Paleoenvironmental significance of clay mineral assemblages in the southeastern Arabian Sea during last 30 kyr

SIDDHARTHA SANKAR DAS^{1,*}, AJAI K RAI¹, VASEEM AKARAM¹, DHANANJAI VERMA¹,
A C PANDEY², Koushik DUTTA^{3,4} and G V RAVI PRASAD^{3,5}

¹*Department of Earth & Planetary Sciences, University of Allahabad, Allahabad 211 002, India.*

²*Department of Ocean & Atmospheric Studies, University of Allahabad, Allahabad 211 002, India.*

³*Institute of Physics, Bhubaneswar 751 005, India.*

⁴*University of Minnesota, Duluth, MN 55812, USA.*

⁵*University of Georgia, Athens, GA 30602, USA.*

**Corresponding author. e-mail: siddharthasankardas.alld@gmail.com*

A gravity core SK-221 recovered from the southeastern Arabian Sea near Laccadive–Chagos Ridge was examined to identify the sources of detrital clay minerals and to decipher paleoenvironmental changes for the last 30 kyr. The clay mineral assemblages predominantly consist of illite, kaolinite and chlorite with small amounts of smectite. Quartz, feldspar and occasionally gibbsite are the clay-sized non-clay minerals present in the examined section. The detrital clay minerals primarily originated from the hinterland and were supplied to the present site by the numerous small rivers draining western India during preglacial and Holocene periods, and partly by the strong reworking of Indian continental shelf during glacial period. The low values of humidity proxies (kaolinite content, kaolinite to illite and smectite to illite ratios) and better illite crystallinity indicate relatively weak summer monsoon condition that resulted in reduced chemical weathering during glacial period, which was interrupted by a discrete event of winter monsoon intensification at ~20–17 ka. The increased kaolinite content, higher values of humidity indices and poorer illite crystallinity reflect high humidity that resulted in strong hydrolysis activity during the preglacial and Holocene periods. The increased CaCO₃ during above periods also indicates less terrigenous dilution and intensified southwest monsoon-led upwelling which result in higher surface biogenic productivity. The characteristic clay mineral associations broadly suggest dry to semi-drier conditions during Heinrich Events H1, H2, and H3 and also during Younger Dryas. The low values of biogenic carbonate and organic carbon also indicate low productivity associated with weak summer monsoons during Heinrich Events. Abrupt increased humidity was recorded at 15–12.7 ka (Bølling/Allerød Event) sandwiched between two lows of Heinrich Events. Cycles of millennial timescale variations 2300, 1800, 1300 and 1000 yr have been observed from the clay mineralogical data. All the cycles observed in the monsoonal climate appear to be part of global oscillations.

1. Introduction

Clay minerals are common components in most marine sediments, especially those deposited on the

continental margins, where there is a significant terrigenous input (Biscaye 1965). The climate change controls continental weathering rates and runoff, which in turn influences soil formation and

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transport of terrigenous material into the sea. The detrital clay minerals, end product of continental weathering processes, are considered a useful key to the past changes in weathering regime (Chamley 1989, 1997; Thiry 2000; Adatte *et al.* 2002). The sedimentation of detrital clay minerals in marine environment takes place mainly through fluvial and/or eolian pathways. About 95% of terrigenous material in the ocean is conveyed by rivers and is deposited on continental margins (Rea and Leinen 1988). In theory, marine sediments may store a record of the environmental conditions and allow comparing these with changes in oceanographic circulation and with global temperatures also (Dou *et al.* 2010). Presuming that geology and geomorphology of the source region remained fairly stable for the time period in consideration in a tropical region, rainfall seems to be main factor determining the composition of clay minerals in marine sediments (Singer 1984; Thamban *et al.* 2002). Clay mineral composition basically indicates the intensity of weathering, especially the degree of hydrolysis at source region which can be used as paleoclimatic indicator (Chamley 1989; Sirocko and Lange 1991; Thiry 2000; Thamban *et al.* 2002; Zhifei *et al.* 2003; Dou *et al.* 2010). The diagenetic effects on Recent marine clay minerals are considered to be insignificant and therefore the provenance of different clay minerals can be identified (Grim 1968).

Previous studies of clay mineral composition of marine sediments from continental margin of western India were mainly based on surficial sediments and were used to understand the provenance and transport pathways of fine-grained terrigenous sediments (Kolla *et al.* 1981; Chauhan 1994; Kessarkar *et al.* 2003). The general relationship between the lithology and weathering regime of a continental source area was established on global scale by Griffin *et al.* (1968) and Kolla *et al.* (1981) to interpret the clay mineral abundance data from surface sediments of the western Indian Ocean and the Arabian Sea. The clay mineralogy of the gravity core samples collected from the continental margin of India near Goa and Cochin have been studied by Thamban *et al.* (2002) and Kessarkar *et al.* (2003). Naidu and Shankar (1999) discussed the paleomonsoon history of late Quaternary based on a pilot study of sediments from the shallower levels of Laccadive Trough. Pattan *et al.* (2003) discussed the productivity variations in the south eastern Arabian Sea for the last 140 ka by using multiple geochemical proxies.

In this paper, we present a high-resolution record of clay mineral assemblages in a gravity core SK-221 collected from southeastern Arabian Sea near Laccadive–Chagos Ridge (8°7.12' N, 73°16.38' E; water depth 2188 m). The main objectives are: (i) to document temporal variation in quantitative input

of terrigenous material to this site, (ii) to record clay mineral assemblages and sources for the past ~30 kyr, and (iii) to correlate between clay mineral assemblages and paleoenvironmental changes during the last 30 kyr.

2. Materials and methods

2.1 Geological and oceanographic settings

The study area lies in the tropical belt and experiences monsoon climate with seasonally reversing wind patterns and large related changes in precipitation (figure 1). Summer monsoons account for the main part of very high rainfall recorded in this region (>3000 mm), whereas winter monsoons are less important as far as rainfall is concerned (Thamban *et al.* 2002). The Western Ghats mountain range is the major physiographic feature all along western peninsular India. The Deccan Trap basalts are the characteristic rocks in the north of Goa; and Precambrian gneisses, schists and charnockites are the predominant rock types in the region between Goa and Cochin. Several small and medium rivers originate from these regions and drain their detritus into the Arabian Sea. The width of continental shelf in the western margin of India varies widely between ~130 km off Ratnagiri and 60 km off Cochin (Rao and Wagle 1997). The

Figure 1. Location map of the core SK-221 in southeastern Arabian Sea with bathymetric contours. Major rock types are shown for peninsular India (redrawn after Thamban *et al.* 2002).

Arabian Sea circulation is greatly influenced by the strong seasonal monsoon wind patterns. During the southwest monsoon, there is clockwise circulation of wind within the basin and it reverses the direction during northeast monsoon period (Honjo *et al.* 1999). A southerly coastal current was reported below 50 m water depth along Indian coast during southwest monsoon and is replaced by a northerly current during the northeast monsoon (Shetye *et al.* 1990).

2.2 Collection of gravity core SK-221

A 5.5-m long gravity core (SK-221) was collected from southeastern Arabian Sea near Laccadive–Chagos Ridge during *ORV Sagar Kanya* cruise 221. The present study is based on the samples taken from the top 1 m of the core. For the purpose of clay mineralogical study, the samples were collected at 1.0-cm interval from top 52 cm of core and at 3.0-cm interval for the rest. The samples for the organic carbon analysis were taken at 1-cm interval.

2.3 Analytical methods for clay mineralogy and organic carbon

All together, 67 samples were used for clay mineralogical study. To minimize the inaccuracies associated with sample mounting procedures, instrumental settings were kept constant and identical sample preparation and quantification procedures were followed. Oven-dried samples were decalcified with 20% hydrochloric acid. Excess acid was removed by repeated rinsing with distilled water and centrifuging. The dried residue was again treated with 30% hydrogen peroxide to remove organic matter. Particles of <2 μm size were separated by using Stoke's Law and concentrated by centrifuging. Oriented clay slides of each sample were prepared by pipetting clay solution on glass slides and air dried prior to X-ray diffraction (XRD) analysis. The XRD analyses were conducted using a Rigaku X-ray Diffractometer (Ni-filtered Cu K-alpha radiation, 40-kV voltage, and 20-mA intensity). The air dried and ethylene-glycolated (glycolated at 100°C for 1 hour) clay slides were scanned from 5° to 40° 2 θ and 5° to 10° 2 θ , respectively, at 2° 2 θ /min. Slow scan XRD from 24 to 26° 2 θ at 1° 2 θ /min was also carried out for each slide. All minerals were identified (Moore and Reynolds 1997) and their peak areas were obtained using DMS software of peak-fitting function. Relative abundance of different minerals such as illite (10 Å), kaolinite (3.57 Å), chlorite (14 Å), smectite (17 Å), quartz (4.26 Å) and plagioclase

feldspar (3.19 Å) was evaluated semi-quantitatively from their peak areas (Robert and Kennett 1997; Mallison *et al.* 2003). As the 3.57 and 3.54 Å doublets of kaolinite and chlorite did not resolve well in each sample, the 3.57 Å peak area was considered for kaolinite. The 14 Å chlorite peak was used for semi-quantitative study. This peak also gives similar result as the 3.54 Å chlorite peak in the relative abundance study (Biscaye 1965). The clay data were thus reported in relative abundance without correction factors. The 'crystallinity', a measure of the lattice ordering was measured as the half-height width of the illite 10 Å peak. Illite chemistry was assessed from the 5/10 Å peak area ratio. Al-rich illites (muscovite) have values >0.5, whereas Mg- and Fe-rich illites have values <0.15 (Esquevin 1969; Gingele 1996). Kaolinite to illite (K/I) and smectite to illite (Sm/I) ratios were also calculated for each sample.

Organic carbon content of 97 samples from this core was also estimated by titration method suggested by Gaudette *et al.* (1974).

2.4 Age-model from ^{14}C dating

The age model is mainly based on the three AMS ^{14}C dates in planktonic foraminifer samples separated from three depths (0–1, 50–51, and 100–102 cm). Sample preparation was carried out at the Institute of Physics AMS Laboratory, Bhubaneswar, India. The planktonic foraminifer samples were first pretreated with 0.1 N HCl for 10 s to remove surficial secondary calcareous deposits, and cleaned in deionized water with ultrasonication. The dried foram samples were then reacted with 85% H_3PO_4 under vacuum. The liberated CO_2 was cryogenically purified and converted to graphite by H_2 reduction over Fe catalyst, and finally pressed into targets for AMS ^{14}C analysis. The ^{14}C measurements were carried out at AMS facility of the Center for Applied Isotope Studies, University of Georgia, Athens, USA, using a NEC 1.5 SDH-1 0.5MV AMS system. The ^{14}C -ages are reported as BP (years before present) after normalizing for the isotopic fractionation of ^{13}C (Stuiver and Polach 1977). Calibration was done using Calib 6.0 radiocarbon calibration software (Stuiver and Reimer 1993), and Marine04 calibration curve (Hughen *et al.* 2004). Mean $\Delta\text{-R}$ correction value of 138 ± 64 years was used for the study region (Dutta *et al.* 2001; Southon *et al.* 2002).

Both the raw ^{14}C ages and the calibrated ages are given in table 1. The age of the core top (0–1 cm) is 2285 ± 60 cal BP and the 100–102 cm level is 30173 ± 180 cal BP. The average sedimentation rate for the core was ~ 3.6 cm ka^{-1} over the period of study.

Table 1. AMS ^{14}C dates in core SK-221.

Sample	Measured ^{14}C -age (BP)	Calibrated ^{14}C -age (cal BP)
SK-221/GC 0–1 cm	2396 $\pm$ 23	2285 $\pm$ 60
SK-221/GC 50–51 cm	21187 $\pm$ 60	25195 $\pm$ 120
SK-221/GC 100–102 cm	25215 $\pm$ 76	30173 $\pm$ 180

2.5 Spectral analysis methods

To determine the major modes of periodicities in the clay mineral concentrations, we have done spectral analysis of the clay mineral data using the software package SPECTRUM (Schulz and Stattegger 1997). This software determines the Lomb–Skargle periodogram from unevenly spaced time series. We have used *O*fac (over sampling factor) of 6 and Welch I window to determine the power spectrum and significance of the spectral coherence, and the corresponding phases at 90% confidence limit. The normalized clay mineral time-series data were used for the analyses. We have also analysed a composite clay mineral time-series obtained by adding the concentrations of chlorite, illite, kaolinite, smectite, and feldspar, to determine the overall periodicities of the clay minerals.

3. Results

3.1 CaCO_3 and organic carbon

The samples are mainly composed of calcareous clayey silt. Grain-size analyses of the terrigenous fraction show dominance of mud over sand throughout the studied section of the core. The carbonate content of the core SK-221 varied between 66% and 86% (av. 77%) and shows stepwise increasing and decreasing pattern for the studied period. It shows relatively higher contents during the ~ 26 , 22–19, 8–6 ka and thereafter it decreased steadily (figure 2). The total organic carbon content (TOC) varies between 0.07% and 2.4% (average 0.6%) in these samples and show variation pattern almost similar to that of carbonate content. The terrigenous matter content varied between 13% and 33% (average 23%) and has negative correlation with the total carbonate content (figures 2 and 3).

3.2 Clay mineral results

Downcore variations of clay and non-clay mineral assemblages are shown in figure 4. The clay mineral assemblages are dominated by illite (19–51%, av. 34%), kaolinite (14–38%, av. 28%) and chlorite (18–41%, av. 28%), while smectite (av. 5%) is less abundant. Quartz (av. 3.5%), feldspar (1.5%)

Figure 2. Temporal variability of biogenic carbonate, terrigenous matter, organic carbon, illite crystallinity, illite chemistry and ratios of kaolinite to illite and smectite to illite. H1, H2, H3: Heinrich Events. YD: Younger Dryas, B–A: Bølling/Allerød, MIS: Marine isotopic stages.

and occasionally gibbsite are the clay-sized non-clay minerals present in the examined section. The downcore variations of clay minerals discussed in terms of three major units: Preglacial period (~ 30 –24 ka), Glacial period (~ 24 –10 ka) and Holocene (figure 4). Illite content shows higher values during glacial period. A marked fall in illite contents is observed between ~ 20 and 17 ka. It is noteworthy that the illite content decreased in a stepwise manner during glacial stage. Overall kaolinite and chlorite have similar variations and show inverse relationships with the illite contents. They show relatively higher contents at preglacial and Holocene periods. However, the kaolinite, in

Figure 3. Scatter plot between biogenic carbonate and terrigenous matter.

particular, shows a fall in its content at ~28 ka and a rise in its content between 20 and 17 ka. Kaolinite and chlorite show stepwise increasing and decreasing patterns for the studied period. Smectite content shows large down-core fluctuations in the studied section. Non-clay minerals, quartz and feldspar have almost identical variation patterns for the studied period. Both the minerals show slightly higher contents during Holocene and Preglacial periods. Gibbsite, another mineral found in some samples is only relatively high at high kaolinite levels. K/I (kaolinite/illite) and Sm/Il (smectite/illite) ratios, which may be used as humidity indices, show relatively lower values during glacial period than preglacial and Holocene (figure 2) periods. However, they show typically higher values between 20 and 17 ka and a downfall at ~28 ka. The K/I ratio, in particular, shows a stepwise increasing pattern in the Holocene period. Since ~5 ka, these indices show significant fall in their values. The illite 5Å/10Å ratio, an index of illite chemistry, varies between 0.07 and 0.52 (average 0.26). The illite data indicate that the illites are relatively rich in Fe–Mg and close to biotite. The ratio shows relatively lower values during glacial period. It also showed significant higher values at ~20–17 ka and lower values at ~28 ka (figure 2).

The present data when compared to *Millennial scale variability* indicates that the biogenic carbonate either remained low or decreased during ages ~29.5–28, 24.5–23, 16–14.8 and 12.7–11 ka corresponding to Heinrich Events 1, 2 and 3 (H1, H2 and H3) and Younger Dryas Event (YD), respectively (figure 2). These ages also recorded relatively lower organic matter contents. There is no distinctive

change in the above materials recorded during 15–12.7 ka, the Bølling/Allerød (B–A) Event. The clay mineralogical signatures and clay-based humidity indices are also correlated with the above events (figure 4). The illite contributed in higher quantities during the H1 and YD Events, but remained low during H2 and H3 events. The kaolinite remained low during the above events. A spike of kaolinite at ~15–12.7 ka corresponds to the B–A Event. Chlorite contribution is relatively higher during H3, H2 and YD. The smectite contribution remained low only during YD. Humidity index K/I either remained low or decreased during the above events.

3.3 Spectral analysis results of the clay mineral data

Results of spectral analysis of the individual clay mineral time series are shown in figure 5. Most robust periodicities common for most clay minerals are seen at about 2.3, 1.8, 1.3 ka and near 1 ka.

Figure 4. Late Quaternary variations in concentrations of illite, kaolinite, chlorite, smectite, feldspar and quartz. H1, H2, H3: Heinrich Events. YD: Younger Dryas, B–A: Bølling/Allerød, MIS: Marine isotopic stages.

Figure 5. Spectral analysis results of individual clay minerals, mineral ratios and composite clay minerals. Periodicities of significant spectral peaks ($>95\%$) are indicated in kilo-years. Horizontal bars (|—|) indicate 6-dB bandwidth of spectral resolution. No significant periodicities observed for organic carbon in the 1–5 ka band. Spectral analysis of the GRIP $\delta^{18}\text{O}$ time series is also shown which is obtained for the exact same time period and resolution as the clay minerals.

Kaolinite shows a significant ~ 6 ka cycle, but we have ignored the cycles longer than 5 ka, which might result from the secular trend in the time series. Feldspar show very prominent 3.6 ka and 1.3 ka cycles, that are also present in the GRIP $\delta^{18}\text{O}$ record for the same time period.

4. Discussion

Accumulation of CaCO_3 on the sea floor is mainly controlled by the biological productivity in surface water, rate of dissolution through the water column as well as on the sea floor and dilution by terrigenous matter. Calcite compensation depth (CCD) in the Arabian Sea lies below 3500 m water depth (Cullen and Prell 1984) and the present sediment core was retrieved from water depth of 2188 m suggesting that dissolution of CaCO_3 is negligible. Relatively high calcium carbonate content in all the samples presumably reflects high biological production. Specifically higher values of CaCO_3 is obtained during parts of preglacial stage, middle parts of glacial period and Holocene. Some sites in the Indian Ocean show Pacific pattern, where CaCO_3 content is higher during glacials and lower during interglacials (Oba 1969; Naidu 1991), whereas others exhibit Atlantic pattern, where CaCO_3 content is generally higher during inter-

glacials and lower during glacials (Peterson and Prell 1985; Naidu *et al.* 1993; Das *et al.* 2008). Some of the sites show both Atlantic and Pacific patterns (Naidu and Malmgren 1999). High CaCO_3 precipitation during glacial period indicates distinct intensification of winter monsoons which led to higher surface productivity in the Arabian Sea (Rai and Das 2011). Decreased CaCO_3 precipitation during glacial period is linked to the weakened summer monsoon leading to reduced upwelling (Nair *et al.* 1989; Narayana *et al.* 2009). Several studies on (Emeis *et al.* 1995; Spaulding and Oba 1992; Ivanova *et al.* 2003; Pattan *et al.* 2003) the Arabian Sea also observed low productivity during last glacial maximum (LGM). However, Gupta *et al.* (2005) observed that in the eastern Arabian Sea, CaCO_3 content changes do not show a consistent pattern with reference to glacial and interglacial climatic fluctuations. High CaCO_3 during preglacial and Holocene periods are related to less terrigenous dilution and enhanced productivity. Moreover, higher contribution of terrigenous matter and lower CaCO_3 values during the Last Glacial Period (LGP) suggest greater terrigenous dilution. Sea level during the LGP was between 100 and 120 m below the present-day level and the rivers were debouching into the sea far away from the present-day coastline (Narayana *et al.* 2009). This lower sea level also favoured erosion of

exposed continental shelf and shortened distance of transportation of suspended load of rivers to the deep sea at this location. The terrigenous sediments of the southwestern continental margin of India consist predominantly of river-borne clays, whereas aeolian fraction is negligible (Rao and Rao 1995). The terrigenous input decreases gradually from ~ 10 to ~ 2 ka. The low terrigenous input during Holocene, particularly at ~ 5 ka suggested weakening of monsoon intensity and stabilized sea level. Pattan *et al.* (2005) also observed similar results in sediment data of the continental margin of India, southeastern Arabian Sea. The high terrigenous influx during certain ages of the preglacial (MIS 3) stage suggests intensive hydrolysis resulting in enhanced erosion due to increased precipitation. Hence, it appeared that the dilution of CaCO_3 in the southeastern Arabian Sea is caused by the influx of terrigenous materials derived from the nearby hinterland.

Several workers used organic carbon as an indicator for paleoproductivity in the eastern Arabian Sea (Thamban *et al.* 2001; Banakar *et al.* 2005). Based on this data, some of the workers suggested higher productivity during LGP than in Holocene (Rostek *et al.* 1997; Schulte *et al.* 1999; Prabhu and Shankar 2005; Banakar *et al.* 2005) and correlated it with strong winter monsoon winds. But, Gupta *et al.* (2005) suggested that the organic carbon content changes in the eastern Arabian Sea were not consistent with the glacial and interglacial conditions. The present investigation at this site shows almost similar variation pattern of TOC with the carbonate for dominant part of the studied section indicating its variability related to the same factors that controlled biogenic productivity.

4.1 Provenance discrimination of the fine-grained terrigenous sediments

Clay minerals in the marine sediments are derived from two main sources: one is terrigenous detritus, which contains most of the provenance information; another is authigenic process which may be caused by alteration of volcanic sea floor basement or hydrothermalism. The provenance discrimination of clay minerals requires a detailed understanding of potential source areas, as well as constraints on transport media and processes (Chamley 1989; Steinke *et al.* 2008). The potential sediment and its transport pathways in the Arabian Sea indicate several source areas, for example, detrital input from Somalia, aeolian contribution from Arabia and Iran–Makran, detrital input from Indus (Himalayan source), Tapti, and Narmada rivers and weathering of Deccan Traps (Western Ghats source), gneissic rocks, laterites

(Precambrian source of southern India) and submarine weathering of Carlsberg Ridge (Nair *et al.* 1982a, 1982b; Shimmield *et al.* 1990; Sirocko and Lange 1991; Pederson *et al.* 1992; Rao and Rao 1995; Sirocko *et al.* 2000; Kessarkar *et al.* 2003; Pattan *et al.* 2005; Thamban *et al.* 2002; Das *et al.* 2008). The aeolian dust input is an important mechanism of sediment contribution in the southwestern and western Arabian Sea (Debrabant *et al.* 1991; Das *et al.* 2008; and others). The Himalayan source is characterized by well crystalline illite and chlorite that are transported by the Indus River to the northern Arabian Sea. The influence of Indus River is largely restricted to the north of Gulf of Kutch because the macro-tidal currents in these zone act as barriers for long shore sediment transport to the south (Nair *et al.* 1982a, b). The illite crystallinity of the present samples varies between moderate to poor, and the illite chemistry indicates that the illites are slightly Fe–Mg rich. The present observations on the illite crystallinity and chemistry data suggest that illite is mostly derived from the southwestern continental margin of India, where Precambrian crystalline rocks are present and have been lateritized at places. Poorly crystalline illites are formed due to intense hydrolysis in the hinterland source area under warm humid climatic conditions. Chauhan and Gujar (1996) reported high illite and chlorite content in the sediments of the southwestern margin of India and identified their source as the waters of the Ganges–Brahmaputra rivers, transported by the Bay of Bengal and intrusion of the southwestern coast during the northeast monsoon. This observation helps in understanding the admixture of Al-rich illite of hinterland with the slightly high Fe–Mg rich illite from the Bay of Bengal that caused imperfect matching of illite crystallinity and chemistry in the present samples.

The occurrence of smectite, a weathered and erosional product of basic volcanic rocks indicate the Western Ghats source of western and central India. These detritus are drained into the eastern Arabian Sea by Narmada, Tapti and several other small streams originating from the steep slopes of Western Ghats mountain (discharge $\sim 100 \text{ km}^3$ water annually). The Western Ghats mountain range with its elevations up to ~ 1000 m, plays a major role in controlling the rainfall intensity (annual rainfall intensity > 3000 mm) over the west coast of India (Thamban *et al.* 2002). Kolla *et al.* (1976, 1981) also reported that smectite-rich sediments in the deeper Arabian Sea are derived from the Deccan Traps.

The weathering of gneissic rocks (Precambrian source of southern India) provides high amount of kaolinite, poorly crystalline illite and gibbsite. These gneissic rocks are lateritized at places.

Intense chemical weathering and leaching of crystalline rocks under tropical humid climate results in the formation of kaolinite and gibbsite (Chamley 1989; Tripathi and Rajamani 2007; Rajamani *et al.* 2009). Kaolinite, dickite and gibbsite are the most common minerals in the lateritic rocks and soils of southern India (Nair and Mathai 1981). Nair *et al.* (1982a, 1982b) and Rao and Rao (1995) also found high kaolinite content in the surficial sediments of upper continental slope of southwestern India. These observations indicate that kaolinite in the present core samples are derived from hinterland source rocks and supplied by the fluvial and cross-shelf transport processes to the southeastern Arabian Sea. The chlorite content remained moderate throughout the examined section. The chlorite contribution almost similarly matched with the kaolinite contribution indicating similar mechanism of derivation. Several workers also reported chlorite in the surficial sediments along the central and southwestern margin of India (Nair *et al.* 1982a, 1982b; Rao and Rao 1995, Thamban *et al.* 2002).

4.2 Paleoenvironmental implications of clay mineralogy

The recognition of provenance of detrital clay minerals and its transport pathways to the present site suggest that the late Quaternary climatic conditions which existed on the hinterland dominantly controlled the contribution of clay minerals. Although several factors such as fluvial discharges, sea level, monsoon climate and oceanic circulation may influence deposition of clays, ultimately climatic signals are given more importance for their derivation in the present site, because the present site lies in the tropical belt and experience monsoon climate with seasonally reversing wind patterns and related large changes in precipitation. Climate controlled aridity and humidity in the source area and ultimately the composition of clay minerals. Similar rock types undergoing weathering in different climatic conditions could give rise to different clay mineral assemblages (Deepthy and Balakrishnan 2005). It is also the climatic change that leads to erosion reactivation and reworking of the soils in the source region (Thiry 2000). To correlate clay mineralogy with paleoclimate, the ratios of specific clay minerals are used instead of only individual clay minerals. This will reduce the effect of mutual dilution on clay mineral assemblages. The kaolinite to illite ratio is a suitable proxy for paleoclimatic reconstruction, because kaolinite is normally formed by intense chemical weathering under warm and humid climate, whereas illite is commonly formed due to physical weathering under cool and dry climate. But, illite may

have also been derived from alteration of micas under warmer and wetter conditions (Birkeland 1984). The smectite to illite ratio (Sm/I) is also considered equally important to decipher paleoclimate because smectite normally forms due to chemical weathering under a warm and humid climate (Ehrmann *et al.* 2007). During the preglacial period (28–24 ka), climate was warm and southwest monsoon was active. This was well-evidenced by relatively higher values of humidity indices (particularly, K/I ratio). During this period, illite showed poorer crystallinity and was Al-rich. The high biogenic carbonate content during this period also conforms to the intense monsoonal activity. The intensification of monsoon during this period is also reported from the laminated sediments of Pakistan margin (von Rad *et al.* 1999) and western margin sediments of India (Thamban *et al.* 2002). The fall in humidity indices to around ~28 ka may be correlated to the stadial (STADIAL 3.1) condition within the preglacial period (Marine Isotopic Stage 3). Lesser contributions of kaolinite and smectite at this age indicate dry period and less chemical weathering of continental rocks. Slightly better crystalline illite with high Fe–Mg content also conforms to this observation. The carbonate contents also indicate less biogenic productivity and poor monsoonal activities at this time. Marked fall in the humidity indices recorded at ~22 ka may be correlated to the LGM. Based on oxygen isotopic data measured in biogenic fossils, reduction of southwestern monsoon wind strength and low precipitation were reported from the western Arabian Sea and equatorial Indian Ocean, respectively during LGM (Prell *et al.* 1980; Duplessy 1982; Van Campo *et al.* 1982; Tiwari *et al.* 2006; Rai and Das 2011; Gunnell 2001a, 2001b). During this period, products of physical weathering have contributed more. The humidity indices typically show higher values during 20–17 ka. During this period, the illite shows poorer crystallinity and is Al-rich. The kaolinite and smectite content were nearly doubled. Gibbsite is also found during this interval. All these data together indicate that punctuation of summer monsoon led to precipitation during glacial period and release of products of chemical weathering from the continental region. This may also occur due to fall in sea level because this period corresponds to the late stages of Pleistocene glaciations (Duplessy 1982; Fairbanks 1992). The products of chemical weathering that were deposited on to the shelf region at higher sea level got eroded by rivers and were directly deposited into deeper sea. Additional sediment might also be deposited due to sustained fluvial erosion and long distance retreat of the Western Ghats escarpment (Kale and Shejwalkar 2007). The Western Ghats escarpment has uniformly retreated at an average rate

of 1–1.5 km Myr⁻¹ (Gunnell 2001a, 2001b). Several workers suggested that erosionally driven isostatic uplift of Western Ghats is occurring since the Tertiary (Gunnell 2001a, 2001b; Kale and Shejwalkar 2008). Valdiya (2001) suggests that such activity occurs due to neotectonic resurgence. It is well-established that Western Ghats are predominantly a denudational feature (Gunnell and Radhakrishna 2001; Valdiya and Narayana 2007; Campanile *et al.* 2008). However, several studies have also indicated that there is no clear relationship between chemical weathering and uplift (White and Blum 1995; Deepthy and Balakrishnan 2005; others). Clay mineral genesis is related to chemical weathering and hence the clay mineralogy of weathering profiles in the hinterland and its contribution into the sea will not directly depend on uplift. The high biogenic carbonate and organic carbon content during the above period may be linked to the intensification of winter monsoons leading to an increased deep-water nutrient injection into photic zone and higher biogenic productivity (Duplessy 1982; Rostek *et al.* 1997; Cayre and Bard 1999; Singh *et al.* 2006, 2011). Based on planktic foraminiferal data (abundance of *Globigerina bulloides* and the ratio between *G. bulloides* and *G. ruber*), Singh *et al.* (2006, 2011) also recorded increased productivity during the above glacial stage in the eastern and western Arabian Sea and postulated that stronger glacial winter monsoon winds were responsible for the above changes.

During 19 to 16 ka, both relatively high and low values of humidity indices are recorded along with the fluctuations of various clay minerals. The carbonate and organic carbon content also fluctuated rapidly. These parameters together indicate that the strength of the monsoon was alternating between strong and weak. The overall lowest values of the humidity indices are observed between 16 and 10 ka indicating lowest intensity of southwest monsoon in the late glacial period. The illite is formed by degradation of gneissic rocks under conditions of limited precipitation. The moderate to good crystalline Fe–Mg-rich illite also conforms to less hydrolysis at the continental source regions. The terrigenous non-clay mineral (quartz and feldspar) input was very low. Although quartz can be derived from a variety of source rocks under a range of weathering conditions, the drier climatic conditions were more favourable for preservation of feldspar. The higher amount of terrigenous input during this period indicates withdrawal of sea and erosion of sediments that were caught in the continental shelf during high stand of sea. Even towards the end of this drier period, humidity indices were again increasing stepwise indicating variation in intensity of monsoon and late deglaciation.

Holocene is marked by distinct increase in kaolinite contribution and humidity indices, poor illite crystallinity and more of Al-rich illite, indicating summer monsoon intensification. The CaCO₃ data, a parameter of biological productivity of this site also indicate intensification of summer monsoons. Based on different proxies, earlier studies indicated intense southwest monsoon during the early Holocene with a maximum at 9 ka (Prell 1984; Van Campo 1986; Naidu and Malmgren 1996; Gupta *et al.* 2003). Whereas, Thamban *et al.* (2002) and Singh *et al.* (2006) reported low intensity of monsoon during the above period and it became intense only after ~9 ka. During the period, between ~10,400 and 5000 calibrated years before present, the Northern Hemisphere temperatures peaked and the Indian, southeast Asian and African monsoons reached their maximum ‘Holocene Climatic Maximum’ (Gupta 2010). Earlier, Alley *et al.* (1997) and others reported cooling phase at 8.2 ka from Indian subcontinent. However, these changes could not be well-identified in the present study.

Millennial scale variability: Clay mineralogical signatures and clay-based humidity index for the ages ~29.5–28, 24.5–23, 16–14.8 and 12.7–11 ka, which correspond to H3, H2, H1 and YD events, respectively show moderate correlation. The low values of humidity index K/I somewhat shows that either very dry or semi-drier conditions were developed during aforesaid events in this region which resulted in more contribution of products of physical weathering, e.g., illite and chlorite. Mineral kaolinite and occasionally gibbsite which is a product of intense chemical weathering either remained low or decreased in content during the above events. The spike in kaolinite content corresponding to 12.7–15 ka (B–A Event) sandwiched between two lows of Heinrich Events indicates sudden change in climate from dry to warm and humid. The low values of biogenic carbonate and organic carbon also indicates low productivity associated with weak summer monsoons during Heinrich Events. The slightly variable behaviour of clay data may be due to the fact that the responses of clay minerals to the sudden change in climatic conditions are not always instantaneous and the clay mineral assemblage deposited at a particular time is the combined effect of weathering during both older and contemporaneous climate. Cycles of millennial timescale variations have been reported by Sirocko *et al.* (1996) from the southwest monsoon records of the last deglaciation in the Arabian Sea (cycles of 2200, 1785, 1450 and 1150 years). South-American pollen records also show periodicities similar to the cycles of 2300, 1800, 1300 and 1000 years obtained from the clay mineralogical record (Mommersteeg *et al.* 1998; cycles of 2200–2300,

1600–1700, 1400–1500 and 1100–1000 years). Pestiaux *et al.* (1988) reported suborbital periodicities at 4600 and 2300 years in the Indian Ocean hydrography, which were explained as combination tones of the orbital precessional and obliquity cycles, representing an internal but nonlinear response of the monsoon system to solar forcing (Hagelberg *et al.* 1994). A similar 2200-year periodicity was reported from Oman margin (Naidu and Malmgren 1995) and attributed to interactions between oceanic circulation changes and atmospheric ^{14}C variations, which show a 2300-year periodicity (Stuiver *et al.* 1991). Higher frequency periodicities (1800, 1300 and 1000 years) were reported from paleo-temperature variations in the upwelling off west Africa (Eglinton *et al.* 1992) and from vegetation changes in the southeastern United States. Thus, all the cycles observed in the monsoonal climate appear to be part of global oscillations.

5. Conclusions

Variations in the clay mineral assemblages at the core SK-221 during the last 30 kyr indicate the following paleoenvironmental changes in the southeastern Arabian Sea:

- The clay mineral assemblages predominantly consist of illite, kaolinite and chlorite with small amounts of smectite. Illite was the dominant mineral during the glacial period; whereas, kaolinite, chlorite and occasionally gibbsite were found during the preglacial and Holocene periods.
- The clay minerals primarily originated from the hinterland and were supplied by numerous small rivers draining the western India. However, strong reworking and erosion of the Indian continental shelf during glacial period were also partly responsible for their deposition.
- The higher kaolinite/illite, smectite/illite ratios along with poor illite crystallinity indicate strengthened summer monsoon precipitation and weakened winter monsoon during preglacial and Holocene. The high CaCO_3 and organic carbon during preglacial and Holocene periods were also related to enhanced biogenic productivity due to strong summer monsoon. In contrast, lower kaolinite/illite, smectite/illite ratios with good illite crystallinity and decreased CaCO_3 precipitation indicate a strongly intensified winter monsoon and weakened summer monsoon during glacial period.
- Typically, higher clay mineral ratios and Al-rich poorly crystalline illites were recorded during 20–17 ka. The fall of sea level during glacial period led to erosion of sediment from the continental shelf regions that were product of chemical weathering deposited during high sea level and transported to the sea by rivers and wind.
- Clay-based proxies indicated dry to semi-dry conditions during Heinrich Events (H1, H2 and H3) and YD; whereas, the B–A Event was recorded as an abrupt increase in humidity. The above proxies also indicated global monsoonal oscillations in millennial timescale variations of 2300, 1800, 1300 and 1000 years.

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