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Heavy metal characterisation in surface sediments off southeast coast of India: Implication on marine pollution

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The present study investigated heavy metals (Mn, Cr, Cu, Ni, Pb, and Zn) distribution in the surface sediments off Karaikal – Velankanni offshore along the east coast of Bay of Bengal as an indicator of marine pollution during 2004-2005. The sediments were analyzed for sand, silt, clay ratio, $CaCO_3$, organic matter, and heavy metal (Mn, Cr, Cu, Ni, Pb, and Zn) contents. The contamination factor (CF) values for the heavy metals are found to be in the order: Mn >Cr >Ni >Zn >Cu >Pb during August 2004; and during January 2005 the CF values were found to be in the order: Zn >Mn >Cu >Ni >Pb >Cr. Zn contamination was found to be low to moderate. 10 samples were found with low and moderate Mn contamination. All the samples displayed moderate contamination with Cu, Ni, and Pb. Among the selected sediments, those which are collected from Karaikal and Velankanni shore were found to be highly contaminated by heavy elements due to the discharge of effluents from petrochemical, oil spills, port activity, and shipping activities. The occurrence of a wide range of elements in coastal sediments is mainly related to the textural variability of sediments during 2004 and 2005.

[**Keywords**: Bay of Bengal, Heavy metals, India, Surface sediments, Texture]

Introduction

Significant settlements, industries, and harbour were established in the coastal areas, and as a result, the coastal areas across the world have witnessed tremendous growth in the human population. Owing to this, the effluents from the domestic and industrial as well as from agricultural activities are being released into the marine environment. The ocean was able to dilute these contaminants during the past, but due to the increased growth in population and industries and thereby increase in per capita waste generation has increased the input of contaminants manifold, and its adverse impacts are felt now along the coastal regions. The contamination of the Minamata Bay, by the release of methyl mercury in the marine environment and the subsequent outbreak of peculiar fatal disease, made the community realize the fact that the seas cannot absorb the pollutants released indiscriminately[1,2]. High level of pollutants is now recorded and published from many areas, and after implementation of monitoring programs and identifying the point sources of pollution, different actions have been taken to protect the environment[3-9]. Sediments constitute a long-term contamination source to the food web[10]. The sediments show less dissimilarity in time and space. They are exceptional monitor tools to allow reliable evaluation of sequential and spatial contamination[11-13].

Hence today the attention of the whole world has turned towards the study of environmental degradation and pollution and their related fields in oceanic science. The adverse impact of pollution on the marine food products on which coastal communities depend upon and on the tourism industry, a primary source of income are threatened as the seas are polluted and hence detailed studies are held to identify the point source of pollution and to understand the related processes occurring in the coastal zone. The contaminants transported from the terrestrial ecosystem are concentrated in the marginal marine and nearshore environments, and as a result, the water and sediments in the nearshore environments are more polluted relative to the open seas. Further, the currents, tides, and waves play a major role in transporting these contaminants to the open ocean.

The coastal marine environment is very important as it forms the sink for toxic pollutants. Estuaries are one of such coastal ecosystem that supports wide array of faunal and floral diversity offering many benefits to human livelihood like renewable food supplies (fishes and marine algae) and tourism.

Several land-based activities on the coast severely damage such type of sensitive environment. The present study is carried out to identify the level of contamination by heavy metals presence in the coastal sediments based on the geochemical characteristics of the sediments.

Materials and Methods

Study area

The 30 km long coastal tract along the Karaikal - Velankanni sector is drained by the distributaries of Cauvery river *viz*., Arasalar, Thirumalairajanar, Vettar, Uppanar, and Vellar river. The major towns located in the coastal zone are Karaikal, T.R. Pattinam, Nagore, Nagapattinam, and Velankanni (Fig. 1). There are several industries including textile, chemical, and a power plant in Karaikal district. Also, there are some oil wells owned by ONGC, which is a major oil field in the Cauvery basin. The coast also has two seaports in Karaikal and Nagapattinam in addition to several fishing harbors that cater to the needs of over 10000 fishermen. Though the Nagapattinam port (minor port) is in existence for over 1000 years from the period of the Cholas dynasty has not been modernized to date. Additionally, the Karaikal port has been developed as a minor port. However, it is operational and the development of this port started in 2005 *i.e.* after the sampling period. The port is expected to accelerate the industrial growth, and might increase the anthropogenic inputs into the marine environment. The Cauvery delta is the primary source of agriculture in Tamil Nadu where agricultural activities are engaged on a massive scale thereby carrying the agricultural effluents to the offshore regions. In this background, the study of the geochemical characteristics of the sediments from the inner shelf of Karaikal - Velankanni coast was carried out and the results are presented herewith. The samples were collected alongshore normal traverses in Karaikal- Velankanni sectors in two seasons. The first sampling was carried out before the onset of NE monsoon and the second sampling was carried out after NE monsoon. Twenty samples were collected during pre-monsoon from four shore normal traverses and were designated as K1 to K20. Pre-monsoon sampling was carried out with the help of Van Veen Grab in the cruises organized by the National Institute of Ocean Technology (NIOT) in their Coastal Research Vessel (CRV) and Sagar Paschimi on August, 2004 while second sampling was done onboard Sagar Purvi from 13th to 18th January 2005. The grab once on board was lowered on a polythene sheet and opened gently. The top layer of the sample was scooped with a PVC spatula. The collected samples were immediately transferred to pre-cleaned PVC containers (Fig S1). Sediments are important hauler of heavy metals in the hydrological cycle, and they effectively gather metals from the adjacent waters[14]. The marine and estuarine regions are the ultimate sinks for the pollutants accumulating these in the bottom sediments[15]. Therefore, coastal marine sediments act as the most concentrated physical pool

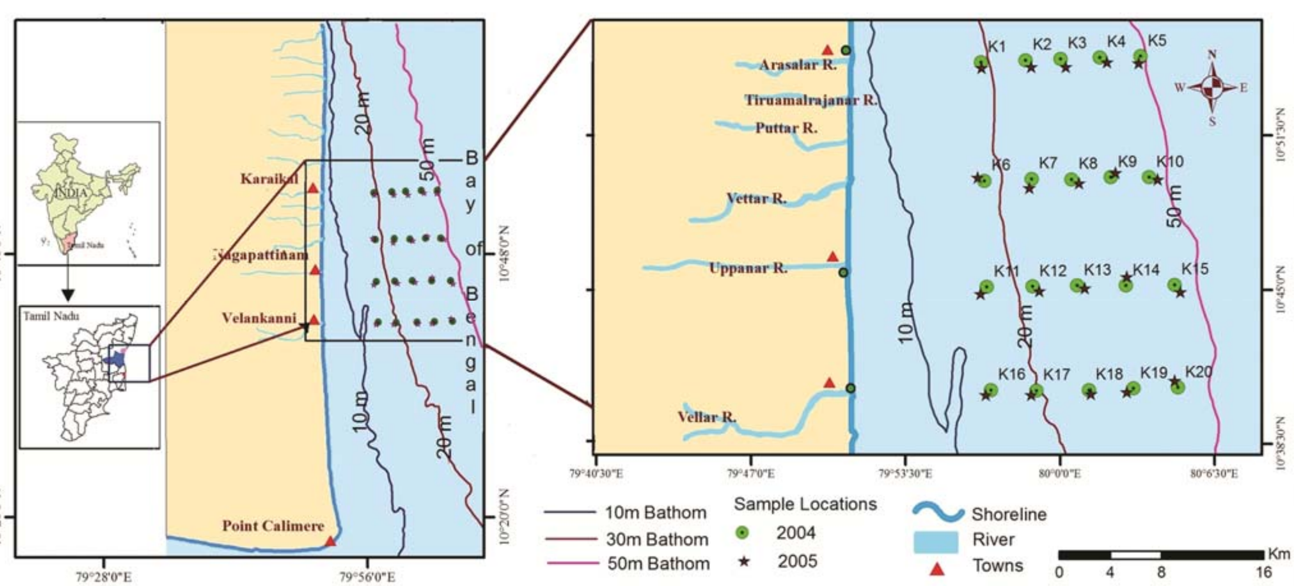

Fig. 1 — Map of the study area showing sample points

of inorganic chemical constituents and particular metals in the aquatic environment.

Results and Discussion

Grain size

The Average sand content in the samples collected in August 2004 in Karaikal-Velankanni offshore areas is 72.5 %. The sediments collected during January 2005 immediately after the Indian Ocean Tsunami (IOT) is characterized by a sand content of 69.4 % (Figs. 2a & b). A comparison of the values 72.5 % and 69.4 % shows that the sand content has decreased significantly in the tsunami sediments. Average Silt content is 23.5 % in sediments collected in August, 2004 while in sediments collected post-tsunami (2005) clay is low (4 %) in sediments collected in August 2004 and it is high (8.21 %) in post-tsunami sediments (Table 1 & 2).

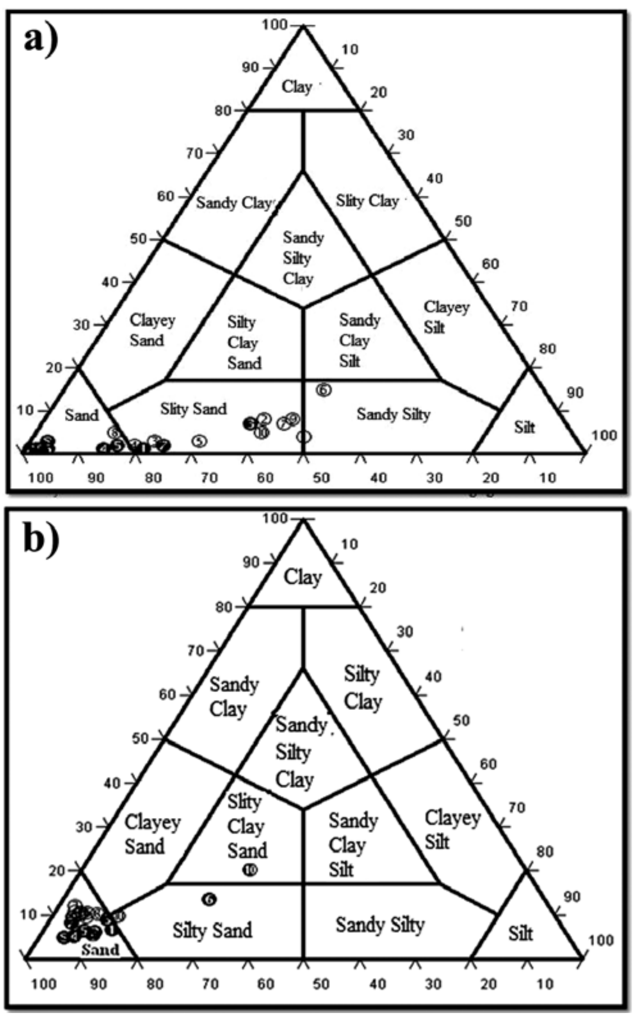

Fig. 2 — Ternary plots of Sand-Silt-Clay components of surface sediment in Karaikal: a) Pre-monsoon; and b) Post-monsoon. Field limits are from Shepard[22]

Organic matter

The data shows that there is an increase in clay content in the tsunami sediments. The increase in clay content is rather uniform and the distribution pattern is strikingly similar. As the tsunami advances towards the land, sediments is found to have a black color due to the suspended load of clay derived by churning the seafloor[16]. From the dating of sediments from off Tagus estuary, off Lisbon, Portugal, it is evident that the advancing tsunami can strip up about 29 cm layer of sediment at about 100 m water depth and the invading waters will be loaded with sediments[17]. The invading tsunami erodes the beach deposits in the foreshore and backshore to redeposit the coarse fraction as on-land tsunami deposits. During backwash, the receding waters return to the sea transporting the fine sediments from marine as well as the terrestrial source. Hence the water in the inner shelf is rich in clay content which immediately settles down due to quick flocculation taking place in saline waters. Experimental studies by Sutherland *et al.*[18] revealed that clay particles flocculate and settle within 10 minutes in saline water filled in a tank 10 cm deep

Table 1 — Sand, silt, clay, $CaCO_3$, Organic Matter, Mud (Silt + Clay) and Sediments type of the sediments off Karaikal-Velankanni, Bay of Bengal (Pre-monsoon)

Sl. No	Sand %	Silt %	Clay %	Mud %	OM %	$CaCO_3$ %	Sediment type
K1	48.00	48.00	4.00	52.00	3.73	2.38	Silty Sand
K2	53.00	39.00	8.00	47.00	3.65	2.56	Silty Sand
K3	75.00	22.00	3.00	25.00	2.87	3.34	Silty Sand
K4	79.00	19.00	2.00	21.00	1.23	3.40	Silty Sand
K5	67.00	30.00	3.00	33.00	1.23	2.95	Silty Sand
K6	39.00	46.00	15.00	61.00	4.32	3.16	Sandy Silty
K7	50.00	43.00	7.00	50.00	3.93	2.76	Silty Sand
K8	81.00	14.00	5.00	19.00	3.12	3.37	Sand
K9	48.00	44.00	8.00	52.00	4.76	2.56	Silty Sand
K10	55.00	40.00	5.00	45.00	3.87	2.68	Silty Sand
K11	78.00	21.00	1.00	22.00	2.45	3.86	Silty Sand
K12	98.00	1.00	1.00	2.00	0.87	4.34	Sand
K13	56.00	37.00	7.00	44.00	3.98	2.88	Silty Sand
K14	85.00	14.00	1.00	15.00	2.32	3.41	Sand
K15	82.00	16.00	2.00	18.00	1.67	3.96	Sand
K16	95.00	4.00	1.00	5.00	1.02	3.90	Sand
K17	74.00	24.00	2.00	26.00	2.56	3.36	Silty Sand
K18	94.00	3.00	3.00	6.00	1.13	3.49	Sand
K19	96.00	3.00	1.00	4.00	0.52	3.94	Sand
K20	97.00	2.00	1.00	3.00	1.24	4.15	Sand
Min	39.00	1.00	1.00	2.00	0.52	2.38	
Max	98.00	48.00	15.00	61.00	4.76	4.34	
Avg	72.50	23.50	4.00	27.50	2.52	3.32	

Table 2 — Estimated values of sand, silt, clay, $CaCO_3$, Organic Matter, Mud and Sediments type of the sediments off Karaikal – Velankanni, Bay of Bengal (Post-Monsoon)

Sl. No	Sand %	Silt %	Clay %	Mud %	OM %	$CaCO_3$ %	Sediment type
K1	46.2	42.0	11.8	53.8	3.56	0.50	Silty Sand
K2	49.0	35.5	15.5	51.0	3.40	0.54	Silty Sand
K3	73.4	19.1	7.5	26.6	2.50	0.91	Silty Sand
K4	72.5	16.5	11.0	27.5	1.52	1.09	Silty Sand
K5	65.0	27.5	7.5	35.0	1.75	0.78	Silty Sand
K6	38.0	44.0	18.0	62.0	4.40	0.70	Sandy Clay Silt
K7	49.0	41.5	9.5	51.0	3.80	0.80	Silty Sand
K8	78.9	15.0	6.1	21.1	2.80	1.20	Silty Sand
K9	45.0	42.0	13.0	55.0	3.20	0.70	Silty Sand
K10	53.2	38.0	8.8	46.8	2.80	0.70	Silty Sand
K11	76.5	20.0	3.5	23.5	2.20	0.98	Silty Sand
K12	89.0	3.0	8.0	11.0	1.20	1.41	Sand
K13	53.0	35.0	12.0	47.0	2.80	0.60	Silty Sand
K14	83.0	12.5	4.5	17.0	2.30	1.30	Sand
K15	80.0	15.0	5.0	20.0	1.50	1.03	Sand
K16	90.5	4.5	5.0	9.5	1.20	1.42	Sand
K17	71.0	23.0	6.0	29.0	2.00	0.87	Silty Sand
K18	91.0	5.0	4.0	9.0	1.13	1.36	Sand
K19	91.0	5.0	4.0	9.0	0.80	1.21	Sand
K20	92.0	4.5	3.5	8.0	1.40	1.21	Sand
Min	38.00	3.00	3.50	8.00	0.80	0.50	
Max	92.00	44.00	18.00	62.00	4.40	1.42	
Avg	69.36	22.43	8.21	30.64	2.31	0.96	

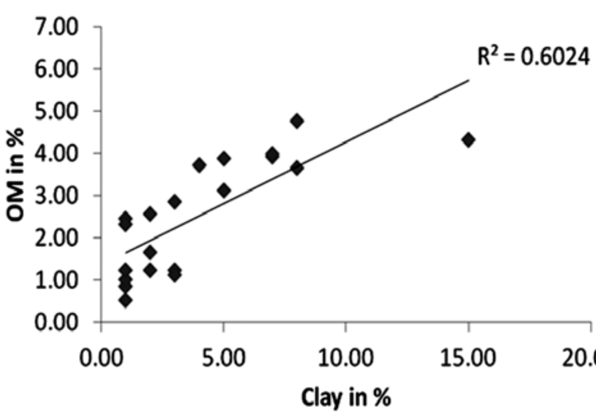

Fig. 3 — Clay and OM in the sediments off Karaikal-Velankanni-Pre-monsoon

while it takes 10 hours in the case of freshwater organic matter shows a good correlation with clay and is similar in Karaikal –Velankanni offshore sediments collected during August 2004, with an average of 2.52 % sediment organic matter (Fig. 3). Organic matter content is more or less equal (average OM is 2.52 % in 2004 and 2.31 % in 2005 (Fig. 4a & b). The OM content is related to clay content which has increased significantly in the tsunami sediments. The high OM content is due to the transportation of OM from the seabed rather than an estuarine source.

The $CaCO_3$ content of the sediments collected during August 2004 shows an average of 3.32 %. The

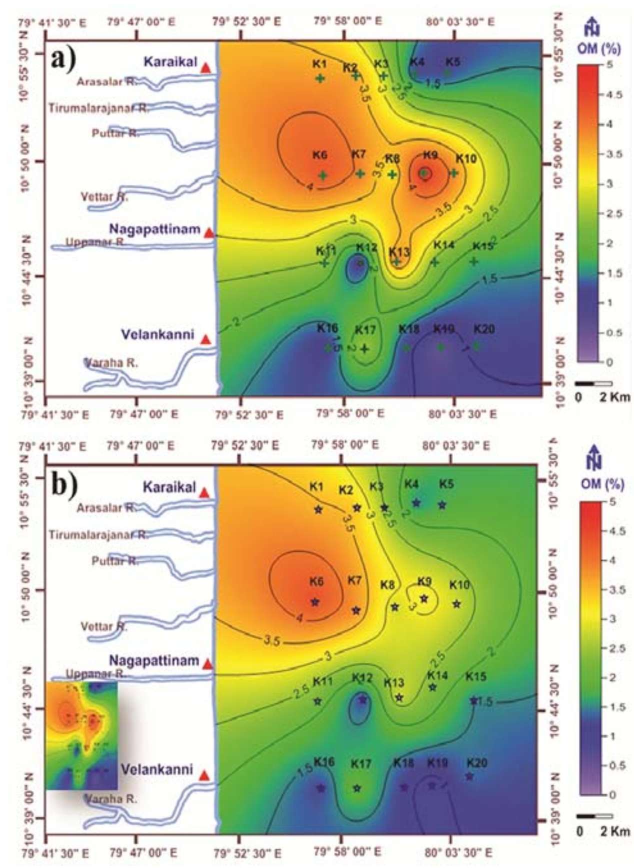

Fig. 4 — Distribution of organic matter percentages of Karaikal-Velankanni: a) Pre-monsoon; and b) Post-monsoon

post-tsunami deposits contain an average $CaCO_3$ content of 0.96 % which is significantly lesser when compared to the pre-tsunami sediments. The concentration of $CaCO_3$ follows the trend of sand content (Fig. S2).

Heavy metal distribution

The heavy elements analysed during the present study are heavy metals that are commonly associated with Mn hydroxide coatings on sand and silt grains, clay fraction, and OM[19]. Mn is the dominant heavy metal found in post-tsunami sediments of 2005 showing an increase of 11.2 % (Figs. 5a & b). In Karaikal – Velankanni sediments, Mn exhibits an excellent positive correlation with OM and good relation with Cr, Cu, mud, and Zn as well as with all other parameters except sand and $CaCO_3$ in 2004 and post-tsunami sediments of 2005 (Fig. S3). The average concentration of Cr in the sediments collected off the Velankanni-Velankanni sector during 2004 and 2005 were 36.9 and 174 µg/g, respectively which showed more than 4 fold rise in the concentration during 2005. The increase is calculated as 37.2 % in Karaikal – Velankanni. The relationship with Mud, OM as well as with all parameters except sand and $CaCO_3$ are excellent to moderate in Karaikal –

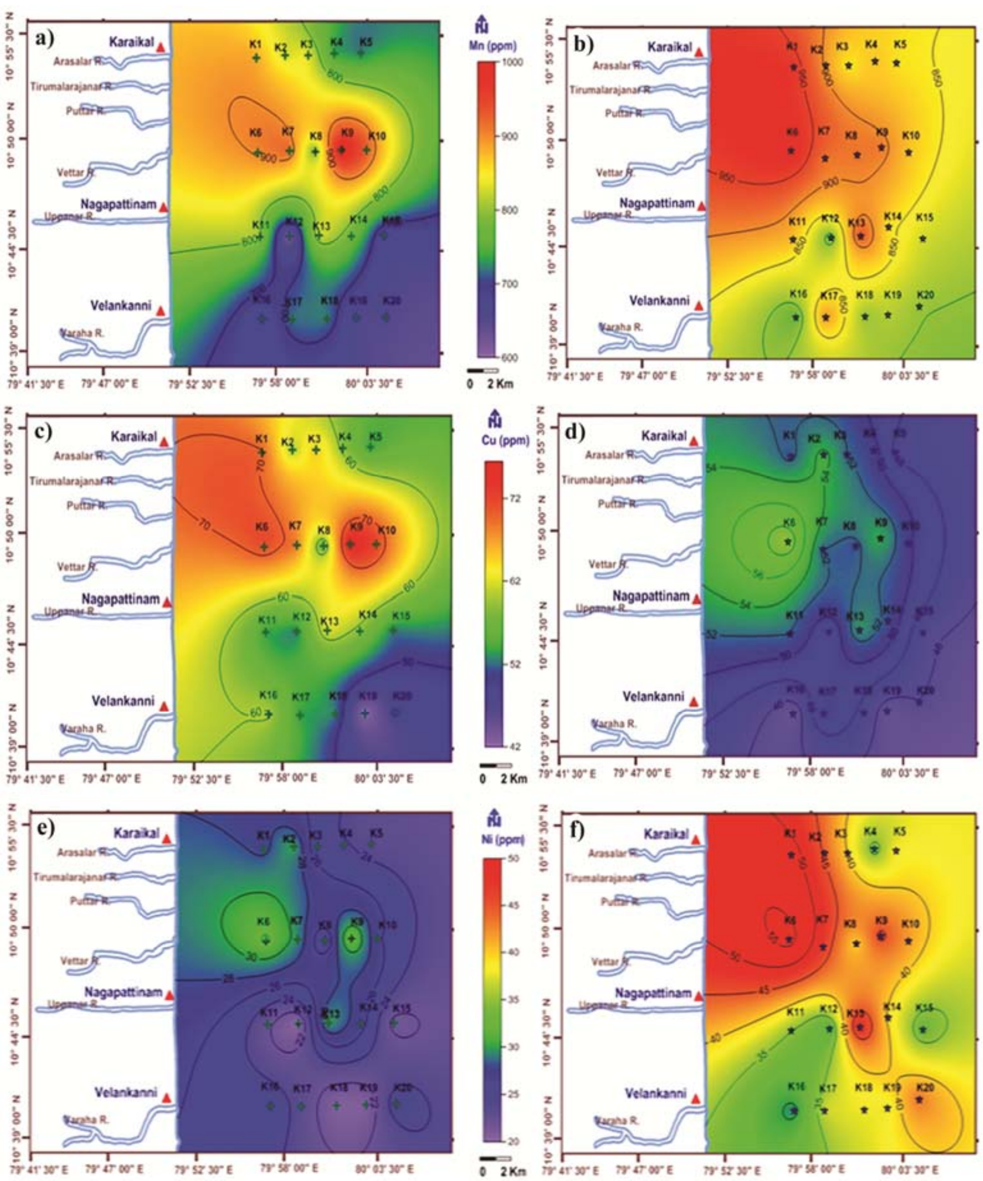

Fig. 5 — Spatial distribution Map of metals in sediments off Karaikal –Velankanni coast: a) Mn-Pre-monsoon; b) Mn- Post-monsoon; c) Cu- Pre-monsoon; d) Cu- Post-monsoon; e) Ni- Pre-monsoon; and f) Ni- Post-monsoon

Velankanni offshore sediments collected during August 2004 and January 2005 (Fig. S4).

Cu is an essential heavy metal and it can form organic Cu complexes with organic matter[20]. In Karaikal – Velankanni offshore sediments Cu exhibits a mean of 59.6 µg/g and 50.4 µg/g, respectively in August 2004 (Table 3) and January 2005 (Table 4) with a decrease in Cu concentration by 15.5 % in 2005 is observed (Figs. 5c & d). Cu shows a good correlation with OM, Mud, Mn, and Ni (Figs. 5e & f).

Table 3 — Analytical results of heavy metal content of surface sediments off Karaikal –Velankanni (Pre-Monsoon)

Sl. No	Mn	Cr	Cu	Ni	Pb	Zn
K1	886	40.60	70.30	27.50	42.30	127.00
K2	829	39.10	60.30	30.00	49.10	117.80
K3	840	40.90	65.30	24.20	39.30	122.20
K4	734	35.70	59.30	24.20	38.60	106.20
K5	720	33.00	54.00	23.20	37.40	112.00
K6	904	48.00	71.70	32.30	49.80	127.30
K7	924	44.70	69.00	29.40	44.50	127.00
K8	792	37.50	57.00	24.00	40.60	112.70
K9	1008	50.00	76.60	32.50	52.60	138.20
K10	932	36.90	72.50	26.40	41.20	120.70
K11	808	38.60	53.70	22.30	40.20	106.90
K12	608	28.00	52.50	19.90	31.40	106.40
K13	829	38.10	62.30	29.90	50.70	126.00
K14	778	39.70	59.00	26.70	39.50	117.10
K15	668	37.00	52.30	21.40	34.60	104.00
K16	660	27.80	60.30	25.40	32.50	93.80
K17	716	33.50	53.60	23.30	37.40	118.70
K18	720	33.30	52.60	20.40	39.10	109.10
K19	653	30.20	41.90	20.50	39.00	106.70
K20	655	26.00	47.90	25.10	39.50	108.00
Min	608	26.00	41.90	19.90	31.40	93.80
Max	1008	50.00	76.60	32.50	52.60	138.20
Mean	783	36.93	59.61	25.43	40.97	115.39

Results are expressed in µg/g

Table 4 — Analytical results of heavy metal content of surface sediments off Karaikal –Velankanni (Post-Monsoon)

Sl. No	Mn	Cr	Cu	Ni	Pb	Zn
K1	967	169.00	51.50	52.00	44.38	57.60
K2	894	207.30	54.60	44.50	43.50	48.00
K3	851	165.00	52.00	39.99	43.72	48.94
K4	871	157.00	50.70	33.99	42.90	44.41
K5	856	169.20	47.40	38.28	43.04	45.80
K6	996	216.00	59.50	56.90	44.09	58.97
K7	915	195.10	52.00	46.90	43.77	59.32
K8	912	173.00	51.50	39.80	43.28	44.90
K9	911	208.10	54.20	45.80	43.73	50.60
K10	871	184.50	50.30	41.90	43.33	46.33
K11	876	181.00	52.00	34.98	43.67	48.12
K12	784	147.00	48.10	31.20	42.41	45.20
K13	935	172.00	53.40	48.99	44.22	52.80
K14	856	169.20	51.50	39.30	42.85	38.00
K15	828	161.50	46.50	32.99	42.58	50.30
K16	775	166.20	44.50	29.28	42.84	34.40
K17	894	163.80	50.00	36.80	42.89	44.00
K18	803	170.70	47.40	37.90	42.19	38.50
K19	816	160.00	44.90	37.80	42.25	31.50
K20	803	148.00	45.98	43.43	43.05	48.00
Min	775	147.00	44.50	29.28	42.19	31.50
Max	996	216.00	59.50	56.90	44.38	59.32
Mean	871	174.18	50.40	40.64	43.23	46.79

Results are expressed in µg/g

The sediments collected off Karaikal – Velankanni in August 2004 show a good correlation with Mn, followed by OM, Mud, and Cr. The average Ni content in the Karaikal – Velankanni sediments for August 2004 is 25.43 µg/g and for January 2005 is 40.64 µg/g (Table 3 & 4). The increase is 45 % and the correlation matrix shows that OM, Cu, and Mn exhibits good correlation and the relationship is moderate with respect to Zn. In Karaikal – Velankanni, Pb, OM, Mud, Mn, Cu, and Zn showed a good correlation in the sediments collected during August 2004. The elements with which moderate correlation is observed are Cr and Ti in 2005 sediments. The relationship is good for OM, Mn, mud, and Pb whereas moderate correlation is noticed in Ti, Cu, Zn, and Cr. Pb is a very toxic element for aquatic organisms and fish[21] and is always determined in the environmental investigations.

The mean concentration of Pb in the sediments collected off the Karaikal – Velankanni sector is 41.0 and 43.2 µg/g during 2004 and 2005, respectively. The mean values of Pb show that the Karaikal – Velankanni sector being an industrial centre is receiving input of anthropogenic lead.

In the case of Zn, in Karaikal – Velankanni samples the mean values decreased from 115 µg/g to 47 µg/g reporting a decrease of 59.5 % during 2005. In 2005, Ni, Pb, mud, Mn, and OM showed good correlation values and moderate values were noticed for Cr. In Karaikal – Velankanni sector, the correlation matrix showed well to moderate correlation with all parameters except sand and $CaCO_3$. The CF values for the trace metals off Karaikal – Velakanni offshore sediments collected during Pre-monsoon (Table S1) is found to be in the order: Mn >Cr >Ni >Zn >Cu >Pb. All the metals in majority of the sediment samples are characterised by moderate CF values (Table S2). 10 samples show low Mn contamination and 7 display Cr contamination. Only for Pb 5 samples and 1 sample for Cu show "considerable" contamination and none of the sediments show very high contamination (Table S3). The characterization of the sediments and examination of the spatial and temporal variation has shown that the sediments have been modified significantly in different seasons but the spatial distribution pattern remains similar.

Conclusion

The textural parameters determined in the study are evaluated to characterize the offshore tsunami deposits. Observations on grain size parameters in the sediments collected off Karaikal – Velankanni area revealed an increase in clay concentration from 4.00 % in August 2004 to 8.21 % in January 2005 registering an average increase of 100 percent. Mud concentration also increases from 27.50 to 30.64 % but silt content has reduced marginally. The increase in clay is attributed to the propagation characteristics of the tsunami. The tsunami grows in height as it approaches the coast due to bottom friction and the bottom friction churns the seafloor and the sediments are scoured by the wave while part of coarse fraction is returned to the seafloor, the fine fraction including silt, clay, and OM remains in suspension and is transported landward. The tsunami devastates the coast by flattening the dunes as they are overtopped and carry the sediments further inland. The tsunami loses its velocity during its landward advancement and deposits the sediments carried from the seafloor and the coastal front. If the tsunami enters a tidal flat or salt marsh or a marginal marine water body its energy gets dissipated and the low-lying area gets flooded. In a moderately sloping coast protected by dunes, the seawater is carried back during backwash into the sea. During backwash, the waters carry the fine sediments leaving sand which forms the sand sheet deposits which are characteristically found in coastal areas affected by the tsunami. Eye witness accounts and video footage taken during tsunami events have shown that the tsunami invading the land as well as returning to the sea is black in color representing rich clay content in suspension. The clay remains in suspension as the tsunami recede it settle down in the seafloor causing an increase in clay content and reduction in the sand. In areas where the seawater is not completely returned to the sea, the increase in clay is expected to occur. This could be the reason for the increase in clay content in Karaikal – Velankanni areas where the onshore topography is flat and brackish water bodies are present close to the coast.

The heavy metals also register a general increase in their concentration in the post-tsunami sediments as their concentration is dependent on increase in clay content. The contamination level has increased due to input from terrestrial sources. There are some studies on the geochemistry of onshore deposits but the geochemical characterization of offshore deposits are lacking. Hence, the pattern on geochemical characters observed in the sediments of the present study cannot be compared with other areas. The inner shelf regions off Porto Novo – Thirumullaivasal and Velankanni are characterized by a high concentration of metals as silt, clay, and organic matter are in higher concentration. The silt rich zone is located to the east of Porto Novo and the high clay zone was found to the east of Thirumullaivasal. Metals like Mn show high concentration in the silt rich zone and on the other hand Pb and Co was found to be high in the clay-rich zones. Metals like Cr, Cu, and Zn form a large patch with high concentrations embracing the silt rich zones and part of clay-rich zones. Examination of temporal variation of the sediments in the Karaikal – Velankanni region with pre and post-tsunami sediments showed a smoothened pattern in the post-tsunami sediments as a result of the agitation of the seafloor consequent to the advancing of the tsunami and backwash. In many locations on the coastal zone, enrichment of Heavy minerals is seen along the beaches.

Supplementary Data

Supplementary data associated with this article is available in the electronic form at http://nopr.niscair.res.in/jinfo/ijms/IJMS_49(12)1810-1818_SupplData.pdf

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Declaration of competing interest

This is to certify that all authors have seen and approved the manuscript being submitted. We warrant that the article is the authors original work. Further, the authors state that the article has not received prior publication and is not under consideration for publication elsewhere.

Author Contributions

KS: Writing-original draft; DP: Investigation, data curation, writing, review & editing; and GN: Conceptualization, resources, writing-review & editing.

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