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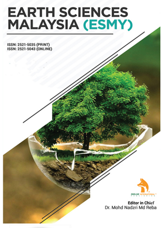

RESEARCH ARTICLE

ELEMENTAL ANALYSIS OF MARINE CORE SEDIMENT BELONG TO CENTRAL WESTERN BAY OF BENGAL BY USING PIXE

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ABSTRACT

The aim of the present work is to estimate the accumulation of various elements related to the core sediment belong to the Krishna-Godavari marginal area with a nuclear analytical technique. Hence, a 4.12 m long marine sediment gravity core was collected from near to Nizampatnam Bay that related to Bay of Bengal and by using particle induced X-ray emission (PIXE) elemental concentrations have been determined. The concentrations of major elements like Fe, Ca, K, Ti, Mn and minor (trace) elements such as Sr, Co, V, Zn, Ni, Cu, Rb, Sc and Cr have been evaluated with the obtained PIXE spectra. These values are interpreted based on the monsoonal runoff coming from the rivers causing to play an important role for bring lot of sedimentary material of the coastal waters to the Bay of Bengal. Further results are interpreted to understand biological and anthropogenic activities, chemical reactions etc during Holocene and Pleistocene periods.

KEYWORDS

Marine sediment, elemental concentration, clay, mineral, Weathering.

1. INTRODUCTION

Sediments (bed or core) provide temporally an indication of aquatic environmental condition; act as a major reservoir for elements and provide information of geochemical distribution along with toxicity of it (Chakraborty, 2019; Caccia et al., 2003). Some of the sediments act as a source of contaminants (Adams et al., 1992; Burton and Scott, 1992). In fact, sediments are the final destination of trace elements, as a result of adsorption, desorption, precipitation, diffusion processes, chemical reactions, biological activity and a combination of all those phenomena takes place (Chatterjee and Massolo, 2009). Sediments are an important sink for heavy elements but when some physical disturbance occurs or there are diagenesis and/or changes in PH or redox potential, they become prime source of elements from the overlying water column (Jones and Turki 1997). Concentration determination of elements that belonging to sediments has also become a common means of assessing the extent of anthropogenic input impact on certain areas indicating sedimentological process along with sediment diagenesis (Hendrizan et al., 2016; Pungky, et al., 2017). The obtained results facilitate to understand paleo-environmental changes of sea (Yao et al., 2012; Rina et al., 2017).

Elements enter into environment and oceans by two ways namely natural sources including erosion of ore-bearing rocks, wind-blown dust, volcanic activity, forest fires and processes derived from human activities by means of atmospheric deposition, through river runoff and direct discharges or dumping (Clark 2001; Noureddine Zaaboub et al., 2014). For some elements, natural and anthropogenic inputs are on the same order of magnitude (for example Hg and Cd), whereas for others (for eg. Pb) inputs due to human activities prevail on natural inputs (Clark, 2001). Very little information is available on elemental concentration of the sediment cores belong to western Bay of Bengal. In the present investigations an attempt has been made to determine the elemental concentrations to understand the sediment depositional environment with reference to modifications of paleoclimatic changes occurred during Holocene and Pleistocene periods of the core.

2. EXPERIMENTAL DETAILS

A 4.12 m long marine sediment gravity core was collected near to Nizampatnam Bay belongs to Bay of Bengal. The core was sub sampled to every 2 cm (gap or space) of the marine sediment. Samples were dried at 60 ^{0}C, grinded and sieved through 230 mesh sizes. Conversion of the samples into fine powder before pelletizing is essential to minimize particle size effects on the results. The fine grinded core sediment samples were mixed with known amount of spec pure graphite powder in 3:2 ratios by weight. The mixed samples were thoroughly grounded, homogenized and finally pressed into pellet form of 10mm diameter. In the present studies, PIXE technique was employed for determination of elemental concentrations of the core sediment samples by using 2.5 MeV proton beam obtained from 3 MV tandem accelerator at Institute of Physics, Bhubaneswar, India.

3. VALIDATION OF THE PRESENT PIXEMETHOD WITH THE CERTIFIED REFERENCE MATERIALS

The observed PIXE spectrum of the certified reference material (CRM) IAEA SL-3 (Lake Sediment) is shown in the Figure 1 and NIST (1515) Pond sediment sample also undertaken in the present work. The elements

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present in these reference materials are identified along with their concentrations those estimated by using GUPIX software program for validation of the presently undertaken experimental facility. These values of NIST (1515) are shown in the table 1. The accuracy of the method is evaluated by using % of deviation. In the present work; measured values of the obtained elements such as K, Ca, Sc, Ti, V, Cr, Mn, Fe, Cu, Zn, Rb, Sr, Zr were compared with the certified values of IAEA and NIST reference materials for their validation. Uncertainty quoted is standard deviation at ± 1σ confidence level (68.3). The experimental values are showing good agreement with the standard certified values. The deviation range of the elements determined with respect to certified values is within ± 8.6 %.

4. RESULTS AND DISCUSSION

Obtained PIXE spectra related to various depth fractions of the sediment core are facilitated to evaluate the elements and their concentrations such as Fe, Ca, K, Ti, Mn (major)and trace elements namely Sr, Co, V, Zn, Ni, Cu, Rb, Sc and Cr (minor)are computed by comparing with the standard spectral values.

4.1 Major elements

The obtained elemental (Fe; Ca; K, Ti and Mn) concentration variation pattern as a function of sediment core depth is shown in the Figures 2 and3 respectively.

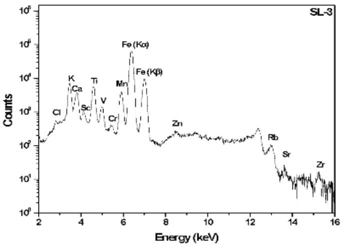

Figure 1: PIXE spectrum of IAEA certified reference material (CRM) SL-3 sample

Table 1: Measured and certified values of NIST (1515) reference material

Element	Certified values	Measured values	% Dev
K	6800 ± 612	6929 ± 78	1.8
Ca	8100 ± 567	7958 ± 106	-1.7
Sc	28	26 ± 0.9	-7.6
Ti	6400	6489 ± 72	1.3
V	250	235 ± 16	-6.4
Cr	75 ± 5.2	80 ± 6	6.2
Mn	1400	1506 ± 46	7.0
Fe	65300 ± 3265	64156 ± 425	-1.8
Cu	210 ± 12.6	196 ± 8	-7.1
Zn	343 ± 17.5	348 ± 19	1.4
Rb	42	39 ± 1.1	-7.6
Sr	110	106 ± 10	-3.8

()-information values, BDL-below detection limit

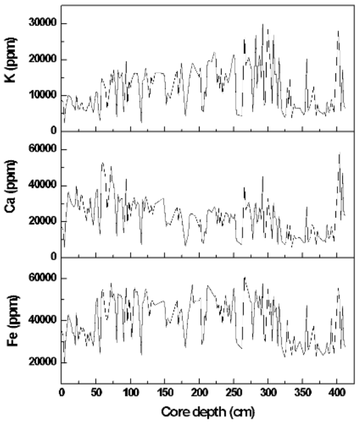

Figure 2: Down core variations of Fe, Ca and K

Based on the observed variation pattern of the elemental concentration that shown in the Figures 2 and 3; the following conclusions are drawn:

i) Low values of elemental concentrations are observed as a function of the core depth regions 0 – 50 cm and 312 – 398 cm that belong to Holocene and Pleistocene periods respectively.

ii) Higher values of elemental concentrations are obtained in the region 52 – 126 cm; which belong to Holocene period while in the Pleistocene period 260 -310 cm and 402 – 404 cm show higher values of concentrations.

iii) Computed concentration values of major elements related to the various regions are displayed in the table 2.

iv) Regions of core depth corresponding to Holocene or Pleistocene period are corroborated by the obtained results of Fe deposition in the present study during the Holocene and Pleistocene periods.

Iron in deep sea sediments generally associates with lithogenous, hydrogenous and metalliferous components (Chester and Aston 1976). Terrigenous solids particularly clay minerals along with other ferro minerals such as augite, feldspar, hornblends, magnetite and ilmenite are the major suppliers of iron (Periakili, 2000). Depositions of manganese nodules, leaching of the more soluble manganese, a volcanic emanation, biogenic or bacterial mat formation and organic precipitates as coating on skeletal parts of marine organisms are other sources of Fe accumulation (Wakeel and Riley 1961; Rina et al., 2017).

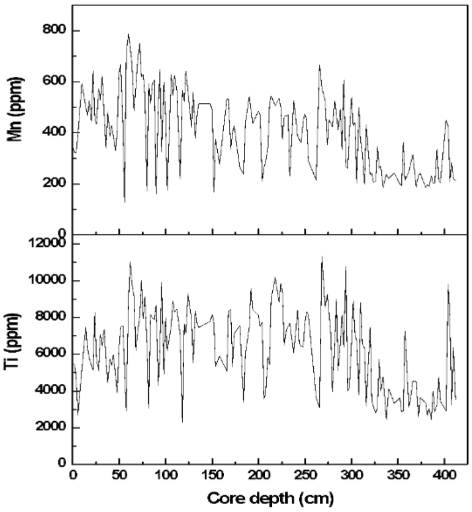

Figure 3: Down core variations of Ti and Mn

In the present work, iron concentration is found to vary from 61594.2 to 19781.3 ppm having an average value 40619.4 ppm. The observed maximum and minimum values of Fe correspond to the locations are at a depth of 266 – 268 cm and 04 – 06 cm respectively. Low values of Fe are obtained at top layer (0 – 50 cm) and also at bottom layer 320 – 398 cm, which may be attributed to early and late diagenetic process (Dymond, 1984). The observed values of iron in the present study show good agreement with the earlier reported values pertaining to the Bay of Bengal sediments that contain to have a range of 2 to 6% of Fe (Sarin, 1979). Relatively lower values (4.06%) are obtained in the present core belong toNizampatnam Bay when compared with the earlier studies (6.51%) (Venkateswara, 2008). The observed lower values of Iron can be attributed to the higher concentrations of obtained calcium carbonate that showing an inverse correlation in between these two constituents (r= -0.61).

This may be due to the dilution factor of calcium carbonate. Iron showed significant positive correlations with Ti (r = 0.93), V (r = 0.93), K (r = 0.87), Rb (r = 0.79), Mn (r = 0.79), Ca (r = 0.71), Ni (r = 0.66), Co (r = 0.43), Sc (r = 0.37), Cu (r = 0.37), Sr (r = 0.34), Cr (r = 0.32), Zn (r = 0.29), OC (r = 0.05). The observed positive correlations of iron with Ca can be understood on the lines those reported earlier by indicating their correlation with sedimentary units (Croudace et al., 2006). A group of researchers mentioned that the iron ratios with Ca and K are most commonly used for paleo climatic reconstructions (Govin et al., 2012). Further they concluded that Fe and Ti are related to the sediment siliciclastic components varying with terrigenous fraction of the sediment. Potassium may be obtained from potassium feldspar or illite. Hence these elements are mostly derived from land sources (terrigenous). Earlier investigators established that fine grained organic residues have a special ability to fix iron by absorption and that iron forms an important micro constituent of all living tissues; which later contribute to the organic matter in the marine sediments (Rankama and Sahama, 1950, Mason, 1958).

Presence of calcium in marine sediments takes place in biogenic form as $CaCO_3$ in shell debris and oolites. Most of the calcium is precipitated as calcite by marine organisms in deep sea sediments having distribution as a function of biological processes and crustal minerals such as clay minerals and feldspars. The concentration of calcium in the present sediment core ranges from 58573.2 to 5804.4 ppm reflecting an average of 23568.3 ppm as shown in the Fig 2. Higher Ca values are observed in the depth range 50-100 cm; at the same time $CaCO_3$ concentrations found to decrease due to dissolution of the $CaCO_3$ as oxic conditions prevailed in the upper 0 -50 cm region. Lower values of Ca are observed at the depth range 320-398 cm due to anoxic conditions prevailed in this range besides the observed increase of organic carbon in the same range. The deposition of Ca found to be higher during Holocene period relative to Pleistocene period.

The observed lower values during Pleistocene period can be derived from the terrigenous source. Observed strong positive correlations of Calcium with Sc (r = 0.84), Mn (r = 0.82), V (r = 0.76), Fe (r = 0.71), K (r = 0.59), Rb (r = 0.51)may be due to biological contribution of these elements while less positive correlation with Ni (r = 0.40), Sr (r = 0.36), Cu (r = 0.33),Ti (r = 0.30), Cr (r = 0.16), $CaCO_3$ (r = 0.44), $MgCO_3$ (r = 0.17) indicates that these are contributed by both biological and terrigenous detrital sources. Negative correlation with OC (r = -0.30) is obtained in the core. According to a study occurrence of carbonaceous shell material is primarily responsible for exhibiting relatively higher concentrations of calcium in the Bay of Bengal sediments (Siddique, 1967). Average value of calcium concentration (2.35%) obtained in the present study seems to be similar with the earlier reported value (2.50%) of sediments belong to Nizampatnam Bay; middle Bengal Fan (2.68%), besides its fairagreement with the Indian River sediments (2.45%) (Venkateswara, 2008; Subramanian, 1985).

Earlier authors, mentioned about association of calcium with Fe and Ti as indication for paleo climatic reconstructions (Govin et al., 2012). Potassium (K) presence in marine sediments can be attributed to detrital feldspars, clays, interstitial pore solutions, potash feldspars, biotite and muscovite besides minor constituent of other silicates (Govin et al., 2012). During weathering, K is dissolved easily and removed from solution, adsorbed on clay minerals. Its concentration in the present core ranges from 29883.6 (292-294 cm) to 2434.8 (116-118 cm) ppm exhibiting an average value 12206.3 ppm as depicted in the Figure 2. The lower values of K are observed in the upper part (0 – 58cm) of the core. It found to increase gradually from 0 – 292 cm, beyond which it decreased up to 396 cm. The observed higher values of K in the depth range 0 to 292 cm can be ascribed to more detrital sources arise from the land, whereas low values after 292 cm may be due to low terrigenous supply during the middle Pleistocene period.

Potassium showed a significant positive correlation with Fe (r = 0.87), V (r = 0.75), Rb (r = 0.65), Ca (r = 0.59), Mn (r = 0.57), Ni (r = 0.52) due to its common source of occurrence i.e., from terrigenous. Less correlation with Co (r = 0.44), Cr (r = 0.38), Zn (r = 0.32), Ti (r = 0.27), Sc (r = 0.25), Sr (r = 0.24) indicates that relative contributions of terrigenous are less. Positive correlation with organic carbon (r = 0.25) and negative correlation with $CaCO_3$ (r = -0.29) shows adsorption in the former and more dissolution in the latter. The average concentration of potassium obtained in the present study (1.22 %) is higher than Indian River sediment average (1.21%) values while it found to be lower than the oceanic sediment average value (2.68%) (Wakeel and Riley 1961; Subramanian 1985). Higher levels of Fe, Ti, Mn and K presence signifies low characteristics in the darker sediments (Dida et al., 2018).

Table 2: Down core significant variations of major elements concentrations (± 8.6 %) in the core sediment

Core depth (cm)	Fe (ppm)	Ca (ppm)	K (ppm)	Ti (ppm)	Mn (ppm)
0	35599.5	17469.2	8932.8	5484.4	352.45
10	42948.0	36678.6	10174.8	5202.4	593.56
14	38032.2	30270.8	9430.8	7511.2	503.31
20	28550.9	29576.4	6026.4	5430.8	446.88
22	43863.8	40058.2	10177.2	5144.4	643.91
30	34968.1	34241.2	7905.6	7091.2	528.96
32	36560.3	28949.2	6159.6	6471.2	619.97
40	30367.6	25180.4	7189.2	5736.4	387.79
42	34858.0	33695.2	10016.4	5406.0	428.45
48	35594.1	28596.4	8251.2	3890.4	414.77
50	48318.9	36002.4	11077.2	6238.8	629.85
52	50804.1	38124.8	10794.0	7486.4	669.37
58	53724.4	51650.2	14821.2	2921.6	710.22
60	54978.3	53102.0	14395.2	8892.4	789.45
62	49122.1	48927.2	13208.4	11046.4	733.02
64	43265.9	44753.8	12021.6	9892.2	676.59
70	49114.2	39061.4	12010.8	7150.8	674.88
72	57870.6	50885.8	16042.8	8086.8	751.45
74	50629.6	41776	13579.2	10009.6	621.3
76	52957.4	41119.4	17125.2	7966	629.47
78	48336.6	28837.2	12436.8	9124.4	552.33
82	52850.9	35260.4	17164.8	3088	594.13
84	51930.4	30788.8	14870.4	8161.2	515.66
86	52265.5	32657.8	15112.8	7972	594.7
88	50118.7	32841.2	16425.6	7906	608.57
94	55567.2	43799	19530	6336.8	649.99
98	50364.9	31476.2	14121.6	4963.2	599.07
106	55976.5	33549.6	16285.2	7547.2	629.47
108	53495.2	27266.4	16088.4	8892.8	548.72
110	53973.1	30920.4	16153.2	8314.4	623.96
120	50266.8	25445	14581.2	7641.2	525.16
122	55212.1	34038.2	17692.8	7170.8	641.06
266	61594.2	40101.6	26466	3117.6	666.9
268	57907.9	26499.2	18489.6	11374.8	578.74
272	52914.7	30972.2	20688	8556.4	525.92
274	49401.4	26240.2	18810	9256.8	354.35
282	54618.3	30545.2	26806.8	6707.6	528.2
284	50305.6	19196.8	12186	9018.4	470.25
288	52111.0	28089.6	21241.2	6581.2	491.15
292	57637.6	45325	29883.6	6414.8	608.38
300	55150.4	32972.8	28480.8	6753.2	541.5
308	51759.9	30311.4	26877.6	3806	499.51
310	44111.4	22419.6	15784.8	8803.6	372.97
312	46290.4	21434	16099.2	6590.8	320.34
316	48067.9	22724.8	20546.4	3195.2	433.01
356	47310.1	28189	20281.2	2926.8	362.52
392	41050.1	17407.6	8973.6	3265.6	338.39
402	55628.1	43880.4	27955.2	2921.6	449.16
404	50073.5	58573.2	22160.4	9838.8	423.51

Titanium (Ti) can be used as an indicator of terrigenous debris. It is chemically un-reactive, usually non anthropogenic element and shows positive correlation with decreasing grain size (Belkin and Sparch, 1993; Cho 1999). The concentration of Ti found to vary from 11374.8 to 2317.2 ppm showing6279.9 ppm as an average value that shown in Figure 3. Ti showed lower values at top layer 0 – 50 cm, further its concentration found to increase up to 318 cm; beyond which it decreases with some fluctuations of its values. Higher Ti values reflected in the range 58 – 318 cm may be derived from the land source that adsorbed on the iron oxide and scavenged from seawater resulting to settle down finally in the sediments. Observed lower Ti values in the top layer (0-50cm) can be ascribed to the decreased terrigenous source due to weak monsoon activity during this period. The deposition of Ti is found to be higher during Holocene period than Pleistocene period due to higher monsoon activity, whereas lower deposition of Ti during Pleistocene period due to decreased terrigenous source is less because of weak monsoon activity.

Significant positive correlations of Ti are observed with V (r = 0.45), Rb (r = 0.45), Fe (r = 0.42), Mn (r = 0.40), Ni (r = 0.32), Ca (r = 0.30), Cu (r = 0.29). It indicates that these metals originated from the same terrigenous source. A group of researchers used Ti/Caratio for understanding precipitation (Fraser et al., 2014). Observed low positive correlations between Ti and K (r = 0.27), Co (r = 0.27), Sr (r = 0.17), Cr (r = 0.17), Sc (r = 0.11) shows that the origination of these metals belongs to either natural or anthropogenic source. The average concentration of Ti in the present core is 0.62%. The similar values (0.65% and 0.56%) were earlier reported with the central Bay of Bengal sediments and found to be higher when compared with sediments of the western continental margin of India (0.42%) and Indus shelf (0.37%) (Satyanarayana and Venkataramana, 1996). Thus, these detrital minerals function as controlling factors for the distribution of Ti. This is authenticated by the significant positive correlations observed earlier between Ti and Fe in the core sediments belong to central Indian Ocean that attributed to the continental origin (Negender and Mudholkar, 1989).

The higher content of Manganese (Mn) in deep sea sediments originates from different sources namely volcanic eruption; sub-marine weathering related to igneous rocks bacterial precipitation, biological extraction and chemical precipitation (Goldberg, 1954; Goldberg and Arrhenius 1958; Peterson and Prell, 1985; Arrhenius, 1952). There are three accretionary ways for manganese in the marine sediment namely hydrogenous precipitation on seafloor that impacted by oxygen content, oxic and sub-oxic diagenesis (Yao et al., 2012). The concentrations of manganese in the present core as a function of depth areshown in the Figure 3. Mn values are found to vary from 789.4 to 129.7 ppm having an average value 411.7 ppm. The region50 – 125 cm of the core is characterized by higher concentrations of Mn with a decreasing trend towards down wards of the core. Lower Mn values are recorded in the depth range 314 -396 cm. The distinct enrichment in the mentioned range of the core may be attributed to the post depositional remobilizations of Mn^{2+}. It migrates in the pore waters to surface where it gets oxidized to insoluble hydrous manganese (Mn^{4+}) oxide.

Positive correlations observed between Mn and Ca (r = 0.82), V (r = 0.81), Fe (r = 0.79), Sc (r = 0.65), Rb (r = 0.58), K (r = 0.57)Ti (r = 0.40), which indicating their source of origin is either from ferro –manganese associated oxides within the sea, biological or terrigenous source (Rina et al., 2017). Less significant correlations are observed between Mn and Sr (r = 0.39), Ni (r = 0.44), Cr (r = 0.29), Cu (r = 0.31), Zn (r = 0.03), it may be ascribed to anthropogenic source. Negative correlations are observed with OC (r = -0.30) due to the oxic and sub-oxic diagenetic processes. In the present core observed higher Mn values may be due to Mn preservation in micro nodules. Presence of Mn micro-nodules over a sizable area in the middle Fan region of the Bay of Bengal as reported confirms that the environment for the formation of these micro-nodules is widespread and suitable conditions prevail, despite high rate of sedimentation in this region (Chauhan et al., 1994).

It is indicated that biological processes; which involve extraction of manganese from water and its incorporation in foraminifers' tests may make a contribution to the process of bringing manganese to the sediment (Goldberg and Arrhenius, 1958). The obtained higher deposition of Mn during the Holocene period relative to Pleistocene period corroborates the above-mentioned results. The geochemistry of marine sediments also found to show effect on Mn concentration due to changes in the environmental conditions (Lynn and Bontti 1965). Concentration average value of manganese in the present core (0.04%) shows an agreement with the core sediments belong to Godavari delta (0.06%) that reported; sediments of eastern continental shelf (0.06) of India and western continental shelf (0.05%) of India (Murthy, 1983; Venkateswara, 2008; Mascarenhas, 1985). Goldschmidt stated that Mn accumulation near shore sediments has the range 0.05 – 0.08% exhibiting lower value relative to deep sea sediments having the ranges 0.3 – 0.8%, hence the observed value can be understood on the same lines as the present studies are also concerned to near shore (Goldschmidt, 1954).

4.2 Trace elements

Accumulation of trace elements and their concentrations in marine sediments are influenced by several physical and chemical parameters namely grain size profile, particle transport, mineralogical composition oxidation and or reduction reactions etc (Villaescusa-Celaya et al., 2000; Angelidis, 2005). Further trace elements are influenced by provenance, weathering from terrigenous particles, biological productivity, and hydrogenous precipitation within the sediments by early and late diagenesis, sediment sorting and the aqueous geochemistry of the individual elements. Chromium (Cr) is mainly associated with micas and clay minerals such as illite in deep-sea sediments (Moore, 1963). Higher chromium contents are common in the sedimentary basins receiving higher supply of ferromagnesian minerals like micas, clay minerals particularly Illite, chromites (Krishnan,1982). Chromium concentration of the core sediment is depicted in the Figure 4. Its value found to be in between 695.6 to 33.4 ppm showing an average value 221.7 ppm. Low values are observed in the sediment depth range 0 to 60 cm and found to increase gradually up to 170 cm due to reducing anoxic conditions.

Lower values in the top core obtained due to the oxidized layer in which the Cr adsorption on to the ferromanganese oxides takes place. Earlier analyzed surface of 45 sediments belong for trace elemental concentrations of three different grain size fractions, fine fraction of Cr average value found to be 218. Average concentration of obtained Cr in the present study 221.7 ppm agrees with the value of Cr obtained for central Mediterranean Sea by Noureddine (Zaaboub et al., 2014). However present value is lower than the Nizampatnam Bay (302 ppm); higher than middle Bengal Fan sediments (188 ppm) and near to Godavari delta core sediment (204 ppm) (Venkateswara, 2008). However, these reported values are higher than the Indian River sediment (87ppm) and world surface rocks (71 ppm) (Subramanian 1985). Chromium is positively correlated with Fe (r = 0.42), V (r = 0.39), K (r = 0.38), Co (r = 0.30), Mn (r = 0.29), Ca (r = 0.24), Rb (r = 0.21), Ti (r = 0.10), due to the influenced ofredox conditions of the sedimentary column and negatively correlated with Cu (r = -0.08), Zn (r = -0.05) in the core indicating their marine origin.

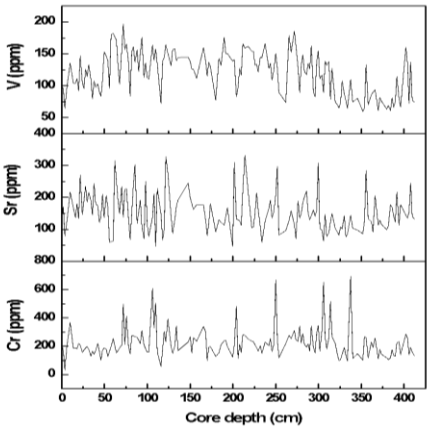

Figure 4: Down core variations of Cr, Sr and V

Strontium (Sr) found to associate with calcium carbonate and clay minerals in marine sediments. Its higher concentrations have been reported in calcareous oozes that strontium could be held in the lattice positions of the lithogenous and hydrogenous components of montmorillonite, zeolites and the skeletal biogenous materials (Wakeel and Riley, 1961; Goldberg and Arrhenius, 1958; Turekian and Kulp, 1956; Riley and Chester, 1971). 80% of strontium is detained in calcareous shell material (Chester and Messiha-Hanna, 1970). Strontium concentration of the core found to lay in the range 332.6 to 46.6 ppmcontaining an average value 153.7 ppm as shown in Figure 4. Obtained higher Sr values in the core upper range 0 – 124 cm relative to the bottom range of the core can be attributed to biological productivity that takes place from the top surface waters due to intensified monsoon during Holocene period. Lower values at the bottom of core may be understood as a cause of the terrestrial source belongs to Pleistocene period. Significant positive correlations of Sr are observed with Mn (r = 0.39), Ca (r = 0.36), Fe (r = 0.34), Sc (r = 0.33), V (r = 0.31), K (r = 0.24), Ni (r = 0.21), Rb (r = 0.18), Ti (r = 0.17) while it negatively correlated with OC (r = -0.19) of the core indicating its association with coarse grained sediments and biogenous components.

Hence Sr positive correlation with the above-mentioned elements in the present core showing that major part of it is associated with biogenic components. Earlier similar results were observed (Mascarenhas et al., 1980). They stated that major contribution of strontium to the deep-sea sediments is due to carbonate shells of coccoliths, corals and foraminifera indicating mixed hydrogenous and biogenous origin. The average concentration of Sr obtained in the present study core 153.7 ppm is higher than those reported for the Bay of Bengal sediments (120 ppm), Godavari delta sediments (115 ppm), lower than the Nizampatnam Bay (189 ppm), middle Bengal fan sediments (189 ppm); average value of Indian river sediments (217 ppm) and also lower than theSr obtained for central Mediterransean sea (Noureddine et al., 2014; Sarin, 1979; Venkateswara, 2008; Subramanian, 1985).

Vanadium (V) may be found as a minor constituent in magnetite, pyroxene, amphibole and biotite minerals (Wedephol, 1974). During weathering of igneous rocks much of vanadium may be incorporated into the resultant clay minerals (Altschuler, 1980). Reinson reported that the fine grained detrital mineral factors exhibit dominant control on the vanadium distribution (Reinson, 1975). Vanadium mainly occurs in the fine-grained terrigenous fraction that associates with aluminum silicates, iron oxides, and additionally with phosphate (Seralathan, 1987; Hartmann and Seralathan, 1986; Noureddine, et al., 2014). The concentration of vanadium in the present study found to vary in the range 196.7 to 59.9 ppm having an average value 120.7 ppm. The observed lower values in the upper part (0 – 48 cm) of the core can be ascribed to anoxic diagenetic process while higher values found in the range 48 – 320 cm with some fluctuations as shown in the Figure 4. The observed smaller increase may be attributed to its reducing conditions of the core. Further lower values are observed in the range 320 – 390 cm of the core.

The obtained average concentration (120.7 ppm) of V in the present study is lower than the core sediment of Nizampatnam Bay (163 ppm), the Godavari delta area (190 ppm), central Mediterransean Sea and equal to Bengal Fan sediments (124 ppm) average value (Venkateswara, 2008; Noureddine et al., 2014). It shows an agreement with the average deep-sea clays 120 ppm and with the igneous rock average values127 ppm (Turekian and Wedephol, 1961; Chester and Aston, 1976). Vanadium (V)found to exhibit positive correlation with Fe (r = 0.93), Rb (r = 0.80), Mn (r = 0.81), K (r = 0.75), Ca (r = 0.76), Ni (r = 0.63), Cu (r = 0.51), Co (r = 0.47), Ti (r = 0.45), Sr (r = 0.31), Cr (r = 0.39), Zn (r = 0.15) indicating its association with terrestrial material. In oxygenated waters, the stable form of vanadium is vanadate and it absorbed into both Fe and Mn oxy-hydroxide phases by having lending support of higher values in the top layers of the core (Wehrli and Stummm 1989). The positive correlation of Vanadium with $CaCO_3$ indicates that part of it may be associated with shells. According to a study, shells in the sediment could provide a major pathway for the depletion of V from seawater (Nicholls et al., 1959). According to a study V is much more strongly adsorbed by hydrous ferric oxide than by hydrated manganese dioxide (Krauskopf, 1956). In present study, vanadium positively correlated with Fe suggesting that part of V is adsorbed on hydrated oxides of iron and manganese.

Zinc associates with Cu, Pb, Ag, Au, Sb, As and Se metal deposits. It presents in mafic minerals. Zn weathering products are zinc sulphates, carbonates, and hydrated silicates besides zinc clay minerals over zinc sulphide ores, Fe, Mn-oxides and organic matter in normal soils. The mobility of Zn is moderately high, limited by its tendency to be absorbed by MnO_2 and by insoluble organic matter (Krauskopf, 1979). A group of researchers mentioned about association of Zn with Fe_2O_3; P_2O_5; MnO and found its enrichment on surface sediments of the Gulf of Tunis having anthropogenic inputs (Noureddine et al., 2014). Its transportation take place along with fine grained particles into the deep oceanic region after crossing the continental shelf (Katz and Kaplan, 1981). In sea water the concentration of Zn ranges from 0.05 x 10 mol kg^{-1}related to the zinc depleted (surface) waters to 9.0 x 10^{-9} mol kg^{-1}; which concerned to zinc enriched (deep) water (Bruland, 1983). Zn shows a strong association with silicic acid in the ocean (Bruland, 1983). It might be a unique proxy for the reconstruction of silic acid concentrations in the past surface waters. A group of researchers observed a correlation between Zn and Si in the north central Pacific location (Bruland et al., 1978). Zn is incorporated into the foraminiferal calcite as solid solution (Reeder, 1983).

In the present study the concentration of Zn found to varyin the range 123 to 32.5 ppm as shown in the Figure 5. Zinc concentrations are lower in the range 0 – 50 cm and further it found to increase with core depth up to bottom exhibiting minor fluctuations. Lower values of zinc in the surface layer is due to its influence of early diagenetic process like copper and control of the surface productivity or may be zinc containing detrital matter; which seems to be lower during late Holocene period. Observed higher concentrations of Zn during Pleistocene period are due to its supply by productivity and also higher contribution of terrestrial detrital origin. The observed positive correlation between Zn and K (r = 0.28), Fe (r = 0.27), Ni (r = 0.25), Cu (r = 0.19), Rb (r = 0.17), V (r = 0.15), Co (r = 0.14) and OC (r = 0.22) indicates that these metals are originated from biological and hydrogenous sources of marine environment besides terrestrial origin. The obtained concentration average (96.8 ppm) value of Zn is higher than the core sediments belong to the Gulf of Mannar, Bay of Bengal (81 ppm) that reported while lower than Gulf of Tunis that mentioned above (Sundararajan and Srinivasalu, 2010).

Rubidium distributional pattern in marine environment could be used as an indicator of transportation pathways relatedto thefine-grained fraction that belong to the bottom of sediments. Rb concentration in the present studies found to vary in between 165.9 to 22.6 ppm containing an average value 88.8 ppm. Lower values are observed at top (0 - 50 cm) and bottom (324 to 390 cm) ranges. The depth wise variation of Rb is shown in the Figure 5. Higher values are observed in the range 51 to 322 cm exhibiting smaller fluctuations due to its inputs from detrital fine-grained fraction and mostly from land source, while the lower values observed in the top layer due to the early diagenesis processes. Rb exhibits positive correlation with V (r = 0.79), Fe (r = 0.77), K (r = 0.65) , Ni (r = 0.61), Mn (r = 0.58), Ca (r = 0.54), Co (r = 0.47), Cu (r = 0.47), Ti (r = 0.45), Zn (r = 0.16), Sc (r = 0.20), Cr (r = 0.19), Sr (r = 0.18) and negative correlations with $CaCO_3$ (r = - 0.06) of the core indicating its association with detrital terrigenous matter and organic matter. A group of researcher observed the association of Rb with total organic carbon (Noureddine et al., 2014). Rbpresent average concentration (88.8 ppm) shows fair agreement with the core sediment of Godavari delta (88 ppm) and lower than the reported value of the Nizampatnam Bay (97.3 ppm), middle Bengal Fan sediments (150 ppm); which was reported and Gulf of Tunis indicating its predominant origin from detrital sources (Venkateswara, 2008; Noureddine et al., 2014).

Nickel (Ni) is a redox sensitive element occurs in deep sea sediments as hydrogenic, diagenetic or hydrothermal source (Pattan, 1995). It is foraged directly from sea water by hydrous manganese oxide (Goldberg, 1954). According to Krauskopf nickel is a major absorbent in the oceanic environment like clay minerals (Krauskopf, 1956). Apatite and organic matter are effective in removing nickel from sea water. Further organic reactions in general, are the controlling factor in the removal of nickel. It is available in the ferromagnesian minerals, pelagic sediments as a colloidal fraction (Chester, 1965). Earlier Ni association in its fine-grained range (<2µm) with Fe_2O_3; P_2O_5; MnO was observed and found its enrichment on surface sediments of the Gulf of Tunis having anthropogenic inputs (Noureddine et al., 2014). Nickel concentrations in the present core lie in the range 123.2 to 40.3 ppm having an average value 85.6 ppm as depicted in the Figure 5. Relatively lower Ni values are observed in the upper range 0 – 42 cm and bottom 326 – 412 cm depths of the sediment core. Lower values in the top oxidized layer (up to 50 cm) may be the loss of Ni along with Mn ions during diagenesis process, whereas at the bottom of the core, the contribution of Ni may be low from the terrestrial clays. The overall distribution of Ni shows that there are no clear-cut maximum and minimum values due to the slope structure of the Nizampatnam Bay.

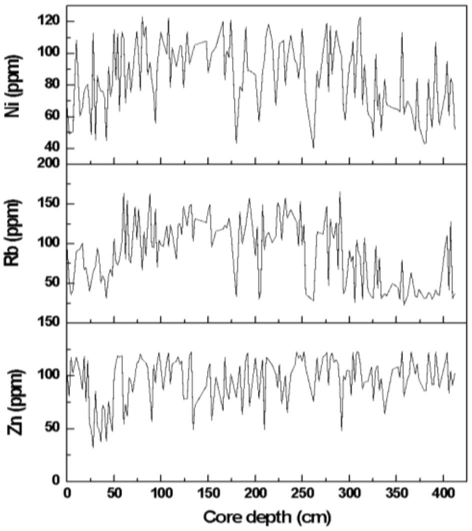

Figure 5: Down core variations of Zn, Rb and Ni

Ni exhibits positive correlations with Fe (r = 0.64), V (r = 0.63), Rb (r = 0.62), K (r = 0.52), Mn (r = 0.44), Co (r = 0.42), Ca (r = 0.40), Ti (r = 0.32), Zn (r = 0.25), Sr (r = 0.21), Cr (r = 0.10), Cu (r = 0.09), OC (r = 0.01), and negative correlations with $CaCO_3$ (r = -0.11), Sc (r = -0.04)in the core. Obtained strong positive correlations indicate about Ni segregation from the source of land derived fine grained sediments. A group of researchers reported presence of nickel higher concentration in the top layer of the core that belongs to the Bay of Bengal (Sarin et al., 1979). It may be as a consequence of post glacial sequence in central Indian Ocean (Borole, 1993). The average concentration of Ni in the present study 85.6 ppm is higher than the values reported earlier for sediments of Bay of Bengal (64 ppm) (Sarin, 1979).

It has good agreement with Nizampatnam Bay (84 ppm), Godavari delta (82 ppm) and middle Bengal Fan (75 ppm) that reported and Gulf of Tunis (Venkateswara, 2008; Noureddine et al., 2014). Copper (Cu) element entry into the marine environment takes place in the form of solution having association with fine-grained solid of colloidal inorganic and organic materials. Presence of copper occurs in igneous ferromagnesian minerals, pelagic sediments and clay minerals. The concentration of copper in the present core ranges between 126.1 to 33.0 ppm with an average value 75.19 ppm as presented in the Figure 6. Lower values are observed in the upper (0 – 52 cm) as well as in the bottom (330 – 412 cm) parts, while higher values found to be presented in the regions of core depth 52 – 84 cm and 272 – 316 cm. For the remaining depths, the concentration of copper is varied with small fluctuations. A number of earlier studies on different kinds of sediments have reported that non-residual fractions of copper are mainly associated with the oxidizable phase, occurring as organically complexed metal species (Pardo, 1993; Li, 2007). This is due to property of copper high affinity with humic substances, which a fraction of natural organic matter chemically active in such complexion metals (Pempkowiak, 1999). In the present study the non-residual fractions of copper show a different pattern, perhaps owing to the influence of early diagenetic processes. A significant fraction of the copper reaching the sediment surface may be returned to the overlying water column by mineralization of the host organic materials at the surface and in the oxic upper layers of the sediments (Petersen, 1995; Xuelu, 2010). Earlier similar to Ni; copper also found to associate with Fe_2O_3; P_2O_5; MnO in its fine grained range (<2µm) and attained its enrichment on surface sediments of the Gulf of Tunis having anthropogenic inputs (Noureddine et al., 2014). Copper exhibits positive correlation with V (r = 0.51), Rb (r = 0.47), Fe (r = 0.44), Co (r = 0.43), Ca (r = 0.33), Mn (r = 0.31), K (r = 0.29), Ti (r = 0.29), Zn (r = 0.19), Sc (r = 0.15) and Ni (r = 0.09) indicating its correlation with detritus minerals that associated with organic matter.

Observed positive correlation with Fe and Mn indicating its adsorption on clay mineral surfaces and also in redox mobilization processes (Wedephol, 1964). Earlier Krauskopf found positive correlation between Cu and Fe, it ascribed that a fraction of copper was foraged from seawater by hydrous ferric oxide precipitate in marine environment (Krauskopf, 1956). Sawam and Murray observed the mutual relationship between P and Cu; which might have originated from biogenic activity (Sawam and Murray, 1983). The obtained average concentration of Cu in the present core (75.19 ppm) shows a good agreement with those values obtained for the eastern continental shelf of Bay of Bengal (75 ppm), Nizampatnam Bay (78 ppm), Bengal Fan (68 ppm) and less when compared with the sediments of western equatorial Indian Ocean (113 ppm), slope sediments of Godavari delta (100 ppm) (Murty, 1983; Mascarenhas, 1985; Venkateswara Rao, 2008).

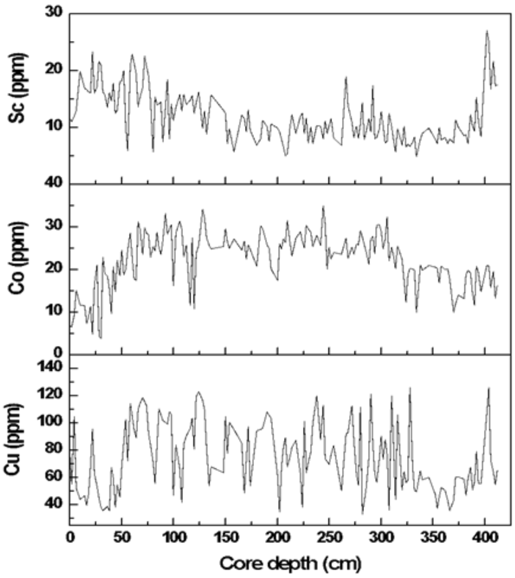

Figure 6: Down core variations of Cu, Co and Sc

Detrital clays and volcanic exhalations are the main contributions of cobalt in the deep-sea sediments. It presents as colloids, and transported in dissolved forms. Cobalt weathering products are carbonates; hydroxides; adsorbed and co-precipitated with Mn oxides or, to a less extent, Fe oxides. In the present study concentration of cobalt found to lie in the range 35 to 3.9 ppm, having an average value 22.2 ppm as shown in Figure 6. The maximum value is observed at 364 – 366 cm while lower values found in the upper part (0 – 54 cm) relative to the other parts of the core. In marine sediments, Co form in the areas of higher biological productivity driven by coastal upwelling. It is often depleted due to the remobilization of the sediment particles in oxygen depleted water and sedimentary environments. Co exhibits positive correlations with Fe (r = 0.50), Rb (r = 0.47), V (r = 0.47), K (r = 0.44), Cu (r = 0.43), Ni (r = 0.42), Cr (r = 0.30), Ti (r = 0.27), Mn (r = 0.21), Zn (r = 0.14), Ca (r= 0.14), OC (r = 0.25) and negative correlations with $CaCO_3$ (r = -0.34), Sc (r = -0.18).

Observed positive correlations indicate regarding cobalt origination from detrital elements and organic material. Chester accounted cobalt association with organic matter in marine sediments (Chester, 1965). The positive correlation between organic carbon and Co (r = 0.25) can be understood about part of the cobalt association with organic matter. Goldberg suggested that Co is scavenged from seawater by hydrous iron oxide (Goldberg, 1954). Cobalt was found with lower concentration on surface sediments of the Gulf of Tunis (Noureddine et al. 2014). Hence in the present study also the positive correlation between Co and Fe, Mn may be understood on same lines as suggested, adsorption to the surface of the fine grains (Goldberg, 1954). The obtained average concentration of Co in the present core (22.2 ppm) shows agreement with the sediments of western continental shelf (22 ppm) and northern Arabian Sea (28 ppm) that reported (Murthy et al., 1983). Similar concentration in the sediment cores of the Nizampatnam Bay (25.4 ppm), middle Bengal Fan (25 ppm) and Godavari delta (27 ppm) are reported (Venkateswara, 2008). However, they are higher than the values reported for western equatorial Indian Ocean (13 ppm) indicating its relative enrichment in the study region sediments (Murthy 1983).

Scandium occurs in very small quantities. The blue color of the aquamarine variety of beryl is thought to be caused by scandium. The concentration of Scandium (Sc) in the present core variesin between 27.06 to 4.84 ppm with an average of 12.07 ppm as displayed in the Figure 6. From 160 to 250 cm and 320 –380 cm it showed lower values while higher values are observed between 0 to 150 cm due to the sediment water interface, at depths corresponds to the oxygen penetration zone. The oxic environment favors the precipitation of Mn and Fe oxides. The Scandium enrichment at the top of the cores probably reflect the association of these elements with Mn and Fe oxy-hydroxides as these are derived from land detrital fraction. The lower values are observed after the depth of 150 cm as like $CaCO_3$. It also proved by obtaining positive correlation between these two (r = 0.65) indicating that these are originating from weathering products. Scandium is positively correlated with Ca (r = 0.84), Mn (r = 0.65), V (r = 0.43), Fe (r = 0.37), Sr (r = 0.33), K (r = 0.25), Ti (r = 0.11) and Cr (r = 0.10) indicating Sc association with coarse grained sediments.

5. CONCLUSION

Concentrations of major and minor elements have been determined along the length of the core sediment by using PIXE technique. The enrichment of elements exhibiting the order of Fe > Ca > K > Ti > Mn along with the order of trace elements Cr> Sr> V > Zn > Rb > Ni> Co > Sc was obtained. Most of the elements showed their enrichment during Holocene period and lower in the Pleistocene period. Upper (0 – 150 cm) and lower (150 - 412 cm) parts of the core sediment belong to Holocene and Pleistocene periods respectively. Holocene enrichment and Pleistocene depletion seem to be directly related to primary productivity changes relating to surface water of ocean.

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