1	Lateral variations in vegetation in the Himalaya since the	
2	Miocene and implications for climate evolution	
3	Natalie Vögeli <sup>1*</sup> , Yani Najman <sup>2</sup> , Peter van der Beek <sup>1</sup> , Pascale Huyghe <sup>1</sup> , Peter M. Wynn <sup>2</sup> , Gwladys	
4	Govin², Iris van der Veen <sup>3,4</sup> , Dirk Sachse <sup>3,4</sup> .	
5	<sup>1</sup> Université Grenoble Alpes, Institut des Sciences de la Terre (ISTerre), CS 40700, 38058	
6	Grenoble Cedex 9, France	
7	<sup>2</sup> Lancaster Environment Centre, Lancaster University, LA1 4YQ, UK	
8	<sup>3</sup> GFZ German Research Centre for Geosciences, Earth Surface Geochemistry, Telegrafenberg,	
9	14473 Potsdam, Germany.	
10	<sup>4</sup> Department of Earth and Environmental Sciences, University of Potsdam, 14476 Potsdam,	
11	Germany.	
12	*Corresponding author: <u>natalievoegeli@gmail.com</u>	 Formatted: Font color: Auto
13		Formatted: Font color: Auto
14		
15	4866 Words in main text, 53 references, 6 figures, 3 Appendices	
16		

# Lateral variations in vegetation in the Himalaya since the Miocene and implications for climate evolution

Natalie Vögeli<sup>1</sup>, Yani Najman<sup>2</sup>, Peter van der Beek<sup>1</sup>, Pascale Huyghe<sup>1</sup>, Peter M. Wynn<sup>2</sup>, Gwladys
Govin<sup>2</sup>, Iris van der Veen<sup>3,4</sup>, Dirk Sachse<sup>3,4</sup>.

<sup>1</sup> Université Grenoble Alpes, Institut des Sciences de la Terre (ISTerre), CS 40700, 38058
 Grenoble Cedex 9, France

23 <sup>2</sup> Lancaster Environment Centre, Lancaster University, LA1 4YQ, UK

<sup>3</sup> GFZ German Research Centre for Geosciences, Earth Surface Geochemistry, Telegrafenberg,
 14473 Potsdam, Germany.

<sup>4</sup> Department of Earth and Environmental Sciences, University of Potsdam, 14476 Potsdam,
Germany.

28

# 29 Abstract

30	The Himalaya has a major influence on global and regional climate, in particular on the Asian Formatted: Font color: Auto
31	monsoon system. The foreland basin of the Himalaya contains a record of tectonics and
32	paleoclimate since the Miocene. Previous work on the evolution of vegetation and climate has
33	focused on the central and western Himalaya, where a shift in vegetation has been observed at Formatted: Font color: Auto
34	~7 Ma and linked to increased seasonality, but the climatic evolution of the eastern part of the <b>Formatted:</b> Font color: Auto
35	orogen is less well understood. In order to track vegetation as a marker of monsoon intensity
36	and seasonality, we analyzed $\delta^{13}$ C and $\delta^{18}$ O values of soil carbonate and associated $\delta^{13}$ C values of <b>Formatted:</b> Font color: Auto
37	bulk organic carbon from previously dated sedimentary sections exposing the syn-orogenic Formatted: Font color: Auto
38	detrital Dharamsala and Siwalik Groups in the west, and, for the first time, the Siwalik Group in
39	the east of the Himalayan foreland basin. Sedimentary records span from 20 to 1 Myr in the west
40	(Joginder Nagar, Jawalamukhi, and Haripur Kolar sections) and from 13 to 1 Myr in the east Formatted: Font color: Auto

41 (Kameng section), respectively. The presence of soil carbonate in the west and its absence in the 42 east is a first indication of long-term lateral climatic variation, as soil carbonate requires 43 seasonally arid conditions to develop.  $\delta^{13}$ C values in soil carbonate show a shift from around -10 % to -2 % at ~7 Ma in the west, which is confirmed by  $\delta^{13}$ C analyses on bulk organic carbon 44 that show a shift from around -23 % to -19 % at the same time. Such a shift in isotopic values is 45 46 likely to be associated with a change from C3 to C4 vegetation. In contrast,  $\delta^{13}$ C values of bulk 47 organic carbon remain at  $\sim$ -23 % in the east. Thus, our data show that the current east-west 48 variation in climate was established at 7 Ma. We propose that the regional change towards a 49 more seasonal climate in the west is linked to a decrease of the influence of the Westerlies, 50 delivering less winter precipitation to the western Himalaya, while the east remained annually 51 humid due to its proximity to the monsoonal moisture source.

# 52 1. Introduction

53 The Himalayan belt has a major influence on global and regional climate, by acting as an orographic barrier for air masses and humidity (Boos and Kuang, 2010; Molnar et al., 2010). 54 55 Modern climate shows significant east-west variation in the Himalaya; both mean-annual and 56 winter precipitation on the plains and foothills are higher in the east, while the west is 57 characterized by more pronounced winter aridity (Figure 1; Bookhagen and Burbank, 2006; 58 2010). This variation is due to the two major atmospheric circulation systems influencing the 59 climate of the Himalayan region: the Indian Summer Monsoon (ISM) and the Westerlies (Kotlia 60 et al., 2015). The ISM takes up moisture in the Bay of Bengal and transports it towards the 61 Himalaya during the northern-hemisphere summer months (e.g., Molnar et al., 2010), whereas 62 the Westerlies bring moisture from the Mediterranean, Black and Caspian Seas and are most 63 efficient in winter (Benn and Owen, 1998; Cannon et al., 2015). Generally, the influence of the 64 Westerlies is greater in the western part of the Himalayan region (Cannon et al., 2015; Caves et al., 2015; Kotlia et al., 2015). The proximity to the moisture source in the Bay of Bengal, makes 65 66 the eastern Himalaya very humid (Bookhagen et al., 2010),

Formatted: Font color: Auto
Formatted: Font color: Auto
Formatted: Font color: Auto
Formatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto
Formatted: Font color: Auto
Formatted: Font color: Auto

67 These lateral variations in modern climate are linked to vegetation patterns, in particular the 68 relative importance of C3 versus C4 plants. C3 plants are favored in a cool and humid climate, 69 whereas C4 plants prefer intense light, warm and water-stressed conditions (Ehleringer, 1988). 70 An additional factor that possibly influences the evolution of C4 plants is growing season 71 temperature and precipitation, favoring C4 plants in drier periods (Cotton et al., 2016). The 72 distinct stable carbon-isotopic signature of C3 versus C4 vegetation allows paleo-vegetation to 73 be tracked from the sedimentary record: pure C3 vegetation has  $\delta^{13}C_{org}$  values between -22 % 74 and -30 %, whereas  $\delta^{13}C_{org}$  values of C4 plants range from -10 % to -14 % (Cerling et al., 75 1997). The lateral variations in modern climate and vegetation are expressed by the signature of modern organic carbon transported in the foreland (Galy et al., 2008a; Fig. 1): sediments 76 77 sampled from Himalayan tributaries at the mountain front have  $\delta^{13}C_{org}$  values around -25 ‰, 78 indicating dominance of C3 plants at higher elevations within the mountain belt. These values 79 remain stable within the eastern Brahmaputra catchment, whereas they increase to values 80 around -22 ‰ in the Western Ganga catchment (Fig. 1), implying laterally varying vegetation (from C4 in the west to C3 in the east) in the floodplain. 81 82 An important question is when and why the modern spatial patterns in climate and vegetation 83 were established in the Himalayan foreland. The onset of the ISM is dated back to at least the 84 middle Miocene (Dettman et al., 2001) and possibly the Late Eocene (Licht et al., 2014). 85 Likewise, the Westerlies have been argued to influence Asian climate since the Eocene (Caves et

al., 2015). However, the evolution of regional climate and vegetation patterns will depend on the 87 relative strength of these two systems through time, which remains largely unknown.

86

88 The foreland basin of the Himalaya contains a sedimentary record of vegetation and 89 paleoclimate since Miocene times, within the continental detrital pre-Siwalik and Siwalik 90 Groups. The record of spatial and temporal variations in vegetation holds information on climate 91 evolution, in particular patterns of atmospheric circulation, seasonality and the origin and 92 transport of humidity (Hoorn et al., 2000; Sanyal et al., 2004; Gupta 2010, amongst others). 93 Carbon and oxygen isotopic compositions of soil carbonates and soil organic matter from preFormatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto Formatted: Font color: Auto

Formatted: Font color: Auto

94	Siwalik and Siwalik sediments in Nepal, Northwest India and Pakistan have been used to	
95	reconstruct changes in vegetation and climate during the Neogene (Quade et al., 1989, 1995a;	
96	Quade and Cerling, 1995; Sanyal et al., 2010; Singh et al., 2013). These records consistently show	
97	a shift in $\delta^{13}C$ values at ${\sim}7$ Ma, which has been interpreted as a change from C3 to C4 vegetation,	
98	which was initially interpreted to be related to an intensification of the ISM (Quade et al., 1989).	Formatted: Font color: Auto
99	Steinke et al. (2010) suggest that this change was rather linked to an increase in aridity, and	
100	therefore a weakening of the ISM. A similar shift has also been recorded in the distal Himalayan-	
101	derived sediments of the Bay of Bengal (France-Lanord and Derry, 1994). It has been argued	
102	that the late-Miocene expansion of C4 plants is a global phenomenon due to a decrease in	Formatted: Font color: Auto
103	atmospheric pCO <sub>2</sub> (Cerling et al., 1997), global cooling and/or increased aridity (Herbert et al.,	Formatted: Font color: Auto
104	2016). Others, however, argue that $pCO_2$ was already at a level favorable for C4 plants during	
105	Oligocene times (Pagani et al., 2005; Beerling and Royer, 2011) and that the spread of C4 plants	
106	during the late Miocene should therefore have other, more regional triggers.	Formatted: Font color: Auto
107	The focus of previous studies on Himalayan climate and vegetation records has been entirely on	Formatted: Font color: Auto
108	the western and central Himalayan foreland; no climate and vegetation data are available east of	
109	Nepal. In order to obtain better spatial insight into the evolution of the monsoon climate,	
110	precipitation patterns and the expansion of C4 plants along strike in the Himalaya, we present	
111	and compare new $\delta^{13}C$ and $\delta^{18}O$ data of pedogenic carbonate and organic matter from the	
112	northwestern and the poorly studied eastern Himalayan foreland basin. Lateral variations in the	Formatted: Font color: Auto
113	evolution of the vegetation yield further insight into how and under what climatic conditions C4	
114	plants developed or not, suggesting that regional influences play a major role.	
115	2. Setting	
116		
116	Neogene Himalayan foreland-basin sediments are composed of the fluvial Dharamsala Group of	
117	Late Oligocene and early Miocene age (Burbank et al., 1996), and the Siwalik Group deposited	
118	since the early Miocene. The Dharamsala rocks consist of continental fluvial, lacustrine or deltaic	Formatted: Font color: Auto
119	sediments, and contain fine-to medium grained sandstones, siltstones and overbank mudstones	

120 with soil carbonate nodules (Raiverman and Seshavataram, 1983). The sediments of the Siwalik

Group are exposed nearly continuously along the front of the Himalayan range, with only minor age variation along strike (Burbank et al., 1996). They were deposited in the foreland before being incorporated in the foothills due to southward propagation of deformation and onset of motion on the Main Frontal Thrust (MFT).

125 The Siwalik Group shows an overall coarsening- and thickening-upward trend, interpreted as 126 recording increasingly proximal deposition (DeCelles et al., 1998), and is divided into the Lower, 127 Middle and Upper Siwaliks (LS, MS, US). The LS were deposited by high-sinuosity streams 128 (Nakayama and Ulak, 1999). The Middle Siwaliks (MS) are characterized by thickly bedded 129 sandstones, which are medium- to coarse-grained and often rich in mica. The MS represent a 130 depositional environment of large braided rivers. The Upper Siwaliks (US) consist of beds of 131 conglomerates alternating with sandstone beds, deposited by gravelly braided rivers. Paleosols 132 are developed throughout most of the Siwalik sections, with lateral and temporal variations in 133 abundance: they are more abundant in the LS and in the west. Paleosols are characterized in 134 western and central Himalayan sections by the presence of soil-carbonate nodules.

135 We sampled three sections exposing Dharamsala and Siwalik deposits in the western Himalaya; 136 the Joginder Nagar (JN), Jawalamukhi (JW) and Haripur Kolar (HK) sections in Himachal 137 Pradesh, and one Siwalik section in the eastern Himalaya; the Kameng River (KM) section in 138 Arunachal Pradesh (Figures 1, 2). All sections have previously been dated by 139 magnetostratigraphy (Meigs et al., 1995; Sangode et al., 1996; White et al., 2001; Chirouze et al., 140 2012). They span a time range of 20-1 Ma in the west and 13-1 Ma in the east. In the western 141 sections, we collected paleosols and associated carbonate nodules, as well as fine-grained 142 mudstone in zones without well-developed paleosols. Carbonate nodules are lacking in the 143 Kameng section (Figure 2); therefore only mudstones, where possible from paleosols, were 144 sampled. Additionally, modern river mud was sampled from riverbanks in proximity to the 145 sections in both the west and the east. Modern river samples in the west (Beas River and Iner 146 Khad River) were collected at an elevation of  $\sim$ 640 m within the sections, whereas the modern 147 Kameng River sample was collected at an elevation of  $\sim 100$  m downstream of the Siwalik.

Formatted: Font color: Auto

### 148 3. Methods

 $^{13}C/^{12}C$  and  $^{18}O/^{16}O$  ratios (expressed as  $\delta^{13}C$  and  $\delta^{18}O$  values respectively) of soil carbonate 149 150 nodules were determined using a multiflow analyser linked to an Isoprime 100 continuous flow 151 mass spectrometer at the Lancaster University, UK. Approximately 600-700 µg of sample 152 powder was drilled from each carbonate nodule and digested online at 90°C with dehydrated 153 phosphoric acid in a He-flushed exetainer. Product CO<sub>2</sub> was analyzed for  $\delta^{13}C_{CO2}$  and  $\delta^{18}O_{CO2}$  and 154 corrected against VPDB and VSMOW, respectively, using within-run analyses of international 155 standards NBS18, LSVEC and CO-1. Within-run standard replication (1  $\sigma$ ) was <0.1 % for both C 156 and O isotope ratios. Sample replication based on separate drill aliquots of powder from the 157 same carbonate nodule was <0.1‰ for  $\delta^{13}$ C and <0.2‰ for  $\delta^{18}$ O (1  $\sigma$ ).

158  $^{13}C/^{12}C$  ratios (expressed as  $\delta^{13}C$  values) of bulk organic matter were determined using an 159 Elementar Vario Micro elemental analyser linked to a VisION continuous flow mass 160 spectrometer at the University of Lancaster. The carbonate content of each sample was removed 161 by acid digestion using 1M ultrapure HCl and the resultant sample washed repeatedly using de-162 ionised water and centrifugation. Approximately 10 mg of each prepared sample was combusted 163 within tin capsules at 960 °C to yield CO2 for determination of  $\delta^{13}$ Corg,  $\delta^{13}$ C values were corrected 164 against VPDB using internal reference materials calibrated to international standards. Within-165 run  $\delta^{13}$ C replication (1  $\sigma$ ) was <0.2 ‰ for standards and <0.25 ‰ for samples.

### 166 4. Results

167 The three sections in Himachal Pradesh (Western Himalaya) provide a continuous age record 168 over the past 20 Ma (Figure 3). Prior to ~7 Ma,  $\delta^{13}$ C values of soil carbonate ( $\delta^{13}$ C<sub>soil carb</sub>) range 169 between -8 ‰ and -13 ‰, whereas at ~7 Ma a shift towards more positive  $\delta^{13}$ C values, ranging 170 from +2 ‰ to -8 ‰, is observed (Figure 3; Appendix 1).  $\delta^{18}$ O values range mostly from -11 ‰ 171 to -4 ‰, except in the older part of the sections, where some values are as low as -14 ‰. A weak 172 trend towards more positive  $\delta^{18}$ O values over time is observed. As noted above, soil carbonate 173 was not present in the eastern Himalayan section. Formatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto
Formatted: Font color: Auto

174	We additionally margured a continuous record of $S^{13}C$ in organic carbon $(S^{13}C)$ in both the	
174	We additionally measured a continuous record of $\delta^{13}$ C in organic carbon ( $\delta^{13}$ C <sub>org</sub> ) in both the	Formatted: Font color: Auto
175	western and eastern sections (Figure 4; Appendix 1). In the west, a clear shift towards more	
176	positive $\delta^{13}C_{org}$ values is observed at ~7 Ma, synchronous with the $\delta^{13}C_{soil carb}$ . Before 7 Ma, $\delta^{13}C_{org}$	
177	values range between -23 $\%_0$ and -27 $\%_0$ , while values are less negative, from -18 $\%_0$ to -23 $\%_0$ ,	
178	after 7 Ma. In the east, in contrast, $\delta^{13}C_{\text{org}}$ values remain constant between -29 $\%$ and -23 $\%$	
179	since the middle Miocene and no shift towards more positive values is observed. Organic matter	
180	from modern river sediments show $\delta^{13}C$ values of approximately -26 $\%$ and -23.5 $\%$ in the	
181	west and in the east, respectively (Figure 4). Total Organic Carbon content (TOC) in the western	Formatted: Font color: Auto
182	samples is mostly <0.5 % although samples from the JN can have up to 15% TOC (Figure 5). TOC	
183	of most Kameng samples is also <0.5 %, with some samples showing values up to 3%. There is	
184	no correlation between $\delta^{13}C_{org}$ values and TOC (Appendix 2).	
185	5. Discussion	
186	5.1. Modern river sediments and vegetation	
100	5.1. Modern river seuments and vegetation	Formatted: Font color: Auto Formatted: Font color: Auto
187	The modern Ganga/Brahmaputra floodplain is widely used for agriculture and is therefore	Formatted. Fort color. Auto
188	predominantly covered in C3 plants such as rice crops, and in the east by tea plantations (Blasco	
189	et al., 1996). This is not reflected by the organic carbon transported in the Ganga and	
190	Brahmaputra Rivers: $\delta^{13}C_{\text{org}}$ values of $\sim\!\!-21.9~\%$ in the modern Ganga floodplain are	
191	representative of a mixture of C4 and C3 plants, suggesting the presence of C4 plants in the west,	
192	whereas the modern Brahmaputra River carries organic carbon with $\delta^{13}C_{org}$ values of -23.0 $\%_0$ in	
193	the range of C3 plants (Galy et al., 2008a).	
194	Measured $\delta^{13}C_{org}$ values of modern river muds in both the west (Beas and Jner Khad River) and	
195	the east (Kameng River) are in the range of C3 plants. However, these modern river sediments	
196	were not collected in the floodplain but at the mountain front, where they will contain detrital	
197	organic carbon of C3 plants transported from higher elevations (Dobremez, 1978) and/or fossil	
198	organic carbon from Himalayan formations (Galy et al., 2008a), which both have a more negative	
199	isotopic signal. Dharamsala and Siwalik sediments were deposited further into the floodplain,	Formatted: Font color: Auto
200	and hence should carry a signal of floodplain vegetation.	Formatted: Font color: Auto
	8	

201

# 202 5.2. Possible factors influencing the isotopic signal

203 Earlier studies (Quade et al., 1995a; Quade and Cerling, 1995; Singh et al., 2007; Sanyal et al., 204 2010) measured  $\delta^{13}$ C on soil carbonate nodules, which can only be found in the western and 205 central Himalayan Siwalik sections, and have consistently shown a change towards more 206 positive values at ~7 Ma from Pakistan to Nepal (Quade et al., 1995a; Quade and Cerling, 1995). 207 This change was interpreted in terms of a shift in vegetation, from initially dominated by C3 208 plants to containing C4 species. In order to compare the western and the eastern Himalaya in 209 this study, we rely on  $\delta^{13}C_{org}$  of bulk organic matter, as soil carbonate nodules are absent in the 210 east. In both the western and the eastern sections,  $\delta^{13}C_{org}$  values range between -23  $\%_0$  and -29 211 % before 7 Ma, indicating vegetation dominated by C3 plants. After 7 Ma,  $\delta^{13}C_{org}$  in the western 212 sections demonstrates an isotopic shift to values enriched in  ${}^{13}C$  (~-19 ‰), suggesting that a 213 component of the organic matter comprises C4 species. Carbonate nodules from western 214 Himalayan sections analyzed in this study show a trend similar to  $\delta^{13}C_{org}$ , demonstrating a shift 215 from C3-dominated vegetation composition, to an increasing proportion of C4 species in the 216 younger sections. In the east, in contrast,  $\delta^{13}C_{org}$  values stay in the range of C3 plants throughout 217 the sedimentary succession (Figure 4).

218 In modern soils, carbonate precipitates in equilibrium with soil CO<sub>2</sub> (Cerling et al., 1989) 219 following an isotopic enrichment in <sup>13</sup>C of 10.36 ‰, (Cerling et al., 1989). Diffusional effects 220 cause soil-respired CO<sub>2</sub> to be further enriched in <sup>13</sup>C by 4.4 %. The total fractionation between 221 soil organic matter and soil carbonate is  $\sim 14$  ‰, at 25°C to  $\sim 17$  ‰ at 0°C (Cerling et al., 1989). 222  $\delta^{13}C_{soil carb.}$  values show a greater (~ +10‰) shift towards more positive values after 7 Ma than 223 the  $\delta^{13}C_{org}$  values (~ + 6 ‰; Figure 6). This discrepancy in the absolute value of the isotopic shift 224 to signatures more enriched in <sup>13</sup>C likely reflects the nature of carbonate nodule production and 225 organic matter source. Whereas soil carbonates reflect only the soil CO<sub>2</sub> characteristics and 226 temperature during formation, the  $\delta^{13}C_{org}$  values are more susceptible to bias by inherited and 227 transported material.



228	Although $\delta^{13}C_{org}$ is usually interpreted to represent isotopic values of vegetation in the floodplain	Forma
229	at the time of sediment deposition, it can potentially be biased by several factors, such as input	Forma
230	of (likely C3 plant dominated) organic carbon from high elevations (Dobremez, 1978) and/or	Forma
231	input of fossil organic carbon. The amount of fossil organic carbon present in Himalayan river	Forma
232	sediments was estimated using the radiocarbon content of total organic carbon (TOC) of modern	
233	suspended and bedload sediments (Galy et al., 2008a; 2008b). Galy et al. (2008b) estimated the	Forma
234	total amount of fossil organic carbon transported in the Ganga and Brahmaputra Rivers between	
235	0.02 and 0.03 $\%$ Sediments of the sampled sections mostly have TOC values >0.1 $\%$ (Figure 5)	Forma
236	suggesting that the proportion of fossil carbon should be low, <30 % at most, if the modern	Forma
237	amounts of transported fossil organic carbon can be extrapolated to the past. $\delta^{13}C_{\text{org}}$ of fossil	
238	organic carbon from Himalayan source rocks varies from -28 to -14.6‰ (Galy et al., 2008a); it is	Forma
239	therefore unclear what the effect of varying proportions of fossil organic carbon on the observed	
240	$\delta^{13}C_{\text{org}}$ signal would be. However, we have no reason to assume the influence of fossil carbon to	
241	be very different from east to west and our data suggest this influence to be rather constant over	
242	time (see below). It is therefore unlikely that the spatial and temporal variations in $\delta^{13}C_{\text{org}}$ values	
243	could be explained by variable fossil organic carbon content.	
244	Additionally, the efficiency of oxidation of organic carbon and hence the replacement of	Forma
245	inherited carbon by floodplain carbon can influence the $\delta^{13}C_{org}$ signal in modern river sediments	
246	(Galy et al., 2008a; 2011). Organic-carbon oxidation varies between the Ganges and	Forma
247	Brahmaputra foreland basin, being more efficient in the Ganges floodplain due to different	
248	hydrological settings: the Ganges is a meandering river, whereas the Brahmaputra is a braided	
249	river with a narrower floodplain (Galy et al., 2008a). Oxidation of inherited organic carbon from	Forma
250	vegetation at higher altitudes, hence with a C3 signal, is efficient in the Ganges floodplain (Galy	Forma
251	et al., 2008a), as shown by the proportion of C4 organic matter increasing downstream in the	
252	Ganges floodplain (Figure 1). This pattern is not present in the Brahmaputra floodplain,	Forma
253	suggesting that the influence of inherited carbon could therefore be greater in the Brahmaputra.	
254	Even though paleosols are less developed in the east, high TOC values indicate the presence of	

Formatted: Font color: Auto
Formatted: Font color: Auto
Formatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto

ormatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto

organic matter acquired from surface organic litter during pedogenesis (Figure 5). Degradation of organic matter in soils could have an influence on the  $\delta^{13}C_{org}$ : detrital organic matter has a ~1-2 %0 more negative  $\delta^{13}C$  signal than soil organic matter (von Fischer and Tieszen, 1995 and references therein). The more negative  $\delta^{13}C$  values in the Kameng section could therefore be explained by the presence of more detrital organic matter. Differences in floodplain dynamics during transport and a greater influence of inherited organic carbon could possibly bias the signal but are unlikely to cancel out the entire C4 signal in the eastern Himalaya,

262 Further information on different organic carbon sources could potentially be derived from lipid 263 biomarker analysis (i.e. compound-specific C- and H-isotope analysis; Freeman et al. 2001; 264 Sachse et al., 2012). We extracted n-alkanes from samples of both the western and the eastern 265 sections (see Appendix 3), but unfortunately n-alkane preservation was generally low in the 266 sediments. Moreover, evidence of diagenetic overprinting was found in sediments with a 267 sufficient concentration, evidenced by an absence of the predominance of odd carbon numbered 268 n-alkane chain lengths (expressed as the carbon preference index, CPI), which is prevalent in 269 modern plant and sediment samples. In modern plant material and immature sediments, CPI 270 values are generally significantly >3 and up to 20, whereas we found values around 1 in the 271 Kameng samples, indicating diagenetic overprinting or addition of fossil carbon at the time of 272 sedimentation. As a result of this, compound-specific hydrogen and carbon stable isotopic values 273 would likely have been altered towards less negative values (Radke et al., 2005). While 274 diagenetic overprinting could also have affected bulk  $\delta^{13}C_{org}$  values by homogenizing the isotopic 275 signal (Cerling et al., 1984; Bera et al., 2010), our CPI data remained uniform at values around 1 276 from the base of the section until ca. 2 Ma, indicating a similar degree of overprinting in these 277 samples. Since we did not find any change in bulk  $\delta^{13}$ Corg values corresponding to changes in CPI 278 values, we argue that any potential overprinting affected all samples equally and as such relative 279 changes can still be interpreted from bulk  $\delta^{13}C_{org}$  values. In addition, the samples presented in 280 Figure 3 are in the same isotopic range as modern soil carbonate nodules, indicating that 281 diagenesis is unlikely to have influenced the isotopic values of the sedimentary samples.

Formatted: Font color: Auto

Formatted: Font color: Auto

282	The isotopic values of pedogenic carbonate are in equilibrium with soil $\ensuremath{\text{CO}}_2$ derived from	
283	irreversible oxidation of organic matter in a diffusion-controlled soil system at different	
284	temperatures, The isotopic equilibrium factor is dependent on temperature, hence if pedogenic	 Formatted: Font color:
285	carbonate precipitates in equilibrium with soil CO2, $\delta^{13}C_{\text{soil carb}}$ should be enriched by ~14 $\%$ at	
286	25 °C and by 17 $\%$ at 0 °C, respectively (Cerling et al. 1984, 1989). Carbonate nodules and their	
287	corresponding organic matter of the Dharamsala and Siwalik paleosols plot mostly at	 Formatted: Font color:
288	temperatures between 0 and $25^{\circ}$ C and are therefore not isotopically altered by diagenesis	
289	(Figure 6), but rather formed within this soil temperature range. Samples below the $0^{\circ}$ C line	
290	could reflect an inconsistency in the enrichment of <sup>13</sup> C in soil carbonate nodules compared to the	
291	co-existing organic matter. Most of the samples plotting under the $0^{\circ}$ C line show a strong C4	
292	signal, and the offset between $\delta^{13}C_{org}$ and $\delta^{13}C_{soil\ carb.}$ in these samples is greater than expected	 Formatted: Font color:
293	from isotopic equilibrium considerations. A possible explanation for this enhanced offset could	
294	be that the carbonate nodule formed in a sediment body (e.g. another paleosol horizon) that was	
295	separate from the parent organic matter. Alternatively, organic matter may be more influenced	
296	by inherited organic matter from C3 vegetation, whereas the $\delta^{13}C_{\text{soil carb}}$ would more directly	
297	represent the local vegetation cover. However, this inconsistency does not reflect a diagenetic	 Formatted: Font color:
298	overprint, as it is found in the youngest samples, where diagenesis is least likely to occur,	 Formatted: Font color:
299	$\delta^{18}O_{\text{soil carb.}}$ values of the three western sections show a slight change towards more positive	
300	values (Figure 3), comparable to $\delta^{18}\text{O}$ values of the Surai Khola section in Nepal (Quade et al.,	
301	1995a). Only $\delta^{18}O_{soil \ carb.}$ values from Pakistan show a clear shift from values < -8 towards more	
302	positive values at $\sim$ 8-6 Ma (Quade and Cerling 1995). In contrast, samples from this study	
303	already show $\delta^{18}O_{\text{soil carb.}}$ values > -8 before 7 Ma (Figure 3). A change in $\delta^{18}O_{\text{soil carb.}}$ which forms	
304	in-situ from soil water, can be associated with a change in either soil temperature $\delta^{18}O_{soil \ carb.}$	 Formatted: Font color:
305	being positively correlated with mean annual temperature; Cerling, 1984) and/or precipitation	 Formatted: Font color:
306	source; $\delta^{18}$ O values of precipitation of moisture transported from the Bay of Bengal are generally	 Formatted: Font color:
307	lighter (more negative) than $\delta^{\rm 18}{\rm O}$ values of moisture transported by the Westerlies (Caves et al.,	
308	2015 and references therein). The isotopic change over time was measured on samples of three	
l		

or: Auto

309 separate sections (Figure 2) at different longitudinal locations; therefore the isotopic signature 310 from precipitation may be location specific rather than representing change over time. However, 311 there is no clear shift in  $\delta^{18}O_{soil carb.}$  values going from one section to another (Figure 3), 312 suggesting this effect to be minimal. As all sediments were deposited in the foreland, the 313 influences of any altitudinal effects (Dansgaard, 1961) can also be excluded.

314 The coarsening-upward trend of sedimentary rocks throughout the sections reflects a change in 315 depositional environment and location in the foreland basin, which varies from a distal 316 floodplain for the Dharamsala and Lower Siwaliks to deposition closer to the mountain front in 317 the Upper Siwaliks. At different locations in the foreland basin, the source of precipitation may 318 vary from moisture influenced by the Westerlies, to moisture sourced from the ISM. More 319 positive  $\delta^{18}O_{soil}$  carb. values over time could therefore indicate an increasing influence of 320 Westerlies with respect to ISM moisture sources, and/or a trend towards a warmer, drier 321 climate, conducive to the growth of C4 vegetation.

# 322 5.3. What caused the change of vegetation at ~7 Ma?

323 C3 and C4 plants grow in different environments and the  $\delta^{13}$ C signal can therefore be used as an 324 indirect climate indicator. Our data show that a change in vegetation occurred at ~7 Ma in the 325 western Himalaya, but not in the east, where C3 plants have been dominant since the middle 326 Miocene. As we have argued above, differences in floodplain setting (Galy et al., 2008a, 2011), 327 while influencing the signal, cannot explain the observed lateral difference and neither can input 328 of fossil organic carbon (Galy et al., 2008b). For this reason, there must be a remarkable lateral 329 variation in the evolution of climate in the Himalayan region. The change at 7 Ma in the west and 330 central Himalaya has been interpreted as resulting from a "stronger monsoon", characterized by 331 greater seasonality (Quade et al., 1989, 1995a; Quade and Cerling, 1995). However, increased 332 seasonality does not necessarily reflect higher amounts of monsoon precipitation; it could also 333 indicate relatively less winter precipitation and thus a more arid (annual-average) climate 334 (Molnar, 2005). C3 plants in the east indicate lower seasonality and higher annually averaged 335 precipitation, consistent with modern precipitation patterns (Bookhagen and Burbank, 2010).

Formatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto

Formatted: Font color: Auto

The expansion of C4 plants in the west could therefore be a consequence of decreased winter precipitation, hence more seasonality associated with less (annually averaged) humidity, leading to a more arid climate. Overall, this difference in the  $\delta^{13}$ C composition post-7 Ma is proposed to reflect water availability, with lower water availability in the west initiating a decline in C3 plants and a rise in C4 species (cf. Freeman and Colarusso, 2001). Dettman et al. (2001) likewise suggest a change in Indian summer monsoon characteristics and drying of the climate at 7.5 Ma. This scenario is supported by a change in  $\delta^{18}O_{\text{soil carb.}}$  towards more positive values.

343 Higher humidity in the east could be explained by the proximity to the Bay of Bengal, which is 344 the major moisture source of precipitation in this area (Bookhagen et al., 2005). The western 345 Himalaya is influenced by the Westerlies (Kotlia et al., 2015), which bring in winter 346 precipitation. A decrease in the intensity of the Westerlies at 7 Ma would lead to more 347 seasonality in the western floodplain, with drier periods in winter. An alternative explanation 348 for a generally more arid climate in the western Himalaya could be a decrease of moisture 349 transport from the Bay of Bengal and the Arabian Sea, possibly linked to a decrease in the 350 intensity of the ISM. However, this would result in less seasonality, hence a less favorable 351 climate for C4 plants. The spatially variable record of  $\delta^{13}C_{org}$  values strongly suggests that the 352 change in vegetation at 7 Ma did not occur simultaneously along the Himalayan foreland, 353 indicating that the change is at least partly driven by regional factors rather than being linked 354 only to a global change in atmospheric  $pCO_2$ . This supports the findings of Pagani et al. (2005) 355 and Beerling and Royer (2011), who noted that atmospheric pCO<sub>2</sub> levels favoring C4 plants were 356 already reached during the Oligocene. Other dry regions such as the Mediterranean have been 357 dominated by C3 plants since the Miocene (Quade et al., 1994, 1995b), also indicating that the 358 late-Miocene expansion of C4 plants was not a global phenomenon. Regionally dependent 359 factors, such as differences in seasonality or humidity, have clearly played a role in determining 360 Himalayan vegetation patterns through time, Lateral variations in vegetation suggest that there 361 is a threshold somewhere along the Himalayan front, where the amount of (either annual or 362 winter) precipitation becomes too large for C4 plants to spread.

Formatted: Font color: Auto

### 363 6. Conclusions

364 Stable carbon and oxygen isotopes were analyzed in carbonate nodules of the Joginder Nagar, 365 Jawalamukhi and Haripur Kolar sections in the western Himalaya.  $\delta^{13}C_{soilcarb}$  values show a clear 366 shift towards more positive values at 7 Ma, similar to the results of earlier studies in the western 367 and central Himalaya. The lack of carbonate nodules in Siwalik sediments of the Kameng section, 368 eastern Himalaya, is a first indicator that the lateral environmental and climatic differences in 369 the modern Himalaya are representative of long-term climatic patterns. In order to directly 370 compare the western and eastern sections, stable carbon isotopes on organic matter were 371 analyzed and show a clear spatial difference. In the west,  $\delta^{13}C_{org}$  values shift towards more 372 positive values at 7 Ma, consistent with the results on carbonate nodules, whereas they remain 373 constant over the last 13 Ma in the east. The  $\delta^{13}$ C of organic matter reflects the evolution of 374 vegetation, with the development of C4 plants in the west and an environment that remains 375 favorable for C3 plants in the east. Such variations in vegetation imply differences in climate, 376 which became more seasonal and overall drier in the west at 7 Ma. The eastern Himalaya is 377 more proximal to the main moisture source for precipitation (the Bay of Bengal); therefore, even 378 though climate may have varied, it remained less seasonal and more humid, inhibiting the 379 evolution of C4 plants. Therefore, the change in climate in the west and the onset of lateral 380 variation is most likely caused by a change in strength of atmospheric circulation, such as a 381 weakening of the influence of the Westerlies. These findings suggest that the late-Miocene 382 expansion of C4 vegetation does not depend solely on atmospheric pCO<sub>2</sub> but also on regional 383 changes in aridity and seasonality. Newly developed methods, such as clumped isotopes or 384 stable isotopes on compound-specific organic carbon, even though unsuccessful in this study, 385 could provide further insight into the climatic evolution and the development of C4 vegetation, 386 both globally and regionally in the Himalayan region. This study has provided the first paleo-387 climate and -vegetation data from the eastern Himalaya; however, more such studies are needed 388 to refine our understanding of the evolution of climate and vegetation in this area.

Formatted: Font color: Auto

Formatted: Font color: Auto

389

# 390 Acknowledgements

Montserrat Auladell-Mestre and David Hughes are thanked for the help preparing and
measuring the samples. We acknowledge financial support from Initial Training Network (ITN)
iTECC funded by the EU REA under the FP7 implementation of the Marie Curie Action, under
grant agreement # 316966. ISTerre is part of the Labex OSUG@2020 (ANR10 LABX56). Reviews
of earlier versions of this manuscript by Greg Retallack, Seema Sing and 3 anonymous reviewers
have helped to significantly improve its clarity and focus.

397

## 398 7. References

- Beerling, D. J., and Royer, D. L., 2011, Convergent Cenozoic CO<sub>2</sub> history: Nature Geoscience, v. 4,
  no. 7, p. 418-420.
- Benn, D. I., and Owen, L. A., 1998, The role of the Indian summer monsoon and the mid-latitude
  westerlies in Himalayan glaciation: review and speculative discussion: Journal of the
  Geological Society, London, v. 155, p. 353-363.
- 404 Bera, M.K., Sarkar, A., Tandon, S.K.; Samanta, A. and Sanyal, P., 2010, Does burial diagenesis reset
  405 pristine isotopic compositions in paleosol carbonates?: Earth and Planetary Science
  406 Letters, v. 300, p. 85-100.
- Blasco F., Bellan M.F. and Aizpuru M., (1996) A vegetation map of tropical continental Asia at
  scale 1.5 million: Journal of Vegetation Science, v. 7, p.623–634.
- Bookhagen, B., Thiede, R. C., and Strecker, M. R., 2005, Late Quaternary intensified monsoon
  phases control landscape evolution in the northwest Himalaya: Geology, v. 33, no. 2, p.
  149-152.
- Bookhagen, B., and Burbank, D. W., 2006, Topography, relief, and TRMM-derived rainfall
  variations along the Himalaya: Geophysical Research Letters, v. 33, no. 8, L08405.

414 Bookhagen, B., and Burbank, D. W., 2010, Toward a complete Himalayan hydrological budget:

- 415 Spatiotemporal distribution of snowmelt and rainfall and their impact on river 416 discharge: Journal of Geophysical Research: Earth Surface, v. 115, F03019.
- Boos, W. R., and Kuang, Z., 2010, Dominant control of the South Asian monsoon by orographic
  insulation versus plateau heating: Nature, v. 463, no. 7278, p. 218-222.
- Burbank, D.W., Beck, R.A. and Mulder, T., 1996, The Himalayan foreland basin. In The Tectonic
  Evolution of Asia, Yin, A and Harrison, TM (eds) Cambridge University press, p. 149-188.
- 421 Cannon, F., Carvalho, L.M.V., Jones, C., and Bookhagen, B., 2015, Multi-annual variations in winter
- 422 westerly disturbance activity affecting the Himalaya. Clim. Dyn. 44, 441–455.

Formatted: Font color: Auto

423	caves, j. K., while K. M. J., Granam, S. A., Sjöström, D. J., Mutch, A., and Granberlam, C. I., 2013,	
424	Role of the westerlies in Central Asia climate over the Cenozoic: Earth and Planetary	
425	Science Letters, v. 428, p. 33-43.	
426	Cerling, T. E., 1984, The stable isotopic composition of modern soil carbonate and its	
427	relationship to climate: Earth and Planetary Science Letters, v. 71, no. 2, p. 229-240.	
428	Cerling, T. E., Harris, J. M., MacFadden, B. J., Leakey, M. G., Quade, J., Eisenmann, V., and	
429	Ehleringer, J. R., 1997, Global vegetation change through the Miocene/Pliocene	
430	boundary: Nature, v. 389, no. 6647, p. 153-158.	
431	Cerling, T. E., Quade, J., Wang, Y., and Bowman, J. R., 1989, Carbon isotopes in soils and palaeosols	
432	as ecology and palaeoecology indicators: Nature, v. 341, no. 6238, p. 138-139.	
433	Chirouze, F., Dupont-Nivet, G., Huyghe, P., van der Beek, P., Chakraborti, T., Bernet, M., and Erens,	
434	V., 2012, Magnetostratigraphy of the Neogene Siwalik Group in the far eastern Himalaya:	
435	Kameng section, Arunachal Pradesh, India: Journal of Asian Earth Sciences, v. 44, p. 117-	
436	135.	
437	Clift, P. D., Hodges, K. V., Heslop, D., Hannigan, R., Van Long, H., and Calves, G., 2008, Correlation	
438	of Himalayan exhumation rates and Asian monsoon intensity: Nature Geoscience, v. 1,	
439	no. 12, p. 875-880.	
440	Cotton, J.M., Cerling, T.E., Hoppe, K.A., Mosier, T.M., Still, C.J., 2016. Climate, CO <sub>2</sub> , and the history	Form
441	of North American grasses since the Last Glacial Maximum. Science Advances 2,	
442	e1501346.	
443	Dansgaard, W., 1961, The isotopic composition of natural waters: Medd. om Gronland, v. 165, no.	
444	2, p. 1-120.	
445	DeCelles, P. G., Gehrels, G. E., Quade, J., Ojha, T. P., Kapp, P. A., and Upreti, B. N., 1998, Neogene	
446	foreland basin deposits, erosional unroofing, and the kinematic history of the Himalayan	
447	fold-thrust belt, western Nepal: Geological Society of America Bulletin, v. 110, no. 1, p. 2-	
448	21.	

- Dettman, D. L., Kohn, M. J., Quade, J., Ryerson, F. J., Ojha, T. P., and Hamidullah, S., 2001, Seasonal
  stable isotope evidence for a strong Asian monsoon throughout the past 10.7 m.y:
  Geology, v. 29, no. 1, p. 31-34.
- 452 Dobremez, J. F. et al., 1978, Carte écologique du Népal 1/250000. University of Grenoble,
  453 Grenoble.
- Ehleringer, J. R., 1988, Carbon isotope ratios and physiological processes in aridland plants. In
  Ehleringer, J.R., Nagy, K.A. (Eds.), Stable Isotopes in Ecological Research.: Springer, New
  York, p. 41-54.
- 457 France-Lanord, C., and Derry, L. A., 1994, δ<sup>13</sup>C of organic carbon in the Bengal Fan: Source
  458 evolution and transport of C3 and C4 plant carbon to marine sediments: Geochimica et
  459 Cosmochimica Acta, v. 58, no. 21, p. 4809-4814.
- Freeman, K. H., and Colarusso, L. A., 2001, Molecular and isotopic records of C4 grassland
  expansion in the late Miocene: Geochimica et Cosmochimica Acta, v. 65, no. 9, p. 14391454.
- Galy, V., France-Lanord, C., and Lartiges, B., 2008a, Loading and fate of particulate organic carbon
  from the Himalaya to the Ganga–Brahmaputra delta: Geochimica et Cosmochimica Acta,
  v. 72, no. 7, p. 1767-1787.
- Galy, V., Beyssac, O., France-Lanord, C., and Eglinton, T., 2008b, Recycling of graphite during
  Himalayan erosion: A geological stabilization of carbon in the crust: Science, v. 322, p.
  943-945.
- Galy, V., Eglinton, T., France-Lanord, C., and Sylva, S., 2011, The provenance of vegetation and
  environmental signatures encoded in vascular plant biomarkers carried by the GangesBrahmaputra rivers: Earth and Planetary Science Letters, v. 304, no. 1–2, p. 1-12.
- 472 Herbert, T.D., Lawrence, K.T., Tzanova, A., Peterson, L.C., Caballero-Gill, R., Kelly, C.S., 2016. Late
  473 Miocene global cooling and the rise of modern ecosystems. Nature Geosci. 9, 843–847.
- 474 Hoorn, C., Ohja T., Quade J., 2000. Palynological evidence for vegetation development and climate
  475 change in the Sub-Himalayan Zone (Neogene, Central Nepal).Palaeogeogr.Palaeoclimatol.
- 476 Palaeoecol. 163, 133–161.

477	Kotlia, B.S., Singh, A.K., Joshi, L.M., Dhaila, B.S., 2015. Precipitation variability in the Indian	Fo
478	Central Himalaya during last ca. 4,000 years inferred from a speleothem record: Impact	
479	of Indian Summer Monsoon (ISM) and Westerlies. Quat. Int. 371, 244–253.	
480	Licht, A., van Cappelle, M., Abels, H. A., Ladant, J. B., Trabucho-Alexandre, J., France-Lanord, C.,	
481	Donnadieu, Y., Vandenberghe, J., Rigaudier, T., Lecuyer, C., Terry Jr, D., Adriaens, R.,	
482	Boura, A., Guo, Z., Soe, A. N., Quade, J., Dupont-Nivet, G., and Jaeger, J. J., 2014, Asian	
483	monsoons in a late Eocene greenhouse world: Nature, v. 513, no. 7519, p. 501-506.	
484	Meigs, A. J., Burbank, D. W., and Beck, R. A., 1995, Middle-late Miocene (>10 Ma) formation of the	
485	Main Boundary thrust in the western Himalaya: Geology, v. 23, no. 5, p. 423-426.	
486	Molnar, P., 2005, Mio-Pliocene Growth of the Tibetan Plateau and Evolution of East Asian	
487	Climate: Palaeontologia Electronica, v. 8, no. 1, p. 1-23.	
488	Molnar, P., Boos, W. R., and Battisti, D. S., 2010, Orographic Controls on Climate and Paleoclimate	
489	of Asia: Thermal and Mechanical Roles for the Tibetan Plateau: Annual Review of Earth &	
490	Planetary Sciences, v. 38, p. 77-102.	
491	Ojha, T. P., Butler, R. F., DeCelles, P. G., and Quade, J., 2009, Magnetic polarity stratigraphy of the	
492	Neogene foreland basin deposits of Nepal: Basin Research, v. 21, no. 1, p. 61-90.	
493	Pagani, M., Zachos, J. C., Freeman, K. H., Tipple, B., and Bohaty, S., 2005, Marked decline in	
494	atmospheric carbon dioxide concentrations during the Paleogene: Science, v. 309, p. 600.	
495	Quade, J., Cater, J. M. L., Ojha, T., Adam, J., and Harrison, T. M., 1995a, Late Miocene	
496	environmental change in Nepal and the northern Indian subcontinent: Stable isotopic	
497	evidence from paleosols: Geological Society of America Bulletin, v. 107, no. 12, p. 1381-	
498	1397.	
499	Quade, J., and Cerling, T. E., 1995, Expansion of C4 grasses in the Late Miocene of Northern	
500	Pakistan: evidence from stable isotopes in paleosols: Palaeogeography,	
501	Palaeoclimatology, Palaeoecology, v. 115, no. 1–4, p. 91-116.	
502	Quade, J., Cerling, T. E., and Bowman, J. R., 1989, Development of Asian monsoon revealed by	
503	marked ecological shift during the latest Miocene in northern Pakistan: Nature, v. 342,	

504 no. 6246, p. 163-166.

505	Quade, J., Cerling, T. E., Andrews, P., and Alpagut, B., 1995b, Paleodietary reconstruction of	
506	Miocene faunas from Paşalar, Turkey using stable carbon and oxygen isotopes of fossil	
507	tooth enamel: Journal of Human Evolution, v. 28, no. 4, p. 373-384.	
508	Quade, J., Solounias, N., and Cerling, T. E., 1994, Stable isotopic evidence from paleosol	
509	carbonates and fossil teeth in Greece for forest or woodlands over the past 11 Ma:	
510	Palaeogeography, Palaeoclimatology, Palaeoecology, v. 108, no. 1, p. 41-53.	
511	Radke J., Bechtel A., Gaupp R., Puttmann W., Schwark L., Sachse D. and Gleixner G. (2005)	Formatted: Fo
512	Correlation between hydrogen isotope ratios of lipid biomarkers and sediment maturity.	
513	Geochim. Cosmochim. Acta 69, 5517–5530.	
514	Rowley, D. B., and Currie, B. S., 2006, Palaeo-altimetry of the late Eocene to Miocene Lunpola	
515	basin, central Tibet: Nature, v. 439, no. 7077, p. 677-681.	
516	Sachse D., Billault I., Bowen G. J., Chikaraishi Y., Dawson T. E., Feakins S. J., Freeman K. H., Magill	Formatted: Fo
517	C. R., McInerney F. A., van der Meer M. T. J., Polissar P., Robins R. J., Sachs J. P., Schmidt H	
518	L., Sessions A. L., White J. W. C., West J. B. and Kahmen A. (2012) Molecular	
519	Paleohydrology: Interpreting the Hydrogen-Isotopic Composition of Lipid Biomarkers	
520	from Photosynthesizing Organisms. Annu. Rev. Earth Planet. Sci. 40, 221–249.	
521	Sangode, S. J., Kumar, R., and Ghosh, S. K., 1996, Magnetic Polarity Stratigraphy of the Siwalik	
522	Sequence of Haripur area (H.P.), NW Himalaya: Journal Geological Society India, v. 47, no.	
523	June 1996, p. 683-704.	
524	Sanyal, P., Bhattacharya, S.K., Kumar, R., Ghosh, S.K., Sangode, S.J., 2004. Mio-Pliocene monsoonal	
525	record from Himalayan foreland basin (Indian Siwalik) and its relation to vegetational	
526	change. Palaeogeogr. Palaeoclimatol. Palaeoecol. 205, 23–41.	
527	Sanyal, P., Sarkar, A., Bhattacharya, S. K., Kumar, R., Ghosh, S. K., and Agrawal, S., 2010,	
528	Intensification of monsoon, microclimate and asynchronous C4 appearance: Isotopic	
529	evidence from the Indian Siwalik sediments: Palaeogeography, Palaeoclimatology,	

530 Palaeoecology, v. 296, no. 1–2, p. 165-173. ont color: Auto

ont color: Auto

- 531 Singh, B. P., Lee, Y. I., Pawar, J. S., and Charak, R. S., 2007. Biogenic features in calcretes developed
- on mudstone: Examples from Paleogene sequences of the Himalaya, India: Sedimentary
  Geology, v. 201, no. 1–2, p. 149-156.
- Singh, S., Awasthi, A. K., Parkash, B., and Kumar, S., 2013, Tectonics or climate: What drove the
  Miocene global expansion of C4 grasslands?: International Journal of Earth Sciences, v.
  102, no. 7, p. 2019-2031.
- Steinke, S., Groeneveld, J., Johnstone, H., and Rendle-Bühring, R., 2010, East Asian summer
  monsoon weakening after 7.5 Ma: Evidence from combined planktonic foraminifera
  Mg/Ca and δ180 (ODP Site 1146; northern South China Sea): Palaeogeography,
  Palaeoclimatology, Palaeoecology, v. 289, no. 1–4, p. 33-43.
- 541 von Fischer, J.C. and Tieszen L.L., 1995, Carbon isotope characterization of vegetation and soil
  542 organic matter in subtropical forests in Luquillo, Puerto Rico: Biotropica, v. 27, p. 138543 148.
- White, N. M., Parrish, R. R., Bickle, M. J., Najman, Y. M. R., Burbank, D., and Maithani, A., 2001,
  Metamorphism and exhumation of the NW Himalaya constrained by U–Th–Pb analyses
  of detrital monazite grains from early foreland basin sediments: Journal of the Geological
  Society, v. 158, no. 4, p. 625-635.
- Wobus, C. W., Hodges, K. V., and Whipple, K. X., 2003, Has focused denudation sustained active
  thrusting at the Himalayan topographic front?: Geology, v. 31, no. 10, p. 861-864.

550

# 551 Figures

Figure 1: Map of the Himalayan region, with  $\delta^{13}$ C of modern river organic carbon from Galy et al., 2008a. The Himalayan range is indicated schematically in grey. Sections are indicated in red: JW: Jawalamukhi; JN: Joginder Nagar; HK: Haripur Kolar; KM: Kameng. Lower plot shows comparison of modern annual precipitation data (TRMM) in proximity to the sampled sedimentary sections in the west and east.

Figure 2: Stratigraphy of sections of the Dharamsala and Siwalik Groups in the west (A) and inthe east (B), with field photos showing sedimentological characteristics of different sub-groups.

Figure 3: A:  $\delta^{13}C_{soil \ carb.}$  and  $\delta^{18}O$  values of soil carbonate in the western Himalaya. Different symbols indicate the different sections (HK: Haripur Kolar; JW: Jawalamukhi; JN: Joginder Nagar). B:  $\delta^{13}C_{soil \ carb.}$  and  $\delta^{18}O$  values of soil carbonate in Pakistan from Quade and Cerling (1995).

Figure 4:  $\delta^{13}C_{org}$  of bulk organic carbon in the western (HK: Haripur Kolar; JW: Jawalamukhi; JN Joginder Nagar) and the eastern (KM: Kameng) Himalayan sections. Light and dark grey shaded bars indicate  $\delta^{13}C_{org}$  values characteristic of C3 and C4 plants, respectively (Cerling et al., 1997).

Figure 5: Total organic carbon content (TOC) vs age [Ma], zoomed in to values below 0.5 % on
the left and values above 0.5 % on the right. TOC values above 0.07% indicate dominant biogenic
Corg, from soil organic matter and floodplain vegetation, rather than detrital and fossil Corg (Galy
et al., 2008b).

Figure 6:  $\delta^{13}C_{soil\ carb.}$  of soil carbonate nodules vs.  $\delta^{13}C_{org}$  of co-existing organic matter of the western Dharamsala and Siwalik Group sections. Solid and dashed lines represent isotopic values of pedogenic carbonate in isotopic equilibrium with the soil CO<sub>2</sub> derived from irreversible oxidation of organic matter in a diffusion controlled soil system at different temperatures (Cerling et al. 1989).

575

# 576 Appendices:

- 577 Appendix 1: Sample overview and results of  $\delta^{13}C_{org}$ ,  $\delta^{13}C_{soil carb.}$  and  $\delta^{18}O_{soil carb.}$
- 578 Appendix 2: Total organic carbon (TOC) vs.  $\delta^{13}C_{org}$  in the western (a) and eastern (b) sections,
- 579 respectively.
- 580 Appendix 3: n-alkane analysis on the Kameng river section
- 581
- 582
- 583