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1 **Tropical and high latitude forcing of enhanced megadroughts in Northern China during**
2 **the last four terminations**

3
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14
15 **ABSTRACT**

16 Understanding the origin and evolutionary history of drought events is of great significance,
17 providing critical insight into future hydrological conditions under the changing climate. Due
18 to the scarcity of drought proxies from northern China, the occurrence and underlying
19 mechanisms of the drought events remains enigmatic on longer timescales. Here we utilize
20 microbial lipid proxies to reconstruct significant drought events over the last four ice age
21 terminations in the southernmost section (Weinan section) of the Chinese Loess Plateau. The
22 abundance of archaeal isoprenoid GDGTs (glycerol dialkyl glycerol tetraethers) relative to
23 bacterial branched GDGTs, measured by $R_{i/b}$ and BIT indices, is diagnostic of enhanced
24 drought conditions. The $R_{i/b}$ (and BIT) indices are stable and low (high) throughout most of
25 the loess section spanning the last 350 thousand years, but they do exhibit sharp transient
26 peaks (valleys) during the intervals associated with the four ice age terminations, and
27 especially Terminations II and IV. These enhanced drought events are, non-intuitively,
28 associated with a significant decrease in the relative abundance of C4 plants, inferred by a
29 decrease in the carbon isotope composition of bulk organic matter. Although the microbial
30 records show some consistency with the Weinan grain size profiles, indicative of Eastern

31 Asian winter monsoon variability, they also show some apparent difference. In fact, some
32 features of the microbial records exhibit strong similarities with marine sediment planktonic
33 foraminiferal $\delta^{13}\text{C}$ records from the western Pacific warm pool, which reflect ENSO-like
34 changes during glacial terminations. Therefore, enhanced droughts immediately before the
35 interglacial warming in northern China could be explained, at least in part, by teleconnections
36 in tropical ocean-atmosphere circulation via shifts in the Intertropical Convergence Zone
37 (ITCZ) and associated Jet Stream over the Asian continent. According to our microbial
38 biomarker data, these enhanced megadroughts are apparently different, both in terms of
39 severity and causal mechanism, from the more commonly discussed dry conditions observed
40 during glacial periods.

41

42 **Keyword:** drought; microbial biomarkers; glacial terminations; Asian monsoon

43

44

45 **Highlights**

- 46 ● Microbial tetraether lipids analyzed for ~350-kyr interval in Chinese Loess Plateau
- 47 ● Megadroughts identified during four glacial terminations on the basis of microbial lipids
- 48 ● Tropical and high latitude forcing proposed for enhanced droughts in North China
- 49 ● Megadroughts during glacial terminations different from regular glacial droughts

50

51 1. INTRODUCTION

52 Drought events exert severe impacts on both terrestrial and aquatic ecosystems, and also
53 society (Webster et al., 1998; Cohen et al., 2007). The history of droughts in the Asian
54 interior has been the focus of much investigation, especially with respect to the impact of the
55 Tibetan Plateau (TP) uplift on enhanced aridity during the late Cenozoic (Manabe et al., 1990;
56 An et al., 2001). Indeed, grain size analyses of Chinese Loess Plateau (CLP) sediments—a
57 proxy for the strength of East Asian winter monsoon (EAWM) winds (e.g., Ding et al.,
58 1995)—has revealed periods of desertification in Central Asia going as far back as the
59 Neogene (Guo et al., 2002). More recently, speleothem records from central (e.g., Wang et al.,
60 2008; Cheng et al., 2009, 2016) and southwest (e.g., Cai et al., 2015) China have shown, via
61 oxygen-isotope ratios ($\delta^{18}\text{O}$) [proxy of Asian summer monsoon (ASM) variability],
62 precession-driven fluctuations in the ASM through glacial-interglacial cycles, as well as
63 millennial-scale perturbations during the last glacial-deglaciation apparently driven by North
64 Atlantic (e.g. Wang et al., 2008) and Antarctic (e.g., Zhang et al., 2016) meltwater events.
65 Records from the western CLP have shown that glacial boundary conditions (i.e. sea ice,
66 atmospheric CO_2) have a more dominant influence on summer precipitation changes in North
67 China (Sun et al., 2015).

68 Despite these advancements in our understanding of orbital- and millennial-scale ASM
69 variability, there still remains a large gap in our knowledge of the spatial homogeneity (or
70 heterogeneity) of monsoon variability in China under varying boundary conditions, and in
71 particular, how changes in the summer monsoon can be manifested as periods of enhanced
72 drought. This is because hitherto most of the longer-term terrestrial monsoon records are
73 sourced from the $\delta^{18}\text{O}$ of the ever-growing speleothem network, despite recent research
74 suggesting that these proxies, particularly those located over central China, primarily reflect
75 large-scale Indian Summer Monsoon (ISM) variability upstream of the cave sites, and not
76 necessarily local precipitation amount (e.g., Pausata et al., 2011; Liu et al., 2014). Therefore,
77 we still lack longer-term records of enhanced drought conditions, or ‘megadroughts’, from
78 the ASM domain, particularly in the CLP region. Recent work by Cook et al. (2010)
79 identified a series of megadroughts [i.e. extreme hydrological events of naturally occurring
80 multidecadal precipitation variations (Meehl et al., 2006)] over the last millennium, which

81 were attributed to summer monsoon failures associated with tropical Pacific sea surface
82 temperature (SST) anomalies. Moreover, Zhang et al. (2008) showed that prolonged periods
83 of monsoon failure occurred over the past millennium, and interestingly, linked these
84 megadrought events with the demise of several Chinese dynasties. Despite these studies
85 shedding light on the magnitude and frequency of these megadroughts in East Asia, their
86 relative brevity precludes a robust assessment of these extreme events on longer time scales
87 (i.e. glacial-interglacial, G-IG time scales). In addition, whilst there are records from the
88 western (e.g., Sun et al., 2006, 2015) and northern CLP (Guo et al., 2009; Hao et al., 2012)
89 which suggest that dry conditions prevailed during glacial times, we still know very little
90 about how the southern sector of the plateau responded to high and low latitude forcing.
91 Hence, gaining a deeper insight into the occurrence of these enhanced megadrought
92 conditions at this location is critical given the importance of monsoon precipitation to the
93 agriculture of the region.

94 In contrast to paleotemperature reconstructions, records of past aridity, particularly on
95 geologic timescales, are especially difficult to obtain given the lack of reliable and well
96 preserved proxies. Glycerol Dialkyl Glycerol Tetraethers (GDGTs, Fig. S1), which are
97 membrane lipids synthesized by archaea and bacteria (Schouten et al., 2013), have been used
98 to reconstruct the paleoclimate history of the CLP, particularly paleotemperature (Gao et al.,
99 2012; Jia et al., 2013; Peterse et al., 2011, 2014; Yang et al., 2014; Thomas et al., 2016).
100 However, in addition to soil alkalinity (Xie et al., 2012; Yang et al., 2014), recent research
101 has shown that the distributions of archaeal isoprenoid GDGTs (iGDGTs) and bacterial
102 branched GDGTs (brGDGTs) are also influenced by soil moisture (Wang et al., 2013;
103 Dirghangi et al., 2013). Most notably, the $R_{i/b}$ ratio (i.e. the abundance of total iGDGTs
104 relative to total brGDGTs) has been shown to significantly increase during extreme arid
105 conditions, and as such, has the potential to be a reliable terrestrial archive of enhanced
106 droughts (Xie et al., 2012). In this study, we show that elevated $R_{i/b}$ values (>0.5) in the
107 southern sector [Weinan section (WS)] of the CLP likely mark intervals of enhanced drought,
108 defined as periods where mean annual precipitation (MAP) is less than 600 mm (Yang et al.,
109 2014, supplemental data Fig. S2). The ‘enhanced drought’ term is used here to discriminate
110 from ‘regular drought’ conditions identified during glacial periods. The term ‘megadrought’

111 is further used to identify enhanced drought conditions (identified by the $R_{i/b}$ ratio) that
112 occurred over long periods of time (e.g., over multiple decades; Meehl et al., 2006).
113 Furthermore, through a survey of the relationship between $R_{i/b}$ values and soil moisture
114 (ranging from 0 to 61%) along three transects perpendicular to the shoreline of Qinghai Lake
115 (located in the transitional zone between the TP and Chinese Loess Plateau), we find that $R_{i/b}$
116 values markedly increase when soil water drops below 30% (Dang et al., 2016),
117 corroborating the reliability of $R_{i/b}$ as an indicator of enhanced drought (supplemental data,
118 Fig. S3).

119 A closely related GDGT-based proxy, the BIT (Branched and Isoprenoid Tetraethers)
120 index, estimates the relative abundance of the main brGDGTs (brGDGTs-I, -II, -III) vs. one
121 specific iGDGT, crenarchaeol, which is biosynthesized by a group of archaea
122 (Thaumarchaeota). Initially, the BIT index was proposed to evaluate the input of terrestrial
123 organic material into immature marine and lake sediments (Hopmans et al., 2004), although
124 later it was found to exhibit a relationship with mean annual precipitation (Dirghangi et al.,
125 2013) and water content (Wang et al., 2013) in soils. In light of these findings, there is strong
126 potential for BIT to be a robust humidity proxy in terrestrial settings (supplemental data, Fig.
127 S2). Our results show an inverse relationship between $R_{i/b}$ and the BIT index, though it is
128 worth noting that the range of $R_{i/b}$ values is much larger when BIT values become relatively
129 low, indicating the potential tandem utility of these proxies in identifying enhanced drought
130 events (Yang et al., 2014). Therefore, we utilized both of these novel soil moisture proxies to
131 identify periods of enhanced aridity in the monsoon-dominated region of the CLP over the
132 past 350,000 years. Our findings will add to the growing body of records derived from
133 loess-paleosol sequences of the CLP for the Quaternary (e.g., An et al., 2001), providing
134 critical new information on past variations in monsoon climate, and the strong links with
135 Earth's changing boundary conditions (e.g., $p\text{CO}_2$, sea level, insolation).

136

137 **2. STUDY SITE AND METHODS**

138 *2.1. Study site and sampling*

139 The loess sequence from Weinan is located at the southern tip of the CLP (34°21.0' N;
140 109°32.0'E), and marks one of the wettest areas of the plateau (Fig.1a). The mean annual air

141 temperature (MAAT) at Weinan is 13.8°C and the mean annual precipitation (MAP) is 570
142 mm (based on China Meteorological Administration climate records during 1981-2010,
143 <http://www.cma.gov.cn>). The modern climate at the site is highly seasonal, with temperatures
144 typically exceeding 20°C between May and September and typically lower than ~5°C
145 between November and January. The annual rainfall is also highly seasonal and largely
146 governed by the strength of the East Asian summer monsoon (EASM), with 70% of the
147 annual precipitation delivered between May and September by moisture-laden air masses
148 sourced from the tropical oceans (Fig. 1c). The end of the EASM season is marked by a shift
149 in wind direction as the East Asian winter monsoon winds from the west bring cold and dry
150 conditions to the region.

151 The Weinan section investigated here contains 34.8 m of loess-paleosol (LPS),
152 extending from the L₄LL₁ loess [the topmost of L₄ phase corresponding to Marine Isotope
153 Stage (MIS) 10] to the Holocene paleosol S₀, covering the last three glacial-interglacial
154 cycles (MIS1-9). The samples were collected at 10cm intervals.

155

156 2.2. Grain size and magnetic susceptibility analysis

157 The magnetic susceptibility (χ) and sediment grain size were analyzed on samples
158 extracted at 10 cm intervals. The magnetic susceptibility and grain size analyses were
159 conducted following the methods of Hao et al. (2012). Specifically, the low-frequency
160 analysis of χ (n=349 samples) was measured at 0.47 kHz using a Bartington Instruments MS
161 2B magnetic susceptibility meter. For grain size analysis, the samples were treated with 10%
162 H₂O₂ and 10% HCl solution to remove organic matter and carbonate, respectively. After
163 dispersal using 0.05 mol/L (NaPO₃)₆, the samples were measured using a Mastersizer 2000
164 analyzer with the range of 0.02-2,000 μ m in diameter, and a precision of \pm 1%.

165

166 2.3. Age model

167 The age model of the Weinan section was obtained by interpolation between
168 geomagnetic polarity boundaries (Ding et al., 2002), using χ as an indicator of accumulation
169 rate (Ding et al., 2002; Kukla et al., 1988). This model is widely used to date the
170 loess-paleosol sections of the CLP. The χ and grain size data, analyzed at 10 cm intervals,

171 represent an average time resolution of 0.3-2.6 kyr.

172

173 2.4. *Lipid extraction*

174 A total of 198 loess-paleosol samples were transported to the lab immediately after
175 collection, and dried in an oven at 45°C. The samples were ground into powder with a mortar
176 and pestle, and passed through a 20-mesh sieve (0.85 mm in diameter) to remove tiny roots
177 and carbonate nodules. An aliquot of each sample (40-50g) was extracted with
178 dichloromethane (DCM): methanol (9:1, v/v) using an accelerated solvent extractor (ASE
179 100, Dionex, USA) at 100°C and 1400psi. The total lipid extracts were concentrated by a
180 rotary evaporator and separated into apolar and polar fractions using flash silica gel column
181 (0.7 cm i.d. and 1.5g activated silica gel) chromatography and with hexane (10ml) and
182 methanol (10ml) as the eluents, respectively. All polar fractions containing GDGTs were
183 passed through 0.45µm PTFE syringe filters and dried under nitrogen gas. The 198 samples
184 for GDGT analysis in this study include 37 samples of the S₀ layer reported by Yang et al.
185 (2014).

186

187 2.5. *GDGT analysis and proxy calculation*

188 Each polar fraction was re-dissolved in 300 µl *n*-hexane: ethyl acetate (EtOA) (84:16,
189 v/v), and a C₄₆ GDGT was added as a synthesized internal standard. 15µl of each sample
190 were injected and analyzed by an Agilent 1200 series liquid chromatography coupled to an
191 Agilent 6460A triple quadruple mass spectrometer (LC-MS/MS). Separation of the brGDGTs
192 was performed on two silica columns (150mm × 2.1mm, 1.9µm, Thermo Finnigan) in tandem.
193 The elution gradients were matched following the description of Yang et al. (2015). The
194 single ion monitoring (SIM) was used, monitoring at *m/z* 1302, 1300, 1298, 1296, 1292, 1050,
195 1048, 1046, 1036, 1034, 1032, 1022, 1020, 1018 and 744. The 5- and 6-methyl brGDGTs
196 were identified by the relative time order of compound peaks. Compound quantification was
197 performed by peak area integration of [M+H]⁺ in the extracted ion chromatogram. MS
198 conditions follow Hopmans et al. (2004). The 6-methylated brGDGTs are identified by an
199 accent after the roman numerals for their corresponding 5-methylated isomers. The typical
200 analytical errors for R_{i/b} and BIT are all better than 0.02.

201 The $R_{i/b}$ proxy was used to identify enhanced aridity conditions (Xie et al., 2012) and
202 calculated as follows:

$$203 \quad R_{i/b} = \frac{\sum(\text{iGDGTs})}{\sum(\text{brGDGTs})}$$

204 BIT is calculated according to the following formula (Hopmans et al., 2004):

$$205 \quad \text{BIT} = (\text{Ia} + \text{IIa} + \text{IIa}' + \text{IIIa} + \text{IIIa}') / (\text{Ia} + \text{IIa} + \text{IIa}' + \text{IIIa} + \text{IIIa}' + \text{crenarchaeol})$$

206 where Roman Numerals indicate the molecular structures of GDGTs shown in supplemental
207 data (Fig. S1).

208

209 2.6. Spectral analysis

210 The Arand software package (Howell et al., 2006) was used to calculate power spectra
211 and phase of time series. The spectral density of magnetic susceptibility and $R_{i/b}$ was
212 analysed at 1 ka interval after all the data were detrended. The Analyseries software was used
213 to conduct f-tests of spectral peak significance (Paillard et al., 1996).

214

215 3. RESULTS AND DISSCUSION

216 3.1. Distribution of GDGTs in the Weinan loess section

217 Both iGDGTs and brGDGTs were detected in all samples. The concentrations of GDGTs
218 are higher in paleosol layers than in adjacent loess layers. In most samples, brGDGTs
219 accounted for a higher proportion of total GDGTs (84.8% in average). Crenarchaeol is the
220 most abundant iGDGT. GDGT-Ia, -Ib, and -IIa' are the most abundant of the brGDGTs and
221 constitute almost 70% of the total brGDGTs on average. The GDGT-IIIc has the lowest
222 concentration in nearly all samples, and is below the detection limit in some cases. The
223 average distribution of GDGTs in loess layers was not significantly different from the
224 paleosols.

225 It is noteworthy that the 6-methyl brGDGTs constitute, on average, 32.9% of the total
226 brGDGTs, which has implications for mean annual air temperature reconstruction (see below).
227 Similarly, the proportion of brGDGT-IIa, one of the main components in brGDGT-based
228 proxies, is lower in the Weinan loess (0.7%-9.6%, 3.6% on average) than in the global soils
229 dataset (0%-24%, 18% on average).

230

231 3.2. *Paleotemperature reconstruction of the Weinan section*

232 In combination with the widely-used age models of the CLP (e.g., Kukla et al., 1988;
233 Porter and An, 1995), paleotemperature records reconstructed from the molecular proxies in
234 the same section, could potentially help to further constrain the timing of glacial terminations
235 in northern China. Indeed, branched GDGTs have been used to reconstruct the MAAT at
236 various locations across the planet, usually based on global MBT (methylation index of
237 branched tetraethers) and CBT (cyclization of branched tetraethers) indices against MAAT
238 and pH (as initially proposed by Weijers et al., 2007, and later refined by Peterse et al., 2012).
239 It is noteworthy that MAATs derived from the global MBT/CBT calibrations are typically too
240 high when applied to arid regions, including the CLP (Gao et al., 2012; Jia et al., 2013;
241 Peterse et al., 2014; Peterse et al., 2011; Dang et al., 2016). However, the relatively new
242 global calibration based primarily on the 5-methylated and tetramethylated brGDGTs appears
243 to minimise the influence of precipitation and to reduce the error in paleotemperature
244 reconstruction in the semi-arid and arid regions (De Jonge et al., 2014):

$$245 \text{ MAT}_{\text{mr}} = 7.17 + 17.1 \times [\text{Ia}] + 25.9 \times [\text{Ib}] + 34.4 \times [\text{Ic}] - 28.6 \times [\text{IIa}]$$

246 where roman numerals correspond to the molecular structures of GDGTs shown in the
247 supplemental Fig. S1.

248 Over the last 350 ka, the WS shows large variations (~10.6 °C range) in MAAT on
249 glacial-interglacial timescales (Fig. 3h). The reconstructed MAAT exhibits a maximum of
250 23.7 °C at the beginning of MIS7 (ca. 250 ka BP), which is slightly warmer than MIS 5 (ca.
251 130 ka BP). This result is somewhat surprising given that MIS5 is generally thought to
252 represent the globally warmest interglacial period of the studied interval. The
253 brGDGT-derived MAAT record also reveals that MIS 5c (ca. 113 ka B.P.) was the warmest
254 within MIS5. This is similar to the results of Lu et al. (2007) and Peterse et al. (2014), also
255 based on brGDGT distributions. As expected, the lowest reconstructed MAATs occur during
256 glacial times (Fig. 3h), with the temporal patterns showing broad similarities to other records.
257 During terminations I, II, III and IV, the MAAT at our site exhibits minimum values. Not
258 surprisingly, these periods of minimum MAATs in the southern CLP coincide with low NH
259 summer insolation (Fig. 3l). Conversely, periods of warming are matched by higher summer
260 insolation. The strong connections between Weinan MAAT and both global ice volume and

261 NH summer insolation highlight the sensitivity of the region to shifts in Earth's boundary
262 conditions. As discussed in more detail below, in most cases the dramatic drop in
263 reconstructed MAATs is associated with very low BIT values (Fig. 3g) and high $R_{i/b}$ ratios
264 (Fig. 3f), lending support to the conclusion that the enhanced drought events occurred during
265 glacial terminations in the CLP. It is worth noting, however, that there are several sudden
266 declines in temperature that do not correspond to changes in BIT and $R_{i/b}$, such as during
267 precession minima through MIS4 and MIS6. The cold climate in the CLP was thus not
268 necessarily accompanied by the occurrence of extreme drought events.

269

270 *3.3. Molecular and sedimentological records of intensified drought events*

271 Our microbial lipid record (n=198 samples) indicates that the $R_{i/b}$ ratio remains relatively
272 low and stable throughout most of the record (Fig. 3f). It is reasonable to assume that the $R_{i/b}$
273 ratios did not change when the precipitation was >600 mm or the soil water content was >
274 30% (Fig. S3), such that intervals with low ratios could have still experienced mildly arid
275 conditions. The exceptions to this overall stability are the very large and abrupt increases that
276 occur during the transitional periods from loess to paleosol (i.e., from L_2 to S_1 , L_3 to S_2 , and
277 L_4 to S_3) (Fig. 3). These intervals correspond with the glacial terminations, including
278 Terminations II-IV (Fig. 3f), where values increased 5-15 fold when local MAAT was
279 ~14-16 °C (Fig. 3f). Specifically, $R_{i/b}$ ratios increase from a baseline value of ~0.2 [typical for
280 soils from non-arid settings (Yang et al., 2014)] to a ratio of ~0.5 during Terminations III and
281 IIIa which is typically characteristic of soils with a pH > 8 (Yang et al., 2014); values were
282 highest during Terminations II and IV where ratios exceeded 0.83. An increase in $R_{i/b}$ ratios
283 (0.40), albeit smaller than that observed during the other terminations (0.40), also occurs at
284 the L_1/S_0 boundary corresponding to the T-I. However, the relatively lower values through T-I,
285 compared with other terminations, merits further investigation. Remarkably, besides the
286 terminations, the only time the $R_{i/b}$ ratios exceed a value 1 is during the late Holocene, which
287 is probably due to land use changes. For example, agricultural practices can lead to the
288 surface soils becoming more loose and porous, and as a result, the ability of the soil to hold
289 water decreases and the evaporation potential increases, ultimately drying out the surface
290 soils.

291 Additional evidence for the increased $R_{i/b}$ ratios reflecting drought conditions is provided
292 by the WS BIT indices, which range from 0.38 to 0.98 and exhibit the same trends as the $R_{i/b}$
293 values throughout the whole sequence. Although the BIT index is also a ratio of isoprenoidal
294 and branched GDGTs, it comprises different GDGTs and therefore different microorganisms,
295 providing additional evidence for profound change in the microbial community.

296 The large changes in $R_{i/b}$ ratios (and BIT indices) provide direct evidence for a more arid
297 North China climate during glacial terminations. It is notable though that the microbial
298 proxies presented here only record very enhanced drought, or megadroughts, but not the more
299 subtle drought events (Xie et al., 2012). Hence, this likely explains the differences between
300 our records and those proxies [e.g. grain size (Ding et al., 2002; Hao et al., 2012) and $WS\chi$
301 (An et al., 1991)] that are associated with more subtle changes in the monsoon system. Our
302 results are in-line with previous findings from the region (e.g., Guo et al., 2009; Hao et al.,
303 2012), suggesting that megadroughts occurred during glacial terminations, when NH ice
304 volume was greatest and NHSI (North Hemisphere Summer Insolation) was generally low
305 (Fig. 4). Moreover, our record likely explains previously reported sedimentological features
306 in central Asia, including the extremely high accumulation rate of loess in the west Kunlun
307 area of the TP (Zan, 2010), the absence of growth intervals in Kesang cave stalagmite records
308 from the western TP (Cheng et al., 2012), and the low values in loess deposits from Jingyuan
309 (Sun et al., 2006) and Chashmanigar, Tajikistan (Ding et al., 2002).

310 The inferred shifts in megadroughts at Weinan (inferred from the BIT and $R_{i/b}$ records)
311 are concentrated at the glacial-interglacial timescale (100-kyr scale) (Fig. 5), whereas the
312 occurrence of pluvials and droughts are also modulated by Earth's precessional cycle, akin to
313 the signals preserved in Chinese speleothem records from Hulu and Sanbao caves (Wang et
314 al., 2008; Cheng et al., 2009, 2016). Indeed, higher (lower) speleothem $\delta^{18}O$ values,
315 indicative of a weaker (stronger) Indian summer monsoon (e.g., Pausata et al., 2011; Liu et al.,
316 2014), are matched by intervals of lower (higher) $WS\chi$ and higher (lower) WS grain size
317 values, suggesting weaker (stronger) EASM and stronger (weaker) EAWM conditions,
318 respectively, during periods of low (high) NHSI. This result suggests that the high $R_{i/b}$ values
319 primarily reflect only the most severe drought events, associated with glacial terminations.

320 The extremely cool and dry conditions during glacial terminations would also have

321 impacts on the vegetation of the CLP. Evidence for this comes from $\delta^{13}\text{C}$ -depleted bulk soil
322 organic matter which indicates, unexpectedly, a sudden decrease in the relative abundance of
323 C4 plants in the Weinan loess-paleosol sequence (Fig. 3c) (Sun et al., 2011). Large $\delta^{13}\text{C}$
324 variations have been used to estimate shifts in the C3/C4 ratio of vegetation because of the
325 different photosynthetic pathways associated with these plant types (O'Leary, 1988). Whilst
326 some studies have interpreted the $\delta^{13}\text{C}$ changes in loess-paleosol sequences to reflect shifts in
327 water use efficiency and aridity (Hatte and Guiot, 2005; Zech et al., 2007), others have
328 proposed that the vegetation changes are primarily governed by temperature. For example,
329 several studies have proposed that cold (warm) climates were characterized by a general
330 expansion (reduction) of C3 (C4) vegetation in the CLP (Zhang et al., 2003; Liu et al., 2005).
331 The consistency between enhanced drought conditions and negative $\delta^{13}\text{C}$ excursions during
332 glacial times contradicts what is expected for an aridity control, and thus favours the latter
333 interpretation.

334

335 *3.4. Mechanism for enhanced aridity during glacial terminations*

336 The enhanced drought events identified by the $R_{i/b}$ and BIT proxies during glacial
337 terminations are generally associated with an higher percentage of grain size $> 32\mu\text{m}$,
338 diagnostic of the intensification of EAWM (An et al., 1991; Ding et al., 2002; Hao et al.,
339 2012). Moreover, the episodic droughts identified by WS — i.e. the monsoon failures
340 typically occurring during periods of reduced NHSI — are coeval with higher WS grain size.
341 Therefore, it is likely that periods of summer monsoon failure were strongly linked with the
342 synchronous increases in winter monsoon winds, which are known to influence hydroclimate
343 in China on G-IG time scales via shifts in the ITCZ (e.g., Yancheva et al., 2007; Cosford et al.,
344 2008); a stronger winter monsoon would push the ITCZ and the rain belt southwards,
345 resulting in increased aridity in northern China. However, there are some apparent differences
346 between the EASM and EAWM proxies (Fig. 3i, j), suggesting that changes in the EAWM
347 cannot fully explain the observed enhanced droughts during glacial terminations.

348 Comparison between the reconstructed southern CLP megadroughts presented here
349 reveal, to an extent, similarities with ice-rafted debris (IRD) records from the North Atlantic
350 (Fig. 3e). The two major $R_{i/b}$ maxima during T-II and T-IV are coincident with significant

351 increases in IRD. The relatively smaller $R_{i/b}$ increases during T-III are associated with
352 similarly small increases in IRD. Previous work has illustrated the strong influence of North
353 Atlantic meltwater pulses (i.e. Heinrich events) on northern China aridity (e.g., Guo et al.,
354 1996). An increased freshwater flux to the North Atlantic during the last deglaciation,
355 associated with enhanced IRD deposition, would have resulted in a slow-down of the Atlantic
356 meridional overturning circulation (AMOC) (e.g., McManus et al., 2004; Böhm et al., 2014).
357 The climate signal of the North Atlantic appears to have been transmitted to the Asian
358 monsoon regions via the northern westerlies, leading to enhanced EAWM winds and reduced
359 summer monsoon precipitation (Sun et al., 2012). However, not all the IRD events are
360 associated with enhanced drought in northern China. For example, the maximum IRD event
361 at ~280 ka BP, corresponding to loess deposition during mid L₃, exhibits no association with
362 both the WS BIT and $R_{i/b}$ indices, and hence no enhanced drought event. In addition, the
363 generally high IRD deposition between 75 and 25 ka BP, corresponding to the loess
364 deposition L₁, is also not matched by anomalous BIT and $R_{i/b}$ values, at least when compared
365 with those events occurring at T-II and T-IV. Thus, we conclude that although North Atlantic
366 freshwater influx events could have brought about CLP enhanced drought, other
367 teleconnections with the NH were also important.

368 The simulations of Sun et al. (2015) and others (e.g., Kutzbach and Guetter, 1986;
369 Kutzbach et al., 2008; Weber and Tuenter, 2011; Lu et al., 2013; Liu et al., 2014), suggest
370 that the dominant forcings imposed on the mid latitude monsoon regions are changing surface
371 boundary conditions (ice sheet extent, sea ice, land albedo), whereas monsoon regions closer
372 to the equator appear to be more influenced by summer insolation. This is certainly apparent
373 in the WS record, along with those of Sun et al. (2015, Fig. 5b), which show a dominant
374 100-kyr signal, whereas the speleothem records from southern China show a dominant
375 precessional (23-kyr) signal (Fig. 5a). Of particular note, the model sensitivity experiments
376 conducted by Sun et al. (2015) demonstrate that the spatial variability is primarily the result
377 of the southern monsoon regions, particularly those sites located near the coast, being
378 dominated by changes in the land-sea thermal contrast, which is modulated by summer
379 insolation (Kutzbach and Guetter, 1986). By contrast, the more northern sites in China are
380 more influenced by the shifting westerlies, and their interaction with the Tibetan Plateau (e.g.,

381 Chiang et al., 2014). Specifically, empirical evidence has shown that increased NH ice sheet
382 extent, such as during glacial maximums, likely increased the hemispheric thermal gradient
383 (NH hemisphere cooler than the SH) (Yanase and Abe-Ouchi, 2007; Jiang et al., 2011),
384 which lead to an increase in the westerlies (e.g., Yanase and Abe-Ouchi, 2007) and therefore
385 strengthened EAWM winds. In addition, the extent of the NH ice sheets also pushes the
386 Siberian High further southwards, which consequently acts to block the northward migration
387 of the Asian Summer monsoon (Peterse et al., 2014; Thomas et al., 2017). At the same time,
388 sea level was lower as was atmospheric CO₂ concentrations. Model simulations suggest that
389 all of these factors could have contributed to preventing the monsoon front from penetrating
390 as far north as the CLP, thus reducing summer monsoon rainfall amount in Northern China
391 (Sun et al., 2015). Whilst the effects of insolation, ice sheet extent, and CO₂ impact all of East
392 Asia, the magnitude of these forcings on the hydroclimate varies from south to north. For
393 example, the model simulations indicate that the magnitude of monsoon reductions induced
394 by increased ice and decreased CO₂ (such as during the LGM), are much greater in Northern
395 China compared with Southern China (Sun et al., 2015).

396 Despite the model simulations described above suggesting that changing
397 glacial-interglacial boundary conditions (e.g., ice sheets, land albedo, and sea ice) impose a
398 greater forcing on mid latitude monsoons than local and/or tropical forcing (e.g., insolation),
399 it is unlikely that the effects of ice volume alone can explain the observed enhanced droughts
400 in the CLP. This is because these effects should be similarly impactful during glacial periods,
401 but they are only observed during terminations. Therefore, other forcing factors must play a
402 role in amplifying the response from NH ice sheet extent. To that end, we find evidence for
403 extreme droughts also being linked with variations in the tropical oceans. In particular, the
404 enhanced aridity records reconstructed here (via molecular proxies) are consistent with *P.*
405 *obliquiloculata* stable carbon isotope minima from the western Pacific warm pool (WPWP;
406 Fig. 3d, Jia et al., 2015). The two major R_{i/b} maxima during T-II and T-IV, are associated with
407 the two largest decreases in δ¹³C values in the WPWP of the past 350 thousand years.
408 Moreover, the relatively smaller R_{i/b} increases during T-III are associated with similarly small
409 decreases in δ¹³C. These phase relationships suggest that, in addition to high northern latitude
410 forcings, the threshold of megadroughts in Northern China could also be connected with

411 changes occurring in the tropical Pacific.

412 The $\delta^{13}\text{C}$ values of planktic subsurface water species *P. obliquiloculata* in the western
413 Pacific MD06-3047B core (Fig. 3d) show highly depleted excursions during T-I, T-II, and
414 T-IV. During terminations, these $\delta^{13}\text{C}$ data suggest that the thermocline was lower in the
415 WPWP compared with the Eastern Pacific (Jia et al., 2015; Farrell et al., 1995), similar to
416 El-Niño conditions today. This teleconnection pattern is proposed to have induced changes in
417 El Niño-Southern Oscillation (ENSO)-like variability, comprising a complicated high- and
418 low-latitude feedback mechanism during glacial terminations (Pena et al., 2008). These
419 meridional teleconnections travel through the atmosphere via latitudinal shifts in wind
420 patterns and through the ocean by circulation changes of intermediate water from the polar
421 regions to the tropical thermocline waters (Pena et al., 2008). Model-proxy syntheses have
422 also suggested an altered ENSO state during the LGM via the first-order influence of the
423 exposed Sunda Shelf landmass on the Walker circulation (DiNezio and Tierney, 2013).
424 Specifically, the models and proxy records highlighted in their study suggest that the exposed
425 Sunda and Sahel Shelves drove reductions in convection over the Indo-Pacific during glacial
426 terminations. Moreover, speleothem $\delta^{18}\text{O}$ records from Borneo (e.g., Meckler et al., 2012;
427 Carolin et al., 2016), which show decreased convection during T-I (and other terminations),
428 appear to align with this Walker circulation mechanism, although as pointed out by Carolin et
429 al. (2013), the timing of Sunda Shelf inundation during T-I and T-II relative to Borneo $\delta^{18}\text{O}$
430 changes are not consistent between the terminations (Fig. 3b). Regardless of the mechanism
431 driving the reduced convection over the Indo-Pacific warm pool (IPWP), it appears that on
432 G-IG time scales, reduced convection in this region during glacial terminations played a
433 critical role in amplifying megadrought conditions over the CLP, possibly due to a reduction
434 in atmospheric heat and vapor transport from the tropics. Under modern conditions, reduced
435 convection over the IPWP, for example during El Niño years, leads to an overall decrease in
436 precipitation over Northern China (Xiao et al., 2000; Gong and Wang, 1999), and thus has led
437 to enhanced droughts in the Northern Chinese Plains (Huang and Wu, 1989). Because the
438 ITCZ tends to be constrained closer to the equator during El Niño events, an equatorward
439 ITCZ shift in East Asia would lead to a moisture deficit in Northern China.

440

441 **4. CONCLUSIONS**

442 We identify enhanced drought events at the last four ice age terminations on the basis of
443 microbial lipid distributions in the southernmost part (Weinan section) of the Chinese Loess
444 Plateau. The abundance of archaeal isoprenoid GDGTs (glycerol dialkyl glycerol tetraethers)
445 relative to bacterial branched GDGTs, measured by $R_{i/b}$ and BIT indices, is diagnostic of
446 extreme drought events. The $R_{i/b}$ (and BIT) indices are stable and low (high) throughout most
447 of the loess section spanning the last 350 thousand years, but they exhibit sharp transient
448 peaks (valleys) during the intervals corresponding to the four ice age terminations, and
449 especially those of Termination II and IV. These enhanced drought events occurring
450 immediately before the interglacial warmings are different from, but much more severe than,
451 the dry conditions during glacial periods. These enhanced megadroughts appear to be
452 controlled by changing glacial-interglacial boundary conditions (e.g., ice sheets, land albedo,
453 and sea ice) affecting the position of westerlies, but also amplified by a reduction in
454 northward heat/moisture transport from the IPWP because of cooler SSTs and a weaker
455 Walker circulation during glacial terminations.

456

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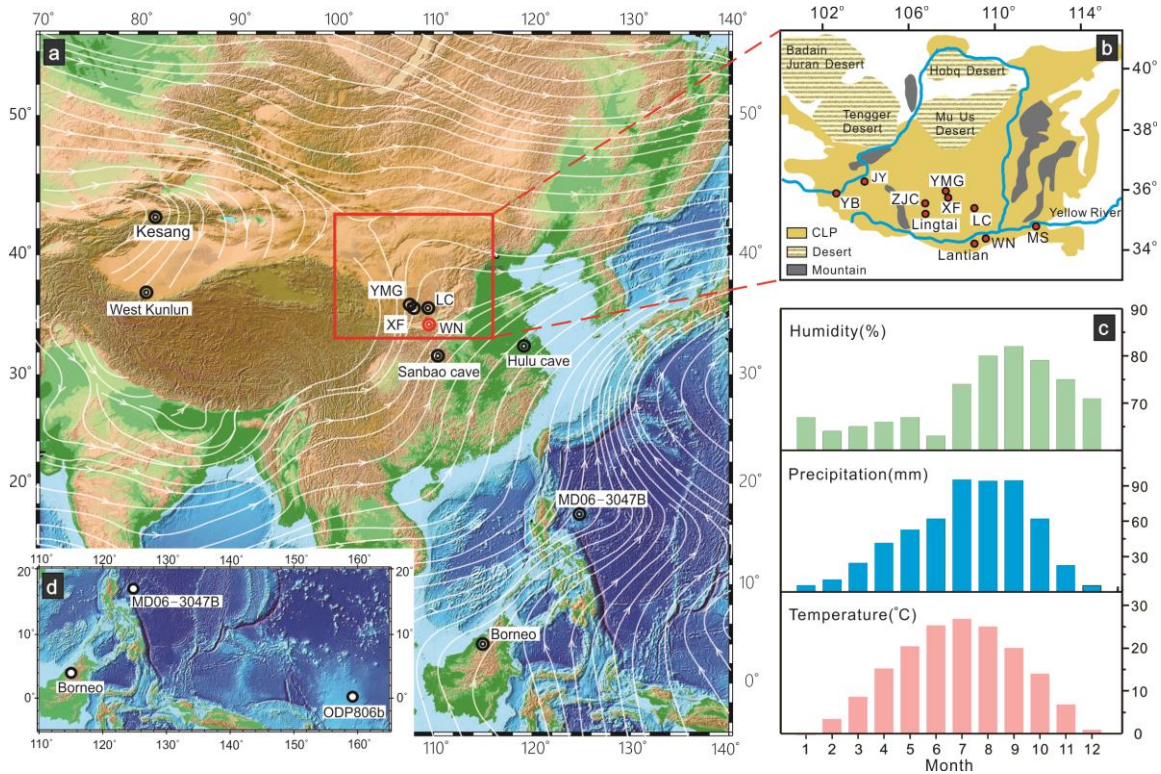
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710

711 **FIGURES**

712 Fig.1



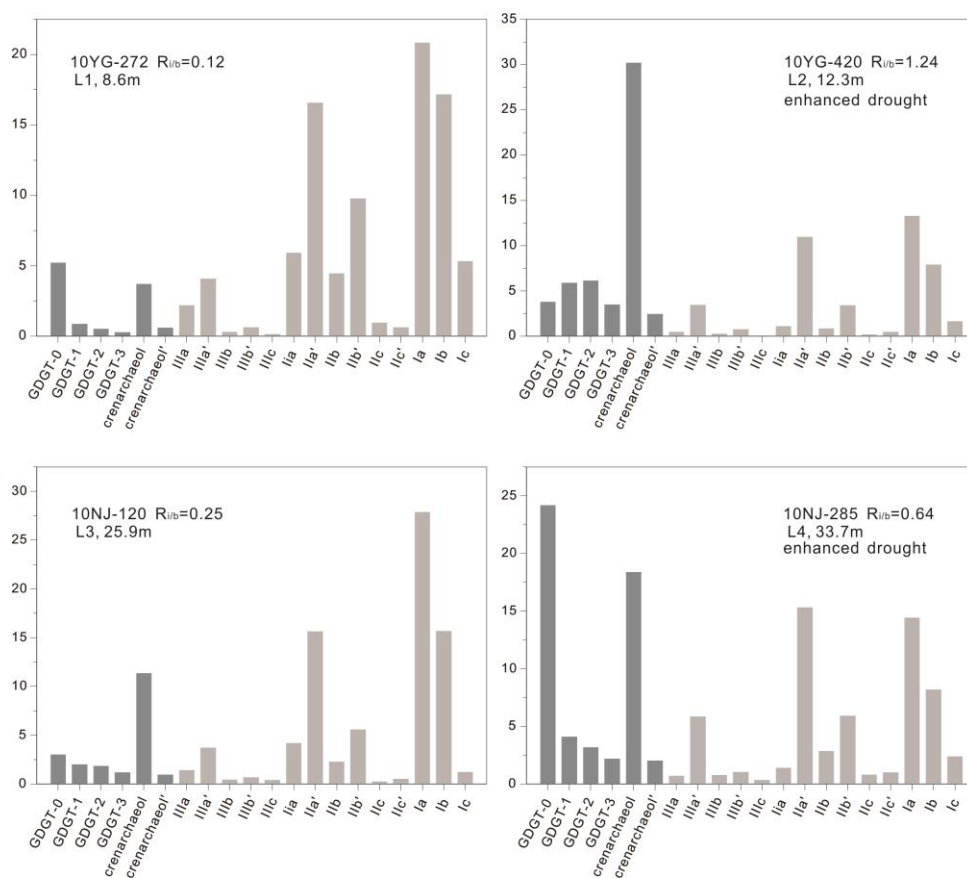
713

714 Fig. 1. The locality (a, b) and modern climatology (c) of the Weinan section and the sites
 715 mentioned in the text, as well as the averaged atmospheric flow fields at 700 hPa isobaric in
 716 summer (JJA) from 1971 to 2000 (a, Kalnay et al., 1996; An et al., 2012). The location of
 717 loess-paleosol sections mentioned in the text (a, b) include: WN (Weinan section, this study
 718 and Thomas et al., 2016, 2017; 34°21' N; 109°32'E), MS (Mangshan, Peterse et al., 2011,
 719 2014; 34°57'N, 113°22'E), Lantian (Gao et al., 2012; 34°12'N, 109°12'E), YB (Yuanbao, Jia
 720 et al., 2013; 103°09'N, 35°38'E), XF (Xifeng, 35°45'N, 107°49'E, Guo et al., 2009), YMG
 721 (Yimaguan, 35°55'N, 107°37'E, Hao et al., 2012), LC (Luochuan, 35°43'N, 109°25'E, Hao et
 722 al., 2012), Lingtai (35°04'N, 107°39'E, Sun et al., 2006), ZJC (Zhaojiachuan, 35°45'N,
 723 107°49'E, Sun et al., 2006), JY (Jingyuan, 36°21'N, 104°4'E, Sun et al., 2006) and west
 724 Kunlun loess site (37°0' N; 80°81'E, Zan, 2010). Chinese caves mentioned in the text include:
 725 Kesang cave (42°87' N; 81°75'E, Cheng et al., 2012) , Sanbao cave (110°26'E, 31°40'N,
 726 Wang et al., 2008), and Hulu Cave (32°30'N, 119°10'E, Cheng et al., 2009, 2016). Also
 727 shown is location of speleothem records from Borneo (4°N, 115°E, Meckler et al., 2012;
 728 Carolin et al., 2016). The location of western Pacific MD06-3047B core (17°00'N; 124°48'E,

729 Jia et al., 2015) and ODP806b (0°11'N, 159°13'E, Lea, 2000) was shown in subfigure d.

730

731 Fig.2



732

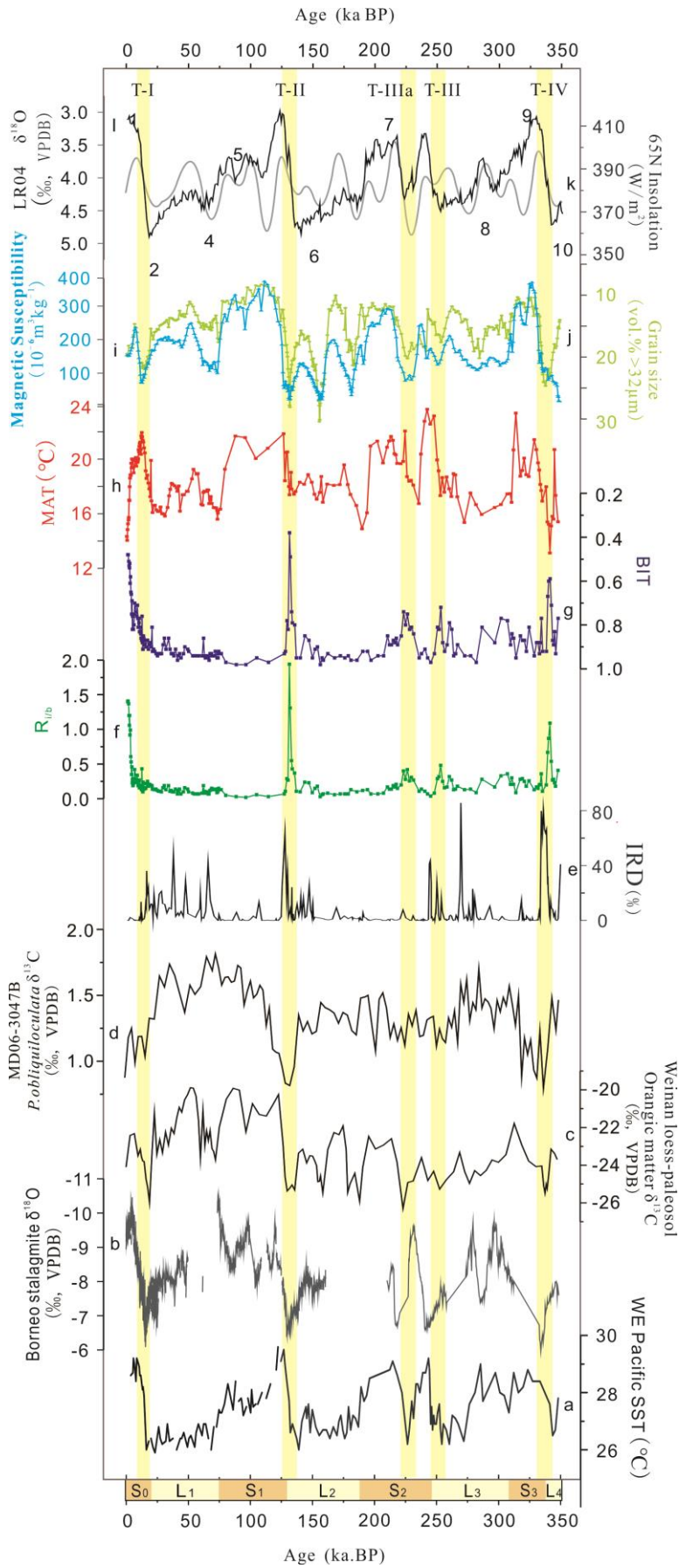
733 Fig. 2. The GDGTs distributions of four typical samples with completely different $R_{i/b}$ values

734 diagnostic of different dry conditions. The roman numerals denote the corresponding GDGT

735 components shown in supplemental data Fig. S1.

736

737 Fig.3



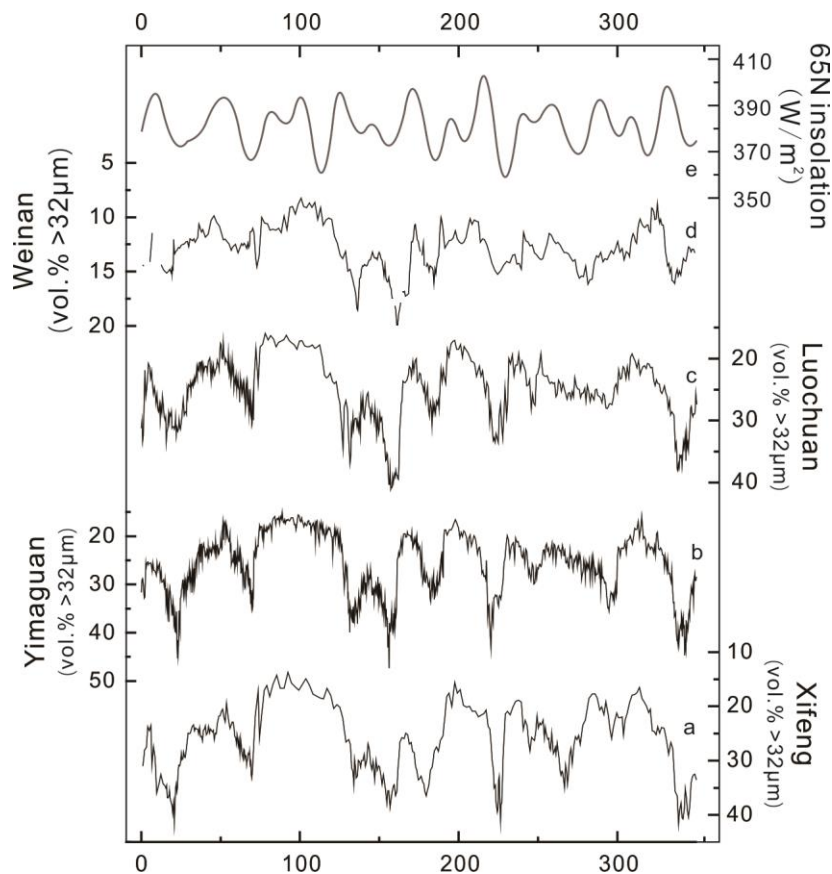
738

739 Fig. 3. Variations of GDGT parameters compared with other records. (a) marine sediment

740 Mg/Ca SST reconstructions from WEP site ODP 806b (Lea, 2000); (b) Speleothem $\delta^{18}\text{O}$
 741 records from Borneo (Meckler et al., 2012; Carolin et al., 2013,2016); (c) $\delta^{13}\text{C}$ of bulk soil
 742 organic matter of Weinan loess-paleosol (Sun et al., 2011); (d) $\delta^{13}\text{C}$ of *P. obliquiloculata* in
 743 western Pacific warm pool (Jia et al., 2015); (e) ice-rafted debris in North Atlantic (McManus
 744 et al., 1999); (f) $R_{i/b}$ and (g) BIT in Weinan, indicative of extreme drought events (this study);
 745 (h) annual mean atmospheric temperature (MAT) estimated by the MAT-mr calibration based
 746 on 5- and 6-methylated brGDGTs (this study, supplementary data, Table S1), (i) magnetic
 747 susceptibility, and (j) loess grain size (vol.% >32 μm) for the loess-paleosol sequences in
 748 Weinan section (this study); (k) benthic foraminifera $\delta^{18}\text{O}$ stack (Lisiecki and Raymo, 2005);
 749 and (l) the 65°N insolation (Berger et al., 2010). All the colored curves (f, g, h, i, j) are from
 750 Weinan section. The highlight yellow bars indicate the termination I, II, IIIa, III and IV
 751 denoted by T-I, T-II, T-IIIa, T-III, T-IV, respectively. The lithologic column shows the loess
 752 (light brown, L) and paleosol (dark brown, S) layers.

753

754 Fig.4



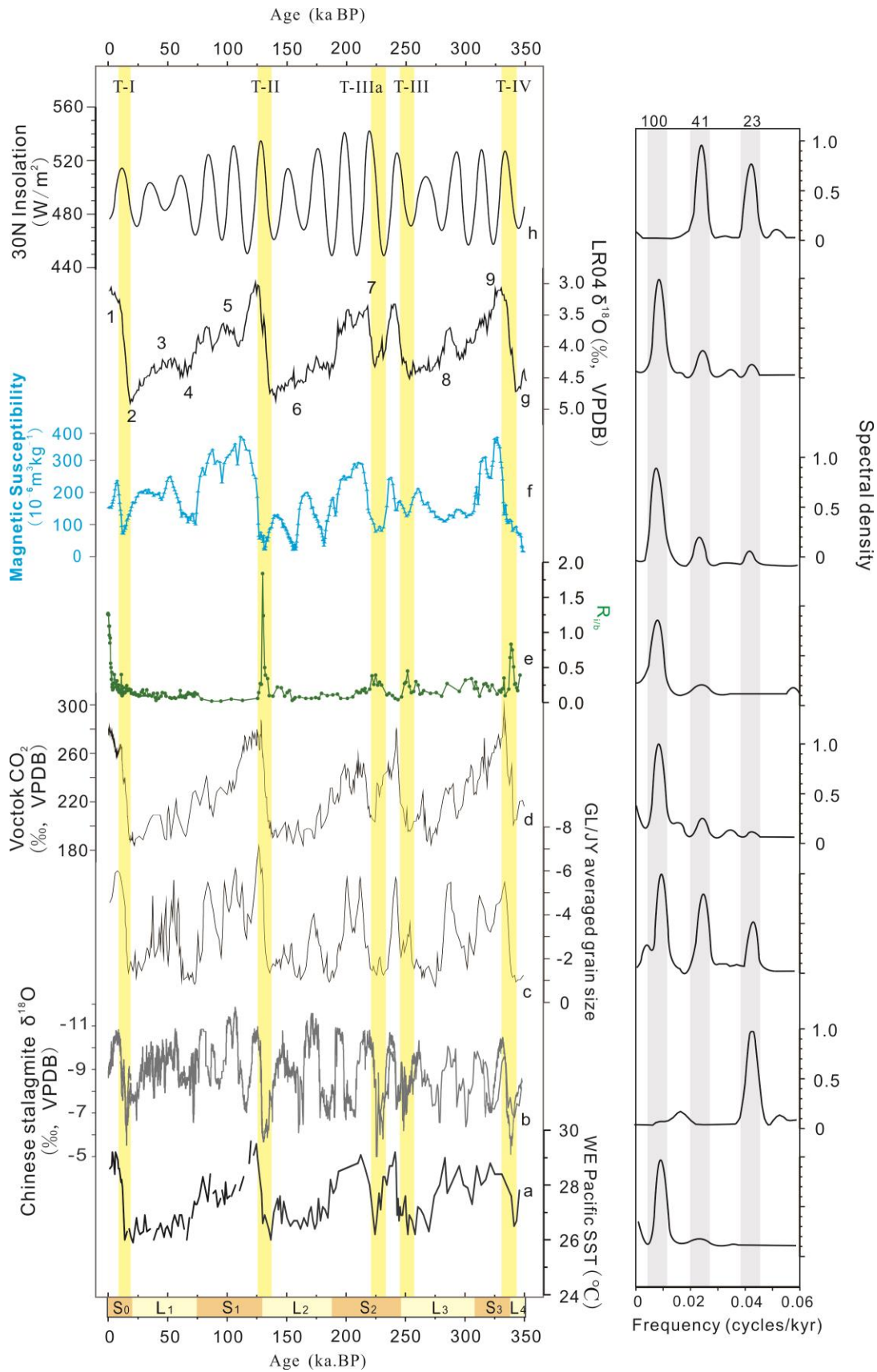
755

756 Fig. 4. Grain size (vol.% >32 μm) (a, b, c, d) variations in CLP along with the 65°N insolation

757 (e) (Berger et al., 2010). (a) Xifeng (35°45'N 107°49'E, Guo et al., 2009); (b) Yimaguan
758 (35°55' N; 107°37'E, Hao et al., 2012); (c) Luochuan (35°43' N; 109°25'E, Hao et al., 2012);
759 (d) Weinan (34°21' N; 109°32'E, this study).

760

761 Fig.5



762

763 Fig.5 Time series and spectral analysis results of monsoonal proxies. (a) speleothem $\delta^{18}\text{O}$

764 from the Hulu and Sanbao caves (Wang et al., 2008; Cheng et al., 2009); (b) averaged $\delta^{13}\text{C}_{\text{IC}}$

765 results of GL/JY sections; (c) CO₂ (Petit et al., 1999); (d) R_{i/b} and (e) magnetic susceptibility
766 from Weinan loess section (this study); (f) benthic δ¹⁸O stack (Lisiecki and Raymo, 2005)
767 and (g) summer insolation (Berger et al., 2010).

768