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Environmental controls on leaf wax δD ratios in surface peats across the monsoonal region of China

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Abstract

Leaf wax molecular and isotopic ratios are generally considered robust isotopic paleohydrologic proxies. Here we evaluate the proxy value of the molecular distributions and hydrogen isotopic compositions of long chain n -alkanes (δD_{alk}) in surface peats collected from peatlands across a range of annual air temperatures from 1 to 15°C and a range of annual mean precipitation from 720 to 2070 mm in the monsoonal region of China. The alkane ratios (ACL and CPI) and δD_{alk} values show relatively large variations in multiple samples from a single site, highlighting the complexity of these ratios at a small spatial scale. In the montane Zoigê peatland, the apparent fractionation between precipitation and δD_{alk} is more positive than in the other six sites, which is possibly an effect of the higher conductivity of the water in this high elevation site (3500 m a.s.l.). At a larger spatial scale, the site-averaged CPI ratios and the δD_{alk} values of n -C₂₉ and n -C₃₁ alkanes show significant correlation with the air temperature and precipitation. These results support the application of the CPI ratio and the δD_{alk} ratios of n -C₂₉ and n -C₃₁ alkanes as sensitive paleohydrologic proxies on millennial and larger timescales.

1 Introduction

Paleohydrologic reconstructions are a very important component in the study of Quaternary paleoclimate changes. During the past several decades, leaf wax hydrogen isotopic compositions have been proven as a powerful isotopic paleohydrologic tool (Sachse et al., 2012 and associated references). Leaf wax δD values generally follow the isotopic compositions of locally meteoric water (δD_p), which is major water source for plant photosynthesis (Sachse et al., 2012). Therefore, leaf wax δD values can imply the δD_p values and their associated environmental factors.

In low latitude areas, the δD_p values are mainly affected by the “amount effect” (Dansgaard, 1964), and therefore leaf wax δD values are generally interpreted as

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recording the precipitation amount or the relative humidity (e.g. Tierney et al., 2008; Schefuß et al., 2011). A few studies have tried to consolidate such an interpretation by using modern surface soils or recent lake sediments (Garcin et al., 2013). In contrast, at high latitude regions, the “temperature effect” mainly controls the δD_p values. Modern lake sediment investigations in west Europe (Sachse et al., 2004) and North America (Hou et al., 2011; Thomas et al., 2012; Shanahan et al., 2013) have confirmed that leaf wax δD values could record past temperature changes. In the middle-latitude regions, such as the monsoonal region of China, Rao et al. (2009) studied the δD compositions of long chain *n*-alkanes across a climate gradient and concluded that the δD values of long chain *n*-alkanes have the potential to record the δD of meteoric water. To date, however, our knowledge about the dominant controllers on leaf wax δD values in this region is still limited.

Peatlands are waterlogged continental settings whose evolutions are closely associated with hydrological conditions. Hence, this kind of archive is well-suited for paleohydrological reconstructions (Charman, 2002). As examples of leaf wax isotopic studies in peatlands, Xie et al. (2000) first described a two-century-long leaf wax δD sequence from the Bolton Fell Moss, Cumbria, UK. Nichols et al. (2010) discussed the differences in hydrogen isotopic ratios of *Sphagnum* and vascular plant biomarkers in common cores from boreal ombrotrophic peatlands. In the monsoonal region of China, Seki and colleagues (2009, 2011) applied the leaf wax δD records from peat sequences to reconstruct paleoclimate changes since the late deglaciation.

In China, peatlands are mainly clustered in the Northeast and Southwest, with a few scattered ones in the vast eastern lowland (Chai et al., 1981). Due to greater human impacts in the eastern lowland, very few sub-alpine peatlands have been preserved in relatively pristine conditions. In this study, we selected seven peatlands, all with relatively low disturbance, across China to study the spatial variations of leaf wax δD ratios in modern conditions, with the aims of investigating the fundamental controls on leaf wax molecular distributions and δD ratios in the monsoonal regions of eastern China.

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2 Material and methods

2.1 Sampling

Surface samples (0–2 cm) were collected at Yichun (272 m above sea level (m.a.s.l.), 48°48′ N, 129°22′ E) in August 2011, Dajiuhu (1700 m.a.s.l., 31°28′ N, 110°00′ E) in July 2012, Tiandouyang (1196 m.a.s.l., 25°55′ N, 117°29′ E) in October 2012, Wangdongyang (1398 m.a.s.l., 27°41′ N, 119°38′ E) in November 2012, and the other three sites, Hani (900 m.a.s.l., 42°13′ N, 126°31′ E), Shiwangutian (1690 m.a.s.l., 26°05′ N, 110°22′ E), Zoigê (3450 m.a.s.l., 33°49′ N, 102°44′ E), in August 2013 (Fig. 1). In the climate zonation of China, Zoigê is characterized as sub-humid, whereas the other six sites are all characterized as humid (Liu and Tian, 2010).

For sampling site descriptions of Hani, Zoigê, Dajiuhu and Shiwangutian, please refer to Huang et al. (2015). For the other three sites (Yichun, Wangdongyang, Tiandouyang), the surface peats were all collected under *Sphagnum* lawns.

The pH, conductivity (Cond), and oxidation-reduction potential (ORP) of the water of the peatlands were measured in the field using portable electrodes (Mettler Toledo, Swiss; Table 1). The depth of the water level was measured as the depth from the water surface to the top of the peat surface (DWT; Table 1). The water levels reported here are based on a single observation for each sample. These data from Hani, Zoigê, Dajiuhu and Shiwangutian have been published in Huang et al. (2015).

2.2 Lipid extraction

The surface peat samples were first freeze-dried and then ground and sieved through an 80-mesh sieve (0.2 mm). About 0.5 g dried powdered peat was ultrasonically extracted 3× with dichloromethane/methanol (9 : 1, v/v) for 10 min. An internal standard of 5β (H)-cholane (Chiron, Norway) was added before extraction. After solvent removal under reduced pressure, the extract was fractionated into aliphatic, aromatic and polar

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fractions using silica gel column chromatography. The aliphatic fraction analyzed here was eluted with *n*-hexane.

2.3 Instrumental analysis

The aliphatic fraction containing *n*-alkanes was analyzed in a Shimadzu GC-2010 gas chromatograph (GC) equipped with a flame ionization detector (FID) and a DB-5 column (30 m × 0.25 mm i.d., 0.25 μm film thickness). The sample was injected in splitless mode with the injector temperature set at 300 °C. The initial oven temperature was 70 °C and held for 1 min, and then ramped to 210 °C at a rate of 10 °C min⁻¹, and finally ramped to 300 °C at a rate of 3 °C min⁻¹ and kept isothermal for 25 min. Quantification was calculated by comparison of peak areas with the internal standard (cholane) and adjusted with the relevant response factors. The alkane concentration results of the batch of surface peats collected from the Dajiuhu peatland were published in Huang et al. (2014).

Compound-specific hydrogen isotope compositions of *n*-alkanes were determined using a Trace GC coupled with a Delta V advantage isotope ratio mass spectrometer. The sample was injected in splitless mode with the injector temperature at 300 °C. The GC oven temperature was initiated at 50 °C and held for 1 min, and then ramped to 210 °C at a rate of 10 °C min⁻¹ (kept 2 min), and further raised to 300 °C at a rate of 6 °C min⁻¹ (kept 2 min), and finally ramped to 310 °C at a rate of 10 °C min⁻¹ and kept isothermal 25 min. The high temperature conversion (HTC) system was operated at 1400 °C. The HTC tube was conditioned with methane. During the sample running interval, the H₃⁺ factor varied between 3.30 and 3.10. To check the system stability, an *n*-alkane mixture (*n*-C₂₃, *n*-C₂₅, *n*-C₂₇, *n*-C₂₉ and *n*-C₃₁ alkane) and the Indiana A4 mixture with known δD values were run between every two samples. Squalane (δD = -167 ‰) was used as the internal standard. Standard deviation for hydrogen isotope analysis was better than 5 ‰, based on at least duplicate analyses. Results are reported in the delta notation (‰) relative to Vienna Standard Mean Ocean Water (VS-MOW) standard.

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2.4 Apparent fractionation calculation

The apparent fractionation ($\varepsilon_{\text{alk/p}}$) between n -alkane δD ($\delta\text{D}_{\text{alk}}$) and precipitation ($\delta\text{D}_{\text{p}}$) was calculated using the following equation (Bowen et al., 2005):

$$\varepsilon_{\text{alk/p}} = [(\delta\text{D}_{\text{alk}} + 1)/(\delta\text{D}_{\text{p}} + 1)] - 1 \quad (1)$$

5 where $\delta\text{D}_{\text{p}}$ is estimated using the precipitation isotope calculator (http://wateriso.utah.edu/waterisotopes/pages/data_access/oipc.html).

3 Results and discussion

3.1 Spatial variations of long chain n -alkanes in peatlands

10 Long chain n -alkanes ranging from C_{21} to C_{35} are the dominant constituents of the aliphatic fractions in all surface peat samples (Fig. 2). These long chain n -alkanes show strong odd-over-even predominance, indicating their principal origin from the leaf waxes of in situ peat-forming plants (Pancost et al., 2002; Huang et al., 2014). In six of these seven peatlands, *Sphagnum* spp. are present; plants of this type are absent only at the sampling region of the Zoigê peatland. Even though most surface samples were
15 collected under *Sphagnum* lawns in these six peatlands, the P_{aq} values vary in a large range, with low average values at Hani (0.16 ± 0.05) and Dajiuhu (0.18 ± 0.07) to values as high as 0.72 and 0.74 in two samples from Yichun. Published *Sphagnum* n -alkane results (Baas et al., 2000; Pancost et al., 2002; Nichols et al., 2006; Bingham et al., 2010; Ortiz et al., 2011), including those from our previous studies in Dajiuhu (Huang et al., 2010, 2014), almost always support high P_{aq} values as a characteristic feature
20 of peat moss n -alkane distributions. An inconsistency between the n -alkane distributions of fresh moss and underlying peats has been described in Huang et al. (2014). One possibility for this apparent discrepancy is the addition of n -alkanes from vascular plants, which normally have n -alkane concentrations one to two orders of magnitude

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higher than those of fresh *Sphagnum* (Pancost et al., 2002; Huang et al., 2014), to the peat distributions. Another possibility is that the intermediate chain $n\text{-C}_{23}$ and $n\text{-C}_{25}$ alkanes have suffered preferential degradation relative to their long chain counterparts, although more information is required to evaluate the latter possibility. Either way, the new and previous results suggest caution in using the P_{aq} values of bulk peat samples as evidence of the contribution from n -alkanes to moss in peat deposits, even though *Sphagnum* can be an important constituent of modern plant assemblages. Furthermore, the relatively high P_{aq} values in the non-*Sphagnum* Zoigê highlight the possible major contribution of medium chain $n\text{-C}_{23}$ and $n\text{-C}_{25}$ alkanes from vascular aquatic plants in peat deposits, especially in relatively cold settings like Zoigê.

In the four peatlands (Hani, Zoigê, Dajiuhu and Shiwangutian) in which more than ten samples were collected, the possible relations between major alkane ratios (CPI, P_{aq} , and ACL) and peat water chemistry can be explored (Tables 2–5). Only in Shiwangutian does the ACL show a strong correlation with peat water chemistry, evident in a strong negative correlation with pH ($r = -0.76$, $p < 0.01$), and a strong positive correlation with the DWT ($r = 0.72$, $p < 0.01$; Table 5). In both Shiwangutian and Zoigê, the CPI values in the surface peat samples vary closely with DWT but in opposite ways. A positive correlation exists between CPI and DWT in Zoigê ($r = 0.45$, $p < 0.05$; Table 2), whereas a negative correlation exists in Shiwangutian ($r = -0.56$, $p < 0.05$; Table 5). In the Hani peatland, the CPI values correlate positively with pH ($r = 0.58$, $p < 0.05$) and negatively with ORP ($r = -0.67$, $p < 0.05$; Table 3). In the batch of surface peats collected from the Dajiuhu peatland, the correlation between alkane ratios and peat water chemistry is always quite weak (Table 4). CPI values are considered to be controlled mainly by microbial activity, which is in turn affected by environment conditions, especially temperature and relative humidity (Xie et al., 2003; Rao et al., 2009). Our results imply that at a small spatial scale, such as within a peatland, the main influences on the CPI values may be site-dependent and hence complicated.

Comparison of the site-averaged CPI values across the monsoonal region of China with the air mean temperature and mean precipitation reveals a general decrease of

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CPI values is related to increasing annual mean air temperature (Fig. 3). The northernmost site (Yichun) and the higher altitude site (Zoigê) have the highest CPI values (8–9), whereas the southernmost site (Tiandouyang) has low averaged CPI values (3.8; Fig. 3). Previous studies have observed a similar trend between CPI of long chain n -alkanes and mean air temperature in surface soils along an eastern China transect (Rao et al., 2009) and also along a transect from southeastern China to the north margin of the Loess Plateau (Luo et al., 2012). Such a close relation between CPI and air temperature probably results from differences in the extent of microbial degradation of long chain n -alkanes. In the warm and humid conditions such as those in the southern peatlands, microbial degradation will decrease the CPI values. In contrast, microbial activity is inhibited in cool and wet conditions such as Yichun and Zoigê, which will preserve plant n -alkanes and yield relatively higher CPI values. In this study, CPI has a weaker correlation with the mean precipitation ($r = -0.79$, $p = 0.04$) than with the mean temperature ($r = -0.87$, $p = 0.01$; Fig. 3).

We calculated ACL values over the same range of C_{21} to C_{33} as we used for the CPI calculations so that contributions from mosses and aquatic plants would be included. Unlike the CPI, the ACL values display a parabolic trend across the temperature gradient (Fig. 3). Relatively low values occur at both the northernmost/high altitude and the southernmost sites. Previous studies have argued that the ACL values should increase as a plant water management response to increasing temperature or decreasing precipitation (Poynter et al., 1989; Schefuß et al., 2003; HUGHEN et al., 2004). In modern geographic conditions in the monsoonal region of China, mean annual temperature and mean annual precipitation vary closely together. Consequently, our field-based study cannot separate the influences of temperature and precipitation on derived n -alkane ratios like the CPI and the ACL. Instead, we interpret the complex relation between ACL and the climate parameters of temperature and precipitation to reflect the additional influence from differences in floral composition. For example, *Sphagnum* species (Bush et al., 2013) and submerged/floating aquatic macrophytes (Ficken et al., 2000) that can populate wet and warm settings typically have relatively low ACL values.

3.2 Differences in long chain *n*-alkane δD values in peatlands across the monsoonal region of China

3.2.1 Intra-peatland differences in long chain *n*-alkane δD values

Here we focus on comparisons of the δD values of the *n*-alkanes in the four peatlands with surface peat samples > 10 . In the batch of samples ($n = 20$) collected from the Zoigê peatland, the δD values of individual *n*-alkane have a relatively large range (Fig. 4). The *n*-C₂₃ and *n*-C₂₅ alkanes have more negative δD values than their *n*-C₂₉ and *n*-C₃₁ homologues, a difference that suggests that the two shorter chain molecules may have different precursors to the longer chain ones. In this non-*Sphagnum* peatland, relatively high P_{aq} values (0.36 ± 0.08) indicate that the *n*-C₂₃ and *n*-C₂₅ alkanes mainly originate from floating aquatic plants. In relation to the water level and chemistry, the δD_{alk} values of the four alkanes correlate significantly only with the DWT (Table 2). The close relation between δD_{alk} and water level in the Zoigê peatland strongly suggests that δD_{alk} values could be a robust proxy for paleohydrological conditions at this location.

The δD_{alk} value distribution is quite different in the batch of surface peats ($n = 12$) collected from the Hani peatland (Fig. 4, Table 3). The range of values is small, the *n*-C₂₃ and *n*-C₂₅ alkanes have δD values similar to those in the Zoigê peatland, and the *n*-C₂₉ and *n*-C₃₁ alkanes have more negative δD values than their counterparts in the Zoigê peatland. In addition, the δD_{alk} values of the *n*-C₂₃ and *n*-C₂₅ alkanes in Hani peatland do not correlate closely with the *n*-C₂₉ and *n*-C₃₁ alkanes (Table 3). Such a pattern probably results from their different sources. The *n*-C₂₃ and *n*-C₂₅ alkanes may principally originate from *Sphagnum*, whereas the *n*-C₂₉ and *n*-C₃₁ alkanes probably come from vascular plants. In the batch of Hani surface peats, the δD_{alk} values correlate poorly with water level and water chemistry; only the δD_{31} has a moderately negative correlation with water pH (Table 3).

In the batch of surface peats ($n = 13$) collected from the Dajihu peatland, the very low concentrations of *n*-C₂₃ and *n*-C₂₅ prevented acquisition of high quality δD values

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of these two compounds. Unlike in the Zoigê and Hani peatlands, the δD value of n -C₂₉ alkane does not correlate well with those of n -C₃₁ alkane in the two southern peatlands (Dajiuhu and Shiwangutian; Tables 4 and 5). This difference between the northern and southern peatlands highlights that the southern peatlands in sub-tropical conditions may have a more complex mixture of sources for their long chain n -alkanes.

This study reveals large amplitudes of the δD values of a single compound within a peatland. A recent study demonstrated that lipid and soil water δD ratios in a salt-marsh differed by up to 35% (Eley et al., 2014). Such a large variation of δD ratios may result from the microtopographical variability within a peatland.

3.2.2 Differences in the long chain n -alkane δD values in the different peatlands

Although the dominant influences on the δD_{alk} values appear to be site-dependent within each peatland, the major controls on the δD_{alk} values peatlands across the monsoonal regional of China may be climate-dependent. As shown in Fig. 5, the site-averaged δD values of n -C₂₉ and n -C₃₁ alkanes follow closely the annual mean air temperature and precipitation. An exception is the Zoigê site, which shows relatively less negative δD_{alk} values. On the basis of its latitude (33°49' N) and high elevation (3500 m a.s.l.), this site should have more negative δD_p and hence more negative δD_{alk} values than the other six sites. Evaporation is probably not the primary reason for the less negative δD_{alk} values in the Zoigê site. The annual precipitation is around 700 mm in Zoigê County, while the annual evaporation is about 260 mm (Guo and Li, 2007). In addition, the δD_{alk} sequence in a Hongyuan peat core (3505 m a.s.l., 32°46' N, 102°30' E; quite near our Zoigê sampling site) was more negative (–20 to –30‰) than that of the Hani peat core (42°13' N, 126°31' E) at the same temporal epoch (Seki et al., 2009, 2011). Also different from the six other sites, Zoigê site has quite high conductivity and basic pH (Tian and Xiong, 2004; Chen et al., 2012; Huang et al., 2015). The higher conductivity in Zoigê results from the high ionic content, es-

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pecially Ca^{2+} , Mg^{2+} and HCO_3^- . The higher conductivity could play a role in the larger n -alkane δD values in the Zoigé peat.

Previous studies have demonstrated that salinity is an important environmental influence on the δD values of lipids in marine and coastal conditions (e.g. Schouten et al., 2006; Sachse and Sachs, 2008). In higher salinity settings, the restricted exchange with extracellular water would result in less hydrogen isotope fractionation during lipid biosynthesis (see Sachse et al., 2012 for a review). The dominant plants surrounding our sampling points at Zoigé peatland are some halophytes (*Glaux maritima*, *Puccinellia* sp., *Halerpestes* sp.), together with *Juncus effuses* and *Pedicularis* sp. Thus it is possible that the relatively less negative $\delta\text{D}_{\text{alk}}$ values in the surface peat samples collected from Zoigé peatland result from a physiological adaptation similar to halophytes to high salinity conditions. However, the salinity in the batch of Zoigé peat samples is less than the detection limitation. Future studies need to investigate why hydrogen isotope fractionation seems to be restricted in this freshwater setting having a high conductivity and a basic pH.

In the six peatlands with relatively low and uniform conductivity, the site-averaged δD values of the $n\text{-C}_{29}$ and $n\text{-C}_{31}$ alkanes correlate positively with air temperature ($r = 0.88$, 0.82 , $p < 0.05$) and precipitation ($r = 0.93$, 0.76 , $p \leq 0.01$). The δD values of the $n\text{-C}_{23}$ and $n\text{-C}_{25}$ alkanes also show increasing trends with air temperature and precipitation (Fig. 5) but the smaller number of these data fails to pass the significance test. The positive correlation between temperature and the δD_{29} and δD_{31} values is consistent with the so-called “temperature effect” (Dansgaard, 1964), which has been observed prominently across regions outside the tropics, including Europe (Sachse et al., 2004) and North America (Hou et al., 2007). In contrast, the positive correlation between precipitation and δD_{29} and δD_{31} is inverse with the so-called “amount effect” (Dansgaard, 1964). Consequently, the positive correlation between precipitation and the δD_{29} and δD_{31} values may indirectly reflect the close relation between temperature and precipitation in modern climate (Johnson and Ingram, 2004).

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The Zoigê site has the least negative $\varepsilon_{\text{alk/p}}$ values of $n\text{-C}_{29}$ and $n\text{-C}_{31}$ alkanes (-126 and -120% ; Fig. 6). In contrast, the other six sites display near constant $\varepsilon_{\text{alk/p}}$ values of $n\text{-C}_{29}$ and $n\text{-C}_{31}$ alkanes (C_{29} -143 – -158% ; C_{31} -148 – -162% ; Fig. 6). In the six sites with low conductivity, the vegetation is dominated by *Sphagnum* and some herb plants (Bu et al., 2011; Luo et al., 2015). In these case, vegetation life forms would not be expected to be a primary effect on the $\varepsilon_{\text{alk/p}}$ values in this study (Sachse et al., 2012). Hence, the fairly narrow range of $\varepsilon_{\text{alk/p}}$ values of $n\text{-C}_{29}$ and $n\text{-C}_{31}$ alkanes in these six locations suggests that evapotranspiration and biosynthesis are not the dominant factors to control the site-averaged δD values of $n\text{-C}_{29}$ and $n\text{-C}_{31}$ alkanes. Instead, the δD value of the water source, i.e. δD_p , is probably the first order control on the large spatial variations of the $\delta\text{D}_{\text{alk}}$ values.

It is interesting to note that the $\varepsilon_{\text{alk/p}}$ values of the $n\text{-C}_{29}$ alkane in this study are more negative than those in surface soil samples across a similar latitude transect in the eastern part of China (-130 to -140% ; Rao et al., 2009). This difference may result from plant physiologic responses to environmental factors in geographically different habits. Alternatively, it may be caused by the different influence of the relative humidity (RH) on leaf wax δD (Sachse et al., 2012). In peatlands, such as the montane Dajiuhu peatland, the RH is normally high. For example, our monitoring of RH at Dajiuhu in 2014 showed that the RH was almost $> 70\%$, even in the warm interval at late July when the water table dropped nearly 1 m (unpublished data).

Relative to the $n\text{-C}_{29}$ and $n\text{-C}_{31}$ alkanes, the medium chain length homologues ($n\text{-C}_{23}$ and $n\text{-C}_{25}$ alkanes) have more scattered but generally more negative $\varepsilon_{\text{alk/p}}$ values (Fig. 6). This pattern is inconsistent with the hypothesis that the different hydrogen isotope ratios of *Sphagnum* and vascular plant biomarkers could be used as a quantitative proxy for precipitation–evaporation balance (Nichols et al., 2010). In fact, the peat cores from both Hani (Seki et al., 2011) and Hongyuan (Seki et al., 2009) consistently showed more negative δD values for the $n\text{-C}_{23}$ and $n\text{-C}_{25}$ alkanes over the Holocene epoch.

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4 Conclusions

We determined the molecular distributions and δD ratios of long chain n -alkanes in surface peats collected from seven peatlands across gradients of mean annual air temperature of 1 to 15 °C and mean annual precipitation of 720 to 2070 mm in the monsoonal region of China. Our results reveal that alkane ratios such as ACL and CPI and δD_{alk} values can have relatively large variations within a single peatland, suggesting that the dominant factors controlling these ratios may be site dependent on a small scale. This finding highlights the complexity of interpreting both alkane ratios and δD_{alk} values on the small spatial scale as paleoclimate proxies. In the batch of surface peats collected from the high elevation Zoigê peatland, the δD_{alk} values of long chain n -alkanes correlate significantly with water level. In addition, the δD_{alk} values in Zoigê peatland are probably affected by the higher conductivity and/or higher pH of this bog, resulting in relatively less negative δD_{alk} values in this high elevation setting.

At a larger spatial scale, the site-averaged CPI ratio and the δD_{alk} values of the n -C₂₉ and n -C₃₁ alkanes show a significant correlation with local air temperature and precipitation. These results support use of the CPI ratio and the δD_{alk} values of the n -C₂₉ and n -C₃₁ alkanes as sensitive proxies for past climate changes on millennial and larger timescales.

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Table 1. Water chemistry and water level for surface peat samples from Yichun (labeled as YC-), Wangdongyang (labeled as WDY-) and Tiandouyang (labeled as TDY-).

Sample	pH	ORP ^a (mv)	DWT-P ^b (cm)	DWT-M ^c (cm)
YC-1	4.78	129.4	n.d. ^d	22.0
YC-2	5.15	109.4	n.d.	23.0
YC-3	4.35	156.4	n.d.	9.0
YC-4	3.93	180.9	n.d.	11.5
YC-5	4.37	155.5	n.d.	11.5
WDY-1	4.47	274.0	5.0	17.0
WDY-2	5.93	224.0	7.0	11.0
WDY-3	n.d.	n.d.	n.d.	n.d.
WDY-4	5.29	247.0	10.5	13.5
WDY-5	5.70	232.0	3.0	13.0
WDY-6	4.84	238.0	9.5	18.5
TDY-1	5.45	210.0	7.0	21.5
TDY-2	6.13	225.0	7.5	14.5
TDY-3	5.70	272.0	0	8.0
TDY-4	6.07	243.0	15.0	19.0

^a Oxidation-reduction potential; ^b Depth from water surface to peat surface;

^c Depth from water surface to moss top. ^d not detected.

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Table 2. Correlation analysis among the δD values of long chain *n*-alkanes and water chemistry in the batch samples collected from Zoigê peatland.

	δD_{23}	δD_{25}	δD_{29}	δD_{31}	pH	Cond	ORP
δD_{25}	0.89**						
δD_{29}	0.79**	0.90**					
δD_{31}	0.76**	0.89**	0.87**				
pH	-0.19	0.04	-0.28	0.08			
Cond	0.31	-0.02	0.21	0.02	-0.72**		
ORP	-0.31	-0.15	-0.35	-0.06	0.75**	-0.63**	
DWT	0.73**	0.80**	0.71**	0.82**	0.00	0.06	-0.08

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Table 3. Correlation analysis among the δD values of long chain *n*-alkanes and water chemistry in the batch samples collected from Hani peatland.

	δD_{23}	δD_{25}	δD_{29}	δD_{31}	pH	Cond	ORP
δD_{25}	0.77*						
δD_{29}	0.06	0.07					
δD_{31}	0.46	0.26	0.88**				
pH	-0.40	-0.43	-0.49	-0.56*			
Cond	-0.34	-0.41	-0.34	-0.32	0.80**		
ORP	0.58	0.51	-0.02	0.05	-0.80**	-0.69**	
DWT	-0.13	0.03	-0.56	-0.50	0.32	0.39	-0.08

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Table 4. Correlation analysis among the δD values of long chain *n*-alkanes and water chemistry in the batch samples collected from Dajiuhu peatland.

	δD_{29}	δD_{31}	pH	Cond	ORP
δD_{31}	0.33				
pH	0.45*	0.20			
Cond	0.39*	0.19	0.50*		
ORP	-0.57**	-0.38	-0.48*	-0.59**	
DWT	-0.11	-0.30	0.08	0.12	0.06

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Table 5. Correlation analysis among the δD values of long chain *n*-alkanes and water chemistry in the batch samples collected from Shiwangutian peatland.

	δD_{29}	δD_{31}	pH	Cond	ORP
δD_{31}	0.52				
pH	-0.10	-0.09			
Cond	-0.03	0.06	0.39		
ORP	-0.42	-0.30	0.13	-0.31	
DWT	0.32	-0.20	-0.75**	-0.26	-0.22

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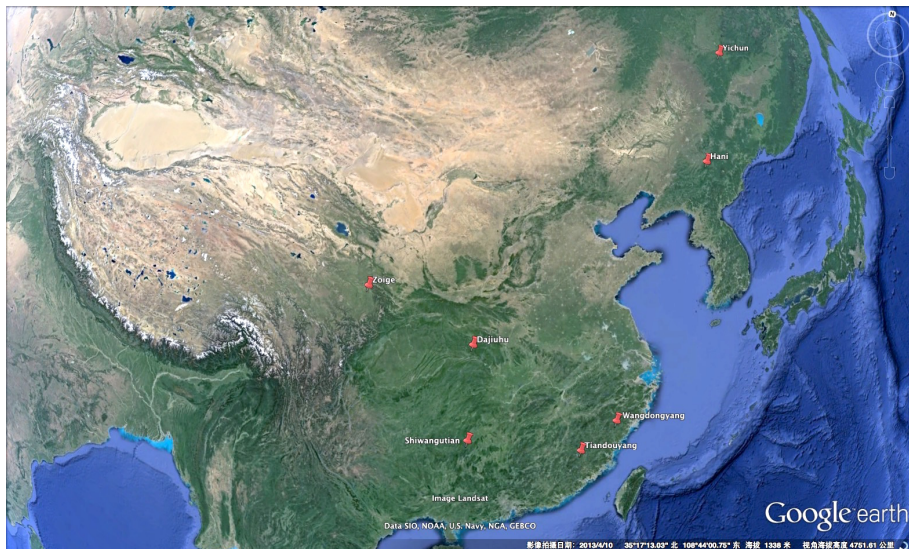



Figure 1. Locations of sampling sites in the monsoonal region of China. Downloaded from Google Earth.

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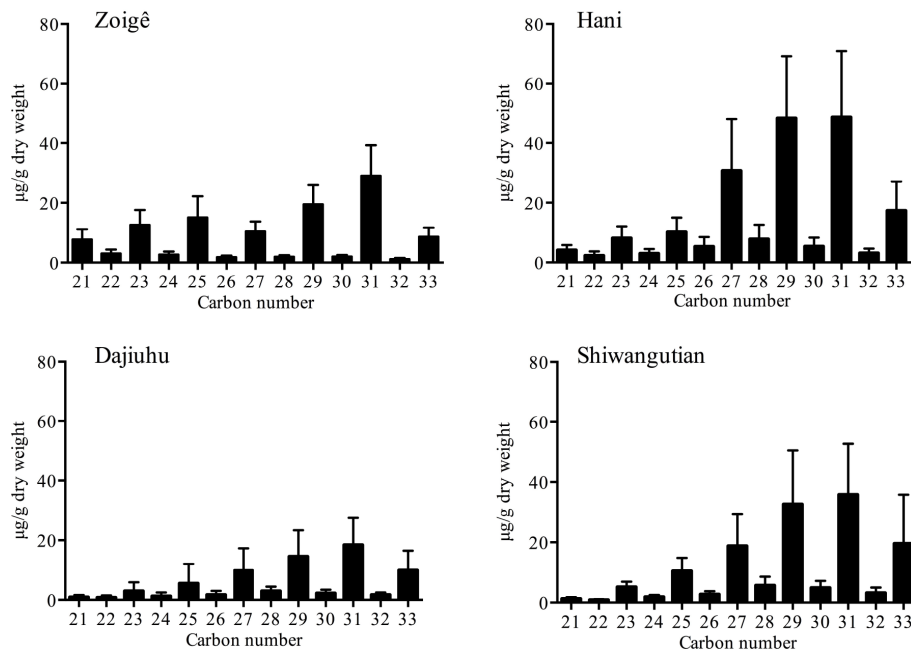
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Figure 2. The averaged concentration of the long chain *n*-alkane homologues in the four peatlands (Zoigê, Hani, Dajiuhu, Shiwangutian) with sampling numbers > 10.

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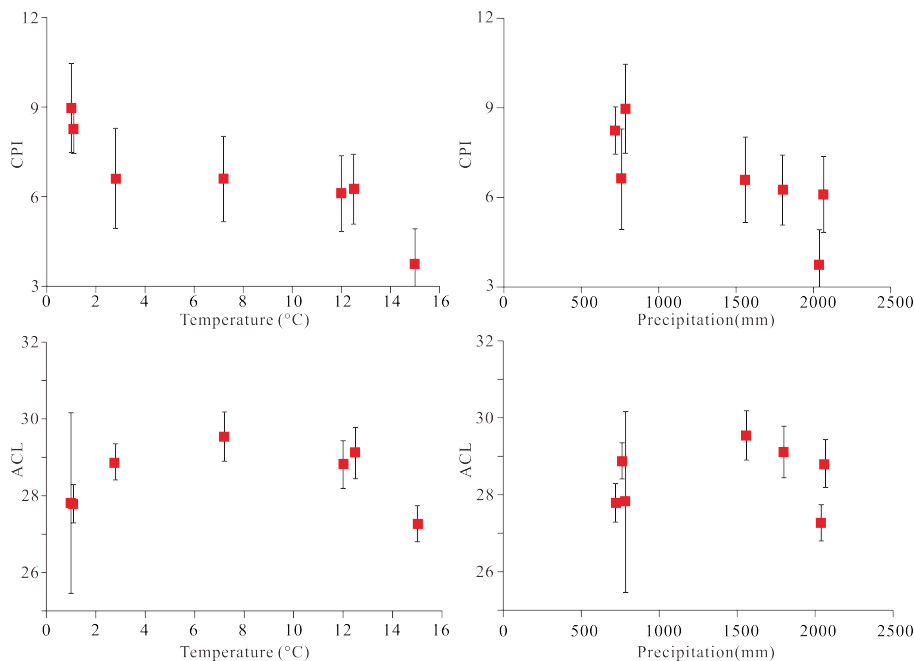


Figure 3. Trends of the site-averaged alkane molecular ratios (CPI, ACL) along the temperature and precipitation gradients.

$$\text{CPI} = \frac{1}{2} \left(\frac{[C_{21}] + [C_{23}] + [C_{25}] + [C_{27}] + [C_{29}] + [C_{31}]}{[C_{22}] + [C_{24}] + [C_{26}] + [C_{28}] + [C_{30}] + [C_{32}]} + \frac{[C_{23}] + [C_{25}] + [C_{27}] + [C_{29}] + [C_{31}] + [C_{33}]}{[C_{22}] + [C_{24}] + [C_{26}] + [C_{28}] + [C_{30}] + [C_{32}]} \right)$$

$$\text{ACL} = \frac{21 \times [C_{21}] + 23 \times [C_{23}] + 25 \times [C_{25}] + 27 \times [C_{27}] + 29 \times [C_{29}] + 31 \times [C_{31}] + 33 \times [C_{33}]}{[C_{21}] + [C_{23}] + [C_{25}] + [C_{27}] + [C_{29}] + [C_{31}] + [C_{33}]}$$

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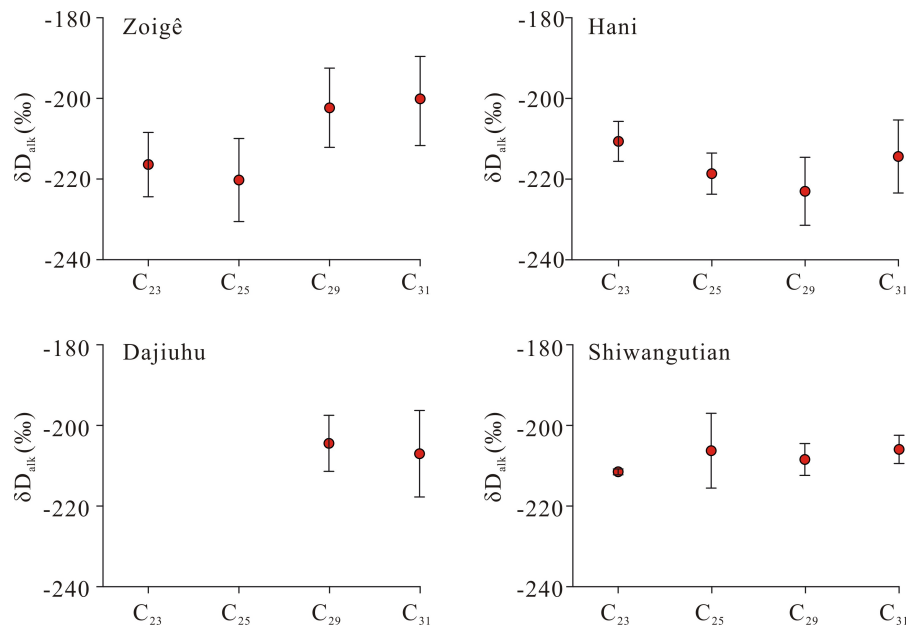


Figure 4. The averaged δD values of the $n\text{-C}_{23}$, $n\text{-C}_{25}$, $n\text{-C}_{29}$ and $n\text{-C}_{31}$ alkanes in the four peatlands (Zoigê, Hani, Dajiuhu, Shiwangutian) with sampling numbers > 10 .

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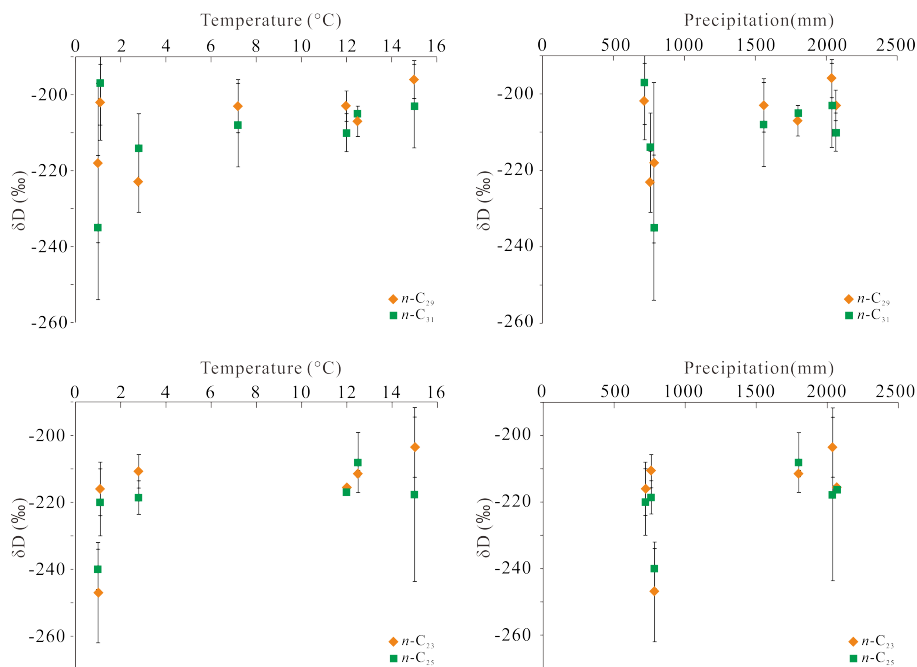


Figure 5. Trends of the site-averaged δD values of the $n-C_{23}$, $n-C_{25}$, $n-C_{29}$ and $n-C_{31}$ alkanes along locational temperature and precipitation gradients.

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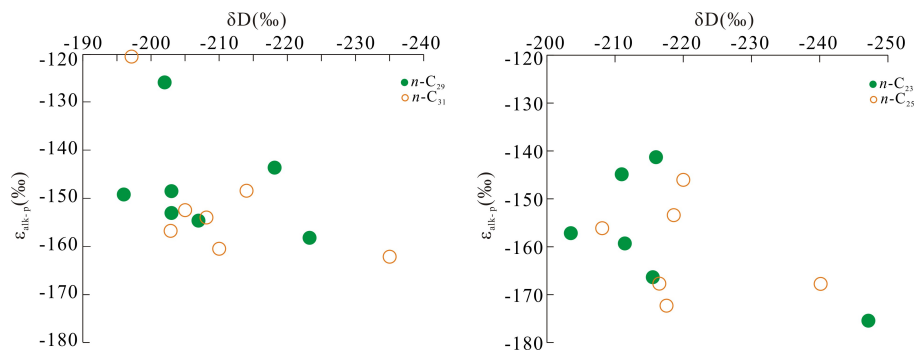


Figure 6. The site-averaged apparent fractionation ($\epsilon_{\text{alk}/p}$) between the n -alkane δD (δD_{alk}) of the $n\text{-C}_{23}$, $n\text{-C}_{25}$, $n\text{-C}_{29}$ and $n\text{-C}_{31}$ alkanes and precipitation (δD_p) calculated after Bowen et al. (2005).