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Coupling between suboxic condition and southwest monsoon intensification in the western Bay of Bengal sediment core: a geochemical study

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ABSTRACT

Reconstruction of paleo-redox conditions in a radiocarbon (^{14}C) dated sediment core (SK-218/1), covering the past 45 ka (thousand calendar years), collected from the western Bay of Bengal (Lat: 14° 02`N; Long: 82° 00`E) at a water depth of 3307 m, has been made based on redox-sensitive elements geochemistry. The high U/Th ratio, Mo enrichment, Mo/U enrichment factor ratio, Ce/Ce* < 1 and lower Mn/Al and Fe/Al ratio, compared to upper continental crust, are all indicative of prevalence of suboxic condition in the benthic environment from 15.2 ka to 4.5 ka, peaking around 9.5 ka. The suboxic condition around 9.5 ka corresponds to the previously recorded southwest (SW) monsoon intensification in response to increase in northern hemisphere summer insulation. However, productivity proxies – organic carbon and nitrogen contents – do not indicate marked increase in productivity at this time. It is proposed that as a result of large increase in lithogenic material supplied from land due to SW monsoon intensification, which is evident by the very high concentration of Al, Zr and Hf, the flux of fresh labile organic matter probably forming dense mineral matter-biogenic aggregates which sinks rapidly to the seafloor and the degradation of labile organic matter might have led to the development of suboxic condition in the benthic environment. There exist a strong positive correlation (r=0.98) between Mo and Zr during 15.2 ka to 4.5 ka suggesting a coupling between suboxic condition and lithogenic flux supply by the intensified SW monsoon. Our results suggest that temporal variability of the ballasting effect of the terrestrially-derived material could play a key role in benthic biogeochemistry and ecology of the Bay of Bengal.

We also provide the first record of the nitrogen isotopic composition ($\delta^{15}N$) of sedimentary organic matter in the western Bay of Bengal, a region where the mesopelagic oxygen minimum zone

(OMZ) is just short of being suboxic (denitrifying) today. The sedimentary $\delta^{15}N$ fluctuated considerably in the past, especially during the Marine Isotope Stage 3. Oscillations in $\delta^{15}N$ were apparently in concert with those in organic carbon and nitrogen contents and could be related to climatic changes (Heinrich and Dansgaard-Oeschger events) in the North Atlantic. The Dansgaard-Oeschger 12 event appears to have exerted the most intense effect on Bay of Bengal biogeochemistry when surface productivity, as inferred from the organic carbon and nitrogen contents, was the highest recorded in the core, and the $\delta^{15}N$ reached up to 6.3‰. Considering the probable dilution by isotopically light terrigenous organic matter, it would appear that OMZ of the Bay of Bengal had turned denitrifying. However, the absence of suboxic conditions in the sediments at this time suggests a decoupling of the benthic processes with those in the mesopelagic water column.

***Keywords*:** Bay of Bengal, Paleoceanography, Redox-sensitive elements, suboxic, Organic carbon, Stable isotopes, Monsoon intensification, Productivity, Denitrification.

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1. Introduction

The Bay of Bengal (BoB) receives a large quantity of suspended particulate matter (2.1×10^{12} kg a^{-1}) and fresh water discharge (1887 km^3 a^{-1}) from the Himalayas and Indian peninsula by major rivers like Ganges–Brahmaputra, Mahanadi, Krishna-Godavari, Cauvery, Irrawadi-Salween (Bird et al., 2008). The northern part of BoB receives maximum particle flux during the SW monsoon that coincides with discharge maxima of the Ganges-Brahmaputra Rivers (Ittekkot et al., 1991, 1992). This discharge reduces the surface water salinity by 7‰ (La Violette, 1967), and also supplies nutrients which may enhance primary productivity (Ramaswamy and Nair, 1994) and cause strong stratification during the SW monsoon (Ostlund et al., 1980). The waters of the BoB have very low (<5 µM) dissolved oxygen content at intermediate depths (200-800 m) resulting a pronounced oxygen minimum zone (OMZ) (Wyrtki, 1971; Rao et al., 1994; Sardessai et al., 2007). A model study revealed that oxygen levels within the OMZ are controlled by a combination of physical and biological processes (Sarma, 2002). Singh et. al., (2011) measured concentrations of dissolved U. Mo and Re in the water column of BoB and found very little variations in these redox-sensitive elements in within the OMZ indicating their conservative behaviour, i.e. these elements co-vary with salinity and are not affected by the large quantity of organic-rich suspended load brought by the rivers. The sediments in the Krishna-Godavari basin supplied by the Krishna-Godavari river systems in the western BoB have smectite as the dominant clay mineral (Subramanaian 1987; Kolla and Rao, 1990) and have silty clay texture (Pattan et al., 2008). Rashid et al., (2007) suggested that the SW monsoon was stronger in the BoB during the Bolling/Allerod event, early Holocene and weaker during the Younger Dryas, and deglacial warming that began around 19 ka was contemporary with deglaciation warming in Antarctica, indicating a strong connection through Indonesian Throughflow or Subantarctic mode water (Naidu and Govil, 2010). Sediments in the western BoB were mainly derived from the Himalayas during the last glacial maximum (LGM) (Tripathy et al., 2011) and there occurred a change in the sedimentary depositional environment around 12 ka (Kessarkar et al., 2005., Prakash Babu et al., 2010). Galy et al. (2007) showed that the amount of terrestrial organic carbon deposited in the Bengal Basin is 10 to 20% of the total terrestrial organic carbon buried in the oceanic sediments and removal of carbon to the seafloor is mostly through continentally derived material (Ittekkot et al., 1991, 1992).

The past depositional environmental conditions and processes involved within marine sediments can be reconstructed by using multiple proxies such as bioturbation index, sediment laminations, trace and ultra-trace metal concentrations, and stable isotope abundances. Trace metals, particularly redox-sensitive elements, in the sediments have been used extensively to reconstruct redox conditions in the bottom water at the time of deposition (Brumsack, 1980; Wignall and Myres, 1988, Calvert and Pedersen, 1993; Dean et al., 1999; Tribovillard et al., 2006, 2008; Pattan and Pearce 2009; Algeo and Tribovillard, 2009; Algeo et al., 2012and reference therein). In the marine environment, the depositional conditions can be oxic, suboxic/dysoxic and anoxic/euxinic depending upon the presence or absence of oxygen and H_2S content (Savrda and Bottjer, 1991; Wignall, 1994). Algeo and Ingall (2007) showed that redox conditions can be highly variable over short time scales and consequently the effects on redox-sensitive trace metals can be rather complex and such effects on the trace metal uptake by sediments are still incompletely understood. As BoB receives large quantity of suspended particulate matter and fresh water discharge mainly from Himalayas and Indian peninsular rivers which supply nutrients and also reduce the surface water salinity. Therefore, it is necessary to understand the implications of these parameters on behaviour and distribution of redox-sensitive elements with time. Till now no sediment core is studied for redox-sensitive elements such as U, Mo, Ce, Mn and Fe in BoB. Therefore, in the present study, we make the first attempt to reconstruct the past depositional environmental conditions in the BoB based on redox-sensitive elements geochemistry using a radiocarbon (^{14}C) dated sediment core that provides insights into factors responsible for the formation of reducing conditions during the past 45 ka.

2. Material and Methods

A 8.2 m long sediment core (SK-218/1) was raised from a water depth of 3307 m (Lat: 14˚ 02' 01"N ; Long: 82˚ 00' 12"E) off Krishna-Godavari basin, western Bay of Bengal (BoB), onboard *ORV Sagar Kanya* cruise (SK-218) during 18th March to 16th April, 2005 (Fig. 1). The core site is well below the present day OMZ water depth (Wyrtki et al., 1971; Olson et al., 1993; Rao et al., 1994). A total of 100 sub-samples from the sediment core were analysed in the present study. Sediment sub-samples were made salt free, dried and powdered. For the chemical analysis, about 50 mg of sediment was weighed in a Teflon beaker. To this 10 ml of acid mixture of HF + HNO_3+ $HClO_4$ (7:3:1 ratio) was added and the contents were concentrated to a paste by placing the beaker on a hot plate. To this paste 4 ml of 1:1 HNO_3 was added. After heating for 5 minutes the material was diluted with ultra pure water (18.2 mega ohm) to a final volume of 100 ml. Similar digestion procedure was followed for standard reference materials (MAG-1, SGR-1) and blanks. These solutions were analysed for a few major, trace and rare earth elements on an Inductively Coupled Plasma-Mass Spectrometer (*Thermo X Series 2*). The accuracy compared to the standard reference material and precision of the data based on duplicate analysis were better than ± 6%. Calcium carbonate content was measured on Coulometrics (UIC, INC-CM5130 Acidification Module). Pure calcium carbonate chemical was used as a standard reference material. Analysis of duplicate samples and standard reference material the precision and accuracy of the analysis was ± 2%.

Aliquots of sediment sub-samples were made carbonate free by treating with 0.1 N HCl. For the analyses of total organic carbon (TOC), total nitrogen (TN) and their isotopes ($\delta^{13}C$ and $\delta^{15}N$), carbonate free sediments (residue) were weighed in tin cups and combusted in a *Euro-Vector* Elemental Analyzer (EA) coupled with a *Delta V plus* stable isotope mass spectrometer (*Thermo*®) in a continuous flow mode. Working standard ε-Amino-n-Caproic Acid (ACA) supplied by Prof. Mark Altabet (SMAST, UMASS) whose reported values of $\delta^{15}N$ and $\delta^{13}C$ are 4.6‰ and −25.3‰ and internal sedimentary standard COD whose long term average values are 7.38‰ and -21.01‰, respectively, were used to check the precision of the instrument. Standard deviation for both $\delta^{15}N$ and $\delta^{13}C$ was less than 0.2‰.

3. Results

Uranium and Mo are the most useful redox sensitive trace elements used as proxies to understand the paleoredox condition in the marine environment. U and Mo concentration in the sediment core reached maximum of 9.4 ppm and 36 ppm at around 9.5ka respectively. Cerium is one of the unique members of rare earth element group due to its occurrence in two valence state (III & IV). Ce/Ce* also used as one of the promising parameter to trace paleo bottom water redox condition. Ce/Ce* vales are generally more than 1 in the entire sediment core (1.2) except from 10.5 ka to 7 ka where it reached < 1. The lowest Ce/Ce* value of 0.71 is recorded at 8.89ka. The terrigenous representing elements such as Al, Zr and Hf also have their highest concentration of 18%, 0.8% and 180 ppm at around 9.5 ka in the sediment core respectively. The productivity indicators such as total organic carbon and total nitrogen varied from 02. % - 2.85% and 0.3% - 0.22% respectively. However, their highest concentration is at 42 ka in the core. $\delta^{13}C$ and $\delta^{15}N$ values fluctuate between -20.26‰ to -13.4‰ and 2.44‰ to 6.31‰ in the sediment core respectively. Highest $\delta^{15}N$ value of 6.33 is recorded at 42 ka.

4. Discussion

4.1 Sedmentation rates

Based on eight ^{14}C dates the 8.2 m long sediment core (SK-218/1) covers the past 45,000 calendar years. The detailed age model for the core has shown in Fig. 2 (Naidu and Govil, 2010; Govil and Naidu, 2011). In the core last glacial maximum (LGM) is between 20 ka and 15 ka (Govil and Naidu, 2011). The sedimentation rate in the last 45 ka varied from 7.08 cm $(ka)^{-1}$ to 27.3 cm $(ka)^{-1}$. During the Marine Isotope Stage (MIS) 1, sediment accumulation rate varied from 15.1 cm $(ka)^{-1}$ to 27.3 cm $(ka)^{-1}$ with an average of 21 cm $(ka)^{-1}$; during MIS 2 the sedimentation rate ranged from 9.9 cm $(ka)^{-1}$ to 11.2 cm $(ka)^{-1}$ with an average of 10.5 cm $(ka)^{-1}$; and during MIS 3, it varied from 7 cm $(ka)^{-1}$ to 22 cm $(ka)^{-1}$ with an average of 17 cm $(ka)^{-1}$. These data suggest higher sedimentation rates during the interglacial as compared to the glacial periods. The mean sedimentation rate in the sediment core during the last 45 ka was 15.6 cm $(ka)^{-1}$.

4.2. Lithological character of the sediment core

To understand the lithological character, we have used concentration of calcium carbonate, total organic carbon (TOC) as a proxy for productivity and Al content as an indicator of lithogenic flux or as a clay mineral (Fig. 3). Calcium carbonate content in the sediment core varies from 0.8% to 50% with an average of 6% (Fig.3). Carbonate content is generally less than 5 % except four high peaks of 25%, 50%, 12% and 15% during 12.4 ka, 20.3 ka, 29 ka and 36 ka respectively. The highest carbonate content of 50 % is only one point at 20.3 ka was repeatedly analysed and is confirmed. The calcium carbonate content is generally low, and does not show any distribution pattern with the age and also can not be used as paleo- productivity proxy. Al content in the marine sediment is an indicator of terrigenous input and indirectly indicates the intensity of monsoonal precipitation. Al content in the sediment core varies from 6.5 % to 18.7 % (Fig. 3) with an average of 9.5 % which is higher than the upper crustal value of 8.04 % (McLennan, 2001). The highest carbonate content of 50 % is associated with lowest Al content of ~4 % at 20.3 ka suggestion dilution effect. The surface sediments from Krishna-Godavari basin showed high content of Al and very high content of clay ranging from 70 to 90 % (Pattan et al., 2008) suggesting Bay of Bengal is completely clay dominated area. The increased Al content in the sediment core of more than 12 % from 10 ka to 6.4 ka and centered around 9.5 ka suggesting intense monsoonal precipitation caused by the increased northern hemisphere summer insulation. Total organic carbon content in the sediment core varies from 0.24% to 2.85% with an average of 1.3 % (Fig. 3) and suggests that it is not high productivity domain. Therefore, the above data shows that present sediment core site is dominated by terrigenous material with less carbonate and organic carbon indicating low biological productivity area.

4.3 Uranium

The concentration of U in marine sediments and sedimentary rocks has been used extensively to trace the paleo-redox conditions of the depositional environment (Dean et al., 1997; Algeo and Maynard, 2004; Tribovillard et al., 2008; Algeo et al., 2012). Uranium behaves conservatively in oxygenated waters and its concentration remains almost uniform in different ocean basins as well as with water depth (Rosenthal et al., 1995; Crusius et al., 1996; Crusius and Thomson 2000; Algeo and Lyons, 2006., Algeo and Tribovillard, 2009; Algeo et al., 2012 and reference therein). U is present as uranyl carbonate complex $UO_2(CO_3)_{4-3}$ and has a residence time of ~500 ka in the ocean (Colodner et al., 1993; 1995; Crusius et al., 1996; Challou et al., 2002). Under anoxic and suboxic conditions, U shows a strong enrichment compared to continental crust values and exhibits a positive covariation with Mo content (Zheng et al., 2000).

The U content in the core varied from 1.4 ppm to 9.4 ppm with an average of 3 ppm which is close to upper continental crust value of 2.8 ppm (McLennan, 2001). Moderately high U content of 9.4 ppm occurred at 9.5 ka suggest an oxygen-depleted environment. Further, there is some indication of beginning of development of reducing condition at 20.5 ka where U content is 6 ppm. Uranium is normalized with Th to eliminate the detrital effect. The moderately high U/Th ratio of 0.6 at 9.5 ka is suggestive of an oxygen-deficient environment probably suboxic condition (Fig. 4) while rest of the core has U/Th ratio less than crustal value (0.26, McLennan, 2001) suggesting oxic depositional condition. High U/Th values (>1.25) are recorded in the sediments exposed to the OMZ in the Arabian Sea, indicative of near anoxia (Nath et al., 1997), whereas very low values (~0.1) in the sediments of Central Indian Ocean Basin south of equator are suggestive of oxic depositional conditions (Pattan et al., 2005). The moderately high U/Th ratio at 9.5 ka is quite lower than sediments of Oxygen Minimum Zone in the Arabian Sea (>1.25) and higher than Central Indian Ocean Basin (0.1), is indicative of suboxic environment. Based on high U and TOC contents in the Arabian Sea, Sarkar et al. (1993) reported prevalence of anoxic conditions in the eastern Arabian Sea whereas, Pattan and Pearce (2009) suggested occurrence of suboxic conditions during LGM in the southeastern Arabian Sea. Sometime, high U content could be due to post depositional diagensis caused by rapid change in sedimentation rate as reported in Atlantic Ocean (Thomson et al., 1990). In the present study, there is no rapid increase in the sedimentation rate (Fig.) which is ruled out.

4.4 Molybdenum

With a residence time of ~780 ka, molybdenum (Mo) behaves conservatively in generally well-oxygenated water column, but shows considerable enrichment relative to other redox-sensitive elements (U, V, Ni and Cu) in reducing sediments compared to crustal abundance (Crusius et al., 1996). Therefore, Mo concentration in sediments and sedimentary rocks deposited under anoxic conditions preserve very well the local redox conditions at the time of sedimentation (Calvert and Pedersen, 1993; Dean et al., 1997; Crusius et al., 1996; Zheng et al, 2000; Yarincik et al., 2000; Tribovillard et al., 2004). Under anoxic or suboxic conditions Mo (VI) is reduced to Mo (IV) and is precipitated from solution (Calvert and Pedersen, 1993; Chaillou et al., 2002). In the presence of H_2S under critical threshold concentration, molybdate is converted to thiomolybdate (Helz et al., 1996; Zheng et al., 2000) that is particle reactive and adsorbed onto organic substances, mineral phases or Mn-Fe-oxyhydroxides and eventually gets buried in sediments (Helz et al., 1996; Tribovillard et al., 2004).

The Mo content in the sediment core varied from 0.3 ppm to 36 ppm with an average of 3.5 ppm and shows moderate enrichment compared to the continental crust value of 1.5 ppm (McLennan, 2001) (Fig.4). Piper and Isaacs (1996) and Crusius et al. (1996) proposed that Mo content of 5-40 ppm in the sediments is indicative of reducing conditions. Such was the case around 9.5 ka where the Mo concentration reached maxima of 36 ppm (Fig. 4). During the prolonged post glacial phase, Mo enrichment started around 15.2 ka and continued up to 4.5 ka. During this period, peaks occurred at 13.1 ka, 11.6 ka, 9.5 ka, 7.3 ka and 5.3 ka suggesting a cyclicity of suboxic conditions at about 2 ka intervals (Fig. 4), with comparatively more suboxic condition centered around 9.5 ka. There is moderate enrichment of Mo (10 ppm) from 20.9 ka to 20.5 ka (lasting very short period of about 0.4 ka), which appears to be the initiation of developing reducing condition or comparatively less oxic condition. During the LGM (from 20 ka to 15 ka - Govil and Naidu, 2011), late Holocene (last 4.5 ka) and from 46 to 21 ka the Mo content was < 5 ppm suggesting oxic conditions. In the southeastern Arabian Sea, Pattan and Pearce (2009) observed that high Mo content (~ 14 ppm) is associated with high Mn during the last 5 ka which could be the result of oxic diagenesis (Crusius et al., 1996). This association is quite different to that observed in the present sediment core. On the Mo content in the Cariaco Basin, and Mesozoic formations in Turkey, France, United Kingdom, and Russia have very high Mo content ranging from 40 to 200 ppm, mostly associated with sulfur-rich organic matter, suggesting highly anoxic basins (Dean et al., 1999; Yarincik et al., 2000; Tribovillard et al., 2004). In the present core Mo content reached to maximum of 36 ppm hence, there is no possibility of any sulfide rich organic matter. Increased Mo concentration up to 60 ppm at Ubra in the Permian-Triassic Panthalastic Ocean was caused by the higher level of productivity resulting suboxic bottom water environment (Algeo et al., 2011). Low carbonate and organic carbon content in the sediment core might not have caused the suboxic condition here. Occurring at 100 ka and 41 ka cyclicity, Mo enrichment in the Cariaco Basin is found during the productive interglacial periods, indicating anoxic bottom waters (Yarincik et al., 2000). Mo is also linked to Mn oxyhydroxides in oxic sediments (Calvert and Pedersen, 1993; Morford and Emerson, 1999) and it is preferentially adsorbed onto MnO_2 phase (Shimmield and Price, 1986). The reduction of Mn-oxyhydroxide phase may release the adsorbed Mo to pore waters. By contrast, in our core the highest Mo contents at 9.5 ka is corresponds to low Mn content suggesting that the ambient conditions were not oxic. The scavenging of Mo from the dissolved phase can take place before other redox sensitive elements, particularly U, leading to elevated Mo/U ratio (Algeo and Tribovillard, 2009). This is evident at 9.5 ka where the highest Mo/U ratio of 10.4 is observed in the present sediment core.

4.5 Mo and U enrichment factor

In order to investigate changes in benthic redox conditions in greater detail, we have calculated the Mo and U enrichment factors using the following equation:

$$X_{EF} = [\,(X/Al)_{sample} / (X/Al)_{crust}]$$

where X and Al are wt% of element X and Al, respectively. The upper continental crust values used are from McLennan (2001). Enrichment factors exceeding 3 indicate detectable authigenic enrichment while, those exceeding 10 indicate substantial enrichment of an element (Algeo and Tribovillard, 2009). The Mo enrichment factor shows a large variation ranging from 0.2 to 17.6 (Fig. 5). This factor remained quite low (~1) during the late Holocene (after 4.5 ka), LGM (20 ka-15 ka) and from 45 ka to 21 ka. The most prominent increase in the factor is recorded between 15.2 ka and 4.5 ka with the maximum of 17.6 located at 9.5 ka. The observed high Mo enrichment at 9.5 ka indicates the prevalence of suboxic condition (Algeo and Tribovillard, 2009). U enrichment factor varied from 0.5 to ~ 3. There is single point increase in U enrichment factor ~2 at 9.5 ka and ~3 at 20.5 ka (Fig. 5) which indicates not much authigenic enrichment (Algeo and Tribovillard, 2009). The Mo/U enrichment ratio ranges from 0.2 to ~20 with the highest value occurs at 9.5 ka. Mo/U enrichment ratio increases from 5 ka , reaches highest at 9.5ka and decreases up to 15 ka. Thus, enrichment ratios of Mo, U and Mo/U indicate suboxic condition for a fairly long period centering around 9.5 ka. Compared to BoB, the Cariaco and Orca basins which are marine systems with restricted deep-water circulation and experience sulfidic conditions, exhibit a high Mo enrichment factor of 10 to 50 and a low U enrichment factor (<2); consequently, the Mo/U enrichment ratio is 2-8 times that of seawater (Tribovillard et al., 2008). Similarly, the Black Sea sediments, particularly those in abyssal plain, are also greatly enriched in Mo as compared to U (Barnes and Cochran, 1991; Lyons, 1992). Based on Mo and U enrichment ratios, a increasing trend of reducing conditions is recorded in southern and central California shelves, southern California basins, Mexico Margin and Peru shelf (Algeo and Tribovillard, 2009 and reference therein).

4.6 Ce/Ce*

Ce/Ce* was calculated following the equation used by Bau and Dulski (1996).

Ce/Ce* value > 1, <1 and = 1 suggest positive, negative and flat type anomaly respectively (Murray et al., 1991). Ce/Ce* > 1 is a positive anomaly generally found in Fe-Mn oxide, hydrothermal deposits and clays with high Fe-Mn content suggesting a well oxidized environment. Ce/Ce* <1 is an indicative of negative anomaly suggesting reducing condition and it can be caused by the presence of calcareous, siliceous shells, phillipsites, phosphorites and authigenic smectite (Courtois and Jafferzic-Renault 1977; Desprairies and Courtois 1980; Tlig and Steinberg 1982, Elderfield et al., 1981., Pattan et al., 2005 and reference therein). Ce/Ce* = 1 is a flat type anomaly which could be due to presence of terrigenous dominated sediment (Nath et al., 1992).

Ce/Ce* values during the last 45 ka in the sediment core varies from 0.71 to 1.32 with an average of 1.18 (Fig. 4). Ce/Ce* in the core is generally high (1.3) during the last 4.9ka and from 10.98ka to 45ka suggestive of oxic depositional condition (Nath et al., 1992). Ce/Ce* value reduced to just below 1 (0.9) at 23.4ka and 39ka which indicates near oxic depositional condition. Ce/Ce* values reduced up to 0.71 from 7.4 ka to 10.46 ka indicate weak reducing or suboxic depositional condition and is interrupted by near oxic condition at 8.4 ka and 9.5 ka where Ce/Ce* value reaches to 1.12 and 0.94 respectively.

Low (<10%) calcium carbonate content (Fig. 3), presence of smectite derived from Deccan Basalts (Pattan et al., 2008) and absence of phillipsite and phosphorites in the study area may not be responsible for the Ce/Ce* values < 1 during 7.4 ka to 10.46 ka. It remains interesting to note that during 9.5 ka, though U/Th and Mo/Al ratio suggests suboxic condition whereas, Ce/Ce* suggest near oxic condition (Fig. 4). This mismatch needs an explanation. During 15 ka to 5 ka, high Zr content of 2000 ppm to 8000 pm is being reported and highest of 8000 ppm is at 9.5ka On the contrary, low Ce/Ce* value at 9.5 ka where highest Zr content is present. Positive Ce-anomaly or Ce/Ce* >1 is very common in all zircons of intermediate and basic rocks (Hoskin and Ireland.,2000; Hoskin and Schaltegger, 2003; Nardi et al., 2013). Further, they suggested that preferential enrichment of Ce compared to its neighboring REE elements is ascribed to the presence of tetravalent Ce, which would be able to form stronger ionic bonds and enriched during earliest stage of crystallization. Therefore, the inherent positive Ce-anomaly of the zircon mineral present within the sediments might have reduced or brought down the intensity of reducing condition ultimately resulting in lower Ce/Ce* values during 15 ka to 5 ka than expected. This is evident by the inverse correlation (r = -0.55) between Zr content and Ce/Ce* during the above period (Fig. 6). Therefore, presence of abundant zircon in the marine sediments may distort the original Ce/Ce* value particularly in the suboxic condition. Hence, usage of Ce/Ce* as an indicator of reducing condition can be used with a caution in an area dominated by zircon in the marine sediment.

4.7 Manganese and Iron

Manganese and iron concentrations have been normalized with that of Al to account for the detrital fraction. The Mn/Al ratio varied from 0.006 to 0.049 whereas the Fe/Al ratio ranged from 0.3 to 0.9 (Fig. 4). For most parts, the Mn/Al and Fe/Al ratios are above the upper continental crustal ratios of 0.0075 and 0.44 (McLennan, 2001) respectively, indicating the presence of structurally unsupported Mn and Fe in the form of Fe-Mn oxy-hydroxides. Lowest Mn/Al ratio of 0.006 and Fe/Al ratio of 0.3 which are less than the upper continental crustal value at around 9.5 ka suggest reducing condition probably suboxic. There is reasonably good positive correlation (r =0.44, n=95) between Fe/Al and Mn/Al ratio in the entire sediment core suggest their presence in the form of Fe-Mn oxyhydroxides (Fig.7). Higher Mn/Al and Fe/Al in the core top indicates diagenetic remobilization of Mn and Fe through pore waters and oxidation of upward-diffusing reduced species. During the decay of organic matter, microbes sequentially utilize oxygen, nitrate, Fe (III) and Mn (IV, III) (Froelich et al., 1979) resulting in reducing or suboxic condition at around 9.5 ka. Further, the lowest Mn/Al and Fe/Al ratios match well with increased Mo content and higher U/Th ratio which is indicative of suboxic condition at around 9.5 ka.

Extensive work has been carried out on the ratio of highly reactive iron to total iron (Fe_{HR}/Fe_T), total Fe to Aluminium (Fe_T/Al), degree of pyritization (DOP) and their enrichment is used as paleoredox proxies in the euxinic basins (Lyons and Severmann, 2006). Black Sea, Effingham Inlet and Orca Basin have high (Fe_T/Al) ratio of 0.6 to 1.2, 0.08 to 0.09 and 0.055 to 0.075 during euxinic condition compared to their oxic shelf of 0.05 to 0.6, 0.55 to 0.65 and 0.4 to 0.5 respectively (Lyons and Severmann, 2006). In BOB high (Fe_T/Al) ratio of 0.5 to 0.088 occur in the oxic sediments except at ~9.5 ka where it reduced to 0.038 due to suboxic condition. The elevated oxic Fe_T/Al values could be due to influence of high detrital sedimentation rates. Further study of Fe_{HR}/Fe_T ratio (oxic sediments also enriched in $Fe_{(HR)}$- Raiswell and Anderson, 2005) and mass balance relationship between source and sink of transported reactive iron needs to be studied (Lyons and Severmann, 2006).

4.8 Moderately redox- sensitive trace elements

Trace elements such as V, Cu, Co and Ni are moderately redox-sensitive trace elements, their abundance and ratios such as Ni/Co, V/Cr, V/Ni and V/(V+Ni) are used to trace the paleoredox conditions (Hatch and Leventhal 1992; Jones and Manning 1994; Wignall et al., 2007; Zhou et al., 2012; Xiong et al., 2012. Cr is normally incorporated in detrital fraction where it may substitute Al within clays. Its content is high at 9.5 ka and follows Al distribution. None of the above trace element ratios indicate any signature of reducing condition at 9.5 ka except Ni/Co which shows some indication (Ni.Co ratio of 4.5) suggesting an occurrence of dysoxic to suboxic conditions (Jones and Manning 1994). The authigenic enrichment of Cu, Ni and Co occurs mainly in euxinic environmental conditions where these trace elements are taken up in iron sulfide phase or precipitated as their own sulfides (Calvert and Pederson, 1993; Algeo and Maynard, 2004; Tribovillard et al., 2006). In the present core there is no euxinic condition as a result these trace elements and their ratio do not exhibit variation at 9.5 ka.

5. Possible causes for the development of reducing conditions

Development of suboxic condition during the periods 15.2-4.5 ka peaking at 9.5 ka could possibly arise from (a) decrease in bottom water oxygen concentration due to changes in circulation, and (b) increase in oxygen demand in the benthic environment due to larger supply of labile organic carbon from the surface layer. While substantial rearrangement in the subsurface water circulation appears to have occurred on a glacial to interglacial time scale, it is highly unlikely that the deep waters in the BoB would have turned anoxic during the time period of interest (Naqvi et al., 1994; Boyle et al., 1995; and references therein). Therefore, the inferred suboxic event should arise primarily from enhanced benthic respiration. An obvious cause of this may be higher primary production that is expected to have led to greater export of organic matter to seafloor. Unlike the Arabian Sea, where past changes in productivity have been investigated quite extensively (Naidu and Malmgren, 1996; Singh et al., 2011; and reference therein), not much is known about paleoproductivity in BoB. One may expect an increase in productivity to have occurred during the early Holocene due to larger flux of nutrients by elevated river runoff and possibly more vigorous wind-driven upwelling along the western boundary of BoB. To test this, we investigated the variability of TOC and TN in our core, along with their stable isotopic composition (Fig. 8). The TOC content in the marine sediments is often taken to serve as a proxy of surface water productivity (e.g., Muller and Suess, 1979; Pedersen and Calvert, 1990). TOC and TN contents in our core vary from 0.2% to 2.8% and from 0.02% to 0.2% with average values of 1.26% and 0.11%, respectively. The two parameters are strongly correlated (r=0.9, n=98). However, surprisingly, both TOC and TN do not show much variability during the past ~ 25 ka except for the outliers around 13 ka. In both cases, relative maxima are recorded around 2-3 ka while the core top values are among the lowest observed in the core. If we ignore the two above-mentioned outliers (single anomalously low values), the TOC and TN records also exhibit relative peaks around 12-14 ka. Significantly, during the period intervening the two relatively higher values, when the SW monsoon is known to have intensified and the redox-sensitive metal records described above show moderately reducing condition in the sediments, both TOC and TN contents remain moderate and close to the long-term average values for the core. Further, down in the core (from between 45 ka to 25 ka), variations observed in the TOC and TN are much larger and mutually consistent. Of particular interest are the peaks observed (highest measured in the core) in both TOC and TN records around 42 ka. The TOC and TN decreased to approximately 10 and 7 times of the peak values in about 2 ka respectively, and continued to oscillate until ~25 ka. Interestingly, the records of redox-sensitive metals exhibited little variability during this period.

The absence of TOC and TN maxima around 9.5 ka, suggests that the higher productivity *per se* is not responsible for the development of suboxic condition in the benthic environment in the BoB. We propose that rather than the total flux of the organic matter reaching the seafloor, it is the flux of labile organic matter that controls benthic respiration and redox-element cycling. In other words, the nature of the organic matter is more important. The stable isotopic composition and C/N ratio in marine sediments have traditionally been used to trace the source of TOC. The C/N ratio of phytoplankon and zooplankton (the Redfield value) is ~6.6; freshly deposited organic matter has a ratio of ~10; and terrigenously derived material has ratios ranging from 20 to 200 (Hedges and Parker 1976; Emerson and Hedges, 1998; Meyer, 1997). The C/N ratio in the core varies from 9 to 16 with an average of 11 which suggests organic carbon is to a large extent of marine origin (Muller, 1977). This is contrary to the results of Galy et al. (2007) according to which the Bengal Fan sediments have terrestrially derived TOC exported by the Ganges-Brahmaputra river system. A large fraction of terrigenous organic matter brought by the numerous rivers including Ganges-Brahmaputra may be deposited over the continental margin itself (Berner and Raisewell 1983; Hedges and Keil, 1995). The view that the TOC in the sediment core is not predominantly of terrigenous origin is also supported by the $\delta^{13}C$ data (Fig. 7) that do not show very negative values that characterize the terrestrially derived carbon (Maya et al., 2011, and references therein).

Despite the dominance of the marine organic matter at our core site, terrigenous inputs could still play a key role in determining the quality, especially the lability, of organic matter reaching the seafloor. Observations during the Geochemical Ocean Sections Study (GEOSECS) revealed an interesting and unique aspect of BoB biogeochemistry: Data from at a station (# 446), that was located close to our core site, showed significant oxygen consumption and nutrient regeneration in ~100 m thick benthic mixed layer compared to overlying water column indicating that fresh organic matter was reaching the deep seafloor supporting elevated rates of respiration and particle dissolution (Broecker et al., 1980). Subsequently, it was proposed that the huge influx of lithogenic material brought by the rivers interacts with labile organic matter, forming dense mineral matter-biogenic aggregates that sink rapidly to the seafloor (Ittekkot et al., 1992). This ballast-hypothesis can explain the relative large export of organic matter to the deep sea as measured by sediment traps (Ittekkot et al., 1991), weak north-south gradients in oxygen and products of microbial degradation of organic matter (such as nutrients and carbon dioxide) (Rao et al., 1994) and low respiration rates in subsurface waters (Naqvi et al., 1996). It is reasonable to expect that during periods when the lithogenic flux to surface waters was higher than it is today, the flux of fresh, labile organic matter reaching the seafloor would also have been higher. Such is expected to have been the case during the period of major change in benthic redox environment (15.2-4.5 ka) for two reasons. First, the abrupt rise of sea level and discharge of melt water would have resulted in greater erosion (from shelves exposed during the LGM), and second, the SW monsoon intensity was also at its maximum due increase in the northern hemisphere summer insulation that peaked around ~11 ka.

Variability of lithogenic elements in our core lends support to the above hypothesis. In the BoB, fresh water discharge during the SW monsoon is associated with large particle flux of up to ~350 mg m^{-2} d^{-1}, of which lithogenic flux is the dominant component (~200 mg m^{-2} d^{-1}) (Ittekott et al., 1991; 1992). In our core, the period centering around 9.5 ka is distinguished by very high contents of Al (18%), Zr (0.8%) and Hf (180 ppm) (Fig.9). These elements are enriched approximately more than 2, 40 and 30 times, respectively, relative to the upper continental crust (McLennan, 2001), pointing to greater supply of lithogenic material to the seafloor during the post-glacial intensification of the SW monsoon. The remarkable similarity of Zr and Hf distribution in the core could be due to their occurrence in the zircon mineral. The period of the major SW monsoon intensification inferred from Al, Zr and Hf distribution in our core (15.2-4.5 ka) is very similar to those reported by previous workers using different proxies. For example, based on the benthic carbon isotope record in the Gulf of Aden (reflecting hydrographic conditions in the Red Sea), Naqvi and Fairbanks (1996) reported that the Afro-Asian monsoon intensified between 15.5 ka and 7.3 ka. Kudrass et al (2001) reported a large decrease in salinity (from ~35 to <31.5) from ~16 ka to 10 ka (31.25‰). The low salinities lasted until 4ka. From the oxygen isotope analysis of planktonic foraminifera in core SK/218/1 itself, Govil and Naidu (2011) showed increase of rainfall/river discharge after the initiation of deglaciation. Within 15.2 ka to 4.5 ka, secondary peaks suggesting relatively higher SW monsoonal precipitation occurred around 13.1, 11.6, 9.5, 7.3 and 5.3 ka, with the most prominent one located at 9.5 ka (Fig. 8). The approximately regular recurrence of events of more intense SW monsoon at about 2 ka intervals, is similar to that of the inferred changes in the benthic redox environment. This is confirmed by the strong positive correlation (r=0.98) between Zr and Mo (Fig. 10) during 15.2 ka to 4 ka while, rest of the core except 15.2 ka to 4 ka, Zr and Mo do not show any correlation (r=0.17) (Fig. 10). Our observation of highest terrigenous material supplied by the fresh water discharge at 9.5 ka is similar to the earlier observations made in BoB (Chauhan and Suneethi.,2001; Reddy and Rao., 2001; Chauhan, 2003).

Galy et al. (2007) noticed a positive relationship between TOC and Al/Si ratio in source rocks, rivers draining from Himalayas and Bengal Fan sediments and concluded that the terrestrial organic matter is efficiently stored in the Bengal Fan. However, as pointed out earlier, in the present sediment core highest concentrations of terrigenous material recorded at 9.5 ka, presumably due to intensification of SW Monsoon, is not associated with increase in TOC content and the TOC is mostly of marine nature. This indicates vertical sedimentation of terrigenous matter at our core site (along with labile organic matter of marine origin) rather than (horizontal) spreading by turbidity currents. This labile organic carbon might have been oxidized quickly without affecting the TOC content of the sediments during this period resulting in suboxic condition close to the sediment-water interface.

Fractionation between light rare earth elements (LREE) and heavy rare earth elements (HREE) can be studied using the $La_{(N)}/Yb_{(N)}$ ratio in the sediments where N is the shale-normalized value. In the present sediment core, $La_{(N)}/Yb_{(N)}$ ratio shows large fluctuation varying from 0.4 to 1.7 with an average of 1.14 (Fig. 8). Surprisingly, suboxic and increased monsoonal precipitation around 9.5 ka is associated with lowest $La_{(N)}/Yb_{(N)}$ ratio (0.4). However, there is one lower ratio at 20.4 ka. The $La_{(N)}/Yb_{(N)}$ ratio of 1 suggests the terrigenous source (Pattan et al., 2005, and reference therein) and the ratio in calcium carbonate is < 1 because it generally follows seawater pattern where HREEs are enriched compared to LREEs. The $La_{(N)}/Yb_{(N)}$ ratio fell below 1 starting from 15.2 ka soon after the initiation of deglaciation and continued to be <1 up to 4.5 ka, after reaching its minimum (0.4) at 9.5 ka. Similarly low $La_{(N)}/Yb_{(N)}$ at 20.5 ka and at 36 ka suggest enrichment of HREEs over LREEs. On the contrary, $La_{(N)}/Yb_{(N)}$ exceeded 1 (1.14 average) during the late Holocene, LGM and from 45 ka to 20 ka reflecting the opposite trend (enrichment of LREEs over HREEs) and appears to be flat which suggest as terrigenous input (Sholkovitz, 1990). Since the carbonate content of the sediments is low (~5%), lower $La_{(N)}/Yb_{(N)}$ could not be caused by the carbonate content. The enrichment of HREE over LREE from 15.2 ka to 4.5 ka particularly highest during 9.5 ka could be due to presence of abundant zircon (Zr content 0.8%) which generally exhibit large enrichment of HREE over LREE (Hoskin and Ireland, 2000; Hoskin and Schaltegger, 2003; Nardi et al., 2013). Therefore, $La_{(N)}/Yb_{(N)} < 1$ from 15.2 ka to 4.5 ka is caused by the presence of zircon with varying abundance supplied by the stronger SW monsoon and rest of the core where, $La_{(N)}/Yb_{(N)}$ is >1 with almost flat type suggest supply of terrestrially derived clay material (Sholkovitz,1990). The distribution of Zr and Hf during 15.2 to 4.5 ka is exactly opposite to the behaviour of $La_{(N}/Yb_{(N)}$ ratio suggesting its control over the ratio.

6. Decoupling between benthic and pelagic processes

One of the most important and interesting findings of our study is that while the period of intense benthic respiration (i.e. development of suboxic condition) did not experience marked accumulation of organic carbon in the sediments, those of substantial organic carbon accumulation did not show any anomalies in redox-sensitive metals. While, the TOC and TN contents generally varied within narrow ranges over the past 25 ka, the amplitudes of variations were much larger in older sediments. During the latter period (45 ka-25 ka), the TOC and TN contents exceeded the long-term averages on several occasions. Such was particularly the case around 42 ka when the two parameters reached their highest values (2.8% and 0.2%, respectively). In most likelihood these events of TOC and TN accumulation in the sediments represent periods of much higher biological production in BoB than today. Singh et al. (2011) have recently reconstructed changes in productivity in the eastern Arabian Sea over the past 80 ka based on planktonic foraminifera abundance and TOC in sediment cores off Goa (central west coast of India). Their results reveal that, in contrast to the trend observed in the northern and western Arabian Sea, productivity in the eastern Arabian Sea was higher during the glacial period than today. However, productivity decreased drastically during the cold Heinrich events, as also happened elsewhere in the Arabian Sea. Our results suggest that the productivity changes associated with the Heinrich and Dansgaard-Oeschger (D-O) events, representing episodes of abrupt cooling and warming, respectively, in the North Atlantic extend to the Bay of Bengal as well. Although the mechanistic link between the abrupt climate change in the North Atlantic and biogeochemistry of the distant North Indian Ocean is not completely known, the association is so good that biogeochemical proxy records (particularly of sedimentary $\delta^{15}N$) have been used to fine-tune age models for sedimentary cores (Altabet et al., 2002;

Singh et al., 2011). The remarkable similarity of the sedimentary $\delta^{15}N$ records from the Arabian Sea with the oxygen isotope records in the Greenland Ice cores (Suthoff et al., 2001; Altabet et al., 2002; Singh et al., 2011) demonstrates how well the global climate subsystems are interconnected, and so it is to be expected that climatic oscillations in the North Atlantic would also affect BoB biogeochemistry. Given the resolution constraints, we cannot precisely identify all Heinrich or D-O events in our core, but from a comparison with the published records from the Arabian Sea, we believe that the maxima in TOC, TN and $\delta^{15}N$ (discussed below) appearing ~42 ka in the core correspond to D-O event 12 in the North Atlantic. This was the period when surface productivity at our core site was the highest during the entire period covered by the core. This increase in productivity does not seem to have resulted from a more intense SW monsoon because of a lack of accumulation of terrigenous elements in the sediments. Instead, other changes in hydrographic conditions, most probably a weaker stratification, could have led to enhanced supply of nutrients to the euphotic zone from the thermocline. It may be pointed out that the pattern of productivity changes in the western BoB bears similarity with that reported from the eastern Arabian Sea (Singh et al., 2011), except for the Holocene period. This is not surprising because like the BoB the eastern Arabian Sea is also affected by fresher water influx due to land runoff during the SW monsoon and the advection of BoB waters by the northward flowing West India Coastal Current during the Northeast Monsoon (Naqvi et al., 2000; Jayakumar et al., 2001).

As pointed out earlier, periods of TOC accumulation in the sediments were not associated with the development of reducing conditions in the benthic environment prior to 25 ka. However, there is some indication from the $\delta^{15}N$ record in the core – the first from the BoB – that the changes in productivity were also associated with biogeochemical changes in the water column (Fig. 7). The $\delta^{15}N$ of sedimentary organic matter has declined steeply over the past ~2 ka with the core top value (~3 ‰) being much lower than the average value of nitrate in seawater (4.7 ‰ - Altabet et al., 1999). Similar declines are also seen in the TOC and TN records. This indicates that the conditions in the present day BoB where the OMZ is just short of being reducing (Rao et al., 1994) have not been not typical for most periods covered by the core, and such low values in the past were probably largely associated with the Heinrich events and the LGM. The scarce organic matter at the core top seems to have substantial terrestrial component. Over much of the Holocene and deglaciation, the $\delta^{15}N$ generally varied within a narrow range (~5-6 ‰) i.e. marginally above the average $\delta^{15}N$ of nitrate in deep water. The $\delta^{15}N$ declined during the LGM, and fluctuated over a relatively larger range during the Marine Isotope Stage

(MIS) - 3. As in case of TOC, peak of $\delta^{15}N$ appear to have been associated with the D-O warm events with the most prominent one corresponding to D-O 12. Thus, changes in $\delta^{15}N$ associated with the stadial and interstadial events of MIS-3 appear to occur all over the North Indian Ocean. It may be debated whether the signal was locally produced in BoB or is the result of advection of intermediate waters from the Arabian Sea where intense denitrification appears to have occurred during the interstadial periods (Suthoff et al., 2001; Altabet et al., 2002; Singh et al., 2011). We believe the former was the case despite the fact that the maximum values of $\delta^{15}N$ (during D-O 12, ~13 ka, and during the mid- to late-Holocene (with the exception of the past 2 ka)) are ~6 ‰. It may, however, be noted that some dilution of sedimentary $\delta^{15}N$ must have occurred by the addition of isotopically light terrestrial nitrogen. If, as a very rough estimate, we assume the terrestrial fraction to be 0.25 (average of all LGM values in the BoB reported by Fontugne and Duplessy (1986)) and take the $\delta^{15}N$ of the terrestrial end member to be 2 ‰, the maximum $\delta^{15}N$ value of the marine end member would be 7.75‰. Although lower than the maximum $\delta^{15}N$ values observed in the Arabian Sea sediments, it does imply that weak denitrification most probably occurred in the BoB in the past. However, the fact that the periods of elevated sedimentary $\delta^{15}N$ did not always correspond to those of more intense mesopelagic OMZ (especially during the D-O events), the pelagic and benthic reducing conditions were temporally separated. This is because they were driven by different forcings: While the reducing conditions in the benthic environment seem to have been driven by intensified SW monsoonal runoff, those in the OMZ appear to be related to enhanced productivity.

7. Conclusions

1. Redox-sensitive element data in the sediment core from Bay of Bengal suggest prevalence of suboxic condition in the benthic environment from 15.2 to 4.5 ka peaking at 9.5 ka. Oxic condition occurred during late Holocene (last 4.5 ka), Last Glacial maximum (20 ka-15 ka), and from 45 ka to 21 ka.
2. Suboxic condition around 9.5 ka is associated with highest concentration of terrestrially-derived elements such as Al, Zr and Hf, which are mostly supplied from weathering and erosion of the Himalayas by the Ganges-Brahmaputra Rivers suggest intensified SW monsoon. During 15.2 ka to 4.5 ka, the increase in precipitation/runoff occurred at regular cyclicity of ~ 2 ka and is similar to that inferred for suboxic condition, strongly pointing to a linkage between monsoonal

runoff and development of suboxic condition. This is evident by the strong positive correlation (r=0.98) between Mo and Zr during 15.2 ka to 4.5 ka.

3. Development of suboxic condition at 9.5 ka is not caused by higher surface productivity because TOC and TN contents do not show any increase at that time. Further, C/N ratio and $\delta^{13}C$ of sediment shows that TOC during the last 45 ka has been of marine origin.
4. Shelf sediments in the BoB showed a strong positive correlation between TOC and terrigenous material, but such a relationship does not extend to our core site. The enormous terrigenous material probably reacts with liable organic matter, forming dense aggregates that sink rapidly to the sea floor creating near-bottom anomalies. Such a ballast effect is expected to have increased during periods of larger supply of terrigenous matter and the decay of labile organic matter would lead to the development of suboxic condition in the benthic environment at 9.5 ka.
5. Increased terrigenous material supply at ~9.5 ka is associated with lowest $La_{(N)/}Yb_{(N)/}$ ratio of 0.4 compared to the average value of ~ 1.2 in the sediment core. This suggests enrichment of heavy REEs over light REEs during 9.5 ka, whereas rest of the core showed enrichment of light REEs over heavy REEs. This could be due to presence of abundant zircon mineral which is characterized by enrichment of HREE over LREE at 9.5 ka.
6. Development of suboxic condition in the sediments seems to be largely decoupled to changes in the water column i.e. the intensity of the OMZ. The latter seems to have varied significantly, as indicated by the sedimentary $\delta^{15}N$ record, especially during MIS-3 (from ~25-45 ka), apparently coeval with the stadial and interstadial periods (Heinrich and Dansgaard-Oeschger events) originating in the North Atlantic. The tele-effects of these events appear to have extended to the entire North Indian Ocean. The most prominent change seems to be associated with D-O 12, around 42 ka BP, when the proxy records show high surface productivity and occurrence of denitrification within the OMZ. Surprisingly, however, there is no evidence for the prevalence of reducing conditions in organic-rich sediments at this time.

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Captions for the figures.

Fig. 1. Map showing location of sediment core (SK/218/1) from western Bay of Bengal.

Fig. 2. Plot of depth (cm) verses calendar age (ka) for the sediment core (SK/218/1), after Govil and Naidu (2011).

Fig. 3. Distribution of lithological parameters such as calcium carbonate (wt%), Al (wt%) and total organic carbon (wt%) in the sediment core.

Fig. 4. Distribution of U/Th ratio, Mo, Ce-anomaly, Mn/Al and Fe/Al ratio in the sediment core during the past 45 ka. Arrows in the lower panel indicate AMS dates.

Fig. 5. Inverse relation between Zr (ppm) and Ce/Ce* for the sediments between 15.2 ka to 4.5 ka in the core.

Fig. 6. Behaviour of Mo, U and Mo/U enrichment factors in the sediment core.

Fig. 7. Correlation between Fe/Al and Mn/Al ratio in the sediment core.

Fig. 8. Distribution of productivity indicative parameters such as bulk TOC, TN, and carbon and nitrogen isotope in the sediment core. (DO-12 is Donsgaard-Oeschger event 12).

Fig. 9. Distribution of Al, Zr, Hf concentration and $La_{(N)}/Yb_{(N)}$ ratio in the sediment core.

Fig. 10. Correlation between Zr (ppm) and Mo (ppm) content in the sediments from 15.2 ka to 4.5 ka (top) and from 45 ka to 15.6 ka in the sediment core (bottom).

Fig. 1

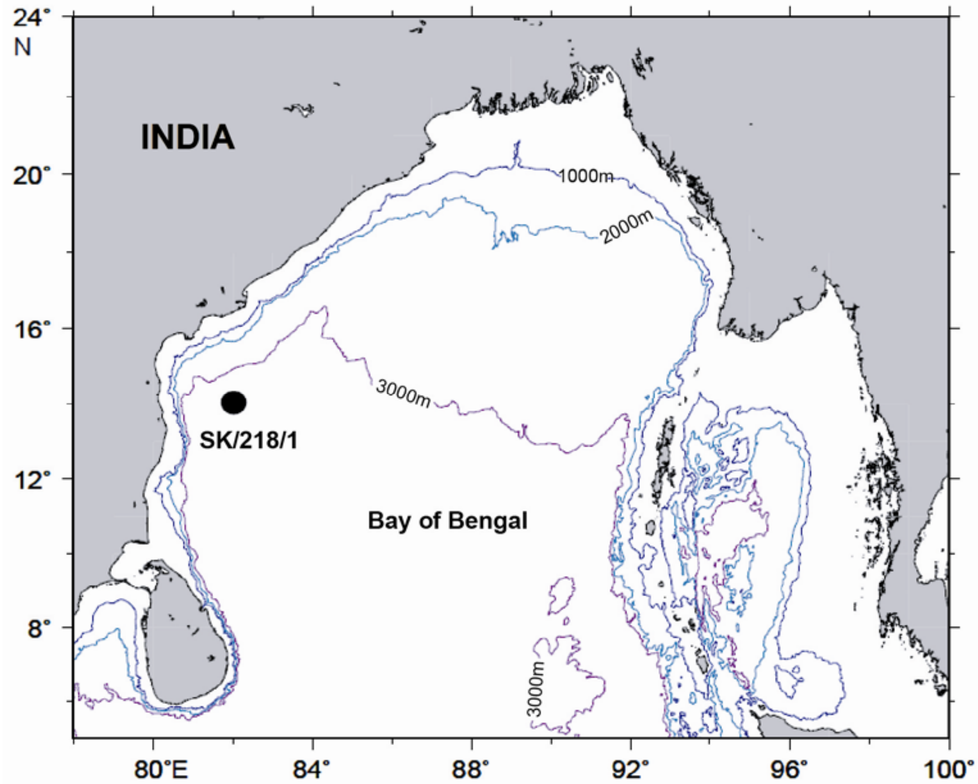

24°
N
20°
16°
12°
8°
INDIA
1000m
2000m
3000m
SK/218/1
Bay of Bengal
3000m
80°E
84°
88°
92°
96°
100°

Fig. 2

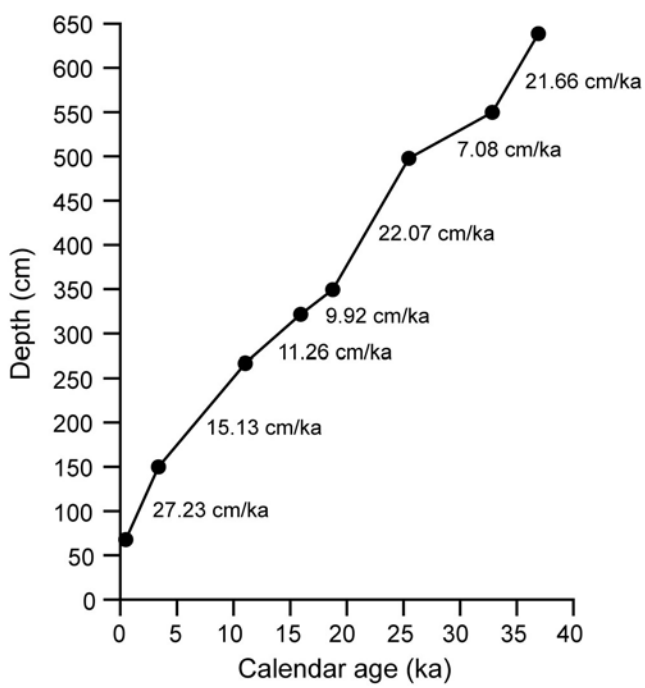
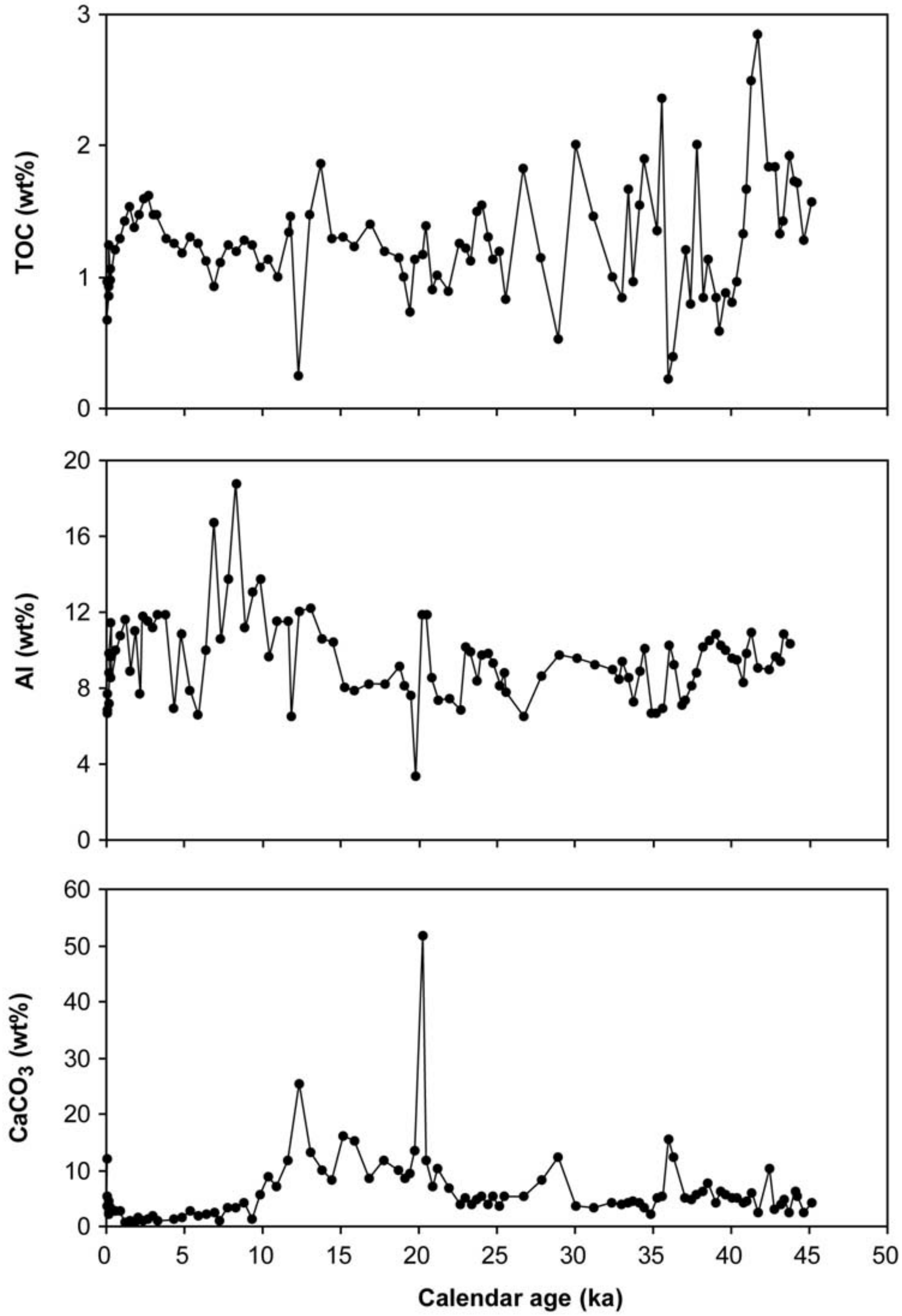

Depth (cm)
Calendar age (ka)
21.66 cm/ka
7.08 cm/ka
22.07 cm/ka
9.92 cm/ka
11.26 cm/ka
15.13 cm/ka
27.23 cm/ka
0
50
100
150
200
250
300
350
400
450
500
550
600
650
0
5
10
15
20
25
30
35
40

Fig. 3



TOC (wt%)
Al (wt%)
$CaCO_3$ (wt%)
Calendar age (ka)
0
1
2
3
0
4
8
12
16
20
0
10
20
30
40
50
60
0
5
10
15
20
25
30
35
40
45
50

Fig. 4

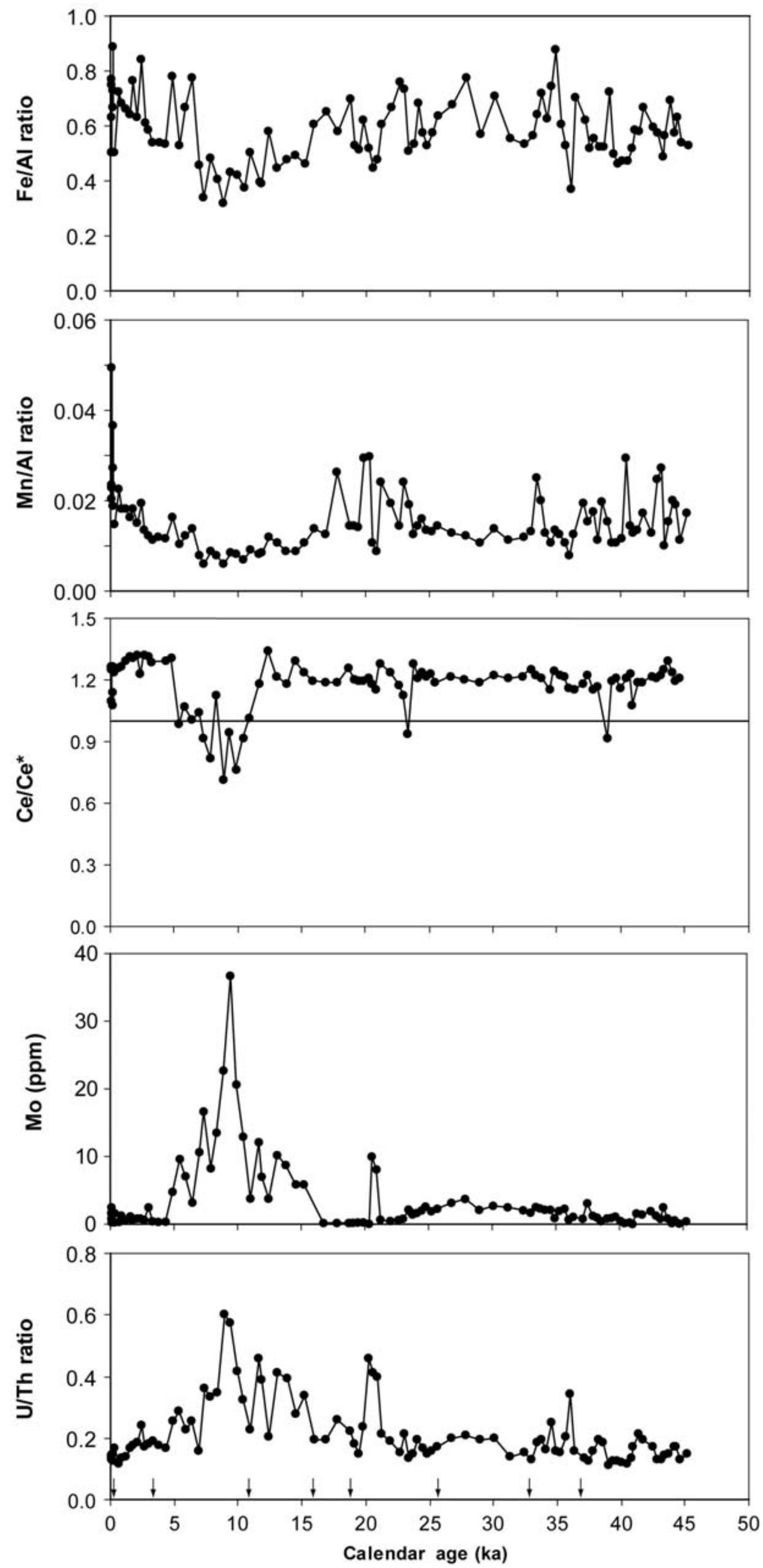
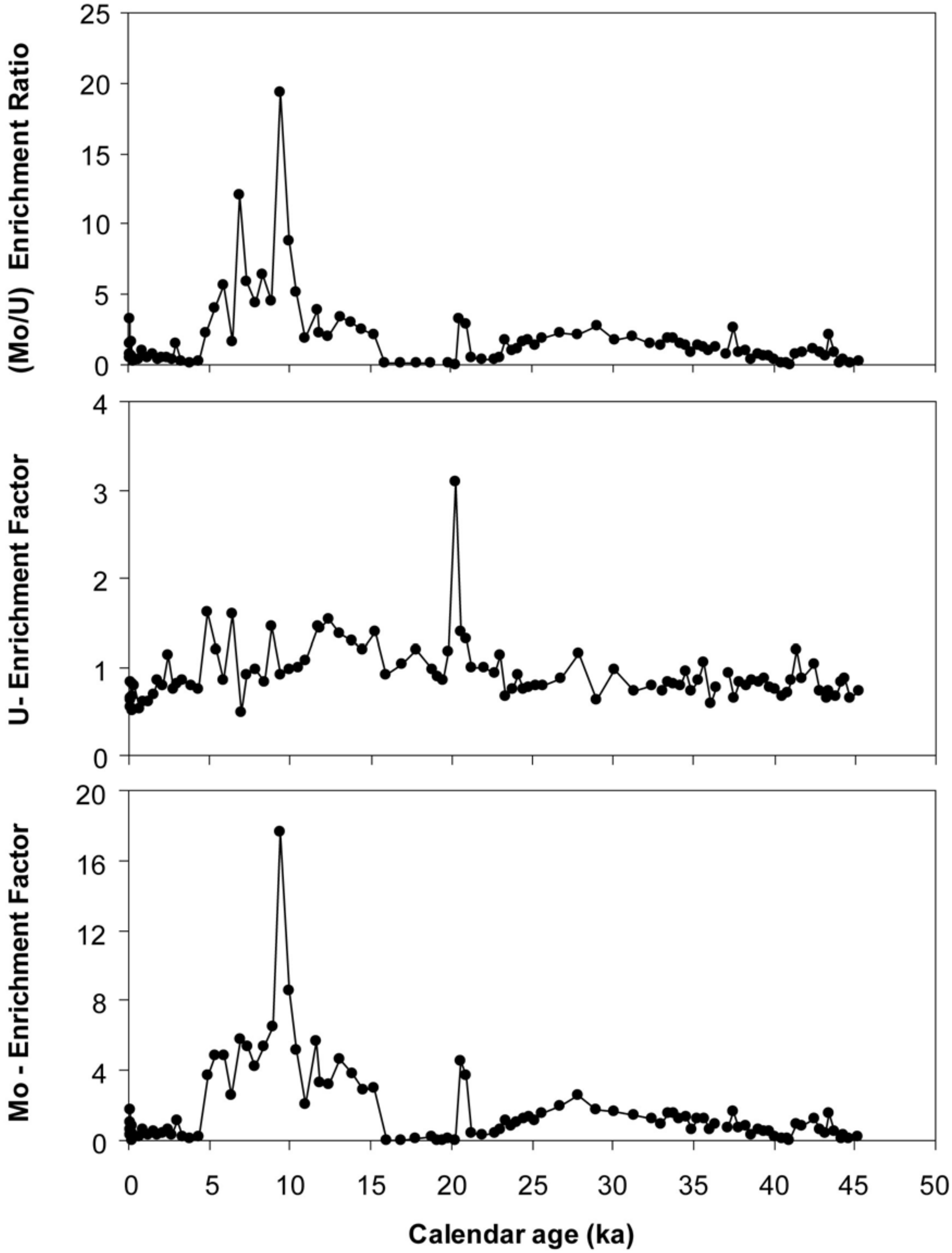
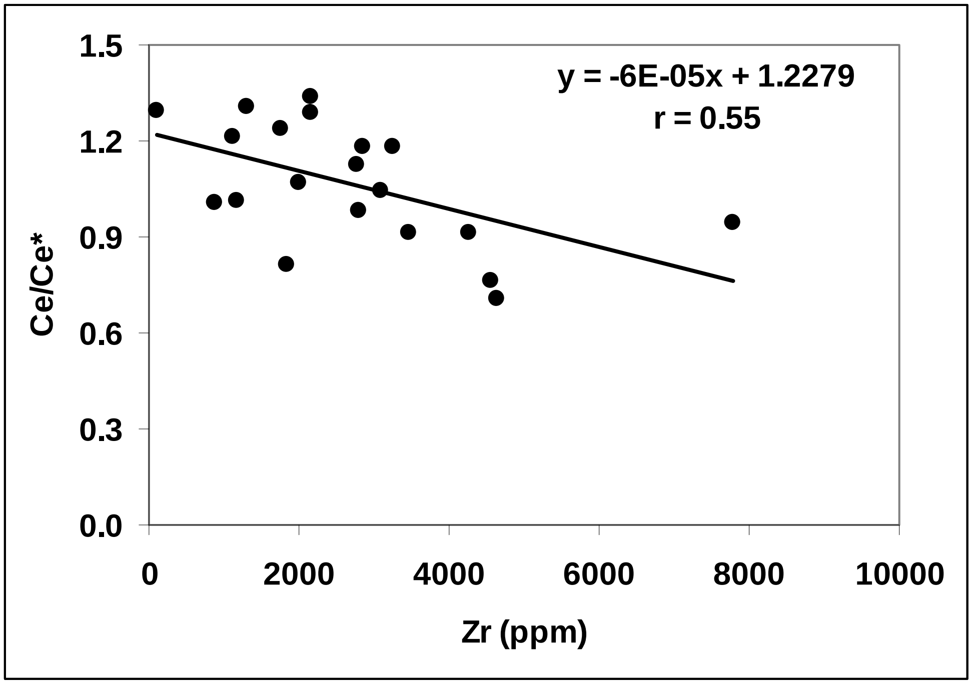


Fig. 5



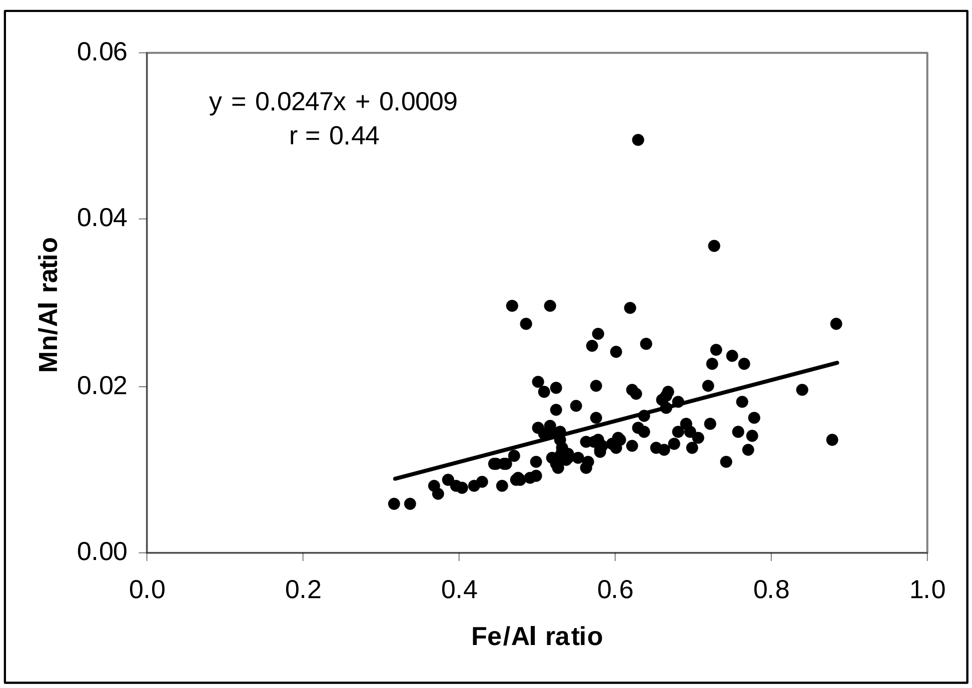
(Mo/U) Enrichment Ratio
U- Enrichment Factor
Mo - Enrichment Factor
Calendar age (ka)
0
5
10
15
20
25
30
35
40
45
50

Fig. 6



y = -6E-05x + 1.2279
r = 0.55
Ce/Ce*
Zr (ppm)
1.5
1.2
0.9
0.6
0.3
0.0
0
2000
4000
6000
8000
10000

Fig.7



y = 0.0247x + 0.0009
r = 0.44
Mn/Al ratio
Fe/Al ratio
0.06
0.04
0.02
0.00
0.0
0.2
0.4
0.6
0.8
1.0

Fig. 8

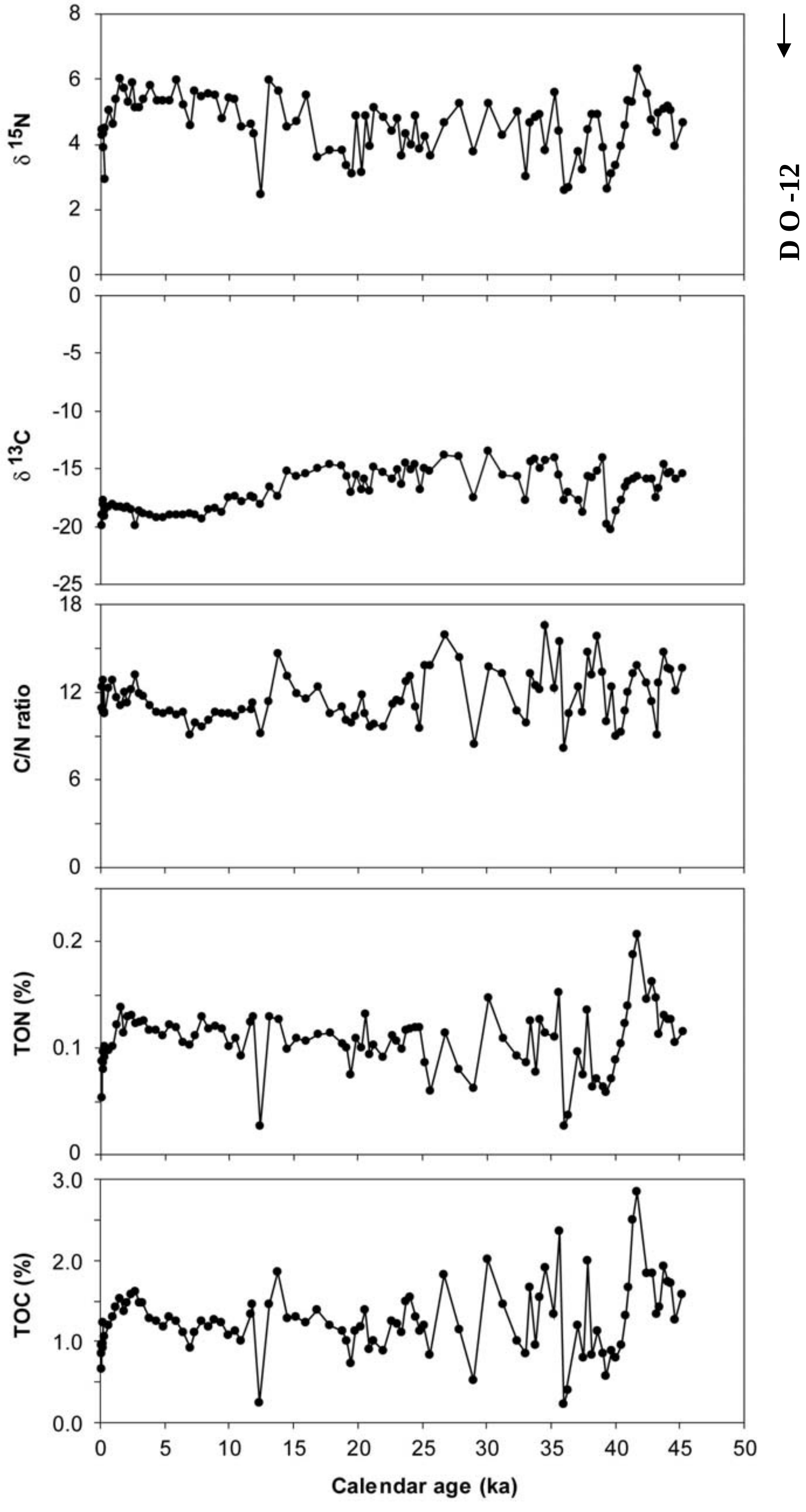
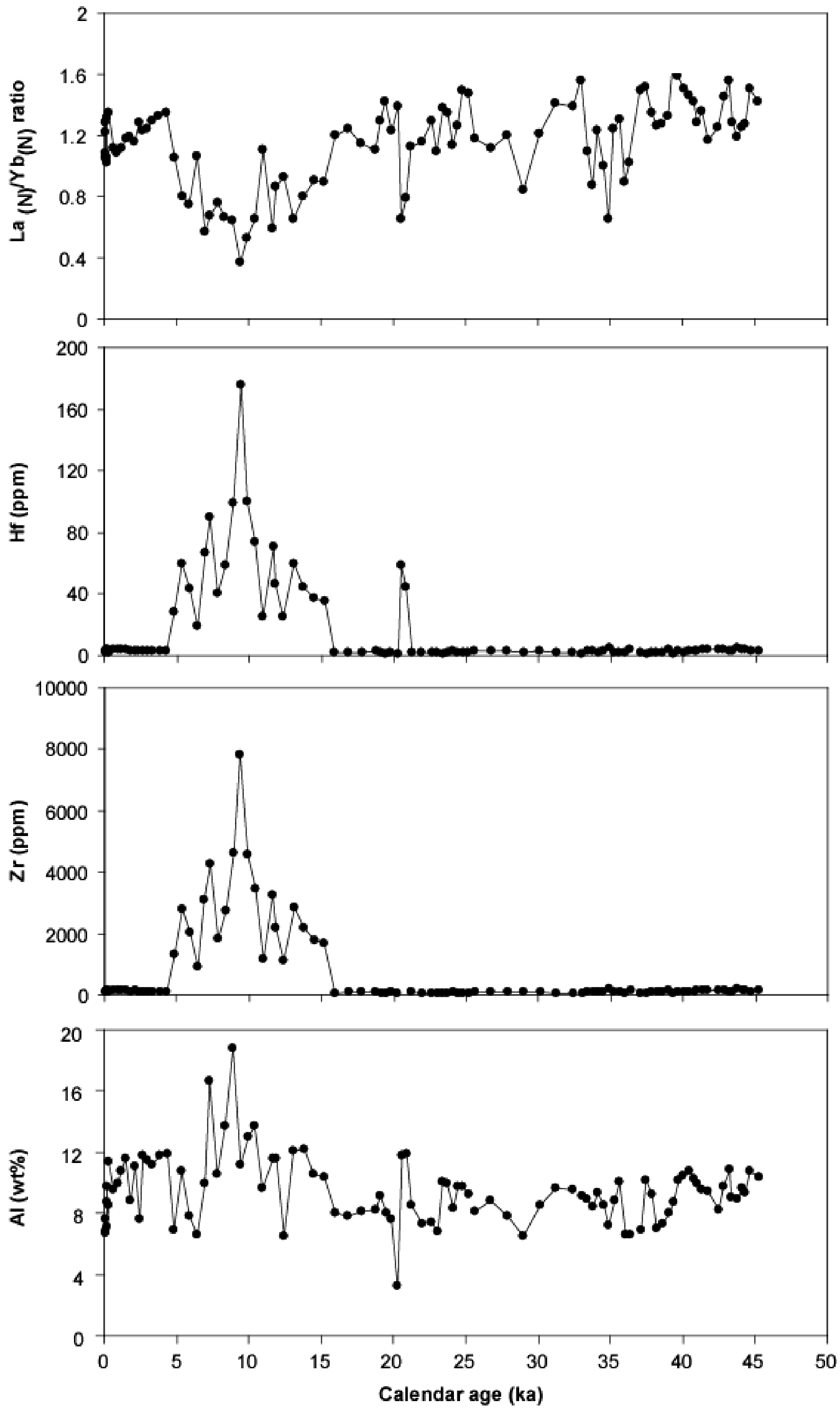
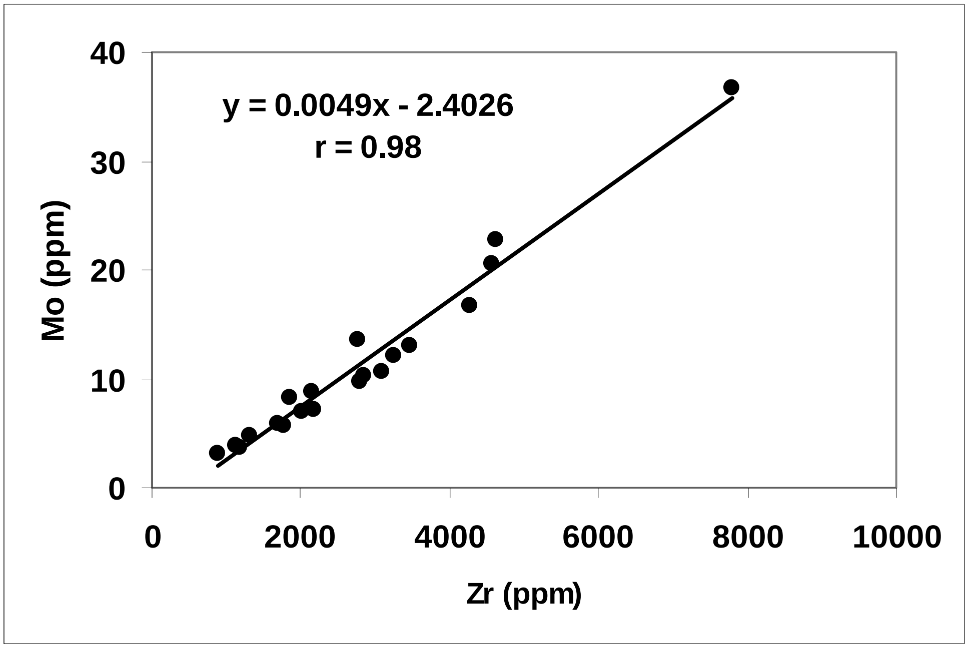


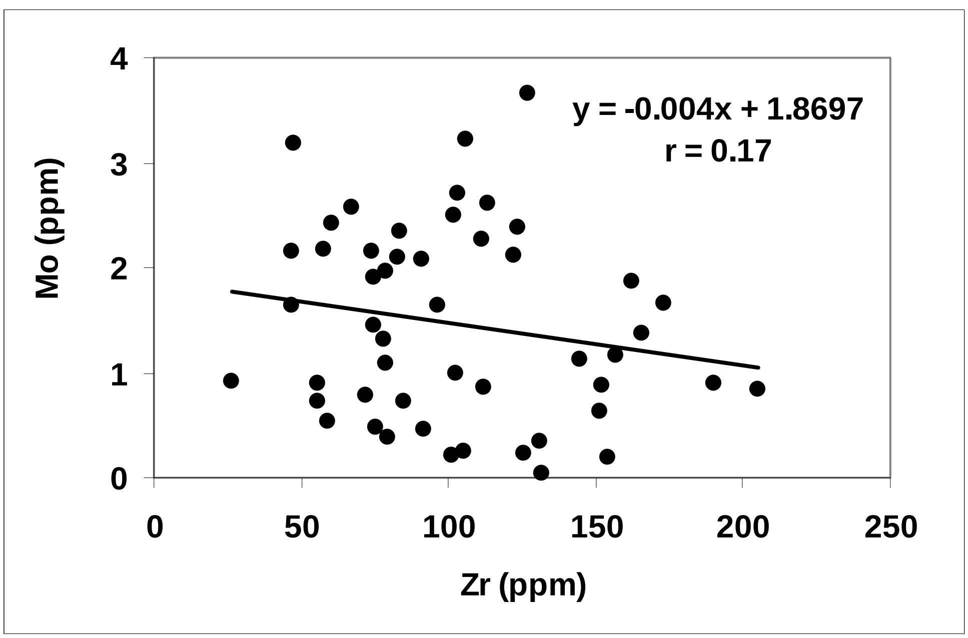
Fig. 9



2
1.6
1.2
0.8
0.4
0
$La_{(N)}/Yb_{(N)}$ ratio
200
160
120
80
40
0
Hf (ppm)
10000
8000
6000
4000
2000
0
Zr (ppm)
20
16
12
8
4
0
Al (wt%)
0
5
10
15
20
25
30
35
40
45
50
Calendar age (ka)

Fig. 10


y = 0.0049x - 2.4026
r = 0.98
Mo (ppm)
Zr (ppm)
0
10
20
30
40
0
2000
4000
6000
8000
10000


y = -0.004x + 1.8697
r = 0.17
Mo (ppm)
Zr (ppm)
0
1
2
3
4
0
50
100
150
200
250