

Author's Response to Reviews of "Pliocene expansion of C₄ vegetation in the core monsoon zone on the Indian Peninsula" by Ann G. Dunlea et al.

Response to Editor Comments

Editor Decision: Reconsider after major revisions (05 Aug 2020) by [Alberto Reyes](#)
Comments to the Author:

Dear Dr. Dunlea,

Thank you for submitting responses to the comments and suggestions of the two reviewers. Both reviewers are positive about the dataset and the underlying science questions. There is also broad consensus from the reviewers that the presentation of methods and discussion can be bulked up, either through additional text or re-organization. Regarding Rev. 2's comment about the age model, I think it should be possible to provide the requested information on age model construction while referring readers to the published source(s) for full details on the tie-points used in your age model.

Your responses indicate that you are willing to undertake these revisions, so I invite you to submit a revised manuscript for further consideration. The revised manuscript will go through another stage of peer-review, which requires that I indicate "major revisions".

Sincerely,
Alberto Reyes (handling editor)

- [Dear Dr. Alberto Reyes,](#)

[Thank you for your comments on our manuscript. Below you can find a point-by-point response to the reviewer's comments and the corresponding revised manuscript with changes tracked. Substantial revisions of the manuscript draft have been made, including an expansion of the discussion section and other edits raised by reviewers. Please let me know if you have any questions regarding the revisions.](#)

[Sincerely,](#)
[Ann Dunlea](#)

Response to Anonymous Referee #1

Dunlea, Giosan and Huang use lipid plant wax biomarkers and their isotope composition to reconstruct the climate and vegetation of the eastern Indian peninsula over the last 6 million year. They find that C₄ vegetation was already

present in the late Miocene and further expanded during the Pliocene. They argue that this expansion was likely caused by changing precipitation patterns during the studied time period as well as a decline in atmospheric CO₂ during the Pliocene. The results are consistent with previous reconstructions from the region that yielded similar results. The manuscript is overall well written. As outlined below there are however shortcomings regarding the description of the methodology that should be addressed prior to its publication and several points where the authors should expand the manuscript and provide further information.

- We thank the anonymous referee for their comments and revisions.

Major comments: The authors should consider moving the method description from the appendix to the main text to have it in chronological order. In the journal format of *Climate of the Past* there is no reason to put the methodology at the end.

- The methods section has been moved from the appendix to the main text.

Lines 82-85: As is, it is confusing to see sample pairs mentioned without proper explanation. I would suggest do give a detailed description of the core and the reasoning behind the sampling strategy beforehand to avoid any confusion.

- A better description of the sampling pairs was added to the methods.

In Fig. 2C and lines 113-115 it is mentioned that the δD values are corrected for physiological effects of C₃ and C₄ photosynthesis. Unfortunately, there is no description available of how this correction was conducted. This should be added in order to be able to reproduce the calculations and presented results.

- We added the explanation of the correction to the methods section.

In lines 147-153, the Pliocene C₄ expansion is explained by a lowering in atmospheric CO₂. It would be useful to show the CO₂ and C₄ vegetation trends together in a figure to illustrate this point.

- Estimates of atmospheric CO₂ have been added to Figure 3.

In the description of the compound-specific isotope analysis it is mentioned that the fatty acids were methylated prior to analysis (i.e. a methyl group added). Since this methyl group changes the isotope composition of the resulting fatty acid methyl esters, the measurements need to be corrected using the isotope composition of the methanol used in the reaction. Without the proper correction the absolute values and associated interpretations are incorrect.

- The correction was performed and we added a description of the correction to the methods section.

Minor comments: Line 27: Add some references backing up the sentence ending at the beginning of this line.

- The following references were added to support the statement: An et al., 2005; Behrensmeyer et al., 2007; Huang et al., 2007; Edwards et al., 2010; Zhou et al., 2014

Line 47: Specify what kind of model was used in the cited study.

- We added details about the type of model used to predict natural flora was added. The reference cited has additional information.

Lines 92-94: Why are the mid-Pliocene (3-5 Myr) and mid-Pleistocene (1.5 Myr) selected in this description. On the figure, the trend in dD seems to be pretty constant and the selection of these time points seems rather arbitrary.

- We selected those time intervals to be consistent with the time intervals we used for the $\delta^{13}\text{C}$ plots. We added text to emphasize the change in δD is gradually increasing and we only remark on the time interval to compare with the $\delta^{13}\text{C}$ record.

Lines 113-114: This sentence on physiological effects of C3 versus C4 plants on dD is not really connected to the previous sentence on air mass mixing. It is therefore confusing to see the word thus at the beginning of this sentence.

- The sentence was edited for better logical flow and includes reference to the methods section that explains the correction for plant physiology.

Line 182: The unit cm³ already implies volume. The word volume after the unit is therefore redundant and can be deleted.

- The redundancy was removed.

Line 210: Provide the isotope composition of the methanol used.

- We have added the isotopic composition of the methanol used and the equations used to correct the data for the addition of methanol.

Lines 225: Specify the standard used. Was it an industry standard with known isotope values?

- Done. It was an in-house lab standard with known values.

Figures: In the text the abbreviation for million years is Myr, while in the figures Ma is used. This should be homogenized.

- We are using “Myr” for “million years”, and “Ma” for “million years ago”, as the USGS does. “Ma” thus stands for an event in the past whereas “Myr” indicates duration or time interval. The text was checked for consistency of this notation.

In the method description, alkenone and alkane measurements are described which are however not mentioned in the rest of the manuscript. Of course, it would be interesting to see these results. So, the authors should either remove reference to these measurements or include them in the manuscripts.

- We removed the reference to the alkenone and alkane measurements.

Supplementary tables S1-S3: In order to facilitate the use of data by other scientists, consider moving the data contained in the tables to a separate file that is in a machine- readable format.

- Yes, we will submit the final tables as .xls or .csv files.

Response to Anonymous Referee #2

Dunlea et al. evaluate new organic geochemical data from the Bay of Bengal (Indian Ocean) in order to investigate changes in terrestrial biomes over the past 6 Ma. Specifically, they carry out: (i) geochemical analyses of major, trace and rare elements on sediments samples from IODP Site U1445 to determine the sediment provenance; (ii) $\delta^{13}\text{C}$ and $\delta^2\text{H}$ analysis on leaf-wax fatty acids (C₃₀) to reconstruct the evolution of C₄ plants since the late Miocene. The authors suggest that the sediment originates from the Mahanadi River in core monsoon area of the Indian Peninsula, and hence the biomarker data allow to better understand the timing of the C₄ plant expansion during the Late Cenozoic in this region. They conclude that although C₄ plants have been growing on the Indian Peninsula already during the late Miocene, they expanded strongly from the mid-Pliocene (at c. 3.5 Ma) onwards in agreement with previous observations from East Africa and NW Australia.

The authors have produced a nice dataset that merits publication in *Climate of the Past* in principle. In its current form, however, the manuscript is poorly structured with most of the important information provided in the supplementary files. It also lacks an in-depth discussion of the results, particularly comparisons with other existing records from both marine and terrestrial settings and an assessment of the potential mechanisms behind the expansion of C₄ plants during the Pliocene. These are dealt with in such a superficial detail that an interested reader from the broad audience of *Climate of the Past* who knows little about C₄ plants and their expansion in the Late Cenozoic, would struggle to follow the arguments. If the authors are interested in greatly expanding the manuscript then I would support acceptance after rewriting.

- We thank the reviewer for their comments and suggested revisions. Substantial portions of the discussion have been re-written.

When revising their manuscript, the authors should carefully address the following points:

1. More information on the age model development is needed. How reliable are the magneto- and biostratigraphic tie points used? Were the turbidite layers removed before developing the age model? What are the sedimentation rates, and how do they change through time? It is difficult to imagine that the sedimentation rates stay ‘fairly constant’ for a such a long time as the authors argue in line 68.

- We added the average sedimentation rate with uncertainty to the main text. We agree that the age model is a critical part of any paleoceanographic study, but the age points we use are published and improving them is not the focus of this study. The line we fit to the previously published age constraints was merely to interpolate the ages to our samples specifically. While we agree that there may have been shorter-term changes in sedimentation rates, Supplemental Figure S1 demonstrates that there are no major or abrupt changes in sedimentation rate over the million year timescales of interest to this study.

2. Please explain what you mean with higher latitudes and elevations in line 152. Reconstructions of C3/C4 vegetation in the Chinese Loess Plateau (An et al. 2005) and palynological records from the Tibetan Plateau (Koutsodendris et al. 2019) – which are arguably from higher latitudes and elevations than the study area – show expansion of C4 plants and arid semi-desert biomes, respectively, during the mid-Pliocene; hence the argument that ecosystems at higher latitudes and elevations remained stable is not correct. By extension, the interpretation that tropical ecosystems adjacent to the Indian Ocean are more sensitive and the CO₂ change is likely not the primary driver of the Pliocene C4 expansion is not fully substantiated.

- Yes, we have clarified this point. We were saying that the Indian Peninsula is more sensitive to monsoon changes than the relatively close records in the India, Pakistan, and the Himalayan regions. We added the Pliocene C4 patterns in Asia to the discussion.

3. The early Pleistocene interval (c. 2 - 1 Ma) is characterized by lower ¹³C values suggesting contraction of C4 vegetation in the study area. Please elaborate on this issue in a revised manuscript. Is a similar pattern also observed in other records? What kind of mechanism could be responsible?

- We added a paragraph discussing the change in the early Pleistocene.

4. The discussion on the global patterns of C4 expansion should be substantially expanded. Please also consider including recently published biomarker data from the western Indian Ocean (e.g., Pollisar et al., 2019) and also comparing the data from Site U1445 with palynological records from adjacent regions to the Indian Ocean (e.g., Miao et al., 2017; Koutsodendris et al., 2019) that also span the time interval from the mid-Miocene to Pleistocene.

- Thank you for these additional references. We included them in the expanded discussion.

5. The influence of precipitation as a trigger for the C4 expansion during the mid-Pliocene is also poorly explained (lines 154-155). The authors simply list several climate components affecting the precipitation variability in the Indian Ocean today without however explaining how they may have influenced the hydroclimate during the mid-Pliocene. They should at least elaborate on whether these climate systems were active during this time interval based on proxy records and model studies, and suggest specific mechanisms responsible for the hydroclimate, and in turn, vegetation dynamics in the study region.

- The explanation of the climate system and possible mechanisms has been expanded.

6. The quality of plots is generally poor and it is difficult to evaluate the proxy records (a prime example is the $\delta^{13}C$ record from Site 231 in Fig. 3c). Please redraw the figures to increase clarity.

- We have revised the plots to increase aesthetic value and clarity.

References: An, Z., Huang, Y., Liu, W., Guo, Z., Clemens, S., Li, L., Prell, W., Ning, Y., Cai, Y., Zhou, W., Lin, B., Zhang, Q., Cao, Y., Qiang, X., Chang, H., Wu, Z., 2005. Multiple expansions of C4 plant biomass in East Asia since 7 Ma coupled with strengthened monsoon circulation. *Geology* 33, 705.

Koutsodendris, A., Allstädt, F.J., Kern, O.A., Kousis, I., Schwarz, F., Vannacci, M., Woutersen, A., Appel, E., Berke, M.A., Fang, X., Friedrich, O., Hoorn, C., Salzmann, U., Pross, J., 2019. Late Pliocene vegetation turnover on the NE Tibetan Plateau (Central Asia) triggered by early Northern Hemisphere glaciation. *Global and Planetary Change* 180, 117-125.

Miao, Y., Warny, S., Clift, P.D., Liu, C., Gregory, M., 2017. Evidence of continuous Asian summer monsoon weakening as a response to global cooling over the last 8 Ma. *Gondwana Research* 52, 48-58.

Polissar, P.J., Rose, C., Uno, K.T., Phelps, S. R., deMenocal, P., 2019. Synchronous rise of African C4 ecosystems 10 million years ago in the absence of aridification. *Nature Geoscience*, 12, 657-660.

Pliocene expansion of C₄ vegetation in the core monsoon zone on the Indian Peninsula

Ann G. Dunlea¹, Liviu Giosan², Yongsong Huang³

¹Marine Chemistry & Geochemistry, Woods Hole Oceanographic Institution, Woods Hole, MA, 02543, USA

²Geology & Geophysics, Woods Hole Oceanographic Institution, Woods Hole, MA, 02543, USA

³Department of Earth, Environmental, and Planetary Sciences, Brown University, Providence, RI, 02912, USA

Correspondence to: Ann G. Dunlea (adunlea@whoi.edu)

Abstract. The expansion of C₄ vegetation during the Neogene was one of the largest reorganizations of Earth’s terrestrial biome. Once thought to be globally synchronous in the late Miocene, site-specific studies have revealed differences in the timing of the expansion and suggest that local conditions play a substantial role. Here, we examine the expansion of C₄ vegetation on the Indian Peninsula since the late Miocene by constructing a ~6 million year paleorecord with marine sediment from the Bay of Bengal at Site U1445 drilled during International Ocean Discovery Program Expedition 353. Analyses of element concentrations indicate the marine sediment originates from the Mahanadi River in the Core Monsoon Zone (CMZ) of the Indian Peninsula. Hydrogen isotopes of the fatty acids of leaf waxes reveal an overall decrease in the CMZ precipitation since the late Miocene. Carbon isotopes of the leaf wax fatty acids suggest C₄ vegetation on the Indian Peninsula existed before the end of the Miocene, but expanded to even higher abundances during the mid-Pliocene to mid-Pleistocene (~3.5 to 1.5 million years ago). Similar to the CMZ on the Indian Peninsula, a Pliocene expansion or re-expansion has previously been observed in northwest Australia and in East Africa, suggesting that these tropical ecosystems surrounding the Indian Ocean remained highly sensitive to changes in hydroclimate after the initial spread of C₄ plants in late Miocene.

1. Introduction

The expansion of plants using the C₄ photosynthetic pathway is one of the most dramatic reorganizations of the global biome during the Neogene. A widespread late-Miocene expansion (8 to 6 Ma) is well documented and many studies have interpreted the broadly synchronous timing as ecosystems adapting to decreasing pCO₂ (e.g., Ehleringer et al., 1991; Ehleringer and Cerling, 1995; Cerling et al., 1993, 1997; Herbert et al., 2016). However, an increasing number of studies have shown that the timing, regional patterns, rate and drivers of C₄ grassland expansion were much more diverse and complex (An et al., 2005; Behrensmeyer et al., 2007; Huang et al., 2007; Edwards et al., 2010; Zhou et al., 2014). Along with low pCO₂, a C₄ photosynthetic pathway is better adapted to higher temperature, aridity, seasonality, and during disturbances such as flood, droughts, and fires (e.g., Edwards et al., 2010, and references therein). The interplay of these parameters varies amongst

Deleted: (

Deleted: Ma

Deleted: climate

Deleted: .

regions. Resolving the precise timing and factors leading to major changes in vegetation demands site-specific studies
35 (Strömberg et al., 2011; Zhou et al., 2014).

Our study provides a novel piece of the puzzle in unraveling the complexities of C₄ expansion by constructing a 6
million year (Myr) record of C₄ vegetation and aridity on the Indian Peninsula. The marine sediment record is from
International Ocean Discovery Program (IODP) Site U1445 (17°44.72'N, 84°47.25'E; 2,503m water depth; Fig. 1A) drilled
in the Bay of Bengal (BoB) close to the mouths of the Mahanadi River. Lithologies at Site U1445 include calcareous fossils,
40 biosilica, silt, and clays (including glauconite), and are overall described as hemipelagic sediment (Clemens et al., 2016). The
Indian Monsoon dictates climate patterns in the Mahanadi River drainage basin: rainy summers, dry winters, and an annual
reversal of wind direction (Gadgil, 2003; Sarkar et al., 2015). Highly sensitive to the seasonal changes, more than 80% of
runoff from the Mahanadi River into the BoB occurs during the summer (Chakrapani and Subramanian, 1990).

Previous reconstructions of Neogene C₄ expansion in regions affected by the Indian Monsoon use deposits originating
45 in the Himalayas and their piedmont regions (France-Lanord and Derry, 1994; Quade and Cerling, 1995; Quade et al., 1995;
Cerling et al., 1997; Freeman and Colarusso, 2001; Sanyal et al., 2004; Behrensmeyer et al., 2007; Galy et al., 2010; Ghosh et
al., 2017). The Mahanadi River drains a relatively low-elevation region of the Indian Peninsula distinct from the nearby
mountain ranges (e.g., the Western Ghats, the Himalaya, Indo-Burman ranges Fig. 1, Xie et al., 2006). With minimal
orographic precipitation in the Mahanadi River basin, rainfall in this “Core Monsoon Zone” (CMZ) represents the mean
50 behavior of the Indian Monsoon (Fig. 1; Ponton et al., 2012; Sarkar et al., 2015; Giosan et al., 2017, and references therein).

Although agriculture dominates present-day vegetation, models of seasonal climate predict the natural flora of the
Mahanadi basin would be closed-canopy, moist deciduous forests and moist-to-dry woodlands with rare open spaces (Fig. 1C,
Zorzi et al., 2015 and references therein). Today the region encompasses a range of C₃ and C₄ vegetation, but proxies and
models suggest that the plant communities are highly sensitive to glacial-interglacial changes with nearly all flora utilizing a
55 C₄ pathway during the last glacial maximum (Galy et al., 2008; Phillips et al., 2014; Zorzi et al., 2015, and references therein).
The behavior of vegetation in the CMZ over million-year timescales is unknown.

Here, we use inorganic bulk geochemical analyses to fingerprint the origin of sediment at Site U1445 to be from the
Mahanadi River. Then we use bulk organic and compound-specific biomarkers at the same site, including carbon and hydrogen
isotope measurements of leaf wax fatty acids, to reconstruct the changes in C₄ vegetation and rainfall in the CMZ of the Indian
60 Peninsula over the last ~6 Myr (Fig. 2).

2. Methods

Over million-year timescales, Site U1445 had a constant sedimentation rate (~115 ± 15 m/Myr; Clemens et al., 2016).
We fit a locally weighted spline to biostratigraphic and magnetostratigraphic age constraints from Hole U1445A (Clemens et
al., 2016) using CLAM software in R (Blaauw, 2010) to estimate the age of our samples (Fig. S1). Our samples and the age
65 constraints were from the same hole. Uncertainty on the ages is estimated to be less than ±0.2 Myr. Shipboard scientists

Deleted: We constructed an age model for

Formatted: English (UK)

Moved (insertion) [1]

Formatted: English (UK)

Deleted: using CLAM software in R (Blaauw, 2010) to

Deleted: ages

Deleted: ;

Deleted: collected

Deleted: in which the age model was constructed. We measured
major, trace, and rare earth element concentrations

Deleted: 30 bulk sediment samples spanning 0-6 Ma

Deleted: determine sediment provenance (Dunlea et al., 2015;
Appendix A). Then we analyzed bulk organics (Appendix B) as well
as compound-specific biomarkers (Appendix C) from 57 samples to
reconstruct hydrological and vegetation changes. Samples for
biomarker analysis were collected from Site U1445 in pairs, visually
targeting relatively light and dark layers at similar depths to capture
the variability range on shorter timescales while characterizing
longer trends.

Moved down [2]: 3. Results

Deleted: --The age model constructed at Hole U1445A suggests
that long-term sedimentation rates have been overall continuous and
fairly constant (Fig. S1).

observed thin turbiditic sequences (~2-20cm thick) throughout Site U1445 and the expansion and dissociation of gas hydrates upon recovery that may muddle a higher-resolution record (Clemens et al., 2016). However, Site U1445 has fewer and smaller turbidite deposits relative to other sites drilled in this region and the records spanning million-year timescales are likely relatively undisturbed.

To determine sediment provenance, we measured major, trace, and rare earth element concentrations on 30 bulk sediment samples spanning 0 to 6 Myr including light and dark layers of sediment (Sec. 2.1.). To reconstruct hydrological and vegetation changes, we analyzed bulk organics (Sec. 2.2.) as well as compound-specific biomarkers (Sec. 2.3.) from 57 samples. We constructed the sampling plan to characterize the differences in organic content and isotope composition between the lighter and darker layers of sediment. As such, samples for organic and biomarker analysis were collected from Site U1445 in pairs, visually targeting relatively light and dark layers at similar depths to capture the variability range on shorter timescales while characterizing longer trends.

2.1. Inorganic analyses of bulk major, trace, rare earth element concentrations

The samples we analyzed for major, trace, and rare earth element concentrations were originally collected for moisture and density (MAD) measurements onboard the JOIDES Resolution during IODP Expedition 353. Each sample was collected with a 2 cm diameter plastic syringe that fits into the top of a 10 cm³ glass vial, allowing for the vial to be completely filled with sediment (Clemens et al., 2016). The samples were dried in a convective oven at 105°C ± 5°C for 24 hours (Clemens et al., 2016). The remaining sample preparation, digestions, and analyses were conducted at Boston University and a detailed description of the analytical geochemical procedures are presented in Dunlea et al. (2015). In summary here, sediment samples were hand-powdered with an agate mortar and pestle. For major elements, sample powders were digested by flux fusion (Murray et al., 2000) and analyzed by inductively coupled plasma-emission spectrometry (ICP-ES). For analysis of trace and rare earth elements, sample powders were dissolved in a heated acid cocktail (HNO₃, HCl, and HF, with later additions of HNO₃ and H₂O₂ after samples were dried down) under clean-lab conditions and analyzed by inductively couple plasma-mass spectrometry (ICP-MS). Three separate digestions of a matrix-matched in-house sediment standard were analyzed with each batch and determined precision [(standard deviation)/(average) x 100] was ~2% of the measured value for each element. The international Standard Reference Material BHVO-2 was analyzed as an unknown with each batch and results were consistently found to be accurate within precision for each element.

2.2. Analyses of carbon and nitrogen content and isotopes

Analyses of the abundance of total carbon (TC), total inorganic carbon (TIC), total organic carbon (TOC), nitrogen (N), and the δ¹³C of the TOC component were performed at Woods Hole Oceanographic Institution and methods are described in Whiteside et al. (2011). In brief here, samples for TOC were weighed into tared silver boats and then acidified to remove carbonates in a closed desiccator for 3 days at 60-65°C over concentrated hydrochloric acid. All samples were flash combusted in a Costech 4010 Elemental Analyzer coupled via a Finnigan-MAT ConFlo-II interface to a Thermo DeltaVPlus isotope ratio

Moved (insertion) [3]

Formatted: English (UK)

Moved (insertion) [4]

Moved (insertion) [5]

Moved (insertion) [6]

Moved (insertion) [7]

mass spectrometer. Data were recorded and integrated using the Isodat software package. Post-run calculations were performed for blank corrections, quantifications, and final calibrations.

2.3. Analyses of compound-specific biomarkers abundances and isotopes

The analyses of compound-specific biomarkers were performed at Brown University (e.g., Daniels et al., 2017). Samples were freeze-dried and lipids were extracted from 3.5 to 4.5 g of sediment using a Dionex 350 Accelerated Solvent Extractor (ASE) with dichloromethane:methanol (9:1 v/v). The fatty acids in the total lipid extract were separated from the neutral lipids using aminopropyl silica gel chromatography, eluting with a dichloromethane:isopropanol solution followed by ether with 5% acetic acid.

The fatty acids were methylated to form fatty acid methyl ester (FAME) by dissolving dried down acid fraction in in ~0.3 mL of toluene and ~1mL of 5:95 acetyl chloride:methanol with known isotopic composition. Nitrogen replaced the headspace in the vial before they were capped tightly and heated at 60°C for 12 hours. Once the reaction was complete, the FAMEs were separated from the water by-products formed during the methylation reaction. Sample received ~1mL of synthetic saline solution (50g NaCl/L of double-distilled water) and ~1mL of hexane, were vigorously shaken, and then allowed to rest until the hexane separated from the water. The hexane fraction was pipetted into a new vial, avoiding the water. Another ~1mL of hexane was added to the sample, shaken, and pipetted into the new vial. To clean the solution and isolate the fatty acids, samples were run through a second silica gel column, eluting with hexane to remove unwanted acids and then DCM to acquire the clean FAME fraction.

The FAME fraction was analyzed on an Agilent 6890 gas chromatograph with a flame ionization detector (GC-FID). Sample blanks were analyzed with every batch. The isotope ratios of the FAME fraction ($\delta D_{n\text{-acid}}$ and $\delta^{13}C_{n\text{-acid}}$) were measured on a Thermo Finnigan Delta + XL isotope ratio mass spectrometer with a HP 6890 gas chromatograph and a high-temperature pyrolysis reactor for sample introduction. For δD , three injections of each sample were analyzed and two injections of each sample were analyzed for $\delta^{13}C$. Between every six injections, an in-house lab standard mixture containing known amounts of various n-acids was analyzed to monitor instrument accuracy and precision. The δD and $\delta^{13}C$ were corrected for the methyl groups added during derivatization using the following equations $\delta D_{corrected} = [(2n+2)*\delta D_{measured} + 123.7*3]/(2n-1)$ and $\delta^{13}C_{corrected} = [(n+1)*\delta^{13}C_{measured} + 36.52]/n$, where n is the carbon chain length of the compound and the methanol added has $\delta D = -123.6\text{‰}$ and $\delta^{13}C = -36.52\text{‰}$. Analytical uncertainty was calculated by [standard deviation/average] of the injections and is typically less than 3% for δD and less than 1% for $\delta^{13}C$. The standard deviations are reported in Table S3. For every instrument run, samples were analyzed in random order.

2.4. Correction of plant physiological effects on δD

We corrected the δD for differences in C_3 versus C_4 plant physiology (Fig. S5; Smith and Freeman, 2006; Chikaraishi and Naraoka, 2007). First, we calculated the fraction of C_3 versus C_4 vegetation with $\delta^{13}C_{FA}$ for each sample, estimating that C_3 and C_4 vegetation have a $\delta^{13}C_{FA}$ of $-37.7 \pm 1.8\text{‰}$ and $-21.1 \pm 1.4\text{‰}$, respectively (Chikaraishi et al., 2004; Ponton et al.,

Moved (insertion) [8]

Moved (insertion) [9]

Moved (insertion) [10]

Moved (insertion) [11]

Moved (insertion) [12]

Moved (insertion) [13]

150 2012). Then we approximate that C₃ plants have a δD that is 30 ‰ lighter than C₄ plants (Smith and Freeman, 2006; Chikaraishi and Naraoka, 2007; Ghosh et al., 2017) and use the fraction of C₃ versus C₄ to correct the δD of each sample for the differences in plant physiology (Fig. S5). The corrected data shows the same overall trend as the uncorrected data, except the values are shifted to be more negative and the overall change in δD is steeper (Fig. S5).

3. Results

155 To determine the provenance of the aluminosilicate fraction, we examined the proportions of Al, Ti, Sc, Nb, La, and Th concentrations, because other elements (e.g., Fe, K, Mg, Si, Zr, Hf) may be affected by continental weathering, sorting during transport, and post-depositional authigenic processes. The results from 30 samples have almost constant element proportions of the selected elements, indicating that the aluminosilicate fraction of sediment did not significantly vary over the past 6 Myr. The composition of the 30 samples, even amongst the light and dark layers, matches the composition of 160 lithologies that comprise the Mahanadi basin such as Precambrian granite and gneisses of the Indian craton and associated sedimentary deposits (Sharma, 2009; Fig. S2; Table S1). Marine sediment deposits in other parts of the Bay of Bengal closer to the Krishna and Godavari Rivers or Ganges-Brahmaputra Rivers have a more mafic or highly variable composition that is not observed at Site U1445 (e.g., Tripathy et al., 2014; Fig. S2). As such, we interpret our results as recording terrestrial changes in the CMZ, specifically the Mahanadi drainage basin.

165 The pairs of samples used for organic analyses are spaced ~28 m apart (~260 kyr intervals) and the adjacent light and dark layers within each pair were 0.2 m to 4.3 m apart in the sediment core (2 kyr to 46 kyr; Fig. S1). The color difference can be related to the total organic carbon content (wt%; TOC) and total nitrogen content with darker layers having 1.0 to 2.8 times more than adjacent lighter layers (Fig. 2A; Fig. S3; Table S2).

Long-chain normal fatty acids of leaf waxes are derived from land plants and are well preserved during transport and 170 burial in marine sediment (Eglinton and Eglinton, 2008). We focus on the C₃₀ chain length to avoid possible contaminations from non-terrestrial sources that contribute shorter chain length fatty acids (Fig. S4; Table S3). The results of the δ¹³C of C₃₀ fatty acid of leaf waxes (δ¹³C_{FA}) show a 5‰ increase from mid-Pliocene to mid-Pleistocene (~3.5 to ~1.5 Ma), after which δ¹³C_{FA} decreases and becomes more variable from ~1.5 Ma to the present (Fig. 2B). The hydrogen isotope compositions of the leaf wax fatty acids (δD_{FA}) increase gradually over the past 6 Myr, but have a wide range amongst light and dark layers and shorter time intervals (Fig. 2C; Table S3). 175 Comparing the earlier and later time intervals when δ¹³C_{FA} changes, before the mid-Pliocene (3.5 Ma) δD_{FA} ranges from ~179‰ to ~147‰ and after the mid-Pleistocene (1.5 Ma) the δD_{FA} increases to between ~166‰ to ~126‰ (Fig. 2C).

Moved (insertion) [2]

Deleted: varied

Deleted: (

Deleted: Before

Deleted: Myr

Deleted: 177

Deleted: 146

Deleted: Myr

Deleted: 163

Deleted: 125

4. Discussion

4.1 C₄ Expansion on the Indian Peninsula

The $\delta^{13}\text{C}_{\text{FA}}$ of terrestrial plants is primarily a function of the photosynthetic pathway and isotopic composition of atmospheric CO₂ (e.g., Farquhar et al., 1989). In this study, the 5‰ increase in $\delta^{13}\text{C}_{\text{FA}}$ is greater than the reconstructed $\delta^{13}\text{C}$ of atmospheric CO₂ ($\leq 1\%$; Tippie et al., 2010), suggesting that a correction for $\delta^{13}\text{C}_{\text{CO}_2}$ would only slightly adjust our results. Thus we interpret $\delta^{13}\text{C}_{\text{FA}}$ as reflecting the amount of C₄ relative to C₃ vegetation produced in the CMZ.

From ~6 Ma until ~3.5 Ma, approximately 51% to 81% (avg. 69% \pm 9% s.d.) of the vegetation in the CMZ utilized a C₄ photosynthetic pathway (Fig. S5). Thus, the environmental threshold for C₄ photosynthetic pathway had already been crossed before the end of the late Miocene. Later in the mid-Pliocene, the reconstruction shows another distinct expansion reaching 64% to 92% (avg. 81% \pm 7% s.d.) of C₄ vegetation in the early Pleistocene (Fig. 2B). The change in vegetation from ~3.5 to ~1.5 Ma suggests multiple steps of C₄ expansion in the CMZ, rather than a singular late-Miocene expansion. From ~1.5 Ma to the present, the average proportion of C₄ vegetation decreased and became more variable (58% to 92%, avg. 76% \pm 10% s.d.; Fig. 2B), which may reflect the sensitivity of the region to glacial-interglacial variations observed in shorter records from this region (e.g., Zorzi et al., 2015, and references therein).

4.2 Aridification of the Indian Peninsula

After correcting for the effects of plant physiology (Sec. 2.4.), the amount of precipitation and mixing of different air masses can each vary the hydrogen isotopic composition of leaf wax fatty acids ($\delta\text{D}_{\text{FA}}$; e.g., Eglinton and Eglinton, 2008). The mixing of two air masses with unique δD values was recently observed to drive δD of rainfall in New Delhi, India, but, similar to the amount of precipitation, the relatively depleted δD corresponded with wetter conditions (Hein et al., 2017). Thus, we interpreted the $\delta\text{D}_{\text{FA}}$ as a qualitative proxy for aridity or the relative amount of precipitation. The $\delta\text{D}_{\text{FA}}$ results suggest an overall drying of the CMZ on the Indian Peninsula over the past 6 Myr. The shorter-term scatter in the $\delta\text{D}_{\text{FA}}$ record may reflect higher frequency variations in aridity or rainfall.

4.3 Patterns of C₄ Expansion surrounding the Indian Ocean

In this section, we compare our record of C₄ expansion with other compound-specific biomarker records of C₄ expansion at sites in the Indian Ocean or adjacent land and seas. Multiple proxy records document a late-Miocene C₄ expansion in the Ganges or Brahmaputra River basins such as the Siwalik Group in northern Pakistan or BoB sites receiving outflow sediment (Fig. 3A France-Lanord and Derry, 1994; Quade and Cerling, 1995; Cerling et al., 1997; Freeman and Colarusso, 2001; Sanyal et al., 2004; Behrensmeyer et al., 2007; Ghosh et al., 2017). Collectively, the reported timing of C₄ expansions in the Himalaya region ranges from 9 to 5 Myr, most commonly 8 to 6 Myr (Behrensmeyer et al., 2007). Rather than a uniform timing, detailed sampling of various deposits around the Siwalik regions shows that C₄ vegetation expansion was staggered amongst nearby sub-environments with different local conditions (Ghosh et al., 2017, and references therein). Another

Moved up [13]: (Chikaraishi et al., 2004; Ponton et al., 2012

Deleted: Approximating typical $\delta^{13}\text{C}_{\text{FA}}$ of C₃ and C₄ plants

Deleted:), we estimate that 36

Deleted: 68

Deleted: 56

Deleted: from 6 Ma until 3.5 Ma

Deleted: 62

Deleted: 88

Deleted: 80% \pm 8

Deleted: substantial

Deleted: 54

Deleted: 87

Deleted: 71% \pm 11

Deleted: The

Deleted: ,

Deleted: , and plant physiology

Deleted: corrected for the physiological effects of C₃ versus C₄ plants on $\delta\text{D}_{\text{FA}}$ (Fig. S5; Fig. 2C), and

Deleted: corrected

Deleted: Global

Deleted: ¶

→ Untangling

Deleted: triggers of C₄ expansions during the Neogene requires compiling records from many sites to identify global versus local trends. Here

Deleted: ¶

→

Deleted: Ma

Deleted: Ma

biomarker record documents a late-Miocene expansion in a wide continental region north and west of the Arabian Sea (Site 722; Fig. 1A, Fig. 3B; Huang et al., 2007). Once C₄ vegetation expanded at each of these sites, the records suggest there is overall little or no systematic change in the amount of C₄ vegetation after the late Miocene.

In contrast, the CMZ of the Indian Peninsula and a few other records around the Indian Ocean document an expansion of C₄ vegetation during the Pliocene (Fig. 3). Marine deposits in the Gulf of Aden originate from northeast Africa and record a late-Miocene C₄ expansion, followed by a relapse to predominantly C₃ vegetation ~4.3 Ma and a re-expansion of C₄ plants in the Pliocene (Site 231; Fig. 1A, Fig. 3C; Feakins et al., 2005, 2013; Liddy et al., 2016). The Pliocene re-expansion is consistent with other records from tropical East Africa (e.g., Levin et al., 2004; Cerling et al., 2011). A C₄ expansion in the Pliocene is also observed in northwest Australia (Site 763A; Fig. 3E; Andrae et al., 2018). There is little evidence of significant C₄ vegetation prior to the Pliocene, suggesting a relatively late onset of C₄ vegetation expansion in northwest Australia (Fig. 3). Additionally, there is evidence that East, South, and Central Asia experienced multiple steps of C₄ expansion through the Pliocene (e.g., An et al., 2005; Passey et al., 2009; Zhou et al., 2014; Miao et al., 2017; Koutsodendris et al., 2019). Collectively, a significant regional expansion in the Pliocene, distinctly after the first late-Miocene expansion, is common at least amongst tropical East Africa, Northwest Australia, Asia, and the Indian Peninsula.

4.4 Triggers of C₄ Expansion in the Pliocene

The adaptations of the C₄ photosynthetic pathway provides a competitive advantage over C₃ vegetation in environmental conditions with low pCO₂, high temperature, high aridity or extreme seasonality that can lead to frequent floods, droughts, and fires (e.g., Edwards et al., 2010, and references therein). Many studies hypothesize that the global expansion of C₄ vegetation in the late Miocene was triggered by pCO₂ decreasing below a temperature-dependent threshold (Herbert et al., 2016 and references therein). During the Pliocene, decreasing pCO₂ may have also contributed to the expansion of C₄ vegetation (Fig. 3a; Pagani et al., 2009; Tripathi et al., 2009; Seki et al., 2010; Bartoli et al., 2011). However, the heterogeneous regional response of the Pliocene C₄ expansion suggests pCO₂ cannot be the only driver. Given that the Pliocene C₄ expansion is observed in multiple regions surrounding the Indian Ocean that have highly seasonal rainfall, regional hydrodynamics likely played a role in the Pliocene C₄ expansion.

In the modern era, a complex interplay of multiple modes of variability dictate the unique seasonal and multi-decadal precipitation patterns in the regions surrounding the Indian Ocean. For example, rainfall on the Indian Peninsula follows quintessential monsoon behavior, but is also tied to the Inter-Tropical Convergence Zone (ITCZ), Walker Circulation, and the Indian Ocean Dipole (IOD; Gadgil, 2003; Wang et al., 2017). The biannual rains of the (semi)arid tropical East Africa are related to the ITCZ, monsoon winds, sea surface temperature (SST), and Walker Circulation (e.g., Williams and Funk, 2011; Tierney et al., 2015; Yang et al., 2015). Monsoon rains annually quench northern Australia, but the El Niño Southern Oscillation (ENSO), Walker Circulation, and the amount of Indonesian Throughflow (ITF) better explain the precipitation in other parts of Australia (Ummenhofer et al., 2009, 2011a, 2011b). Previous studies reason that changes in monsoon precipitation or frequency of fires likely triggered local expansion of C₄ vegetation (An et al., 2005; Passey et al., 2009; Zhou

Moved (insertion) [14]

Deleted: –Collectively, a significant regional expansion in the Pliocene, distinctly after the first late-Miocene expansion, is common at least amongst tropical East Africa, Northwest Australia, and the Indian Peninsula. Farther from the Indian Ocean, there is also evidence that East and Central Asia also experienced multiple steps of C₄ expansion through the Pliocene, depending on local sub-climates (e.g.,

Moved up [14]: An et al., 2005; Passey et al., 2009; Zhou et al., 2014).

Deleted:). The modes of climate variability around the Indian Ocean likely differed throughout the Pliocene and may have set the stage for a regional, multi-step reorganization of the terrestrial biome. ¶

Deleted: widespread

Deleted: Since C₄-dominated ecosystems are favored under low pCO₂ and higher temperatures, the expansion was proposed to occur earlier at lower latitudes and later cross the pCO₂ threshold at cooler, higher latitudes (e.g., Cerling et al., 1997).

Deleted: pCO₂ also decreased (Fedorov

Deleted: 2013), but ecosystems at higher latitudes or elevations remained relatively stable, perhaps sufficiently past

Deleted: pCO₂ threshold. In contrast, the tropical ecosystems adjacent to the Indian Ocean seem to be more sensitive, suggesting pCO₂ is likely not the primary driver

Deleted: .

Deleted: –Other possible triggers for C₄ expansion (aridity, seasonality, flood, droughts, and fires) depend on precipitation patterns. ...

et al., 2014; Miao et al., 2017; Andrae et al., 2018). However, the sampling resolution of our million-year study cannot resolve these seasonal, decadal, or centennial rhythms in precipitation, so we examine the underlying processes driving variations in each of these modes of variability.

The physical mechanism of each of these modes of variability can be related back to atmospheric pressure gradients. Monsoons are classically defined as a seasonal wind reversal induced by the pressure gradient caused by differential heating of land and sea (Wang et al., 2017). Broadly, the ITCZ is formed by the convergence and uplift of air masses near the equator due to differential heating between low and high latitudes and the formation of Hadley Cells. The IOD and ENSO are cyclic changes in the zonal gradient across the Indian and Pacific Oceans, respectively, and affect Walker circulation. Variable Walker circulation affects ocean upwelling and thermocline depth, which in turn will influence sea surface temperature. Sea surface temperatures can interact with the atmosphere and reinforce or dampen the atmospheric pressure gradients affiliated with the monsoon, ITCZ, IOD, or ENSO dynamics. Changes in any combination of these atmospheric gradients may have altered the seasonal precipitation in the regions surrounding the Indian Ocean during the Pliocene (Wang et al., 2017).

What could have changed atmospheric pressure gradients in the Pliocene? Indonesian Throughflow was being restricted from 5 to 3 Myr and there is evidence for strengthening SST gradients during the Pliocene (Cane and Molnar, 2001; Wara et al., 2005; Karas et al., 2009; Ford et al., 2012; Zhang et al., 2014; Burls and Fedorov, 2017; Christensen et al., 2017; White and Ravelo, 2020). Changes in the zonal and meridional SST gradients would have affected atmospheric pressure gradients in and surrounding the Indian Ocean and modified seasonal precipitation patterns. At the same time, the onset of northern hemisphere glaciation likely also helped reorganize the atmospheric pressure gradients surrounding the Indian Ocean (e.g., Koutsodendris et al., 2019). Before the intensification of glaciation at 2.7 Ma, there is evidence of four early local glaciation events at 4.9–4.8 Myr, 4.0 Myr, 3.6 Myr and 3.3 Myr (De Schepper et al., 2014). The latter two events are within age model uncertainty of 3.5 Myr when C₄ vegetation expands in various regions around the Indian Ocean. Thus, it is possible that the early local growth of ice, not the intensification of glaciation, altered the Siberian high or other atmospheric pressure gradients near the Indian Ocean changing cyclic precipitation patterns that lead to a regional expansion in C₄ vegetation.

On the Indian Peninsula, C₄ vegetation expands gradually from 3.5 Ma until the Mid-Pleistocene when the range of variability in C₄ vegetation increases. The timing of the change coincides within uncertainty with the Mid-Pleistocene transition (~1.3 Ma) where the glacial periods transitioned from 41 kyr cycles to 100 kyr cycles. Although the trigger for the switch in cyclicity is unknown, the processes responsible for the change or the ice sheets themselves may have affected atmospheric gradients and the modes of climate variability that altered the dynamics of C₄ expansion on the Indian Peninsula. The sampling resolution from 1.5–0 Ma in our study is too low to resolve the change in cyclicity. However, previous studies documenting variations in C₄ vegetation relative to C₃ since the last glacial maximum demonstrate the sensitivity of plant communities to recent glacial-interglacial cycles (Galy et al., 2008; Phillips et al., 2014; Zorzi et al., 2015, and references therein).

Deleted: –Since a single process cannot explain the precipitation patterns surrounding the Indian Ocean, the interplay of many processes likely changed during the Pliocene. Along with decreasing pCO₂, the ITF constricted (Cane and Molnar, 2001; Karas et al., 2009; Christensen et al., 2017) and SST gradients changed (Wara et al., 2005; Zhang et al., 2014; Burls and Fedorov, 2017), which would have modulated the monsoon dynamics and their interaction with the ITCZ, ENSO, IOD, and other modes of variability. Manifesting as increased aridity, seasonality, droughts, flooding, or fires, the changes in the hydroclimate variability over the Pliocene led to conditions more conducive to C₄ vegetation in certain tropical regions adjacent to the Indian Ocean.

5. Conclusion

Our study provides a piece of the puzzle in unraveling the complexities of C₄ vegetation expansion. Although C₄ vegetation was established in the CMZ on the Indian Peninsula before the end of the Miocene, the results of this study show another significant expansion in the Pliocene (~3.5 to 1.5 Myr). The latter expansion is not observed in many records from the orographically-wet Himalaya emphasizing the spatial heterogeneities in C₄ vegetation response – even within the same monsoon system. However, other regions adjacent to the Indian Ocean, such as tropical East Africa, Asia, and Northwest Australia, corroborate the observed expansion in the CMZ of the Indian Peninsula and show C₄ vegetation patterns sensitive to the changes in hydroclimate during the Pliocene. The heterogeneous response suggests that pCO₂ cannot be the exclusive driver of the expansion of the C₄ vegetation in the Pliocene and changes in regional hydrodynamics likely contributed. Restriction of Indonesian Throughflow and the onset on Northern Hemisphere glaciation may have altered the atmospheric pressure gradients and the modes of variability that determine seasonal precipitation patterns in the continents surrounding the Indian Ocean, which may have caused the regional expansion of C₄ vegetation in the Pliocene.

Data Availability

Data is included in the supplementary tables and will be made publically available in the NSF funded EarthChem database (<https://www.earthchem.org/>) maintained by the Interdisciplinary Earth Data Alliance (IEDA) at the Lamont-Doherty Earth Observatory of Columbia University.

Author Contribution

A.G.D. participated in the IODP Expedition 353 sampling party, performed the geochemical analyses, and lead writing and revisions of the manuscript. L.G. participated on IODP Expedition 353, was involved in the project’s conceptualization, sample acquisition, and provided supervision. Y.H. also participated on IODP Expedition 353, was involved in the project’s conceptualization, provided resources and funding acquisition, advised on the methodology, supervision, and aided in interpretation of data.

Competing Interests

The authors declare that they have no conflict of interest.

Deleted: and adds nuance to the discussion of triggering mechanism...

Formatted: Normal

Deleted: Ma

Deleted: ¶

6. Appendices ¶

Appendix A: Analytical Procedures - Inorganic Analyses

Moved up [3]: of bulk major, trace, rare earth element concentrations ¶

¶ The samples we analyzed for major, trace, and rare earth element concentrations were originally collected for moisture and density (MAD) measurements onboard the JOIDES Resolution during IODP Expedition 353. Each sample was collected with a 2 cm diameter plastic syringe that fits into the top of a 10 cm³

Formatted: English (UK)

Moved up [4]: 2016). The samples were dried in a convective oven at 105°C ± 5°C for 24 hours (Clemens et al., 2016). The remaining sample preparation, digestions, and analyses were conducted at Boston University and a detailed description of the analytical geochemical procedures are presented in Dunlea et al. (2015). In summary here, sediment samples were hand-powdered with an agate mortar and pestle.

Deleted: For major and certain trace

Moved up [5]: elements, sample powders were digested by flux fusion (Murray et al., 2000) and analyzed by inductively coupled

Moved up [8]: specific biomarkers abundances and isotopes ¶

Moved up [9]: Nitrogen replaced the headspace in the vial before they were capped tightly and heated at 60°C for 12 hours.

Deleted: ¶ The neutral lipid fraction of the total lipid extract was further separated into the n-alkane fraction (N1), ketone/sterol ... [2]

Moved up [10]: Sample blanks were analyzed with every batch.

Moved up [11]: The isotope ratios of the FAME fraction (δD_{n-acid} and δ¹³C_{n-acid}) were measured on a Thermo Finnigan Delta + XL

Deleted: For analysis of additional

Moved up [6]: trace and rare earth elements, sample powders were dissolved in a heated acid cocktail (HNO₃, HCl, and HF, with

Moved up [7]: (2011). In brief here, samples for TOC were weighed into tared silver boats and then acidified to remove

Deleted: ¶

→

Deleted: Between every six injections, a standard mixture containing C₁₆, C₁₈, C₂₂, C₂₆, and C₂₈ n-acids was analyzed to ... [3]

Moved up [12]: Analytical uncertainty was calculated by [standard deviation/average] of the injections and is typically less

Deleted: volume glass vial, allowing for the vial to be completely filled with sediment (Clemens et al.,

Deleted: Appendix B: Analytical Procedures - Carbon and nitrogen content and isotopes ¶ ... [1]

Deleted: Appendix C: Analytical Procedures - Compound

Acknowledgements

We thank Raj Kumar Singh (IIT Bhubaneswar, India) for providing Mahanadi sediment samples, X. Wang, R. Taroza, and M. Da Rosa Alexandre at Brown Univ. and T. Ireland at Boston Univ. for their analytical assistance and as well as S. Clemens, K. Thirumalai, V. Galy, and C. Ummenhofer for discussions and advice. This research used samples and data provided by the International Ocean Discovery Program. Funding for this research was provided by the Ocean and Climate Change Institute Postdoctoral Scholarship at Woods Hole Oceanographic Institution to AGD, and the U.S. National Science Foundation to LG (NSF OCE-0652315). USSSP post-cruise support was provided to Exp. 353 shipboard participants LG and YH.

References

- An, Z., Huang, Y., Liu, W., Guo, Z., Clemens, S., Li, L., Prell, W., Youfeng, N., Yanjun, C., Weijian, Z., Benhai, L., Qingle, Z., Yunning, C., Xiaoke, Q., Hong, C. and Zhenkun, W.: Multiple expansions of C4 plant biomass in East Asia since 7 Ma coupled with strengthened monsoon circulation, *Geology*, 33(9), 705, doi:10.1130/g21423.1, 2005.
- Andrae, J. W., McInerney, F. A., Polissar, P. J., Sniderman, J. M. K., Howard, S., Hall, P. A. and Phelps, S. R.: Initial Expansion of C4 Vegetation in Australia During the Late Pliocene, *Geophys. Res. Lett.*, 45(10), 4831–4840, doi:10.1029/2018GL077833, 2018.
- Bartoli, G., Hönisch, B. and Zeebe, R. E.: Atmospheric CO2 decline during the Pliocene intensification of Northern Hemisphere glaciations, *Paleocean.*, 26(4), 253–14, doi:10.1029/2010PA002055, 2011.
- Behrensmeyer, A. K., Quade, J., Cerling, T. E., Kappelman, J., Khan, I. A., Copeland, P., Roe, L., Hicks, J., Stubblefield, P., Willis, B. J. and Latorre, C.: The structure and rate of late Miocene expansion of C4 plants: Evidence from lateral variation in stable isotopes in paleosols of the Siwalik Group, northern Pakistan, *Geol. Soc. Am. Bull.*, 119(11-12), 1486–1505, doi:10.1130/B26064.1, 2007.
- Blaauw, M.: Methods and code for 'classical' age-modeling of radiocarbon sequences, *Quat. Geochron.*, 5, 512–518, doi:10.1016/j.quageo.2010.01.002, 2010.
- Burls, N. J. and Fedorov, A. V.: Wetter subtropics in a warmer world: Contrasting past and future hydrological cycles, *PNAS*, 114(49), 12888–12893, doi:10.1073/pnas.1703421114, 2017.
- Cane, M. A. and Molnar, P.: Closing of the Indonesian seaway as a precursor to east African aridification around 3–4 million years ago, *Nature*, 411(6834), 157–162, doi:10.1038/35075500, 2001.
- Cerling, T. E., Wang, Y. and Quade, J.: Expansion of C4 ecosystems as an indicator of global ecological change in the late Miocene, *Nature*, 361(6410), 344–345, doi:10.1038/361344a0, 1993.
- Cerling, T. E., Harris, J. M., MacFadden, B. J., Leakey, M. G., Quade, J., Eisenmann, V. and Ehleringer, J. R.: Global vegetation change through the Miocene/Pliocene boundary, *Nature*, 389(6647), 153–158, doi:10.1038/38229, 1997.

- 535 Cerling, T. E., Wynn, J. G., Andanje, S. A., Bird, M. I., Korir, D. K., Levin, N. E., Mace, W., Macharia, A. N., Quade, J. and
Remien, C. H.: Woody cover and hominin environments in the past 6 million years, *Nature Geosci.*, 476(7358), 51–
56, doi:10.1038/nature10306, 2011.
- Chakrapani, G. J. and Subramanian, V.: Factors controlling sediment discharge in the Mahanadi River Basin, India, *Journal
of Hydrology*, 117(1-4), 169–185, doi:10.1016/0022-1694(90)90091-b, 1990.
- 540 Chikaraishi, Y., Naraoka, H. and Poulson, S. R.: Hydrogen and carbon isotopic fractionations of lipid biosynthesis among
terrestrial (C3, C4 and CAM) and aquatic plants, *Phytochemistry*, 65(10), 1369–1381,
doi:10.1016/j.phytochem.2004.03.036, 2004.
- Christensen, B. A., Renema, W., Henderiks, J., De Vleeschouwer, D., Groeneveld, J., Castañeda, I. S., Reuning, L., Bogus,
K., Auer, G., Ishiwa, T., McHugh, C. M., Gallagher, S. J., Fulthorpe, C. S., IODP Expedition 356 Scientists:
545 Indonesian Throughflow drove Australian climate from humid Pliocene to arid Pleistocene, *Geophys. Res. Let.*,
44(13), 6914–6925, doi:10.1002/2017GL072977, 2017.
- Clemens, S. C., Kuhnt, W., LeVay, L. J., Anand, P., Ando, T., Bartol, M., Bolton, C. T., Ding, X., Gariboldi, K., Giosan, L.,
Hathorne, E. C., Huang, Y., Jaiswal, P., Kim, S., Kirkpatrick, J. B., Littler, K., Marino, G., Martinez, P., Naik, D.,
Peketi, A., Phillips, S. C., Robinson, M. M., Romero, O. E., Sagar, N., Taladay, K. B., Taylor, S. N., Thirumalai, K.,
550 Uramoto, G., Usui, Y., Wang, J., Yamamoto, M. and Zhou, L.: Indian Monsoon Rainfall, *Proc. of IODP*, 353,
doi:10.14379/iodp.proc.353.101.2016, 2016.
- Daniels, W. C., Russell, J. M., Giblin, A. E., Welker, J. M., Klein, E. S. and Huang, Y.: Hydrogen isotope fractionation in leaf
waxes in the Alaskan Arctic tundra, *Geochim. Cosmochim. Acta*, 213, 216–236, doi:10.1016/j.gca.2017.06.028,
2017.
- 555 De Schepper, S., Gibbard, P. L., Salzmann, U. and Ehlers, J.: A global synthesis of the marine and terrestrial evidence for
glaciation during the Pliocene Epoch, *Earth-Sci Rev.*, 135(C), 83–102, doi:10.1016/j.earscirev.2014.04.003, 2014.
- Dunlea, A. G., Murray, R. W., Sauvage, J., Spivack, A. J., Harris, R. N. and D'Hondt, S.: Dust, volcanic ash, and the evolution
of the South Pacific Gyre through the Cenozoic, *Paleocean.*, 30, 1078–1099, doi:10.1002/2015PA002829, 2015.
- Edwards, E. J., Osborne, C. P., Strömberg, C., Smith, S. A. and Consortium, C. G.: The origins of C4 grasslands: integrating
560 evolutionary and ecosystem science, *Science*, 328(5978), 587–591, doi:10.1126/science.1177216, 2010.
- Ehleringer, J. R. and Cerling, T. E.: Atmospheric CO2 and the ratio of intercellular to ambient CO2 concentrations in plants,
Tree Physiology, 15, 105–111, 1995.
- Ehleringer, J. R., Sage, R. F., Flanagan, L. B. and Pearcy, R. W.: Climate change and the evolution of C4 photosynthesis, *Mar.
Geol.*, 6(3), 95–99, doi:10.1016/0169-5347(91)90183-X, 1991.
- 565 Eglinton, T. I. and Eglinton, G.: Molecular proxies for paleoclimatology, *Earth Planet. Sci. Let.*, 275(1-2), 1–16,
doi:10.1016/j.epsl.2008.07.012, 2008.
- Farquhar, G. D., Ehleringer, J. R. and Hubick, K. T.: Carbon isotope discrimination and photosynthesis, *Annu. Rev. Plant
Physiol. Plant Mol. Biol.*, 40(1), 503–537, doi:10.1146/annurev.pp.40.060189.002443, 1989.

Deleted: .

- 570 Feakins, S. J., DeMenocal, P. B. and Eglinton, T. I.: Biomarker records of late Neogene changes in northeast African vegetation, *Geology*, 33(12), 977–4, doi:10.1130/G21814.1, 2005.
- Feakins, S. J., Levin, N. E., Liddy, H. M., Sieracki, A., Eglinton, T. I. and Bonnefille, R.: Northeast African vegetation change over 12 m.y, *Geology*, 41(3), 295–298, doi:10.1130/G33845.1, 2013.
- Fedorov, A. V., Brierley, C. M., Lawrence, K. T., Liu, Z., Dekens, P. S. and Ravelo, A. C.: Patterns and mechanisms of early Pliocene warmth, *Nature Geosci.*, 496(7443), 43–49, doi:10.1038/nature12003, 2013.
- 575 Ford, H. L., Ravelo, A. C. and Hovan, S.: A deep Eastern Equatorial Pacific thermocline during the early Pliocene warm period, *Earth Planet. Sci. Let.*, 355-356(C), 152–161, doi:10.1016/j.epsl.2012.08.027, 2012.
- France-Lanord, C. and Derry, L. A.: $\delta^{13}\text{C}$ of organic carbon in the Bengal Fan: Source evolution and transport of C3 and C4 plant carbon to marine sediments, *Geochim. Cosmochim. Acta*, 58(21), 4809–4814, doi:10.1016/0016-7037(94)90210-0, 1994.
- 580 Freeman, K. H. and Colarusso, L. A.: Molecular and isotopic records of C4 grassland expansion in the late miocene, *Geochim. Cosmochim. Acta*, 65(9), 1439–1454, doi:10.1016/s0016-7037(00)00573-1, 2001.
- Gadgil, S.: The Indian Monsoon and its Variability, *Annu. Rev. Earth Planet. Sci.*, 31(1), 429–467, doi:10.1146/annurev.earth.31.100901.141251, 2003.
- 585 Galy, V., France-Lanord, C., Peucker-Ehrenbrink, B. and Huyghe, P.: Sr–Nd–Os evidence for a stable erosion regime in the Himalaya during the past 12Myr, *Earth Planet. Sci. Let.*, 290(3–4), 474–480, doi:10.1016/j.epsl.2010.01.004, 2010.
- Galy, V., François, L., France-Lanord, C., Faure, P., Kudrass, H., Palhol, F. and Singh, S. K.: C4 plants decline in the Himalayan basin since the Last Glacial Maximum, *Quat. Sci. Rev.*, 27(13–14), 1396–1409, doi:10.1016/j.quascirev.2008.04.005, 2008.
- 590 Ghosh, S., Sanyal, P. and Kumar, R.: Evolution of C4 plants and controlling factors: Insight from n-alkane isotopic values of NW Indian Siwalik paleosols, *Org. Geochem.*, 110, 110–121, doi:10.1016/j.orggeochem.2017.04.009, 2017.
- Giosan, L., Ponton, C., Usman, M., Blusztajn, J., Fuller, D. Q., Galy, V., Haghipour, N., Johnson, J. E., McIntyre, C., Wacker, L. and Eglinton, T. I.: Massive erosion in monsoonal central India linked to late Holocene land cover degradation, *Earth Surf. Dynam.*, 5(4), 781–789, doi:10.5194/esurf-5-781-2017, 2017.
- 595 Hein, C. J., Galy, V., Galy, A., France-Lanord, C., Kudrass, H. and Schwenk, T.: Post-glacial climate forcing of surface processes in the Ganges–Brahmaputra river basin and implications for carbon sequestration, *Earth Planet. Sci. Let.*, 478, 89–101, doi:10.1016/j.epsl.2017.08.013, 2017.
- Herbert, T. D., Lawrence, K. T., Tzanova, A., Peterson, L. C., Caballero-Gill, R. and Kelly, C. S.: Late Miocene global cooling and the rise of modern ecosystems, *Nat. Geosci.*, 9(11), 843–847, doi:10.1038/ngeo2813, 2016.
- 600 Huang, Y., Clemens, S. C., Liu, W., Wang, Y. and Prell, W. L.: Large-scale hydrological change drove the late Miocene C4 plant expansion in the Himalayan foreland and Arabian Peninsula, *Geology*, 35(6), 531–534, doi:10.1130/G23666A.1, 2007.

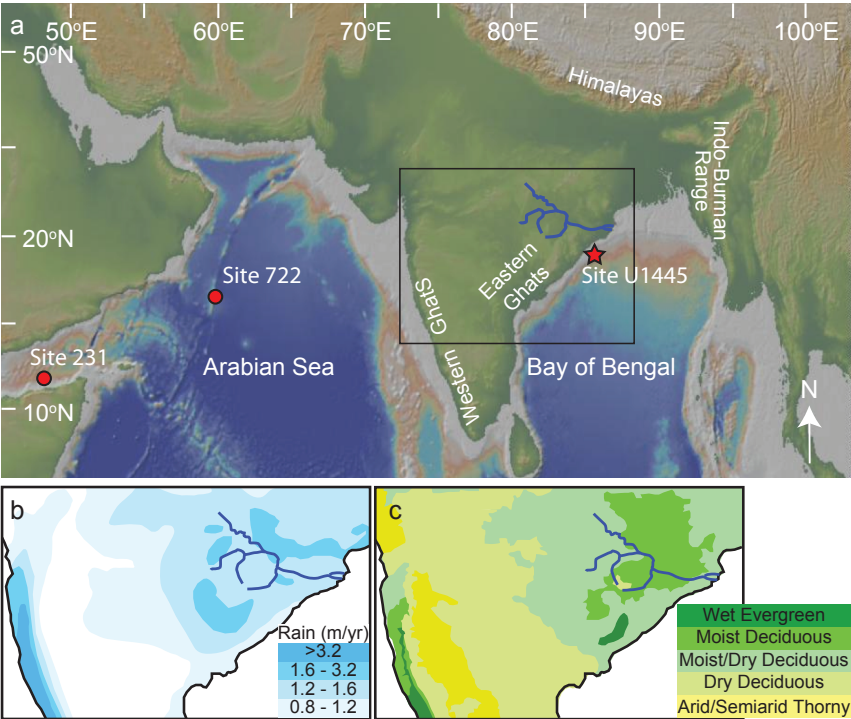
- Karas, C., Nürnberg, D., Gupta, A. K., Tiedemann, R., Mohan, K. and Bickert, T.: Mid-Pliocene climate change amplified by a switch in Indonesian subsurface throughflow, *Nat. Geosci.*, 2(6), 434–438, doi:10.1038/ngeo520, 2009.
- 605 Koutsodendris, A., Allstädt, F. J., Kern, O. A., Kousis, I., Schwarz, F., Vannacci, M., Woutersen, A., Appel, E., Berke, M. A., Fang, X., Friedrich, O., Hoorn, C., Salzmann, U. and Pross, J.: Late Pliocene vegetation turnover on the NE Tibetan Plateau (Central Asia) triggered by early Northern Hemisphere glaciation, *Glob. Planet. Change*, 180, 117–125, doi:10.1016/j.gloplacha.2019.06.001, 2019.
- Levin, N. E., Quade, J., Simpson, S. W., Semaw, S. and Rogers, M.: Isotopic evidence for Plio–Pleistocene environmental change at Gona, Ethiopia, *Earth Planet. Sci. Lett.*, 219(1–2), 93–110, doi:10.1016/S0012-821X(03)00707-6, 2004.
- 610 Liddy, H. M., Feakins, S. J. and Tierney, J. E.: Cooling and drying in northeast Africa across the Pliocene, *Earth Planet. Sci. Lett.*, 449, 430–438, doi:10.1016/j.epsl.2016.05.005, 2016.
- Lisiecki, L. E. and Raymo, M. E.: A Pliocene–Pleistocene stack of 57 globally distributed benthic δ 18O records, *Paleocean.*, 20(1), PA1003, doi:10.1029/2004PA001071, 2005.
- 615 Miao, Y., Warny, S., Clift, P. D., Liu, C. and Gregory, M.: Evidence of continuous Asian summer monsoon weakening as a response to global cooling over the last 8 Ma, *Gondwana Research*, 52, 48–58, doi:10.1016/j.gr.2017.09.003, 2017.
- Murray, R., Miller, D. J. and Kryc, K.: Analysis of major and trace elements in rocks, sediments, and interstitial waters by inductively coupled plasma–atomic emission spectrometry (ICP–AES), *ODP Tech. Note*, 29, 1–27, 2000.
- 620 Pagani, M., Liu, Z., LaRiviere, J. and Ravelo, A. C.: High Earth-system climate sensitivity determined from Pliocene carbon dioxide concentrations, *Nat. Geosci.*, 3(1), 27–30, doi:10.1038/ngeo724, 2009.
- Passey, B. H., Ayliffe, L. K., Kaakinen, A., Zhang, Z., Eronen, J. T., Zhu, Y., Zhou, L., Cerling, T. E. and Fortelius, M.: Strengthened East Asian summer monsoons during a period of high-latitude warmth? Isotopic evidence from Mio-Pliocene fossil mammals and soil carbonates from northern China, *Earth Planet. Sci. Lett.*, 277(3–4), 443–452, doi:10.1016/j.epsl.2008.11.008, 2009.
- 625 Phillips, S. C., Johnson, J. E., Giosan, L. and Rose, K.: Monsoon-influenced variation in productivity and lithogenic sediment flux since 110 ka in the offshore Mahanadi Basin, northern Bay of Bengal, *Marine and Petroleum Geology*, 58(PA), 502–525, doi:10.1016/j.marpetgeo.2014.05.007, 2014.
- Polissar, P. J., Rose, C., Uno, K. T., Phelps, S. R. and deMenocal, P.: Synchronous rise of African C4 ecosystems 10 million years ago in the absence of aridification, *Nat. Geosci.*, 12, 657–660, doi:10.1038/s41561-019-0399-2, 2019.
- 630 Ponton, C., Giosan, L., Eglinton, T. I., Fuller, D. Q., Johnson, J. E., Kumar, P. and Collett, T. S.: Holocene aridification of India, *Geophys. Res. Lett.*, 39(3), L03704, doi:10.1029/2011GL050722, 2012.
- Quade, J. and Cerling, T. E.: Expansion of C4 grasses in the Late Miocene of Northern Pakistan: evidence from stable isotopes in paleosols, *Palaeogeogr. Palaeoclimatol. Palaeoecol.*, 115, 91–116, doi:10.1016/0031-0182(94)00108-K, 1995.
- Quade, J., Cater, J. M. L., Ojha, T. P., Adam, J. and Harrison, T. M.: Late Miocene environmental change in Nepal and the northern Indian subcontinent: Stable isotopic evidence from paleosols, *Geol. Soc. Am. Bull.*, 107(12), 1381–1397, doi:10.1130/0016-7606(1995)107<1381:LMECIN>2.3.CO;2, 1995.
- 635

- Sanyal, P., Bhattacharya, S. K., Kumar, R., Ghosh, S. K. and Sangode, S. J.: Mio–Pliocene monsoonal record from Himalayan foreland basin (Indian Siwalik) and its relation to vegetational change, *Palaeogeogr. Palaeoclimatol. Palaeoecol.*, 205(1-2), 23–41, doi:10.1016/j.palaeo.2003.11.013, 2004.
- 640 Sarkar, S., Prasad, S., Wilkes, H., Riedel, N., Stebich, M., Basavaiah, N. and Sachse, D.: Monsoon source shifts during the drying mid-Holocene: Biomarker isotope based evidence from the core 'monsoon zone' (CMZ) of India, *Quat. Sci. Rev.*, 123(C), 144–157, doi:10.1016/j.quascirev.2015.06.020, 2015.
- Seki, O., Foster, G. L., Schmidt, D. N., Mackensen, A., Kawamura, K. and Pancost, R. D.: Alkenone and boron-based Pliocene pCO₂ records, *Earth Planet. Sci. Lett.*, 292(1-2), 201–211, doi:10.1016/j.epsl.2010.01.037, 2010.
- 645 Sharma, R. S.: Cratons of the Indian Shield, in *Cratons and Fold Belts of India*, vol. 127, pp. 41–115, Springer Berlin Heidelberg, Berlin, Heidelberg. 2009.
- Strömberg, C. A. E.: Evolution of Grasses and Grassland Ecosystems, *Annu. Rev. Earth Planet. Sci.*, 39(1), 517–544, doi:10.1146/annurev-earth-040809-152402, 2011.
- Tierney, J. E., Ummenhofer, C. C. and DeMenocal, P. B.: Past and future rainfall in the Horn of Africa, *Science Advances*, 1(9), e1500682–9, doi:10.1126/sciadv.1500682, 2015.
- 650 Tipple, B. J., Meyers, S. R. and Pagani, M.: Carbon isotope ratio of Cenozoic CO₂: A comparative evaluation of available geochemical proxies, *Paleocean.*, 25(3), 129–11, doi:10.1029/2009PA001851, 2010.
- Tripathi, A. K., Roberts, C. D. and Eagle, R. A.: Coupling of CO₂ and Ice Sheet Stability Over Major Climate Transitions of the Last 20 Million Years, *Science*, 326(5958), 1394–1397, doi:10.1126/science.1178296, 2009.
- 655 Tripathy, G. R., Singh, S. K. and Ramaswamy, V.: Major and trace element geochemistry of Bay of Bengal sediments: Implications to provenances and their controlling factors, *Palaeogeogr. Palaeoclimatol. Palaeoecol.*, 397(C), 20–30, doi:10.1016/j.palaeo.2013.04.012, 2014.
- Ummenhofer, C. C., Gupta, Sen, A., Briggs, P. R., England, M. H., McIntosh, P. C., Meyers, G. A., Pook, M. J., Raupach, M. R. and Risbey, J. S.: Indian and Pacific Ocean Influences on Southeast Australian Drought and Soil Moisture, *J. Climate*, 24(5), 1313–1336, doi:10.1175/2010JCLI3475.1, 2011a.
- 660 Ummenhofer, C. C., Gupta, Sen, A., Li, Y., Taschetto, A. S. and England, M. H.: Multi-decadal modulation of the El Niño–Indian monsoon relationship by Indian Ocean variability, *Environ. Res. Lett.*, 6(3), 034006–9, doi:10.1088/1748-9326/6/3/034006, 2011b.
- Ummenhofer, C. C., Gupta, Sen, A., Taschetto, A. S. and England, M. H.: Modulation of Australian Precipitation by Meridional Gradients in East Indian Ocean Sea Surface Temperature, *J. Climate*, 22(21), 5597–5610, doi:10.1175/2009JCLI3021.1, 2009.
- 665 Wara, M. W., Ravelo, A. C. and Delaney, M. L.: Permanent El Niño-Like Conditions During the Pliocene Warm Period, *Science*, 309(5735), 758–761, doi:10.1126/science.1112596, 2005.
- Wang, P. X., Bin Wang, Cheng, H., Fasullo, J., Guo, Z., Kiefer, T. and Liu, Z.: The global monsoon across time scales: Mechanisms and outstanding issues, *Earth-Sci Rev.*, 174, 84–121, doi:10.1016/j.earscirev.2017.07.006, 2017.
- 670

- White, S. M. and Ravelo, A. C.: Dampened El Niño in the Early Pliocene Warm Period, *Geophys. Res. Lett.*, 47(4), 40–15, doi:10.1029/2019GL085504, 2020.
- Williams, A. P. and Funk, C.: A westward extension of the warm pool leads to a westward extension of the Walker circulation, drying eastern Africa, *Climate Dynamics*, 37(11–12), 2417–2435, doi:10.1007/s00382-010-0984-y, 2011.
- 675 Whiteside, J. H., Olsen, P. E., Eglinton, T. I., Cornet, B., McDonald, N. G. and Huber, P.: Pangean great lake paleoecology on the cusp of the end-Triassic extinction, *Palaeogeogr. Palaeoclimatol. Palaeoecol.*, 301(1–4), 1–17, doi:10.1016/j.palaeo.2010.11.025, 2011.
- Xie, S.-P., Xu, H., Saji, N. H., Wang, Y. and Liu, W. T.: Role of Narrow Mountains in Large-Scale Organization of Asian Monsoon Convection, *J. Climate*, 19(14), 3420–3429, doi:10.1175/jcli3777.1, 2006.
- 680 Yang, W., Seager, R., Cane, M. A. and Lyon, B.: The Annual Cycle of East African Precipitation, *J. Climate*, 28(6), 2385–2404, doi:10.1175/JCLI-D-14-00484.1, 2015.
- Zhang, Y. G., Pagani, M. and Liu, Z.: A 12-Million-Year Temperature History of the Tropical Pacific Ocean, *Science*, 344(6179), 84–87, doi:10.1126/science.1246172, 2014.
- Zhou, B., Shen, C., Sun, W., Bird, M., Ma, W., Taylor, D., Liu, W., Peterse, F., Yi, W. and Zheng, H.: Late Pliocene–Pleistocene expansion of C4 vegetation in semiarid East Asia linked to increased burning, *Geology*, 42(12), 1067–1070, doi:10.1130/g36110.1, 2014.
- 685 Zorzi, C., Goñi, M. F. S., Anupama, K., Prasad, S., Hanquiez, V., Johnson, J. and Giosan, L.: Indian monsoon variations during three contrasting climatic periods: The Holocene, Heinrich Stadial 2 and the last interglacial-glacial transition, *Quat. Sci. Rev.*, 125(C), 50–60, doi:10.1016/j.quascirev.2015.06.009, 2015.

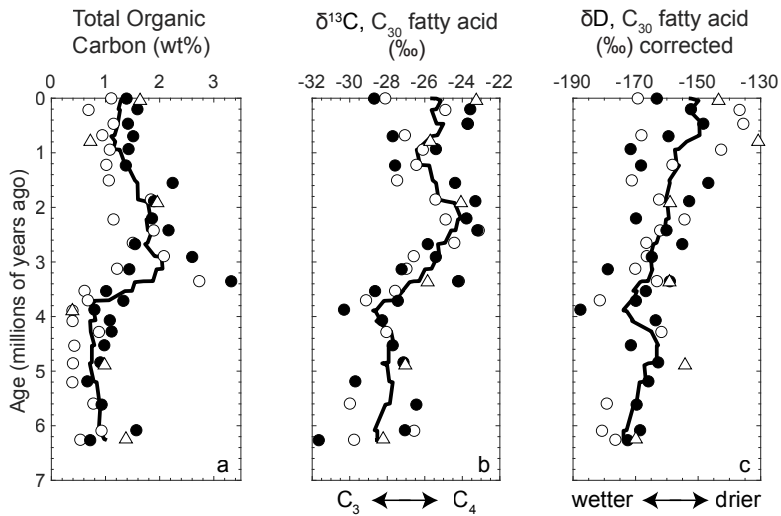
690 Figure 1. (a) Location of IODP Site U1445 in the Bay of Bengal (red star). Site 231 in the Gulf of Aden and Site 722 in the Arabian
 Sea are plotted for reference (red dots). Topography and bathymetry are represented in the background map. The Mahanadi River
 and main tributaries are traced in dark blue and the region outlined by the box is zoomed-in for Figures 1a and 1c, which are
 modified from Ponton et al. (2012). (b) average annual rainfall (m/year) and (c) natural ecosystems in the region including the
 Mahanadi River drainage basin.

695



700

Figure 2. Analyses of 57 samples from Site U1445 in the Bay of Bengal, plotted with age (Ma) and (a) total organic carbon (wt. %), (b) carbon isotope values of C₃₀ fatty acids from leaf waxes ($\delta^{13}\text{C}_{\text{FA}}$, per mil), and (c) hydrogen isotope values of C₃₀ fatty acids from leaf waxes ($\delta\text{D}_{\text{FA}}$, per mil). Black and white dots are pairs of samples from relatively dark and light layers, respectively, at a similar depth. Triangles are samples not in pairs. Black curves are a 9-point moving average of all samples.



705

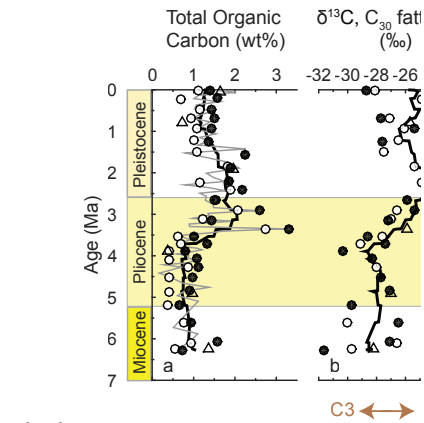
Deleted: .

Deleted: Black text labels the data and brown text is our interpretations. (a) total organic carbon (wt. %). The grey line represents shipboard measurements (

Moved up [1]: Clemens et al., 2016).

Formatted: English (UK)

Deleted: (b) carbon isotope values of C₃₀ fatty acids from leaf waxes ($\delta^{13}\text{C}_{\text{FA}}$, per mil), (c) hydrogen isotope values of C₃₀ fatty acids from leaf waxes ($\delta\text{D}_{\text{FA}}$, per mil). The yellow horizontal band highlights the Pliocene epoch.

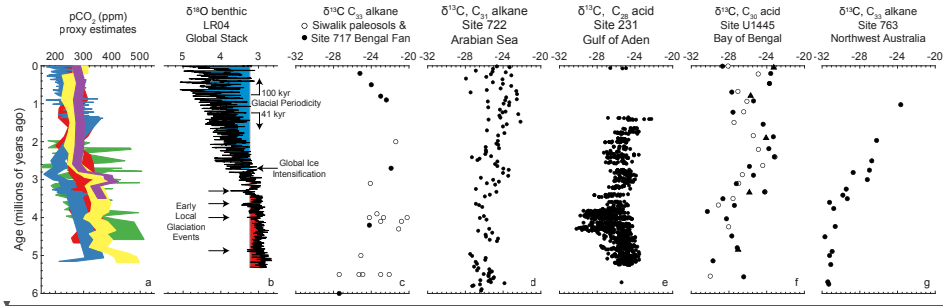


Deleted:

Page Break

Figure 3. (a)

Figure 3. (a) Proxy estimates of pCO₂ over the past 6 Myr (green, $\delta^{11}\text{B}$, Bartoli et al., 2011; red, alkenone, Pagani et al., 2009; blue, B/Ca Tripati et al. (2009); yellow, alkenone, Seki et al., 2010; purple, $\delta^{11}\text{B}$, Seki et al., 2010). (b) LR04 global stack of benthic $\delta^{18}\text{O}$ records as a proxy for global ice volume (Lisiecki and Raymo, 2005) with arrows marking early glaciation events (De Schepper et al., 2014). Blue and red highlight values above and below modern $\delta^{18}\text{O}$, respectively. (c) $\delta^{13}\text{C}$ of C₃₃-alkanes from Siwalik paleosols in Northern Pakistan (white dots) and from sediment at Site 717 in the Bengal Fan (black dots; Freeman and Colarusso, 2001). (d) $\delta^{13}\text{C}$ of C₃₁-alkanes at Site 722 in the Arabian Sea (Huang et al., 2007), which integrates vegetation variability from north and east of the Arabian Sea. (e) $\delta^{13}\text{C}$ of C₂₈-fatty acids at Site 231 in the Gulf of Aden, which records vegetation in East Africa (Feakins et al., 2013; Liddy et al., 2016). (f) $\delta^{13}\text{C}$ of C₃₀-fatty acid at Site U1445 in the Bay of Bengal, which records vegetation from the Mahanadi basin on the Indian Peninsula (this study). (g) $\delta^{13}\text{C}$ of C₃₃-alkanes from northwest Australia (Andrae et al., 2018).



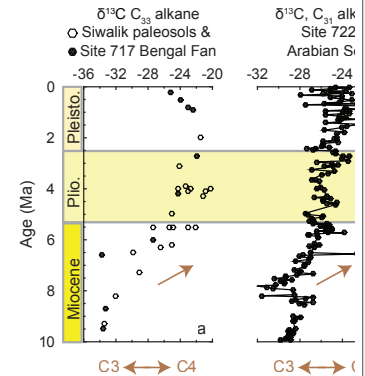
Deleted: b

Deleted: c

Deleted: d

Deleted: e

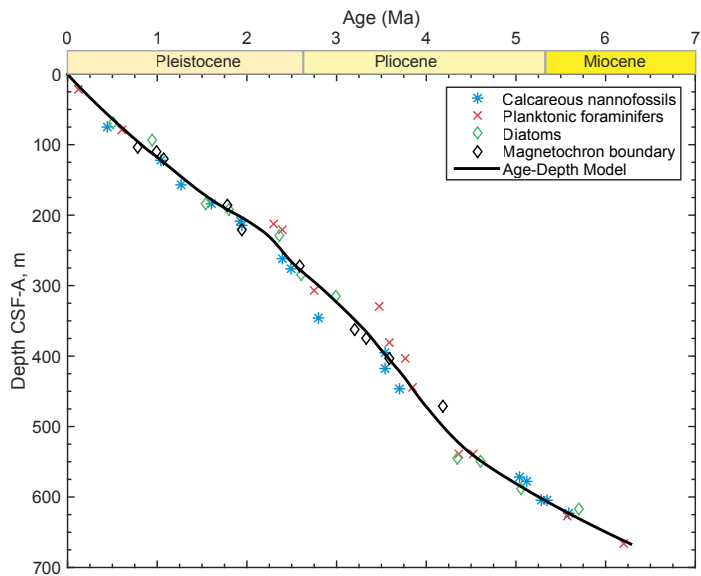
Deleted: Brown arrows point out the late-Miocene C₄ expansion in Northern Pakistan, Arabian Sea, and Gulf of Aden as well as the Pliocene C₄ expansion in northeast Africa and the Indian Peninsula.



Deleted:

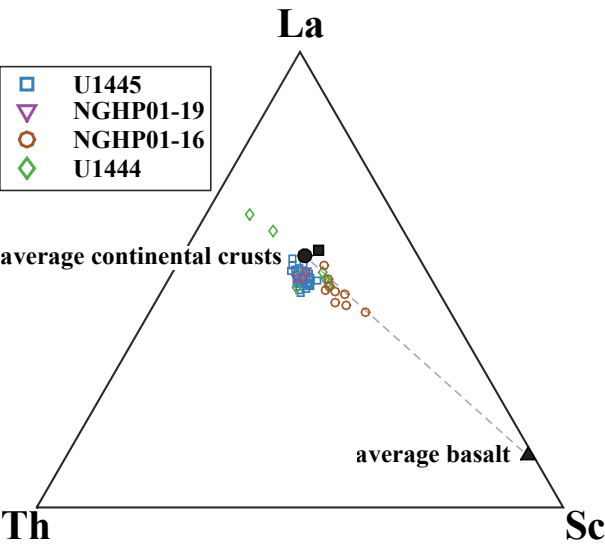
Supplemental Material

Figure S1. Age-depth model for Site U1445. To determine the ages of our samples, we fit the biostratigraphic and magnetostratigraphic age constraints (Clemens et al., 2016) with an age-depth model using CLAM software in R (Blaauw, 2010). We ran iterations of the model with different types of fit and levels of smoothing, and identified a locally weighted spline with 0.4 smoothing to best represent the trends observed in the age constraints. The differences between the age models iterations are not significant and would not change the interpretations of this study.



750

Figure S2. La-Th-Sc diagram of 30 sediment samples from the Bay of Bengal. Samples from IODP Site U1445 (blue squares) are plotted as well IODP Site U1444 (green diamonds), NGHP Site 19 (purple triangles), and NGHP Site 16 (brown circles) in the Bay of Bengal. Average upper continental crust (black square, Rudnick and Gao, 2014), post-Archean average Australian Shale (black dot, Taylor and McLennan, 1985), and average mid-ocean ridge basalt (Gale et al., 2013) compositions are plotted for reference.

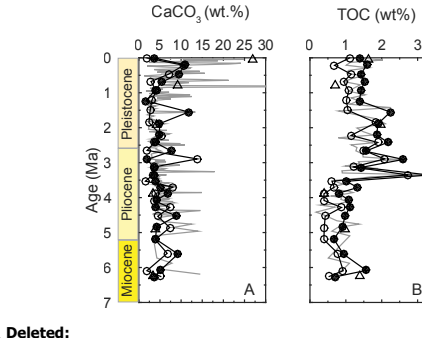
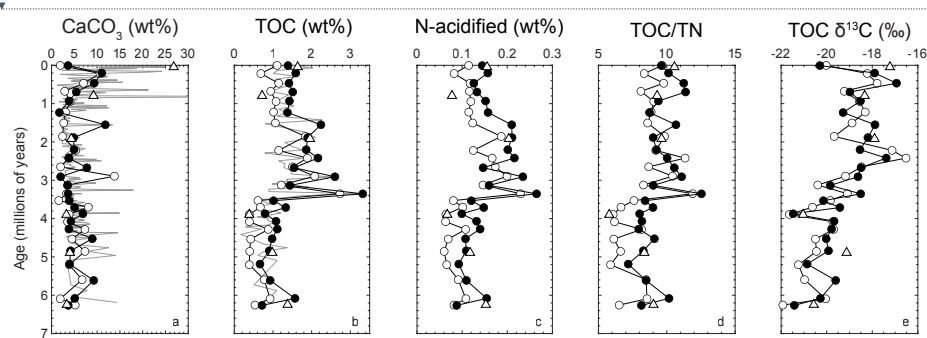


755

760

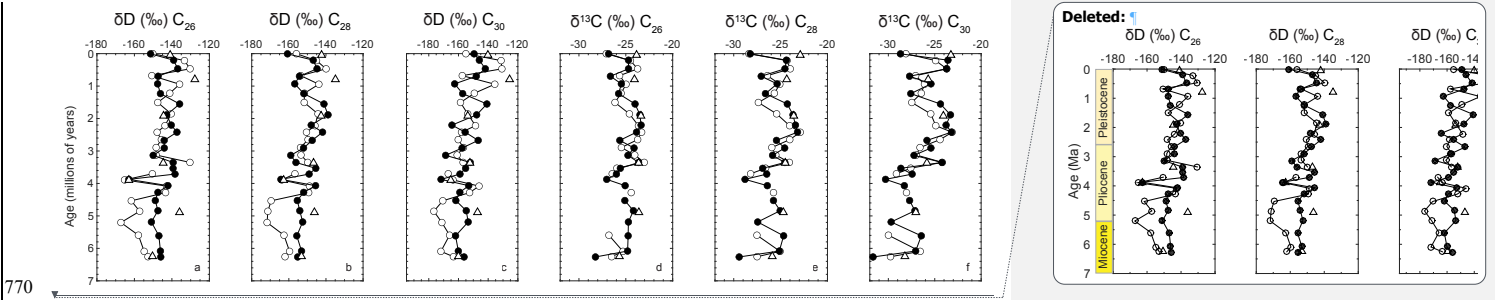
Figure S3. Carbonate and bulk organic analyses at Site U1445. Analysis of 57 samples at IODP Site U1445 for (a) bulk calcium carbonate content (weight %) calculated as (total inorganic carbon x (8.33313 CaCO₃ wt/C wt)), (b) total organic carbon concentration (weight %), (c) total acidified nitrogen content (weight %), (d) Ratio of total organic carbon to total nitrogen (TOC/TN, wt.%/wt.%) and (e) carbon isotopes of the total organic carbon (per mil). Black dots represent visually darker layers relative to a lighter layer (white dot) at a similar depth. TOC/TN shows a distinct increase in the mid-Pliocene, but remains within the range of TOC/TN expected for marine organic material.

765

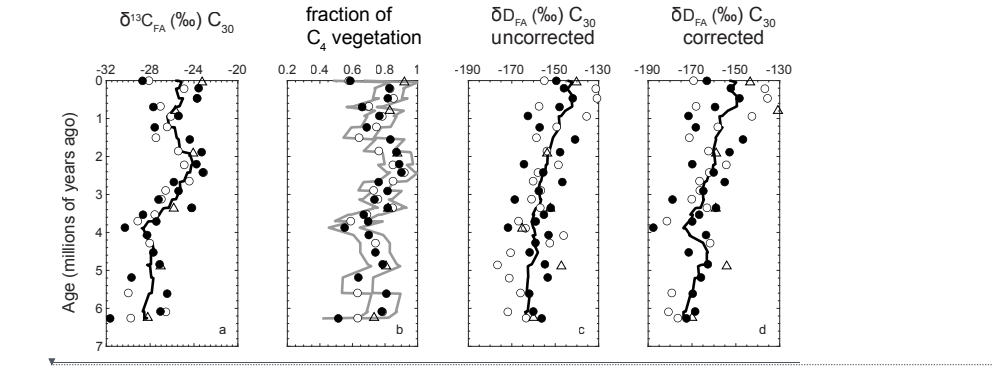


Deleted:

Figure S4. Long-chain fatty acids from leaf waxes extracted from Site U1445. Plotted from left to right are hydrogen isotopes and then carbon isotopes of leaf wax fatty acids from chainlengths C₂₆, C₂₈, and C₃₀.

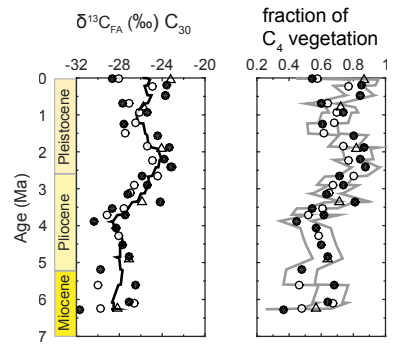


775 Figure S5. Correcting δD for differences in fractionation due to plant physiology. (a) Raw, uncorrected $\delta^{13}C_{FA}$ plotted for comparison. (b) Calculated fraction of C_4 vegetation with grey lines indicating the maximum and minimum boundaries. (c) Uncorrected δD data, plotted here for comparison. (d) The δD data corrected for differences between C_3 and C_4 plant physiologies.



780

Deleted: We used the $\delta^{13}C_{FA}$ to calculate the relative percentages of C_3 and C_4 vegetation and then correct the δD for differences in plant physiology (Smith and Freeman, 2006; Chikaraishi and Naraoka, 2007). For consistency with other studies in proximity to our marine sediment site, we used the end-member values of Ponton et al. (2012). They used values reported in Chikaraishi et al. (2004) extracted from 52 measurements of $\delta^{13}C$ for n-alkanoic acids isolated from different plant species of C_3 plants between C_{26} and C_{32} averaging -37.7 ± 1.8 ‰ and 16 measurements of the same compounds in C_4 plants with an average of -21.1 ± 1.4 ‰. We use these same end-member values [-37.7 ‰ = 0% C_4 plants; -21.1 ‰ = 100% C_4], to calculate our $\delta^{13}C$ plant wax values as a percentage of C_4 plants. The maximum and minimum fractions of C_4 vegetation (gray lines) were calculated with the standard deviation of $\delta^{13}C$ within the C_3 and C_4 groups (Ponton et al., 2012).
We estimate the offset in δD between C_3 and C_4 plants to be ~ 25 ‰ (Smith and Freeman, 2006; Ghosh et al., 2017) where C_3 is on average 25 ‰ lighter than C_4 vegetation. Using the fraction of C_4 relative to C_3 estimated from the $\delta^{13}C_{FA}$, we correct the δD of each sample for the differences in plant physiology.
The corrected data shows the same overall trend as the uncorrected data, except the values are shifted to be more negative and the change in δD between ~ 3.5 and ~ 1.5 Ma is steeper. Thus, the correction emphasizes the drying trend that we interpret in the data. Since we are interpreting δD as a qualitative proxy for aridity or the relative amount of precipitation, the slope of the δD change has no impact on our interpretations of the data. The corrected δD data is discussed in the main text of the manuscript.



815 Table S1. Concentrations of major, trace, and rare earth elements. Inorganic analyses of major, trace, and rare earth element concentrations for 30 bulk sediment samples at Site U1445 and additional samples from other sites in the Bay of Bengal for reference. For methods see Appendix A or details in Dunlea et al. (2015).

Table S2. Carbonate and bulk organic analyses data at Site U1445. Analysis of 57 samples at Site U1445 for bulk calcium carbonate, total organic carbon, total carbon, total acidified nitrogen, carbon isotopes of the total organic carbon, and the designation of visually lighter versus darker samples at similar depths.

820 Table S3. Carbon and hydrogen isotope analysis of leaf wax fatty acids at Site U1445. Hydrogen isotopes and carbon isotope analyses of leaf wax fatty acids extracted from 57 samples at Site U1445. Measurements from fatty acid chainlengths C₂₆, C₂₈, and C₃₀ are reported with their standard deviation. The correction for C₃-C₄ physiological differences in the hydrogen isotopes of C₃₀ fatty acids is reported, estimating C₃ vegetation as having a $\delta^{13}\text{C}$ of -35.4 ‰ and C₄ vegetation as -21.4‰ (Fig. S5).

References for Supplemental Material

Blaauw, M.: Methods and code for 'classical' age-modeling of radiocarbon sequences, *Quat. Geochron.*, 5, 512-518, doi:10.1016/j.quageo.2010.01.002, 2010.

Chikaraishi, Y. and Naraoka, H.: $\delta^{13}\text{C}$ and δD relationships among three n-alkyl compound classes (n-alkanoic acid, n-alkane and n-alkanol) of terrestrial higher plants, *Org. Geochem.*, 38(2), 198–215, doi:10.1016/j.orggeochem.2006.10.003, 2007.

Chikaraishi, Y., Naraoka, H. and Poulson, S. R.: Hydrogen and carbon isotopic fractionations of lipid biosynthesis among terrestrial (C_3 , C_4 and CAM) and aquatic plants, *Phytochemistry*, 65(10), 1369–1381, doi:10.1016/j.phytochem.2004.03.036, 2004.

835 Clemens, S. C., Kuhnt, W., LeVay, L. J., Anand, P., Ando, T., Bartol, M., Bolton, C. T., Ding, X., Gariboldi, K., Giosan, L., Hathorne, E. C., Huang, Y., Jaiswal, P., Kim, S., Kirkpatrick, J. B., Littler, K., Marino, G., Martinez, P., Naik, D., Peketi, A., Phillips, S. C., Robinson, M. M., Romero, O. E., Sagar, N., Taladay, K. B., Taylor, S. N., Thirumalai, K., Uramoto, G., Usui, Y., Wang, J., Yamamoto, M. and Zhou, L.: Indian Monsoon Rainfall, *Proc. of IODP*, 353, doi:10.14379/iodp.proc.353.101.2016, 2016.

840 Dunlea, A. G., Murray, R. W., Sauvage, J., Spivack, A. J., Harris, R. N. and D'Hondt, S.: Dust, volcanic ash, and the evolution of the South Pacific Gyre through the Cenozoic, *Paleocean.*, 30, 1078–1099, doi:10.1002/2015PA002829, 2015.

Gale, A., Dalton, C. A., Langmuir, C. H., Su, Y. and Schilling, J.-G.: The mean composition of ocean ridge basalts, *Geochem. Geophys. Geosy.*, 14(3), 489–518, doi:10.1029/2012GC004334, 2013.

Ghosh, S., Sanyal, P. and Kumar, R.: Evolution of C_4 plants and controlling factors: Insight from n-alkane isotopic values of NW Indian Siwalik paleosols, *Org. Geochem.*, 110, 110–121, doi:10.1016/j.orggeochem.2017.04.009, 2017.

845 Ponton, C., Giosan, L., Eglinton, T. I., Fuller, D. Q., Johnson, J. E., Kumar, P. and Collett, T. S.: Holocene aridification of India, *Geophys. Res. Lett.*, 39(3), L03704, doi:10.1029/2011GL050722, 2012.

Rudnick, R. L. and Gao, S.: Composition of the Continental Crust, in *Treatise on Geochemistry*, edited by H. Holland and K. Turekian, pp. 1–51, Elsevier Ltd., 2014.

850 Smith, F. A. and Freeman, K. H.: Influence of physiology and climate on δD of leaf wax n-alkanes from C_3 and C_4 grasses, *Geochim. Cosmochim. Acta*, 70(5), 1172–1187, doi:10.1016/j.gca.2005.11.006, 2006.

Taylor, S. R. and McLennan, S. M.: *The Continental Crust: Its Composition and Evolution*, Blackwell Scientific Publications Inc., Oxford. 1985.

Deleted: ¶

Deleted: ¶
Page Break
¶
Page Break
¶
Page Break
¶

Deleted: ¶
Page Break
¶
Page Break
¶
Page Break
¶