

Evaluation of Metal Contamination in Coastal Sediments of East Coast of Tamilnadu, India; Geochemical and Statistical Approaches

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Abstract: Sediment samples were collected by a Peterson grab sampler along the Bay of Bengal coastline, from Poombuhar to Karaikal along the South East Coast of Tamilnadu, India to evaluate the degree of contamination and pollution status of the study area. The determined mean metal concentration is in the order of Ti > Fe > Al > Ca > Mg > K > Mn > V > Cr > La > Ba > Pb > Zn > Ni > Co > As. The pollution indices such as enrichment factor (EF), contamination factor (CF) and pollution load index (PLI) were calculated in the sediments to differentiate the origin of metals between anthropogenic and geogenic sources. Using multivariate statistical analysis (correlation coefficients, factor and cluster analysis) the interrelationships among elements are determined. The concentration of the trace elements reported in this work is useful as baselines analysis for sediment quality studies in future.

Keywords: Sediment samples, Pollution indices, Enrichment factor (EF), Contamination factor (CF), Pollution load index (PLI).

1 Introduction

Pollution of the natural environment by metals is becoming a potential global problem. Coastal and estuarine regions are the important sinks for many persistent pollutants and they accumulate in organisms and bottom sediments. Sediment pollution by heavy metals has been regarded as a critical problem in marine environment because of their toxicity and bioaccumulation [1-6]. Sediment quality has been recognized as an important indicator of water pollution [7] since sediments are the main sink for various pollutants, including metals discharged into the environment [8-12]. Sediments also play a significant role in the remobilization of contaminants in aquatic systems under favorable conditions and in interactions between water and sediment.

The geochemical characteristics of the sediments can be used to infer the weathering trends and the sources of pollution [13-15]. Comprehensive methods for identifying and assessing the severity of sediment contamination have

been introduced in order to protect the aquatic life Community [16].

Multi-elemental analysis of sediment may reveal the presence of heavy metals which are contaminants and may have toxic influence on ground water and surface water and also on plants, animals and humans [17]. Heavy metals may accumulate to a toxic level in sediments without visible signs. Sediment analysis is vital to assessing qualities of total ecosystem of a water body in addition to water sample analysis practiced for many years.

In this work, sediments have also been employed for the monitoring and assessment of metal pollution from Poombuhar to Karaikal of East Coast of Tamilnadu, India. This coast is a densely populated area with variety of industrial activities (such as metal smelting, pharmaceuticals etc) and agriculture activities (which include maize, cassava, sugarcane and vegetables farming). All these activities release toxic and potentially toxic metals to the environment. This research therefore aims at assessing the influence and sources of these toxic and potentially toxic metals on these sediments from East Coast of Tamilnadu.

This study aims at assessing the level of heavy metal

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enrichment in the sediments as well as the contamination status. The main objectives of the current study are: (1) to determine concentrations of metals existing in the sediments from Poombuhar to Karaikal of East Coast of Tamilnadu and (2) to assess the degree of contamination by heavy metals in sediments using indices of contamination (3) to define the natural and/or anthropogenic sources of these metals using statistical techniques.

2 Materials and Methods

2.1 Study Area

Table 1: represents the geographical latitude and longitude for the sampling locations.

S.No	Locations	Location ID	Latitude (N)	Longitude (E)
1	Poombuhar	PPR	11° 9' 1.0836" N	79° 50' 37.248" E
2	ChinnaVaangiri	PCV	11° 7' 5.5776" N	79° 50' 49.2828" E
3	Vaangiri	PVG	11° 7' 10.7652" N	79° 50' 47.328" E
4	Chinnangudi	PCG	11° 5' 32.1504" N	79° 50' 32.5212" E
5	Pillaiperumalnallur	PPM	11° 4' 21.8028" N	79° 49' 59.1492" E
6	Vellaikoil	PVK	11° 2' 3.2496" N	79° 51' 15.5484" E
7	Tharangambadi	PTP	11° 1' 37.182" N	79° 50' 13.6752" E
8	Cheranampadi	PCP	10° 59' 59.658" N	79° 51' 0.7164" E
9	Mandapaputhur	PMP	10° 59' 8.2464" N	79° 51' 4.4244" E
10	Akkampettai-I	PAP-1	10° 56' 56.3784" N	79° 50' 54.6144" E
11	Akkampettai-II	PAP-2	10° 56' 56.0724" N	79° 51' 17.172" E
12	Keelakasakudimedu-I	PKM-1	10° 56' 26.5684" N	79° 51' 10.7384" E
13	Keelakasakudimedu-II	PKM-2	10° 56' 10.5681" N	79° 51' 6.7382" E
14	Kilinjalmadu	PKJ	10° 56' 23.6148" N	79° 50' 50.9064" E
15	Karaikalmedu	PKL	10° 56' 24.2196" N	79° 51' 11.9196" E
16	Ammantherumedu-I	PAT-1	10° 55' 54.2192" N	79° 51' 07.9136" E
17	Ammantherumedu-II	PAT-2	10° 55' 43.2188" N	79° 51' 10.9192" E
18	Karaikal-I	PKK-1	10° 55' 31.584" N	79° 50' 16.8216" E
19	Karaikal-II	PKK-2	10° 55' 30.432" N	79° 50' 16.8396" E
20	Karaikal-III	PKK-3	10° 55' 30.1296" N	79° 50' 18.8484" E

Table 1 represents the geographical latitude and longitude for the sampling locations at the study area.

2.2 Sample Collection

The Peterson grab sampler collects sediment layer from the seabed along the 20 stations. Uniform quantity of sediment samples were collected from all the sampling stations. Each sample of about 2 kg was kept in a thick plastic bag. The samples were air dried at 105°C for 24 hrs to a constant weight. Care was taken to ensure that the collected sediments were not in contact with the metallic dredge of the sampler and the top sediment layer was scooped with an acid washed plastic spatula. Sediment samples were stored in plastic bags and kept in Sediment

samples were collected by a Peterson grab sampler along the Bay of Bengal coastline, from Poombuhar to Karaikal along the South East Coast of Tamilnadu, India during premonsoon on condition. The sampling locations were selected based on the prevailing stresses and included areas near the urban and domestic effluent discharge point. The Fig. 1 shows the sampling location of the Study area. The samples were collected in premonsoon season, when sediment texture and ecological conditions can be clearly observed, when erosional activities are predominant and sediments were not transported from the river and estuary towards the beach.

refrigeration at -4°C until analysis. Then pebbles, leaves and other foreign particles were removed.

2.3 Sample Preparation

The samples were air dried at 105°C for 24 hrs to a constant weight and sieved using a 63 µm sieve in order to identify the geochemical concentrations. The grain size <63 µm, which presents several advantages: (1) heavy metals are mainly linked to silt and clay; (2) this grain size is like that of the suspended matter in water; and (3) it has been used in many studies on heavy metal contamination. Then samples were ground into a fine powder for 10–15 min, using an agate mortar. All powder samples were stored in desiccators until they were analyzed. One gram of the finely ground sample and 0.5 g

of the boric acid (H3BO3) were mixed. The mixture was thoroughly ground and pressed to a pellet of 25 mm diameter using a hydraulic press (20 tons).

2.4 EDXRF Technique

The prepared pellets were analyzed using the EDXRF available at Environmental and Safety Division, Indira Gandhi Centre for Atomic Research (IGCAR), Kalpakkam, Tamilnadu. The instrument used for this study consists of an EDXRF spectrometer of model EX-6600SDD supplied by Xenometrix, Israel. The spectrometer is fitted with a side window X-ray tube (370 W) that has Rhodium as anode. The power specifications of the tube are 3-60 kV; 10-58331A. Selection of filters, tube voltage, sample position and current are fully customizable. The detector SDD 25mm² has an energy resolution of 136 eV ± 5 eV at 5.9 keV Mn X-ray and 10-sample turret enables keeping and analyzing 10 samples at a time. The quantitative analysis is carried out by the In-built software nEXT. A standard soil (NIST SRM 2709a) was used as reference material for standardizing the instrument. This soil standard obtained from a follow field in the central California San Joaquin valley. Fig 2 shows the typical spectrum of EDXRF. The soil standard (reference material) (NIST SRM 2709a) analysis value are given in Table 2.

Tamilnadu, southeastern India is presented in Table 3. The range and mean value of heavy metal concentration, 242-13861 mg kg⁻¹(7223 mg kg⁻¹) for Mg; 27937-57567 mg kg⁻¹ (41725 mg kg⁻¹)for Al; 1100-7415 mg kg⁻¹ (5246 mg kg⁻¹) for K; 10392-29069 mg kg⁻¹ (20015 mg kg⁻¹) for Ca; 4343-135568 mg kg⁻¹ (43241 mg kg⁻¹)for Ti; 24413-129254 mg kg⁻¹ (66216mg kg⁻¹)for Fe; 108-1701.3 mg kg⁻¹(610 mg kg⁻¹) for V; 112-947 mg kg⁻¹ (383 mg kg⁻¹) for Cr; 545-3289 mg kg⁻¹ (1641 mg kg⁻¹) for Mn; 9-45 mg kg⁻¹ (23 mg kg⁻¹)for Co; 29-50 mg kg⁻¹ (42 mg kg⁻¹) for Ni; 46-136 mg kg⁻¹ (90 mg kg⁻¹) for Zn; 0-6 mg kg⁻¹ (4 mg kg⁻¹)for As; 25-269 mg kg⁻¹ (159 mg kg⁻¹)for Ba; 12-391 mg kg⁻¹ (146 mg kg⁻¹) for La and 9-158 mg kg⁻¹ (50 mg kg⁻¹) for Pb. The determined mean metal concentration is in the order of Ti> Fe > Al > Ca > Mg > K > Mn > V > Cr > La > Ba > Pb > Zn > Ni > Co > As. Among the heavy metals detected, Aluminum (Al) is the most abundant metal in the sediments. The mean concentration values of heavy metals in sediments do not exceed the natural background levels of heavy metals given by [18]. This indicates that study area is dominated with large amounts of natural sediment with low heavy metal content. Fig 3 shows the different location with heavy metal concentration.

Table 2: Results obtained from the analysis of soil standard-2709a reference sample using EDXRF (in mg kg⁻¹).

Element	Certified Values	EDXRF values
Mg	14600	14900 ± 1000
Al	72100	68400 ± 2300
K	20500	19100 ± 700
Ca	19100	16500 ± 500
Ti	3400	3100 ± 100
Fe	33600	33900 ± 1200
V	110	98.8 ± 6.59
Cr	130	112.1 ± 4.01
Mn	529	568.2 ± 19.85
Co	12.8	12.8 ± 0.55
Ni	83	69.3 ± 2.98
Zn	107	127.9 ± 4.88

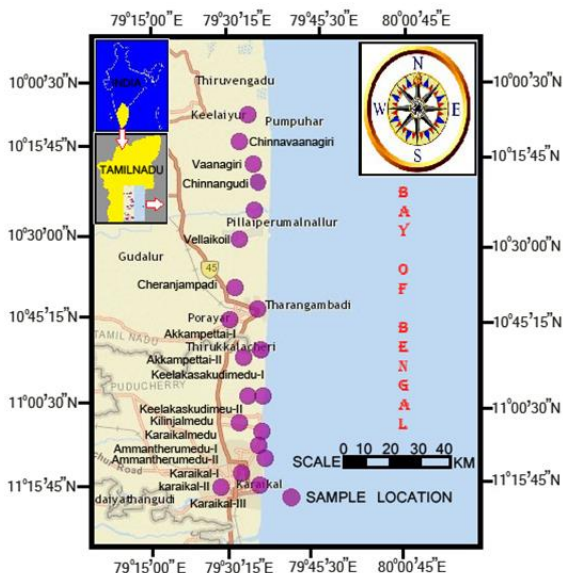


Fig. 1: Location Map of collected sediment samples of East Coast of Tamilnadu, India

3 Results and Discussion

3.1 Metal Contents in Surface Sediments

The heavy metal concentrations of elements in sediments from Poombuhar to Karaikal along the East Coast of

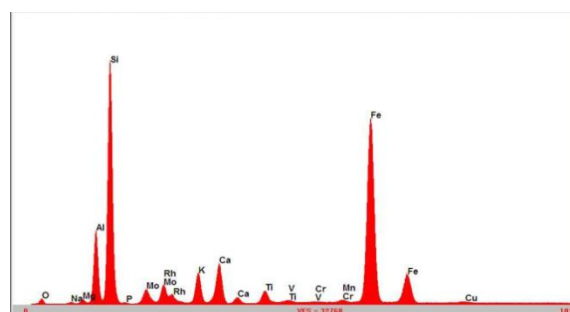


Fig. 2: Typical EDXRF spectrum of sediment sample.

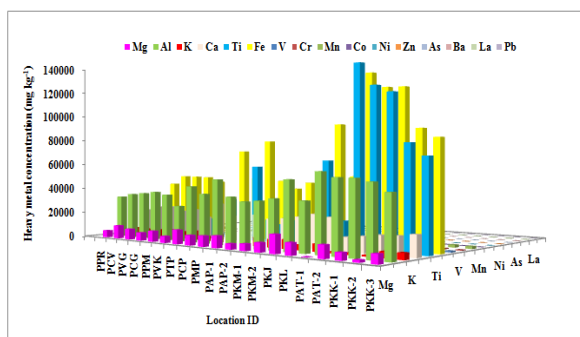


Fig. 3: Plot of locations versus heavy metals concentration in sediment of East Coast of Tamilnadu, India

The location of Kilinjalmedu (PKJ) is characterized by higher concentrations of Ti, Fe, V, Cr, Mn, Co, Zn, La and Pb when compared with other locations. This may be due to the high tourists' boat activities and other anthropogenic activities like shipping and harbor activities, industrial and urban wastage discharges, dredging, etc., such findings are in agreement with the results of earlier workers [19-21].

The accumulations of heavy metals in sediments might be due to point sources such as direct discharge of large amounts of industrial and domestic sewages into rivers and/or seas [22-24]. There are many chemical and pharmaceutical factories located along the east coast of Tamilnadu whose discharge can heavily pollute the soils with heavy metals. Additionally, these enriched metals may also have originated from non-point sources such as agricultural pollution (e.g. fertilizers and livestock manure), atmospheric transport and other industrial activities [24]. Overall, our data indicates that the elevated heavy metal levels in the sediments resulted partially from the anthropogenic activities, such as wastewaters, aquaculture activities and shipping.

3.2 Enrichment Factor (EF)

A common approach to estimating the anthropogenic impact on sediments is to calculate a normalized enrichment factor (EF) for metal concentrations above uncontaminated background levels [25-27]. Measuring enrichment factor (EF) is an essential part of geochemical studies and is generally used to differentiate between the metals originating from anthropogenic (non-crustal) and geogenic (crustal) sources, and to assess the degree of metal contamination. Enrichment factor (EF) is a useful tool for determining the degree of anthropogenic heavy metal pollution.

The EF is computed using the relationship below

$$EF = \frac{\left(\frac{C_x}{C_{Al}}\right)_{\text{sample}}}{\left(\frac{C_x}{C_{Al}}\right)_{\text{reference}}} \quad \text{--- (1)}$$

Where C_x and C_{Al} denote the concentrations of metals X

and Al and EF is their ratio in the samples of interest to their average background shale. [18].

The Enrichment factor in sediments from Poombuhar to Karaikal is presented in Table 4. In this study, aluminum (Al) was used as the reference element for geochemical normalization for the following reasons: (1) Al is associated with fine solid surfaces; (2) its geochemistry is similar to that of many trace metals and (3) its natural concentration tends to be uniform. However, an $EF > 1.5$ indicates that a significant portion of the trace metals was delivered from non-crustal materials so, these trace metals were delivered by other sources, like point and non-point pollution and biota. With EF index, soil quality state can be indicated by different classes (Table 4) ranging from $EF < 2$ (Deficiency to minimal enrichment) to $EF > 40$ (Extremely high enrichment).

The EF levels of heavy metals vary as follow: 0.022–1.561 (mean of 0.959) for Mg, 0.062–0.798 (mean of 0.414) for K, 0.653–2.427 (mean of 1.838) for Ca, 2.704–40.95 (mean of 15.890) for Ti, 1.481–3.806 (mean of 2.577) for Fe, 2.386–18.186 (mean of 8.101) for V, 3.563–14.623 (mean of 7.689) for Cr, 1.837–5.506 (mean of 3.524) for Mn, 1.282–3.283 (mean of 2.216) for Co, 0.625–1.656 (mean of 1.235) for Ni, 1.397–2.184 (mean of 1.799) for Zn, 0.447–1.136 (mean of 0.722) for As, 0.065–1.264 (mean of 0.585) for Ba, 0.379–6.605 (mean of 2.711) for La and 1.315–10.946 (mean of 4.300) for Pb.

The minimum EFs obtained for some elements (Mg, K, Ni, As and Ba) are less than unity implying that these elements are depleted in some phases relative to crustal abundance in the study area [28]. The EF values for Mg, K, Ni, As and Ba were less than 1.5, which indicates dominant metal enrichments from natural sources in the study area. EF values greater than 1.5 that were obtained for Ca (PPR, PCV, PVG, PCG, PPM, PVK, PTP, PCP, PAP-1, PAP-2, PKM-1, PKM-2, PKJ and PKL), Ti (PPR, PCV, PVG, PCG, PPM, PVK, PTP, PCP, PMP, PAP-1, PAP-2, PKM-1, PKM-2, PKJ, PKL, PAT-1, PAT-2, PKK-1, PKK-2 and PKK-3), Fe (PPR, PCV, PVG, PCG, PPM, PTP, PCP, PMP, PAP-1, PAP-2, PKM-1, PKM-2, PKJ, PKL, PAT-1, PAT-2, PKK-1, PKK-2 and PKK-3), Zn (PPR, PCV, PVG, PCG, PPM, PTP, PCP, PMP, PAP-1, PAP-2, PKM-1, PKM-2, PKJ, PKL, PAT-1, PAT-2, PKK-1, PKK-2 and PKK-3), La (PCV, PCG, PTP, PCP, PMP, PKM-1, PKJ, PAT-1, PAT-2, PKK-1, PKK-2 and PKK-3), Pb (PPR, PCV, PVG, PCG, PPM, PTP, PCP, PMP, PAP-1, PAP-2, PKM-1, PKM-2, PKJ, PKL, PAT-1, PAT-2, PKK-1, PKK-2 and PKK-3) suggest that these levels of enrichment might have originated from sources that are of non-crustal origin including anthropogenic sources.

The results suggest that sediments in these areas are contaminated with heavy metals, whose major source is anthropogenic input from industrial activities [29].

Table 3: Heavy metal concentration (mg kg⁻¹) in sediments along the East Coast of Tamilnadu, India.

S. No.	Location ID	Locations	Mg	Al	K	Ca	Ti	Fe	V	Cr	Mn	Co	Ni	Zn	As	Ba	La	Pb
1	PPR	Poombuhar	5549	33090	6099	20812	14221	42432	244.4	204.5	1027.1	14.6	45.2	69	5	221.2	34	19.2
2	PCV	ChinnaVaaganai	10435	35643	5691	23900	15861	49400	275.3	271.5	1131.9	17.3	50.2	75.8	5.8	169.4	70.8	23.7
3	PVG	Vaaganai	8416	36848	5999	24547	13899	49284	242.6	254.9	1142.3	17.3	49.7	75.7	5.4	191	53.7	25.1
4	PCG	Chinnangudi	6360	38287	6742	21512	20263	48993	313.3	283.8	1186.1	17	43.8	74.3	6.1	165.2	86.4	23.6
5	PPM	Pillaipeenamalalur	8493	36497	6096	23278	14762	45541	246.2	207.8	1103.2	15.9	46.3	69.6	5.1	268.9	52.7	16.8
6	PVK	Vellaikoil	5300.5	27937	7415	17755	4343	24413	108.3	112	545.1	8.5	38	46.3	5.2	256	12.2	9.2
7	PTP	Tharangambadi	11014	43954	4148	29069	31910	71316	490	416	1977.1	24.6	49.6	100.2	5.1	114.9	83	45.3
8	PCP	Cheranjampadi	7820	38465	6557	20092	23447	49788	364.9	267.1	1213.1	17.3	41.3	72.3	5.3	193.1	99.3	21
9	PMP	Mandapaputhur	8279	49957	5822	18629	59071	79515	813.3	415.7	1900.8	26.8	36.7	92.2	5.2	135.2	269.7	52.4
10	PAP-1	Akkampettai-I	9197	37240	6909	22208	17243	47694	301.5	230.9	1103.1	16.5	44.7	96.6	5.3	220	41.7	19.2
11	PAP-2	Akkampettai-II	4335.5	34182	6827	19783	14699	41810	258.4	220	1033	14.3	40.8	70.6	5.3	218	47.2	16.7
12	PKM-1	Keelakasakudimedu-I	5247.5	35271	6226	20857	18065	46824	291.6	267.6	1086.7	16.3	45.6	71.6	6.3	220.1	80.2	21.6
13	PKM-2	Keelakasakudimedu-II	7089.5	37442	6997	22751	18833	48986	323.5	242.8	1164.6	16.7	44.7	77.7	6.1	165.9	60.4	27.4
14	PKJ	Kilinjalmedu-I	13860.5	51440	4277	25400	64431	91716	876.7	489.9	2459.9	32.1	45.9	127.7	3.8	108.6	204	75.7
15	PKL	Karaikalmedu-II	9051.5	36902	5798	23885	20200	52531	337.5	331.9	1325	18.4	47.6	84	5.4	181.2	52	27.8
16	PAT-1	Ammantherumedu-I	242	57567	1370	10392	135568	129254	1701.3	947	3289.4	44.9	31	135.9	-	54	352.4	157.5
17	PAT-2	Ammantherumedu-II	9598.5	53769	1500	11913	118710	118006	1571.5	751.3	3145.6	40.3	28.6	133.5	-	34.8	272.1	123.6
18	PKK-1	Karaikal-I	5554	53585	1100	13646	113399	117696	1498.1	708	3133.2	40.5	32.4	128.8	-	25.3	331	138.9
19	PKK-2	Karaikal-II	1804.5	51412	4545	14100	77522	87817	1025.2	592	1984.9	30.4	30.8	101.2	3.7	88.5	390.5	87.2
20	PKK-3	Karaikal-III	6819	45022	4800	15783	68384	81300	920.1	452.9	1873.8	28.4	37.4	99.7	5.1	150.2	322.6	74.2
Average			7223	41725	5246	20015	43241	66216	610	383	1641	23	42	90	4	159	146	50
Minimum			242	27937	1100	10392	4343	24413	108	112	545	9	29	46	3.7	25	12	9
Maximum			13861	57567	7415	29069	135568	129254	1701	947	3289	45	50	136	6	269	391	158
Crustal Average (Turekian And Wedepohl, 1961)			15000	80000	26600	22100	4600	47200	130	90	850	19	68	95	13	580	92	20

Table 4: The EF values of heavy metals in sediments from East coast of Tamilnadu, India.

S. No.	Location ID	Locations	Mg	K	Ca	Ti	Fe	V	Cr	Mn	Co	Ni	Zn	As	Ba	La	Pb
1	PPR	Poombuhar	0.894	0.554	2.277	7.474	2.173	4.546	5.495	2.921	1.862	1.606	1.757	0.924	0.922	0.893	2.316
2	PCV	ChinnaVaaganai	1.561	0.480	2.427	7.739	2.349	4.754	6.771	2.989	2.049	1.656	1.790	0.999	0.656	1.728	2.658
3	PVG	Vaaganai	1.218	0.490	2.411	6.560	2.267	4.052	6.148	2.918	1.972	1.587	1.730	0.909	0.715	1.268	2.721
4	PCG	Chinnangudi	0.886	0.530	2.034	9.204	2.169	5.036	6.590	2.916	1.870	1.346	1.633	0.974	0.595	1.963	2.464
5	PPM	Pillaipeenamalalur	1.241	0.502	2.309	7.034	2.115	4.151	5.060	2.845	1.830	1.492	1.606	0.853	1.016	1.256	1.840
6	PVK	Vellaikoil	1.012	0.798	2.301	2.704	1.481	2.386	3.563	1.837	1.282	1.600	1.397	1.136	1.264	0.379	1.315
7	PTP	Tharangambadi	1.336	0.284	2.394	12.626	2.750	6.861	8.413	4.234	2.355	1.327	1.920	0.708	0.361	1.643	4.124
8	PCP	Cheranjampadi	1.084	0.513	1.891	10.601	2.194	5.837	6.173	2.968	1.893	1.264	1.583	0.850	0.692	2.244	2.187
9	PMP	Mandapaputhur	0.884	0.350	1.350	20.564	2.698	10.019	7.397	3.581	2.259	0.865	1.554	0.637	0.373	4.695	4.197
10	PAP-1	Akkampettai-I	1.317	0.558	2.159	8.052	2.171	4.981	5.512	2.788	1.870	1.411	2.184	0.881	0.815	0.973	2.063
11	PAP-2	Akkampettai-II	0.676	0.601	2.095	7.478	2.073	4.653	5.722	2.844	1.762	1.403	1.739	0.955	0.880	1.201	1.950
12	PKM-1	Keelakasakudimedu-I	0.793	0.531	2.141	8.907	2.250	5.088	6.743	2.900	1.942	1.521	1.708	1.105	0.861	1.976	2.448
13	PKM-2	Keelakasakudimedu-II	1.010	0.562	2.200	8.747	2.217	5.317	5.765	2.927	1.874	1.405	1.747	1.010	0.611	1.403	2.931
14	PKJ	Kilinjalmedu	1.437	0.250	1.787	21.783	3.022	10.489	8.466	4.501	2.624	1.051	2.090	0.453	0.291	3.448	5.885
15	PKL	Karaikalmedu	1.308	0.473	2.343	9.520	2.413	5.629	7.995	3.379	2.095	1.519	1.918	0.900	0.677	1.226	3.011
16	PAT-1	Ammantherumedu-I	0.022	0.072	0.653	40.956	3.806	18.186	14.623	5.378	3.283	0.633	1.988	-	0.129	5.323	10.946
17	PAT-2	Ammantherumedu-II	0.952	0.084	0.802	38.396	3.720	17.986	12.420	5.506	3.156	0.625	2.091	-	0.089	4.400	9.199
18	PKK-1	Karaikal-I	0.553	0.062	0.922	36.805	3.723	17.205	11.745	5.503	3.184	0.712	2.024	-	0.065	5.372	10.372
19	PKK-2	Karaikal-II	0.187	0.266	0.993	26.224	2.895	12.271	10.235	3.634	2.492	0.704	1.657	0.447	0.237	6.605	6.783
20	PKK-3	Karaikal-III	0.808	0.321	1.269	26.416	3.061	12.577	8.941	3.917	2.658	0.977	1.864	0.702	0.460	6.231	6.591
Minimum			0.022	0.062	0.653	2.703	1.481	2.386	3.563	1.837	1.282	0.625	1.397	0.447	0.065	0.379	1.315
Maximum			1.561	0.798	2.427	40.95	3.806	18.186	14.623	5.506	3.283	1.656	2.184	1.136	1.264	6.605	10.946
Average			0.959	0.414	1.838	15.890	2.577	8.101	7.689	3.524	2.216	1.235	1.799	0.722	0.585	2.711	4.300

However, sediments from some stations outside these locations were either slightly or not contaminated with these heavy metals. Finally, the levels of heavy metal enrichment obtained in sediments in the East Coast of Tamilnadu are “minimal to moderate”. The order of total EF are of the order Ti> V> Cr >Pb>Mn>La > Fe > Co > Ca > Zn >Ni > Mg > As >Ba > K . Variation in heavy metal enrichment factor along the east coast of Tamilnadu is shown in Fig 4.

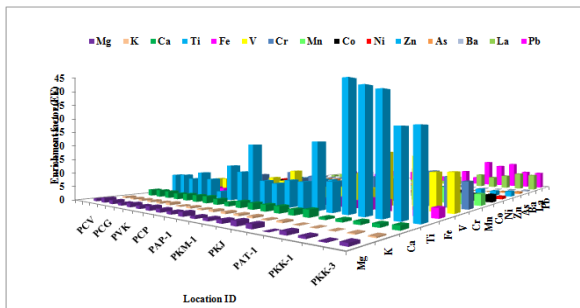


Fig. 4: Plot of locations versus EF values of heavy metals in sediment of East Coast of Tamilnadu, India.

3.3 Contaminant Factor (CF)

The assessment of sediment contamination was also carried out using the contamination factor. The contamination factor is used to determine the contamination status of the sediment in the present study. Contaminant factor (Cf) is the ratio obtained by dividing the concentration of each metal in the sediment by the background value [30]. CF is considered to be an effective tool in monitoring the pollution over a period of time and is given by the formula,

$$CF = \frac{C_{\text{heavymetal}}}{C_{\text{background}}} \quad \text{--- (2)}$$

“C_{background}” refers to the concentration of metal of interest in the sediments when there was no anthropogenic input. According to Hakanson, 1980 [31]:Cf<1 indicates low contamination; 1<Cf<3 is moderate contamination; 3<Cf<6 is considerable contamination; and Cf> 6 is very high contamination. The contaminant factor values in sediments are presented in Table 5.

The range and mean value of Contamination Factors are 0.016-0.924 (average 0.482) for Mg; 0.349-0.72 (average 0.522) for Al;0.041-0.279 (average 0.197) for K; 0.47-1.315 (average 0.906) for Ca;0.944-29.471 (average 9.4)for Ti; 0.517-2.738 (average 1.403) forFe;0.833-13.087 (average 4.694) for V;1.244-10.522 (average 4.26) for Cr; 0.641-3.87 (average 1.931) for Mn; 0.448-2.362 (average 1.205) for Co; 0.420-0.738 (average 0.61) for Ni; 0.488-1.431 (average 0.949) for Zn; 0.287-0.487 (average 0.343) for As; 0.044-0.464 (average 0.274) for Ba; 0.132-4.245 (average 1.585) for La and 0.459-7.876 (average 2.515) for Pbrespectively.Values of CF for all samples are less

than 1 except Titanium, Vanadium Cadmium, Chromium and Lead. The results indicating that sediments were low contaminated with these elements. Fig 5shows the variation in CF values of heavy metals with locations.

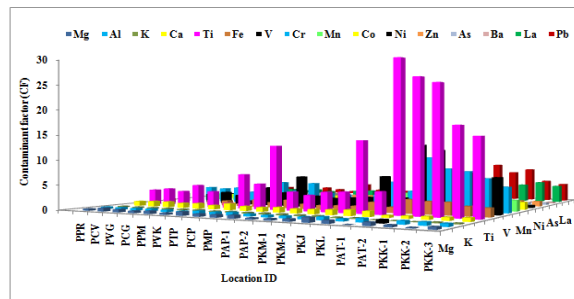


Fig. 5: Plot of locations versus CF values of heavy metals in sediment of East Coast of Tamilnadu, India.

3.4 Geo-Accumulation Index (I_{geo})

A common approach to estimating the enrichment of metal concentrations above background or baseline concentrations is to calculate the geoaccumulation index (I_{geo}) as proposed by Mullar, (1979) [32].It is used to determine metals contamination in sediments and can be calculated using the following formula:

$$I_{\text{geo}} = \text{Log}_2 \left(\frac{C_n}{1.5 \times B_n} \right) \quad \text{--- (3)}$$

Here, C_nis the measured concentration in the sediment for metal n, B_nis the background value for the metal n [18], The factor 1.5 is introduced to minimise the effect of possible variations in the background values which may be attributed to lithologic variations in the sediments [33]. According to the scale established by [32], 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 I_{geo} values for each element at each sampling site were calculated using background values. The calculated I_{geo}values based on the world shale average [18]. Table 6 presents the geo-accumulation index for the quantification of heavy metal accumulation in the study area.

The I_{geo} values of coastal sediments are -1.968 to -0.210(average -0.579) for Mg, -0.633 to -0.319 (average -0.467) for Al, -1.560 to -0.731(average -0.931) for K, -0.504 to -0.057(average -0.233) for Ca, -0.201 to 1.293(average 0.626) for Ti, -0.462 to 0.261 (average -0.067) for Fe, -0.255 to 0.941(average 0.371) for V, -0.081to -0.846(average 0.394) for Cr, -0.369 to 0.412 (average 0.064) for Mn, -0.525 to 0.197 (average -0.133) for Co, -0.553 to -0.308(average -0.396) for Ni, -0.488 to -0.021 (average -0.215) for Zn, -0.718 to -0.488 (average -0.488) for As,, -1.537 to -0.510 (average -

0.803) for Ba, -1.055 to 0.452 (average -0.149) for La, -0.514 to 0.720 (average -0.081) for Pb respectively. The averaged pollution degree of these metals decreased in the following order of Ti > Cr > V > Pb > Mn > Fe > Co > La > Zn > Ca > Ni > Al > As > Mg > Ba > K. The variation of index (I_{geo}) values with the locations is shown in Fig 6.

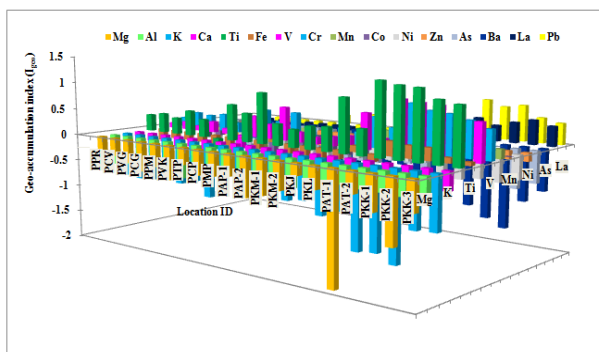


Fig. 6: Plot of locations versus I_{geo} values of heavy metals in sediment of East Coast of Tamilnadu, India.

According to the Muller scale [32], the I_{geo} values of the present study indicate no pollution of the investigated metals in the sampling location of the study area. The results of I_{geo} in all the sampling sites were more restrictive, characterizing the sediments as unpolluted with I_{geo} values <1. Thus the I_{geo} values of heavy metals indicate that sediment is not much polluted from anthropogenic inputs.

3.5 Pollution Load Index (PLI)

Pollution severity and its variation along the sites were determined with the use of pollution load index. This index is a quick tool in order to compare the pollution status of different places. The pollution load index (PLI) provides a simple, comparative means for assessing the level of heavy metal pollution [34]. PLI is determined as the nth root of the product of nCf

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{1/n} \quad \text{--- (4)}$$

Where Cf is the contamination factor and n is the number of metals. According to Tomlison, 1980 [34], $PLI > 1$ means that pollution is present; otherwise, if it is below 1, there is no metal pollution.

The PLI results range from 0.492 to 1.402 with a mean of 0.809 (Table 5), thus indicating that the area is practically not polluted. Criteria of all the pollution indicators in sediment based on pollution indices are given in the Table 7. The variation of pollution load index with locations is given in Fig 7.

4 Multivariate Statistical Analysis

Multivariate statistical analysis such as correlation analysis, principal component analysis (PCA) and

Hierarchical cluster analysis (HCA) are Powerful tool used to give the relationship between the variables. Statistical Package social science (SPSS) software of version 16.0 issued to obtain the relationship between the variables in this study.

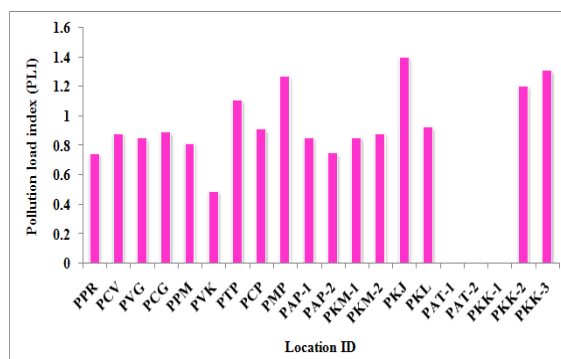


Fig. 7: Shows the variation of PLI with locations ($r=0.938$), Fe ($r=0.972$), V ($r=0.941$), Cr ($r=0.939$)Ti($r=0.938$), Fe ($r=0.972$), V($r=0.941$), Cr ($r=0.939$).

4.1 Pearson Correlation Coefficient Analysis

The Pearson Correlation produces correlation coefficient, r , which measures the strength and direction of linear relationships between pairs of continuous variables. By extension, the Pearson Correlation evaluates whether there is statistical evidence for a linear relationship among the same pairs of variables. Positive correlation indicates that both variables increase or decrease together, whereas negative correlation indicates that as one variable increase, so the other decreases and vice versa. It has a value between +1 and -1, where 1 is total positive linear correlation, 0 is no linear correlation, and -1 is total negative linear correlation. The Pearson correlation (Table 8) shows a positive correlation between the heavy metals, except that between Mn($r=0.955$), Co($r=0.971$), Zn($r=0.932$), La($r=0.906$) and Pb(0.922) were above 0.5 while the others were below 0.5, K($r=-0.863$), Ca($r=-0.546$), Ni(-0.659), As($r=-0.787$) and Ba(-0.918). At the 0.05 or 95% is the confidence level (2-tailed, $P < 0.01$). The geochemical behaviors of Al and Ti, Fe, V, Cr, Mn, Co, Zn, La and Pb are known to be similar in most natural processes [35]. This could explain the high correlation and suggest minimal or no anthropogenic inputs [36]. The levels of these heavy metals pose no environmental concern. Though the sediments receive effluents from industries, surface runoffs and domestic wastes, the levels of input of these heavy metals are low [37]. Thus the heavy metal concentrations are likely to be a result of natural background levels rather than pollution [38].

Table 5: Contamination factor (Cf), Contamination Degree (Cd) and Modified Degree of Contamination (mCd) of sediments along the East Coast of Tamilnadu, India.

Table with 20 columns: S. No., Location ID, Locations, Mg, Al, K, Ca, Ti, Fe, V, Cr, Mn, Co, Ni, Zn, As, Ba, La, Pb, PLI. Rows include locations like PPR (Roombuhar), PCV (Chinnamagan), PVG (Vannagiri), PCG (Chinnangudi), PPM (Pillaiarumalnallur), PVK (Vellaikoil), PTP (Tharangambadi), PCP (Cheranampadi), PMP (Mandapattur), PAP-1 (Akkampattai-I), PAP-2 (Akkampattai-II), PKM-1 (Keelakasa kudimedu-I), PKM-2 (Keelakasa kudimedu-II), PKJ (Kilinjalmadu), PKL (Karakalmedu), PAT-1 (Ammantharumadu-I), PAT-2 (Ammantharumadu-II), PKK-1 (Karakal-I), PKK-2 (Karakal-II), PKK-3 (Karakal-III), AVERAGE, MINIMUM, MAXIMUM, Cd, mCd, and Cp.

Table 6 :The Igeo values of heavy metals in sediments from East coast of Tamilnadu, India.

Table with 20 columns: S. No., Location ID, Locations, Mg, Al, K, Ca, Ti, Fe, V, Cr, Mn, Co, Ni, Zn, As, Ba, La, Pb. Rows include the same 20 locations as Table 5, followed by Total, Average, Minimum, and Maximum rows.

4.2 Principal Component Analysis (PCA)

Principal component analysis (PCA) is a statistical procedure that uses an orthogonal transformation to convert a set of observations of possibly correlated variables into a set of values of linearly uncorrelated variables called principal components. PCA can be used to reduce data and to extract a smaller number of independent factors (principal components) to find the relationship among observed variables [39]. Principal component analysis (PCA) was applied in the Study to have high quality experimental results.

Principal component analysis (PCA) applied on the data accounts for 92.46% of the total variance and set the data through two factors with eigen values greater than unity (Table 9). Based on rotated component matrix using principle component analysis with Varimax rotation, Al(0.955), Ti (0.998), Fe (0.993), V (0.998), Cr (0.983), Mn (0.978), Co (0.992), Zn (0.923) La (0.904) and Pb (0.990) (with high loadings) are placed together within PC1 explaining 71.98 % of total variance. This factor is considered as “natural factor”. This indicates that heavy metals are due to earth crustal materials. PC2 which explains 20.48% of the total variance is mainly composed Co, Zn, La and Pb. Cluster II includes Mg, K, Ca, Ni, As

And Ba. The strong similarity between cluster – I metals of Mg(-0.228),K(-0.106),Ca(-0.712),Ni(0.546), As(0.141) and Ba(-0.202)(with poor loadings). This factor is a “anthropogenic factor”, since sediments receive effluents from industries, surface runoffs and domestic wastes, the levels of input of these heavy metals. [40].The results of principal component analysis are in agreement with Pearson correlation analysis.

4.3 Hierarchical Cluster Analysis (HCA)

Cluster analysis is a data exploration (mining) tool for Dividing a multivariate dataset into “natural” clusters (groups).Hierarchical cluster analysis was also used in this study to identify the relatively homogeneous groups of heavy metals. The cluster analysis of the heavy metals was performed according to the average linkage method with Euklidian distance as similarity measure [41]. Fig 8 shows a derived dendrogram summarizing all heavy metals.

Cluster analysis for heavy metals could be grouped into two clusters. Cluster I includes Al, Ti, Fe, V, Cr, Mn, Co, Zn, La and Pb. Cluster II includes Mg, K, Ca, Ni, As

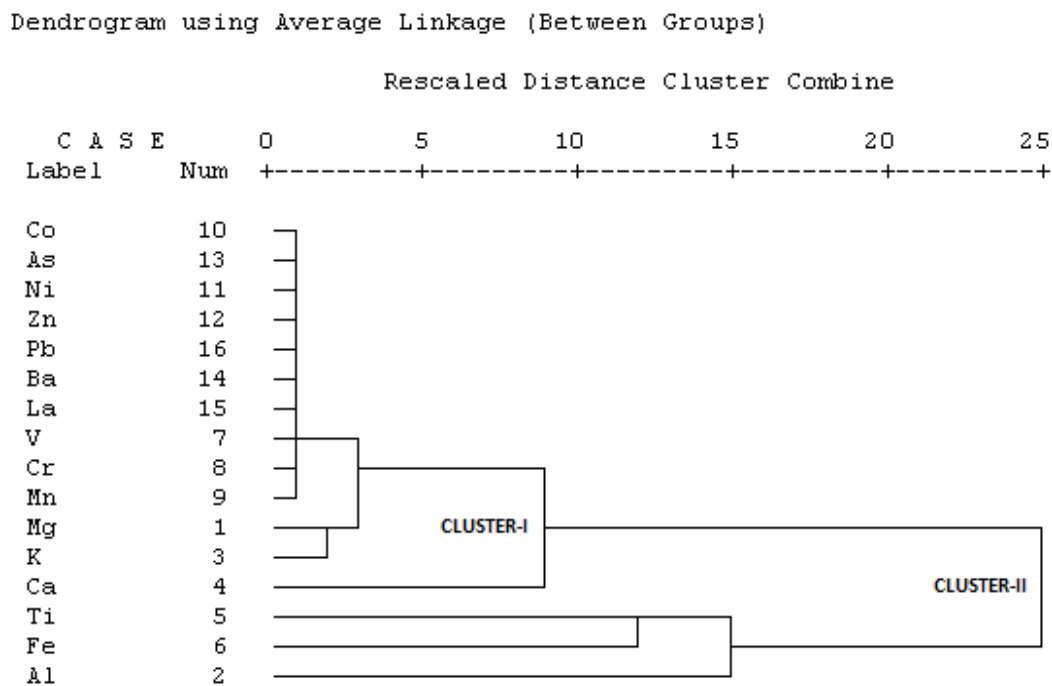


Fig. 8: Dendrogram shows the clustering of heavy metals.

Table 8: The correlation of heavy metals in sediment from East Coast of Tamilnadu, India.

Variables	Mg	Al	K	Ca	Ti	Fe	V	Cr	Mn	Co	Ni	Zn	As	Ba	La	Pb
Mg	1															
Al	-0.093	1														
K	0.122	-0.863	1													
Ca	0.674	-0.546	0.573	1												
Ti	-0.271	0.938	-0.93	-0.755	1											
Fe	-0.142	0.972	-0.948	-0.623	0.982	1										
V	-0.249	0.941	-0.933	-0.745	0.999	0.985	1									
Cr	-0.268	0.939	-0.941	-0.672	0.978	0.981	0.976	1								
Mn	-0.079	0.955	-0.962	-0.57	0.965	0.992	0.969	0.967	1							
Co	-0.141	0.971	-0.949	-0.618	0.981	1	0.983	0.982	0.991	1						
Ni	0.559	-0.659	0.609	0.948	-0.802	-0.692	-0.797	-0.715	-0.651	-0.684	1					
Zn	0.05	0.932	-0.894	-0.45	0.903	0.952	0.91	0.914	0.958	0.953	-0.537	1				
As	0.255	-0.787	0.929	0.722	-0.916	-0.884	-0.916	-0.892	-0.898	-0.883	0.75	-0.821	1			
Ba	0.078	-0.918	0.892	0.518	-0.9	-0.935	-0.908	-0.919	-0.935	-0.933	0.619	-0.894	0.8	1		
La	-0.333	0.906	-0.76	-0.732	0.906	0.885	0.904	0.869	0.829	0.885	-0.794	0.72	-0.703	-0.819	1	
Pb	-0.281	0.922	-0.952	-0.718	0.99	0.98	0.989	0.982	0.968	0.98	-0.755	0.907	-0.928	-0.91	0.88	1

Table 9: Varimax rotated components of heavy metals in sediment samples of east coast of Tamilnadu, India

S.No	Elements	Component - I	Component - II
1	Mg	-0.228	0.778
2	Al	0.955	0.213
3	K	-0.938	-0.106
4	Ca	-0.712	0.69
5	Ti	0.998	-0.065
6	Fe	0.993	0.12
7	V	0.998	-0.048
8	Cr	0.983	0.023
9	Mn	0.978	0.172
10	Co	0.992	0.123
11	Ni	-0.767	0.546
12	Zn	0.923	0.297
13	As	-0.907	0.141
14	Ba	-0.916	-0.202
15	La	0.904	-0.087
16	Pb	0.99	-0.039
% of Variance Explained		71.98%	20.48%

Note: Bold values indicates the significant correlation between the heavy metal

Showed relationship of these metals comes from the same origin or earth crustal materials. The result obtained from the hierarchical cluster analysis is very well in agreement with Pearson correlation and principal component analysis.

5 Conclusions

Concentration of heavy metals were measured in the coastal sediment using EDXRF technique and twenty sampling sites were predefined in different locations of East coast of Tamilnadu. The heavy metals and major elements concentration of the sediments were found to decrease in sequence of Ti > Fe > Al > Ca > Mg > K > Mn > V > Cr > La > Ba > Pb > Zn > Ni > Ca > As. Results of pollution indices of heavy metals indicated that Industrial activities and vehicle emissions represented the most important sources of Pb and Zn contamination. The results of this study indicated that a general absence of serious pollution in East Coast of Tamilnadu. The treatment of 16 variables sampled at twenty sites by the factor and cluster analyses provided a possible interpretation of the collective data. The result obtained from the hierarchical cluster analysis is very well in agreement with Pearson correlation and principal component analysis. Finally, the statistical tools provided interpretation of the obtained data in a more systematic way. The results in this paper will establish an initial

View of sediment pollution and the status of the contamination in the study area.

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