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Isotope Geochemistry of Siwalik sediments and signature of past climate change

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Summary

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Introduction

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Asian monsoon is an important climate system of the tropical region. A seasonally reversing wind system with moist oceanic air from southwest during summer and cold dry continental air from northeast during winter characterizes the monsoon climate. This system is responsible for heavy rains during June, July, August and September in the Indian sub-continent. It is believed that the monsoon system got initiated relatively recently in the geological past (about 20 Myr ago) due to uplift of Himalayas beyond a critical height (Harrison et al., 1992). Heating of land in summer at high elevation intensifies low pressure over the Tibetan plateau creating a powerful pump for moist air from oceans, which results in heavy rainfall during summer months. Reverse circulation of wind occurs during winter; the radiative cooling of elevated landmass is responsible for flow of cold dry continental air towards the ocean.

Monsoon variation record from ocean is well constrained in terms of wind strength. The monsoon intensity variation in ocean should also reflect in continent in terms of variation of rainfall amount. However, only limited information is available about monsoonal rainfall variation from continental archives (Quade et al., 1989).

Uplift of Himalaya and Tibet may also have an indirect but potentially more extensive effect on global climate through the action of carbon cycle (Ruddiman and Prell, 1997). Chemical weathering of silicate rocks on tectonically active Himalaya is thought to be the primary sink for atmospheric CO_2 for Cenozoic period.

CO_2 decrease in atmosphere and change in rainfall pattern during Cenozoic time should have had substantial effect on vegetation (Cerling et al., 1997; Pagani et al., 1999). Pre-Miocene Epoch was dominated by trees. Evolution of grasses and expansion of grasslands are observed during late Miocene indicating possible atmospheric control on vegetation.

Monsoon rainfall signature during Miocene can be reconstructed from Siwalik sediments, which are derived from higher reaches of Himalaya and deposited by various rivers in the Himalayan foreland basin stretching from Arunachal Pradesh of India in east and Potwar Plateau of Pakistan in the west (Fig.1). The Siwalik basin is a syn-sedimentary basin where sediments were deposited during the last 20 Myr forming a sediment pile as thick as 5 km in some places (Johnson et al., 1985). The lower and middle Siwalik sediments are characterized by intercalation of palaeosols with sandstones while the upper part is characterized by intercalation of conglomerate and palaeosols with occasional lenses of sandstone. These sediments contain a variety of components such as soil carbonate, organic matter, early diagenetic carbonate cement and clay minerals which are sensitive tools for reconstructing rainfall variation, vegetation, atmospheric CO_2 concentration and climate change. Besides, the huge thickness of sediments is ideal for studying geo-chemical changes associated with burial diagenesis.

The first attempt towards reconstruction of monsoonal rainfall was done based on oxygen isotope ratio of soil carbonate from Potwar Plateau of Pakistan Siwalik (Quade et al., 1989). Enrichment in ^{18}O of soil carbonate from Potwar Plateau at around 8 Ma was interpreted as intensification of monsoon in the Indian subcontinent. However, $\delta^{18}O$ of soil carbonate is a function of several variables, such as temperature of carbonate formation, amount effect in rainfall, source of moisture, shifts in seasonality etc. (Quade et al., 1995; Quade and Cerling, 1995; Stern et al., 1997; Sanyal et al., 2004). In addition, the amount of precipitation and $\delta^{18}O$ of the corresponding soil carbonate is governed by geographical configuration of the site during the formation of palaeosol. Thus the interpretation of $\delta^{18}O$ change in soil carbonates is quite complex and it is not clear if $\delta^{18}O$ increase can be explained by an increase in rainfall. Normally, an increase in rainfall is associated with a decrease in $\delta^{18}O$ (Yurtsever and Gat, 1981) and not an increase as observed by Quade et al. (1989).

Later study by Quade et al (1995) from Surai Khola section of Nepal Siwalik showed that the enrichment in ^{18}O of soil carbonate took place at 6 Ma i.e. it was delayed by 2 Myr relative to Pakistan Siwalik indicating that climate variation implied by $\delta^{18}O$ change did not occur concurrently across the Himalayan belt.

The enrichment of ^{18}O in soil carbonate in Potwar Plateau was followed by dramatic change in carbon isotope ratio at around 7.8 Ma indicating change in vegetation from pure C_3 type to a mixed C_3-C_4. The change in vegetation was thought to be a result of monsoon intensification mentioned above (Quade et al., 1989). Later studies from different sections in low latitude areas showed that the timing and nature of C_4 plant expansion varied from place to place. Studies on carbon isotope ratio of tooth enamel from East Africa showed that around 8 Ma mammals had a significant fraction of C_4

biomass in their diet. Tooth enamel data from North America show that samples older than 7 Ma indicate a C_3 dominant diet. The modern distribution of C_4 plants in North America, including the south to north gradient from C_3 to C_4 in the Great Plains is recognizable only from about 4 Ma ago (Cerling et al., 1997). Overall, it was found that the appearance of C_4 plants was earlier in tropical region compared to the high latitude belts.

A major vegetational change of the type mentioned above must have been caused by change in some component of climate system affecting plant world. Some ideas in this field are discussed below.

Cerling et al. (1997) suggested that at temperatures typical of low latitudes, lowering of atmospheric CO_2 concentration acted as the trigger for sudden expansion of C_4 plants. In this context, it may be mentioned that increased weathering rate during the past 40 Myr, especially in the tectonically active Himalayan-Tibetan region, was propose to be a cause for lowering of CO_2 (Raymo and Ruddiman, 1992). It is also likely that the lowering of CO_2 was not gradual throughout the whole period. Recent studies on alkenones from Pacific sediments showed that atmospheric CO_2 concentration increased from 15 Ma and stabilized at the pre-industrial value (~280 ppm) at around 9 Ma (Pagani et al., 1999). This seems to suggest that C_4 plant expansion is not related to CO_2 change. It might have been driven by enhanced low-latitude aridity or changes in seasonal precipitation pattern because in most regions, C_4 grass expansion is seen to be associated with changes in rainfall (Pagani et al., 1999).

Seasonality model of Pagani et al. (1999) supports the proposition of Quade et al (1989) that change in rainfall pattern was the cause for vegetational change observed in Pakistan Siwalik. However, the timings of rainfall variation in Pakistan Siwalik and Nepal Siwalik do not agree. In Pakistan Siwalik, the vegetational change is preceded by rainfall change whereas in Nepal Siwalik, the reverse was observed. Pakistan Siwalik region is situated at the far end of Indian monsoonal rainfall track and as a result, receives only moderate amount of rainfall. It is obvious that rainfall reconstruction can be better done from Siwalik zones situated in monsoon sensitive region. In this context, presence of extensive Siwalik exposures in India (Fig.1), which experiences high monsoon rainfall in present day condition offers an excellent choice of samples for reconstructing rainfall variation.

Timing of vegetational change in Pakistan Siwalik was also challenged by other workers. Carbon isotope ratio of tooth enamel showed presence of C_4 plants as early as 10 Ma (Morgan et al., 1994). The carbon isotope ratio of long chain alkane from organic matter also showed that C_4 plants were around at about 9 Ma (Freeman and Colarusso, 2001). Again, existence of different Siwalik sections in Indian Himalayan belt, which are well dated, provides a good opportunity for further study on this topic.

Objectives of present study

(1) Reconstruction of monsoonal rainfall variation: As mentioned previously, monsoonal climate in Indian subcontinent evolved with the rising of Himalaya. While the onset of monsoon is somewhat clear, the variation of monsoon rainfall in geological past is poorly known. The best way to investigate palaeo-monsoon is to study rainfall proxies from monsoon sensitive regions. Kangra valley and Haripur Khol Siwalik sections in Himachal Pradesh, India are two areas characterized by high seasonal rainfall (mean 550 mm during southwest monsoon; Indian Meteorological Department, 1970) with closely spaced isohyets (Rao, 1976). In this zone, the mean rainfall changes by about 350 mm if one moves over a distance of only 50 km towards north (Rao, 1976). Isotopic studies from such areas could help in deciphering past change in monsoon and assessing their relation with the expansion of C_4 plants. The proxies that offer promises to reconstruct the monsoon are oxygen isotope ratio of soil carbonate and hydrogen isotope ratio of pedogenic clay minerals.

(2) Reconstruction of vegetation from well-dated sections: Since the timing of appearance of C_4 plants in Pakistan Siwalik is debatable well-dated sections from Indian Siwalik could help in clarifying the issue. The proxies that can be used to reconstruct vegetation are carbon isotope ratio of soil carbonate, organic matter associated with soil carbonate nodules and early diagenetic carbonate cement of sandstone nodules.

(3) Estimation of atmospheric CO_2 in geological past: The CO_2 concentration in earth's atmosphere has varied enormously through the geological time (Berner, 1991; Retallack, 2002; Ekart et al., 1999). The Paleozoic to Mesozoic era was a period of extreme fluctuations when CO_2 concentration changed from a level of 250 ppmV to ten times that level. In contrast, the Cenozoic era was characterized by a decrease in CO_2 with the minimum concentration at the Neogene Period (Ekart et al., 1999; Retallack, 2002; Ghosh et al., 2001; Pagani et al., 1999). However, there are significant

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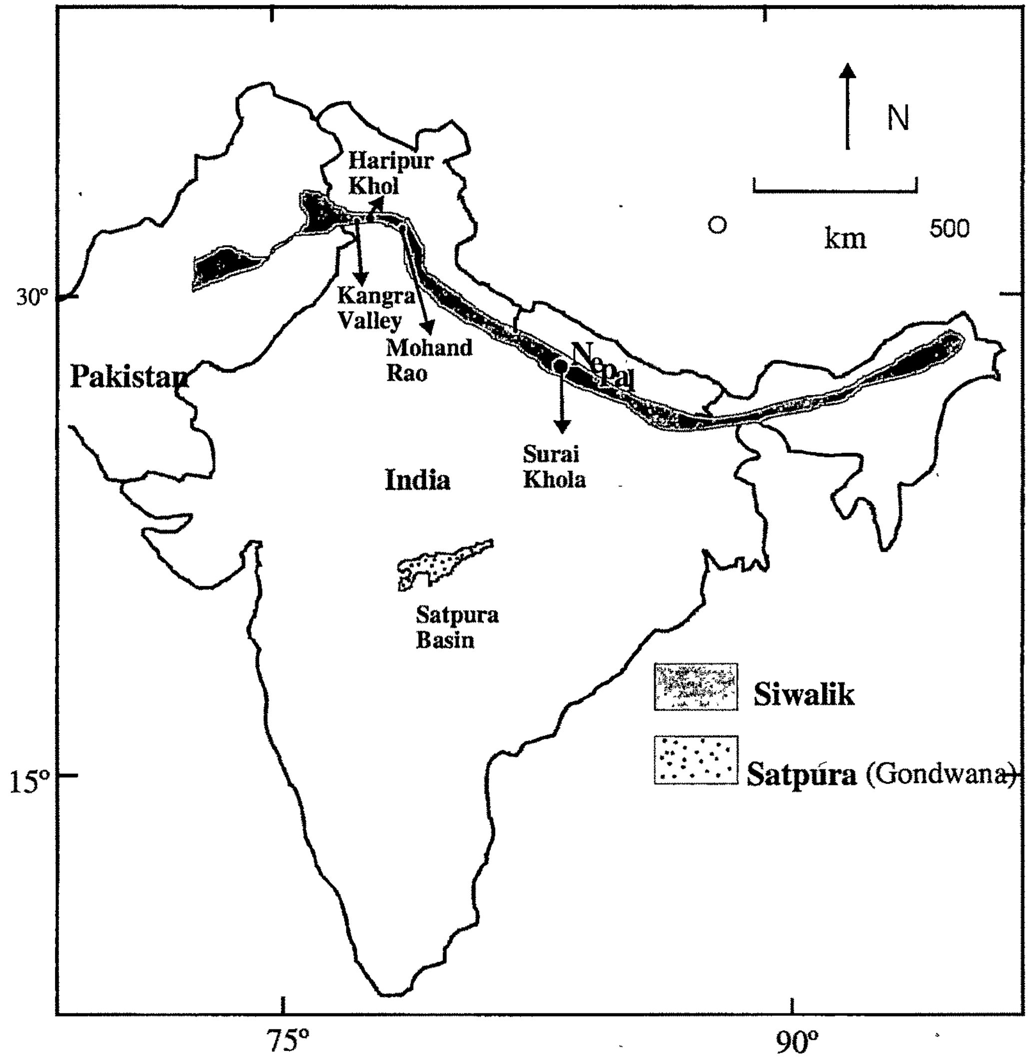

Fig.1 *Geographical extension of Siwalik and Satpura (Gondwana) Basins. Location different study areas in the Siwalik basin are shown.*

discrepancies between estimates derived from different proxies and the data is not well constrained for some geologically important time periods. Based on carbon isotope ratio in palaeosols and Cerling's model (1991) an attempt has been made in the present thesis to estimate the atmospheric CO_2 concentration for the late Neogene Period. A comparative study on CO_2 concentration for the Mesozoic time was also done based on palaeosols from Denwa and Bagra Formation of central India. There are excellent soil exposures associated with these two Formations containing well-preserved soil carbonate nodules. They provide contrasting picture of a high CO_2 concentration era in the geological history. These studies are particularly important since CO_2 concentration estimation along with monsoonal reconstruction can provide insight about the controlling factors for vegetational change.

(4) Study of diagenesis of sediments: Siwalik basin is a syn-sedimentary basin, which sank continuously by the overburden of depositing sediments. During burial, sediments undergo physical and chemical changes collectively known as diagenesis. An attempt has been made here to study diagenetic processes through isotopic changes in carbonate cement of sandstone, change in the assemblages of clay minerals and feldspar in sandstone. These studies dealing with diagenesis of sediments are necessary for proper interpretation of geochemical data.

Field areas

The Siwalik sections selected for the present study are Haripur Khol section (Age: 6 to 0.5 Ma) of Subathu sub-basin, Ranital and Kotla section (Age: 11 to 6 Ma) of Kangra Valley, Mohand Rao section (Age: 10 to 5 Ma) of Dehra Dun Valley and Surai Khola section (Age: 13 to 1 Ma) of Western Nepal. For a comparative study Gondwana sediments from Satpura Basin of Central India has also been studied (Fig.1). Age of the Siwalik sections are based on palaeomagnetic method and Gondawana sections are based on fossil assemblages and field relationships.

Results and discussions

Carbon and oxygen isotope ratios of soil carbonate nodules and carbon isotope ratio of associated organic matter were measured from Ranital, Kotla and Haripur Khola sections in order to reconstruct vegetational history and change in contemporaneous rainfall (Fig.2). $\delta^{13}C$ values of soil carbonate show that from 10.5 Ma to 6 Ma the vegetation was C_3 type and that around ~6 Ma C_4 grasses appeared. The $\delta^{18}O$ variations of soil carbonate suggest that the monsoon system intensified, with one probable peak at around 10.5 Ma and a clear onset at 6 Ma, with peak at 5.5 Ma. After 5.5 Ma monsoon strength decreased and attained the modern day condition with minor fluctuations, which is supported by marine proxy of upwelling in the Arabian Sea and sedimentary morphology in Siwalik. The covariation between $\delta^{18}O$ and $\delta^{13}C$ data suggests that a change in precipitation pattern was partly responsible for expansion of C_4 grasses.

It was also noted that in a mixed C_3-C_4 environment, estimation of abundance of

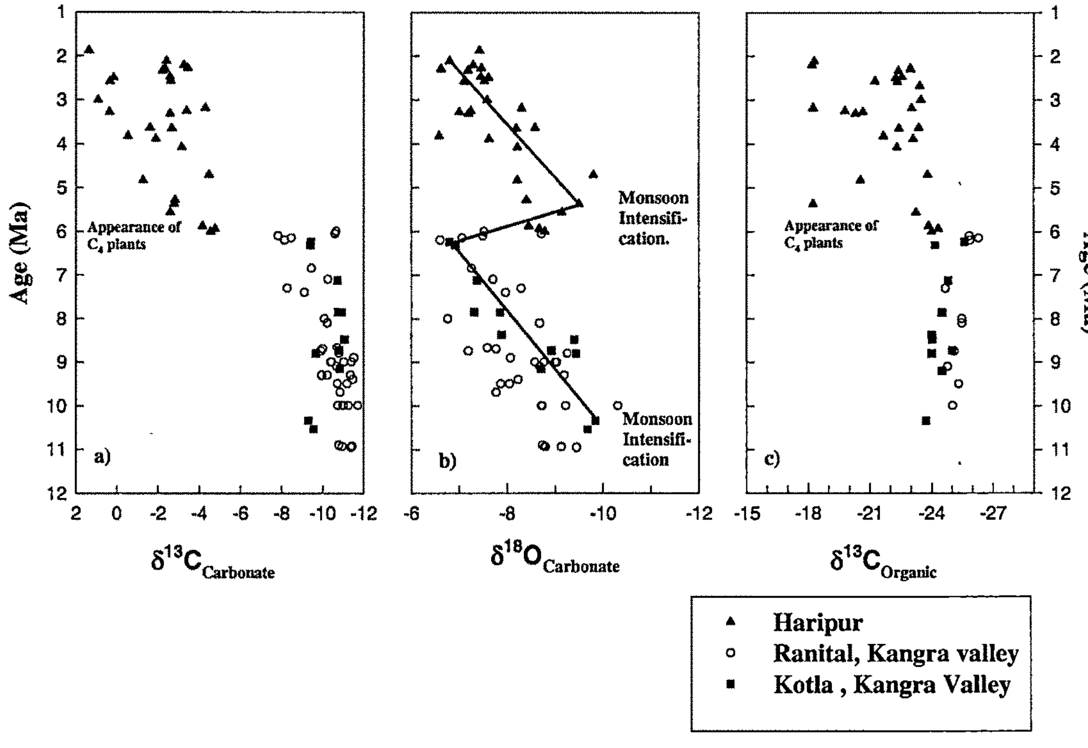

Fig.2 *Composite plot of isotope ratios against age of soil carbonate nodules from three sections (Ranital, Kotla and Haripur Khol) a) Carbon b) Oxygen and c) Organic matter from the same nodules. Carbon isotope ratio of soil carbonate and associated organic matter indicates that vegetation was entirely of C_3 type from 11 to 6 Ma. Post 6 Ma time is marked by appearance and expansion of C_4 grass. Oxygen isotope composition shows stepwise variation with probable peak at ~10.5 Ma and a clear peak at ~6 Ma. These variations are attributed to change in monsoon rainfall. The solid lines are drawn to guide the eye.*

C_3 and C_4 plants using soil carbonate in one case, and residual soil organic matter in another, may differ. This can be explained by assuming that the plant-respired CO_2 is the main contributor of carbon in soil carbonate and may have different isotopic composition from that of residual organic matter in soil. Since plant-respired CO_2 from C_3 or C_4 plants depends on response of each of them on growing season-the net effect may not be representative of organic matter abundance from their residual in soil. In addition, abundance estimate of C_3-C_4 plants also shows variation with time, probably caused by change in growing season conditions through time.

Carbon isotope ratio of early diagenetic carbonate cement from sandstone (DCCN) was measured from Mohand Rao and Haripur Khol section to reconstruct palaeovegetation (fig.3). The $\delta^{13}C$ of DCCN from Mohand Rao section varies from –10.5 ‰ to –0.2 ‰ with progressive enrichment in $\delta^{13}C$ values from 9 Ma to 7.3 Ma indicating gradual change of C_3 type of vegetation to C_4 type vegetation. Post 7.3 Ma, the $\delta^{13}C$ value is anchored around zero per mil indicating mixed C_3-C_4 environment with C_4 dominating the ecosystem. In Haripur Khol section, the $\delta^{13}C$ value of DCCN indicates presence of both C_3 and C_4 type of plants with the dominance of C_4 in ecosystem.

The oxygen isotope ratio of DCCN does not show any systematic variation with time. The $\delta^{18}O$ of DCCN from Mohand Rao section ranges from –8.9 to –13.6 ‰ and in Haripur Khol section $\delta^{18}O$ ranges from –9.9 to –13.6 ‰. At a given stratigraphic level average $\delta^{18}O$ value of DCCN is depleted (maximum depletion up to 4‰) compared to the average $\delta^{18}O$ of soil carbonate from the corresponding level. The depletion in $\delta^{18}O$ may be due to contribution from river water infiltrating the groundwater system in post-monsoon period.

Carbon and oxygen isotope ratio of soil carbonate nodules and carbon isotope ratio of associated organic matter from the same nodules were also measured from Mohand Rao section in few cases (n=9). From 9 to 8 Ma the carbon isotope ratio of soil carbonate varies from –10.8 to –7.8 ‰ indicating the vegetation in the flood plain was characterized by C_3 dominated plants and from 5.4 to 4.8 the $\delta^{13}C$ ranges from 0.1 to –4.3 ‰ indicating vegetation was characterized by mixed C_3-C_4 plants with C_4 dominating the ecosystem. The carbon isotope ratio of the organic matter from same soil carbonate nodule ranges from –25.2 to –24.4 ‰ (from 9 to 8 Ma) and –17.4 to –24.6 ‰ (from 5.4 to 4.8 Ma) corroborating the above results. The average $\delta^{18}O$ value of soil carbonate nodule for the time period 9 to 8 Ma is –8.8 ‰ and for 5.4 to 4.8 Ma the value

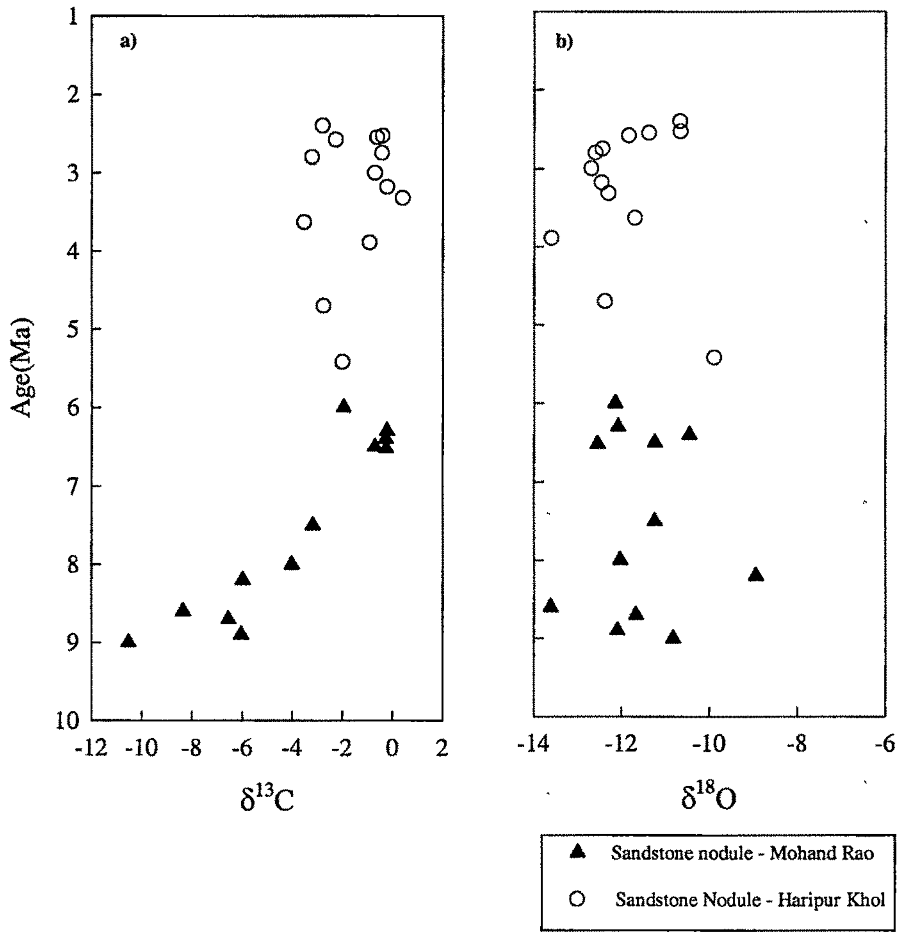

Fig.3 *Composite plot of isotope ratios against age for sandstone nodules from Mohand Rao and Haripur Khol sections. a) Carbon isotope ratios of DCCN show that at around 9 Ma the vegetation was mainly C_3 type and subsequently C_4 plants started appearing. By 6 Ma abundance of C_4 plants reaches a high value and remains so thereafter. b) Oxygen isotope ratios of DCCN do not show any systematic variation. The dispersion in $\delta^{18}O$ may be due to various contribution of river water into shallow groundwater*

is –7.9 ‰. These average $\delta^{18}O$ values are comparable with the oxygen isotope data of soil carbonate from Haripur Khol and Kangra valley.

Hydrogen isotope ratio of OH group of pedogenic clays was measured in Siwalik sediments samples from Haripur Khol section (Fig.4). X-ray diffraction analysis of clays separated from pedogenic nodules shows presence of illite as the most dominant species followed by smectite and chlorite. The relative abundance of the three species is nearly

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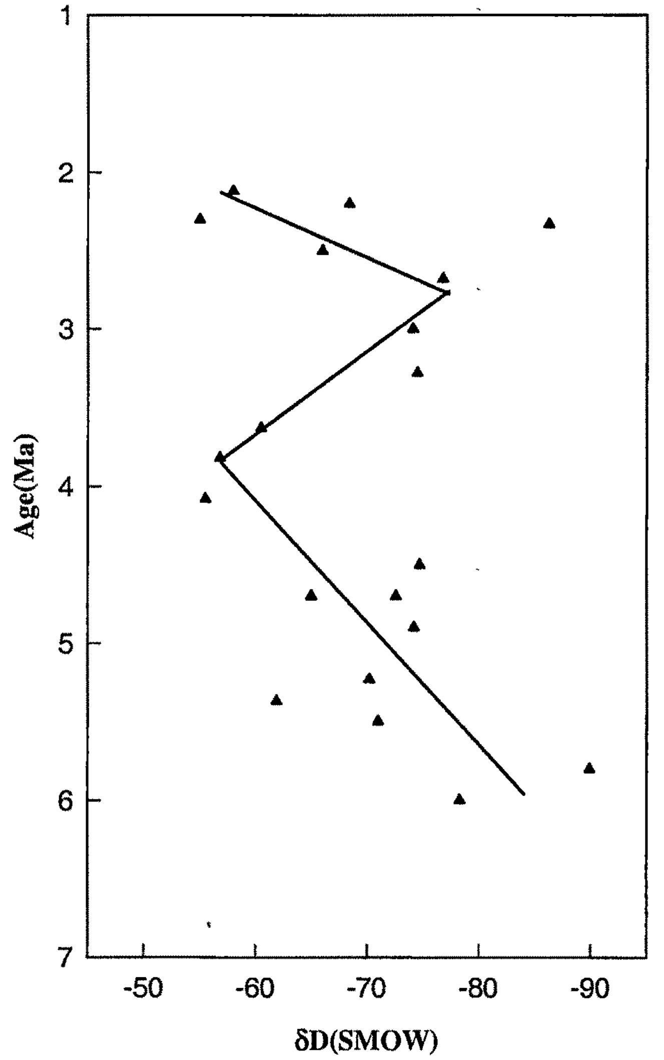

Fig.4 *Hydrogen isotope ratio of pedogenic clays from Haripur Khol section. The profile is characterized by lower values at around 6 Ma and 3 Ma punctuated by higher value at around 4 Ma. The two phases of lower δD probably indicate intensified monsoonal rainfall. It could be mentioned that oxygen isotope ratio of soil carbonate from Haripur Khol section also shows intensification of monsoon at around 6 Ma but the peak at 3 Ma was not observed from oxygen isotope ratio of soil carbonate. Solid lines are drawn to guide the eye.*

constant throughout the section. Hydrogen isotope ratio of the clays (combined illite, smectite and chlorite) varies from –55 ‰ to –90 ‰ (relative to VSMOW). At around 6 Ma and 3 Ma, the hydrogen isotope ratios are characterized by lower δD values (–80 to –

90‰), which probably indicates high rainfall; relatively higher δD (–55‰) values occur at around 4 Ma and 2 Ma, which may indicate low rainfall regime. It seems that monsoonal intensity for the last 6 Ma varied with two clear peaks occurring at 6 Ma and 3 Ma punctuated by decrease at 4 Ma and post 3 Ma. This variation is, in general, consistent with other proxies like vegetation and sediments architecture.

Carbon and oxygen isotope ratio of soil carbonate and carbon isotope ratio of

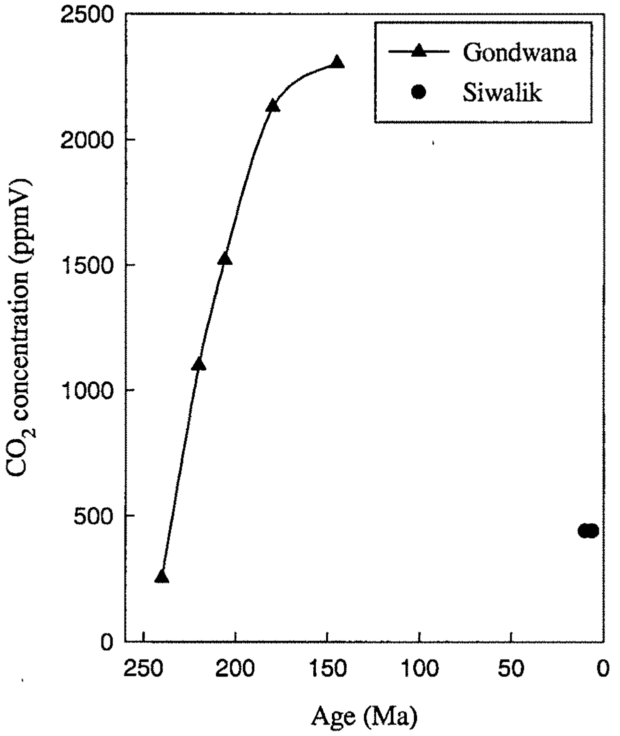

Fig.5 *Atmospheric CO_2 concentration variation through time based on $\delta^{13}C$ of paleosols from Gondwana deposit of Satpura basin. Lower Triassic CO_2 concentration was about 255 ppmV and it reached a maximum of 1520 ppmV during upper Triassic through an intermediate concentration of 1100 ppmV. During the Jurassic period the average concentration was about 2190 ppmV. During Siwalik time (11-6 Ma) CO_2 concentration was 455 ppmV.*

associated organic matter from two stratigraphically superimposed Formations, Denwa and Bagra from Gondwana Supergroup were measured. The $\delta^{13}C$ value of the oldest soil carbonate from Denwa Formation is about –10.5 ‰, the youngest sample is –6.4 ‰ and the average $\delta^{13}C$ value of the in-between samples is –8.4 ‰. The mean $\delta^{13}C$ value of the associated organic matter from the above samples is about –24.9 ‰. The $\delta^{13}C$ value of soil carbonate from the different exposure of Bagra Formation is almost similar (–6.7‰) and is close to the $\delta^{13}C$ value of the youngest sample collected from Denwa Formation (contact of the Bagra Formation). The average $\delta^{13}C$ value of associated organic matter from Bagra Formation is –25.9 ‰. Calculations of atmospheric CO_2 concentration using the carbon isotope ratio of Denwa and Bagra soil carbonate shows that at the beginning of the Denwa Formation CO_2 concentration was about 255 ppmV and it reached a high value of 1520 ppmV at the end of Denwa Formation through an intermediate concentration of 1100 ppmV. During Bagra Formation the concentration was about 2190 ppmV. The progressive increase in CO_2 concentration during the Denwa and constant concentration at Bagra is conformable with the available CO_2 variation record from other parts of world for the Permo-Triassic boundary to Jurassic time period. The CO_2 concentration during late Miocene (10 to 6 Ma) time was 455 ppmV (Fig.5).

Like carbon isotope ratio, the $\delta^{18}O$ value of soil carbonate in individual soil profile of Gondwana and Siwalik is almost constant though the Siwalik samples show little enrichment toward surface. The average $\delta^{18}O$ value of the Siwalik soil carbonate is –9 ‰ and those of the Bagra and Denwa are –6.7 ‰ and –5.2 ‰ respectively. The $\delta^{18}O$ of soil carbonate from the Gondwana and Siwalik probably reflect the difference in the rainfall pattern for these two time periods. In addition, $\delta^{18}O$ of Bagra and Denwa indicate that amount effect in rainfall played a major role in determining the oxygen isotope ratio of soil carbonate.

Carbon and oxygen isotope ratios of carbonate cement from Siwalik sandstone were measured from Surai Khola section of western Nepal (Fig.6). The $\delta^{18}O$ values of cement show three evolutionary phases. From 12 Ma to ~6 Ma, the average $\delta^{18}O$ is around –13.6±1.9 ‰ (n=114) with a large spread from –10 to –18 ‰. This large spread probably indicates dissolution and re-precipitation of carbonate at various stages during burial. Subsequent to 6 Ma, $\delta^{18}O$ shows sudden swing towards enriched values with less scatter in data; the enrichment continues up to 4 Ma with maximum $\delta^{18}O$ value around –

7 ‰. The average $\delta^{18}O$ value for this period is –10.7 ±1.6 ‰ (n=25). From 4 Ma to 2 Ma, $\delta^{18}O$ remains fairly uniform with an average value of –8.8±1.2 ‰ (n=17). The increase in

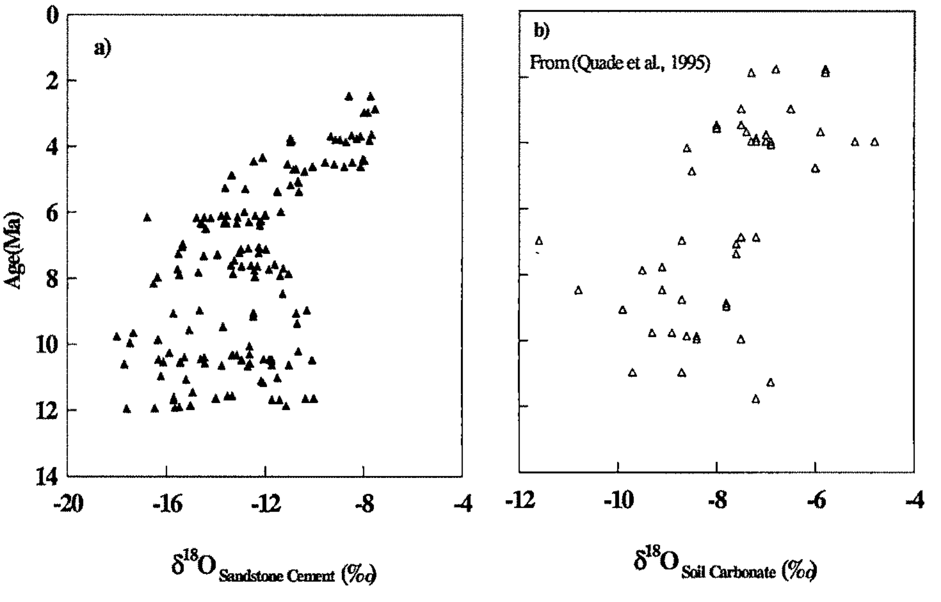

Fig.6 *Oxygen isotope ratio of diagenetic calcite cement from sandstone samples of Surai Khola showing three evolutionary phases. From 12 Ma to 6 Ma, oxygen isotope ratio is characterized by large spread: –10 to –18 ‰. The large spread is probably due to dissolution and re-precipitation of carbonate at different stages of burial. At around 6 Ma, the ^{18}O shows sudden swing towards enriched values and the maximum enrichment is attained near 4 Ma. The sudden swing is in tandem with enrichment of oxygen isotope ratio in soil carbonate* ***(Fig.b; Data from Quade et al., 1995)*** *from the same section, which indicates change in precipitational pattern. The similar trend of isotope ratio for cement and soil carbonate (for post 6 Ma period) suggests meteoric water has played an important role in determining the ratio during diagenesis. However, the ratio of sandstone cement is depleted compared to the ratio of soil carbonate (b). The depletion may be due to precipitation of cement at higher temperature compared to that of soil carbonate. Subsequent to 4 Ma, the $\delta^{18}O$ is nearly constant.*

oxygen isotope ratio of carbonate cement in sandstone occurs concurrently with that of soil carbonate, indicating a major role of meteoric water in changing oxygen isotope ratio of diagenetic carbonate cement. However, the oxygen isotope ratio of carbonate cement of sandstone is relatively depleted compared to that of soil carbonate in 6 Ma to 4 Ma time range. The depletion probably indicates precipitation of sandstone cement at a temperature higher than that of soil carbonate.

$\delta^{13}C$ values of calcite cement do not show any definite trend with time. From 12 Ma to 7 Ma, the $\delta^{13}C$ ranges between –3.3 to –9.9 ‰ with an average of –7.1 ± 1.5‰ (n=91). Subsequent to 7 Ma, the number of relatively enriched ^{13}C values increases. From 7 Ma to 2 Ma, the $\delta^{13}C$ varies from –2.8 to –9.2 ‰ with an average of –5.7±1.5 ‰ (n=65). Relatively enriched $\delta^{13}C$ for post 7 Ma period is due to appearance of C_4 plants, which have enriched ^{13}C, compared to C_3 plants. The large spread in $\delta^{13}C$ of the cement probably indicates production of CO_2 at various stages of diagenesis of organic matter. Diagenesis of organic mater at various depths produces CO_2, which could have large range of carbon isotope ratio depending on the depth-level of CO_2 generation.

Mineralogical assemblages show increase in diagenesis with increase in depth. Clay minerals (<2μ) separated from sandstone comprises of smectite, illite, chlorite and kaolinite. Relative increase in the abundance of illite and decrease in the abundance of smectite indicate illitization of smectite with increase in burial, which is compatible with observed absence of K-feldspar in lower stratigraphic succession. Dissolution of K-feldspar might have supplied potassium for illite formation. In the younger samples K-feldspar is present but shows corrosive features.

Conclusions

1) Monsoon evolution in Indian sub-continent was not smooth. The monsoon strength intensified at around 10, 6 and 3 Ma. The appearance of C_4 plants probably related to compound effects of increase in monsoonal strength and lowering of pCO_2 in the atmosphere.

2) The timing of C_4 plants expansion ranges from 9-6 Ma in Indian Siwalik. Also the nature of transition from pure C_3 type of vegetation to C_3-C_4 mixed vegetation varies from section to section. In some section the transition is rapid and in some section it is gradual.

3) CO_2 concentration estimation showed that lower Triassic CO_2 concentration was about 255 ppmV and it reaches maximum up to 1520 ppmV during upper Triassic through an intermediate concentration of 1000 ppmV. During the Jurassic time the

concentration was between 2110 and 2275 ppmV. Late Miocene CO_2 concentration was 455 ppmV.

4) Meteoric water has played a major role during diagenesis of sandstone. Clay minerals assemblages show illitization of smectite with increase in depth. Abundance of K-feldspar also decreases with depth, which is compatible with the illitization of smectite. The K-feldspar might have acted as a supplier of potassium during illitization process.

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