350 ka Organic $\delta^{13}C$ record of the monsoon variability on the Oman continental margin, Arabian Sea

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The stable isotope compositions of sedimentary organic carbon and content of organic carbon for sediment cores recovered at two sites (sites 724C and 725C) during Ocean Drilling Program (ODP) Leg. 117 on the Oman continental margin are used to document variability of the monsoon winds for the past 350 ka. Although both sites have a mean $\delta^{13}C$ value of $-20.1‰$, three zones depleted in ^{13}C are observable at site 724C during isotope stages 3, 8 and 10, while only one zone is recognizable at site 725C. Increased coastal upwelling during isotope stage 3 owing to intense SW monsoon winds resulted in higher concentration of CO_2 in the water column causing the formation of organic matter that was depleted in ^{13}C. The other two zones deposited during oxygen isotope stages 8 and 10, which are also characterized by low values of organic carbon, nitrogen and C/N ratios, could be attributed to the dilution by terrestrial material derived from paleosol by transported by northwesterlies. Because of utilization of ^{13}C enriched dissolved CO_2 during the last glacial maximum Holocene sedimentary organic materials are depleted in ^{13}C relative to the the fomer. The content of residues organic carbon (ROC) is higher at site 724C (with an average of $2.3 \pm 1.2\%$) relative to site 725C, which averages to $0.9 \pm 0.4\%$ probably because of differences in the degree of preservation. Organic material deposited at site 725C has undergone more degradation relative to site 724C as reflected by a systematic downcore decrease in ^{13}C resulting from a loss of ^{13}C enriched organic compounds. Owing to lack of good chronology at site 725C, a zone that is characterized by low $\delta^{13}C$ values it could not be correlated with the other three zones observed at Site 724C.

1. Introduction

A large part of the Indian Ocean and its surrounding continents are affected by seasonally changing monsoon winds. The monsoon wind regime is inferred to have been established during the middle Miocene (Leclaire 1974; Kroon *et al* 1991). From the geological records of mineralogy, contents of organic carbon and micro-fossils, it has been found that the intensity of the monsoon winds has changed considerably since its establishment (Van Campo *et al* 1982; Fontugne and Duplessy 1986; Prell and Van Campo 1986; Sarkar *et al* 1990; Clemens and Prell 1991; Sirocko *et al* 1993; Naidu and Malmgren 1996; Schulz *et al* 1998). Documentation of variations in the intensity of the monsoon winds through the use of temporal variations in the abundance of foraminifer species indicative of upwelling, pollen grain and other geological records, has demonstrated that the southwest (SW) monsoon winds were stronger during interglacial periods, while northeast (NE) monsoon winds were stronger during glacial periods (Duplessy 1982; Van Campo *et al* 1982; Prell and Van Campo 1986; Sarkar *et al* 1990; Krissek and Clemens 1991; Naidu and Malmgren 1996; Schulz *et al* 1998). Variability in the monsoon winds has been found to correlate well with Greenland ice records, with stronger SW monsoons corresponding to a milder environmental conditions in Greenland (Schutz *et al* 1998). Furthermore, various records indicated intensification of monsoon winds at a frequency of 23 kyr (Steens *et al* 1991). Variations in the strength of monsoon winds between the SW and NE directions imply variations in the rate of input of terrestrial material and primary productivity. This is because enhanced upwelling, which causes high nutrient concentrations in the photic zone, is currently intense

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during the SW monsoon (Wyrtki 1973; Prell and Streeter 1982). Also precipitation is considered to have been higher during interglacial periods (Holmgren and Karlén 1998), and it has been inferred to be associated with the stronger SW monsoon winds in the Indian Ocean region (interglacial periods) (Peterson *et al* 1979; Kolla *et al* 1981a,b; Pokras and Mix 1985), thus affecting terrestrial vegetation cover. If indeed there was a significant difference in the intensity of the monsoon winds between NE and SW, then it is likely that such changes were preserved/recorded in marine and continental sediments. One way of finding out whether such changes were recorded in the sediments is through the use of productivity indicators and the stable isotopes of organic carbon.

Although many workers have addressed the issue of monsoonal variability since its establishment through the use of abundance of benthic and planktonic foraminifera, pollen and spores, clay minerals, and stable isotopes of inorganic carbon and oxygen (e.g. Kolla and Biscaye 1977; Kolla *et al* 1981a,b; Van Campo 1986 1991; Sarkar *et al* 1990; Kroon *et al* 1991; Zahn and Pedersen 1991; Sirocko *et al* 1993; Naidu and Malmgren 1996; Schulz *et al* 1998), only a few studies have used stable isotopes of organic carbon and nitrogen in conjunction with other geochemical data to infer Pliocene-Holocene fluctuations in the intensity of monsoon winds (Fontugne and Duplessy 1986; Muzuka *et al* 1991; Sarkar *et al* 1993; Muzuka and Macko 1995). Land-derived organic material in the Arabian Sea is expected to be a good indicator of changes in the intensity of the monsoon winds because the terrestrial source with a mean δ^{13}C value of -27‰ is isotopically different from that of marine source that averages -20‰ (Deines 1980 for review), and any variations in the relative proportion of the two fractions could be easily detected. Therefore, the present work uses multiple parameters indicative of variations in the source strength of marine particles in response to changes in the intensity of the monsoon winds to document monsoonal variability for the past 350 ka. To this end, residual organic carbon (ROC) not corrected for the $CaCO_3$ content, and stable isotope compositions of sedimentary organic carbon for sediments recovered at ODP sites 724C and 725C, Oman Margin are used. Supplementary data used in this study are C/N ratios, contents of organic carbon and nitrogen, the stable oxygen isotope compositions of planktonic foraminifer as reported by Zahn and Pedersen (1991), and weight percentages of aluminium (Al), silica (Si), and titanium (Ti) reported by Shimmield and Mowbray (1991) for a core recovered at site 724C.

2. Methodology

Upper 32 m of the sediment cores collected at two sites (724C and 725C) during the Ocean Drilling Programme in the Arabian Sea (Leg 117) (figure 1) are used in this study. From the δ^{18}O, magnetostratigraphy, and biostratigraphy (Hermelin 1991; Hayashinda and Bloemendal 1991; Zahn and Pedersen 1991) the analysed sediments are estimated to have been deposited over the past 350 ka.

Determination of stable isotope compositions of sedimentary organic carbon, and the contents of ROC was performed on sediments that were initially frozen. However, some of the samples (sediment samples recovered at depth greater than 20 m) thawed during shipment from the vessel to the laboratory. These samples were frozen upon receipt at -10°C until processing, and it is unlikely that thawing of the samples caused a significant isotopic fractionation. Aliquots for the stable isotopes and ROC were dried at 40°C and then acidified using 30% HCl to remove inorganic carbon (carbonate). Because of the high content of carbonate, the acidified samples were washed of all salts and this practice could have led to a loss of some soluble organic compounds. A portion of the dried material was then weighed and combusted in quartz tubes for 1 hour at 850°C in the presence of purified cupric oxide wire and high purity granular copper (Macko 1981). The CO_2 evolved was cryogenically isolated from other combustion products and analysed on V.G. micro-mass PRISM stable isotope ratio mass spectrometer. On the basis of replicate analysis of samples ($n = 10$), the reproducibility in combustion and measurement is within ± 0.2‰. Isotope data are in δ-notation and reported with reference to the V-PDB (Coplen-Tyler 1995). Because of the lack of δ^{18}O for core 725C, correlation between two cores is made based on the general downcore trends of δ^{13}C.

3. Results

The δ^{13}C values at site 724C range from -18.4% to -24.7‰, with an average of -20.1 ± 0.9‰. The δ^{13}C result shows a slight enrichment in ^{13}C during the isotope stage 2 (last glacial maximum) when compared to the isotope stage 1 (figure 2). Also, three zones characterized by low stable isotope values relative to other sections are observable at the following intervals: 4–6 m (event 1), 24–26 m (event 2), and 28.5–30.5 m (event 3) (figure 2). These three intervals depleted in ^{13}C fall within the oxygen isotope stages 3, and at or near the transition from glacial to interglacial periods during isotope stages 8 and 10 (figure 2). Two out of these three zones (zone 2 and 3) are associated with relatively lower values of organic carbon, nitro-, ROC, and C/N ratio (figure 3). Zone 1 is characterized by slightly high C/N ratios and more variable contents of organic carbon nitrogen, and ROC. Moreover, the three zones are associated with relatively high weight percentages of Al, Si, and Ti

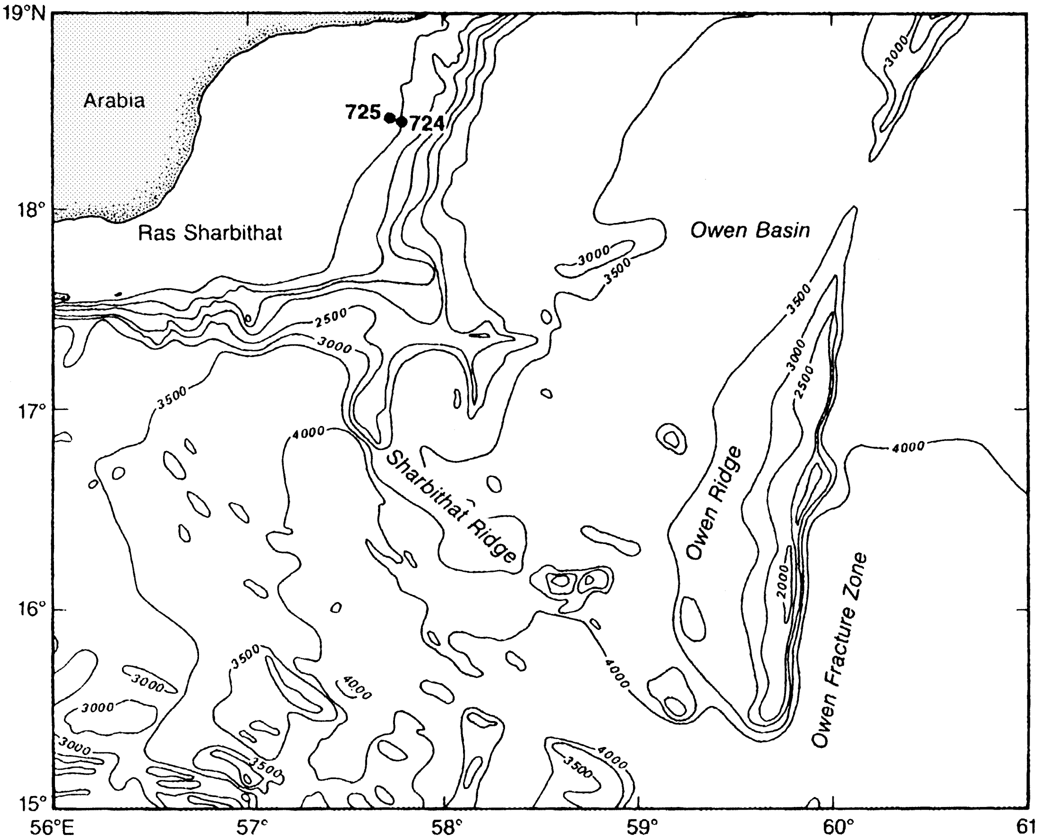

Figure 1. A map showing bathymetry and location of ODP (Ocean Drilling Program) Leg 117, sites 724 and 725. The upper 35 m of the cores recovered at these sites are used in this study. Contours represent water depths in meters.

(figure 4). The ROC range from 0.1% to 6.5% and averages $2.3 \pm 1.2\%$. Last glacial maximum has slightly higher C/N ratio values relative to the Holocene. The contents of organic carbon, nitrogen and to a lesser degree the ROC, are relatively lower at about 15 m (figure 3) but with a lack of corresponding decrease in ^{13}C record.

At site 725, the δ^{13}C values, which show a general decreasing trend downcore (figure 2), range from -17.6‰ to -25.6‰ and averages -20.1 ± 1.3‰. A similar downcore decrease in δ^{13}C values, though to a lesser degree, is observable at site 724C (figure 2). A trend of downcore decrease in δ^{13}C at site 725C is associated with a downcore decrease in the ROC (figure 3). A zone (zone 1) with the lowest stable isotope values at site 725C is observable between 28 and 31 m (figure 2) and is associated with low contents of ROC, which range from 0.2% to 2.1% and averages $0.9 \pm 0.4\%$. (figure 3). Zone 1 marked for site 725C is not stratigraphically the same as the one for site 724C.

Low concentrations of Si, Al, and Ti, are observable during interglacial stages, except during isotope stage 3 (figure 4). Furthermore, inter-site comparison shows that site 725C has low content of ROC relative to site 724C (figure 3). When the stratigraphies established using different methods pointed out previously are transformed into absolute values, there is a mismatch of events at the two sites. At site 724C an event that has the lowest stable isotope values is located at about 340 ka, while at site 725C it is located at about 260 ka. Because of this the two zones cannot be correlated.

4. Discussion

The isotopic compositions of organic carbon for sediments deposited in the Arabian Sea during the Pleistocene-Holocene period average -20‰ with few time intervals characterized by low isotope values (figure 2). This mean value, common to both sites is within the range of the Holocene δ^{13}C values reported by Fontugne and Duplessy (1986) for this basin. The observed mean δ^{13}C value may either indicate little influence of terrestrial organic material or at least 50% input of terrestrial material of the C_4 type which has been reported to have a mean isotope value of -12‰ (see Deines 1980; Gearing 1988 for reviews). Since C/N ratio values are not higher than 20, it is unlikely that there is a significant contribution from land in time intervals that are enriched in ^{13}C. Little influence is the most likely factor because the Arabian Sea lacks any major river inflow that may transport a significant quantity of organic matter to the area particularly off the Oman Margin. A major river that has a

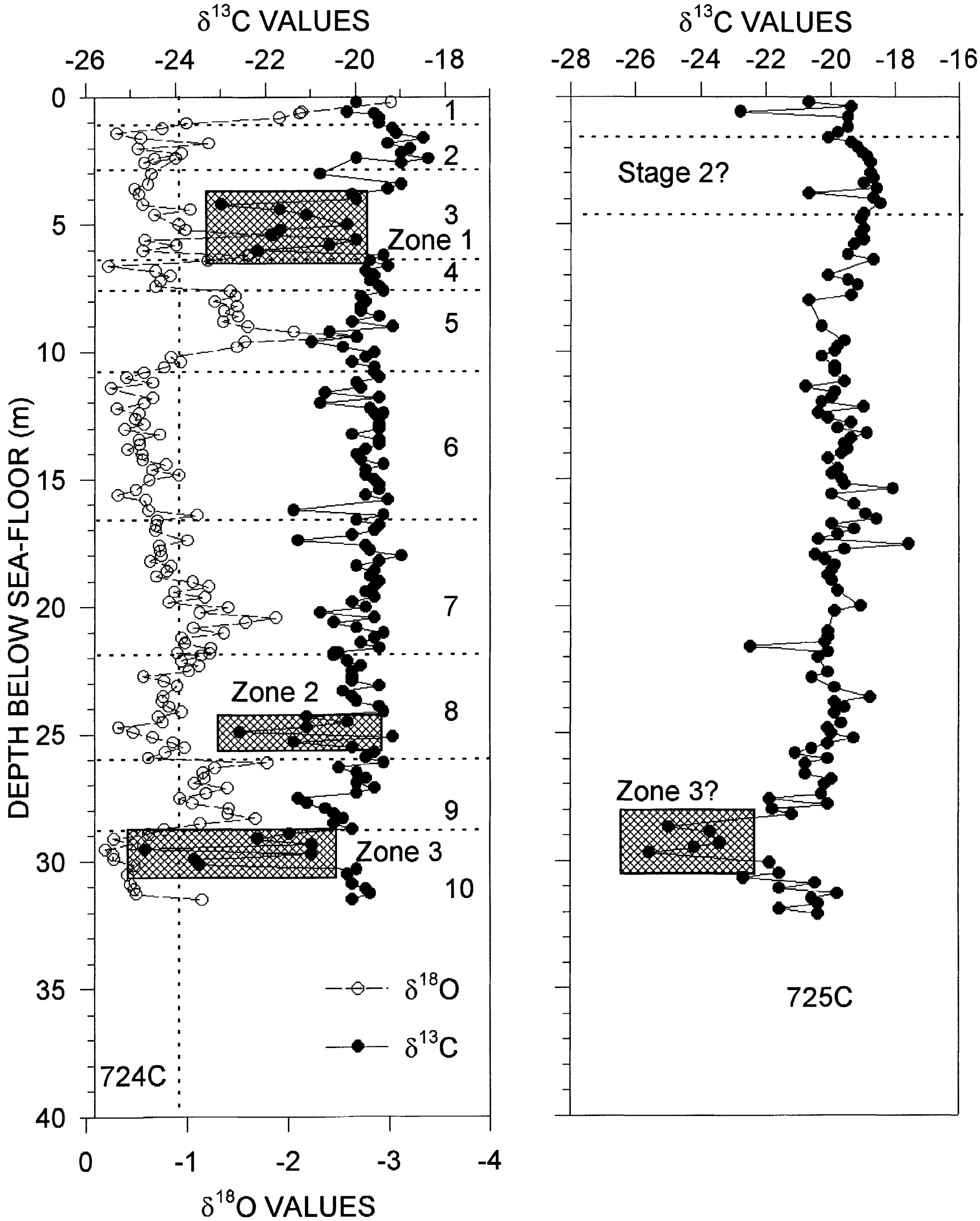

Figure 2. Downcore variations in organic carbon isotope values for the past 350 ka at sites 724C (left) and 725C (right), and oxygen isotope stratigraphy for site 724C (modified after Zahn and Pedersen 1991).

potential of supplying large quantity of particulate material to the area is the Indus, which is located far from the location of the sampling sites. A low input of terrestrial organic matter, mainly through wind transport, is further supported by a relatively low concentration of siliciclastic material during interglacial periods (figure 4). Moreover, elevated percentages in Al, Si, and Ti during isotope stage 2 is in agreement with the observed high concentrations of quartz and clay minerals in the Arabian Sea during glacial period (Kolla and Biscaye 1977; Kolla *et al* 1981a,b).

The last glacial maximum is characterized by enrichment in ^{13}C relative to the Holocene. This is probably due to either elevated input of C_4 plant particulate matter, or utilization bicarbonate during synthesis of organic matter. As pointed out above, high contribution of terrestrial material, particularly C_4 type of organic matter during glacial period, is unlikely. This is because glacial conditions were associated with aridity (drier continental conditions) and hence low availability of terrestrial material for wind transport (Peterson *et al* 1979; Kolla *et al* 1981b;

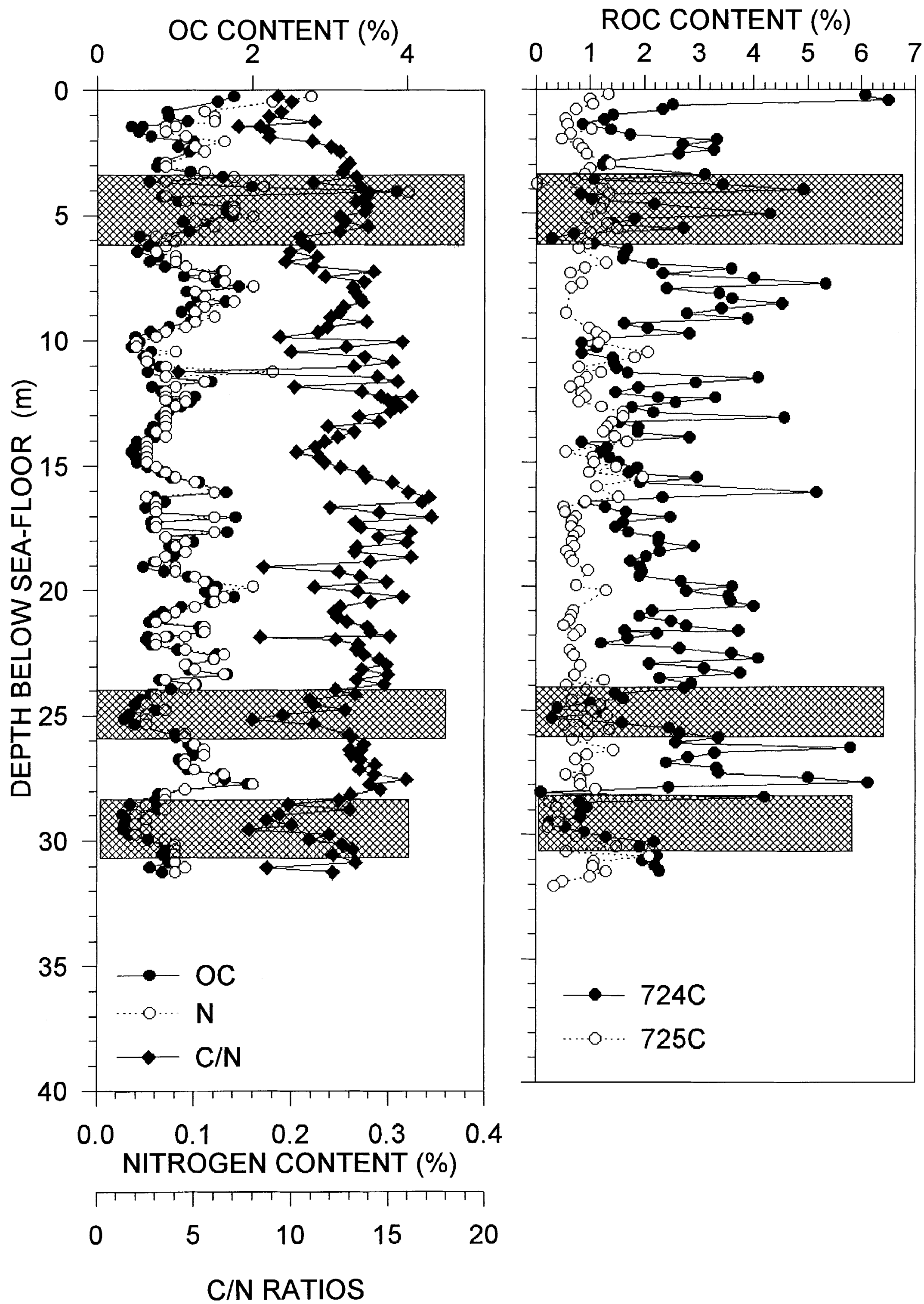

Figure 3. Downcore variations in the contents of organic carbon, nitrogen, and C/N ratios for site 724C (left); and contents of residue organic carbon at sites 724C and 725C (right).

Pokras and Mix 1985). Therefore, ^{13}C enrichment during glacial period could have resulted from utilization of isotopically enriched bicarbonate in response to seasonal variations in temperature associated with shallower continental shelves and low precipitation (Spero *et al* 1997).

The stable isotope compositions of sedimentary organic material collected from the Oman Margin show three zones that are depleted in ^{13}C (figure 2). The first zone represents sediments that were deposited during isotope stage 3, and is associated with a slight increase in the C/N ratios and more variable contents of organic carbon, nitrogen and ROC. The observed depletion in ^{13}C during isotope stage 3 (zone 1) cannot be attributed to input of terrestrial material from the surrounding continents. This is

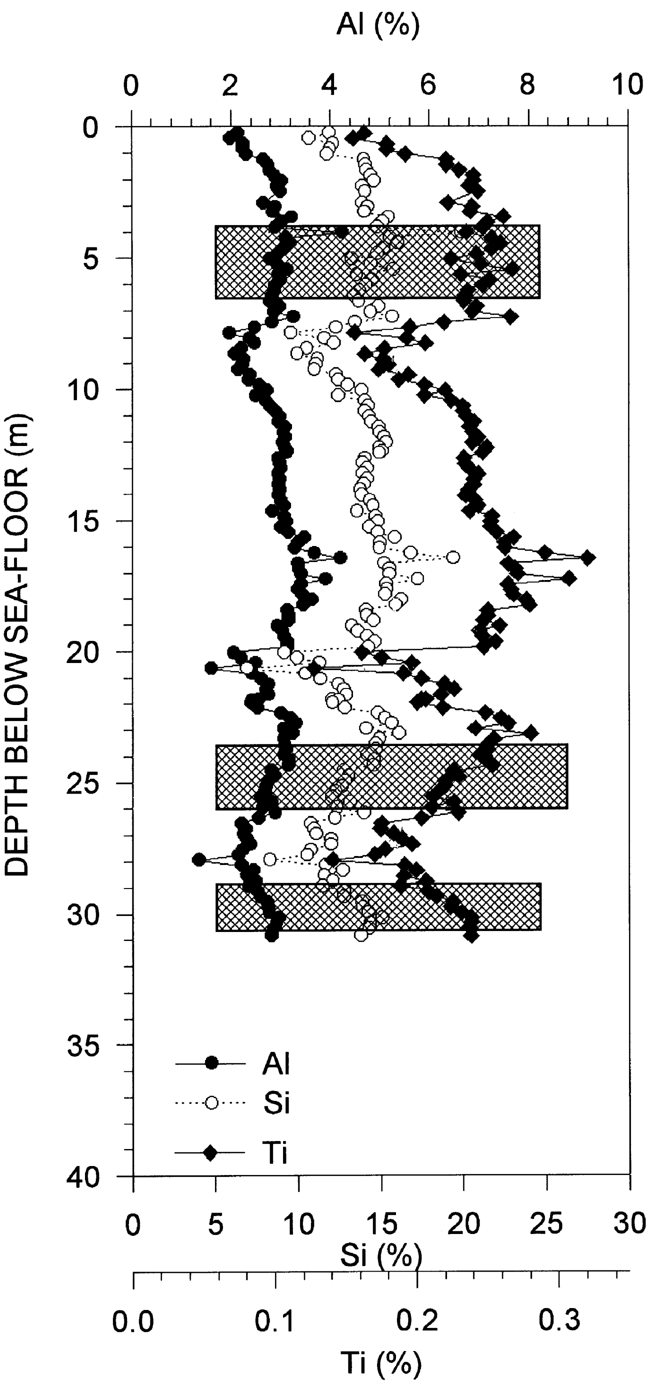

Figure 4. Distribution of weight percentage of Al, Si, and Ti at site 724C for the past 350 ka (modified after Shimmield and Mowbray 1991).

because inferred increase in the intensity of the SW monsoon winds during interglacial periods (Duplessy 1982; Van Campo *et al* 1982; Prell and Van Campo 1986; Sarkar *et al* 1990; Krissek and Clemens 1991; Naidu and Malmgren 1996; Schulz *et al* 1998), could have increased the rate of upwelling and thus increasing rate of CO_2 dissolution. An increase in the rate of CO_2 dissolution implies higher concentration of CO_2 in the euphotic zone. Owing to higher concentration of CO_2 in the photic zone fractionation became high and hence leading to lower isotope values (Francois *et al* 1993; Hinga *et al* 1994). Therefore, incomplete utilization of available CO_2 associated with increase in primary productivity resulting from intense upwelling may be responsible for the observed low stable isotope values during isotope stage 3. A slight increase in the contents of organic carbon and nitrogen could be an additional evidence of increased primary productivity in the area during isotope stage 3.

Slightly higher C/N ratio values associated with low isotope values during isotope stage 3 (figure 3) may be attributed to diagenetic processes. Under normal circumstances, autochthonous marine organic materials have C/N ratios that range from 6 to 10 (Tissot 1984; Meyers 1994). But in case of a significant diagenetic alteration of autochthonous marine OM, C/N ratios are higher than 10. Diagenetic process could have caused preferential loss of nitrogen bearing compounds thus leading to an increase in the C/N ratios.

Other zones that are characterized by low isotope values at site 724C are found at/or near the isotope stages 8 and 10, and are associated with decreases in the contents of organic carbon, nitrogen and C/N ratios (figures 2 and 3). Depleted isotope values during glacial periods or close to glacial-interglacial transition boundaries of isotope stages 8 and 10 (figure 2) indicate that changes from glacial to interglacial conditions were abrupt in agreement with previous reports (Jouzel 1994; Johnsen *et al* 1995). These zones could have resulted from high primary productivity or input of terrestrial material. Primary productivity is unlikely, as the rate of input of nutrients to the photic zone was low owing to weaker SW monsoons. Lower contents of organic carbon and nitrogen could be an indication that primary productivity was low, and the deposited material most likely was refractory derived from continental palaeosol and exposed continental shelf areas. Lower C/N ratios observed in these two zones may indicate the presence of wind-born palaeosol refractory material. This is because C/N ratios for highly degraded material are intermediate between marine and terrestrial materials (Hedges *et al* 1986).

Inferred elevated input of terrestrial organic material during isotope stages 8 and 10 leads to major questions of where this material was derived from, and whether the NE monsoon winds were stronger than the SW monsoon winds. Low contents of organic carbon and nitrogen during intervals of low isotope values (figures 2 and 3) suggest that either primary productivity was low or preservation conditions were poor. As stronger SW monsoon winds at present cause upwelling and thus high primary productivity in the area, low contents of organic carbon and nitrogen further suggest that the strength of the SW monsoon winds was weaker relative to the NE monsoon winds further. Since glacial periods are considered to have been associated with dry conditions (Peterson *et al* 1979; Kolla *et al* 1981a,b; Pokras and Mix 1985), low contents of organic carbon and nitrogen during periods of low stable isotope values may suggest low primary productivity and low availability of material for transport from the surrounding land masses. Also owing to weaker SW monsoon winds during isotope stages 8 and 10, it is unlikely that material deposited in the Arabian Sea during these periods was derived from the African continent. The present day dust input to the western Arabian Sea is generally controlled by the northwesterlies (Shamal winds) whose southward extent is controlled by the strength of SW monsoon. A stronger SW monsoon pushes the winds northwards while weaker SW monsoons allow the influence of the Shamal winds be recorded further southwards, thus leading to enhanced dust input from the Arabian deserts to the western Arabian Sea (Sirocko *et al* 1993). Therefore, a weaker SW monsoon winds during glacial periods could have allowed significant transport of palaeosol organic material by Shamal winds from the adjacent land mass.

Terrestrial sources of organic material during isotope stages 8 and 10 is supported by the contents of the siliciclastic material deposited in the Arabian Sea (figure 4) that is high during periods of low isotope values. Siliciclastic material in the marine environment may originate from land or alteration of basalts, which are well distributed in the ocean particularly near the mid-oceanic ridges. In the Arabian Sea, the bulk sediment composition is controlled by both lithogenic and biogenic input, with virtually no hydrothermal component (Shimmield and Mowbray 1991). Thus, high concentrations of Al, Ti, and Si during the isotope stages 8 and 10 (figure 4) associated with high content of palygorskite (Debrabant *et al* 1991) may suggest that the material actually was derived from the continents.

The δ^{13}C values on the Oman margin show a downcore decrease particularly at site 725C. Such a downcore trend could be a result of

- diagenetic loss of ^{13}C enriched compounds,
- systematic decrease or increase in the relative proportion of the ^{13}C depleted-enriched C_3/C_4 terrestrial material, or
- increase in the utilization of available nutrients (primary productivity).

Relatively high C/N ratios above 10 for the large part of core 724C may be an indication of diagenetic alteration that is likely to have been associated with a loss of ^{13}C enriched compounds such as carbohydrate (Macko *et al* 1994). Although all factors are possible, a downcore decrease in δ^{13}C values associated with a downcore decrease in the contents of ROC may suggest an increase in the nutrient utilization or primary productivity towards the Holocene period.

When the stratigraphies established, using different methods pointed out previously are transformed into absolute values, there is a mismatch of events at the two sites. At site 724C an event that has the lowest stable isotope values is located at about 340 ka, while at site 725C it is located at about 260 ka. This mismatch is most likely due to differences in methods used in establishing chronology. The chronology for the past 400 ka at site 725C has been established based on the first appearance datum of *Emiliania huxleyi* and the last appearance datum of *Pseudoemiliania lacunosa* (Hermelin 1991). At site 724C, the chronology was established based on δ^{18}O and magneto-stratigraphic methods (Hayashida and Bloemendal 1991; Zahn and Pedersen 1991). Furthermore, it has been assumed that the sedimentation rate of 11.1 cm/ka at site 725C has been constant for the past 460 ka. As it has been observed elsewhere (Hillaire-Marcel *et al* 1994; Wu and Hillaire-Marcel 1994; de Vernal *et al* 1996) such assumption is incorrect and because of this the age assignment of events at these two sites is a first-order approximation. Therefore, because of this mis-match factors leading to very low stable isotope compositions relative to other parts of the core at site 725C cannot be positively identified to be that of glacial or interglacial. Also because of the mis-match the data are plotted against depth below seafloor.

5. Conclusion

Sediments deposited over the past 350 ka have preserved climatic changes in the Arabian Sea area. The stable isotopes of sedimentary organic carbon in combination with other geochemical measurements are good indicators of variations in the intensity of the monsoon winds as well as the sources of OM in the Arabian Sea. The Arabian Sea experienced input of terrestrial organic matter during glacial-interglacial transitions of oxygen isotope stages 8, and 10. Lower stable isotope ratios during isotope stage 3 could be attributed to an increase in primary productivity associated with increase in coastal upwelling. Generally, glacial periods particularly stages 8 and 10 were associated with stronger NE monsoon winds, and most of the materials were derived from the north and northeast of the Arabian Sea. Also the stable isotope ratios suggest that climatic conditions change abruptly from glacial to interglacial conditions. Moreover, diagenetic alteration has contributed to the general downcore decrease in the content of ROC at site 725C.

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