Title
A Reassessment of the Chronostratigraphy of Late Miocene C3–C4 Transitions

Permalink
https://escholarship.org/uc/item/3b5912rr

Journal
Paleoceanography and Paleoclimatology, 35(7)

ISSN
2572-4517

Authors
Tauxe, L
Feakins, SJ

Publication Date
2020-07-01

DOI
10.1029/2020pa003857

Peer reviewed
A re-assessment of the chronostratigraphy of late Miocene C$_3$-C$_4$ transitions

L. Tauxe$^1$ and S.J. Feakins$^2$

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

Key Points:

- A change from forest to grassland ecologies began in the late Miocene but chronologies of the published records are based on different time scales.
- Recalibration to a consistent time scale demonstrates the diachronocity of the ecological change in different places around the globe.
- New constraints link a major C$_4$ expansion in the late Miocene with evidence for carbon cycle perturbations.

Corresponding author: Lisa Tauxe, lttxe@ucsd.edu
Abstract

Combining magnetostratigraphic and carbon isotopic data for the late Miocene can provide a temporal framework for an isotopic shift first documented in soil carbonate nodules of northern Pakistan. The shift has been interpreted as a change in vegetation from trees and shrubs (using the C3 photosynthetic pathway) to grassland (using the C4 pathway). The cause of the event has been hotly debated and its timing is close to a shift in carbon isotopes in the marine realm. Further understanding depends critically on timing.

Unfortunately, temporal calibration of the various records published over decades relied on different time scales. To address the lack of a consistent chronology, we have re-evaluated the constraints for the carbon isotopic shifts recorded from the Indian subcontinent. These show a diachronous transition ranging in age from about 7.8 Ma in Pakistan to as late as 6 Ma in Nepal. The record from IODP Expedition 355 Site U1457, drilled on the Indus fan shows that the transition in peninsular India began at about 7.2 Ma. Similar records from the African margin saw an earlier shift to C4 dominance starting around 10 Ma and those from Australia and South America transitioned later, during the Pliocene. The diachronicity around the globe does not invalidate pCO2 as a driver, but is consistent with it being one of several drivers of the global C4 expansion.

Plain Language Summary

Grasslands expanded on the Indian sub-continent in the late Miocene. Precise chronological control is critical to compare the timing of the expansion between regions and to evaluate the possible causes (and consequences) of the ecological transformation. Here we take a new look at published records from Pakistan, India and Nepal and update the age models to a modern geomagnetic reversal timescale. We then compare these to new records from the marine sediments obtained during IODP Expedition 355 to the Indus Fan and others from the African margin. Based on microfossil appearance and paleomagnetic constraints, the timing of the transition to C4 vegetation is found to be asynchronous across the globe, probably initiating in Africa after 10 Ma, then proceeding to Pakistan at around 7.8 Ma and reaching Nepal by ∼6 Ma. Elsewhere in the world (Australia and Argentina) the transition happened several million years later. We also review recent model and proxy evidence to find the causes of the C4 grassland expansion.

1 Introduction

The classic papers of Cerling et al. (1993) and Cerling et al. (1997) (based on earlier work by Quade et al. (1989)) introduced the hypothesis that an expansion of C4 grasses in the late Miocene was linked to a global driver: a putative drop in pCO2 in the atmosphere. However constraining pCO2 in the ancient atmosphere is notoriously difficult (see, e.g., Cerling (1992)), and alkenone carbon isotopic records published by Pagani et al. (1999) shortly after that by Cerling et al. (1997) found no evidence for changes in pCO2 in the late Miocene. Thus the CO2-hypothesis as a driver for change in vegetation in the late Miocene fell out of favor, and other drivers including aridity, herbivory and fire were considered more likely (e.g., Osborne (2008)). However, a global pCO2 change would not trigger a synchronous C4 expansion as thresholds exist in other climatic variables including temperature, rainfall and fire as demonstrated for Africa by Higgins and Scheiter (2012).

Zhou et al. (2018) recently modeled the role of various climatic drivers (CO2, temperature, water and light) in controlling the competitive advantage of C3 versus C4 vegetation globally using a fully coupled climate model under mid Miocene conditions run with three pCO2 concentrations to bracket the range of estimates for the Oligocene to late Miocene (see Figure 1). Simulations run at 600 ppm pCO2, suggested that water
limitation determined a few ‘hot spots’ where C₄ adaptations could emerge including in northern Africa and central Asia and to a lesser extent western North America, southern South America and Africa and western Australia (Figure 1a) and these could be locations were early C₄ evolution innovations occurred (Edwards et al., 2010). For pCO₂ of 400 ppm, the optimality of C₄ plants increased in these regions and was favored over much larger regions, driven primarily by lower pCO₂ with feedbacks of increased light as the canopy opened (Figure 1b). Under low pCO₂ (270 ppm) shown in Figure 1c, optimal conditions for C₄ dominance would have intensified and expanded. However, this scenario sees increases in C₄ coverage beyond that reported in late Miocene reconstructions (especially in places like East Asia and Australia) and thus may be a sign that such low pCO₂ levels were not reached in the late Miocene. Testing the completeness of the forcing assumptions in these simulations requires both a knowledge of CO₂ variations and an accurate chronology for when vegetation changes occurred around the globe.

Figure 1. Map of predictions of C₃ versus C₄ dominance under various pCO₂ scenarios: a) 600 ppm, b) 400 ppm and c) 270 ppm CO₂. [Figure modified from Zhou et al. (2018)].

Estimating pCO₂ in the past is a difficult problem (see for example commentary by Beerling and Royer (2011)). Many proxies have been developed and some have data in the late Miocene interval (δ¹¹B; Sosdian et al. (2018), B/Ca; Tripati et al. (2009), alkenones; Pagani et al. (1999), leaf stomata; Van der Burgh et al. (2009), ; diatom frustules; Mejia et al. (2017), foraminiferal ∆¹³C; Holbourn et al. (2018) and paleosol carbonates (Cerling, 1992). These data were reported on different time scales and the details of precise dates are frequently obscure (e.g., which time scale was used). For example, the leaf stomata data of Van der Burgh et al. (2009) are poorly calibrated with respect to age and it is difficult to tie them to the tight temporal framework required for inter-calibration. A larger problem is that none of the pCO₂ proxies agree with each other (see compilation of Foster et al. (2017) in Figure 2).

Figure 2. CO₂ 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.
The interval of interest here of the late Miocene (shown in the box in Figure 2) has a very sparse data set and little agreement among the various types of proxies. According to the compilation of Foster et al. (2017), there is no convincing evidence for a significant change in pCO₂ since the early Miocene except for the high values found in between 15 and 17 Ma and those from about 3 to 2.5 Ma which have been associated with the mid-Miocene Climatic Optimum and the Pliocene warm interval respectively. More recent records, such as the data from Δ¹³C of organic matter within diatom frustules of Mejia et al. (2017) suggest high pCO₂ and a drop from a high of 800 ppm to 200 ppm with the peak at around 11 Ma and the drop beginning at around 8 Ma.

A recent data set from (Holbourn et al., 2018) based on foraminiferal Δ¹³C records from planktic and benthic foraminifera from ODP Site 1146 (shown in Figure 3) are the most detailed for the interval of interest here and can be placed on the time scale used here (Gradstein et al., 2012). These data provide no quantitative estimate of pCO₂, however. Nonetheless Δ¹³C data are interpreted to indicate a drop in pCO₂ values between ~7 and ~7.5 Ma, possibly synchronous with part of the carbon isotopic shift in plant proxies discussed in this paper. Thus, these new pCO₂ proxies and the modeling efforts of (Zhou et al., 2018) reawaken a possibility of pCO₂ mediating the regional C₄ expansion in a diachronous progress as modification of the mechanism for a global synchronous transition envisioned by Cerling et al. (1993) and Cerling et al. (1997).

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

Once the timing of the change in vegetation on the Indian sub-continent is secured, we can ask what combination of climate and CO₂ factors drove the C₄ expansion. We can also ask whether there is positive C₄ feedback on the carbon cycle. Commonalities of the timing of C₄ expansion and a shift in the δ¹³C of marine carbonates (Feakins et al., This volume) have been noted. Grasslands can be quite efficient at sequestering CO₂ in soils (Fornara & Tilman, 2008; Spiesman et al., 2018; Yang et al., 2019) in comparison to the thin soil under the C₃ forests they replaced in the Indian sub-continent (although elsewhere replaced C₃ grasses (Feakins et al., 2013)). This positive feedback in turn could enhance global cooling (e.g., Retallack (2013); Retallack et al. (2018)).

2 Timing of the C₃-C₄ transition

2.1 The marine realm

In addition to the carbon isotope shift on land first detected by Quade et al. (1989) and expanded on by Cerling et al. (1993), there is also a marked carbon isotope transition recorded in deep sea sediments at about the same time. In fact, the story of the late Miocene shift in carbon isotopes began with a study by Keigwin of deep sea sediments (Keigwin, 1979). He documented a mysterious decrease of up to 0.8% in the δ¹³C in benthic foraminifera in Deep Sea Drilling Project (DSDP) cores recovered at Site 158 in the Panama Basin (Easternmost Pacific) and Site 310 (Hess Rise) in the North Pacific (Figures 4a and 5 and Table 1). Keigwin inferred an age for the shift of ∼ 6.5 Ma based on correlation of the biostratigraphy (including the first occurrence (FO) of Cer-
Figure 3. a) Benthic (red) and planktic (blue) δ¹³C 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 δ¹³C (Δ¹³C), plotted with lower values to the top. Higher values have been interpreted as resulting from lower atmospheric pCO₂ concentrations. The decrease in pCO₂ inferred between 7.2 and 7.1 Ma is as in Holbourn et al. (2018).

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

Shortly after the discovery by Keigwin, Vincent et al. (1980) found similar shifts in the Indian Ocean at DSDP Site 238 (Figures 4a and 5), also closely associated with the FO of *A. primus*. Vincent et al. (1980) echoed Keigwin in explaining the shift as resulting from “changes in ocean circulation”. The Vincent et al. study was immediately followed by the study of Haq et al. (1980) who found the same shift occurring just after the FO of *Amaurolithus spp.* (i.e., *A. primus*) in the world’s oceans.
More recently, Cramer et al. (2009) compiled records from Deep Sea Drilling Project (DSDP) and Ocean Drilling Program (ODP) cores with benthic foraminiferal carbon isotopic data ($\delta^{13}C$) organized by ocean basin. In this way, water mass differences between basins resulting from ocean circulation patterns are held separate, allowing for detection of global carbon isotopic transitions rather than artifacts of the averaging of a varied number of records with ocean basin bias. Because we are interested in the precise timing of the carbon isotope shifts relative to each other, we consider only those records that also have the FO of *Amaurolithus* spp. (see Figure 5 and Table 1). We plot the different records against stratigraphic depth relative to the FO of *Amaurolithus* spp. in Figure 5. There does appear to be a decrease in $\delta^{13}C$ associated with the FO of *Amaurolithus* spp. in many records, but some occur prior to the biostratigraphic datum (e.g., ODP1088) while others show no change at all (DSDP 552).

### 2.2 Records from the continents

Starting with Quade et al. (1989), the focus of Miocene carbon isotope studies shifted to the continents. They found a large (~10%) increase in $\delta^{13}C$ values in paleosol carbonate 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).

<table>
<thead>
<tr>
<th>Site/Section</th>
<th>Lat.</th>
<th>Lon.</th>
<th>FO A. spp. (mbsf/mcd)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>DSDP077</td>
<td>-0.48</td>
<td>133.23</td>
<td>130</td>
<td>Keigwin and Corliss (1986), Woodruff et al. (1981), Woodruff and Savin (1989)</td>
</tr>
<tr>
<td>DSDP158</td>
<td>6.63</td>
<td>-85.24</td>
<td>150</td>
<td>Keigwin (1979)</td>
</tr>
<tr>
<td>DSDP207</td>
<td>36.96</td>
<td>165.43</td>
<td>94</td>
<td>Haq et al. (1980)</td>
</tr>
<tr>
<td>DSDP208</td>
<td>-26.11</td>
<td>161.22</td>
<td>180</td>
<td>Haq et al. (1980)</td>
</tr>
<tr>
<td>DSDP214</td>
<td>-11.34</td>
<td>88.72</td>
<td>128</td>
<td>Haq et al. (1980)</td>
</tr>
<tr>
<td>DSDP231</td>
<td>-11.89</td>
<td>48.25</td>
<td>384.1</td>
<td>Feakins et al. (2013), Fisher et al. (1974)</td>
</tr>
<tr>
<td>DSDP235</td>
<td>3.23</td>
<td>52.68</td>
<td>273</td>
<td>Uno et al. (2016), Party (1974)</td>
</tr>
<tr>
<td>DSDP238</td>
<td>-11.15</td>
<td>70.53</td>
<td>188</td>
<td>Haq et al. (1980)</td>
</tr>
<tr>
<td>DSDP289</td>
<td>-0.50</td>
<td>158.51</td>
<td>224</td>
<td>Woodruff et al. (1981)</td>
</tr>
<tr>
<td>DSDP310</td>
<td>36.85</td>
<td>176.90</td>
<td>68</td>
<td>Keigwin (1979)</td>
</tr>
<tr>
<td>DSDP357</td>
<td>-30.00</td>
<td>-35.56</td>
<td>24.135</td>
<td>Cramer et al. (2009)</td>
</tr>
<tr>
<td>DSDP360</td>
<td>-35.85</td>
<td>18.10</td>
<td>139.5</td>
<td>Wright et al. (1992)</td>
</tr>
<tr>
<td>DSDP525</td>
<td>-29.07</td>
<td>2.99</td>
<td>98.6</td>
<td>Shackleton et al. (1984)</td>
</tr>
<tr>
<td>DSDP527</td>
<td>-28.04</td>
<td>1.76</td>
<td>104.5</td>
<td>Shackleton et al. (1984)</td>
</tr>
<tr>
<td>DSDP552</td>
<td>56.04</td>
<td>-23.23</td>
<td>127</td>
<td>Cramer et al. (2009)</td>
</tr>
<tr>
<td>DSDP659</td>
<td>18</td>
<td>-21</td>
<td>180.24</td>
<td>Polissar et al. (2019), Ruddiman et al. (n.d.)</td>
</tr>
<tr>
<td>ODP0846</td>
<td>-3.09</td>
<td>-90.82</td>
<td>251</td>
<td>Diester-Haass et al. (2006)*</td>
</tr>
<tr>
<td>ODP0667</td>
<td>4.57</td>
<td>-21.91</td>
<td>107</td>
<td>Curry and Miller (1989)</td>
</tr>
<tr>
<td>DSDP959</td>
<td>3.5</td>
<td>-2.6</td>
<td>107.35</td>
<td>Polissar et al. (2019), Backman et al. (2012)</td>
</tr>
<tr>
<td>U1457</td>
<td>17.17</td>
<td>67.93</td>
<td>645</td>
<td>This study, Feakins et al.(this volume)</td>
</tr>
<tr>
<td>Kaulial Kas</td>
<td>333.34</td>
<td>72.70</td>
<td></td>
<td>Tauxe and Opdyke (1982), Quade et al. (1995)</td>
</tr>
<tr>
<td>Rhotas:</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dhabwala Kas</td>
<td>32.95</td>
<td>73.58</td>
<td></td>
<td>Behrensmeier et al. (2007), Opdyke et al. (1979)</td>
</tr>
<tr>
<td>Basawa Kas</td>
<td>32.95</td>
<td>73.579</td>
<td></td>
<td>Behrensmeier et al. (2007)</td>
</tr>
<tr>
<td>Jalalpur</td>
<td>32.75</td>
<td>73.42</td>
<td></td>
<td>Quade and Cerling (1995), Johnson et al. (1982)</td>
</tr>
<tr>
<td>Jawalamukhi</td>
<td>31.8</td>
<td>76.39</td>
<td></td>
<td>Vogeli et al. (2017), Meigs et al. (1995)</td>
</tr>
<tr>
<td>Haripur Kolar</td>
<td>30.46</td>
<td>77.39</td>
<td></td>
<td>Vogeli et al. (2017), Sangode et al. (1996)</td>
</tr>
<tr>
<td>Surai Khola</td>
<td>27.82</td>
<td>82.79</td>
<td></td>
<td>Rosler et al. (1997), Neupane et al. (2019), Ojha et al. (2009), Quade et al. (1995)</td>
</tr>
<tr>
<td>Bakiya Khola</td>
<td>27.17</td>
<td>85.173</td>
<td></td>
<td>Quade et al. (1995), Harrison et al. (1993)</td>
</tr>
<tr>
<td>PCQ</td>
<td>-26.6</td>
<td>-66.9</td>
<td></td>
<td>Latorre et al. (1997), Butler et al. (1984)</td>
</tr>
</tbody>
</table>
Figure 5. a) Carbon isotopic shift in marine sediment cores (see Table 1 for locations and references). Values for δ\(^{13}\)C (normalized) are δ\(^{13}\)C - the mean value, plotted with more negative values up. Black bars to the left are ±0.5‰. X axis shows depth relative to the FO of *Amaurolithus* 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.

A change in vegetation from what is known as the “C\(_3\) photosynthetic pathway” plants using the C\(_3\) photosynthetic pathway to grasses using the C\(_4\) pathway (see e.g., Tipple and Pagani 2007). Quade et al. (1989) tentatively suggested that the shift could be related to an intensification of the Asian monsoon. Later, Cerling et al. (1993) reinterpreted the shift as resulting from a drop in atmospheric pCO\(_2\) which would favor C\(_4\) vegetation.

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

The approach of tying carbon isotopic data from continental sections to the GRTS has since been replicated elsewhere in Pakistan, India and Nepal as well as in Africa and South America. Latorre et al. (1997) and Singh et al. (2011) compiled data from continental sections, some of which were magnetostratigraphically calibrated. Their compilations suggested that the shift ranged in age from 4 Ma in North America to as early
as 8 Ma in East Africa, with the Asian data from the Siwaliks of India, Pakistan and Nepal ranging in age from 6 to 7.4 Ma. However, the calibration of the GRTS itself has evolved significantly over the nearly four decades of research on the topic. Moreover, the different carbon isotopic records have in some cases an ambiguous relationship to magnetostratigraphic calibration (e.g., the East African and North American data) making temporal inferences much more difficult.

What is of critical importance in assessing potential causes and effects of the $C_3$-$C_4$ transition is the relative timing of events on land (the expansion of $C_4$ grasses) compared to the marine records of carbon isotopes and to proxies for atmospheric $p$CO$_2$. These three different types of records have been dated using a variety of methods and the time scales have changed considerably over the years resulting in uncertainties of millions of years in the exact timing of the disparate records. In this paper we attempt to re-calibrate the different records in terms of a consistent time scale, which will enable us to assess the relative timing of the $C_4$ expansion, climatic drivers and carbon cycle responses. To do this, we first find those continental records that have a reasonable tie to magnetostratigraphic companion records and recalibrate them to a single time scale (here, the GTS12 of Gradstein et al. (2012)). In Section 3.1 we begin with a recalibration of the data from Pakistan, India and Nepal. In Section 3.2 we discuss the constraints from South America. Unfortunately, there are no results from North American Miocene with both soil carbonate isotopes and published magnetostratigraphic data and the magnetostratigraphies from Africa are similarly ambiguous in their relationship to the isotopic records. In Section 3.3, we turn to a new marine record of carbon isotopes from the Indus Fan (IODP Site U1457) that has both the biostratigraphic (including the FO of *Amaurolithus*) and magnetostratigraphic constraints. We then consider data from leaf waxes found by drilling on the African continental margin that also are tied the FO of *Amaurolithus* and the last occurrence of *Discoaster hamatus* (which has an age of $\sim$10.5 Ma.) Finally we discuss the implications of our newly calibrated isotopic shifts compared to atmospheric $p$CO$_2$ proxies and consider possible feed-backs in the system.

### 3 Revisiting the timing of the $C_3$-$C_4$ transition in continental settings

#### 3.1 Pakistan, India, Nepal

Quade et al. (1989) and Quade and Cerling (1995) reported carbon isotopic data from paleosol carbonate nodules from many sections from Pakistan in order to determine the age of the $C_3$-$C_4$ ecological transition: the Chinji-Nagri section of Johnson et al. (1985), the Kaulil Kas section of Tauxe and Opdyke (1982), the Mirpur section of Opdyke et al. (1979), the Jalalpur section of Johnson et al. (1982), the Pabbı Hills section of Opdyke et al. (1979) and Gabhir Kas or Johnson et al. (1982). These were all converted to ages using the GRTS of Berggren et al. (1985). A section studied initially by Quade et al. (1989) was resampled for paleomagnetic analysis in order to provide a tighter age constraint and the updated section was published by Behrensmeyer et al. (2007). Of the Pakistani sections, only two span the transition in a single (in some cases composite) section: the Rhotas (R) (composite) section of Behrensmeyer et al. (2007) (a combination of Dhabawal Kas and Basawa Kas sections with magnetostratigraphy from Opdyke et al. (1979) and Behrensmeyer et al. (2007)), and Jalalpur (JP) (Quade & Cerling, 1995) with magnetostratigraphy from Johnson et al. (1982). Kaulil Kas (KK) (Quade et al., 1995), with magnetostratigraphy from Tauxe and Opdyke (1982) has the onset of the transition and is also included in the present study.

Vögeli et al. (2017) compiled records from a variety of sections in NW India including the Jawalamukhti, Haripur Kolar, Jogindernagar, Kaming, Jammm Hills, the Parmandal-Utterbeni and Kangra sections. The magnetostratigraphy for Jogindernagar was based on unpublished data of Maithani and Burbank and the original data could not be located for the present study. The Kaming section has an excellent magnetostratigraphic con-
Figure 6. Magnetostratigraphic constraints for carbon isotopic data on the Indian subcontinent. 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.
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.

Isotopic data from Nepal were published by Quade et al. (1995) who investigated soil carbonates from the Surai Khola (SK) and Bakiya Khola (BK) sections in Nepal. More recently, Neupane et al. (2019) published new compound-specific data from leaf wax n-alkanes for Surai Khola. The Surai Khola data from Quade et al. (1995) and Neupane et al. (2019) have magnetostratigraphic constraints from Ojha et al. (2009) and Rösler et al. (1997) respectively and the Bakiya Khola isotopic data from Quade et al. (1995) have magnetostratigraphic data from Harrison et al. (1993). Quade et al. (1995) also presented data from Muksar Khola and Neupane et al. (2019) presented data from the Karnali River, but these sections do not show a shift in the carbon isotopes and in the case of Muksar Khola, they were not derived from pedogenic carbonate, the focus of the current study. Quade et al. (1995) also showed data from the Katari Khol section, but these
were only plotted versus stratigraphic height and we were unable to locate the original magnetostratigraphic data.

Most of the studies concerned with the C\textsubscript{3}-C\textsubscript{4} transition in Asia plotted the isotopic data in terms of age and the exact relationship to the supporting magnetostratigraphic age control can be obscure. Therefore in order to recalibrate the age for the isotopic data using a consistent time scale, we digitized the magnetostratigraphic and isotopic data, converting both to a common stratigraphic height scale in order to obtain age-height relationships using the GTS12 time scale of Gradstein et al. (2012). C\textsubscript{3} plants have a range of carbon isotopic values from -38 to -18 \%/o (mean -26.7 ± 2.3\%/o) and C\textsubscript{4} plants have values of -16 to -9 \%/o (mean -12.5 ± 1.1 \%/o; Cerling et al. (1997)) with corresponding boundaries in plant waxes (more depleted) and soil carbonates (more enriched). These can be used to place bounds on the stratigraphic position of the change in isotopic values, with the caveat that dry, open C\textsubscript{3} woodland can be indistinguishable from C\textsubscript{3} forest with some C\textsubscript{4} understory, thus there is always ambiguity in the interpretation of the first appearance of C\textsubscript{4} as appreciated in the discussion by Fox and Koch (2003), but often missed in presentations of C\textsubscript{4} %. Here the case is simplified as we are not looking to detect first appearance of C\textsubscript{4} or low proportions of C\textsubscript{4} in ecosystems, but rather focus on the shift to C\textsubscript{4} dominance and thus seek large positive isotope shifts that are unambiguous.

Our reconstructed magnetostratigraphic and isotopic data are shown in Figure 6. Using the information in the figure, bounds on the stratigraphic position of the isotopic shifts from C\textsubscript{3} dominated ecosystems to those dominated by C\textsubscript{4} vegetation can be converted to ages from GTS12 in a consistent manner.

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

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

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

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

In the NW Indian composite, the magnetic stratigraphy in the lower section (Jawalamukhi of Meigs et al. (1995)) is based on an undersampled section and the lab procedures followed the early protocol whereby extremely scattered within site directions were used, leading to the possibility of incorrect polarity assignments. The upper section at Haripur (Sangode et al., 1996) used a more sophisticated approach and reported within site statistics. While the lower part of the section had poor within site reproducibility in general, the upper part (relied upon here for the age model) can be considered "re-
liable". The isotopes of Vögel et al. (2017) were sampled much later than the magnetostratigraphic sample and plotted against age. We combined the magnetostratigraphic data with the isotopic data by using the age model of Vögel et al. (2017) to convert their ages back to stratigraphic height. Then, by pairing the inferred heights with the heights from the magnetostratigraphic studies, we attempted to recalibrate the ages for isotopic data. However, there is an added uncertainty in this process and condition 4 above was not met.

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

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

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

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

An alternative approach to calculating an age model for these sections would be to assume all four of the conditions above are met and interpolate between magnetostratigraphic tie points using a straight-line calculated between the bounds. This assumes that sediment accumulation is essentially linear between each bounding tie point and can result in abrupt changes in rate between adjacent segments. We recalculated all ages using this approach and found that the difference between the smoothly varying approach (polynomial assumption) and the piece-wise linear approach was ± ~100,000 years and much better than that near the interval of interest here (a few 10s of thousands of years). Given the additional uncertainties and assumptions associated with the second approach, we adopt the first, smoothly varying, approach in the following.

The first question we wish to address is whether there is a single age or age range for the isotopic transition that is consistent with all of the available data from the Indian sub-continent. We replot the data from Figures 6 and 7 as isotopes versus inferred age in Figure 8. In Figure 8b we plot the data from India and Pakistani sections (Kaulial Kas, Rhotas, Jawalamukhti/Haripur and Jalalpur) against the revised ages. The transition began perhaps as early as C4n (~7.8 Ma) as suggested by a single point in the Jalalpur section, but certainly by C3Bn (~7.2 Ma) in the Kaulial and Rhotas sections.
In Figure 8c we plot the data from Nepal (Surai and Bakiya Kholo). The transition recorded in Nepal started later than in Pakistan and India, perhaps as young as 6 Ma as recorded in Surai Kholo. This diachronocity within a single subcontinent could argue against pCO$_2$ as the sole or even dominant driver of the process, although tipping points may differ across climatic regimes (Higgins & Scheiter, 2012). For example, the western side of the subcontinent (Pakistan) is considerably drier and thus may pass climatic thresholds that promote C$_4$ grasslands to replace forests earlier than the eastern side (Nepal). This observation is also supported by the modeling efforts of (Zhou et al., 2018) (Figure 1) which show considerable regional variability within a constant atmospheric pCO$_2$ 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 Kholo 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.

3.2 Other continental sections

To place the data from the Indian subcontinent into a global context, we look first to other records with good magnetostratigraphic control. While there are studies from Chinese Loess Plateau (e.g., Dong et al. (2018)), these have not yet been placed in a magnetostratigraphic context. In any case, the study focused on the Mid-Miocene Climatic Optimum (much earlier than the focus of this paper) and demonstrated that there may have been a slight increase in the percentage of C$_4$ plants at around 15 Ma as two data points are close to a 50% C$_4$ fraction.
Ségalen et al. (2007) cited data from Africa but those do not show the clear transition to C\textsubscript{4} values and the data are not clearly tied to the available magnetostratigraphic sections (which are in an unpublished master’s thesis and do not stand up to scrutiny). Uno et al. (2011) published isotopic data from mammal teeth but the magnetostratigraphic data (as opposed to the interpretations) were apparently also never published. Similarly, Morgan et al. (1994) present data from mammal teeth from the Tugen Hills section (near the sections studied by Tauxe et al. (1985), but include no information about the relationship between their fossil localities and any magnetostratigraphic constraints.

North America appears to have no magnetostratigraphically calibrated isotopic data either. However, Tipple and Pagani (2010) published isotopic data from DSDP Site 94 from the Gulf of Mexico. Unfortunately, the latter section lacks the FO of \textit{A. primus} with which to tie the record to the present study. Moreover, it has only weak support for the onset of C\textsubscript{4} vegetation in the Miocene. Similarly, Chen et al. (2015) sampled sections in Montana relying on an age of 10.42 Ma for an ash bed reported by (Re tallack, 2007) who in turn cited an unpublished field guide. The isotopes (which show a dominance of C\textsubscript{3} grasses throughout the section) are thought to range from ∼ 9 to ∼ 10.4 Ma. Finally, Fox and Koch (2003) published a set of soil carbonate isotopic data calibrated with the North American Land Mammal Ages. This data set suggests that by the Pliocene, the vegetation in western North America was dominated by C\textsubscript{4} vegetation.

The picture in South America, is much better constrained than for North America. Latorre et al. (1997) published isotopic data from soil carbonates from the Puerta de Corral Queulado (PCQ) section in Argentina which has excellent magnetostratigraphy control from Butler et al. (1984), which we show in Figure S8. The increasing abundance of C\textsubscript{4} plants is suggested by δ\textsubscript{13}C values higher than -7.5 ‰ and the transition is well constrained to be younger than 4 Ma. This result agrees with other sections from Argentina that unfortunately lack magnetostratigraphic constraints (Kleinert & Strecker, 2001). The results from Argentina are similar to those of Andrae et al. (2018) who found a Pliocene onset for the C\textsubscript{3}-C\textsubscript{4} transition as recorded in leaf wax data found in samples from ODP Site 763A off NW Australia. Therefore the C\textsubscript{3}-C\textsubscript{4} transition in terrestrial sequences cannot be globally synchronous, in agreement with predictions by the modeling efforts of, for example, Zhou et al. (2018) (see Figure 1).

### 3.3 U1457: A Rosetta stone for carbon isotopes

Returning to the paleoceanographic carbon isotopic shift first noted by Keigwin (1979), the question arises as to how the transition in the Indian subcontinent relates to the paleoceanographically observed carbon shift. Until now, deep sea records of the carbon shift have been tied to the first occurence (FO) of \textit{Amaurolithus spp}. The age of \textit{Amaurolithus spp.} in GTS12 is stated to be 7.4 Ma. However, it was found in two Ocean Drilling Program (ODP) holes: 844B and 710B. The FO in 844B was found at 844B-5H-5:29 cm which, according to Schneider (1995), is in the middle of C3Br.2r or about 7.3 Ma using the timescale of Gradstein et al. (2012). It was also found in 710B-7H-5.30/7H-4, 130 or 60.5 mbsf, an interval between C3Ar (y) and C4n (y). Assuming a linear sedimentation rate gives an approximate age for this datum of about 7.35 Ma. We adopt the former age of 7.3 Ma here as it is better constrained. What is required to tie the continental records to the marine records is a core in which the isotopic shift and the FO of \textit{Amaurolithus spp}. both occur in a magnetostratigraphic context. IODP Site U1457 provides such an opportunity.

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

<table>
<thead>
<tr>
<th>Datum</th>
<th>Type</th>
<th>Event</th>
<th>Age (Ma)</th>
<th>Max. (m)</th>
<th>Min. (m)</th>
<th>Midpoint (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>24</td>
<td>CN</td>
<td>T Sphenolithus spp.</td>
<td>3.540</td>
<td>517.45</td>
<td>513.09</td>
<td>515.27</td>
</tr>
<tr>
<td>26</td>
<td>CN</td>
<td>B Discoaster tamalis</td>
<td>4.130</td>
<td>539.15</td>
<td>539.65</td>
<td>539.40</td>
</tr>
<tr>
<td>28</td>
<td>CN</td>
<td>T Discoaster quinqueramus</td>
<td>5.590</td>
<td>539.65</td>
<td>539.15</td>
<td>539.40</td>
</tr>
<tr>
<td>29</td>
<td>PF</td>
<td>T Globorquadrina dehiscens</td>
<td>5.920</td>
<td>526.64</td>
<td>513.09</td>
<td>519.87</td>
</tr>
<tr>
<td>30</td>
<td>CN</td>
<td>T Nicklithus amplificus</td>
<td>5.940</td>
<td>610.36</td>
<td>610.05</td>
<td>610.21</td>
</tr>
<tr>
<td>32</td>
<td>MR</td>
<td>C3Ar</td>
<td>6.733</td>
<td>624.23</td>
<td>625.42</td>
<td>624.83</td>
</tr>
<tr>
<td>33</td>
<td>PF</td>
<td>B Pulleniatina primalis</td>
<td>6.600</td>
<td>615.50</td>
<td>621.36</td>
<td>618.43</td>
</tr>
<tr>
<td>35</td>
<td>CN</td>
<td>B Nicklithus amplificus</td>
<td>6.910</td>
<td>626.34</td>
<td>629.53</td>
<td>628.94</td>
</tr>
<tr>
<td>36</td>
<td>MR</td>
<td>C3Br.2r</td>
<td>7.285</td>
<td>643.04</td>
<td>644.16</td>
<td>643.60</td>
</tr>
<tr>
<td>38</td>
<td>CN</td>
<td>B Amaurolithus spp.</td>
<td>7.300</td>
<td>645.05</td>
<td>644.76</td>
<td>644.91</td>
</tr>
<tr>
<td>39</td>
<td>MR</td>
<td>C3Br.2n</td>
<td>7.454</td>
<td>662.92</td>
<td>664.92</td>
<td>663.92</td>
</tr>
<tr>
<td>41</td>
<td>MR</td>
<td>C4n.1r</td>
<td>7.642</td>
<td>674.47</td>
<td>675.62</td>
<td>675.05</td>
</tr>
<tr>
<td>42</td>
<td>CN</td>
<td>B Discoaster quinqueramus</td>
<td>8.120</td>
<td>832.85</td>
<td>845.62</td>
<td>839.24</td>
</tr>
<tr>
<td>43</td>
<td>CN</td>
<td>T Minylitha convallis</td>
<td>8.680</td>
<td>845.62</td>
<td>856.50</td>
<td>851.06</td>
</tr>
<tr>
<td>44</td>
<td>CN</td>
<td>T Discoaster bohlii</td>
<td>9.690</td>
<td>864.64</td>
<td>865.62</td>
<td>865.57</td>
</tr>
<tr>
<td>45</td>
<td>CN</td>
<td>T Catinaster coalitus</td>
<td>9.790</td>
<td>864.18</td>
<td>866.20</td>
<td>865.19</td>
</tr>
<tr>
<td>46</td>
<td>MR</td>
<td>C5n</td>
<td>9.830</td>
<td>885.02</td>
<td>894.30</td>
<td>889.66</td>
</tr>
<tr>
<td>47</td>
<td>PF</td>
<td>B Neogloboquadrina acostaensis</td>
<td>10.400</td>
<td>1001.08</td>
<td>1005.11</td>
<td>1003.10</td>
</tr>
<tr>
<td>48</td>
<td>CN</td>
<td>B Discoaster bellus</td>
<td>10.890</td>
<td>1001.08</td>
<td>1005.11</td>
<td>1003.10</td>
</tr>
<tr>
<td>50</td>
<td>CN</td>
<td>Absence of Fasciculithus spp.</td>
<td>62.130</td>
<td>1068.63</td>
<td>1067.35</td>
<td>1067.99</td>
</tr>
</tbody>
</table>

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

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

Using the age model shown in Figure 9c, we plot the magnetostratigraphic and isotopic data for Unit 4b in Figure 10. The C3-C4 transition occurs just above the FO of Amaurolithus spp and within a normal interval interpreted here as C3Br.2n or at about 7.2 Ma. This is within the age interval for the transition in India and Pakistan estimated 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.
The carbon isotopes were measured on leaf waxes whose inputs to the hemipelagic units are interpreted as being wind-transported based on a lack of associated lignin (both are present in the fluvially-export carried in turbidic units Feakins et al. (This volume). The wind-blown waxes are thought to derive from peninsular India based on the proximity of the continent, the dominant easterly winds in October-to-January and modern evidence from coretops across the Arabian Sea (Dahl et al., 2005).

Figure 11. Leaf wax $\delta^{13}$C C$_{35}$ n-alkane (a-c) and d) C$_{30}$ 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

As noted earlier, data from continental sections in Africa are insufficient to constrain the C$_3$-C$_4$ transition. However, analysis of leaf waxes found in DSDP and ODP cores, in connection with the FO of Amaurolithus spp can provide some clues. We plot the data from leaf wax $\delta^{13}$C C$_{35}$ n-alkanes from Uno et al. (2016) and Polissar et al. (2019) and $\delta^{13}$C C$_{30}$ n-alkanoic acids from Feakins et al. (2013) in Figure 11 against depth. We also show the FO of Amaurolithus spp and the last occurrences (LO) of Discoaster hamatus with an age estimated at ~10.5 Ma (Gradstein et al., 2012). These data suggest that the change in vegetation from C$_3$ dominated ecosystems to those dominated by C$_4$ occurred between these two tie points, sometime after about 10 Ma as suggested by (Polissar et al., 2019). These are the earliest ages for the transition that we know of and are consistent with the model of (Zhou et al., 2018) (Figure 1) whereby eastern Africa is one of the first places predicted to favor C$_4$ vegetation.
4 Discussion

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

Raymo and Ruddiman (1992) made the case that uplift of the Himalayan region and the Tibetan Plateau could have resulted in higher chemical weathering which in turn could have resulted in a drawdown of atmospheric CO$_2$. While the Himalayas are broadly believed to have been at high elevations much earlier than the shifts examined here, there are some signs of uplift in the late Miocene. Harrison and Yin (2004) suggested a major phase of uplift at around 9 Ma and Molnar et al. (1993) interpret the onset of normal faulting in Tibet at around 8 Ma as evidence of an uplift of the plateau. Interestingly, Tremblay et al. (2015) call for decreased erosion in southern Tibet caused by enhanced uplift at about 10 Ma, an effect opposite to the mechanism envisioned by Raymo and Ruddiman.

Changes in the carbon budget may also arise from often overlooked adjustments in the global organic carbon cycle and the case has been made that enhanced organic carbon burial in the Bengal Fan could outweigh any changes in silicate weathering (Derry & France-Lanord, 1996). It is also possible that the rise of C$_4$ grasslands led to a change in the carbon cycle with attendant climate feedbacks. A large-scale change in ecology could shift the balance of carbon storage as forests have generally thin soils whereas grasslands can build deep soils. Limited evidence from experimental farms in Minnesota, (Fornara & Tilman, 2008) estimated that C$_4$ vegetation pulls down 193% more carbon into the soil than C$_3$ vegetation. More recently, Spiesman et al. (2018) found that, depending on the quality of the soil, a higher proportion of C$_4$ grasses relative to C$_3$ grasses can indeed enhance carbon storage; they attribute this to the higher efficiencies in nitrogen and water usage by the C$_4$ photosynthetic pathway.

Even if only part of the Indian sub-continent were covered by C$_4$ grasses, and the resulting paleosols buried (as they were in the Siwaliks), this could provide a positive feedback as lower pCO$_2$ would in turn favor the C$_4$ grasses. Whatever the mechanism, the current array of pCO$_2$ reconstructions do leave open the possibility of a late Miocene pCO$_2$ drop that would have enhanced the viability of C$_4$ plants.

4.1 Conclusions

In this study we reconsider the timing of the shift to C$_4$ dominance in ecosystems around the world. We have been able to realign records where the following conditions are met:

1. Marine data include the first appearance of *Amaurolithus* spp.
2. Magnetostratigraphic records are provided including the depth in section of the chronostratigraphic markers in order for alignment as chronostratigraphy evolves.
We emphasize that making the age model data available alongside new carbon isotope records is particularly valuable for ensuring that updated comparisons can be made as chronologies evolve. Some regions that are missing from the present comparison are on land in North America where magnetostratigraphic age control would greatly improve interpretations.

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

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

Acknowledgments

This research was supported in part by the US National Science Foundation (EAR-1547263 and Consortium for Ocean Leadership, sub-award GG0093093-01 to LT; OCE 14-50528 and to Consortium for Ocean Leadership, sub-award GG0093093-01 to S.F.). This research used samples collected by the International Ocean Discovery Program (and earlier programs), supported by funding from the US National Science Foundation and other member nations. We thank all participants of the shipboard science party and crew on Expedition 355. We thank Christeanne Santos who performed some of the shorebased paleomagnetic analyses. The terrestrial records were compiled from the literature, supplemented by data made available by Jay Quade, Anna K. Behrensmeyer, John Barry and Prabhat Neupane. We thank them for providing data associated with their publications enabling this re-evaluation. We are also very grateful for the comments of Jay Quade and the Editor and Associate Editor of Paleoceanography & Paleoclimate as well as one anonymous reviewer whose efforts greatly improved the manuscript. All magnetostratigraphic and carbon isotopic data used in this study will be available from the MagIC database at https://earthref.org/MagIC/DOI/10.1029/2020PA003857 upon acceptance of this article. For the purposes of review, the link to the private workspace that contains the data is: https://earthref.org/MagIC/16737/ec119793-abd4-41aa-8884-28b5fe0d0f09
References


5 Supplemental Information

Figure S1

Details for sections in Figure 6. See Table 1 for data sources. left: Magnetic polarity time scale of Gradstein et al. (2012). middle: Virtual Geomagnetic Pole (VGP) positions for sites in the Kaulial Kas Section plotted against stratigraphic height. Those in parentheses were added or modified in this paper; those without are as in the original publication. Solid green lines are the Chron boundaries as identified to the right. c) Carbon isotopes from paleosol carbonate nodules. Dotted lines are C₃ and C₄ end-
members for soil carbonates. C$_3$-C$_4$ transition boundary (boundaries) as dashed cyan horizontal line(s).

Figure S2

Same as Figure S1 but for the Rhotas Section.
Same as Figure S1 but for the Jawalamukhi/Haripur Sections.
Same as Figure S1 but for the Jalalpur Section.
Same as Figure S1 but for the Surai Khola Section. Middle panel is declination (not VGP). Isotopic data are C$_{27}$ $n$-alkane data. Dotted lines are the limits of C$_3$, C$_4$ for plant waxes.
Same as Figure S1 but for the alternate Surai Khola Section of Ojha et al. (2009) and Quade et al. (1995). Middle panel is declination (not VGP).
Same as Figure S1 but for the Bakiya Khola Section. Middle panel is declination (not VGP).
Same as Figure S1 but for the Puerta de Corral Quermado (PCQ) Section.
Figure 1.
600 ppm

C3 dominant C3 + forest C3=C4 C3>C4 C4>C3 C4 dominant
Figure 2.
Figure 3.
Benthic δ¹³C ‰

Planktonic δ¹³C ‰

LMCIS

Age (Ma)

∆¹³C ‰

pCO₂ decrease
Figure 4.
Figure 5.
Figure 7.
Figure 8.
a) GTS12

b) Pakistan/India

c) Nepal

- Age (Ma): GTS12
- δ¹³C
- R, KK, JH, JP, BK
Figure 9.
Figure 10.
Figure 11.
δ¹³C (C₃₅ n-alkane)

a) ODP 659

b) ODP 959

c) DSDP 235

d) DSDP 231
Figure S1.
Figure S3.
Figure S6.
Figure S7.