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ORGANIC MATTER VARIATIONS IN THE NORTHWESTERN ARABIAN SEA SEDIMENTS

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ABSTRACT: In this study composition of organic matter and its variations in marine sediments of northwestern Arabian sea has been discussed. This paper presents the geochemical analysis of organic carbon content, C/N and δ^{13} stable carbon isotope. The primary objective was to investigate the organic matter in sediments below an upwelling area. Undisturbed sediments (Piston core NIOP-486) of late Pleistocene time was collected during Netherlands Indian Ocean Program (NIOP-1992-93). The core NIOP-486 was raised from a depth of 2077 meters near the Owen ridge. This core records deposition history of last 200,000 years and includes 4 warm and 3 cold periods. The distribution of organic carbon content in studied core shows cyclicity during glacial and interglacial stages. The source of organic matter and variations in glacial/interglacial stages are discussed. C/N ratio and δ^{13} C_{org} isotope results are described to assess the relative proportions of terrestrial and marine organic matter.

KEYWORDS: Organic Carbon, C/N ratio, δ^{13} stable carbon isotope, Terrestrial and Marine organic matter, Sediment core, Glacial/interglacial climate, northwestern Arabian sea, Netherlands Indian Ocean Program (NIOP-92-93).

INTRODUCTION

The distribution of organic carbon in modern marine sediments is very complex. In general, shelf/upper slope sediments are enriched in organic carbon, whereas sediments from the open ocean show lower organic carbon content. Organic carbon is higher in marine sediments accumulating beneath biologically productive water. Generally it reaches concentrations >1%. These organic rich sediments (black shales) are potential petroleum source rocks and have a great geological significance. The difference in both amount and composition of the organic matter occurs because different mechanisms control the accumulation of organic matter in the marine realm. These mechanisms are environment-dependent and are controlled by climatic and oceanographic factors: (i) high plankton productivity in the surface waters (ii) high terrigenous input, (iii) high sedimentation rates which results in rapid burial and prevent oxidation of organic matter (iv) sluggish circulation which causes in extreme cases a layering and stagnation of deeper water masses. To understand the distribution and composition of organic carbon in modern sediments and its changes though time is of special interest for several reasons:

(i) It may give information about changes in surface water productivity and /or changes in oxygen content of deep water. Since surface-water productivity influences the exchange of CO_2 between ocean and atmosphere, changes in bioproductivity may affect the concentration of atmospheric CO_2 which is an important factor controlling the global climate.

(ii) The study of terrigenous organic matter in marine sediments and its changes through time may give information about the climate evolution of the surrounding continents.

(iii) The investigation of the quality and quantity of organic matter in marine sediments may yield depositional models, which may help to explain the formation of fossil organic carbon rich sediments and sedimentary rocks (i.e.; black shales). Since these black shales are major petroleum source rocks,understanding their formation has not only a scientific value, but may also be of interest for petroleum prospects.

Organic matter in marine sediments may be of either marine or terrestrial origin. Land-derived organic matter includes recently biosynthesized plant debris and dissolved humic substances (Thurman, 1985, Hedges *et al.*, 1986). Marine derived organic matter is formed by phytoplankton which are particularly rich in proteins. In offshore areas, marine sediments indicate that the predominant organic carbon source is plankton (Degens, 1969; Hedges and Parker, 1976). In certain marginal areas of oceans, terrigenous carbon may contribute a significant proportion of the carbon fraction of the sediments (Lee and Wakeham, 1988). A number of chemical parameters have been used to distinguish between marine and terrestrial organic matter. These are C/N, C/H, Br/ C_{org} (Bordovsky, 1965; Pocklington and Leonard, 1979; Mayer *et al.*, 1981) and δ^{13} C, δ^{15} N stable isotope ratios (Cline and Kaplan, 1975; Sweeny and Kaplan, 1980).

In the northwestern Arabian sea sediments it has been shown that the historical changes in the intensity of monsoon winds have affected the distribution and quantity of organic carbon (Khan, 1989; Reichart *et al.*, 1997). The amount of organic matter produced over time should be related to the development of regional upwelling, which depends directly on the intensity of monsoon winds in the northwestern Arabian sea. During warm (interglacial) periods, southwest monsoon winds were stronger, resulting in enhanced upwelling whereas upwelling tended to be week during cold (glacial) events (Prell and Curry, 1981; Prell and Kutzbach, 1987; Khan, 1989). As a result of stronger SW monsoon upwelling increased paleoproductivity and organic carbon accumulation occurs in interglacial stages whereas low bio-productivity and lower organic carbon occurs in glacial stages (Khan, 1995). During glacial stages increased terrigenous input (Khan and Price, 1991) may contribute significant land derived organic carbon. In order to investigate the variations in organic matter composition during glacial and interglacial time in the northwestern Arabian sea a sediment core (NIOP-486) collected during Netherlands Indian Ocean Program (NIOP-92-93) has been analysed.

Geological Setting:

The sediment core site is located on the Owen ridge in the northwestern Arabian sea (Fig. 1) The Owen ridge is an asymmetric, northeast-trending ridge near the Owen Fracture Zone, and extends to about 20°N, 61´E, where it merges with the Murray Ridge. The western flank of the ridge dips more gently (about 4°) and merges in to the Owen basin at about 3500 m. The ridge dips steeply (about 15°) to the east, where it abuts the Indus Fan at about 4000 m. The crest of the ridge varies from 1900 m to 2100 m below

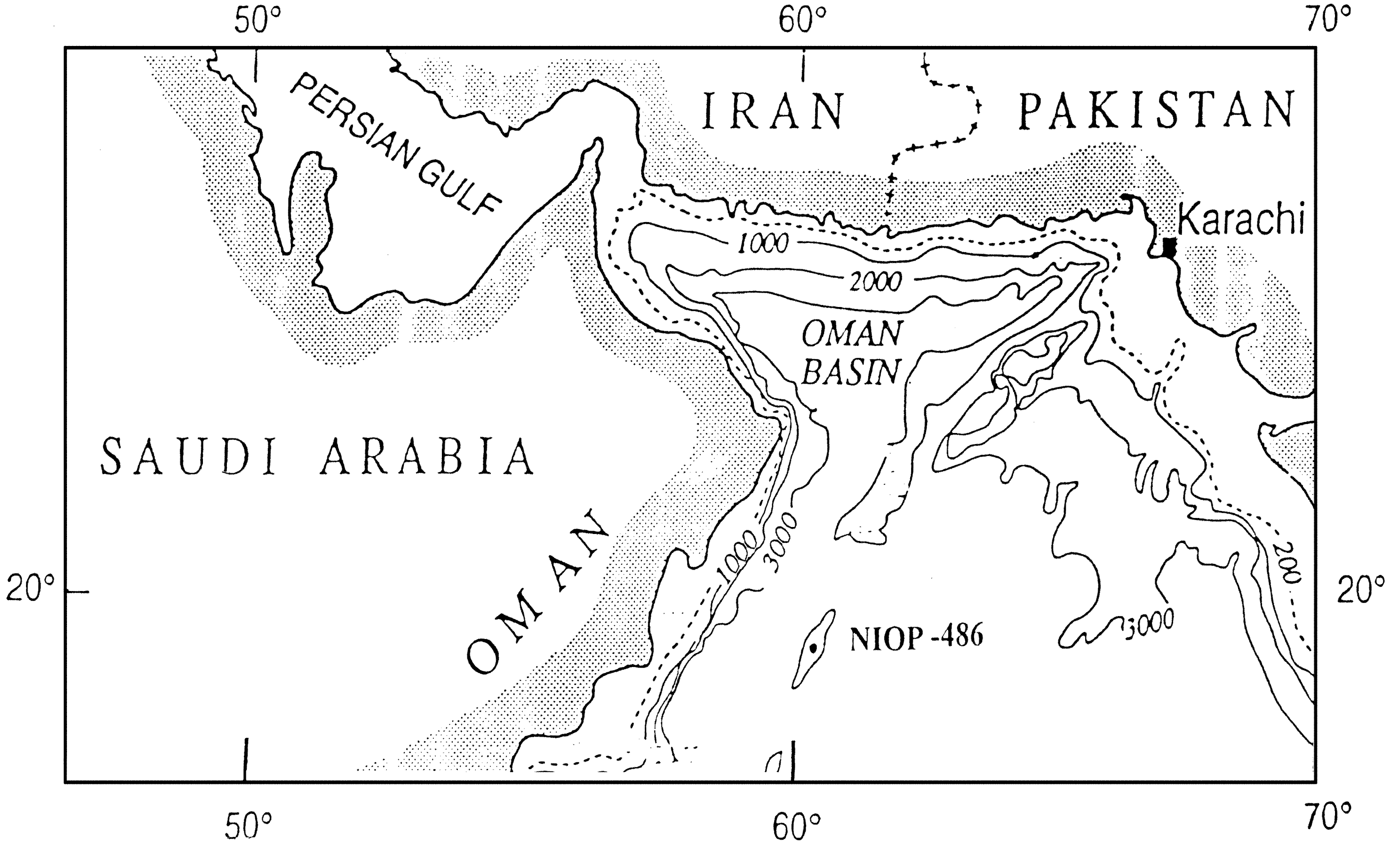

Fig. 1. Location map of core NIOP-486 collected during Netherlands Indian Ocean Program (1992-93).

sea level and has thick, smooth and subhoriozontal pelagic sediments. The origin of the Owen ridge and the age of the underlying basement rocks are associated with both the early separation of Madagascar and India and to the middle Tertiary reorganisation of seafloor spreading in the Indian Ocean. Since the Owen Fracture Zone has been the major transform between spreading centers to the north and south of the Arabian margin, the plate geometry places the Owen ridge and Owen basin on the passive Arabian margin and assigns them a Jurassic age. However, the DSDP drilling in the area has questioned the age and nature of the basement underlying the Owen ridge and basin. The uplift of the Owen ridge has been attributed to the compression along the Owen Fracture Zone caused by changes in spreading direction associated with the continued collision of India and Asia and the opening of the Gulf of Aden (Whitmarsh, 1979). Following its uplift above the reach of turbidite deposition, the ridge crest has accumulated predominantly pelagic, carbonate rich sediments during the late Neogene (Whitmarsh *et al.*, 1974; Prell, 1984).

Oceanographic Setting:

The Arabian sea is characterized by variable seasonal winds due to monsoon climate. This brings large changes in oceanography. Southwesterly winds associated with the summer monsoon cause upwelling of nutrient rich water along the coast of Arabia. These nutrient rich water sustain high levels of oceanic productivity. In summer months the surface productivity, up to 400 gC/m^2/yr making northwestern Arabian sea one of the worlds most productive areas (Kabanova, 1968; Codispoti, 1991). During the winter monsoon productivity is generally low. The Arabian sea oxygen minimum zone (OMZ) is one of the most pronounced low oxygen environments in the open ocean (Swallow, 1984 You and Tomczak, 1993). High primary productivity and limited ventilation of the thermocline leads to an intense Oxygen Minimum Zone. In the OMZ oxygen concentrations reaches values < 0.05ml/l (Van Bennekom and Heihle, 1994). It has been shown that the OMZ has varied considerably in the past (Hermelin, 1991; Ten Kate *et al.* 1991; Altabet *et al.* 1995; Reichart *et al.* 1997).

MATERIALS AND METHOD

The piston core described in this study was recovered during the Netherlands Indian Ocean Program (1992-1993). The location of the sediment core is shown in Fig. 1. The core site NIOP-486 is located near the crest of the Owen ridge. A long piston core (10m) NIOP-486 raised from the northwestern part of the Arabian sea (19°.09´.11¨N, 60°.37´.0¨E water depth 2077m). The sediments consists of hemipelagic foram bearing mud. The colour of the sediments ranges from light olive gray, gray and dark greenish gray (7,5y 5/2, 5/3,6/2 and 10Gy, 7/1 6/2).

Analytical methods:

The sediments were sampled at 10 cm interval. The sediments were weighed before and after drying at 80° C for 24 hours. The dried sediment sample was homogenised in an agate mortar. For the analysis of organic carbon and isotopic composition of organic carbon δ^{13} C 1 gram of dry sediment was weighed in centrifuge tube. Carbonate was dissolved in 1 M HCl by mechanical shaking during 12 hours after which the samples were rinsed in demineralised water in order to remove $CaCl_2$ and subsequently dried. Volumetrically the organic carbon content was determined, followed dry oxidation with CuO at 900°C in a closed circulation system at 0.2 atm oxygen. The released CO_2 gas was cryogenically separated from the other gases. The δ^{13} C was measured with a VG SIRA 24 mass spectrometer with a precision better than 0.1%. The isotope data reported are relative to the PDB standard. Another aliquot of dry sediment was also used to determine carbon organic and N total, using a Carlo Erba NA 1500- CNS analyser with a relative precision of better than 10% for N and better than 3% for organic carbon.

Chronostratigraphy:

A time scale developed for the investigated core is based on correlation with the carbonate stratigraphy of core ODP 722 (16°30´.3 N; 59°47´.8 E; 2027 m water depth). Such an approach has limitations but has been used satisfactorily in other areas where oxygen isotope or carbon dating is unavailable (vander Gasst *et al.*, 1984; Lyle *et al.*, 1988).The core ODP 722 has also been dated by tuning the oxygen isotope signal recorded in the planktonic foraminifera with SPECMAP δ^{18} O (Imbrie *et al.*, 1984) (see Fig. 2). The age model of core NIOP-486 shows depositional history of about 200 k.yrs of Pleistocene period. The chronostratigraphy shows the isotopic stages 1 through 7 of Emiliani (1955). The odd number shows the warm interglacial period and even number denotes the cold glacial times. The age boundaries of different isotopic stages have been placed developed by Martinson *et al.*, (1987).

RESULTS

Total Organic Carbon (TOC):

The total organic carbon (TOC) content in sediments ranges between 0.8% to 4.0%. Down the core sediments show cyclicity during glacial and interglacial isotopic stages (Fig. 3). The lowest organic carbon content of < 0.8% is seen in glacial stage 2. Glacial stages 4 and 6 suggest an increasing trend. The organic carbon content in sediments of these stages is relatively high between 1% and 4%. In stage 4 TOC varies between 1% and 3%. In the upper part TOC value is high i.e. 3.5%. The highest organic carbon content >4.0% occurs in upper part of stage 6. In Holocene stage 1 and interglacial stages 3, 5 and 7 organic carbon content in sediments shows some increase but less conspicuous than glacial stage 6. In interglacial stages 1, 3 and 7 TOC values range between 2% to 3%. In stage 5 TOC value is between 1.5% to 2%.

Composition of Organic Matter:

This study uses C/N and organic carbon δ^{13} stable isotope composition to delineate the origin of organic matter in sediments from northwestern Arabian sea.

(i) C/N:

The proteins are the most important contributors of organic nitrogen (Wakeham *et al.*, 1984) and as such may be used to characterise different types of organic matter. Protein has a C/N atomic ratio of about 3.0, and so organisms rich in protein have a low

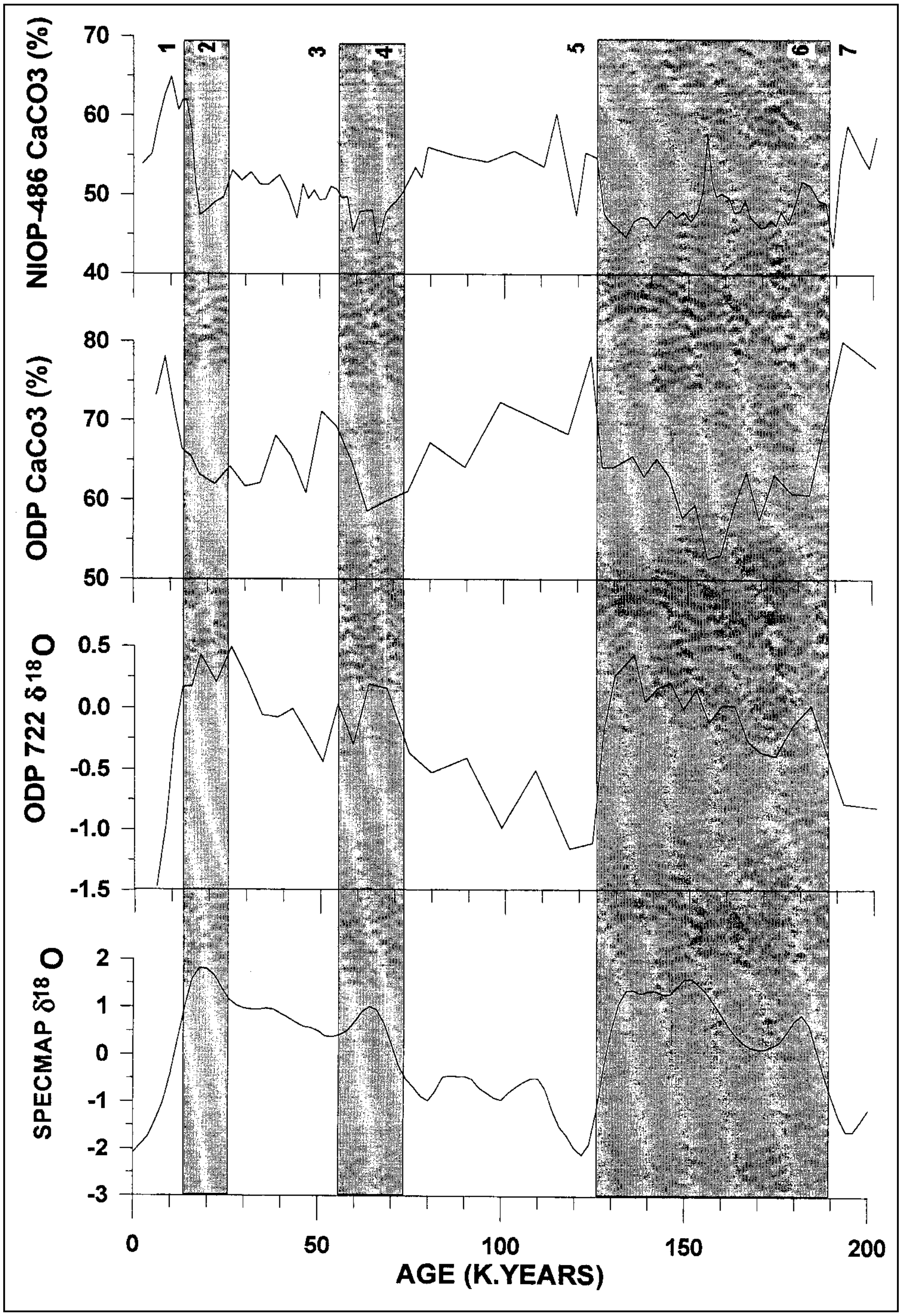

Fig. 2. Specmap, ODP 722 and NIOP-486 Chronostratigraphic Framework.

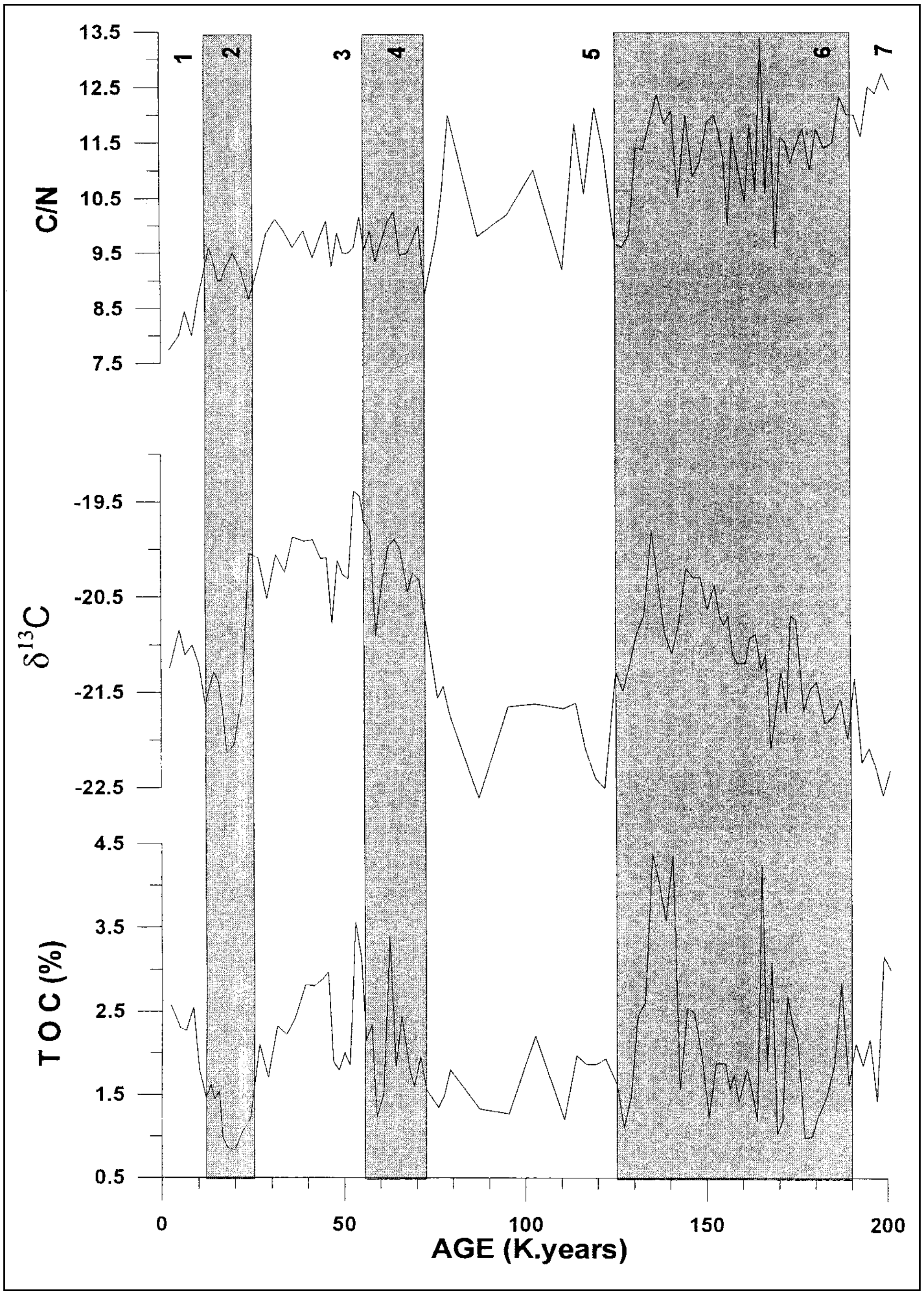

Fig. 3. Total Organic Carbon (T O C %), 13 C and C/N variations in core NIOP-486 from Northwestern Arabian Sea.

C/N ratio, e.g. polychaetes (3.4), fish and fish bones (3.9) and copepods (4.3) (Beers, 1966). In contrast, zooplankton and phytoplankton have higher atomic ratio of 6 to 7 respectively. Benthic organisms are also found to be rich in protein, giving C/N ratios of 4-5 (Bordovsky, 1965). Higher land plants are low in proteins and therefore display very high C/N ratios of 25-30 (Muller, 1977). Hence C/N ratio has been used to characterise the nature of the organic matter within sediments (Badar, 1955; Bordovsky, 1965; Pocklington and Leonard, 1979). The C/N ratio for marine plankton of 6.6 given by Redfield *et al.*, (1963) forms the basis for distinguishing between terrestrial and marine organic matter.

In the present study C/N ratios of Arabian Sea sediments range from 6-13. Variation in C/N ratios with depth in core NIOP-486 is shown in Fig. 3. The pattern of the C/N variations resembles the organic carbon pattern. Highest C/N ratios corresponds to the highest organic carbon content. Average values of C/N in different isotopic stages remains fairly uniform. For example in stages 3 and 4 C/N values fluctuate around 9 and 10. The C/N ratio in upper stage 3 shows upward small decrease. In glacial stage 2 it is 8 and decreases upward. In Holocene sediments C/N ratio is about 6. In lower part of the core C/N ratio is relatively higher. C/N ratios (11-12) are seen in stages 5, 6 and 7. For much of the sediments in different isotopic stages C/N values largely fall in the range 9-12. The validity of using C/N ratios in order to identify the sources of organic matter in core NIOP-486 may be in question. For this reason an attempt has been made to show the nature of the organic matter and its relative variation with time using δ^{13} C stable carbon isotope variations. This is presented in the following section.

(ii) Carbon Isotopes δ^{13} C:

Carbon isotope composition helps in distinguishing marine and terrestrial organic carbon. The carbon isotopic ratio of organic matter in marine sediments primarily depends on the percentage of continental to marine carbon. Studies have shown that the carbon in terrestrial plants is depleted in δ^{13} C compared to that of marine plants (Sackett and Rankin, 1970; Newman *et al.*, 1973). The value δ^{13} C of typical terrestrial organic carbon is known to lie within the range, -26.0‰ to 28.0‰ (Hunt, 1970; Sackett and Rankin, 1970; Newman *et al.*, 1973; Gearing *et al.*, 1977; Fontugne and Jouaneau, 1981). This is distinct from marine organic matter which shows δ^{13} C values lying in the range of -18.0‰ to -22‰ (Fontugne and Duplessy, 1986). δ^{13} C values of plankton in northern Indian ocean are stated to lie around -20‰ (Fontugne and Duplessy, 1981).

In this study, δ^{13} C results from core NIOP 486 are described. The purpose of this study was to ascertain whether or not land derived organic matter has influenced the organic carbon content of the sediments. During glacial episodes, stronger northerly or northwesterly winds may have transported more land derived organic matter with high terrigenous input from surrounding land masses. Hence, the pattern of the isotopic composition of organic carbon in northwestern Arabian sea sediments during glacial and interglacial times should reflect the relative proportions of marine and terrigenous organic matter. δ^{13} C values of the core TY-486 fall between –20‰ and –23‰ relative to PDB. The variation in δ^{13} C values, although small however appear to show some cyclicity (Fig.3). For instance, an increase in δ^{13} C from -22‰ -21‰ occurs between the Holocene and the last glacial maximum (18,000 years BP). Surface sediment and some other Holocene horizons show δ^{13} C of -21.0‰ which is very similar to that of modern plankton . This implies that organic carbon is of marine origin. In glacial stage 2 and interglacial stage 5 δ^{13} C organic carbon values are relatively more negative i.e. –22‰. The lowest δ^{13} C value (i.e. -22.0‰) occurs in sediments of stages 5 and 7. In stage 5 δ^{13} C values are relatively low (-21.5‰ to -22.5‰) especially in its lower and upper parts. Interglacial stage 3 and glacial stages 4 show more or less similar δ^{13} C values i.e. -20‰. For stages 3 and 4, the δ^{13} C is between -20‰ and -21.0‰. An enrichment of δ^{13} C from values –22‰ to –19.5‰ has also been observed in stage 6.

DISCUSSION

Organic carbon content in core NIOP-486 ranges between 0.8 to 4.0%. This range is similar to most other oceanic sediments associated with high biological productivity in overlying waters (Van Andel; 1964; Listizin 1972; Degens and Ross; 1974; Fontugne and Duplessy, 1986). In core NIOP-486 at depth, in different climatic stages considerable variations, in organic carbon have been noted (Fig. 3). The variation of organic carbon is likely to be associated with SW monsoon upwelling. During warm interglacial periods strong SW monsoon induce upwelling along the Arabian coast (Prell, 1984). Nutrient rich water will be brought to the surface enhancing the primary productivity and contributing organic input to the underlying sediments. Nair *et al.*, (1989) have shown that organic flux from sediment trap is higher during SW monsoon. In this study of northwestern Arabian sea sediments a coherent pattern is seen in organic carbon. This supports the higher organic carbon content in sediments during warm periods of increased productivity. However, certain inconsistencies are also observed for example in stage 6. Concentration of total organic carbon generally remain between 0.8% and 4.0%. A significant enrichment in organic carbon has been noted in sediments of glacial stage 6 deposited between 125 and 180 K.yrs ago. This maximum zone corresponds to maximum C/N ratios and enrichment in δ^{13} C values (Fig. 3).

The C/N ratio and δ^{13} C data of the anlysed sediments of core NIOP—486 from northwestern Arabian sea implying a dominantly marine source. Variations in C/N and δ^{13} C trends shown in figure 3 follow organic carbon pattern and hence appears to be related. The isotopic values falls with in the range of marine plankton (-20‰ and -23‰), but the C/N ratios are much higher than expected for algal organic matter. The increase in the C/N ratio from its normal value of 6.0 to 8.0 or more suggests that organic matter may have land derived organic matter or have undergone selective mechanical sorting or chemical degradation. This is particularly noticeable in stages 5 and 6. Here C/N ratios are about 11-12. The elevated C/N ratios in glacial stages 2, 4 and 6 would normally indicate that land plant derived organic matter is contained in these sediments but several evidence contradict this conclusion. (i)The sediments are biogenous ooze with little clastic input (ii)the surrounding desert has no vegetation to provide organic matter (iii)the isotopic values indicate a marine algal source of organic matter. Meyers (1991) has shown that productivity under limited nitrogen availability may result in elevated C/N ratios. The organic matter produced under such conditions would be lipid-rich and nitrogen poor, there by having higher than expected C/N ratios. Goldman *et al.*, (1979) showed that under nutrient depleted conditions the C/N raios of marine algal matter rise, but they never reach the elevated values characteristic of land plants.

δ^{13} C profile shown in figure 2 though suggests small changes down the core but the values indicate uniform source. δ^{13} C values are between –20‰ and –23‰. These values are consistent with the reported marine derived organic carbon in the Arabian sea (Fontugne and Duplessy, 1981). An increase in δ^{13} C in the Holocene may be a result of productivity change associated with SW monsoons as observed by Fontugne and Duplessy (1986). δ^{13} C values in interglacial stage 5 are slightly lighter (-22‰) than stage 6 (-20‰), and this difference could be due to a minor contribution of isotopically lighter terrestrial organic matter. During last interglacial stage 5 (127,000 years) it has been reported that global sea level was high (Chappel and Shackleton, 1986). The inundation of exposed paleoshelf areas could have transported seaward much land derived material and resulted in lighter δ^{13} C organic carbon. In glacial stage 6 sediments δ^{13} values increases from -22‰ to -20‰. From carbon isotopic studies of Oman margin sediments (Mazuka and *et al.*, 1991) it has been noted that carbon isotope composition of organic matter is more enriched during ice stages 2 and 6. During glacial periods, the areas surrounding the Arabian sea were drier than during interglacial periods (Sirocko, 1989; Khan; 1989; Khan and Price; 1991). According to Mazuka *et al.*, (1991) decrease in precipitation and diminished upwelling of deeper, cooler water may have caused surface water temperature to be variable with season and dissolved carbon dioxide concentrations to be slightly enriched in δ^{13} C from bicarbonate. This will result in organic matter enrichments in δ^{13} C during glacial periods. An increase in carbon isotope values during oxygen isotope stage 2 and 6 is also noted by Zahn and Pedersen, (1991). Thus it is possible that the δ^{13} C enrichment with higher organic carbon content in sediments of isotopic stage 6 and 4 of core NIOP-486 may be a result of glaciation effects as noted in other studies (Mazuka and *et al.*, 1991and Zahn and Pedersen, 1991).

This study of C/N ratios, δ^{13} C data of NIOP-486 core and other studies (Fontugne and Duplessy, 1986; Reichart *et al.*, 1997) show that organic matter in the NW Arabian Sea is predominantly marine in origin. Thus it is believed that organic matter preserved in the sediments of the Arabian sea is derived from the marine source as a result of biological productivity above in the surface water. From the above discussion it can be inferred that increasing trends of C/N ratio can not be solely from land derived organic matter. High C/N values for marine organic matter and delineation of smaller variations in isotopic composition and organic carbon contents needs more detailed analysis.

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