sub>3</sub>, Organic matter, Cuddalore.**1. INTRODUCTION**

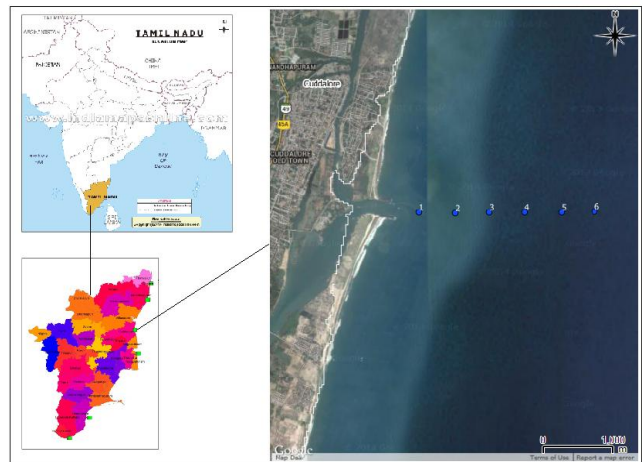
Human kind is becoming aware of the complexity of nature and the delicate balance that exists in the global ecosystem. Every action taken towards modifying the environment has countless repercussions. Geoscientists seek to understand these complex interactions so that an assessment of the present condition of the ambient environment may be known and measures may be taken in the hopes of preventing or minimizing future degradation.

In this study a methodological acid leachable extraction approach for assessment of heavy metals is proposed to achieve the hypothesis

**2. MATERIALS AND METHODS****Study Area**

The present study covers Cuddalore area, which is situated in the coastal stretch of the East coast, Tamil Nadu and the study area was selected based on the industrial activity, land use, oceanographic conditions and geological conditions. The study area forms the part of the topo – sheet bearing number 58 M/ 10 and M/ 14 (Fig 1). The study area experiences a tropical sub - humid type of climate with an annual mean temperature of 26° C and annual precipitation of 1160mm.

11°43'N-79°49'E

**Figure 1. Map showing the Study area and sampling location****Sampling stations**

The area of investigation in the Bay of Bengal and sampling locations are shown in the (Figure 1). Total number of six surface sediment samples were collected in a transect pattern in Cuddalore using Petersen grab sampler. The samples were collected using the coastal research vessel 'Sagar Paschimi' in collaboration with National Institute of Ocean Technology, Taramai, Chennai.

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### Geochemical analysis of sediments

Geochemical analysis of sediments involves procedures for their decomposition and related chemical measurements. Samples were collected in a transect pattern. The samples were analyzed for acid leachable trace elements study after proper preliminary treatment. All the elemental analyses were carried out on total unfractionated sediment samples. This means that no separation of specific sediment sizes was performed prior to the chemical analyses. To determine acid leachable trace metal concentrations of collected sediment samples, method by Loring and Rantala, 1992 is followed.

### Sand silt and clay determination by Pipette method

Inexpensive size analysis of naturally occurring fine sediments or of material obtained from wet sieving of sediment containing significant fines is most satisfactorily achieved by the use of the pipette method (Krumbein and Pettijohn, 1938; Galehouse, 1971 and Folk, 1974). This technique relies on the fact that in a dilute suspension, particle settles through a column of water at velocities which are dependent up on their size. As particles decrease in sizes they become increasingly cohesive as surface ionic charges grow on relative significance (McManus, 1988). To determine the size of the flocculated sediments it is necessary to introduce a dispersing agent. Sodium Hexa-metaphosphate is the commonest dispersing agent used for sedimentation analysis. A 20 g portion of each sample is treated with dilute 4 N HCl to remove carbonate materials and is washed through a 230 ASTM sieve until clear solution passed through. A pinch of sodium-hexa-meta-phosphate (dispersant) is added to isolate clay bounded strongly with silt. The washings are collected carefully in a container and transferred to a 1000 ml measuring jar of de-ionized water. Care is taken to keep the washing to a minimum, so that it did not exceed 1000 ml, and thoroughly mixed by means of manual stirrer which travels from the base to the top of the fluid column. As suspension begins, a timing device is started. Pipette is inserted slowly into the fluid so that its inlet is 20 cm below the surface of fluid and a volume of 20 ml is withdrawn after 1 minute. In each case the pipette is gently inserted and withdrawn from the fluids in order to minimize disturbances. Withdrawn aliquot transferred to pre-weighed beaker and dried at 90°C. Then weight of the withdrawn sample is measured to the nearest milligram. In calculating the weight of sediment retained in each withdrawn aliquot, allowance also given for the dispersant.

The other raw sub-sample are separated as sand (>63  $\mu$ m) and mud (<63  $\mu$ m) fractions through wet sieving with the help of 230 ASTM mesh. The separated sand fraction is dried and calculated separately as sand fraction. The mud fraction of each sample is collected in a 1 liter graduated measuring jar and filled with distilled water. The suspension in the measuring jar is then well agitated using a stirring device, in order to obtain a uniform distribution of particles. As soon as the agitation is stopped, the time is noted and after exactly 2 hours and 3 minutes, a 20 cc pipette is inserted up to a depth of 10 cm in the solution and from that level the sample is withdrawn with uniform suction. The pipette out sample is transferred to a 50 cc beaker and dried in an oven. Care is taken to prevent boiling and splattering. Later, the material retained on the sieve is dried and weighed as the sand fraction. The silt and clay that are collected in washings are further discriminated as silt and clay fractions by the pipette

method. After complete drying the weights of sand, silt and clay are converted into weight percentages and plotted on a trilinear diagram after Trefethen (1950).

### Calcium Carbonate ( $\text{CaCO}_3$ )

Calcium carbonate contents are determined by digesting the sample in HCl according to the procedures of Muller (1979) as follows: 5 g of sediment sample in 150 ml of beaker is added 100 ml of 1 N HCl (87.28 ml of con. HCl make up to 1 l with distilled water) and mixed thoroughly for 1 hour. Then mixture is allowed to settle down and overlying acidic solution is siphoned of (20 ml) into a conical flask. After adding 6-8 drops of Bromothymol Blue indicator. The solution is titrated against 1N NaOH and the disappearance of blue color is noted as end point. Blank titration is also carried out using same procedure.

### Organic Matter (OM)

The readily oxidizable organic carbon content is determined by oxidizing the sediments in  $\text{K}_2\text{Cr}_2\text{O}_7$  and Sulphuric Acid and then titrated with Ferrous Ammonium Sulphate solution by using Diphenylamine indicator to get clear end point as discussed in detail by Gaudette et al. (1974). A 0.5 g of dried and powdered sediment sample is placed in a 500 ml conical flask. Exactly 10 ml of 1 N  $\text{K}_2\text{Cr}_2\text{O}_7$  solution and 20 ml of concentrated  $\text{H}_2\text{SO}_4$  with  $\text{Ag}_2\text{SO}_4$  (dissolved 2.5 g  $\text{Ag}_2\text{SO}_4$ ) in 1 L of  $\text{H}_2\text{SO}_4$  are added with the help of burette and mixed the mixture by gently rotating the flask for 1 minute. The mixture is allowed to stand for 30 minutes. After 30 minutes, 200 ml of distilled water; 10 ml of 85%  $\text{HPO}_3$  and 0.2 g NaF are added sequentially. Then, 15 drops (0.5 ml) of the diphenylamine indicator are added to the sample flask. Finally, the solution mixture is back titrated with 0.5 N Ferrous Ammonium Sulphate solutions to a one-drop brilliant green end point. A blank without sediment is run along with each new batch of the samples.

### Acid leachable trace metal analysis

The acid leachable fraction extracts almost the whole degree of fraction of elements simply absorbed by sediments depicting the contamination of an area (Agemian and Chau, 1976; Taliadouri, 1995). Extraction of acid leachable metals was done by taking 5 g of dry sediment sample in a 100 ml plastic bottle in which 75 ml of 0.5 N HCl was added and after mechanically shaking for 16 h it is filtered with Whatman Grade "A" filter paper. The filtered solution was analyzed for acid leachable Fe, Mn, Cr, Cu, Ni, Co, Pb, Zn and Cd in flame Atomic Absorption Spectroscopy (GBC 920 *plus*) and graphite furnace was in use for determination of Cd due to the low level.

Chemicals of high purity (Merck, Germany) were used and standard solutions were prepared from 1000 mg/L stock solution of each metal. Analysis of triplicates for every fifth sample suggests that precision of analysis lies in the following coefficient of variation Fe 3.1%, Mn 1.5%, Cr 3.4%, Cu 1.9%, Ni 2.4%, Co 2.4%, Pb 2.1%, Zn 3.9% and Cd 3.1%.

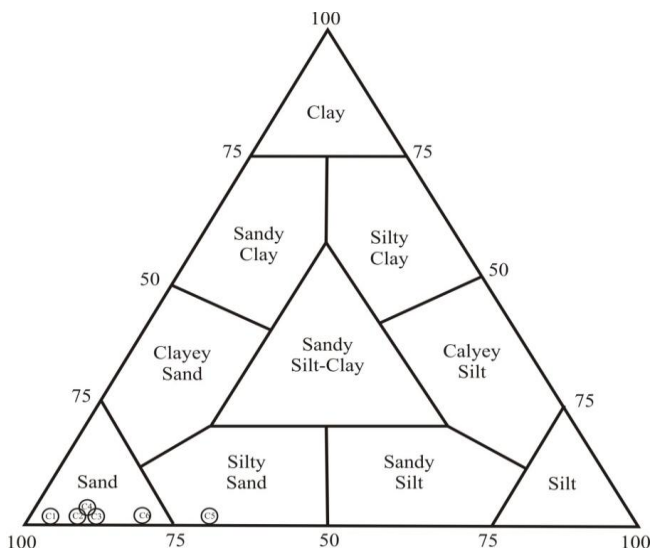
## 3. RESULTS AND DISCUSSION

A total of 6 surface sediment samples were collected from a transect perpendicular to the coast. The textural characters, calcium carbonate, organic matter distribution and the sediment type in surface are given in Table 1 and the sample stations are shown in Figure 1. Sand content (Figure 2)

dominate in the Cuddalore region with sand content ranging between 70.48% and 96.40% with an average of 86.37% indicating a relatively higher energy regime that prevents sedimentation of fine-grained particles and silt between 3.09% and 28.52% with an average of 12.62% (Table 1). Clay content is less than 1.5%.

**Table 1 Sample locations, textural characters, calcium carbonate and organic matter distribution in surface sediments of the Cuddalore**

Sample ID	Sand %	Silt %	Clay %	CaCO <sub>3</sub> %	OM %	Sediment Type
C1	96.40	3.09	0.50	16.75	4.24	Sand
C2	91.00	8.00	1.00	25.00	3.48	Sand
C3	89.96	9.54	0.50	24.50	4.46	Sand
C4	89.28	9.22	1.50	21.25	5.22	Sand
C5	70.48	28.52	1.00	32.50	3.48	Silty sand
C6	81.12	17.38	1.50	36.25	1.96	Sand
Minimum	70.48	3.09	0.50	16.75	1.96	
Maximum	96.40	28.52	1.00	36.25	5.22	
Average	86.37	12.62	1.00	26.04	3.80	



**Figure 2 Ternary plots of sand-silt-clay components of surface sediments in Cuddalore. Field limits are from Shepard (1954)**

The CaCO<sub>3</sub> in the surface sediments ranges from 16.75% to 36.25% with an average of 26.04% indicating moderate enrichment of carbonates in the reduced layers due to precipitation and increase in alkalinity generated by Sulphate reduction (Gaillard et al., 1989). The OM content in the sediments is low and ranges from 0.5% to 0.9%. Similar low OM content was also reported from the Krishna Godavari basin (Subba Rao, 1960) and in the Tuticorin shelf region (Jonathan and Ram Mohan, 2003) of the Gulf of Mannar indicating that the shelf sediments of east coast of India are generally low in OM. The strong positive correlation between CaCO<sub>3</sub> with silt ( $r=0.82$ ) (Table 3) suggest that the carbonate and the silt content are related to each other.

The concentration of acid leachable trace metal distribution in surface sediments off the Cuddalore are given in the Table 2

In the analyzed sample, the Fe concentration ranges from 1445  $\mu\text{g g}^{-1}$  to 5754  $\mu\text{g g}^{-1}$  with an average concentration of 3029  $\mu\text{g g}^{-1}$ . The minimum concentration is found in station C1 and the highest is found at C4. The concentration gradually increases towards the sea from C1 to C4 and then it decreases. The Mn concentration varies from 65  $\mu\text{g g}^{-1}$  at station C1 to 200  $\mu\text{g g}^{-1}$  at C4. It also shows the similar trend like the Fe. The Cr concentration ranges from the minimum of 4  $\mu\text{g g}^{-1}$  at C2 and the maximum of 12  $\mu\text{g g}^{-1}$  at C6 with an average concentration of 8  $\mu\text{g g}^{-1}$ . The Cu concentration ranges from the minimum of 1  $\mu\text{g g}^{-1}$  at C2, C5 and C6 and the maximum of 13  $\mu\text{g g}^{-1}$  at C4 with an average concentration of 4  $\mu\text{g g}^{-1}$ . The Ni concentration varies from 49  $\mu\text{g g}^{-1}$  at station C5 to 243  $\mu\text{g g}^{-1}$  at C4. The Co concentration ranges from the minimum of 2  $\mu\text{g g}^{-1}$  at C1 and C5 and the maximum of 8  $\mu\text{g g}^{-1}$  at C2 with an average concentration of 5  $\mu\text{g g}^{-1}$ . The Pb concentration ranges from the minimum of 6  $\mu\text{g g}^{-1}$  at C3 and the maximum of 10  $\mu\text{g g}^{-1}$  at C5 with an average concentration of 7  $\mu\text{g g}^{-1}$ . The Zn concentration from 3  $\mu\text{g g}^{-1}$  at station C2 and C5 to 25  $\mu\text{g g}^{-1}$  at C4. The Cd concentration is less than 1  $\mu\text{g g}^{-1}$ .

The Fe shows very good correlation with Mn ( $r=0.90$ ), Cu ( $r=0.85$ ) and Zn ( $r=0.86$ ). Mn shows good relationship with Cu ( $r=0.96$ ), Ni ( $r=0.93$ ) and Zn ( $r=0.99$ ). Cu is strongly correlated with Ni ( $r=0.96$ ) and Zn ( $r=0.98$ ). Ni also shows good correlation with Zn ( $r=0.95$ ).

In order to establish the geochemical processes and enrichment of metals (Bridgman 1992), correlation coefficient matrix and R-mode factor analysis with Varimax rotation were applied separately to know the processes operating in different environmental conditions (SPSS 1995). To test the elemental associations, the compositional data is subjected to Varimax factor loading. The first factor accounts for 43% of the total variance (Figure 3) and depicts the strong influence of OM, Fe, Mn, Cu, Ni, and Zn. This explains the general coherence of transition metals probably resulting from a single source. The second factor accounts for 34% of variance incorporating strong positive loading by mud, CaCO<sub>3</sub>, Cr and Cd. The third factor accounts for 14% of the total variance and is loaded only by silt and Pb (Figure 3). These factor loading results are further supported by the elemental associations obtained by regression analysis.

#### Normalization and Evaluation of Sediment Pollution

It is difficult to make an overall assessment of the degree of metal contamination in estuarine and marine sediments. This is a consequence of variations in analytical procedures between studies and the presence of an unknown natural background in the sediments. In the present study, Geoaccumulation index ( $I_{geo}$ ) was used to normalize the total metal concentration and to evaluate the extent of pollution. It was originally defined by Muller (1979) in order to determine and define metals contamination in sediments, by comparing current concentrations with pre-industrial levels. Muller has distinguished seven classes of index (Muller 1981) which is given in Table 4.

Factor 1.5 is used because of possible variations in background values for a given metal in the environment, as well as very small anthropogenic influences.

Table 2 Acid leachable trace metal distribution in surface sediments off the Cuddalore

Sample ID	Concentrations in $\mu\text{g g}^{-1}$								
	Fe	Mn	Cr	Cu	Ni	Co	Pb	Zn	Cd
C1	1445.0	66.0	5.0	3.0	126.0	2.0	7.0	5.0	0.0
C2	2215.0	65.0	4.0	1.0	63.0	8.0	7.0	3.0	1.0
C3	3090.0	67.0	6.0	4.0	76.0	7.0	6.0	5.0	0.0
C4	5754.0	200.0	11.0	13.0	243.0	4.0	7.0	25.0	1.0
C5	3388.0	70.0	7.0	1.0	49.0	2.0	10.0	3.0	1.0
C6	2281.0	65.0	12.0	1.0	58.0	6.0	7.0	5.0	1.0
Minimum	1445.0	65.0	4.0	1.0	49.0	2.0	6.0	3.0	0.0
Maximum	5754.0	200.0	12.0	13.0	243.0	8.0	10.0	25.0	1.0
Average	3029.0	89.0	8.0	4.0	103.0	5.0	7.0	8.0	0.0

Table 3 Correlation coefficient matrix ( $R^2$ ) of acid leachable elements in surface sediments off the Cuddalore, southeast coast of India ( $p < 0.05$ )

	Sand	Silt	Clay	CaCO <sub>3</sub>	OM	Fe	Mn	Cr	Cu	Ni	Co	Pb	Zn	Cd
Sand	1.00													
Silt	-1.00	1.00												
Clay	-0.39	0.35	1.00											
CaCO <sub>3</sub>	-0.83	0.82	0.51	1.00										
OM	0.47	-0.46	-0.31	-0.80	1.00									
Fe	-0.19	0.17	0.52	-0.05	0.57	1.00								
Mn	0.13	-0.16	0.54	-0.32	0.62	0.90	1.00							
Cr	-0.35	0.31	0.82	0.47	-0.23	0.52	0.52	1.00						
Cu	0.31	-0.33	0.34	-0.48	0.76	0.85	0.96	0.43	1.00					
Ni	0.43	-0.45	0.30	-0.60	0.73	0.70	0.93	0.36	0.96	1.00				
Co	0.26	-0.27	0.09	0.19	-0.21	-0.11	-0.18	-0.08	-0.15	-0.28	1.00			
Pb	-0.82	0.82	0.16	0.42	-0.22	0.10	-0.09	0.00	-0.27	-0.26	-0.61	1.00		
Zn	0.21	-0.24	0.52	-0.35	0.62	0.86	0.99	0.56	0.98	0.95	-0.16	-0.19	1.00	
Cd	-0.57	0.54	0.87	0.58	-0.38	0.39	0.32	0.47	0.06	0.02	0.10	0.47	0.24	1.00

Table 4 Muller's classification for the geoaccumulation index (Muller, 1981)

$I_{geo}$ value	Class	Quality of sediment
$\leq 0$	0	Unpolluted
0-1	1	From unpolluted to moderately polluted
1-2	2	Moderately polluted
2-3	3	From moderately to strongly polluted
3-4	4	Strongly polluted
4-5	5	From strongly to extremely polluted
$> 5$	6	Extremely polluted

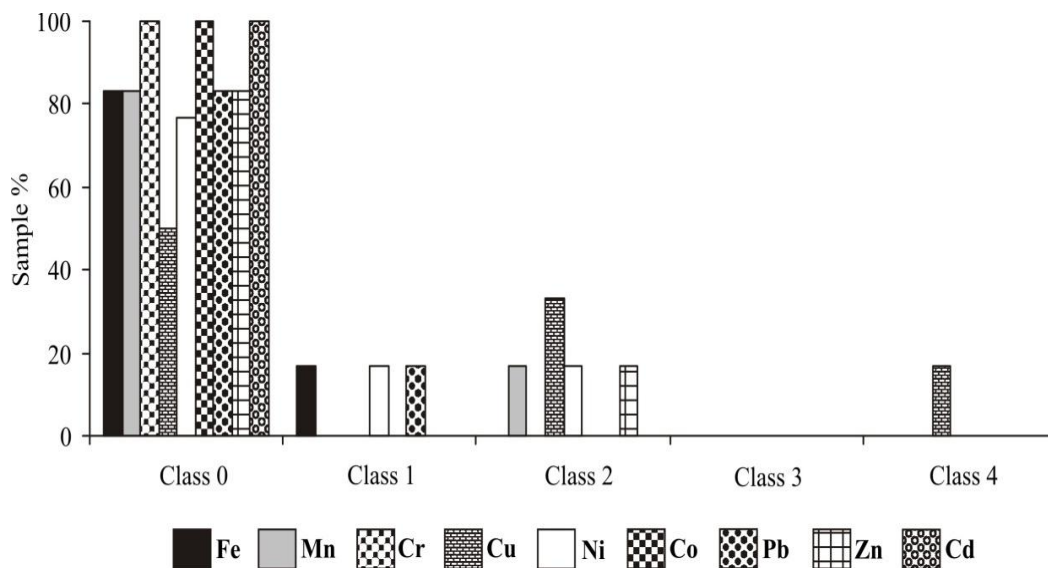


Figure 3 Percentage of samples in Muller's classes for Fe, Mn, Cr, Cu, Ni, Pb and Zn using the background values for Cuddalore

A number of methods are available for assessing sediment quality and each has its own pros and cons (Caeiro et al., 2005). Enrichment indices (EF and  $I_{geo}$ ) provide a simple way of comparing the extent of metal pollution among different sites. However, the main criticism against calculation of such indices is that they do not aggregate all the contaminants into one variable and it becomes difficult to compare the sites and interpret the data when multiple metals are in question (Caeiro et al., 2005). The enrichment factor further suffers from the fact that there are no threshold values for denoting the pollution level.  $I_{geo}$  on the other hand provides a classification system for the degree of pollution when compared to the background (Ruiz, 2001):  $I_{geo} < 1$  – unpolluted;  $1 < I_{geo} < 2$  – very lightly polluted;  $2 < I_{geo} < 3$  – lightly polluted;  $3 < I_{geo} < 4$  – moderately polluted;  $4 < I_{geo} < 5$  – highly polluted;  $I_{geo} > 5$  – very highly polluted.

Geoaccumulation index ( $I_{geo}$ ) has been calculated for analyzed metals. Muller has distinguished seven classes of geoaccumulation index (Muller, 1981) (Table 4). In Figure 4, we show sample percentages in Muller's classes (Muller 1981) for Fe, Mn, Cr, Cu, Ni, Co, Pb, Zn and Cd using the background values. Figure 4 shows that, for all the background values, 100% of samples fall in class 0 for Cr, Co and Cd. Therefore, Cuddalore is not a contaminated area for these metals. For Fe, 83% of the samples fall in class 0 and 17% in class 1, by using our background values. For Mn, about 83% of samples are included in class 0 and 17% falls under class 2. Cu concentrations fall for 50% in class 0, for 33% in class 2 and 17% in class 4, suggesting that the quality of sediments in this area is strongly polluted. Ni concentrations fall for 77% in class 0, for 17% in class 1 and 16% in class 2, suggesting that the quality of sediments in this area is moderately polluted. Pb concentrations fall for 83% in class 0, for 17% in class 1. Zn concentrations fall for 83% in class 0, for 17% in class 2, suggesting that the quality of sediments in this area is moderately polluted.

In conclusion geoaccumulation index allows us to deduce the quality of sediments in Cuddalore moderately polluted to strongly polluted for metals considered.

## 4. CONCLUSION AND RECOMMENDATIONS

The major conclusions derived from this study are given below. The heavy metal concentration for the environmentally sensitive area (Cuddalore) has been evaluated and compared.

- The sediments in Cuddalore are mostly sandy silt and silty sand. The correlation matrix exhibits poor correlation between these components and minor metals indicating an insignificant role of  $CaCO_3$  and OM in enriching minor metals.
- Sediment quality is from moderately polluted to strongly polluted with reference to Cu and moderately polluted with reference to Mn, Ni and Zn.

Presently as the coastal areas of Tamil Nadu State are subjected to developments with construction of harbor, chemical industries, power plants and other large industries, exploration of petroleum in both onshore and offshore regions, the assessment of dissolved trace metals, physico chemical parameter conditions acting in the region are to be studied in detail and continuous pollution monitoring studies should be carried out for improving the aquatic environment. These types of studies are required to implement the regulations from the international conventions which intend to reduce the trace metal discharge in to the sea.

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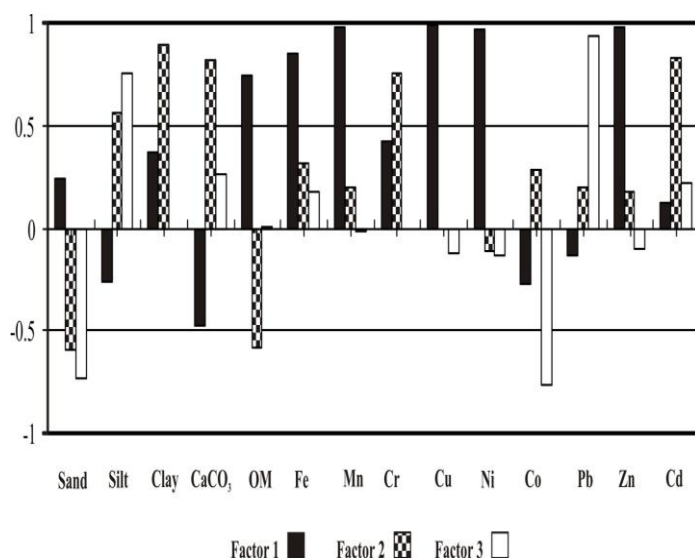


Figure 4 Results of factor analysis (R-mode) showing three primary factors (F1, F2 and F3) for surface sediments

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