

Distribution of Heavy Metals in the Marine Sediments of Various Sites in Karaichalli Island, Tuticorin, Gulf of Mannar, India

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Abstract The study of heavy metals in the sediments of coral islands is significant in understanding the distribution levels and the cause of anthropogenic impacts along the marine environment. A large number of heavy metals is discharged through domestic and industrial effluents along the coastal area of the southeast coast of India. In this work the sediments from Karaichalli island of Tuticorin were collected and the samples subjected to a total digestion technique and major and trace elements concentration estimated. To interpret and assess the contamination status for heavy metals in sediments, four metal pollution indices were assessed using an enrichment factor, a geo-accumulation index, contamination factor and pollution load index. The results were treated with SPSS 16.0 software for statistical analysis like Correlation matrix and Principal component analysis. The average metal accumulation levels in coral samples of the study area is in the following order: Fe > Mn > Zn > Cu > Ni > Pb > Cr > Cd > As > Co.

Keywords Karaichalli island · Trace elements · Correlation matrix · Principal component analysis

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1 Introduction

Coral reefs are quite common in the tropical seas and oceans. They have been designated as the marine equivalent of the tropical rain forest. Due to rapid development in industrialization and modern expansion along the coast of Tuticorin, the discharge of heavy metals in the coastal environment has increased. Heavy metals are naturally occurring elements in the environment and differ in concentrations along the earth crust. Even though the heavy metals are natural components of our earth, their concentrations have been drastically altered by human activities [1]. Besides the natural process several metals compounds enter the aquatic environment through various factors like industrial effluent, domestic sewage disposal, dumping, surface run-off, atmospheric fallout, etc [2, 3]. It was a huge task for the industries to increase the productivity and profit without contaminating the environment. Heavy metals are considered to be the most poisonous due to their high toxicity, abundance, and are found to have harmful effect on living organisms as they are hard to metabolize [4, 5]. Higher heavy metals concentrations in the coastal environment are the pointers of anthropogenic influence and prospective danger to the natural environment [6]. Coastal sediments act as the ultimate sink for metals that are discharged into the coastal environment [7]. The spatial distribution of heavy metals in marine sediments is of major importance for determining the pollution history of an aquatic environment [8, 9] and also provides basic information for identifying the possible sources of contamination and to depict the areas where the metal concentration exceeds the threshold values and the strategies of site remediation [10]. Therefore, understanding the mechanisms of accumulation and geochemical distribution of heavy metals in sediments is crucial for the management of coastal environments.

2 Materials and Methods

2.1 Description of the Study Area

The Gulf Of Mannar consists of several coral reef islands and of the 21 coral islands present along the coastline between Tuticorin and Pamban, most of them are close (2 to 18 km) to the mainland. The Gulf of Mannar is a transitional zone lying between the Arabian Sea and the Indian Ocean. The study area Karaichalli island is 18 km away from the northeast of Tuticorin, in the Gulf of Mannar. The area under investigation consists of many major chemical industries like SPIC, a copper smelting plant, Dharangadhara Chemicals, salt pans and several small scale industrial units found in the Thoothukudi SIPCOT complex. Tuticorin is one of the major harbors and the movement of the ships may release oil effluents and petrochemical products into the sea. Ash from a thermal power station is directly dumped into the sea and the other industries also discharge their wastes into the sea. The Google pictorial map of Karaichalli island of Tuticorin is shown in Fig. 1.

2.2 Sample Collection and Sediment Analysis

The sediments were collected using stainless steel coated equipment and transferred to polythene bags to avoid the metal contaminations. The sediment samples were initially washed with sodium hypochloride for 24 h, and then with distilled water. The samples were oven-dried at 60 °C and powdered in an agate mortar. The Loring and Rantala (1992) method was followed for analyzing trace elements in the sediment samples. 0.5 g of the powdered sample was

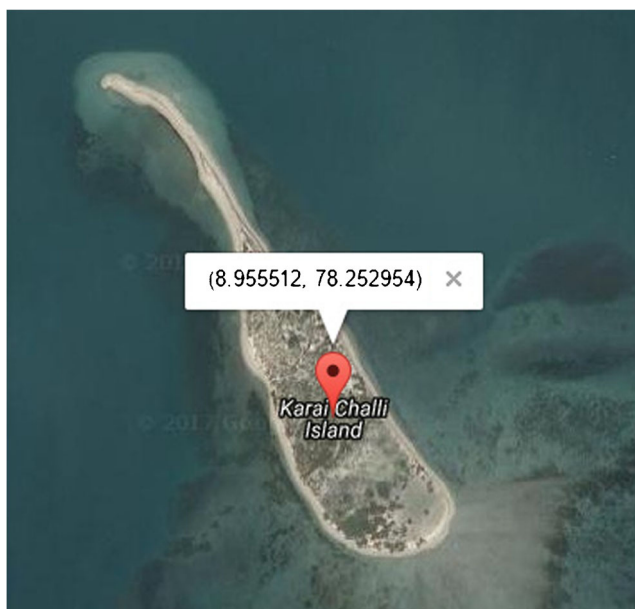


Fig. 1 Google pictorial map of Karaichalli island of Tuticorin

digested in 25 ml of HCl and 15 ml HNO₃–HClO₄ (5:1) acid mixture at 80 °C [11]. The digested sample was centrifuged at 200 rpm and the centrifuged liquid was used for the determination of trace elements using an Inductively Coupled Plasma Atomic Emission Spectrometer (ISA JOBIN YVON 24 MODEL). Metals like Fe, Zn, Cu, As, Cd, Mn, Ni, Pb, Co, Cr were analyzed in each sample. The heavy metal concentrations in sediments were expressed as ppm.

2.3 Pollution Indices

Different methods have been developed to measure the extent of heavy metal pollution in the sediments [12]. In the present study various pollution indicators such as Contamination factor (CF), Pollution Load Index (PLI), Enrichment factor (EF) and Geo-accumulation index (I_{geo}) were used to express the heavy metals contamination in the sediments and its possible sources. For successful interpretation of geochemical data, the choice of background values plays an important role. Many authors have used the average crustal abundance data as reference baselines [13]. In the present study, the average metal concentrations in sediments reported by Taylor, 1964 were taken as the representative background values in the sediments [14].

2.4 Statistical Analysis

Statistical analysis was performed using SPSS 16.0 software. Principle Component Analysis (PCA) was carried out to determine the most significant factors which influence heavy metal concentration in sediment samples and Pearson correlation matrix was performed to evaluate the relation between various metals.

3 Result and Discussion

The concentration of trace elements in sediments helps to monitor terrestrial inputs and anthropogenic pollution. The result of minimum, maximum, mean and standard deviation of trace elements concentration in sediments are shown in Table 1. The concentration of Fe ranges from 7612 to 7713 ppm with an average value of 7670. Fe is the fourth most abundant element in the earth's crust (after oxygen, silicon and aluminum) and its concentration may be due to the weathering of rocks. The concentration of Zn ranges from 121.18 to 127.32 ppm with an average of 124.58. The concentration of As ranges from 1.46 to 2.91 ppm with an average of 2.23. As is a naturally occurring toxic element and it is widely distributed in natural ecosystems. The concentration of Cd ranges from 1.96 to 2.93 ppm with an average value of 2.42. The concentrations of Pb range widely from 31.99 to 36.84 ppm with an average value of

Table 1 Trace element concentrations (ppm) in Karaichalli island of Tuticorin, India

Sampling sites	Fe	Zn	Cu	As	Cd	Mn	Ni	Pb	Co	Cr
KCT 1	7612	123.01	48.92	1.46	1.96	686.30	35.13	32.95	0.56	11.66
KCT 2	7713	126.42	55.63	2.29	2.39	712.66	39.98	34.62	0.99	10.38
KCT 3	7659	127.04	47.82	1.66	2.93	676.68	36.31	33.33	0.83	10.88
KCT 4	7634	123.34	54.38	2.58	2.16	682.41	36.74	35.98	0.96	11.85
KCT 5	7694	126.68	52.92	2.13	1.96	714.65	37.08	35.64	0.95	11.95
KCT 6	7710	127.32	56.97	2.35	2.11	699.98	36.96	31.99	0.97	11.83
KCT 7	7693	122.64	55.48	2.12	2.61	680.84	38.79	33.52	1.06	11.47
KCT 8	7655	123.28	49.54	2.03	2.85	702.36	36.37	34.86	0.97	10.89
KCT 9	7638	124.44	53.22	2.91	2.25	724.44	35.96	35.96	1.19	10.89
KCT 10	7695	121.18	54.19	2.34	2.66	686.42	38.23	36.84	0.75	11.56
KCT 11	7639	126.76	53.91	2.36	2.58	682.28	39.43	35.35	1.23	10.99
KCT 12	7698	122.88	51.36	2.52	2.53	672.88	37.16	35.39	1.06	10.98
Minimum	7612	121.18	47.82	1.46	1.96	672.88	35.13	31.99	0.56	10.38
Maximum	7713	127.32	56.97	2.91	2.93	724.44	39.98	36.84	1.23	11.95
Mean	7670.00	124.58	52.86	2.23	2.42	693.49	37.35	34.70	0.96	11.28

34.7. Pb and Cd in sediments are well known indicators of anthropogenic activity [15]. Severe contamination of Cd gives rise to itai-itai disease [16]. The concentration of Mn ranges widely from 672.88 to 724.44 ppm with an average value of 693.49. The concentration of Ni ranges widely from 35.13 to 39.98 ppm with an average value of 37.35. The concentration of Co ranges from 0.56 to 1.23 ppm the average being 0.96. The concentration of Cr ranges widely from 10.38 to 11.95 ppm with an average value of 11.28. Cu was detected in all the sediments and its concentration ranges from 47.82 to 56.97 ppm with an average of 52.86.

3.1 Enrichment Factor (EF)

EF is a normalized method to separate metals of natural changeability from the metal fraction that is related with

sediments because of anthropogenic activities. In this study, Fe has been used as a normalizing element because of its enormous availability. The EF for each element was calculated to estimate the anthropogenic effects on heavy metals in sediments using the following equation [17, 18].

$$EF = (M_c|Fe_c) / (M_b|Fe_b)$$

Where M_c and Fe_c are the examined metal and iron concentration in the sediments; M_b and Fe_b are the background values of examined metal and iron respectively. EF groupings recognized are as follows: $EF < 1$, background concentration; 1–2, depletion to minimal enrichment; 2–5, moderate enrichment; 5–20, significant enrichment; 20–40, very high enrichment; and >40 , extremely high enrichment [19]. The average EF value for Co and Cr is <1 , which shows that

Table 2 Enrichment factors of heavy metals

Sampling sites	Zn	Cu	As	Cd	Mn	Ni	Pb	Co	Cr
KCT 1	13.00	6.58	7.20	72.48	5.34	3.46	19.50	0.17	0.86
KCT 2	13.18	7.38	11.14	87.23	5.48	3.89	20.22	0.29	0.76
KCT 3	13.34	6.39	8.13	107.69	5.24	3.56	19.60	0.24	0.80
KCT 4	12.99	7.29	12.68	79.65	5.30	3.61	21.23	0.28	0.87
KCT 5	13.24	7.04	10.39	71.71	5.50	3.62	20.86	0.28	0.87
KCT 6	13.28	7.56	11.44	77.04	5.38	3.60	18.69	0.28	0.86
KCT 7	12.82	7.38	10.34	95.50	5.24	3.79	19.62	0.31	0.84
KCT 8	12.95	6.62	9.95	104.80	5.44	3.57	20.51	0.29	0.80
KCT 9	13.10	7.13	14.30	82.92	5.62	3.53	21.21	0.35	0.80
KCT 10	12.67	7.21	11.41	97.31	5.29	3.73	21.56	0.22	0.85
KCT 11	13.35	7.22	11.60	95.07	5.29	3.87	20.84	0.36	0.81
KCT 12	12.84	6.83	12.29	92.52	5.18	3.62	20.71	0.31	0.803
Average	13.06	7.05	10.91	88.66	5.36	3.65	20.38	0.28	0.828

these metals have no enrichment and Ni shows moderate enrichment in the sediments. Significant enrichment was observed for Mn, Zn, Cu and As. Very high enrichment was observed for Pb and extremely high enrichment was found for Cd. The order of average EF values of metals was: Cd > Pb > Zn > As > Cu > Mn > Ni > Cr > Co. The calculated EF of heavy metals is given in Table 2.

3.2 Geo-accumulation Index (I_{geo})

Geo-accumulation index is used to find the possible metal enhancement in marine sediments and it is calculated by comparing the current and crustal average value of metal concentrations by using the following equation:

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

where, C_n is measured concentration of metal; B_n is background value of the same metal. In order to reduce the possible variation in background values for a given metal in the environment, as well as very small anthropogenic influences, the concentrations of the geochemical background value are multiplied by the factor 1.5 [20]. According to Muller (1979), a sediment can be classified as, non-polluted ($I_{geo} < 1$), very slightly polluted ($1 < I_{geo} < 2$), slightly polluted ($2 < I_{geo} < 3$), moderately polluted ($3 < I_{geo} < 4$), highly polluted ($4 < I_{geo} < 5$) and very highly polluted ($I_{geo} > 5$). The geo-accumulation index of Fe, Cu, As, Mn, Ni, Co, Cr, Zn and Pb shows that sediments were not polluted by these metals and Cd shows slight pollution in all the sites. The calculated I_{geo} of heavy metals is given in Table 3.

3.3 Contamination Factor (CF)

The Contamination Factor is used to express the level of contamination by each metal in the sediment of the sample. It is calculated by the following equation

$$CF = C_{metal} / C_{background}$$

Where $C_{background}$ is the concentration of the background value of the metal and C_{metal} is the concentration of the metal in the sediments. Four grades are considered for the classification of sediment pollution, $CF < 1$ refers to low contamination, $1 < CF < 3$ indicates moderate contamination, $3 < CF < 6$ implies considerable contamination, and $CF > 6$ denotes high contamination [21]. The average CF of Fe, Mn, Ni, Co and Cr was < 1 which indicates that sediment samples are slightly contaminated by these metals. The average CFs of Zn, Pb, As > 1 shows that the sediments are moderately contaminated. The average CF of Cd was greater than 6, which indicates the sediments are highly contaminated by Cd. The average CFs of metals are in the following order: Cd > Pb > Zn > As > Cu > Mn > Ni > Fe > Cr > Co.

3.4 PLI-Pollution Load Index

The level of heavy metal pollution was assessed by using the Pollution Load Index (PLI) and it is computed using the following equation [22].

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

where CF_n is the CF value of metal n. PLI values were deduced in two levels such that $PLI > 1$ is polluted and PLI

Table 3 Geo-accumulation index of heavy metals

Sampling sites	Fe	Zn	Cu	As	Cd	Mn	Ni	Pb	Co	Cr
KCT 1	-3.472	0.23	-0.75	-0.62	2.71	-1.05	-1.68	0.81	-6.07	-3.69
KCT 2	-3.453	0.27	-0.57	0.03	2.99	-1.00	-1.49	0.88	-5.24	-3.85
KCT 3	-3.463	0.27	-0.79	-0.44	3.29	-1.07	-1.63	0.83	-5.50	-3.79
KCT 4	-3.468	0.23	-0.60	0.20	2.85	-1.06	-1.61	0.94	-5.29	-3.66
KCT 5	-3.456	0.27	-0.64	-0.08	2.71	-1.00	-1.60	0.93	-5.30	-3.65
KCT 6	-3.453	0.28	-0.53	0.06	2.81	-1.03	-1.61	0.77	-5.27	-3.66
KCT 7	-3.456	0.22	-0.57	-0.09	3.12	-1.07	-1.54	0.84	-5.14	-3.71
KCT 8	-3.464	0.23	-0.74	-0.15	3.25	-1.02	-1.63	0.89	-5.27	-3.78
KCT 9	-3.467	0.25	-0.63	0.37	2.91	-0.98	-1.65	0.94	-4.98	-3.78
KCT 10	-3.456	0.21	-0.61	0.06	3.15	-1.05	-1.56	0.97	-5.64	-3.70
KCT 11	-3.467	0.27	-0.61	0.07	3.10	-1.06	-1.51	0.91	-4.93	-3.77
KCT 12	-3.456	0.23	-0.68	0.16	3.08	-1.08	-1.60	0.92	-5.14	-3.77
Average	-3.461	0.25	-0.644	-0.04	2.997	-1.04	-1.592	0.89	-5.315	-3.73

Table 4 Correlation coefficient matrix of trace element concentration in Karaichalli island of Tuticorin, India

	Fe	Zn	Cu	As	Cd	Mn	Ni	Pb	Co	Cr
Fe	1									
Zn	0.158	1								
Cu	0.526	0.117	1							
As	0.195	−0.066	0.609*	1						
Cd	0.106	−0.146	−0.308	−0.115	1					
Mn	0.122	0.315	0.255	0.324	−0.399	1				
Ni	0.538	0.158	0.623*	0.258	0.269	−0.049	1			
Pb	−0.065	−0.379	0.082	0.577*	0.096	0.152	0.197	1		
Co	0.146	0.294	0.436	0.722**	0.16	0.231	0.432	0.276	1	
Cr	−0.045	−0.162	0.211	−0.102	−0.58	−0.132	−0.326	−0.093	−0.365	1

*Correlation is significant at the 0.05 level (2-tailed)

**Correlation is significant at the 0.01 level (2-tailed)

< 1 is unpolluted. The calculated PLI values for all the samples was less than 1 which indicates that the studied area has not been severely affected by anthropogenic contamination.

3.5 Correlation Matrix

The Pearson correlation matrix gives the relation among the heavy metals present in sediment samples as given in Table 4. The correlation that was significant at $p < 0.05$ is noted

between As vs Cu, Ni vs Cu, and Pb vs As. Also the correlation between Cu vs Co was significant at $p < 0.01$ level.

3.6 Factor Analysis

The PCA was applied to further analyse the sources of heavy metals. The sources of heavy metals may be classified as either natural or anthropogenic, where the natural sources are mainly coastal erosion and rock weathering

Table 5 Varimax rotated factor matrix of partially extractable metals in sediments of the study area

Component	Rotation Sums of Squared Loadings			
	Eigen value	% of Variance	Cumulative %	
1	2.261	22.606	22.606	
2	2.190	21.904	44.510	
3	1.926	19.260	63.770	
4	1.720	17.200	80.970	
Elements	Component			
	Factor 1	Factor 2	Factor 3	Factor 4
Fe	0.826	−0.079	0.012	0.037
Zn	0.195	−0.352	0.135	0.799
Cu	0.804	0.340	−0.349	0.193
As	0.295	0.867	−0.054	0.218
Cd	0.078	−0.038	0.855	−0.370
Mn	−0.053	0.306	−0.188	0.751
Ni	0.824	0.168	0.332	−0.017
Pb	−0.075	0.853	0.080	−0.221
Co	0.338	0.573	0.355	0.433
Cr	0.010	−0.118	−0.880	−0.242

Extraction Method: Principal Component Analysis

Rotation Method: Varimax with Kaiser Normalization

Rotation converged in 5 iterations

and anthropogenic contributions are mainly from industrial effluents and domestic sewage discharge and other human activities [23–26]. The factor analysis identified, four principal components accounting to 80.97% of the total variance of the parameters and is shown in Table 5. The first principal component (PC 1) was used to infer the factors contributing to sediment concentration in the area. Component 1 was heavily loaded with high concentration of Fe, Cu, Ni. This indicates more quantities of these metals originated from natural sources such as rock weathering and coastal erosion. Component 2 accounts for 21.904% and had high positive loadings for As, Pb and Co. As was mainly derived from

products of human activities such as pesticides, fertilizers and industrial effluents. Co and Pb were partly derived from anthropogenic sources. Component 3 accounts for 19.26% and shows high positive loading for Cd and high negative loading for Cr, indicating that both metals may be derived from different pollution sources. Component 4 accounts for 17.2% and shows high positive loading for Zn and Mn representing the combination of both anthropogenic sources and natural sources.

From principal component analysis the group plot was plotted for PC 1 vs PC 2, PC 2 vs PC 3 and PC 1 vs PC 3 as shown in Fig. 2a, b and c respectively. Since the first three factors are considered as major factors which influence the metal concentration variation in sediment samples, the group plot was limited to the first three factors obtained from principal component analysis. The PC 1 vs PC 2 plot shows the elemental variation in two groups where group 1 and 2 elements are (Zn, Cd, Cr, Fe) and (Pb, Mn) respectively. The PC 2 vs PC 3 plot shows the elemental variation in two groups where group 1 and 2 elements are (Pb, As, Mn, Cu, Cr) and (Fe, Cd, Zn) respectively. The PC 1 vs PC 3 plot shows the elemental variation in two groups where group 1 and 2 elements are (As, Cr, Cu, Fe, Mn) and (Pb, Cd) respectively. The PC 2 vs PC 3 plot shows a similar grouping as results obtained from the PC 1 vs PC 3 plot. The cumulative results obtained from the group plot indicate that As, Cu, Fe, Cr, Cd, Mn and Pb come under the same group, whereas Ni, Co and Zn lie in different groups. This explains the metal concentration variation in sediment samples [27–29].

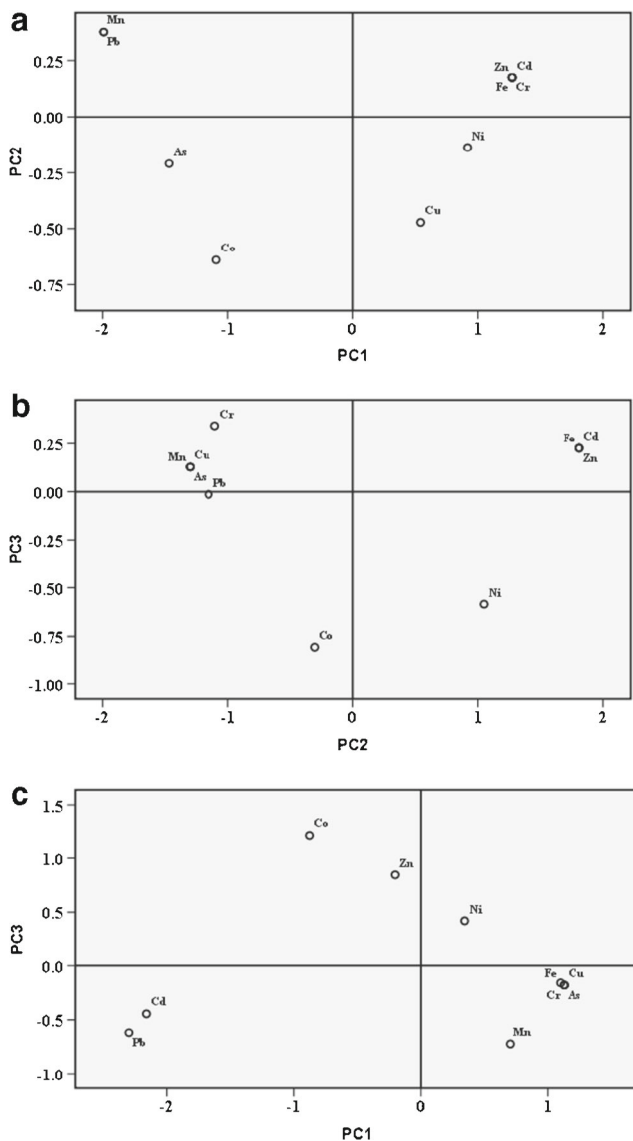


Fig. 2 a Principal component analysis group plot of PC 1 vs PC 2 (Derived stimulus configuration, Euclidean distance model). b Principal component analysis group plot of PC 2 vs PC 3 (Derived stimulus configuration, Euclidean distance model). c Principal component analysis group plot of PC 1 vs PC 3 (Derived stimulus configuration, Euclidean distance model)

4 Conclusions

The average concentration of Fe is 7670 ppm and may be due to it being the fourth most abundant element (after oxygen, silicon and aluminum) in the earth's crust. The concentration of As ranges from 1.46 to 2.91 ppm with an average of 2.23. The concentration of Cd ranges from 1.96 to 2.93 ppm. The concentration of Co ranges from 0.56 to 1.23 ppm and the concentration of Cr ranges widely from 10.38 to 11.95 ppm with an average value of 11.28. Cu was detected in all the sediments, the concentration ranges from 47.82 to 56.97 ppm with an average of 52.86. The average concentration of Mn ranges from 672.88 to 724.44 ppm with an average of 693.49. The concentration of Pb ranges from 31.99 to 36.84 ppm with an average of 34.7, while Zn ranges from 121.18 to 127.32 ppm with an average of 124.58. The concentration of Ni ranges widely from 35.13 to 39.98 ppm with an average value of 37.35. The low levels of nickel in the coastal environment may be due to the chemical weathering of rocks. The average metal accumulation levels in coral samples of the study area is in the following order:

Fe > Mn > Zn > Cu > Ni > Pb > Cr > Cd > As > Co. Results obtained from EF, I_{geo} and CF indicate that sediments are contaminated by Cd in most of the sites. Pollution load index is less than one indicating that the contamination of Cd was suppressed by the presence of other elements. The cumulative results obtained from PC 1 vs PC 3 and PC 2 vs PC 3 explain the metal concentration variation in sediment samples.

It is evident that industrial effluents and riverine inputs have contributed to trace elements accumulation in coral sediments. The anthropogenic activities taking place in the area over the last five decades have a damaging effect on the marine ecosystem due to the large quantities of industrial waste water discharge and the domestic sewage through rivers joining the study area. Hence the study area is getting contaminated by trace elements and if the levels of trace elements continue to increase, the toxic effect on the marine ecosystem will also be increased. Therefore, the trace element accumulation in the coral sediment is a direct indicator of industrial effluents discharge. The pre-treatment of industrial and domestic effluents before discharge into the coastal area of Gulf of Mannar is warranted to protect the marine ecosystem.

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