

# Supporting Information

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## SI Text

**Age Model of GeoB9528-3.** The age model of GeoB9528-3 was constructed by visual correlation of the benthic foraminifer *C. wuellerstorfi*  $\delta^{18}\text{O}$  record with the  $\delta^{18}\text{O}$  records of marine sediment core MD95-2042 (1) and the global benthic  $\delta^{18}\text{O}$  stack of Lisiecki and Raymo (2). The software package AnalySeries 1.1 (3) was used to perform peak-to-peak correlation, which was done on the smoothed (five-point running mean)  $\delta^{18}\text{O}$  records. The  $\delta^{18}\text{O}$  record of GeoB9528-3 was first correlated to the record of MD95-2042 (1) in the time interval spanning from 0 to  $\approx 140$  ka, and the older part of the record was correlated to the global benthic stack (2) in the time interval from  $\approx 140$  to 192 ka (Fig. S1). The benthic  $\delta^{18}\text{O}$  record of GeoB9528-3 exhibits a close correlation with both the records of MD95-2042 (1) ( $R^2 = 0.91$ ) and the global benthic stack (2) ( $R^2 = 0.93$ ) (Fig. S1), attesting to the strength to the chronology.

**Long-Chain *n*-Alkane  $\delta^{13}\text{C}$  Values for  $\text{C}_3$  and  $\text{C}_4$  Plants.** To estimate the past contribution of  $\text{C}_3$  and  $\text{C}_4$  vegetation to the different *n*-alkanes, we compiled a list of *n*-alkane  $\delta^{13}\text{C}$  values reported for  $\text{C}_3$  and  $\text{C}_4$  plants in the literature (Table S1 and Table S2) (4–7). The mean values of  $\text{C}_{29}$  *n*-alkanes are  $-34.7\text{‰}$  ( $n = 50$ ) and  $-21.4\text{‰}$  ( $n = 49$ ) for  $\text{C}_3$  and  $\text{C}_4$  plants, respectively. The mean values of  $\text{C}_{31}$  *n*-alkanes are  $-35.2\text{‰}$  ( $n = 52$ ) and  $-21.7\text{‰}$  ( $n = 50$ ) for  $\text{C}_3$  and  $\text{C}_4$  plants, respectively. We use these mean end-members for  $\text{C}_3$  and  $\text{C}_4$  plants to create a binary mixing models to estimate the contribution of  $\text{C}_4$  plants to the  $\text{C}_{29}$  and  $\text{C}_{31}$  *n*-alkanes, respectively. There is likely substantial error associated with the  $\% \text{C}_4$  estimates. The largest *n*-alkane  $\delta^{13}\text{C}$  standard deviation of 2.6, which is reported for the  $\text{C}_{29}$  and  $\text{C}_{31}$  *n*-alkanes of  $\text{C}_3$  plants (Table S1 and Table S2), translates to a maximum error estimate of  $\pm 20\%$   $\text{C}_4$  plants. However, for the purpose of our study, the overall trends in the *n*-alkane  $\delta^{13}\text{C}$  records are more important than the  $\% \text{C}_4$  estimates because relatively enriched (depleted) *n*-alkane  $\delta^{13}\text{C}$  values indicate increased (decreased) inputs from  $\text{C}_4$  plants. Therefore, shifts to relatively enriched or depleted *n*-alkane  $\delta^{13}\text{C}$  values provide important information on past vegetation shifts in central North Africa.

**A Dust Source for Long-Chain Odd-Numbered *n*-Alkanes.** Long-chain, odd-numbered *n*-alkanes ( $\text{C}_{25}$ – $\text{C}_{35}$ ) are a main component of plant epicuticular leaf waxes (8), and these compounds are generally well preserved in sediments. The carbon isotopic composition of *n*-alkanes can be used to distinguish between plants using the different photosynthetic pathways because  $\text{C}_4$  plants, such as warm-season grasses, possess a  $\text{CO}_2$  concentrating mechanism, which causes them to be isotopically enriched in  $^{13}\text{C}$  compared with  $\text{C}_3$  plants, which include most trees, cool-season grasses, and sedges. A third photosynthetic pathway, the CAM pathway, has isotopic values intermediate between those of  $\text{C}_3$  and  $\text{C}_4$  plants but CAM plants are not a significant component of northwest African vegetation (9).

Although long-chain *n*-alkanes are produced by higher plants, these compounds are also found in petroleum. However, the *n*-alkane carbon preference index (CPI) (10), which is used to examine odd over even carbon number predominance, can be used to distinguish terrestrial plant from petroleum sources. Terrestrial plants are characterized by CPIs of  $>3$ , whereas mature hydrocarbons have CPIs of  $\approx 1$  (10). Core GeoB9528-3 is characterized by high CPI values, which range from 3.5 to 8.3

with a mean value of 6.2, indicating a source from terrestrial higher plants throughout the entire record (8).

Plant leaf waxes (*n*-alkanes) can be transported to marine sediments by wind or water (runoff or riverine inputs); however, fluvial transport is not likely at site GeoB9528-3. The coring site is located offshore on the continental slope with no major rivers in close vicinity. Furthermore, we analyzed core GeoB9528-3 for branched glycerol dialkyl glycerol tetraethers (GDGTs), which are lipids produced by soil bacteria that are transported solely by runoff/riverine input (11). However, branched GDGTs were not detected in GeoB9528-3. Thus, the main supply of *n*-alkanes to GeoB9528-3 is via wind erosion.

## Origin of Dust and Associated Leaf Wax *n*-Alkanes Transported to Site GeoB9528-3.

The main source of *n*-alkanes to site GeoB9528-3 is from the central Africa near the boundary of the Sahara with the Sahel (Fig. 1). Isotopic mapping of *n*-alkanes in dust (12) and surface sediments (13) collected off the coast of Northwest Africa supports this hypothesis. The isotopic pattern in surface sediments of Northwest Africa depends in the latitudinal distribution of vegetation and the transport pathways of the wind systems. Enriched *n*-alkane  $\delta^{13}\text{C}$  values, indicating high  $\text{C}_4$  contributions, are noted in both dust and surface sediments offshore Northwest Africa between  $\approx 0$  and  $10^\circ\text{N}$  (12, 13), although at this latitude the adjacent continent is covered by tree savanna or rainforest, which is  $\text{C}_3$  dominated. This pattern is attributed to long-distance transport of Saharan dust to offshore Northwest Africa (12, 13). Modeling of backwards trajectories indicates that the dust reaching  $10^\circ\text{N}$  (near the location of GeoB9528-3) originates from central Africa near the boundary of the Sahara with the Sahel (12). Thus, this region of central North Africa is likely the main source of *n*-alkanes to site GeoB9528-3.

**Relationship Between  $\delta^{13}\text{C}_{\text{benthic}}$  and *n*-Alkane  $\delta^{13}\text{C}$ .** Throughout the past 200 ka, a significant negative linear relationship is observed between  $\delta^{13}\text{C}_{\text{benthic}}$  and the *n*-alkane  $\delta^{13}\text{C}$  records of GeoB9528-3 (Fig. S2;  $R^2 = -0.79$  for the  $\text{C}_{29}$  *n*-alkane and  $-0.82$  for the  $\text{C}_{31}$  *n*-alkane;), indicating a strong association between AMOC strength and vegetation type in the Sahara/Sahel region.

**Alkenone SST Estimates.** Molecular identification of the  $\text{C}_{37:2}$  and  $\text{C}_{37:3}$  alkenones was performed on a Thermo Finnigan Trace Gas Chromatograph Ultra coupled to Thermo Finnigan DSQ mass spectrometer. A 25-m CP Sil-5 fused silica capillary column was used (25 m  $\times$  0.32 mm; film thickness = 0.12  $\mu\text{m}$ ) with helium as the carrier gas. The column was directly inserted into the electron impact ion source of the DSQ quadrupole mass spectrometer. Mass scans were made in the range of  $m/z = 50$ – $800$  with three scans per s and an ionization energy of 70 eV. The temperature program initiated at  $70^\circ\text{C}$ , increased first at a rate of  $20^\circ\text{C}$  per min to  $130^\circ\text{C}$ , and next at a rate of  $4^\circ\text{C}$  min to the final temperature of  $320^\circ\text{C}$ , which was held for 10 min.

For quantification, samples were analyzed on an HP 6890 GC using a 50-m CP Sil-5 column (0.32-mm diameter, film thickness of 0.12  $\mu\text{m}$ ) and helium as the carrier gas. The oven program initiated at  $70^\circ\text{C}$  and increased by a rate of increased by  $20^\circ\text{C}$  per min to  $200^\circ\text{C}$  and next by a rate of  $3^\circ\text{C}$  per min until  $320^\circ\text{C}$ . The final temperature of  $320^\circ\text{C}$  was held for 25 min. Compound concentrations were determined by relating chromatogram peak areas to the concentration of the internal standard.

The  $U_{37}^k$  Index, defined as  $\text{C}_{37:2}/(\text{C}_{37:2} + \text{C}_{37:3})$ , was used to

estimate SSTs (14).  $U_{37}^k$  values were converted to SSTs by using the core top calibration of Müller et al. (15). Of the 193 samples analyzed, 74 were run in duplicate and 18 were run in triplicate.

The reproducibility of these analyses is always better than  $\pm 0.6$  °C.

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Table S1. Carbon isotopic values ( $\delta^{13}\text{C}$ ) of the  $\text{C}_{29}$  and  $\text{C}_{31}$   $n$ -alkanes of  $\text{C}_3$  plants

Plant	Type	$\delta^{13}\text{C}_{29}$ , $n = 50$	$\delta^{13}\text{C}_{31}$ , $n = 52$	Ref.
<i>Psidium cattleionum</i>	C3	-38.6	-38.8	4
<i>Jacobinia cornea</i>	C3	-36.1	-36.2	4
<i>Cyperus diffusus</i>	C3	-36.2	-36.5	4
<i>Dendrocalamus stricus</i>	C3	-35.3	-36.4	4
<i>Cyprus alternifolius</i>	C3	-34.2	-35.3	4
<i>Fagus sylvatica</i>	C3			4
<i>Acer campestre</i>	C3	-35.0	-35.0	4
<i>Magnolia delabayi</i>	C3	-34.0	-34.3	4
<i>Quercus turneri</i>	C3	-32.9	-35.7	4
<i>Quercus rober</i>	C3	-36.4	-36.9	4
<i>Euphorbia pulcherrima</i> Willd.	C3	-38.0	-37.4	5
<i>Codiaeum variegatum</i> (L.) Bl. Var. <i>pictum</i> M.-A. forma <i>crispum</i> Pax	C3		-35.1	5
<i>Ficus altissima</i> Bl.	C3	-33.9	-36.1	5
<i>Ficus microcarpa</i> Linn. f.	C3	-31.0	-33.4	5
<i>Osmanthus fragrans</i> Lour.	C3	-35.7	-37.0	5
<i>Kigelia africana</i> (am.) Benth.	C3	-33.1	-33.3	5
<i>Syzygium cumini</i> (L.) Skeels	C3	-37.2	-35.5	5
<i>Swietenia mahagoni</i> (L.) Jacq.	C3	-34.1	-35.9	5
<i>Pistia stratiotes</i>	C3	-36.6	-37.1	5
<i>Caryota mitis</i> Lour.	C3		-35.5	5
<i>Cinnamomum burmanni</i> (Nees) Bl.	C3	-33.3	-37.2	5
<i>Araucaria cunninghamii</i> Sweet	C3	-30.1	-30.5	5
<i>Alternanthera dentata</i> 'Rubiginosa'	C3	-36.6	-37.2	5
<i>Alternanthera versicolor</i> Regel	C3	-36.7	-37.5	5
<i>Alternanthera bettzickiana</i> (Regel) Nichols.	C3	-36.5	-37.2	5
<i>Holmskioldia sanguinea</i> Retz.	C3	-35.3	-33.8	5
<i>Quercus acutissima</i>	C3	-34.7	-34.8	6
<i>Camellia sasanqua</i>	C3	-31.3	-33.1	6
<i>Chamaecyparis obtusa</i>	C3	-30.6	-30.0	6
<i>Pinus thunbergii</i>	C3	-33.5	-34.1	6
<i>Colocasia esculenta</i>	C3	-33.2	-34.0	6
<i>Lycoris radiata</i>	C3	-28.0	-28.4	6
<i>Albizia julibrissin</i>	C3	-35.9	-37.8	6
<i>Benthamidia japonica</i>	C3	-36.5	-36.5	6
<i>Cryptomeria japonica</i>	C3	-32.3	-30.4	6
<i>Acer carpiniifolium</i>	C3	-35.5	-35.4	6
<i>Acer argutum</i>	C3	-35.9	-36.4	6
<i>Phrogmites communis</i>	C3	-34.6	-38.1	6
<i>Benthamidia japonica</i>	C3	-38.8	-37.3	6
<i>Prunus jamasakura</i>	C3	-34.2	-33.5	6
<i>Cryptomeria japonica</i>	C3	-32.9	-32.5	6
<i>Acer carpiniifolium</i>	C3	-37.3	-37.1	6
<i>Acer argutum</i>	C3	-35.6	-36.0	6
<i>Taraxacum officinale</i>	C3	-37.0	-36.4	6
<i>Plantago asiatica</i>	C3	-39.6	-39.8	6
<i>Artemisia princeps</i>	C3	-36.5	-35.2	6
<i>Acer palmatum</i>	C3	-40.5	-41.8	6
<i>Quercus mongolica</i>	C3	-33.4	-32.2	6
<i>Quercus dentata</i>	C3	-33.5	-34.4	6
<i>Manihot utilissima</i>	C3	-30.8	-32.0	6
<i>Bromus</i> sp.	C3	-35.7	-36	7
<i>Festuca orthophylla</i>	C3	-30.3	-31.3	7
<i>Festuca orthophylla</i>	C3	-31.2	-32.2	7
Average		-34.7	-35.2	
SD		2.6	2.6	

