



## Atmospheric circulation patterns during late Pleistocene climate changes at Lake Malawi, Africa

Bronwen L. Konecky <sup>a,\*</sup>, James M. Russell <sup>a</sup>, Thomas C. Johnson <sup>b,c</sup>, Erik T. Brown <sup>b,c</sup>, Melissa A. Berke <sup>b,c</sup>, Josef P. Werne <sup>b,d</sup>, Yongsong Huang <sup>a</sup>

<sup>a</sup> Department of Geological Sciences, Brown University, Box 1846, Providence, RI 029012, United States

<sup>b</sup> Large Lakes Observatory, University of Minnesota Duluth, Duluth, MN 55812, United States

<sup>c</sup> Department of Geological Sciences, University of Minnesota Duluth, Duluth, MN 55812, United States

<sup>d</sup> Department of Chemistry & Biochemistry, University of Minnesota Duluth, Duluth, MN 55812, United States

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### ABSTRACT

The climate of tropical Africa transitioned from an interval of pronounced, orbitally-paced megadroughts to more humid and stable conditions approximately 70,000 years ago (Scholz et al., 2007). The regional atmospheric circulation patterns that accompanied these climatic changes, however, are unclear due to a paucity of continental paleoclimate records from tropical Africa extending into the last interglacial. We present a new 140-kyr record of the deuterium/hydrogen isotopic ratio of terrestrial leaf waxes ( $\delta D_{wax}$ ) from drill cores from Lake Malawi, southeast Africa, that spans this important climatic transition.  $\delta D_{wax}$  shifts from highly variable and relatively D-depleted to more stable and D-enriched around 56 ka, contemporary with the onset of more humid conditions in the region. Moisture source and transport history dominate the  $\delta D_{wax}$  signal at Lake Malawi, with local rainfall amount playing a secondary role for much of the paleorecord. Analysis of modern moisture sources for Lake Malawi suggests that D-depletion of waxes during the megadroughts may have been caused by an enhanced contribution of the drier, D-depleted air mass currently located in central southern Africa to the Lake Malawi catchment. This D-depleted air mass is associated with the descending limb of the Hadley cell, which implies significant changes in the Hadley circulation during the megadroughts and related changes in the position of the Intertropical Convergence Zone over Africa. These findings demonstrate the ability of  $\delta D_{wax}$  to serve as an atmospheric tracer when used in conjunction with additional proxy records for moisture balance, and elucidate potential mechanisms for pronounced hydrological change in southeast Africa during the late Pleistocene.

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### 1. Introduction

Variations in the climate of East Africa likely influenced the migration of our hominin ancestor populations during the late Pleistocene (Cohen et al., 2007), and threaten to impact highly vulnerable human populations today. However, the causes of East African climate changes and the relationship between African hydrologic change and global temperature increases remain poorly understood. On millennial to orbital timescales, droughts in many tropical regions correspond to high latitude temperature patterns, insolation forcing, and associated migrations of the Intertropical Convergence Zone (ITCZ) and its corresponding tropical rain belt (Wang et al., 2006). Recent paleoclimate records from Southeast Africa suggest that its climate exhibits more complex behavior driven by interactions between the African and Indian monsoon systems and zonal sea surface temperature (SST) patterns

(Barker and Gasse, 2003; Tierney et al., 2008). To further elucidate the patterns of paleoclimate variations in the region, we present new compound-specific stable isotope data from Lake Malawi, situated at the southernmost extent of the ITCZ over East Africa and just east of the Congo Air Boundary (CAB) that divides the Indian and Atlantic Ocean monsoon systems.

Southeast Africa is a climatically complex sub-region of the continent. Studies of modern climate variability reveal that interannual precipitation anomalies across Africa follow spatially diverse modes (Nicholson, 1986). Southeast African precipitation anomalies can either be in phase or out of phase with those of equatorial tropical East Africa, including the rest of the Great Lakes region. The lateral boundary between the positively and negatively correlated regions of East Africa lies anywhere from 10° to 20°S—between Lakes Malawi and Tanganyika, or even bisecting Lake Malawi itself (e.g. Nicholson, 1986; Ropelewski and Halpert, 1987). This has been referred to by some authors as the “meteorological equator” or the “climatic hinge zone” dividing tropical and subtropical East Africa (Barker et al., 2002; Garcin et al., 2006).

\* Corresponding author. Tel.: +1 401 863 2810; fax: +1 401 863 2058.

E-mail address: [bronwen\\_konecky@brown.edu](mailto:bronwen_konecky@brown.edu) (B.L. Konecky).

The spatial complexity observed in the instrumental record is also reflected in the paleoclimatology of the region. Paleoclimate records from Southeast Africa show significant disagreement over the nature of hydrological change over the last ~60 kyr. For example, during the Last Glacial Maximum (LGM) various records from Lake Malawi indicate relative aridity (Castañeda et al., 2007; Johnson et al., 2002), yet pollen assemblages yield little indication of difference between modern and LGM moisture balance (Beuning et al., 2011; DeBusk, 1998), while paleolimnological records from sites to the north provide overwhelming evidence for a dry LGM (Gasse et al., 2008, and references therein). These complexities make it difficult to assess the controls on long-term precipitation change in southeast Africa. Many paleolimnological records from Lake Malawi such as  $\text{CaCO}_3$  precipitation and benthic/epibenthic ostracod assemblages, show little change during the LGM (Scholz et al., 2007). However, these proxies are sensitive to intense aridity leading to closed basin conditions, but not to wet or moderately dry conditions, highlighting the need for a proxy that records the full range of climatic variations.

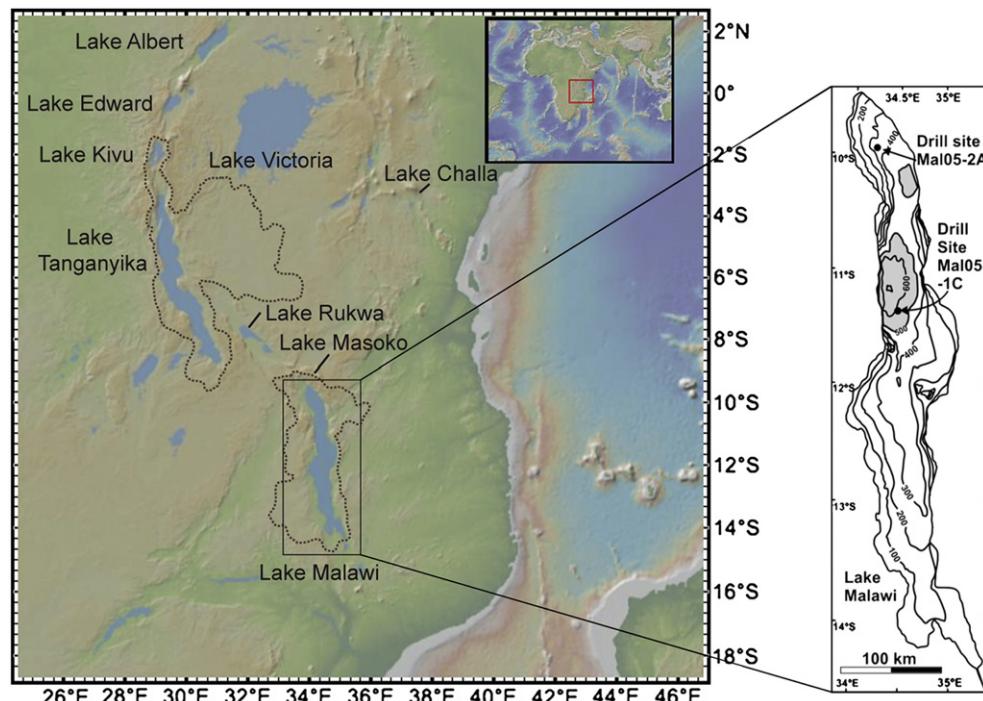
This study presents a 140,000-year record of leaf wax  $\delta\text{D}$  ( $\delta\text{D}_{\text{wax}}$ ) from Lake Malawi (Fig. 1).  $\delta\text{D}_{\text{wax}}$  measures the deuterium/hydrogen isotopic ratio of terrestrial leaf waxes found in lake sediments, and has been shown to record variations in the  $\delta\text{D}$  of precipitation ( $\delta\text{D}_{\text{precip}}$ ) through time in many systems (Hou et al., 2008; Huang et al., 2004). Observational data and isotope-enabled climate models indicate that  $\delta\text{D}_{\text{precip}}$  reflects the integrated atmospheric history of a water parcel, including source region, transport history, and convective, evaporative, and distillation processes (Dansgaard, 1964; LeGrande and Schmidt, 2009; Vuille et al., 2005). Here we examine the relative influence of these controls on  $\delta\text{D}_{\text{wax}}$  in the region by using the constraints of previous paleohydrological reconstructions, thereby providing insight into the climatic reorganizations that have taken place over the last 140 kyr. These results demonstrate the utility of  $\delta\text{D}_{\text{wax}}$  to delineate changes in atmospheric circulation patterns in addition to hydrologic history.

## 2. Methods

### 2.1. Sample preparation and analysis

The Lake Malawi Drilling Project recovered cores GLAD7-MAL05-2A and GLAD7-MAL05-1C in 2005 from 361 m and 593 m water depth in Lake Malawi's northern and central basins, respectively (MAL05-2A: 10°01.06' S, 34°11.16' E; MAL05-1C: 11°17.66 S, 34°26.15 E; Cohen et al., 2007). A multi-centennial resolution record (average 320 years) was constructed using the 76 kyr of nearly continuous sedimentation from MAL05-2A (Scholz et al., 2007), and a multi-millennial resolution  $\delta\text{D}_{\text{wax}}$  record (average 2640 years) was constructed from core MAL05-1C to examine longer-term  $\delta\text{D}_{\text{wax}}$  trends since 140 ka.

Sediment samples were freeze-dried and homogenized, and lipids were extracted from approximately 1 g dry sediment using a DIONEX Accelerated Solvent Extractor with dichloromethane:methanol (9:1). The lipid extract was separated into neutral and acid fractions using dichloromethane:isopropanol (2:1) and ether:acetic acid (96:4) over an aminopropyl silica gel column. Acid fractions were methylated using 5% acetyl chloride in methanol of a known isotopic composition, and fatty acid methyl esters (FAMEs) were further purified via silica gel chromatography. The  $\delta\text{D}$  of the  $\text{C}_{28}$  *n*-alkanoic acid, the dominant homologue in all samples, was measured using gas chromatography-pyrolysis-isotope ratio-mass spectrometry at Brown University. Samples were run in duplicate or triplicate to achieve precision of less than 3‰ with a  $1-\sigma$  error of 2.25‰ determined from standards.  $\delta\text{D}_{\text{wax}}$  values are reported relative to Vienna Standard Mean Ocean Water, and have been corrected for the methyl group added during methylation.  $\delta\text{D}_{\text{wax}}$  is also corrected for ice volume effects on the  $\delta\text{D}$  of seawater by assuming the ocean at the LGM was 1‰ more enriched in  $\delta^{18}\text{O}$  than the present (Schrag et al., 1996), and scaling this shift to patterns of ice volume changes represented in the LR04 benthic isotope stack (Lisiecki and Raymo, 2005) using the D-O relationship of global meteoric water.

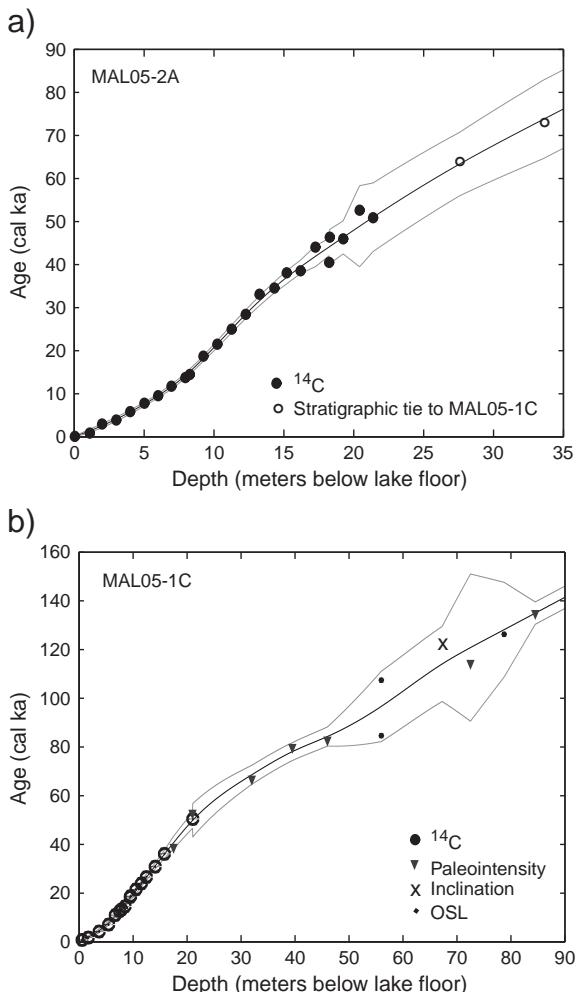


**Fig. 1.** Location of Lake Malawi (black box) and other lakes within the Great Rift Lakes region of East Africa discussed in the text, and location of drill sites MAL05-2A and MAL05-1C (after Scholz et al., 2007 and <http://www.geomapapp.org>). The dashed lines denote the approximate hydrological catchment areas of Lakes Malawi and Tanganyika (after Otu et al., 2011, and Bergonzini et al., 1997).

The  $\delta^{13}\text{C}$  of the  $\text{C}_{28}$  *n*-acid was measured on 11 randomly selected samples via gas chromatography/isotope-ratio mass spectrometry at the Large Lakes Observatory/University of Minnesota, Duluth.  $\delta^{13}\text{C}$  values are reported relative to Vienna Pee Dee Belemnite (VPDB) and have been corrected for the methyl group added during methylation.

## 2.2. Age model

The age model for MAL05-1C is based on 16 radiocarbon dates as well as optically-stimulated luminescence and paleomagnetic ties for sediments outside the radiocarbon dating range (Scholz et al., 2007). The age model for MAL05-2A (Brown et al., 2007) is based on 24 radiocarbon dates covering the past 50 kyr, and stratigraphic tie points to core MAL05-1C in the central basin for older sediments. We applied an updated mixed-effect regression age model to these data in order to quantify age model uncertainty (Heegaard et al., 2005), which is between 52 and 4799 years in MAL05-2A and between 96 and 15,412 years in MAL05-1C (Fig. 2). The age model is reasonably linear from 0 to 20 ka in MAL05-2A and 0–50 ka in MAL05-1C, with lower sedimentation rates in older sediments due to compaction. This age model differs slightly from previously published polynomial age models; the largest difference is at the base of core MAL05-1C, which is up to 3500 years younger than that of



**Fig. 2.** Age models for (a) MAL05-2A and (b) MAL05-1C based on a mixed-effect regression (Heegaard et al., 2005). Age control points are taken from previously published age model data in Brown et al., 2007 (MAL05-2A) and Scholz et al., 2007 (MAL05-1C). Gray lines represent lower and upper age limits. Note that errors at the base of MAL05-1C are underestimated due to a lack of error estimates on basal age control points.

Cohen et al., 2007. These differences, however, fall well within age model uncertainties.

## 3. Results and discussion

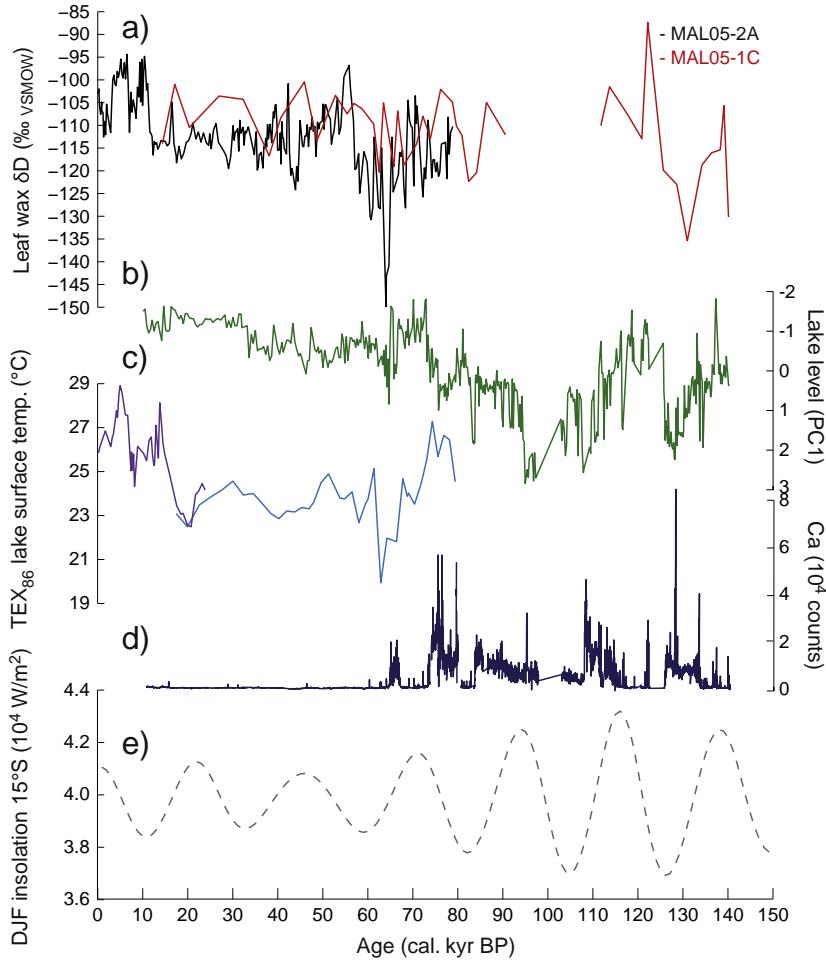
### 3.1. Long-term trends in $\delta\text{D}_{\text{wax}}$ and the East African megadroughts

The  $\delta\text{D}_{\text{wax}}$  records from both MAL05-2A and MAL05-1C show a long-term trend toward D-enriched leaf waxes with decreasing age, as well as a reduction in  $\delta\text{D}_{\text{wax}}$  variance around the Marine Isotope Stage (MIS) 4/3 boundary ~60 ka (Fig. 3). MAL05-1C  $\delta\text{D}_{\text{wax}}$  varies between  $-135.4\text{\textperthousand}$  and  $-87.4\text{\textperthousand}$ , with variations on the order of  $\sim 20\text{--}50\text{\textperthousand}$  through MIS 4–5, and variations on the order of  $\sim 10\text{--}15\text{\textperthousand}$  through MIS 2–3. The higher resolution MAL05-2A  $\delta\text{D}_{\text{wax}}$  record is marked by high variability prior to 56 ka, with a mean  $\delta\text{D}_{\text{wax}}$  of  $-118.6\text{\textperthousand}$  ( $1\sigma = 8.8\text{\textperthousand}$ ) and millennial-scale excursions of  $\sim 15\text{--}40\text{\textperthousand}$ . From 56 ka to present Lake Malawi  $\delta\text{D}_{\text{wax}}$  varies less, with quasi-periodic shifts on the order of  $10\text{--}20\text{\textperthousand}$  and a mean  $\delta\text{D}_{\text{wax}}$  of  $-109.9\text{\textperthousand}$  ( $1\sigma = 6.3\text{\textperthousand}$ ). The decrease in the amplitude of variability after 56 ka is accompanied by a shift toward more positive values.  $\delta\text{D}_{\text{wax}}$  is moderately D-depleted and less variable during MIS 2 approximately 30 ka–11.5 ka, with an average of  $-113.8\text{\textperthousand}$  ( $1\sigma = 2.6\text{\textperthousand}$ ).  $\delta\text{D}_{\text{wax}}$  during the Holocene is significantly more enriched than majority of the record, with an average of  $102.8\text{\textperthousand}$  ( $1\sigma = 4.9\text{\textperthousand}$ ).

The  $\delta\text{D}_{\text{wax}}$  transition at 56 ka occurs amidst a well-documented transition in tropical African climate from a series of pronounced, orbital-scale megadroughts to a period of relative climate stability (Cohen et al., 2007; Johnson et al., 2011; Lyons et al., 2011; Scholz et al., 2007). This transition is attributed to a shift toward lower eccentricity, which weakened precessional insolation forcing, resulting in a relatively stable hydrologic cycle compared to the high amplitude variability from 145 ka to 60 ka. At Lake Malawi, evidence from geophysical, geochemical, and paleoecological studies suggests that the transition out of the megadroughts interval progressed in several stages from ~75 ka to ~30 ka. Seismic stratigraphy and authigenic calcite preservation delineate at least two initial, multi-millennial shifts to higher lake levels and more hydrologically open conditions at ~75 ka and ~65 ka, returning to more closed-basin conditions in between (Brown et al., 2007; Scholz et al., 2007). The  $\delta\text{D}_{\text{wax}}$  transition occurs at 56 ka, after these fluctuations, and is followed by a pronounced lake level rise to near-modern levels between 50 and 35 ka based on seismic data (Scholz et al., 2007). The onset of increasingly stable conditions took place around 31 ka, according to paleoecological indicators (Stone et al., 2011). This sequence of events indicates that the 56 ka  $\delta\text{D}_{\text{wax}}$  transition records a climatic adjustment that followed the initial onset of hydrologically open, but still highly variable, conditions, but preceded and likely influenced the switch to more stable and wet conditions. Increasing stability was likely due to decreasing orbital eccentricity, which reached a minimum around 45 ka.

Interestingly, the dominant mode of the  $\delta\text{D}_{\text{wax}}$  variability is millennial in scale, a trend that persists through the Holocene despite comparatively low orbital eccentricity. The high amplitude of millennial variations prior to 56 ka in the Malawi basin contrasts with isotopic records from other monsoon regions that show high amplitude millennial-scale variations during MIS 3 (30–60 ka; Wang et al., 2001), and suggests that the orbital configuration that influenced Southeast African climate instability produced climate instability at a range of timescales.

Although our  $\delta\text{D}_{\text{wax}}$  record thus supports and augments our understanding of this transition in southeast African climate, the modest shift toward more D-enriched  $\delta\text{D}_{\text{wax}}$  at 56 ka appears inconsistent with numerous paleolimnological and paleoecological studies at Lake Malawi showing that the average lake level rose over 300 m after the termination of the East African megadroughts. Many tropical studies interpret fluctuations in  $\delta\text{D}_{\text{wax}}$  to primarily reflect local rainfall amount, based on seminal observations of the enormous influence



**Fig. 3.** (a) Lake Malawi  $\delta D_{\text{wax}}$  (MAL05-2A, black; MAL05-1C, red); (b) lake level (high lake level oriented up; Stone et al., 2011); (c) TEX<sub>86</sub> lake surface temperature (purple, Powers et al., 2005; blue, Woltering et al., 2011); (d) Ca counts measured by X-ray fluorescence (Scholz et al., 2007); (e) integrated mean Dec–Feb insolation at 15°S (Huybers and Eisenman, 2006). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

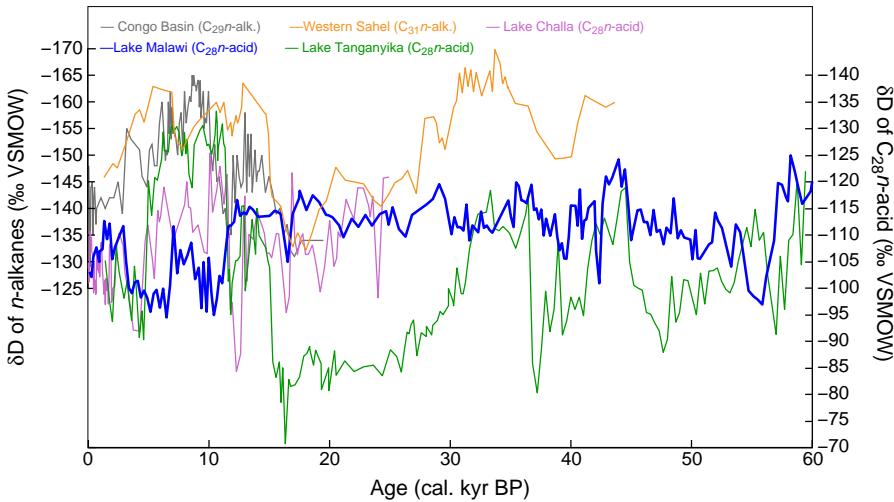
of the amount effect on  $\delta D_{\text{precip}}$  at tropical rainfall stations (e.g. Dansgaard, 1964). This process should yield D-enriched waxes during arid periods and D-depleted waxes during rainy periods. Although the exact timing of the transition from lowstand to moderate highstand at Lake Malawi during the late Pleistocene has not been constrained, the lake level rise itself has been robustly demonstrated using seismic reflection profiles and sedimentological changes in core MAL05-2A, which penetrates to the depth of an exposure surface marked by nearshore sands deposited after a severe lowstand prior to 76 ka (Lyons et al., 2011). The long-term enrichment in  $\delta D_{\text{wax}}$  with the onset of wetter conditions indicates that the amount effect on the  $\delta D$  of precipitation is likely to be a secondary influence on  $\delta D_{\text{wax}}$  at Lake Malawi, with other processes, most likely changes in moisture transport history, being the dominant control on  $\delta D_{\text{wax}}$ .

Though long  $\delta D_{\text{wax}}$  records from Africa are scarce, comparison of the Lake Malawi  $\delta D_{\text{wax}}$  record to the few existing reconstructions indicates significant differences in trend and amplitude among the records. Records from Lake Tanganyika to the north of Lake Malawi, Lake Challa on Mount Kilimanjaro, and the Congo Basin in central Africa show D-enriched waxes during the LGM, D-depleted waxes in the early Holocene, and D-enriched waxes in the late Holocene (Schefuß et al., 2005; Tierney et al., 2008, 2011), consistent with the dry glacial, wet early Holocene, and dry late Holocene moisture history for tropical Africa north of Lake Malawi (Gasse, 2000; Fig. 4). Lake Malawi  $\delta D_{\text{wax}}$  differs considerably from this, with relatively little orbital-scale variability and moderately D-depleted waxes during the LGM compared to the Holocene. Moreover, the amplitude of variability in Lake Malawi  $\delta D_{\text{wax}}$  is

relatively low for the region (Fig. 4). In the last 60 kyr,  $\delta D_{\text{wax}}$  at Lake Malawi varies by approximately 30‰ while  $\delta D_{\text{wax}}$  at Lake Tanganyika varies by ~60‰ (Tierney et al., 2008). In the last 20 kyr,  $\delta D_{\text{wax}}$  from the Congo Basin and Lake Challa vary by approximately 30‰ and 40‰, respectively (Schefuß et al., 2005; Tierney et al., 2011), higher than Lake Malawi's ~25‰ but much lower than the 60‰ at Lake Tanganyika during the same time period. These data suggest regionally diverse changes in, and likely controls on,  $\delta D_{\text{precip}}$  over the last 60 kyr, with Lake Malawi and Lake Tanganyika representing very different isotopic sensitivities to hydrological change. Absolute  $\delta D_{\text{wax}}$  values for Lakes Malawi, Tanganyika, and Challa – all measured on the C<sub>28</sub> n-acid – converge around 4.2 ka, indicating that  $\delta D_{\text{precip}}$  in the late Holocene is more regionally homogeneous than during the late Pleistocene and early Holocene (see Section 3.5).

### 3.2. Controls on $\delta D_{\text{wax}}$ at Lake Malawi

Our findings indicate that the long-chain fatty acids in Lake Malawi sediments are derived from terrestrial plants, particularly the C<sub>28</sub> n-acid on which our D/H isotopic measurements are based. The average  $\delta^{13}\text{C}$  of C<sub>28</sub> n-acid from 11 randomly selected samples is −32.08‰ (ranging −28.41‰ to −34.98‰), typical for fatty acids derived from terrestrial plant material (Chikaraishi and Naraoka, 2007, and references therein). The average chain length of long-chain n-acids (C<sub>24</sub>–C<sub>34</sub> n-acid) in our study ranges between 26.9 and 28.4, also typical for terrestrial plant waxes (Eglinton and Hamilton, 1967) and similar to the range of ~26 to ~28.5 reported for C<sub>23</sub>–C<sub>33</sub> plant wax n-alkanes in Malawi (Castañeda



**Fig. 4.**  $\delta D_{\text{wax}}$  from late Quaternary African records, measured from  $C_{28}$  *n*-acid from Lakes Malawi (blue), Tanganyika (green; Tierney et al., 2008), and Challa (purple; Tierney et al., 2011); *n*- $C_{29}$  alkane from the Congo Basin (gray; Schefuß et al., 2005); and  $C_{31}$  *n*-alkane from the western Sahel (orange; Niedermeyer et al., 2010). Note that the Y-axes are reversed, and  $\delta D_{\text{wax}}$  measured from *n*-alkanes and *n*-acids are plotted on separate y-axes. These axes are offset to account for the ~25% D-depletion of *n*-alkanes relative to their corresponding carbon-numbered *n*-alkanoic acids (e.g.  $C_{27}$  *n*-alkane relative to  $C_{28}$  *n*-acid; Chikaraishi and Naraoka, 2007). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

et al., 2009). These conclusions are in agreement with previous studies that found that the  $\delta^{13}\text{C}$  of long-chain *n*-alkanes ( $\delta^{13}\text{C}_{\text{alk}}$ ) reflected regional terrestrial vegetation shifts and moisture balance at Lake Malawi (Castañeda et al., 2007), and that  $\delta^{13}\text{C}_{\text{alk}}$  was closely related to variations in terrestrially derived lignin phenols and to bulk  $\delta^{13}\text{C}$  (Castañeda et al., 2009), affirming the integrity of long-chain leaf wax compounds in this system. Feakins et al. (2007) suggested a potential bacterial source for the  $C_{28}$  *n*-acid in lake sediments from Lake Turkana, Kenya/Ethiopia, but this source has yet to be confirmed in lacustrine sediments in East Africa or in culture studies, and our results indicate little, if any, bacterial contamination in Lake Malawi waxes. If present at Lake Malawi, bacterial or other microbial sources would likely be minor compared to terrestrial inputs from the large catchment (Volkman et al., 1998). The close relationship between *n*-alkane ACL and other terrestrial vegetation proxies (Castañeda et al., 2009) also indicates that reworking of fossil waxes from the landscape is not likely a major source of error on the millennial to orbital timescales addressed in this study, as has been recently suggested for marine sediments in the Cariaco and Santa Barbara basins (Drenzek et al., 2009; Li et al., 2009, 2011).

Changes in ambient air temperature and biosynthetic processes are not likely to significantly influence long-term trends or millennial excursions in  $\delta D_{\text{wax}}$ . The influence of temperature on  $\delta^{18}\text{O}$  and  $\delta\text{D}$  is relatively weak at the high temperatures observed in the tropics (Rozanski et al., 1993; Vuille et al., 2005), and temperature has been shown to have only a minor impact on seasonal  $\delta^{18}\text{O}$  and  $\delta\text{D}$  in low-latitude monsoon regions such as New Delhi (Rozanski et al., 1993). Even during the glacial period, temperature fluctuations at Lake Malawi were too small to significantly alter the precipitation  $\delta\text{D}$  signal. For example, the largest change in  $\text{TEX}_{86}$  lake surface temperature prior to the deglaciation is a 4 °C cooling from 64.6 to 60.8 ka (Woltering et al., 2011); the effect of an equivalent change in air temperature on the kinetic D/H fractionation factor is <6‰, too small to explain the observed 20–40‰  $\delta D_{\text{wax}}$  fluctuations (Majoube, 1971).

We estimate the biosynthetic apparent fractionation  $\epsilon$  between precipitation and the  $C_{28}$  *n*-acid to be ~85‰. This estimate is based on modern precipitation at northern Lake Malawi, which is approximately −17‰ according to an interpolation between isotopic measurements of precipitation (Bowen and Revenaugh, 2003), and leaf wax  $\delta\text{D}$  values from the uppermost sediment samples at Lake Malawi of approximately −102‰. Little is known about the apparent fractionation ( $\epsilon$ ) between precipitation and the  $C_{28}$  *n*-acid in African ecosystems, but ~85‰ is comparable to  $\epsilon_{\text{precip}-C28}$  estimates of ~99‰ ±

8‰ from arid and semi-arid regions of the southwestern U.S., with slightly lower values corresponding to areas with lower relative humidity (Hou et al., 2008), such as the modern semi-arid Malawi basin. We cannot rule out an effect of changing vegetation on the degree of D/H fractionation during fatty acid biosynthesis, as grass pollen fluctuates throughout the record (Beuning et al., 2011). However, the recently documented ~40‰ difference in apparent fractionation of fatty acids synthesized by trees and grasses, including both C3 and C4 varieties (Gao et al., 2011; Hou et al., 2008), would necessitate a total replacement of woody species with grasses—a shift not supported by fossil pollen data (Beuning et al., 2011) except between 90.6 and 111.8 ka when leaf wax abundance was too low for isotopic analysis. Moreover, the ACL of  $C_{24}$ – $C_{34}$  *n*-acids, which can indicate changes in the type of vegetation producing long chain fatty acids in leaf waxes, is not significantly correlated with  $\delta D_{\text{wax}}$  in our record ( $R^2 = 0.028$ ), indicating that changes in the type of vegetation producing waxes are not the primary control on the  $\delta D_{\text{wax}}$ .

Thus, the Lake Malawi  $\delta D_{\text{wax}}$  likely reflects the  $\delta\text{D}$  of precipitation and associated changes in precipitation and vapor transport, not biosynthetic or temperature effects. The few instances of discrepancies in  $\delta D_{\text{wax}}$  values between coring sites 2A and 1C likely reflect inter-basinal variations in water balance or leaf wax sources due to the complex topography around the north basin of Lake Malawi and the central basin's sensitivity to strong southerly winds, which have been shown in previous studies to affect inter-basinal variations in pollen deposition (DeBusk, 1997).

### 3.3. Regional controls on the $\delta\text{D}$ of precipitation at Lake Malawi

The most likely explanation for the more depleted  $\delta D_{\text{precip}}$  during arid paleoclimatic conditions at Lake Malawi is a change in the source area and transport history of moisture advected into the Malawi watershed. Recent studies with isotope-enabled general circulation models have confirmed that changes in moisture source region can be a significant component of  $\delta D_{\text{precip}}$  in Africa (Lewis et al., 2010; Tierney et al., 2011). Sources of continental precipitation in tropical and southeastern Africa are a complex mixture of Indian Ocean, Atlantic Ocean, and recycled continental moisture (Gimeno et al., 2010). Three main air masses converge over modern-day Southeast Africa during the austral summer rains (November–February). Vapor derived from the Northern Indian Ocean travels in a moderately humid air mass borne by the winter Indian monsoon. Southeasterly

winds transport vapor from the southern Indian Ocean, although this air mass loses much of its moisture in the Madagascar highlands before reaching the main African continent. Northwesterly winds carry extremely humid and unstable air from the Atlantic and the Congo basin, converging with Indian Ocean vapor at the CAB adjacent to Lake Malawi during austral summer (McGregor and Nieuwolt, 1998; Fig. 5).

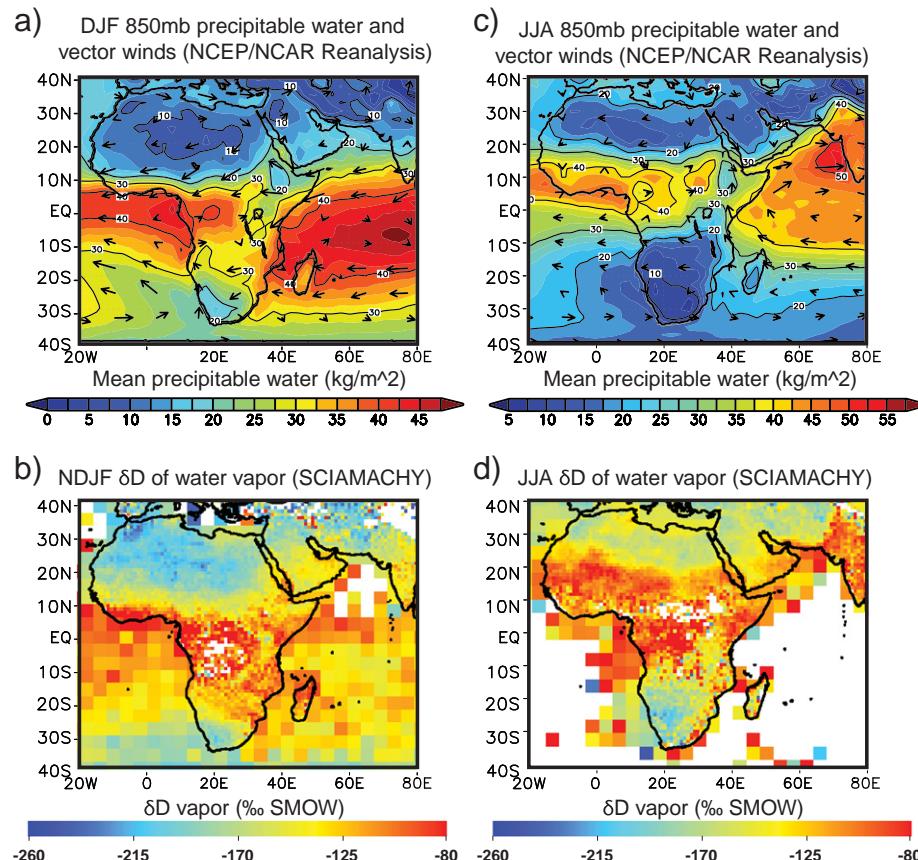
Modern H-isotopic composition of precipitation from these three moisture sources differs considerably (data retrieved from <http://ndds121.iaea.org/wiser/>). Mean December–February (DJF) precipitation at Dar-es-Salaam, Tanzania, approximately 650 km northeast of Lake Malawi, has a  $\delta D_{\text{precip}}$  of  $-10$  to  $-20\text{\textperthousand}$ , derived mostly from the winter Indian monsoon. Modern mean DJF precipitation at Ndola, Zambia, southwest of Lake Malawi's north basin, is derived from a mixture of southeasterly and northwesterly sources and has a  $\delta D_{\text{precip}}$  of  $-37$  to  $-45\text{\textperthousand}$ . Although there are few measurements of the isotopic composition of the Atlantic-sourced precipitation recycled over the Congo, satellite-based studies of the  $\delta D$  of atmospheric water vapor ( $\delta D_{\text{vapor}}$ ) indicate highly D-enriched vapor over the Congo basin due to the weak net isotopic fractionation associated with transpiration in humid tropical forests (Frankenberg et al., 2009; Worden et al., 2007), delivering D-enriched precipitation to East Africa (Levin et al., 2009; Rozanski et al., 1996). These patterns of  $\delta D_{\text{vapor}}$  track the isotopic composition of African rainfall both seasonally and annually (Fig. 5), as discussed below, confirming that drier air masses/D-depleted vapor/low precipitation and wetter air masses/D-enriched vapor/high precipitation tend to co-occur on a regional scale.

As described above, the modern isotopic composition of precipitation (approximately  $-17\text{\textperthousand}$ ) should yield  $\delta D_{\text{wax}}$  values around  $-102\text{\textperthousand}$  with an  $\varepsilon_{\text{precip-C28}}$  of  $\sim 85\text{\textperthousand}$ . Assuming that this apparent

fractionation is constant through time,  $\delta D_{\text{wax}}$  should generally fall between  $-95\text{\textperthousand}$  and  $-105\text{\textperthousand}$  when tracking a winter Indian monsoon signal, and between  $-122\text{\textperthousand}$  and  $-130\text{\textperthousand}$  when tracking a mixture of southeasterly/northwesterly sources. If variations in Lake Malawi  $\delta D_{\text{precip}}$  are largely driven by changes in the admixture of precipitation from northeasterly and southeasterly/westerly moisture sources, then  $\delta D_{\text{precip}}$  and  $\delta D_{\text{wax}}$  variations should thus be on the order of  $\sim 17\text{\textperthousand}$  to  $35\text{\textperthousand}$ . The majority ( $1\sigma$ ) of the Lake Malawi  $\delta D_{\text{wax}}$  record falls between  $-104\text{\textperthousand}$  and  $-120\text{\textperthousand}$  (Fig. 3), reflecting a predominant moisture source from the winter Indian monsoon as exists today. Variations in  $\delta D_{\text{wax}}$  range between  $\sim 15\text{\textperthousand}$  and  $30\text{\textperthousand}$ , with higher amplitude variations strongly skewed toward depleted  $\delta D_{\text{wax}}$  values (see Sections 3.4–3.6). Complete replacement of the winter Indian monsoon source with a southeasterly/northwesterly source would deplete  $\delta D_{\text{wax}}$  by about  $35\text{\textperthousand}$ , which is the amplitude of change observed between the megadroughts interval and the Holocene. These lines of evidence lend further support to our interpretation of  $\delta D_{\text{precip}}$ , and thus  $\delta D_{\text{wax}}$ , at Lake Malawi as a recorder of changes in moisture source during the late Pleistocene and most of the Holocene.

### 3.4. Atmospheric circulation during the African megadroughts 140–70 ka

The relative contribution of each of the three moisture sources to Lake Malawi precipitation depends on the position of regional convergence boundaries and the relative strength of the winter monsoons. During the African megadroughts, we argue that a northward shift of the ITCZ and a weakening of the Indian winter monsoon reduced the contribution of relatively D-enriched precipitation to the Lake Malawi basin, resulting in an expansion of the D-depleted,



**Fig. 5.** (a) Average 850 mb vector wind strength and direction during austral summer (Dec.–Feb.; NCEP/NCAR Reanalysis, <http://www.esrl.noaa.gov/psd/>). (b) Satellite-based observations of the  $\delta D$  of water vapor over the African continent (Nov.–Feb.; Frankenberg et al., 2009).  $\delta D$  values represent an average of a 1–2 km vertical atmospheric column in the lower troposphere, weighted by the concentration of water vapor within the column (Frankenberg et al., 2009, supporting online material). (c) As in (a) but for June–August. (d) As in (b) but for June–August. Customized datasets provided courtesy of Christian Frankenberg (NASA/JPL) and Remco Scheepmaker (SRON).

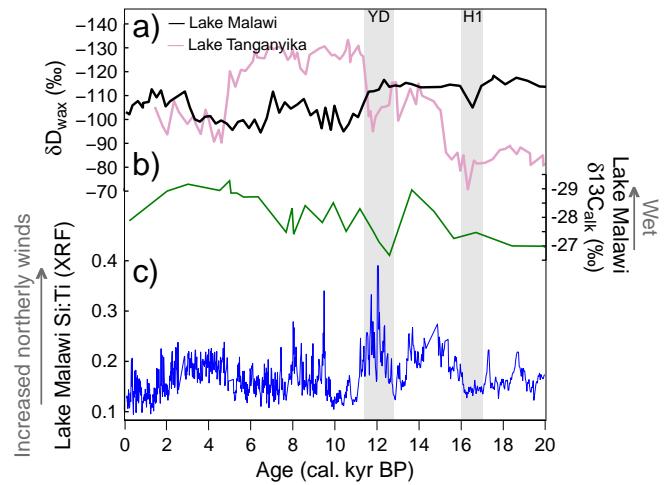
drier air mass that sits over much of southern Africa. A similar dynamic exists in modern austral winter, when the dry, D-depleted air mass over southwestern Africa expands north- and eastward (Fig. 5). After the termination of the megadroughts the relative contribution of this D-depleted air mass to Lake Malawi precipitation weakened considerably, explaining the long-term subsequent rise in  $\delta D_{wax}$  values during the late Pleistocene and into the Holocene. The ~20–40‰ overall difference in  $\delta D_{wax}$  between the megadrought interval and the early Holocene reflects this weakened contribution of the D-depleted air mass.

The latitudinal migration of the ITCZ and the expansion of the dry, D-depleted air mass may point to a modified Hadley circulation during the megadrought interval, as around 30° latitude in Africa, dry, D-depleted air is a product of the descending limb of the Hadley circulation (Worden et al., 2007). This would imply that Lake Malawi was located south of the ITCZ during the megadrought interval, influencing the  $\delta D$  of precipitation year-round. A northward migration of the southern Hadley cell would correspond to an expansion of the dry subtropical steppes and deserts of southern Africa. Subsequent southward migration would enable the ITCZ to penetrate further south into the African subtropics. This mechanism would explain evidence for dry and wet intervals at the Tswaing crater in eastern South Africa (~25°S) in-phase with the megadroughts recorded in the rift lakes (Kristen et al., 2007). Evidence for concurrent wetter conditions at paleolake Makgadikgadi, the catchment of which spans 12° of latitude northwest of Botswana (Burrough et al., 2009), may point to the influence of Atlantic-derived moisture over modern-day Angola during the megadroughts. However, hiatuses and age model uncertainties in the Makgadikgadi record preclude the interpretation of these behaviors as in-phase or out of phase with the tropical African lakes.

### 3.5. Paleohydrological variations at Lake Malawi during MIS 2 and 3

During MIS 3, orbital-scale variations in Lake Malawi  $\delta D_{wax}$  generally track those of Lake Tanganyika (Tierney et al., 2008), albeit with reduced amplitude of variability and with generally more depleted  $\delta D_{wax}$  at Lake Malawi (Fig. 4). However, no significant perturbations occur in Lake Malawi during the Heinrich events 4 and 5 (H4 and H5), despite  $\delta D_{wax}$  enrichments exhibited at Lake Tanganyika. Similarly, a significant  $\delta D_{wax}$  enrichment at Lake Malawi from 57.2 to 54.0 ka has no counterpart in the Lake Tanganyika record. This suggests that multi-millennial scale climatic perturbations were different between the two lakes, with North Atlantic cooling events having less of an impact on Lake Malawi. After ~32 ka the two records diverge, with Lake Tanganyika  $\delta D_{wax}$  exhibiting more extreme  $\delta D_{wax}$  enrichment and variability reflecting, primarily, Northern Hemisphere insolation forcing, while Lake Malawi  $\delta D_{wax}$  remains more stable throughout MIS 2. An abrupt, short-lived  $\delta D_{wax}$  enrichment at 16.5 ka during the H1 cooling event in the North Atlantic occurs at both sites (Fig. 6). Recent isotope-enabled general circulation model experiments suggest that D-enrichments during Heinrich events in East Africa may reflect an increase in the eastward propagation of enriched Congo vapor into the region without major changes in precipitation (Lewis et al., 2010). D-enriched Congo source moisture may also contribute to the extremely enriched  $\delta D_{wax}$  signal at Lake Tanganyika during MIS 2 and parts of MIS 3, relative to other records from the region (Fig. 4); Lake Malawi's greater distance from the Congo basin would make this influence more limited. The unique character of Lake Malawi  $\delta D_{wax}$  during MIS 2 and 3 suggests that long-term controls on precipitation in the Lake Malawi region are distinct from those at sites to the north, in keeping with a climatic "hinge zone" separating southeast and equatorial Africa.

Notably, no significant shift in  $\delta D_{wax}$  occurs during the Last Glacial Maximum (LGM, ~22 ka) at MAL05-2A. A small decrease in lake level (75–100 m), a decrease in total Pollen Accumulation Rate (PAR), an increase of benthic diatoms, and ~2.5‰ enrichment in the  $\delta^{13}\text{C}$  of



**Fig. 6.** (a)  $\delta D_{wax}$  (‰ VSMOW) from the north basin of Lake Malawi (MAL05-2A; black) and Lake Tanganyika (Tierney et al., 2008; purple); (b) Lake Malawi  $\delta^{13}\text{C}$  of  $\text{C}_{29}\text{--C}_{33}$  *n*-alkanes (‰ PDB; Castañeda et al., 2007); (c) Lake Malawi Si:Ti ratio measured by X-ray fluorescence (Brown et al., 2007). Gray bars indicate the Younger Dryas and Heinrich 1 events in the North Atlantic (Hemming, 2004). Note that the Y-axes are reversed. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

terrestrial *n*-alkanes indicate that cooler and slightly drier conditions prevailed (Beuning et al., 2011; Castañeda et al., 2007; Johnson et al., 2002; Scholz et al., 2007; Woltering et al., 2011), but these were unremarkable compared to other droughts in the last 76 kyr, as evidenced by the relatively minor change in lake level and a lack of carbonate preservation (Brown et al., 2007; Stone et al., 2011; Fig. 3).  $\delta D_{wax}$  from the low-resolution central basin record is more D-enriched than in the north basin during the same time period, but not significantly more enriched than the rest of MIS 2 and 3 in the central basin. This evidence, along with our  $\delta D_{wax}$  data, indicates that moisture balance and atmospheric circulation variations associated with the LGM in Lake Malawi were relatively minor in the context of the last 140 kyr.

Similarly, no significant  $\delta D_{wax}$  excursion occurs at Lake Malawi during the Younger Dryas (YD; 11.5–12.8 ka). This contrasts with several proxy reconstructions showing paleoenvironmental change during this period (Fig. 6). A 2‰ increase in  $\delta^{13}\text{C}$  of  $\text{C}_{29}\text{--C}_{33}$  *n*-alkanes indicates a change in plant community composition (Castañeda et al., 2007), although this excursion is minor in the context of C3/C4 variations recorded at other East African sites (e.g. Tierney et al., 2010). Paleoecological reconstructions show that lake level fluctuated within 100 m of modern lake level (Stone et al., 2011). Northerly winds were strong, as evidenced by an upwelling-induced increase in biogenic silica mass accumulation rate (Johnson et al., 2002) and increased volcanic Zr transported from north of the lake (Brown et al., 2007). The lack of a significant  $\delta D_{wax}$  excursion during the YD suggests that while this cooling event corresponded to changes in wind field at Lake Malawi, which heavily affected lacustrine productivity, overall moisture balance did not impact terrestrial plants as much as other droughts recorded at the lake. In this context it should be noted that Lake Masoko, located to the north of Lake Malawi, shows evidence for a wet Younger Dryas (Garcin et al., 2006), although localized hydrology and age model uncertainties limit the applicability of these results to the whole region.

### 3.6. The Holocene

Lake Malawi  $\delta D_{wax}$  is approximately 10‰ more enriched during the Holocene, on average, than the earlier parts of the record (Fig. 2). Mean  $\delta D_{wax}$  during the Holocene is 102.8‰ ( $1\sigma=4.9\text{‰}$ ), reflecting a primarily Indian winter monsoon signal (see Section 3.3), but with four prominent shifts in  $\delta D_{wax}$  suggesting significant

fluctuations in the region's source moisture (Fig. 6).  $\delta D_{wax}$  increases by 17‰ at the beginning of the Holocene, while sites to the north experience marked depletions in  $\delta D_{wax}$  (Fig. 4), likely reflecting the well-documented intensification of monsoon circulation experienced north of the climatic "hinge zone" (deMenocal et al., 2000; Schefuß et al., 2005; Tierney et al., 2011).  $\delta D_{wax}$  shifts to more negative values from 9.4 to 7.1 ka, followed by an abrupt 17‰ increase from 7.1 to 6.5 ka and an abrupt return to more negative values after 3.4 ka through 1.2 ka. These distinct, multi-millennial fluctuations in  $\delta D_{wax}$  are consistent with the moderate range of climatic variability suggested by other Holocene proxy records from Lake Malawi (Fig. 6). Previous diatom-based estimates of the  $\delta^{18}\text{O}$  of lake water at Lake Malawi also reveal little change from the early to the late Holocene (Barker et al., 2007). Palynological data indicate three distinct zones, with dry early Holocene conditions until 6.15 ka, wetter than present conditions from 6.15 to 3.0 ka, followed by a transition to drier conditions similar to modern at 3.0 ka (DeBusk, 1998). The  $\delta^{13}\text{C}$  of *n*-alkanes also suggests a relatively wet mid-Holocene from 7.7 to 2.0 ka (Castañeda et al., 2007; Fig. 6). However, these findings contrast with previous suggestions for a gradually increasing moisture balance at Lake Malawi from the early to the late Holocene resulting from increasing southern hemisphere insolation and a southward shift of the ITCZ (e.g. Finney et al., 1996; Gasse, 2000). Biogenic silica mass accumulation (BSi MAR) and the related Si:Ti ratio fluctuate throughout the Holocene, indicating that the strength and intensity of northerly winds were variable (Fig. 6). The non-stationary relationship between Si:Ti and  $\delta D_{wax}$  suggests that significant changes in the east–west component of regional circulation must have also taken place concurrently.

Together, these multiple proxies for winds and moisture balance suggest that the hydrological history of Lake Malawi during the Holocene was distinct from that of sites to the north. Lake Malawi  $\delta D_{wax}$  is more D-enriched than that sites north of the climatic "hinge zone," especially during the early Holocene when the equatorial region experienced more humid conditions and more D-depleted waxes (Fig. 4). Changes in moisture balance at Lake Malawi were more moderate than those experienced at some other East African rift lakes (e.g. Lakes Edward and Rukwa; Barker et al., 2002; Beuning and Russell, 2004) and the climatic factors affecting these moderate changes in moisture balance were more complex than a gradual increase in austral summer insolation. The  $\delta D_{wax}$  signal integrates the isotopic imprints of these changes.

In the late Holocene the ITCZ attained its current southern mean position (Haug et al., 2001), and an increase in the subtropical–tropical SST and sea level pressure gradient in the southeast Atlantic Ocean strengthened the trade winds off of southwestern Africa (Schefuß et al., 2005). This may have limited the extent of the D-depleted air mass over southern Africa, explaining its current range today. In this context, relatively depleted  $\delta D_{wax}$  values at Lake Malawi from ~3.1 ka to present likely reflect the modern hydrological controls on  $\delta D_{precip}$ , with source changes constituting a more limited role than the amount effect and overall intensity of the hydrological cycle. The absolute  $\delta D_{wax}$  values of Lakes Malawi and Tanganyika converge at this time, supporting the hypothesis that while the mean ITCZ position plays a major role in modern Southeast African precipitation, the controls on hydrology were more complex prior to the late Holocene.

#### 4. Conclusions

Our  $\delta D_{wax}$  record from Lake Malawi indicates that a reorganization of atmospheric circulation over southeast Africa may explain the transition into relative climatological stability at Lake Malawi after ~56 ka. Changes in vapor transport history and source region dominate the  $\delta D_{wax}$  signal for the majority of the record, with local rainout playing a secondary role. This is most obvious during the climatically unstable period prior to 56 ka, when lake levels and  $\delta D_{wax}$  exhibit a strong positive relationship. D-depleted leaf waxes and more extreme

$\delta D_{wax}$  variability at this time suggest that a weakening of the Indian monsoon and/or a modification in the Hadley circulation may have played critical roles in the late Pleistocene African megadroughts, an idea that merits further investigation with additional long records. Subsequently, from 56 ka through the deglaciation, a balance of southwesterly, northeasterly, and northwesterly moisture helped maintain higher lake levels while stabilizing the  $\delta D_{wax}$  signal. This balance may explain why proxies sensitive to northerly wind strength (e.g. Zr:Ti, BSi MAR) exhibit great variability in MIS 2–3 while proxies more sensitive to hydrologic balance (e.g. fossil pollen,  $\text{CaCO}_3$  preservation, and seismic reflection) indicate relatively stable conditions. Heightened  $\delta D_{wax}$  variability during the Holocene and a mid-Holocene D-isotopic enrichment do not resemble other stable isotopic patterns from elsewhere in East Africa, highlighting this region's spatial complexity. Further work is needed to delineate these circulation patterns, particularly the east–west component, through novel proxy reconstructions of wind field and experiments with general circulation models.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at doi:10.1016/j.epsl.2011.10.020.

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