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Reply to: Multiple drivers of Miocene C$_4$ ecosystem expansions

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We thank Schefuß and Dupont[1] for their interest in our research and respond to their comments below.

Identifying the emergence of C$_4$ ecosystems

We identify the emergence and expansion of C$_4$ ecosystems in four different sediment cores from two oceans, including regions offshore Northwest, West and East Africa. The patterns of δ^{13}C change are common to all sites, synchronous at ~10 Myr ago (Ma), replicated in timing by the C$_4$ dietary signature of East African mammals[2,3] and consistent with what is known about leaf-wax *n*-alkanes in modern African vegetation (see refs. [4,5] and references therein). Any alternative interpretation of the data must address these observations.

We identified two distinct events using the four sediment cores. The first was the emergence of C$_4$ vegetation at ~10 Ma, which was followed by the second event: their rise to dominance at the ecosystem scale. It is the totality of the δ^{13}C patterns in all homologues that we use to identify the initial increase in contributions from C$_4$ plants at all four sites. As described in our paper[6], we observe the greatest increase in *n*-C$_{35}$ δ^{13}C values after ~10 Ma and progressively smaller increases in sequentially shorter odd-numbered homologues (*n*-C$_{33}$, *n*-C$_{31}$). The δ^{13}C changes in the longer chain lengths are substantial and statistically significant: at ODP 659 the isotopic value of *n*-C$_{35}$ at 10.07 Ma is +1.3‰ from the mean of older samples, and at 9.9 Ma is +1.7‰, +3.0 and +4.1 s.d. from the mean, respectively. Changes in *n*-C$_{33}$ are similarly significant: +1.67‰ from the long-term mean by 9.68 Ma (+3.2 s.d.). These positive δ^{13}C changes mark the beginning of almost 5 Myr of increasing δ^{13}C values and indicate the onset of C$_4$ ecosystem expansion in Northwest Africa. Furthermore, the timing of detected δ^{13}C shifts across homologues (first *n*-C$_{35}$, then *n*-C$_{33}$, then *n*-C$_{31}$) is exactly as expected from the greater contributions of C$_4$ vegetation to longer-chain *n*-alkanes in modern African vegetation: modern C$_4$ grasses in Africa make more *n*-C$_{35}$ and *n*-C$_{33}$ than C$_3$ plants (see ref. [4]). In contrast, *n*-C$_{31}$ or weighted average δ^{13}C values will miss the early presence of C$_4$ plants because they require substantial contributions from C$_4$ vegetation before any isotopic change can be confidently identified. For example, the *n*-C$_{31}$ δ^{13}C values do not significantly increase until 9.26 Ma, reaching +1.5‰, or +3.7 s.d. above the mean from pre-10.1 Ma samples.

The sensitivity of longer-chain *n*-alkanes to the contributions of C$_4$ plants is further supported by the first evidence of C$_4$ diets in equids by 9.9 Ma, the same time as the onset of C$_4$ expansion identified by *n*-alkane δ^{13}C in East Africa[2,3,6]. The dietary δ^{13}C signature in the tooth enamel of a grazing taxon is more sensitive to a low abundance of C$_4$ vegetation in the landscape due to their selective feeding on grasses, similar to the amplified contribution of African C$_4$ grasses to the *n*-C$_{33}$ and *n*-C$_{35}$ long-chain *n*-alkanes in sediments. By 9.3 Ma, the presence of C$_4$ vegetation in East Africa supported a variety of mixed and C$_4$-dominated diets (50–100% C$_4$) in equid, rhino, bovid, hippo, suid and gomphothere lineages, demonstrating their role as an important dietary resource for large and megaherbivores[2].

We observe increasing abundances of longer-chain C$_{33}$ and C$_{35}$ *n*-alkanes at the two east African core sites (DSDP 235 and 241), matching the increasing C$_4$ contribution in the δ^{13}C values. We discussed the *n*-alkane distributions at ODP 659 and 959 in the supplementary information of our study[6]. Briefly, ODP 659 and 959 *n*-alkane distributions are similar across the onset and expansion of C$_4$ ecosystems. However, the δ^{13}C values of all chain lengths at all four sites clearly demonstrate a greater abundance of the longer-chain *n*-C$_{33}$ and *n*-C$_{35}$ alkanes in the C$_4$ plant contribution to the samples (as observed in modern African plants and Holocene sediments)[4,5,7]. Therefore, the data from ODP 659 and 959 simply require that C$_4$ plants are replacing one of the C$_3$ sources with similar *n*-alkane distributions (as shown by the δ^{13}C signatures), perhaps reflecting an already dry climate as suggested by ref. [8].

We observe statistically significant increases in *n*-alkane δ^{13}C values starting at ~10 Ma, and sustained increases after this without a return to the pre-10 Ma values (except one sample at ~8 Ma from ODP 959). We therefore conclude that the signature of increasing δ^{13}C values reflects the contributions of C$_4$ plants, as corroborated by the inclusion of C$_4$ plants in herbivore diets beginning at 9.9 Ma. If this were not the case, these results would require an initial increase in the δ^{13}C values of C$_3$ plant contributions to the longer-chain *n*-alkanes followed by a replacement with increasing amounts of C$_4$ plants. In bulk leaf tissues from C$_3$ plants, such δ^{13}C increases can occur by decreasing mean annual precipitation[9,10]. However, the magnitude of the required changes in mean annual precipitation is substantial. For example, an increase of +1.3‰ as observed in the *n*-C$_{35}$ alkane at 10.07 Ma at ODP 659 would require a 42% decrease in mean annual precipitation[9] over 0.7 Myr that was then sustained for several millions of years. Such a change is not supported by other proxy data, including the leaf-wax δD values. Other factors influencing bulk leaf tissue δ^{13}C values, such as a canopy effect[10], are unlikely, as the middle Miocene vegetation in Northwest and East Africa was already a dry woodland with grass

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elements, and in these environments such effects are minimal (as discussed in our original contribution[6]).

We recognize the possible inclusion of lycopane in the n-C_{35} alkane $\delta^{13}C$ measurement. However, we find no detectable contribution from this molecule through sensitive screening by ion ratios in gas chromatography–mass spectrometry analysis. Furthermore, the $\delta^{13}C$ patterns across n-alkane molecules are consistent with a modern C_4 grass plant-wax source but not with lycopane co-elution. These leaf-wax $\delta^{13}C$ patterns also occur at all four core sites despite very different oceanographic settings, indicating a plant-wax signal rather than a marine algal source. Finally, as noted above, the timing of the $\delta^{13}C$ increase in East Africa identified in the n-C_{35} (and other) alkanes coincides with the presence of dietary C_4 in East African equids, which are also more sensitive to a low abundance of C_4 vegetation in the landscape.

Absence of aridification

We used the δD values of leaf-wax n-alkanes (accounting for vegetation change) to detect changes in rainfall patterns based on theoretical grounds for this relationship (see ref. [11] and references therein) and empirical evidence from modern Africa[12,13]. The observation from modern and historical African sediments that plant-wax (and rainfall) δD is highly correlated with spatial and temporal patterns in measured rainfall amount should directly inform the interpretation of rainfall δD on longer timescales. That other factors such as moisture source region might affect the baseline or magnitude of this relationship is understood, but unlikely to have changed in our study locations. Moisture in Northwest Africa is sourced from the subtropical Atlantic Ocean, and moisture in East Africa from the Indian Ocean. The major wind patterns and the substantial east–west extent of the African continent at these latitudes precludes major contributions from other sources, in contrast with the core site in southwest Africa mentioned by Schefuß and Dupont[1]. Furthermore, climate model studies of highly altered palaeogeography (such as ref. [14]) suggest that these sources remained stable through time. Other effects, such as temperature, reflect the underlying atmospheric water balance (see ref. [15]), and are largely non-existent in the tropics. For example, the spatial rainfall isotope–temperature relationship[16] overestimates the temporal slope, and the slope in tropical regions may in fact be close to zero[17]. Finally, the increase in dust flux that we document occurs much later than the initiation of C_4 ecosystem expansion. As has been shown for more recent time periods, African dust fluxes largely reflect increased wind strength and transport, and are not a reliable indicator for aridity[18].

Sampling resolution

Our sampling resolution is sufficient to detect long-term changes in plant-wax δD (and therefore hydroclimate) that would be required to sustain any permanent shift towards a C_4 ecosystem. We demonstrated through detailed analysis that the variability we observe in δD is consistent with Pleistocene and Pliocene orbitally driven variability at ODP 659[19,20] and that we would be able to detect any substantial shift or trend in δD values required for a hydrologic driver for C_4 ecosystem establishment (see supplementary figs. 9 and 10 in ref. [6]).

Drivers of C_4 ecosystem establishment

In conclusion, we regard the consistent data from four different sites in two distinct geographic regions as evidence for the early establishment of C_4 ecosystems in Northwest and East Africa in the absence of aridification. The parsimonious explanation is that declining CO_2 levels drove the emergence of these ecosystems. The divergence of proxy CO_2 estimates through this time interval is a central problem in palaeoclimate studies that many research groups are working on from different angles. A decline in CO_2 preceding and accompanying the emergence of C_4 ecosystems is consistent with recent CO_2 estimates for this time period[21,22]. Because we rule out any substantial temperature or hydrologic change, our data support the argument of Herbert et al.[23] and the hypothesis of Cerling et al.[24] and Ehleringer et al.[25]: the late Miocene temperature decline and early African C_4 ecosystem expansion were driven by declining CO_2.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41561-020-0591-4.

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