Transition from a warm and dry to a cold and wet climate in NE China across the Holocene

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Abstract

Northeast (NE) China lies in the northernmost part of the East Asian Summer monsoon (EASM) region. Although a series of Holocene climatic records have been obtained from lakes and peats in this region, the Holocene hydrological history and its controls remain unclear. More specifically, it is currently debated whether NE China experienced a dry or wet climate during the early Holocene. Here we reconstruct changes in mean annual air temperature and peat soil moisture across the last ~ 13,000 year BP using samples from the Gushantun and Hani peat, located in NE China. Our approach is based on the distribution of bacterial branched glycerol dialkyl glycerol tetraethers (brGDGTs) and the abundance of the archaeal isoprenoidal (iso)GDGT crenarchaeol. Using the recently developed peat-specific $MAAT_{peat}$ temperature calibration we find that NE China experienced a relatively warm early Holocene (~5-7 °C warmer than today), followed by a cooling trend towards modern-day values during the mid- and late Holocene. Moreover, crenarchaeol concentrations, brGDGT-based pH values, and the distribution of 6-methyl brGDGTs, all indicate an increase in soil moisture content from the early to late Holocene in both peats, which is largely consistent with other data from NE China. This trend towards increasing soil moisture/wetter conditions across the Holocene in NE China records contrasts with the trends observed in other parts of the EASM region, which exhibit an early and/or mid-Holocene moisture/precipitation maximum. However, the Holocene soil moisture variations and temperature-moisture relationships (warm-dry and cold-wet) observed in NE China are similar to those observed in the core area of arid central Asia which is dominated by the westerlies. We therefore propose that an increase in the intensity of the westerlies across the Holocene, driven by increasing winter insolation, expanding Arctic sea ice extent and the enhanced Okhotsk High, caused an increase in moisture during the late Holocene in NE China.

Keywords: Peatland, GDGTs, Holocene hydrological evolution, air temperature, NE China

1. Introduction

Climate in northeastern (NE) China is influenced by the interplay of different atmospheric circulation patterns, predominantly the Asian monsoon system and the northern-part of the Westerlies. The climate evolution in the region since the last deglacial period has been reconstructed using various types of paleoclimatic archives, such as lake sediments (e.g., Stebich et al., 2015; Zhou et al., 2016), peats (e.g., Zhou et al., 2010; Zheng et al., 2017), and speleothem oxygen isotope records (e.g., Wu et al., 2011). Several of these paleoclimatic studies have suggested that the climate of this region since the last deglaciation differed from that of other East Asian monsoon regions (e.g., Zhou et al., 2010; Stebich et al., 2015; Zheng et al., 2017).

Although these paleoclimate studies have improved our understanding of Holocene climate and environmental change, the various reconstructed patterns of hydrological change in NE China are inconsistent. For example, using *n*-alkane ratios in peat, Zhou et al. (2010) suggested that NE China was characterized by a dry early Holocene (~10.5 to 6 ka), attributed to enhanced evaporation caused by high sea surface temperatures (SSTs) from the nearby Japan Sea, and a wet late Holocene (after ~6 ka). This is consistent with pollen records from lake sediments from the Sihailongwan Maar and Tianchi lake (see compilation of Fig. 1) that indicate wettest conditions after 5 ka (Stebich et al., 2015; Zhou et al., 2016). However, a climatic evolution from a dry early Holocene to a wet late Holocene is unexpected, because the intensity of the

EASM is controlled by local summer insolation, with high insolation warming the continent and leading to a stronger EASM (Wang et al., 2005a; Wang et al., 2005b). Summer insolation was highest during the early Holocene and decreased since then (Berger and Loutre, 1991). Indeed, there are other records from the region that indicate a wet early Holocene and dry late Holocene (Li et al., 2017), more in-line with the expected evolution of the EASM based on the local insolation. The contrasting response recorded in different proxies and in different regions indicates that the climatic evolution of NE China and especially the EASM across the Holocene remains poorly constrained. This highlights a fundamental gap in our understanding of the processes and mechanisms that drive the expression of the Monsoon in NE China.

Over the last decade, peats have become an important archive for the reconstruction of terrestrial climate change in Asia (e.g., Barber et al., 2003; Xie et al., 2004; Hong et al., 2005; Zheng et al., 2007, 2015, 2017; Dise, 2009). The rate of peat accumulation and water table position are sensitive to changes in precipitation and temperature (Barber et al., 2000; Ise et al., 2008). Peat deposits are widespread in NE China and can extend back into the last deglaciation, representing the potential to constrain the deglacial evolution of climate. Previous peat-based palaeoclimate studies in NE China have focused predominantly on the Hani peatland using a range of proxies including *n*-alkane δD and $\delta^{13}C$ values (Seki et al., 2009; Yamamoto et al., 2010), peat cellulose $\delta^{13}C$ and $\delta^{18}O$ records (Hong et al., 2005; Hong et al., 2009), compositional changes in *n*-alkanes, *n*-alkanoic acids and *n*-alkanols (Zhou et al., 2010), *n*-alkan-2-one distributions (Zheng et al., 2011), glycerol dialkyl glycerol tetraethers (GDGTs) (Zheng et al., 2017), and macrofossil analysis (Schröder et al., 2007). However, biomarker records are currently lacking from other peats in NE China that span the deglaciation such as the Gushantun peat deposits. Although pollen and grain sizes have been used to reconstruct Holocene climate and vegetation changes in the Gushantun peat (Liu et al., 1989; Zhao et al., 2015; Li et al., 2017), the temperature and paleohydrological variations in this peatland during the Holocene are currently unknown. To provide new information on the paleoclimate history of NE China, and dynamics of the EASM, our study employs high temporal resolution (~100-200 year resolution) paleoclimatic proxies based on the abundance and distribution of GDGTs, similar to that of previous Holocene studies (Zheng et al., 2014, 2015, 2017).

There are two main classes of GDGTs and both are abundant in peat: i) branched (br)GDGTs, membrane lipids of bacteria that occur ubiquitously in mineral soils and peats (Weijers et al., 2006, 2007; Sinninghe Damsté et al., 2000; Naafs et al., 2017a), and ii) isoprenoidal (iso)GDGTs, membrane lipids of Archaea that are present in mineral soils and peat but typically dominate the GDGT pool in aquatic (marine) settings (Schouten et al., 2000, 2013). At present 15 different brGDGTs have been identified, bearing 0 to 2 extra methyl groups at either the C-5 or C-6 position and/or up to two cyclopentane moieties (De Jonge et al., 2013, 2014). The distribution of brGDGTs in mineral soils can be used to reconstruct past air temperatures and soil pH (Weijers et al., 2007; Peterse et al., 2012; De Jonge et al., 2014; Naafs et al., 2017b). Although most work on brGDGTs is based on mineral soils and lake sediments, peat-specific temperature and pH calibrations have recently been developed (Naafs et al., 2017a). The peat-specific proxies allow us to reconstruct temperature and pH variations over the Holocene in the Gushantun peat sequence. In addition to brGDGTs, changes in the relative abundance of crenarchaeol, a biomarker so far known only to be biosynthesized by *Thaumarchaeota* (Sinninghe Damsté et al., 2002; Schouten et al., 2013), have been used to identify past dry periods in peat (Zheng et al., 2015).

The GDGT-based records of temperature, pH, and aridity from the Gushantun and Hani peat are compared with those from other sites in NE China, as well as other Asian summer monsoon-dominated regions and arid central Asia such as the Xingjiang region, in order to confirm that our data are representative of Holocene hydrological and temperature evolution across NE China. Based on this, we offer new perspectives on Holocene climate changes and mechanisms driving climate in NE China.

2. Material and methods

2.1 Study Site

The Gushantun peat deposit (42°18′N, 126°17′E) is situated in Huinan County in Jilin Province at an elevation of 500 m on the western flank of the Changbai Mountains (Fig. 1; see Zheng et al. (2017) for precise location of the Hani peat). It is surrounded by a basalt platform that is more than 600 m high. It is suborbicular with a diameter of about 1 km and slopes from north to south. The ground is perennially saturated with water, so a swampy, peat-forming environment has been sustained in this region since the last deglaciation, and a sequence of peat of around 1-2 m in average thickness (8-9 m in maximum thickness) has accumulated. At present, the annual mean temperature is ~3 °C, with monthly mean temperatures that range from −16 °C in January to 21 °C in July. The annual mean precipitation is about 700 mm (Liu et al., 1989).

Our samples are from a 735 cm long core collected from near the center of the Gushantun peat. The core consists of 655 cm of brown to dark brown peat containing a large amount of non-degraded plant residue. Below 655 cm depth, the sediment is grayish-green to dark brown mud, representing lacustrine depositional conditions. After collection, the core was transported intact to the laboratory where it was subsampled at 1-cm intervals. All samples were stored at – 20 °C until analyses, and a total of 93 samples were analysed for their GDGT distribution.

2.2. Chronology of the peat core

Sample pretreatment, AMS-target preparation and AMS measurement were all conducted at the Xi'an AMS Laboratory. The pre-treatment of 8 peat samples for ^{14}C dating was performed using the method of Zhou et al. (2002): plant fragments with a size ranging from 90 to 300 μm were isolated from peats by wet sieving and then subjected to an Acid-Alkali-Acid (HCl-NaOH-HCl) treatment. Two samples of total organic carbon (TOC) from bulk mud sediments at the bottom of the lacustrine layers was processed using 10% HCl to remove all carbonate content before graphitization (Zhou et al., 2004). AMS-targets were prepared from the pretreated samples, which were then placed with CuO powder into 9 mm quartz tubes, evacuated to $<10^{-5}$torr, and then combusted. The CO_2 was converted catalytically to graphite using Zn (Zn powder with added Fe powder as a catalyst) (Slota et al., 1987). The calibrated ages were obtained from the ^{14}C ages using the Northern Hemisphere INTCAL13 curve (Stuiver et al., 1993; Reimer et al., 2013). In order to produce the reliable ages for all depths in the Gushantun peat core, we used Bayesian age-depth modeling software Bacon (Blaauw et al., 2011) to estimate ages and uncertainties for each sample (Fig. 2 and Table 1). The model using a Bacon approach provides a chronological framework for the past 13,000 years. Two samples were excluded as outliers based on a student-t model (Blaauw et al., 2011). We also note that the shallowest sample, from 8 cm, has a modern age, which could indicate mixture of carbon-ages in the peat profile, i.e. via root production; in the absence of high resolution approaches such as wiggle matching, these processes cannot be resolved and represent a small additional source of error in our age model.

2.3. GDGT extraction and analysis

Freeze-dried, homogenized samples (including the Hani peat samples see Zheng et al., 2017 for details) were extracted ultrasonically with a sequence of increasingly polar solvents; three times with dichloromethane (DCM), three times with

DCM/methanol (1:1, v/v) and two times with methanol. The total lipid extract was then base hydrolyzed in 1M KOH/methanol (5% H_2O in volume) at 80℃ for 2 h. The solution was extracted at least 6 times with *n*-hexane, and the combined extracts were dried under a stream of N_2 gas. Extracts were separated into a saturated hydrocarbon and a polar fraction on a short silica gel column using *n*-hexane and methanol as eluents, respectively. Half of the polar fraction was filtered through 0.45 μm PTFE syringe filters and dried under nitrogen gas and used to analyze the GDGT distribution.

The GDGTs were analyzed using an Agilent 1200 series liquid chromatography and triple quadruple mass spectrometry (LC–MS^2) system, equipped with an autosampler and ChemStation manager software. Samples were spiked with an internal C_{46} GDGT standard (Huguet et al., 2006) and re-dissolved in 300μl *n*-hexane/isopropanol (99:1, v/v). Samples (10μl) were injected and separation of GDGTs, including 5-and 6-methyl brGDGTs, was achieved using two silica columns in tandem (150 mm ×2.1 mm, 1.9 μm, Thermo Finnigan; USA) maintained at 40 °C. EtOAc was used instead of the widely used isopropanol (IPA) as it has lower polarity, leading to better separation of GDGT isomers. GDGTs eluted isocratically for the first 5 min with 84% A and 16% B, where A = n-hexane and B = EtOAc. The following elution gradient was used: 84/16 A/B to 82/18 A/B from 5–65 min and then to 100% B in 21 min, followed by 100% B for 4 min to wash the column and then back to 84/16 A/B to equilibrate the column for 30 min. We used a constant flow rate of 0.2 ml/min throughout. GDGTs were ionized in an atmospheric pressure chemical ionization (APCI) chamber with single ion monitoring at *m/z* 1050, 1048, 1046, 1036, 1034, 1032, 1022, 1020 and 1018 for the brGDGTs and *m/z* 1292 for crenarchaeol. The MS conditions followed Hopmans et al. (2000). GDGTs were quantified from integrated peak areas of the [M+H]+ ions. The relative response ratio of the GDGTs relative to the internal C_{46} GDGT standard was set at 1:1, allowing for semi-quantitative concentrations.

In addition to new data from the Gushantun peat deposit, we also use GDGT data (some previously published such as $MAAT_{peat}$) from the nearby Hani peat deposit. For details on the age model and sample preparation, see Zheng et al. (2017).

2.4. BrGDGT-based climate proxies

For this study, the global peat-specific brGDGT calibrations of Naafs et al. (2017a) were used to reconstruct mean annual air temperature (MAAT) and pH. The standard (or root mean square) errors of the temperature and pH calibrations are 4.7 ºC and 0.8, respectively. These proxies are based on the degree of methylation (MBT_{5me}') and cyclisation (CBT_{peat}) of brGDGTs and built on the original work done using mineral soils (Weijers et al., 2007; De Jonge et al., 2014). Labeling of brGDGTs follows the established protocols with Roman numbers indicating none (I), one (II) or two (III) additional methyl groups at the C5 or C6 (′) position and letters indicating none (a), one (b), or two (c) cyclopentane moieties (See the supplementary file for the structures; De Jonge et al., 2014; Naafs et al., 2017a):

$$(1)\ MBT'_{5ME} = \frac{(Ia + Ib + Ic)}{(Ia + Ib + Ic + IIa + IIb + IIc + IIIa)}$$

$$(2)\ MAAT_{peat} = 52.18 \times MBT'_{5me} - 23.05 \qquad (R^2 = 0.76, RMSE = 4.7\ ℃)$$

$$(3)\ CBT_{peat} = \log\left(\frac{Ib + IIa' + IIb + IIb' + IIIa'}{Ia + IIa + IIIa}\right)$$

$$(4)\ pH = 2.49 \times CBT_{peat} + 8.07 \qquad (R^2 = 0.58, RMSE = 0.8)$$

To reconstruct changes in the relative abundance of 5-methyl over 6-methyl brGDGTs, we used:

$$(5)\ IR_{6me} = \left(\frac{IIa' + IIb' + IIc' + IIIa' + IIIb' + IIIc'}{IIa + IIa' + IIb + IIb' + IIc + IIc' + IIIa + IIIa' + IIIb + IIIb' + IIIc + IIIc'}\right)$$

$$(6)\ f(6 - Methyl\, brGDGTs) = \frac{\left(IIa' + IIb' + IIc' + IIIa' + IIIb' + IIIc'\right)}{\left(Ia + Ib + Ic + IIa + IIa' + IIb + IIb' + IIc + IIc' + IIIa + IIIa' + IIIb + IIIb' + IIIc + II\right.}$$

3. Results

The full suite of 15 brGDGTs was present in the Gushantun peat sequence. 5-methyl brGDGTs were more abundant than 6-methyl brGDGTs in all samples. 6-methyl brGDGTs were most abundant in the lacustrine section at the bottom of the core. Generally brGDGT-Ia was the dominant brGDGT, but brGDGT-IIa and -IIIa were also present in significant amounts. The cyclopentane-containing brGDGTs, especially IIIb, IIIb', IIIc and IIIc', occurred in very low abundance or were not detected. brGDGT IIa' was the most abundant of the 6-methyl brGDGTs, followed by IIIa' and IIb'.

MBT_{5me}' and reconstructed mean annual air temperatures based on the global peat-specific calibration ($MAAT_{peat}$) have been applied to the Hani peat and previously published (Zheng et al., 2017). For Gushantun, values for the MBT_{5me}' index ranged from 0.36 to 0.64, with a mean of 0.51. $MAAT_{peat}$-based temperatures range from -4.2 to 10.2 ± 4.7 °C with a mean value of 3.4 °C (Fig. 3a). Temperatures are higher during the early Holocene (about 11 to 6 cal kyr BP), with values between 3.1 and 10.2 ± 4.7 °C , and then decrease to values between 0.8 and 4.1 ± 4.7 °C during the late Holocene. The lower $MAAT_{peat}$ values during the late Holocene correspond to higher fractional abundances of 5-methyl brGDGTs.

The CBT_{peat} index at Gushantun ranges from -1.52 to -0.12 with a mean of -0.79. Reconstructed pH ranges from 4.3 to 7.8 ± 0.8 with a mean value of 6.1 and displays a general decrease from the early Holocene to the late Holocene (Fig. 4c). The pH values covary with the IR_{6me}, and the fractional abundances of 6-methyl brGDGTs (Fig. 4a, b and c). The overall decrease in pH coincides with a general decrease in the abundance of crenarchaeol (Fig. 5c and d). The CBT_{peat} and pH values vary from -0.52 to -1.12 and 5.2 to 6.7± 0.8, respectively, at Hani peat (Fig. 5b). Crenarchaeol concentrations mostly range from and 0 to 73 ng/g at Hani (Fig. 5a).

4. Discussion

4.1 Holocene temperature variations in the Gushantun peat core

The global peat calibration of Naafs et al. (2017a) is predominantly based on low-temperature peats from the regions between 40 ºN and 60 ºN, which covers the latitude and temperature of our peat in Northeast China. Crucially, the $MAAT_{peat}$ in the top ~50 cm of Gushantun peat core (~3 to 7 °C with most values around 3 °C) fits well with the observed instrumental yearly mean air temperature in the region of between 3 and 7 °C from 1951 to 2013 and the modern-day MAAT of ~3 °C (Liu, 1989). As such this global peat calibration is well-suited to reconstruct temperature in this region. Although care has to be taken to use the global peat-specific calibration to reconstruct small-scale (1-2 °C) and brief (< 1 kyr) temperature anomalies (Naafs et al., 2017a), here we apply the calibration across the last 13,000 years to determine whether the several thousand years long period of the early Holocene is different from the several thousand years long period of the late Holocene.

We do not have data from before 13 kyr BP, but the lowest temperatures around ca. 12.7 kyr BP could be related to the global cooling during the Younger Dryas. However, we must emphasize that these sediments represent lacustrine rather than peat deposition and changes in depositional environment almost certainly have an impact on brGDGT-based temperature proxies (Sun et al., 2011; Peterse et al., 2012). For the Holocene our mean annual air temperature reconstruction indicates a warm early Holocene and a colder late Holocene climate (Fig. 3a). The reconstructed difference in temperature between the late and early Holocene is around 5-7 °C, large enough to be accurately captured by the $MAAT_{peat}$ record. The observation that the highest temperatures occurred during the early Holocene at Gushantun is consistent with other records from China that indicate highest MAAT during the early Holocene (He et al., 2004; Gao et al., 2012; Jia et al., 2013; Peterse et al., 2014; Zheng et al., 2017). The degree of warming reconstructed using $MAAT_{peat}$ at Gushantun during the early Holocene (5-7 °C) is similar to that observed at the nearby Hani peat (6-7 °C) using $MAAT_{peat}$ (Fig. 3b) (Zheng et al., 2017). They are also consistent with the relatively higher percentages of the thermophilous broadleaf trees, including *Quercus*, *Corylus*, *Juglans* and *Ulmus*, during this interval in the same peat (e.g., Liu, 1989, Zhao et al., 2015) and multiproxy based temperature reconstructions that indicate higher than modern MAAT between 8 and 3 ka in NE China (Shi et al., 1994). Similarly, pollen records from nearby Mount Changbai and Sihailongwan Maar lake (Fig. 3c) indicate higher temperatures during the early Holocene compared to modern (He et al., 2004; Stebich et al., 2015). Furthermore, MAAT records from the Chinese Loess Plateau also suggested temperature maxima 7-9 °C higher than modern during the early Holocene (Peterse et al., 2014; Gao et al., 2012; Jia et al., 2013). Consequently, we consider the temperatures obtained using the global peat calibration to be representative of climate in (NE) China.

The highest temperatures occurred between ca. 8 and 6.8 kyr BP, with occasional annual mean temperatures > 8.0 ± 4.7 °C, compared to the modern-day MAAT of ~3 °C. These relatively high temperatures were interrupted by slightly lower values between 10.5-10.2 kyr BP with temperatures ca. 5 ± 4.7 °C and between 8.7-8.3 kyr BP with temperatures ca. 6 ± 4.7 °C. Although well within the calibration error of our proxy, these two brief cool intervals could correspond with the '10.3 ka event' and '8.2 ka event' recorded in climatic records from the North Atlantic (Bond et al., 2001). From ca. 6 kyr BP, $MAAT_{peat}$-derived temperatures are colder with most values below ~4 °C, and reconstructed temperatures for the last millennium are close to the present-day mean annual air temperature in the region of ~3 °C.

The overall MAAT pattern at Gushantun peat – with a clear early Holocene maximum and cold conditions during the mid- and late Holocene – is broadly consistent with other climatic records from (NE) China. However, there are some discrepancies in the trends recorded in the Gushantun and nearby Hani peats. The Gushantun $MAAT_{peat}$ record does not exhibit a cooling during the last 2 kyr and the reconstructed temperature in the very top sample is ~7 °C. A potential seasonal bias at the very top of the peat core may be responsible for the different trends, as has been observed for some high-latitude peats due to intense summer warming (Naafs et al., 2017a). Furthermore, small discrepancies in $MAAT_{peat}$ between Gushantun and the nearby Hani peat could be related to peat soil heterogeneity (Weijers et al., 2007), difference in vegetation cover (Peterse et al., 2012), and water content (Dang et al., 2016) that can affect the brGDGTs distributions. Despite the discrepancy, the absolute MAAT estimates in the top ~ 50 cm are close to the present-day temperature in two peats, providing confidence in our absolute MAAT estimates.

4.2 Holocene moisture patterns in NE China, other Asian monsoon regions and arid central Asia

4.2.1 Holocene moisture variations in NE China

In addition to the temperature reconstructions, the Gushantun peat GDGT distributions are characterized by changes in concentrations of crenarchaeol (Fig. 5d), a biomarker specific to *Thaumarchaeota* (Sinninghe Damsté et al., 2002). *Thaumarchaeota* generally account for the majority of the archaeal community in dry soils (Timonen and Bomberg, 2009; Bates et al., 2011), but are generally less proportionally abundant in peat, which also contains abundant *Euryarchaeota* as methanogens (Zheng et al., 2015). Consequently, the relative abundance of crenarchaeol tends to be relatively high in mineral soils (depending on temperature; Xie et al., 2012; Yang et al., 2014), but low in peat (Pancost and Sinninghe Damsté, 2003; Zheng et al., 2015).

Therefore, we interpret the higher concentrations of crenarchaeol at Gushantun during the early Holocene (> 5-6 ky BP) as evidence for drier conditions in this peat-forming environment and interpret lower concentrations during the late Holocene as indicative of wetter, more typical peat-forming conditions (Fig. 5d). This interpretation is consistent with drying events in other peats also being associated with increased crenarchaeol concentrations (Zheng et al., 2015). The nearby Hani peat core exhibits some similar features in the crenarchaeol concentration profile, i.e. a long-term (but irregular) decrease over the past 6 to 8 kyr. However, concentrations are higher at Hani throughout the records, and there are strong differences in the profiles prior to 8 kyr (Fig. 5a).

This appears to reflect differences in the thaumarchaeotal population between the two peats, likely arising from hydrology and vegetation, and these differences might have been greater during the early formation of the peat, i.e. from 8 to 10 kyr. Crenarchaeol is more abundant in warm settings than in low temperature settings (Schouten et al., 2000; Zhang et al., 2006), so the low absolute and relative crenarchaeol abundances from the Hani peats between ca. 8-10 kyr BP are possibly due to the lower temperature at Hani at this time (see Fig. 3b). So although in general both peat cores indicate higher concentrations of crenarchaeol during the early Holocene, which we interpret to reflect drier conditions, there is the need for further investigation into crenarchaeol as an indicator of moisture in other peat settings from around the world.

Further evidence for changes in wetland hydrology in NE China across the Holocene comes from the brGDGT-reconstructed pH at both Hani and Gushantun, with high values during the early Holocene and low pH during the late Holocene (Fig. 5b and c). This trend is the most obvious at Gushantun. In general, low effective precipitation results in dry bog conditions, which suppresses the production of organic acids (e.g., Clymo, 1984) and yields high pH values. This is in agreement with the previous results from other peats where elevated pH values correspond to low monsoon precipitation (a dry climate) (Zheng et al., 2015; Wang et al., 2017). However, the pH values diverge between the two peats after ~4 kyr BP, increasing at Hani and continuing to decrease at Gushantun. This discrepancy in the pH variations between the two peats could be attributed to hydrological conditions and vegetation changes. The Hani river and its tributaries go through Hani peatland, thereby affecting the sedimentary environment and water content of Hani peatland, whereas Gushantun peatland does not have such an influence. Additionally, the abundance of *Sphagnum* vegetation decreases at Hani after 4 ky BP as *Betula*, *Potentilla*, and *Carex* become more abundant (Schröder et al., 2007); the decrease in *Sphagnum* abundance could be associated with an increase in pH (Gagnon et al., 1992). Thus, the dissimilarity in small-scale pH variations during the (late) Holocene between the two peats might result from different hydrological conditions, vegetation and the specific sediment settings and features, although the same calibration and method have been used. This does illustrate the complexity of peat as an archive of hydrological change and dictates caution in our discussion of Holocene change.

More evidence for changes in the moisture content across the Holocene comes from the high IR_{6me} values during the early Holocene (Fig. 4a). High IR_{6me} values occur in mineral soils and peat characterized by (arid) alkaline conditions (De Jonge et al., 2014; Dang et al., 2016; Naafs et al., 2017a). The higher IR_{6me} values during the early Holocene provide complementary evidence for drier conditions during the early Holocene. The highest values between 12 and 14 kyr BP likely reflect higher pH values during the lake phase.

The conclusion that the Gushantun (and Hani) peat and surrounding region became wetter through the Holocene (although note the complexity of the pH record at Hani) is supported by the rise in conifer tree percentages such as *Pinus* (which grow in humid settings (Sun et al., 1996)) from the same peatland and nearby lake sediments (Liu et al., 1989; Zhao et al., 2015; Stebitch et al., 2015), as well as the increase in effective precipitation across the Holocene indicated by the *n*-alkane C_{27} δD and P*aq* records at Hani (Seki et al., 2009; Zhou et al., 2010). Pollen-based mean annual precipitation (Pann) estimates continuously increased from the early Holocene and reached a maximum value around 4000 cal yr BP in Sihailongwan Maar lake (Stebich et al., 2015; Fig. 6h); Pann variations do indicate that mean annual precipitation decreased after 4 kyr BP (Fig. 6h), but it remained at a relatively high level in comparison with that of the early Holocene (Stebich et al., 2015). Similarly, Zhou et al. (2016) proposed that relatively higher pollen percentages of trees including *Pinus* and *Betula* indicate wetter climate conditions during the mid- to late Holocene (after 5 kyr BP) in Tianchi Lake compared to the early Holocene (Fig. 6g). Thus, our results are consistent with other climate reconstructions from the region that indicate an increase in effective precipitation from the early Holocene to late Holocene in NE China (Liu et al., 1989; Zhou et al., 2010; Zhou et al., 2016; Stebich et al., 2015), although the exact timing differs between different settings and records. We also note that although many of these records are for annual precipitation, that is today derived primarily from summer monsoon precipitation in NE China.

4.2.2 Comparison of Holocene moisture patterns in NE China with East Asian monsoon regions and arid central Asia

Leaf wax (*n*-alkane) δD values, GDGT distributions and pollen records, from a combination of peats and lake sediments, all indicate a dry early Holocene and a humid middle to late Holocene in NE China (Fig. 6g-i). However, this evolution differs from that known from northern China, including EASM margin regions and monsoonal eastern China (Liu et al., 2015). It is also opposite to that recorded by the high-resolution stalagmite $\delta^{18}O$ records from northeast China, northern China and Southern China (Fig. 6b; Wu et al., 2011; Cai et al., 2010; Tan, 2009; Dykoski et al., 2005; Wang et al., 2005a) and the biomarker records from the Pearl River Estuary (Strong et al., 2013). Liu et al. (2015) summarized pollen-based proxy evidence from lake sediments, paleosol development, and other proxies from loess-paleosol sequences and aeolian activity in the northern Chinese sandlands from various geographical regions and concluded that the EASM precipitation maximum occurred during the mid-Holocene (ca. 8-3 kyr BP).

Stalagmite $\delta^{18}O$ records from Dongge cave in southern China indicate an early Holocene (ca. 10-5 kyr BP) EASM maximum (Fig. 6b; e.g., Dykoski et al., 2005; Wang et al., 2005a). In particular, the Dongge Cave $\delta^{18}O$ record was originally interpreted to record elevated monsoon precipitation (wet) between 10-6 kyr (An et al., 2000). However, subsequent studies on moisture/precipitation from different sites have challenged this conclusion and the record has been re-interpreted as indicating maximum effective precipitation during the early/mid-Holocene in EASM regions (Herzschuh et al., 2006; Wang et al., 2010; Zhao et al., 2009; Zhang et al., 2011; Chen et al., 2015). More recently, Zhou et al. (2016) proposed that the Holocene Optimum, defined as a period with high monsoon precipitation, began ca. 6 kyr BP in NE China, although it is likely that a slightly northward transgression of the high monsoon precipitation occurred (Zhou et al., 2016). Nonetheless these studies collectively indicate that the Holocene moisture optimum occurred largely during the early/mid-Holocene with effective moisture or monsoon precipitation decreasing during the late Holocene in the EASM region. NE China is influenced by the East Asian monsoon system and that seems to be manifested in some aspects of our records (i.e. Hani peat pH) and other records (Pollen and Pann records from lakes; Fig. 6g and h) from 4 kyr. But the long-term decline in moisture over the past 8 kyr recorded in EASM-dominated regions is not seen in most records and it appears that other climatic factors have influenced the moisture history of NE China (Fig. 6).

A regional synthesis from the North Xinjiang area based on fifteen published climate proxy records also indicates a persistent increase towards wetter conditions from the early Holocene to late Holocene, similar to our findings (Fig. 6k; Wang et al., 2013). Recently, four well-dated Holocene loess-paleosol sequences from the northern slopes of the Tienshan Mountains and the Yili River valley of Xinjiang, located in the core area of arid central Asia, also indicated increasing moisture in the region from about 8 kyr BP (Fig. 6j) (Chen et al., 2016). These moisture records from central Asia are consistent with our findings from NE China (Fig. 6g-k), but differ significantly from the trends of EASM evolution during the Holocene (Fig. 1 and Fig. 6b-d).

In addition, the $MAAT_{peat}$ record from Gushantun and Hani indicate highest temperatures from 11-6 kyr BP, which broadly corresponds with a period of relatively warm climate in the middle and high latitudes of the Northern Hemisphere, including the classical ‘Holocene Optimum’ defined by high monsoon precipitation in both northern and southern China (e.g., Zhou et al., 2007; Zhao et al., 2009; Ran et al., 2013). Thus, it appears that the thermal maximum during the early Holocene corresponds to low effective precipitation in NE China but high effective precipitation in other EASM-influenced regions: i.e. the temperature-moisture patterns in NE China were mainly dominated by warm-dry, cold-wet episodes during the Holocene. This is similar to climate relationships in the core area of arid central Asia that also exhibit a transition from a warm-dry early Holocene to cold-wet late Holocene (Huang et al., 2009; Jiang et al., 2013); but it is clearly different from other East Asian monsoon regions including South China and North China which show warm-wet and cold –dry climate Holocene patterns (e.g., Wang et al., 2005a).

4.3 Possible forcing mechanisms of Holocene climate evolution in NE China

The Western Pacific Subtropical High (WPSH) is the most important component of the EASM and governs modern summer precipitation in NE China (Chu et al., 2014). Considering the linkage between SSTs in the subtropical west Pacific and the WPSH (Chu et al., 2014), we propose that increasing SST in the northern East China Sea and Sea of Japan during the early Holocene (Ishiwatari et al., 2001; Kubota et al., 2015) induced a northward shift of the WPSH. Consequently, the monsoon precipitation band extended into NE China and increased rainfall in NE China. As SSTs decreased during the late Holocene, the WPSH would have shifted southwards, weakening its influence in NE China and presumably leading to a decrease in precipitation in this region. However, our results (and those of others) indicate the opposite – an increase in effective precipitation from the early Holocene to late Holocene in NE China. Thus, the EASM precipitation driven by summer insolation might not solely control Holocene moisture variations (from dry to wet) in NE China (Fig. 6a-d and g-i). In particular, this partial discrepancy between NE China climate relationships with other Asian monsoon regions implies the influence of additional forcing mechanisms, as well as a possible link between NE China and the core area of arid central Asia. Previous research attributed this difference to local SST changes in the Sea of Japan (Zhou et al., 2010): high SSTs in the Sea of Japan during the early Holocene could have amplified the evaporation rate, resulting in the drier climate conditions. Lower SSTs and the cold climate (low MAAT) during the mid- and late Holocene caused less evaporation, resulting in wet climate conditions. Although the relatively dry climate in NE China could be partly attributed to enhanced evaporation related to the additional warming caused by higher SST from the Sea of Japan (Zhou et al., 2010), less evaporation and decreased EASM precipitation seems insufficient to explain the increasingly wet conditions during the late Holocene. Nor can wetter conditions be explained by strengthening of the East Asian winter monsoon (EAWM) during the late Holocene, as it carries a relatively dry air mass to NE China (Zhang et al., 2016).

The temperature-moisture evolution patterns in NE China, however, are coincident with those observed in the core area of arid central Asia; these also exhibit a transition from a warm-dry early Holocene to cold-wet late Holocene (Huang et al., 2009; Jiang et al., 2013; Chen et al., 2016). These relationships are also observed in the mid-latitudes of Europe between ca. 50°N and 43°N during the Holocene (Magny et al., 2003). These evidently resulted from variations in the strength of the Westerly jet, in turn related to the thermal gradient between high and low latitudes. Similarly, increased Holocene moisture/precipitation in the core area of the arid central Asia has been attributed to an increase in the strength of the westerlies (Chen et al., 2016). Thus, this suggests that the westerlies might be a major link between NE China, the core area of arid central Asia and mid-latitude Europe, as we have also suggested based on enhanced MAAT variability recorded in the Hani peat (Zheng et al., 2017). Indeed, the strength of the westerlies, inferred from the insolation gradient between 35°N and 55°N, could have gradually increased since the early Holocene (Rossby et al., 1939; Chen et al., 2016). In this scenario, increasing winter insolation caused an increase in winter temperature from the early Holocene to the late Holocene in Northern Europe (Fig. 6e; Davis et al., 2003), leading to enhanced water vapor evaporation over the Mediterranean, Black and Caspian Seas. Previous research has confirmed that this air with elevated water vapor contents could have been delivered by strengthened mid-latitude westerlies to the Xinjiang region (Northern China) in the core area of arid central Asia (Fig. 6f; Zhang et al., 2016; Chen et al., 2016; Long et al., 2017). In fact, the westerlies could have penetrated even further eastward (Vandenberghe et al., 2006), even to Japan (Yamada, 2004). Therefore, both higher water vapor contents and strengthened westerly winds could have brought more moisture to NE China, thereby causing wetter conditions, during the late Holocene

(Fig. 6f).

In addition, changes in Arctic sea ice extent and shifts in the position of the Okhotsk High also play an important role in regulating climate (including moisture) in NE China (e.g., Guo et al., 2014; Chu et al., 2014). According to Guo et al. (2014), lower spring Arctic sea ice extent is associated with less rainfall in the Northern EASM region, and vice versa. Consistent with this scenario, the relative abundance of sea ice-related diatoms from the West Okhotsk Sea shows a long-term increase through the Holocene (Harada et al., 2014). Thus, increased sea ice extent during the late Holocene could have caused high precipitation in NE China in comparison with the early Holocene. Furthermore, decreasing SST in the Okhotsk Sea from the early Holocene to late Holocene could have strengthened the Okhotsk high that brings moisture into the Far East including NE China (Kakei and Sekine, 2004; Harada et al., 2014). Clearly, our records cannot unravel these complex and multiple climatic controls on rainfall and peat water balance. They do, however, clearly indicate a decoupling between NE China and other EASM-dominated regions that requires further critical analysis and could have implications for our understanding of how the EASM evolves in the future.

5. Conclusions

In this study we present a detailed GDGT data set covering the last 13,000 years from a peat sequence in the Changbai Mountain in NE China. The brGDGT-based temperature reconstruction from Gushantum peat indicates that mean annual air temperatures in NE China during the early Holocene were 5-7 ºC higher than today. A constantly high air temperature is reconstructed between ca. 8 and 6.8 kyr BP, with maximum annual mean temperatures exceeding ca. 8.0 ºC. Lower temperatures are recorded from around ca. 6 kyr BP onwards, with most values $< 4 \pm 4.7$ ºC. Crenarchaeol concentrations, brGDGT-based pH values, and relative abundance of 6-methyl brGDGTs obtained from both the Gushantun and nearby Hani peat generally indicate that peat soil moisture, and by inference effective precipitation, increased from the early Holocene to the late Holocene in NE China. Therefore, the temperature-moisture patterns in NE China appear to be dominated by warm-dry and cold-wet alternations during the Holocene, which is largely consistent with other data from NE China. Comparisons with other proxy records from the EASM regions reveal that the reconstructed climate development differs from the Holocene moisture evolution in Southern/Eastern China and Northern China, but is consistent with the core area of Arid Central Asia such as Xinjiang. We suggest that changes in i) the intensity of the mid-latitude westerlies associated with winter insolation and EASM, ii) SST-modulated evaporation in the Japan Sea, iii) Arctic sea ice extent and iv) the shift of the Okhotsk High all could have played an important role in the out-of-phase relationship in the moisture evolution between NE China and other EASM regions and in the strong climatic similarities between NE China and the core area of arid central Asia during the Holocene.

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Figure captions

Figure 1: Location of the Gushantun peat (yellow star) and other sites in arid central Asia and the East Asian Monsoon region: Huguangyan Maar lake (Wang et al., 2007); Dahu peat (Zhou et al., 2004); Dongge Cave (Dykoski et al., 2005); Dajiuhu peat (Zhu et al., 2010); Sanbao Cave (Wang et al., 2008); Hongyuan peat (Zheng et al., 2007); Yaoxian Loess (Zhao et al., 2007); Xunyi Loess (Stevens et al., 2008); Luochuan Loess (Lu et al., 2013); Yulin Loess (Lu et al., 2013); Lake Dalianhai (Cheng et al., 2013); Lake Qinghai (Shen et al., 2005; An et al., 2012; Wang et al., 2014); Lake Gonghai (Chen et al., 2015); Lake Daihai (Xiao et al., 2004); Lake Bayancha.(Bayanchagan, Jiang et al., 2006); Lake Tianchi (Zhou et al., 2016); Lake Moon (Liu et al., 2010); Lake Sihailongwan (Stebich et al., 2015); Hani peat (Zhou et al.,2010); Xinjiang loess (Chen et al., 2016); Bayanbulak (Long et al., 2017); Lake Balikun (Tao et al., 2010); Lake Aibi (Wang et al., 2013); Lake Sayram (Jiang et al., 2013); Lake Wulungu (Liu et al., 2008). Also shown are the dominant atmospheric circulation systems: the EASM-East Asian summer monsoon, EAWM-East Asian winter monsoon, WJ-Westerly jet.

Figure 2: The age-depth model of Gushantun peats using a Bacon-depth method.

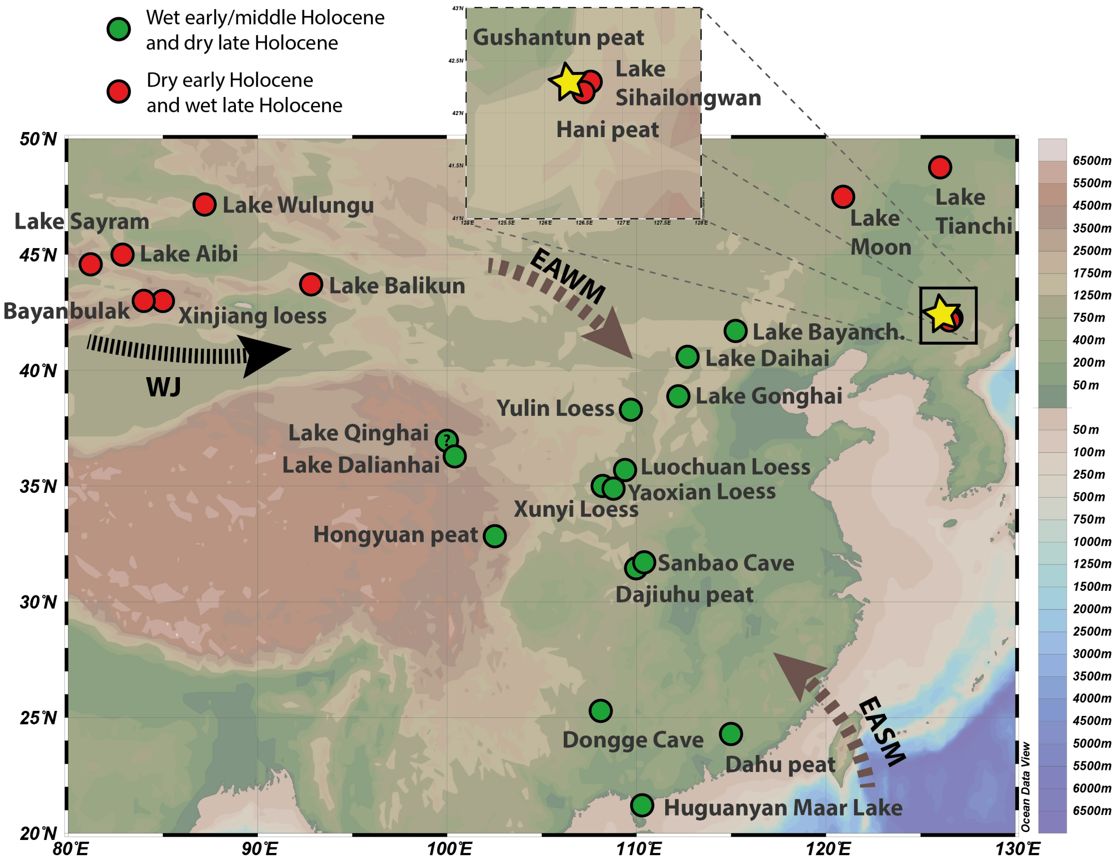
Figure 3: Comparison of reconstructed proxies based on brGDGTs in Gushantun peat sequence with other temperature variations. (a). $MAAT_{peat}$ variations in Gushantun peat. The gray line is modern MAAT (~3 °C); (b). $MAAT_{peat}$ variations in Hani peat The gray line is modern MAAT (~5 °C) (Zheng et al., 2017); (c). A pollen-derived mean warmest month (July) temperature changes (Mtwa) of the Sihailongwan lake sequence (Stebich et al., 2015. The gray bar shows lacustrine deposits at Gushantun peat.

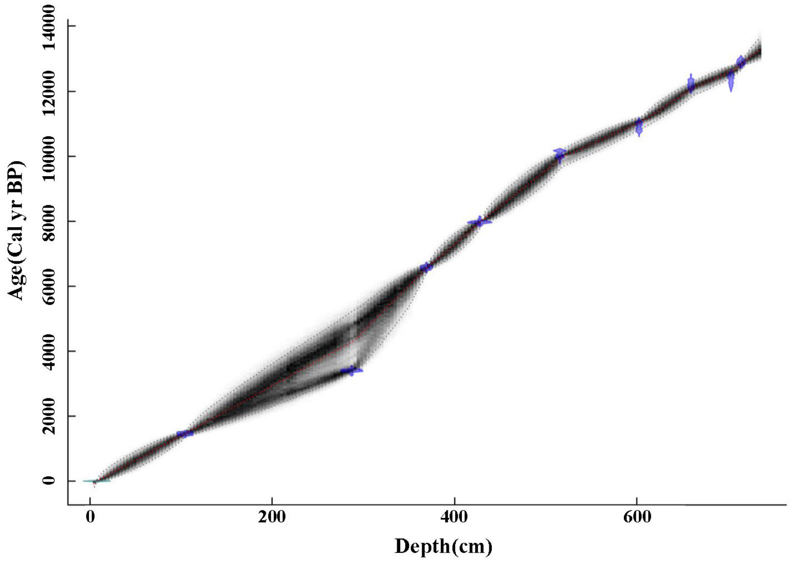
Figure 4: Reconstructed proxies in Gushantun peat sequence. (a). IR_{6me}; (b). Fractional abundance of 6-methyl brGDGTs; (c). pH in Gushantun. The gray bar shows lacustrine deposits at Gushantun peat.

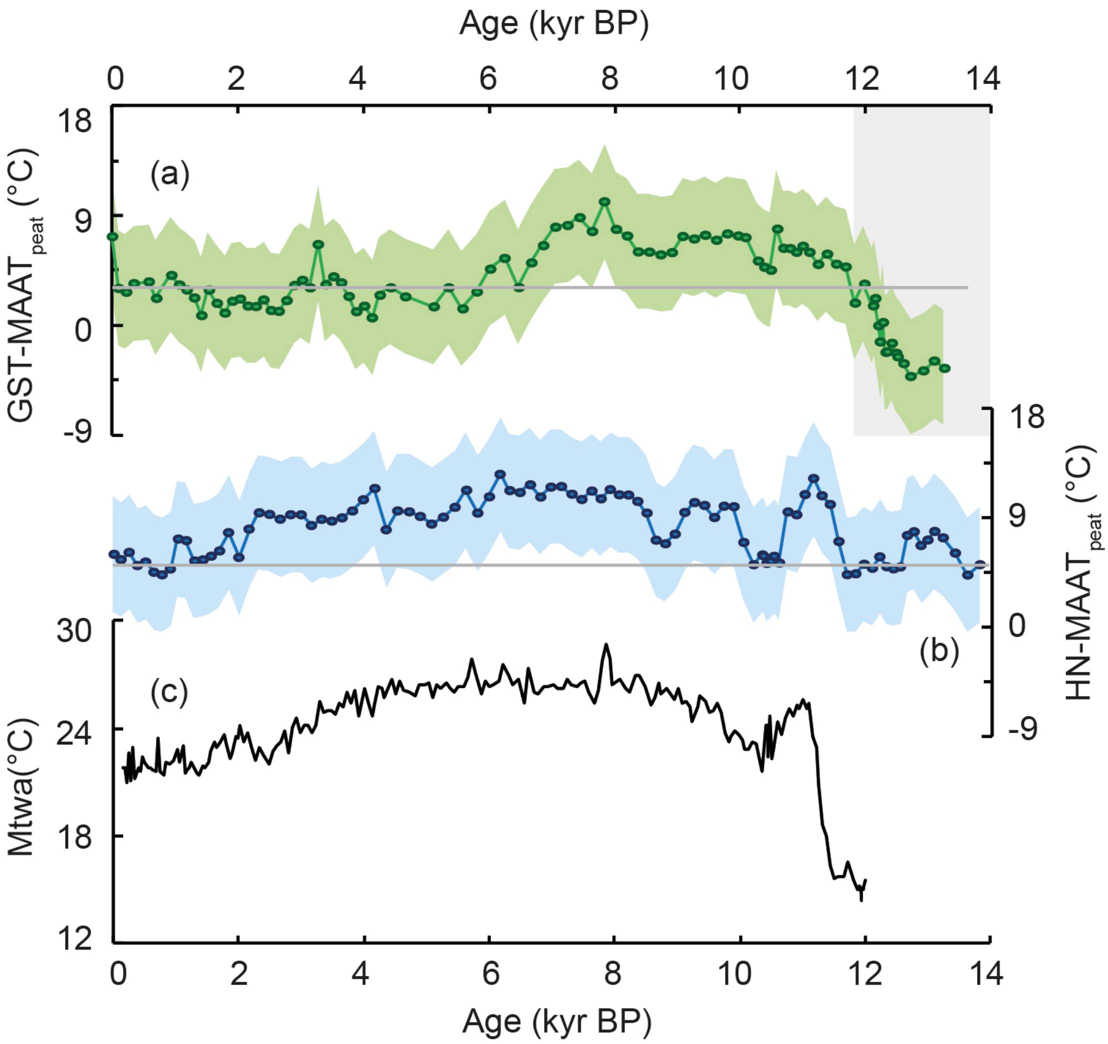
Figure 5: Comparison of Crenarchaeol and pH variations in Gushantun and Hani peat sequence. (a) Crenarchaeol concentrations in Hani; (b). pH in Hani; (c). pH in Gushantun; (d) Crenarchaeol concentrations in Gushantun.

Figure 6: Holocene moisture changes represented by pH in Gushantun peats and its comparison with winter and summer insolation and other moisture changes from NE China, the arid central Asia and other East Asian monsoon regions. (a) Northern Hemisphere summer insolation (Berger and Loutre, 1991); (b) $\delta^{18}O$ record from Dongge cave (Dykoski et al., 2005); (c) Pollen-based moisture index synthesized from the East Asian summer monsoon rainfall belt over northern China (Wang and Feng, 2013); (d) EASM index synthesized from monsoonal eastern China (Wang et al., 2010);(e) Northern Hemisphere winter insolation (Berger and Loutre, 1991); (f) Strength of the westerlies represented by the winter insolation gradient between 35°

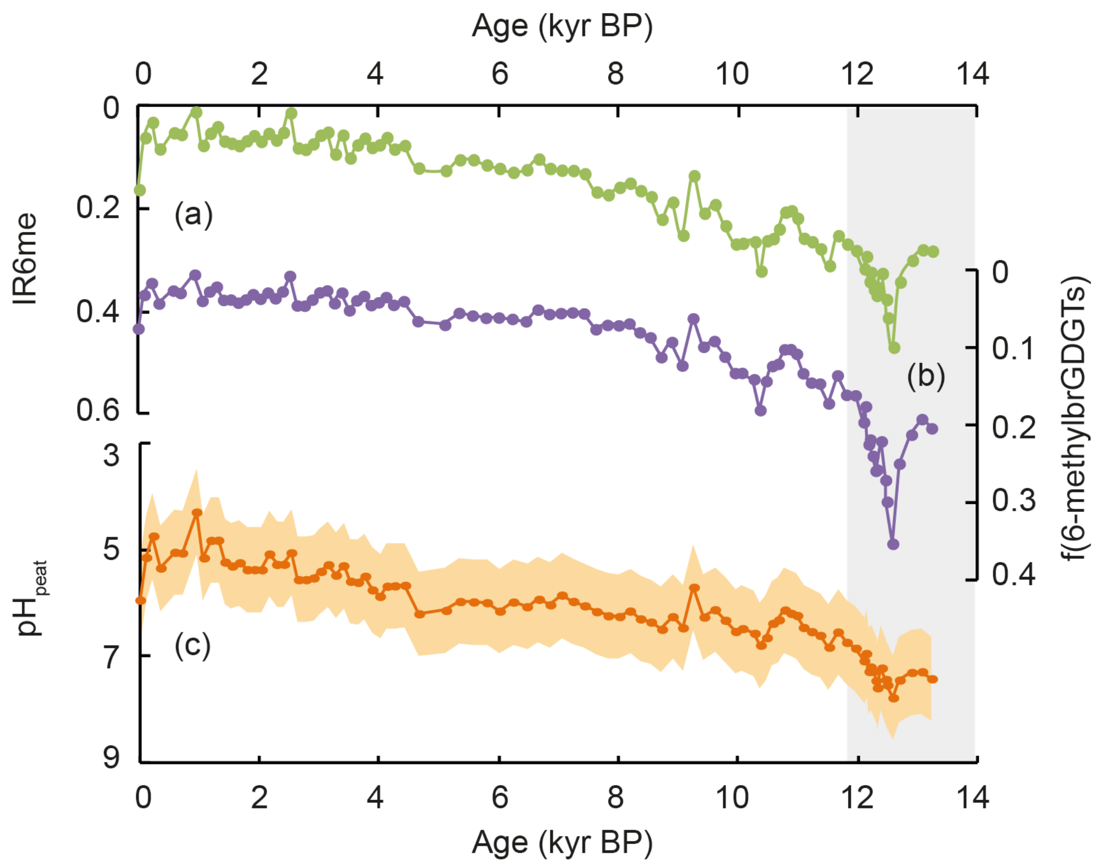
and 55ºN (Rossby et al., 1939; Chen et al., 2016); (g) The tree pollen percentages in Tianchi lake (Zhou et al., 2016); (h) The pollen-derived Pann (mean annual precipitation) in Sihailongwan Maar lake (Stebich et al., 2015); (i) pH values from Gusnantun (this study; The black line is a polynomial regression trendline.); (j) The moisture changes from the LJW10 section of the Xinjiang Loess in the core area of Arid central Asia (Chen et al., 2016); (k) Synthesis of records of moisture variations in the Xinjiang region (Wang and Feng, 2013).



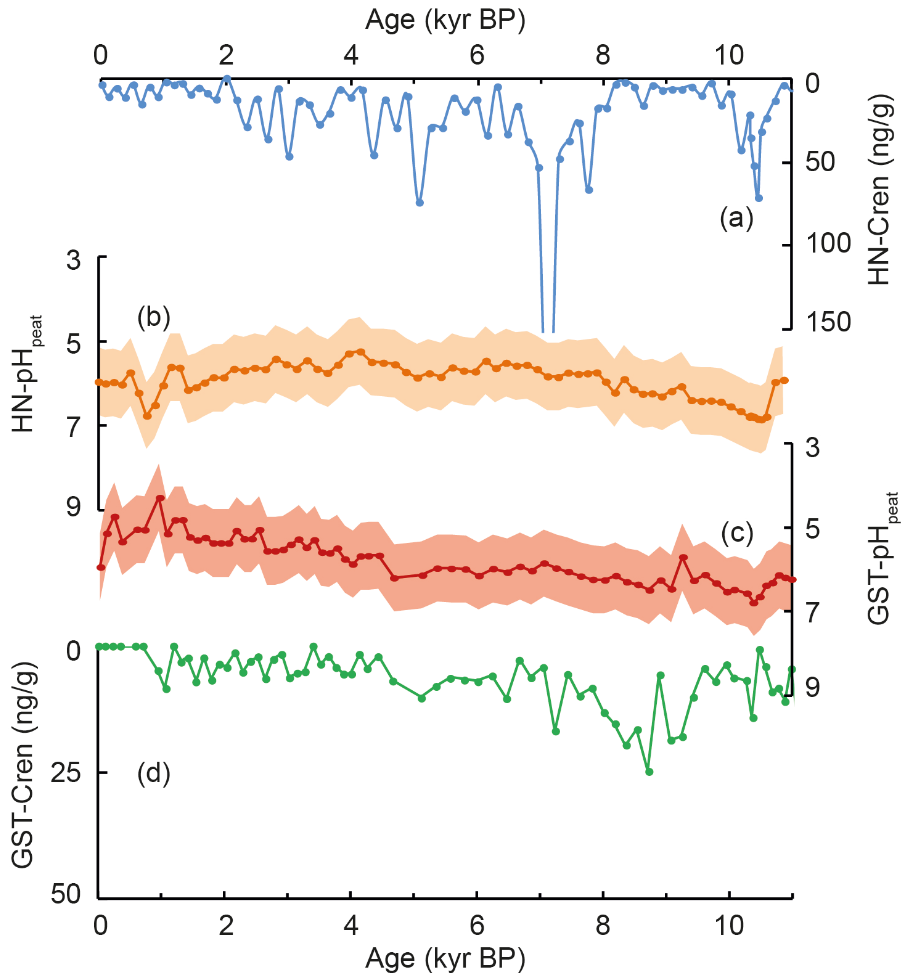

Age(Cal yr BP)
0
2000
4000
6000
8000
10000
12000
14000
0
200
400
600
Depth(cm)

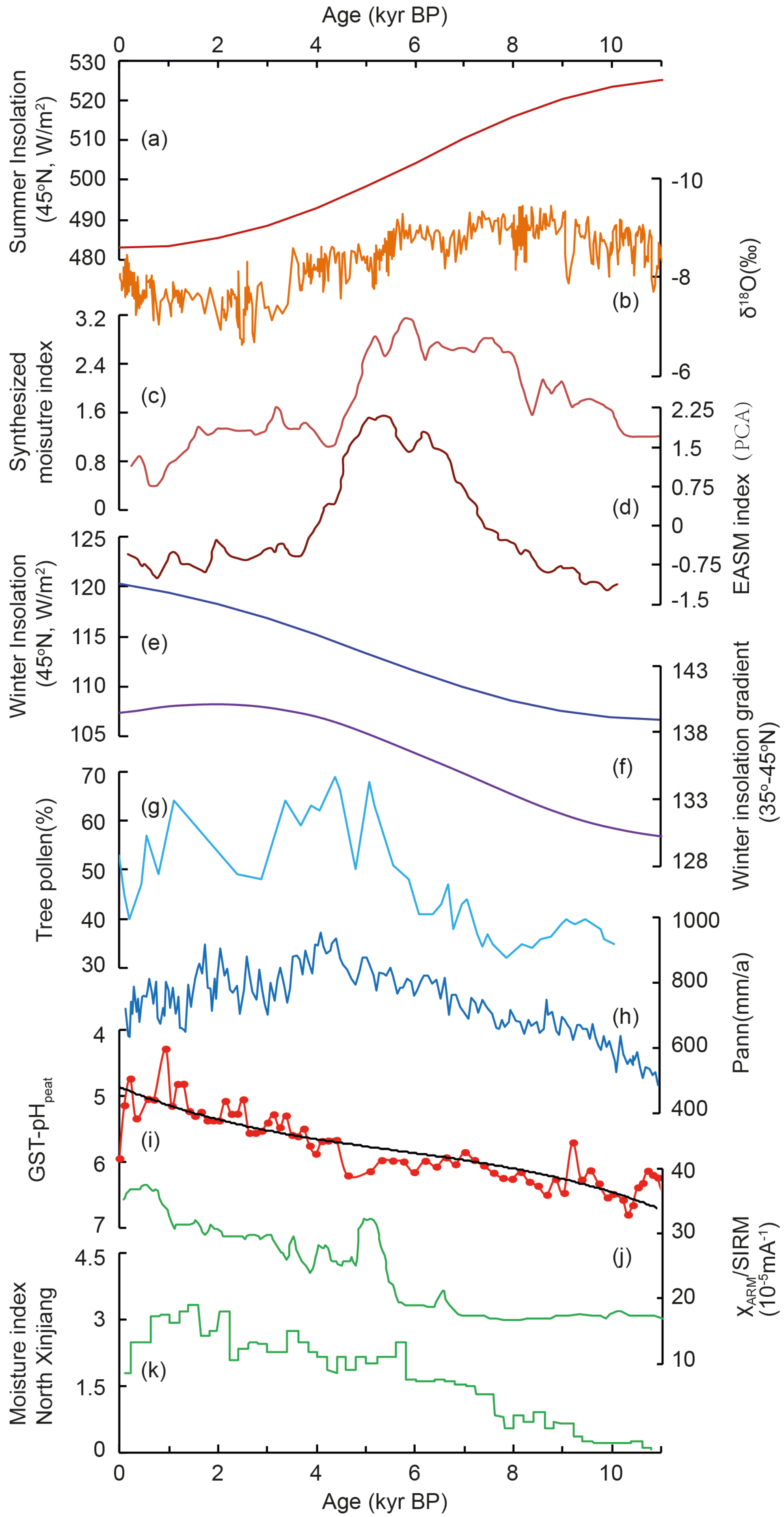

Age (kyr BP)
0
2
4
6
8
10
12
14
18
9
0
-9
(a)
GST-MAAT$_{peat}$ (°C)
HN-MAAT$_{peat}$ (°C)
(b)
30
24
18
12
(c)
Mtwa(°C)
Age (kyr BP)

966


Age (kyr BP)
0 2 4 6 8 10 12 14
IR6me
0 0.2 0.4 0.6
(a)
(b)
f(6-methylbrGDGTs)
0 0.1 0.2 0.3 0.4
pH$_{peat}$
3 5 7 9
(c)
0 2 4 6 8 10 12 14
Age (kyr BP)

967


Age (kyr BP)
0 2 4 6 8 10
0 50 100 150
HN-Cren (ng/g)
(a)
HN-pH$_{peat}$
3 5 7 9
(b)
3 5 7 9
GST-pH$_{peat}$
(c)
GST-Cren (ng/g)
0 25 50
(d)
0 2 4 6 8 10
Age (kyr BP)



Age (kyr BP)
Summer Insolation (45°N, W/m²)
(a)
δ18O(‰)
(b)
Synthesized moisutre index
(c)
EASM index （PCA）
(d)
Winter Insolation (45°N, W/m²)
(e)
Winter insolation gradient (35°-45°N)
(f)
Tree pollen(%)
(g)
Pann(mm/a)
(h)
GST-pH peat
(i)
XARM/SIRM (10-5mA-1)
(j)
Moisture index North Xinjiang
(k)
Age (kyr BP)