

1 **A re-assessment of the chronostratigraphy of late**
2 **Miocene C₃-C₄ transitions**

3 **L. Tauxe¹ and S.J. Feakins²**

4 ¹Scripps Institution of Oceanography, University of California San Diego, La Jolla, CA, 92093-0220
5 ²Dept. Earth Sciences, University of Southern California, Los Angeles, CA, 90089

6 **Key Points:**

- 7 • A change from forest to grassland ecologies began in the late Miocene but chrono-
- 8 logies of the published records are based on different time scales.
- 9 • Recalibration to a consistent time scale demonstrates the diachroneity of the eco-
- 10 logical change in different places around the globe.
- 11 • New constraints link a major C₄ expansion in the late Miocene with evidence for
- 12 carbon cycle perturbations.

Corresponding author: Lisa Tauxe, 1tauxe@ucsd.edu

13 **Abstract**

14 Combining magnetostratigraphic and carbon isotopic data for the late Miocene can
 15 provide a temporal framework for an isotopic shift first documented in soil carbonate nod-
 16 ules of northern Pakistan. The shift has been interpreted as a change in vegetation from
 17 trees and shrubs (using the C_3 photosynthetic pathway) to grassland (using the C_4 path-
 18 way). The cause of the event has been hotly debated and its timing is close to a shift
 19 in carbon isotopes in the marine realm. Further understanding depends critically on tim-
 20 ing.

21 Unfortunately, temporal calibration of the various records published over decades
 22 relied on different time scales. To address the lack of a consistent chronology, we have
 23 re-evaluated the constraints for the carbon isotopic shifts recorded from the Indian sub-
 24 continent. These show a diachronous transition ranging in age from about 7.8 Ma in Pak-
 25 istan to as late at 6 Ma in Nepal. The record from IODP Expedition 355 Site U1457,
 26 drilled on the Indus fan shows that the transition in peninsular India began at about 7.2
 27 Ma. Similar records from the African margin saw an earlier shift to C_4 dominance start-
 28 ing around 10 Ma and those from Australia and South America transitioned later, dur-
 29 ing the Pliocene. The diachroneity around the globe does not invalidate pCO_2 as a driver,
 30 but is consistent with it being one of several drivers of the global C_4 expansion.

31 **Plain Language Summary**

32 Grasslands expanded on the Indian sub-continent in the late Miocene. Precise chrono-
 33 logical control is critical to compare the timing of the expansion between regions and to
 34 evaluate the possible causes (and consequences) of the ecological transformation. Here
 35 we take a new look at published records from Pakistan, India and Nepal and update the
 36 age models to a modern geomagnetic reversal timescale. We then compare these to new
 37 records from the marine sediments obtained during IODP Expedition 355 to the Indus
 38 Fan and others from the African margin. Based on microfossil appearance and paleo-
 39 magnetic constraints, the timing of the transition to C_4 vegetation is found to be asyn-
 40 chronous across the globe, probably initiating in Africa after 10 Ma, then proceeding to
 41 Pakistan at around 7.8 Ma and reaching Nepal by ~ 6 Ma. Elsewhere in the world (Aus-
 42 tralia and Argentina) the transition happened several million years later. We also review
 43 recent model and proxy evidence to find the causes of the C_4 grassland expansion.

44 **1 Introduction**

45 The classic papers of Cerling et al. (1993) and Cerling et al. (1997) (based on ear-
 46 lier work by Quade et al. (1989)) introduced the hypothesis that an expansion of C_4 grasses
 47 in the late Miocene was linked to a global driver: a putative drop in pCO_2 in the atmo-
 48 sphere. However constraining pCO_2 in the ancient atmosphere is notoriously difficult (see,
 49 e.g., Cerling (1992)), and alkenone carbon isotopic records published by Pagani et al. (1999)
 50 shortly after that by Cerling et al. (1997) found no evidence for changes in pCO_2 in the
 51 late Miocene. Thus the CO_2 -hypothesis as a driver for change in vegetation in the late
 52 Miocene fell out of favor, and other drivers including aridity, herbivory and fire were con-
 53 sidered more likely (e.g., Osborne (2008)). However, a global pCO_2 change would not
 54 trigger a synchronous C_4 expansion as thresholds exist in other climatic variables includ-
 55 ing temperature, rainfall and fire as demonstrated for Africa by Higgins and Scheiter (2012).

56 Zhou et al. (2018) recently modeled the role of various climatic drivers (CO_2 , tem-
 57 perature, water and light) in controlling the competitive advantage of C_3 versus C_4 veg-
 58 etation globally using a fully coupled climate model under mid Miocene conditions run
 59 with three pCO_2 concentrations to bracket the range of estimates for the Oligocene to
 60 late Miocene (see Figure 1). Simulations run at 600 ppm pCO_2 , suggested that water

61 limitation determined a few ‘hot spots’ where C_4 adaptations could emerge including in
 62 northern Africa and central Asia and to a lesser extent western North America, south-
 63 ern South America and Africa and western Australia (Figure 1a) and these could be lo-
 64 cations where early C_4 evolution innovations occurred (Edwards et al., 2010). For pCO_2
 65 of 400 ppm, the optimality of C_4 plants increased in these regions and was favored over
 66 much larger regions, driven primarily by lower pCO_2 with feedbacks of increased light
 67 as the canopy opened (Figure 1b). Under low pCO_2 (270 ppm) shown in Figure 1c, op-
 68 timal conditions for C_4 dominance would have intensified and expanded. However, this
 69 scenario sees increases in C_4 coverage beyond that reported in late Miocene reconstruc-
 70 tions (especially in places like East Asia and Australia) and thus may be a sign that such
 71 low pCO_2 levels were not reached in the late Miocene. Testing the completeness of the
 72 forcing assumptions in these simulations requires both a knowledge of CO_2 variations
 73 and an accurate chronology for when vegetation changes occurred around the globe.

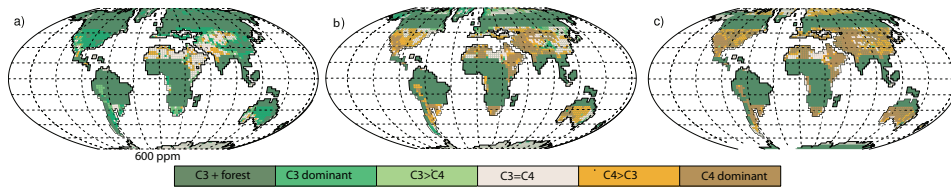


Figure 1. Map of predictions of C_3 versus C_4 dominance under various pCO_2 scenarios: a) 600 ppm, b) 400 ppm and c) 270 ppm CO_2 . [Figure modified from Zhou et al. (2018)].

74 Estimating pCO_2 in the past is a difficult problem (see for example commentary
 75 by Beerling and Royer (2011)). Many proxies have been developed and some have data
 76 in the late Miocene interval ($\delta^{11}B$; Sosdian et al. (2018), B/Ca; Tripathi et al. (2009), alkenones;
 77 Pagani et al. (1999), leaf stomata; Van der Burgh et al. (2009), ; diatom frustules; Mejia
 78 et al. (2017), foraminiferal $\Delta^{13}C$; Holbourn et al. (2018) and paleosol carbonates (Cerling,
 79 1992). These data were reported on different time scales and the details of precise dates
 80 are frequently obscure (e.g., which time scale was used). For example, the leaf stomata
 81 data of Van der Burgh et al. (2009) are poorly calibrated with respect to age and it is
 82 difficult to tie them to the tight temporal framework required for inter-calibration. A
 83 larger problem is that none of the pCO_2 proxies agree with each other (see compilation
 84 of Foster et al. (2017) in Figure 2).

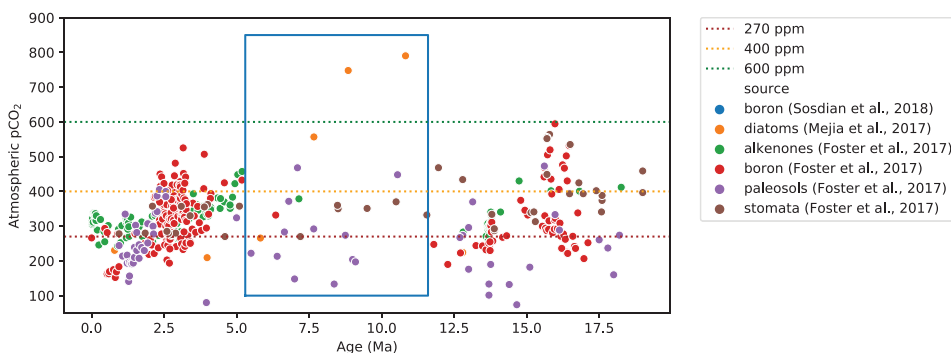


Figure 2. CO_2 proxy data synthesized in Foster et al. (2017), and updated with data from Mejia et al. (2017), and Sosdian et al. (2018), colored by proxy type and data source. The box is the late Miocene interval of interest in this paper.

85 The interval of interest here of the late Miocene (shown in the box in Figure 2) has
 86 a very sparse data set and little agreement among the various types of proxies. Accord-
 87 ing to the compilation of Foster et al. (2017), there is no convincing evidence for a sig-
 88 nificant change in pCO₂ since the early Miocene except for the high values found in be-
 89 tween 15 and 17 Ma and those from about 3 to 2.5 Ma which have been associated with
 90 the mid-Miocene Climatic Optimum and the Pliocene warm interval respectively. More
 91 recent records, such as the data from $\Delta^{13}\text{C}$ of organic matter within diatom frustules
 92 of Mejia et al. (2017) suggest high pCO₂ and a drop from a high of 800 ppm to 200 ppm
 93 with the peak at around 11 Ma and the drop beginning at around 8 Ma.

94 A recent data set from (Holbourn et al., 2018) based on foraminiferal $\Delta^{13}\text{C}$ records
 95 from planktic and benthic foramifera from ODP Site 1146 (shown in Figure 3) are the
 96 most detailed for the interval of interest here and can be placed on the time scale used
 97 here (Gradstein et al., 2012). These data provide no quantitative estimate of pCO₂, how-
 98 ever. Nonetheless $\Delta^{13}\text{C}$ data are interpreted to indicate a drop in pCO₂ values between
 99 ~ 7 and ~ 7.5 Ma, possibly synchronous with part of the carbon isotopic shift in plant
 100 proxies discussed in this paper. Thus, these new pCO₂ proxies and the modeling efforts
 101 of (Zhou et al., 2018) reawaken a possibility of pCO₂ mediating the regional C₄ expan-
 102 sion in a diachronous progress as modification of the mechanism for a global synchronous
 103 transition envisioned by Cerling et al. (1993) and Cerling et al. (1997).

104 In this paper, we revisit the timing of the C₄ expansion around the globe by com-
 105 piling published records and updating the magnetostratigraphic framework for their col-
 106 lective re-interpretation. This is an important step as the age of a single reversal in the
 107 time period of interest, for example the termination of Chron C4n, changed from ~ 6.3
 108 Ma in the time scale of LaBrecque et al. (1977) to ~ 7.5 Ma in the current standard of
 109 Gradstein et al. (2012). In addition, we present a new record from IODP Site U1457 which
 110 has carbon isotopes from leaf waxes (Feakins et al., This volume), and reasonable chronos-
 111 tratigraphic constraints from biostratigraphy and magnetostratigraphy (Routledge et al.,
 112 2019), updated here. We also consider other recent records of leaf wax carbon isotopes
 113 recovered in DSDP and ODP cores taken around Africa (Feakins et al., 2013; Uno et al.,
 114 2016; Polissar et al., 2019) that have common biostratigraphic tie points.

115 Once the timing of the change in vegetation on the Indian sub-continent is secured,
 116 we can ask what combination of climate and CO₂ factors drove the C₄ expansion. We
 117 can also ask whether there is positive C₄ feedback on the carbon cycle. Commonalities
 118 of the timing of C₄ expansion and a shift in the $\delta^{13}\text{C}$ of marine carbonates (Feakins et
 119 al., This volume) have been noted. Grasslands can be quite efficient at sequestering CO₂
 120 in soils (Fornara & Tilman, 2008; Spiesman et al., 2018; Yang et al., 2019) in compar-
 121 ison to the thin soil under the C₃ forests they replaced in the Indian sub-continent (al-
 122 though elsewhere replaced C₃ grasses (Feakins et al., 2013)). This positive feedback in
 123 turn could enhance global cooling (e.g., Retallack (2013); Retallack et al. (2018)).

124 2 Timing of the C₃-C₄ transition

125 2.1 The marine realm

126 In addition to the carbon isotope shift on land first detected by Quade et al. (1989)
 127 and expanded on by Cerling et al. (1993), there is also a marked carbon isotope tran-
 128 sition recorded in deep sea sediments at about the same time. In fact, the story of the
 129 late Miocene shift in carbon isotopes began with a study by Keigwin of deep sea sedi-
 130 ments (Keigwin, 1979). He documented a mysterious decrease of up to 0.8‰ in the $\delta^{13}\text{C}$
 131 in benthic foraminifera in Deep Sea Drilling Project (DSDP) cores recovered at Site 158
 132 in the Panama Basin (Easternmost Pacific) and Site 310 (Hess Rise) in the North Pa-
 133 cific (Figures 4a and 5 and Table 1). Keigwin inferred an age for the shift of ~ 6.5 Ma
 134 based on correlation of the biostratigraphy (including the first occurrence (FO) of *Cer-*

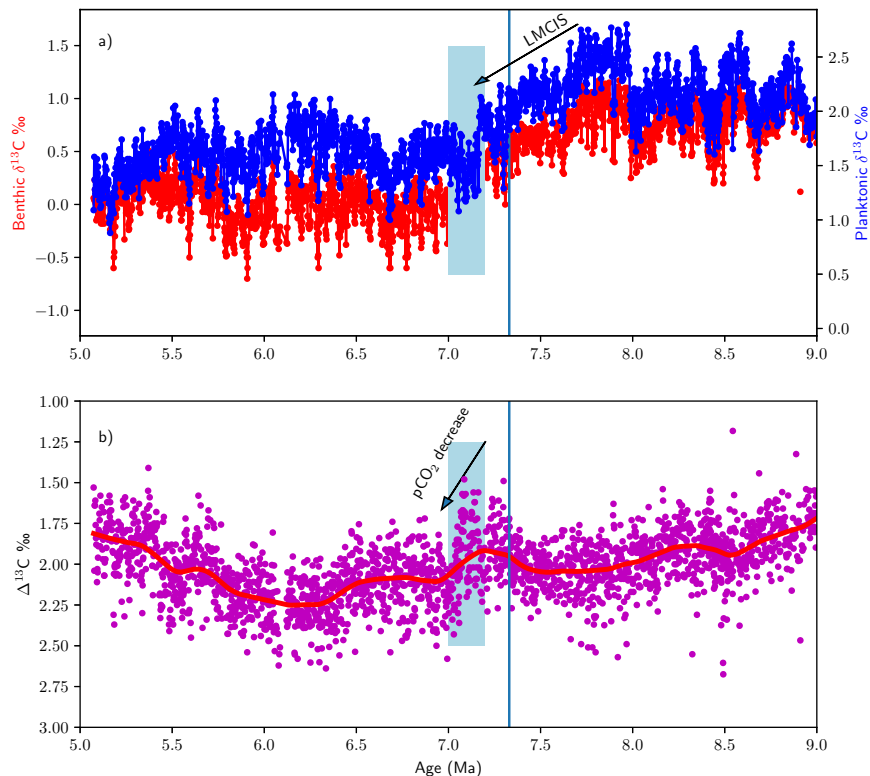


Figure 3. a) Benthic (red) and planktic (blue) $\delta^{13}\text{C}$ data of Holbourn et al. (2018) from ODP Site 1146. Blue vertical line is the FO of *Amaurolithus primus* at that site. The “Late Miocene carbon isotope shift” (LMCIS) from 8 Ma to 6.8 Ma is shown as in Holbourn et al. (2018). b) Difference between planktic and benthic $\delta^{13}\text{C}$ ($\Delta^{13}\text{C}$), plotted with lower values to the top. Higher values have been interpreted as resulting from lower atmospheric pCO_2 concentrations. The decrease in pCO_2 inferred between 7.2 and 7.1 Ma is as in Holbourn et al. (2018).

135 *atolithus primus* (now known as *Amaurolithus primus*), a diagnostic nannofossil. This
 136 datum had been tied to the middle of “Epoch 6” (Theyer & Hammond, 1974) or in mod-
 137 ern parlance Chron C3B, now estimated at 7.4 Ma in the GTS12 time scale of Gradstein
 138 et al. (2012) (see also Raffi et al. (2006)). We note that Schneider (1995) tied this marker
 139 to the middle of C3Br.2r so it is more likely to be 7.3 Ma, the age adopted here. Bender
 140 and Keigwin (1979) suggested that the shift might reflect “either a global decrease in
 141 upwelling rate or a different abyssal circulation pattern before the shift.”

142 Shortly after the discovery by Keigwin, Vincent et al. (1980) found similar shifts
 143 in the Indian Ocean at DSDP Site 238 (Figures 4a and 5), also closely associated with
 144 the FO of *A. primus*. Vincent et al. (1980) echoed Keigwin in explaining the shift as re-
 145 sulting from “changes in ocean circulation”. The Vincent et al. study was immediately
 146 followed by the study of Haq et al. (1980) who found the same shift occurring just af-
 147 ter the FO of *Amaurolithus* spp. (i.e., *A. primus*?) in the world’s oceans.

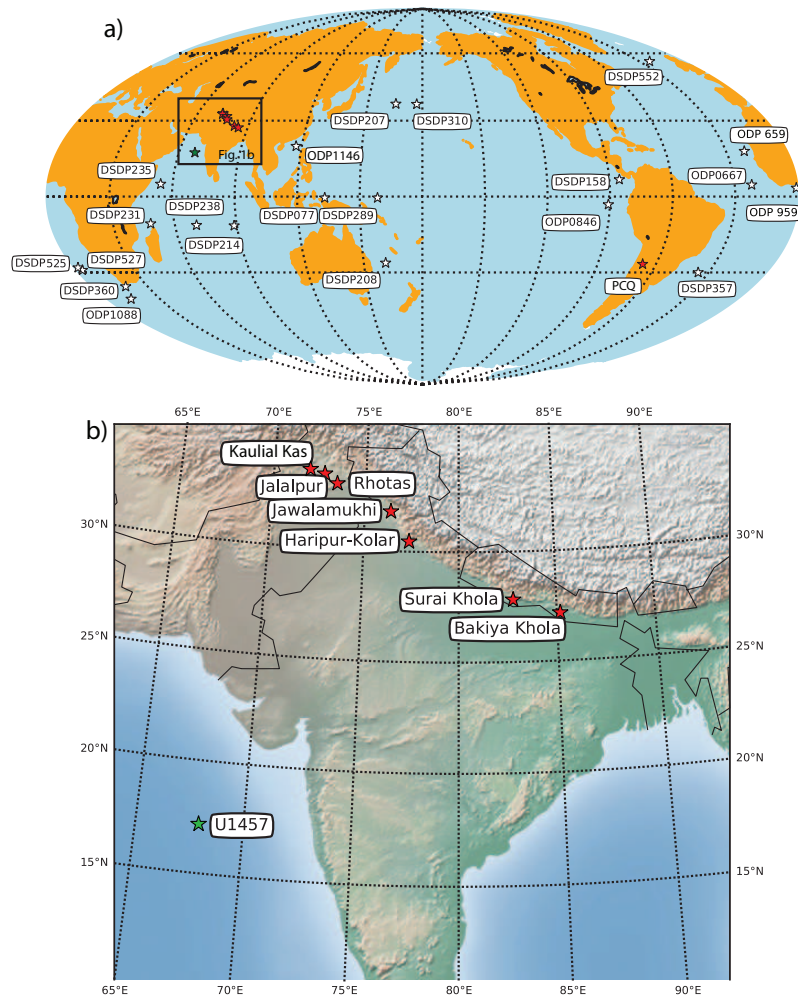


Figure 4. Map of locations of studies cited in this paper showing a) global compilation, b) Indian subcontinent and margins. See Table 1 for references.

148 More recently, Cramer et al. (2009) compiled records from Deep Sea Drilling Project
 149 (DSDP) and Ocean Drilling Program (ODP) cores with benthic foraminiferal carbon iso-
 150 topic data ($\delta^{13}\text{C}$) organized by ocean basin. In this way, water mass differences between
 151 basins resulting from ocean circulation patterns are held separate, allowing for detection
 152 of global carbon isotopic transitions rather than artifacts of the averaging of a varied num-
 153 ber of records with ocean basin bias. Because we are interested in the precise timing of
 154 the carbon isotope shifts relative to each other, we consider only those records that also
 155 have the FO of *Amaurolithus* spp. (see Figure 5 and Table 1). We plot the different records
 156 against stratigraphic depth relative to the FO of *Amaurolithus* spp. in Figure 5. There
 157 does appear to be a decrease in $\delta^{13}\text{C}$ associated with the FO of *Amaurolithus* spp. in
 158 many records, but some occur prior to the biostratigraphic datum (e.g., ODP1088) while
 159 others show no change at all (DSDP 552).

160 2.2 Records from the continents

161 Starting with Quade et al. (1989), the focus of Miocene carbon isotopes studies shifted
 162 to the continents. They found a large ($\sim 10\%$) increase in $\delta^{13}\text{C}$ values in paleosol car-
 163 bonate of the Siwalik sequence of Pakistan which they interpreted as the signature of

Table 1. Locations of studies cited in the text. Rhotas is a composite of two sections (Dhabwala & Basawa Kas).

Site/Section	Lat.	Lon.	FO <i>A. spp.</i> (mbsf/ <i>mcd</i>)	Ref.
DSDP077	-0.48	133.23	130	Keigwin and Corliss (1986), Woodruff et al. (1981), Woodruff and Savin (1989)
DSDP158	6.63	-85.24	150	Keigwin (1979)
DSDP207	36.96	165.43	94	Haq et al. (1980)
DSDP208	-26.11	161.22	180	Haq et al. (1980)
DSDP214	-11.34	88.72	128	Haq et al. (1980)
DSDP231	-11.89	48.25	384.1	Feakins et al. (2013), Fisher et al. (1974)
DSDP235	3.23	52.68	273	Uno et al. (2016), Party (1974)
DSDP238	-11.15	70.53	188	Haq et al. (1980)
DSDP289	-0.50	158.51	224	Woodruff et al. (1981)
DSDP310	36.85	176.90	68	Keigwin (1979)
DSDP357	-30.00	-35.56	24.135	Cramer et al. (2009)
DSDP360	-35.85	18.10	139.5	Wright et al. (1992)
DSDP525	-29.07	2.99	98.6	Shackleton et al. (1984)
DSDP527	-28.04	1.76	104.5	Shackleton et al. (1984)
DSDP552	56.04	-23.233	127	Cramer et al. (2009)
DSDP659	18	-21	180.24	Polissar et al. (2019). Ruddiman et al. (n.d.)
ODP0846	-3.09	-90.82	251	Diester-Haass et al. (2006)*
ODP0667	4.57	-21.91	107	Curry and Miller (1989)
DSDP959	3.5	-2.6	107.35	Polissar et al. (2019), Backman et al. (2012)
ODP1088	-41.14	13.56	71	Billups (2002), Hodell et al. (2002)
U1457	17.17	67.93	645	This study, Feakins et al. (this volume)
Kaulial Kas	333.34	72.70		Tauxe and Opdyke (1982) Quade et al. (1995)
Rhotas:				
Dhabwala Kas	32.95	73.58		Behrensmeyer et al. (2007) Opdyke et al. (1979)
Basawa Kas	32.95	73.579		Behrensmeyer et al. (2007)
Jalalpur	32.75	73.42		Quade and Cerling (1995) Johnson et al. (1982)
Jawalamukhi	31.8	76.39		Vögeli et al. (2017) Meigs et al. (1995)
Haripur Kolar	30.46	77.39		Vögeli et al. (2017) Sangode et al. (1996)
Surai Khola	27.82	82.79		Rösler et al. (1997); Neupane et al. (2019), Ojha et al. (2009); Quade et al. (1995)
Bakiya Khola	27.17	85.173		Quade et al. (1995) Harrison et al. (1993)
PCQ	-26.6	-66.9		Latorre et al. (1997) Butler et al. (1984)

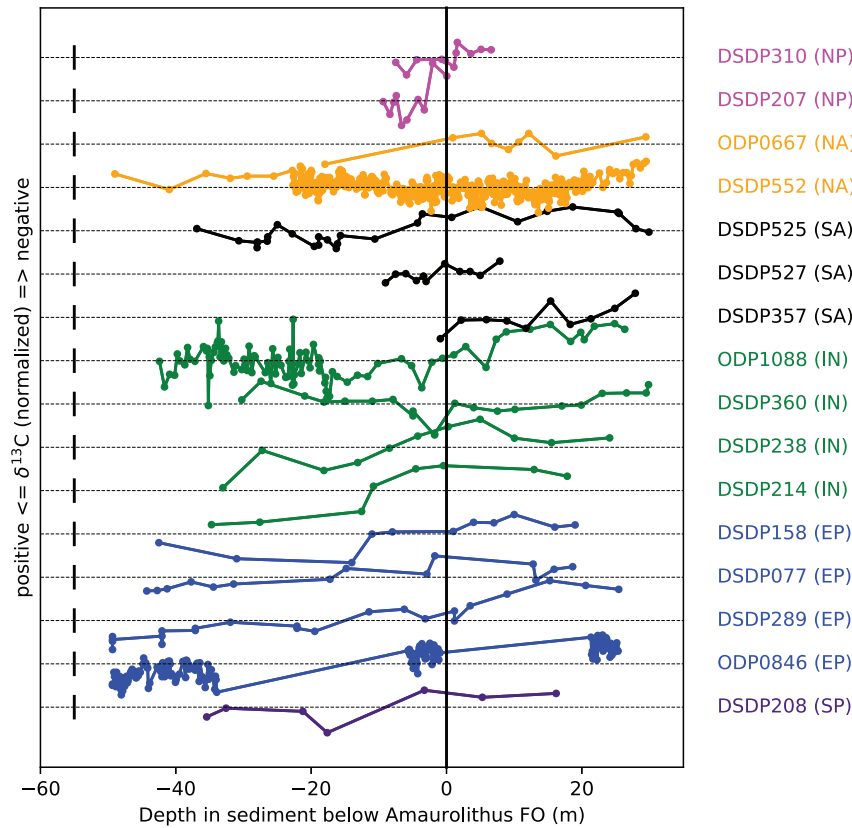


Figure 5. a) Carbon isotopic shift in marine sediment cores (see Table 1 for locations and references). Values for $\delta^{13}\text{C}$ (normalized) are $\delta^{13}\text{C}$ - the mean value, plotted with more negative values up. Black bars to the left are $\pm 0.5\text{‰}$. X axis shows depth relative to the FO of *Ammonium spp.*, which occurs within Chron C3Br, now estimated to be 7.3 Ma (see text). Colors reflect regions where NP: North Pacific (magenta), NA: North Atlantic (orange), SA: South Atlantic (black), IN: Indian Ocean (green), EP: East Pacific (blue), SP: South Pacific (indigo). DSDP/ODP Site same as in Figure 4.

164 a change in vegetation from what is known as the “C₃ photosynthetic pathway” plants
 165 using the C₃ photosynthetic pathway to grasses using the C₄ pathway (see e.g., Tiple
 166 and Pagani (2007)). Quade et al. (1989) tentatively suggested that the shift could be re-
 167 lated to an intensification of the Asian monsoon. Later, Cerling et al. (1993) reinterpreted
 168 the shift as resulting from a drop in atmospheric pCO₂ which would favor C₄ vegeta-
 169 tion.

170 The Siwalik isotopic data were directly tied to magnetostratigraphic sections, con-
 171 straining the shift to be within “Chron 6” which was renamed C3Ar and C3B. The ge-
 172 omagnetic reversal time scale (GRTS) used by Quade and Cerling (1995) was that of Berggren
 173 et al. (1985), hence their estimated age for the transition of between about 7.4 Ma and
 174 6 Ma.

175 The approach of tying carbon isotopic data from continental sections to the GRTS
 176 has since been replicated elsewhere in Pakistan, India and Nepal as well as in Africa and
 177 South America. Latorre et al. (1997) and Singh et al. (2011) compiled data from con-
 178 tinental sections, some of which were magnetostratigraphically calibrated. Their com-
 179 pilations suggested that the shift ranged in age from 4 Ma in North America to as early

180 as 8 Ma in East Africa, with the Asian data from the Siwaliks of India, Pakistan and Nepal
 181 ranging in age from 6 to 7.4 Ma. However, the calibration of the GRTS itself has evolved
 182 significantly over the nearly four decades of research on the topic. Moreover, the differ-
 183 ent carbon isotopic records have in some cases an ambiguous relationship to magnetostrati-
 184 graphic calibration (e.g, the East African and North American data) making temporal
 185 inferences much more difficult.

186 What is of critical importance in assessing potential causes and effects of the C₃-
 187 C₄ transition is the relative timing of events on land (the expansion of C₄ grasses) com-
 188 pared to the marine records of carbon isotopes and to proxies for atmospheric pCO₂. These
 189 three different types of records have been dated using a variety of methods and the time
 190 scales have changed considerably over the years resulting in uncertainties of millions of
 191 years in the exact timing of the disparate records. In this paper we attempt to re-calibrate
 192 the different records in terms of a consistent time scale, which will enable us to assess
 193 the relative timing of the C₄ expansion, climatic drivers and carbon cycle responses. To
 194 do this, we first find those continental records that have a reasonable tie to magnetostrati-
 195 graphic companion records and recalibrate them to a single time scale (here, the GTS12
 196 of Gradstein et al. (2012)). In Section 3.1 we begin with a recalibration of the data from
 197 Pakistan, India and Nepal. In Section 3.2 we discuss the constraints from South Amer-
 198 ica. Unfortunately, there are no results from North American Miocene with both soil car-
 199 bonate isotopes and published magnetostratigraphic data and the magnetostratigraphies
 200 from Africa are similarly ambiguous in their relationship to the isotopic records. In Sec-
 201 tion 3.3, we turn to a new marine record of carbon isotopes from the Indus Fan (IODP
 202 Site U1457) that has both the biostratigraphic (including the FO of *Amaurolithus*) and
 203 magnetostratigraphic constraints. We then consider data from leaf waxes found by drilling
 204 on the African continental margin that also are tied the FO of *Amaurolithus* and the last
 205 occurrence of *Discoaster hamatus* (which has an age of ~10.5 Ma.) Finally we discuss
 206 the implications of our newly calibrated isotopic shifts compared to atmospheric pCO₂
 207 proxies and consider possible feed-backs in the system.

208 **3 Revisiting the timing of the C₃-C₄ transition in continental settings**

209 **3.1 Pakistan, India, Nepal**

210 Quade et al. (1989) and Quade and Cerling (1995) reported carbon isotopic data
 211 from paleosol carbonate nodules from many sections from Pakistan in order to determine
 212 the age of the C₃-C₄ ecological transition: the Chinji-Nagri section of Johnson et al. (1985),
 213 the Kaulial Kas section of Tauxe and Opdyke (1982), the Mirpur section of Opdyke et
 214 al. (1979), the Jalalpur section of Johnson et al. (1982), the Pabbi Hills section of Opdyke
 215 et al. (1979) and Gabhir Kas or Johnson et al. (1982). These were all converted to ages
 216 using the GRTS of Berggren et al. (1985). A section studied initially by Quade et al. (1989)
 217 was resampled for paleomagnetic analysis in order to provide a tighter age constraint and
 218 the updated section was published by Behrensmeyer et al. (2007). Of the Pakistani sec-
 219 tions, only two span the transition in a single (in some cases composite) section: the Rho-
 220 tas (R) (composite) section of Behrensmeyer et al. (2007) (a combination of Dhabawal
 221 Kas and Basawa Kas sections with magnetostratigraphy from Opdyke et al. (1979) and
 222 Behrensmeyer et al. (2007)), and Jalalpur (JP) (Quade & Cerling, 1995) with magne-
 223 tostratigraphy from Johnson et al. (1982). Kaulial Kas (KK) (Quade et al., 1995), with
 224 magnetostratigraphy from Tauxe and Opdyke (1982) has the onset of the transition and
 225 is also included in the present study.

226 Vögeli et al. (2017) compiled records from a variety of sections in NW India includ-
 227 ing the Jawalamukhti, Haripur Kolar, Jogindernagar, Kaming, Jammu Hills, the Parmandal-
 228 Utterbeni and Kangra sections. The magnetostratigraphy for Jogindernagar was based
 229 on unpublished data of Maithani and Burbank and the original data could not be located
 230 for the present study. The Kaming section has an excellent magnetostratigraphic con-

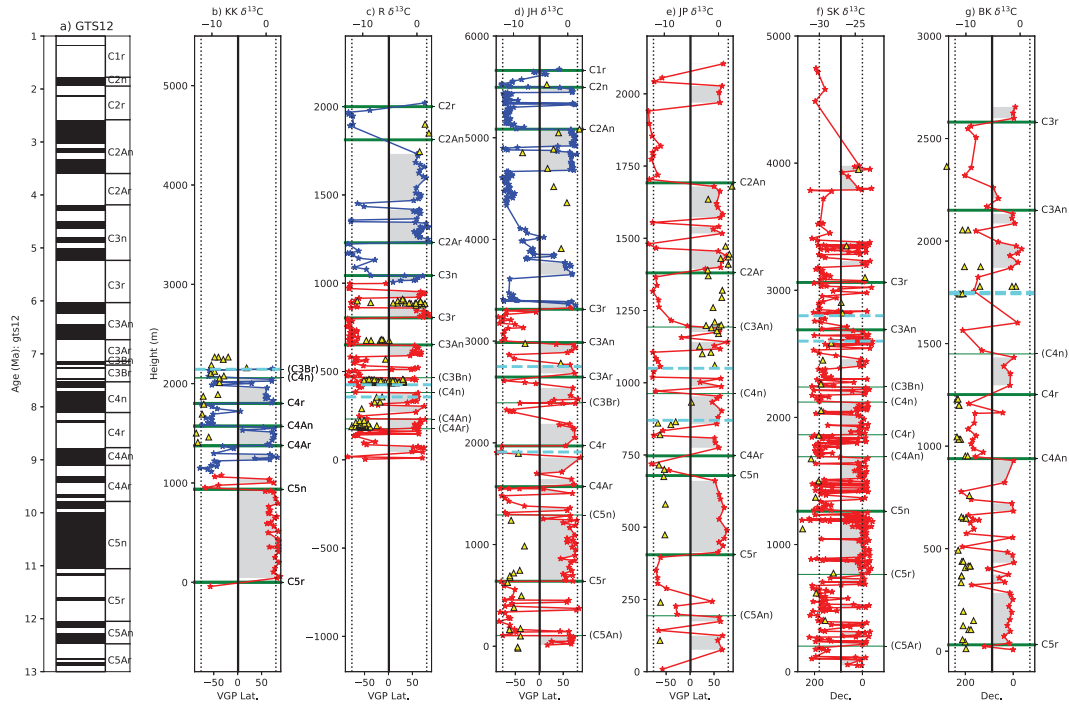


Figure 6. Magnetostratigraphic constraints for carbon isotopic data on the Indian sub-continent. a) GTS12 time scale of Gradstein et al. (2012) with the current Chron nomenclature. The magnetostratigraphy for each section is shown as virtual geomagnetic pole latitudes (VGP Lat.) or declination (Dec.) as a function of stratigraphic height in meters. Chrons are identified as in the original papers with heavy green horizontal lines, except for those in thin lines which are re-interpreted here (also placed in parenthesis). Isotopic data for each section are plotted as yellow triangles. The expected values for $\delta^{13}C$ (VPDB) for pure C_3 and C_4 biomass (-12 and 1.8‰ respectively from Quade (2014)) are plotted as vertical dotted lines in all sections except for SK which uses the bounds appropriate for plant wax n -alkanes (-30‰ is the upper limit of the C_3 plant waxes and -24‰ is the lower limit of C_4 for plant waxes). The stratigraphic bounds of the isotopic transitions are shown as cyan horizontal dashed lines. The sections are b) Kaulial Kas (KK), c) Rhotas composite (R), d) Jawalamukhi/Haripur (JH), e) Jalalpur (JP), f) Surai Khola (SK), g) Bakiya Khola (BK). See Table 1 for locations and references. More detailed views of these sections are shown in Figures S1-S7 in the Supplemental Information.

231 text (Chirouze et al., 2012) but the isotopes show no transition. The Jammu section only
 232 has data through the Pliocene and the older data come from 50 miles away at Nurpur
 233 with no published relationship to the magnetostratigraphy at Jammu. The data for Parmandal-
 234 Utterbeni (magnetostratigraphy from Rao (1993)) only goes back through the Pliocene
 235 and do not record the shift. The Kangra section (data in Sanyal et al. (2004)) come from
 236 two different sections with no clear correlation to each other. Ghosh et al. (2017) pub-
 237 lished isotopic data from several sections in India (Naladkhad, Ranital, Jabbarkhad and
 238 Haripur Khol, or Haripur Kolar here). The magnetostratigraphic context for the Nal-
 239 adkhad section is from Brozovic and Burbank (2000) and the isotopic sampling termi-
 240 nated at about 1600 m, or C4An. The original magnetostratigraphic data from Rani-
 241 tal are not in the reference cited (Sanyal et al., 2004) and the isotopic data show no change
 242 from a C_3 dominated ecosystem throughout the section. They also report data from the
 243 upper part of Jabbarkhad (with magnetostratigraphy from Rao (1993)) and Haripur (with

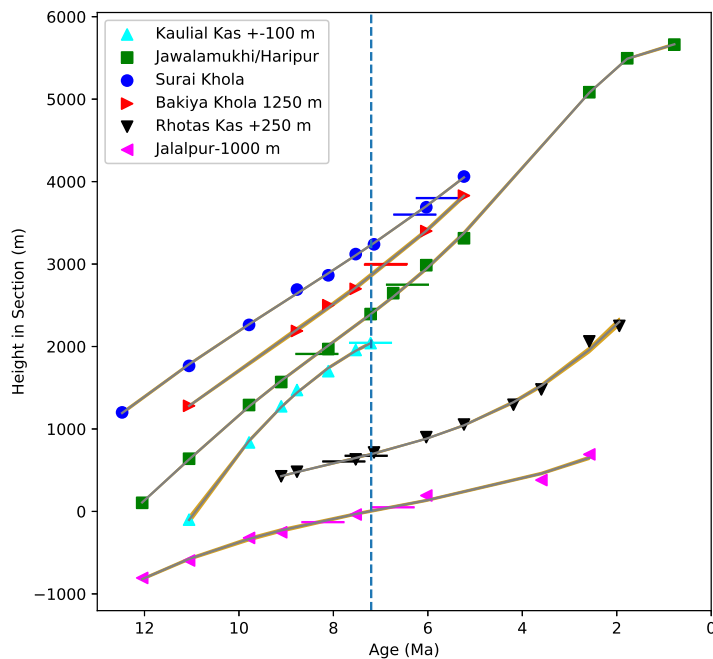


Figure 7. Plots of stratigraphic heights of magnetostratigraphic tie points (Chron boundaries) for each section from Figure 6. Stratigraphic bounds on the isotopic shifts for each section (cyan horizontal lines in Figure 6) are shown as short horizontal bars for each section. A nominal age of 7.2 Ma is shown as a dashed vertical line, which passes through all the bounds except for those in the two Nepali sections (Bakiya and Surai Khola), whose bounds are younger. Reference heights for the different sections have been adjusted by various offsets to allow inter-section comparison. Kaulial Kas was shifted down by 100 m; Rhotas was shifted up by 250 meters; Bakiya Khola was shifted up by 1250 meters; Jalalpur was shifted down by 1000 meters. Polynomial fits are shown as yellow lines which are 1000 estimates using a Monte Carlo resampling scheme.

244 magnetostratigraphy from Sangode et al. (1996) and Sanyal et al. (2004)). The transition
 245 is not contained in any one of these sections. Therefore we focus on the Jawalamukhi
 246 and Haripur Kolar (JH) sections for the present study. These relied on magnetostrati-
 247 graphic control from Meigs et al. (1995) and Sangode et al. (1996) respectively.

248 Isotopic data from Nepal were published by Quade et al. (1995) who investigated
 249 soil carbonates from the Surai Khola (SK) and Bakiya Khola (BK) sections in Nepal.
 250 More recently, Neupane et al. (2019) published new compound-specific data from leaf
 251 wax *n*-alkanes for Surai Khola. The Surai Khola data from Quade et al. (1995) and Neupane
 252 et al. (2019) have magnetostratigraphic constraints from Ojha et al. (2009) and Rösler
 253 et al. (1997) respectively and the Bakiya Khola isotopic data from Quade et al. (1995)
 254 have magnetostratigraphic data from Harrison et al. (1993). Quade et al. (1995) also pre-
 255 sented data from Muksar Khola and Neupane et al. (2019) presented data from the Kar-
 256 nali River, but these sections do not show a shift in the carbon isotopes and in the case
 257 of Muksar Khola, they were not derived from pedogenic carbonate, the focus of the cur-
 258 rent study. Quade et al. (1995) also showed data from the Katari Khola section, but these

259 were only plotted versus stratigraphic height and we were unable to locate the original
260 magnetostratigraphic data.

261 Most of the studies concerned with the C₃-C₄ transition in Asia plotted the iso-
262 topic data in terms of age and the exact relationship to the supporting magnetostrati-
263 graphic age control can be obscure. Therefore in order to recalibrate the age for the iso-
264 topic data using a consistent time scale, we digitized the magnetostratigraphic and iso-
265 topic data, converting both to a common stratigraphic height scale in order to obtain
266 age-height relationships using the GTS12 time scale of Gradstein et al. (2012). C₃ plants
267 have a range of carbon isotopic values from -38 to -18 ‰ (mean $-26.7 \pm 2.3\text{‰}$) and C₄
268 plants have values of -16 to -9 ‰ (mean $-12.5 \pm 1.1 \text{‰}$; Cerling et al. (1997)) with cor-
269 responding boundaries in plant waxes (more depleted) and soil carbonates (more enriched).
270 These can be used to place bounds on the stratigraphic position of the change in isotopic
271 values, with the caveat that dry, open C₃ woodland can be indistinguishable from C₃
272 forest with some C₄ understory, thus there is always ambiguity in the interpretation of
273 the first appearance of C₄ as appreciated in the discussion by Fox and Koch (2003), but
274 often missed in presentations of C₄ ‰. Here the case is simplified as we are not looking
275 to detect first appearance of C₄ or low proportions of C₄ in ecosystems, but rather fo-
276 cus on the shift to C₄ dominance and thus seek large positive isotope shifts that are un-
277 ambiguous.

278 Our reconstructed magnetostratigraphic and isotopic data are shown in Figure 6.
279 Using the information in the figure, bounds on the stratigraphic position of the isotopic
280 shifts from C₃ dominated ecosystems to those dominated by C₄ vegetation can be con-
281 verted to ages from GTS12 in a consistent manner.

282 The logic for calculation of an age model for magnetostratigraphic sections relies
283 on several key assumptions:

- 284 1. We must assume that all magnetic directions (from which we determine polarity)
285 are reliable.
- 286 2. In order to correlate a given section to the polarity time scale we must assume a
287 quasi-linear or slowly varying sedimentation rate.
- 288 3. Sections must be sampled at a sufficient density to insure that all (or at least most)
289 of the polarity intervals are represented.
- 290 4. To transfer ages from the magnetostratigraphic correlations to the isotopic data,
291 the isotopic sampling must be done in close coordination with the magnetostrati-
292 graphic sampling, so that the stratigraphic section heights are the same.

293 In Kaulial Kas, the magnetic stratigraphy is densely sampled. Multiple samples
294 per site allowed rejection of highly scattered (random) directions and the age model is
295 fairly robust. The isotopic sampling was done on the same section as the magnetostrati-
296 graphy. Therefore all four conditions are met.

297 At Rhotas, the magnetic stratigraphy was sampled at sufficient density and was
298 based on a reasonable laboratory protocol (step wise thermal demagnetization of mul-
299 tiple specimens per site) to insure that polarity identifications are robust. The isotopic
300 sampling was done along side the magnetic stratigraphy so there is no ambiguity about
301 the relationship of the two data sets.

302 In the NW Indian composite, the magnetic stratigraphy in the lower section (Jawala-
303 mukhi of Meigs et al. (1995)) is based on an undersampled section and the lab proce-
304 dures followed the early protocol whereby extremely scattered within site directions were
305 used, leading to the possibility of incorrect polarity assignments. The upper section at
306 Haripur (Sangode et al., 1996) used a more sophisticated approach and reported within
307 site statistics. While the lower part of the section had poor within site reproducibility
308 in general, the upper part (relied upon here for the age model) can be considered “re-

309 liable". The isotopes of Vögeli et al. (2017) were sampled much later than the magne-
 310 tostratigraphic sample and plotted against age. We combined the magnetostratigraphic
 311 data with the isotopic data by using the age model of Vögeli et al. (2017) to convert their
 312 ages back to stratigraphic height. Then, by pairing the inferred heights with the heights
 313 from the magnetostratigraphic studies, we attempted to recalibrate the ages for isotopic
 314 data. However, there is an added uncertainty in this process and condition 4 above was
 315 not met.

316 In the Jalalpur section, the original magnetostratigraphy of Johnson et al. (1982)
 317 was severely undersampled so condition 3 above was not met. Also, a very high degree
 318 of scatter ($k > 10$) was allowed and many sites that were based on random directions
 319 were deemed acceptable so condition 1 was not met. However, the isotopes were taken
 320 in coordination with the magnetic sampling sites so condition 4 was met.

321 There are several different data sets from Surai Khola. The most recent is the iso-
 322 topic data set of Neupane et al. (2019). This was tied to the magnetostratigraphy of (Rösler
 323 et al., 1997) (which itself was updated from (Appel et al., 1991)). The section was densely
 324 sampled, but only one specimen per horizon (site) was measured so there are no within
 325 site statistics on which to assess condition 1. In the Kaulial section it was found that some
 326 40% of sites had random within site directions and could be eliminated by using within
 327 site statistics, but that is not possible in the SK section of Rösler et al. (1997). However,
 328 stratigraphic height information relating the isotopic data to the SK section was made
 329 available by the authors of Neupane et al. (2019), so condition 4 was met.

330 At Bakiya Khola, the magnetostratigraphy was apparently based on single spec-
 331 imens per site so condition 1 was not met. Furthermore, the section is undersampled so
 332 condition 3 was also not met. However, the isotopic samples were taken in coordination
 333 with the magnetic sampling so condition 4 was met.

334 We plot stratigraphic height against the ages inferred from the correlation to GTS12
 335 in Figure 7. Given the above mentioned caveats, we estimate sediment accumulation rates
 336 for each section by calculating a 3^{rd} (or in the case of the NW Indian composite a 5^{th})
 337 order polynomial. To estimate the uncertainties for each curve we adopt a Monte Carlo
 338 resampling scheme whereby the depths of the tie points are drawn from uniform distri-
 339 butions between the upper and lower bounds for each calculate a new curve for each re-
 340 sampled set of tie points, repeating the process 1000 times. We plot each of the Monte
 341 Carlo curves in yellow on the figure. The stratigraphic bounds for the isotopic shift from
 342 each section are shown as horizontal lines.

343 An alternative approach to calculating an age model for these sections would be
 344 to assume all four of the conditions above are met and interpolate between magnetostrati-
 345 graphic tie points using a straight-line calculated between the bounds. This assumes that
 346 sediment accumulation is essentially linear between each bounding tie point and can re-
 347 sult in abrupt changes in rate between adjacent segments. We recalculated all ages us-
 348 ing this approach and found that the difference between the smoothly varying approach
 349 (polynomial assumption) and the piece-wise linear approach was $\pm \sim 100,000$ years and
 350 much better than that near the interval of interest here (a few 10s of thousands of years).
 351 Given the additional uncertainties and assumptions associated with the second approach,
 352 We adopt the first, smoothly varying, approach in the following.

353 The first question we wish to address is whether there is a single age or age range
 354 for the isotopic transition that is consistent with all of the available data from the In-
 355 dian sub-continent. We replot the data from Figures 6 and 7 as isotopes versus inferred
 356 age in Figure 8. In Figure 8b we plot the data from India and Pakistani sections (Kau-
 357 lial Kas, Rhotas, Jawalamukhti/Haripur and Jalalpur) against the revised ages. The tran-
 358 sition began perhaps as early as C4n (~ 7.8 Ma) as suggested by a single point in the Jala-
 359 pur section, but certainly by C3Bn (~ 7.2 Ma) in the Kaulial and Rhotas sections.

360 In Figure 8c we plot the data from Nepal (Surai and Bakiya Khola). The transi-
 361 tion recorded in Nepal started later than in Pakistan and India, perhaps as young as 6
 362 Ma as recorded in Surai Khola. This diachroneity within a single subcontinent could ar-
 363 gue against $p\text{CO}_2$ as the sole or even dominant driver of the process, although tipping
 364 points may differ across climatic regimes (Higgins & Scheiter, 2012). For example, the
 365 western side of the subcontinent (Pakistan) is considerably drier and thus may pass cli-
 366 matic thresholds that promote C_4 grasslands to replace forests earlier than the eastern
 367 side (Nepal). This observation is also supported by the modeling efforts of (Zhou et al.,
 368 2018) (Figure 1) which show considerable regional variability within a constant atmo-
 369 spheric $p\text{CO}_2$ regime.

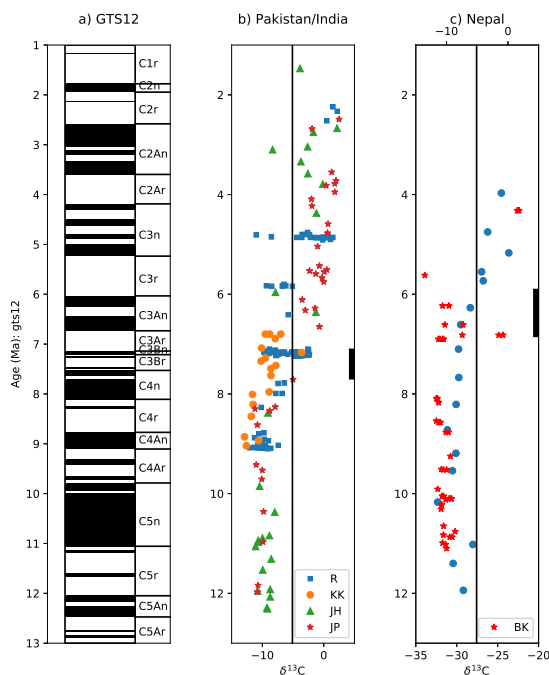


Figure 8. Isotopic data from Figure 6 plotted against age from Figure 7. Section abbreviations same as in Figure 6. See Table 1 for references. a) Time scale of (Gradstein et al., 2012). b) data from Pakistan and India. c) data from Nepal. Surai Khola has the latest onset of all sections. Black bars on side of b-c) are range for onset of C_4 dominated ecosystems in each region.

370 3.2 Other continental sections

371 To place the data from the Indian subcontinent into a global context, we look first
 372 to other records with good magnetostratigraphic control. While there are studies from
 373 Chinese Loess Plateau (e.g., Dong et al. (2018)), these have not yet been placed in a mag-
 374 netostratigraphic context. In any case, the study focused on the Mid-Miocene Climatic
 375 Optimum (much earlier than the focus of this paper) and demonstrated that there may
 376 have been a slight increase in the percentage of C_4 plants at around 15 Ma as two data
 377 points are close to a 50% C_4 fraction.

378 Ségalen et al. (2007) cited data from Africa but those do not show the clear tran-
 379 sition to C₄ values and the data are not clearly tied to the available magnetostratigraphic
 380 sections (which are in an unpublished master's thesis and do not stand up to scrutiny).
 381 Uno et al. (2011) published isotopic data from mammal teeth but the magnetostratigraphic
 382 data (as opposed to the interpretations) were apparently also never published. Similarly,
 383 Morgan et al. (1994) present data from mammal teeth from the Tugen Hills section (near
 384 the sections studied by Tauxe et al. (1985), but include no information about the rela-
 385 tionship between their fossil localities and any magnetostratigraphic constraints.

386 North America appears to have no magnetostratigraphically calibrated isotopic data
 387 either. However, Tipple and Pagani (2010) published isotopic data from DSDP Site 94
 388 from the Gulf of Mexico. Unfortunately, the latter section lacks the FO of *A. primus* with
 389 which to tie the record to the present study. Moreover, it has only weak support for the
 390 onset of C₄ vegetation in the Miocene. Similarly, Chen et al. (2015) sampled sections in
 391 Montana relying on an age of 10.42 Ma for an ash bed reported by (Retallack, 2007) who
 392 in turn cited an unpublished field guide. The isotopes (which show a dominance of C₃
 393 grasses throughout the section) are thought to range from ~ 9 to ~ 10.4 Ma. Finally,
 394 Fox and Koch (2003) published a set of soil carbonate isotopic data calibrated with the
 395 North American Land Mammal Ages. This data set suggests that by the Pliocene, the
 396 vegetation in western North America was dominated by C₄ vegetation.

397 The picture in South America, is much better constrained than for North Amer-
 398 ica. Latorre et al. (1997) published isotopic data from soil carbonates from the Puerta
 399 de Corral Quemado (PCQ) section in Argentina which has excellent magnetostratigra-
 400 phy control from Butler et al. (1984), which we show in Figure S8. The increasing abun-
 401 dance of C₄ plants is suggested by $\delta^{13}\text{C}$ values higher than -7.5 ‰ and the transition is
 402 well constrained to be younger than 4 Ma. This result agrees with other sections from
 403 Argentina that unfortunately lack magnetostratigraphic constraints (Kleinert & Strecker,
 404 2001). The results from Argentina are similar to those of Andrae et al. (2018) who found
 405 a Pliocene onset for the C₃-C₄ transition as recorded in leaf wax data found in samples
 406 from ODP Site 763A off NW Australia. Therefore the C₃-C₄ transition in terrestrial se-
 407 quences cannot be globally synchronous, in agreement with predictions by the model-
 408 ing efforts of, for example, Zhou et al. (2018) (see Figure 1).

409 3.3 U1457: A Rosetta stone for carbon isotopes

410 Returning to the paleoceanographic carbon isotopic shift first noted by Keigwin
 411 (1979), the question arises as to how the transition in the Indian subcontinent relates
 412 to the paleoceanographically observed carbon shift. Until now, deep sea records of the
 413 carbon shift have been tied to the first occurrence (FO) of *Amaurolithus spp.* The age of
 414 *Amaurolithus spp.* in GTS12 is stated to be 7.4 Ma. However, it was found in two Ocean
 415 Drilling Program (ODP) holes: 844B and 710B. The FO in 844B was found at 844B-5H-
 416 5; 29 cm which, according to Schneider (1995), is in the middle of C3Br.2r or about 7.3
 417 Ma using the timescale of Gradstein et al. (2012). It was also found in 710B-7H-5,30/7H-
 418 4, 130 or 60.5 mbsf, an interval between C3Ar (y) and C4n (y). Assuming a linear sed-
 419 imentation rate gives an approximate age for this datum of about 7.35 Ma. We adopt
 420 the former age of 7.3 Ma here as it is better constrained. What is required to tie the con-
 421 tinental records to the marine records is a core in which the isotopic shift and the FO
 422 of *Amaurolithus spp.* both occur in a magnetostratigraphic context. IODP Site U1457
 423 provides such an opportunity.

424 The nannofossil, foraminiferal, strontium isotope and paleomagnetic age constraints
 425 for Site U1457 were recently published by Routledge et al. (2019). Here we use their bios-
 426 tratigraphy and a revised interpretation of a few of the paleomagnetic age constraints
 427 (see Table 2 and Figure 9). The lithology of Site U1457 is shown in Figure 9. Routledge
 428 et al. (2019) defined six lithologic units at Site U1457 and used the age model shown in

Datum	Type	Event	Age (Ma)	Max. (m)	Min. (m)	Midpoint (m)
24	CN	T Sphenolithus spp.	3.540	517.45	513.09	515.27
26	CN	B Discoaster tamalis	4.130	539.15	539.65	539.40
28	CN	T Discoaster quinqueramus	5.590	539.65	539.15	539.40
29	PF	T Globoquadrina dehiscens	5.920	526.64	513.09	519.87
30	CN	T Nicklithus amplificus	5.940	610.36	610.05	610.21
32	MR	C3Ar	6.733	624.23	625.42	624.83
33	PF	B Pulleniatina primalis	6.600	615.50	621.36	618.43
35	CN	B Nicklithus amplificus	6.910	628.34	629.53	628.94
36	MR	C3Br.2r	7.285	643.04	644.16	643.60
38	CN	B Amaurolithus spp.	7.300	645.05	644.76	644.91
39	MR	C3Br.2n	7.454	662.92	664.92	663.92
41	MR	C4n.1r	7.642	674.47	675.62	675.05
42	CN	B Discoaster quinqueramus	8.120	832.85	845.62	839.24
43	CN	T Minylitha convallis	8.680	845.62	832.85	839.24
44	CN	T Discoaster bollii	9.210	856.50	845.62	851.06
45	CN	T Catinaster coalitus	9.690	864.64	856.50	860.57
46	MR	C5n	9.790	864.18	866.20	865.19
47	PF	B Neogloboquadrina acostaensis	9.830	885.02	894.30	889.66
48	CN	B Discoaster bellus	10.400	1001.08	1005.11	1003.10
49	CN	B Catinaster coalitus	10.890	1001.08	1005.11	1003.10
50	CN	Absence of Fasciculithus spp.	62.130	1068.63	1067.35	1067.99

Table 2. Age model revised from Routledge et al. (2019). B: bottom (or first occurrence), T: top (or last occurrence). Ages are as in Gradstein et al. (2012) except for *Amaurolithus spp.* (see text). All depths are in composite meters depth (CCSF) as in Routledge et al. (2019).

429 Figure 9b. Based the fact that Unit 4 contains several subunits with more or less car-
430 bonate or turbiditic sandstones (which substantially change the sediment accumulation
431 rates), we now further subdivide Unit 4 into three packages (4a-c, see Figure 9c). The
432 upper package (Unit 4a) is dominated by sandy layers, the middle package (Unit 4b) is
433 dominated by carbonates and clays and the lower unit (Unit 4c) has a higher sand con-
434 tent as in Unit 4a. There are pronounced changes in sediment accumulation rates with
435 inflection points at the unit boundaries. We have modified the age model from Routledge
436 et al. (2019) to take into account the lithological changes and the resulting revised tie
437 points are listed in Table 2 and on Figure 9c.

438 The sediment accumulation rate for Unit 4c was estimated by a linear fit to the two
439 bounding tie points (28 and 30 in Table 2) as there are no additional internal controls.
440 For Unit 4b, we used a 3rd order polynomial as with the Indian subcontinent analysis,
441 with the Monte Carlo estimates shown in yellow. The higher sediment accumulation rate
442 in the lower part of the unit is caused by the higher proportion of sandy layers). Unit
443 4a is dominated by turbidites and we again simply use a linear fit between the two bound-
444 ing tie points (41 and 41). Unit 3 is clay/claystone and the age model is based on a lin-
445 ear fit between tie points 42 and 46. Unit 2 is a mass transport deposit.

446 Using the age model shown in Figure 9c, we plot the magnetostratigraphic and iso-
447 topic data for Unit 4b in Figure 10. The C₃-C₄ transition occurs just above the FO of
448 *Amaurolithus spp* and within a normal interval interpreted here as C3Br.2n or at about
449 7.2 Ma. This is within the age interval for the transition in India and Pakistan estimated
450 in Figure 8.

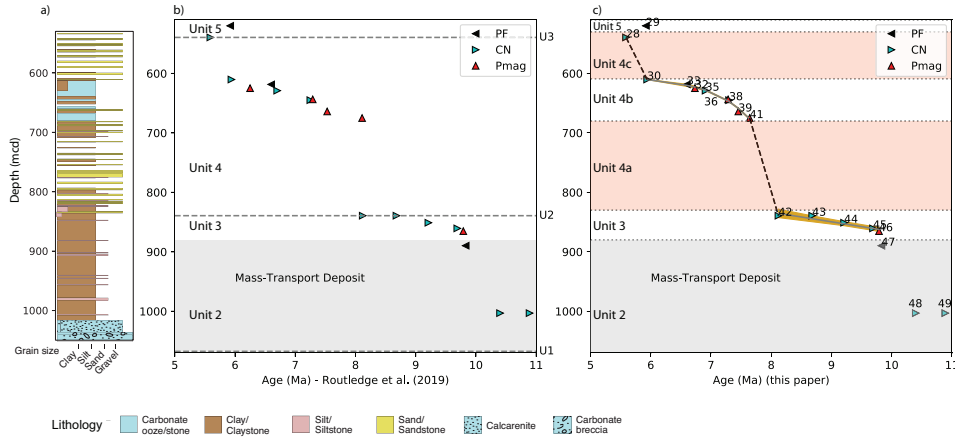


Figure 9. a) Lithostratigraphy for U1457 modified from Pandey et al. (2016) in meters composite depth (CCSF). b) Age model from Routledge et al. (2019). c) Revised age model for this study (see Table 2). PF: planktonic foraminifera, CN: calcareous nannofossils, Pmag: paleomagnetic Chron identifications.

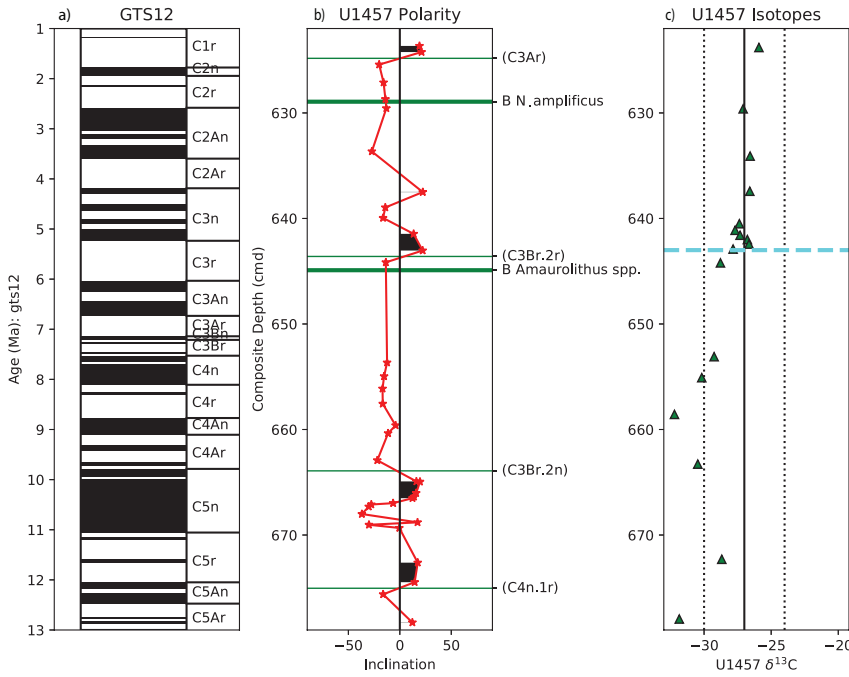


Figure 10. a) Geomagnetic reversal time scale of Gradstein et al. (2012). b) Magnetostratigraphic data for U1457 (Indus Fan) are from Routledge et al. (2019) as re-interpreted in Table 2. The Chron boundary picks minimize changes in sedimentation rate between hiatuses and minimize the discrepancy between the nannofossil identifications. Some Chron boundary picks and were adjusted slightly from the original publication (shown in parentheses). c) Isotopic data are from Feakins et al. (This volume). Magnetostratigraphic and isotopic data are plotted on the composite depth scale. Bounds for upper limit of C₃ and lower limit of C₄ (vertical dotted lines). Horizontal cyan dashed line is the transition between C₃ and C₄ vegetation.

451 The carbon isotopes were measured on leaf waxes whose inputs to the hemipelagic
 452 units are interpreted as being wind-transported based on a lack of associated lignin (both
 453 are present in the fluviably-export carried in turbidic units Feakins et al. (This volume).
 454 The wind-blown waxes are thought to derive from peninsular India based on the prox-
 455 imity of the continent, the dominant easterly winds in October-to-January and modern
 456 evidence from coretops across the Arabian Sea (Dahl et al., 2005).

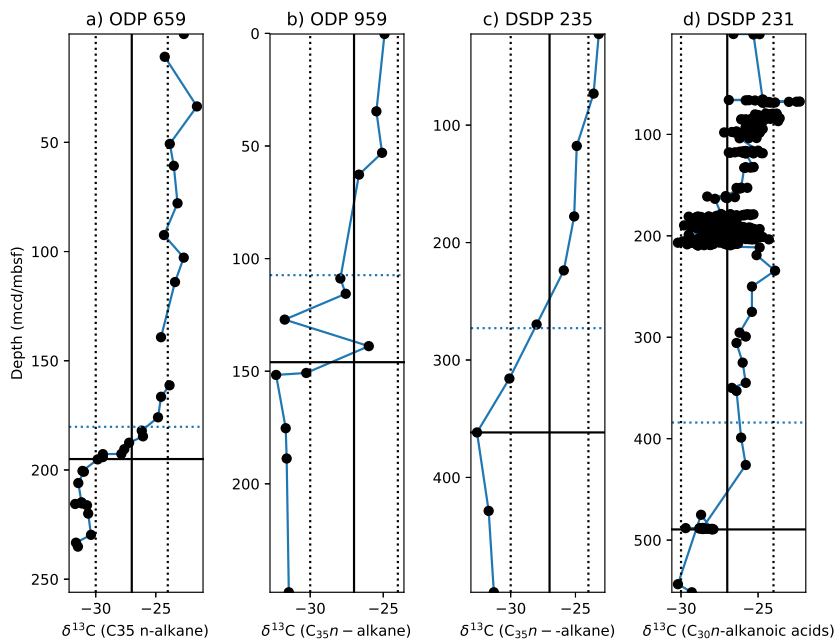


Figure 11. Leaf wax $\delta^{13}\text{C}$ C₃₅ n-alkane (a-c) and d) C₃₀ n-alkanoic acid data from deep sea cores around Africa (see Table 1 and Figure 4). Dotted horizontal lines are the first occurrences of *Amaurolithus* spp. in each core. Solid lines are the last occurrences of *Discoaster hamatus* with an age of 10.5 Ma. a) ODP Site 659; data from Polissar et al. (2019). b) ODP Site 959; data from Polissar et al. (2019). c) DSDP Site 235; data from Uno et al. (2016). d) DSDP Site 231 from Feakins et al. (2013).

3.4 Africa

457
 458 As noted earlier, data from continental sections in Africa are insufficient to con-
 459 strain the C₃-C₄ transition. However, analysis of leaf waxes found in DSDP and ODP
 460 cores, in connection with the FO of *Amaurolithus* spp can provide some clues. We plot
 461 the data from leaf wax $\delta^{13}\text{C}$ C₃₅ n-alkanes from Uno et al. (2016) and Polissar et al. (2019)
 462 and $\delta^{13}\text{C}$ C₃₀ n-alkanoic acids from Feakins et al. (2013) in Figure 11 against depth. We
 463 also show the FO of *Amaurolithus* spp and the last occurrences (LO) of *Discoaster* *hama-*
 464 *tus* with an age estimated at ~10.5 Ma (Gradstein et al., 2012). These data suggest that
 465 the change in vegetation from C₃ dominated ecosystems to those dominated by C₄ oc-
 466 curred between these two tie points, sometime after about 10 Ma as suggested by (Polissar
 467 et al., 2019). These are the earliest ages for the transition that we know of and are con-
 468 sistent with the model of (Zhou et al., 2018) (Figure 1) whereby eastern Africa is one
 469 of the first places predicted to favor C₄ vegetation.

470 4 Discussion

471 From the re-analysis of age constraints for isotopic data from marine and continen-
 472 tal sections it appears that there was at least a regional shift in carbon isotopes in Africa
 473 beginning sometime after 10 Ma, but well before 8 Ma. Data from U1437 (Indus Fan)
 474 suggests an age of 7.2 Ma for peninsular India. In Northern Pakistan and NW India, the
 475 change began perhaps as early as 7.8 Ma (based on a single data point from Jalalpur)
 476 but certainly by about 7.2 Ma. Vegetation in Nepal shifted somewhat later, after 7 Ma,
 477 and data from Argentina and Australia indicate that the shift occurred in the Pliocene.
 478 Moreover, using the first occurrence of *Amaurolithus spp.* as a temporal marker, we can
 479 tie the record from U1457 to other marine records of carbon isotopes (for example, the
 480 leaf wax data of Uno et al. (2016) recovered at DSDP Site 235, of Feakins et al. (2013)
 481 at DSDP Site 231 and of Polissar et al. (2019) from ODP Sites 659 and 959) which show
 482 that the shift in carbon isotopes in Africa likely pre-dated the marine carbon isotope shift
 483 first recognized by Keigwin (1979) and was well before that recorded elsewhere. The ques-
 484 tion remains as to how these ecological changes fit with possible drivers, a topic to which
 485 we now turn.

486 Raymo and Ruddiman (1992) made the case that uplift of the Himalayan region
 487 and the Tibetan Plateau could have resulted in higher chemical weathering which in turn
 488 could have resulted in a drawdown of atmospheric CO₂. While the Himalayas are broadly
 489 believed to have been at high elevations much earlier than the shifts examined here, there
 490 are some signs of uplift in the late Miocene. Harrison and Yin (2004) suggested a ma-
 491 jor phase of uplift at around 9 Ma and Molnar et al. (1993) interpret the onset of nor-
 492 mal faulting in Tibet at around 8 Ma as evidence of an uplift of the plateau. Interest-
 493 ingly, Tremblay et al. (2015) call for decreased erosion in southern Tibet caused by en-
 494 hanced uplift at about 10 Ma, an effect opposite to the mechanism envisioned by Raymo
 495 and Ruddiman.

496 Changes in the carbon budget may also arise from often overlooked adjustments
 497 in the global organic carbon cycle and the case has been made that enhanced organic
 498 carbon burial in the Bengal Fan could outweigh any changes in silicate weathering (Derry
 499 & France-Lanord, 1996). It is also possible that the rise of C₄ grasslands led to a change
 500 in the carbon cycle with attendant climate feedbacks. A large-scale change in ecology
 501 could shift the balance of carbon storage as forests have generally thin soils whereas grass-
 502 lands can build deep soils. Limited evidence from experimental farms in Minnesota, (Fornara
 503 & Tilman, 2008) estimated that C₄ vegetation pulls down 193% more carbon into the
 504 soil than C₃ vegetation. More recently, Spiesman et al. (2018) found that, depending on
 505 the quality of the soil, a higher proportion of C₄ grasses relative to C₃ grasses can in-
 506 deed enhance carbon storage; they attribute this to the higher efficiencies in nitrogen and
 507 water usage by the C₄ photosynthetic pathway.

508 Even if only part of the Indian sub-continent were covered by C₄ grasses, and the
 509 resulting paleosols buried (as they were in the Siwaliks), this could provide a positive
 510 feedback as lower pCO₂ would in turn favor the C₄ grasses. Whatever the mechanism,
 511 the current array of pCO₂ reconstructions do leave open the possibility of a late Miocene
 512 pCO₂ drop that would have enhanced the viability of C₄ plants.

513 4.1 Conclusions

514 In this study we reconsider the timing of the shift to C₄ dominance in ecosystems
 515 around the world. We have been able to realign records where the following conditions
 516 are met:

- 517 1. Marine data include the first appearance of *Amaurolithus spp.*
- 518 2. Magnetostratigraphic records are provided including the depth in section of the
 519 chronostratigraphic markers in order for alignment as chronostratigraphy evolves.

520 We emphasize that making the age model data available alongside new carbon isotope
521 records is particularly valuable for ensuring that updated comparisons can be made as
522 chronologies evolve. Some regions that are missing from the present comparison are on
523 land in North America where magnetostratigraphic age control would greatly improve
524 interpretations.

525 Leaf wax data from offshore of Africa (Feakins et al., 2013; Uno et al., 2016; Polissar
526 et al., 2019) suggest an age of around ~ 10 Ma for beginning of the shift to dominance
527 of C_4 ecosystems. A reconsideration of the magnetostratigraphic constraints for the car-
528 bon isotopic records of pedogenic carbonate from the Siwaliks in Pakistan and India show
529 that vegetation changed from a C_3 photosynthetic pathway to C_4 grasslands perhaps as
530 early as 7.8 Ma and certainly by about 7.2 Ma. The shift in Nepal occurred somewhat
531 later, starting after 7 Ma, and perhaps as late as 6 Ma. This ecological change was also
532 recorded at about 7.2 Ma in leaf waxes recovered from the peninsular India at IODP Site
533 U1457. Finally, magnetostratigraphic data from Argentina argue for a much later shift
534 to C_4 dominance in South America and Australia in the Pliocene.

535 While it was previously hypothesized that a shift in vegetation on the Indian sub-
536 continent could have been caused by uplift in the Himalayas through a change in local
537 climate (enhanced Asian monsoon), Feakins et al. (This volume) show that there was
538 no change in precipitation related isotopes through this interval. The connection to at-
539 mospheric CO_2 as a major driver is therefore of renewed interest for this regional tran-
540 sition, and some new pCO_2 reconstructions do indicate the possibility of a late Miocene
541 drop, but more reconstructions are needed to secure such an interpretation. On land,
542 rapid sediment accumulation rates in the Siwaliks in the Miocene could have led to en-
543 hanced carbon sequestration and the Siwaliks deposition ends abruptly soon after the
544 C_4 transition in many sequences pointing to a change in basin dynamics. We wonder if
545 the vegetation shift itself could be a feedback on the draw-down of atmospheric CO_2 ,
546 perhaps via changing fluvial erosion on a grassy floodplain with enhanced erosion result-
547 ing in increased carbon burial (e.g., France-Lanord and Derry (1997); Derry and France-
548 Lanord (1996)). In soils, the expansion of C_4 vegetation could have played a role in de-
549 creasing atmospheric CO_2 as C_4 grasslands can be more efficient at transferring CO_2 into
550 soil than C_3 tropical forests. Understanding the terrestrial and marine organic carbon feed-
551 backs on the vegetation expansion look to be a promising direction for future enquiry.

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569 tains the data is: <https://earthref.org/MagIC/16737/ec119793-abd4-41aa-8884-28b5fecd0f09>

NOTE TO REVIEWERS: THE REFERENCE LIST UNCAPITALIZES WORDS THAT SHOULD BE CAPITALIZED- THIS IS CAUSED BY A CHANGE IN THE AGU TEMPLATE - I HAVE NOTIFIED AGU OF THIS PROBLEM AND IT WILL BE FIXED PRIOR TO PUBLICATION.

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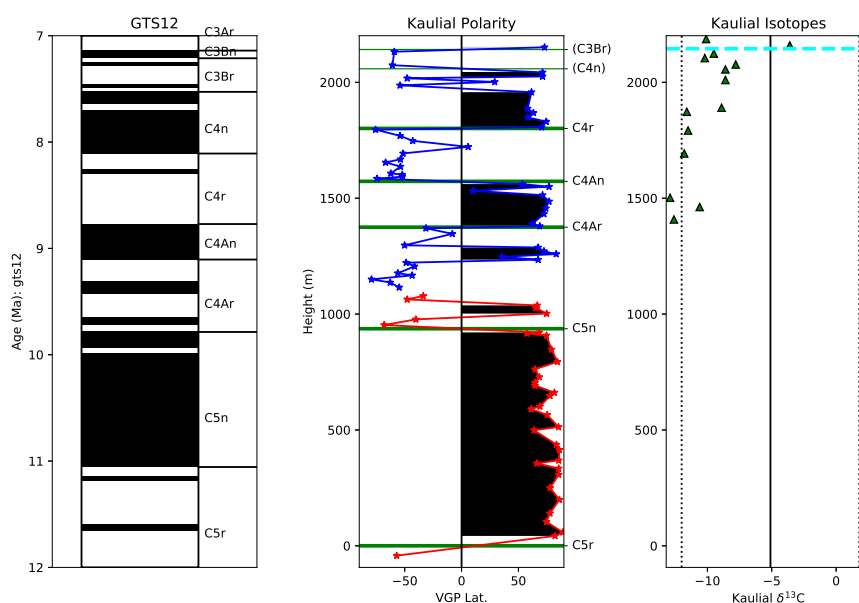
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864 5 Supplemental Information

865 Figure S1

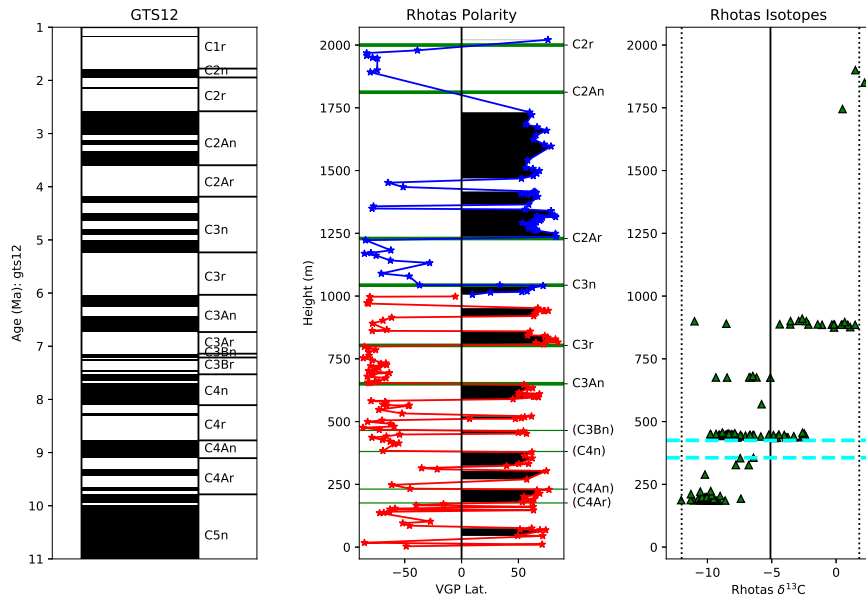


866

867 Details for sections in Figure 6. See Table 1 for data sources. left: Magnetic polarity
868 time scale of Gradstein et al. (2012). middle: Virtual Geomagnetic Pole (VGP)
869 positions for sites in the Kaulial Kas Section plotted against stratigraphic height. Those
870 in parentheses were added or modified in this paper; those without are as in the origi-
871 nal publication. Solid green lines are the Chron boundaries as identified to the right.
872 c) Carbon isotopes from paleosol carbonate nodules. Dotted lines are C_3 and C_4 end-

873 members for soil carbonates. C₃-C₄ transition boundary (boundaries) as dashed cyan
 874 horizontal line(s).

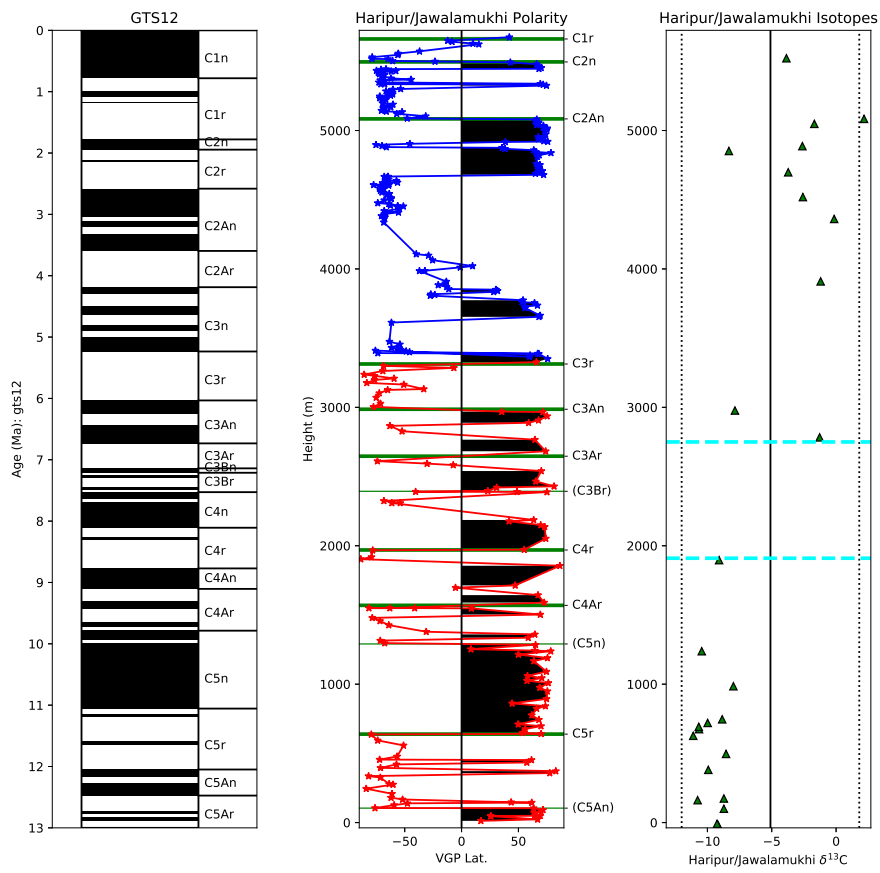
875 Figure S2



876

877 Same as Figure S1 but for the Rhotas Section.

878 Figure S3

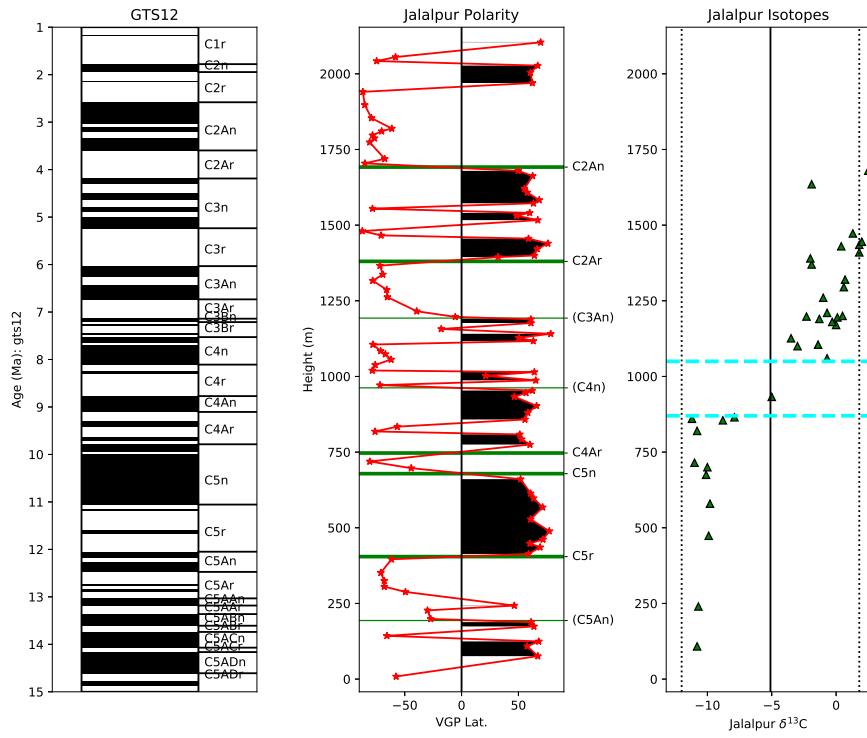


879

880

Same as Figure S1 but for the Jawalamukhi/Haripur Sections.

881 Figure S4

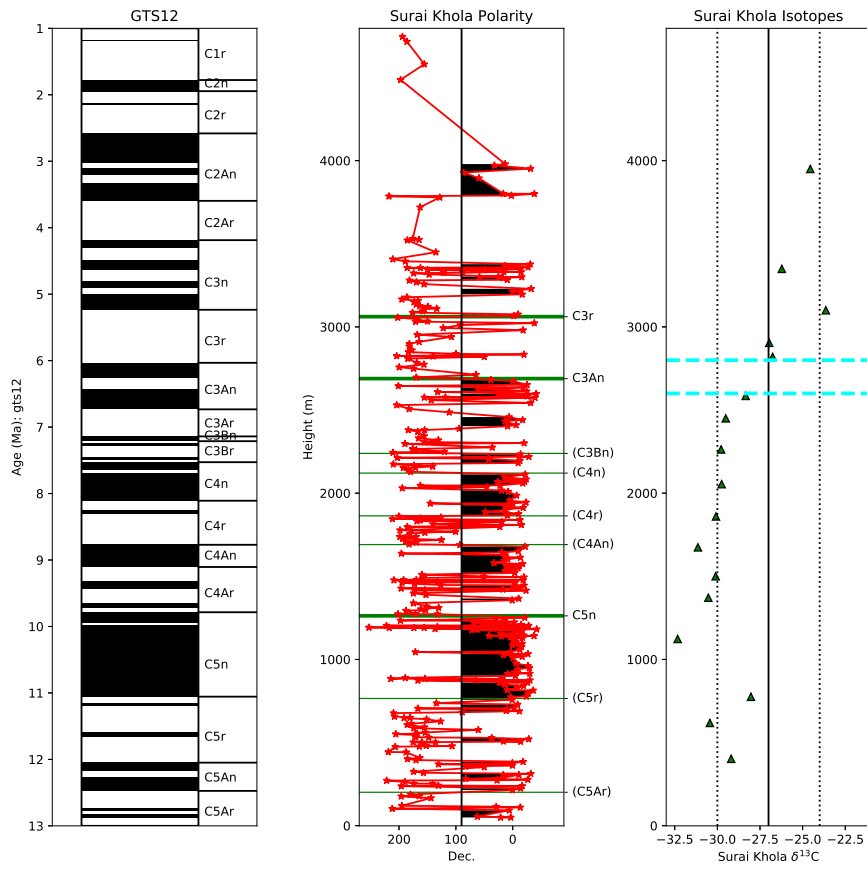


882

883

Same as Figure S1 but for the Jalalpur Section.

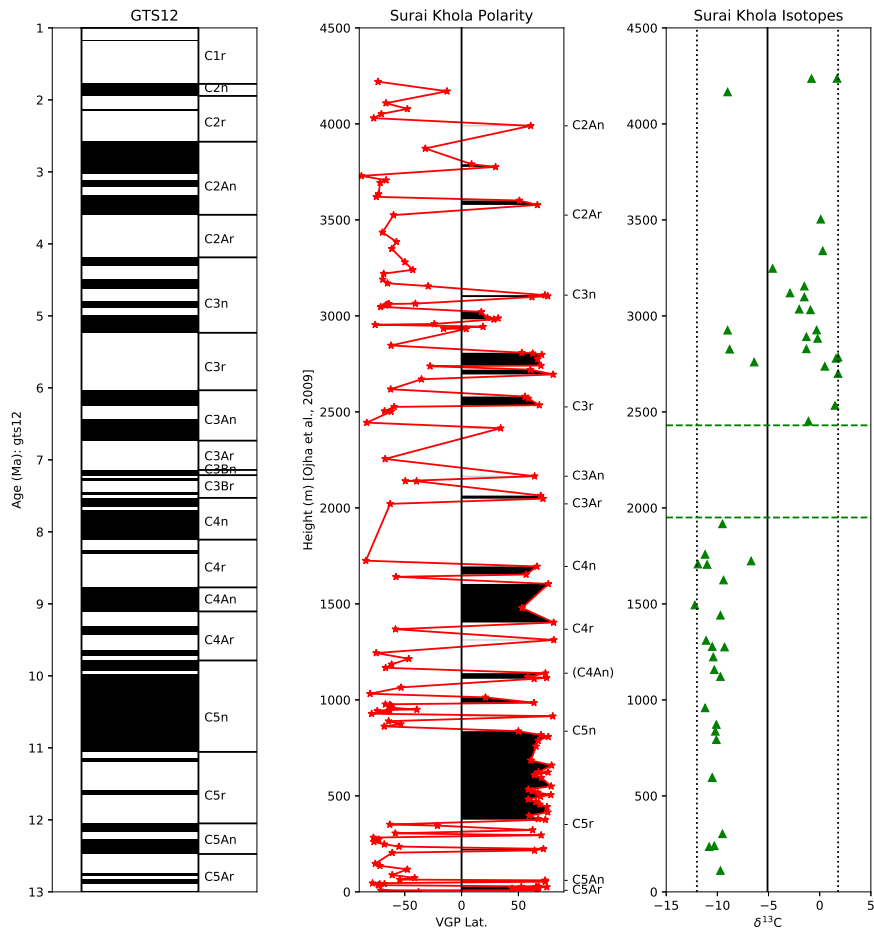
884 Figure S5



885

886 Same as Figure S1 but for the Surai Khola Section. Middle panel is declination (not
 887 VGP). Isotopic data are C₂₇ *n*-alkane data. Dotted lines are the limits of C₃, C₄ for plant
 888 waxes.

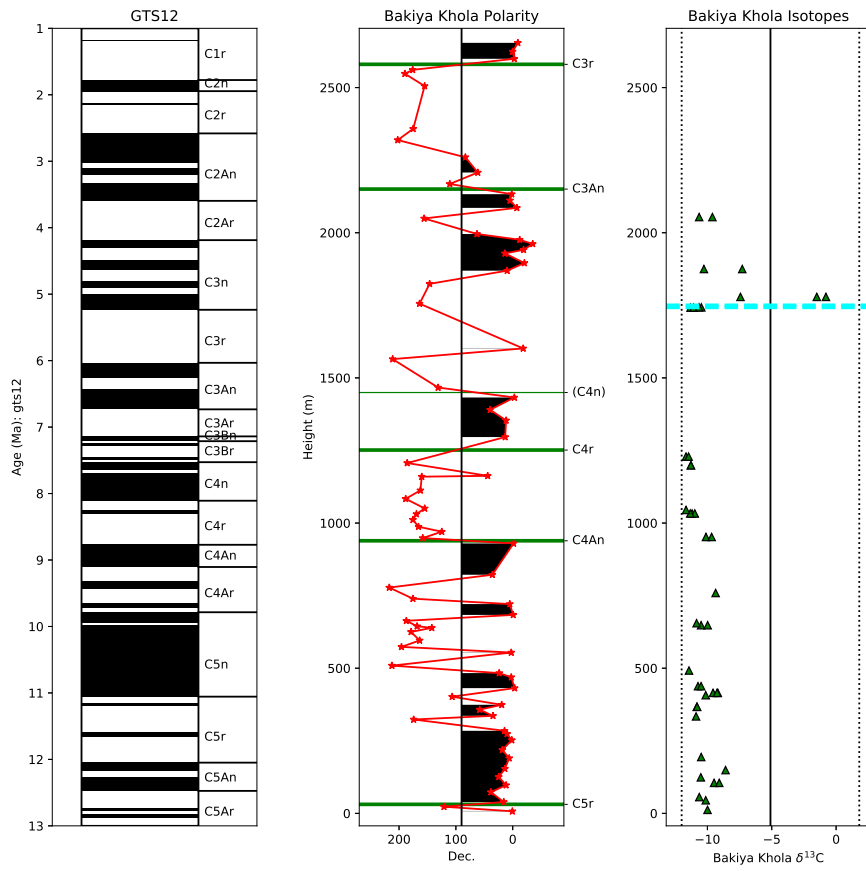
889 Figure S6



890

891 Same as Figure S1 but for the alternate Surai Khola Section of Ojha et al. (2009)
 892 and Quade et al. (1995). Middle panel is declination (not VGP).

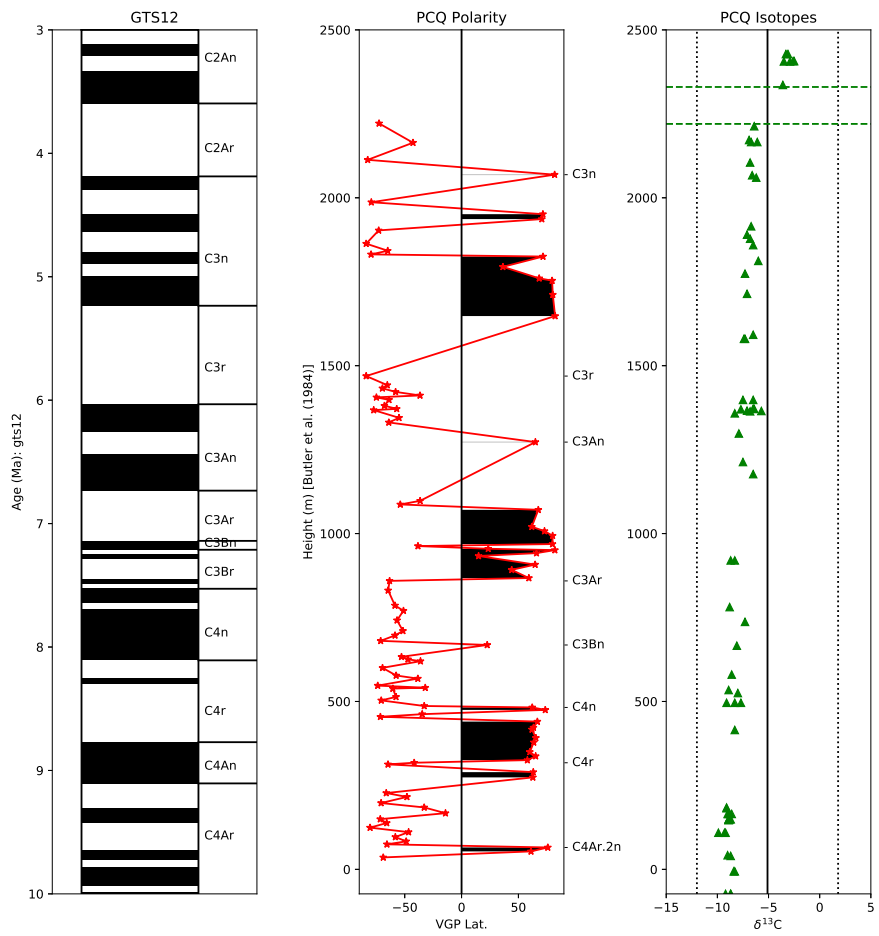
893 Figure S7



894

895 Same as Figure S1 but for the Bakiya Khola Section. Middle panel is declination
 896 (not VGP).

897 Figure S8



898

899

Same as Figure S1 but for the Puerta de Corral Quermado (PCQ) Section.

Figure 1.

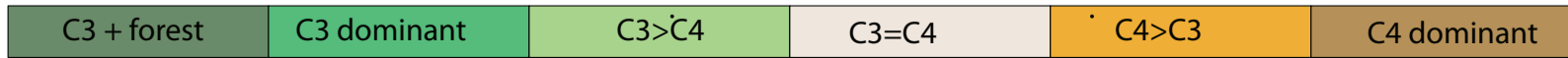
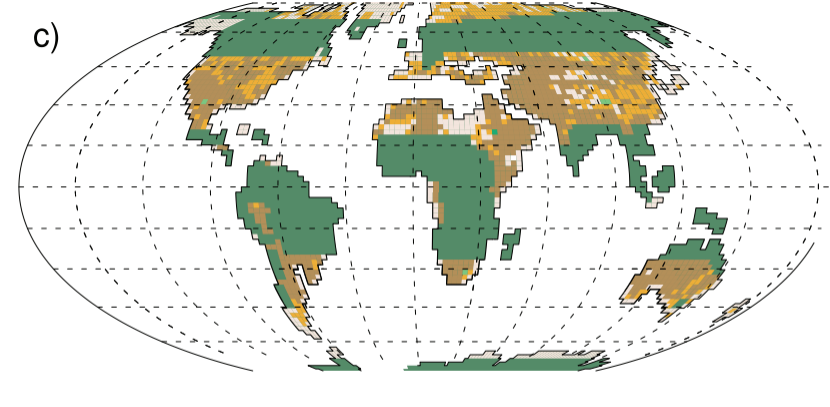
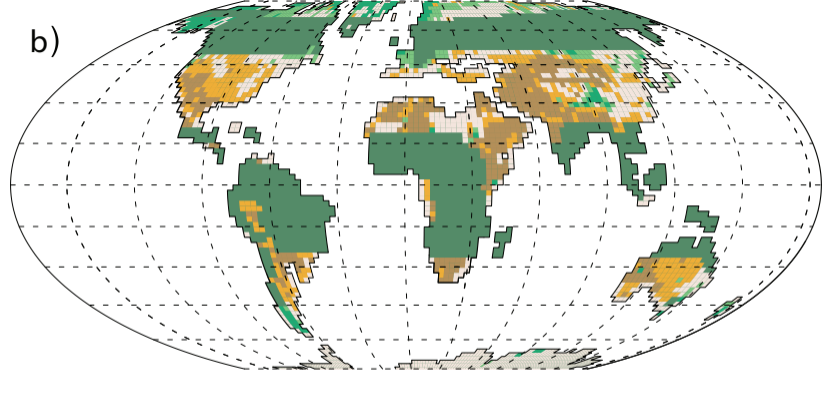
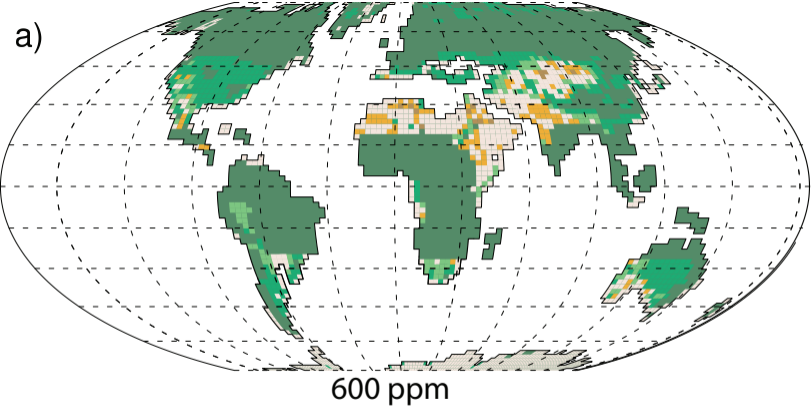


Figure 2.

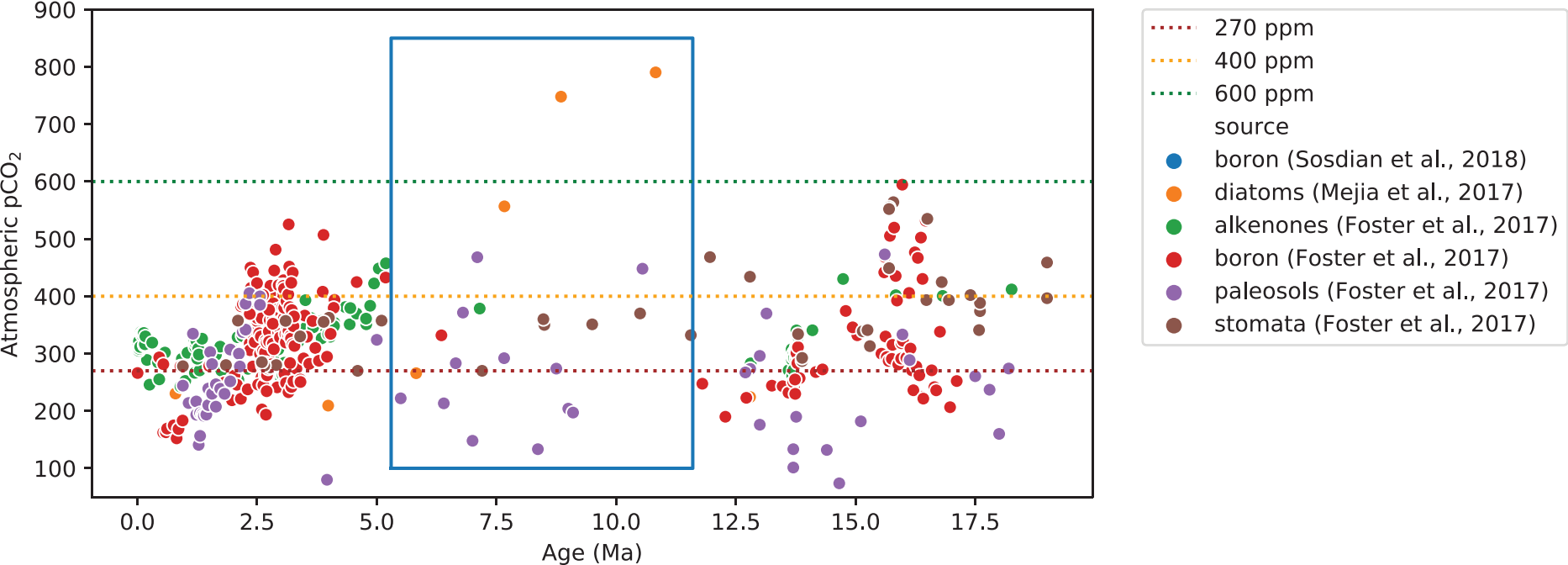


Figure 3.

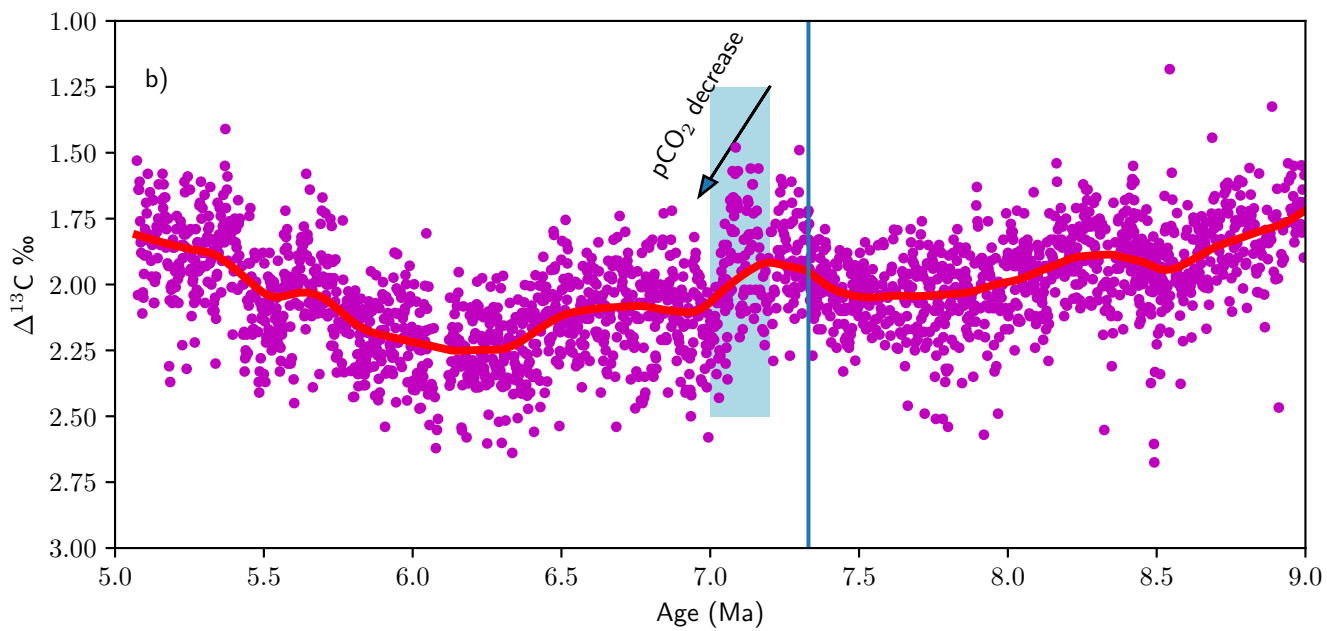
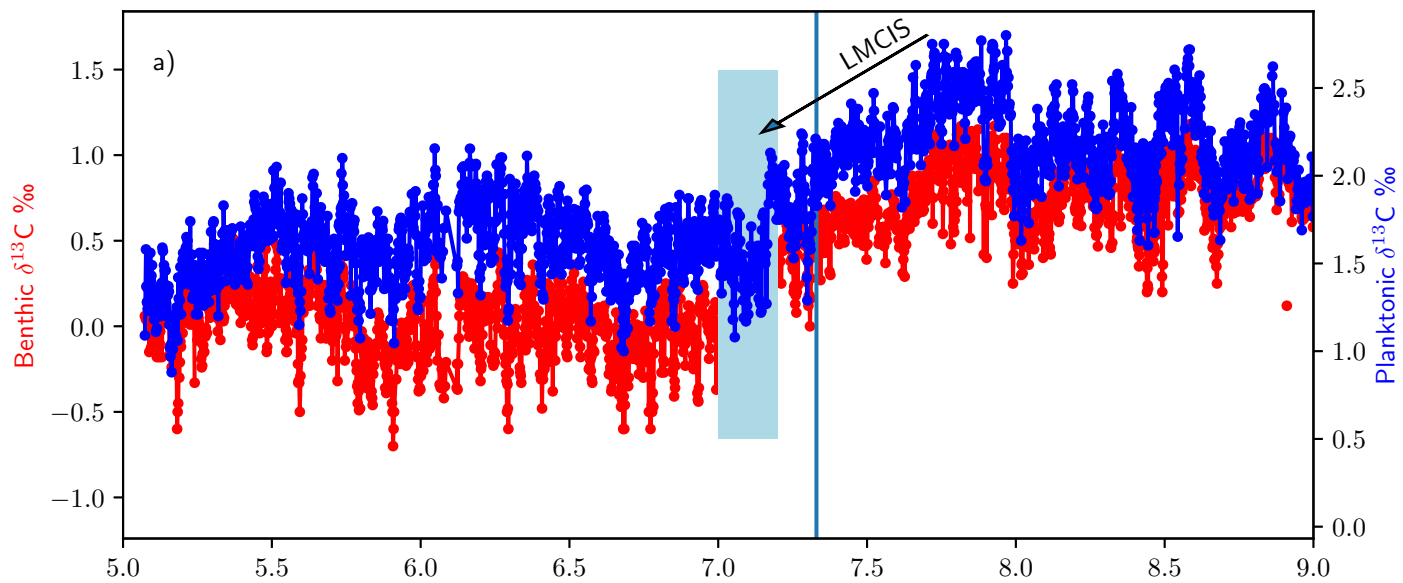
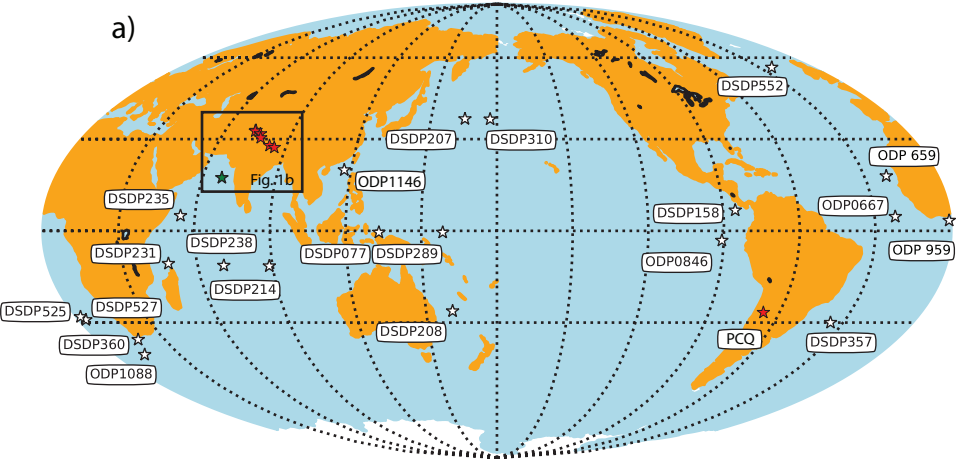


Figure 4.

a)



b)

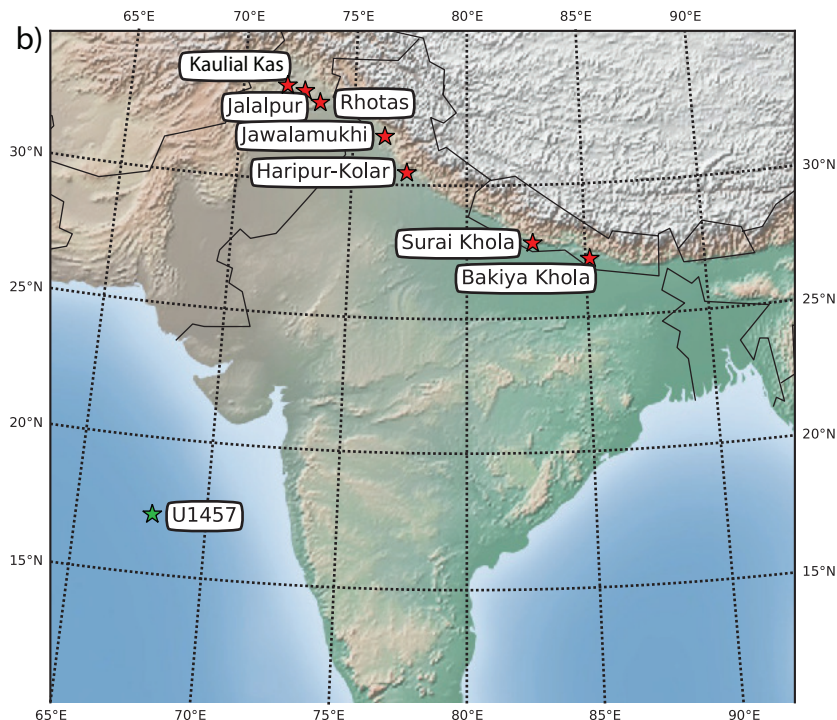


Figure 5.

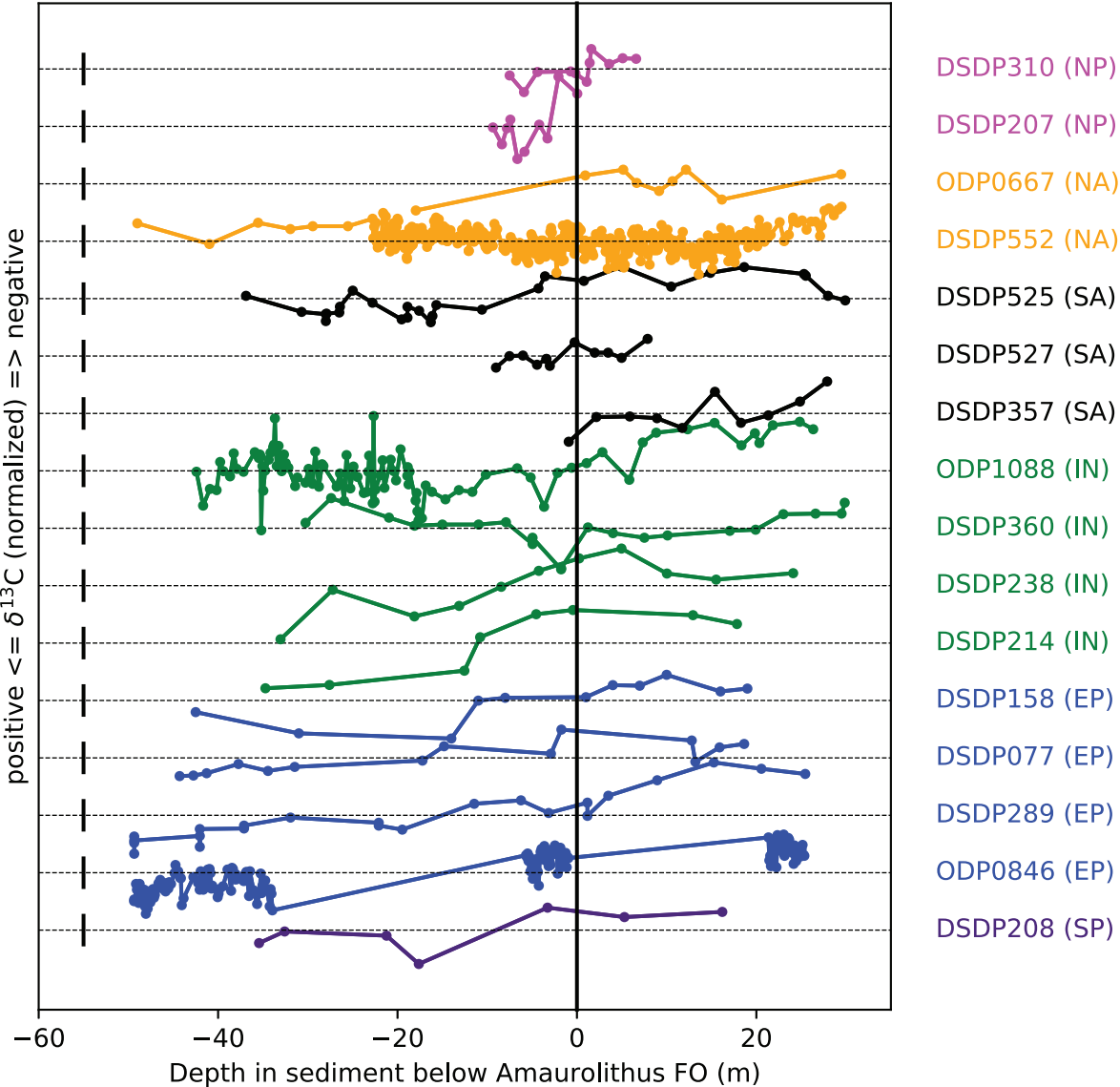


Figure 6.

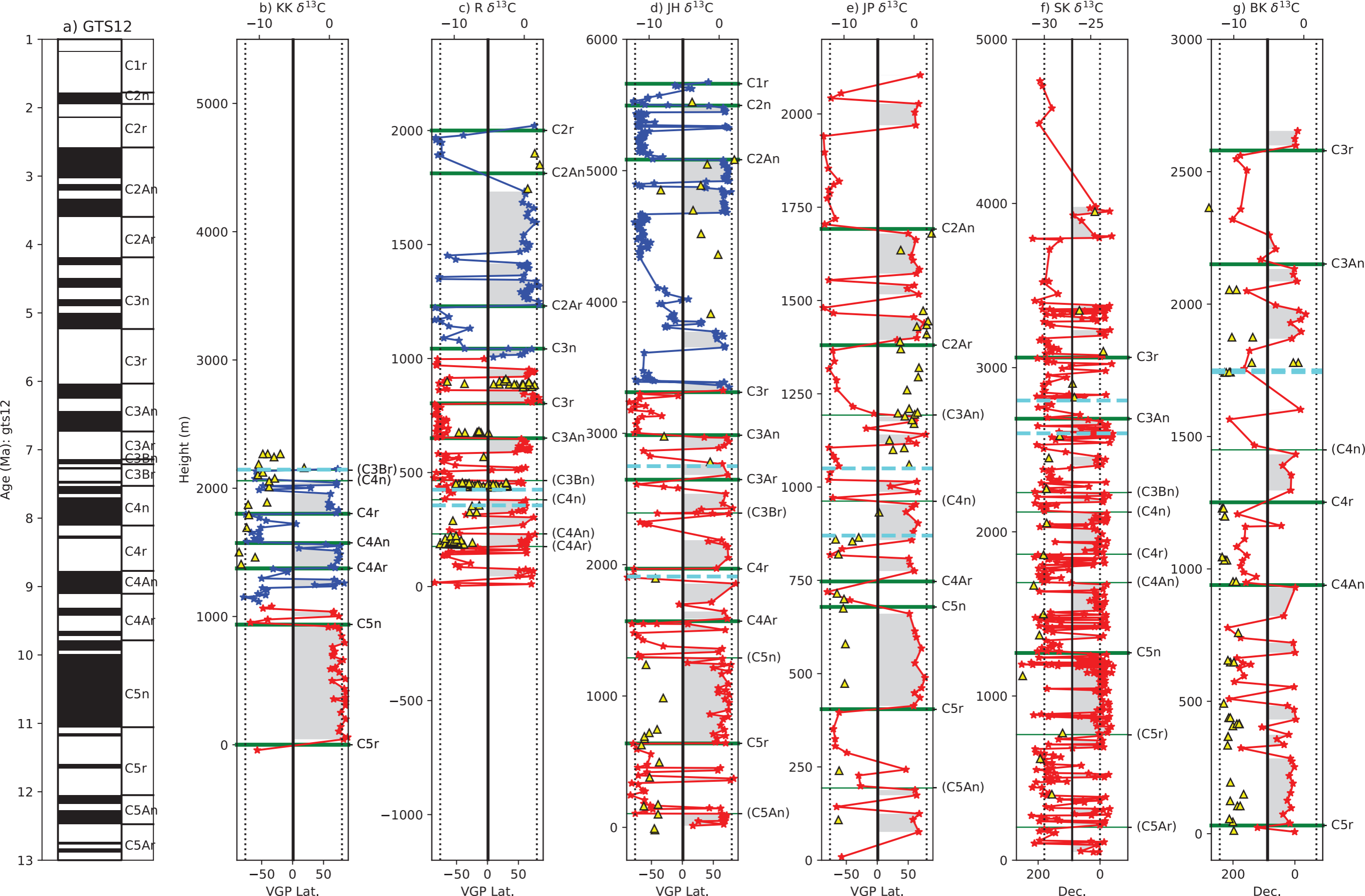


Figure 7.

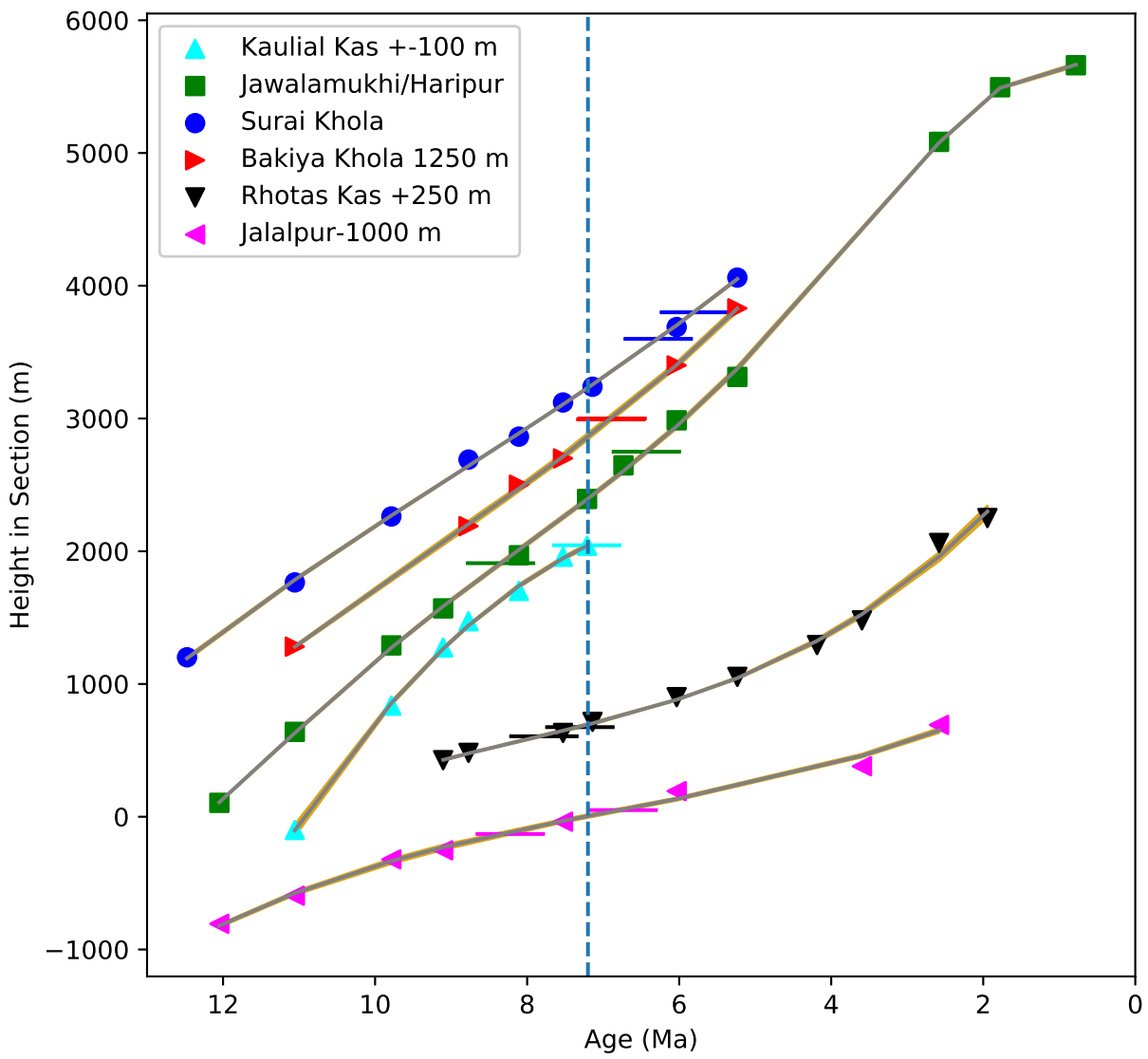
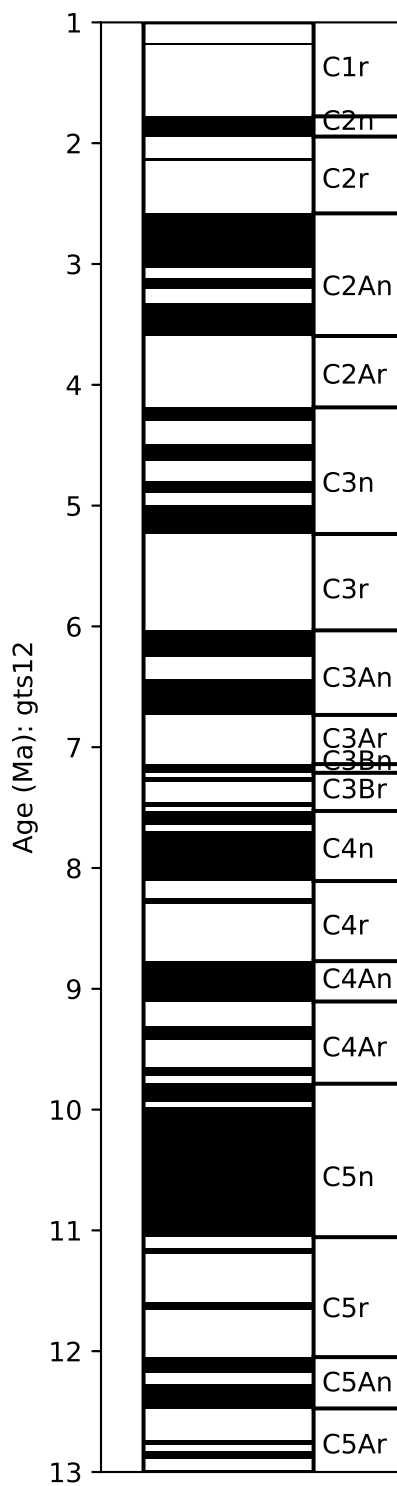
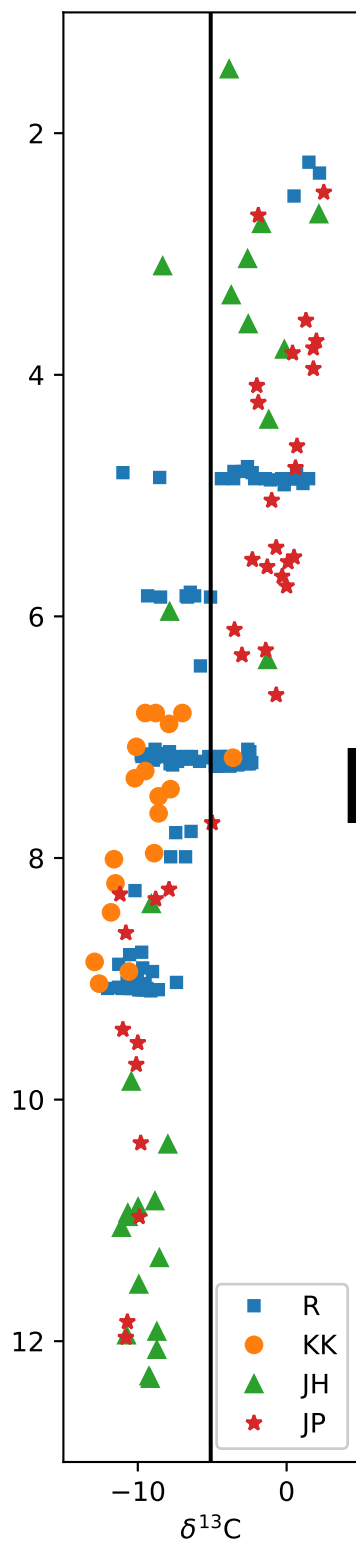


Figure 8.

a) GTS12



b) Pakistan/India



c) Nepal

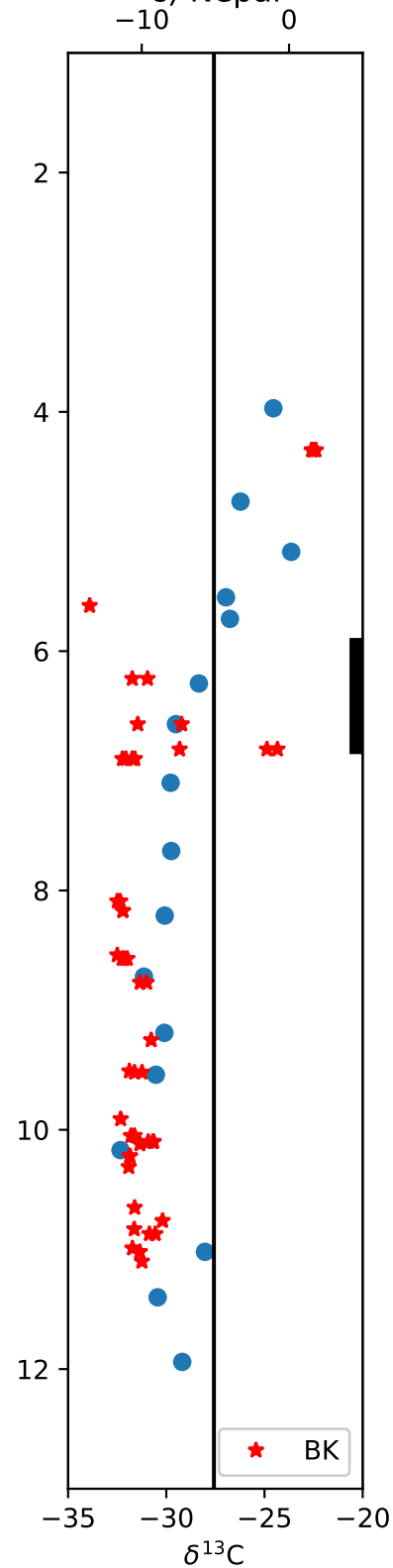
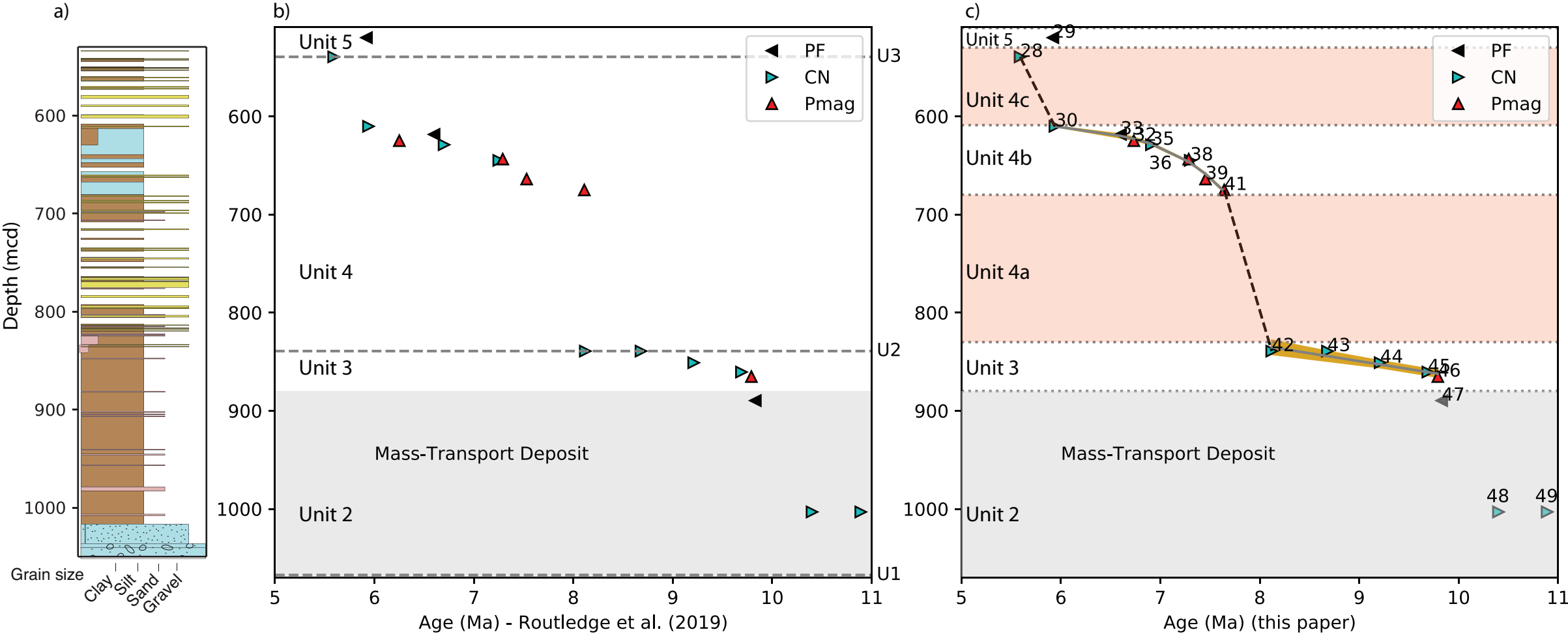


Figure 9.



Lithology

 Carbonate ooze/stone

 Clay/ Claystone

 Silt/ Siltstone

 Sand/ Sandstone

 Calcarenite

 Carbonate breccia

Figure 10.

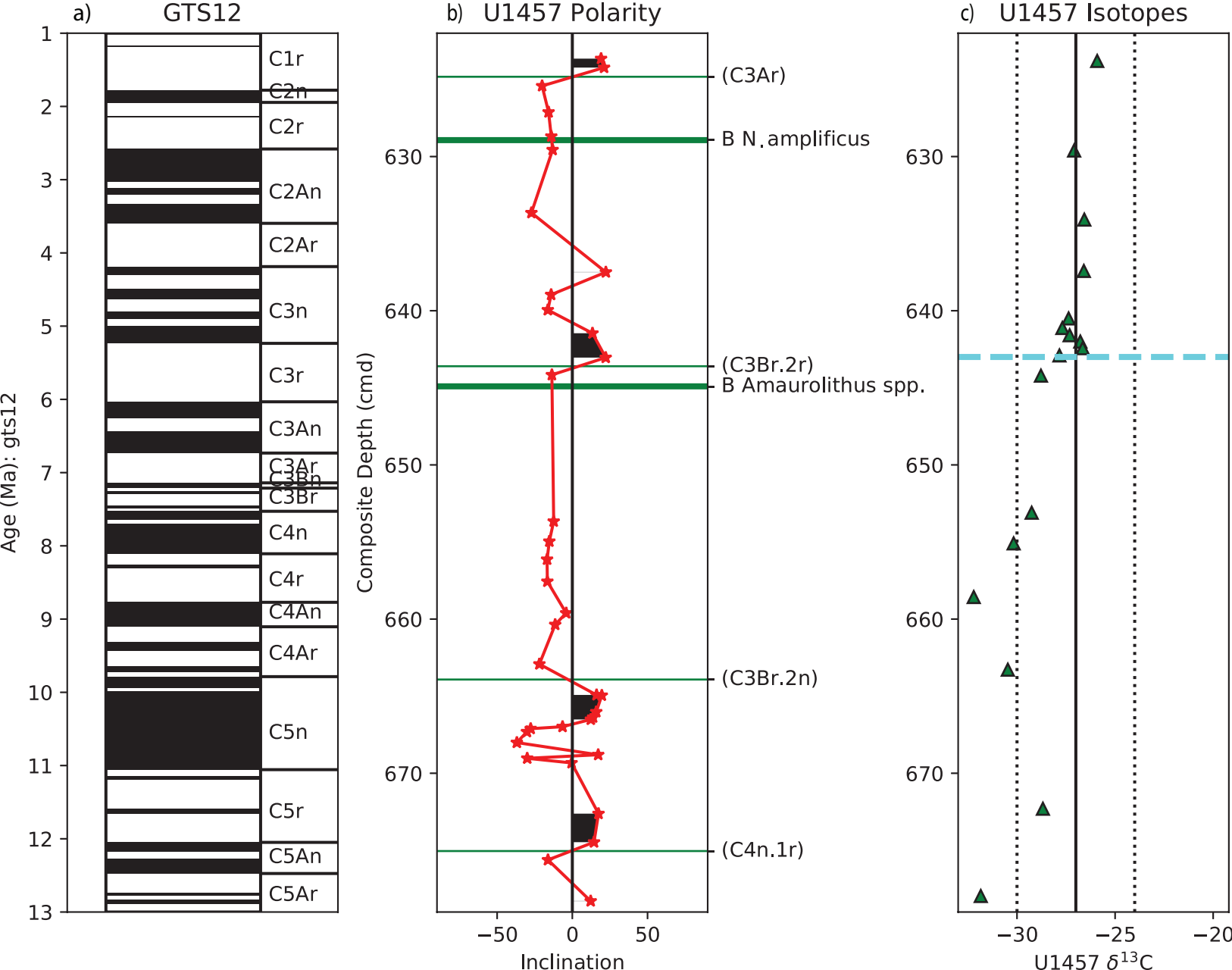


Figure 11.

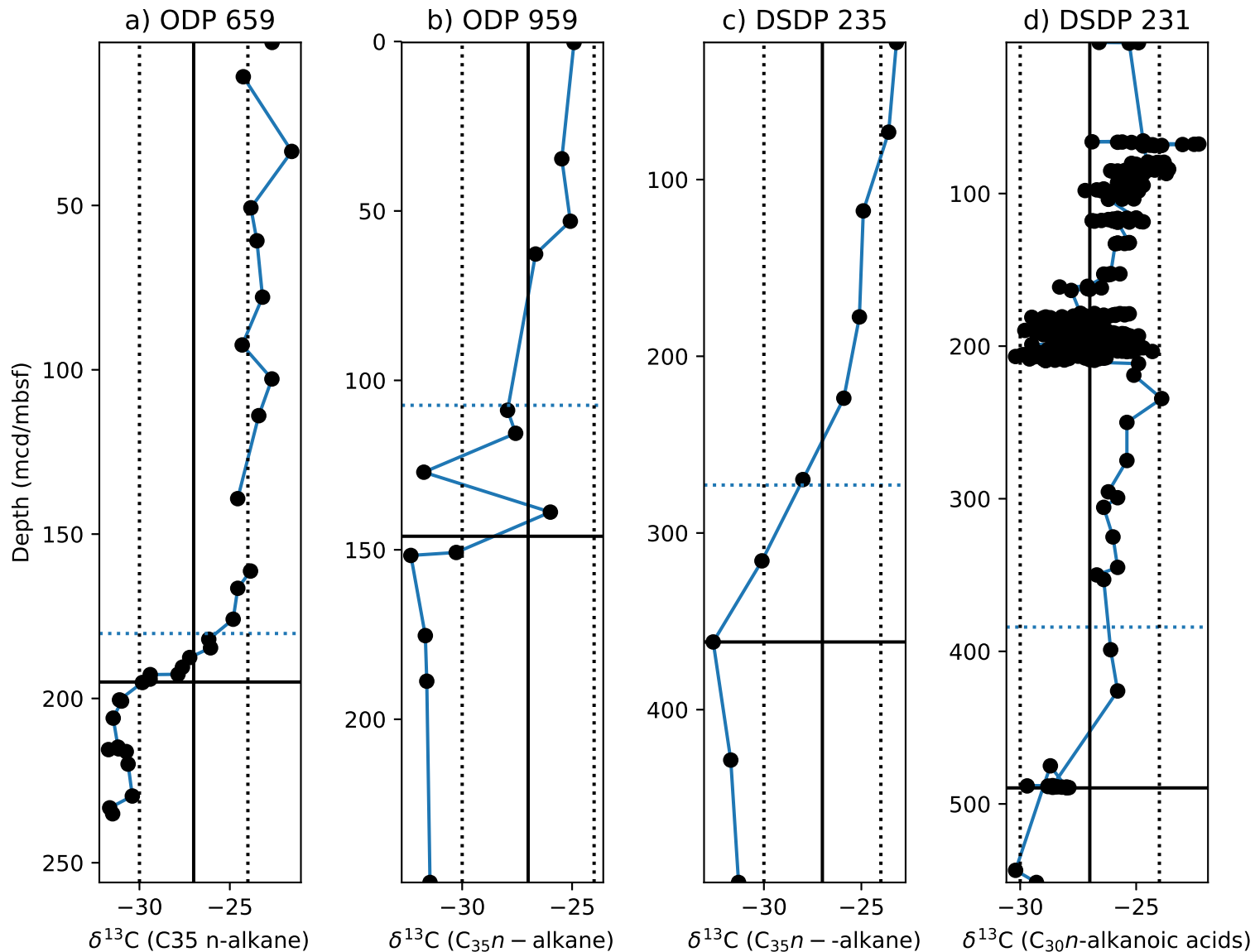


Figure S1.

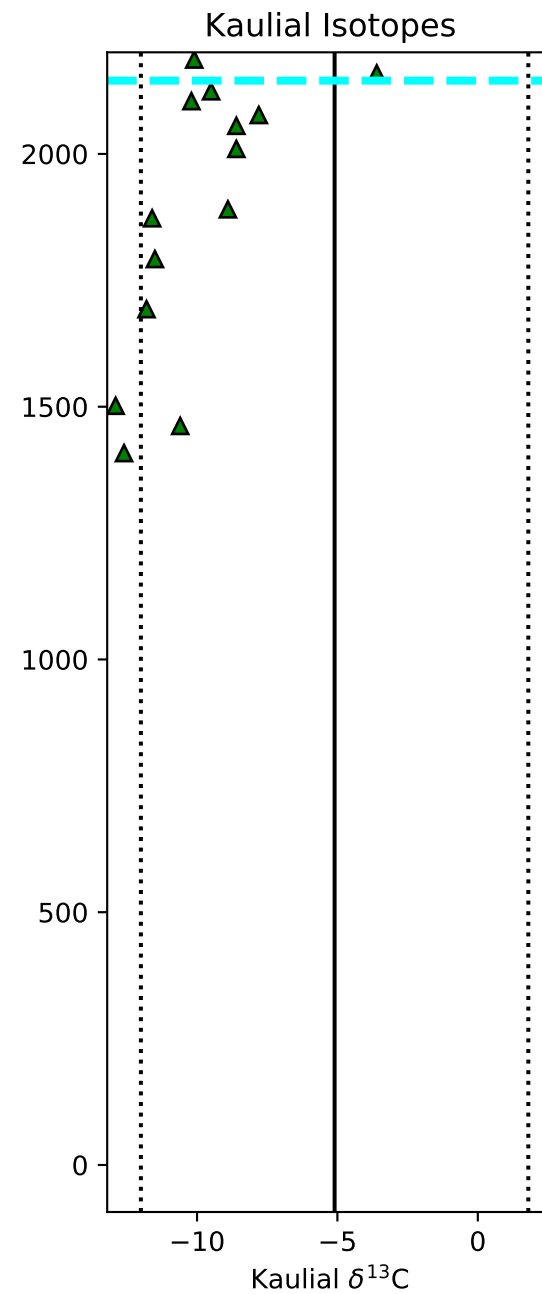
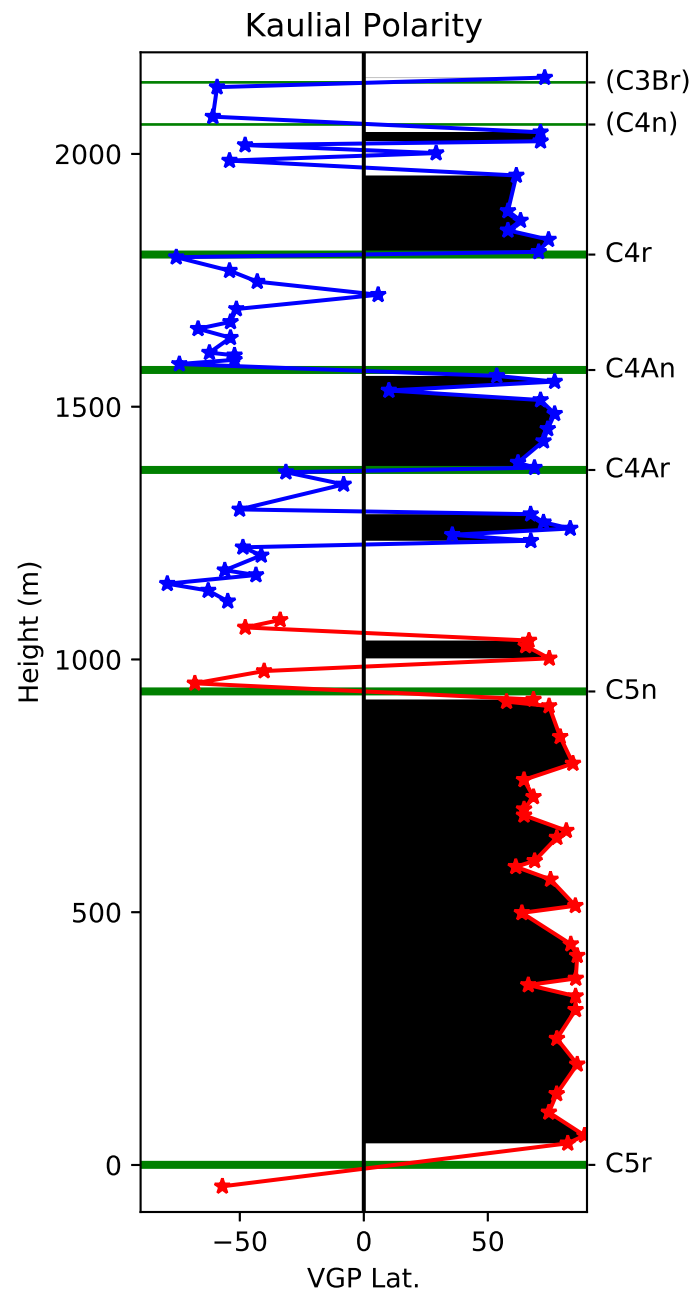
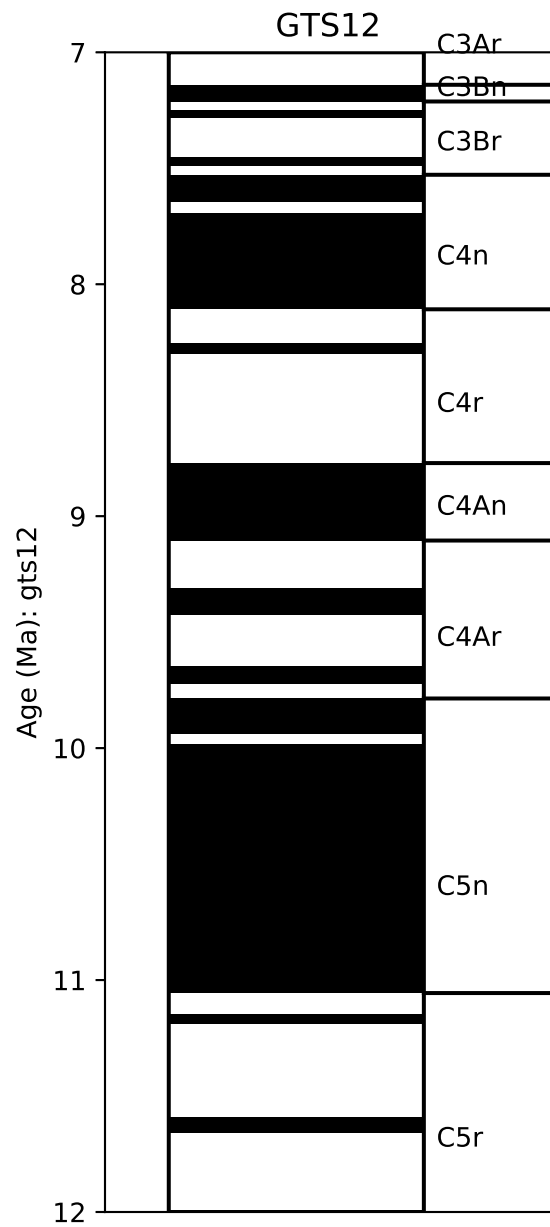


Figure S2.

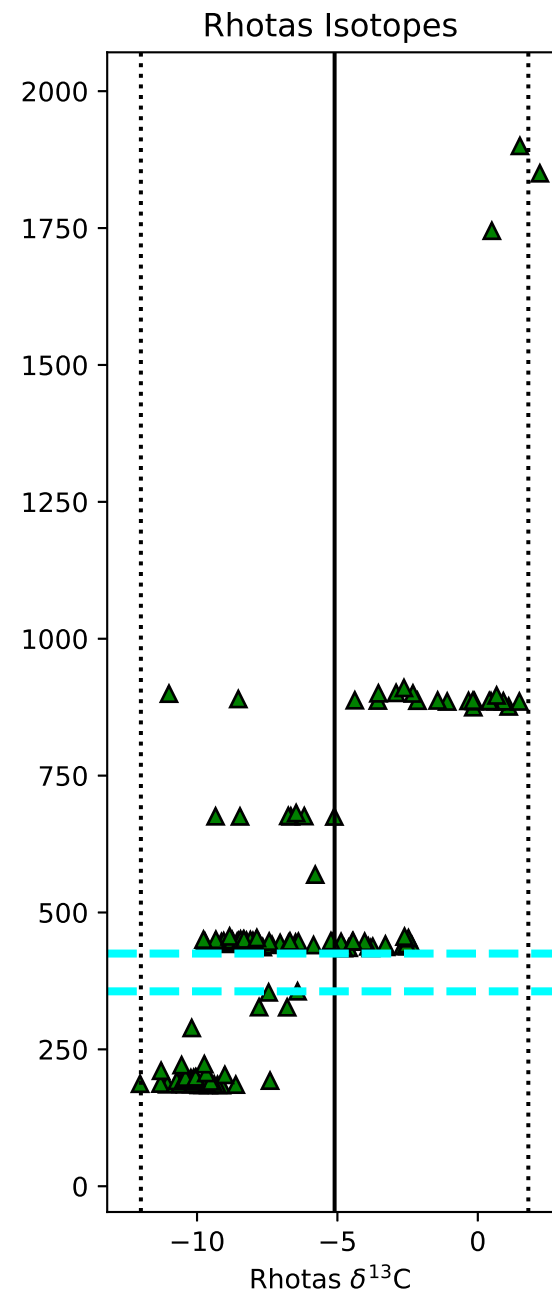
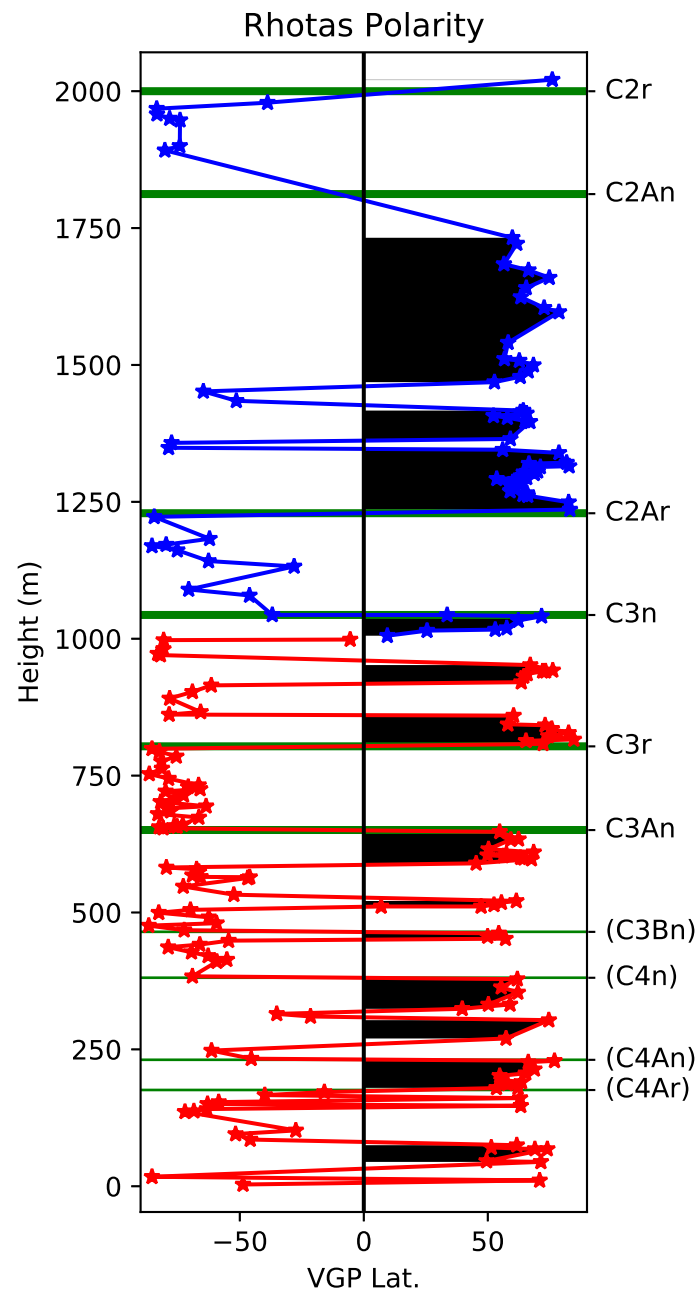
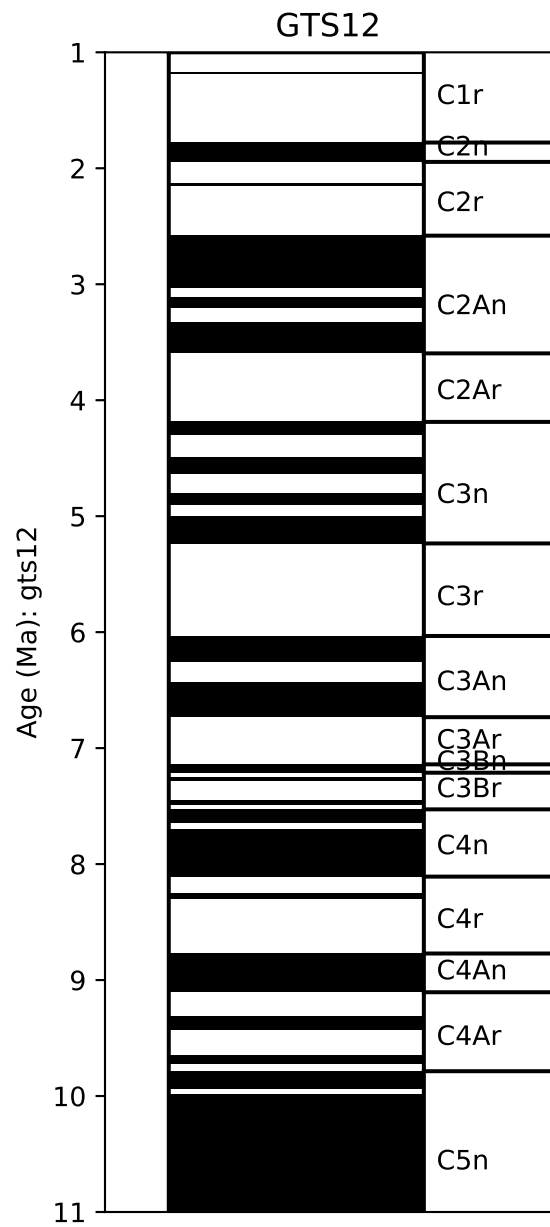


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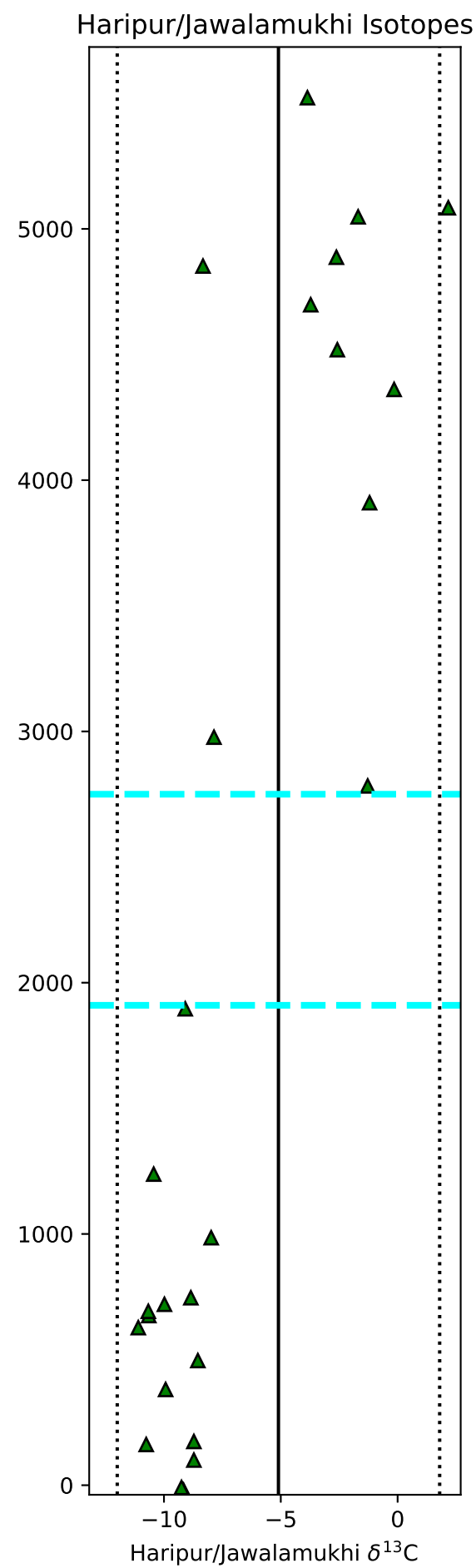
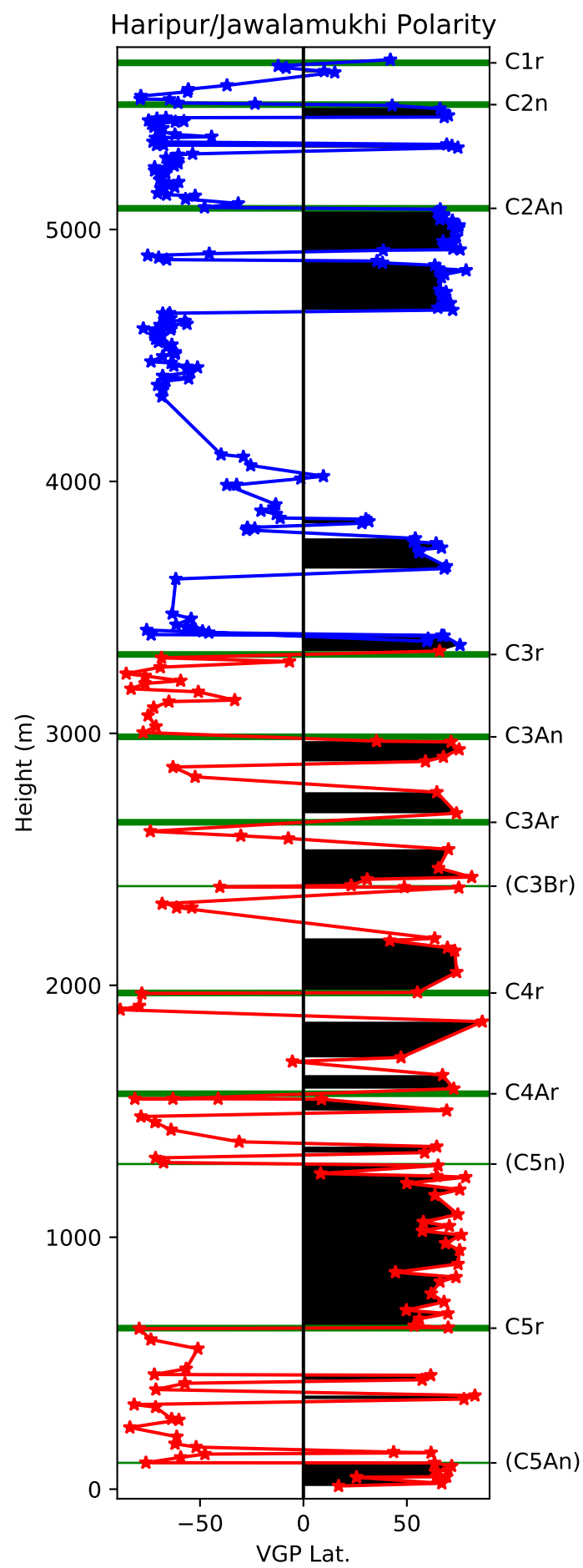
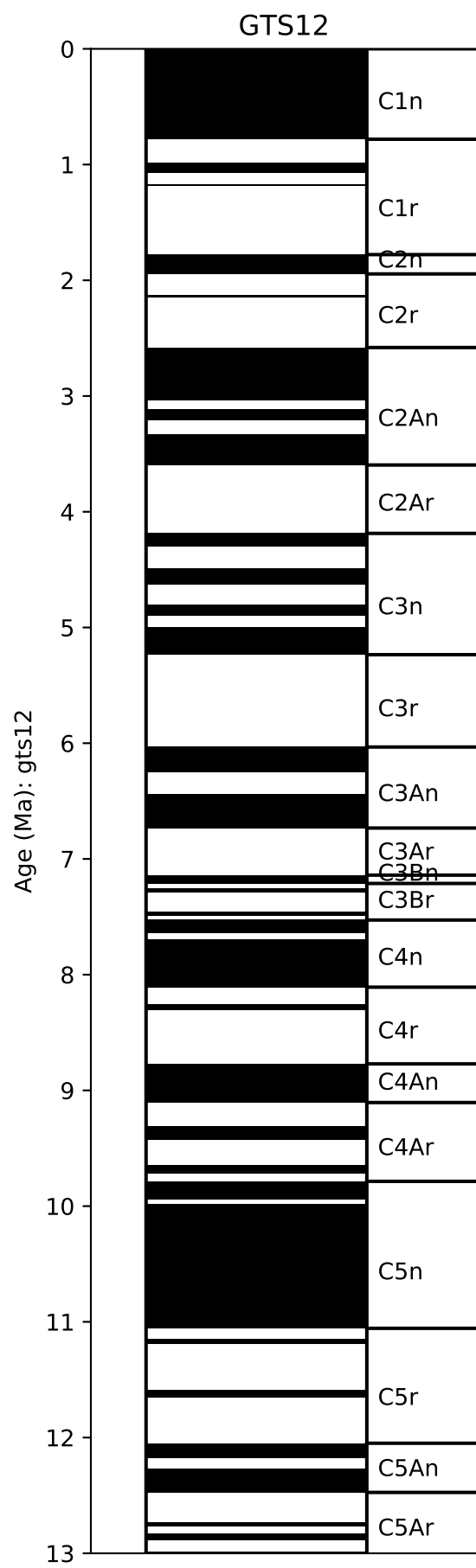


Figure S4.

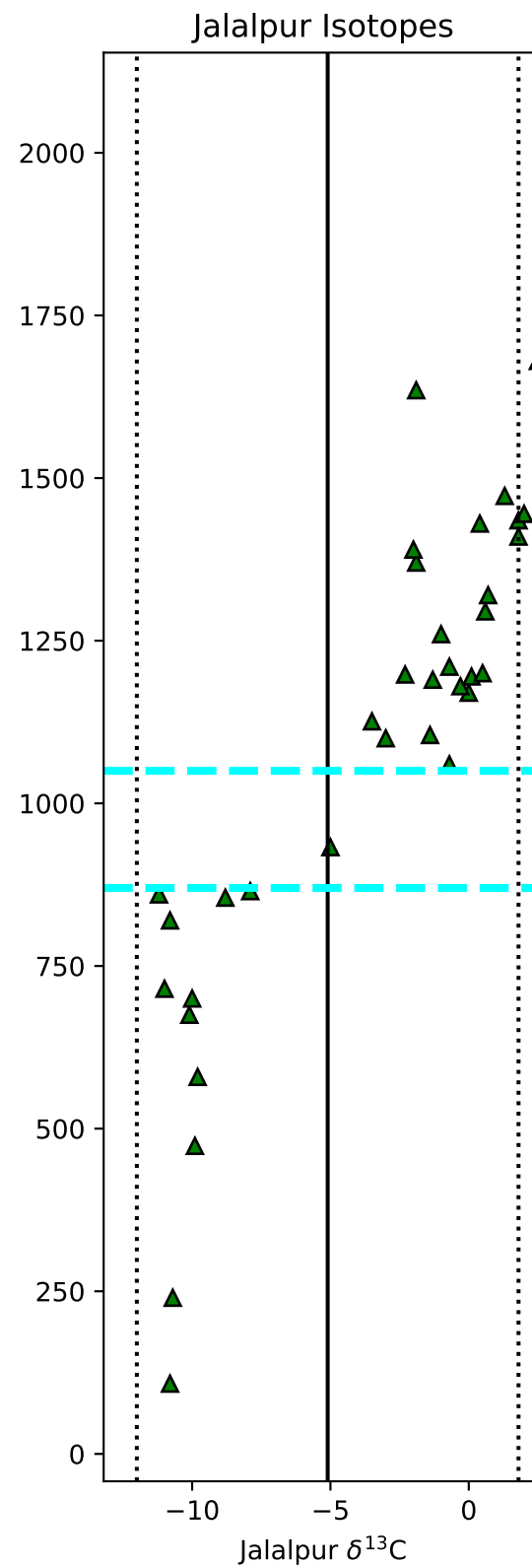
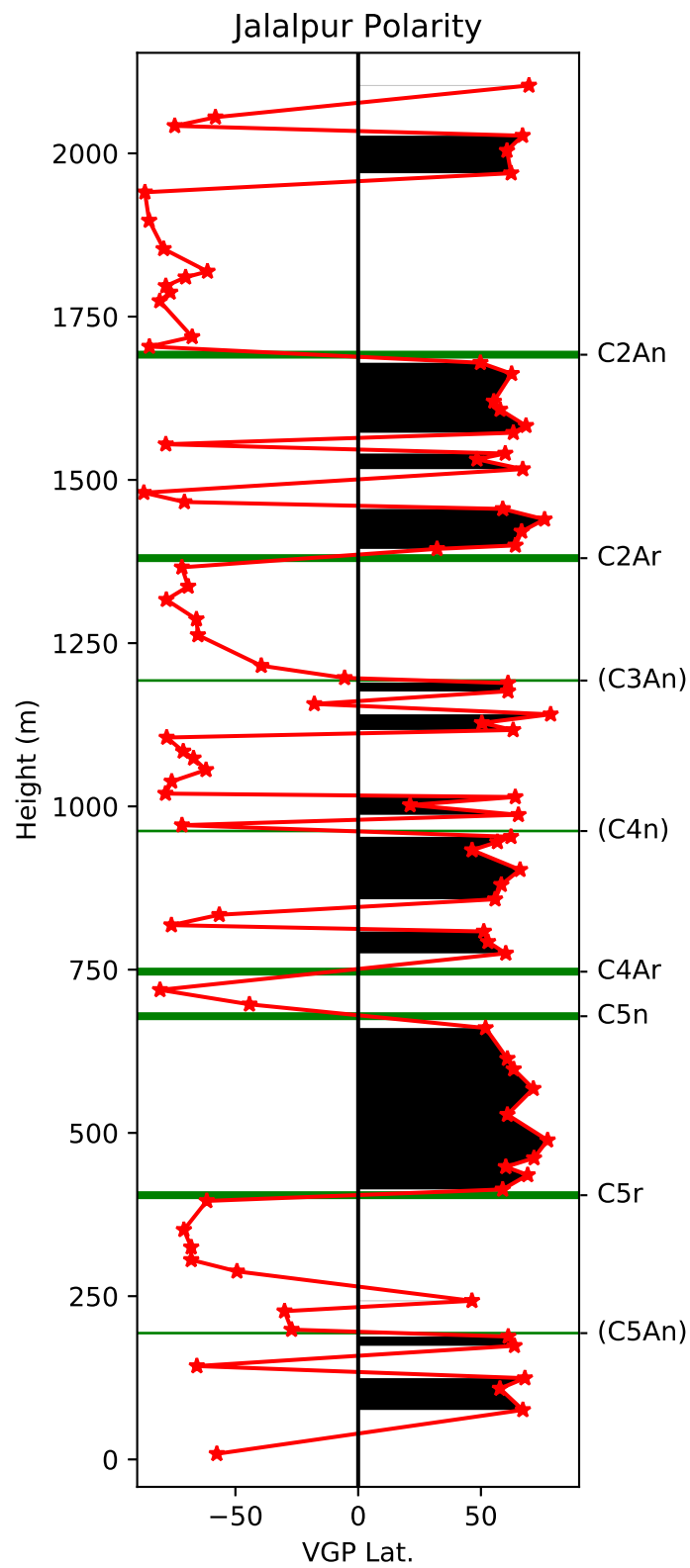
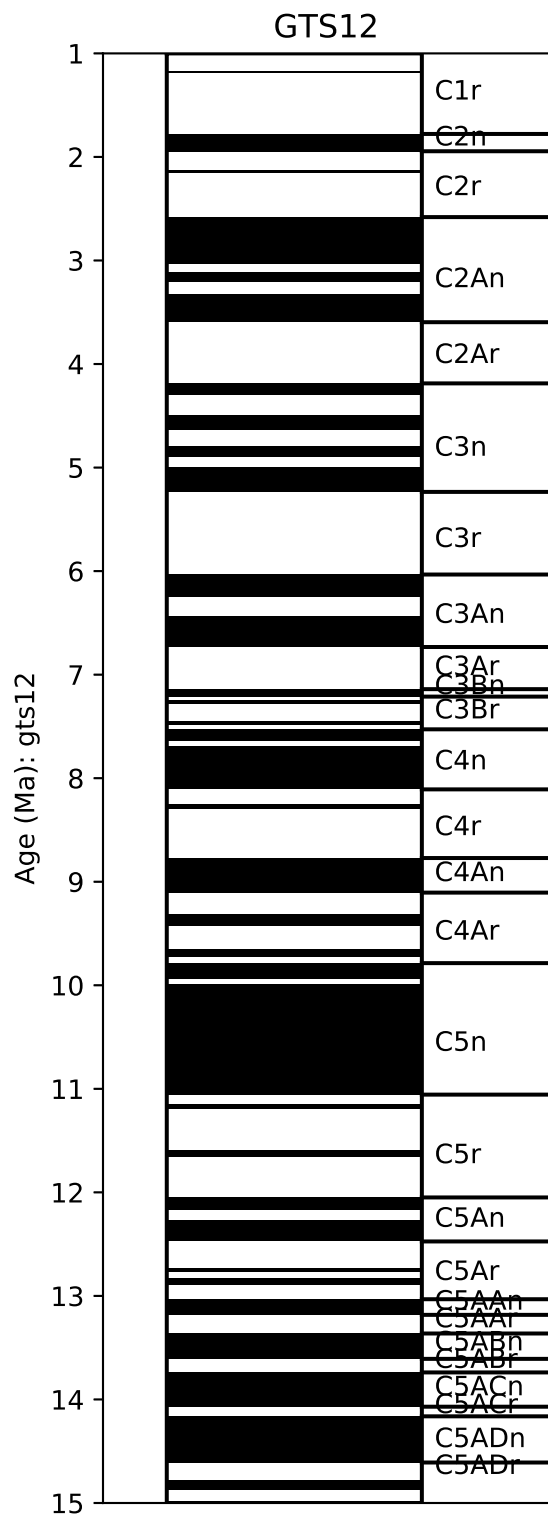


Figure S5.

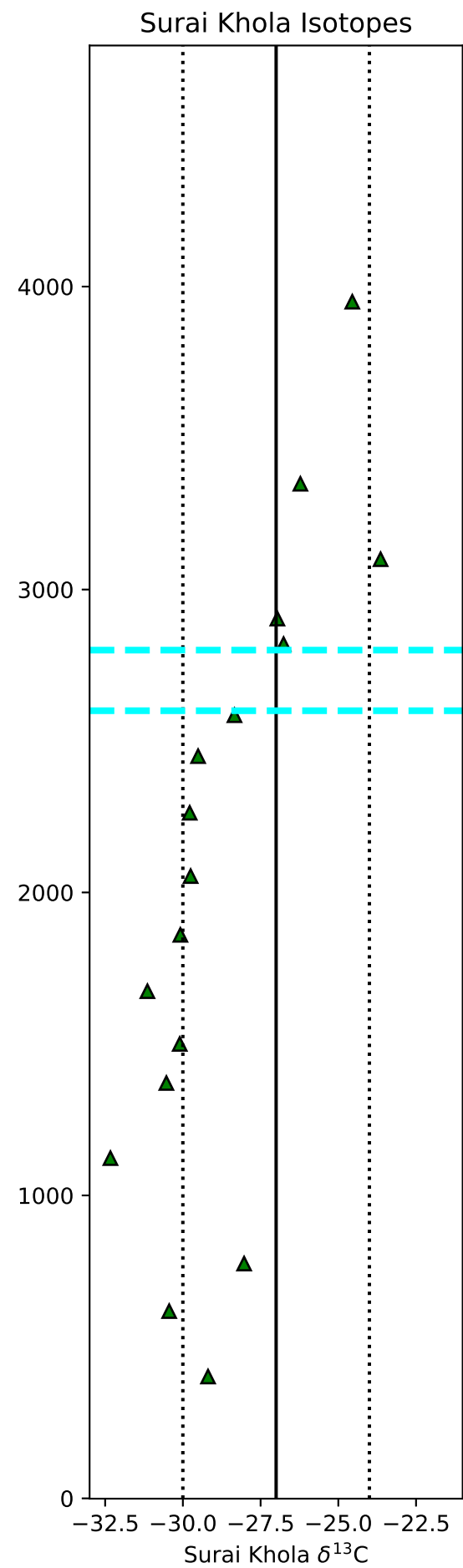
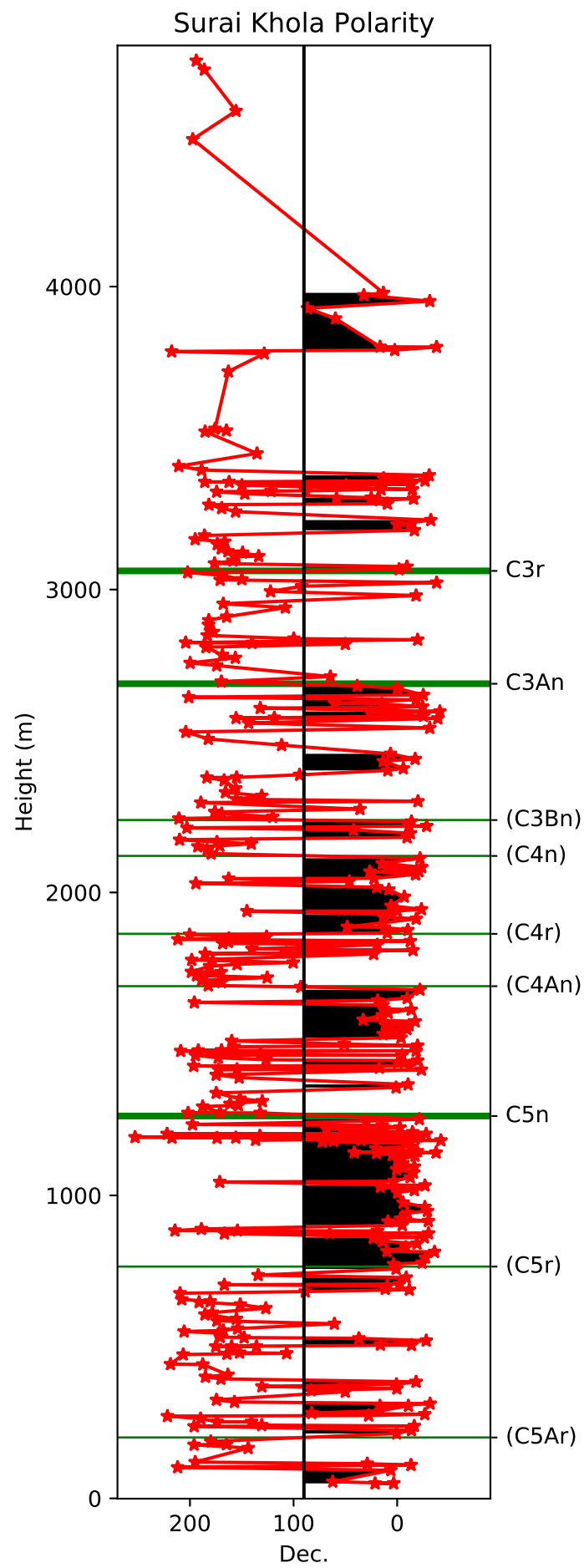
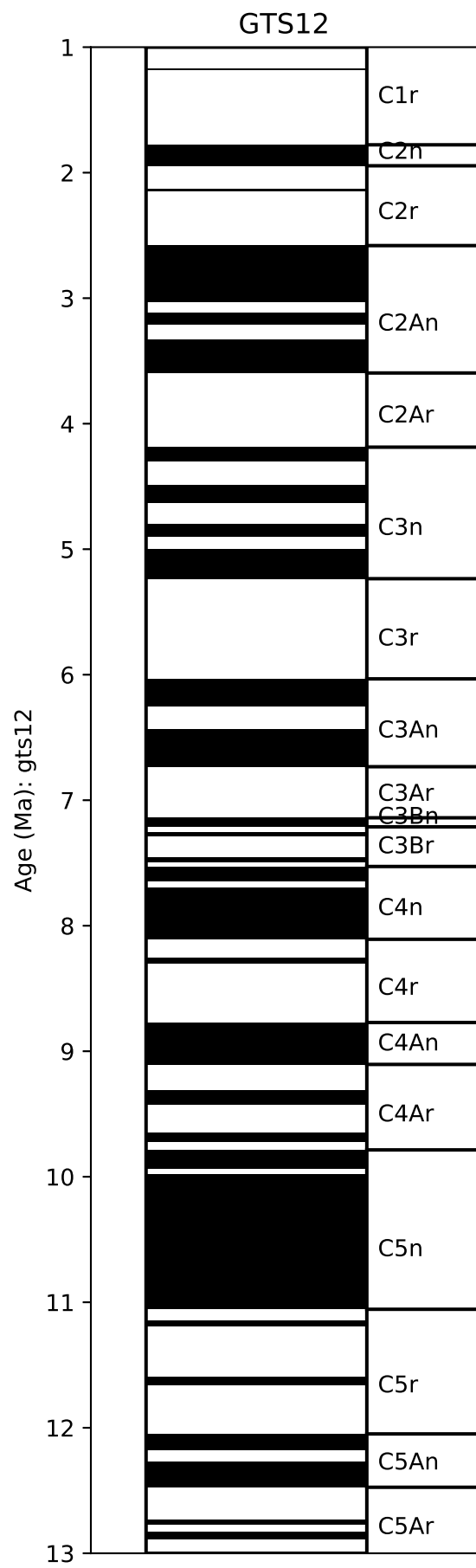


Figure S6.

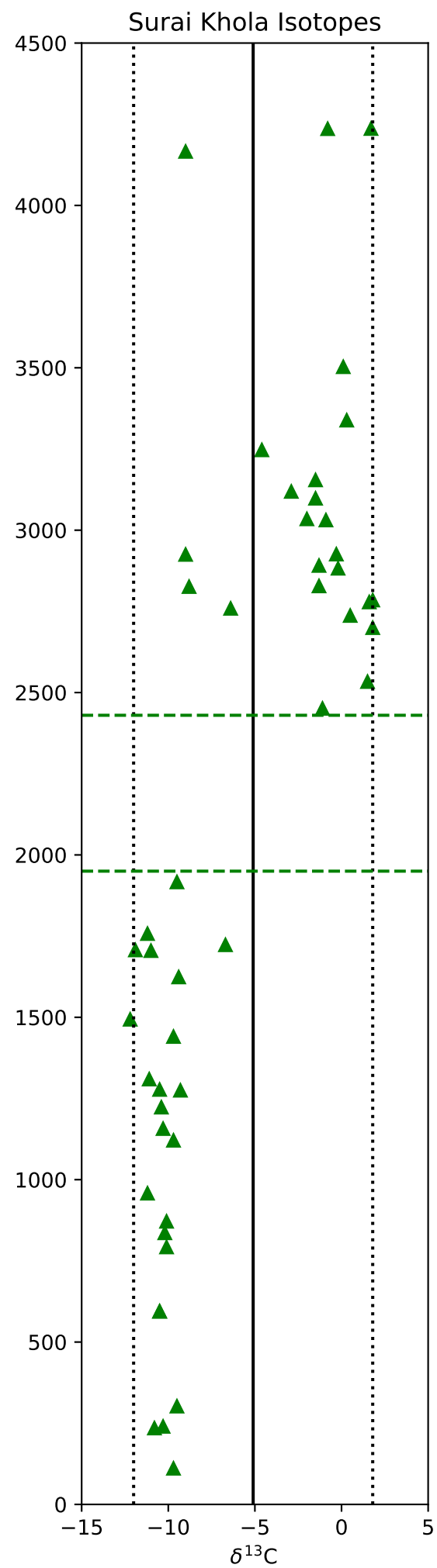
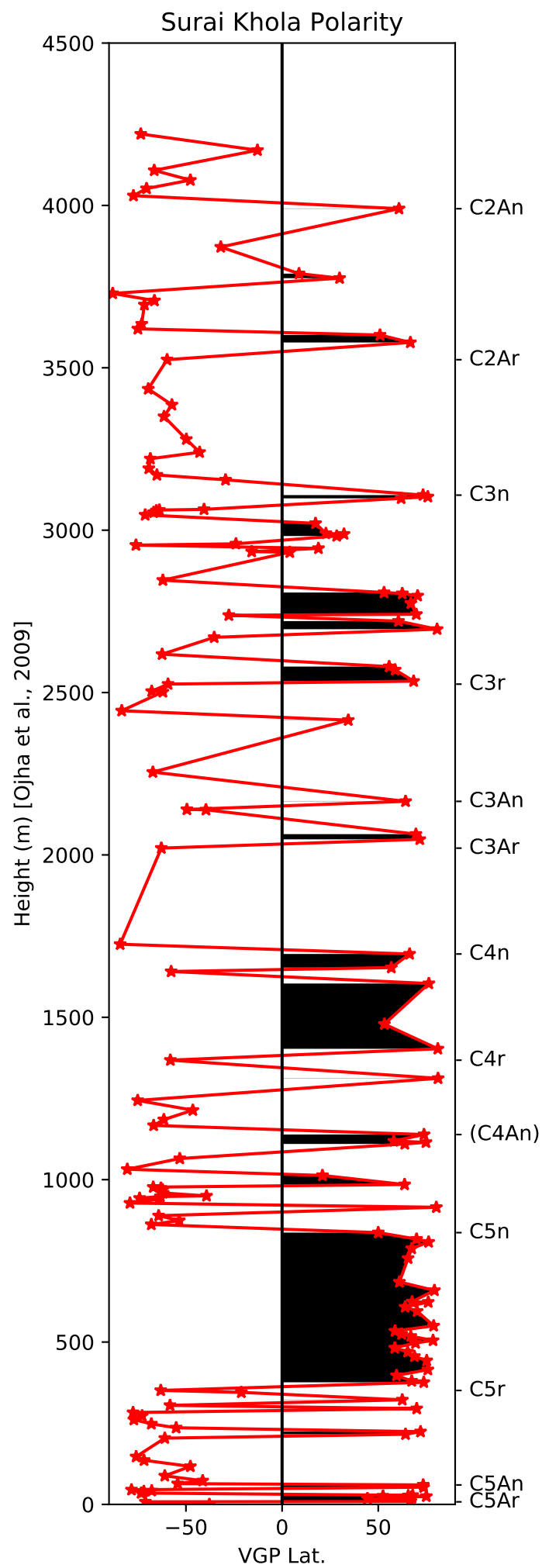
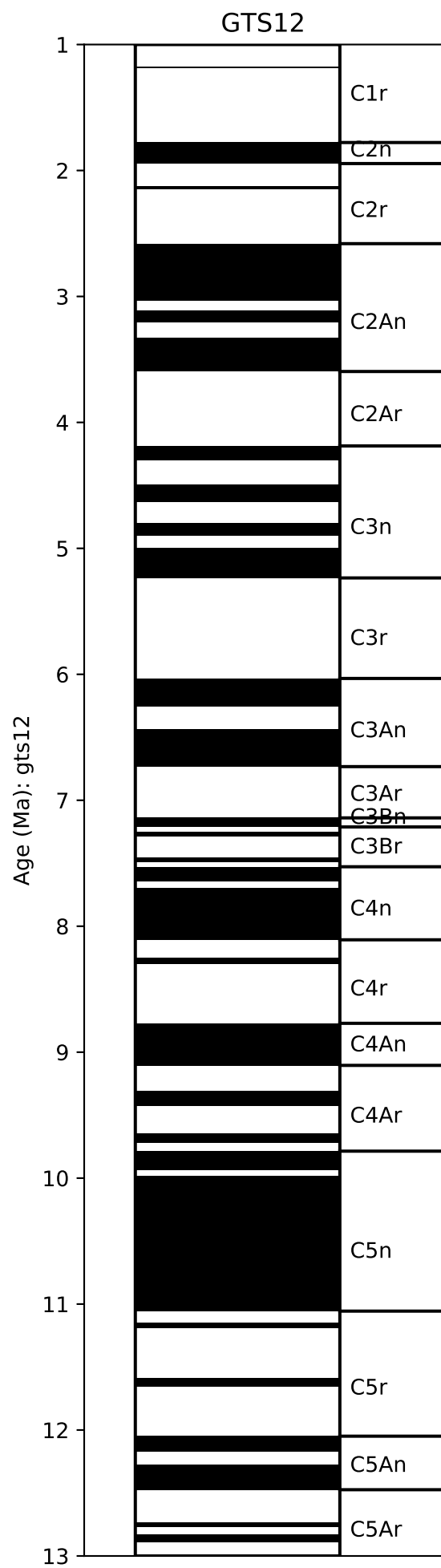


Figure S7.

