

Geochemical distribution of forms of phosphorus in marine sediment of Bay of Bengal, southeast coast of India

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Received 27 October 2016; revised 14 December 2016

Sediment samples were collected from three transects in the continental shelf region (up to 30km offshore) between Chennai to Marakkanam. Sedimentary parameters such as sand, silt, clay and organic carbon percentage were analyzed in order to find out their relation with various phosphorus fractions. Sediment was found to be predominantly sandy in nature with low silt and clay content. Among all the fractions (loosely bound- Lo-P, calcium bound- Ca-P, iron bound- Fe-P, aluminum bound- Al-P and organically bound- OP), Ca-P fraction constituted the largest portion (71.38%) followed by OP (16.16%). Bioavailable fractions of phosphorus ranged from 9.3 to 36.2% (avg. 20.9%) of the total phosphorus (TP) during the present study. Total phosphorus content in the offshore sediments ranged between 469.35 and 1219.34 mg/kg with an average of 824.66 mg/kg. The abundance of the major forms of P in the surface sediments of Kalpakkam coast followed the order: Ca-P > OP > Al-P > Lo-P > Fe-P. Cluster analysis clearly showed that all the three transects were different from each other as they formed three different clusters.

[Keywords: Phosphorus speciation; offshore sediment; East coast of India; Bay of Bengal]

Introduction

Sediment plays an important role in the overall nutrient dynamics of the marine environment. Among other nutrients, phosphorus has been recognized as the most essential and critical nutrient in the terrestrial and aquatic environments¹. Phosphorus along with the other nutrients such as nitrate and silicate coupled with physical parameters control the oceanic productivity. It is known to be the ultimate limiting nutrient element over geological time scale²⁻³. Biogeochemical cycle of phosphorus in the marine sediment is mainly controlled by a combination of physical, chemical and biological properties and processes⁴⁻⁵. Phosphorus can occur in the sediment associated with calcium or iron or aluminum hydroxides or can be absorbed on the surface of minerals⁶ or present in the form of organic compounds.

Phosphorus gets into the marine environment through sources particularly agricultural fertilizers, pesticides, etc and point and non-point sources such as treated and untreated sewage effluents originating from households and industries⁷. Consequently, monitoring of phosphorus content in the water and sediment is essential to control and avoid

eutrophication of the coastal aquatic environment. Current abundance of phosphorus in the earth crust is approximately 0.12%, however, almost all the phosphorus on the earth crust is found in the form of minerals including apatite (chloro and fluoro), vivianite, wavellite and phosphorite. Apatite {Ca (PO₄)₃[F, OH, Cl]}, the largest reservoir of phosphate on Earth, is relatively insoluble in water. Most of the phosphorus in the current natural environment is present in particular form which is not biologically available, thus limiting primary production. The sorption of phosphate on sediments has major influences on transport, degradation and ultimate fate of phosphorus in marine ecosystems⁴⁻⁶. Phosphorus fractionation in the sediments can provide valuable information on the origin of phosphorus, the degree of pollution from anthropogenic activities, the bioavailability and also the burial and diagenesis of phosphorus in the sediments⁸⁻⁹. The importance of the effect of sediments on the marine ecosystem depends on the chemical form of phosphorus in which this element is combined⁹⁻¹⁰. For this reason, the study of phosphorus speciation is particularly interesting.

Scrutiny of literature shows plethora of report is available regarding ecological and environmental

studies from Bay of Bengal large marine ecosystem¹¹⁻¹⁴. However, there is a conspicuous lack of information about concentration and variability of different phosphorus fractions from offshore sediment samples in general and from the Indian coast in particular. The present study is to investigate the distribution of phosphorus bioavailability (total phosphorus, organic and inorganic phosphorus) in the offshore marine sediment of Tamil Nadu coast and to sequentially extract different forms of inorganic phosphorus (IP) such as; loosely bound phosphorus (Lo-P), aluminium bound phosphorus (Al-P), calcium bound phosphorus (Ca-P) and iron bound phosphorus (Fe-P).

Materials and Methods

The continental shelf and slope region along the southeast coast of India is a unique environment due to the substantive riverine inputs. Monsoon wind reversal and subsequent changes in the coastal current pattern are observed twice in a year during inter-monsoon periods¹⁵⁻¹⁶. Bathymetry data shows that east coast of Tamil Nadu is shallow in nature and the topography of the abyssal plane is fairly monotonous¹⁷. Along these near-shore waters and estuaries, the biogeochemical parameters exhibit considerable seasonal variations depending on the local conditions, such as rainfall, tidal incursions, various abiotic and biotic processes, and quantum of freshwater inflow that affects the nutrient cycle of the coastal environment¹⁸. The SW to NE monsoon transition occurs during September/October and the NE to SW transition occurs during February/March. Subsequent to the monsoon transition between NE and SW monsoon periods, current pattern changes causing alterations to the coastal water quality^{15, 19}. The sediment and the freshwater influx during the SW and NE monsoon period influence the coastal ecosystem and cause a substantial change in the biogeochemistry along the continental shelf along this coastal stretch²⁰⁻²¹. It has been reported that the littoral drift or the long-shore sediment transport is generally high during the SW monsoon as compared to that of the NE monsoon period²². According to the climatology of this area, the whole year has been divided into three seasons viz: i. Post-monsoon/Summer (February- May), ii. pre-monsoon or SW monsoon (June- September), and iii. Monsoon or NE monsoon (October-January).

Surface sediment samples were collected in the continental shelf area along the coast of Tamil Nadu from three transects perpendicular to Chennai city,

DAE Township Kalpakkam and Marakkanam (Figure 1) during July 2012 onboard CRV Sagar Purvi. Three samples were collected from each transect at a distance of 5, 15 and 30 km offshore using a Van Veen grab sampler. Samples were preserved at 4°C in sealed plastic bags prior to processing for further analysis. For the determination of sand, silt and clay, accurately about 20 gm of sediment samples were washed with 30% H₂O₂ and 1N HCl to remove organic matter and carbonate, respectively²³. Then the samples were washed in a 230 ASTM sieve (mesh size 63µm) repeatedly till all the silt and clay were removed. The washings were collected in a 1000 ml measuring cylinder, which were further categorized as silt and clay by pipette analysis method²⁴⁻²⁵. The material retained in the sieve was dried and weighed for sand fraction.

For analysis of phosphorous, samples were freeze dried and powdered by a mixer mill. Trace metal grade or analytical grade chemicals (Fisher Scientific) were used for preparation of reagents. Milli-Q (Millipore) water was used for all the analyses. Working standard solutions were prepared from a

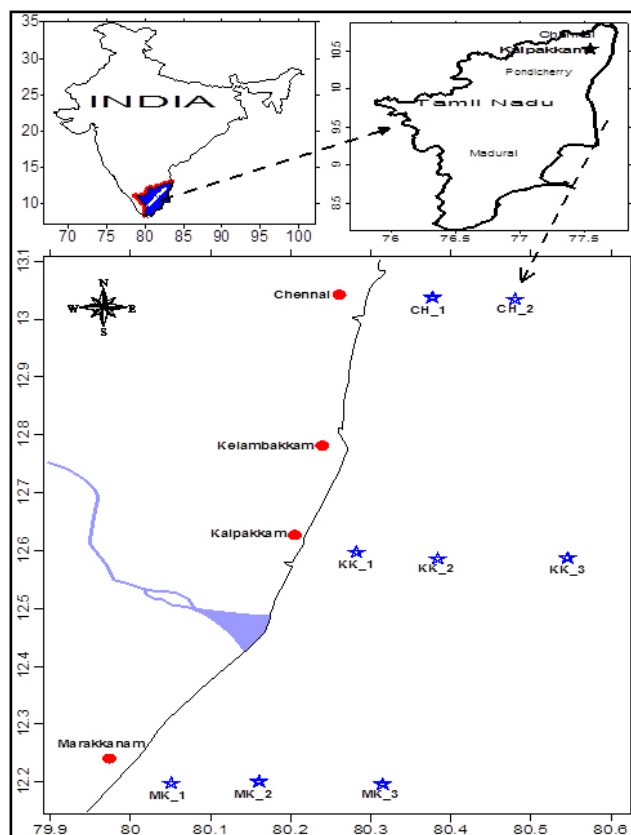


Fig. 1 — Study area showing the sampling locations, Tamil Nadu coast, Bay of Bengal

stock standards solution (1000 mg/l) of phosphate (Merck- Certipur). Certified reference material (CRM) sediment, CRM 7001 from the CZECH Metrology Institute, Czech Republic, was used to find out the analytical and instrument accuracy of the methods. The recovery was found to be 95% of the certified value. Phosphorus fractions in the sediments have been characterized by their differential solubility in various chemical extractants. The phosphorus extraction method used in the present study determined five phases of sedimentary phosphorus²⁶⁻²⁷. The extraction of the sediment samples was carried out with 1.0 M NH_4Cl , 1.0 M NH_4F , 0.1 M NaOH , 1.0 M H_2SO_4 for loosely bound phosphorus (Lo-P), Aluminum bound phosphorus (Al-P), Iron bound phosphorus (Fe-P) and calcium bound phosphorus (Ca-P), respectively. Total phosphorus (TP) in the sediment was determined by igniting the sediment at $550 \pm 10^\circ\text{C}$ in a muffle furnace followed by shaking of the sample with 1N H_2SO_4 for 16 h²⁸. The total inorganic phosphorus (TIP) was the sum of all inorganic bound phosphorus. Organic phosphorus was calculated as the difference between TP and TIP²⁹. The phosphate in the extracts was estimated with the phosphomolybdate blue method³⁰. Organic carbon was analysed using Shimadzu TOC analyzer (model – TOC VCP-H-SSM-500). Correlation analysis was carried out using XLSTAT-Pro from Statsoft. In order to find out the distribution pattern of phosphorous, contour maps were plotted using Surfer (ver. 8.02) from Golden software.

Results

Summary of the results of physical parameters and various phosphorus fractions in the offshore marine sediment off south east coast of India are presented in Table-1. The textural composition of the sediment

Table 1 — Summary of phosphorus fraction and physical parameter results obtained during the study

Parameters	Minimum	Maximum	Avg. ($\pm$ SD)
Lo_P(mg/kg)	12.63	45.62	26.28 $\pm$ 11.55
Al_P (mg/kg)	45.42	87.59	66.56 $\pm$ 15.26
Fe_P (mg/kg)	0.23	15.30	5.79 $\pm$ 5.60
Ca_P (mg/kg)	298.75	998.57	588.60 $\pm$ 288.11
TIP (mg/kg)	376.34	1109.24	687.24 $\pm$ 296.88
OP(mg/kg)	40.19	263.16	137.42 $\pm$ 79.74
TP(mg/kg)	469.35	1219.34	824.66 $\pm$ 303.33
Sand (%)	80.94	94.41	87.22 $\pm$ 5.01
Silt (%)	3.69	9.98	6.85 $\pm$ 2.15
Clay (%)	1.24	12.22	6.44 $\pm$ 4.49
OC (%)	0.16	1.06	0.47 $\pm$ 0.32
IC (%)	0.54	7.38	2.29 $\pm$ 2.32

showed a distinct spatial variation. The sand fractions ($> 63\mu\text{m}$) were dominant in all the station and showed a decreasing trend towards offshore region (Figure 2, Figure 3a). As per the classification of Folk³¹, contribution of sand, silt and clay to the marine sediment was 87%, 7% and 6%, respectively (Figure 2). The mud (silt+clay) content was found to be the lowest at the Chennai transect and the highest at Kalpakkam transect (Figure 3b). The organic carbon (OC) content showed a wide variation, ranging from 0.16% to 1.06% with an average of 0.47% (± 0.32).

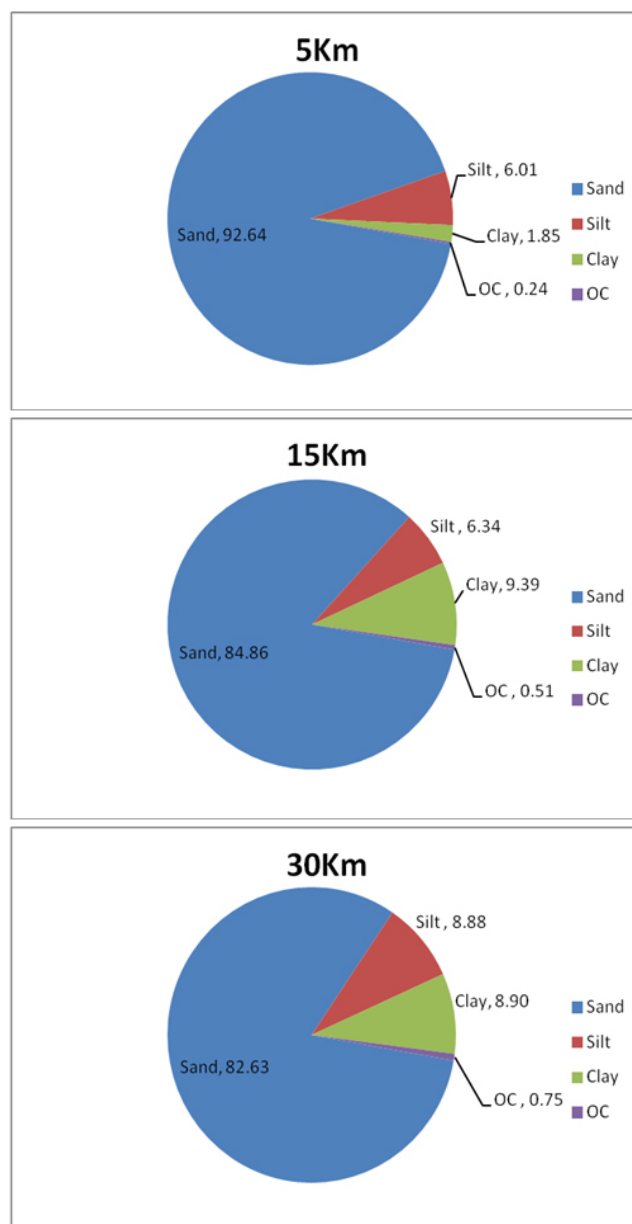


Fig. 2 — Percentage contribution (average) of sand, silt, clay and organic carbon at 5km, 15 km and 30km sampling stations off the Tamil Nadu coast

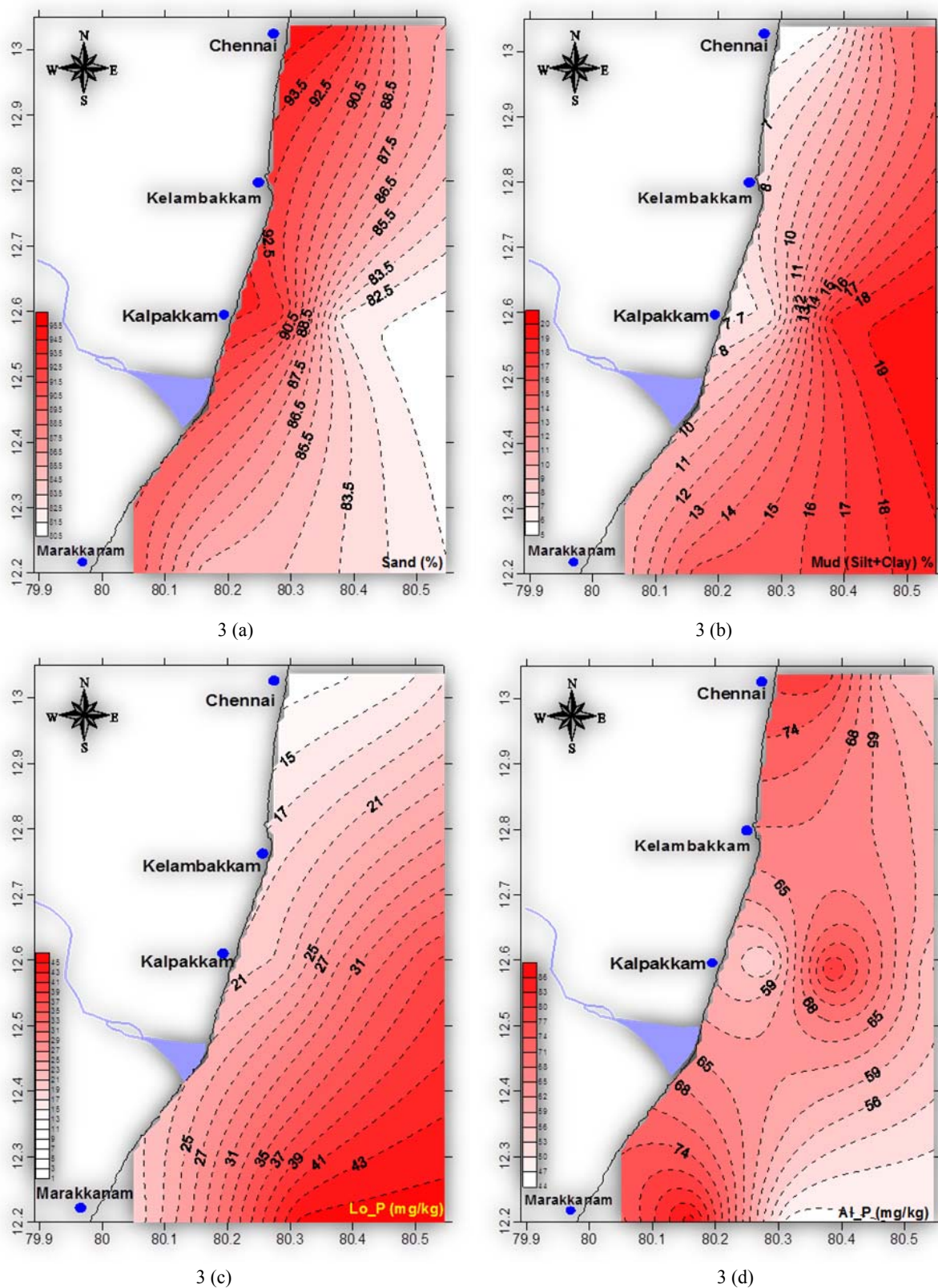
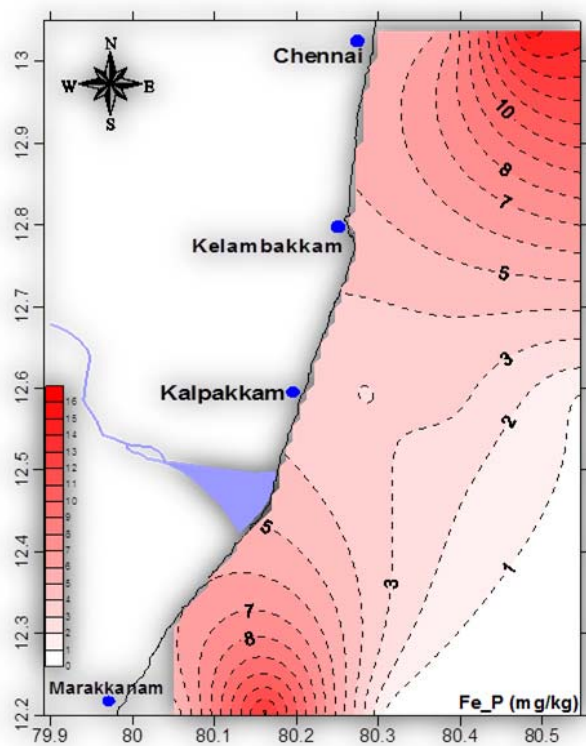
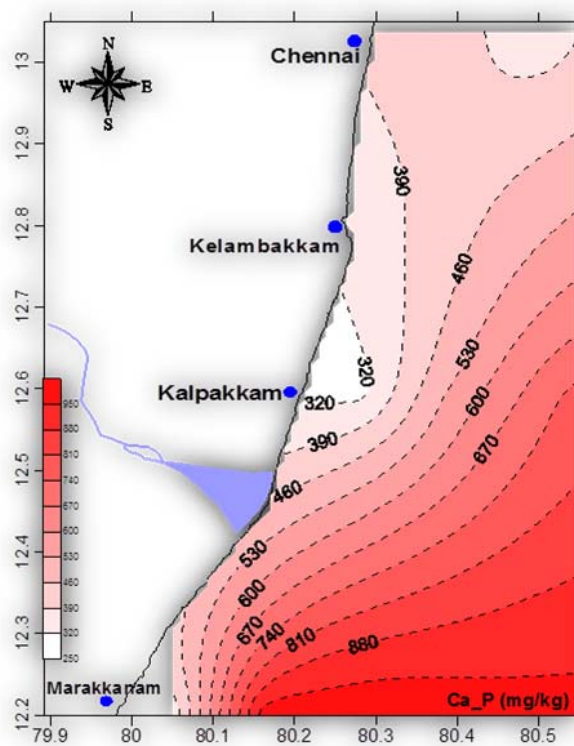


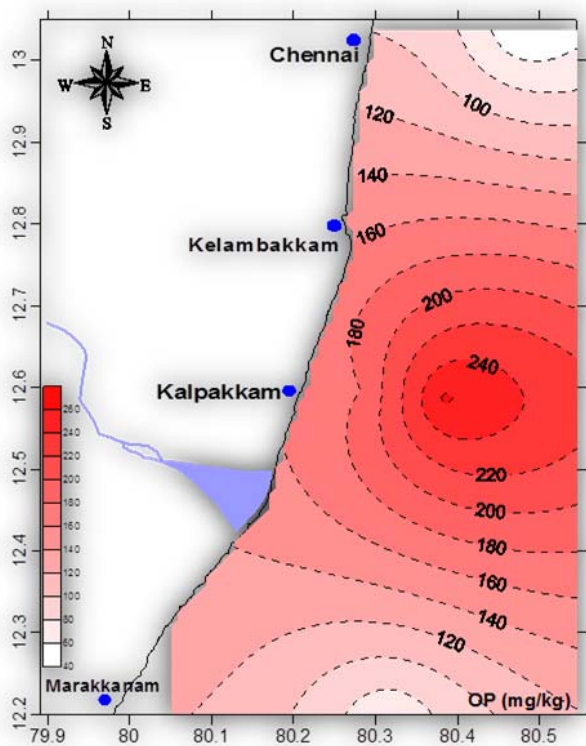
Fig. 3a-d — Contour maps showing spatial distribution of sand percentage, mud (silt+clay) percentage, loosely bound P (LoP) and aluminum bound P (AlP) in the surface sediments off Tamil Nadu coast



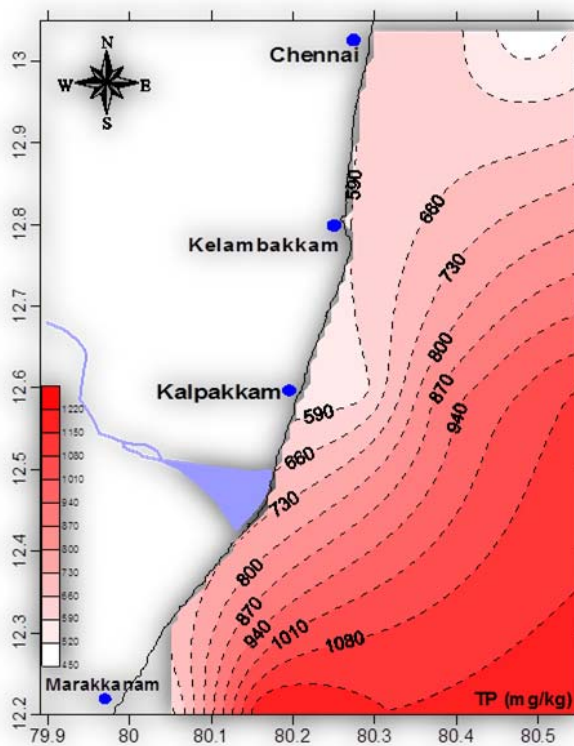
3 (e)



3 (f)



3 (g)



3 (h)

Fig. 3e-h — Contour maps showing spatial distribution of iron bound P (FeP), calcium bound P (CaP), organic P (OP), total P (TP) in the surface sediments off Tamil Nadu coast

It also showed a similar increasing trend from near shore to offshore regions. Distribution pattern of inorganic carbon (IC) content showed similar characteristics to that of OC and ranged from 0.54% to 7.38% with an average of 2.29% (± 2.32).

Total phosphorus content in the offshore sediments ranged between 469.35 and 1219.34 mg/kg with an average of 824.66 mg/kg. Among the inorganic phosphorus fractions, Lo-P (mg/kg) ranged between 12.63 and 45.62 with an average of 26.28, contributing 3.19% to TP (Figure 4). A gradual increase in contents of Lo-P from near shore to offshore region was observed at all the three transects (Figure 3c). The Al-P (mg/kg), Fe-P (mg/kg) and Ca-P (mg/kg) contents ranged from 45.42-87.59 (average: 66.56), 0.23-15.30 (average: 5.79) and 298.75-998.57 (average: 588.60), respectively. The contribution of Al-P, Fe-P and Ca-P to TP content was 8.07%, 0.70% and 71.38%, respectively. A similar spatial distribution trend as that of Lo-P was observed for Ca-P (Figure 3f). However, Al-P and Fe-P did not show any particular trend with almost similar distribution pattern (Figure 3d, e). Total inorganic phosphorus (TIP) represented the sum of the four fractions such as Co-P, Al-P, Fe-P and Ca-P. The range and average concentration of the OP in the present study was 40.19-263.16 and 137.42 mg/kg, respectively. Contribution of OP was found to be 16.16% of the TP content. It showed a clear decreasing trend from near shore to offshore region (Figure 3g). The TIP content was much higher than organic phosphorus (OP) in the study area and ranged from 376.34-1109.24 mg/kg with an average of 687.24 mg/kg. TIP contributed 83.34% of the TP content. Relative abundances of the different forms of phosphorus followed the order: Ca-P > OP > Al-P > Lo-P > Fe-P.

Discussion

The shelf sediment off Tamil Nadu coast was found to be sand dominated with relatively low silt and clay content corroborating earlier observations from this region³². Absence of perennial riverine system in this region, which otherwise could have caused the silt and clay deposition, could be attributed to the above observation. Similarly, relatively high sand contents observed at near-shore locations as compared to that of the offshore locations could be attributed to the continuous wave action and churning of the shallow water near the coast, which prohibits the settlement of fine particles. Distribution of these parameters with respect to each transect showed that,

sand percentage was the highest at Chennai (90.90) followed by Marakkanam (85.39) and Kalpakkam (86.6). However, the Kalpakkam transect showed relatively high mud (15.14%) and organic carbon (0.60%) content as compared to Chennai and Marakkanam transects. Organic carbon content also exhibited a similar increasing trend towards the

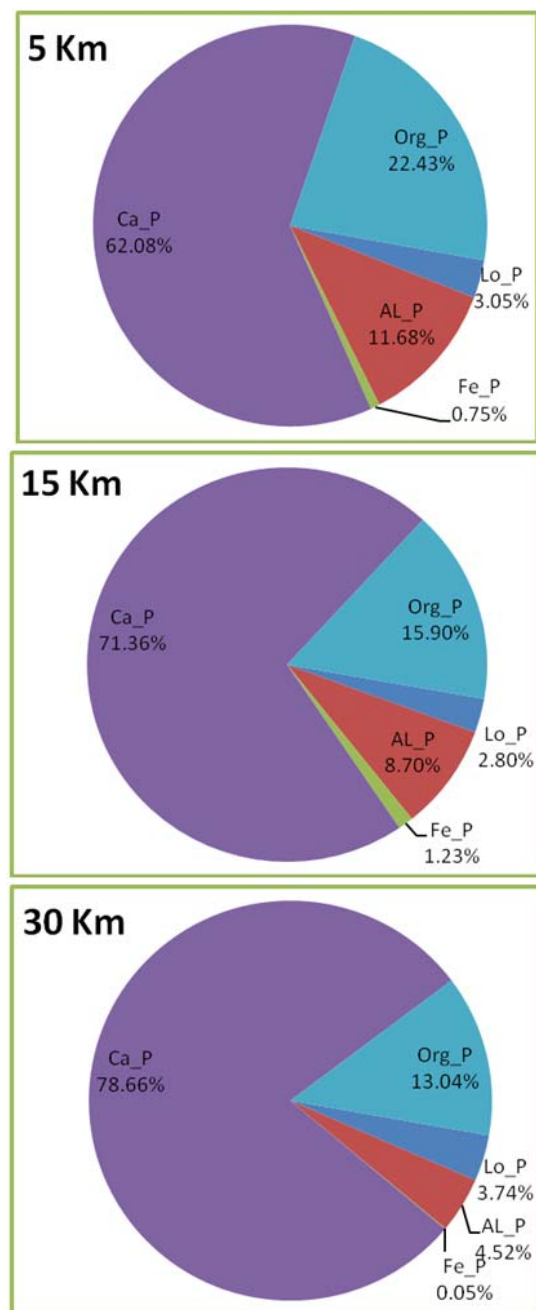


Fig. 4 — Percentage contribution (average) of loosely bound P (LoP), calcium bound P (CaP), iron bound P (FeP), aluminum bound P (AlP) and organic P (OP) in surface sediments off Tamil Nadu coast

offshore region. The range of OC content observed at this location was comparable to the values reported from Krishna-Godavari basin and Tuticorin shelf region of the Gulf of Mannar³³⁻³⁴.

Phosphorus is associated with and liberated by sediment particles through a complex series of absorption and disassociation³⁵. Its concentration in the seawater is regulated mainly by the release from sediments which is further dependent on the contents of different P fractions. However, all P fractions are not released from the sediments to be easily available for the biota³⁶. In the present study, Ca-P emerged as the dominant fraction of P present at this location which has also been reported for other coastal regions³⁶. Generally Ca-P is a highly stable mineral in alkaline environment³⁷. In coastal sediments, P associated with Ca is present in solids of various types and origins. Though there could be two groups of Ca associated P such as: i) the detrital fluoro apatite of igneous & metamorphic origin and ii) Ca bound P in biogenic skeletal debris & carbonate fluoro apatite precipitations³⁸. The Ca-P in this study represented all the above forms. Authigenic and biogenic Ca-P in sediments have been shown to represent a sink for reactive P³⁸. Those forms like detrital are almost insoluble under the physicochemical conditions of marine waters and remains stable under both oxidizing and reducing conditions and cannot be released again from the sediment to the overlying water³⁹⁻⁴⁰. Thus, P concentration in this extraction step will not be bioavailable so easily; being considered permanent burial. Therefore, knowledge of the level of Ca-P concentration is useful to evaluate the proportion of bioavailable P in coastal areas. The high Ca-P is related mainly with the over saturation of calcite and co-precipitation of phosphate⁷. The high adsorption capacities of carbonates also contribute to this form of P in some extent⁴¹. It is well known that calcite is produced at high salinities through precipitation reactions and biological activity, both in estuarine and marine environments, which forms an adsorption substrate for dissolved phosphate. This could be a plausible reason behind the observed dominance of Ca-P in the study area, as the coastal water at this location is sparsely influenced by freshwater input throughout the year. However, the gradual increase in Ca-P contents from near shore to offshore region could be attributed to the above fact that, the influence of freshwater, through land runoff, riverine discharge etc during monsoon, gradually

decreases from coastal to offshore areas which ultimately leads to production calcite in the offshore waters at a higher rate as compared to that of the near shore water.

Phosphorus bound to Fe and Al through chemisorptions⁴² acts as indicator of algal available P⁴³. These P fractions could be released for the growth of phytoplankton when anoxic conditions prevail at sediment– water interface⁴⁴. However, according to Hedley et al.⁴⁵ this fraction is less plant-available and usually associated with humic compounds and amorphous and crystalline Al and Fe oxides. Some studies have indicated that Fe/Al-P was the major sink for the available P and was in equilibrium with some other fractions⁴⁶. Moreover, P fractions in surface sediments were in a dynamic equilibrium with overlying water⁴⁷, which can exchange P with each other under certain environmental conditions⁴⁸. In general, dominance of Fe-P and Al-P has been reported from areas of high freshwater influence⁴⁹ as phosphorus gets adsorbed to Fe and Al oxides present in the freshwater⁵⁰. Thus, low Fe/Al-P contents observed here could be attributed to the above reason. Furthermore, as a general rule, concentrations and proportions of Fe/Al-P is higher in the finer sediment⁸. Stone and English⁵¹ in a specific study of Fe/Al-P concentrations in different grain-size fractions have observed an inverse relationship between Fe/Al-P concentrations and particle size. Hence, the low levels of Fe/Al-P found in this area were therefore attributed to the sandy nature of the sediments.

Concentration of loosely adsorbed P (also called water soluble; labile; exchangeable + carbonate-associated; hydrolyzed P) directly determines the bioavailability of P⁵². It gives an estimate of the immediately available phosphorus for consumption. Contribution of Lo-P was the third highest among the IP fractions in the present investigation. Lo-P showed positive correlations ($p \geq 0.005$) with Ca-P, IP and TP (Table-2), which implied that, it maintained a constant proportion with all these fractions. Some of the studies have reported increased Lo-P with increase in fine grain particles^{8, 52}. The present study corroborated the above observations as an increase in Lo-P contents was observed with increase in the mud percentage, which was further confirmed from the observed strong positive correlation between Lo-P and mud and its strong negative correlation with sand. An increase in Lo-P content in the offshore region can

also be attributed to the above reason. Moreover, its increasing trend towards offshore, could also be ascribed to the fact that high water turbulence near to the coast enhance high exchange of Lo-P from sediment to water column as compared to that of the offshore regions.

OP in marine sediment includes apatite bounded phosphate and biochemical components such as nucleic acid, Lipid and sugar that are bound to phosphorus⁵³. According to Berner⁵⁴, OP has been considered as a source of dissolved phosphate present in interstitial water in sediment due to bacterial regeneration. Contribution of OP to TP was the second highest in the present study which suggested that mineralization of the phosphate containing organic matter at the sediment surface⁵⁵ is predominant process at this location. Similar phenomenon has also been reported from the coastal waters of South India⁵⁶. Increase in organic matter with that of fine sediment fractions leading to increase in OP has been reported from some areas⁵⁷. In the present investigation, OP neither yielded any significant correlation with sediment particle size nor with any other parameter. Relatively low OP contents observed at offshore locations (Figure 3g) could be due to the fact that, quantum of accumulation of anthropogenic inputs in the near shore area are relatively high as compared to the offshore regions. Moreover, the high organic phosphorus contained in the near shore sediments might be due to flocculation and precipitation processes involving humic acids from advective mixing of sea water with the nutrient rich fresh water inputs.

Even though, TP is not representative of the reactive fraction of phosphorus in sediments, it is usually the only component that can be compared to existing data based on lesser defined geochemical determinations. TP showed positive correlation with almost all the inorganic fractions (Lo-P- $r=0.964$; Ca-P- $r=0.964$; IP- $r=0.981$) whereas it did not yield any significant correlation with the organic fraction. TP as well as the OP contents observed in the present study were comparable to that of the other marine environments of India and elsewhere^{22, 58}.

Considering the important role of phosphorus in marine productivity and its existence in various fractions (soluble and non-soluble), the notion of bioavailability must be taken into account. Bioavailable phosphorus in sediments corresponds to the amount that can be released easily, for algal

growth⁵⁹. Though bioavailability can be assessed by bioassays⁶⁰, knowledge of the various forms of phosphorus is useful in determining the upper limit of the potentially bioavailable phosphorus. Ca-P, which remains insoluble in marine conditions and Fe/Al-P, which cannot be released unless anoxic environment prevails, practically do not contribute to the bioavailable fraction. The present study area being an open shallow coastal environment that supports turbulence and mixing, the Ca-P and Fe/Al-P fractions cannot be bioavailable under normal circumstances. However, Lo-P can progressively be released when the phosphate concentration in the water column is lower than that in the pore water. Similarly, OP can be progressively bioavailable due to remineralization. Therefore, initially only Lo-P and OP should be considered as potentially bioavailable fractions, which contributed from 9.3 to 36.2% (avg. 20.9%) of the TP during the present study.

Distribution of phosphorus fractions and other sedentary parameters with respect to different transects showed a distinct pattern and the same has been confirmed by the cluster analysis. Three different clusters representing three transects were evident from this analysis with only exception of MK1 which combined with the Kalpakkam cluster (Figure 5). It showed that geochemical characteristics of the sediment of these transect were entirely different from each other. Relatively high OP content

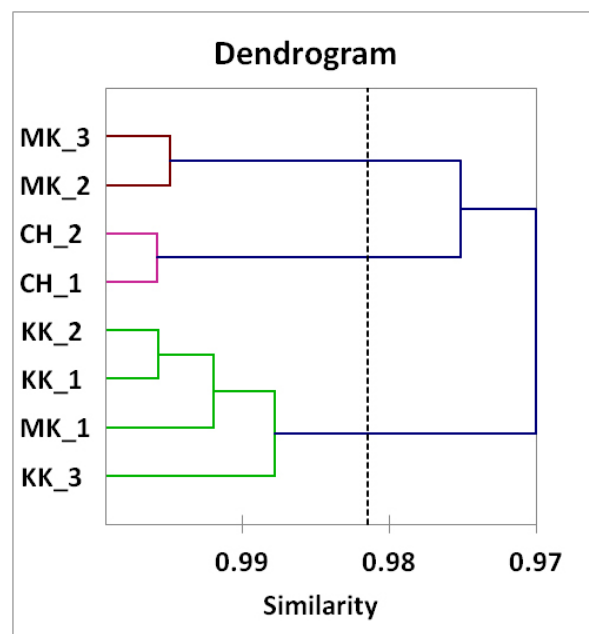


Fig. 5 — Cluster analysis showing three distinct clusters formed by sampling locations from three transects.

was observed in the Kalpakkam region which can be attributed to the fact that organic carbon as well as mud content of this region was relatively high as compared to other transects. It is well known that sediment with finer particles contains relatively high organic matter which leads to increase in OP content⁵⁷. Similarly, relatively high TP content was observed at Kalpakkam and Marakkanam as compared to that of the Chennai which can also be attributed to the above reason²⁷.

Conclusion

The shelf sediment off the Tamil Nadu coast was found to be sand dominated with gradually increasing mud and organic carbon content from near shore to offshore regions. Almost all the inorganic phosphorus fractions showed an increasing trend towards offshore. A reverse trend, i.e. gradual decrease in content of organic phosphorus fraction was observed for all the three transects. The most abundant form of inorganic phosphorus in the surface sediment was calcium bound phosphorus which contributed 71.38% of the total phosphorus content. Relative abundance of the phosphorus fractions in the marine offshore sediments followed the order: Ca-P > OP > Al-P > Lo-P > Fe-P. Bioavailable phosphorus fractions contributed from 9.3 to 36.2% (avg. 20.9%) of the TP in the sediment samples. Cluster analysis clearly showed that all the three transects were different from each other as they formed three different clusters.

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