Orbital forcing and the spread of C₄ grasses in the late Neogene: stable isotope evidence from South African speleothems

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Abstract

Reconstructing Plio-Pleistocene African paleoenvironments is important for models of early hominin evolution, but is often hampered by low-resolution or discontinuous climatic data. Here, we present high-resolution stable oxygen and carbon isotope time series data from two flowstones (secondary cave deposits) from the South African hominin-bearing Makapansgat Valley. The age of the older of the two flowstones (Collapsed Cone) is constrained by magnetostratigraphy to approximately 4–5 Ma; the younger flowstone (Buffalo Cave) grew between 2.0–1.5 Ma, as determined by magnetostratigraphy and orbital tuning of the isotopic data.

The carbon isotope data is used as a proxy for the proportion of C₄ grasses in the local environment and the oxygen isotope data reflects monsoon rainfall intensity. The carbon isotope evidence indicates that in the late Miocene/early Pliocene, the local environment was dominated by C₃ vegetation, whereas, in the Plio-Pleistocene, it was composed of a mixture of C₃ and C₄ vegetation. This suggests that C₄ grasses became a significant part of the Makapansgat Valley ecosystem at approximately 4–5 Ma, towards the end of the late Neogene global expansion of C₄ grasses. After this initial expansion, South Africa experienced further fluctuations in the proportion of C₃ and C₄ vegetation during the Plio-Pleistocene, in response to regional and global climatic changes. Most notably, the Buffalo Cave flowstone provides evidence for C₄ grass expansion at ca. 1.7 Ma that we suggest was a response to African aridity caused by the onset of the Walker Circulation in the Pacific Ocean at this time.

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Introduction

Global climatic oscillations with Milankovitch periodicities are a dominant feature of the late Neogene and have been viewed as driving forces in the evolution of African mammalian faunas, including hominins (e.g., Bromage and Schrenk, 1995; Vrba, 1995a; Potts, 1998; Zeitoun, 2000; Bobe and Eck, 2001; Barry et al., 2002; Bobe et al., 2002). In the absence of a long and detailed terrestrial record of global change, these models of climatically forced macroevolution have used the marine paleotemperature record (e.g., Shackleton, 1995) as the template onto which trends in mammalian evolution are fitted. The focus has been either on directional trends of global cooling (Vrba, 1992, 1995a) or on the magnitude and frequency of climatic variability (Potts, 1996, 1998). These global climatic trends have been held responsible for episodes of hominin speciation, extinction, and migration. While there
are sound theoretical and empirical reasons to link events in hominin evolution to periods of climatic change (Vrba, 1995b), it is simplistic to assume that the late Neogene marine paleoclimate record is a faithful proxy for subtropical and tropical African paleoclimates. On the contrary, there is mounting evidence to show that tropical terrestrial climatic change is decoupled from the global ice volume record of deep-sea δ¹⁸O (e.g., Pokras and Mix, 1987; Clemens et al., 1991; Partridge et al., 1997; Trauth et al., 2005; Hopley et al., 2007). Previous proxy records of African climate change have been constructed from terrestrial components within marine sediment cores (deMenocal and Bloemendal, 1995; Dupont and Leroy, 1995) that produce space-averaged terrestrial records that cannot be linked to specific hominin habitats.

Neogene spread of savanna grasses and the faunal response

It is widely accepted that the mid-Miocene African faunas differ significantly from the Plio-Pleistocene African faunas in terms of their species composition and habitat preferences (e.g., Hill, 1987; Andrews, 1992; Vrba, 1995a), and that the interim period represents a gross restructuring of the African terrestrial communities in response to the developing savanna biome. However, refining the details of this transition from the mid-Miocene “C₃-world” to the Plio-Pleistocene “C₄-world” has been problematic. The Cerling et al. (1997) interpretation of the carbon isotope data from fossil herbivore teeth restricts the global expansion of C₄ biomass to one event between 8 Ma and 6 Ma. However, as noted by Köhler et al. (1998), this single pulse of C₄ grass expansion is not always accompanied by an increase in faunal turnover in the “C₄-world”. Köhler et al. (1998) highlight the fact that faunal turnover events also occur in purely C₃ environments in the late Neogene, indicating that faunal turnover is not unique to tropical savanna-woodland mosaics.

Numerous episodes of increased rates of faunal turnover have been identified within terrestrial late Neogene mammalian lineages, each showing replacement of woodland fauna by more savanna-adapted species. These events occur at different times in different regions of the world and are uniformly distributed within the last ten million years. Examples of documented late Neogene faunal turnover events have been recorded at 10 Ma, 7.8 Ma, and 7.3–7.0 Ma in Pakistan (Barry et al., 2002; Patnaik, 2003; Raza et al., 2002); 10–9 Ma and 6.5 Ma in Spain (Pickford et al., 1995; Köhler et al., 1998); approximately 7.4 Ma–6.0 Ma in the Nawata Formation of the Lothagam succession, Kenya (Leakey and Harris, 2003; McDougall and Feibel, 2003); and between 2.8 and 2.3 Ma in the Omo sequence of Ethiopia (Bobe and Eck, 2001; Bobe et al., 2002). There is no global faunal turnover event significantly above background rates at 8–6 Ma (Köhler et al., 1998) that can be related to the rapid global expansion of C₄ grasses as described by Cerling et al. (1997). These studies indicate there was a series of turnover events in the late Neogene which, if they are linked with episodes of climatic change, are related to regional rather than global events.

Southern African late Neogene paleoclimates

The relative abundance of C₃ and C₄ plants in modern-day South Africa is strongly influenced by the existence of two seasonally distinct climate systems (Vogel et al., 1978; Lee-Thorp and Talma, 2000). C₄ grasses dominate the grassy vegetation of those areas receiving summer rainfall in the eastern and interior regions of South Africa, with the exception of the high mountains along the eastern escarpment (Vogel et al., 1978). C₃ grasses occur in the Fynbos region to the southwest of South Africa, which receives the majority of its rainfall in the winter. A mixture of C₃ and C₄ grasses is found in the transition zone between the winter and summer rainfall zones. A shift in the range and intensity of the dominant atmospheric circulation systems would, therefore, change the distribution of these two vegetation systems (Lee-Thorp and Talma, 2000). As a consequence of the C₄ photosynthetic pathway, the carbon isotope composition of C₄ plant matter is enriched in ¹³C (O’Leary, 1981, 1988), enabling the presence of C₄ photosynthesis to be detected in organic and inorganic carbon species derived from biological processes. The proportion of C₃ and C₄ vegetation in past ecosystems can be determined in fossil teeth (e.g., Lee-Thorp et al., 1994, 2000) and sedimentary carbonates (e.g., Cerling et al., 1989), as long as the effects of diagenetic alteration can be discounted.

Data on the paleoenvironments of South African hominins are derived either from the fauna, flora, or geology of cave breccias. Many of the faunal specimens in the South African hominin sites were found in lime-dumps after the caves were mined in the early twentieth century, and lack stratigraphic context. In situ bone-breccias that survived the lime-mining intact tend to be poorly stratified, and represent unknown periods of deposition of tens- to hundreds-of-thousands-of years (McFadden et al., 1979; Herries, 2003). Both in situ and mined bone-breccias are given Member status within the stratigraphy of each cave deposit (e.g., Partridge, 1978, 1979) and are defined on broad differences in sedimentological and paleontological characteristics. Previous paleoenvironmental studies based on these bone-breccias have, therefore, lacked the temporal resolution required to investigate climatic change and early hominin paleoecology on the timescale of orbital cycles.

Stable carbon isotopes of fossil teeth from South African Plio-Pleistocene cave breccias have shown the presence of both C₃ and C₄ vegetation in the local environment of each faunal member studied (van der Merwe and Thackeray, 1997; Sponheimer et al., 1999; Lee-Thorp et al., 2000). However, as discussed in Kingston (1999), selective feeding and taphonomic factors (Vrba, 1980) make converting isotopic paleodietary data into meaningful paleovegetational reconstructions problematic. Fossil macrofloras have only been found in Sterkfontein Member 4 (Bamford, 1999) and fossil pollen studies have been hindered by modern-day contamination (Scott, 1995, 2002).

This study investigates the late Neogene paleoclimates of South Africa using time series of carbon and oxygen isotopes in flowstone sequences from the hominin-bearing Makapansgat Valley. In contrast to bone breccias, the speleothems offer a high-resolution and continuous stratigraphy.
Carbon and oxygen isotope studies of speleothems offer the potential of high-resolution, semi-quantitative records of paleovegetation, rainfall amount, and paleotemperature.

**Locality and stratigraphy**

Two continuous sequences of primary calcite flowstone were obtained from paleocaves in the Makapansgat Valley (Limpopo Province) of South Africa. The flowstones were collected from Collapsed Cone locality of the Makapansgat Limeworks and from Buffalo Cave, both within the Makapansgat Valley (24°08’S, 29°11’E), Limpopo Province, South Africa (Fig. 1). Buffalo Cave has yielded an early Pleistocene fauna from ex situ blocks (Kuykendall et al., 1995) and recent in situ excavations (Herries et al., 2006). Situated 1.5 km to the east of Buffalo Cave is the Plio-Pleistocene hominin-bearing Makapansgat Limeworks (Tobias, 2000; see Fig. 1).

**Makapansgat Limeworks — Collapsed Cone flowstone**

A 1.22 m-long sequence (the Collapsed Cone flowstone; see Fig. 2a) was taken from the basal flowstone deposit of the Makapansgat Limeworks (referred to as Member 1b in Partridge, 2000; Partridge et al., 2000) of Member 1b (Partridge et al., 2000); this block of flowstone fell from the roof of the cave after it was mined, and at this locality is overlain unconformably by the Member 5 breccia of Partridge (2000). Member 1b directly underlies the Member 2 red mud deposits dated by magnetostratigraphy to 4 Ma (Herries, 2003). Photograph facing west. b) Photograph of the Buffalo Cave flowstone (basal deposit of the Buffalo Cave sedimentary sequence) showing the stable isotope sampling strategy. Horizontal broken line indicates a change in the color of the flowstone, which coincides with an observed shift in carbon isotope values, Mg/Ca ratios, and the growth-rate of the flowstone (Hopley, 2004; Hopley et al., 2007). Photograph facing east.

The Makapansgat Limeworks (referred to as Member 1b in Partridge, 2000; Partridge et al., 2000). This basal flowstone unit originally consisted of two flowstone bosses of up to 20 m wide and approximately 10 m high (Latham et al., 1999, 2003). The deposit was extensively mined in the early twentieth century, leaving disconnected outcrops of the flowstone with uncertain stratigraphic relationships (Latham et al., 1999, 2003).

Faunal correlation with radiometrically dated deposits from East Africa gives the Member 3 fauna of the Makapansgat Limeworks an age of 2.8 Ma to 2.6 Ma (Vrba, 1995a; Partridge et al., 2000; Herries, 2003; see Fig. 3). Constrained
by this faunal age-estimate, a series of paleomagnetic polarity reversals have been identified and used to generate a timescale for the sedimentary sequence underlying the Member 3 faunal deposit (Herries, 2003; see Fig. 3). The majority of the Member 1b flowstone is very pure calcite that lacks the detrital iron oxides required to provide stable paleomagnetic directions. However, Member 1b flowstones from the North Alcove and Classic Section of the Main Quarry (Latham et al., 2003) show evidence of a normal polarity event (see Fig. 3), bound by reversed polarity periods, which is interpreted as the Cochiti (4.19–4.3 Ma; Herries, 2003). These samples were sourced from among the youngest layers of the Member 1b sequence, indicating that the Member 1b flowstones are likely to have a minimum age of approximately 4 Ma.

Buffalo Cave

The Buffalo Cave flowstone is the basal unit of the Buffalo Cave sedimentary sequence (Kuykendall et al., 1995; Herries et al., 2006). The age of the sedimentary sequence has been constrained by faunal correlation with radiometrically dated faunal deposits from East Africa and by a series of paleomagnetic polarity reversals (Herries et al., 2006) that place the Olduvai event towards the base of the flowstone deposit. The timescale for the Buffalo Cave flowstone was further constrained by Hopley et al. (2007) by matching the amplitude modulation of the flowstone oxygen isotope record to that of the Pliocene/early Pleistocene precession index (Laskar et al., 2004). Tuning proceeded as though maximum δ^{18}O corresponds to maximum precession and maximum solar insolation, based on our interpretation of the late Pleistocene to Recent speleothem record from Cold Air Cave (Holmgren et al., 1999, 2003).

The orbital tuning of Hopley et al. (2007) is used throughout this article and is featured in Figs. 7 and 8. We are confident in our assignment of the oxygen isotope record amplitude modulation to that of the Pliocene/early Pleistocene precession index (Laskar et al., 2004), within the age constraints provided by paleomagnetic analysis. Given inherent uncertainties, we cannot exclude the possibility of alternative chronological interpretations, although we consider our age model to be by far the best interpretation of the available data. Alternative age models do not significantly affect the discussion of orbitally forced climate change nor do they alter the broad conclusions of this work.

Methods

Stable isotope and trace element methods

The Buffalo Cave flowstone (2.41 m long, 482 samples) and Collapsed Cone flowstone (1.22 m long, 239 samples) were sampled continuously for stable isotope analysis at 5 mm intervals using a diamond-tipped microdrill with a diameter of 1.5 mm. The powdered samples were pretreated in an oxygen plasma asher to remove organic matter. The samples were then reacted to completion at 90 °C with 100% phosphoric acid using an automated VG Isocarb preparation system. The CO2 released by the reaction was cryogenically purified prior to the measurement of carbon and oxygen isotope ratios on an automated VG SIRA 12 mass spectrometer at the
University of Liverpool. All data were corrected for 17O effects following Craig (1957). Carbon and oxygen isotope data are reported in conventional delta (δ) notation in "parts per mil" (‰) relative to V-PDB (Pee Dee Belemite; Coplen, 1995). Accuracy and reproducibility of the isotopic analyses was assessed by replicate analysis of BCS2 (internal calcite standard) against NBS-19 and two internal calcite standards. Long-term laboratory reproducibility is better than 0.1‰ for both isotope ratios.

δ13C values of organic matter were also determined for samples of both flowstones. The Buffalo Cave flowstone contained acid insoluble organic matter (e.g., humic acids) that was extracted by centrifugation, following dissolution of 5–15 g of flowstone (approximately 1 cm in vertical thickness) in 2 M HCl. Carbon dioxide for carbon isotope ratio measurement was obtained by combusting the freeze-dried humic fraction with Cu(II)O and silver wire in quartz tubes at 850 °C for two hours prior to isotopic analysis. The resultant CO2 was separated cryogenically and carbon isotope ratios were measured on an automated VG SIRA 12 mass spectrometer at the University of Liverpool. Accuracy and reproducibility were determined by replicate analysis of USGS 24 graphite standard and an internal humic acid standard. Long-term reproducibility (1σ) is ±0.25‰.

The Collapsed Cone flowstone contained acid soluble organic matter (e.g., fulvic acids) that remained in solution after dissolution of the flowstone samples in 2 M HCl. The solution was subjected to wet oxidation using the persulfate wet oxidation method which converts dissolved organic compounds to CO2 by chemical decomposition (Darnifer et al., 1999; Peltzer et al., 1996). The isotopic composition of dissolved organic carbon was measured using a modified wet oxidation total organic carbon analyzer coupled with a continuous flow Finnigan MAT Delta Plus mass spectrometer at the University of Ottawa, Canada (St-Jean, 2003). Samples were calibrated against NBS 21 graphite and IAEA CH-6 sucrose international standards. Analytic precision (1σ) of dissolved organic carbon is approximately ±0.5‰.

For trace element analysis, aliquots of approximately 5 mg of powdered speleothem were dissolved in dilute nitric acid and analyzed for calcium (Ca), magnesium (Mg), and strontium (Sr) on a Perkin Elmer Optima 3300RL inductively coupled plasma atomic emission spectrometer (ICP-AES) at Royal Holloway, University of London. Analytical precision was determined using an internal speleothem standard. Reproducibility (2σ) of Mg/Ca molar ratios is ±0.00065.

Time series analysis methods

The relationship between the δ13C and δ18O data was assessed using coherency and phase spectra for both the Collapsed Cone and Buffalo Cave flowstones. Coherency is a measure of the similarity of the amplitude variations in two time series determined at particular frequencies and ranges from 0 (zero coherency) to 1.0 (perfectly coherent). A phase spectrum describes the average difference in phase between two time series at different frequencies. Phase difference ranges from +180° to −180°, with a zero phase difference indicating oscillations that are “in phase” and ±180° for oscillations that are in “antiphase”. Since phase errors are very large for low coherency, phase values have only been plotted at frequencies where the coherency is significant. Positive phase indicates that the first series leads the second, and negative phase that the first series lags the second. Coherency and phase spectra were based on the discrete Fourier transform spectral estimates for irregularly spaced data, and were generated using a modification (by G.P.W.) of the Bloomfield (1976) algorithm.

Results

Carbon isotopes in organic matter

Collapsed Cone flowstone organic matter δ13C ranges from −31.6 to −25.5 with a mean value of −28.8 (n = 7; see Fig. 4). As shown in Fig. 4, the Collapsed Cone δ13C organic matter values lie entirely within the range of modern-day C3 vegetation (Vogel et al., 1978; O’Leary, 1981). The observed variation in δ13C of organic matter in the Collapsed Cone flowstone is indicative of the natural δ13C variation of C3 vegetation within the soil-zone (Ehleringer et al., 2000; Powers and Schlesinger, 2002). Low δ13C values (<−30‰) are typical for C3 forest plants that are affected by canopy or irradiance effects (Farquhar et al., 1989). This suggests that the low δ13C values of the Collapsed Cone flowstone organic matter may be indicative of a closed or forested environment. However, δ13C values of organic matter are known to vary depending on the organic fraction analyzed (e.g., Rieley et al., 1991; Freeman and Colarusso, 2001). Therefore, if the 1.2‰ depletion of humic and fulvic acids relative to total soil organic matter (Lichtfouse et al., 1995a,b) is accounted for, the estimated mean δ13C value of bulk soil organic matter is −27.6, close to the mean C3 vegetation δ13C value of −28‰ (O’Leary, 1988).

Buffalo Cave flowstone organic matter δ13C values ranged from −27.3 to −21.1 with a mean value of −23.7 (n = 27; see Fig. 4). These δ13C values are intermediate between the C3 and C4 vegetation end-members (see Fig. 4), in contrast to the purely C3 vegetation values of the Collapsed Cone flowstone. This reconstruction is consistent with the mixed savanna-woodland, composed of both C3 and C4 vegetation, which is present in northern South Africa in both the modern-day and in the Plio-Pleistocene (Sponheimer et al., 1999; Lee-Thorp and Talma, 2000).

Carbon isotopes in speleothem carbonate

The speleothem carbonate δ13C values of the Collapsed Cone flowstone (mean of −8.1‰) are significantly lower than those of the Buffalo Cave flowstone (mean of −5.7‰), as shown in Fig. 5. The δ13C values of the Collapsed Cone flowstone are relatively invariant (range of 1.7‰) when compared with the Buffalo Cave flowstone (range of 5.6‰), also shown in Fig. 5. It is likely that the different carbon isotope signatures of the two flowstones indicate that they were
formed in different vegetation regimes. Carbon isotopes in speleothem carbonate can be used as a paleovegetation proxy, provided that the isotopic signal transferred via soil CO$_2$ can be distinguished from the carbon component sourced from dissolution of the carbonate host-rock (Coplen et al., 1994). This method has the potential of providing a far higher resolution paleovegetation record than that derived from humic acid $\delta^{13}$C, due to the smaller sample sizes required. If we assume that the carbonate paleovegetation signal has been diluted with a typical host-rock proportion of 15% (e.g. Beck et al., 2001; Genty et al., 2001) and that the host-rock has a mean $\delta^{13}$C value of $-0.9\%$ (Veizer et al., 1992), this produces corrected $C_3$ and $C_4$ plant end-members of $-8.0\%$ and $+1.3\%$, respectively (following the methodology of

![Graph](image-url)

Fig. 4. $\delta^{13}$C values of organic matter within the Collapsed Cone and Buffalo Cave flowstones. $C_3$ and $C_4$ plant distributions (grey) are a combination of data from Vogel et al. (1978) and O’Leary (1988).

![Graph](image-url)

Fig. 5. (a) Carbon and oxygen isotope cross-plot for the Collapsed Cone and Buffalo Cave flowstones indicating their mutually exclusive isotopic composition. (b) 1000 Mg/Ca and 1000 Sr/Ca molar ratios for the Collapsed Cone and Buffalo Cave flowstones indicating greater trace element variability in the Buffalo Cave sequence.
Genty et al., 2001). Using these end-members, the carbon isotope values of the Collapsed Cone flowstone (ranging from $-8.8_{\text{oo}}$ to $-7.1_{\text{oo}}$ with a mean value of $-8.1_{\text{oo}}$, n = 239), are indicative of a paleoenvironment composed entirely of C3 vegetation (in agreement with the organic matter carbon isotope data; see Fig. 4). The samples taken from elsewhere in the Makapansgat Limeworks Member 1 sequence (Original Ancient Entrance, North Alcove, and the Entrance Quarry) also indicate a purely C3 environment ($\delta^{13}$C values ranging from $-8.6$ to $-7.8_{\text{oo}}$; n = 5). The low degree of carbonate $\delta^{13}$C variation (range of 1.7$_{\text{oo}}$) in the Collapsed Cone flowstone occurs despite more variable $\delta^{18}$O values (see Fig. 5), as is typically observed in purely C3 environments (e.g., Quade et al., 1989; Cerling, 1992; Cerling et al., 1993; France-Lanord and Derry, 1994).

The range of measured $\delta^{13}$C carbonate values for the Buffalo Cave flowstone ($-8.2_{\text{oo}}$ to $-2.6_{\text{oo}}$) can be converted to a percentage of C4 plants ranging from 0% to 67%, with an uncertainty of approximately 10% (see Fig. 7). The lower part of the Buffalo Cave carbonate record (1.99 to 1.70 Ma) has significantly lower $\delta^{13}$C values (mean = $-5.7_{\text{oo}}$, s.d. = 1.0$_{\text{oo}}$) than the upper part of the record (1.70 to 1.52 Ma; mean = $-4.4_{\text{oo}}$, s.d. = 0.7$_{\text{oo}}$). The rapid increase in average carbon isotope values of 1.3$_{\text{oo}}$ between 1.78 Ma and 1.69 Ma is interpreted to reflect an increase in the mean proportion of C4 grasses (see Fig. 7). The marked difference in the isotopic composition and isotopic variability of the two flowstones is indicative of deposition under two contrasting climatic regimes, before and after the spread of C4 grasses in ecosystems (see Fig. 8).

Oxygen isotope results

It is likely that the long-term increase in marine $\delta^{18}$O values from the late Neogene to the Holocene, caused by increasing global ice volume, is responsible for the increase in the $\delta^{18}$O values of the Makapansgat speleothems over this time period. The mean $\delta^{18}$O value of the Collapsed Cone flowstone is $-5.7_{\text{oo}}$ (ranging from $-7.0_{\text{oo}}$ to $-4.5_{\text{oo}}$), whereas the mean $\delta^{18}$O value of the Buffalo Cave flowstone is significantly higher at $-4.9_{\text{oo}}$ (ranging from $-6.5_{\text{oo}}$ to $-3.4_{\text{oo}}$). Both of these speleothems have significantly lower mean $\delta^{18}$O values than that of the Holocene stalagmite T8 from Cold Air Cave (mean $\delta^{18}$O of $-3.7_{\text{oo}}$; Holmgren et al., 2003). The glacial-interglacial variability of the ice volume effect at and around the Pliocene-Pleistocene boundary was less than 1$_{\text{oo}}$ (Shackleton and Kennett, 1975; Shackleton, 1995, 1997), indicating that ice volume effects alone are not sufficient to explain the 1 to 2$_{\text{oo}}$ variability of the $\delta^{18}$O record from Buffalo Cave (see Fig. 7). The remaining $\delta^{18}$O variability must be explained by changes in the isotopic composition of rainfall.

Insights into oxygen isotope variability of African rainfall (Vuille et al., 2005) demonstrate a link between the $\delta^{18}$O composition of rainfall over eastern and southern Africa and the interannual variability of circulation in the Indian Ocean, known as the Indian Ocean Dipole (IOD). The anomalous years of the IOD are characterized by a reversal in the sign of the Sea Surface Temperature (SST) gradient (from east to west) and change in the direction of surface zonal winds from westerlies to easterlies across the ocean basin (Saji et al., 1999; Webster et al., 1999). Associated with the anomalously warm SSTs in the western Indian Ocean is an increase in monsoon rainfall over tropical East Africa and a significant increase in the $\delta^{18}$O of this rainfall (Vuille et al., 2005). During southern hemisphere precessional maxima, when the Intertropical Convergence Zone (ITCZ) had migrated southward, the Makapansgat Valley of South Africa (24°S) would have received heavier $\delta^{18}$O values with increased rainfall amounts, as occurs in eastern Africa today. During precessional minima, the northward expansion of the ITCZ and the Equator-ward expansion of the westerlies would have brought cooler and drier air from the Atlantic to northeastern South Africa (Cockcroft et al., 1987; Tyson, 1999). With a large landmass to the southwest of the Makapansgat Valley, it is likely that the continental effect would have influenced the oxygen isotopic composition of rainfall during these precessional minima, resulting in lower $\delta^{18}$O values. On the basis of these arguments, we suggest that the precessional cycle in the Makapansgat Valley was characterized by increased $\delta^{18}$O during humid insolation maxima and lower $\delta^{18}$O during dry insolation minima. The most likely interpretation of the Buffalo Cave and Collapsed Cone flowstone $\delta^{18}$O time series is that they are predominantly records of monsoon intensity over precessional cycles, and indicators of global ice volume source effects over longer time frames.

Time series analysis results

The Collapsed Cone flowstone has spectral peaks above the 95% confidence level with wavelengths of 34 cm for $\delta^{18}$O and 23 cm for $\delta^{13}$C within the 122-cm-long record (see Fig. 6). The coherency and phase relationships between $\delta^{18}$O and $\delta^{13}$C do not exceed the 90% confidence level at the 34 cm or the 23 cm cycle frequencies (see Fig. 6). At least six cycles at a particular wavelength are required to demonstrate regular cyclicity (Weedon, 2003), so with only 3.6 and 5.3 oscillations, respectively, of these low frequency cycles, it is not possible to identify regular cyclicity within the Collapsed Cone flowstone depth series at these frequencies. Time series analysis of the Collapsed Cone flowstone cannot be taken any further with the existing dataset, but there is plenty of scope for extension of the stable isotope record in the future.

Band-pass filtering (Weedon, 2003) of the Buffalo Cave flowstone $\delta^{18}$O data was used to isolate the cycles associated with the shortest wavelength spectral peaks. The filtered data show strong amplitude modulations reminiscent of the orbital precession index that permitted orbital tuning as described in Hopley et al. (2007). The tuning indicates that the sampled flowstone dates from 1.990 to 1.518 Ma. Power spectra for the $\delta^{18}$O and $\delta^{13}$C records from the flowstone carbonate have evidence for the same orbital cycles. However, $\delta^{18}$O was dominated by precession cycles (22.8 and 19.4 kyr), whereas $\delta^{13}$C was dominated by obliquity cycles (37.5–40.2 kyr).
A visual comparison of the δ¹³C and δ¹⁸O records for the Buffalo Cave flowstone (Fig. 7) indicates an inverse correlation (phase of ±180°) after 1.77 Ma and the lack of simple relationship prior to this time. Therefore, to investigate the relationship between δ¹³C and δ¹⁸O using cross-spectral analysis, the tuned Buffalo Cave record was divided into two sections above and below 1.77 Ma. Figure 7 shows that after 1.77 Ma, the two isotopic records are coherent at both the precession (ca. 21 kyr) and obliquity (41 kyr) frequencies. At the precession frequency the two records are in antiphase (phase = ±180°), but at the obliquity frequency δ¹⁸O leads δ¹³C by a quarter of a cycle (phase = −90°). Before 1.77 Ma (Fig. 7), both the precession and obliquity frequencies are coherent and show different phase relationships. δ¹⁸O lags δ¹³C by a quarter of a cycle (phase = +90°) at the precessional frequency, whereas at the obliquity frequency, the two records are approaching antiphase (phase = +130° to +180°; Fig. 7).

The increase in phase difference at ca. 1.77 Ma indicates that either the monsoon rainfall or the Northern Hemisphere ice volume was nonstationary relative to orbital forcing, most likely the latter (Ruddiman, 2006; Wang et al., 2005). Clemens et al. (1996) showed a similar systematic increase in phase difference between the Asian monsoon and Northern Hemisphere ice volume over the last 2.6 Ma at both the precession and obliquity bands, including an increase in phase difference at approximately 1.7 Ma. This is further evidence for the restructuring of global and tropical climate at and around 1.7 Ma.
Fig. 7. Buffalo Cave flowstone $\delta^{18}O$ and $\delta^{13}C$ time series and associated spectra. a) Buffalo Cave flowstone $\delta^{18}O$ time series (see Hopley et al., 2007) filtered and tuned to the orbital solution from 1.52 Ma to 1.98 Ma (Laskar et al., 2004). b) Orbitally tuned $\delta^{18}O$ and $\delta^{13}C$ time series filtered at precessional frequencies (19.4–22.8 kyr). c) Orbitally tuned $\delta^{18}O$ and $\delta^{13}C$ time series filtered at the obliquity frequency (40 kyr). d) Buffalo Cave flowstone $\delta^{13}C$ time series (see Hopley et al., 2007) filtered and tuned to orbital precession (Laskar et al., 2004). The 40-kyr obliquity cycle is the dominant periodicity in the $\delta^{13}C$ record, whereas the ca. 20-kyr precessional cycles dominate the $\delta^{18}O$ record. Thick lines on the $\delta^{18}O$ and $\delta^{13}C$ plots indicate the changing average values as determined by the RAMPFIT program (Mudelsee, 2000); there is a rapid change in $\delta^{18}O$ and $\delta^{13}C$ values between 1.8 Ma and 1.7 Ma. The % C$_4$ grasses values are determined using organic matter $\delta^{13}C$ values to correct for the host-rock carbon contribution to carbonate $\delta^{13}C$. The % C$_4$ grasses values indicate that the Buffalo Cave flowstone formed in a mixed C$_3$ and C$_4$ plant environment, in which the proportion of the two plant types was highly variable. e) Spectral analysis and phase relationships of $\delta^{13}C$ and $\delta^{18}O$ for the bottom half of the flowstone (1.99–1.77 Ma). At the 22-kyr periodicity, $\delta^{13}C$ leads $\delta^{18}O$ by approximately a quarter of a cycle ($+90^\circ$). e) Spectral analysis and phase relationships of $\delta^{13}C$ and $\delta^{18}O$ for the top half of the flowstone (1.77–1.52 Ma). At the 19-kyr periodicity, $\delta^{18}O$ leads $\delta^{13}C$ by approximately a quarter of a cycle ($-90^\circ$). BW = bandwidth; CL = confidence level.
Discussion

Orbital forcing in the Plio-Pleistocene of South Africa

The Buffalo Cave δ18O time series is dominated by the 19-kyr and 23-kyr precession signal, and also contains a minor 40-kyr obliquity component (Hopley et al., 2007; see Fig. 7). Precessional forcing of monsoon intensity in subtropical terrestrial environments is both an observed and modelled phenomenon throughout the late Neogene (Rossignol-Strick, 1983; Prell and Van Campo, 1986; Pokras and Mix, 1987; Clemens et al., 1991, 1996; deMenocal and Bloemendal, 1995; Partridge et al., 1997; Tyson, 1999; Wang et al., 2001; Cruz et al., 2005). In contrast to the obliquity cycle (40 kyr), with the 23-kyr and 19-kyr precessional cycles occurring as minor components (see Fig. 7). The obliquity cycle is known to force high-latitude climates, as variation in insolation over the obliquity cycle is greatest at high latitudes (Ruddiman et al., 1986; Raymo and Nisancioglu, 2003). Obliquity is the dominant periodicity in marine δ18O variation during the late Pliocene and early Pleistocene (Ruddiman et al., 1986; Raymo and Nisancioglu, 2003), rather than the dominant eccentricity cycles observed in the late Pleistocene (Shackleton, 1995). The presence of the obliquity cycle in the Buffalo Cave δ13C time series suggests that the proportion of C4 grasses may be responding to changes in the atmospheric partial pressure of CO2, in agreement with carbon isotope proxies from the late Pleistocene (Collatz et al., 1998; Boom et al., 2001) and the West African Plio-Pleistocene pollen record of Dupont and Leroy (1995).

The negative correlation between δ18O and δ13C in the Buffalo Cave flowstone at the precessional frequencies (see Fig. 7) provides further support for a link between the proportion of C4 grasses and the amount of precipitation in southern Africa. Sankaran et al. (2005) demonstrate that modern-day savannas of tropical Africa are stable in regions that receive less than 650 mm of mean annual precipitation (MAP); however, above this precipitation threshold, the savanna environment is unstable and heads towards the new equilibrium state of a closed canopy forest. Over the last 200 kyr in Pretoria, mean annual precipitation has oscillated between 800 mm and 500 mm at the precessional frequencies (Partridge et al., 1997), indicating that the 650 mm MAP threshold of Sankaran et al. (2005) was routinely crossed in the past. This explains the repeated cycle in vegetation observed in the Buffalo Cave flowstone at the precessional frequencies (see Fig. 7) in which drier conditions (lower δ18O values) were associated with an increased percentage of savanna grasses (higher δ13C values).

Climatic variability and early hominin habitats in South Africa

Isotopic shifts between 1.8 Ma and 1.7 Ma are evident in both proxy records, the carbon isotopic shift being of greater magnitude than the oxygen (see Fig. 7). This is interpreted as a reduction in precipitation and a corresponding increase in C4 grasses, similar in magnitude and polarity to the relationship observed over the precessional frequencies (see Fig. 7). A similar shift in C4 grass biomass proportions between 1.8 and 1.6 Ma has also been recorded at Sterkfontein, 200 km to the south of Buffalo Cave, and in equatorial Africa (Bonnefille, 1995; deMenocal, 2004; Bobe and Behrensmeyer, 2004; Wynn, 2004). This event has been linked to a period of faunal turnover (Vrba, 1995a; deMenocal, 2004) that is characterized by an increased proportion of savanna-adapted species (Vrba, 1995a). These observations indicate a pan-African shift in the composition of vegetation in the early Pleistocene, between 1.8 and 1.7 Ma, supporting the hypothesis that a restructuring of tropical climate (Ravelo et al., 2004; Wara et al., 2005) led to greater aridity and increased savanna grassland at this time (Wynn, 2004).

Previous studies have assumed that the South African faunal deposits were formed under constant ecological conditions, and consequently the paleoenvironment has been interpreted as a highly mixed composite of forest, grasslands, and wetlands (Reed, 1997, 1998; Sponheimer et al., 1999, 2001). While the present study indicates a mixed environment during the Plio-Pleistocene, it also highlights the rapidity with which the environment oscillated between forest-dominated and grass-dominated end-members. Faunal deposits rarely have temporal resolutions better than 100 kyr (Reed, 1997; Sponheimer et al., 1999; Luyt and Lee-Thorp, 2003), which suggests that most Plio-Pleistocene African faunal deposits are likely to be time-averaged amalgamations derived from both forest-dominated and grass-dominated environmental end-members. The Buffalo Cave flowstone time series offers a far greater temporal resolution than the coeval macrofaunal evidence, aiding our ability to assess to what degree climatic forcing was responsible for the speciation and extinction events within local mammalian communities.

In addition to evidence for mammalian turnover in Africa between 1.8 Ma and 1.7 Ma (Vrba, 1995a; Bobe and Behrensmeyer, 2004; deMenocal, 2004), the evolution of African Homo erectus must also be considered within the context of increasing savanna grassland distribution. It has been suggested that H. erectus was the first hominin to develop a savanna-adapted morphology (Ruff, 1991), as indicated by the similarity of its body shape and limb bone proportions to those of modern people living in tropical savanna grasslands. The earliest occurrence of H. erectus in East Africa is from Koobi Fora, Kenya, at ca. 1.7 Ma (based on KNM-ER 3733 and the chronology of Gathogo and Brown, 2006), and the earliest record in South Africa is at ca. 1.7–1.4 Ma from Sterkfontein (Kuman and Clarke, 2000). Both dates coincide with the spread of savanna grassland discussed in this study, which we attribute to the onset of the Walker Circulation and increased African aridity at this time. While it is currently speculative to invoke climatic forcing of events in the hominin fossil record, it is apparent that there is both a temporal correlation and a plausible mechanism (obligate bipedalism and body shape modification as adaptations to the savanna environment) to link the evolution of H. erectus in Africa with the restructuring of tropical climate and the spread of savanna grasslands.
The Collapsed Cone flowstone has low carbon isotope values indicative of a purely C3 vegetation environment (see Figs. 4–6, 8). Given the warmer conditions of the late Neogene (Billups and Schrag, 2002), it is unlikely that this represents the vegetation response to a significant northward expansion of the winter rainfall zone (currently limited to the Cape region of South Africa). Instead, the lack of C4 grasses signifies a difference in the underlying values of atmospheric pCO2, temperature, and rainfall amount (e.g., Ehleringer et al., 1997; Cerling et al., 1998; Huang et al., 2001) in the late Miocene of South Africa, which combined to produce conditions that were not favorable for the growth of C4 grasses. For example, the warmer conditions of the late Miocene would have increased photorespiration, favoring plants with the C4 photosynthetic pathway; however, the increased temperatures were likely to have increased monsoon rainfall intensity, perhaps promoting the growth of C3 vegetation. With the combined rainfall and temperature effects resulting in little overall change in vegetation type, it is also necessary to invoke an increase in atmospheric pCO2 during the late Miocene (relative to the Plio-Pleistocene) to explain the lack of C4 vegetation at this time. Current estimates suggest that atmospheric pCO2 concentrations were higher in the late Miocene than in the Pleistocene (Royer et al., 2001); this would have led to an increased photosynthetic yield amongst the C3 plants, and to their competitive advantage over C4 plants. We suggest that the C3-dominated environment of the late Miocene of South Africa was sustained through both higher levels of atmospheric pCO2 and higher rainfall amounts during the austral summer growing season, relative to the modern day.

The carbon isotope evidence presented in this study indicates that the transition from C3 to C4 vegetation in the summer rainfall zone of southern Africa occurred between the late Miocene and the late Pliocene (between approx. 4–6 Ma; see Fig. 9). The earliest evidence for the presence of C4 vegetation at the Makapansgat Limeworks comes from carbon isotope data for the Member 3 (ca. 2.6–3.0 Ma) bovid fauna (Sponheimer et al., 1999), and the micromammal faunas (ca. 3.3–3.5 Ma) from the Exit Quarry and Rodent Corner deposits (Hopley et al., 2006). The absence of C4 grasses towards the base of the Makapansgat Limeworks sedimentary sequence (Member 1b) is indicated by the low carbon isotope values of the Collapsed Cone flowstone (see Fig. 8). This flowstone has a minimum age of 4 Ma on the basis of magnetostratigraphy constrained by faunal correlation. A maximum age for the flowstone is currently unknown, but with the lack of an observable hiatus, it is unlikely to be greater than 5 Ma. The age estimate for the first C4 grasses in South Africa of 4–6 Ma is similar to the first occurrence of C4 grasses at 2–3.5 Ma in Namibia (see Fig. 9) at a latitude of 25–28°S (Ségalen et al., 2002).

The Makapansgat speleothems provide further evidence that the expansion of C4 grasses into Southern Africa occurred later than the global expansion of C4 biomass between 8 Ma
and 6 Ma documented by Cerling et al. (1997). In combination with evidence from other mid-latitude sites, such as northern China (Ding and Yang, 2000; Jiang et al., 2002) and northern North America (Cerling et al., 1997), it is apparent that the expansion of C4 grasses extended towards the end of the Neogene (see Fig. 9). This can be explained in the context of gradual global cooling as atmospheric pCO2 decreased, the Northern Hemisphere ice sheet expanded, ocean circulation was modified, and rainfall over eastern and southern Africa was reduced. The reduction in atmospheric pCO2 during the Neogene reduced the temperature threshold of photorespiration in C3 plants (Ehleringer et al., 1991, 1997; Ehleringer and Monson, 1993), and pushed the range of C4 grasses into mid-latitudes. Regions that experienced a reduction in rainfall would have undergone further C4 grass expansion. The premise of a gradual expansion of C4 grassland is supported by faunal evidence for prolonged faunal turnover (Köhler et al., 1998; Barry et al., 2002) and gradual increases in hypsodonty (Jernvall and Fortelius, 2002) in the late Neogene. The faunal evidence suggests that the late Neogene faunal response to the spread of the savanna biome occurred gradually over millions of years with periods of peak turnover that varied in timing between regions. The overall result of this gradual change was a Plio-Pleistocene savanna-woodland-adapted fauna that differed markedly from the mid-Miocene forest-adapted faunas.

**Conclusions**

Two vegetational shifts towards increased savanna grasslands are observed within the Makapansgat Valley flowstones between the late Miocene/early Pliocene (see Fig. 8) and at about 1.7 Ma (see Fig. 7). Both of these events would have led to a restructuring of terrestrial ecosystems of importance to mammalian evolution, but pass largely unnoticed in the marine δ18O record. The late Miocene/early Pliocene vegetation shift represents the first occurrence of C4 grasses in the South African summer rainfall zone and was likely to have been triggered by falling levels of atmospheric pCO2, and, to a lesser extent, a reduction in rainfall over southern Africa. The increase in C4 grasses at 1.7 Ma is related to the reorganization of tropical climate at this time, specifically the onset of the Walker Circulation (Ravelo et al., 2004; Wara et al., 2005), which led to drier conditions over eastern and southern Africa. These two climatic and vegetational events coincide with key evolutionary changes such as the origin of bipedalism and the evolution of early Homo, respectively, and are likely to have played a role in these evolutionary developments.

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