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       Late Quaternary climate variability at Mfabeni peatland, eastern South Africa
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       Abstract
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15 The scarcity of continuous, terrestrial, palaeoenvironmental records in eastern South Africa leaves the evolution of late Quaternary climate and its driving mechanisms uncertain. Here we use a 16 17 ~7-m long core from Mfabeni peatland (KwaZulu-Natal, South Africa) to reconstruct climate 18 variability for the last 32 thousand years (cal ka BP). We infer past vegetation and hydrological variability using stable carbon ( $\delta^{13}C_{wax}$ ) and hydrogen isotopes ( $\delta D_{wax}$ ) of plant-wax *n*-alkanes and use 19 20 Pag to reconstruct water table changes. Our results indicate that late Quaternary climate in eastern South Africa did not respond directly to orbital forcing nor to changes in sea surface temperatures 21 (SSTs) in the western Indian Ocean. We attribute the arid conditions evidenced at Mfabeni during the 22 23 Last Glacial Maximum (LGM) to low SSTs and an equatorward displacement of: i) the southern hemisphere westerlies, ii) the subtropical high-pressure cell and iii) the South Indian Ocean 24 25 Convergence Zone (SIOCZ), which we infer was linked to increased Antarctic sea-ice extent. The northerly location of the high-pressure cell and the SIOCZ inhibited moisture advection inland and 26 27 pushed the rain-bearing cloud band north of Mfabeni, respectively. The increased humidity at Mfabeni between 19–14 cal kyr BP likely resulted from a southward retreat of the westerlies, the 28 29 high-pressure cell and the SIOCZ, consistent with a decrease in Antarctic sea ice extent. Between 14-5 cal kyr BP, when the westerlies, the high-pressure cell and the SIOCZ were in their southernmost 30 31 position, local insolation became the dominant control, leading to stronger atmospheric convection 32 and an enhanced tropical easterly monsoon. Generally drier conditions persisted during the past c. 5 cal ka BP, probably resulting from an equatorward return of the westerlies, the high-pressure cell and 33 the SIOCZ. Higher SSTs and heightened ENSO activity may have played a role in enhancing climatic 34 variability during the past c. 5 cal ka BP. Our findings highlight the influence of the latitudinal position 35

of the westerlies, the high-pressure cell and SIOCZ in driving climatological and environmental
 changes in eastern South Africa.

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Key words: Mfabeni; eastern South Africa; *n*-alkanes; hydrogen isotopes; carbon isotopes; southern
 hemisphere westerlies; tropical easterlies

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#### 42 **1.** Introduction

43 Eastern South Africa is an important region for scientific focus, specifically for furthering our 44 understanding of regional and global climate dynamics. The region is particularly dynamic and 45 sensitive to long-term climate change as it lies within a climatic transition zone, where it is strongly influenced by both temperate (southern westerlies) and tropical (tropical easterlies) climate systems. 46 47 In eastern South Africa, modelled precipitation reductions and projected regional warming (3–6°C by 2099), threaten the stability of current ecosystems in a region populated by communities already 48 49 economically vulnerable to the effects of climate change (IPCC, 2013). Past climate and environmental reconstruction and the determination of climate driving mechanisms will provide 50 valuable information for assessing future climate and environmental trends in the region. 51

Changes in vegetation, precipitation and temperature from the beginning of the Last Glacial 52 53 Maximum (LGM; c. 26.5 ka; Clark et al., 2009) to the present-day are poorly constrained in eastern 54 South Africa. Whether this region was characterized by aridity or increased humidity during the last 55 glacial period remains unclear. Proxy data show spatial complexity (e.g., Baker et al., 2016; Chase et 56 al., 2017; Chevalier and Chase, 2015 & 2016, Dupont et al., 2011; Schefuß et al., 2011; Scott et al., 2012; Scott, 2016; Schmidt et al., 2014; Simon et al., 2015), and modelled LGM (26.5–19 ka; Clark et 57 58 al., 2009) precipitation patterns for the region are highly variable and often do not even agree on the sign of precipitation change. For example, the PMIP3 model ensemble mean suggests increased LGM 59 60 precipitation in the east of South Africa with dry conditions towards the north (compared to the present day; Braconnot et al., 2007; Chevalier et al., 2017). Conversely, the NCAR CCSM3 model 61 62 indicates drier than present conditions in the centre of South Africa and along the eastern coast (Otto-Bliesner et al., 2006). These contrasting simulations for the last glacial period highlight the 63 difficulty in simulating past precipitation in South Africa, with a lack of a comprehensive 64 understanding regarding the relevant climate processes involved (Stone, 2014). 65

The mechanisms driving Quaternary climate variability in South Africa are complex and spatially heterogeneous. For example, hydroclimate may be paced by austral summer insolation fluctuations, resulting from changes in the Earth's orbital precession on 23–19 ka timescales. Strong summer insolation (during precession maxima) causes stronger atmospheric convection and an increase in the land/ocean temperature contrast, which results in higher moisture transport by the 71 tropical easterlies and higher precipitation in eastern South Africa (e.g., Simon et al., 2015; Chevalier and Chase, 2015). Climate may also be influenced by high-latitude forcing related to changes in the 72 73 Earth's orbital obliquity and eccentricity on longer, i.e., glacial-interglacial timescales, which may 74 result in the latitudinal contraction and expansion of the climatic belts (e.g., Dupont, 2011). The 75 model of Nicholson and Flohn (1980) suggests an equatorward displacement of the tropical rainbelt (Nicholson, 2008) during the last glacial period, although proxy data from South Africa provide no 76 77 conclusive support for this scenario. In addition, during glacial periods, the Walker Circulation may 78 have been weaker with its ascending limb further to the east, over the Indian Ocean (e.g., DiNezio et 79 al., 2018). This possibly resulted in an eastward displacement of the cloud band (SIOCZ) and thus a 80 drier summer rainfall zone (SRZ; Tyson, 1999). Furthermore, changes in the latitudinal position of the 81 southern hemisphere westerlies (as a response to fluctuations in Antarctic sea ice extent) have been 82 invoked to influence climate in South Africa (Chase and Meadows, 2007; Chevalier and Chase, 2015; 83 Chase et al., 2017). The western South African region has received most focus regarding the southern 84 hemisphere westerly influence in controlling climate variability (e.g., Stuut et al., 2004; van Zinderen 85 Bakker, 1976). Some studies also suggest possible mechanistic links between SSTs in the Agulhas Current and the Indian Ocean and rainfall variability in South Africa, with high SSTs linked to 86 87 increasing South African summer precipitation (e.g., Baker et al., 2017; Chevalier and Chase, 2015; Dupont, 2011; Dupont et al., 2011; Reason and Mulenga, 1999). Climate forcing experiments also 88 89 indicate that changes in greenhouse gas concentrations may have driven eastern South African 90 rainfall changes, increasing precipitation between 17-11 kyr (Otto-Bliesner et al., 2014).

91 The spatially heterogeneous nature of climate variability in South Africa from the last glacial 92 period to the present-day, and the multiple possible climate drivers render the region an important 93 focus for palaeoclimate research. Two important questions remain: i) what was the climate like in 94 eastern South Africa during the last glacial period? and, ii) what were the causes for the climate 95 variability? These questions are difficult to answer with the majority of long, continuous, terrestrial records situated further north, within the range of the modern tropical rainbelt (e.g., Barker et al., 96 97 2007; Tierney et al., 2008), making it hard to assess the long-term climate drivers in the south, in 98 particular in eastern South Africa. In this area, terrestrial sediment archives suitable for 99 palaeoenvironmental reconstruction are scarce, in particular those extending to the LGM. Marine and speleothem archives have hitherto mostly formed the basis of Quaternary climate research in 100 101 this region (e.g., Dupont et al., 2011; Holmgren et al., 2003). Here we provide stable carbon ( $\delta^{13}$ C) 102 and hydrogen ( $\delta D$ ) isotope records of terrestrial plant-waxes (long-chained *n*-alkanes) from Mfabeni 103 peatland, one of the longest continuous terrestrial archives from South Africa. Our vegetation and 104 hydroclimate reconstructions are compared with a previous biomarker-palaeoclimate study from 105 Mfabeni (Baker et al., 2014, 2016 & 2017). We more than double the temporal resolution of the 106 previous plant-wax  $\delta^{13}$ C record from Baker et al. (2017), from *c*. 1200 to *c*. 500 years, revealing 107 important and previously undocumented environmental variability.

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## 109 2. Regional setting

110 The mid-latitude westerlies, in association with the subtropical high (and low) pressure cells 111 and the SIOCZ, play critical roles in determining climatic conditions across the whole South African 112 continent (Fig. 1; Dyson and van Heerden, 2002; Macron et al., 2014; Munday and Washington, 2017). During austral winter, an anti-clockwise rotating subtropical high-pressure cell is located over 113 114 southern Africa, which drives large-scale subsidence and suppresses rainfall (Fig. 1b). This high-115 pressure cell creates a blocking effect over the continent, which stops moisture advection inland over the majority of South Africa during winter (Dedekind et al., 2016), except for in the winter rainfall 116 117 zone (WRZ), where the westerlies bring rainfall. During summer, the high-pressure cell shifts to the 118 south, and the Angola and Kalahari low pressure cells dominate the continent, enabling monsoonal 119 systems (tropical easterlies) to penetrate southern Africa, bringing rainfall to the summer rainfall 120 zone (SRZ; Fig. 1a; Tyson and Preston-Whyte, 2000; Munday and Washington, 2017). The dominant 121 rain-producing mechanism in the SRZ during the summer are tropical temperature troughs (TTTs), 122 which are embedded within the SIOCZ and form a northwest-southeast orientated cloud band, 123 extending over the continent into the southwest Indian Ocean (Fig. 1a; Todd and Washington 1999; Tyson and Preston-Whyte, 2000). TTTs form from interactions between tropical convection and mid-124 125 latitude perturbations, which result in heavy precipitation events (Tyson, 1986; Macron et al., 2014; Chase et al., 2017). A combination of strong easterly flux from the Indian Ocean and low pressure 126 127 over the continent during the summer results in the development of TTTs (Fig. 1a; Cook, 2000; 128 Macron et al., 2014; Rácz and Smith, 1999; Todd and Washington 1999).

129 South Africa can be divided into several climate zones: the SRZ lies in the north and east where 130 66 % of the mean annual precipitation falls between October and March (Fig. 1a; Chase and Meadows, 2007). Based on late Quaternary precipitation reconstructions, further subdivisions of the 131 SRZ (northern SRZ, central/eastern SRZ) have been suggested by Chevalier and Chase (2015). In the 132 133 extreme south and west of South Africa lies the WRZ (Fig. 1a), where 66 % of the mean annual 134 precipitation falls between April and September (Chase and Meadows, 2007). This rainfall is associated with temperate frontal systems related to the southern hemisphere westerlies (Fig. 1b; 135 136 Mason and Jury, 1997; Tyson, 1986; Tyson and Preston-Whyte, 2000). In between the SRZ and WRZ lies the year-round rainfall zone (YRZ) which receives precipitation both in summer and winter 137 138 seasons (Fig. 1a; Chase and Meadows, 2007). This zone comprises much of the southern Cape of South Africa and is highly heterogeneous in terms of precipitation seasonality and amount, spanning 139

some of the wettest (e.g., along the south coast), and driest (e.g., Namib Desert) regions in SouthAfrica.

142 Mfabeni peatland is located within the SRZ, on the coastal plain of northern KwaZulu-Natal 143 (28°09'8.1"S; 32°31'9.4"E; 9 m above sea level; Fig. 1; Fig. 2). The dominating subtropical high-144 pressure cell across the majority of South Africa during the austral winter months leads to mild and dry winter conditions at Mfabeni. Occasional rainfall during the winter months at Mfabeni is 145 associated with the passage of cold fronts, which develop in the western Atlantic and move across 146 147 southern Africa (Fig. 1b; Grab and Simpson, 2000). These cold fronts trigger rainout of atmospheric 148 moisture, which is sourced from the Indian Ocean and Agulhas region (Gimeno et al., 2010). When 149 the subtropical high-pressure cell has moved south during the austral summer, the tropical easterlies 150 dominate, TTTs form and conditions at Mfabeni are hot and humid. The average annual rainfall 151 amount between 2010 and 2018 at Mfabeni in the winter months (June-August) was measured at 152 134 mm compared to 426 mm during the summer months (December-February), meaning the majority of rainfall (76 %) falls during the summer months (data from World Weather Online). A 153 154 northeast-southwest precipitation gradient is present across the peatbog, with 1200 mm year<sup>-1</sup> of precipitation in the east decreasing to 900 mm year<sup>-1</sup> westwards towards Lake St. Lucia (Fig. 1; Fig. 2; 155 Taylor et al., 2006). The main source of water to Mfabeni is precipitation, predominantly provided by 156 157 the tropical easterlies and TTTs, sourced from the Indian Ocean and Agulhas Current region (Fig. 1; 158 Tyson, 1999; Gimeno et al., 2010). Mean summer temperatures (November to March) surpass 21 °C. 159 The wind regime is characterised by moderate northeasterly winds during the summer and more 160 intense southwesterly winds during winter.

161 Mfabeni is one of the oldest, continuously growing peatlands in South Africa (Grundling et al., 162 2013). It lies within a topographical inter-dunal depression between the Indian Ocean to the east and Lake St. Lucia to the west (Fig. 2; Grundling et al., 2013). Towards the ocean, it is bordered by an 80-163 164 100 m high vegetated dune barrier and to the west by the 15–70 m high Embomveni sand dune ridge (Fig. 2). Over the last 44 ka, the mire accumulated c. 11 m of peat, deposited on top of a basal clay 165 layer (Grundling et al., 2015). This clay layer was crucial in the formation and development of the 166 167 mire, limiting water loss during low sea level stands (Grundling et al., 2013). Mfabeni is bound to the north and south by beach ridges isolating it from Lake Bhangazi and Lake St. Lucia, respectively (Fig. 168 169 2; Grundling et al., 2013). When lake levels in Lake Bhangazi are high, minor water exchange 170 between Mfabeni and Bhangazi occurs, but there are no fluvial inputs to either system. Surface 171 drainage occurs southwards towards Lake St Lucia (Fig. 2). The peatland receives groundwater via the 172 swamp forest and the western dunes. This groundwater, which is important in keeping the mire wet during the dry season, discharges towards the center of the peatland and then flows within a sub-173 surface layer towards the east (Grundling et al., 2015). In the northern and eastern part of the 174

peatland, the vegetation is sedge and reed fen (comprising of sedges and grasses). In the westernand southern parts of Mfabeni is swamp forest (Venter, 2003).

177 The modern water balance at Mfabeni is dominated by the interplay between 178 evapotranspiration (ET; 1035 mm) and precipitation (1053 mm). Groundwater inflow (14 mm) and 179 stream outflow (9 mm) have a minor contribution to the modern water balance (all measured between May 2008 and April 2009; Grundling et al., 2015). Changes in regional climate have much 180 potential to influence the fine balance between ET and precipitation. For example, ET is suppressed 181 182 when cloud cover is increased during the summer months and increased during times of higher wind 183 speed (Grundling et al., 2015). ET is higher in the swamp forest than in the sedge and reed fen, 184 therefore a change in vegetation composition also has the potential to impact ET rates. The 185 depositional setting of the Mfabeni peatland provides a unique opportunity to reconstruct past 186 eastern South African climate variability at centennial-scale resolution from the Late Pleistocene to 187 the present day.

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## 189 **3.** Methodological background

To reconstruct past vegetation and hydroclimate we use the distribution, and the carbon and hydrogen isotopic composition, of long chain *n*-alkanes derived from plant-waxes.

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#### 193 3.1 Distributions of plant-waxes

To obtain information on water table variations, we quantify the relative contribution of plant-waxes derived from submerged and floating macrophytes relative to that of emergent and terrestrial plants ( $P_{aq}$ ). Odd-numbered *n*-alkanes ( $C_{25}$ - $C_{35}$ ) are derived from the epicuticular wax coating of terrestrial higher plants (Eglinton and Hamilton, 1967). Conversely, aquatic plant-waxes (of submerged macrophyte origin) are dominated by mid-chain *n*-alkanes (typically  $C_{23}$  and  $C_{25}$ ; e.g., Baker et al., 2016; Ficken et al., 2002). Thus we quantify  $P_{aq}$  using Equation 1 (Ficken et al., 2000).

$$P_{aq} = (C_{23} + C_{25}) / (C_{23} + C_{25} + C_{29} + C_{31})$$

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$$aq = (0.23 + 0.25)/(0.23 + 0.25 + 0.29 + 0.3)$$

Eq. 1

201 with  $C_x$  the amount of each homologue.

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To assess *n*-alkane degradation we used the carbon preference index (CPI; Bray and Evans, 1961). The CPI reflects the molecular distribution of odd-to-even *n*-alkanes, within a certain carbon number range (here, n-C<sub>26</sub> to n-C<sub>34</sub>; Equation 2). High CPI values indicate a higher contribution of oddnumbered *n*-alkanes (relative to even), indicating the *n*-alkanes are derived from higher terrestrial plants. Low CPI values indicate either low contribution from terrestrial higher plants or high organic matter degradation (Eglinton and Hamilton, 1967).

 $CPI_{27-33} = 0.5 * (\Sigma C_{odd27-33} / \Sigma C_{even26-32} + \Sigma C_{odd27-33} / \Sigma C_{even28-34})$ 210 211 Eq. 2 212 with  $C_x$  the amount of each homologue. 213 214 3.2 Carbon and hydrogen isotopes of terrestrial plant-waxes To reconstruct vegetation changes, we use the carbon isotopic composition of terrestrial plant-215 waxes ( $\delta^{13}C_{wax}$ ). On late Quaternary timescales, the primary factor determining the amplitude of 216 fractionation between the  $\delta^{13}C$  of atmospheric CO\_2 ( $\delta^{13}C_{atm}$ ) and the carbon isotopic composition of 217 the plant ( $\delta^{13}C_{plant}$ ) is the plant carbon fixation pathway ( $C_3/C_4/CAM$ ; e.g., Diefendorf and Freimuth, 218 2017). On these timescales, changes in the  $\delta^{13}C_{atm}$  are too small to significantly influence  $\delta^{13}C_{wax}$ 219 (Tipple et al., 2010). Shrubs and trees use the C<sub>3</sub> photosynthetic pathway and show the largest 220 221 fractionation. Grasses utilize either the  $C_3$  or the  $C_4$  pathway, with  $C_4$  plants having the smallest net 222 fractionation (Collister et al., 1994). The differences in carbon isotope fractionation during carbon uptake leads to different  $\delta^{13}C_{wax}$  signatures, and allows the determination of past vegetation types: *n*-223 alkane  $\delta^{13}$ C values of C<sub>3</sub> plants are c. -36‰ VPDB (Vienna Pee Dee Belemnite) and c. -20‰ VPDB for 224 225 C<sub>4</sub> plants (e.g., Diefendorf and Freimuth, 2017).

The hydrogen isotope composition of plant-waxes ( $\delta D_{wax}$ ) reflects the isotopic composition of 226 227 the water used during lipid biosynthesis (Sachse et al., 2012), rendering it a valuable tool for 228 reconstructing past hydrological conditions (e.g., Collins et al., 2013; Schefuß et al., 2005).  $\delta D_{wax}$  is 229 influenced by three main factors: i) the isotopic composition of precipitation; ii) enrichment of soil 230 and leaf water due to ET; and iii) differences in the apparent isotopic fractionation between source water and plant-waxes due to differences in vegetation type. The importance of each factor varies by 231 study site and with time. The detailed interpretation of the Mfabeni  $\delta^{13}C_{wax}$  and  $\delta D_{wax}$  is discussed in 232 233 section 6.1.

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## 4. Methods: compound specific C and H isotope analyses

236 Core MF4-12 (6.96 m recovery, 8.77 m penetration) was recovered from the centre of 237 Mfabeni peatland during January 2012 using a vibrocoring device (Fig. 2). The chronology of the core 238 is established by 24 <sup>14</sup>C AMS (accelerator mass spectrometry) dates from bulk peat (Fig. 3, S1). The 239 chronology is extended from that published in Humphries et al. (2017) and the age model is made 240 using Bacon 2.2 program (Blaauw and Christen, 2011). Radiocarbon ages were calibrated using the 241 southern hemisphere calibration curve, ShCal13 (Hogg et al., 2016) and the post-bomb southern 242 hemisphere curve, zone 1–2, for the uppermost modern dates (Hua et al., 2016).

Freeze-dried, bulk peat samples were ground and homogenized using a pestle and mortar 243 and lipids were extracted from c. 2 g of peat using a DIONEX Accelerated Solvent Extractor (ASE 200) 244 245 at 100 °C and at 1000 psi for 5 minutes (repeated 3 times) using a dichloromethane (DCM):methanol 246 (MeOH) (9:1, v/v) mixture. Prior to extraction, squalane was added as an internal standard. Copper turnings were used to remove elemental sulfur from the total lipid extract (TLE). To remove water, 247 the TLE was passed over a Na<sub>2</sub>SO<sub>4</sub> column (eluting with hexane). Subsequent to saponification (by 248 adding 6 % KOH in MeOH) and extraction (with hexane), the neutral fractions were split into a further 249 250 three fractions: hydrocarbon, ketone, and polar, by silica gel column chromatography (mesh size 60 251 µm) and elution with hexane, DCM and DCM:MeOH (1:1), respectively. By eluting the hydrocarbon 252 fractions with hexane over AgNO3-impregnated silica columns we obtained the saturated hydrocarbon fractions. The saturated hydrocarbon fractions were measured using a Thermo Fischer 253 254 Scientific Focus gas-chromatograph (GC) with flame-ionization-detection (FID) equipped with a 255 Restek Rxi 5ms column (30 m x 0.25 mm x 0.25  $\mu$ m), in order to determine the concentrations of long-chain *n*-alkanes. The GC oven temperature was set at 60 °C, held for 2 minutes, increased at 20 256 257 °C/minute to 150 °C and then at 4 °C/minute to 320 °C and held for 11 minutes. The split/splitless inlet temperature was 260 °C. To estimate the sample concentrations needed for isotope analyses, 258 259 samples were compared with an external standard that was run every 5 samples, which contained nalkanes ( $C_{19}-C_{34}$ ) at a concentration of 10 ng/µl. A quantification uncertainty of <5% was yielded 260 through replicate analyses of the external standard. 261

The  $\delta^{13}$ C values of the long-chain *n*-alkanes were measured using a Thermo Trace GC Ultra 262 equipped with an Agilent DB-5 column (30m x 0.25mm x 0.25µm) coupled to a Finnigan MAT 252 263 isotope ratio mass spectrometer (IR-MS) via a combustion interface operated at 1000 °C. The GC 264 265 temperature was programmed from 120 °C (hold time 3 min), followed by heating at 5 °C/minute to 320 °C (hold time 15 minutes). The external CO<sub>2</sub> reference gas was used to calibrate the  $\delta^{13}$ C values 266 and they are reported in % VPDB. Samples were analysed in duplicate when *n*-alkane concentrations 267 were adequate for multiple runs. The internal standard (squalane,  $\delta^{13}C=$  -19.9‰), yielded an 268 accuracy of 0.6‰ and a precision of 0.2‰ (n=37). The external standard mixture was analysed every 269 6 runs. The long-term precision and accuracy of the external *n*-alkane standard was 0.2 and 0.15‰, 270 respectively. For  $\delta^{13}$ C the average precision of the *n*-C<sub>29</sub> and *n*-C<sub>31</sub> alkane in replicates was 0.2‰ and 271 0.1‰ (n=22), respectively. 272

The  $\delta D$  compositions of long-chain *n*-alkanes were measured using a Thermo Trace GC coupled via a pyrolysis reactor (operated at 1420 °C) to a Thermo Fisher MAT 253 IR-MS. The GC column and temperature program was similar to that used for the  $\delta^{13}C$  analysis. The external H<sub>2</sub> reference gas was used to calibrate the  $\delta D$  values and they are reported in ‰ VSMOW. The H<sup>3+</sup> factor was monitored daily and fluctuated around 5.2 ppm nA<sup>-1</sup> during analyses. After every sixth measurement, an *n*-alkane standard of 16 externally calibrated alkanes was measured. The long-term precision and accuracy of the external *n*-alkane standard was 2.7 and 2‰, respectively. Samples were analysed in duplicate when *n*-alkane concentrations were adequate for multiple runs. The internal standard (squalane,  $\delta D$ = -180‰; ±2), yielded an accuracy of 0.9‰ and a precision of 1.9‰ (n=36). For  $\delta D$  the average precision in replicates was 1‰ for both *n*-C<sub>29</sub> and *n*-C<sub>31</sub> alkanes (n=52).

The last glacial period Mfabani  $\delta D_{wax}$  values were corrected to account for the effect of changes in global ice volume (Collins et al., 2013; Schefuß et al., 2005). For this, the benthic foraminifera-based oxygen isotope curve (Waelbroeck et al., 2002) was interpolated to each sample age and then converted to  $\delta D$  values using the global meteoric water line (Craig, 1961).

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## 288 **5. Results**

This study focusses on the last 32 cal ka BP (c. 590 cm). The average temporal resolution 289 between the 62 samples analysed for  $\delta^{13}$ C and  $\delta$ D is c. 500 years. From 590 to 70 cm (c. 32–2 cal kyr 290 291 BP) the core is very dark brown in colour containing peat with humus, fine detritus and silt. From 70 cm to core top, the sediments are similar in colour to the peat below and contain fibrous peat with 292 293 humus and herbaceous fine detritus (Humphries et al., 2017). Between 457 and 358 cm (c. 23-14 cal 294 kyr BP; comprising the LGM) mean grain sizes average at 110 µm, with smaller diameters averaging 295 at 50 µm between 298 and core top (c. 11 cal kyr BP–present, Holocene; Fig. 4g). The lithology of 296 core MF4-12 does not exactly match with that observed from core SL6 (Baker et al., 2014; 2016; 2017), although sandy peat is observed during the LGM at both locations. This result is not surprising 297 as multiple cores taken in transects across the bog indicate peat heterogeneity (Grundling et al., 298 299 2013).

Long chain *n*-alkane CPI values are generally around 6 (ranging from 2–13), indicating good *n*-300 301 alkane preservation. The two samples with CPI values of 2, potentially containing more degraded n-302 alkanes, are highlighted in red (Fig. 4b & c; Fig. 5b & c; Fig. 6f & g). However, the in- or exclusion of 303 these samples does not affect the observed pattern of changes and we thus consider the record to 304 be suitable for palaeoclimate reconstruction. The samples contain *n*-alkanes with carbon chain lengths ranging from  $C_{17}\!\!-\!\!C_{35}$ , with  $C_{29}$  and  $C_{31}$  generally having the highest abundance. The high 305 abundances of  $C_{29}$  and  $C_{31}$  enabled reliable isotopic analyses. The relationship between the  $\delta D$  and 306  $\delta^{13}$ C of the C<sub>29</sub> and C<sub>31</sub> *n*-alkanes is strong, with R<sup>2</sup> values of 0.8 and 0.9, respectively. Consequently, 307 for the  $\delta^{13}C_{wax}$  and  $\delta D_{wax}$ , we use the amount-weighted mean of the  $C_{29}$  and  $C_{31}$  *n*-alkanes. 308

The  $\delta^{13}C_{wax}$  values range from -29‰ to -21‰ (Fig. 4b). The ice volume  $\delta D$  correction decreases the glacial Mfabeni  $\delta D_{wax}$  values by <8 ‰ (Fig. 4c). The ice-corrected  $\delta D_{wax}$  values of the *n*- $C_{29}$  and *n*- $C_{31}$  alkanes range from -181‰ to -128‰ (Fig. 4c). P<sub>aq</sub> values range from 0.02–0.7, averaging at 0.2 (Fig. 4f).

During the LGM,  $\delta^{13}C_{wax}$  and  $\delta D_{wax}$  values are relatively high averaging at -23‰ and *c*. -136‰, 313 respectively (Fig. 4b & c) and P<sub>aq</sub> values are low (c. 0.24; Fig. 4f). At c. 19 cal ka BP a 4‰ negative shift 314 in  $\delta^{13}C_{wax}$  values occurs (Fig. 4b). This negative shift in  $\delta^{13}C_{wax}$  is concurrent with a gradual shift to 315 lower  $\delta D_{wax}$  values (Fig. 4c) and an increase in  $P_{aq}$  values (Fig. 4f). Between 14 and 5 cal kyr BP,  $\delta^{13}C_{wax}$ 316 values are relatively stable and average at -28‰ (Fig. 4b).  $\delta D_{wax}$  values become gradually lower 317 during this period reaching -173‰ at 7.5 cal ka BP. At 5 cal ka BP,  $\delta D_{wax}$  values shift towards more 318 positive values by 16‰ (Fig. 4c). Relatively high  $P_{aq}$  values occur between 14–5 cal kyr BP (Fig. 4f). 319 After c. 5 cal ka BP several high amplitude millennial-scale fluctuations in both  $\delta^{13}C_{wax}$  and  $\delta D_{wax}$ 320 values are evident. These fluctuations interrupt a trend where the isotope values of both  $\delta^{13}C_{wax}$  and 321 322  $\delta D_{wax}$  gradually increase towards present day. A pronounced shift to higher  $\delta^{13}C_{wax}$  and  $\delta D_{wax}$  values occurs at 2.8 cal ka BP. From c. 900 cal yr BP,  $\delta^{13}C_{wax}$  and  $\delta D_{wax}$  values become higher reaching core 323 324 top values of -21 and -128‰, respectively (Fig. 4b & c). Generally high, but variable and rapidly fluctuating P<sub>aq</sub> values are evident between c. 5–0 cal kyr BP. P<sub>aq</sub> values decrease substantially after 325 326 1.3 cal ka BP from 0.6 to a core top value of c. 0 (Fig. 4f).

327

#### 328 **6.** Discussion

## 329 6.1 Interpretation of the proxy signals

The relatively high CPI<sub>27-33</sub> values indicate that the long-chain *n*-alkanes within the peat are 330 331 derived from terrestrial higher plants and are relatively non-degraded. The long-chain *n*-alkanes are 332 likely sourced directly from the local vegetation surrounding the coring location. It is possible that 333 during times of stronger wind strength (i.e., during the LGM; Humphries et al., 2017) increased aeolian transport resulted in a higher biomarker contribution from more distal sources (i.e., the 334 335 surrounding dune vegetation). Good preservation of *n*-alkanes in Mfabeni peat was also observed in nearby core SL6, but this was based on a CPI calculated using n-C<sub>21-31</sub> (Baker et al., 2016). No 336 relationship exists between the CPI and  $P_{aq}$  (R<sup>2</sup> = 0.11), which suggests that CPI variations at the 337 location of core MF4-12 are not related to changes in organic matter preservation due to water table 338 339 level variations.

The main source of carbon for terrestrial higher plants (the source of the  $C_{29}$  and  $C_{31}$  *n*-alkanes) is 340 atmospheric CO<sub>2</sub>, whereas aquatics also assimilate dissolved carbon, complicating the interpretation 341 of their carbon isotope signal. We thus focus solely on  $C_{29}$  and  $C_{31}$  *n*-alkanes that are predominantly 342 343 derived from terrestrial plants (Eglinton and Hamilton, 1967). The majority of the samples (67 %) 344 have dominant *n*-alkane chain lengths of C<sub>29</sub> and C<sub>31</sub>. For the remaining 33 % of the samples, concentrated between 6 and 1.1 cal kyr BP, the dominant chain length switched to n-C<sub>25</sub>, indicating a 345 higher *n*-alkane input from submerged macrophytes (Ficken et al., 2000). The *n*-C<sub>25</sub> are unlikely to be 346 sourced from mosses, as mosses are rare in subtropical peatland environments (Baker et al., 2016). 347

Instead, the C<sub>25</sub> is likely mainly derived from aquatic plants, which produce mid-chain *n*-alkanes as dominant homologues (C<sub>20</sub>-C<sub>25</sub>; Ficken et al., 2000). This increase of *n*-alkanes sourced from aquatic plants *c*. 6–1.1 cal kyr BP is unlikely to have had any impact on the isotopic composition of the longchain *n*-alkanes (C<sub>29</sub> and C<sub>31</sub>) as these are minor components in aquatic plants (e.g., Aichner et al., 2010). Therefore, we interpret the  $\delta^{13}C_{wax}$  as changes in the C<sub>3</sub>/C<sub>4</sub> ratio of terrestrial higher plants.

Grasses exhibiting the C<sub>4</sub> or C<sub>3</sub> photosynthetic pathway in South Africa are largely 353 geographically separated, with C<sub>4</sub> grasses dominant within the SRZ and C<sub>3</sub> grasses more prevalent in 354 355 the YRZ, WRZ and at higher altitudes (Vogel et al., 1978). As  $C_4$  grasses require less water to fix  $CO_2$ , thus having greater water-use efficiency than  $C_3$  grasses,  $C_4$  photosynthesis is favored in arid regions 356 357 (e.g. Downes, 1969; Osborne and Sack, 2012). C<sub>4</sub> grasses also have the potential to achieve higher rates of photosynthesis than  $C_3$  particularly at high irradiance and temperature levels (Black et al., 358 359 1969; Monteith, 1978), as their more efficient carbon fixation has a higher energy demand (Sage, 2004). Today growing season temperatures are a controlling factor for the distribution of  $C_4$  and  $C_3$ 360 grasses (with  $C_4$  grasses having an advantage over  $C_3$  grasses at higher temperatures; Sage et al., 361 362 1999). Consequently C<sub>4</sub> grasses are mainly found in warm and dry environments such as the African savannas (Beerling and Osborne, 2006). Furthermore, under reduced atmospheric (i.e., glacial) CO<sub>2</sub>, 363 the higher carbon fixation efficiency of  $C_4$  grasses provides an important advantage over  $C_3$  grasses 364 (Sage, 2004; Pinto et al., 2014). Previous palynological studies indicate that the dominant 365 366 components of the pollen assemblage at Mfabeni are Poaceae and Cyperaceae (Finch and Hill, 2008). 367 Although Cyperaceae species can be either  $C_3$  or  $C_4$ , most Cyperaceae in eastern South Africa (67 %) are of the  $C_4$ -type (Stock et al., 2004). The  $C_4$  vegetation at Mfabeni is thus mostly Poaceae or 368 369 Cyperaceae from the sedge and reed fen. The  $C_3$  vegetation at Mfabeni is comprised of arboreal taxa 370 from the swamp forest (e.g., Myrtaceae and Ficus) and locally distributed Podocarpus (Finch and Hill, 2008; Venter, 2003). Shifts to higher  $\delta^{13}C_{wax}$  values (more C<sub>4</sub>-type vegetation) at Mfabeni could 371 372 indicate an expansion of grassland (at the expense of arboreal taxa), or a shift from C<sub>3</sub> to C<sub>4</sub> grasses, resulting from: i) less precipitation, ii) a longer/more intense dry season, iii) heightened ET, iv) 373 reduced water table height, v) higher temperatures, vi) reduced atmospheric CO<sub>2</sub>, or vii) increased 374 375 insolation levels (or any combination of the above).

The  $\delta D_{wax}$  reflects the  $\delta D_{precip}$ , ET and vegetation type. The  $\delta D_{precip}$  can be influenced by changes in air temperature, with an estimated temperature effect of *c*. 0.5‰ per 1°C for  $\delta^{18}O_{precip}$ (Dansgaard, 1964). The maximum estimated temperature change of *c*. 6 °C in the SRZ of South Africa from the LGM to Holocene (Gasse et al., 2008), would thus correspond to a change in  $\delta^{18}O_{precip}$  of 3‰. Conversion to changes in  $\delta D_{precip}$  using the global meteoric water line would thus lead to a potential LGM to Holocene  $\delta D_{precip}$  enrichment of 24‰ (Craig, 1961). However, the Mfabeni  $\delta D_{wax}$ record shows a depletion in  $\delta D_{wax}$  from the LGM to the Holocene, rather than an enrichment. The observed glacial  $\delta D$  depletion is therefore a conservative estimate. Consequently, changes in air temperature from the LGM to the Holocene did not exert a dominant control on Mfabeni  $\delta D_{wax}$ .

Changes in vegetation type ( $C_3/C_4$ ) have the potential to reduce or exaggerate shifts in  $\delta D_{wax}$ . 385 386 There are differences in the apparent fractionation (the integrated isotopic fractionation between precipitation and plant-wax lipids) between plant types using different photosynthetic pathways. C<sub>3</sub>-387 388 type shrubs and trees fractionate the least,  $C_4$ -type grasses slightly more, while  $C_3$ -type grasses show the highest apparent fractionation (Sachse et al., 2012). The difference in  $\delta D_{wax}$  between dicots (C<sub>3</sub>, 389 shrubs, trees and forbs) and monocots (C<sub>4</sub>, grasses) is likely the result of leaf architecture and the 390 391 nature of water movement in the leaf. Monocots display progressive evaporative enrichment along 392 parallel veins along the leaf, which does not occur in dicots. This grass-blade enrichment results in 393 higher  $\delta D_{wax}$  values in C<sub>4</sub> grasses (Helliker and Ehleringer, 2000). However, recent data suggest that the effect of C<sub>3</sub>-tree to C<sub>4</sub>-grass vegetation type changes on  $\delta D_{wax}$  is likely relatively small (Collins et 394 395 al., 2013; Vogts et al., 2016).

The  $\delta D_{\text{precip}}$  is strongly controlled by the 'amount effect', where there is a negative correlation 396 between monthly precipitation amount and  $\delta D_{\text{precip}}$  (Dansgaard, 1964). Close to the equator, passage 397 398 of the tropical rainbelt can result in precipitation that is extremely depleted in D. Conversely, in arid 399 regions, rainfall tends to be enriched in D, because of enhanced evaporation of the raindrops as they fall (Risi et al., 2008). Studies investigating the present-day relationship between precipitation 400 amount and the isotopic variations in rainfall indicate shifts in  $\delta^{18}$ O of up to 15‰ (c. 120‰ in  $\delta$ D) 401 with the passage of the tropical rainbelt and shifts in  $\delta^{18}$ O of 7‰ (c. 56‰ in  $\delta$ D) with the passage of 402 convective storms (Gat et al., 2001). During times of heightened ET and/or lower precipitation 403 404 amount, soil waters become enriched in D (Sprenger et al., 2017). In addition, under conditions of low ambient relative humidity, leaf water becomes enriched in D through increased transpiration 405 (Kahmen et al., 2013). Large values of isotopic enrichment (c. 40‰ in  $\delta^{18}$ O, 180‰ in  $\delta$ D) are 406 associated with the effects of evaporation (e.g., Cappa et al., 2003). The control of precipitation 407 408 amount and ET on D operates in the same direction, and thus the mechanisms are not easily 409 disentangled.

410 Mfabeni has high rates of ET, which can equal, or even exceed precipitation during dry 411 periods (Grundling et al., 2015). Consequently, both precipitation amount and ET are likely to control 412 the isotopic composition of soil and leaf waters, and subsequently of the leaf waxes at Mfabeni. High 413  $\delta D_{wax}$  values at Mfabeni likely result from decreased summer precipitation amount and/or 414 heightened ET. The similarity between the  $\delta D_{wax}$  pattern and the regional precipitation/aridity stacks 415 (Fig. 4d & e; Chevalier and Chase, 2015; 2016) supports the inference that precipitation amount and 416 ET drive Mfabeni  $\delta D_{wax}$ . Furthermore, this similarity indicates that the hydrological fluctuations in the 417 Mfabeni record represent hydrological change at a broader spatial scale (Fig. 4c–e), but also suggest

that the pollen-based precipitation stacks may also include an element of ET variability.

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# 6.2 Climatic and environmental conditions at Mfabeni over the last 32 cal ka BP

The  $\delta^{13}C_{wax}$ ,  $\delta D_{wax}$  and  $P_{ag}$  data from Mfabeni indicate that the vegetation, hydrology and the 421 water table varied considerably over the last 32 cal ka BP (Fig. 4 &. 5). The high  $\delta^{13}C_{wax}$  values during 422 the LGM indicate that the vegetation was likely dominated by more drought-tolerant C4 plant types 423 (Fig. 4b). Similar LGM  $\delta^{13}C_{wax}$  depletion was observed previously at Mfabeni (Fig. 4h; Baker et al., 424 425 2017). Drier conditions during the LGM correspond with low P<sub>ag</sub> values that indicate a higher relative 426 contribution of terrestrial-over-aquatic *n*-alkanes, likely a consequence of a lower water table (Fig. 427 4f). The high  $\delta D_{wax}$  values during the LGM suggest decreased precipitation amount and/or higher ET, 428 which are both consistent with a drier environment (Fig. 4c). We cannot completely rule out the 429 possible impact of increased drainage of the peatbog during the LGM due to low eustatic sea level 430 (Grundling et al., 2013). A lower water table during the LGM would likely serve to further soil water D 431 enrichment. Nevertheless, the fact that the peat continued to grow during the LGM suggests that the sea level effect was minor. The organic geochemical proxies agree with palynological data indicating 432 regional grassland dominance (high Poaceae, Cyperaceae and Asteraceae) with low amounts of 433 434 arboreal taxa (Fig. 6; Finch and Hill, 2008). Regional aridity and increased wind strength during the 435 LGM at Mfabeni are also indicated by increased mean grain size of the lithogenic sediment fraction 436 (Fig. 4g), and the modal grain size of the distal aeolian component (Humphries et al., 2017). Evidence 437 for reduced precipitation (from the regional precipitation stack; Fig. 4d) and high aridity (from the 438 regional aridity stack; Fig. 4e) during the LGM, provide evidence that the dry conditions at Mfabeni 439 appear to be part of a wider eastern South African pattern (Fig. 4d & e).

The shift to more negative  $\delta^{13}C_{wax}$  values following the LGM, at *c*. 19 cal ka BP, indicating that 440 the vegetation at Mfabeni changed to more C<sub>3</sub>-type (Fig. 4b), is also evident in Mfabeni core SL6 (Fig. 441 4h; Baker et al., 2017). This change is thus likely representative of a  $C_4$ – $C_3$  change across the peat bog. 442 The palynological record indicates no shift towards arboreal taxa at this time but instead a 443 444 continuation of grasslands (Fig. 6a & b; Finch and Hill, 2008). A decrease in Cyperaceae percentages (as most Cyperaceae is of C<sub>4</sub> type), may be responsible for the C<sub>4</sub> to C<sub>3</sub> shift observed in the  $\delta^{13}C_{wax}$ 445 record, but the gradual nature of the Cyperaceae decrease points to an additional driver (Fig. 6c). The 446 447 shift is more likely the result of a switch from C4 to C3 grasses. If temperature was driving the vegetation shifts at Mfabeni, we would expect a shift from  $C_3$  to  $C_4$  grasses from the LGM to the 448 Holocene (with a c. 6°C increase in temperature). Nevertheless, the LGM to Holocene shift from C<sub>4</sub> to 449  $C_3$  grasses suggests that temperature did not drive the vegetation change at Mfabeni. We suggest 450 that the shift from  $C_4$  to  $C_3$  grasses may have been caused instead by i) more precipitation, ii) a 451

452 shorter/less intense dry season, iii) lower ET, and/or iv) increased water table height. Furthermore, 453 with  $C_3$  vegetation favored under lower insolation conditions, a decrease in local summer insolation 454 from the LGM to Holocene (Fig. 4a) could have played a role in driving the vegetation shifts.

After c. 19 cal ka BP, the  $\delta^{13}C_{wax}$  values continue to decrease to -29‰ until they stabilize at c. 455 14 cal ka BP. This trend in  $\delta^{13}C_{wax}$  values between c. 19 and 14 cal kyr BP, indicating an expansion of 456  $C_3$  vegetation, corresponds well with the  $\delta^{13}C_{wax}$  record from Mfabeni core SL6 (Baker et al., 2017; 457 Fig. 4b & 4h). There are, however, some minor differences between the two  $\delta^{13}C_{wax}$  records. We 458 attribute these to small-scale variations in vegetation across the peatbog, the lower sampling 459 resolution of core SL6 and to dating uncertainties in both records. The shift to lower  $\delta^{13}C_{wax}$  values at 460 461 c. 19 cal ka BP occurs at the same time as a rise in the water table as documented by an increase in P<sub>ag</sub> values (Fig. 4f). An abrupt increase in precipitation amount and a decrease in aridity is evident in 462 463 the precipitation and aridity stacks at c. 19 cal ka BP. All proxy records for precipitation (the regional stacks and the Mfabeni  $\delta D_{wax}$  data; Fig. 4) strongly suggest a switch to wetter conditions after c. 19 464 465 cal ka BP.

466 The  $\delta^{13}C_{wax}$  values between 14–5 cal kyr BP reflect a stable period of C<sub>3</sub>-type vegetation (Fig. 4b). At the same time, gradually decreasing  $\delta D_{wax}$  values indicate increasing humidity. The gradual 467 increase in precipitation is also evident in the precipitation stack, but this trend is interrupted by an 468 abrupt return to aridity at c. 14.2 cal ka BP, coinciding with the Antarctic Cold Reversal (Chase et al., 469 470 2017). This abrupt arid event is only evident in one sample at Mfabeni and thus higher resolution 471 sampling is needed across this interval. The aridity stack indicates low aridity during this interval, but 472 high variability suggests a complex interplay between high ET (from increased temperatures, 473 resulting in less effective precipitation) and generally more precipitation (Fig. 4e). Pollen data from 474 Mfabeni provide evidence for an expansion of arboreal type vegetation at c. 12 cal ka BP (Fig. 6a; Finch and Hill, 2008). The pollen data thus suggest the establishment of swamp forest vegetation 475 476 during the early Holocene, indicative of a moist climate (Fig. 6a). Mfabeni aeolian sediment flux is low and stable throughout this period, also suggesting a moist climate (Humphries et al., 2017). The 477 moist climate likely resulted in vegetated dunes, reducing the amount of material available for 478 aeolian transport. The relatively high P<sub>ag</sub> values between 14–5 cal kyr BP indicate a high and stable 479 480 water table at this time (Fig. 4f). Elevated total organic carbon percentages within Mfabeni core SL6 during the Holocene, also suggest increased water levels (Baker et al., 2017). 481

Between *c*. 5–0 cal kyr BP several high-amplitude millennial-scale  $C_3/C_4$  vegetation changes are evident superimposed on an overall shift from predominantly  $C_3$  to more  $C_4$ -type vegetation towards the present-day (Fig. 5b). This variability contrasts with the more gradual  $C_4/C_3$  vegetation transition from the glacial period to Holocene. The  $\delta^{13}C_{wax}$  values from Mfabeni core SL6 between *c*. 6–1 cal kyr BP also indicate a period of predominantly  $C_4$ -type vegetation, implying arid conditions 487 during this time (Baker et al., 2017; Fig. 4h). A similar pattern of a long-term trend with superimposed short-term variability is visible in the in  $\delta D_{wax}$  record. The general enrichment in D 488 489 reflects gradual drying, punctuated by millennial-scale pulses of aridity, with the most pronounced 490 arid event at c. 2.8 cal ka BP (Fig. 5c). Counterintuitively, the high abundance of  $n-C_{25}$  alkanes and high but variable P<sub>aq</sub> values between c. 5–0 cal kyr BP indicate a generally high water table, 491 interrupted by brief periods of a lower water table (Fig. 5d). After 2.3 cal ka BP, both  $\delta^{13}C_{wax}$  and 492  $\delta D_{wax}$  values become higher and  $P_{aq}$  values lower (Fig. 5b–d). This suggests increased C<sub>4</sub>-type 493 494 vegetation cover, decreased summer precipitation amount and/or higher ET and low water table 495 levels. A slight increase in precipitation followed by gradually decreasing precipitation over the last c. 496 5 ka is evidenced in the precipitation stack (Fig. 4d). This initial increase in precipitation at c. 5 cal ka 497 BP corresponds to an abrupt decrease in aridity (Fig. 4d & e). The increased variability observed in 498 our records between 5–0 cal kyr BP could be an artefact of the high temporal resolution of our record during this interval (~220 vs ~700 years per sample for the remainder of the record). 499 500 Nevertheless, (Fig. 4d) other data from the region (e.g., Baker et al., 2017, Humphries et al., 2017; 501 2016, Finch and Hill, 2008, Neumann et al., 2010) also indicate climatic instability and arid climatic conditions during the last c. 5 cal ka BP, suggesting that the observed variability is likely real (Fig. 5e). 502

503 It is interesting that modern  $\delta D_{wax}$  values and those during the LGM appear similar (Fig. 4c), 504 implying similarly arid conditions during both periods. The southern aridity stack also indicates 505 extremely arid conditions during the last few thousand years and the authors stress the importance 506 of temperature in controlling aridity (Fig. 4e; Chevalier and Chase, 2016). It is possible that modern 507 high mean annual temperatures drove these modern-day  $\delta D_{wax}$  values to appear similar to those 508 from the LGM. High modern day temperatures, increase ET and result in less 'effective precipitation' 509 and arid conditions, even when rainfall is high (Chevalier and Chase, 2016). During the LGM, lower 510 temperatures would have reduced ET, leading to apparent humid conditions, despite reduced rainfall 511 amount.

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## 513 6.3 Climate driving mechanisms

514 Modern observations suggest that high SSTs within the Mozambique Channel and Agulhas Current induce increased evaporation (e.g., Walker, 1990), resulting in higher rainfall in the SRZ 515 (Tyson, 1999). Variations in local SSTs are thus thought to be an important driver of hydroclimate in 516 517 eastern South Africa. This mechanism may also play a role on longer timescales. Indeed, Chevalier and Chase (2015) invoke SSTs as the dominant driver of precipitation variability during the LGM. 518 Mfabeni vegetation and hydrology reconstructions over the last 32 cal ka BP do not show a clear 519 relationship with changes in southwest Indian Ocean SSTs (Fig. 4j, Sonzogni et al., 1998). For example 520 521 if SSTs drove the climate at Mfabeni then the abrupt shift to more  $C_3$  type vegetation and the gradual

522 shift to a wetter climate at c. 19 cal ka BP would be expected to correspond with an increase in SSTs. 523 This is not the case, and SSTs do not increase until c. 15.7 ka (Sonzogni et al., 1998; Fig. 4). The lowest temperatures within the Mozambique Channel correspond to Heinrich Event 1 (SSTs c. 3°C colder 524 525 than present day), an event which is not evident as a particularly arid period in the Mfabeni dataset. 526 Mozambique Channel SSTs thus do not fully explain the variability observed in the records comprising the precipitation stack. These differences, as proposed previously by Chevalier and Chase 527 (2015), suggest that SST variability is unlikely to be the sole driver of the changes in hydroclimate 528 529 within this part of the SRZ. Chevalier and Chase (2015) proposed that the differences observed between SSTs and the records comprising the precipitation stack is due to the modulation of 530 531 precipitation by the position of the westerlies.

We attribute the arid climate and the associated expansion of drought tolerant  $C_4$  plants and 532 533 a low water table at Mfabeni during the LGM, in part, to a northward displacement of the westerlies, 534 the SIOCZ and the subtropical high-pressure cell, shifting the hydroclimate to a more evaporative regime, where ET exceeds precipitation. In addition, lower SSTs (Fig. 4j) in the Mozambique Channel 535 536 at this time likely reduced moisture availability. It is possible that the combination of a northward displacement of these three systems (the westerlies, SIOCZ and subtropical high-pressure cell) and 537 lower SSTs shifted the fine balance between precipitation and ET at Mfabeni towards higher ET rates 538 during the LGM. 539

540 Palaeoenvironmental studies (e.g., Lamy et al., 2001; Lamy et al., 2010; Stuut and Lamy, 541 2004), climate model simulations (e.g., Rojas et al., 2009; Toggweiler et al., 2006) and theoretical 542 models (e.g., Cockcroft et al., 1987) provide evidence for an equatorward migration and 543 strengthening of the southern hemisphere westerlies in response to the increased extent of Antarctic 544 sea ice during the LGM. Records from the present WRZ such as Elands Bay Cave (Baxter, 1996), 545 Pakhuis Pass (Scott, 1994) and Driehoek Vlei (Meadows and Sugden, 1993) indicate increased winter 546 rainfall, interpreted as a northward shift and strengthening of the westerlies during the LGM (Chase and Meadows, 2007). An equatorward migration of the westerlies may have expanded the limit of 547 the WRZ in South Africa northward, to around 25°S in the west and 30°S in the east (Cockcroft et al., 548 549 1987). This would have put Mfabeni (at 28°S) within the range of the southern westerlies. Although during the LGM the westerlies were in a more northerly position, and had the potential to provide 550 551 rainfall (via the passage of more cold fronts; Nkoana et al., 2015), we do not see any evidence for 552 increased precipitation at Mfabeni. Today mid-latitude cyclones (frontal systems; Fig. 1b) associated with the westerlies trigger rainout of atmospheric moisture, sourced from the Indian Ocean and 553 Agulhas Current, during the winter months (Gimeno et al., 2010). However, the co-occurring 554 555 subtropical high-pressure cell over the South African interior may have limited the amount of moisture advection towards Mfabeni, thus even with increased cyclone occurrence, arid conditions 556

557 persisted. Furthermore, with a northerly displaced subtropical high-pressure cell inhibiting 558 monsoonal penetration, the duration of the dry season at Mfabeni may have been extended, 559 shortening the rain season and heightened ET rates.

A northward migration and strengthening of the westerlies is also associated with a northerly displaced and weaker South Indian Anticyclone (Fig. 1; Cohen and Tyson, 1995). A weakening of the western portion of the South Indian Anticyclone results in a northeastward shift of the SIOCZ (and the rain-bearing cloud band associated with TTTs; Cook, 2000). This northeastward shift results in higher precipitation over coastal Africa (around 15°N) and Madagascar and lower than normal precipitation to the south, in eastern South Africa (Cook, 2000). We propose that a northeastward shift of the SIOCZ during the LGM may have also played a key role in driving aridity at Mfabeni.

The latitudinal position of the subtropical high-pressure cell is highly correlated to rainfall 567 568 variability along the eastern coast of South Africa (Dyson and van Heerden, 2002). Multivariate 569 analysis of zonal moisture fluxes in South Africa indicates that the latitudinal position of the 570 subtropical high-pressure cell directly controls the amount of moisture advection (monsoonal 571 penetration) towards the southern African interior during the summer months (Vigaud et al., 2009). When the cell is shifted southward, during the summer, the tropical easterlies are able to penetrate 572 further inland, resulting in higher continental moisture availability (Vigaud et al., 2009). Conversely, 573 574 when the cell is shifted northward, during the winter, monsoonal circulation south of 25°S is 575 impeded, creating a deficit in moisture advection from the ocean to the continent (Tyson and 576 Preston-Whyte, 2000; Vigaud et al., 2009). A more northerly location of the subtropical high-pressure 577 cell, during the LGM, would have lengthened the dry season, resulting in aridity at Mfabeni.

578 We suggest that the shift to more humid conditions at c. 19 cal ka BP was related to the 579 retreat of the westerlies, the subtropical high-pressure cell and the SIOCZ, as Antarctic sea ice began 580 to retreat poleward (Fig. 4k), allowing an increased influence of the moist tropical easterlies. With 581 the subtropical high-pressure cell further south, stronger easterly flux from the Indian Ocean likely enhanced the development of TTTs in the region leading to increased precipitation. This shift at c. 19 582 cal ka BP was unlikely driven by a change in local summer insolation because insolation was 583 584 decreasing at this time. We suggest that the abrupt shift to more  $C_3$  vegetation was a non-linear response to increasing moisture availability in the region (Fig. 4c). Precipitation amount may have 585 586 reached a critical threshold at c. 19 cal ka BP for the establishment of  $C_3$  type vegetation, resulting in 587 the observed abrupt vegetation shift (Fig. 4b).

588 Between 14–5 kyr BP, a reduced extent of Antarctic sea ice (Fig. 4k & 5g), resulted in a more 589 poleward position of the westerlies and the subtropical high-pressure cell. The diminished effect of 590 the westerlies and the subtropical high-pressure cell in eastern South Africa at this time permitted 591 the tropical systems (easterlies), to dominate the climatic regime at Mfabeni. With a strengthened

592 (but poleward displaced) South Indian Anticyclone the SIOCZ was likely situated over Mfabeni 593 resulting in increased rainfall. Strong easterly flux would have increased the development of TTTs in 594 the region, resulting in higher humidity at Mfabeni. Increasing humidity at Mfabeni during the 595 Holocene, corresponds with increasing southern hemisphere summer insolation (Fig. 4a). The 596 importance of insolation for South African climate variability during the late Quaternary has been 597 suggested before (e.g., Partridge et al., 1997; Simon et al., 2015). Our results support the hypothesis that insolation control on precipitation variability was only significant during the Holocene (e.g., 598 599 Schefuß et al., 2011; Chevalier and Chase, 2015). We suggest that direct local insolation forcing is only dominant in this region when the westerlies and subtropical high-pressure cell are located far 600 601 south, which allows monsoonal precipitation to penetrate into the continent during the summer months. 602

To explain the millennial-scale climatic variability over the glacial-interglacial transition within their central and eastern African sites (which also includes Mfabeni), Chevalier and Chase (2015) and Chase et al. (2017) suggest that this region may be influenced by the position and the intensity of the westerlies, and the interactions between the westerlies and the tropical easterlies (resulting in TTT development). We highlight the importance of the location of TTT development (i.e., the SIOCZ) and stress the interconnections between TTT development, the latitudinal position of the westerlies and the subtropical high-pressure cell on glacial-interglacial timescales.

610 After c. 5 cal ka BP, palaeoenvironmental records from both the WRZ and YRZ, such as from 611 Verlorenvlei (Fig. 1; Fig. 5f; Carr et al., 2015), Seweweekspoort (Fig. 1; Fig. 4i; Chase et al., 2017), Klaarfontein (Fig. 1; Meadows and Baxter, 2001), Cecilia Cave (Fig. 1; Baxter, 1989) and Eilandvlei 612 613 (Wündsch et al., 2018), document increased moisture availability, implying a recurring more 614 northerly location of the westerlies. Chevalier and Chase et al. (2015) propose that increased 615 precipitation in the WRZ during the late Holocene was due to both the warmer interglacial climate 616 and the northward expansion of the westerly storm tracks. Although no indication for an increase in sea ice is evident from EPICA salt concentration data (Fig. 4k), diatom data (Fragilariopsis curta and F. 617 cylindrus) from PS2090/ODP1094 in the southern South Atlantic document an increase in sea ice 618 619 during the late Holocene (Fig. 5g), which may have pushed the southern westerlies equatorward. In addition, climate modelling results imply a northward shift of the southern westerlies at this time 620 621 (Hudson and Hewitson, 2001). Consequently, in a comparable way to the LGM, the increased sea ice 622 during the late Holocene (Fig. 5g), may have displaced (and strengthened) the westerlies, the South African high-pressure system and the SIOCZ equatorward, resulting in higher aridity at Mfabeni. A 623 624 slight decrease in Mozambique Channel SSTs may have also played a role in the generally arid climate 625 at Mfabeni during the last c. 5 cal ka BP (Fig. 4j; Sonzogni et al., 1998). Interestingly, the hydrological variability at Mfabeni (Fig. 5c) during the last c. 5 cal ka BP, is not present in the central and eastern 626

South African precipitation stack (Fig. 4d). We attribute this to the highly sensitive balance between
ET and precipitation at Mfabeni (Grundling et al., 2015), and the fact that the precipitation stack
smooths local hydrological variability.

630 It is possible that anthropogenic influences also played a role in shaping the environment at Mfabeni, at least, during the late Holocene. However, unequivocal agricultural and exotic pollen 631 indicators are absent from the pollen record and although pollen data indicate that forest decline 632 occurred during the late Holocene, it is unclear whether this was related to human influence or 633 634 regional climate change (Fig. 6; Finch and Hill, 2008). The forest decline could have affected the water table and increased the relative amount of C<sub>4</sub>-type vegetation. The appearance of Morella and 635 Acacia in the late Holocene may indicate the development of open vegetation or secondary forest 636 due to fire disturbance (Fig. 6e; Finch and Hill, 2008). Human activities or climate change may be 637 638 responsible for changes in fire regime. With no palaeo-charcoal data available for Mfabeni yet, no 639 direct evidence for increased fire activity during the late Holocene exists. In addition, the palaeoenvironmental evidence available suggests that the arid conditions during the late Holocene 640 641 were regional in nature (Scott, 1999; 2003; Humphries et al., 2016, Neumann et al., 2010). Thus, any human activity was unlikely the primary cause of the late Holocene regional aridity and the large 642 643 magnitude of environmental changes observed at Mfabeni.

Today ENSO activity is one of the most important driving mechanisms for inter-annual 644 645 climatic variability in South Africa. Southern Africa's seasonal rainfall is linked to ENSO, with dry (wet) 646 conditions associated with El Niño (La Niña) events (Archer et al., 2017; Mason and Jury, 1997). 647 Interannual variability in the strength and position of the SIOCZ is linked to ENSO variability (Cook, 648 2000). During La Niña years, the SIOCZ is located over the continent, resulting in wet conditions in 649 eastern South Africa. During El Niño, the SIOCZ shifts northeastward over the Indian Ocean and as a 650 consequence, dry conditions prevail in eastern South Africa (Lindesay, 1988; Cook, 2001; Hart et al., 651 2018). Furthermore, during El Niño events, a northward shift of the westerlies may occur, which could increase rainfall over western South Africa but lead to aridity in the east (i.e., at Mfabeni; 652 Lindesay, 1988). Palaeoenvironmental studies in the Pacific Basin and South America indicate that 653 654 during the early Holocene El Niño events were smaller and occurred less frequently, with a shift to stronger ENSO activity after c. 5 cal ka BP (Fig. 5h, Moy et al., 2002; Huffman, 2010; Rodbell et al., 655 656 1999; Sandweiss et al., 1996). It is difficult to disentangle the possible potential drivers of climate 657 variability during the last c. 5 cal ka BP at Mfabeni. We therefore invoke a possible combination of northerly-displaced westerlies, lower SSTs and the impact of ENSO variability as potential climatic 658 659 drivers during this time.

660

# **661 7. Conclusions**

Compound specific carbon and hydrogen isotope data and *n*-alkane distributions (P<sub>aq</sub>) from 662 Mfabeni peatbog are used to reconstruct climatic conditions, over the last 32 cal ka BP in eastern 663 South Africa. The LGM at Mfabeni was characterized by a high contribution of C<sub>4</sub> grasses, low 664 665 precipitation amount/high ET and a low water table. We attribute the arid LGM conditions to an equatorward displacement (and strengthening) of the southern hemisphere westerlies, the SIOCZ 666 and the subtropical high-pressure cell, which may have extended the length and increased the 667 intensity of the dry season, as well as shifted the location of TTT formation northeast of Mfabeni. 668 669 These mechanisms for driving LGM climate in South Africa are consistent with an increase in Antarctic sea ice extent. Between c. 19–5 cal kyr BP an expansion of  $C_3$ -type vegetation occurred, 670 with more rainfall and a higher water table at Mfabeni. At c. 19 cal ka BP, a southward retreat of the 671 westerlies, the SIOCZ and the subtropical high-pressure cell occurred, coincident with a retreat in 672 673 Antarctic sea ice. This ice retreat combined with an increase in local summer insolation, after c. 12 cal 674 ka BP, may have resulted in more precipitation and an increased wet season length at Mfabeni. When the westerlies, the SIOCZ and the subtropical high-pressure cell were in their southernmost 675 676 position (c. 14–5 cal kyr BP), local insolation became the dominant control on Mfabeni climate, leading to stronger convection and enhanced monsoonal precipitation from the tropical easterlies. 677 The late Holocene (c. <5 cal ka BP) was characterized by increased environmental instability and 678 increasingly arid conditions. We attribute these trends to concurring low SSTs, and the recurring 679 680 influence of the southern westerlies and/or heighted ENSO activity.

The Mfabeni record indicates that climate and environmental variability in eastern South Africa over the last 32 cal ka BP were driven by a combination of i) enhanced/reduced moisture transport by the tropical easterlies, driven by variations in southern hemisphere summer insolation, and ii) latitudinal displacements (and the strengthening/weakening) of the westerlies, the SIOCZ and the subtropical high-pressure cell. With the expansion and retreat of Antarctic sea ice ultimately responsible for the displacement of these systems, we invoke high-latitude climate forcing as an important driver of climate in eastern South Africa.

688

Data availability: Supplementary data for the depth-age model (S1) is available with this manuscript.
 A new depth-age model of core MF1 (Finch and Hill, 2008), produced by Bacon, can be found within
 the supplementary information (S2). Other data is available on PANGAEA.

692

693 **Author contributions:** CM and ES conducted  $\delta^{13}C_{wax}$  and  $\delta D_{wax}$  analyses. Interpretation was carried 694 out by CM, JF, TH, FP, MH, MZ and ES.

695

696 **Competing interests:** The authors declare no competing financial interests.

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701

## 702 Figure captions

703 Figure 1. Map of South Africa in austral summer (a) and winter (b) showing the major oceanic and 704 atmospheric currents and the position of the Congo Air Boundary (CAB). H (L) = high (low)-pressure 705 systems. BC = Benguela Current. AC = Agulhas Current. Rainfall zones are shown in (a): WRZ = winter 706 rainfall zone, YRZ = year-round rainfall zone, SRZ = summer rainfall zone. SIA = South Indian Anticyclone. SAA = South Atlantic Anticyclone. SIOCZ = South Indian Ocean Convergence Zone. Note, 707 708 the westerlies move north during austral winter and the high-pressure system dominates over much of the continent, suppressing rainfall in the SRZ. Squares represent the key study sites mentioned in 709 710 the text (and shown in Fig. 4 and 5): a) MD79257 (Sonzogni et al., 1998). b) Mfabeni, this study (red 711 square). c) Lake St Lucia (Humphries et al., 2016). d) Seweweekspoort (Chase et al., 2017). e) Cecilia 712 Cave (Baxter, 1989). f) Klaarfontein (Meadows and Baxter, 2001) and Verlorenvlei (Carr et al., 2015), 713 one location. Figure modified from Gasse et al. 2008.

714

Figure 2. Mfabeni peatland and its regional geomorphological features, indicating the location of
core MF4-12 (red circle, this study) and the location of core SL6 (black circle, Baker et al., 2014; 2016;
2017). Map is courtesy of B. Gijsbertsen, UKZN Cartography Unit.

718

Figure 3. Depth-age model of Mfabeni core MF4-12 produced using Bacon, based on 24 <sup>14</sup>C AMS dates (S1). Blue symbols are AMS dates and grey shading indicates 95% confidence interval on the mean age (red line).

722

723 Figure 4. Climate and environmental change at Mfabeni compared with regional records and orbital insolation. a) December-January-February (DJF) insolation for 28°S (blue line; Laskar et al., 2011). b) 724 Stable carbon isotope composition (weighted mean) of  $C_{29}-C_{31}$  *n*-alkanes from Mfabeni, reflecting 725 changes in  $C_3/C_4$  vegetation type. c) Hydrogen isotope composition (weighted mean) of  $C_{29}-C_{31}$  n-726 727 alkanes from Mfabeni, reflecting changes in precipitation amount and ET. Red is the  $\delta D_{wax}$  corrected 728 for ice volume changes. Error bars on isotope data reflect analytical uncertainty of duplicate analyses. d) Central and eastern South African regional precipitation stack (red line; Chevalier and 729 Chase, 2015). e) Southern African regional aridity stack (Chevalier and Chase, 2016). f) Pag at Mfabeni, 730 731 indicating the amount of aquatic vs. terrestrial *n*-alkanes (high/low water table). g) Mean grain size data of the lithogenic sediment fraction from Mfabeni (Humphries et al., 2017). h) Mfabeni core SL6 stable carbon isotope composition (weighted mean) of  $C_{29}$ – $C_{31}$  *n*-alkanes (Baker et al., 2017). i) Combined nitrogen isotope data from Seweweekspoort rock hyrax middens, reflecting changes in humidity (Chase et al., 2017). j) U<sup>K'</sup><sub>37</sub> derived SSTs from core MD79257 in the Mozambique Channel (Sonzogni et al., 1998). k) Sea salt sodium concentrations from the EPICA DML ice core in Antarctica, reflecting changes in sea ice coverage (Fischer et al., 2007). The two Mfabeni samples with CPI values of *c*. 2 are highlighted in red (4b & c). Blue shading = Mfabeni wet, orange = Mfabeni arid.

739

740 Figure 5. Comparison of Mfabeni data with other records of environmental variability over the last 15 741 cal kyr BP. a) DJF insolation for 28°S (black line; Laskar et al., 2011). b) Carbon isotope composition (weighted mean) of  $C_{29}$ - $C_{31}$  *n*-alkanes from Mfabeni, reflecting changes in  $C_3/C_4$  vegetation type. c) 742 743 Hydrogen isotope composition (weighted mean) of C<sub>29</sub>-C<sub>31</sub> n-alkanes from Mfabeni, reflecting changes in summer precipitation amount and ET. d) P<sub>aq</sub> at Mfabeni, indicating the amount of aquatic 744 745 vs. terrestrial n-alkanes (high/low water table). Blue dashed lines highlight trends. e) Mfabeni 746 calcium/scandium ratio, indicating changes in water table (Humphries et al., 2017). f) Bulk carbon 747 isotope data from Verlorenvlei (Carr et al., 2015). g) An estimation of the extent of Antarctic sea ice 748 based on the abundance of Fragilariopsis curta and Fragilariopsis cylindrus at site PS2090/ODP1094 (SW of Cape Town; Bianchi and Gersonde, 2004). h) Red colour intensity time-series from Laguna 749 750 Pallcacocha. High values are light coloured inorganic clastic laminae, which were deposited during 751 ENSO-driven episodes (Moy et al., 2002). The Mfabeni sample with a CPI value of c. 2 is highlighted in 752 red (5b & c).

753

754 Figure 6. Summary figure highlighting the main climate phases and driving mechanisms at Mfabeni. All pollen data is from Finch and Hill (2008). Note, the new age model for pollen % data is in the 755 756 supplementary material (S2). a) Podocarpus % data from Mfabeni. b) Poaceae % data from Mfabeni. c) Cyperaceae % data from Mfabeni. d) Asteraceae % data from Mfabeni. e) Morella serrata % data 757 from Mfabeni. Poaceae and Cyperaceae were excluded from the regional pollen sum so their 758 759 percentages are based on total pollen frequencies. Podocarpus, Asteraceae and M. serrata 760 percentages are based on regional frequencies. See Finch and Hill (2008) for more details. f) Stable carbon isotopic composition (weighted mean) of C<sub>29</sub>-C<sub>31</sub> *n*-alkanes from Mfabeni. g) Hydrogen 761 762 isotope composition (weighted mean) of  $C_{29}$ - $C_{31}$  *n*-alkanes from Mfabeni. Red is the  $\delta D_{wax}$  corrected 763 for ice volume changes. The two Mfabeni samples with CPI values of c. 2 are highlighted in red. Blue shading = Mfabeni wet, orange = Mfabeni arid. 764

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