

Late Miocene record of palaeovegetation from Siwalik palaeosols of the Ramnagar sub-basin, India

Seema Singh^{1,*}, B. Parkash¹, A. K. Awasthi¹ and S. Kumar²

¹Department of Earth Sciences, Indian Institute of Technology Roorkee, Roorkee 247 667, India

²National Institute of Hydrology, Roorkee 247 667, India

Stable carbon isotope analyses of pedogenic carbonates were conducted on two Siwalik sections, Jammu–Nandni and Purmandal–Uttarbehani, covering a lateral stretch of ~ 25 km, in Ramnagar sub-basin, in order to reconstruct palaeovegetation history over the past ~ 12 Ma. The results revealed that the carbon isotopic record in studied sections fell in two broad divisions, an older part (> 7 Ma) characterized by $\delta^{13}\text{C}$ -depleted isotopic values and a younger part (< 5 Ma) consisting of $\delta^{13}\text{C}$ -enriched values, indicating exclusive presence of C_3 and C_4 vegetation respectively. The present work provides an important link to the extensive palaeovegetational studies done in the Pakistan and Nepal Siwaliks.

Keywords: Carbon isotopes, palaeovegetation, pedogenic carbonates, photosynthetic pathway.

THE Sub-Himalaya forms the outermost foothills of the Himalaya and preserves record of post-collision sediments produced by weathering and erosion of debris of the rising Himalayan front. These sediments were carried and deposited in a foredeep basin called the Himalayan Foreland Basin (HFB). The HFB is divided into a number of sub-basins separated from each other by sub-surface basement highs and ridges¹. The Neogene Siwalik Group sediments of HFB have received much attention from the geologists and enormous work has been done in palaeontology, biostratigraphy, sedimentology, geomorphology and magnetostratigraphy in the Indian part of the Siwalik belt since the early eighties^{2–10}. In comparison to this, meagre amount of work has been accomplished in palaeopedology, palaeoclimatic and palaeoecological reconstructions^{2,8,11–16}.

Many faunal and floral changes are apparent from the Siwalik sediments and the change is largely attributed to uplift of the earth's youngest and dynamic orogenic belt called the 'Himalaya' during the Cenozoic, which has had a profound effect in regulating climate, atmospheric CO_2 , and flora and fauna on the Asian continent^{17–19}. This may be possible because the Asian monsoon climate got estab-

lished only when the Himalaya attained a critical height and weathering in the tectonically active Himalaya caused a lowering of atmospheric CO_2 , which had substantial effect on the nature of the photosynthetic pathway or the type of vegetation¹⁷. A large number of workers have illustrated that the Himalaya and the Tibetan Plateau attained sufficient elevations to initiate major palaeoclimatic changes only during Late Miocene. For example, fossil evidences indicate that no significant physical barrier to the migration of mammal faunas existed prior to Late Miocene^{18,19}. Palaeoecological reconstructions have long been consistently based on the study of vertebrate and invertebrate fossil evidences, but due to their poor preservation in various Siwalik sections and the recent emphasis on more refined techniques, it becomes natural to switch over to some other reliable studies besides fossil evidences.

Palaeosols have proved to be beneficial in solving various geological problems, especially palaeoclimatic reconstructions. Siwalik palaeosols are associated with alluvial deposits forming a part of thick aggradational pedocomplexes within the alluvial strata and commonly do not occur as single isolated units of palaeosol profiles, but rather as multiple sequences. Stable isotope ratios of carbon and oxygen of pedogenic carbonates (or soil carbonates or pedogenic calcretes) preserved in ancient soils are being used to track vegetation and climate changes through time within many different Cenozoic rock sequences²⁰. Quade and co-workers²¹ first analysed the carbon (^{13}C) and oxygen (^{18}O) isotope ratios of pedogenic carbonates in the Himalayan Cenozoic sediments from the Potwar Plateau of Pakistan Siwaliks, ranging in age from 18 Ma to Recent. Thereafter, a large amount of work on palaeovegetation aspects has been carried out in Pakistan and Nepal Siwaliks^{20–23}, followed by some palaeovegetational reconstructions using stable isotope ratios of pedogenic carbonates in the Indian Siwalik belt restricted to the Kangra, Subathu and Dehradun sub-basins^{14,15,24}. Despite the fact that there are numerous exposures of Siwaliks in the Ramnagar sub-basin and also that these have several palaeosol assemblages containing pedogenic carbonate nodules, no work is available on their palaeovegetational reconstructions using stable isotope compo-

*For correspondence. (e-mail: geoseema05@yahoo.co.in)

sition. In view of this, for the present study two Siwalik sections in the Ramnagar sub-basin have been considered, viz. Jammu–Nandni (J–N) and Purmandal–Uttarbehani (P–U) in Jammu and Kashmir, India.

An attempt has been made to use carbon isotope ratios of pedogenic carbonates as proxy palaeo-records in order to reconstruct palaeovegetational history of the Ramnagar sub-basin since Late Miocene. The present work may provide a good opportunity for more studies in the Jammu region, which could reveal important and significant information on past climate and vegetation history of the NW part of the foreland basin and consequently offer a good link for global palaeovegetation correlation since late Miocene.

Study area

The study area comprises low-lying Siwalik hills sloping gently towards south, but gradually becoming steep as we move northward. The Siwalik Group has developed all along the southern margin of the Himalayan mountain belt as the folded, faulted and uplifted part of the Himalayan foreland basin and is stratigraphically divided into Lower, Middle and Upper Siwaliks ranging in age from Mid-Miocene to Lower Pleistocene. We carried out carbon isotope analysis of soil carbonate, largely nodules, from two Indian Siwalik sections: J–N and P–U,

which are placed laterally ~25 km from each other (Figure 1). Both the studied sections fall in Ramnagar sub-basin of the Outer Himalaya between lat. 32°34' and 32°80', and long. 74°50' and 75°12' (Figure 1). The complete Siwalik Group stratigraphy of the Jammu area is given in Table 1.

Methodology

Analysis of soil carbonate in palaeosol beds

In order to use stable isotope ratios of carbon and oxygen in the soil carbonate mineral to study its relationship with climate, it is necessary to eliminate the problem of carbonate or calcrete not formed by pedogenic processes in the soil, because the stable isotope ratios of carbon and oxygen in both organic and inorganic components of the plant soil environment can only record and integrate information relating to ecosystem/vegetation and climatic parameters, whereas groundwater carbonates do not have such a reliable mechanism¹⁴. It is therefore a necessity and prerequisite action to first distinguish pedogenic and groundwater carbonates (non-pedogenic) in the palaeosols.

Various field and micromorphological evidences have been used by different workers to distinguish pedogenic carbonates from non-pedogenic carbonates^{25–28}, many of which are used in the present study and are also illustrated in Figures 2 and 3. For example, Birkeland²⁵ suggested that pedogenic carbonate is mainly micrite and has dry colours of 7.5YR7/4 to 10YR8/3, and the original grains have been forced apart (Figures 2 and 3). Pedogenic carbonate may be distinguished in the field from shallow groundwater carbonate by gradational contacts, palaeosol horizons above the zone of carbonate accumulation (particularly the presence of B/Bt horizon overlying a Bk or K horizon) and a vertical arrangement of peds, root traces, desiccation cracks and tubules, and a groundwater rather than soil origin of carbonate is indicated by sharp contacts and lack of associated soil horizons²⁶ (Figure 2). Similarly, Pal *et al.*²⁸ concluded that pedogenic calcrete can be micritic, micro-sparitic or sparitic occurring as coatings, fillings and nodules affecting weathering of primary minerals, but has fabric similar to adjacent soil fabric, i.e. inclusion of primary minerals and usually occurs together with illuvial clay pedofeatures (Figure 3).

Field analysis

Extensive field work was done along road and river-cut exposures in the J–N and P–U sections. The identification and characterization of Siwalik palaeosols has been done by recording horizon-wise field properties²⁹. Pedogenic carbonate is usually nodular, but is also present in forms like powdery and pedotubule (Figure 2a–g), as defined

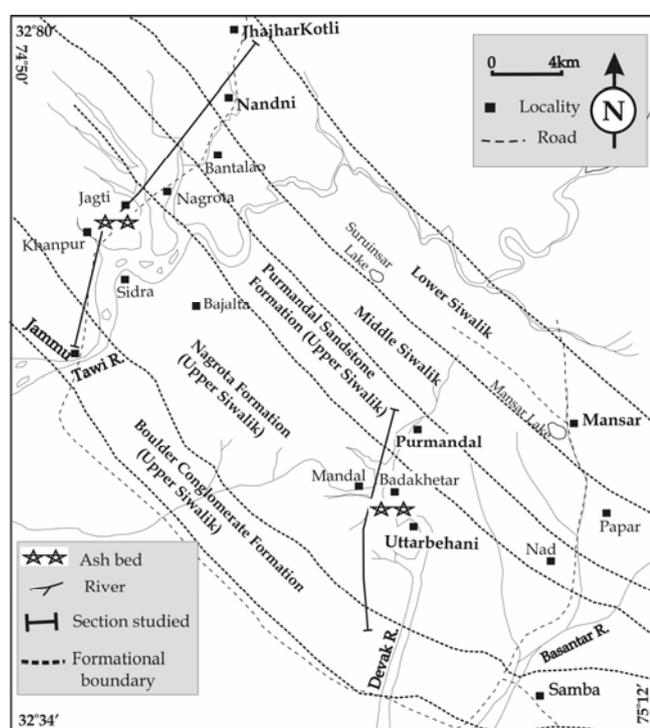


Figure 1. Geological map showing location of the studied sections: Jammu–Nandni (J–N) and Purmandal–Uttarbehani (P–U) (modified after Dey *et al.*⁵³ and Pandita and Bhat⁶).

Table 1. Siwalik Group stratigraphy of Jammu region on the basis of palaeontology, lithology and palaeomagnetic studies (modified after Dasarathi³¹, Ranga Rao *et al.*³ and Aggarwal *et al.*⁵)

Siwalik	Potwar/Chandigarh, etc.	Tawi Valley, Jammu
Upper	Boulder Conglomerate Formation Pinjor Formation Tatrot Formation Dhokpathan Formation	Tawi/Bahu Conglomerate Formation Nagrota Formation Purmandal Sst. Formation
Middle	Nagri Formation Chinji Formation	Bantalao/Mansar Formation Nandini Formation
Lower	Kamlial Formation	Jahjjar Formation

by Wright and Tucker²⁷, in the palaeosols and was collected from the respective pedons for analysis. Dilute (about 10%) hydrochloric acid carried in an eyedropper bottle was needed for testing the carbonate content of the samples by their effervescence during reaction with the acid²⁹. It is important to mention here that the sampling for pedogenic carbonates was confined to the B/Bk/BC-horizons and at least more than 30 cm deep from the surface of the palaeosol profile to avoid any possible surficial alteration of CO₂ composition in the carbonate due to exchange with the atmosphere. Sampling of carbonate nodules from the C-horizon was avoided in order to exclude the possibility of mixing of the detrital non-pedogenic carbonates present in the parent material with the pedogenic carbonates. The pedogenic carbonates were distinguished from the groundwater carbonates while sampling on the basis of one or more of the field characters as described earlier (Figure 2).

Laboratory analysis

Micromorphological analysis: Before proceeding to the laboratory techniques for isotopic analysis, the collected pedogenic carbonate samples from the field were checked for their pedogenic nature. This was done by cross-checking with thin-section study of samples, and the presence of various pedogenic micromorphological features in accordance with studies by various workers^{26–28,30} further confirmed the pedogenic origin of calcium carbonate (Figure 3).

Micromorphological features often serve as first-order criteria in evaluating diagenesis of sediments. Preservation of micrite and absence of obvious recrystallization should indicate relatively lesser occurrence of diagenetic alteration, and the measured $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values are primary. The dominantly micro-sparitic groundmass, partial replacement of micrite by microspar, and nonluminescence of all the groundmass components suggest that diagenetic alteration (post-calcrete nodule formation) is insignificant in altering the isotopic values³¹. Singh *et al.*³² have shown the occurrence of diagenesis of Lower Siwalik palaeosols in the Katilu Khad section from the Kangra sub-basin. In a similar manner, we have found the

presence of sparite calcite veins and less micritic textures in the oldest analysed Lower Siwalik sediments in the J–N section, and as we move from the Lower Siwalik to Middle Siwalik sediments, finer-grained micritic textures become abundant, ultimately dominating the Upper Siwalik sediments (Figure 3).

However, various workers have cited that the diagenesis of sediments is more likely to alter the $\delta^{18}\text{O}$ values, rather than $\delta^{13}\text{C}$ values^{33–37}. The reasons for contrasting preservation of oxygen versus carbon isotopic compositions are well known and relate to the fact that a typical molar water/rock ratio between natural waters and limestone is 10³–10⁴ times larger for oxygen than for carbon³⁸. Hence, fluids are far more likely to alter $\delta^{18}\text{O}$ than $\delta^{13}\text{C}$ values of carbonates. This means $\delta^{18}\text{O}$ values of palaeosol carbonates will have larger probability of alteration by diagenesis than $\delta^{13}\text{C}$ values from the same bed. For example, co-existing micrite and sparite from buried palaeosol nodules in Eocene sediments in Wyoming showed that $\delta^{13}\text{C}$ values are essentially unchanged by recrystallization, but $\delta^{18}\text{O}$ values were often 4–8‰ depleted in ¹⁸O in sparite³³. In a similar manner, the palaeosol carbonates from the Eocene Willwood Formation show that the effects of recrystallization during diagenesis do not significantly shift the ¹³C content of nodules, although these nodules become depleted by upto 10‰ in ¹⁸O during diagenetic recrystallization³⁴. Also, Cerling *et al.*³⁹ illustrated that the original $\delta^{13}\text{C}$ values of organic matter and pedogenic carbonate from palaeosols of Pleistocene to late Miocene in Pakistan were not altered by diagenesis. In view of this, we assume insignificant alteration of $\delta^{13}\text{C}$ isotopic values due to diagenesis, if any, of the Lower Siwalik palaeosols.

Isotopic analysis: For isotopic analyses of the nodules, they were first washed in mild HCl to remove any modern carbonate sticking to the outer surface. The nodules were then powdered in agate mortar and left for drying in an oven prior to conversion to CO₂ with phosphoric acid (specific gravity 1.88). Isotopic analyses were performed on G. V. Instruments Isoprime Continuous Flow Mass Spectrometer at the National Institute of Hydrology, Roorkee, India. Samples were reacted in individual vials,

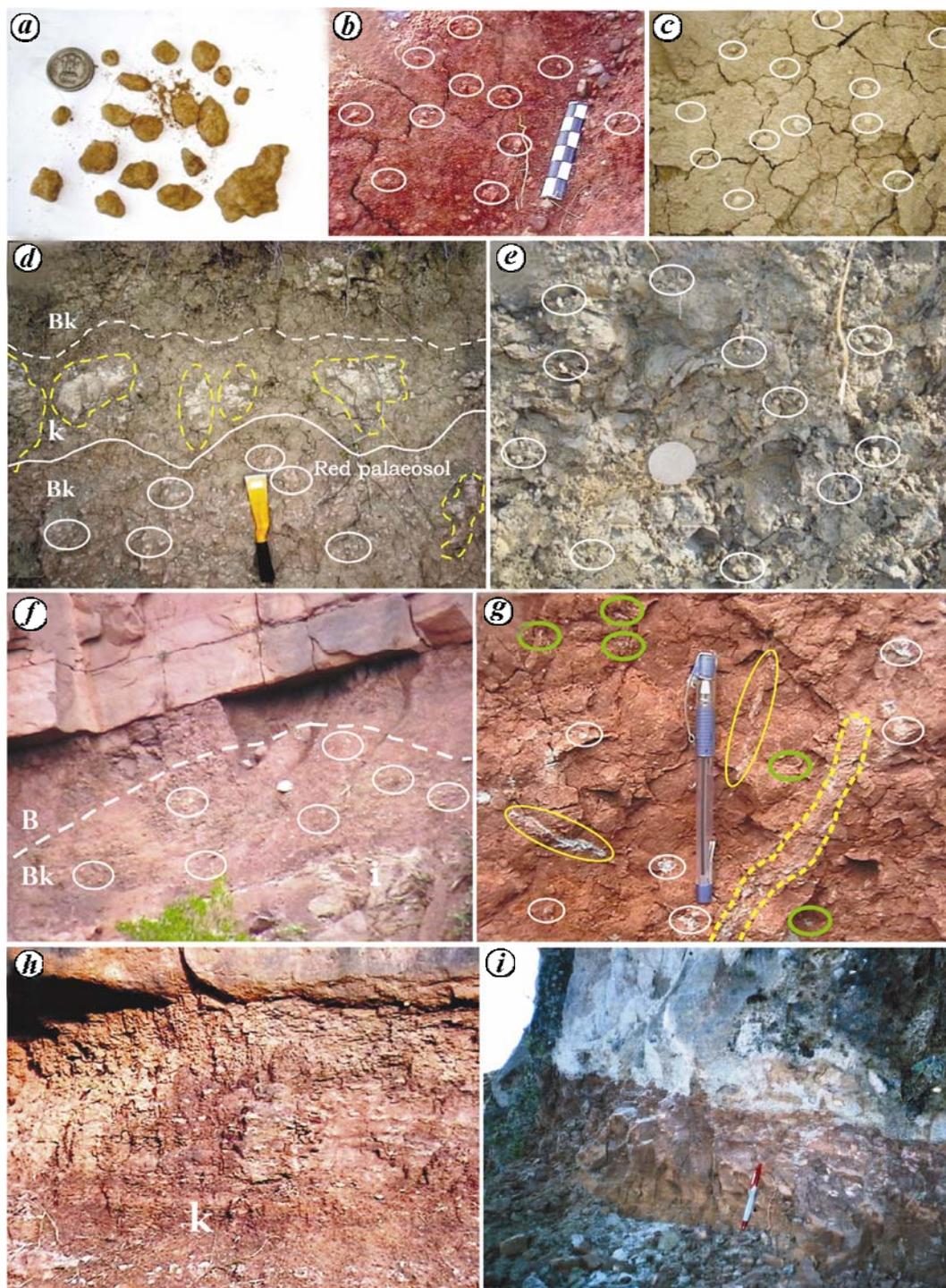


Figure 2. Different forms of pedogenic carbonate nodules in the palaeosols of the studied sections. The white, green and yellow ellipses indicate nodular, powdery and tubular/rhizome forms of carbonates respectively. Pedogenic carbonate nodules: (a) having pale-brown colour (10YR7/3) in the Upper Siwalik subgroup of the J–N section. Diameter of coin used as scale is ~2.2 cm; (b) In the red palaeosol of the Lower Siwalik subgroup in the J–N section, Jhajharkotli. Each white and black unit in the scale is ~1 cm; (c) In the yellow palaeosol of the Upper Siwalik Nagrota Formation in the J–N section; (d) In the lower red and upper gleyed palaeosol of the Upper Siwalik Nagrota Formation in the P–U section. Note the gradational nature of palaeosol horizons. Spade used as scale is ~26 cm; (e) In the gleyed palaeosol of the Upper Siwalik Nagrota Formation in the P–U section. Diameter of coin used as scale is ~2.4 cm; (f) In the moderately developed light-brown palaeosol of the Middle Siwalik subgroup in the J–N section. Note the presence of B horizon overlying the carbonate horizon (i.e. Bk); (g) In the palaeosol of the Upper Siwalik subgroup in the J–N section. Pen used as scale is ~15 cm; (h) Light brownish-red palaeosol showing shallow groundwater carbonate nodules in the Siwalik Group of the J–N section. Note the almost parallel alignment of the groundwater nodules along the bedding, indicating the direction of groundwater flow and the sharp contact between the lithologies, and (i) Shallow groundwater carbonate nodules in the Siwalik Group. Note the presence of sharp contacts and lack of associated soil horizons. Pen used as a scale is ~15 cm.

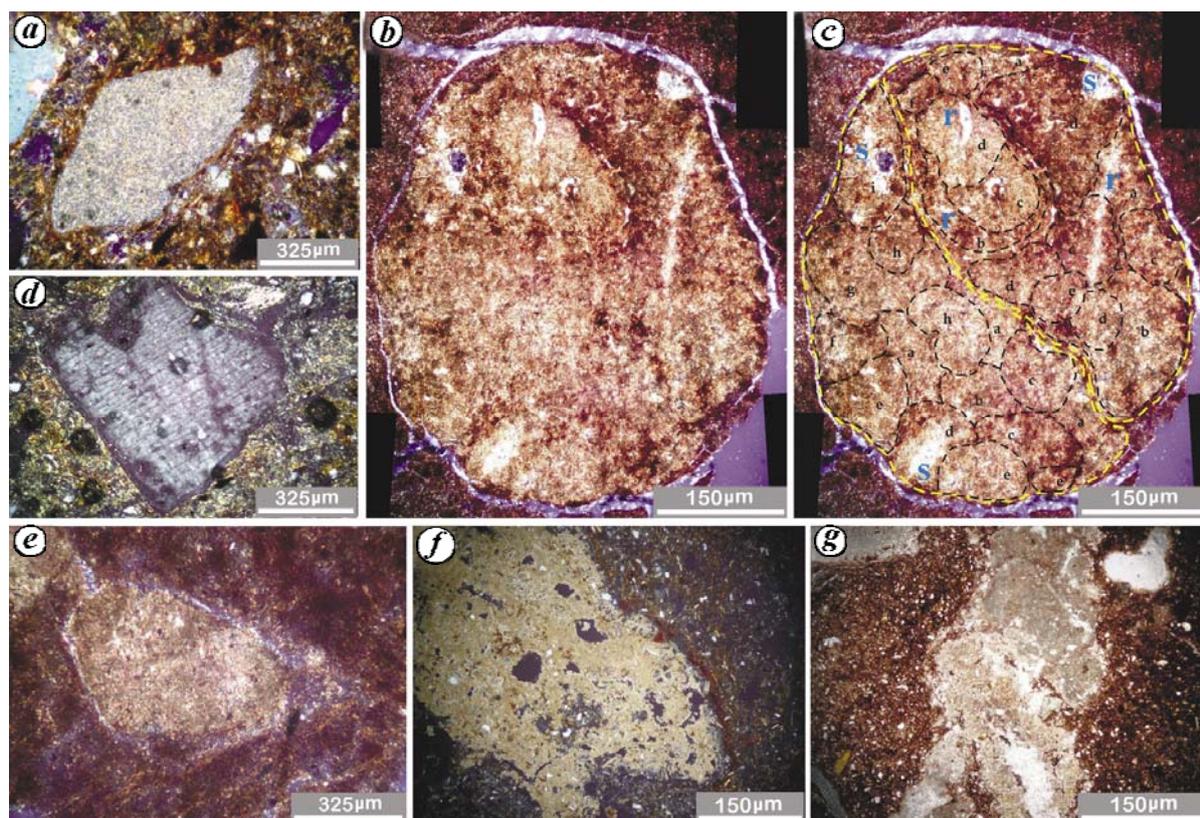


Figure 3. *a*, Typical micritic pedogenic carbonate nodule in the youngest analysed Upper Siwalik palaeosol, XPL. *b* and *c*, Large carbonate nodules present in well-developed ped consisting of micritic (m) to sparitic (s) textures during different stages of carbonate formation in the Upper Siwalik palaeosol. Note the sparitic to micro-sparitic filled abundant root traces (r). Smaller alphabets written in black represent the different stages of formation of the carbonate nodules, XPL. *d*, Well-preserved calcite grain in the Upper Siwalik palaeosol, XPL. *e*, Well-developed, largely micritic to micro-sparitic, carbonate nodule in the upper Middle Siwalik palaeosol, XPL. *f*, Micro-sparitic pedogenic carbonate nodule in the Middle Siwalik palaeosol. Note the ferran hypocoating and presence of primary minerals (largely quartz grains) in the nodule, XPL. *g*, Presence of abundant sparry calcite vein fillings in a channel/root in the palaeosols of the Lower Siwalik Formation in the J–N section, PPL. Note the presence of soil groundmass matrix in the carbonate.

eliminating the risk of cross-contamination from one sample to the next. Instrumental precisions are $\pm 0.12\text{‰}$ for $\delta^{13}\text{C}$ value and $\pm 0.15\text{‰}$ for $\delta^{18}\text{O}$ value.

Measurements of stable isotopes were done in terms of abundance ratios, i.e. atomic mass of heavy atom to the atomic mass of light atom. Only the relative difference in the ratio of the heavy isotope to the more abundant light isotope of the sample with respect to a standard (or reference) was determined. The difference was designated by δ . If the δ value is positive, it refers to the enrichment of the sample in the heavy-isotope species with respect to the standard, whereas a negative value corresponds to the sample depleted in the heavy-isotope species. Generally, δ units are quoted relative to an internationally recognized standard, which is arbitrarily set to 0‰. In the present study, the isotopic results for carbon are presented in the usual notation as the per mil (‰) deviation of the sample CO_2 from the VPDB standard, where $R = {}^{13}\text{C}/{}^{12}\text{C}$, using

$$\delta = [(R_{\text{sample}}/R_{\text{standard}}) - 1] \times 1000.$$

Other reference materials used in the isotopic analyses, whose values are referenced with respect to VPDB, are IAEA marble CO-1 ($\delta^{13}\text{C} = +2.48\text{‰}$; $\delta^{18}\text{O} = -2.44\text{‰}$); IAEA calcite CO-8 ($\delta^{13}\text{C} = -5.75\text{‰}$; $\delta^{18}\text{O} = -22.7\text{‰}$); BaCO_3 –Loba ($\delta^{13}\text{C} = -11.89\text{‰}$; $\delta^{18}\text{O} = -5.66\text{‰}$) and Na_2CO_3 –Loba ($\delta^{13}\text{C} = -25.62\text{‰}$; $\delta^{18}\text{O} = -17.46\text{‰}$).

Age assignments: A total of 93 samples (59 and 34 pedogenic carbonate nodules from J–N and P–U respectively) have been analysed. The analysed samples cover all the three subgroups of the Siwalik Group spanning a period from ~ 12 to ~ 0.4 Ma. Age estimates for different horizons in the studied Siwalik sections are based on palaeomagnetic dating^{3,40}. In both the studied sections, soil carbonate nodules were collected from the palaeosol horizons and their ages assigned from those of the corresponding palaeosol beds in the respective stratigraphic lithosections, whose ages are based on palaeomagnetic dates. The Lower–Middle Siwalik and Middle–Upper Siwalik boundaries in the J–N and P–U sections fall at 10.8 and 4.92 Ma respectively^{6,40}.

Results

The carbon isotopic record of pedogenic carbonates in palaeosol beds of the studied sections when plotted against age falls into two broad divisions; an older part characterized by $\delta^{13}\text{C}$ -depleted isotopic values ($< -6\text{‰}$) and a younger part showing $\delta^{13}\text{C}$ -enriched isotopic values ($> -6\text{‰}$), which are named as Zones I and III respectively (Figure 4). This change in $\delta^{13}\text{C}$ -values from the $\delta^{13}\text{C}$ -depleted isotopic values in Zone I to $\delta^{13}\text{C}$ -enriched isotopic values comprises a transition period, named as Zone II, of approximately 2 Ma (Figure 4). The period 12–7 Ma consists of Zone I comprising samples from the Lower Siwalik and the lower part of the Middle Siwalik subgroups having $\delta^{13}\text{C}$ values between -11.36 and -7.48‰ . The isotopic transition zone, i.e. Zone II, has $\delta^{13}\text{C}$ values -9.43 to -4.88‰ and consists of samples from the upper part of the Middle Siwalik subgroup covering an approximate age between 7 and 5 Ma. The younger part of the studied sections ranging in age between 5 and 0.40 Ma, named as Zone III, comprises samples from the Upper Siwalik subgroup. The $\delta^{13}\text{C}$ isotopic values in Zone III vary from -6.18 to $+2.18\text{‰}$.

Discussion and conclusion

A large number of workers have done palaeovegetational reconstructions from the carbon isotope ratios of

pedogenic carbonates in soils. But how the $\delta^{13}\text{C}$ values are useful in palaeovegetational reconstructions and their application in the present study is discussed below in detail.

Selective intake of ^{12}C and ^{13}C by different types of vegetation

The stable carbon isotope ratio of soil organic matter/humus and pedogenic carbonate in palaeosols can be directly used to reconstruct palaeovegetation present during pedogenesis^{33,39}. This is because plants metabolize and in the process fractionate carbon isotopes along three distinct photosynthetic pathways^{41,42}. This means different types of vegetation present during soil formation can be distinguished broadly into three plant groups on their characteristic carbon isotope ratios.

The stable isotopes ^{12}C and ^{13}C are found in rocks and palaeosols of all geological ages⁴³. These carbon isotopic values ($\delta^{13}\text{C}$) are affected by a variety of physical, chemical and biological processes. The key photosynthetic enzyme of plants, Rubisco, selects the light isotope (^{12}C) preferentially to the heavy isotope (^{13}C), so that plant organic matter is isotopically much lighter ($\delta^{13}\text{C}$ more negative) than the atmospheric or oceanic CO_2 from which it was derived. The $\delta^{13}\text{C}$ values of pedogenic carbonate are 14–17‰ enriched than the plant organic matter. Majority of the continental plants (i.e. virtually all trees, shrubs, herbs and a few grasses favoured by a cool growing season) employ a photosynthetic pathway creating at first a three-carbon phosphoglyceric acid using C_3 or Calvin–Benson photosynthetic cycle. Plants which use a photosynthetic pathway creating at first a four-carbon malic and aspartic acid are called C_4 plants, as they use C_4 or Hatch–Slack photosynthetic cycle. C_4 plants include most of the grasses favoured by a warm growing season and a few shrubs in the families Euphorbiaceae and Chenopodiaceae. The C_3 plants fractionate carbon isotopes more intensely, and so have more negative $\delta^{13}\text{C}$ values than the C_4 plants⁴³. The $\delta^{13}\text{C}$ values for C_3 plants range from -22‰ to -38‰ with an average of -27‰ , whereas values for C_4 plants range from -9‰ to -21‰ with an average value of -13‰ , and hence these distinct $\delta^{13}\text{C}$ isotopic values between C_3 and C_4 plants make them different from each other^{44–46}. The third type of plants have crassulacean acid metabolism (CAM) photosynthetic cycle, which includes desert succulents⁴⁷ and shows a natural range of $\delta^{13}\text{C}$ values between -10‰ and -20‰ . This photosynthetic pathway is in some ways a combination of C_3 and C_4 pathways, but these plants are geographically restricted⁴⁸ and are not important components of any ecosystem outside deserts²¹. In view of this and the fact that the analysed samples do not form part of the desert ecosystem, these CAM plants are not taken into account in the present study.

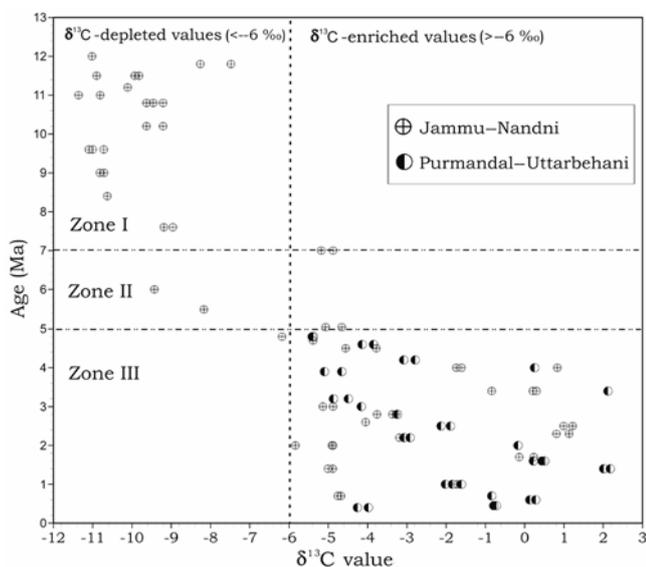


Figure 4. Plot showing variation of $\delta^{13}\text{C}$ values of pedogenic carbonates of the studied sections against age. It divides the $\delta^{13}\text{C}$ values into two broad divisions; an older part characterized by $\delta^{13}\text{C}$ -depleted isotopic values ($< -6\text{‰}$) and a younger part showing $\delta^{13}\text{C}$ -enriched isotopic values ($> -6\text{‰}$), which are named as Zones I and III respectively. This change in $\delta^{13}\text{C}$ values with age from the $\delta^{13}\text{C}$ -depleted isotopic values in Zone I to $\delta^{13}\text{C}$ -enriched isotopic values comprises a transition period, named as Zone II, of approximately 2 Ma. The $\delta^{13}\text{C}$ values are expressed in per mil (‰).

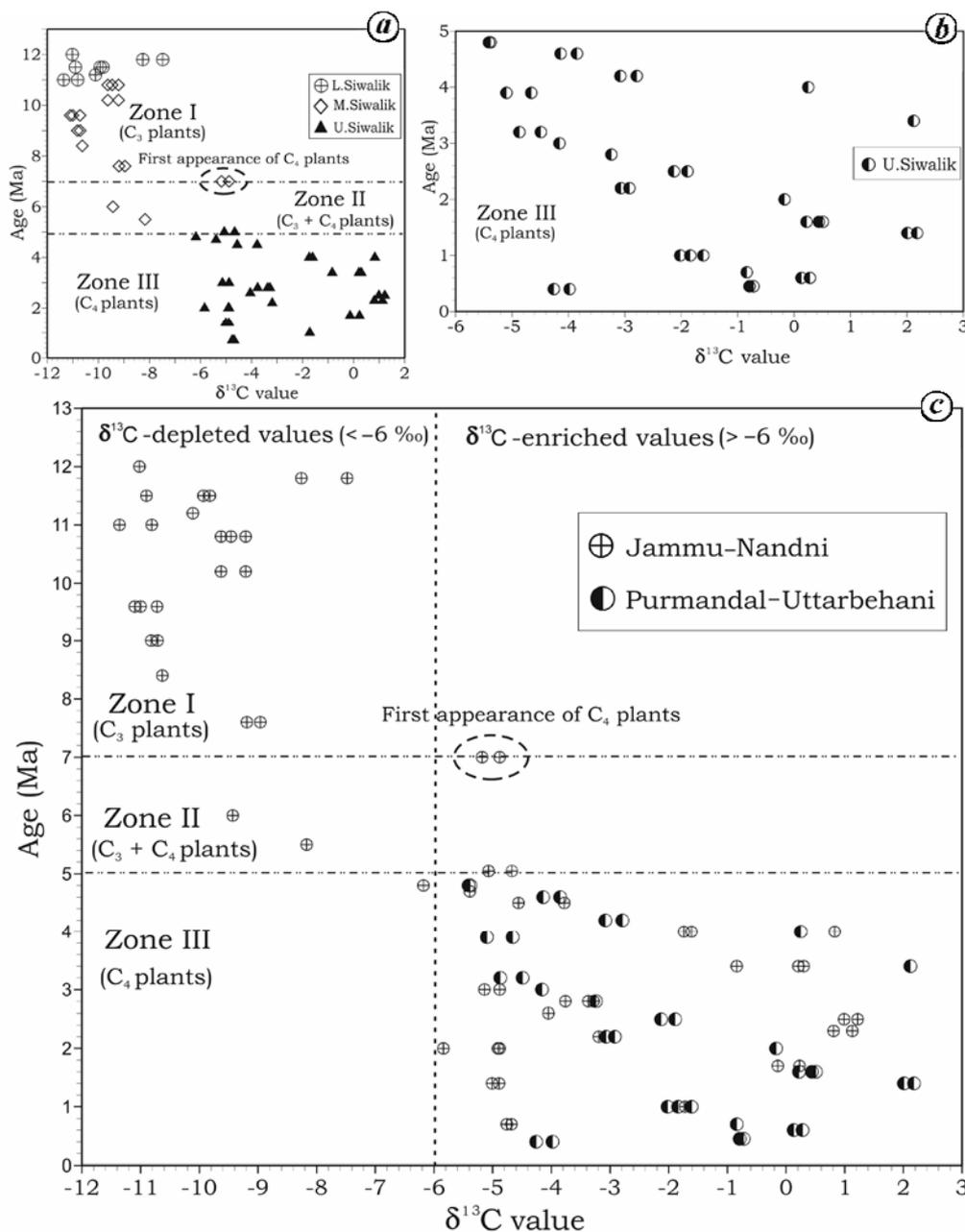


Figure 5. The $\delta^{13}\text{C}$ values of pedogenic carbonate nodules plotted against age in the (a) J-N and (b) P-U sections. Dotted ellipse marks the first appearance of C_4 plants at 7 Ma in the J-N section. (c) Composite plot of $\delta^{13}\text{C}$ values against age from the studied sections. It divides the $\delta^{13}\text{C}$ values into two broad divisions; an older part characterized by $\delta^{13}\text{C}$ -depleted isotopic values ($< -6\text{‰}$) and a younger part showing $\delta^{13}\text{C}$ -enriched isotopic values ($> -6\text{‰}$), which are named as Zones I and III respectively. Zone I indicates the exclusive presence of C_3 plants, whereas Zone III indicates the exclusive presence of C_4 plants on the basis of $\delta^{13}\text{C}$ values. This change in vegetation with age from C_3 to C_4 plants comprises a transition period, named as Zone II, in which the presence of both C_3 and C_4 plants has been found. Dotted ellipse marks the first appearance of C_4 plants at 7 Ma in the J-N. The $\delta^{13}\text{C}$ values are expressed in per mil (‰).

Palaeovegetational reconstruction in the studied sections

J-N section: A total of 59 pedogenic carbonate samples were analysed from the J-N section, which ranges in age between ~ 12 and 0.73 Ma. The $\delta^{13}\text{C}$ values vary from -11.36‰ to $+1.22\text{‰}$, and when plotted against age fall into three distinct zones: Zones I–III, characterized by the

presence of $\delta^{13}\text{C}$ -depleted isotopic values, both $\delta^{13}\text{C}$ -depleted and -enriched isotopic values, and $\delta^{13}\text{C}$ -enriched isotopic values respectively (Figure 5a). The $\delta^{13}\text{C}$ values vary in age from ~ 12 to 7.5 Ma. The depleted or negative nature of $\delta^{13}\text{C}$ values in this zone imply vegetation composed of pure or nearly pure C_3 plants and is also corroborative with $\delta^{13}\text{C}$ value range for C_3 plants. The $\delta^{13}\text{C}$ values

Table 2. Approximate time of first appearance of C₄ plants in different parts of the earth, which shows the global nature of expansion of C₄ plants during late Miocene

Place and latitude	Time of first appearance of C ₄ plants (Ma)	Reference
Asia (Siwalik)		
Jammu–Nandni, Jammu and Kashmir, India; 32°N	7	Present study
Kangra Valley, Ranital–Kotla section, India; 32°N	6	15
Potwar Plateau, Pakistan; 32°–33°N	7.4–7	21
Surai Khola, Nepal; ~27°N	6.8	23
East Africa; 3°S–5°N		
Central Africa; ~16°N	8	48
South Africa; 24°–25°S	7	
North America; 20°–37°N		
North America; 40°–43°N	5	
South America; 28°–32°S	6.8–5.5	50
	4	
	7.3–6.7	52

vary between -9.43% and -4.88% in Zone II, ranging in age from 7 to 5 Ma. These $\delta^{13}\text{C}$ values indicate the presence of both C₃ and C₄ plants, and the first appearance of C₄ plants started in this transition period at ~7 Ma, with $\delta^{13}\text{C}$ value of -4.88% (Figure 5a). Zone III comprising 5–0.7 Ma shows the presence of $\delta^{13}\text{C}$ -enriched isotopic values varying between -6.18% and 1.22% . The $\delta^{13}\text{C}$ values indicate the complete dominance of C₄ vegetation after 5Ma (Figure 5a).

P–U section: A total of 34 pedogenic carbonate samples were analysed from this section, which ranges in age between ~4.8% and ~0.40 Ma. The $\delta^{13}\text{C}$ values are highly positive or enriched, and when plotted against age fall in Zone III of the J–N section. The $\delta^{13}\text{C}$ values vary between -5.41% and $+2.18\%$, and indicate the exclusive presence of C₄ plants (Figure 5b).

The composite plot of $\delta^{13}\text{C}$ values against age in the J–N and P–U sections indicate that an older part characterized by $\delta^{13}\text{C}$ -depleted isotopic values ranging in age between 12 and 7 Ma comprises exclusively of C₃ vegetation (i.e. Zone I; Figure 5c). In a similar manner, the younger part ranging in age between 5 and 0.4 Ma, named as Zone III, consists of $\delta^{13}\text{C}$ -enriched values and hence indicates exclusive presence of C₄ vegetation (Figure 5c). Hence, the study area was dominated by C₃ and C₄ vegetation pre-7 Ma and post-5 Ma respectively, with a narrow transition period of 2 Ma (i.e. 7–5 Ma) consisting of both C₃ and C₄ plants (i.e. Zone II; Figure 5c).

Palaeovegetational change during late Miocene: local or global

It has been shown earlier that pre-7 Ma there was exclusive presence of C₃ plants and then at 7 Ma the first appearance of C₄ plants took place, which became dominant by 5 Ma. What becomes interesting is that this dramatic change in vegetation during late Miocene was not a local phenomenon, but rather global. This is

because it has been shown by many workers that the carbon isotopic trends of palaeosol carbonate, fossil mammal tooth enamel and terrestrially derived organic matter from Pakistan, South America and North America, and Africa indicate a period of substantial ecological change to C₄-dominated vegetation between 8 and 4 Ma (ref. 49). The present data also reveal a pronounced change in vegetation from C₃ to C₄ after 7 Ma. This striking change from C₃ to C₄ vegetation was first noted in palaeosol carbonates in the Siwalik sediments of the Potwar Plateau, Pakistan²¹. Subsequent studies from other parts of the earth showed that this change in vegetation from C₃ to C₄ at different places did not occur abruptly, but gradually within a narrow time range largely from 8 to 5 Ma (Table 2). This difference is attributed to latitudinal variation of places on the earth. Compared to high-latitude areas, the change occurred earlier at lower latitudes where the threshold for C₃ photosynthesis was higher at elevated temperatures⁵⁰. Table 2 shows this latitudinal variation and global nature of C₄ expansion during the Upper Miocene.

The evidences thus far indicate that the change from C₃ to C₄ vegetation in the late Miocene was a major palaeo-ecologic event in the earth's terrestrial history and the causes behind this evolutionary phenomenon have remained controversial since its discovery. To find out the possible drivers of C₄ expansion, we must first have more such studies from the Indian Siwaliks which are well exposed all along the Himalayan belt. The present work constitutes a small part from the Siwaliks in the Ramnagar sub-basin and provides opportunities for further studies in this region as it offers an important link to the extensive palaeovegetational studies done in the Pakistan and Nepal Siwaliks.

1. Raiverman, V., *Foreland Sedimentation in Himalayan Tectonic Regime: A Relook at the Orogenic Process*, Bishen Singh Mahendra Pal Singh, Dehradun, 2002, p. 378.
2. Shukla, A., *Palaeopedology of the Overbank intervals of the Lower Siwalik Subgroup (Kathgodam–Amritpur Section of Kumaun Himalaya, India)*. M.Sc. dissertation, University of Delhi, Delhi, 1984, p. 57.

3. Ranga Rao, A., Aggarwal, R. P., Sharma, U. N., Bhalla, M. S. and Nanda, A. C., Magnetic polarity stratigraphy and vertebrate paleontology of the Upper Siwalik Subgroup of Jammu Hills, India. *J. Geol. Soc. India*, 1988, **31**(4), 361–385.
4. Kumar, R. and Nanda, A. C., Sedimentology of the Middle Siwalik Subgroup of Mohand area, Dehradun valley, India. *J. Geol. Soc. India*, 1989, **34**(6), 597–616.
5. Aggarwal, R. P., Nanda, A. C., Prasad, D. N. and Dey, B. K., Geology and biostratigraphy of the Upper Siwalik of the Samba area, Jammu foothills. *J. Himalayan Geol.*, 1993, **4**(2), 227–236.
6. Pandita, S. K. and Bhat, G. M., Temporal patterns of palaeoflow of Middle and Upper Siwalik Subgroups, Jammu. *J. Geol. Soc. India*, 1996, **48**, 211–219.
7. Bhat, G. M., Pandita, S. K. and Singh, R., Record of anastomosing fluvial and lacustrine deposition in the Upper Siwalik of Jammu, NW Himalaya, India. *J. Nepal Geol. Soc.*, 1997, **16**, 63–64.
8. Thomas, J. V., Parkash, B. and Mohindra, R., Lithofacies and palaeosol analysis of the Middle and Upper Siwalik Groups (Plio–Pleistocene), Haripur–Kolar section, Himachal Pradesh, India. *Sediment. Geol.*, 2002, **150**, 343–366.
9. Singh, T., Sharma, U. and Kumar, R., Soft sediment deformation in the Morni area, NW Sub-Himalaya. *Curr. Sci.*, 2007, **93**, 1151–1155.
10. Singh, T., Tectonic implications of geomorphometric characterization of watersheds using spatial correlation: Mohand Ridge, NW Himalaya, India. *Z. Geomorphol.*, 2008, **52**, 489–502.
11. Johnson, G. D., Palaeopedology of Ramapithecus-bearing sediments, North India. *Geol. Rundsch.*, 1977, **66**, 192–216.
12. Retallack, G. J., Paleosols of the Siwalik Group as a 15 Myr record of South Asian palaeoclimate. *Mem. Geol. Soc. India*, 1995, **32**, 36–51.
13. Ganjoo, R. K., Verma, R. K. and Achyuthan, H., Lower Pleistocene palaeosols of the Upper Siwaliks in Jammu: evidence of seasonality. *J. Geol. Soc. India*, 2002, **60**, 371–379.
14. Ghosh, P., Padia, J. T. and Mohindra, R., Stable isotopic studies of paleosol sediment from Upper Siwalik of Himachal Himalaya: evidence from high monsoonal intensity during late Miocene? *Palaeogeogr. Palaeoclimatol. Palaeoecol.*, 2004, **206**, 103–114.
15. Sanyal, P., Bhattacharya, S. K., Kumar, R., Ghosh, S. K. and Sangode, S. J., Mio–Pliocene monsoonal record from Himalayan foreland basin (Indian Siwalik) and its relation to the vegetational change. *Palaeogeol. Palaeoclimatol. Palaeoecol.*, 2004, **205**, 23–41.
16. Kumaravel, V., Sangode, S. J., Siddaiah, N. S. and Kumar, R., Major element geochemical variations in a Miocene–Pliocene Siwalik paleosol sequence: implications to soil forming processes in the Himalayan foreland basin. *J. Geol. Soc. India*, 2009, **73**(6), 759–772.
17. Raymo, M. E. and Ruddiman, W. F., Tectonic forcing of late Cenozoic climate. *Nature*, 1992, **359**, 117–122.
18. Wang, C. Y., Shi, Y. L. and Zhou, W. H., Dynamic uplift of the Himalayas. *Nature*, 1982, **289**, 553–556.
19. Conroy, G., West, R. J. and Munthe, J., The Siwaliks of Nepal: recent contributions to vertebrate paleontology and biostratigraphy. In *Contributions to Himalayan Geology. 3. Geology of the Western Himalayas* (ed. Gupta, V. J.), Hindustan Publ., New Delhi, 1985, pp. 52–61.
20. Behrensmeyer, A. K. *et al.*, The structure and rate of late Miocene expansion of C₄ plants: evidence from lateral variation in stable isotopes in palaeosols of the Siwalik Group, northern Pakistan. *Geol. Soc. Am. Bull.*, 2007, **119**, 1486–1505.
21. Quade, J., Cerling, T. E. and Bowman, J. R., Development of Asian monsoon revealed by marked ecological shift during the latest Miocene in the northern Pakistan. *Nature*, 1989, **342**, 163–166.
22. Quade, J. and Cerling, T. E., Expansion of C₄ grasses in the Late Miocene of northern Pakistan: evidence from stable isotopes in palaeosols. *Palaeogeol. Palaeoclimatol. Palaeoecol.*, 1995, **115**, 91–116.
23. Quade, J., Cater, M. L. J., Ojha, P. T., Adam, J. and Harrison, M. T., Dramatic carbon and oxygen isotopic shift in paleosols from Nepal and late Miocene environmental change across the northern Indian sub-continent. *Geol. Soc. Am. Bull.*, 1995, **107**, 1381–1397.
24. Thomas, J. V., Parkash, B., Mohindra, R., Bhattacharya, S. K. and Jani, R. A., Syndepositional tectonics and paleoclimatic implications of paleosols of the Plio–Pleistocene Siwalik Group, Haripur–Kolar section, Himachal Pradesh, India. *Himalayan Geol.*, 2005, **26**, 327–345.
25. Birkeland, P. W., *Soils and Geomorphology*, Oxford University Press, New York, 1984.
26. Mack, G. H., Cole, D. R. and Trevino, L., The distribution and discrimination of shallow, authigenic carbonate in the Pliocene–Pleistocene Palomas Basin, southern Rio Grande rift. *Geol. Soc. Am. Bull.*, 2000, **112**, 643–656.
27. Wright, V. P. and Tucker, M. E., *Calcretes*, International Association of Sedimentologists Reprint Series, Blackwell, Scientific, Oxford, 1991.
28. Pal, D. K., Dasog, G. S., Vadivelu, S., Ahuja, R. L. and Bhattacharya, T., Secondary calcium carbonate in soils of arid and semi-arid regions of India. In *Global Climate Change and Pedogenic Carbonates* (eds Lal, R. *et al.*), Lewis, USA, 2000, pp. 149–185.
29. Retallack, G. J., Field recognition of palaeosols. *Geol. Soc. Am. Spec. Publ.*, 1988, **216**, 1–20.
30. Sehgal, J. L. and Stoops, G., Pedogenic calcite accumulation in arid and semi-arid regions of the Indo-Gangetic alluvial plain of Erstwhile Punjab (India)—their morphology and origin. *Geoderma*, 1972, **8**, 59–72.
31. Andrews, J. E., Singhvi, A. K., Kailath, A. J., Kuhn, R., Dennis, P. F., Tandon, S. K. and Dhir, R. P., Do stable isotope data from calcrete record late Pleistocene monsoonal climate variation in the Thar desert of India. *Quaternary Res.*, 1998, **50**, 240–251.
32. Singh, S., Parkash, B. and Awasthi, A. K., Origin of red colour of Lower Siwalik palaeosols: A micromorphological approach. *J. Mt. Sci.*, 2009, **6**, 147–154.
33. Cerling, T. E., The stable isotopic composition of modern soil carbonate and its relationship to climate. *Earth Planet. Sci. Lett.*, 1984, **71**, 229–240.
34. Cerling, T. E., Carbon dioxide in the atmosphere: evidence from Cenozoic and Mesozoic paleosols. *Am. J. Sci.*, 1991, **291**, 377–400.
35. Driese, S. G. and Mora, C. I., Physico-chemical environment of pedogenic carbonate formation in Devonian vertic palaeosols, central Appalachians, USA. *Sedimentology*, 1993, **40**, 199–216.
36. Morrill, C. and Koch, P. L., Elevation or alteration? Evaluation of isotopic constraints on paleoaltitudes surrounding the Eocene Green River Basin. *Geology*, 2002, **30**, 151–154.
37. Garzzone, C. N., Dettman, D. L. and Horton, B. K., Carbonate oxygen isotope paleoaltimetry: evaluating the effect of diagenesis on paleoelevation estimates for the Tibetan plateau. *Palaeogeogr. Palaeoclimatol. Palaeoecol.*, 2004, **212**, 119–140.
38. Banner, J. L. and Hanson, G. N., Calculation of simultaneous isotopic and trace-element variations during rock–water interaction with applications to carbonate diagenesis. *Geochim. Cosmochim. Acta*, 1990, **54**, 3123–3137.
39. Cerling, T. E., Quade, J., Wang, Y. and Bowman, J. R., Carbon isotopes in soils and palaeosols as ecology and paleoecology indicators. *Nature*, 1989, **341**, 138–139.
40. Rao, R. A., Magnetic polarity stratigraphy of the Upper Siwalik of northwestern Himalayan foothills. *Curr. Sci.*, 1993, **64**, 863–872.
41. Smith, B. N. and Epstein, S., Two categories of ¹³C/¹²C ratios for higher plants. *Plant Physiol.*, 1971, **47**, 380–384.
42. Bender, M. M., Rouhani, I., Vines, H. M. and Black, C. C., ¹³C/¹²C ratio changes in crassulacean acid metabolism. *Plant Physiol.*, 1973, **52**, 427–430.

RESEARCH ARTICLES

43. Retallack, G. J., Soils and global change in the carbon cycle over geological time. *Treatise Geochem.*, 2003, **5**, 581–603.
44. Raven, P. H., Evert, R. F. and Curtis, H., *Biology of Plants*, Worth Publishers, New York, 1981, pp. 106–112.
45. Salisbury, F. B. and Ross, C. W., *Plant Physiology*, Wadsworth Publishing Company, Belmont, CA, 1985, pp. 197–225.
46. O'Leary, M. H., Biochemical basis of carbon isotope fractionation. In *Stable Isotopes and Plant Carbon–Water Relations* (eds Ehleringer, J. R., Hall, A. E. and Farquhar, G. D.), Academic Press, 1993, pp. 19–28.
47. Lerman, J. C., Soil-CO₂ and groundwater: carbon isotope composition. In *Proceedings of the 8th International Conference on Radiocarbon Dating*, Lower Hutt, New Zealand, 1972, pp. H16–H28.
48. Selagen, L., Lee Thorp, J. A. and Cerling, T. E., Timing of C₄ grass expansion across sub-Saharan Africa. *J. Hum. Evol.*, 2007, **53**, 549–559.
49. Pagani, M., Freeman, K. H. and Arthur, A. A., Late Miocene atmospheric CO₂ concentrations and the expansion of C₄ grasses. *Science*, 1999, **285**, 876–879.
50. Cerling, T. E. *et al.*, Global vegetation change through the Miocene/Pliocene boundary. *Nature*, 1997, **389**, 153–157.
51. Dasarathi, N., A note on certain geological aspects of Tawi Valley Tertiaries. *Kashmir Sci.*, 1968, **5**, 222–232.
52. Latorre, C., Quade, J. and McIntosh, W. C., The expansion of C₄ grasses and global change in the late Miocene: stable isotope evidence from the Americas. *Earth Planet. Sci. Lett.*, 1997, **146**, 83–96.
53. Dey, B. K., Mitra, D. S., Agarwal, R. P., Dotiwala, S. F. and Prasad, D. N., Spectral mapping of Siwalik sediments of a part of Jammu foothills. *J. Geol. Soc. India*, 1994, **44**, 193–202.

ACKNOWLEDGEMENTS. We thank the Head, Department of Earth Sciences, Indian Institute of Technology, Roorkee and the Director, National Institute of Hydrology, Roorkee, for providing the necessary facilities for this research work. We also thank Dr Bishm Kumar and Dr M. S. Rao, NIH, Roorkee and Dr Tejpal Singh, Centre for Mathematical Modelling and Computer Simulation, Bangalore for his extensive help during field work and informative discussions. S.S. thanks CSIR, New Delhi for providing a fellowship.

Received 25 November 2009; revised accepted 3 November 2010
