**Asynchronous evolution of the isotopic composition and
amount of precipitation in north China during the Holocene
revealed by a record of compound-specific carbon and
hydrogen isotopes of long-chain *n*-alkanes from an alpine
lake**

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ABSTRACT

Both the timing of the maximum East Asian summer monsoon (EASM) intensity
in monsoonal China and the environmental significance of the Chinese stalagmite

oxygen isotopic record ($\delta^{18}\text{O}$) have been debated. Here, we present a ca. 120-year-resolution compound-specific carbon ($\delta^{13}\text{C}$) and hydrogen (δD) isotopes of terrestrial long-chain *n*-alkanes extracted from a well-dated sediment core from an alpine lake in north China. Our $\delta^{13}\text{C}$ data, together with previously reported pollen data from a parallel core, demonstrate a humid mid-Holocene from ca. 8–5 ka BP. Assuming that the climatic humidity of north China is an indicator of the EASM intensity, then the maximum EASM intensity occurred in the mid-Holocene. Our δD data reveal a similar long-term trend to the $\delta^{18}\text{O}$ record from nearby Lianhua Cave, indicating that the synchronous δD and $\delta^{18}\text{O}$ records faithfully record the δD and $\delta^{18}\text{O}$ of precipitation, respectively. The most negative δD and $\delta^{18}\text{O}$ values occur in the early-mid Holocene, from ca. 11–5 ka BP. This contrast in the timing of isotopic variations demonstrates a complex relationship between the isotopic composition of precipitation and precipitation amount, or EASM intensity. Further comparisons indicate a possible linkage between the precipitation amount in north China and the west-east thermal gradient in the equatorial Pacific. In addition, the temperature of the moisture source area may play an important role in determining the isotopic composition of precipitation in monsoonal China.

Keywords: long-chain *n*-alkanes; $\delta^{13}\text{C}$ and δD ; precipitation amount; north China; isotopes in precipitation; Holocene

1. Introduction

The evolution history of the East Asian summer monsoon (EASM) has long been of major research interest (e.g. Shi et al., 1994; An et al., 2000). Recent debate is focused on whether the maximum EASM intensity occurred in the early Holocene or the mid-Holocene (e.g. Chen F et al., 2015; Liu et al., 2015). Recent independently-dated climatic records from loess deposits in north China demonstrate that peak EASM precipitation occurred from ca. 8–3 ka BP (Lu et al., 2013). However, results from stalagmites (Heshang Cave, HS) and peat deposits (Dajiuahu, DJH) in central China (Fig. 1) indicate that the mid-Holocene interval of ca. 7–3 ka BP was relatively arid (Xie et al., 2013).

As illustrated in Fig. 1, the results of a recent analysis of modern observation data from 1951–2012 from 753 meteorological stations across China indicate a trend of decreasing summer (June, July, August) precipitation in north China, but a trend of increasing summer precipitation in south and central China (Wu et al., 2014; Liu et al., 2015). This dipolar mode of precipitation seems to have commenced during the past ca. 1 ka, according to a recent synthesis of paleoclimatic records (Chen J et al., 2015). An increased EASM intensity would be expected to result in a northward movement of the rainfall belt in China and thus to a simultaneous rainfall increase in north China and a decrease in central and south China, i.e., a humid mid-Holocene from ca. 8–3 ka BP in north China (Lu et al., 2013) and a relatively arid mid-Holocene from ca. 7–3 ka BP in central China (Xie et al., 2013). This spatial pattern of rainfall on an orbital scale in the Holocene is implicit in the aforementioned dipole mode, with overall maximum precipitation in the mid-Holocene in north China. Furthermore, the

precipitation amount in north China is being increasingly emphasized as an indicator of the EASM intensity (Liu et al., 2015; Chen F et al., 2015).

Because of their precise and absolute chronology, high-resolution oxygen isotope ($\delta^{18}\text{O}$) records from stalagmites in monsoonal China have been widely used as indicators of the EASM intensity (Wang et al., 2008; Dong et al., 2010, 2015). Consequently, the interval of most negative $\delta^{18}\text{O}$ values, in the early-mid Holocene, is widely regarded as corresponding to peak EASM intensity (Dong et al., 2010, 2015). However, these records clearly exhibit an apparent time lead relative to the aforementioned maximum EASM intensity during the mid-Holocene (Lu et al., 2013; Xie et al., 2013). With regard to the paleoenvironmental significance of the Chinese stalagmite $\delta^{18}\text{O}$ records, it has been increasingly argued that they may be affected by several factors, including different vapor source ratios and winter temperatures (Clemens et al., 2010), Indian summer monsoon intensity (Pausata et al., 2011; Liu et al., 2015), and the circulation effect (Tan, 2014; Caley et al., 2014).

Although the stalagmite $\delta^{18}\text{O}$ record is widely regarded as a recorder of the local $\delta^{18}\text{O}$ in precipitation (Zeng et al., 2015), it is not a direct recorder since the $\delta^{18}\text{O}$ values may potentially be biased by local effects such as the "buffering effect" of soil water (Genty et al., 2014). Therefore, more records related to the past variation of the isotopic composition of precipitation are needed, especially δD records since they can be directly compared with $\delta^{18}\text{O}$ records. Similarly, records of the past variation of local precipitation amount and the isotopic composition of precipitation from the same sediment core, or the same site, are needed in order to reveal the relationship

between precipitation amount and the isotopic composition of precipitation.

In the present paper, we present the results of analyses of compound-specific $\delta^{13}\text{C}$ and δD values of long-chain *n*-alkanes sourced from land plants and extracted from a well-dated sediment core from an alpine lake in north China. The $\delta^{13}\text{C}$ data are compared with previously-reported pollen data from the same lake (Chen F et al., 2013, 2015), and provide further confirmation of changes in the terrestrial vegetation in the lake catchment. Both of these variables have a direct relationship with local precipitation amount, as controlled by the EASM intensity. Our δD data can also be compared with previously reported $\delta^{18}\text{O}$ data from nearby Lianhua cave (Dong et al., 2015; Fig. 1), and from central China (Wang et al., 2008; Dong et al., 2010; Fig. 1), since both the δD and $\delta^{18}\text{O}$ datasets may be related to the isotopic composition of local precipitation.

2. Materials and methods

2.1. Gonghai Lake

Gonghai Lake (GH; 38°54' N, 112°14' E; 1,860 m a.s.l; Fig. 1) is located close to both the modern northern boundary of the EASM and the northeastern margin of the Chinese Loess Plateau. GH has an area of ca. 0.36 km², a maximum water depth of ca. 10 m and a flat lake bed. It is a small closed-basin alpine lake whose principal water source is precipitation. The present-day local climate is dominated by the EASM; instrumental data from the nearby Ningwu Weather Station indicate an annual mean temperature of ca. 6 °C and an annual mean precipitation of 445 mm, with ~65% of

annual precipitation falling between June and August. The local vegetation type is transitional forest-steppe, dominated by *Larix principisrupprechtii*, *Pinus tabulaeformis* and *Populus davidiana* forest, with *Hippophae rhamnoides* scrub, *Bothriochloa ischaemum* grassland and *Carex spp.* widely distributed on the planation surface, including in the GH catchment (Chen F et al., 2013).

2.2. Lithology of sediment core GH09C

In January 2009, two parallel sediment cores of 9.8 m (GH09B) and 9.3 m length (GH09C) were obtained from the centre of GH at water depths of 8.96 m and 9.21 m (Fig. 1), respectively, using a Uwitec Piston Corer. The cores were transported to the laboratory and stored in a freezer at a temperature < -4 °C. As shown by Fig. 2, core GH09C can be divided into three lithological units: Unit 1 (ca. 9.3–7.5 m) consists of alternating layers of dark silty clay and light–dark silt with the occasional occurrence of yellow sand; Unit 2 (ca. 7.5–2.5 m) consists of dark organic mud interlayered with light grey mud from ca. 4.5–2.5 m; and Unit 3 (ca. 2.5–0 m) consists of silty clay containing occasional plant macrofossils.

The results of detailed magnetic measurements (Chen F et al., 2013) and high resolution (1 cm per sample) pollen analyses, and subsequently a pollen-based quantitative annual precipitation reconstruction, for core GH09B (Chen F et al., 2015) have been reported previously. A comparison of the simplified lithology of cores GH09B and GH09C, and carbonate content, grain size, organic matter content, and magnetic susceptibility data from core GH09B and magnetic susceptibility data from

core GH09C are illustrated in [Fig. 2](#) ([Chen F et al., 2015](#)).

2.3. Chronology

Details of the chronology of both cores GH09B and GH09C are presented elsewhere ([Chen F et al., 2015](#)). Briefly, the chronology of the upper 35 cm of core GH09B was determined using ^{210}Pb and ^{137}Cs dating. Seven terrestrial plant macrofossil samples from core GH09C and 18 terrestrial plant macrofossil samples from core GH09B ([Fig. 2](#)) were picked for AMS radiocarbon dating at the Xi'an AMS Laboratory (China) and at Beta Analytic Inc. (Florida, USA). All of the radiocarbon ages were calibrated to calendar years using the IntCal09 calibration curve ([Reimer et al., 2009](#)) and are expressed in 1,000 years (ka) before present (BP) where "present" is defined as AD 1950. Bayesian age-depth modelling for core GH09B based on all 25 AMS dating results was performed using the OxCal v4.2.2 Program designed for the Poisson-process (P-sequence) depositional model ([Ramsey, 2008](#)), at 1 cm increments. Finally, a detailed chronology was transferred back to core GH09C using detailed magnetic susceptibility correlations ([Fig. 2](#)).

According to the chronology results ([Chen F et al., 2015](#)), core GH09C spans the past ca. 14.7 ka ([Supplementary Table S1](#)). According to the final age model and the sediment accumulation rate of core GH09C, 126 subsamples from core GH09C at intervals of 3-20 cm (average of ca. 7.4 cm per sample) were used for organic geochemical analyses ([Supplementary Table S1](#)), with an average time resolution of ca. 120 years.

2.4. Analytical methods

All the 126 subsamples from core GH09C were freeze-dried, lightly ground and ultrasonically extracted. The concentrated free lipids were extracted using pure *n*-hexane and then separated via silica gel column chromatography. After isolation by urea adduction, the *n*-alkanes fraction was analyzed using an HP 6890 gas chromatograph (GC). The carbon numbers and concentrations were determined by comparing their retention times and peak areas with those of *n*C₃₆ alkanes (internal standard substances, [Fig. 3](#)). Compound-specific $\delta^{13}\text{C}$ values were determined using an Agilent 7890A GC connected to an IsoPrime100 isotope ratio mass spectrometry (IRMS) via a GCC III interface (GC-C-IRMS). The resultant $\delta^{13}\text{C}$ values were expressed relative to the Vienna Pee Dee Belemnite standard; the average standard deviation for duplicate analyses of samples and multianalyses of standard substances was less than $\pm 0.3\%$. Compound-specific δD values were determined using an HP 6890 GC connected to a Finnigan DELTA^{plus} XL IRMS via a thermal conversion (GC-TC-IRMS). The final δD values were expressed relative to the standard mean ocean water, and most standard deviations for repeated analyses of the standard substances and samples were less than $\pm 4\%$. Finally, the weighted average long-chain $\delta^{13}\text{C}_{n\text{-alkanes}}$ and $\delta\text{D}_{n\text{-alkanes}}$ values of the *n*C₂₇, *n*C₂₉, and *n*C₃₁ alkanes were calculated using conventional methods ([Ratnayake et al., 2006](#)) and the results are listed in [Supplementary Table S1](#).

3. Results

In all of the subsamples, an odd number of *n*-alkanes of nC_{23} to nC_{33} , which are normally dominated by nC_{27} or nC_{29} homologues, exhibit higher relative abundances and significant odd-to-even preferences, as shown by the typical gas chromatogram in Fig. 3. The calculated carbon preference index values for nC_{26} to nC_{34} alkanes (Ratnayake et al., 2006) are in the range 5.1–13.1, with most of them greater than 9, consistent with the values for modern higher land plants (Rieley et al., 1993) and clearly indicating the terrestrial higher plant origins of the obtained long-chain *n*-alkanes. Significant positive correlations between compound-specific $\delta^{13}C$ and δD data of the nC_{27} , nC_{29} , and nC_{31} alkanes further confirm that the long-chain *n*-alkanes (nC_{27} , nC_{29} , nC_{31}) in the sediments of core GH09C are mainly derived from a single source, i.e., higher land plants (Fig. 4). Therefore, we use the weighted average $\delta^{13}C_{n\text{-alkanes}}$ and $\delta D_{n\text{-alkanes}}$ values of nC_{27} , nC_{29} and nC_{31} alkanes to represent the overall $\delta^{13}C$ and δD signals of higher land plants (expressed as $\delta^{13}C$ and δD respectively).

Compound-specific $\delta^{13}C$ data were obtained from 124 of the 126 subsamples. The $\delta^{13}C$ data range from ca. -36.8‰ to -27.8‰ , with an average value of ca. -30.8‰ (Supplementary Table S1). As a whole, the $\delta^{13}C$ data can be divided into two intervals with relatively negative values, and three intervals with relatively positive values. The two negative intervals are ca. 13–12 and 8–5 ka BP, and the three positive intervals are ca. 14.7–13, 12–8, and 5–0 ka BP (Fig. 5).

Compound-specific δD data were obtained from 113 of the 126 subsamples. The

δD data range from ca. −199‰ to −163‰, with an average value of ca. −185‰ (Supplementary Table S1). The δD values exhibit a positive excursion in the interval of ca. 13–12 ka BP and then rapidly become more negative. The most negative interval of δD values is the early-mid Holocene, from ca. 11–5 ka BP, with three obvious positive excursions at ca. 10, 8 and 6 ka BP. Subsequently, towards the late Holocene, the δD values exhibit an overall trend of more positive values with some minor fluctuations superimposed (Fig. 5).

4. Discussion

4.1. Isotopic composition of precipitation in north China

Results from modern higher land plants (Smith and Freeman, 2006), lacustrine surface sediments (Garcin et al., 2012) and soil surface samples (Jia et al., 2008; Rao et al., 2009; Tipple and Pagani, 2013) all demonstrate that long-chain δD values derived from higher land plants primarily record the δD signals of local precipitation (Sachse et al., 2012), while they may also be secondarily affected by variations in the relative abundance of C₃/C₄ plants (Chikaraishi and Naraoka, 2003; Smith and Freeman, 2006) and ecological life form (i.e., trees, shrubs, grasses; Liu et al., 2006).

Interestingly, the long-chain δD data from core GH09C exhibit similar trends to the δ¹⁸O data from nearby Lianhua Cave (Dong et al., 2015): there are positive offsets during the Younger Dryas (YD) interval, with most of the negative values occurring during the early-mid Holocene (ca. 11–5 ka BP), followed by a return to positive levels in the late Holocene (Fig. 5). In terms of millennial- and centennial- scale

variations, there are short-term synchronous positive and negative excursions in both the δD and $\delta^{18}\text{O}$ records, such as the positive excursions at ca. 10 and 8 ka BP (which may related to the so-called 8.2 ka event) and at 6, 4.5 and 2.5 ka BP (indicated by the vertical grey bars in [Fig. 5](#)).

The greater amplitude of variation of the long-chain δD data from core GH09C compared to the $\delta^{18}\text{O}$ record may result mainly from the greater mass differentials in hydrogen isotopes (1 vs. 2) relative to oxygen isotopes (16 vs. 18). The minor differences in the δD and $\delta^{18}\text{O}$ records may result mainly from the different time resolution, and from insignificant differences in the recording mechanisms. For example, the $\delta^{18}\text{O}$ signal in caves may have been affected by the buffering effect of the overlying soil water on the local $\delta^{18}\text{O}$ in precipitation ([Genty et al., 2014](#)), and the δD data may be affected by minor effects from C_3/C_4 relative abundance ([Chikaraishi and Naraoka, 2003](#); [Smith and Freeman, 2006](#)) and in ecological life forms ([Liu et al., 2006](#)). Usually, however, δD and $\delta^{18}\text{O}$ values co-vary in modern precipitation ([Craig, 1961](#)); and therefore the similar overall pattern of variations in the δD record from core GH09C and in the nearby $\delta^{18}\text{O}$ record from Lianhua Cave ([Fig. 5](#)), determined by careful cross-validation, demonstrates that both of these variables are indicators of the δD and $\delta^{18}\text{O}$ values of local precipitation. Local effects, such as those of soil water buffering for $\delta^{18}\text{O}$ and C_3/C_4 relative abundance and ecological life forms for long-chain δD data, exert minimal influence at the study sites of GH and Lianhua Cave.

In summary, the GH long-chain δD record is an indicator of the δD signal of

local precipitation and exhibits a similar pattern of variations to the $\delta^{18}\text{O}$ record from nearby Lianhua Cave. Both the GH δD record and the Lianhua $\delta^{18}\text{O}$ record demonstrate that the most negative stage of the isotopic composition of precipitation in north China occurred during the interval of ca. 11-5 ka BP.

4.2. Precipitation amount in north China

The higher land plants assimilate atmospheric CO_2 principally via two photosynthetic pathways, termed the C_3 and C_4 pathways, with $\delta^{13}\text{C}$ values mainly ranging from ca. -22‰ to -30‰ and ca. -9‰ to -19‰ , respectively (Farquhar et al., 1989). In addition, it has long been demonstrated that different $\delta^{13}\text{C}$ ranges of long-chain n -alkanes also occur in C_3 and C_4 plants (Rieley et al., 1993). Therefore, long-chain $\delta^{13}\text{C}$ data extracted from lacustrine sediments have been widely used to trace past C_3/C_4 variations (e.g. Huang et al., 2001; Jia et al., 2015).

The $\delta^{13}\text{C}$ values in core GH09C fall principally into the data range for modern C_3 plants (Rieley et al., 1993), which indicates that during the past ca. 1.47 ka the local vegetation was dominated by C_3 plants (Fig. 5). In a previous comparative study of long-chain $\delta^{13}\text{C}$ and $\delta^{13}\text{C}_{\text{TOC}}$ (total organic matter) data from 100 surface soils distributed in eastern China ($18^\circ\text{N} - 50^\circ\text{N}$), -32‰ was suggested as the discriminator between pure C_3 and mixed C_3/C_4 vegetation (Rao et al., 2008). If this is the case, the $\delta^{13}\text{C}$ data from core GH09C indicate that the local vegetation in the GH catchment area was overwhelmingly dominated by pure C_3 plants around 13–12 ka BP, corresponding to the YD interval, as well as from ca. 8–5 ka BP, both these two

intervals exhibit a negligible C₄ signal (Fig. 5). During the other intervals, the local vegetation consisted of mixed C₃/C₄ types dominated by C₃ plants (Fig. 5).

Both the low temperature and high precipitation have an adverse effect on the growth of C₄ plants (Farquhar et al., 1989; Sage et al., 1999). Independent data from core GH09B demonstrate that tree pollen percentages (all trees are C₃ plants; Farquhar et al., 1989) were normally greater than 70% from ca. 8–5 ka BP (Chen F et al., 2015; Fig. 5). Investigations of modern C₃ plants demonstrate that their $\delta^{13}\text{C}$ values become more negative under more humid climatic conditions (Diefendorf et al., 2010; Kohn, 2010). The very negative long-chain $\delta^{13}\text{C}$ data from core GH09C during this interval probably reflect the response of $\delta^{13}\text{C}$ of the C₃ forest to humid climatic conditions, supporting the results of the quantitative reconstruction of precipitation amount based on pollen assemblages in core GH09B (Chen F et al., 2015; Fig. 5). In core GH09B tree pollen percentages were normally less than 30% during the YD interval (Chen F et al., 2015; Fig. 5). With regard to the negative long-chain $\delta^{13}\text{C}$ data from core GH09C during the YD, this interval is widely accepted as being cold (North Greenland Ice Core Project members, 2004) and low temperatures generally inhibit the growth of C₄ plants (Zhang et al., 2003; Rao et al., 2012). In fact, almost no C₄ species occur below 46 °S, and only a few C₄ species occur above 60 °N (Sage et al., 1999) and on the Tibetan Plateau (Wang et al., 2004). High latitude and high altitude regions are characterized by low temperatures, and therefore it seems that the low temperatures during the YD interval severely limited the growth of C₄ plants, further reinforcing the proposition that the local vegetation during this period was dominated

by C₃ grasses, with a negligible C₄ component (Fig. 5). During the other intervals, such as the early (ca. 12–8 ka BP) and late (ca. 5–0 ka BP) Holocene, the mixed C₃/C₄ vegetation exhibited a complex relationship with climatic conditions which apparently resulted from the contrary effects of increasing precipitation and temperature on the C₄ relative abundance; and thus increased precipitation adversely affected the growth of C₄ plants, but higher temperatures favored their growth (Farquhar et al., 1989; Rao et al., 2012). In other words, during the early Holocene, the increased precipitation adversely affected the growth of C₄ plants, and the increased temperature favored their growth; during the late Holocene, the decreased precipitation favored the growth of C₄ plants, and the decreased temperature adversely affected their growth.

In summary, the $\delta^{13}\text{C}$ data from GH support the previously-reported pollen data from a parallel sediment core. Together, both datasets demonstrate the occurrence of a relatively humid interval from ca. 8-5 ka BP during the Holocene.

4.3. Asynchronous evolution of the amount and isotopic composition of precipitation

The relatively humid interval from ca. 8-5 ka BP, as indicated by the GH $\delta^{13}\text{C}$ and pollen data (Fig. 5), is widely supported by recent results from the Chinese Loess Plateau (Lu et al., 2013), sand lands (Wang et al., 2014), and deserts (Li et al., 2014), implying that it was a widespread phenomenon in north China (Liu et al., 2015). During the past ca. 14.7 ka in north China, there are several intervals during which changes of isotopic composition and amount of precipitation occur synchronously: from ca. 14-13 (roughly the Bølling/Allerød interval), 13-11 (roughly the YD interval)

and from 5-0 ka BP (late Holocene) (Fig. 5). An inconsistent relationship occurred mainly during the interval from ca. 11-5 ka BP when the precipitation isotopes were generally negative. However, precipitation amount exhibits an increasing trend from ca. 11-8 ka BP, reaching a peak from ca. 8-5 ka BP (Fig. 5). All of this evidence demonstrates a complex relationship between the isotopic composition of precipitation and precipitation amount in north China, and thus the former is influenced by one or more factors in addition to precipitation amount. In other words, the isotopic records related to past variations in precipitation (such as the lacustrine δD record and the stalagmite $\delta^{18}O$ record) in monsoonal China cannot be used directly as an indicator of precipitation amount, or of EASM intensity, on the orbital scale of the Holocene.

In addition to the asynchronous temporal evolution of precipitation amount and isotopic composition of precipitation in north China, as demonstrated by the GH δD and $\delta^{13}C$ data, there is also a spatial inconsistency in the two records in China. Previously reported results from DJH peat bog and a stalagmite from HS Cave (Fig. 1) reveal a relatively arid interval from ca. 7-3 ka BP in central China (Xie et al., 2013; Fig. 5), which contrasts markedly with the relatively humid interval from ca. 8-5 ka BP in north China. However, the stalagmite $\delta^{18}O$ record from central China, such as the Sanbao record (Wang et al., 2008; Dong et al., 2010), is largely consistent with the GH δD record and with the Lianhua $\delta^{18}O$ record from north China (Fig. 5).

Therefore, the Holocene evolution of precipitation amount was roughly asynchronous in central and north China, but the evolution of the isotopic

composition of precipitation was roughly synchronous in these regions (Fig. 5). As noted in section 1, the northward movement of the rain belt under enhanced EASM intensity can simultaneously result in increased precipitation amount in north China and a decrease in central China; and therefore both a relatively arid mid-Holocene in central China (Xie et al., 2013) and a relatively humid mid-Holocene in north China are compatible with an intensified EASM during this interval. The synchronous evolution of the isotopic composition of precipitation in both central and north China, in particular the timing of the interval of the most negative isotopic values during the early-mid Holocene, in contrast to the timing of the peak EASM intensity in the mid-Holocene, further indicate a complex relationship between the isotopic composition of precipitation and climatic conditions. Modern observation data demonstrate that the effects of temperature and rainfall on the isotopic composition of precipitation are more pronounced at high latitudes (such as the Polar Regions) and in tropical areas, respectively. In the middle latitudes, the isotopic composition of precipitation may be influenced by both effects (Dansgaard, 1964). Therefore, given the mid-latitudinal location of central and north China (30-60 °N), a complex relationship between the isotopic composition of precipitation and local climatic conditions, in particular, precipitation amount, is unsurprising.

4.4. Potential factors influencing the amount and isotopic composition of precipitation in monsoonal China during the Holocene

The important influence of the west-east thermal gradient in the equatorial

Pacific on climate in monsoonal China has long been observed (Huang and Wu, 1989). Its importance is further confirmed by recent work on various spatiotemporal scales, including a study of its effects on present-day conditions on a global scale (Ward et al., 2014) and on millennial-scale (Chen J et al., 2015) and Holocene orbital-scale (Jia et al., 2015) variations in EASM intensity. The reconstructed mean SST records obtained from seven marine sediment cores from the western equatorial Pacific Warm Pool and from two marine sediment cores from the eastern equatorial Pacific clearly highlight an enhanced west-east thermal gradient in the equatorial Pacific during the mid-Holocene, from ca. 8-5 ka BP (Koutavas and Joannides, 2012), indicating an overall La-Niña status consistent with a relatively humid mid-Holocene in north China (Fig. 5). Both precipitation amount in north China and the west-east thermal gradient in the equatorial Pacific exhibit increasing trends during the early Holocene, from ca. 11-8 ka BP, reaching a peak stage in the mid-Holocene from ca. 8-5 ka BP, followed by a decrease in the late Holocene (Fig. 5). This evidence suggests that the west-east thermal gradient in the equatorial Pacific played an important role in modulating variations in EASM intensity during the Holocene. The important influence of the west-east thermal gradient in the equatorial Pacific on monsoonal precipitation in east China is also supported by recent paleoclimate simulation results which suggest that the intensified mid-Holocene EASM was caused by an enhanced land-sea thermal contrast and sea level pressure gradient during that period (Jiang et al., 2013). Another simulation study also indicates that internal forcing, such as that caused by equatorial Pacific SSTs, is critical for driving monsoonal precipitation

variations in the Asian monsoonal region (Caley et al., 2014).

It also noteworthy that a synthesis of SST records from seven marine sediment cores from the western equatorial Pacific (Koutavas and Joanides, 2012) exhibits a similar pattern of change to that of the isotopic composition of precipitation in monsoonal China, i.e. both records exhibit a peak stage during the early-mid Holocene, ca. 11-5 ka BP, followed by a decreasing trend towards the late Holocene (Fig. 5). As it is one of the moisture source areas, this may partially explain the variations in the isotopic composition of precipitation in monsoonal China. When the SST in the west equatorial Pacific is relatively high, more moisture will be evaporated resulting in a depleted isotopic composition and finally in more negative isotope ratios in monsoonal China, irrespective of where the precipitation occurred. The occurrence of precipitation may be controlled by other factors, such as the aforementioned west-east thermal gradient in the equatorial Pacific. Due to the fact that potential moisture source areas for monsoonal China also include the South China Sea and the Indian Ocean, a more complete understanding of the indicative significance of the isotopic composition of precipitation in monsoonal China requires further research.

5. Conclusions

We have presented a ca. 120-year-resolution record of compound-specific carbon ($\delta^{13}\text{C}$) and hydrogen (δD) isotopic data from terrestrial long-chain *n*-alkanes extracted from a well-dated sediment core (GH09C) from Gonghai Lake in north China. Combined with previously reported pollen assemblage data from a parallel core

(GH09B) from the same lake, the $\delta^{13}\text{C}$ record clearly demonstrates the presence of C_3 -dominated forest vegetation during the mid-Holocene, ca. 8-5 ka BP, in the Gonghai catchment. This indicates a relatively humid mid-Holocene in north China and is widely supported by other results from north China. The humid mid-Holocene in north China is asynchronous with a previously reported relatively arid mid-Holocene of ca. 7–3 ka BP in central China, suggesting a “dipole mode” of precipitation amount in north and central China. Therefore, both the relatively arid mid-Holocene in central China and humid mid-Holocene in north China can be regarded as a consequence of enhanced EASM intensity during the mid-Holocene.

During the past ca. 14.7 ka, the δD data exhibit a similar long-term trend to that of $\delta^{18}\text{O}$ values from nearby Lianhua cave and from Sanbao Cave in central China. This indicates that the δD and $\delta^{18}\text{O}$ data essentially record the δD and $\delta^{18}\text{O}$ of precipitation, respectively; and that there are insignificant local effects on these two precipitation isotopic proxies at these study sites. The most negative δD and $\delta^{18}\text{O}$ values occur in the early-mid Holocene, from ca. 11–5 ka BP, and demonstrate a complex relationship between the isotopic composition of precipitation and precipitation amount, or EASM intensity. In contrast to the close relationship between precipitation amount in north China and the west-east thermal gradient in the equatorial Pacific, the isotopic composition of precipitation in monsoonal China is closely related to the SST anomaly in the western equatorial Pacific, implying the potentially important role of the temperature of the moisture source area on the isotopic composition of precipitation in monsoonal China.

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Figure captions:

Fig. 1. Spatial distribution of the study sites mentioned in the text (GH-Gonghai Lake; LH-Lianhua Cave; SB-Sanbao Cave; HS-Heshang Cave; DJH-Dajiuhu peat bog) and the decreasing (yellow background) and increasing (blue background) trends of summer (JJA) precipitation in north China and in central (areas between the middle and lower reaches of the Yangtze and Yellow River valleys) and south China respectively, for the period of 1951 to 2012 (modified from Liu et al., 2015). The panel to left shows the bathymetry of Gonghai Lake and the locations of cores GH09B and GH09C (modified from Chen F et al., 2013).

Fig. 2. Simplified stratigraphic units of cores GH09B and GH09C (the locations of macrofossil samples for AMS radiocarbon dating are indicated by black dots), and comparison of the proxy records: (a) carbonate content; (b) grain size; (c) organic matter content; (d) and (e), magnetic susceptibility (curves a-d are from core GH09B and curve e is from core GH09C; modified from the Supplementary Information of Chen F et al., 2015).

Fig. 3. Typical gas chromatogram of the *n*-alkanes fraction extracted from a sediment sample from the core GH09C. Numbers indicate carbon numbers (chain length) of the corresponding peaks. The peak marked with (●) denotes the internal standard substance (*n*C₃₆ alkanes).

Fig. 4. Illustration of the significant positive correlations between compound-specific carbon (A and B) and hydrogen (C and D) isotopes of *n*C₂₇, *n*C₂₉ and *n*C₃₁

alkanes extracted from the core GH09C.

Fig. 5. Comparison of records reflecting the evolution of precipitation amount (left column) and of the isotopic composition of precipitation (right column) during the past 15 ka. A, pollen-based quantitative annual precipitation record from core GH09B (Chen F et al., 2015); B, record of tree pollen percentages from core GH09B (Chen F et al., 2015) (the horizontal dashed grey line indicates values of 70% and 30%, respectively); C, the $\delta^{13}\text{C}$ record from the core GH09C (the grey line represents the measured values, the red line represents the 500-year running mean, and the horizontal dashed black line represents the value of -32‰ as the discriminator between pure C_3 and mixed C_3/C_4 vegetation (Rao et al., 2008)); D, record of the accumulation rates of hopanoids from the Dajiuhu peat bog (Xie et al., 2013); E, record of the west-east thermal gradient in the equatorial Pacific (Koutavas and Joanides, 2012); F, the δD record from core GH09C (the grey line represents the measured values, the red line represents the 500-year running mean); G, the $\delta^{18}\text{O}$ record from Lianhua Cave (Dong et al., 2015) (different colours represent different stalagmite samples); H, the $\delta^{18}\text{O}$ record from Sanbao Cave (Wang et al., 2008) (different colours represent different stalagmite samples); I, SST anomaly record from the western equatorial Pacific (Koutavas and Joanides, 2012). The blue background in the left column represents the relatively humid mid-Holocene in north China, the yellow background represents the relatively arid mid-Holocene in central China, and the green background represents the

overall La-Niña status during the mid-Holocene. In the right column, the light blue background represents the early-mid Holocene with the most negative isotopic composition of the precipitation, and the vertical grey bars represent positive excursions of the isotopic composition of precipitation. The locations of the sites are shown in Fig. 1. The bold and dashed lines and arrows at the bottom indicate simplified Holocene long-term trends of climatic conditions (left) and isotopic composition of precipitation (right).

Figures (provided separately):

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5