Neogene vegetation shift in the Nepalese Siwalik, Himalayas: A compound-specific isotopic study of lipid biomarkers

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Abstract
The late Neogene vegetation change of C4 plants replacing C3 plants is widely documented across the world. This vegetation shift has been particularly well-documented in the Himalayan foreland based on δ13C isotopic data from palaeosol carbonates and bulk organic matter in the Siwalik sedimentary rocks of Pakistan, India and Nepal, showing asynchronous expansion of C4 plants between 8 and ~5 Ma, 9 and 6 Ma, and around 7 Ma, respectively. In this study, compound-specific isotopic analysis of lipid biomarkers extracted from shale and palaeosols in the palaeomagnetically age-constrained Nepalese Siwalik is utilized to better understand this vegetation shift. This is the first comprehensive lipid biomarker study in the Nepalese Siwalik, with new isotopic results from the previously undocumented Karnali River section. The δ13C n-alkane (C27-31) results from the Surai Khola section suggest C3 plants were dominant between 12 and 8.5 Ma. A stepwise expansion of C4 plants that started gradually at 8.5 Ma escalated rapidly after 5.4 Ma, so that by 5.2 Ma C4 vegetation dominated the landscape. This dramatic ecological shift at the Miocene-Pliocene boundary was likely linked with an intriguing tectonic-climate coupling in the Himalayan-Tibetan region, prompting wetter summers and drier winters that drove C4 grass expansion. However, in the Karnali River section, the data show no clear sign of C4 plant expansion until 5.2 Ma (youngest sample age). In the two study locations separated laterally by ~200 km along tectonic-strike, this different trend of vegetation change likely indicates local controls like river-catchment influence. However, it is possible that, similar to the Surai Khola section, C4 plants dominated after 5.2 Ma in the Karnali River section. This study also suggests that the past vegetation makeup of an area is better reconstructed using isotopic signatures of molecular markers than of bulk organic matter or pedogenic nodules.

KEYWORDS
Carbon isotope, Himalayan foreland, Karnali River, Siwalik, Surai Khola
1 | INTRODUCTION

Late Miocene global expansion of C₄ grasses replacing C₃ trees is extensively documented from various parts of the world including the Siwalik ranges, Himalayan foreland. Although the grass expansion likely started between 32 and 23 Ma (Fox and Koch, 2004; Edwards et al., 2010; Urban et al., 2010), the main expansion occurred in the late Miocene (Quade et al., 1989, 1995: Cerling et al., 1993, 1997).

The expansion of C₄ plants tends to be asynchronous from one continent to another and even regionally within one continent. For example, this vegetation shift started between 6.4 and 4 Ma in the North American Great Plains (Fox and Koch, 2004) and around 2.6 Ma in the Gulf of Mexico region (Tipple and Pagani, 2010). The shift in East Africa occurred between 11 and 5 Ma (Feakins et al., 2013), but more likely over the past 6 Ma (Cerling et al., 2011). The timing of C₄ plant expansion in the Himalayan foreland has been variously reported as between 7.4 and 7 Ma (Quade et al., 1989, 1995), 8 and 6.5 Ma (Hoorn et al., 2000) and between 7.9 and 5.5 Ma (Huang et al., 2007).

In Central Asia, north of the Tibetan Plateau, this vegetation shift occurred at 5.3 Ma (Sun et al., 2013; Shen et al., 2018).

Most studies of the Neogene vegetation shift utilize the carbon isotopic signature (δ¹³C) of pedogenic carbonate nodules or bulk organic matter (Quade et al., 1989, 1995; Cerling et al., 1993; Fox and Koch, 2004), whereas only a few are based on compound-specific isotopic analysis (CSIA) of molecular carbon (Huang et al., 2007; Tipple and Pagani, 2010). In the bulk carbon isotope study, the organic matter in the soil can receive input from organisms other than the vegetation, including insects, cyanobacteria, fungi and soil bacteria, and such incidental contamination has been reported from the Pakistani Siwalik (Freeman and Colarusso, 2001). Similarly, as reported in the Siwalik succession in the Surai Khola section of Nepal (Sanyal...)
et al., 2005), the diagenetic alteration of pedogenic carbonate nodules can be a problem for an isotopic study.

In CSIA, one can specifically target long-chain (>24 carbons) lipid biomarkers—derived primarily from higher (i.e., vascular) plants—that are widely distributed in terrestrial, lacustrine and marine sediments (Tipple and Pagani, 2007). Moreover, CSIA allows us to analyse leaf wax lipids, which are largely resistant to diagenetic alteration (Sessions et al., 2004). The extraction of lipids following different chemical treatments allows individual carbons in a carbon chain to be analysed. This technique was applied in the Tibetan Plateau by Polissar et al. (2009) and Liu et al. (2005), and in the Arabian Sea by Huang et al. (2007) to understand Neogene vegetation change.

For the Himalayan foreland, Freeman and Colarusso (2001) applied CSIA in the Pakistani Siwalik, plus an additional two samples from the Nepalese Siwalik. The first extensive study using CSIA in the Nepalese Siwalik is presented here (Figure 1), including the analysis of organic matter from palaeomagnetically age-constrained mudstones and palaeosols deposited in the Himalayan foreland basin during the middle Miocene to early Pleistocene. The objective of this study is to test the applicability of CSIA in the Nepalese Siwalik, and compare CSIA results with previously published pedogenic bulk-carbon isotope results from the same locality (Quade et al., 1995) for a better understanding of the late Neogene expansion of C4 plants. This study also reports new isotopic results from a previously undocumented locality, the Karnali River section of the Nepalese Siwalik (Figure 1).

1.1 | Geology of the study area

The Himalayan rock successions are divided into four tectonostratigraphic units (Tethys Himalaya, Higher Himalaya, Lesser Himalaya and Siwalik), which are separated from each other by the north-dipping thrust fault systems (Figure 1; Gansser, 1964; Hodges and Silverberg, 1988; Hodges et al., 1996; DeCelles et al., 1998). The youngest of these units, the Siwalik, is separated by the Main Boundary Thrust (MBT) from the Lesser Himalayan rocks to the north, and the Main Frontal Thrust (MFT) from the Gangetic plain in the south (present-day foreland basin).

Of the ~2,000 km long Siwalik outcrop belt in the Himalayan foreland, the Nepalese Siwalik occupies ~800 km in an E-W trending exposure (Figure 1C), with an average total thickness of ~6 km (Schelling et al., 1991; Gautam et al., 2000). The molasse sedimentary rocks of the Siwalik succession were deposited during the Miocene and early Pleistocene (Prakash et al., 1980; Tokuoka et al., 1986; Appel et al., 1991; Burbank et al., 1996; Gautam and Fujiwara, 2000; Ojha et al., 2000), and are now exposed in the foothills of the Himalayas (Figure 1). Based on lithology, the Siwalik succession is divided into three units: Lower Siwalik, Middle Siwalik and Upper Siwalik (Auden, 1935; Hagen, 1969; Yoshida and Arita, 1982; Ulak, 2009). The Lower Siwalik consists of interbedded fine to medium grained, grey sandstone and variegated or grey mudstone (Figure 2) deposited in fluvial and floodplain environments (Gautam and Rosler, 1999; Ulak, 2009). The medium to coarse grained, ‘salt and pepper’ sandstones, which are often multistory in nature and intercalated with mudstones (Figure 2), indicate braided river deposition in the Middle Siwalik (Gautam and Rösler, 1999; Nakayama and Ulak, 1999; Ojha et al., 2000; Ulak, 2009). Conversely, the Upper Siwalik consists of bedded pebble and cobble conglomerates intercalated with loosely packed sandstone and claystone layers that are interpreted to represent alluvial fan environments deposited closer to the mountain belt (Gautam and Rosler, 1999; Ojha et al., 2000; Ulak, 2009).

FIGURE 2 Siwalik fluvial sedimentary rocks in Nepal. (A) Lower Siwalik exposure in the Surai Khola section, showing sandstones interbedded with mudstones. (B) Close-up view of a mudstone bed. (C) Middle Siwalik exposure in the Karnali River section, showing interbedded sandstones and mudstones. (D) Close-up view of a floodplain mudstone bed.
Several palaeomagnetic studies were conducted in the Nepalese Siwalik to reveal high-resolution ages of the Siwalik succession (Figure 3). These studies indicate that the Siwalik ranges in age from 16 to 2 Ma (Appel et al., 1991; Gautam and Rosler, 1999; Gautam and Fujiwara, 2000; Ojha et al., 2009). Two well-exposed and palaeomagnetically age-constrained river sections were selected for the present study (Figures 1C and 3), where organic-rich sedimentary rock samples were collected for carbon isotope analysis. These two along-strike sections are Surai Khola, which exposes strata dated between 13 and 2 Ma (Figure 3; Appel et al., 1991; Rösler et al., 1997), and Karnali River, where the exposed strata is determined to be between 16 and 5.2 Ma (Figure 3; Gautam and Fujiwara, 2000).

2 | METHODOLOGY

2.1 | Sampling procedures

With the help of detailed stratigraphic logs generously provided by the senior authors of previous palaeomagnetic studies at the Surai Khola and Karnali River sections (Appel et al., 1991; Rösler et al., 1997; Gautam and Fujiwara, 2000), it was possible to identify most of the palaeomagnetic sample holes, while a few missing holes were located with the help of bearing and thickness data. These palaeomagnetic ages are based on the modified geomagnetic polarity time scale (GPTS) of Cande and Kent (1995).

A total of 47 samples (28 from Karnali River and 19 from Surai Khola), mostly floodplain mudstones and a few palaeosols, were collected employing both regular and high-resolution sampling intervals, averaging 0.5–0.25 Myr (Figure 3). Samples were analysed in the organic geochemistry laboratory at Brown University using CSIA.

2.2 | Sample preparation and isotopic analysis

Sample preparation and chemical treatment followed the procedures described in Huang et al. (2007, 2015) and Gao et al. (2011). Samples were first cleaned three times with dichloromethane to remove any possible surface contamination and freeze-dried for 12 h. The dried samples were ground in a ceramic mortar and pestle, weighed to collect 40–60 g and then mixed with baked sands (2:1) to enhance the mobility of organic compounds. Prepared samples were

**FIGURE 3** Palaeomagnetic chronology of the Surai Khola and Karnali River sections (after Gautam and Fujiwara, 2000) showing stratigraphic depths and geologic ages of the samples analysed in this study.
placed in an ASE 200 (Accelerated Solvent Extractor), and organic materials were collected in 40 ml glass vials using dichloromethane:methanol (9:1) solution at 120°C with 1,200 psi. Extracted organic materials were separated into acid and neutral fractions using a silica-based LC-NH₂ column and dichloromethane:isopropanol (2:1) and ethyl-ether:acetic-acid (96:4) solutions. The neutral fractions of organic compounds were again eluted in a silica gel column (LC-SiO₂) with a hexane solution to obtain n-alkanes. These solutions were quantified using a gas chromatograph and flame ionization detector (GC-FID). Identification of specific compounds was based on a comparison of mass spectra from the GC-MS (gas chromatography-mass spectrometry) with respect to GC retention times (Gao et al., 2011).

Organic compounds (n-alkane fractions) were run in a GCIRMS to obtain compound-specific δ¹³C values. To monitor the analytical accuracy, a laboratory standard (CO₂) with known δ¹³C value (−29.735‰) was measured after every 10 samples. The repeated (two to four times) sample analyses showed a standard deviation of <±0.3‰.

2.3 | Interpreting δ¹³C values

The δ¹³C composition of various materials, including bulk organic matter, lipid biomarker and soil carbonate, has been used as a proxy in published literature to study the shift from C₃ to C₄ plants. In soil carbonates (carbonate nodules), δ¹³C values range from −14‰ to −8‰ for C₃ plants, and >−8‰ for C₄ plants (Quade et al., 1989). In bulk organic matter, modern C₃ plants have a wide range of δ¹³C values ranging from −35‰ to −20‰, whereas C₄ plants have a narrower range between −14‰ and −10‰ (data compilation of Cerling and Harris, 1999). In lipid biomarkers, n-alkane δ¹³C shows an average value of −36‰ for modern C₃ plants, whereas the average value ranges from −24‰ to −20‰ for C₄ plants (Collister et al., 1994; Tipple and Pagani, 2007). These values are in line with the statistical norms of an extensive set of published modern data (δ¹³C of soil carbonate, organic matter and leaf-lipid biomarkers) presented in Cerling et al. (2011) and Magill et al. (2013). These values are used as a general guideline in this study, assuming that the δ¹³C of Miocene atmospheric CO₂ was similar to preindustrial values (Freeman and Colarusso, 2001).

FIGURE 4 Representative chromatographs of n-alkanes for Karnali River (A) and Surai Khola (B) samples, both of which are floodplain mudstones.
(Eglinton and Hamilton, 1967; Tipple and Pagani, 2007; Sachse et al., 2012), as opposed to lower chain-length n-alkanes (<C_{25}) that are commonly sourced from aquatic plants and algae (Ficken et al., 2000; Gao et al., 2011). In terms of relative abundance and in comparison to n-alkonic acid, it was shown that n-alkanes better represent a catchment-integrated signal (Hemingway et al., 2016). In this study, the δ^{13}C values of C_{27}, C_{29} and C_{31} n-alkanes are strongly correlated with each other (Figure 5A,B). The δ^{13}C values, are reported here in reference to the value of C_{27} (Figure 5C). The chromatographs of n-alkane lipids show regular peaks with a strong odd-over-even preference (Figure 4), indicating insignificant thermal alteration of the analysed organic matter (Polissar et al., 2009).

In the Surai Khola section, δ^{13}C values of C_{27} range from −32.3‰ to −18.7‰ with an average value of −25.5‰ (Table 1; Figure 5C). The oldest sample (12 Ma) has a δ^{13}C value of −29.2‰, with the most negative values recorded

**Figure 5** (A) and (B) δ^{13}C n-alkane of C_{27}, C_{29} and C_{31} from the Surai Khola and Karnali River sections, respectively. (C) Comparison of δ^{13}C values of C_{27} from the Surai Khola and Karnali River sections.
around 10 Ma (−32.3‰). From 12 to 8.5 Ma, the δ13C values show a fluctuating trend with an overall decrease of ~2‰. From 8.5 to 6.5 Ma, there is a slow, yet steady increase by ~2‰. The δ13C values start to increase abruptly at 6.5 Ma (Figure 5C), with a dramatic ~6‰ increase to reach −23.6‰ at 5.2 Ma. From 5.4 to 2.5 Ma, the data show a higher-amplitude δ13C variability (Figure 5C) with an overall increase of ~8‰ to reach −19.1‰ at 2.5 Ma.

In the Karnali River section, δ13C values of n-alkane C27 range from −32.8‰ to −27.8‰ with an average of −30.3‰ (Table 2; Figure 5C). The oldest sample, dated to 16 Ma, shows a δ13C value of −29.6‰ that decreases by ~2‰ by 14.5 Ma. The isotopic values start to increase after 14.5 Ma but remain around −28‰ until 9.5 Ma. From 9.5 to 5.2 Ma (the youngest sample age), the values fluctuate by ~3‰ (Figure 5C) within an overall ~3‰ depleting trend.

## DISCUSSION

In the Himalayan foreland, previous isotopic studies of the Neogene vegetation shift from C3 plants to C4 plants used Siwalik palaeosols or fossil teeth, which are generally limited in their stratigraphic occurrence. To overcome this limitation, some isotopic studies used marine sediments from the Bay of Bengal and Arabian Sea (Freeman and Colarusso, 2001; Huang et al., 2007). However, organic materials transported to marine basins from upstream continental lands represent a widely integrated signal of the vast terrestrial ecosystem (Tipple and Pagani, 2007). In contrast, this study used floodplain mudstone and palaeosol samples collected from the terrestrial Siwalik strata at regular and high-resolution intervals of 0.5–0.25 Myr. Moreover, compound-specific (as opposed to bulk) carbon isotopic analysis was applied. This study, therefore, has a greater potential to reveal location-specific as well as direct evidence for the

### TABLE 1

| Sample #      | Age (Ma) | δ13C n-alkane (‰) |
|---------------|---------|-------------------|
| 13-NP-SK-29   | 2.50    | −19.09 −19.72 −18.95 |
| 13-NP-SK-30   | 3.00    | −24.55 −26.79 −24.87 |
| 12-NP-SK-26   | 3.85    | −18.67 −18.96 −18.86 |
| 12-NP-SK-25   | 4.70    | −26.22 −28.12 −25.27 |
| 12-NP-SK-24   | 5.20    | −23.64 −25.62 −25.96 |
| 12-NP-SK-16   | 5.40    | −26.96 −26.84 −33.92 |
| 12-NP-SK-15   | 5.55    | −26.76 −25.65 −23.34 |
| 12-NP-SK-14   | 6.05    | −28.34 −27.22 −25.31 |
| 12-NP-SK-13   | 6.50    | −29.51 −30.39 −31.77 |
| 12-NP-SK-11   | 7.00    | −29.78 −30.01 −29.33 |
| 12-NP-SK-10   | 7.50    | −29.75 −30.56 −31.18 |
| 12-NP-SK-9    | 8.00    | −30.08 −30.05 −29.89 |
| 12-NP-SK-7    | 8.50    | −31.14 −31.45 −32.59 |
| 12-NP-SK-5    | 9.00    | −30.10 −30.47 −32.42 |
| 12-NP-SK-4    | 9.50    | −30.53 −30.68 −31.76 |
| 12-NP-SK-2 (palaeosol) | 10.00 | −32.33 −32.95 −34.31 |
| 12-NP-SK-23   | 10.80   | −28.03 −29.21 −31.80 |
| 12-NP-SK-22   | 11.20   | −30.44 −30.76 −31.89 |
| 12-NP-SK-20   | 11.95   | −29.19 −29.21 −30.45 |

### TABLE 2

| Sample #      | Age (Ma) | δ13C n-alkane (‰) |
|---------------|---------|-------------------|
| 13-NP-KR-50   | 5.20    | −30.02 −31.91 −34.30 |
| 13-NP-KR-49   | 5.25    | −32.78 −32.43 −46.33 |
| 13-NP-KR-48   | 5.50    | −30.46 −33.50 −36.14 |
| 13-NP-KR-47   | 5.90    | −31.92 −32.80 −34.15 |
| 13-NP-KR-46   | 6.00    | −31.79 −33.34 −35.19 |
| 13-NP-KR-44   | 6.25    | −31.90 −31.64 −34.08 |
| 13-NP-KR-43   | 6.40    | −28.57 −30.85 −33.43 |
| 13-NP-KR-42   | 7.00    | −28.15 −29.19 −30.54 |
| 13-NP-KR-41   | 7.25    | −31.76 −32.81 −35.41 |
| 13-NP-KR-40   | 7.50    | −31.43 −32.12 −33.18 |
| 13-NP-KR-39   | 7.75    | −28.96 −30.17 −32.03 |
| 13-NP-KR-38   | 8.00    | −29.67 −30.72 −31.89 |
| 13-NP-KR-37   | 8.25    | −30.79 −30.90 −32.85 |
| 13-NP-KR-36   | 8.50    | −31.05 −31.68 −32.76 |
| 13-NP-KR-35   | 8.75    | −31.75 −31.69 −34.11 |
| 13-NP-KR-34   | 9.00    | −30.14 −31.27 −33.05 |
| 13-NP-KR-32   | 9.50    | −28.04 −28.95 −29.83 |
| 13-NP-KR-29   | 10.25   | −27.80 −28.68 −30.63 |
| 13-NP-KR-28   | 10.50   | −28.06 −28.28 −28.68 |
| 13-NP-KR-25   | 11.25   | −28.30 −29.18 −32.15 |
| 13-NP-KR-22   | 12.00   | −28.87 −29.52 −30.78 |
| 13-NP-KRa-22  | 13.00   | −28.46 −29.54 −31.10 |
| 13-NP-KRa-20  | 13.50   | −28.20 −28.31 −31.06 |
| 13-NP-KR-18   | 14.00   | −28.76 −29.12 −29.90 |
| 13-NP-KR-16 (palaeosol) | 14.50 | −31.22 −31.25 −31.36 |
| 13-NP-KR-14   | 15.00   | −31.02 −32.21 −32.72 |
| 13-NP-KR-13   | 15.40   | −30.19 −32.11 −32.15 |
| 13-NP-KR-11   | 16.00   | −29.63 −33.19 −32.79 |
character of ancient vegetation and their temporal changes in the Himalayan foreland.

Many previous studies have used δ13C values as a proxy for the types of terrestrial vegetation in the Himalayas (Quade and Cerling, 1995; Quade et al., 1995; Freeman and Colarusso, 2001; Huang et al., 2007; Behrensmeyer et al., 2007; Sanyal et al., 2010). Similarly, it is argued here that the δ13C values of n-alkane lipids extracted from floodplain strata reflect the character of past vegetation in the region. The δ13C values of C27 from the Surai Khola section suggest C3 plant domination between 12 and 8.5 Ma (Table 1; Figure 5C). The C4 plants likely started to expand gradually at 8.5 Ma and then rapidly at 6.5 Ma. By 5.2 Ma, C4 vegetation clearly dominated the landscape, as suggested by a heavily enriched δ13C value of −23.6‰. Maintaining this dominance, C4 plants continued to expand between 5.2 and 2.5 Ma (youngest sample age). However, higher-amplitude δ13C fluctuations during this time (Figure 5C) may indicate that patches of C3 forest expanded and contracted several times.

Overall, the lipid biomarker study of the Surai Khola section validates the results of Quade et al. (1995), who used the δ13C values of pedogenic carbonate nodules and bulk organic matter from the same section to document the late Miocene vegetation shift (Figure 6). However, several differences are notable. The data from this study have relatively less scatter with a clearer trend of vegetation change, which likely reflects the fact that compound-specific biomarker proxies can specifically target organic matter sourced from terrestrial higher plants (Freeman and Colarusso, 2001; Tipple and Pagani, 2007). In this respect, this study supports the findings of Freeman and Colarusso (2001) that the terrestrial vegetation record is better reconstructed with isotopic signatures of molecular markers. Although Quade et al. (1995) data suggest a dramatic expansion of C4 plants replacing C3 plants around 7 Ma, the data from this study indicate a stepwise expansion of C4 plants starting gradually at 8.5 Ma and culminating rapidly at 5.2 Ma (Figure 6), by which time C4 vegetation dominated the landscape. Notably, this C4 plant domination since the Miocene-Pliocene boundary (5.3 Ma) has recently been documented in Central Asia (Sun et al., 2013; Shen et al., 2018).

This is the first study to present isotopic data from the Karnali River section (Table 2) with the δ13C values of C27 suggesting a somewhat different vegetation history than that interpreted from the Surai Khola data (Figure 5C). No clear sign of C4 plant expansion is indicated in the Karnali River section until 5.2 Ma, which is the youngest sample from this section. However, slightly more C4 plants may have contributed to a predominately C3 biomass between 14.5 and 9.5 Ma (Figure 5C). While the Surai Khola data suggest expansion of C4 plants over the past 8.5 Ma, the Karnali River data likely indicates expansion of C3 plants between 9.5 and 5.2 Ma. This opposite trend in the vegetation around 9 Ma in two different localities separated laterally by ~200 km along tectonic-strike is intriguing. However, without the δ13C data of the past 5.2 Ma, it is not possible to fully test the concept of a late Neogene vegetation shift in the Karnali River section at this point.

The growing literature regarding the late Neogene vegetation shift increasingly advocates for the asynchronous and spatially heterogeneous expansion of C4 plants replacing the

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**FIGURE 6** Correlation of lipid biomarker data of this study (A) with Quade et al. (1995) data (B) and (C) from the Surai Khola section.
C₃ community in the Himalayan foreland. In the Pakistani Siwalik, previous studies based on the δ¹³C values of soil carbonate and bulk organic matter, showed a transition from C₃ to C₄ vegetation between 8 and ~5 Ma (Quade and Cerling, 1995; Behrensmeyer et al., 2007). Similarly, the δ¹³C values of pedogenic carbonate nodules in the Indian Siwalik suggests the asynchronous appearance of C₄ plants in various areas between 9 and 6 Ma (Sanyal et al., 2004, 2005, 2010). In this study, the lipid biomarkers in the Nepalese Siwalik suggest a transition from C₃ to C₄ plant domination between 8.5 and 5.2 Ma in the Surai Khola section, yet there is no clear indication of C₄ plant expansion in the Karnali River section where data younger than 5.2 Ma are not available (Figure 5C).

With a declined atmospheric CO₂ concentration as a likely necessary precondition (since low CO₂ favours C₄ photosynthetic pathway), the asynchronous and spatially heterogeneous late Neogene expansion of C₄ vegetation can be better explained by region-specific climates (seasonal precipitation, aridity, temperature) and disturbances (fire, grazing) (Tipple and Pagani, 2007; Stromberg, 2011). In addition, local environments like riparian vegetation can be responsible for the anomalous presence of C₃ vegetation in the Siwalik floodplain (Behrensmeyer et al., 2007). For the Karnali River section, it is possible that such local factors, including the influence of the palaeo-Karnali catchment area, might play a role for the likely continued dominance of C₃ vegetation as late as 5.2 Ma.

It is particularly noteworthy that this high-resolution isotopic study reveals a dramatic expansion of C₄ plants since 5.4 Ma in the Himalayan foreland (at the Surai Khola section). A significant expansion of C₄ plants since 5.3 Ma has also been documented in Central Asia, north of the Tibetan Plateau (Sun et al., 2013; Shen et al., 2018). This critical ecological shift at the Miocene-Pliocene boundary (5.3 Ma) suggests an intriguing tectonic-climate link for the Himalayan-Tibetan region. The winter-season precipitation in the region, which is linked to westerlies carrying water vapour from the eastern Eurasian Paratethys Ocean, started to decline after ~8 Ma (Fortelius et al., 2014). Collision and associated uplift of the Pamir Plateau and Tian Shan Ranges further blocked the eastward transport of the winter westerly moisture at the end of the Miocene (Thompson et al., 2015). Here, it is inferred that a critical elevation-gain of the Himalayas around 5.4 Ma likely blocked winter westerly moisture as well as strengthening summer monsoon precipitation significantly. This dramatic shift of climate at the Miocene-Pliocene boundary that is characterized by increased summer precipitation and drier winters likely drove the C₄ plant expansion in the region.

5 | CONCLUSIONS

There are two main advantages in using compound-specific lipid biomarkers when reconstructing past vegetation history: firstly the ability to specifically target terrestrial higher (i.e., vascular) plants, and secondly that sampling is not restricted to palaeosols or fossil teeth. Here, the δ¹³C of n-alkanes (C₂₇₋₃₁) extracted from floodplain shale and palaeosols in the Nepalese Siwalik are used to provide a better knowledge of the Neogene vegetation shift from C₃ to C₄ plants.

Broadly, the data from the Surai Khola section support the results of Quade et al. (1995), who used the δ¹³C values of pedogenic carbonate nodules and bulk organic matter from the same section to document the vegetation shift. However, unlike a dramatic expansion of C₄ plants around 7 Ma that was suggested previously, the data presented here indicate a stepwise expansion of C₄ vegetation starting gradually at 8.5 Ma and culminating rapidly at 5.2 Ma, by which time C₄ vegetation dominated the landscape. This dramatic vegetation shift at the Miocene-Pliocene boundary was likely linked with an intriguing tectonic-climate coupling in the Himalayan-Tibetan region that prompted wetter summers and drier winters.

The first isotopic data from the Karnali River section are also presented. While the Surai Khola data suggest expansion of C₄ plants for the past 8.5 Ma, the Karnali River data show no clear indication of C₄ plant expansion until 5.2 Ma (youngest sample age). This somewhat different vegetation history in two different locations separated laterally by ~200 km along the tectonic-strike is worth exploring in future studies.

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CONFLICT OF INTEREST

The authors have no conflict of interest to declare.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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