

Biomarker records of late Neogene changes in northeast African vegetation

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ABSTRACT

Open savannah grasslands (dominated by C_4 plants) became a significant component of northeast African vegetation during the late Neogene. We present molecule-specific carbon isotopic measurements of terrestrial plant biomarkers preserved in marine sediments off northeast Africa that allow reconstruction of orbital-scale vegetation changes in short time windows over the past 9.4 m.y. The biomarker data show large-amplitude vegetation variability as early as 3.8 Ma, with the greatest C_4 expansion occurring after 3.4 Ma. We sampled orbital-scale oscillations of up to 5‰, almost as large as the observed late Neogene range of 7‰, suggesting that large and repeated oscillations between more open and more closed landscapes were an important aspect of northeast African vegetation change during the past 4 m.y.

Keywords: $\delta^{13}C$, carbon isotopes, biomarker, Africa, vegetation, C_4 .

INTRODUCTION

Aridity-controlled transitions between forest and grassland vegetation cover in northeast Africa during the late Neogene (Fig. 1A) represent a shift in the dominance of plants utilizing C_3 and C_4 photosynthetic pathways. C_3 and C_4 plants exhibit distinctive stable carbon isotopic ($\delta^{13}C$) signatures (O'Leary, 1981). The former, which include nearly all trees and shrubs, have bulk leaf tissue $\delta^{13}C$ of -31.4‰ to -24.6‰ , whereas $\delta^{13}C$ values of C_4 plants, mostly grasses adapted to water-limited or warm-season precipitation conditions, are within -14.1‰ to -11.5‰ in East Africa (Cerling et al., 2003). The range of isotopic values depends on water availability and on the openness of the canopy; C_3 plants in more open or water-stressed conditions display more enriched values (Cerling et al., 1997). Leaf wax lipids record isotopic differences produced by different photosynthetic pathways (Collister et al., 1994; O'Leary, 1981). These leaf wax lipids are

readily abraded from the plant leaf surface and transported by wind to marine sediments, where they are well preserved with a distribution and composition that is representative of vegetation source (Freeman and Colarusso, 2001; Schefuß et al., 2003a). Molecule-specific $\delta^{13}C$ measurements of marine sediments allow carbon exclusively from terrestrial plants to be sampled (Freeman and Colarusso, 2001), avoiding the problems of interpreting $\delta^{13}C$ of bulk organic carbon that may include carbon from multiple sources of varying isotopic composition (Pearson and Eglinton, 2000). Biologically specific marker molecules (biomarkers) diagnostic of a terrestrial plant leaf wax source include C_{24} - C_{32} n -alkanoic acids with an even chain length predominance (Eglinton and Hamilton, 1967; Kolattukudy, 1969). As shown in studies off West Africa (Huang et al., 2000; Schefuß et al., 2003a, 2003b), these biomarkers offer a promising tool for reconstructing African vegetation from marine sequences.

We present the first biomarker record of northeast African vegetation changes, using marine sediments from Deep Sea Drilling Project Site 231 recovered from the Gulf of Aden. Site 231, with a sedimentary record of environmental change spanning the late Neogene, is the closest marine site to the northeast African hominin fossil sites (Fig. 1B) that record critical periods of faunal and hominin evolution. Atmospheric circulation patterns suggest the principal vegetation sources to Site 231 are east of the Rift Valley, including the Horn of Africa and northern Ethiopia, with transport by strong, lower tropospheric, southwest monsoon winds during May to September (Kalnay et al., 1996) (Fig. 1A) and maximum dust activity recorded in May to July (Prospero et al., 2002). Weaker winter winds and upper tropospheric transport may contribute leaf wax lipids from other sources surrounding the northwestern Arabian Sea, although their lower vegetation density and weaker transports suggest that such contributions will be minor.

MATERIALS AND METHODS

To develop a biomarker record of late Neogene vegetation change in northeast Africa, we analyzed 100 samples in 9 time slices near 9.4,

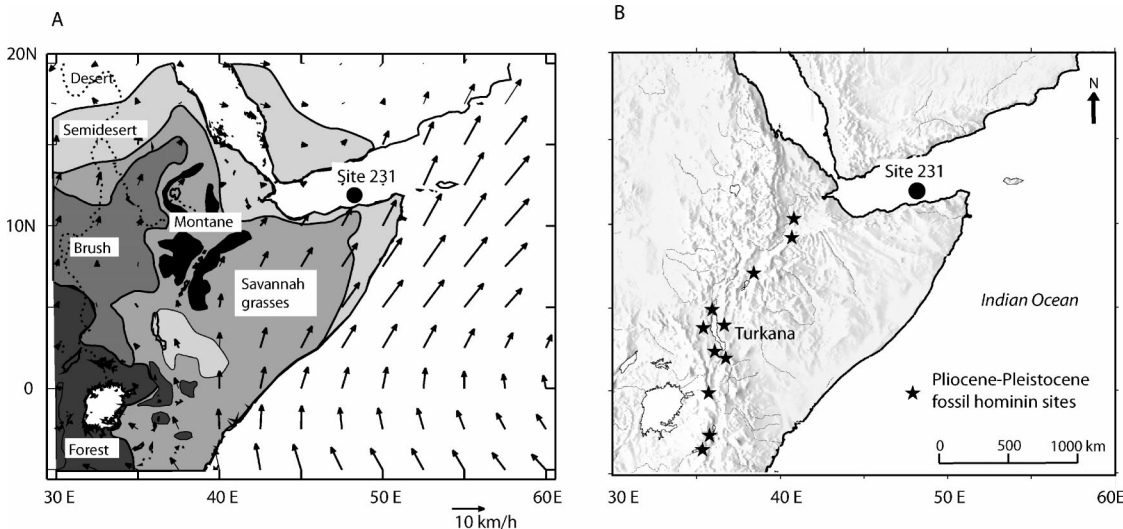


Figure 1. A: Location of Deep Sea Drilling Project Site 231 in relation to major present-day vegetation zones in northeast Africa (White, 1983) and average winds at 1000 hPa (Kalnay et al., 1996) during period (May to July) of maximum eolian dust production and transport from regional dust source areas (Prospero et al., 2002). **B:** Key northeast African hominin fossil sites and regional topography.

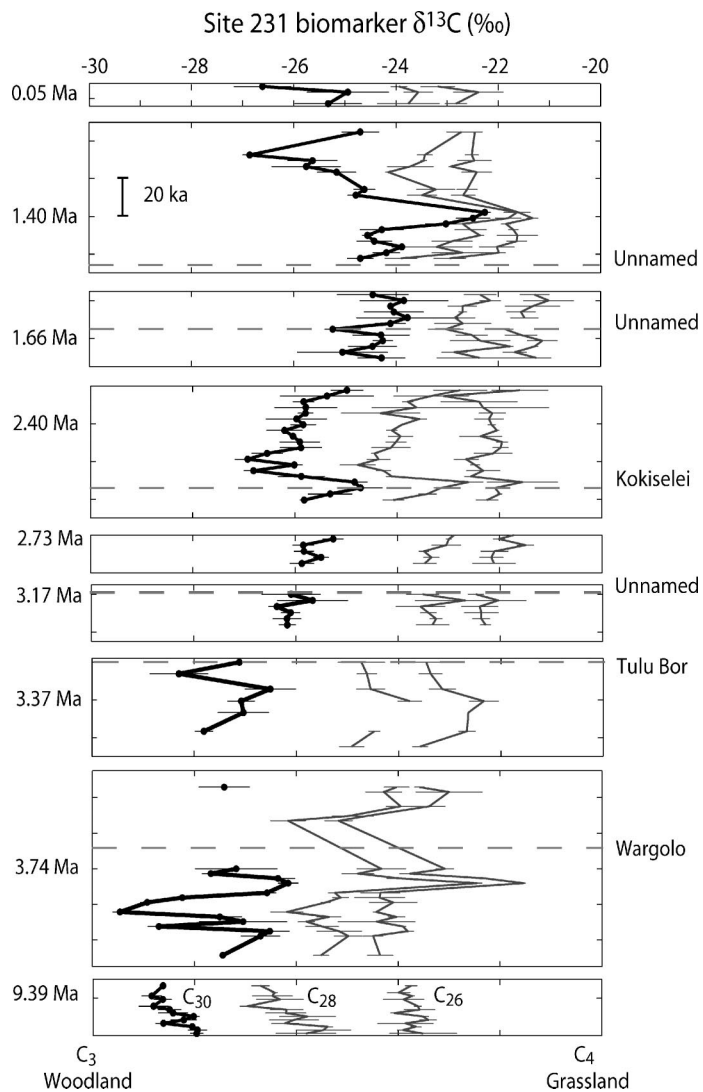


Figure 2. Carbon isotopic values of *n*-alkanoic acids from Deep Sea Drilling Project Site 231 for 9 20–100 k.y. time intervals spanning late Neogene (near 9.4, 3.8, 3.4, 3.2, 2.7, 2.4, 1.7, 1.4, and 0.1 Ma). Mean $\delta^{13}\text{C}$ and 1σ analytical errors are shown for C_{30} , C_{28} , and C_{26} homologs; inferred vegetation changes are based on C_{30} $\delta^{13}\text{C}$. Tephra age constraints are shown with dashed gray line (Brown et al., 1985; Sarna-Wojcicki et al., 1985). Foraminiferal $\delta^{18}\text{O}$ suggests interglacial timing of upper interval rather than glacial age (0.05 Ma) indicated by interpolated age model.

3.8, 3.4, 3.2, 2.7, 2.4, 1.7, 1.4, and 0.1 Ma, each of which spans 20–100 k.y. in duration and was sampled at 2–3 k.y. resolution to resolve orbital-scale variability (Fig. 2). Molecular-level biomarker isotopic data were measured in triplicate on prepared samples using an automated gas chromatograph–isotope ratio monitoring–mass spectrometer. Details of the method are in the GSA Data Repository.¹ Age control is provided by the published tephrostratigraphic age model for the core based on the geochemical correlation of five tephra layers within the core to widely dispersed eruptions recorded at hominin sites and one nannofossil datum (Brown et al., 1985; Rahman and Roth, 1989; Sarna-Wojcicki et al., 1985). Time slices were selected to take advantage of intervals where core recovery was optimal and where chronological tie points from tephra horizons were available (Brown et al., 1985; Mc-

Dougall et al., 1992; Sarna-Wojcicki et al., 1985). These time slices constrain the timing of vegetation changes relative to important hominin evolutionary events (Kimbel, 1995; Potts, 1998; Vrba, 1995) (Figs. 2 and 3).

RESULTS AND DISCUSSION

Molecular distributions (e.g., the carbon preference index) of putative leaf wax biomarkers show that for paleoecological reconstructions at Site 231, long-chain *n*-alkanoic acids are the most reliable proxies of terrestrial vegetation. C_{26} , C_{28} , and C_{30} *n*-alkanoic acids have molecular abundance distributions diagnostic of a plant leaf wax source and exhibit carbon isotopic values within the range of modern C_3 and C_4 vegetation end members. We focus on the C_{30} *n*-alkanoic acid for interpretation of the Neogene reconstruction because this homolog displays the largest dynamic range in carbon isotopic values (Fig. 2). While these differential offsets between homologs are not yet fully documented, they are consistent with evidence for physiological fractionation pathways in plant wax biosynthesis (Chikaraishi et al., 2004). Shorter chain lengths are also more likely to be influenced by marine algal contributions, whereas longer chain lengths are thought to be exclusively derived from terrestrial plants (Volkman et al., 1980).

Relatively invariant, ^{13}C -depleted isotopic compositions of the long-chain fatty acids from 12 ca. 9.4 Ma samples (Fig. 2) indicate a predominantly C_3 , woody or herbaceous vegetation community in the late Miocene. This stable C_3 composition persists despite color cyclicity in the core, recording significant environmental variability at that time, including changes in terrestrial biomarker abundance. The latter may reflect either greater primary productivity on land or enhanced transport of biomarkers to marine sediments. Any variability in C_3 floral composition or density prior to the 6–8 Ma emergence of C_4 vegetation in East Africa (Cerling et al., 1997) is expected to be undetected by isotopic techniques. In contrast to the isotopic stability reconstructed for this short interval of the Miocene at 9.4 Ma, biomarkers show large-amplitude isotopic variations on precessional time scales during a 100 k.y. interval in the mid-Pliocene (3.69–3.79 Ma). Isotopic compositions oscillate between values as depleted as the late Miocene to values 3‰ more enriched, indicating a more open, water-stressed or expanded C_4 vegetation component. These results reveal large-amplitude vegetation variability occurring as early as 3.8 Ma, prior to the onset of Northern Hemisphere glaciation and during a period when African climate was dominated by precessional-frequency wet-dry oscillations (deMenocal, 1995).

Although the biomarker data show that C_4 vegetation is present in the sampled interval between 3.69 and 3.79 Ma, the isotopic enrichment relative to 9.4 Ma is modest compared to the step-like 10‰ enrichment in fossil tooth enamel between 8 and 6 Ma in northeast Africa (Cerling et al., 1997). Tooth enamel $\delta^{13}\text{C}$ reflects the diet of a sample of mammals, and these records have been interpreted as showing a dramatic expansion of C_4 grasslands in northeast Africa at that time (Cerling et al., 1997). A more conservative interpretation would be that the large isotopic shift reflects dietary bias rather than a quantitative measure of vegetation replacement across the landscape (Cerling, 1992). Initial biomarker data suggest that while C_4 vegetation became a detectable component of northeast African vegetation by 3.8 Ma, its contribution was highly variable and represented a modest ~5% average increase in C_4 vegetation cover between the sampled intervals at 9.4 and 3.8 Ma. Initial reconciliation of these data sets implies that while C_4 plants expanded to become a significant dietary component for certain grazing mammals, they remained far from dominant components of the northeast African landscape through the mid-Pliocene, in agreement with soil carbonate $\delta^{13}\text{C}$ data (Cerling et al., 1993; Levin et al., 2004).

The molecular $\delta^{13}\text{C}$ record indicates that only in the late Pliocene

¹GSA Data Repository item 2005188, methods, is available online at www.geosociety.org/pubs/ft2005.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301-9140, USA.

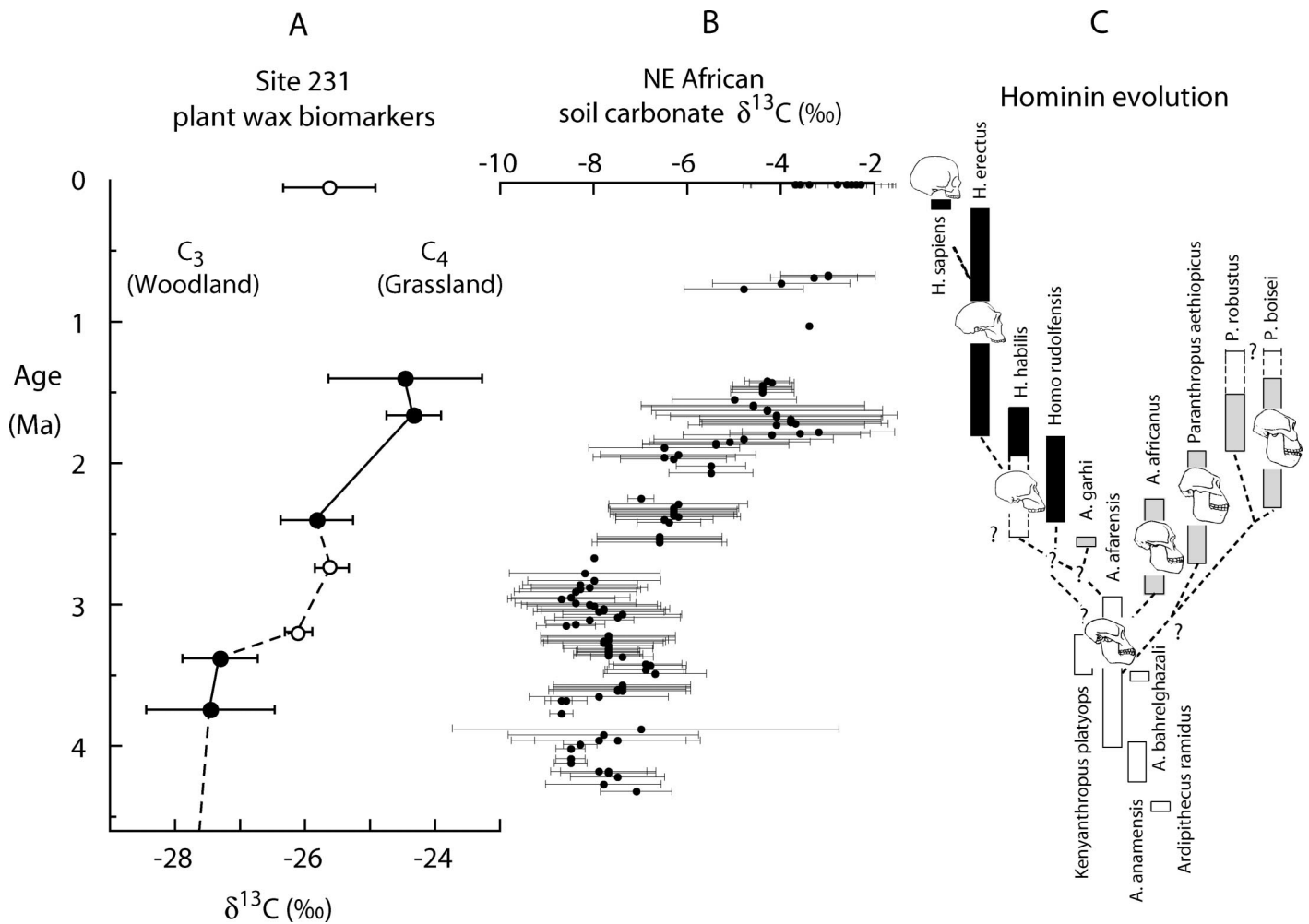


Figure 3. Summary figure comparing records of northeast African vegetation and hominin evolution. **A:** Interval means and (1σ) standard deviations of C_{30} *n*-alkanoic acid $\delta^{13}C$, for intervals >40 k.y. in duration (filled circles), and for intervals <40 k.y. in duration (open circles, dashed lines). **B:** Soil carbonate $\delta^{13}C$ from Turkana Basin (northern Kenya), means and (1σ) standard deviations from individual stratigraphic layers (Cerling, 1992; Cerling and Hay, 1986; Wynn, 2004). **C:** Phylogeny of major hominin lineages throughout Pliocene–Pleistocene (from sources in deMenocal, 2004).

to early Pleistocene did C_4 plants expand to be a dominant component of the landscape. Biomarker $\delta^{13}C$ shows an enrichment of $\sim 2\text{‰}$ between the 3.4 and 3.2 Ma time slices (Figs. 2 and 3A), slightly earlier than observed in the soil carbonate record (Fig. 3B). The C_4 expansion is approximately coincident with the onset of high-latitude glacial cycles (Shackleton, 1990) and related increase in subtropical North African aridity (deMenocal, 1995). Further C_4 expansion occurred between the 2.4 and 1.7 Ma time slices, indicating that progressively more open vegetation continued to spread well after the mid-Pliocene cooling and drying event. This continued grassland expansion is also detected in soil carbonate isotopic records (Fig. 3B) and may be linked to the 1.5–2.0 Ma onset of increased tropical Pacific Ocean temperature gradients (Ravelo et al., 2004; Wara et al., 2005), a surface ocean boundary condition known to directly affect East African rainfall (Godard and Graham, 1999).

Overall, the terrestrial biomarkers indicate a late Neogene carbon isotopic enrichment of 4‰ across the means of the 9 time slices (Fig. 3A), with variations between individual samples exceeding 7‰ , roughly equivalent to a 50% range in C_4 vegetation cover (Fig. 2). This range is comparable to the 6‰ range between means of soil carbonate $\delta^{13}C$ for a given time and site (Cerling, 1992; Cerling and Hay, 1986; Wynn, 2004) (Fig. 3B). Individual biomarker samples in this study span 3 k.y. and integrate a region-wide vegetation signal that is expected to dampen the isotopic range. Terrestrial soil carbonates record

a localized signal, as seen by the large standard deviations in isotopic values at each site; however, that signal is integrated over the long time scale of soil carbonate nodule formation. Given the different geochemical, spatial, and temporal bases for the soil carbonate and biomarker proxies, the records for the past 4 m.y. are remarkably consistent and together provide strong evidence for a late Pliocene shift toward a greater C_4 contribution, which commenced near the mid-Pliocene (by 3.2 Ma) and continued into the early Pleistocene (until at least 1.4 Ma).

In contrast to soil carbonates, $\delta^{13}C$ measurements on plant wax biomarkers preserved in marine sediments enable vegetation variations to be resolved at orbital time scales (Fig. 2). We find precessional-duration $\delta^{13}C$ cycles that capture nearly half of the full range of the late Neogene vegetation shift from 3.79 to 3.69 Ma in the mid-Pliocene. Isotopic values range over 3‰ , roughly equivalent to 20%–25% variability in C_4 vegetation cover. Longer duration and larger amplitude variability are observed in the sampled interval between 1.42 and 1.36 Ma, with very large (5‰) swings in $\delta^{13}C$, equivalent to a 30% range in C_4 vegetation cover (Fig. 2). In contrast, biomarker isotopes exhibit low-amplitude oscillations from 2.44 to 2.38 Ma (Fig. 2), an interval when large glacial climate cycles were already established at high latitudes, suggesting that northeast African vegetation may not have followed the high-latitude trend toward increasing variability during the Pliocene–Pleistocene. The data instead suggest that changes in northeast African vegetation were primarily related to the amplitude of

subtropical orbital insolation variations, since large-amplitude vegetation variability ca. 3.7 and ca. 1.4 Ma coincided with high orbital precession variability, and low variability ca. 2.4 Ma occurred during a precessional minimum.

CONCLUSIONS

Orbital-scale biomarker data provide new evidence for the timing and nature of northeast African vegetation change during the late Neogene. Our approach of high-resolution sampling in brief time slices provides a record of C₄ expansion consistent with soil carbonate and fossil tooth carbon isotopic data that in addition provides evidence for orbital-scale vegetation variability. Carbon isotopic compositions of leaf wax biomarkers indicate exclusively C₃ vegetation at 9.4 Ma, consistent with fossil tooth data. In the past 4 m.y., biomarker data show C₄ expansion principally after 3.4 Ma, slightly earlier than the observed shift in soil carbonate data. Our record demonstrates orbital-scale carbon isotopic variability as early as 3.8 Ma, demonstrating that large-amplitude shifts in the proportion of C₄ grasslands began prior to the onset of Northern Hemisphere glaciation during a period when African climate was dominated by precessional-frequency wet-dry oscillations (deMenocal, 1995). These initial biomarker records demonstrate that for fossil faunal and hominin species whose ranges typically spanned 10⁵–10⁶ yr, vegetation changes were experienced as large, abrupt, and high-frequency shifts in the landscape.

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REFERENCES CITED

- Brown, F., McDougall, I., Davis, T., and Maier, R., 1985, An integrated Plio-Pleistocene chronology of the Turkana Basin, *in* Delson, E., ed., *Paleoanthropology: The hard evidence*: New York, Alan R. Liss, p. 82–90.
- Cerling, T.E., 1992, Development of grasslands and savannas in East Africa during the Neogene: *Palaeogeography, Palaeoclimatology, Palaeoecology*, v. 97, p. 241–247.
- Cerling, T.E., and Hay, R.L., 1986, An isotopic study of paleosol carbonates from Olduvai Gorge: *Quaternary Research*, v. 25, p. 63–78.
- Cerling, T.E., Wang, Y., and Quade, J., 1993, Expansion of C₄ ecosystems as an indicator of global ecological change in the late Miocene: *Nature*, v. 361, p. 344–345.
- Cerling, T.E., Harris, J.M., MacFadden, B.J., Leakey, M.G., Quade, J., Eisenmann, V., and Erleringer, J.R., 1997, Global vegetation change through the Miocene/Pliocene boundary: *Nature*, v. 389, p. 153–158.
- Cerling, C., Harris, J., and Passey, B., 2003, Diets of East African Bovidae based on stable isotope analysis: *Journal of Mammalogy*, v. 84, p. 456–470.
- Chikaraishi, Y., Naraoka, H., and Poulson, S., 2004, Carbon and hydrogen isotopic fractionation during lipid biosynthesis in a higher plant (*Cryptomeria japonica*): *Phytochemistry*, v. 65, p. 323–330.
- Collister, J., Rieley, G., Stern, B., Eglinton, G., and Fry, B., 1994, Compound-specific $\delta^{13}\text{C}$ analyses of leaf lipids from plants with differing carbon dioxide metabolisms: *Organic Geochemistry*, v. 21, p. 619–627.
- deMenocal, P.B., 1995, Plio-Pleistocene African climate: *Science*, v. 270, p. 53–59.
- deMenocal, P.B., 2004, African climate change and faunal evolution during the Pliocene-Pleistocene: *Earth and Planetary Science Letters*, v. 220, p. 3–24.
- Eglinton, G., and Hamilton, R., 1967, Leaf epicuticular waxes: *Science*, v. 156, p. 1322.
- Freeman, K., and Colarusso, L., 2001, Molecular and isotopic records of C₄ grassland expansion in the late Miocene: *Geochimica et Cosmochimica Acta*, v. 65, p. 1439–1454.
- Goddard, L., and Graham, N.E., 1999, Importance of the Indian Ocean for simulating rainfall anomalies over eastern and southern Africa: *Journal of Geophysical Research*, v. 104, p. 19,099–19,116.
- Huang, Y., Dupont, L., Sarnthein, M., Hayes, J., and Eglinton, G., 2000, Mapping of C₄ plant input from North West Africa into North East Atlantic sediments: *Geochimica et Cosmochimica Acta*, v. 64, p. 3505–3515.
- Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G., Woolen, J., Zhu, Y., Chelliah, M., Ebisuzaki, W., Higgins, W., Janowiak, J., Mo, K., Ropelewski, C., Wang, J., Leetma, A., Reynolds, R., Jenne, R., and Joseph, D., 1996, The NCEP/NCAR 40 year reanalysis project: *American Meteorological Society Bulletin*, v. 77, p. 437–471.
- Kimbel, W., 1995, Hominid speciation and Pliocene climatic change, *in* Vrba, E., et al., eds., *Paleoclimate and evolution with emphasis on human origins*: New Haven, Connecticut, Yale University Press, p. 425–437.
- Kolattukudy, P., 1969, Plant waxes: Lipids, v. 5, p. 259–275.
- Levin, N., Quade, J., Simpson, S., Semaw, S., and Rogers, M., 2004, Isotopic evidence for Plio-Pleistocene environmental change at Gona, Ethiopia: *Earth and Planetary Science Letters*, v. 219, p. 93–110.
- McDougall, I., Brown, F.H., Cerling, T.E., and Hillhouse, J.W., 1992, A reappraisal of the geomagnetic polarity time scale to 4 Ma using data from the Turkana Basin, East Africa: *Geophysical Research Letters*, v. 19, p. 2349–2352.
- O’Leary, M., 1981, Carbon isotope fractionation in plants: *Phytochemistry*, v. 20, p. 553–567.
- Pearson, A., and Eglinton, T., 2000, The origin of n-alkanes in Santa Monica Basin surface sediment: A model based on compound-specific $\delta^{14}\text{C}$ and $\delta^{13}\text{C}$ data: *Organic Geochemistry*, v. 31, p. 1103–1116.
- Potts, R., 1998, Environmental hypotheses of hominin evolution: *Yearbook of Physical Anthropology*, v. 41, p. 93–136.
- Prospero, J., Ginoux, P., Torres, O., Nicholson, S., and Gill, T., 2002, Environmental characterization of global sources of atmospheric solid dust intensified with the Nimbus 7 Total Ozone Mapping Spectrometer (TOMS) absorbing aerosol product: *Reviews of Geophysics*, v. 40, doi:10.1029/2000RG000095.
- Rahman, A., and Roth, P.H., 1989, Late Neogene calcareous nannofossil biostratigraphy of the Gulf of Aden region: *Marine Micropaleontology*, v. 15, p. 1–27.
- Ravelo, A., Andreassen, D., Lyle, M., Olivarez-Lyle, A., and Wara, M., 2004, Regional climate shifts caused by gradual global cooling in the Pliocene epoch: *Nature*, v. 429, p. 263–267.
- Sarna-Wojcicki, A.M., Meyer, C.E., Roth, P.H., and Brown, F.H., 1985, Ages of tuff beds at East African early hominid sites and sediments in the Gulf of Aden: *Nature*, v. 313, p. 306–308.
- Scheffuß, E., Ratzmeyer, V., Stuut, J.B.W., Jansen, J.H.F., and Damste, J.S.S., 2003a, Carbon isotope analyses of n-alkanes in dust from the lower atmosphere over the central eastern Atlantic: *Geochimica et Cosmochimica Acta*, v. 67, p. 1757–1767.
- Scheffuß, E., Schouten, S., Jansen, J.H.F., and Damste, J.S.S., 2003b, African vegetation controlled by tropical sea surface temperatures in the mid-Pleistocene period: *Nature*, v. 422, p. 418–421.
- Shackleton, N.J., 1990, An alternative astronomical calibration of the lower Pleistocene timescale based on ODP Site 677: *Royal Society of Edinburgh Transactions, Earth Sciences*, v. 81, p. 251–261.
- Volkman, J., Johns, R.B., Gillan, F.T., Perry, G.J., and Bavor, H.J., 1980, Microbial lipids of an intertidal sediment—I. Fatty acids and hydrocarbons: *Geochimica et Cosmochimica Acta*, v. 44, p. 1133–1143.
- Vrba, E., 1995, The fossil record of African antelopes (Mammalia, Bovidae) in relation to human evolution and paleoclimate, *in* Vrba, E., et al., eds., *Paleoclimate and evolution with emphasis on human origins*: New Haven, Connecticut, Yale University Press, p. 385–424.
- Wara, M.W., Ravelo, A.C., and Delaney, M.L., 2005, Permanent El Niño-like conditions during the Pliocene Warm Period: *Science*, v. 309, p. 758–761.
- White, F., 1983, *Vegetation of Africa—A descriptive memoir to accompany the UNESCO/AETFAT/UNSO vegetation map of Africa*; Natural Resources Research Report XX; Paris, France, U.N. Educational, Scientific and Cultural Organization, p. 356.
- Wynn, J.G., 2004, Influence of Plio-Pleistocene aridification on human evolution: Evidence from paleosols of the Turkana Basin, Kenya: *American Journal of Physical Anthropology*, v. 123, p. 106–118.

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