Major and Trace element concentration of Late Quaternary sediments of South Eastern Arabian Sea

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ABSTRACT: *In order to understand the paleo-environmental and paleo-climatic changes in the South-Eastern Arabian Sea during the late Quaternary period, the present study was taken up. To achieve the objective, major and trace element concentration along with total organic carbon percentage of top 4 m of the sediment core PC-01/SR 004 recovered from the continental slope region off Cochin in the South-Eastern Arabian Sea was studied in detail, The main findings of this work indicates that the core site has recorded higher productivity with increased fluvial input during the warm, humid interglacial periods and interstadials than the stadials and glacial period. Two warmer and one colder period define the MIS 3 phase of the core site. The high concentration of Si, Al, Fe, and Ti indicates a high terrigenous influx between 340 and 300 cm and 150 and 100 cm. Comparatively low concentrations of these elements are found between these two lengths, or between 300 and 150 cm of the core length. The geochemical proxies show two distinct phases in MIS 2: (1) a high concentration of terrigenous elements such as Si, Al, Fe, Ti, and K from 22.3 to about 17 ka, and (2) a progressive decrease in terrigenous element concentration from 17 to 11.7 ka. CaCO₃, Sr, and TOC concentrations reciprocate these values in the corresponding manner. The temperature gradient developed during the Holocene due to the nearly full melting of ice sheets intensified southwest monsoonal wind brought moisture to the Indian subcontinent, resulting in high precipitation. Through either upwelling or deepening of the mixed layer, this powerful wind adds nutrients to the Arabian Sea's surface mixed layer. This is demonstrated by a matching rise in the TOC and calcium carbonate content, and the early Holocene increase in these elements is indicative of the high terrestrial input to the core site.*

## I. INTRODUCTION

Changing chemical weathering patterns and provenance during the Cenozoic over various timescales has been reconstructed by several workers through analysis of elemental ratios or isotopic systems of the Arabian Sea sediments [1-5]. Studying the major element geochemistry of sediments is crucial because it reveals details about the origin of sediments and the mechanisms involved in sedimentation. Since the majority of chemical changes occur at or around the sediment/water interface, it is crucial to investigate the geochemical makeup of bulk sediments [6]. After deposition, chemical processes may have taken place that alters the bulk chemical composition. Following their release into the water, elements quickly attach themselves to particles and become part of bottom sediments. Sediments in marine aquatic systems can both carry and potentially be a source of several elements, especially metals. Trace elements are essential, particularly in the oxygen minimum zone (OMZ), which has a significant effect on ecosystems in terms of adding greenhouse gases (GHGs) to the atmosphere during the decomposition of organic matter [7]. The present paper deals with the influence of

climate in major and trace element concentration in south eastern Arabian sea. Bulk sediment geochemical data was utilized in deciphering the sedimentation history of the area.

II.   GEOLOGICAL SETTING

The study area is located in the mainland slope region between Lakshadweep Island and the southwestern part of the west coast of India (Fig 1).
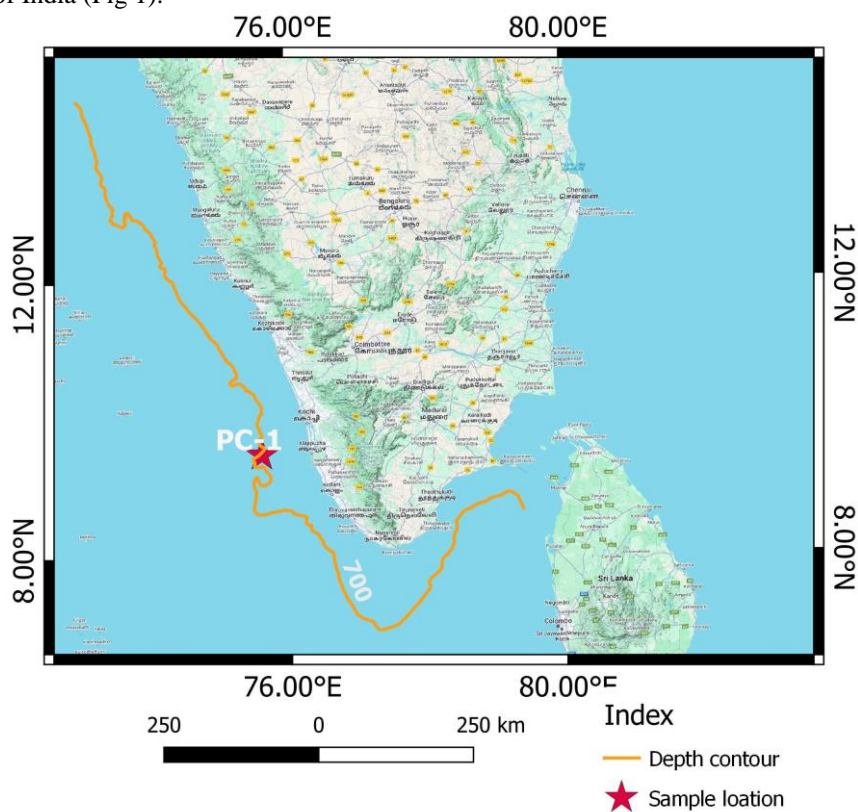
Figure1: Location of the core SR-004/PC-001, off Kochi.

The region has a tropical climate with an annual rainfall of 1500 mm. The continental shelf width ranges from 70 to 78 km, with water depths ranging from 1650 to 95 m. The slope is westward, with the eastern part having shallower terrain and steeper water depths. The southwestern part is shallower and features a channel feature. The shelf edge is straight and sharp in the northern part, while less defined in the southern part. Bathymetric contours indicate steep slopes.

III.   MATERIALS AND METHODS
3.1 MAJOR OXIDES AND TRACE ELEMENTS

The sediments analyzed in this study is from the top 4 m of a 28.7 m-long piston core that was taken off Kochi, India, during the cruise SR-004 by the Geological Survey of India's state-of-the art vessel Samudra Ratnakar. The water depth was 635 m, which is situated in the Arabian Sea's current oxygen minimum zone, which is between 150 and 1200 m. On-board, the core was inspected, and the physical characteristics were recorded. The top four meters of the core, which was kept in the core lab at M & CSD, Mangalore, was subsampled at interval of one centimeter for this study. Salt content was removed from the subsamples, dried and powdered in agate mortar and 0.1 g of the sample was transferred to a dry polypropylene bottle and 5 ml of aqua regia was added very slowly followed by addition of 5 ml of hydrofluoric acid; the bottle was immediately closed firmly, swirled intermittently and kept overnight. 50 ml of saturated boric acid solution was then added, and the solution was kept for an hour. It was then transferred into 100 ml volumetric flask (plastic) and made up with distilled water. Solutions of standard samples and sample blank were prepared in the same way. For trace metal analysis 0.2 g sample was transferred into a dry PTFE beaker and 10 ml of aqua regia was added very slowly followed by addition of 5 ml of hydrofluoric acid. The beaker was heated gently to dryness on a hot plate, after cooling 5 ml of hydrofluoric acid was added further and again heated to dryness. It was followed by

addition of perchloric acid (2 ml) and heating again to dryness. 5 ml of nitric acid (1:1 V/V) was then added and warmed; the solution was then transferred to a graduated tube and made up to 20 ml. AAS was calibrated for the individual elements by using standard solutions. Absorbance was measured for known concentration of the element. The sample solution was then aspirated into AAS (VARIAN make, AA240FS) to measure the absorbance. From the absorbance, concentration of the element was calculated. For analysing $CaCO_3$ & $MgCO_3$, 0.1 g sample were treated with acetic acid (4% V/V) and kept overnight. The solution was used for the estimation of Ca and Mg by flame AAS. Standard solutions and sample blank were prepared. AAS instrument was calibrated for the individual elements using standard solutions. The sample solutions were then aspirated into AAS to measure the absorbance.

3.2 TOTAL CARBON AND INORGANIC CARBON

TOC refers to a total organic carbon analyzer, which utilizes a catalytic oxidation combustion technique at high temperature (the temperature raises up to 900°C), to convert carbon present (organic carbon and inorganic carbon) into CO_2. The CO_2 generated by oxidation is measured with a non-dispersive infra-red sensor. Here known amount of finely powdered sediment samples is heated to 900°C in furnace in presence of catalyst. At high temperature all the carbon present in sediment samples is converted into CO_2, it is quantified using the non-dispersive infra-red sensor. This gives total carbon content in the sediment sample. Here dextrose (AR grade) is used as standard to check the accuracy of the measurement.

Unlike the previous case, for finding out inorganic carbon, known quantity of the sediment sample is treated with required quantity of the phosphoric acid. The sample will be heated to 200°C in furnace. Due to reaction with acid all the carbonate will be dispersive infra-red sensor. This will give inorganic carbon (IC). Here anhydrous Na_2CO_3 (AR grade) is used for calibration and well as check standard to check the accuracy of the measurement.

$$\% total\ organic\ carbon = \%TC - \%IC$$

3.3 GEOCHRONOLOGY

Two samples from the core, from 99-100 cm and 199-200 cm ware dated by Accelerator Mass Spectrometry (AMS)[14]C method at Physical Research Laboratory, Ahmadabad, published in [8]. Top 100 cm of the core represents 31,433 years and at 200 cm the age of the sediments is 42,569 years. The $\delta^{18}O$ data of this core is correlated and matched with SPECMAP data [9,10] and the marine isotope stage (MIS) boundaries of the core have been demarcated and reported in [8]. According to this, MIS 2/MIS 3 boundary is identified at 81.5 cm core depth, based on large fluctuation of $\delta^{18}O$ values of the order of 1.3‰. Another Oscillation in $\delta^{18}O$ values observed at ~20 cm core depth, where fluctuation of $\delta^{18}O$ values of the order of 0.8‰ observed which marks the MIS1/MIS2 boundary. Variation in the geochemical data is described as per these boundaries. As per these data the core represents Quaternary Period and the down core variation of elements are described as MIS III, MIS II and Holocene.

IV.   RESULTS

Reversal of monsoonal winds in the Arabian sea plays a major role in seasonal variations in upwelling and related primary productivity, which has a large impact on the distribution of elements in the sediment. So a high resolution chemical analysis of sediment cores can provide valuable information regarding palaeoclimate changes [11].

4.1 MAJOR OXIDES

Information about the relative amounts of the three main components of marine sediment—diagenetic products, biogenous material, and terrigenous detritus—is provided by the major element composition. Terragenous material-related elements and element ratios also aid in determining provenance traits and, consequently, shifts in the climate and/or sediment supply.

A. SiO_2, Al_2O_3, Fe_2O_3, TiO_2 AND K_2O

The down-core variations of SiO_2, Al_2O_3, Fe_2O_3, TiO_2, and K_2O exhibit a consistent pattern throughout the core (Fig. 2).
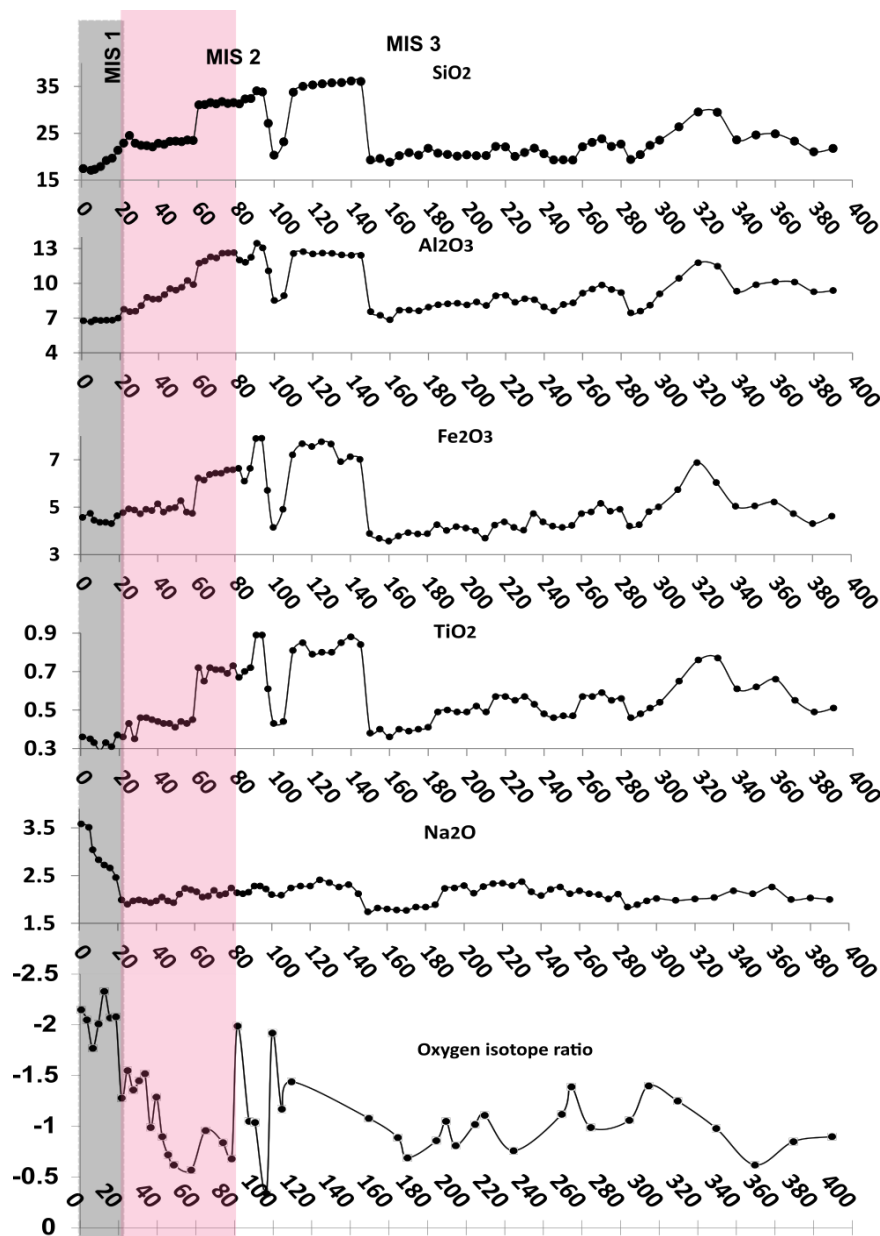
Figure2: Major element concentration and $\delta^{18}O$ of the core SR-004/PC-001, off Kochi.

These components signify the terrestrial contribution to the sediments. The concentration of these elements in the core is mostly influenced by past sediment supply. The concentration of SiO_2 ranges from 17.06% to 36.17%, with a mean of 24.57%. The highest concentration of silica is seen in MIS-III at a depth of 139-140 cm, while the lowest concentration is found in the Late Holocene at a depth of 3-4 cm. In MIS 3, the SiO_2 value fluctuates from 21.75% to 24.86% between depths of 390 and 340 cm. Subsequently, at a depth of 330 cm, there is a rapid rise in the value from 23.59% to 29.47%, which persists until a length of 310 cm. The readings
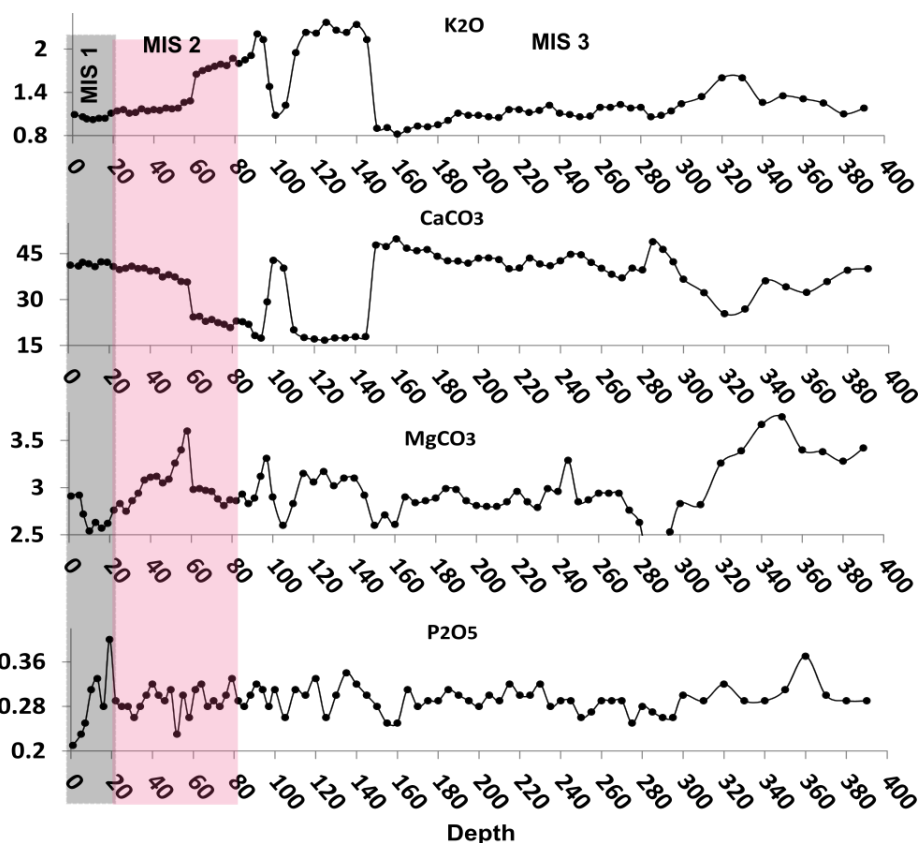
Figure2: Major element concentration of the core SR-004/PC-001, off Kochi (continued).

fluctuate between 18% and 23% from 310 cm to 150 cm. At a depth of 145 cm, there is a significant increase in the value from 19.28% to 36.17%, followed by a slow decline to 33.78% at a depth of 110 cm. The concentration decreases from 23% to 27% when the measurement transitions from 105 cm to 97 cm. A rapid rise in value to 33.83% occurs at 93 cm, fluctuating between 34% and 31% up to 60 cm. The SPECMAP oxygen isotope curve establishes the MIS2/MIS3 boundary at a core depth of 81.5 cm, determined by significant fluctuations in $\delta^{18}O$ values, around 1.3‰. The interval from 82 to 20 cm of the core delineates the MIS 2 phase. In MIS 2, the SiO_2 levels range from 31% with slight changes at depths of 80 to 60 cm, whereas at depths of 60 to 21 cm, the values fluctuate between 23% and 22%. A sharp decline in SiO_2 concentration occurs beyond 60 cm, with no significant fluctuation in values at the beginning of the Holocene. A steady decline from 19.65% to 17.42% is seen. The decreased Al levels in the sediments of the eastern Arabian Sea may result from dilution with lithic components that possess low Al concentration. The alumina concentration ranges from 6.7% to 13.5%, with a mean of 9.5%. The highest concentration occurs during the MIS 3 phase at a depth of 90-91 cm, while the lowest concentration is seen at 3-4 cm in the Holocene. The maximum and lowest concentrations of Fe_2O_3 are seen in MIS 3 at depths of 93-94 cm and 159-160 cm, respectively with an average concentration of 5.16%. The minimum concentration of TiO_2 is 0.29% in the Holocene at 9-10 cm, while the greatest concentration is 0.89% at 93-94 cm during the MIS 3 phase. The maximum and lowest concentrations of K_2O are 2.34% at 124-125 cm and 0.82% at 159-160 cm during the MIS 3 phase, respectively. The mean concentration of K_2O in the core is 1.35%.Upon examining the concentrations of SiO_2, Al_2O_3, Fe_2O_3, and TiO_2, a total of three sedimentation cycles may be seen in MIS 3 phase. The computed average concentration of Al is lower than that of normal shale (8.84%), suggesting that the sediments are diluted with a phase, most likely quartz, which lacks Al and trace elements.

    B. NA_2O

Na_2O content in igneous rocks is largely controlled by the abundance of Na-rich feldspars, clinopyroxenes or clinoamphiboles. Clastic sediments usually show low Na_2O contents, because Na is completely dissolved during weathering of the source rock and dissolved ions are concentrated in the river runoff to the ocean and its salt content. Deep sea shales reveal higher Na_2O levels because salt-derived Na is enclosed in the pore water of the sediments [12]. Maximum concentration of Na_2O is 3.58% in the top sample (Holocene) and minimum

concentration is 1.74 at 150 cm in MIS 3 with an average of 2.18%. Na_2O concentration is not showing much variation upto the beginning of Holocene. From the boundary of Holocene it is gradually increasing and the maximum concentration is at the top most sample.

C. $CaCO_3$

Calcium carbonate concentration ranges from 16.71 to 49.75%. The average concentration of calcium carbonate is 35.55%. Upto 60 cm depth from the top the $CaCO_3$ content in the core ranges from 35.6 to 42.2% and there is a gradual decrease in the concentration. From 390 to 340 cm $CaCO_3$ concentration fluctuates between 32 and 40%, but at 330 there is a sudden decrease in concentration from 36 to 26% and the lower concentration continues upto a length of 310 cm. From 310 cm to 150 cm the concentration remains in the higher side as it fluctuates between 36 and 47% but after that there is a sudden decrease at 145 cm to 17.89%. Upto a length of 110 cm it fluctuates between 17.89 and 20% and again at 105 cm length there is a sudden increase in the concentration to 40.22%. But after 10 cm there is again a sudden decrease in the concentration and the lesser concentration continues upto a length of 60 cm.

According to the $\delta^{18}O$ record of *G. ruber* in the core the MIS 2/MIS 1 boundary is marked at 81.5 cm. In MIS 2 from 22.3 to 18.5 ka (81 to 60 cm length) the concentration of $CaCO_3$ is low (20.81 to 24.48%). From 18 ka to 11.7 ka (at about 21.5 cm) the $CaCO_3$ concentration is gradually increasing to the recent and at MIS2/MIS1 boundary there is no change in the general trend.

D. $MgCO_3$

The down core variation of $MgCO_3$ shows more fluctuations than the other major oxides though the general trend is same. Minimum concentration of $MgCO_3$ is 2.12% at 284 to 285 cm and the maximum 3.75 is at 349-350 cm. Average concentration of $MgCO_3$ is 2.95%. In MIS 3 phase from 390 to 320 cm length the concentration fluctuates between 3.48 and 3.75%, whereas from 310 to 140 cm length it is ranging from 2.12 to 2.99% except at 145 cm where the concentration is 3.29%. From 140 to 115 cm length the concentration is high, i.e., above 3%, and next 10 cm the values are low i.e., 2.6 to 2.9%. At 97 to 93 cm length the concentration is high and after that from 90 to 60 cm it is again less. In MIS 2, from 22.3 to 18.5 ka the concentration of $MgCO_3$ is less and after 60 there is sudden increase from 2.98 to 3.6%, then it is decreasing with fluctuations.

E. P_2O_5

Distribution of P_2O_5 is entirely different from all other major elements in the core. Minimum concentration of P_2O_5, 0.21% (P – 0.09%) is at the top sample and maximum concentration 0.4% (0.15%) is in the beginning of Holocene, at 18-19 cm depth. Thus the concentration is falling within the limit of marine sediments. In MIS 3 P_2O_5 concentration varies between 0.25 and 0.37% and at the beginning of Holocene (about 11.7 ka) there is a sudden increase in the value from 0.29 to 0.4%. In Holocene generally the concentration shows a decreasing trend.
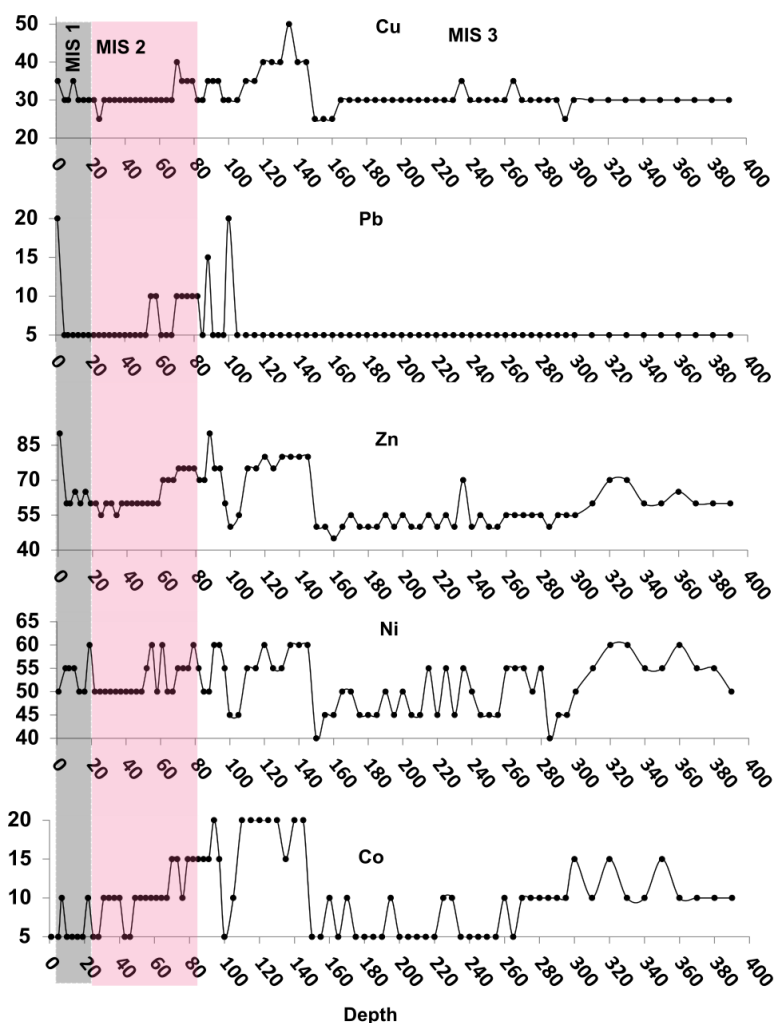
4.2 TRACE ELEMENTS
The chemical composition of marine sediments is controlled by the relative contributions of particulate materials derived from different sources having variable compositions. Trace elements are initially supplied to the site of deposition from terrestrial sources via rivers or atmosphere, and from biological productivity in the ocean. Usually degree of enrichment or depletion of the trace element in the sample is evaluated for paleo-environmental reconstruction [13-15]. The distribution and vertical flux of most of the trace elements are controlled by the production, sinking and deposition of the biogenic particles [16]. The monsoon winds transporting dust are found to be responsible for the spatial and seasonal variations in fluxes of particles in the Arabian Sea [17,18]. Seasonally varying particle flux patterns make the southeastern Arabian Sea an ideal site to study the trace metal fluxes and their implications to the paleoclimate.
Cu concentration is not showing much variation and the maximum concentration of Cu is 50 ppm at 134-135 cm and minimum is 25 ppm. Only one sample is showing value above average shale value (45 ppm). Concentration of lead is also not showing much variation. The values range from >5 to 20 ppm, which is at the top of the core and at 99-100 cm depth. After 100 cm depth it is less than 5 ppm. As the behavior of Nickel (Ni) is similar to that of organic carbon, Ni/Al ratio can be related to the nutrient cycling and reflects the accumulation of organic matter, i.e., productivity. Concentration of Ni is not showing much variation it is varying between 40 and 60 ppm. All the samples are showing values below average shale value (68 ppm). Cobalt values vary from >5 to 20 ppm and all samples Cd concentration is either 5 or >5. Chromium content in

the core varies from 90 to 145 ppm, maximum concentration is at 134-135 cm and minimum concentration is at 21-22 and 149-150 cm depth.
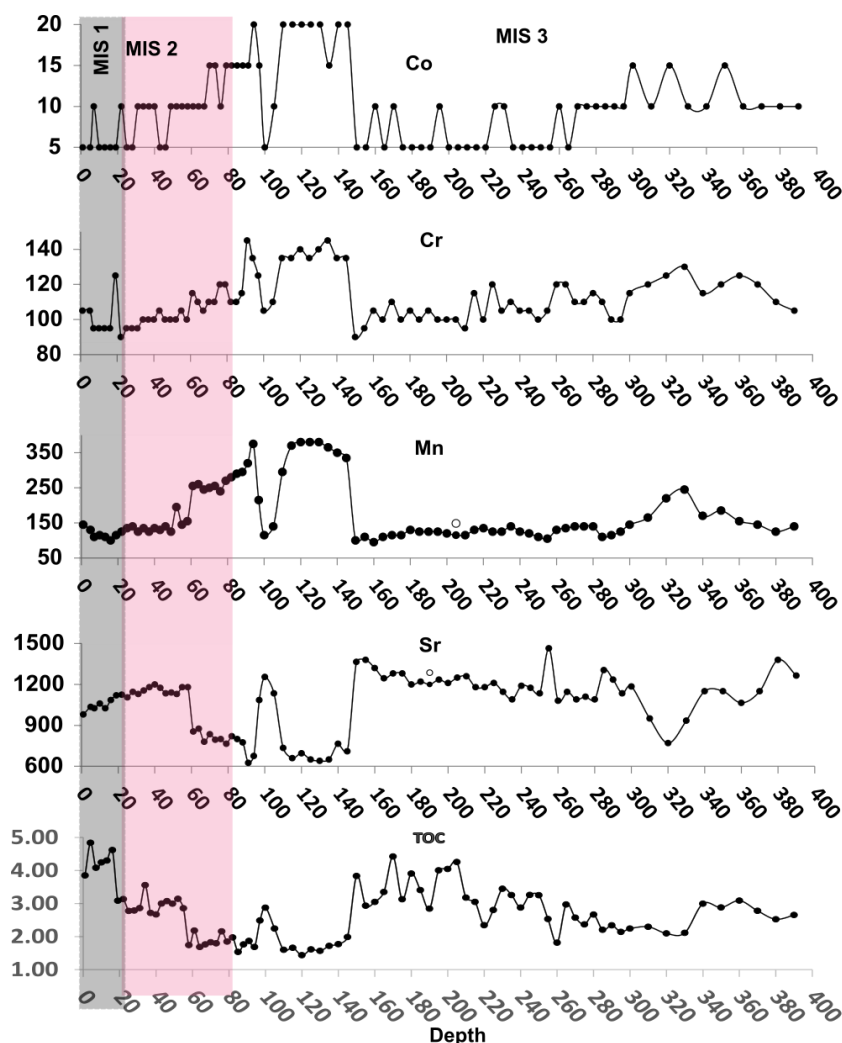

Figure3: Trace element TOC concentration of the core SR-004/PC-001, off Kochi

In MIS 3 phase, from 400 cm to 160 cm concentration of Cr is above 100 ppm and from 155 to 150 cm depth it is less than 100 ppm. At 145 cm there is a sudden increase in the concentration from 90 to 135 ppm and this high concentration remains up to 110 cm and from 105 to 100 cm the values are 110 and 105 ppm again at 97 cm it is suddenly increasing to 125 ppm and this trend of higher values are continuing for the next 7 cm. From the beginning of Pliestocene (approximately 22 ka) Cr values are above 100 ppm except in the Late Pliestocene the concentration is 95 ppm. In early to mid Pliestocene it is fluctuating slightly and before the MIS2/MIS3 boundary 90-91 cm depth there is a sudden increase in the value from 115 to 145 ppm. Manganese is showing the same pattern of Cr. Minimum concentration (95 ppm) is at 159-160 cm and maximum concentration (380 ppm) is at 129-130 cm. Sr concentration is very high in this core. It varies between 625 ppm at 90-91 cm and 1465 ppm at 254-255 ppm. All samples show values higher than that of average shale (300 ppm) (Fig. 3).

4.3 ELEMENTAL RATIOS
Reconstructing the paleoclimate during the Quaternary depends on both lithogenous input and productivity. When evaluating compositional variance, normalizing elemental concentration to Al is useful since the amount of Al in the sediment record typically indicates the concentration of terrigenous aluminosilicate detritus. [19-21]. Therefore, Si, Ti, K, Mg and Fe of the core is calculated as ratios to Al. Si/Al ratio ranges from 2 to 2.86 and, the average is 2.29 which is lower than the world average shale value of 3.31. The Si/Al shows low values towards the bottom of the core in

Figure3: Trace element and TOC concentration of the core SR-004/PC-001, off Kochi (continued).

MIS 3 phase, high values are noticed at a length of 295 to 285 cm, 245 to 230 cm and from 180 to 105 cm in MIS 3 phase. From 88 to 79 cm again the values are high and it decreases from the beginning of Pliestocene (81 cm). In Late Pliestocene it is increasing gradually and it reaches the maximum value of 2.86 just before the beginning of present interglacial—Holocene. In the Holocene the ratio is showing a decreasing trend to the top of the core.

Ti/Al ranges from 0.05 to 0.08 with an average of 0.06. Ti/Al ratio is higher than the average shale value, 0.053. Ti is associated with certain minerals found in the coarse grained sediments, so the concentration of Ti may have a relation with the grain size of the sediments. Slightly higher average value of Ti/Al may be due to the distinctly different lithological units on the bordering continents. The lowest value is noticed in the beginning of Holocene and in Holocene it is varying between 0.05 and 0.06. Fe/Al ranges from 0.6 (209 to 210) to 0.94 (4 to 5) with average of 0.72, which greater than the average shale value of 0.55. Na/Al ranges from 0.12 (72 to 76) to 0.39 (0 to 1) with an average of 0.18. K/Al ranges from 0.18 (164 to 165) to 0.30 (139 to 140) with average of 0.22. Mg/Al ranges from 0.12 to 0.34 with an average of 0.17. It is showing an increasing trend from LGM to most recent sediments. Average value of Mg/Al is slightly lower than that of world average shale (Fig 4).

4.4 Organic carbon

Information on organic carbon helps to understand biological productivity in the aquatic regions as organic carbon in marine sediments is primarily derived from settling biogenic debris from the water column and their abundances serve as indicators of primary productivity [22]. Oceanic CO_2 concentration is supposed to be in equilibrium with the CO_2 concentration of the atmosphere, so for understanding the global carbon cycle it is important to know how much carbon is buried in marine sediments and how much has been withdrawn from the carbon cycle.
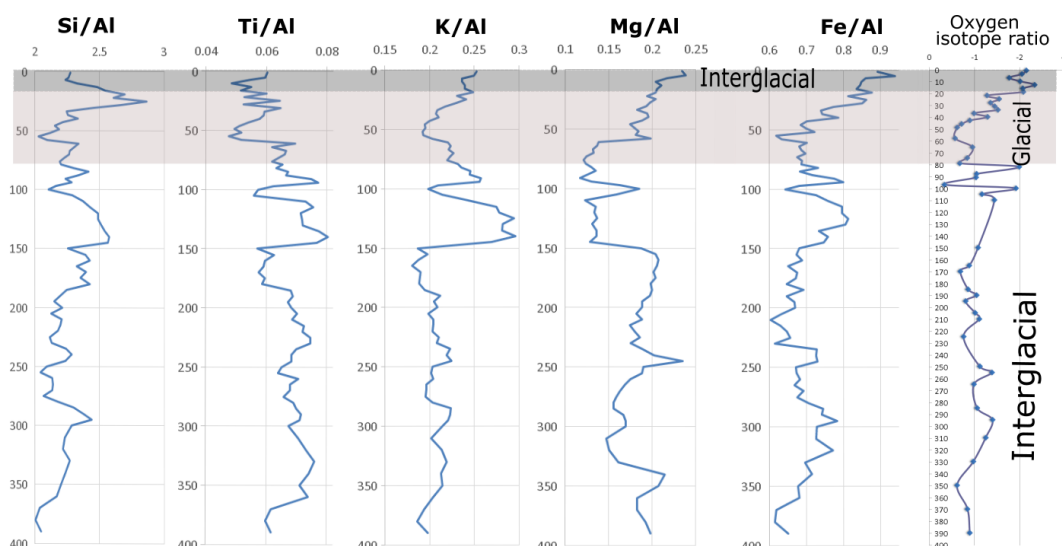
Figure4: Ratios of major elements to Al of the core SR-004/PC-001, off Kochi.

V.   DISCUSSION AND CONCLUSION

In the present study the down core variation of total organic carbon (TOC) follows the pattern same as that of $CaCO_3$. Maximum concentration of TOC is 4.84 at 3 to 4 cm depth and minimum concentration is 1.44 at 190 to 120 cm depth. Monsoonal winds are reversing in the Arabian Sea; according to serveral workers, the summer monsoon is stronger during interglacial periods than during glacial ones [23,24]. In early deglacial period [25] and LGM [26] winter monsoon was stronger than the present. So glacial period is characterized by low terrigenous input and interglacial period icharacterized by a high terrigenous input here. Warmer interstadials correlate with higher siliciclastic concentrations than colder stadials.

Abundances of major elements Si, Al, Fe and Ti in the core are employed to investigate the changes in supply and source of detrital material to the eastern Arabian sea during the Late Quatenary. Al and Ti concentrations in marine sediments are the best indicators of lithogenous material flux to the marine environment [9]. Both these elements may have been derived from the extensive laterite deposits present in the hinterland of southwestern India and are supplied to the southeastern Arabian Sea. High Al and Ti values in the early bv Holocene lends support to the argument suggesting greater terrigenous dilution at this core site during this period.

MIS 3 phase is characterized by two warmer and one colder period. From 340 cm to 300 cm and 150 to 100 cm are characterized by a high terrigenous input as indicated by the high concentration of Si, Al, Fe and Ti. In between these two lengths, i.e., from 300 to 150 cm of the core length is characterized by comparatively low concentration of these elements. Element ratios like Ti/Al, Mg/Al, which are not influenced by productivity or redox conditions, follow a similar pattern like Ti- and Mg-element concentrations. This suggests that the millennial-scale pattern in the siliciclastic fraction represents a shift in input, source, or transportation circumstances rather than merely being the consequence of dilution due to biogenic carbonate input or preservation. High CIA values from 340 to 300 cm length of the core also indicates a warmer period though it is not clearly identified from 150 to 100 cm length. In the trace elements the EF values of Cu, Zn, Ni and Cr are low from 340 to 300 cm and 150 to 100 cm length of the core (Bhadra et al, in preparation). These two periods are marked by low $CaCO_3$ concentration, low EF for Sr and low TOC values. All these evidences indicate an increased terrigenous input and a reduced organic production during these periods.

Based on the $\delta^{18}O$ record of G. ruber in the core boundary between MIS 2 and MIS 3 is identified at 81 cm and in MIS 2 period (from 22.3 to 11. 7 ka) two phases can be identified from the geochemical proxies, (1) from 22.3 to approximately 17 ka where the concentration of terrigenous elements like Si, Al, Fe, Ti and K is high and (2) from 17 to 11.7 in which a gradual reduction in the concentration of terrigenous elements is noticed. Concentration of $CaCO_3$, Sr and TOC are reciprocating these values in the accordingly.

As melting of ice sheets was almost complete during Holocene, the thermal gradient started developing between land and ocean resulting in enhanced southwest monsoonal wind which carries moisture towards the Indian subcontinent and results in heavy precipitation. This strong wind introduces nutrients in the surface

mixed layer of Arabian Sea either by the upwelling or by deepening of mixed layer [17]. This is reflected in corresponding increase in the TOC and calcium carbonate content and this ocean wind interaction is reflected in this core as marked by an increase in the TOC and calcium carbonate in the early Holocene.

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