

# Clay minerals as palaeomonsoon proxies: Evaluation and relevance to the late Quaternary records from SE Arabian Sea

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## Abstract

Palaeoclimatic studies are largely dependant on the quality of records and a prudent use of environmental proxy indicators. In this study, the factors affecting the detrital clay minerals in marine environment and their utility and reliability as palaeoclimatic proxies are evaluated and discussed. Systematic investigations using several sediment cores from the SE Arabian Sea reveal that despite the influence of several complicating factors, variations in clay mineral composition during the late Quaternary are essentially climate dependant. Palaeoclimatic reconstruction based on clay mineral proxies suggests that the intensity of hydrolysis at source regions was relatively weak throughout the last glacial cycle and was minimum during the last glacial maximum (LGM). The early deglacial monsoon intensification started at ~16 ka B.P., which was further reduced during 12.5- 11.5 ka B.P., synchronous with the global and regional climatic fluctuations. Maximum humidity was inferred between 9 and 6 ka B.P. and is comparable with similar records from this region. Regional correlations and consistency among various proxy indicators support the utility of clay minerals as a palaeoclimatic proxy.

## Introduction

It is well known that the climate varies substantially on all time scales and the palaeoclimatic records provide the only opportunity for testing the ability of state-of-the-art models to simulate climatic conditions. Unlike the present day, past global changes inferred from various natural archives indicate that the climate system has experienced rapid, high amplitude variations over large regions, oscillating between fundamentally different modes of operations (Schulz *et al.*, 1998). The reliability and accuracy of the palaeoclimatic reconstructions mainly depends on the quality of database and a discreet use of environmental indicators or proxies. "Proxy" as used in palaeoclimatic terminology is a measurable descriptor which stands in for the desired, but unobservable

environmental variables such as temperature, wind-speed, precipitation, etc. (see Wefer et al. 1999 for an overview). Since the concept of proxy presumes the existence of a target parameter, each proxy is associated with a fundamental set of rules, indicating how the proxy is linked to the target. However, the correlation between target parameter and proxy variable is less than perfect as several poorly understood parameters could interfere and the resultant environmental reconstruction tends to be more qualitative.

Although useful information for reconstructing the climate history is preserved in different kinds of archives on land and ocean, marine sediments provide superior information as sedimentation in the ocean is fairly continuous and chronological assignment is more reliable. Palaeoclimatic investigations employ a wide spectrum of proxies and the existing proxies are continuously evolving and being strengthened (Wefer et al. 1999; Henderson, 2002). Proxies can be classified based on the type of sediment property they describe (biological, sedimentological, geochemical, isotopic, magnetic, etc.) or according to the target parameter they stand for (e.g., temperature, precipitation, wind-strength, productivity, etc.). Although the organic and inorganic constituents of marine sediment are the most widely used proxies in climate research, the terrigenous fractions of the sediments are very useful especially in reconstructing the terrestrial environments (Chamley, 1989; Chauhan, 1999; Thamban et al. 2002).

### **Clay mineral as a palaeoclimatic proxy**

Clay mineral characteristics of the aquatic sediments reflect the prevailing climatic conditions, hydrography, geology and topography of the continental source area (Chamley, 1989). The variations in clay mineral abundance, therefore, are a tool for deciphering the sediment sources and transport vectors to an area. Clay minerals in the marine environment are found to be largely detrital and widely dispersed (Biscaye, 1965). Since the clay minerals are ubiquitous in the marine sediments and their measurement is relatively easier and economic than the other sophisticated techniques, they provide a potential tool for palaeoclimatic reconstruction. Presuming that the geology and geomorphology of the source region remained fairly stable for the time period in consideration in a tropical region, rainfall seems to be the main factor determining the composition of clay minerals in the marine sediments (Singer, 1984; Chamley, 1989; Thamban et al. 2002). Clay mineral composition basically indicates the intensity of weathering, especially the degree of hydrolysis, at source rock regions that can be used as palaeoclimatic indicators (Chamley, 1989).

Diagenetic effects on recent marine clays are considered to be insignificant and therefore the provenance of different clay minerals can be identified (Grim, 1968). Nevertheless a cautious approach is warranted as many other processes act on clay minerals simultaneously or immediately after reaching the marine

environment, suppressing the climate-induced differentiation of clay minerals. These processes include: size sorting of clay minerals during transportation (Gibbs, 1977), flocculation at lower salinities (Grim, 1968), selective deposition of certain clay minerals in relation to organo-mineral interactions (Degens and Ittekkot, 1984), texture-related clay mineral variations (Maldonado and Stanley, 1981), dispersal of clay minerals due to the prevalence of regional and local currents (Kolla et al. 1981; Naidu et al. 1985), redistribution of settled clays during reworking and methods followed for sample preparation and clay mineral quantification (Biscaye, 1965; Pierce and Seigal, 1969). As a consequence of these factors, either selective transportation and deposition of clay minerals or mixing of clay minerals from different sources take place. It is therefore likely that the distribution may not show distinctive variations to differentiate the climatic record. A proper understanding of the provenance, regional oceanography and other factors mentioned above are thus essential before any meaningful climatic interpretation is attempted.

Previous studies of clay mineral composition of marine sediments from the 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 (e.g., Biscaye, 1965; Kolla et al. 1981; Nair et al. 1982 a&b; Chauhan, 1994; Rao and Rao, 1995; Rao and Wagle, 1997, Kessarkar et al. 2003). The objective of the present study is to evaluate the use of clay mineral variations along the southwestern continental margin of India as palaeoclimatic proxies and apply them to reconstruct the late Quaternary monsoon conditions on Indian subcontinent.

## Sample and Techniques

In order to achieve the above, four gravity cores (GC-2, GC-3, GC-4 & GC-5) were carefully selected along the western continental margin of India between Goa and Cochin (Fig. 1). These cores were collected during the sixth cruise of A. A. *Siderenko*, a research vessel chartered by the Department of Ocean Development (DOD), New Delhi. Detailed information like the core location, depth of collection, length of the cores, onboard lithological observations and sub-sampling intervals are given by Thamban et al. (1997). Although all these cores were raised from shallow depths (280-355 m) along the continental slope, they depict diverse depositional settings and reveal contrasting sedimentation patterns (Thamban et al. 1997, Thamban, 1998). Lithology and grain size characteristics of these cores were also discussed elsewhere (Thamban et al. 1997). Age assessment for core GC-2 and GC-4, the bulk carbonate  $^{14}\text{C}$  dates were obtained from Physical Research Laboratory, Ahmedabad and for core GC-5, detailed AMS  $^{14}\text{C}$  dates were obtained from the Leibniz Laboratory of University of Kiel, Germany. The chronological control for core GC-3 was based on the detailed oxygen isotope stratigraphy.

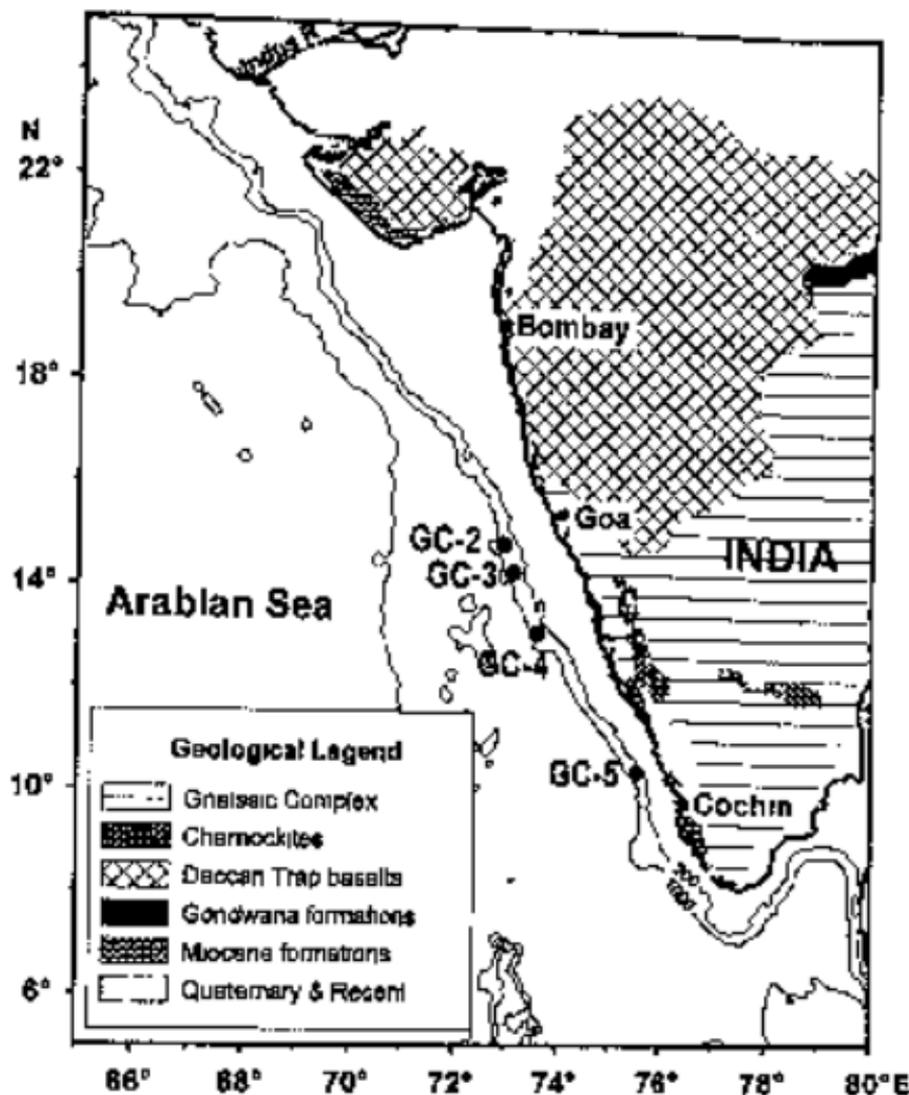


Fig. 1. Location of the sediment cores used in this study and the geological map of the hinterland region

The study area extends along the western Indian margin between Goa and Cochin (Fig. 1). The general geology of the hinterland is well known and consists predominantly of Precambrian gneisses, schists and charnockites, which are lateritised at places (see Thamban et al 2002). North of Goa the characteristic rocks are the Deccan Trap basalt and along the coastal regions of southern India Vaikala and Quilon beds of Miocene age are exposed. Humid tropical climatic

conditions exist in the coastal belt and the rainfall over this region is controlled by the seasonal reversals in monsoon winds, with the summer monsoon (June-August) brings maximum precipitation (~2700 mm; India Meteorological Department). The drainage patterns are characterised by numerous small seasonal rivers and streams, discharging large quantity of water (~95.58 km<sup>3</sup>) annually to the coastal Arabian Sea (Rao, 1979).

To minimise the inaccuracies associated with sample mounting procedures, the instrumental settings were kept constant and the same sample preparation and quantification procedures were followed throughout. The 2  $\mu$ m size clay fraction was separated from the sand-free sediment based on the settling velocity principle (Folk, 1968). The sample was then made free of carbonate and organic matter by treating the clay solution with 5 ml acetic acid and 10 ml of 50% hydrogen peroxide, respectively. The excess acid was removed by washing with distilled water. Oriented slides were then prepared by pipetting 1 ml of the concentrated clay aliquot on glass slides and allowing them to dry in air. X-ray diffraction studies were carried out on the air dried samples from 3° to 22° 2 $\theta$  at 1.2° 2 $\theta$ /minute on a Philips X-ray diffractometer (1840 Model) using nickel-filtered Cu K $\alpha$  radiation, operated at 20 mA and 40 kV. The samples were then glycolated by exposing the slides to ethylene glycol vapours at 100°C for 1 hour and rescanned. In order to differentiate the kaolinite and chlorite peaks, the slides were also scanned from 24° to 26° 2 $\theta$  at 1/2° 2 $\theta$ /minute. The areas of major clay minerals and weighted peak area percentages were calculated following the semi-quantitative method of Biscaye (1965). Crystallinity of illite was measured as the half height width (HHW) of the 10A peak and the illite chemistry was assessed using the ratio of intensity of 5A and 10A peak areas of illite in the X-ray diffractograms (Esquevin, 1969; Gingele, 1996).

## Results

The major clay mineral groups present in all the sediment cores are: smectite (S), illite (I), kaolinite (K) and chlorite (C) (Fig. 2 & 3). Gibbsite (G) is also identified in sediment cores off southwest India (see Fig. 2). The relative abundance of major clay minerals varied significantly both temporally and spatially (Fig. 4-6). Kaolinite and illite are the dominant clay minerals in cores GC-4 and GC-5 from the upper slopes off Cochin and Mangalore (Fig. 4 & 5). Both kaolinite (21-42%) and illite (10-47%) showed large amplitude variations within the cores with an antithetic relationship to each other. Although smectite showed wide variations (7-41%), it did not show any consistent relation with other minerals. Chlorite showed opposite trend to that of kaolinite in both the cores. Prominent reflections of gibbsite are found at intervals of high kaolinite content (see Fig. 3).

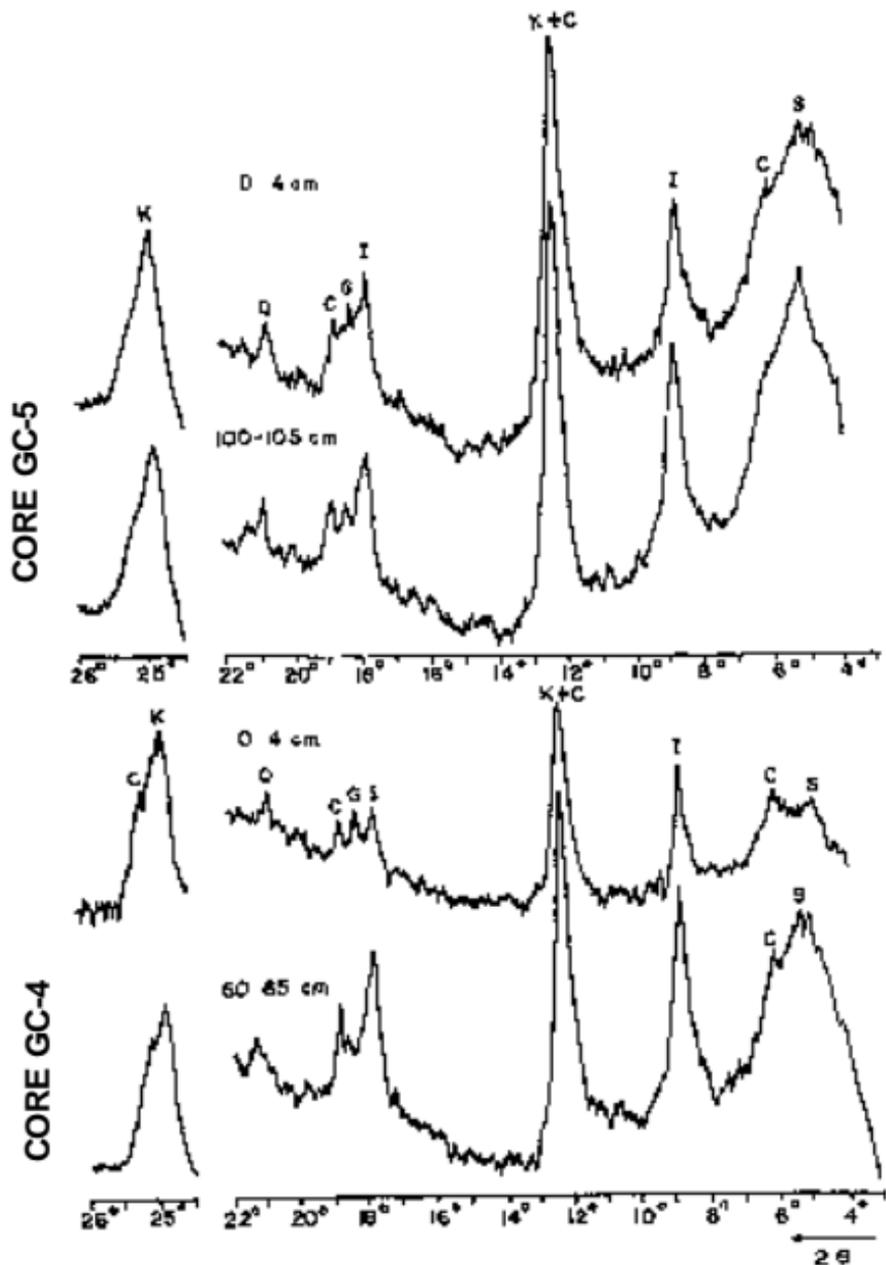


Fig. 2. Representative X ray diffractograms showing major clay minerals at two different depth intervals in cores GC-5 and GC-4 from the upper continental slope of India (southern cores)  
S-smectite; I - illite; K-kaolinite; C - chlorite; G - gibbsite;

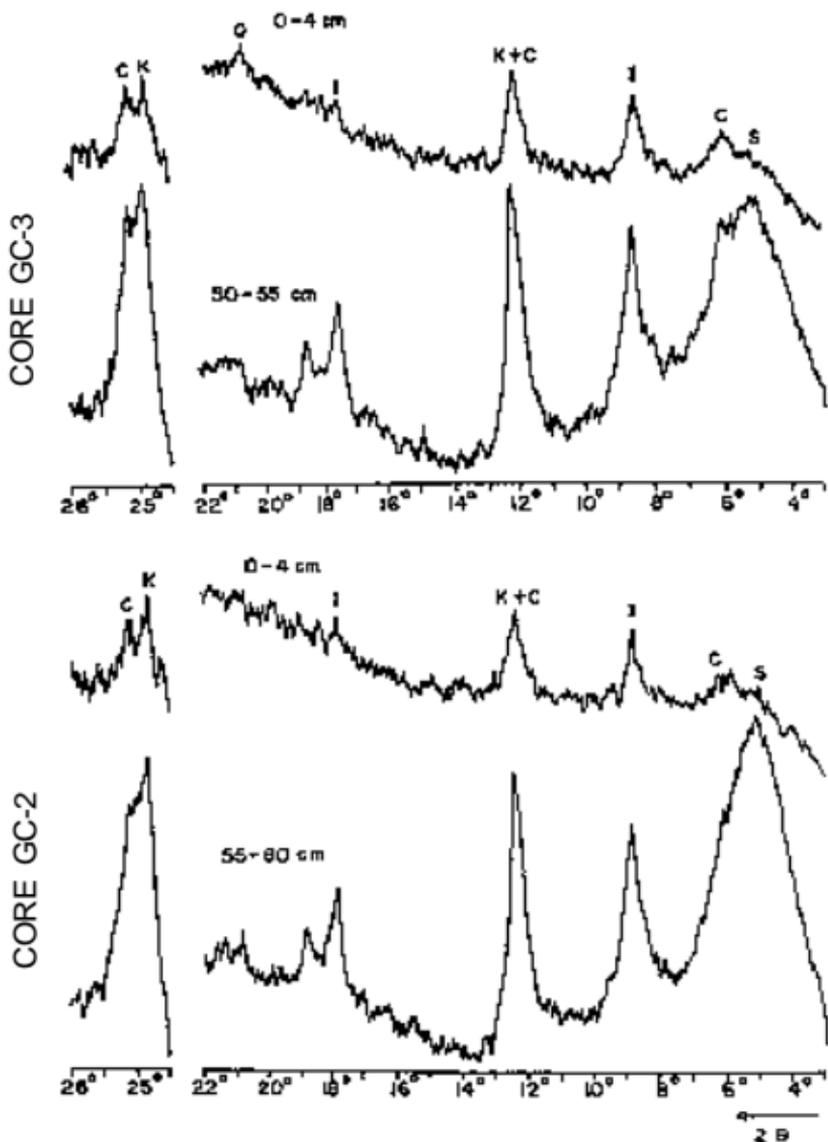


Fig. 3. Representative X-ray diffractograms for cores GC-3 and GC-2 from the topographic highs off Goa (northern cores)

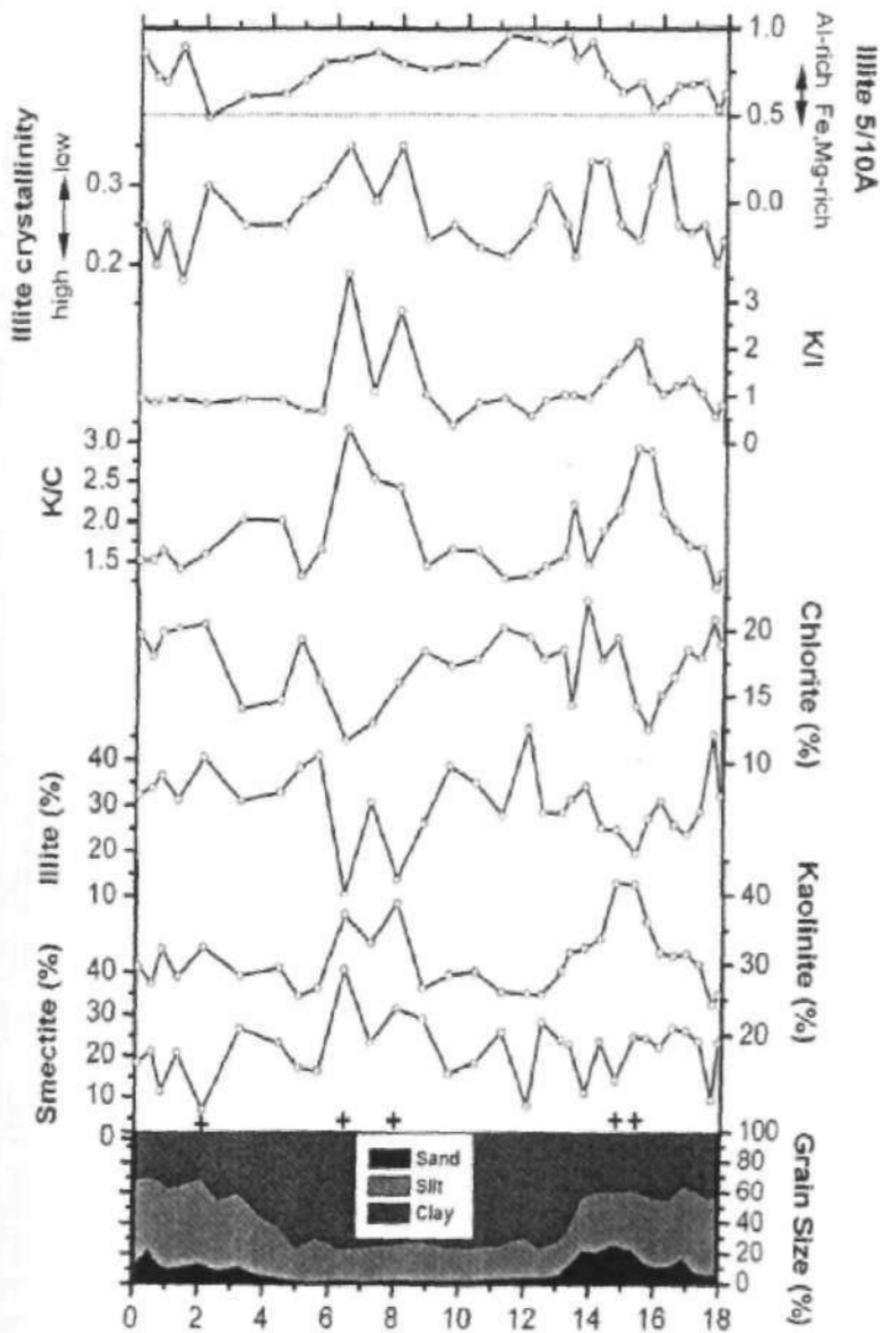


Fig 4 Clay mineral data and grain size parameters for the core GC-5 Intervals showing enhanced gibbsite peaks are shown by + marks. Total length of the core is 333 cm. Age model is based on five AMS  $^{14}\text{C}$  dates (after Thamban et al., 2002).

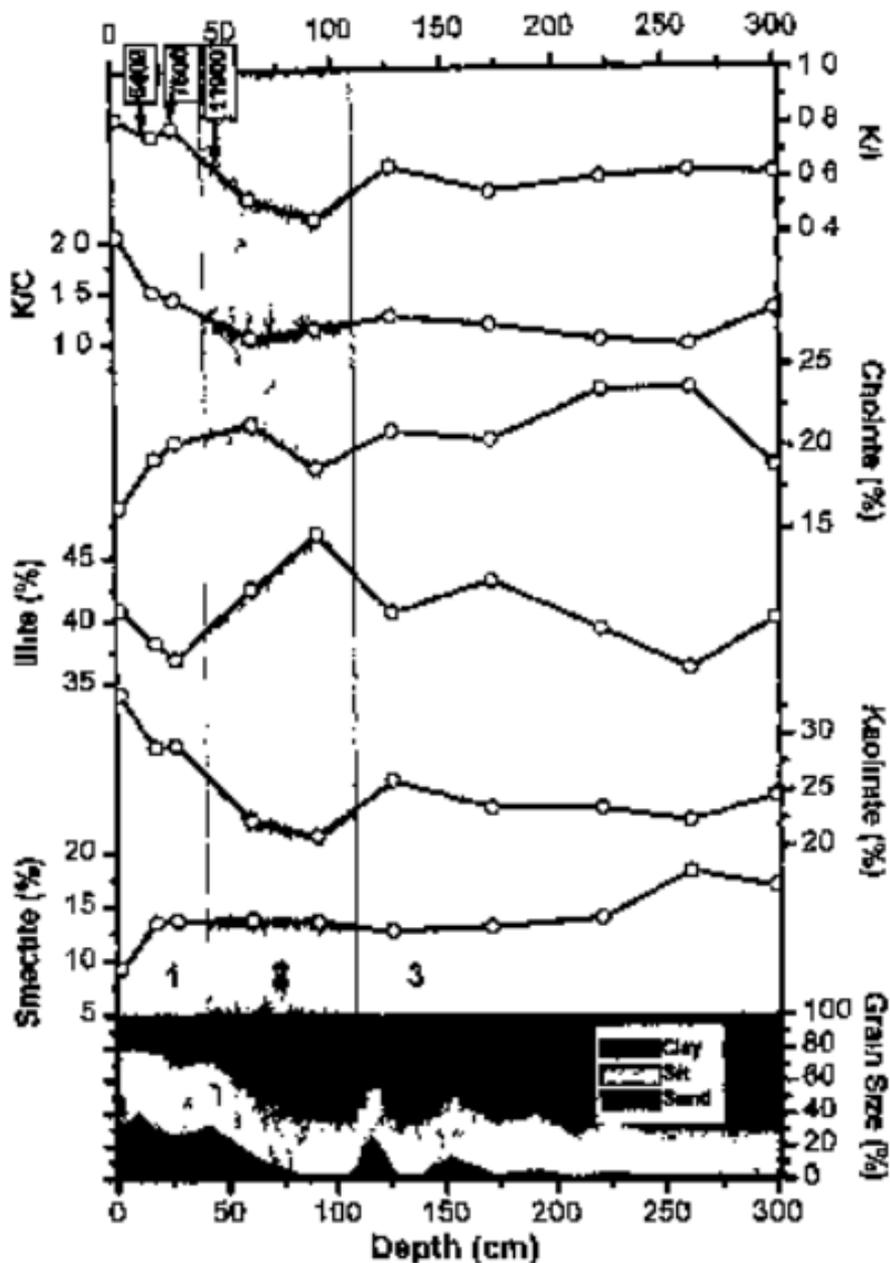


Fig. 5. Clay mineral data and grain size parameters for the core GC-4. Age controls based on bulk carbonate  $^{14}\text{C}$  dates are given by arrows and the shaded interval is the marine isotope stages based on the  $^{14}\text{C}$  dates.

Illite is the most dominant (31-63%) clay mineral in two cores (GC 3 & GC 2) from the topographic highs off Goa (Fig 6 for GC 3 data refer Thamban et al 2002). Chlorite content in the cores varied between 13 and 23% and shows a negative relation with kaolinite. Smectite (6-29%) and kaolinite (6-23%) contents are lower in both the records. Gibbsite peaks are not prominent in these cores and where they present are associated with high kaolinite content.

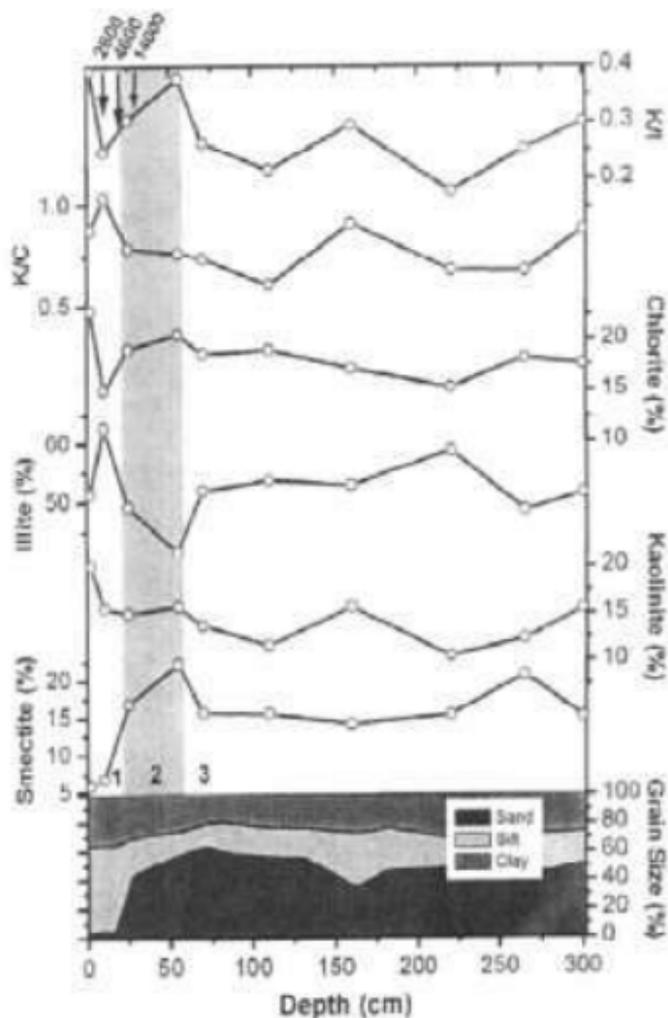


Fig 6 Clay mineral data and grain size parameters for core GC3. The model is based on the three <sup>14</sup>C dates (shown by arrows) on bulk carbonate

## Discussion

Pioneering studies on the modern distribution of the clay minerals along the eastern Arabian Sea (western continental margin of India) revealed the following: a) The Indus river is the major source of illite and chlorite to the eastern Arabian Sea and are basically originated from the physical weathering of Precambrian terrain in the Himalayas (Kolla et al. 1981; Nair et al. 1982 b; Rao and Rao, 1995; Kessarkar et al. 2003); (b) Principal source of smectite to Arabian Sea is the weathering of Deccan Trap basalts under a semi-arid climate and is transported mainly by the Narmada and Tapti river systems (Subramanian, 1980; Naidu et al. 1985; Rao and Rao, 1995; Kessarkar et al. 2003); (c) Kaolinite and gibbsite along the western margin of India have their source in the Precambrian gneissic rocks and laterites of Western Ghats and are formed by the intense chemical weathering under humid, tropical conditions (Nair et al. 1982 b; Rao and Rao, 1995; Kessarkar et al. 2003). Kaolin deposits of the Varkala formation, which outcrop along the southwest coast of India, could act as an additional local source for kaolinite. Although wind-borne particles rich in illite, chlorite and palygorskite are abundant in the surface sediments of northwestern India (Sirocko and Lange, 1991), their accumulation along the western continental margin of India is negligible (Kolla et al. 1981; Rao and Rao, 1995). As the major dust storms of northwestern India has an ENE and NNW direction (Middleton, 1989), significant aeolian input from Thar Desert during present day is also negated.

## Factors controlling the spatial and temporal variations in clay mineral distribution

Illite concentrations in cores from the topographic highs off Goa are significantly higher than in the cores of the upper continental slope off southwest India. This may indicate that either the illite at these locations has different sources or physical processes may have favoured the sorting of these clay minerals leading to high illite in the sediment. High illite contents at the core top samples of GC-2 and GC-3 are associated with organic-rich ( $C_{org} \sim 8.7\%$ ) and mud-dominated (clay+silt  $\sim 97\%$ ) sediments (Thamban et al. 1997). Therefore, size sorting during transportation or reworking may not have taken place and the high illite values in the northern cores appear to suggest a predominantly northern source for illite. Moreover, there is a clear gradation in the mean values of illite in four cores from north to south (51, 45, 40 and 31% for GC-2, GC-3, GC-4 and GC-5, respectively), confirming a decreasing supply of illite from north. It was also suggested that some illite could have formed by the strong hydrolysis activities along the coastal parts of southwest India. This is supported by an overwhelmingly Al-rich illite (illite  $5\text{\AA}/10\text{\AA}$  ratios  $>0.5$ ; Thamban et al. 2002) in the southern cores (Fig. 4). Intense hydrolysis leads to

stripping of cations from the illite structure and become Al-rich (Esquevin, 1969; Gingele, 1996).

Clay mineral data presented here are in conformity with the earlier findings on the provenance of the clay minerals in the SE Arabian Sea (Nair et al. 1982b; Rao and Rao, 1995; Thamban et al. 2002) and identifies the hinterland source regions. The high amount of kaolinite and gibbsite found in the southern cores (GC-4 & GC-5) compared to the northern ones (GC-3 & GC-2), suggest the influence of hinterland source rocks and climate. Relatively low concentrations of chlorite in all the cores are consistent with the prevailing humid climatic conditions in the hinterland region, which destabilises chlorite (Chamley, 1989). Influence of early diagenesis on the clay mineral variations can be considered as negligible as revealed from the spectroscopic studies on the Quaternary sediments from the northern Indian Ocean (Colin et al. 1999). Further, clay mineral variations are independent of grain size variations in all the sediment cores (Fig. 4-7), negating any texture-induced clay mineral variations in the study area.

### **Relevance of clay mineral proxies in palaeo-monsoon reconstruction**

A careful evaluation of the modern distribution and possible factors affecting the clay mineral abundance along the eastern Arabian Sea indicate that the past variations of clay mineral proxies may be used to reconstruct the palaeoclimatic conditions on the Indian subcontinent (Thamban et al. 2002). Since lithogenic sedimentation along the SE Arabian Sea is mainly runoff-related, the clay minerals would carry signatures of the intensity of hydrolysis on land and the fluvial strength. The uncertainty related to sample treatments and mutual dilution of minerals is minimised by calculating the ratios of clay minerals rather than their relative abundance. In order to cross correlate and have confidence in different proxies, the results obtained from clay mineral proxies were compared with stable oxygen isotope records wherever available.

Since kaolinite (K) is formed under warm, humid conditions and chlorite (C) and illite (I) under arid and cold climate respectively, an increase in the ratio of kaolinite/chlorite (K/C) and kaolinite/illite (K/I) may indicate enhanced humidity. Illite crystallinity and chemistry provide independent proxies for the intensity of precipitation and hydrolysis at the weathering sites (Thamban et al. 2002). The crystallinity of illite indicates the degree of "opening" of illite structure and is considered as an index for the hydrolysmg power of the environment of source area (Singer, 1984; Chamley, 1989). The presence or absence of cations in the illite structure defines the illite chemistry and an illite  $5\text{\AA}/10\text{\AA}$  ratio below 0.5 represent Fe, Mg-rich illite, which is characteristic of unweathered rocks and ratios above 0.5 indicate Al-rich illites formed by hydrolysis (Esquevin, 1969). Enhanced rainfall and temperature are conducive

for a strong hydrolization of illite, leading to the opening of structure (poor crystallinity) and stripping of cations from its structure (become Al-rich). The presence of gibbsite can also be taken as a reliable indicator of humid environment as gibbsite is an exclusive weathering product of laterite rocks under humid conditions (Sirocko and Lange, 1991).

During most of the last glacial period, the eustatic sea level remained lower than the present level with a lowest value (~120 m) during the LGM (Fairbanks, 1989). Widespread arid conditions (Prell, 1984) in association with the lowered sea level during the LGM would have led to the enlargement of desert area of the northwestern India and the dust-laden coastal plains. Clay mineral data suggest that the illite (Fe,Mg- rich and better crystallised) concentration increased during the glacial period (Fig. 4-6). The increased illite and chlorite input and decreased kaolinite and gibbsite concentration could be attributed to an enhanced dust input related to an expanded desert region and reduced fluvial supply during LGM. Smectite in the longer sediment records (GC-2, GC-3, GC-4) showed a substantial increase in its concentration during the last glaciation compared to Holocene. This could be related to a lowered sea level and resultant exposure of the large areas of modern shelf region, flooded by the Decan Traps as well as the effect changes in the drainage patterns of the rivers.

It was suggested that during LGM, the precipitation over Arabian Sea and Indian subcontinent was diminished and/or the evaporation over Arabian Sea had increased, both indicating a weaker SW monsoon and prevalence of more arid conditions (Duplessy, 1982). The oxygen isotope records of core GC-5 showed high amplitude LGM to Holocene  $^{18}\text{O}$  fluctuations which has been attributed to the large changes in evaporation-precipitation (E-P) related to the monsoonal oscillations in this region (Thamban et al. 2000). The palaeoclimatic conditions inferred based on the clay mineral variations are therefore, consistent with the stable isotope records and also with the regional palaeoclimatic reconstructions.

The clay mineral abundances and proxy ratios revealed a marked deglacial increase in the values of humidity proxies (K/C, K/I, gibbsite and poorly crystalline, Al-rich illite) around 16 ka B.P. An increase in precipitation can also be inferred from the planktic  $^{18}\text{O}$  records, supporting an intensified summer monsoon activity during this interval. Highly increased values of humidity indicators (high K/C, K/I, gibbsite and poorly crystalline, Al-rich illite) are obtained between 9 and 6 ka B. P. (Fig. 4 & 5). This is the most prominent feature in all the cores and is best resolved in core GC-5. A substantial increase in hydrolysis and fluvial strength is evidenced, which was also inferred from the planktic  $^{18}\text{O}$  records (Thamban *et al.*, 2000).

In general, the isotope Stage 3 in cores GC 4 was characterised by relatively low values of humidity proxies K/C and K/I ratios and high Illite crystallinity values compared to the Holocene (Fig 5) This suggest that climate was less humid (deduced summer monsoons) during this period with a reduced hydrolysis at the source region Based on the detailed records of core GC-3, Thamban et al (2002) drew similar inferences and suggested that within this generally weak monsoon period, there were distinct events of monsoon intensification as indicated by sharp increases in K/C, K/I and low crystallinity values at certain intervals These events are nearly synchronous with the enhanced monsoon intervals revealed in marine cores off Pakistan (von Rad et al 1999)

A schematic diagram depicting the contrasting distribution characteristics of clay minerals along the eastern Arabian Sea in relation to the large glacial interglacial changes in climate and sea level are shown in Fig 7 During Holocene, Illites at the different sites indicate two main sources with the Indus source (transported via the southernly surface currents) dominating at the northern core sites and a hinterland source at the south The poorly crystalline and Al-rich Illite in the south and a Fe, Mg rich Illite in the northern sites

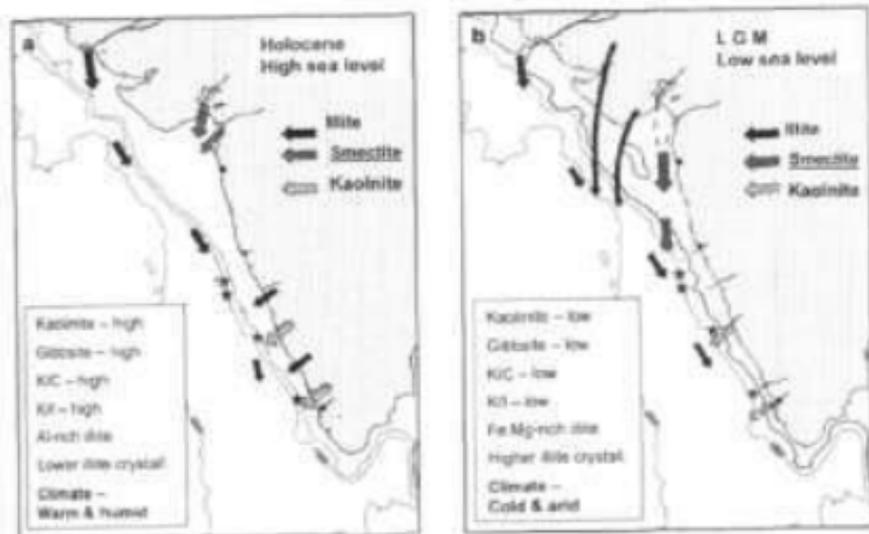


Fig 7 Schematic summary of the clay mineralogical inferences on the provenance and palaeodimatic conditions during Holocene (a) and LGM (b) in the study area. The LGM coastline is taken as the 120 m isobath below the present level (Fairbanks 1989). The arrowhead shows the direction of pathways of individual clay minerals and the thickness of the arrows suggests the estimated relative contribution of each source

corroborates this. Kaolinite and gibbsite were predominant in the southern cores, clearly suggesting a hinterland source for the clays during this period. Presence of smectite was characteristically low at all the sites and is consistent with the modern conditions wherein the smectite that supplied mainly by the Narmada and Tapti rivers are being trapped in the inner shelf itself. It is also clear that even though quantity and distribution pattern of various clay minerals seems to have modified in relation to the climatic changes in the past, the source regions remained more or less same as during modern times.

The overall consistency among individual clay mineral records and correlation with other independent proxies like oxygen isotope records from same cores, clearly suggest the reliability of the palaeoclimatic reconstruction based on clay mineral proxies. These interpretations also correspond exceptionally well with the regional monsoon reconstructions from the Indian subcontinent and Arabian Sea. It is therefore proposed that down core variations in clay mineral parameters provide a simple and reliable proxy for the late Quaternary palaeomonsoonal reconstruction. With further refinements in the approach and higher temporal resolution in measurements, it is expected that clay mineral proxies would strengthen and support the past monsoon reconstructions.

## **Summary and Conclusions**

In this study the relevance of detrital clay minerals as palaeoclimatic proxies has been evaluated using late Quaternary sediment records from the western continental margin of India. It is suggested that even though clay mineral variations in marine environment are influenced by other complicating factors, with a careful assessment of various factors and by minimising analytical uncertainties, it can be used as a reliable proxy for the source area and climatic conditions on land. The present reconstruction based on temporal variations in clay mineral records suggest that there had been significant shifts in the climatic regimes and source regions during the last glacial to present interglacial period. During LGM, hydrolysis on hinterland source rocks reduced substantially and the dust input from northwestern India to the eastern Arabian Sea appears to have increased. Subsequent deglaciation was characterised by an enhanced hydrolysis related to intensified summer monsoons starting at ~16 ka B. P. Intense hydrolysis conditions on hinterland were inferred between ~9 and 6 ka B.P., synchronous with the regional monsoonal intensification reconstructed for this Indian monsoon regime.

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