Abstract

Uplift of Himalaya in Cenozoic has had a profound effect in regulating climate, vegetation and atmospheric CO_2. It is now clear that Indian monsoon climate got established only when Himalaya attained a critical height. Weathering in tectonically active Himalaya caused lowering of atmospheric CO_2. Establishment of monsoon and lowering of CO_2 should have had substantial effect on vegetation. The main emphasis of this thesis is to glean out the monsoonal rainfall variation, quantification of atmospheric CO_2 and build the vegetation scenario for last 11 Myr from Siwalik sediments of Himalayan foothills. 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 have also been analyzed.

Carbon and oxygen isotope ratios of soil carbonate nodules and carbon isotope ratio of associated organic matter were measured from Ranital, Kotla and Haripur Khol sections in order to reconstruct vegetational history and change in contemporaneous rainfall. $\delta^{13}C$ 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. This finding 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 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. 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 plants dominating the ecosystem. In Haripur Khol section (from 6 Ma onwards), 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 that 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 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 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 sediment samples from Haripur Khol section. 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 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 from these data that monsoonal intensity for the last 6 Ma varied with two clear peaks occurring at 6 Ma and 3 Ma punctuated by a 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 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 –10.7 ‰, 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 (11 to 6 Ma) time was 455 ppmV.

Like carbon isotope ratio, the $\delta^{18}O$ value of soil carbonate in individual soil profiles 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 $\delta^{18}O$ values of soil carbonates 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. 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 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 ($<2\mu$) 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.