Paleoclimatic, paleovegetational and provenance change in the Ganga Plain during the late Quaternary

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Present study aims at reconstructing the paleomonsoonal rainfall, paleovegetation and provenance change during the late Quaternary. Towards this, Bhognipur core, collected from the southern Ganga Plain, have been sampled for soil carbonate (SC) and soil. The δ^{18}O values of SC ($\delta^{18}O_{SC}$) range from −7.6 to −4.9‰. The variations in $\delta^{18}O_{SC}$ values suggest that during the late Quaternary, the monsoon intensified during MIS 3 and MIS 1 and the maximum lowering of rainfall intensity is observed during MIS 2. The δ^{13}C value of SC ($\delta^{13}C_{SC}$), organic matter dispersed in the soil ($\delta^{13}C_{SOM}$) and occluded in the carbonate nodules ($\delta^{13}C_{NOM}$) ranges from −4.1 to +1.4‰, −25.6 to −16.3‰, and −27.7 to −25.0‰, respectively, implies mixed C_3–C_4 vegetation over the Ganga Plain. Variations in $\delta^{13}C_{SOM}$ and $\delta^{13}C_{NOM}$ values at same depth imply preservation problem of pristine organic matter signature. Therefore, it is important to assess the preservation of residual organic matter before using it for paleovegetational reconstruction. The monsoon-vegetation relationship indicates that relative abundances of C_3–C_4 vegetation were mainly driven by variations in monsoonal rainfall intensity. Using ^{87}Sr/^{86}Sr in SC, we show that the Himalayan river was supplying sediments in the southern part of the Ganga Plain during MIS 3.

1. Introduction

The paleoclimate reconstruction for the late Quaternary period (last 500 ka) from marine sediments showed that glacial periods (marine isotope stages 2, 4, 6) were characterized by weaker summer monsoon and interglacial periods (marine isotope stages 1, 3, 5) by stronger summer monsoon (Prell and Kutzbach 1987; Clemens and Prell 1990; Clemens *et al.* 1991, 1996). Most of these studies used proxy records that respond to wind-induced upwelling, and therefore essentially relate to the variation of wind speed. However, the amount of rainfall depends on the moisture content and transportation path of the monsoon wind rather than the wind speed (Clemens *et al.* 1996; Sarkar *et al.* 2000). Studies pertaining to the past monsoon reconstruction based on continental sequences (a direct proxy for rainfall) are limited (Andrews *et al.* 1998; Sinha *et al.* 2006; Agrawal *et al.* 2012).

Keywords. Ganga Plain; monsoonal rainfall; isotopic ratio; C_3–C_4 plants; late Quaternary; soil carbonate.

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In this situation, sediments of the Ganga Plain provide an excellent opportunity to reconstruct past rainfall variations as it falls on the track of Indian summer monsoon. The sediments in the Ganga Plain are mainly deposited by various perennial and monsoon-fed rivers (Ganga, Yamuna, Chambal, Betwa, etc., figure 1a) which originate from the Himalaya and peninsular India. During the pause in sedimentation, authegenic minerals like soil carbonate (SC) gain the oxygen isotopic signature of rain water and provide useful information regarding the past rainfall variations (Cerling 1984; Quade *et al.* 1989; Stern *et al.* 1997).

It would also be interesting to see the effect of monsoonal rainfall variations on vegetation. Photosynthetic pathways of plants have evolved with time. Early vegetation was characterized by Calvin cycle (C_3 plants). During the late Miocene, a new photosynthetic pathway, i.e., Hatch-Slack cycle (C_4 plants) evolved in low-latitude areas across the globe (Hatch and Osmond 1976; Taiz and Zeiger 1998; Sage 2001). The C_4 plants outcompete C_3 plants when water is limited, temperature is high, and pCO_2 is low. The relative importance of these factors for the evolution and expansion of C_4 plants during the late Miocene time is still a matter of debate (Cerling *et al.* 1997; Pagani *et al.* 1999, 2005; Sanyal *et al.* 2004, 2005, 2010) mostly due to the lack of high resolution temporal pCO_2 data. But, the pCO_2 variations are well constrained from ice core records (Ahn and Brook 2008) for the late Quaternary period. Therefore, the late Quaternary fluvial sedimentary deposits can provide opportunity to understand the role of pCO_2 and monsoonal rainfall on the relative abundance of C_3 and C_4 plants (Andrews *et al.* 1998; Huang *et al.* 2001; Galy *et al.* 2008; Sarkar *et al.* 2009; Agrawal *et al.* 2012). The reconstruction of vegetation can be done using carbon isotope ratio of SC and organic matter trapped within soil carbonate nodules (NOM) (Cerling 1984; Cerling *et al.* 1989, 1993). However, limited occurrence of carbonate nodules within the soil profile is a potential obstacle for such reconstruction

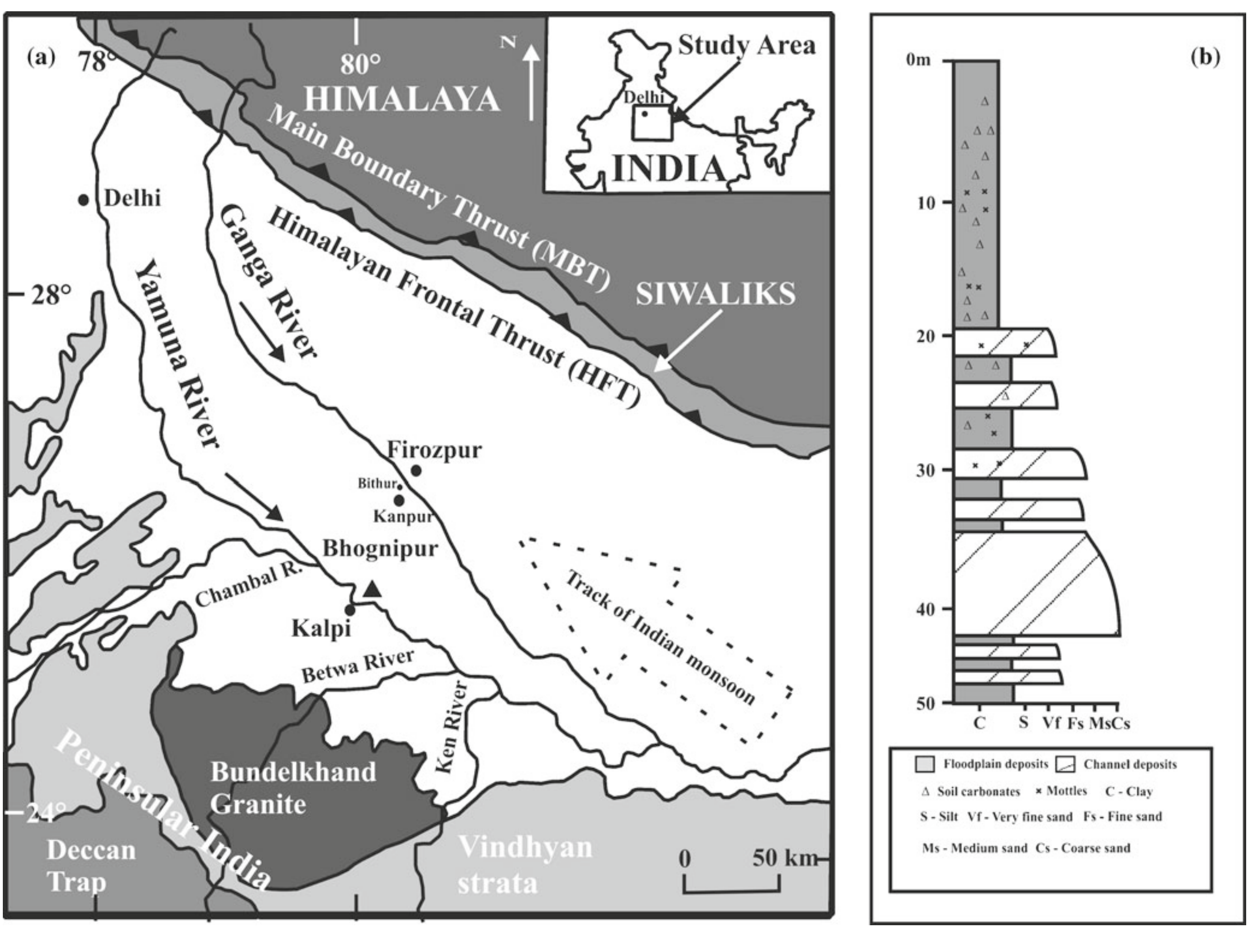

Figure 1. (**a**) Part of the Ganga Plain in Himalayan foreland basin (after Gibling *et al.* 2005) showing position of Himalayan and peninsular rivers and location of the Bhognipur core (solid triangle). (**b**) Litholog of the Bhognipur core collected from the southern part of the Ganga Plain (modified after Srivastava *et al.* 2010).

because calcium carbonates do not precipitate under tropical conditions where annual rainfall excess of 2000 mm/yr (Birkeland 1999). Thus, carbon isotope ratio of residual organic matter dispersed in paleosols (SOM) can be used to reconstruct past vegetation. Therefore, $\delta^{18}O_{SC}$ based monsoon rainfall reconstruction, and $\delta^{13}C_{SC}$, $\delta^{13}C_{SOM}$ and $\delta^{13}C_{NOM}$ values-based vegetation reconstructions can be used to glean out the effect of monsoonal rainfall on vegetation during the late Quaternary time.

It is also important to understand the fluvial response to the climatic changes. The Himalaya, a young orogenic belt, transfers large amounts of particulate and dissolved materials to the Ganga Plain and to the Bay of Bengal via Himalayan rivers (Galy and France-Lanord 2001; Singh *et al.* 2008). A significant amount of sediment is also supplied from the peninsular Indian rivers like Chambal and Betwa which drain through strikingly different lithologies. The Sr isotope compositions of the Himalayan and peninsular Indian rocks are quite distinct which is reflected by the Himalayan and peninsular India river water (Krishnaswami *et al.* 1992; Palmer and Edmond 1992; Dalai *et al.* 2003; Rengarajan *et al.* 2009; Rai *et al.* 2010). For instance, the Sr isotope ratio of the Yamuna river water at the foot hills of Himalaya (Saharanpur) is 0.7266 (Dalai *et al.* 2003) whereas Chambal, a peninsular river, is quite unradiogenic in Sr (Rengarajan *et al.* 2009). The Chambal river contributes 84 ± 17% of the dissolved strontium to the Yamuna river (Rengarajan *et al.* 2009) with a $^{87}Sr/^{86}Sr$ ratio 0.7111–0.7115 (Palmer and Edmond 1992). It causes dilution of the $^{87}Sr/^{86}Sr$ ratio in the Yamuna river which in turn lowers the $^{87}Sr/^{86}Sr$ ratio of the Yamuna river water to 0.7135–0.7149 (Rai *et al.* 2010). However, information related to past configuration of Himalayan and peninsular rivers within the Ganga Plain and their relative contribution in the geological past is not available to understand the evolution of sedimentary sequences in the Ganga Plain. Thus, characterizing sediments by the Sr isotope ratios in soil carbonate, which represents the contemporaneous $^{87}Sr/^{86}Sr$ ratio of river water, the source rock can be indentified (Quade *et al.* 1997). Characterization of sources of sediments along with rainfall variation data can serve opportunity to delineate the past configuration of the Himalayan and peninsular Indian rivers and relative role of climate and tectonics on evolution of sedimentary sequences of the Ganga Plain. Therefore, in this study, Bhognipur core (figure 1b) which represents southern interfluve margin area (northern bank of Yamuna river; figure 1a) of the Ganga Plain have been sampled for SC nodules (figure 2a) and soil. $\delta^{18}O$, $\delta^{13}C$ values, concentration of Sr and $^{87}Sr/^{86}Sr$ ratios are measured in SC. In addition, $\delta^{13}C$ values in SOM and NOM have been measured. The main objectives of this study are:

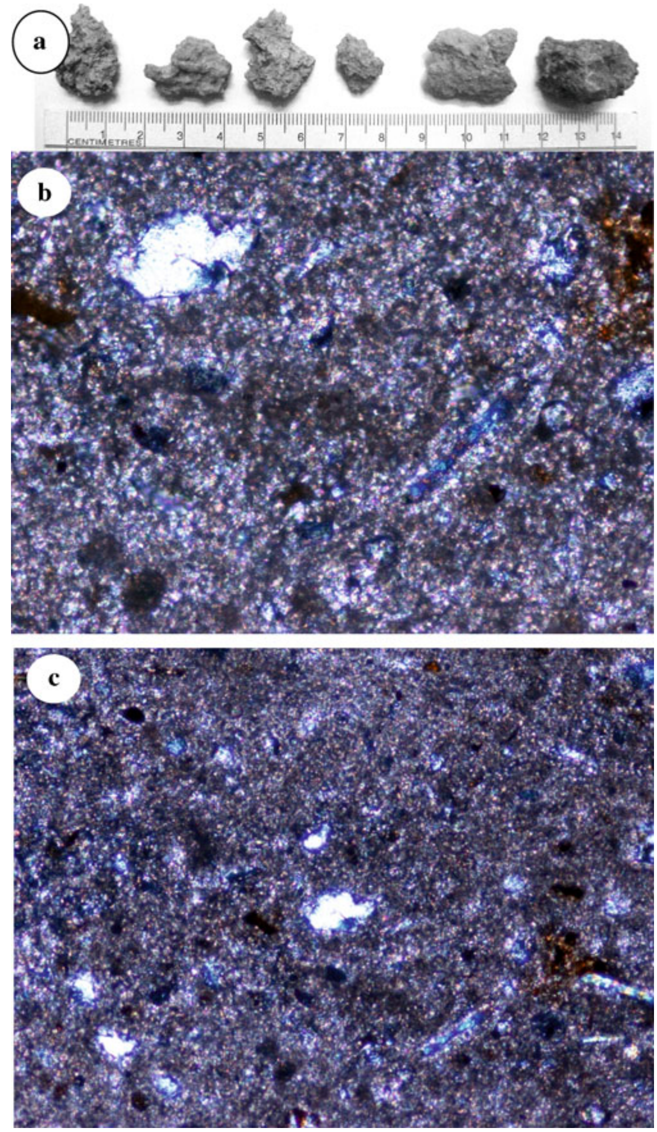

Figure 2. (**a**) SC nodule from the Bhognipur core. Photomicrograph of SC nodule displaying mostly micritic nature (**b**) and (**c**) indicates absence of major post-diagenetic recrystallization.

- to reconstruct the monsoon rainfall variations during the late Quaternary,
- to check whether the inferences from carbon isotope ratios of inorganic (SC) and organic fractions (SOM and NOM) are consistent or not, thereby
- reconstruction of the paleovegetation history over the Ganga Plain, and
- to delineate the relative contribution of sediments from the Himalaya and peninsular India.

Finally, $\delta^{18}O_{SC}$ values-based rainfall reconstruction along with $\delta^{13}C$ values-based vegetation reconstruction and Sr isotope ratio-based provenance reconstruction are used to assess relative role of monsoonal rainfall on the relative abundance of C_3–C_4 and to understand processes regulating the sediment supply.

2. Study area

The Ganga Plain, a younger Himalayan foreland basin, occupying the central position in the Indo–Gangetic Plains covers an area of 250,000 km^2, lies roughly between longitudes 77°–88°E and latitude 24°–30°N (figure 1a). It receives huge amount of sediments by various rivers, i.e., Ganga, Yamuna, Chambal, Betwa, etc. The alluvial deposits in the Ganga Plain are characterized by alternate floodplain and channel sand deposits, and are mostly exposed along the cliff sections. These sediments in the Ganga Plain have suffered pedogenesis during the pauses in sedimentation and the pedogenic intervals are characterized by presence of soil carbonate nodules and soil. The inorganic and organic fractions in the carbonate nodules and organic fractions within the soil are potential proxies for past rainfall and vegetational reconstruction. The study area falls under the sub-tropical monsoonal regime, with average precipitation of 800–1000 mm/year, mean maximum temperature of 30°–32°C, and mean minimum temperature of 15°–18°C (Singh 1994). The monsoon typically arrives in mid-June, and provides more than 85% of the annual rainfall by September.

For the present study, SC and soil samples were collected from the Bhognipur core (figure 1b). The Bhognipur core is marked by alternate floodplain and channel sand deposits. Lower part of the core is characterized by feldspar-rich sand that gradually changes to quartz- and mica-dominated sand with a prominent erosional surface (Srivastava *et al.* 2010). The upper part of the core is mainly dominated with floodplain deposits. Presence of Fe–Mn nodules, pedogenic carbonate, illuvial clay coatings and relict pedofeatures suggest pedogenic activity during pauses in sedimentation. Strongly developed pedofeatures are observed between 10 and 14 m depth (Srivastava *et al.* 2010) which indicates strong pedogenic activity. Carbonate nodules are preferentially concentrated on the top part of the core.

3. Methodology

3.1 *Procedure of stable isotopic analysis of carbonates*

For isotopic analysis of inorganic fraction (carbonate), each carbonate nodule was divided into two parts. One part was used for making thin section for microscopic study and the remaining part was used for isotopic analysis of carbonates and organic matter. Thin sections of the nodules show that the calcium carbonate is mostly micrite (figure 2b, c). Absence of any sparry calcite indicates that no major recrystallization has taken place after the precipitation of carbonates. This ensures preservation of pristine stable isotope composition in the Bhognipur core nodules.

For isotopic analysis, powdered samples from fresh surface of individual nodule were collected by dental drill. For δ^{13}C and δ^{18}O values ~100–300 μg of powdered carbonate samples were put into individual screw capped glass vials. The vials were systematically kept into the Gas Bench along with in-house $CaCO_3$ standards BDH (procured from University College London) and Z-Carrara (procured from Physical Research Laboratory, Ahmedabad). Subsequently, the vials were flushed with pure He gas. After flushing, ~100% orthophosphoric acid was injected manually into each vial which was kept at 72°C temperature bath. The evolved CO_2 was purified by Nafion tube and Porapack column in Gas Bench and allowed into Continuous Flow Isotope Ratio Mass Spectrometer (CFIRMS) for analysis. Each measurement comprised of four pulses of reference followed by six pulses of sample CO_2 gas. Long-term (~1 yr) measurements of BDH and Z-Carrara that were calibrated via NBS-19 provide an external precision of ±0.1‰ (2σ) for both the δ^{18}O and δ^{13}C values in VPDB scale.

3.2 *Procedure of stable isotopic analysis of organic matters*

To analyze the carbon isotope ratio in both SOM and NOM, powder of individual samples were produced and treated with 0.5N HCl to remove carbonate. Acid and soluble salts were removed by centrifuging (~3000 rpm). After centrifuging, the decarbonated samples were dried in oven at about 70°C temperature. The dried samples were crushed to make fine powder. Powdered sample of 15–25 mg was loaded into a tin capsule and introduced into pre-filled and conditioned reactor of Elemental Analyzer (Flash EA 1112) through an auto sampler. The sample filled tin capsule was flash combusted at ~1050°C in an oxygenated environment. Due to this combustion, N and C of the sample were converted to their respective gaseous oxides. The gas mixture was then allowed to pass successively through chromium oxide, reduced copper and silver cobaltous. Chromium oxide was used for complete oxidation of sample, while reduced copper removes unused oxygen. Any CO produced during reaction with reduced copper was further oxidized to CO_2 by reacting with silver cobaltous. Sample gas (CO_2) was allowed to pass through Magnesium per chlorate [$Mg(ClO_4)_2$] filled trap to remove the moisture and was introduced into the isothermal Gas Chromatographic Column for separating the other gases (N_2, NO_x, O_2, etc.). After the purification, the sample gas was introduced into the CFIRMS. Two pulses of reference CO_2 are allowed to enter into Mass Spectrometer through reference open split, followed by single pulse of sample CO_2. The raw delta value of sample was calculated with respect to this reference CO_2 gas. Standards (C-3 cellulose and Acetanilide) were measured in the same way. The system was calibrated by analyzing IAEA standard C-3 Cellulose. Analytical precision was monitored by running both C-3 and UBC-ACE (Acetanilide). A routine precision of $\sim \pm 0.1‰$ (2σ) has been obtained for $\delta^{13}C$. All isotopic data are reported against VPDB. All carbonates and organic matter samples were analyzed in the Stable Isotope Laboratory National facility of the Indian Institute of Technology, Kharagpur, India.

3.3 *Procedure of isotopic analysis of Sr in SC*

To determine $^{87}Sr/^{86}Sr$ ratio in carbonate fraction, 10 mg of powdered samples of SC was reacted with 1M acetic acid for 30 seconds. Subsequently, the solution was filtered to remove silicate minerals and dried at 60°C. Sr was purified and concentrated by the cation-exchange resin chromatography and Sr isotope ratios were measured using a multicollector Thermal Ionization Mass Spectrometer (TRITON, Thermo-Finnigan) at National Facility for Isotope Geosciences, Pondicherry University, India.

Multiple measurements of International Standard SRM 987 were carried out during the course of analysis of samples to check the accuracy and precision. The mean value of $^{87}Sr/^{86}Sr$ ratio of 0.710255 with external precision of 0.000007 was obtained from 20 standard measurements. The recommended $^{87}Sr/^{86}Sr$ ratio for SRM 987 is 0.710244.

Sr concentration in SC nodules was measured by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES = 0.001 ppm, 2σ) at National Facility for Isotope Geosciences, Pondicherry University, India. The ICP-AES was calibrated using different salt solution of known concentration.

4. Results

4.1 *$\delta^{18}O$ values in SC*

The $\delta^{18}O_{SC}$ values from the Bhognipur core samples range from $-7.6‰$ to $-4.9‰$ ($n = 21$; table 1), covering 2.7‰ variation. At about 25.5 m depth, the observed $\delta^{18}O_{SC}$ value is $-6.3‰$ (figure 3a). Subsequently, gradual enrichment in ^{18}O is observed up to 10 m depth with fluctuation. The variation in $\delta^{18}O_{SC}$ values above 10 m is characterized by moderate fluctuations with most negative phase observed at ∼3.5 m with values $\sim -7.6‰$. The most negative interval is again followed by phase of less negative interval with $-5.9‰$ value at about 2.5 m depth (figure 3a).

4.2 *$\delta^{13}C$ values in SC*

The $\delta^{13}C_{SC}$ values range from -4.1 to $1.4‰$ ($n = 21$, table 1), covering 5.5‰ variation. Same as the $\delta^{18}O_{SC}$ value, between 25 and 9.9 m depth, $\delta^{13}C_{SC}$ values record an overall trend towards higher values from -1 to $1.4‰$ (figure 3b) followed by lowering. In the top part of the core, at about 4.7 m, a value of $-4.1‰$ is observed. Samples above 4 m are characterized by an increase in $\delta^{13}C_{SC}$ value up to 2.5 m depth (figure 3b).

4.3 *$\delta^{13}C$ values in SOM and NOM*

Carbon isotope ratio of SOM as well as NOM from the same soil horizon was measured from the Bhognipur core. The $\delta^{13}C_{SOM}$ values range from -25.6 to $-16.3‰$ ($n = 21$, table 1), covering a wide range of 9.3‰. The limited $\delta^{13}C_{NOM}$ values are less variable and range from -27.7 to $-25.0‰$ ($n = 5$, table 1). The $\delta^{13}C_{SOM}$ values show isotopically more negative intervals at about 10.7 and 4.7 m depth and less negative intervals at 25, 7.7 and 1 m depth (figure 3c). The $\delta^{13}C_{NOM}$ values of samples between 2.5 and 10 m depth are more depleted in ^{13}C than the $\delta^{13}C_{SOM}$ values (figure 3c).

4.4 *Sr isotope ratio in SC*

$^{87}Sr/^{86}Sr$ ratios and Sr concentration in SC of the Bhognipur core ranges from 0.71232 to 0.71867 ($n = 12$) and 163 to 456 ppm ($n = 8$), respectively (table 1). The lower part between 25.5 and 20.3 m depth is characterized by low $^{87}Sr/^{86}Sr$ ratios (0.71277–0.71232). The 19.4 to 2.4 m depth interval is characterized by monotonous increase in $^{87}Sr/^{86}Sr$ ratios (figure 3d).

5. Discussion

5.1 *$\delta^{18}O$ values of SC*

The $\delta^{18}O$ values of soil carbonate have been used to reconstruct paleomonsoonal rainfall from different

Table 1. *$\delta^{18}O$, $\delta^{13}C$, $^{87}Sr/^{86}Sr$ and Sr concentration in soil carbonate (SC) and $\delta^{13}C$ values in organic matter dispersed in the soil (SOM) and occluded within the soil carbonate nodules (NOM) from the Bhognipur core.*

Sample no.	Depth (m)	$\delta^{18}O_{SC}$ (‰; VPDB)	$\delta^{13}C_{SC}$ (‰; VPDB)	$\delta^{13}C_{SOM}$ (‰; VPDB)	$\delta^{13}C_{NOM}$ (‰; VPDB)	$^{87}Sr/^{86}Sr$ in SC	Sr concentration (ppm)	1/Sr
BP 1	1.0	–	–	−19.7	–	–	–	–
BP 2	2.0	–	–	−19.9	–	–	–	–
BP 3	2.4	−6.8	−1.8	−17.9	−26.6	0.71853	195.0	0.005
BP 4	2.5	−5.9	−1.8	−19.1	−25.0	–	–	–
BP 5	2.8	−7.1	−2.0	−21.7	–	0.71867	–	–
BP 6	3.2	−6.4	−1.5	–	–	–	–	–
BP 7	3.5	−7.6	−3.1	−21.3	−27.7	0.71764	172.2	0.006
BP 8	4.2	−6.0	−2.9	−24.7	−27.6	–	–	–
BP 9	4.7	−6.3	−4.1	−25.6	−26.2	0.71796	163.2	0.006
BP 10	5.2	−5.8	−3.8	−21.0	–	–	–	–
BP 11	7.2	–	–	−21.3	–	–	–	–
BP 12	7.7	−6.2	−2.4	−17.4	–	0.71465	454.9	0.002
BP 13	8.6	−5.0	0.8	–	–	0.71580	383.8	0.003
BP 14	9.5	−5.5	1.3	–	–	–	–	–
BP 16	9.9	−5.3	1.4	−21.0	–	–	–	–
BP 17	10.0	−4.9	1.1	−23.7	–	0.71643	306.3	0.003
BP 18	10.7	−5.0	−0.1	−24.4	–	–	–	–
BP 19	11.2	–	–	−23.4	–	–	–	–
BP 20	14.0	−5.6	1.1	−20.5	–	0.71727	–	–
BP 22	17.7	−5.3	−0.7	−16.3	–	–	–	–
BP 23	18.0	−5.0	−0.2	–	–	0.71546	456.1	0.002
BP 24	19.3	–	–	−17.5	–	–	–	–
BP 25	19.4	−5.4	0.4	−19.8	–	0.71640	391.8	0.003
BP 26	20.3	−5.6	0.1	–	–	0.71232	–	–
BP 27	24.0	−6.0	−1.1	−17.6	–	–	–	–
BP 28	25.5	−6.3	−0.8	−18.7	–	0.71277	–	–

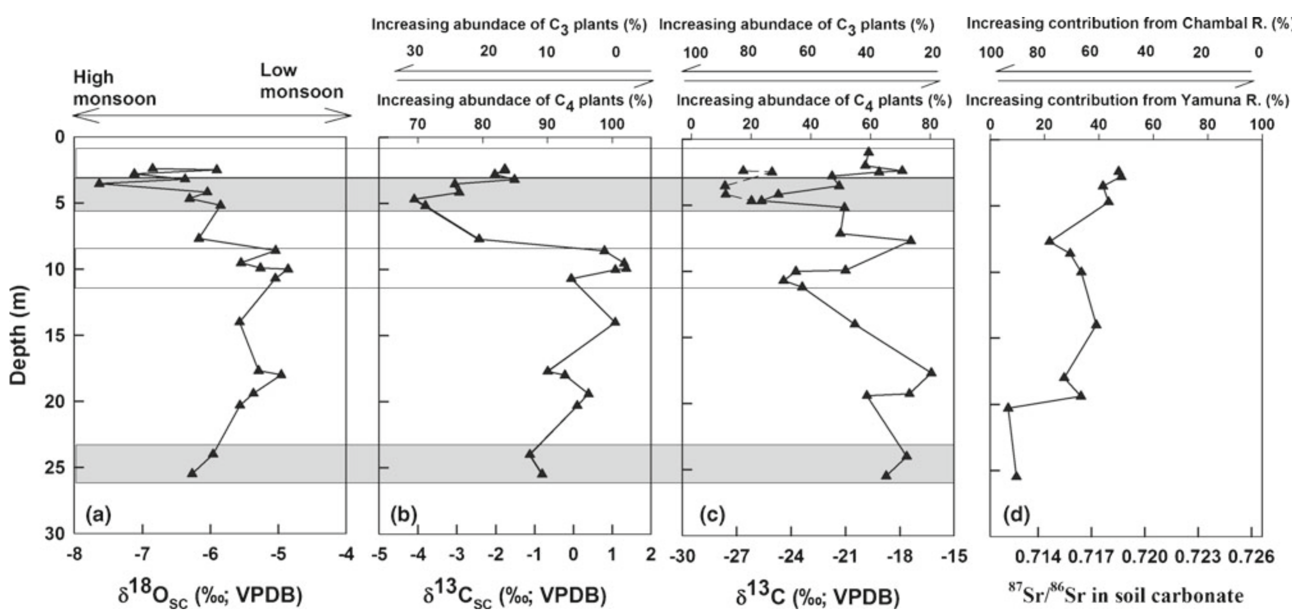

Figure 3. Depth variations in (**a**) $\delta^{18}O_{SC}$ value, (**b**) $\delta^{13}C_{SC}$, (**c**) $\delta^{13}C_{SOM\ and\ NOM}$ values and (**d**) $^{87}Sr/^{86}Sr$ ratios in soil carbonate. (**a**) The $\delta^{18}O_{SC}$ value shows isotopically more negative and less negative intervals at 25.5 and 3.5 m depth (long dark grey boxes) and 10 and 2.5 m depth, respectively (long light grey boxes). (**b**) The $\delta^{13}C_{SC}$ and (**c**) $\delta^{13}C_{SOM}$ values show negative intervals around 25.5 and 4.5 m depth (long dark grey boxes), which suggest higher abundance of C_3 plants, almost coinciding with high monsoon intervals. (**d**) Sr isotopic values suggest fluctuation of sediment supply from peninsular India and Himalayan rocks.

parts of the Indian subcontinent. Recent work based on $\delta^{18}O$ values of soil carbonates by Agrawal *et al.* (2012), using samples from three drill cores raised from the southwest to northeast transect of the Ganga Plains, showed three periods of monsoonal intensification at 100, 40 and 25 ka and ~20% decrease in rainfall during the LGM. Synchronicity of such monsoonal variations can also be deduced from different parts of the Ganga Plain to understand regional climate prevailed during the late Quaternary time. Oxygen isotope ratios of SC is mainly dependent on the isotopic ratio of soil water, which is derived from local rain water that percolates through the soil profile, and temperature of carbonate precipitation (Cerling 1984; Quade *et al.* 1989). The $\delta^{18}O$ values of local rain water are affected by factors like temperature, latitude, elevation, the distance from the moisture source, seasonality, and the amount of rainfall per event (the 'amount effect') (Nativ and Riggio 1990; Lawrence and White 1991; Rozanski *et al.* 1993). Near-surface, the $\delta^{18}O$ value of soil water is enriched in ^{18}O compared to the $\delta^{18}O$ values of rain water as the lighter isotope (^{16}O) is preferentially removed from the soil water during evaporation. Evaporation effect is most distinct in closed-basin like lakes where water is lost by infiltration and evaporation (Talbot 1990), as compared to open paleosols. Soil carbonates form in seasonally dry environments (Sparks 2003) and during evaporation soil water can get enriched by 1‰ as observed by Salomons *et al.* (1978). It implies that the $\delta^{18}O$ value of soil carbonate can be used to decipher paleoprecipitation.

Studies based on the seasonal variations in $\delta^{18}O$ values of rain water show large annual variation with a value of 11‰ (VSMOW) in Kolkata and 8‰ (VSMOW) in New Delhi (Sengupta and Sarkar 2006). This kind of seasonal isotopic pattern can also be observed in spelothem archives (Fleitmann *et al.* 2004; Breitenbach *et al.* 2010). Soil carbonates can take 10^2–10^5 years to form (Jenny 1980; Machette 1985; Retallack 2005). Therefore, it is not possible to find out seasonal variations but a long term average monsoonal rainfall variations can be deduced from $\delta^{18}O$ value of soil carbonate (Sanyal *et al.* 2004, 2005; Agrawal *et al.* 2012). It has been observed in low latitude sites that $\delta^{18}O$ value of rain water decreases by 1.5‰ for every increase in 100 mm of rainfall (Yurtsever and Gat 1981), a phenomenon commonly called 'amount effect' (Dansgaard 1964). Hence, periods of more negative $\delta^{18}O_{SC}$ values are interpreted as intervals of high rainfall and periods of less negative $\delta^{18}O_{SC}$ values as intervals of relatively low rainfall. Based on these assumptions, oxygen-isotope-based reconstruction of rainfall intensity of the southwest monsoon has been discussed below. In Bhognipur core, distinct lowering in $\delta^{18}O_{SC}$ value around 25.5 and 3.5 m suggests increase in monsoon rainfall intensity (figure 3a). Whereas higher $\delta^{18}O_{SC}$ value around 10 and 2.5 m depth indicates decrease in monsoonal rainfall (figure 3a).

5.2 $\delta^{13}C$ *values of SC, SOM and NOM*

On the basis of photosynthetic pathway, plant biomass is mainly divided into two groups, C_3 (Calvin cycle) and C_4 (Hatch-Slack cycle) plants. These plant types have distinct carbon isotope signature. The plant bio-organic residues incorporated into soil environment after death of the plant, form a fraction of the SOM, is assumed to preserve the mean carbon isotopic composition of the contemporary vegetation, with little or no fractionation. In contrast, soil carbonate forms from soil solution mixed with soil CO_2, derived from decomposition of SOM and plant respiration during a dry phase (Wang and Follmer 1998). The $\delta^{13}C$ values of SC is 14–17‰ enriched (0 to 25°C) compared to $\delta^{13}C$ values of coexisting organic matter (SOM or NOM; $\Delta(\delta^{13}C)$) if the SC forms equilibrium with plant-derived CO_2 (Cerling 1984). Agrawal *et al.* (2012) reported that $\delta^{13}C$ value of C_3 plants ranges from −32.6 to −19.2‰ with average -29.5 ± 1.2‰ and for C_4 plants the value ranges from −16.6 to −10.5‰ with average -13 ± 1.2‰. Therefore, with 100% C_3 plant in soil, $\delta^{13}C$ value of SC will be −15.5‰ and 100% C_4 plant will correspond to $\delta^{13}C$ value of SC about +1‰. On the basis of these end member values we have calculated relative abundance of C_3 and C_4 plants from the SC, SOM and NOM.

The $\delta^{13}C_{SC}$, $\delta^{13}C_{SOM}$ and $\delta^{13}C_{NOM}$ values mainly represent mixed C_3–C_4 vegetation over the Ganga Plain (figure 3b, c). At about 25.5 m depth $\delta^{13}C_{SC}$ and $\delta^{13}C_{SOM}$ show 90 and 65% abundance of C_4 plants, respectively (figure 3b, c). From 25.5 m onwards, abundance of C_4 plants has gradually increased and at about 9.9 m depth $\delta^{13}C_{SC}$ values suggest 100% abundance of C_4 plants (figure 3b). In contrast, the $\delta^{13}C_{SOM}$ shows 30% abundance of C_4 plants at about 10 m depth and ~75% abundance of C_4 plants is observed at about 7.7 m depth (figure 3c). Subsequently, $\delta^{13}C_{SOM}$ values show increment to 50% in abundance of C_3 plants up to 4.7 m depth. On the other hand, $\delta^{13}C_{SC}$ values show increment to 70% in abundance of C_3 plants at about 4.7 m depth. Following this C_4 plants reached its acme at ~2.5 m depth with 75–85% abundance (figure 3b). In contrast, $\delta^{13}C_{NOM}$ values show 10–30% abundance of C_4 plants between 4.5 and 1 m depth (figure 3c).

The paleovegetation data obtained using $\delta^{13}C_{SC}$, $\delta^{13}C_{SOM}$ and $\delta^{13}C_{NOM}$ values from the Bhognipur core suggest that the southern Ganga Plain was characterized by mixed C_3–C_4 vegetation. Although trend of temporal change in vegetation obtained from $\delta^{13}C_{SC}$, $\delta^{13}C_{SOM}$ and $\delta^{13}C_{NOM}$ are more or less similar, estimates of C_3 or C_4 plants from these proxies vary. For example, about 4.5 m $\delta^{13}C_{SC}$ shows 70% abundance of C_4 plants whereas $\delta^{13}C_{SOM}$ and $\delta^{13}C_{NOM}$ show 20–25% abundance of C_4 plant (figure 3b, c). Overall, the abundance calculation shows relatively higher percentage of C_4 plants in SC compared to SOM and NOM with higher difference in the latter one. The cause of this discrepancy has been discussed below.

5.3 *Relationship between the $\delta^{13}C$ of SC, SOM and NOM*

SC, SOM and NOM are the common constituents in soil and their carbon isotope ratio is directly related to the plants grew in the soil. As mentioned earlier, the $\delta^{13}C$ value of SC is 14–17‰ enriched relative to the coexisting $\delta^{13}C$ value of SOM or NOM if the soil CO_2 is high enough to prevent infiltration of atmospheric CO_2 (Cerling 1984). The $\Delta(\delta^{13}C)$ values for SC-SOM of the Bhognipur core ranges from 15.0 to 24.8‰ and for SC-NOM ranges from 22 to 25‰. The difference in $\Delta(\delta^{13}C)$ values between SC, SOM and NOM can be explained by the source-controlled model proposed by Wang and Follmer (1998), which modifies the CO_2 source assumption of Cerling (1984).

SOM progressively accumulate in the soil due to decomposition of plant over time whereas plants' respired CO_2 is the major carbon source for soil carbonate (Deines 1980). In general, $\Delta(\delta^{13}C)$ value is $\leq$14‰, if soil carbonate carbon is derived from respiration dominated by C_3 plants, and $\geq$17‰ when soil carbonate derived carbon mainly from C_4 dominated plants. The higher proportion of C_4 plants estimated from SC of the Bhognipur core suggests that C_4 plant respiration dominated during carbonate formation, i.e., during dry period and C_3 plant derived organic carbon contributed greater towards SOM and NOM, thereby making $\Delta(\delta^{13}C)$ value >14‰. The statistical analysis shows that $\Delta(\delta^{13}C)$ is negatively correlated with $\delta^{13}C_{SOM}$ value (figure 4a; $n = 16$, $r^2 = 0.70$, $p = <0.0001$), which also suggests that estimation of C_4 biomass from SOM does not matches with the C_4 biomass as indicated by SC. It strongly implies the large variation of $\Delta(\delta^{13}C)$ is due to different carbon sources for SOM, NOM and SC.

The deviation of estimation can be compounded if there is additional source of organic matter in soil. Agrawal (2011) performed compound specific

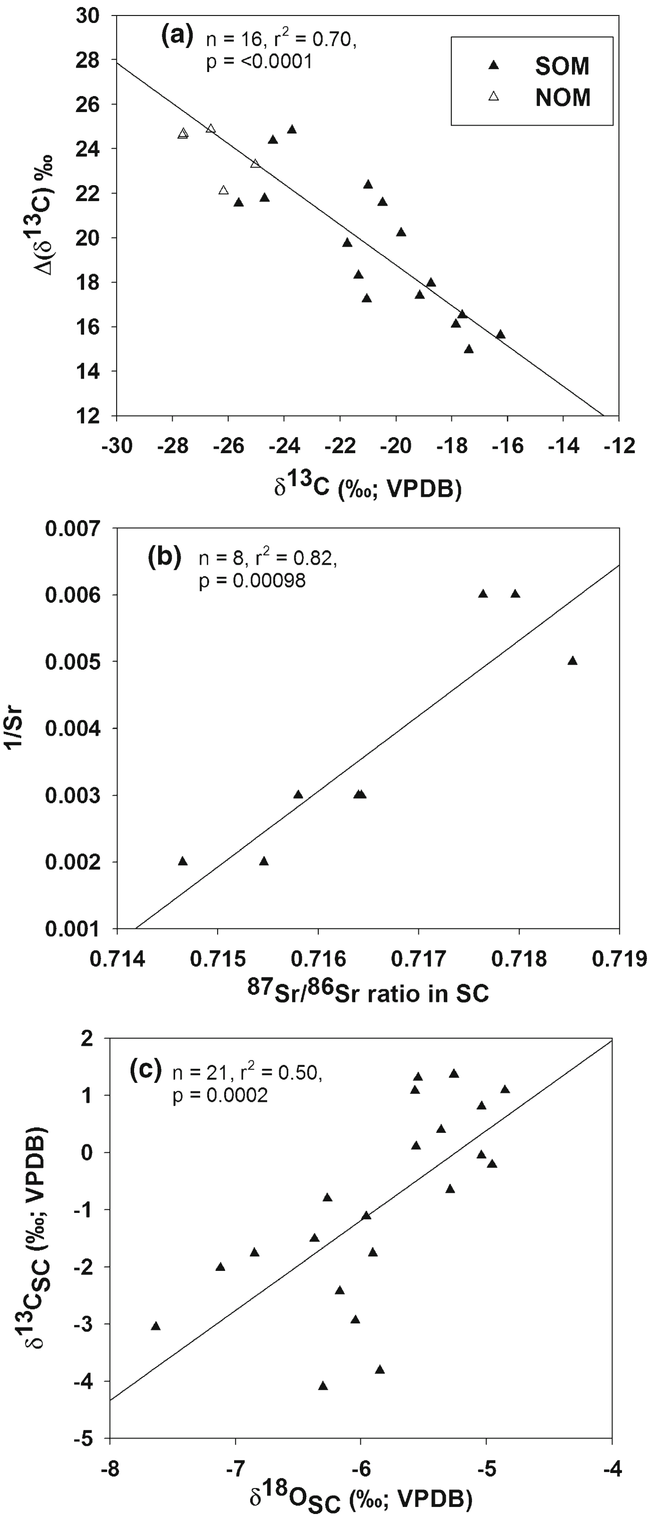

Figure 4. Regression of (**a**) $\Delta(\delta^{13}C)$ and $\delta^{13}C_{SOM}$ shows negative correlation which indicates the increase of C_4 biomass determined by SOM is not proportional with increase of C_4 biomass determined by SC. (**b**) Regression of $^{87}Sr/^{86}Sr$ ratios and 1/Sr in SC. The positive correlation between $^{87}Sr/^{86}Sr$ ratios and 1/Sr in SC of the Bhognipur sample indicates two components mixing, one with high $^{87}Sr/^{86}Sr$ ratio and low Sr concentration and other with low $^{87}Sr/^{86}Sr$ ratio and high Sr concentration. The positive slope of the (**c**) $\delta^{18}O_{SC}$–$\delta^{13}C_{SC}$ values indicates decrease in rainfall intensity which caused an increase in the abundance of C_4 plants.

analysis in SOM from the Ganga Plain to understand source of organic matter and organic matter dynamics. The abundance and distribution of *n*-alkanes and saturated fatty acids reveals that SOM in the Ganga Plain sediments is composed of a complex mixture of petrogenic, microbial and vascular plant inputs. The isotopic composition of long-chain saturated fatty acids suggests that SOM in the Ganga Plain sediments is formed by partial degradation of a Himalayan riverine petrogenic organic carbon component followed by addition of the local Ganga Plain microbial and vascular plant organic matter. Therefore, the discrepancy of C_4 plant estimates from the SC, SOM and NOM indicate selective preservation of lipids but this explanation need to be tested using data from other part of the Ganga Plain which can further resolve this issue.

5.4 *Sr isotope ratio in SC*

The rivers that drain the Himalaya and peninsular India carry variable amount of sediments and these sediments are deposited into the Ganga Plain and Bay of Bengal. Pauses in sedimentation in the plain area causes pedogenic activity and ultimately produces soils. SC, common constituents of the buried soil (paleosols) in the Ganga Plain, precipitates from soil solution whose chemistry is determined by weathering in sediments. These soil solutions also percolate through the base of the soil and finally go into river through lateral movement of groundwater which in turn controls the river water chemistry (Quade *et al.* 1997). As the SC nodules represent a time window of 10^{2-5} years, the soil solution from which it is precipitating provides a time averaged chemistry of sediments as well as river water. Sr is one such element whose isotopic signature in SC can be used to characterize the river water chemistry as well as the source of sediments.

The $^{87}Sr/^{86}Sr$ ratios of Bhognipur core SC samples show overall increasing trend from bottom to top part of the core. The low Sr isotope ratio is observed between 25.5 and 20.3 m depth whereas between 19.4 and 2.4 m depth the Bhognipur core is characterized by high Sr isotope ratio (figure 3d). Positive correlation between $^{87}Sr/^{86}Sr$ ratios and 1/Sr for SC (figure 4b, $n = 8$, $r^2 = 0.82$, $p = 0.00098$) suggests two components mixing, one with high $^{87}Sr/^{86}Sr$ ratio and low Sr concentration and other with low $^{87}Sr/^{86}Sr$ ratio and high Sr concentration. The peninsular rivers like Chambal have low Sr isotope ratio (0.7111–0.7115; Palmer and Edmond 1992) and high Sr concentration. In contrast, Himalayan river, Yamuna, has high Sr isotope ratio (0.7266; Dalai *et al.* 2003). The Sr isotopic signature of SC falls in-between these two categories. Therefore, it can be suggested that the isotopic signature of SC at the bottom part of the core (between 25 and 20 m depth) represents exclusive peninsular Indian river source. With the help of modern day Sr isotope ratio and relative contribution of Sr from Chambal and Yamuna river water, mass balance equation has been used to quantify the relative contribution from the Himalayan and peninsular rivers in geological past for the Bhognipur core. Such calculation suggest that if Yamuna and Chambal rivers contribute 50–50% dissolve Sr with an average value of 0.72624 (Dalai *et al.* 2003) and 0.7113 (Palmer and Edmond 1992), respectively then only 0.719 values can be achieved (figure 3d). Hence, high Sr isotope ratios, above 20 m depth, suggest high radiogenic Sr contribution from the Himalayan river, i.e., from Yamuna. The abrupt increase of the $^{87}Sr/^{86}Sr$ ratios of SC from 20.3 m onwards reflects change in source from peninsular India to Himalaya. Possible causes of change in source are discussed below.

6. Paleoclimatic implication

Recent studies showed that stable isotope ratios of paleosol from the Himalayan foreland provide important paleoenvironmental information related to climatic fluctuations and its relation with marine isotope stages (MIS 5-1) during the late Quaternary (Sinha *et al.* 2006; Agrawal *et al.* 2012). In this context, Agrawal *et al.* (2012) also showed periodic change in monsoonal rainfall during last 100 ka over the Ganga Plain. However, due to absence of reliable age estimates, the ages of the Bhognipur core were obtained by depthwise comparison against the Kalpi core (Supplementary data, figure S1; Sinha *et al.* 2009; Agrawal *et al.* 2012), which is located on the southern bank of Yamuna river. Such exercise suggests ∼20 ka (MIS 2) and ∼46 ka age (MIS 3) for the Bhognipur core sediments at about 12.5 and 32.5 m depth (figure S1). Overall, comparison between the Kalpi and Bhognipur cores suggests that the bulk of the strata in the Bhognipur section are in the age range of <20 ka to >45 ka. Thus the section represents late Quaternary 'window' through southern interfluve margin deposits of the Ganga Plain. Based on these dates, the Bhognipur core is inferred to span part of the late Quaternary (MIS 1-3). In view of the few ages and large error bars for the Optically Stimulated Luminescence dates, the age model for the Bhognipur core is only broadly constrained. Hence, we have interpreted our isotope data with respect to age of the core to broadly mark the different events over the Ganga Plain. Overall, the $\delta^{18}O_{SC}$ values suggest periodic change in monsoonal rainfall over the Ganga Plain during the late Quaternary. On the other hand, the $\delta^{13}C_{SC}$ values suggest mixed C_3–C_4 vegetation over the Ganga Plain during the late Quaternary. Interestingly, the $\delta^{13}C_{SC}$ values are changing simultaneously with the $\delta^{18}O_{SC}$ and making positive correlation. For example, at ∼25.5 (MIS 3) and 3.5 m depth (MIS 1) $\delta^{18}O_{SC}$ values suggest high monsoonal rainfall and at the same depth $\delta^{13}C_{SC}$ values show higher abundance of C_3 plants whereas the low monsoon condition at about 10 m depth (MIS 2) is characterized by higher abundance of C_4 plants (figure 3a, b). The positive slope of the $\delta^{18}O_{SC}$–$\delta^{13}C_{SC}$ plot (figure 4c; $n = 21$, $r^2 = 0.50$, $p = 0.0002$) also indicates decrease in rainfall intensity caused an increase in the abundance of C_4 plants and suggest casual relationship between type of plants and monsoonal rainfall.

We also investigated the possible configuration of the Himalayan river system with the help of Sr isotope ratio in SC. The Sr isotope ratio in SC represents the river water chemistry and used as provenance proxies that have already been successfully applied to the Himalayan drainage to identify river water chemistry in millennium as well as millennial time scale (Quade *et al.* 1997; Agrawal *et al.* 2013). The $^{87}Sr/^{86}Sr$ ratios in SC from the Bhognipur core show substantial changes in sediment sources during the late Quaternary. The high $^{87}Sr/^{86}Sr$ ratios in SC suggest existence of the Yamuna river at southern part of the Ganga Plain during MIS 3. Recent study by Agrawal *et al.* (2013) suggests Himalayan river activity (Yamuna river) in the southern part of the Ganga Plain at about 80 ka (i.e., near Kalpi region) (Supplementary data, figure S1). The high Sr isotope ratio at the bottom part of the Kalpi core (Agrawal *et al.* 2013) and the top part of the Bhognipur core suggests connection between the Kalpi and Bhognipur regions (figure S1) related to change in provenance. But, more detailed sub-surface investigations and isotopic analysis are necessary to identify the course of Yamuna river in the Ganga Plain.

7. Conclusions

Monsoonal rainfall reconstruction shows that it has varied with two peaks of intensification during MIS 3 and MIS 1. The $\delta^{13}C_{SC}$, $\delta^{13}C_{SOM}$ and $\delta^{13}C_{NOM}$ suggest that during the late Quaternary, sediments of the Ganga Plain were characterized by mixed C_3–C_4 vegetation. Variations in carbon isotope ratio in SOM and NOM at same depth implies preservation problem of pristine organic matter signature and vegetational reconstruction (absolute percentage of C_3–C_4 plants) using paleosol organic matter (both SOM and NOM) should be done after assessing the pristine character. During the high monsoon time C_3 plants were dominant. With decrease in rainfall, vegetation has changed from C_3 to C_4 dominant ecosystem. The change in vegetation pattern in response to rainfall indicates that the variation in C_3–C_4 plants during the late Quaternary was determined by rainfall variations. The Sr isotope ratio in SC suggests existence of Yamuna river at the southern part of the Ganga Plain during MIS 3.

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