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Mid- to- late Holocene hydroclimatic changes on the Chinese Loess Plateau: evidence from n-alkanes from the sediments of Tianchi Lake --Manuscript Draft--

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Abstract:	the Chinese Loess Plateau using n-alkane We used Paq (the proportion of aquatic ma reflect changes in lake water level, with a h macrophytes indicating a lower water level hydrological reconstruction agrees with va (average chain length), CPI (carbon prefer molecular distribution of the sediments in T water level was relatively high during 5.7 to thereafter. Our paleohydrological reconstru- reconstructions from the Loess Plateau, w asynchronous with paleoclimatic records fr Holocene. Overall, our results confirm that EASM (East Asian summer monsoon) is a	higher abundance of submerged and vice versa. The Paq -based rious other lines of evidence, including ACL rence index), C/N ratio and the n-alkane Fianchi Lake. The results reveal that the lake to 3.2 ka BP, and decreased gradually action is consistent with existing paleoclimate hich suggest a humid mid-Holocene, but is rom central China which indicate an arid mid- the intensity of the rainfall delivered by the n important factor in affecting nd can be considered as further evidence for us "northern China drought and southern
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Response to Reviewers:	Dear Professor Whitmore, Thanks you so much for your corrected my manuscript. I accepted all the corrections and "accept all changes". Best regards! Sincerely, Aifeng

1		Mid- to- late Holocene hydroclimatic changes on the Chinese Loess
2 3 4 5		Plateau: evidence from <i>n</i> -alkanes from the sediments of Tianchi Lake
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49 50 51	16	
52 53	17	Key words
54 55 56	18	<i>n</i> -Alkanes $\cdot P_{aq} \cdot Lake$ level \cdot Mid-late Holocene \cdot Loess Plateau
57 58 59	19	
60 61 62		
63 64		
65		

20 Abstract

22	We have reconstructed the history of mid-late Holocene paleohydrological changes in
23	the Chinese Loess Plateau using <i>n</i> -alkane data from a sediment core in Tianchi Lake.
24	We used P_{aq} (the proportion of aquatic macrophytes to the total plant community) to
25	reflect changes in lake water level, with a higher abundance of submerged
26	macrophytes indicating a lower water level and vice versa. The P_{aq} -based
27	hydrological reconstruction agrees with various other lines of evidence, including
28	ACL (average chain length), CPI (carbon preference index), C/N ratio and the
29	<i>n</i> -alkane molecular distribution of the sediments in Tianchi Lake. The results reveal
30	that the lake water level was relatively high during 5.7 to 3.2 ka BP, and decreased
31	gradually thereafter. Our paleohydrological reconstruction is consistent with existing
32	paleoclimate reconstructions from the Loess Plateau, which suggest a humid
33	mid-Holocene, but is asynchronous with paleoclimatic records from central China
34	which indicate an arid mid-Holocene. Overall, our results confirm that the intensity of
35	the rainfall delivered by the EASM (East Asian summer monsoon) is an important
36	factor in affecting paleohydrological changes in the region and can be considered as
37	further evidence for the development of a spatially asynchronous "northern China
38	drought and southern China flood" precipitation pattern during the Holocene.
39	
40	
41	

42 Introduction

44	Climatic and environmental changes in the Chinese Loess Plateau are mainly
45	controlled by the EASM, which directly affects almost all aspects of the hydrology
46	and ecology of East Asia (Clift and Plumb 2008). An increase in EASM intensity
47	would be expected to result in a northward movement of the rainfall belt in China and
48	a corresponding rainfall increase in the Loess Plateau (Chen et al. 2008). Many
49	regional paleoclimatic records have been produced from this semi-arid, monsoon
50	marginal zone (Zhao et al. 2010; Dong et al. 2012; Liu and Feng 2012; Lu et al. 2013;
51	Qiang et al. 2013). However, regional high-resolution paleohydrological
52	reconstructions are extremely limited because proxies or archives that record ancient
53	hydrological conditions, with good age control, are scarce on the Loess Plateau. A
54	humid mid-Holocene has been proposed based on a pollen-based record (Chen et al.
55	2015a) and a hydrogen isotope reconstruction of long-chain <i>n</i> -alkanes (Rao et al.
56	2016) from Gonghai Lake, one of the few natural lakes on the Loess Plateau. Their
57	paleohydrological reconstruction is inconsistent with records from the core
58	monsoon-controlled regions of central China. It shows an arid interval from 7.0-3.0 ka
59	BP (Xie et al. 2013; Zhu et al. 2017). Therefore, more high-resolution lacustrine
60	reconstructions of hydroclimatic variations during the mid-late Holocene are needed
61	to explore the underlying mechanism of this asynchronous hydroclimatic variability.
62	Here, a high-resolution lacustrine record based on <i>n</i> -alkanes of sediments from
63	Tianchi Lake on the Loess Plateau will be discussed.

1 2	64	<i>n</i> -Alkanes
3 4	65	composition a
5 6 7	66	are widely pre
8 9 10	67	lacustrine sedi
11 12	68	the Average C
13 14 15	69	Index (CPI) (N
16 17 18	70	indices, have
19 20 21	71	He et al. 2014
22 23	72	dominated by
24 25 26	73	al. 2011), whi
27 28 29	74	C ₂₅ <i>n</i> -alkanes
30 31 32	75	bacteria (Cran
33 34	76	considered to
35 36 37	77	from an increa
38 39 40	78	terrestrial plar
41 42 43	79	related to the
44 45	80	and lake level
46 47 48	81	distribution ar
49 50 51	82	1986; Hudon
52 53 54	83	and Liu et al.
55 56	84	produced by s
57 58 59	85	understand the
60 61		
62 63		
64 65		

1	<i>n</i> -Alkanes preserved in lake sediments can be used to infer variations in the
5	composition and origin of organic inputs to the lacustrine environment, because they
3	are widely preserved in various environmental contexts, such as plants, soils and
7	lacustrine sediments, and can resist degradation actions (Meyers 1997). In particular
3	the Average Chain Length (ACL) (Poynter and Eglinton 1990), Carbon Preference
)	Index (CPI) (Meyers and Ishiwatari 1993), and P_{aq} (Ficken et al. 2000) <i>n</i> -alkane
)	indices, have been widely used in paleoenvironmental research (Nichols et al. 2006;
Ĺ	He et al. 2014). In general, terrestrial plants and emergent macrophytes are typically
2	dominated by the long-chain length homologues (C_{27} - C_{33}) (Ficken et al. 2000; Gao et
}	al. 2011), while submerged and floating-leaved macrophytes mainly produce C_{23} and
1	C_{25} <i>n</i> -alkanes (Ficken et al. 2000), and short chain ones are produced by algae and
5	bacteria (Cranwell et al. 1987). Consequently, higher ACL and CPI are commonly
3	considered to be predominantly produced by terrestrial plants. A higher P_{aq} may result
7	from an increase in submerged macrophytes in combination with a recession of the
3	terrestrial plants around the lake. Moreover, the biomass of submerged macrophytes is
)	related to the variation of the water table (Wagner and Falter 2002; Liu et al. 2015),
)	and lake level fluctuations have the potential to simultaneously constrain the spatial
L	distribution and the biomass of submerged macrophytes in a lake (Duarte and Kalf
2	1986; Hudon 1997; Middelboe and Markager 1997). Howerver, Aichner et al. (2010)
3	and Liu et al. (2015) found that higher amounts of long chain <i>n</i> -alkanes can be
1	produced by submerged macrophytes in several lakes. Therefore, it is necessary to
5	understand the extent to which long chain <i>n</i> -alkanes in lacustrine sediments are

86 influenced by terrestrial plants and submerged macrophytes in a study lake when
87 reconstructing the paleoenvironments.

In this study, we first define the potential sources and the contributions from the various plants (e.g. terrigenous plants vs. submerged macrophytes) in Tianchi Lake. Second, we give an interpretation of the proxies (Paq, ACL, CPI of *n*-alkanes, and C/N), especially P_{aq} as an effective indicator of lake level changes in Tianchi Lake. Additionally, we seek to compare regional climate reconstructions with those from Tianchi Lake and other nearby sites to confirm a spatially asynchronous hydroclimatic variability occurred in China during the Holocene. Study site Tianchi Lake (lat. 35°15'55"N, long. 106°18'43"E, elevation 2430 m a.s.l.) is a small freshwater alpine lake located in the Liupan Mountains, southwestern Loess Plateau, northwest China (Fig. 1a). The length of the lake from east to west is 250 m and the width is 120 m. The maximum water depth is 8.2 m, and the lake covers an area of 2×10^4 m² (Fig. 1b). The lake receives no surface run off, and it is fed by meteoric water and groundwater recharge. There is no apparent surface outflow, except for a possible transient outflow in the western part of the lake basin, which is possibly active during the rainy season. The mean annual temperature is 8.2 °C and mean annual precipitation is 677 mm based on data from the nearest meteorological station (Liupan Mountain station, at 2845 m a.s.l.). Most of the precipitation occurs as

108	rainfall during summer, accounting for nearly 72.2% of the annual total. The
109	vegetation of the upland slopes of the lake is dominated by shrubs and steppe. Grassy
110	steppe with sparse shrub covers the north slopes, and shrubs dominate the south
111	slopes (Zhao et al. 2010). Emergent (Phragmites australis (Cav.) Trin. ex Steud) and
112	submerged (Potamogeton sp. and Chara sp.) macrophytes are widely distributed in
113	the shallow areas of the lake (Fig. 1d), but floating-leaved macrophytes are absent,
114	based on our field observations in 2010.
115	
116	Materials and methods
117	
118	Field sampling
119	
120	Two parallel sediment cores of lengths 11.2 m (GSA07-1) and 10.4 m (GSB07-1)
121	were collected using a UWITEC piston corer system (6 cm in diameter) from the lake
122	center in 2007 (Fig. 1b). The lithology of core GSB07-1 consisted of alternating
123	brown-colored sandy clay and grey-brownish clay between 1040-746 cm, and
124	grey-brownish clay above 746 cm. The sediments are characterized by 1 to
125	2-mm-thick organic detritus-rich laminations (Fig. 1c), which yielded abundant
126	terrestrial macrofossils for radiocarbon dating. Fifty-six down-core sedimentary
127	samples were taken at 15-cm intervals throughout core GSB07-1. Additionally, we
128	collected six surface soil samples, three surface lake sediments, nine dominant
129	terrestrial plant samples (Cedrus sp., Larix sp., Abies sp., Betula sp., Rosa sp., Rubus

sp., Salix sp., Berberis sp. and Artemisia sp.), which surround the lake, and one emergent macrophyte (P. australis) and two submerged macrophytes (Potamogeton and Chara) within the lake for modern process study. All the above samples were carried out for TOC, TN analyses, and lipid extraction. Laboratory analyses Samples for TOC and TN measurements were pretreated with 10 ml of 10% HCl to remove carbonates, washed with distilled water until the pH was neutral, and then measured using a CE Model 440 Elemental Analyzer. The C/N ratio was derived from the ratio of TOC and TN. n-Alkanes were extracted based on methods described previously (Kawamura et al. 2003) in G-MOL lab of the University of Glasgow. Briefly, 2-10 g of freeze-dried, homogenized sediment were transferred to a test tube and hydrolyzed with 15 ml of 0.3 M KOH dissolved in 95:5 methanol/dichloromethane-extracted water. The samples were then hydrolyzed and centrifuged and the supernatant and pipetted into a round-bottomed flask. The sediment was then extracted three times with 10 ml dichloromethane/methanol (3:1) using ultrasonication. The extracts were combined and concentrated, using a rotary evaporator, under vacuum and then separated into neutral and acidic fractions using the methods of Kawamura (1995). The neutral fraction was further separated using silica gel column chromatography to get *n*-alkane fraction. Dried *n*-alkane fraction was redissolved in hexane and analyzed using a gas chromatograph (GC; Shimadzu

152	2010) with a flame ionization detector (FID) and hydrogen as carrier gas at constant	
153	pressure (190 kPa). Separation of the different compounds was achieved using an	
154	identical column (length: 60 m, diameter: 0.25 mm, film thickness: 0.25 μ m, coating:	
155	100 % dimethyl-polysiloxane). The gas chromatograph temperature program was set	
156	to increase from 50 -120 °C at 30 °C min ⁻¹ , then 120 -310 °C at 5 °C min ⁻¹ , with a	
157	final isothermal time of 20 min at 300 °C. Compound identification was confirmed by	
158	GC/MS (Shimadzu OP2010-Plus Mass Spectrometer (MS) interfaced with a	
159	Shimadzu 2010 GC) based on retention times and mass spectra.	
160	The <i>n</i> -alkane proxies (equation (1) from Poynter and Eglinton (1990); equation (2))
161	from Marzi et al. (1993); and equation (3) from Ficken et al. (2000) were calculated	
162	as follows:	
163	$ACL = (19*C_{19}+20*C_{20}+21*C_{21}+\dots+33*C_{33})/(C_{19}+C_{20}+C_{21}+\dots+C_{33}) $ (1)	
164	$CPI = \frac{7}{8} (C_{19} + C_{21} + C_{23} + \dots + C_{33}) / (C_{20} + C_{22} + C_{24} + \dots + C_{32}) $ (2)	
165	$P_{aq} = (C_{23} + C_{25})/(C_{23} + C_{25} + C_{29} + C_{31}) $ (3)	
166	where C_i is the concentration of <i>n</i> -alkane of i number of carbon.	
167		
168	Age model	
169		
170	The chronology of core GSA07-1 used in this study mainly consists of 19 dates from	
171	Zhao et al. (2010) and 6 new dates (Table 1). All 14 C dates were measured in the AMS	
172	Dating Laboratory of Beijing University and are based on the leaves of terrestrial	
173	plants. The ages were calibrated to calendar years before present (AD 1950) using the	

174	program CALIB Rev. 5.0.1 with the IntCal04 calibration data set (Reimer et al. 2004).
175	The depths of characteristic laminations in cores GSA07-1 and core GSB07-1 are
176	consistent. Therefore, the chronology of core GSB07-1 was calibrated based on the
177	corresponding depths in GSA07-1 (Table 1). The chronology indicates that the age of
178	core GSB07-1 spans the past 5720 years (Fig. 2). The average accumulation rate
179	based on the age-depth model is about 1.85 mm a ⁻¹ .
180	
181	Results
182	
183 184 185	<i>n</i> -Alkane distributions and P_{aq} variations in modern vegetation
186	The P_{aq} index has been proposed as an indicator of the relative contributions of
187	<i>n</i> -alkanes from submerged/floating aquatic plants versus those from emergent and
188	terrestrial plants in the lake. Generally, $P_{aq} < 0.1$ corresponds to terrestrial plants,
189	0.1-0.4 to emergent macrophytes, and 0.4-1.0 to submerged/floating macrophytes
190	(Ficken et al. 2000). In this study, average P_{aq} values and <i>n</i> -alkane molecular
191	distribution patterns vary considerably in the three types of plant material (terrestrial,
192	and emergent and submerged macrophytes: Fig. 3a-c). Terrestrial plants (Fig. 3a),
193	which have a lower average P_{aq} value (0.18), are dominated by the <i>n</i> -C ₃₁ homologue.
194	Emergent macrophytes (Fig. 3b) growing in the near-shore environment are mainly
195	dominated by the <i>n</i> -C ₂₇ homologue and have a higher P_{aq} value (0.65). In contrast,
196	n-C ₂₃ is the dominant homologue in the submerged macrophytes (Fig. 3c) with a
197	secondary peak at <i>n</i> -C ₂₅ . The average P_{aq} value of submerged macrophytes is 0.93. In

198	addition, a bimodal <i>n</i> -alkane distribution pattern with high abundances at n -C ₂₃ and
199	<i>n</i> -C ₃₁ is observed in the surface sediments of Tianchi Lake which have an average P_{aq}
200	value of 0.51 (Fig. 3d), indicating a specific mixture of inputs from terrestrial plants
201	and submerged macrophytes. The distribution pattern for the surface soil has an
202	overwhelming preponderance of the n -C ₃₁ homologue, and the average P _{aq} value of
203	the surface soils is 0.25 (Fig. 3e).
204	
205	<i>n</i> -Alkane proxies and C/N ratios in the down-core sediments
206	
207	Time series of the various sedimentary parameters are illustrated in Fig. 4. The
208	records span the last 5.7 ka BP. P_{aq} (Fig. 4a) ranges from 0.32 to 0.78 with a mean of
209	0.56. The average P_{aq} value is 0.46 during 5.7-3.2 ka BP, and 0.65 from 3.2 ka BP to
210	the present. It is noteworthy that prior 3.2 ka BP most of the P_{aq} values are less than
211	0.52, while subsequently they are greater than 0.52. ACL ranges from 25 to 29 with a
212	mean of 27.4 (Fig. 4b). The CPI values range from 1.7 to 8.8 with a mean of 5.2 over
213	the last 5.7 ka BP (Fig. 4c). The C/N ratios (Fig. 4d) range from 9.5 to 26 with a mean
214	of 15. The ACL, CPI, and C/N ratios exhibit similar patterns of variation, and they all
215	exhibit an obvious shift at 3.2 ka BP, as do the P_{aq} values. The threshold values of
216	ACL, CPI, and C/N ratios are almost the same as their average values.
217	
218	Discussion
219	

222	The organic component of lake sediments represents a pool of organic matter derived
223	from the decomposing detritus of aquatic plants growing in the littoral and marginal
224	zone of the lake and from terrestrial plants growing in the catchment (Meyers and
225	Ishiwatari 1993; Meyers 1997). Lacustrine sediment <i>n</i> -alkanes often have multiple
226	sources, including terrestrial plants, aquatic macrophytes and lower organisms.
227	Generally, <i>n</i> -alkane distributions of terrestrial and emergent plants tend to exhibit
228	high proportions of the n -C ₃₁ homologue (Rielley et al. 1991; Ficken et al. 2000;
229	Sachse et al. 2006), whereas those of submerged and floating plants are generally
230	dominated by n -C ₂₃ and n -C ₂₅ homologues (Ficken et al. 2000; Gao et al. 2011; Seki
231	et al. 2012). Therefore, <i>n</i> -alkanes can be used to identify local and regional sources of
232	organic matter. However, recent studies have indicated that aquatic plants also make a
233	large contribution to the long chain <i>n</i> -alkanes in lake sediments (Aichner et al. 2010;
234	Liu et al. 2015; Liu and Liu 2016). For example, Liu et al. (2015) found that the long
235	chain <i>n</i> -alkanes produced by submerged plants in Qinghai Lake had a significant
236	influence on n -C ₂₇ and n -C ₂₉ alkanes in sediments. Even for the same submerged
237	plant (Potamogeton sp.) from 16 Tibetan Plateau lakes, the distribution patterns of all
238	
	the <i>n</i> -alkane homologs show obvious differences (Liu and Liu 2016). It is thus
239	the <i>n</i> -alkane homologs show obvious differences (Liu and Liu 2016). It is thus necessary to make a distinction between the various sources that contribute to the
239 240	

Sources of organic matter to the lake

242	<i>n</i> -alkanes (<i>n</i> -C ₂₃ and <i>n</i> -C ₂₅) (Fig. 3c). The average P_{aq} value is as high as 0.93. There
243	is no evidence that they exhibit relatively high abundance of long chain <i>n</i> -alkanes as
244	Liu and Liu (2016) described. The unimodal distribution pattern with the maxima at
245	n-C ₃₁ alkanes and relatively low P _{aq} values of modern terrestrial plants (Fig. 3a) and
246	surface soils (Fig. 3e) in Tianchi Lake, suggest again that $n-C_{31}$ alkanes can be traced
247	to terrestrial plant inputs and not to lake macrophytes. In addition, a bimodal
248	molecular distribution pattern with major peaks at the n -C ₂₃ and n -C ₃₁ homologues in
249	the surface lake sediments (Fig. 3d) probably represents a combination of inputs from
250	submerged macrophytes (Fig. 3c) and terrestrial plants (Fig. 3a)/emergent (Fig. 3b).
251	Our observations are consistent with those of previous studies (Cranwell 1984; Ficken
252	et al. 2000; Gao et al. 2011; Street et al. 2013), which indicate that P_{aq} can be used to
253	reflect the contribution from submerged macrophytes.
254	
255	Interpretation of <i>n</i> -alkane indices
256	
257	It has been demonstrated that the abundance of submerged macrophytes in lake is
258	affected by irradiance and the littoral slope (Hudon 1997; Hudon et al. 2000;
259	Cheruvelil and Soranno 2008). Thus, lake level has the potential to constrain the
260	spatial distribution of submerged macrophytes via both a reduction in light intensity
261	(Duarte and Kalf 1986; Middelboe and Markager 1997) and a change in the spatial
262	extent of the littoral habitat (Hudon 1997). Hence, changes in the relative inputs of
263	submerged macrophytes can potentially be ascribed to fluctuations in lake level. Most

264	of the submerged macrophytes in Tianchi Lake are distributed in a shallow area close
265	to the shoreline and very few floating macrophytes can be observed (Fig. 1d).
266	Potamogeton and Chara are the two dominant submerged species, which grow in a
267	narrow zone down to a depth of ~1.2 m. A bathymetric survey of Tianchi Lake (Fig.
268	1b) reveals that the shoreline forms a narrow shelf from a depth of 0.5 m down to 2.8
269	m, followed by a steep slope that causes a decrease in the occurrence of submerged
270	macrophytes. Assuming that the basic bathymetry of the basin has remained similar
271	through time, the reductions in lake level would result in a relatively larger shelf area,
272	which would produce an expansion of the shallow-water habitat for submerged
273	macrophytes. Accordingly, the lower P_{aq} values in Tianchi Lake could be interpreted
274	as reflecting less abundance of submerged macrophytes and the raising of lake level.
275	On the other hand, the contribution from terrestrial plants can also exert an influence
276	on P_{aq} values since the P_{aq} index is a proxy for evaluating the contribution of
277	<i>n</i> -alkanes from submerged/floating aquatic plants relative to emergent and terrestrial
278	plants (Ficken et al. 2000). During intervals of high rainfall and lake level, an
279	increased contribution of terrestrial plant material delivered by increased catchment
280	rain and runoff could lower P_{aq} values, and vice versa. This coincides with the
281	findings of Liu and Liu (2016), which indicated a negative relationship between the
282	P_{aq} value of surface lake sediments and the water level of Qinghai Lake.
283	As an important parameter of <i>n</i> -alkanes, the climate implications of ACL have
284	been discussed a lot in the literature, but there is no unified agreement as to their
285	interpretation because ACL often appears highly specific to regional or local

286	conditions (Ling et al. 2017). Furthermore, ACL can not be used to reconstruct
287	temperature or precipitation change if the plant species or sedimentary environment in
288	the catchment area underwent considerable change (in parallel with or forced by
289	climatic variation) (Pu et al. 2010). CPI is another <i>n</i> -alkane index, which has been
290	widely accepted as an indicator for terrestrial sources of sedimentary organic matter.
291	Terrestrial plants have abundant long-chain <i>n</i> -alkanes, and show distinct odd-even
292	predominance, thus their CPI is always greater than 5. On the contrary, CPI values of
293	the aquatic plants and planktonic bacteria are considerably lower than those usually
294	reported for terrestrial plant sourced <i>n</i> -alkanes (Cranwell 1987).
295	
296	Reconstruction of the lake-level evolution
297	
297 298	<i>n</i> -Alkane based records and C/N ratios from Tianchi Lake are presented in Fig. 4.
	<i>n</i> -Alkane based records and C/N ratios from Tianchi Lake are presented in Fig. 4. Overall, there is an obvious shift at ~3.2 ka BP among the various proxies. During
298	
298 299	Overall, there is an obvious shift at ~3.2 ka BP among the various proxies. During
298 299 300	Overall, there is an obvious shift at ~3.2 ka BP among the various proxies. During 5.7-3.2 ka BP, the ACL (Fig. 4b) and CPI (Fig. 4c) proxies show relatively high
298 299 300 301	Overall, there is an obvious shift at ~3.2 ka BP among the various proxies. During 5.7-3.2 ka BP, the ACL (Fig. 4b) and CPI (Fig. 4c) proxies show relatively high values in the core. CPI values almost greater than 5 and ACL values range from 27 to
 298 299 300 301 302 	Overall, there is an obvious shift at ~3.2 ka BP among the various proxies. During 5.7-3.2 ka BP, the ACL (Fig. 4b) and CPI (Fig. 4c) proxies show relatively high values in the core. CPI values almost greater than 5 and ACL values range from 27 to 29, likely indicate a predominance of terrestrial plant inputs to the lake basin. The
 298 299 300 301 302 303 	Overall, there is an obvious shift at ~3.2 ka BP among the various proxies. During 5.7-3.2 ka BP, the ACL (Fig. 4b) and CPI (Fig. 4c) proxies show relatively high values in the core. CPI values almost greater than 5 and ACL values range from 27 to 29, likely indicate a predominance of terrestrial plant inputs to the lake basin. The results are supported by the higher C/N ratios (mean >15, with occasional values up to
 298 299 300 301 302 303 304 	Overall, there is an obvious shift at ~3.2 ka BP among the various proxies. During 5.7-3.2 ka BP, the ACL (Fig. 4b) and CPI (Fig. 4c) proxies show relatively high values in the core. CPI values almost greater than 5 and ACL values range from 27 to 29, likely indicate a predominance of terrestrial plant inputs to the lake basin. The results are supported by the higher C/N ratios (mean >15, with occasional values up to 26) (Fig. 4d) in this phase since the C/N ratios from terrestrial plants and emergent

308	Tianchi Lake. Based on interpretations discussed above, less abundance of submerged
309	macrophytes input and more abundance of terrestrial plants input to the sediments are
310	likely a response to relatively high lake levels during this phase in Tianchi Lake.
311	We interpret the increase in average P_{aq} values (0.51-0.78) and a decrease in ACL
312	(25-27) and CPI (1.6-5.4) after 3.2 ka BP (Fig. 4a) as corresponding to an increase in
313	the proportion of submerged and floating-leaved macrophytes, and a decrease in
314	terrestrial inputs. Furthermore, the C/N ratios are generally low (<15) during this
315	interval. In view of the absence of floating-leaved macrophytes in Tianchi Lake,
316	based on our field observations, the increasing contribution from submerged
317	macrophytes accordingly indicates a gradually falling lake level from 3.2 ka BP.
318	The variations in lake level inferred by the <i>n</i> -alkanes record is also supported by a
319	shift in pollen assemblages from Tianchi Lake, which indicate that closed canopy
320	forest was replaced by an open landscape at around 3.0 ka BP (Zhao et al. 2010).
321	Another high-resolution pollen record from the Dadiwan peatland (Fig. 1a), 50 km
322	southwest of Tianchi Lake on the Loess Plateau, also reveals a significant decrease in
323	tree pollen frequencies at around 3.0 ka BP (An et al. 2003).
324	
325	Asynchronous hydroclimatic variability
326	
327	The P_{aq} record from Tianchi Lake reveals a transition from higher lake levels to lower
328	lake levels after 3.2 ka BP, and thus wetter conditions during 5.7-3.2 ka BP and drier
329	conditions after 3.2 ka BP (Fig. 5d). This accords with other paleoclimatic records

1 2	330	from the nearby C
3 4	331	2015; Rao et al. 20
5 6 7	332	Tianchi Lake sugg
8 9 10	333	et al. 2015a). Ano
11 12 13	334	Gonghai Lake (Fi
14 15	335	A recent study of
16 17 18	336	(Wang et al. 2014
19 20 21	337	Plateau. In additio
22 23	338	similar pattern of
24 25 26	339	al. 2013). This evi
27 28 29	340	widespread pheno
30 31 32	341	in accord with the
33 34	342	paleohydrological
35 36 37	343	middle reaches of
38 39 40	344	previous paleoclir
41 42 43	345	in Dajiuhu peatlar
44 45 46	346	relatively wet con
47 48	347	al. 2015). Another
49 50 51	348	grain-size and <i>n</i> -a
52 53 54	349	the Yangtze River
55 56 57	350	and a humid interv
58 59	351	5a) and the flux of
60 61		
62 63		
64 65		

330	from the nearby Chinese Loess Plateau (Lu et al. 2013; Chen et al. 2015a; Liu et al.
331	2015; Rao et al. 2016). The pollen-based annual precipitation reconstruction from
332	Tianchi Lake suggests a rapid precipitation decrease since ~3.3 ka BP (Fig. 5e; Chen
333	et al. 2015a). Another pollen-based annual precipitation reconstruction from nearby
334	Gonghai Lake (Fig. 5f) reveals a humid interval around 8-3 ka BP (Chen et al. 2015a).
335	A recent study of palaeosol development as an indicator of the strength of the EASM
336	(Wang et al. 2014) suggests a wet interval during 8.6-3.2 ka BP in the Chinese Loess
337	Plateau. In addition, a TOC record from the Dadiwan peat profile also revealed a
338	similar pattern of wet and dry episodes as at Tianchi Lake (Zhou et al. 1996; Huang et
339	al. 2013). This evidence supports the contention that a moist climate was a
840	widespread phenomenon on the Chinese Loess Plateau during the mid-Holocene. It is
841	in accord with the gradually decreasing solar insolation (Fig. 5g). However, the
342	paleohydrological conditions reconstructed from Dajiuhu peatland (Fig. 5c) in the
843	middle reaches of the Yangtze River of central China are in contrast with these
844	previous paleoclimatic records. Changes in the aerobic bacteria-derived hopanoid flux
345	in Dajiuhu peatland (Fig. 5c) imply relatively arid conditions from 7.0-3.0 ka BP and
846	relatively wet conditions from 3.0-1.0 ka BP (Xie et al. 2013; Huang et al. 2013; He et
847	al. 2015). Another late-Holocene paleohydrological reconstruction based on sediment
848	grain-size and <i>n</i> -alkane data from Longgan Lake in the middle and lower reaches of
849	the Yangtze River (Xue et al. 2017), indicated drought conditions from 4 to 2.7 ka BP
850	and a humid interval from 2.7 to 1.2 ka BP. In addition, the studies on the $\delta^{18}O$ (Fig.
851	5a) and the flux of soil-derived magnetic minerals preserved (Fig. 5b) in stalagmite

1 2	352	HS4 from Heshang cave in central China also revealed a relatively arid interval from
3 4	353	6.7 to 3.4 ka BP (Hu et al. 2008; Zhu et al. 2017). Therefore, it seems that mid-late
5 6 7	354	Holocene paleohydrological evolution was asynchronous in the middle reaches of the
8 9 10	355	Yangtze River of central China and in the Yellow River region of north China.
11 12 13	356	Tianchi Lake is located in the 'far-field' northwestern marginal region of the
14 15	357	EASM, whereas the Dajiuhu peatland (Fig. 1a) is located in the 'core' monsoonal area
16 17 18	358	of the EASM (Qian et al. 2007). Summer rainfall is the predominant contributor to the
19 20 21	359	annual precipitation at both sites (Gao and Xie 2014). The northwards advance of the
22 23 24	360	rainfall front resulting from an enhanced EASM intensity could result in increased
25 26	361	precipitation in the marginal region of the EASM but decreased precipitation in the
27 28 29	362	core monsoonal area of EASM (Ding et al. 2008; Rao et al. 2016). The occurrence of
30 31 32	363	this contrasting spatial pattern of moisture conditions, with more frequent droughts in
33 34 35	364	north China and more frequent floods in the mid-low Yangtze River valley during
36 37	365	summer, has also been observed during the last few decades (Gemmer et al. 2004;
38 39 40	366	Qian and Lin 2005; Zhai et al. 2005). It has been designated the "northern China
41 42 43	367	drought and southern China flood" precipitation pattern (Zhou et al. 2009), and is also
44 45 46	368	evident on millennial and centennial time scales (Chen et al. 2015b).
47 48	369	Previous workers have analyzed the main factors responsible for the
49 50 51	370	asynchronous pattern of hydroclimatic variability between the marginal and core
52 53 54	371	monsoonal area of EASM in China. For example, He et al. (2014) suggested that
55 56 57	372	terrestrial temperature-induced evaporation changes and the extent of the Asian
58 59 60 61 62 63 64	373	monsoonal front could potentially explain the out-of-phase pattern of hydrological
65		

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errestrial temperature-induced evaporation changes and the extent of the Asian	

374	changes during the mid-Holocene. Chen et al. (2015a) emphasized that insolation
375	forcing, especially the tropical ocean conditions might be responsible for the abrupt
376	decline at 3.3 ka. Chen et al. (2015b) concluded that ENSO is one of the most
377	important factors affecting the precipitation of monsoonal northern and central China
378	on the centennial scale. Rao et al. (2016) emphasized the important influence of the
379	west-east thermal gradient in the equatorial Pacific on the climate of monsoonal China.
380	Zhu et al. (2017) concluded that a mid-Holocene reduction in ENSO intensity was
381	related to a decrease in storm frequency in the middle reaches of Yangtze River
382	between 6.7 and 3.4 ka BP. Finally, it is likely that the sea surface temperature (SST)
383	anomaly in the equatorial Pacific during the mid-Holocene probably played a key role
384	in facilitating the influence of ENSO on the asynchronous pattern of precipitation in
385	the marginal and core monsoonal area of the EASM in China.

387 Conclusions

We have used the record of *n*-alkanes extracted from a lacustrine sediment core from Tianchi Lake on the Chinese Loess Plateau to reconstruct lake-level variations during the past 5.7 ka BP. P_{aq} values and C/N ratios through the sequence in general exhibit a gradually increasing trend through the past 5.7 ka BP, indicating an increasing (and more variable) abundance of submerged macrophytes in response to a falling lake level. Terrestrial plants dominated the record before 3.2 ka BP, and subsequently there was a shift to the dominance of submerged macrophytes. The predominance of

396	terrestrial plants agree with higher ACL, higher CPI, and lower P_{aq} values from
397	5.7-3.2 ka BP, whereas the dominance of submerged macrophytes resulted in lower
398	ACL, lower CPI, and higher P_{aq} values after 3.2 ka BP. These changes indicate a
399	relatively humid interval during 5.7-3.2 ka BP and a drier but more variable interval
400	after 3.2 ka BP on the Chinese Loess Plateau. These findings are consistent with
401	previous paleoclimatic reconstructions for the Loess Plateau which indicate a humid
402	mid-Holocene. However, they are in disagreement with paleoclimatic records from
403	central China, which indicate an arid mid-Holocene. Overall, this spatial pattern
404	indicates that an enhanced intensity of monsoon rainfall delivered by the EASM
405	during the mid-Holocene was an important factor in affecting paleohydrological
406	changes in the region.
407	
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 409 410 411 412 413 414 415 	We thank Dr.Christopher Gallacher and Dr. Heiko Moossen for their training and help with laboratory analyses. This research was supported by grants from the National Science Foundation of China (NSFC Grants 41761044 and 41771208). We thank the China Scholarship Council (CSC) for funding a 20-month visit (File no. 2009618032) by Huiling Sun to work with Dr. James Bendle (now at the University of Birmingham) as a joint Ph.D. student (Lanzhou-Glasgow) at the G-MOL

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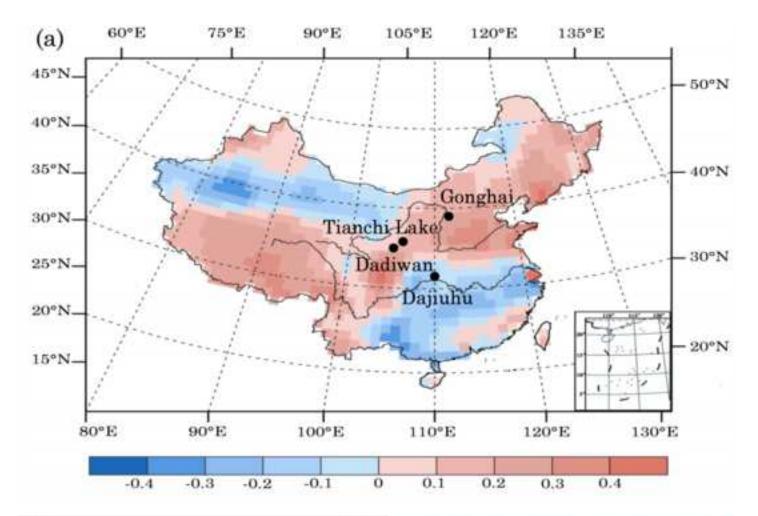
573	Fig. 1. (a) Location of Tianchi Lake in North China. Solid dots represent the study
574	area and other study sites referenced in the text. The map shows the correlation
575	coefficients between summer precipitation in China and summer monsoon intensity
576	from 1951-2000 (Wang et al. 2008), (b) schematic representation of the bathymetry of
577	Tianchi Lake (depths in m), (c) laminated structure of the sediment cores from
578	Tianchi Lake, (d) photo of submerged macrophytes in the shallow area of Tianchi
579	Lake
580	
581	Fig. 2. Age-depth model for core GSB07-1 from Tianchi Lake
582	
583	Fig. 3. Histogram of the molecular distributions of <i>n</i> -alkanes from (a) modern
584	terrestrial plants, (b) modern emergent macrophytes, (c) modern submerged
585	macrophytes, (d) surface lake sediments and (e) surface soils from around Tianchi
586	Lake. Only odd carbon number distributions are shown for the <i>n</i> -alkanes
587	
588	Fig. 4. Time series of sedimentary parameters for core GSB07-1 from Tianchi Lake
589	over the past 5.7 ka BP. (a) P_{aq} values based-on <i>n</i> -alkanes, (b) <i>n</i> -alkane ACL, (c)
590	<i>n</i> -alkane CPI, (d) C/N ratios
591	
592	Fig. 5. Comparison of regional paleohydrological records. (a) Heshang cave
593	speleothem δ^{18} O records (Hu et al. 2008), (b) the flux of soil-derived magnetic
594	minerals (IRM _{soft-flux}) preserved in stalagmite HS4 (Zhu et al. 2017), (c) hopanoids

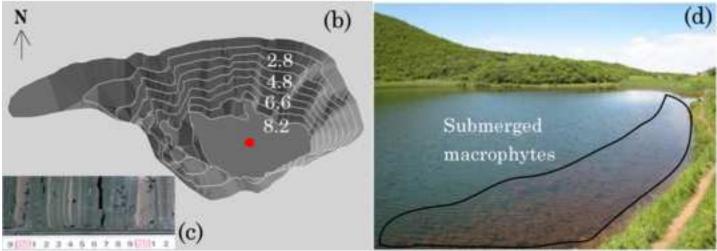
595	flux in Dajiuhu peatland (Xie et al. 2013), (d) P_{aq} values based-on <i>n</i> -alkanes in
596	Tianchi Lake, (e) pollen-based reconstruction of mean annual precipitation (MAP)
597	from Tianchi Lake (Chen et al. 2015a), (f) pollen-based reconstruction of mean annual
598	precipitation (MAP) from Gonghai Lake (Chen et al. 2015a), (g) July Insolation at
599	30 °N (Berger and Loutre 1991)
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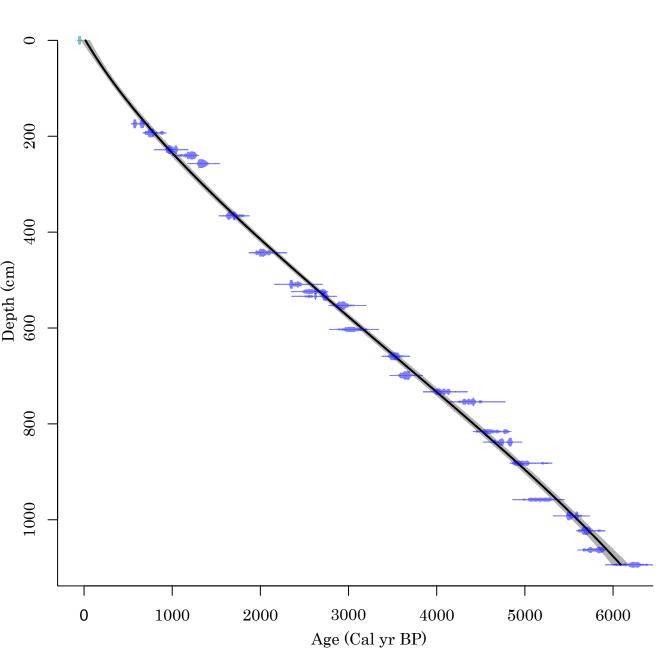
Table 1. AMS radiocarbon dates of core GSA07-1 in Tianchi Lake and the chronology

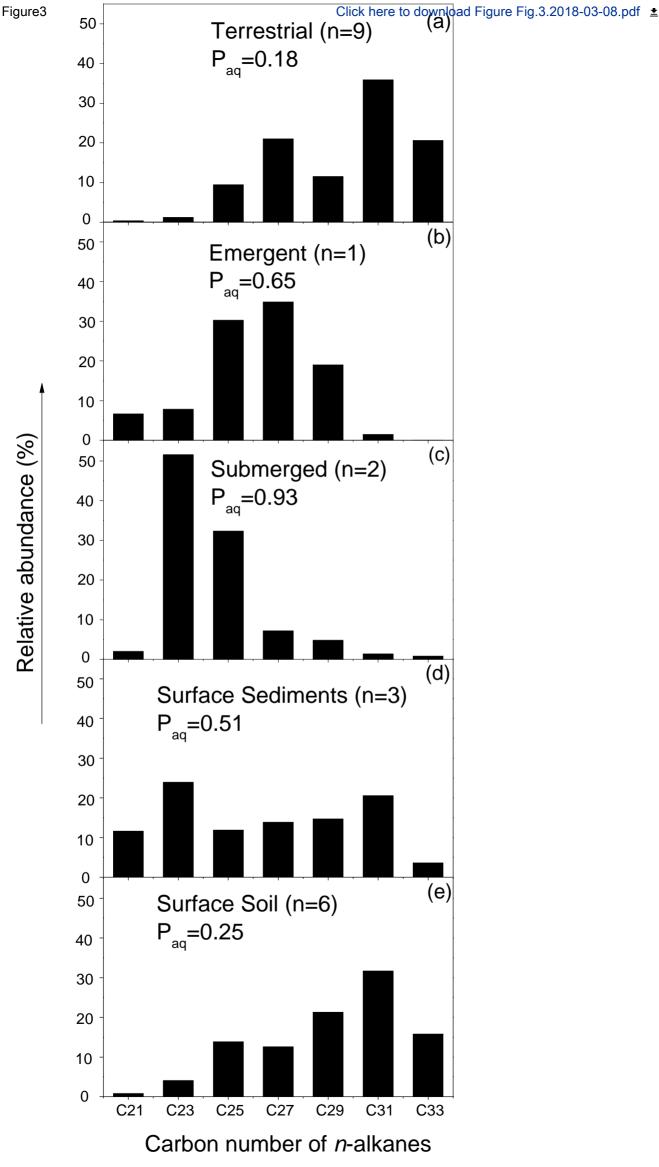
of core GSB07-1 based on depth calibration with core GSA07-1

		Core	Core GSB07-1				
Depth (cm)	Material dated	δ ¹³ C (‰ VPDB)	¹⁴ C date (yr BP)	Error (±yr)	Calibrated age (Cal yr BP-2σ range)	Calibrated depth (cm)	Calibrated ag (Cal yr BP)
162	Tree leaves	-29.0	680	30	619±56	174	662±21
183	Tree leaves	-26.9	855	35	740±47	193	776±27
221	Tree leaves	-21.1	1080	35	963±35	228	1009±33
260	Tree leaves	-11.6	1255	30	1169±42	240	1088±23
302	Tree leaves	-18.7	1440	45	1378±37	257	1192±14
383	Tree leaves	-21.0	1775	30	1793±30	366	1768±58
436	Tree leaves	-24.3	2060	30	2089±42	443	2217±82
489	Tree leaves	-23.1	2355	30	2398±52	509	2633±37
510	Tree leaves	-17.6	2520	35	2537±49	526	2719±26
518	Tree leaves	-12.7	2585	40	2580±52	532	2754±25
554	Tree leaves	-26.3	2415	35	2789±51	545	2830±25
570	Tree leaves	-25.4	2830	30	2898±45	554	2891±30
600	Tree leaves	-16.6	2895	45	3097±51	593	3130±43
660	Tree leaves	-19.7	3300	30	3520±43	658	3538±38
701	Tree leaves	-30.3	3400	30	3793±36	700	3810±19
732	Tree leaves	-24.4	3720	35	4002±33	730	4012±28
751	Tree leaves	-20.2	3935	35	4149±45	754	4174±21
815	Tree leaves	-14.7	4100	40	4547±33	816	4540±29
848	Tree leaves	-19.5	4230	35	4726±33	838	4653±28
893	Tree leaves	-18.3	4400	40	4937±41	882	4899±33
966	Tree leaves	-16.3	4495	40	5277±33	957	5290±25
1014	Tree leaves	-25.6	4820	40	5513±32	992	5497±26
1049	Tree leaves	-27.1	4970	35	5689±34	1023	5673±33
1086	Tree leaves	-25.4	5030	35	5870±33		
1114	Tree leaves	-24.4	5440	70	6038±41		

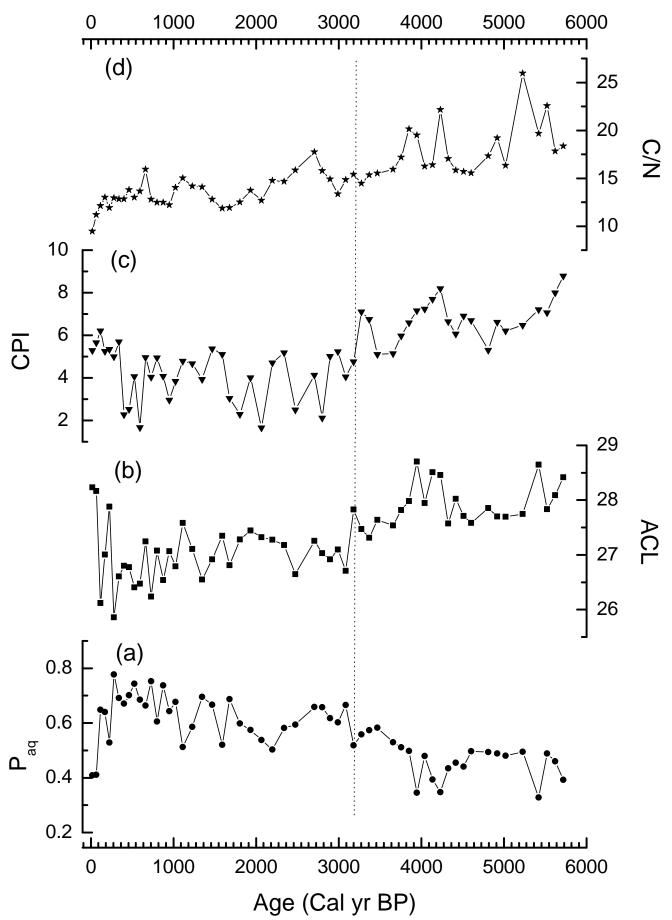


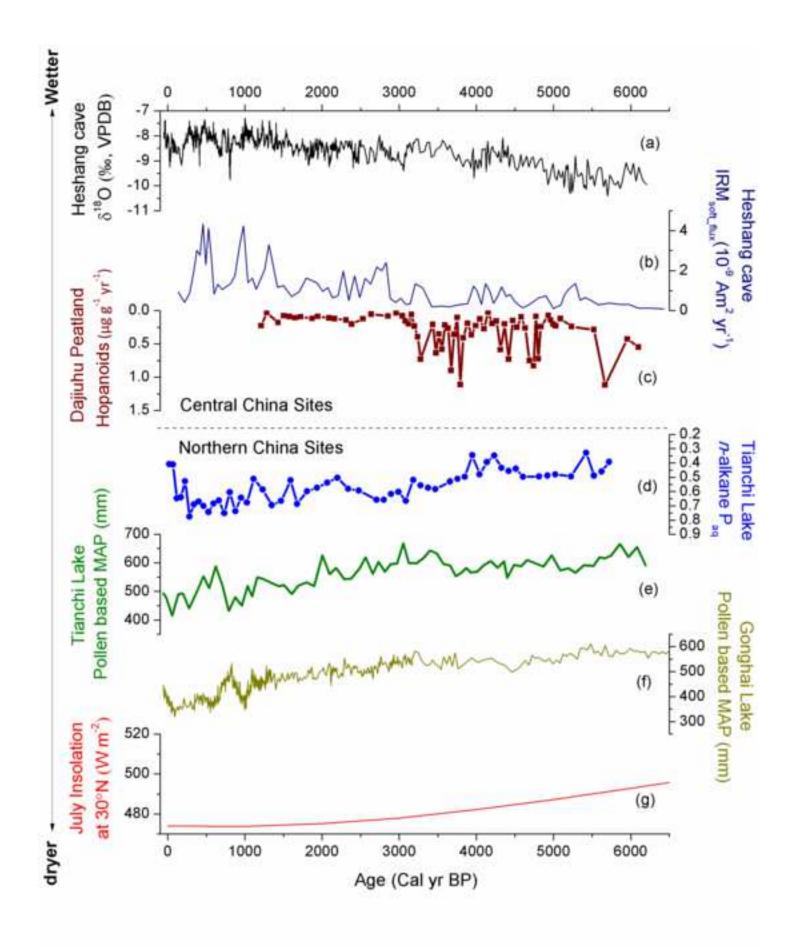






Relative abundance (%)





		Core GSB07-1					
Depth (cm)	Material dated	δ13C (‰ VPDB)	14C date (yr BP)	Error (±yr)	Calibrated age (Cal yr BP-2σ range)	Calibrated depth (cm)	Calibrated age (Cal yr BP)
162	Tree leaves	-29.0	680	30	619±56	174	662±21
183	Tree leaves	-26.9	855	35	740±47	193	776±27
221	Tree leaves	-21.1	1080	35	963±35	228	1009±33
260	Tree leaves	-11.6	1255	30	1169±42	240	1088±23
302	Tree leaves	-18.7	1440	45	1378±37	257	1192±14
383	Tree leaves	-21.0	1775	30	1793±30	366	1768±58
436	Tree leaves	-24.3	2060	30	2089±42	443	2217±82
489	Tree leaves	-23.1	2355	30	2398±52	509	2633±37
510	Tree leaves	-17.6	2520	35	2537±49	526	2719±26
518	Tree leaves	-12.7	2585	40	2580±52	532	2754±25
554	Tree leaves	-26.3	2415	35	2789±51	545	2830±25
570	Tree leaves	-25.4	2830	30	2898±45	554	2891±30
600	Tree leaves	-16.6	2895	45	3097±51	593	3130±43
660	Tree leaves	-19.7	3300	30	3520±43	658	3538±38
701	Tree leaves	-30.3	3400	30	3793±36	700	3810±19
732	Tree leaves	-24.4	3720	35	4002±33	730	4012±28
751	Tree leaves	-20.2	3935	35	4149±45	754	4174±21
815	Tree leaves	-14.7	4100	40	4547±33	816	4540±29
848	Tree leaves	-19.5	4230	35	4726±33	838	4653±28
893	Tree leaves	-18.3	4400	40	4937±41	882	4899±33
966	Tree leaves	-16.3	4495	40	5277±33	957	5290±25
1014	Tree leaves	-25.6	4820	40	5513±32	992	5497±26
1049	Tree leaves	-27.1	4970	35	5689±34	1023	5673±33
1086	Tree leaves	-25.4	5030	35	5870±33	1060	5871±35
1114	Tree leaves	-24.4	5440	70	6038±41	1094	6078±58

Table	<u>- 1</u>
Idult	Ξ <u>Τ</u>