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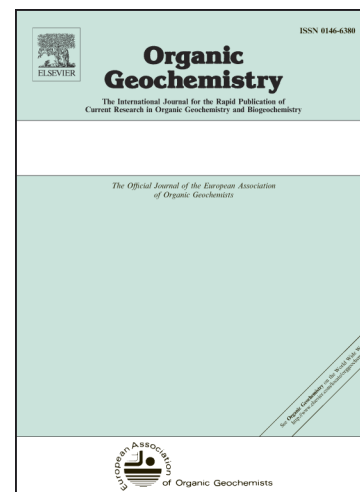
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Glacial-interglacial vegetation changes in northeast China inferred from isotopic composition of pyrogenic carbon from Lake Xingkai sediments

Weiwei Sun^{a*}, Enlou Zhang^a, Enfeng Liu^b, Jie Chang^a, Rong Chen^a, Ji Shen^{a, c*}

^a *State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, Nanjing 210008, China*

^b *College of Geography and Environment, Shandong Normal University, Jinan 250014, China*

^c *College of Earth Sciences, University of Chinese Academy of Sciences, Beijing 100049, China*

*Corresponding authors. *E-mail addresses:* wwsun@niglas.ac.cn;
jishen@niglas.ac.cn

ABSTRACT

Understanding the changes in monsoon intensity and ecosystem response at different timescales is crucial for the well-being of humans, yet the paleoclimatic interpretation of stable carbon isotope ($\delta^{13}\text{C}$) values from northeast China records is debatable. In this study, reported $\delta^{13}\text{C}$ data from 76 surface soils in northeast China are compiled, and a $\delta^{13}\text{C}$ record of pyrogenic carbon ($\delta^{13}\text{C}_{\text{PyC}}$) from Lake Xingkai in northeast

China since the last interglacial period is presented. The aim was to investigate the orbital timescale environmental implication of geological $\delta^{13}\text{C}_{\text{PyC}}$ data for northeast China. The results showed a distinct increase in $\delta^{13}\text{C}$ values of surface soils, which correlated with increasing temperature of the warmest month. Higher temperature favored the expansion of C_4 plants, while precipitation had only a weak correlation with $\delta^{13}\text{C}$ values of surface soils in the region. On an orbital timescale, the $\delta^{13}\text{C}_{\text{PyC}}$ record from Lake Xingkai generally reflected paleovegetation change, suggesting that the abundance of C_4 plants was relatively high during the warm periods, changing to almost purely C_3 plants during the cold periods. Both modern and geological analyses suggest that the climatic factor determining the $\delta^{13}\text{C}$ in northeast China was temperature of the warmest month. This is similar to the situation for mid-latitudes such as the Chinese Loess Plateau, in contrast to low latitudes such as southern China.

Keywords: Asian summer monsoon; stable carbon isotopes; C_3/C_4 plants; pyrogenic carbon; lacustrine sediments

1. Introduction

The Asian summer monsoon is an important component of the global climate system that transports heat and moisture from the warmest part of the tropical ocean to higher latitudes, playing a significant role in the socio-economic and agricultural development over a densely populated region (An et al., 2000). Northeast China is in

the current East Asian summer monsoon (EASM) margin and particularly sensitive to monsoon variation. However, there remain conflicting interpretations regarding the variation in monsoonal precipitation on decadal to orbital timescales for the region. For example, a large number of stable carbon isotope ($\delta^{13}\text{C}$) records from this sensitive zone have been developed to reconstruct the centennial to millennial abrupt monsoon change events since the last deglaciation (Hong et al., 2005; Hong et al., 2010; Chu et al., 2014; Sun et al., 2016). These records show more negative $\delta^{13}\text{C}$ values during cold conditions, suggesting that the abrupt changes in monsoon intensity are closely linked to the climatic anomalies in the Northern Hemisphere at high latitude. However, some pollen and sedimentary records from northeast China suggest that the EASM was weakened in response to the cold climate conditions at the high latitudes (Stebich et al., 2009; Xiao et al., 2009; Chen et al., 2015a). Therefore, it is important to clarify these inconsistent interpretations of paleo-records from northeast China.

The $\delta^{13}\text{C}$ data from organic matter (OM) derived from terrigenous higher plants have been widely used for paleoenvironmental and paleoclimatic reconstruction in the Asian summer monsoon region in the past two decades (Zhang et al., 2003; Vidic and Montañez, 2004; An et al., 2005; Liu et al., 2005; Zhou et al., 2012; Jia et al., 2015; Yang et al., 2015; Zhang et al., 2015; Rao et al., 2016). However, there is a lack of consistent regional climatic significance of the $\delta^{13}\text{C}$ data, examples being that surface soil $\delta^{13}\text{C}$ values correlate positively with precipitation in the central of Chinese Loess Plateau, but the correlation is the opposite of that in the sand fields of northern China

and Mongolia (An et al., 2005; Feng et al., 2008; Lu et al., 2012; Chen et al., 2015b).

In addition, the climatic factor that controls the modern $\delta^{13}\text{C}$ values in northeast China has not been well investigated despite its important geographic location.

Pyrogenic carbon (PyC) represents a wide spectrum of carbon-rich material produced either naturally or anthropogenically by combustion or pyrolysis, including partly charred biomass, charcoal, carbonaceous spherules, soot and microcrystalline graphite (Masiello, 2004; Bird and Ascough, 2012). Slightly charred biomass with low aromaticity and high reactivity is formed at low temperature, whereas particulate black carbon with high aromaticity and low reactivity is condensed at high temperature (Bird and Ascough, 2012). The $\delta^{13}\text{C}$ values of PyC ($\delta^{13}\text{C}_{\text{PyC}}$) is unlikely to be subject to significant alteration during pyrolysis, so is likely to record the original $\delta^{13}\text{C}$ characteristics of the combusted biomass (Bird and Ascough, 2012; Liu et al., 2013; Wang et al., 2013b). It has been widely used as a proxy to reconstruct paleovegetation and palaeoclimate in southern China (Jia et al., 2003; Zhou et al., 2014, 2017; Sun et al., 2015; Zhang et al., 2015; Sun et al., 2017). In order to clarify the environmental implications of geological $\delta^{13}\text{C}_{\text{PyC}}$ data for northeast China, we have compiled reported $\delta^{13}\text{C}$ values of top soil OM from northeast China under different environments (Rao et al., 2008, 2017; Lu et al., 2012; Chen et al., 2015b; Jia et al., 2016). We then present a $\delta^{13}\text{C}_{\text{PyC}}$ record from Lake Xingkai in northeast China, covering the last interglacial period and compared the record with a range of other $\delta^{13}\text{C}$ records from the Asian summer monsoon region on an orbital timescale.

2. Study region

Northeast China (115°E to 135°E and 38°N to 53°N, Fig. 1a and b) is characterized by plains and sand fields separated by three major mountain systems: The Great Khingan Mountains, the Lesser Khingan Mountains and the Changbai Mountains. The western part is covered mainly by the Hulun Buir sand field and Otindag sand field, and the south-central part and the northeast corner comprise Songnen/Liaohe Plain and Sanjiang Plain, respectively. The climate is influenced by both the EASM and the polar climate system, with a semi-humid to arid pattern. During the winters, the cold and dry northwesterly airflows generated by the Mongolian High prevail in the study region, while during the summers, the warm and moist southerly air masses driven by the pressure gradient between the northwest Pacific and the Eurasian continent interact with cold air from the northwest and produce most of the annual precipitation. The mean annual temperature (MAT) ranges from -4.7 to 10.7 °C and the mean summer temperature ranges from 14.7 to 23.8 °C. Mean annual precipitation (MAP) decreases from the southeast (1000 mm) to the northwest (ca. 150 mm), 80% of which falls in the period of May to September. The vegetation in northeast China is characterized by arid to semi-arid grassland and shrubs in the sand fields, the plains being covered by mixed C₃ and C₄ plants and temperate mixed forest dominated by C₃ plants prevailing in the mountain regions (Qian et al., 2003). Most of the C₄ plants in northeast China belong to the Poaceae, Cyperaceae, Amaranthaceae and Chenopodiaceae; the habitats are dry steppe and saline grassland (Yin and Wang, 1997).

Lake Xingkai (44°32"- 45°21"N, 131°58"- 132°51"E, 65 m above sea level, Fig. 1c) is in a graben basin that formed in the Cenozoic (Wan and Zhong, 1997). It is the largest freshwater lake in northeastern Asia, with a surface area of 4190 km² and a maximum water depth of 10 m. It has a catchment of 16,890 km² and is fed mainly by five rivers. It has only one outflowing river (the Songacha River) in the northeast, which flows into the Ussuri River (Fig. 1c). The meteorological records from the adjacent Jixi meteorological station (ca.120 km east of the lake) indicate a MAT of 3 °C, a maximum monthly mean temperature of 21 °C in July and a minimum monthly mean temperature of -18 °C in January. The MAP is ca. 540 mm. The modern natural vegetation of the lake catchment is categorized as temperate mixed forest. However, there is a strong altitudinal zonation of vegetation in the region: the landscape is occupied by helophytes, composed primarily of *Alnus sibirica*, *Salix brachypoda*, *Betula fruticosa*, *Spiraea salicifolia*, *Phragmites communis*, *Carex appendiculata*, *Carex lasiocarpa* and *Carex pseudocuraica* below ca. 200 m, while in the mountainous region, the vegetation is dominated by broadleaved forest and *Pinus koraiensis* forest.

3. Material and methods

3.1. $\delta^{13}\text{C}$ data for topsoil and climatic data

All the $\delta^{13}\text{C}$ data for topsoil samples were obtained directly from the publications or from the related supplementary information (Fig. 1b). The samples were collected

at a depth of ca. 2,- 4 cm, where there was no significant disturbance from human activity. The climate variables for each sampling site were estimated from WorldClim version 2 (<http://worldclim.org/version2>), which provides gridded climate data at a spatial resolution of 1 km × 1 km. To identify the relationship between $\delta^{13}\text{C}$ data of topsoil and climate, we used MAT, MAP, mean temperature and precipitation for the growing season (May to October).

3.2. Coring and dating

In summer of 2008, two parallel and overlapping sediment cores (XK08-A1 and XK08-A2) were collected at a water depth of 7 m near the China-Russia boundary (Fig. 1c, 45°12'21"N, 132°30'33" E) using a UWITEC piston corer. In the laboratory, core correlation between XK08-A1 (308 cm) and XK08-A2 (336 cm) was carried out using surface scanning magnetic susceptibility. Core XK08-A1 was split under subdued red light in the dark room and sectioned at 5-10 cm intervals for optically stimulated luminescence dating. Core XK08-A2 was split into two halves and optically described in the field. The cores were composed mainly of fine-grained, grayish, minerogenic, organic-poor sediments. One half of XK08-A2 was used here and sectioned at 1 cm intervals, and the samples were stored at 4 °C in the repository prior to analysis. Its chronology is based on the correlation of the magnetic susceptibility with XK08-A1, which was dated using the OSL method (Fig. 2a, Long et al., 2015; Sun et al., 2018). It was supported by an AMS ^{14}C age from twigs at 64 cm depth, which was dated to ca. 26.5 cal ka BP in Beta Analytic Inc., Miami, USA. The age-depth model for the upper 70-cm of Lake Xingkai sediment is produced by

Bacon software (Fig. 2b).

3.3 Extraction of pyrogenic carbon and analysis of $\delta^{13}\text{C}_{\text{PyC}}$

Samples at 1 cm intervals above 70 cm depth and 4 cm intervals below this were used for the analysis of $\delta^{13}\text{C}_{\text{PyC}}$, yielding a total of 136 samples. Due to that continuum containing slightly charred biomass at low temperature, the PyC in the bulk sediment was extracted using the dichromate oxidation method of Lim and Cachier (1996): Freeze-dried bulk sample (ca. 1.0 g) was weighed and treated with HCl (3 mol/l); a mixed solution of 3 mol/l HCl and 22 mol/l HF (1: 2, v/v), and HCl (10 mol/l) was added to remove carbonate and part of the silicate fraction. The acid-treated sample was then oxidized using a mixture of $\text{K}_2\text{Cr}_2\text{O}_7$ (0.2 mol/l) and H_2SO_4 (2 mol/l) at 55 °C for 60 h to remove soluble OM and kerogen, the residual refractory carbon being regarded as PyC. The dried sample was then crushed to powder using an agate mortar. The $\delta^{13}\text{C}_{\text{PyC}}$ value was determined using a Thermo Delta Plus mass spectrometer coupled to an elemental analyzer (Flash EA 1112) at the Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences. The carbon isotope results are expressed in conventional delta notation, as per mil (‰) deviation from the Vienna Peedee Belemnite (VPDB) standard. The calibration and assessment of the reproducibility and accuracy of the isotopic analyses were based on replicate analyses of external working standard materials, and the precision was better than 0.2‰.

4. Results

The $\delta^{13}\text{C}$ values ranged from -27.7 to -19.1‰ with a mean of -23.9‰. Regression analysis showed that there was a significant and moderate correlation of $\delta^{13}\text{C}$ values with MAT and temperature of the warmest season (Fig. 3a and b, $y = 0.261x - 24.345$, $r^2 = 0.28$, $p < 0.001$ and $y = 0.422x - 32.586$, $r^2 = 0.30$, $p < 0.001$, respectively), and a significant negative correlation between the topsoil $\delta^{13}\text{C}$ values and MAP and growing season precipitation (Fig. 3c and d, $y = -0.004x - 22.124$, $r^2 = 0.07$, $p < 0.05$ and $y = -0.004x - 22.096$, $r^2 = 0.06$, $p < 0.05$). The results show clearly that the relationship between topsoil $\delta^{13}\text{C}$ values and temperature is stronger than the relationship with precipitation.

As shown in Fig. 4a, the $\delta^{13}\text{C}_{\text{Pyc}}$ values from the lake exhibited wide variation from -29.1 to -21.7‰, with a mean of -24.4‰ along the 336 cm profile. On the basis of the correlated age with OSL dating, the sequence could be divided into three stages: the $\delta^{13}\text{C}_{\text{Pyc}}$ values tended to be more positive in the interglacial stages (MIS 5 and 1) than the last glacial period (MIS 4 to 2). During the last interglacial period, $\delta^{13}\text{C}_{\text{Pyc}}$ values ranged from -24.8 to -21.7‰, with a mean of -23.3‰. During the last glacial period, they decreased significantly during MIS 4 and early MIS 3, ranging from -27.1 to -23.9‰, with a mean of -24.9‰, while they ranged from -29.1 to -24.0‰ also with a mean of -24.9‰, during MIS 2. During the Holocene, the values fluctuated frequently, ranging from -28.6 to -22.0‰, with a mean of -24.1‰.

5. Discussion

5.1. Climatic influence on $\delta^{13}\text{C}$ composition of topsoil OM

At a given location, $\delta^{13}\text{C}$ values for topsoil samples inherited mainly the overlying vegetation isotopic signature, and were potentially affected by the early decomposition of OM. Generally speaking, terrestrial higher plants can be classified into two principal functional forms: Calvin-Benson (C_3 plants) and Hatch-Slack (C_4 plants), according to the carbon fixation pathway in the process of photosynthesis (Farquhar et al., 1989). The $\delta^{13}\text{C}$ values of plants using the C_3 pathway, such as all trees and most shrubs, range from -37‰ to -20‰, with a mean of -27‰, omitting the samples in low-light tropical forest under modern atmospheric CO_2 conditions (Farquhar et al., 1989; Kohn, 2010). C_4 plants, including most warm season grasses/sedges have, however, higher $\delta^{13}\text{C}$ values in the range -16‰ to -10‰, with a mean of -13‰ (Farquhar et al., 1989). Similar values were reported in north China, with -26.7‰ for C_3 plants and -12.8‰ for C_4 plants in the Chinese Loess Plateau (Wang et al., 2008); and with -27.8 ± 1.4 ‰ for C_3 plants and -13.5 ± 1.9 ‰ for C_4 plants in the region with 400 mm isoline of mean annual precipitation (Wang et al., 2013). Field and laboratory investigations suggested that ^{13}C enrichment would occur during decomposition of the plant residue in soil, with a magnitude of 1- 3‰ (Connin et al., 2001; Wang et al., 2008). In this study, the mean $\delta^{13}\text{C}$ values of the topsoil OM is about -24‰, with the potentially carbon isotopic fractionation when plant residue decomposes in soil, suggesting a predominance of C_3 plants and low abundance of C_4 plants in northeast China.

The abundance of C₃ and C₄ plants can be estimated by applying the following isotope mass balance equations, using the $\delta^{13}\text{C}$ values of the topsoil OM proposed by Wang et al. (2008):

$$\delta^{13}\text{C} = \delta^{13}\text{C}_{\text{C}_4} \times \text{C}_4\% + \delta^{13}\text{C}_{\text{C}_3} \times (100\% - \text{C}_4\%) + 1.8 \quad (1)$$

$$\text{C}_4\% = (\delta^{13}\text{C} - 1.8 - \delta^{13}\text{C}_{\text{C}_3}) / (\delta^{13}\text{C}_{\text{C}_4} - \delta^{13}\text{C}_{\text{C}_3}) \times 100 \quad (2)$$

where $\delta^{13}\text{C}_{\text{C}_3}$ is the global mean value of $\delta^{13}\text{C}$ for C₃ plants, $\delta^{13}\text{C}_{\text{C}_4}$ the end member value of $\delta^{13}\text{C}$ for C₄ plants, $\delta^{13}\text{C}$ the measured $\delta^{13}\text{C}$ value of OM in topsoil and 1.8 the mean value for carbon isotope fraction during decomposition of OM; C₄% is the relative abundance of C₄ plants, and 100% - C₄% is the relative abundance of C₃ plants. The estimated relative abundance of C₄ plants in northeast China ranges from 0 to 43.6%, with a mean of 11.0%. The result is consistent with the study of modern C₄ biomass in northeast China, which showed that C₄ plants contributed almost 40% of the total biomass in the dry grassland communities and are rare in the alpine forest (Han et al., 2006). Therefore, soil OM preserves the large isotopic contrast in C₃/C₄ plants, despite minor fractionation between topsoil and the corresponding overlying vegetation.

Temperature and precipitation are the most important climatic factors that can affect the carbon isotopic fractionation during the photosynthetic processes of both C₃ and C₄ plants. More negative $\delta^{13}\text{C}$ values of C₃ plants normally occur under relatively humid conditions, resulting from the relatively high stomatal conductance of C₃ plants, therefore increasing intercellular partial pressure of CO₂ under such conditions

(Farquhar et al., 1989). Globally, the $\delta^{13}\text{C}$ values of C_3 plants generally decrease with increasing MAP ($-0.2\text{‰}/100\text{ mm}$), while C_4 plants show a weak positive relationship with MAP (only $0.06\text{‰}/100\text{ mm}$). However, the coefficient is largely dependent on plant species (Rao et al., 2017). Wang et al. (2008) measured the $\delta^{13}\text{C}$ values of hundreds of C_3 species in north China and found that the value decreased 0.4‰ for every 100 mm increase in MAP. In this study, the $\delta^{13}\text{C}$ value of topsoil OM in northeast China also decreases 0.4‰ for every 100 mm increase in MAP; however, that is insignificant. Furthermore, studies of C_3 plants showed mostly negative correlations between $\delta^{13}\text{C}$ values and MAT, without controlling MAP (Rao et al., 2017). After correction for the effect of MAP, temperature exerts only a minimal effect on $\delta^{13}\text{C}$ values of C_3 plants, with $0.1\text{‰}/^\circ\text{C}$ in north China (Diefendorf et al., 2010; Wang et al., 2013). Therefore, the effect of the main climatic factors on $\delta^{13}\text{C}$ values of pure C_3 plants could not explain the relationship between $\delta^{13}\text{C}$ values of topsoil OM and temperature and precipitation in northeast China.

The significant positive correlation between temperature and the $\delta^{13}\text{C}$ values of topsoil samples is consistent with studies of the regions with mixed C_3/C_4 vegetation or pure C_4 vegetation, such as the Chinese Loess Plateau (An et al., 2005; Rao et al., 2017). However, the weak negative correlation between precipitation and $\delta^{13}\text{C}$ values of topsoil samples in northeast China contrasts with the significant negative correlation between rainfall and the $\delta^{13}\text{C}$ values of topsoil samples in the low latitudes (Rao et al., 2010 and 2017). There is a general agreement that C_4 plants are more competitive than C_3 plants in a warm and dry environment (Farquhar et al., 1989;

Sage et al., 1999); however, higher water use efficiency of C₄ plants than C₃ plants cannot explain the weak negative correlation between precipitation and the δ¹³C values of topsoil samples. One of the alternative interpretations is that the growth of C₄ plants can be limited in regions with a cold climate (Sage et al., 1999). Globally, only 3 to 5 modern C₄ species are found in the region above 60 °N and the most typical C₄ vegetation is distributed in relatively hot and dry areas in the low latitudes (Sage et al., 1999). Investigations of C₄ plants in the Qinghai-Tibetan Plateau also demonstrated that very few C₄ species exist in the high altitude regions (Wang et al., 2004; Li et al., 2009). Laboratory data also indicate a close correlation between the distribution of C₄ plants and growing season temperature, and C₄ photosynthesis is inhibited in regions where the mean growing season temperature is < 16 °C under modern atmospheric CO₂ conditions (McWilliam and Naylor, 1967). The summer temperature in northeast China ranges from 14.7 to 23.8 °C, above the threshold temperature of 16 °C on average. This possibly explains the minor contribution of C₄ plants in this region, and the abundance of C₄ plants positively correlated with the temperature of the warmest month temperature. In addition, different carbon isotope fractionation response of C₃ and C₄ plants to precipitation may also weaken the correlation between rainfall and the δ¹³C values of topsoil samples in northeast China.

5.2. Variation in relative abundance of C₃/C₄ plants in the late Quaternary

In natural burns and controlled field burns, the carbon isotope fractionation of PyC from C₃ plant combustion varies from -3 to 3‰ with a mean of -0.3 ± 1.0 ‰, while the fractionation for PyC from C₄ plants combustion ranges from -10 to 3‰

with a mean of $-1.7 \pm 2.4\%$ (Wang et al., 2013b). This may be attributed to differences in the proportion of isotopically distinct volatile and refractory compounds between plants and pyrolysis temperature, and the more negative fractionation for PyC from C_4 plants may result from the more depleted carbon being protected in phytoliths (Krull et al., 2003; Das et al., 2010; Bird and Ascough, 2012). In general, biochemical fractions of plant material such as lignin, cellulose and lipids are depleted in ^{13}C compared with the whole plant, while hemicellulose, sugars, amino acids and pectin are enriched in ^{13}C (Deines, 1980). For example, monosaccharides were generally enriched in ^{13}C by 1 to 16% compared with lipids within single organisms (Van Dongen et al., 2002). Thus, the preferential loss of ^{13}C -enriched biochemical fractions of plants during biomass burning might result in depletion of ^{13}C in the PyC (Das et al., 2010). Some recent work showed that waxes and other paraffinic components can survive dichromate oxidation, suggesting that the method might not have completely removed organics (Knicker et al., 2007; Ascough et al., 2008). However, this might also be a result of the refractory compounds retained within the charcoal structure due to incomplete burning. Although the carbon isotope fractionation of plants during pyrolysis is variable, $\delta^{13}\text{C}_{\text{PyC}}$ and $\delta^{13}\text{C}$ values of the bulk soil OM is strongly positive and the difference between them is also within the small range -1.5 to 1.3% in the Chinese Loess Plateau (Liu et al., 2013). Therefore, $\delta^{13}\text{C}_{\text{PyC}}$ may generally reflect $\delta^{13}\text{C}$ of paleovegetation change at a given locality.

On an orbital timescale, large changes in temperature, precipitation, $p\text{CO}_2$ level and $\delta^{13}\text{C}$ value of atmospheric CO_2 may directly influence the $\delta^{13}\text{C}$ values of plant

tissues. The $\delta^{13}\text{C}$ values of C_3 plants might increase with increasing temperature (ca. $0.1\text{‰}/^\circ\text{C}$) and decrease with increasing precipitation (ca. $0.4\text{‰}/100\text{ mm}$) in northern China (Wang et al., 2013a). However, after correction of both temperature and precipitation, climate exerts a negligible effect on the $\delta^{13}\text{C}$ values of C_3 plants (Wang et al., 2013; Yang et al., 2015). Due to the lack of long term quantitative climatic data for northeast China, we suggest that the effect of the response of $\delta^{13}\text{C}$ values of plants to different climatic conditions is also insignificant in this study. During the last 130 ka, $p\text{CO}_2$ level in the interglacial periods was ca. 80 ppmv higher than in the last glacial period, and $p\text{CO}_2$ has increased by 70 ppmv since the industrial revolution (Fig. 4e, Friedli et al., 1986; Monnin, 2001, 2004). Although some studies suggested a significant negative relationship between $p\text{CO}_2$ and $\delta^{13}\text{C}$ of C_3 plants (Feng et al., 1995; Schubert and Jahren, 2012), the estimated depletion of $\delta^{13}\text{C}$ values of C_3 plants is close to the $\delta^{13}\text{C}$ value of atmospheric CO_2 , which has decreased during the last two centuries due to fossil combustion and vegetation destruction (Friedli et al., 1986; Keeling et al., 2017). Therefore, we ignored the effect of changes in $p\text{CO}_2$ on the $\delta^{13}\text{C}$ value of C_3 plants. The $\delta^{13}\text{C}$ value of atmospheric CO_2 has decreased to -8.4‰ recently, which is about 2.0‰ lower than that during the pre-industrial period (Keeling et al., 2017). Hence, the mean $\delta^{13}\text{C}$ value of C_3 and C_4 plants would increase by 2.0‰ during the period before the industrial revolution. The $\delta^{13}\text{C}$ value of atmospheric CO_2 and fractionation during pyrolysis, the $\delta^{13}\text{C}$ value of PyC derived from C_3 would become $-25.3 \pm 1.7\text{‰}$, while the corrected $\delta^{13}\text{C}$ value for PyC derived from C_4 plants is $-12.7 \pm 3.1\text{‰}$. Therefore, the abundance of C_3 and C_4 plants can be

estimated by the modified equation using the $\delta^{13}\text{C}_{\text{PyC}}$ values:

$$C_4\% = (\delta^{13}\text{C}_{\text{PyC}} - \delta^{13}\text{C}_{\text{PyC-C}_3}) / (\delta^{13}\text{C}_{\text{PyC-C}_4} - \delta^{13}\text{C}_{\text{PyC-C}_3}) \times 100$$

(3)

where $\delta^{13}\text{C}_{\text{PyC-C}_3}$ is the end member value of $\delta^{13}\text{C}$ for PyC derived from C_3 plants, $\delta^{13}\text{C}_{\text{PyC-C}_4}$ the end member value of $\delta^{13}\text{C}$ for PyC derived from C_4 plants and $\delta^{13}\text{C}_{\text{PyC}}$ the measured $\delta^{13}\text{C}$ value of PyC in Lake Xingkai sediment.

The estimated change in the relative abundance of C_4 plants since the last interglacial period is shown in Fig. 4b. The long term trend suggests a C_4 plant expansion during warm periods and purely C_3 plants during cold periods. During the last interglacial period, the relative abundance of C_4 plants ranged from 3.8 to 28.7% with a mean of 16.3%, suggesting a mixed C_3/C_4 plant ecosystem but dominated by C_3 plants in and around the catchment of Lake Xingkai. During the last glacial period, the mean $\delta^{13}\text{C}_{\text{PyC}}$ values were nearly same as $\delta^{13}\text{C}_{\text{PyC-C}_3}$, indicating that the land was nearly covered by pure C_3 plants, with only about 4.8% C_4 plants. During the Holocene, C_4 plants reappeared with an abundance up to 26.3% and a mean of 11.7%. The accuracy of the estimates could also be influenced by the uncertainty in the $\delta^{13}\text{C}$ values of both C_3 and C_4 plants during the pyrolysis process. However, our estimate of the C_4 plant abundance during the interglacial periods is close to the modern abundance in the plains and grasslands in northeast China (Han et al., 2006).

5.3. Regional comparison of C_3/C_4 plant evolution

The increase in the relative abundance of C_4 plants from northeast China since

the last glacial maximum is consistent with the record of compound-specific carbon isotope composition of long chain *n*-alkanes ($\delta^{13}\text{C}_{n\text{-alkanes}}$) from Huola Basin and Wudalianchi in Songnen Plain. These data show that the *n*-alkanes were more ^{13}C depleted during the late glacial period (Fig. 5b, Wei et al., 2015; Wang et al., 2017). Similarly, the record of $\delta^{13}\text{C}_{n\text{-alkanes}}$ from the Sea of Japan also shows that *n*-alkanes are depleted in ^{13}C by ca. 1‰ in colder climates than in a warmer climate (Yamada and Ishiwatari, 1999). In the Chinese Loess Plateau, the relative abundance of C_3 and C_4 plants has been extensively investigated using $\delta^{13}\text{C}_{n\text{-alkanes}}$ or $\delta^{13}\text{C}$ of bulk OM from the loess-paleosol profiles during the last decades (Fig. 5c, Zhang et al., 2003; Vidic and Montañez, 2004; Liu et al., 2005; Yang et al., 2015). These records show that the $\delta^{13}\text{C}$ values along a temporal sequence were more positive in paleosol layers and more negative in loess layers. Along a spatial gradient, the estimated relative abundance of C_4 plants increased from < 5% in the northwest to 10-20% in the southeast during the last glacial maximum. During the mid-Holocene, the estimated relative abundance of C_4 plants increased from 10% to 20% in the northwest to > 40% in the southeast (Yang et al., 2015). However, in the low latitudes of the Asian summer monsoon regions, decreases in the relative abundance of C_4 plants from the last glacial maximum to the Holocene were recorded by way of the $\delta^{13}\text{C}_{\text{PyC}}$ record from lacustrine sediments in southwest China (Fig. 5d, Zhang et al., 2015), agreeing well with the $\delta^{13}\text{C}_{n\text{-alkanes}}$ records from the northern Bay of Bengal and the northern South China Sea (Fig. 5e and f, Zhou et al., 2012; Contreras-Rosales et al., 2014). The different glacial-interglacial vegetation changes between the mid and low latitudes

can be attributed mainly to a different response of C₄ plants to the regional climate change instead of global *p*CO₂ (Huang et al., 2001; Rao et al., 2017).

As discussed above, growing season temperature might be the dominant factor controlling the distribution and relative abundance of C₄ plants in mid latitudes (Rao et al., 2017). During the last glacial maximum, pollen records for south China suggest that the temperature was 2- 3 °C lower than at present (Wang et al., 2012; Chen et al., 2014), while the reconstructed summer air temperature based on gastropods, pedogenic carbonate, phytoliths and fossil branched tetraether membrane lipids of soil bacteria from the Chinese Loess Plateau suggest a 6- 7 °C cooling in the summer (Fig. 4d, Lu et al., 2007; Eagle et al., 2013; Peterse et al., 2014). The magnitude and pattern of cooling during the late glacial period is in agreement with the paleoclimate modeling, suggesting in the Northern Hemisphere, 5- 10 °C cooling in the mid high latitudes, and 2- 5 °C cooling in the tropical continent and the ocean (Braconnot et al., 2007). Therefore, we infer that the growing season temperature might be lower than the threshold temperature in northeast China, but still high enough for the growth of C₄ plants at low latitudes. During the last glacial period, the weakened Asian summer monsoon and lower temperature led to a southward shift of C₄ plants, and their general disappearance from northeast China. In contrast, in the low latitudes, weakened Asian summer monsoon and low *p*CO₂ were favorable for the expansion of C₄ plants during the last glacial maximum, and the vegetation composition in the Holocene (dominated by C₃ plants) was driven mainly by the strengthening of the Asian summer monsoon (Fig. 5d to f, Zhou et al., 2012; Contreras-Rosales et al., 2014;

Zhang et al., 2015).

6. Conclusions

We compiled reported $\delta^{13}\text{C}$ values for topsoil OM from northeast China and presented a $\delta^{13}\text{C}_{\text{Pyc}}$ record from Lake Xingkai in northeast Asia since the last interglacial period in order to investigate the environmental implication of geological $\delta^{13}\text{C}_{\text{Pyc}}$ data for northeast China. The results show that warmest month temperature is the dominant climatic factor determining the $\delta^{13}\text{C}$ values of surface soil, with higher temperature favoring the expansion of C_4 plants, while precipitation had only a weak correlation with $\delta^{13}\text{C}$ values of surface soils in northeast China. On an orbital timescale, the $\delta^{13}\text{C}_{\text{Pyc}}$ record from Lake Xingkai suggests that the abundance of C_4 plants was relatively high during warm periods and ranged to almost purely C_3 plants during cold periods. Both modern and geological analysis suggest that the climatic implication of $\delta^{13}\text{C}$ of OM derived from terrestrial higher plants in northeast China is similar to that of the Chinese Loess Plateau, in contrast to that for low latitudes.

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Fig. 1. (a) Location of Lake Xingkai (1, triangle) and the other paleoclimatic sites (circles) mentioned in Fig. 4. Arrows indicate the dominant atmospheric circulation systems in the region. 2, Wudalianchi in northeast China (Wang et al., 2017); 3, Xifeng profile in the Chinese Loess Plateau (Liu et al., 2005); 4, Lake Tengchongqinghai in southwest China (Zhang et al., 2015); 5, Core MD05-2905 in the South China Sea (Zhou et al., 2012) and 6, Core SO188-342KL in the Bay of

Bengal (Contreras-Rosales et al., 2014). (b) Map of study region showing locations of reported $\delta^{13}\text{C}$ values of surface soils in northeast China, and (c) location of coring site in Lake Xingkai.

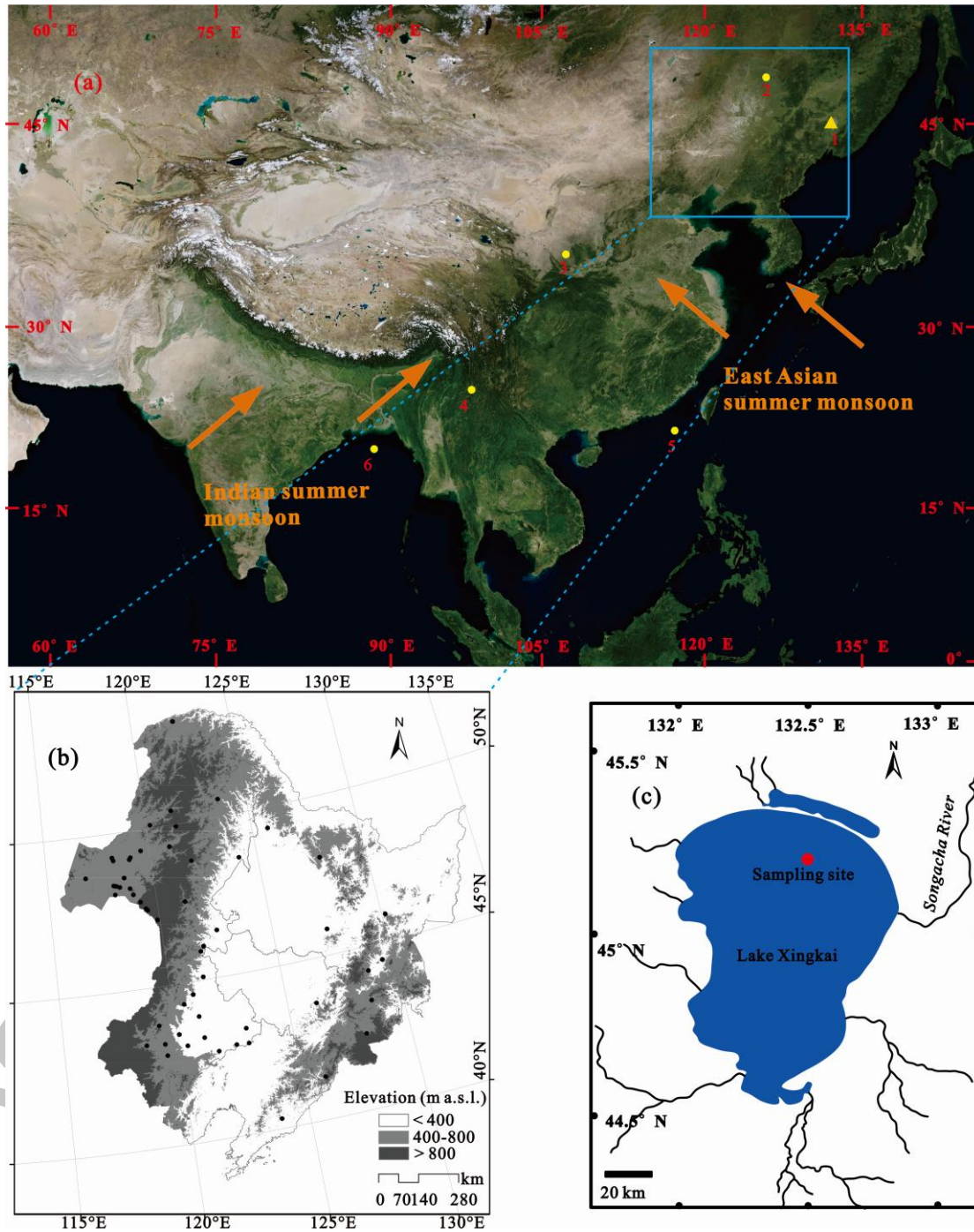
Fig. 2. (a) Stratigraphy correlation of core XK08-A2 (red), XK-1 (blue), and XK08-A1 (purple), dated using the optically stimulated luminescence method (Long et al., 2015; Sun et al., 2018); (b) Age-depth model for the Lake Xingkai upper 70-cm sediment produced with Bacon software. Dotted lines indicate the 95% confidence range and the solid line indicates the weighted mean ages for each depth;

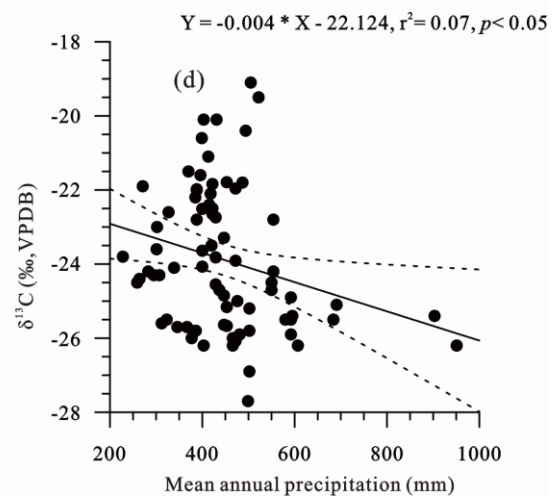
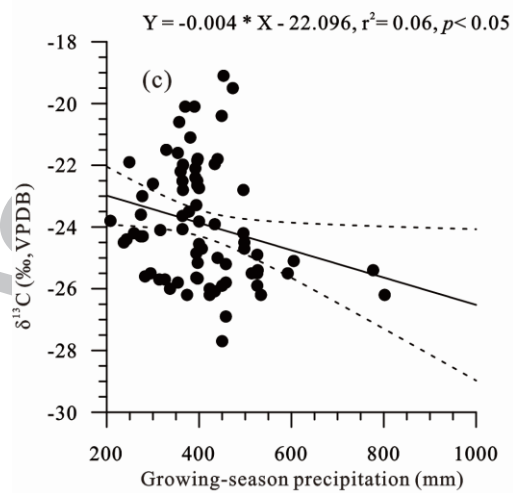
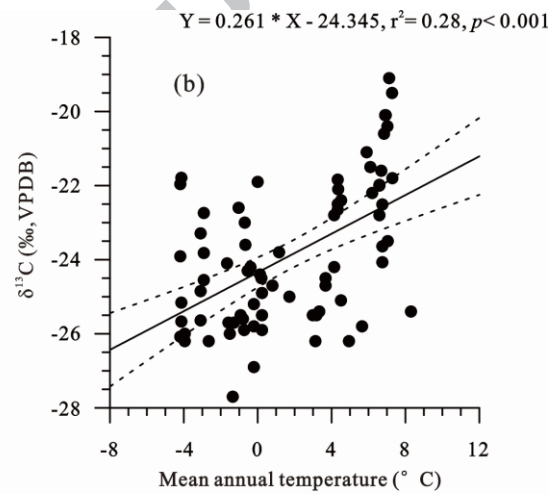
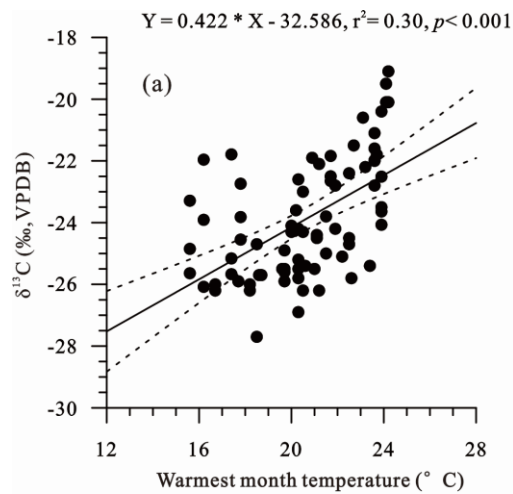
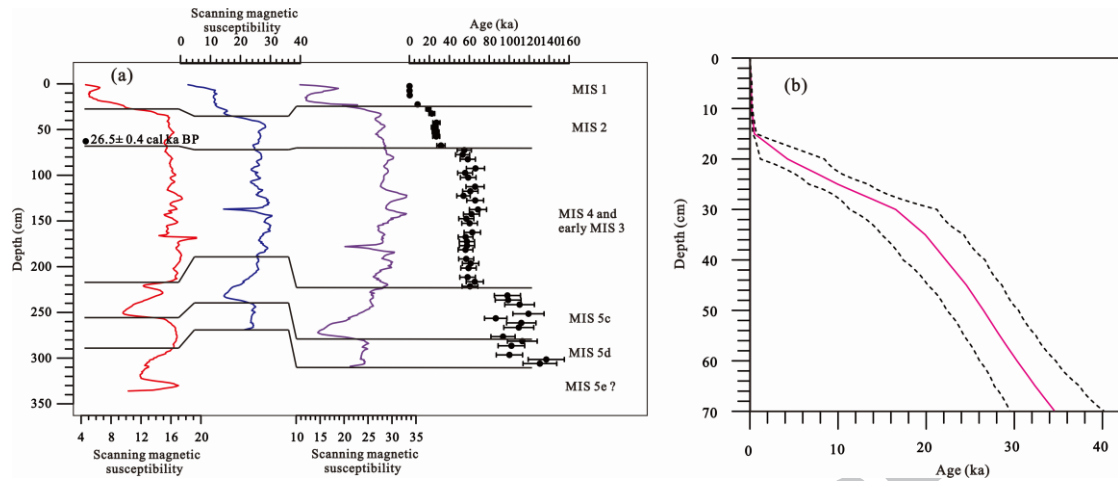
Fig. 3. Linear correlations between $\delta^{13}\text{C}$ values of surface soils and (a) corresponding MAT, (b) warmest month temperature, (c) MAP and (d) growing season precipitation.

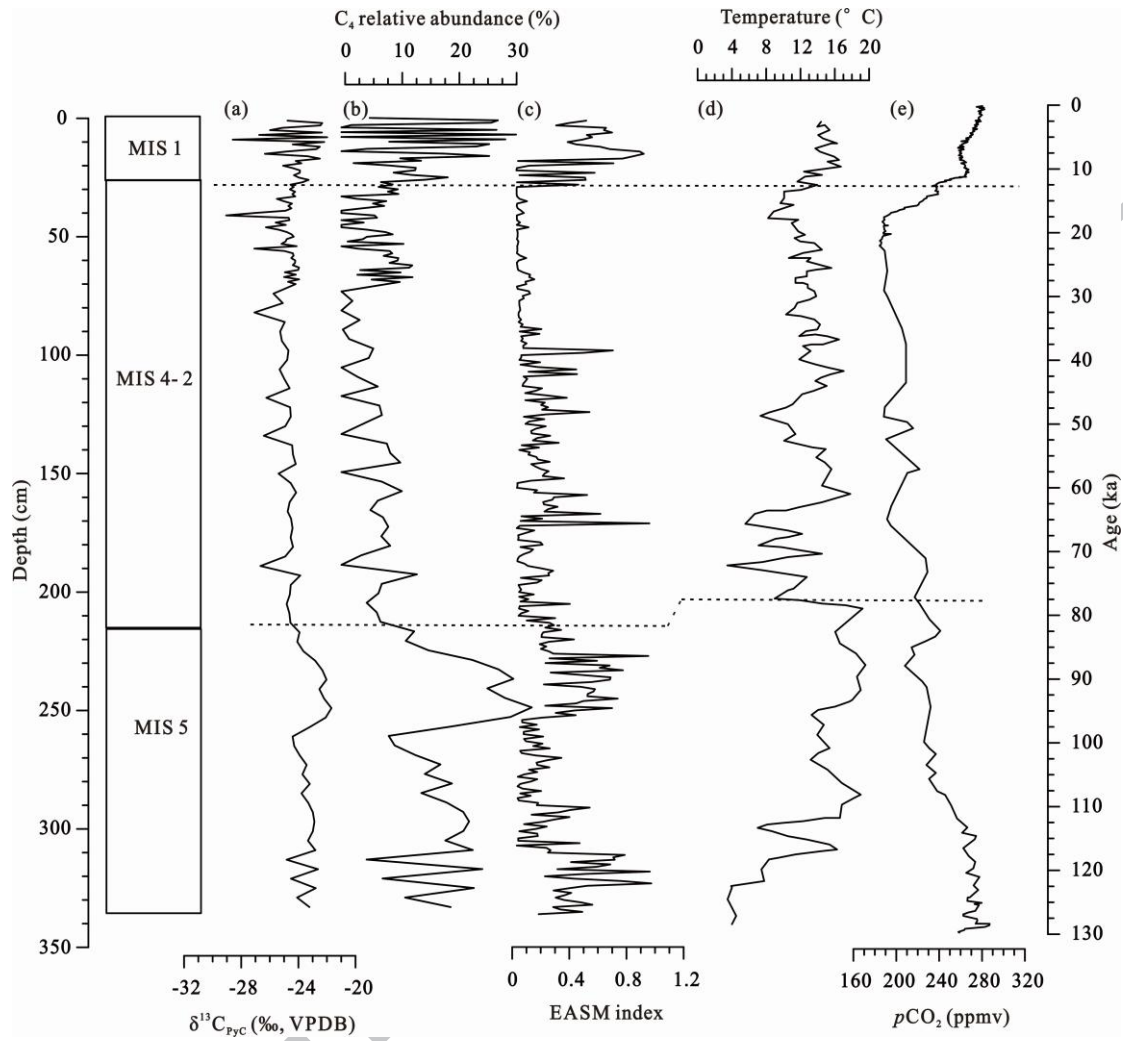
Fig. 4. Comparison of (a) $\delta^{13}\text{C}_{\text{PyC}}$ record from Lake Xingkai with other environments. (b) Estimated C_4 plants relative abundance around Lake Xingkai, the standard deviation (σ) is ca. 15%; (c) EASM index derived from core XK08-A2 grain size end members (Sun et al., 2018); (d) MAT inferred from phytoliths at Weinan section in the Chinese Loess Plateau (Lu et al., 2007); (e) Vostok ice core CO_2 concentration (Jouzel et al., 2007; Luthi et al., 2008).

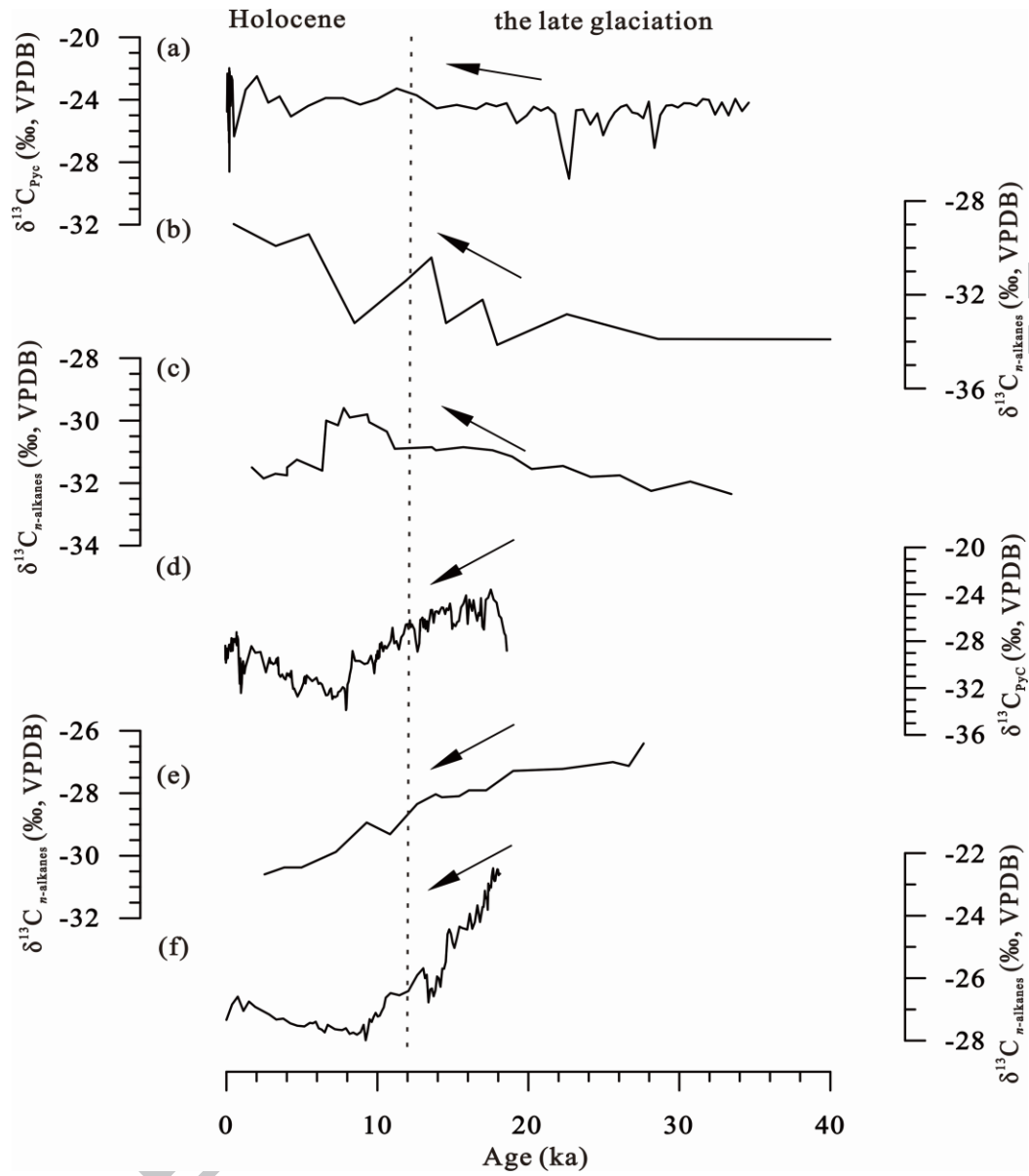
Fig. 5. Latitudinal comparison of C_3/C_4 plants relative abundance change since the last 40 ka. (a) $\delta^{13}\text{C}_{\text{PyC}}$ record from Lake Xingkai; (b) weighted mean average $\delta^{13}\text{C}$ of long chain n -alkanes from Wudalianchi in northeast China (Wang et al., 2017); (c) $\delta^{13}\text{C}$ of C_{31} n -alkane from Xifeng profile in the Chinese Loess Plateau (Liu et al., 2005); (d) $\delta^{13}\text{C}_{\text{PyC}}$ record from Lake Tengchongqinghai in southwest China (Zhang et al., 2015); weighted mean average $\delta^{13}\text{C}$ of specific n -alkanes from Core MD05-2905 in the South China Sea (e, Zhou et al., 2012) and Core SO188-342KL in the Bay of

Bengal(f, Contreras-Rosales et al., 2014).









Highlights

- $\delta^{13}\text{C}$ of surface soil organic matter significantly influenced by temperature.
- $\delta^{13}\text{C}$ of pyrogenic carbon increased during the interglaciations in northeast China.
- Different climatic implication of $\delta^{13}\text{C}$ record between middle and low latitude.

ACCEPTED MANUSCRIPT