Three and half million year history of moisture availability of South West Africa: Evidence from ODP site 1085 biomarker records

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1. Introduction

While the late Cenozoic tectonic and moisture history of East Africa is well constrained (Levin et al., 2004; Wynn, 2004; Peakens et al., 2005; Trauth et al., 2005; 2007; Sepulchre et al., 2006), this is not the case for southern Africa (Dupont et al., 2005; Maslin and Christensen, 2007). Far from the major rifting centres, the region lacks the volcanic (basalt and ash) deposits that are characteristic of East Africa and permit the development of radiometric-based chronologies. Instead, most of the continental evidence for past climate change is derived either from estimations of denudation rate and landscape development using fission-track thermochronology and/or cosmogenic isotope analysis or by reconstructing regional geomorphological features associated with specific tectonic events. Estimations of denudation rate and landscape development for South West Africa (Brown et al., 1994; 2000; Cockburn et al., 1999; 2000; Gallagher and Brown, 1999; Raab et al., 2002; 2005) and South East Africa (Fleming et al., 1999; Brown et al., 2002; van der Beek et al., 2002) indicate denudation rates peaking at between 100–200 m/Myr during the late Cretaceous (80 to 60 Ma). Subsequent denudation rates for Namibia are 15 m/Myr or lower, and are typical for a passive margin setting (Raab et al., 2005); similar results have also been found for the Drakensberg Escarpment (Brown et al., 2002). This suggests that since 60 Ma there has been little or no uplift in South West Africa and that tectonic activity has been quiescent. Continental palaeoclimate records are also limited due to a lack of chronological control, e.g., Vaal River terraces, Florisbad Pan deposits, Tswaing Crater or lake deposits (Partridge et al., 1997; de Wit et al., 2000).

At present it is believed that a major change in southern African climate occurred during the late Cenozoic, resulting from the development of a strong cross-contintental temperature gradient associated with the onset of the Benguela Current (BC) (Tyson and Partridge, 2000; Christensen et al., 2002). The upwelling associated with the BC initiated arid conditions in the western region, but there was substantial climate amelioration during the Pliocene when streams were rejuvenated and grasslands expanded (Tyson and Partridge, 2002). Uplift of the south-eastern and eastern hinterlands, including East Africa and Zimbabwe, created rain-shadow areas to the west of these regions (Tyson and Preston-White, 2000) and is thought to have further enhanced aridification. Tyson and Preston-White (2000) suggest the timing of this uplift was not well constrained and that it occurred at approximately the same time as the late Pliocene intensification of Northern Hemisphere glacialization (INIG; Li et al., 1998; Maslin et al., 1998). Subsequently Trauth et al. (2005) have shown East African uplift
started at about 12 Ma in northern Ethiopia and progressively shifted southward and occurred between ~4.5 Ma to 3.5 Ma in central Kenya and between 3.2 Ma to 2.3 Ma in southern Kenya and Tanzania. This coincides with a long-term drying trend in East Africa as well as that inferred from pollen records between 3.5 Ma and 1.7 Ma from ODP Site 1082 off the coast of the Namib Desert (Fig. 1). This record also suggests a possible rapid increase in local aridity at about 2.2 Ma (Dupont et al., 2005). Arguments to the contrary suggest that the late Miocene was more arid and that there is no indication of major change during the Pliocene (Diester-Haass et al., 2002; Giraudet et al., 2002).

The most widely used method of reconstructing moisture availability is through the reconstruction of palaeovegetation using pollen analysis (e.g., Dowsett and Willard, 1996; Shi et al., 2000; Dupont et al., 2005). An alternative useful tool is carbon isotopic analysis (Hayes et al., 1990) of soil carbonates (Kingston et al., 1994; Morgan et al., 1994; Levin et al., 2004; Ségalen et al., 2007) or terrestrial higher plant biomarkers (e.g., Bird et al., 1995; Yamada and Ishiwatari, 1999; Huang et al., 2000). This allows the discrimination of inputs from isotopically distinct C3 and C4 plants. Average δ13C values of organic carbon in land plants (higher plants) vary according to the chemical pathway of photosynthesis (Ehleringer and Monson, 1993). Plants using the C3 (Calvin-Benson or non-Kranz) photosynthesis pathway have a δ13C range between −22‰ to −30‰ (average −27‰) (Ehleringer and Monson, 1993), compared with an average atmospheric isotopic composition of −6‰ to −7‰. Plants using the C4 (Hatch-Slack or Kranz) pathway, including tropical and marsh grasses, have a δ13C range of between −9‰ to −15‰ (average −13‰) (Ehleringer and Monson, 1993). C4 plants re-evolved in the Miocene (Edwards et al., 2010; Brown et al., 2011) and the first proliferation of C4 plants into Africa was between 12 and 15 Ma and then they became abundant between 7 and 8 Ma (Kingston et al., 1994; Morgan et al., 1994; Cerling et al., 1997; Ségalen et al., 2007). There is also a third type of plant, CAM (Crassulacean Acid Metabolism), which uses either C3 or C4 pathways depending on water availability. In terms of occurrence it is minor compared with the other two pathways, but is very important in the context of SW Africa due to their unusually high abundance (ca. 60% near the SW African coast; Werger and Ellis, 1981). In this study we use the distributions and δ13C values of n-alkanes from marine sediments recovered from Ocean Drilling Program (ODP) Leg 175 in the South East Atlantic Ocean to provide a long-term estimate of the vegetation water stress and thus moisture availability of the region over the last 3.5 million years.

2. Site location

The location of Ocean Drilling Program Site 1085 in the Benguela Current (BC) is shown in Fig. 1. The Site is located 260 km away

Fig. 1. Schematic representation of Southern African vegetation and rivers, including estimated average C11 n-alkane δ13C values (adapted from Rommerskirchen et al., 2003), estimated surface ocean hydrography, and locations of ODP Sites 1082 and 1085 (adapted from Ettwein et al., 2001 and Christensen et al., 2002). BCC = Benguela coastal current, BOC = Benguela ocean current, ABF = Angola-Benguela Front, AC = Angola Current and SECC = South East Counter Current.
from the mouth of the Oranje River, 410 km away from the Olifant system and over a 1000 km from the Cunene River mouth. According to the review by Dupont (2011) if we were studying pollen ~300 km is the break between mixed river and aeolian input (~300 km) compared with just aeolian input (>300 km). Even if we assume fluvial transported biomarkers can travel further in the upper ocean, only input from the Oranje River need be considered. However the sediment from the Oranje River is deposited as a mud blanket along the continental shelf and very little of it moves off shore into the open Ocean (Meadows et al., 2002). Moreover, Site 1085 is directly opposite the mouth of the Oranje River and thus the predominate ocean currents will move sediment northward along the coast away from Site 1085. Hence we suggest that terrigenous material is delivered mainly to the southern shelf and very little of it moves off shore into the open Ocean (Rommerskirchen et al., 2003). Aolian deposition is driven by the Southern Hemisphere trade wind system operating at the same latitude as the BC system. These predominantly south-westerly winds entrain terrestrial material from the adjacent continental areas of southern Africa and deposit them into the BC system (Shi and Dupont, 1997; Dupont and Wypputa, 2003).

The vegetation of Africa south of the equator is a complex mosaic of phytogeographies ranging from tropical rainforest to desert, with a high degree of diversity and endemism. Fig. 1 summarises the general vegetation types, the relative occurrence of C₃, C₄ and CAM plants (Werger and Ellis, 1981; Rommerskirchen et al., 2003; Denison et al., 2005), and the average Holocene n-alkane δ¹³C values for each region (Rommerskirchen et al., 2003). The continental margin adjacent to ODP Site 1085 is dominated by desert and woody savannah. The Karoo-Namib Desert vegetation is dominated by C₃ plants, whereas the Zambezian and Kalahari-Highveld regions are mainly savannah and woodland and have a mixture of C₃ and C₄ plants with C₃ being more common (White, 1983). Large dust plumes occur during austral winter with winds blowing off the continent. There is also a small input of material from the Succulent Karoo during austral summer (Rommerskirchen et al., 2003). The Succulent Karoo is dominated by CAM plants, which can change their photosynthetic pathway between C₃ and C₄ depending on water availability, which potentially provides a wider range of n-alkane δ¹³C values.

3. Methodology

Based on the age model of Christensen et al. (2002), material from ODP Site 1085 was sampled every ~100 ka, generating 35 samples for n-alkane δ¹³C analysis representing the last 3.5 Ma and 55 samples for biomarker characterisation representing the last 5.5 Ma. Total Organic Matter (TOC), was determined by weighing and drying sediments overnight in an oven at 110 °C, then placing them in a furnace at 550 °C for 3 h; the loss-on-ignition is taken as the amount of organic material present in the sample. Samples to be analysed for biomarker distributions were freeze-dried, ground and extracted via ultrasonication in a sequence of solvents with increasing polarity: hexane; a 1:1 (v/v) azoteore of dichloromethane (DCM); methanol (MeOH); acetate, each sample was injected into a Carlo Erba 5300 series High Resolution Gas Chromatograph fitted with a CP-Cil 5CB silica column (internal diameter 0.32 mm, length 50 m, df 0.11 μm), which was heated from 40 °C to 140 °C at 20 °C/min, then at 4 °C/min to 300 °C at which temperature it was held for 20 min. Compounds were quantified against the known concentrations of internal standards described above. All fractions were subsequently analysed using gas chromatography/mass spectrometry (GC/MS) with a Thermoquest Finnigan Trace chromatograph interfaced to a Thermoquest Finnigan Trace mass spectrometer operating with electron ionization at 70 eV and scanning an m/z range of 50 to 850. GC conditions for GC/MS were identical to those used for GC analysis.

Compounds were assigned by comparing mass spectra and relative retention times with those in the literature. Sedimentation rates derived from the AMS age model described above were used along with the dry bulk density to calculate MARs for the various molecular components along with the average chain length (ALC) for the n-alkanes (Boot et al., 2006):

\[
\text{MAR}_{\text{component}} = \left( \text{Conc}_{\text{component}} \times \text{SR} \times \text{DBD} \right),
\]

where \( \text{MAR}_{\text{component}} \) is the MAR in ng cm⁻² kyrt⁻¹, Concₜₐₜₜₜ is the mass of component in ng g⁻¹ dry sediment, SR the sedimentation rate in cm kyrt⁻¹ and DBD is the dry bulk density in g cm⁻³. Average chain lengths (ACLS) for the n-alkanes were calculated as:

\[
\text{ACL}_{27-33} = \frac{(C_{27} + C_{28} + C_{31} + C_{33})}{(C_{27} + C_{28} + C_{31} + C_{33})}
\]

with \( C_j \) signify the concentration of the n-alkane with \( j \) carbon atoms. Carbon preference indices (CPIs) were calculated for n-alkanes and n-alkanols as:

\[
\text{CPI} = \left[ 0.5 \sum \left( X_j + X_{j+2} + \ldots + X_n \right) / \sum \left( X_{j-1} + X_{j+1} + \ldots + X_{n+1} \right) \right]
\]

\[
+ 0.5 \sum \left( X_j + X_{j+2} + \ldots + X_n \right) / \sum \left( X_{j-1} + X_{j+1} + \ldots + X_{n+1} \right) \right],
\]

where \( i = 25 \) and \( n = 33 \) in the case of n-alkanes and \( i = 21 \) and \( n = 31 \) in the case of n-alkanols.

Gas Chromatography-Isotope Ratio Monitoring Mass Spectrometry (GC-IRMMS) was used to measure the carbon isotopic composition of compounds in the n-alkane fraction. Each sample was dissolved in 100 μl ethyl acetate and placed in 1 ml auto-sample vials before being analysed on a Varian GC fitted with a 2B-1 column linked to a ThermoFinnigan Mat DeltaS mass spectrometer via a combustion interface; the GC temperature program was the same as that used for standard GC analysis. Each sample was analysed twice and values are expressed in standard δ³¹C notation as parts per mil (‰) deviations from the Vienna Pee Dee Belemnitte (VPDB) standard, measured during analyses against a pulsed reference gas input whose isotopic composition relative to VPDB had been determined by an external facility. Fatty acid and n-alkane mixtures, for which the individual compounds δ¹³C values had been determined offline by external facilities, were analysed every day, revealing instrumental precision better than 0.3‰. (see errors bars on Figs. 2 and 3). This is consistent with the error recorded by duplicate measurement of the samples. Only 25 out of the 35 samples taken from Site 1085 yielded sufficient quantities of n-alkanes for isotope analysis.

4. Concentrations and distributions of n-Alkanes

N-alkanes, along with long-chain acids and alcohols, occur in vascular plant leaf extracts with an odd-over-even predominance (OEP) and carbon chain-lengths ranging from C₁₃ to C₉₅ (Eglinton et al., 1962; Eglinton and Hamilton, 1967). In Site 1085 sediments, the high-molecular-weight (HMW) n-alkanes are characterised by a
predominance of the C27, C29, C31 and C33 homologues, exhibiting a strong OEP (CPI values range from 4 to 10, with no systematic variation through the section) and consistent with a leaf wax origin. Total HMW n-alkane concentrations vary by about two orders of magnitude, from ca 10 to 1000 ng g\(^{-1}\) sediment (Fig. 2). In general, a decrease in total and individual HMW n-alkane concentrations occurs over the 5.5 Ma study interval despite an increase in TOC contents. In detail, this occurs as a gradual decline from the base of the study interval at 5.5 Ma to 3.5 Ma; concentrations then decrease dramatically at ca 3 Ma and remain low (<100 ng g\(^{-1}\) sediment) until ca 2.5 Ma when they increase to intermediate values (100 to 500 ng g\(^{-1}\) sediment). These trends are still apparent or even enhanced if n-alkane concentrations are normalized to TOC contents or converted to mass accumulation rates (Fig. 2). N-alkanes are resistant to degradation and relatively easily

Fig. 2. Total organic carbon contents (A) and n-alkane concentration (B-D) and distribution (E) parameters in ODP Site 1085 over the past 5.5 million years. Specifically, (B) shows the C\(_{31}\) n-alkane concentrations normalized to TOC (ng g\(^{-1}\) of OC), (C) shows concentrations of the C27, 29, 31 & 33 n-alkane homologues as ng g\(^{-1}\) of dry sediment and (D) shows summed n-alkane concentrations (C27-33) in ng g\(^{-1}\) of dry sediment. Average Chain Lengths (ACLs) of the HMW n-alkanes are shown in (E).

Fig. 3. Comparison of eccentricity variations (Berger and Loutre, 1991) with ODP Site 1085 n-alkane \(\delta^{13}C\) values (this study), East African soil carbonate \(\delta^{13}C\) values (light dots = Levin et al., 2004; dark dots = Wynn, 2004), East African lake occurrence (Trauth et al., 2005, 2007) and Hominin Evolution Transitions (see full references in Trauth et al., 2007).
transported from continental to marine (or lacustrine) settings via either aeolian or fluvial mechanisms (see Pancool and Boot, 2004 review and references therein). As described above, n-alkanes at Site 1085 almost certainly derive from aeolian processes. Thus, the changes in concentration could reflect changes in wind strength. However, such an interpretation should be done cautiously as the controls on wax concentrations or the distribution of n-alkanes relative to functionalised leaf waxes (n-alkanols and n-alkanoic acids) remain poorly understood and do vary amongst plant types. Biomarker concentration changes can also reflect OM degradation, although we consider that less likely due to the lack of similar changes in TOC contents. Changes in the distribution of n-alkanes can be evaluated by the average chain length (ACL). ACLs are generally stable, ranging from 29.9 to 30.7 (except for a single sample with an ACL of 29.5). However, average ACLs are generally lower from ca. 3.6 to 2.5 Ma, having an average value of 30.0 compared to an average of 30.5 over the rest of the record. Interpretation of n-alkane ACLs is unclear, with suggestions that higher values are associated with hotter or more arid conditions (Hinrichs and Rullkötter, 1997; Hinrichs et al., 1998). However, n-alkane ACLs in plant extracts are rather variable and interpretation remains unclear.

5. Long-term Plio-Pleistocene trends in SW African moisture availability

Palaeovegetation and relative moisture availability can be reconstructed using n-alkane δ13C values. C₃ and C₄ plant species, as discussed above, each have distinct isotopic signatures (Pancool and Pagani, 2006). The species balance can be recorded by bulk sedimentary organic matter (e.g. Kastner and Göth, 2003), but bulk OM δ13C values can be skewed by contributions from marine inputs. Compound-specific δ13C analyses of refractory n-alkanes of exclusively higher plant origin avoids this problem (e.g. Eglington et al., 1962; Eglington and Hamilton, 1967), and they have been shown to record C₃ and C₄ species differences with fidelity (e.g. Collister et al., 1994; Scheufuß et al., 2003; Pancool and Boot, 2004). Collister et al. (1994) observed that in C₃ and C₄ species, the n-alkanes δ13C values were depleted relative to biomass by −5.9‰ and −9.9‰ making the observed C₃ range −28 to −36‰ (average −33‰) and C₄ range −19 to −26‰ (average −13‰) in n-alkanes. Consequently, they have been used widely to determine the relative contribution of C₃ and C₄ plants to lacustrine and marine sediments (e.g. Scheufuß et al., 2003; Street-Perrott et al., 2004; Feakins et al., 2007). The late Holocene range of n-alkane δ13C values from South Africa, from −26‰ to −28‰ (Rommerskirchen et al., 2003), reflects the mixed vegetation of South Africa and provides a baseline for comparison to the long-term record from Site 1085.

The ODP Site 1085 n-alkane δ13C values are presented in Fig. 3 and exhibit no long-term trend over the last 3.5 Ma. In fact, the total range over the last 3.5 Ma years is between −25.5‰ and −28.5‰, with similar values and only a slightly greater range than that observed for the Holocene. Interpretation of n-alkane δ13C values (as with those of bulk organic or paleosol carbon) as indicators of C₃-C₄-CAM vegetation shifts needs to be done cautiously because water-stress of C₃ and CAM plants can also produce heavier carbon isotope values (Diefendorf et al., 2010). The Site 1085 n-alkane δ13C record, therefore, implies that the vegetation in southern South West Africa under went very little or no change in the water stress. This may reflect very little change in the proportion of C₃ to CAM plants in SW Africa over the last 3.5 Ma. We note, however, that our record could be consistent with shifts in the proportion of C₃ to C₄ to CAM, which result in the same average n-alkane δ13C values.

Recently, Tipple et al. (2010) suggested that the δ13C of atmospheric CO₂ could have become up to 1‰ more negative over the last 4 million years. If so then this should be reflected in our long-term n-alkane δ13C records. In fact this is exactly what is observed, with slightly higher δ13C values between 3.5 Ma and 3 Ma (−26‰) and slightly lower values between 0.9 Ma to 0.4 Ma (−27‰). We suggest that further work is required to validate the proposed decrease in atmospheric δ13C values but if confirmed it would indicate that there was even less variation in the n-alkane δ13C record of southern SW Africa, implying that very stable conditions persisted throughout the last 3.5 million years.

Although interpretation of n-alkane δ13C values should be done cautiously, it is clear that the 1085 record provides no evidence for a long-term decrease or increase in SW Africa moisture availability over the last 3.5 Ma. In fact, moisture availability appears to have been incredibly stable, despite large-scale tectonic activity in East Africa and major shifts in global climate. This is in contrast with the pollen record of ODP Site 1082 (see Fig. 1), about 10⁴ N further north, which suggests a long-term trend towards drier conditions between 3.5 Ma and 1.7 Ma (Dupont et al., 2005). Dupont et al. (2005) suggest this aridification of the Namib Desert is due to the suppression of local sea surface temperatures caused by increased upwelling of cold water and less convective atmospheric moisture transport towards land.

Despite its overall stability, the Site 1085 n-alkane δ13C record does exhibit some variations. Between 3.5 Ma and 2.7 Ma, n-alkane δ13C values are less negative than average suggesting slightly drier conditions. Intriguingly, this interval also corresponds to the interval characterised by the lowest n-alkane ACLs. Although low ACLs have been interpreted as reflecting wetter conditions, we caution that interpretation of such signatures remains unclear. Instead, both the high δ13C values and low ACLs might reflect a different vegetation regime (Boot et al., 2006).

Between 2.5 and 2.7 Ma n-alkane δ13C values become more negative, and this interval contains the most negative values of the whole record. The same interval is also characterised by the lowest n-alkane concentrations (Fig. 2), suggesting decreased aeolian transport of n-alkanes (e.g. Pancool and Boot, 2004; Scheufuß, et al., 2005). Although both signatures could have multiple interpretations, both suggest that slightly wetter conditions in SW Africa coincided with the intensification of Northern Hemisphere Glaciation (INHG). This is in agreement with the Site 1082 pollen record further north which shows an increase in Stoebe-type pollen between 3 Ma and 2.3 Ma. This pollen is indicative of the fynbos biome in southwest Africa, and Dupont et al. (2005) suggest this indicates a northward extension of the winter rainfall area.

From 2.4 Ma until 0.4 Ma (the end of the record) the n-alkane δ13C values are relatively consistent, varying by less than ±0.5‰ suggesting that there has been little or no change in SW African moisture availability over the last 2.4 million years. This stability of SW African climate suggests that changes in the ocean currents both in the South East Atlantic and Indian Oceans had little or no effect on continental climate, contrary to the suggestions of Tyson and Partridge (2002) and Tyson and Preston-White (2000).

6. Comparison between SW and E African moisture availability

The long-term moisture history of SW Africa is radically different from that of East Africa. Our n-alkane δ13C record suggests that moisture availability in SW Africa was characterised by long-term stability, with no discernable trend. This suggests that the stable, if not quiescent, tectonic activity in SW Africa (Raab et al., 2005) provided a stable geography leading to a stable climate over the last 3.5 million years. In contrast, soil carbonate and n-alkane δ13C records (Levin et al., 2004; Feakins et al., 2005; 2007; Ségalen et al., 2007) from East Africa and the adjacent Indian Ocean show a significant 10% increase over the last 2 million years indicating a strong increase in the aridity of extreme periods due to the continued development of the East African Rift system (Trauth et al., 2005; 2007), see Fig. 4. Using Plio-Pleistocene terrestrial dust flux records to the Indian and Atlantic Oceans and the Mediterranean Sea, Trauth et al. (2009) have shown that the aridity trend (deMenocal, 2004) may in fact be a trend towards increasing environmental variability through time. They postulate that after approximately 2 Ma, dry periods became drier whilst wet periods may have remained similar in character. There is strong evidence that this aridity...
trend in East Africa was punctuated by episodes of precessionally-forced extreme wet and dry periods lasting a few hundred thousand years (see Fig. 2). These periods are characterised by the appearance and disappearance of large deep lakes within the East African Rift Valley (Kingston et al., 2007; Trauth et al., 2007) in conjunction with corresponding changes in the flux of terrigenous material reaching the oceans (deMenocal, 2004; Trauth et al., 2009). In contrast to these marked extremes in climatic variability observed in East Africa over the last 3 million years ago, the results from ODP Site 1085 suggest that SW African climate was relatively stable during this time. Our data suggest that SW Africa, like East Africa, seems to be wetter during the INHG but currently there is no evidence that this region responded to either the onset of Walker Circulation at ca. 1.9–1.8 Ma (Ravelo et al., 2004) or the Middle Pleistocene Revolution at ca. 1.0–0.8 Ma. However, in order to further evaluate past climate variability in SW Africa, it is necessary to improve the quantity and resolution of available palaeoclimate records.

7. Conclusions

The long-term moisture history of SW Africa over the last 3.5 million years is one of stability with no discernable trend. This suggests that previously inferred late Pliocene tectonic changes must have occurred much earlier and that large global climate shifts and local oceanic changes had little or no effect on the region. These results have interesting implications for the study of early hominin evolution and migration. The climatic stability of the region, even on short precessional time scales (Denson et al., 2005), contrasts sharply with East Africa which experienced not only a long-term increase in aridity, but alternating periods of extreme wet and dry climatic conditions (Maslin and Brown, 2004) on the same scale. Note that the moisture availability reconstructed for South West Africa is essentially flat, while in East Africa the range of variability increases from approximately 2 millions years BP.

**Fig. 4.** Comparison of SW African ODP Site 1085 n-alkane δ13C values (this study) and East African soil carbonate δ13C values (light dots = Levin et al., 2004; dark dots = Wynn, 2004) on the same scale. Note that the moisture availability reconstructed for South West Africa is essentially flat, while in East Africa the range of variability increases from approximately 2 millions years BP.

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