Holocene climate controls on water isotopic variations on the northeastern Tibetan Plateau

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ABSTRACT

On the Tibetan Plateau, applications of δD and $\delta^{18}\text{O}$ values in paleoclimate studies tend to be complicated due to multiple processes influencing isotopic compositions in paleoclimatic archives. In this study, isotopic compositions of modern waters in the eastern Qaidam Basin on the northeastern Tibetan Plateau, and δD values of n -fatty acids (n -FA δD) from a sediment core at Hurler Lake were systematically analyzed to infer hydroclimate controls during the Holocene. The modern water isotopic results show a major contribution of snowmelt water originating from high-elevation mountains to the north of the Qaidam Basin via river and groundwater discharge, and the importance of evaporation in affecting lake water budget in this region. n -C₂₆ FA δD values tend to be more negative at millennial-scale warm-dry periods during the Holocene, and vice versa, opposite to what is commonly expected. Assisted with modern water isotopic results, we infer amplified contribution of snowmelt water to the soil water around this open lake system at warm-dry periods. Meanwhile, changes in n -C₁₆ FA δD values at Hurler Lake reflect the evolution of isotopic compositions of lake water, thus we use the isotopic difference between n -C₂₆ and n -C₁₆ FA ($\Delta\text{D}_{\text{C16-C26}}$) to infer hydroclimate and evaporation variations in this region. Based on our data, relatively low n -C₂₆ FA δD and n -C₁₆ FA δD values at 10–6 cal ka BP indicate large contribution of snowmelt water into the lake during the Holocene Climate Optimum. After 6 cal ka BP, changes in evaporation became the major control on lake hydrology and led to larger fluctuations of $\Delta\text{D}_{\text{C16-C26}}$. Our study highlights the importance of systematic analysis on modern processes before using stable isotopes for paleoclimate reconstructions, and demonstrates that δD difference between long-chain and short-chain n -FA might be an effective way to better understand the controlling factor of hydrological variations in a climatic complex region like the Tibetan Plateau.

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1. Introduction

Stable isotopes serve as a powerful tool for understanding past climatic changes. In polar regions, $\delta^{18}\text{O}$ records from ice cores have been widely interpreted as past temperature variations (e.g. Petit et al., 1999; Johnsen et al., 2001). In tropical monsoon regions, $\delta^{18}\text{O}$ in climate archives including speleothems have been used as proxy for monsoon

intensity and thus monsoon precipitation (e.g. Dykoski et al., 2005; Wang et al., 2005). Around the high-altitude Tibetan Plateau, however, interpretation of stable isotopes in paleoclimatic archives is more complicated. For instance, the $\delta^{18}\text{O}$ of some ice cores from the Tibetan region (e.g. Dunde ice core) is not only controlled by air temperature as conventionally interpreted, but also influenced by monsoon precipitation (Davis et al., 2005). Also, $\delta^{18}\text{O}$ data from Kesang Cave (Cheng et al., 2012) at Tianshan Mountain to the north of the Tibetan Plateau shows an anti-phased relation with those from the nearby Guliya ice cap (Thompson et al., 1997) at the beginning of interglacials, and thus cannot be simply interpreted by the temperature effect only as the Guliya ice cap record does (Thompson et al., 1997). It is possible that incursions of Asian summer monsoon rainfall and moisture in the region during

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the high insolation times may play an important role in changing the $\delta^{18}\text{O}$ signals in Kesang Cave (Cheng et al., 2012). Although stable isotopes derived from lake sediments in this arid region have also been widely used for reconstructions of past hydroclimate variations around the Tibetan Plateau, the interpretation of the isotopic data is also further complicated by multiple factors influencing isotopic fractionation, such as temperature effect and amount effect on precipitation isotopes, evaporation effect under dry climate (including evaporation on lake water, rain drops, and vegetation, etc.) and probable contributions of snow or glacial melt water input from nearby high-elevation mountains (e.g. Liu et al., 2008a; Henderson et al., 2010; Wang et al., 2013).

In order to better understand past hydroclimate variations in this climatically sensitive region, information about inputs from various water sources and effects of evaporation need to be separated. Compound specific δD of lipids preserved in lake sediments might serve as a potential proxy for past hydroclimate changes, since terrestrial and aquatic plants/algae use their own ambient water for lipid biosynthesis (e.g. Yapp and Epstein, 1982; Sessions et al., 1999; Sachse et al., 2004, 2012; Aichner et al., 2010; Duan et al., 2014; Rao et al., 2014; Yu et al., 2014; Aichner et al., 2015; Yao et al., 2015; Thomas et al., 2016). Many studies have used δD values of mid- and long chain *n*-alkanes to reconstruct individual isotopic information of aquatic and terrestrial source water (e.g. Mügler et al., 2008; Duan et al., 2014; Rao et al., 2014). It has been suggested that there exists more distinguished difference in the chain-length distribution of *n*-fatty acid (FA) between the aquatic and terrestrial plants (Wang and Liu, 2012). In lake sediments, δD values of long-chain *n*-FA (e.g. *n*-C₂₆ to *n*-C₃₀) can be used to track the isotope signal of terrestrial source water and thus local precipitation (Sternberg, 1988; Sachse et al., 2004; Liu and Huang, 2005; Smith and Freeman, 2006; Huang et al., 2007; Wang and Liu, 2012), while δD values of short-chain *n*-FA (e.g. *n*-C₁₆ to *n*-C₂₀) could record the isotopic composition of lake water which carries the isotopic imprints of both inflowing source water and evaporative enrichment (Cranwell et al., 1987; Huang et al., 2002; Wang and Liu, 2012). Since the biosynthetic fractionation of the lipids to source water is similar in both terrestrial plants and aquatic organisms (Sachse et al., 2006), the δD difference between short-chain *n*-FAs and long-chain *n*-FAs can potentially remove the isotopic information of source water from the isotopic signal, and assess the magnitude of lake water evaporation relative to source water, especially in arid regions with limited precipitation but strong evaporation (Sachse et al., 2006; Mügler et al., 2008). This method is particularly helpful for open-lake systems in arid areas where lake hydrology could be dominated by either evaporation or inflow water input, since lake water isotope composition alone won't be able to determine which factor is more important.

A systematic analysis on isotopic processes of regional hydrology is necessary before using *n*-FA δD values for paleoclimate reconstructions in a specific region. Previous studies of *n*-FA δD values of different chain lengths have been largely focused on modern surface samples from lake sediments, soils and plants (e.g. Sachse et al., 2006; Wang and Liu, 2012). δD values of both short-chain and long-chain *n*-FA are influenced by multiple factors, making it difficult to fully delineate the mechanism of isotopic fractionation process (e.g. Duan et al., 2014; Yu et al., 2014; Aichner et al., 2015; Yao et al., 2015), which in turn would prevent us from applying down-core isotopic records to paleohydroclimatic reconstruction. However, if we have already extracted climatic information from a sedimentary core using different proxies, we may infer controlling factors of isotopic fractionation process through time using the same core.

Climate in the Holocene has been characterized by a gradual cooling trend in the Northern Hemisphere high and mid-latitude regions and by a gradual drying trend in the monsoonal region (Mayewski et al., 2004; Dykoski et al., 2005). However, the Arid Central Asia experienced increased humidity in the mid-Holocene, which was anti-phased with changes in the Asian monsoonal region (Chen et al., 2008). In addition to the long-term trend, many studies on lakes in the Arid Central Asia

have revealed hydrological variations at millennial-scale (e.g. Rhodes et al., 1996; Mischke et al., 2005; Mischke and Wünnemann, 2006). In previous studies, Holocene temperature (Zhao et al., 2013) and moisture (Zhao et al., 2010) reconstructions based on a sediment core taken from Hurlig Lake suggested a distinct millennial warm-dry association throughout the Holocene and an increase in regional climate variability after the mid-Holocene. In this study, we examine δD and $\delta^{18}\text{O}$ compositions of modern water samples from freshwater and saline lakes, groundwater, rain, puddles, and rivers in the eastern Qaidam Basin on the northeastern Tibetan Plateau. Then, we report δD records of *n*-C₁₆ and *n*-C₂₆ FAs extracted from a Holocene sediment core at Hurlig Lake. Together with the alkenone-based salinity record from %C_{37:4} and the previously published temperature (Zhao et al., 2013) and lake-level (Zhao et al., 2010) data from the same sediment core, we aim to infer main water sources to Hurlig Lake, to reconstruct hydrogen isotopic signals in terrestrial (long-chain *n*-FA) and lacustrine (short-chain *n*-FA) source waters, to infer controlling factors of δD values in terrestrial and lacustrine source waters, and to better understand Holocene hydroclimate variations on the northeastern Tibetan Plateau.

2. Material and methods

Hurlig Lake (37°17' N, 96°54' E; elevation 2817 m above sea level, Fig. 1A) is a freshwater lake located in the Qaidam Basin on the northeastern Tibetan Plateau. The lake is mainly fed by the Bayin River, the Balegen River (prior to the reservoir construction), and groundwater originating in the high mountains to the north of the basin. This region is likely influenced by the westerly rather than the Asian Summer Monsoon since the early Holocene, as indicated by modern meteorological studies (Tian et al., 2003) and proxy records (Chen et al., 2010; Zhao et al., 2010, 2013; He et al., 2013). Mean annual air temperature, precipitation and potential evaporation in this region are 1.4 °C, ~160 mm and ~2000 mm, respectively, as documented by the instrumental data from the nearby Delingha meteorological station (~30 km to the east, Fig. 1B; Zhao et al., 2010).

A total of 56 water samples were collected from rain, lakes, rivers, springs/wells, and puddles around the Hurlig Lake region in the summers of 2005, 2006 and 2007 (Fig. 1C, Supplemental Table S1). Core HL06-1 was obtained in July 2006 at 7.6 m water depth using a UWITEC corer. Detailed chronological information has been described previously (Zhao et al., 2010, 2013). The age model was established through AMS-¹⁴C dates on the aquatic plant *Ruppia* from selected intervals, after correction of a 2758-year reservoir effect determined by the age difference between ¹⁴C and ²¹⁰Pb dates from the top part of core HL06-1 (Zhao et al., 2010). Based on the derived chronology published previously (Zhao et al., 2013), core HL06-1 contains continuous sequence through the past 10,500 years, with a mean sedimentation rate of ~0.083 cm/year. As core HL06-1 lacks dating control before 7.4 cal ka BP, here we mainly focus on the isotopic variability over the last 8000 years. Consequently, we achieved centennially-resolved records, with ~120-year sampling resolution for the last 7400 years and ~500-year sampling resolution before 7.4 cal ka BP.

A total of 243 sediment samples were analyzed for *n*-FAs and alkenones and ~70 samples were further chosen for compound specific δD analysis. The samples were prepared following the standard procedures from Wang and Liu (2012). The total lipids were extracted from the freeze-dried sediments with organic solvents (dichloromethane:methanol = 9:1, v/v). After being hydrolyzed by 6% potassium hydroxide in methanol solution, the neutral fraction was extracted with *n*-hexane. The basic solution was acidified to pH < 1 to extract the carboxylic acid fraction with *n*-hexane. The neutral fraction was then separated into three fractions with silica gel column chromatography using eluents of *n*-hexane, dichloromethane and methanol, while the dichloromethane fraction contains alkenones. The acid fraction samples were heated to 60 °C with the 5% acetyl chloride in

Fig. 1. (A) Map showing location of Hurler Lake on the northeastern Tibetan Plateau. The bold grey line depicts the outline of the Tibetan Plateau and the dashed line indicates the Qaidam Basin. The East Asian summer monsoon, Indian monsoon, and westerlies are also indicated. The modern East Asian monsoon limit is roughly shown by the dark blue dashed line. (B) Monthly temperature, precipitation and evaporation at the nearby Delingha weather station. (C) Locations of coring sites of core HL06-1 and HL05-2 (Zhao et al., 2010) and the surface water samples taken in the summers of 2005, 2006 and 2007 from rain, river and lakes around the Lake Hurler region.

methanol (v/v) for 12 h, extracted with *n*-hexane, and then further purified with silica gel chromatography. The alkenone and *n*-FA fractions were analyzed on Gas Chromatography equipped with a flame ionization detector (Agilent 7890 at HKU for alkenone analysis and Agilent 6890 at IEECAS for *n*-FA analysis), using *n*-C₃₆ alkane as internal standard for quantification and the lab standards of alkenones and *n*-FAs for identification. The %C_{37:4} was calculated using the equation (Rosell-Melé, 1998):

$$\%C_{37:4} = C_{37:4} / (C_{37:2} + C_{37:3} + C_{37:4}) * 100\%$$

where C_{37:2}, C_{37:3} and C_{37:4} are the concentrations of di-, tri- and tetra-unsaturated C₃₇ alkenones respectively. The analytical error is within 1.5% for %C_{37:4}.

The ACL_{*n*-FA} was calculated with the equation (Poynter and Eglinton, 1990; Wang and Liu, 2012):

$$ACL_{n-FA} = (16 * n-C_{16} + 17 * n-C_{17} + 18 * n-C_{18} + \dots + 28 * n-C_{28}) / \sum n-C_{16-28}$$

where *n*-C_{*i*} is the concentration of *n*-FA of *i* number carbon. Analytical uncertainty for quantification of individual *n*-FA compounds is within 5% and the uncertainty for these ratio-based proxies is even smaller.

The water oxygen and hydrogen isotopes were analyzed by isotope ratio mass spectrometry (IRMS) at the University of Arizona Isotope Lab. Isotope values were reported in conventional δ notation relative to VSMOW (Vienna Standard Mean Ocean Water) for water δD and δ¹⁸O. The analytical precisions are 0.09‰ for water δ¹⁸O and 0.9‰ for water δD. Compound specific hydrogen values of *n*-FAs were measured by

the TRACE GC Ultra-Delta V IRMS in the State Key Laboratory of Loess and Quaternary Geology, CAS and reported in conventional δ notation relative to VSMOW. Only δD values in samples with signal intensity higher than a threshold level are reported in this manuscript, resulting in different number of n -FA δD values of different chain length. The measured δD values were further corrected by mathematically removing the isotopic contribution from the added methyl during the sample preparation (Hou et al., 2008). The precision was routinely checked by injection of the laboratory isotopic standards, after every ~ 4 –6 sample injections. The 1σ precision of the analysis was generally within $\pm 3\%$ indicated by standard deviation of duplicate analysis of the lab standard (Fig. 3).

3. Results

The water isotope values of lakes, rivers, wells/springs, and rain samples show a clear enrichment gradient from rivers and ground water, to open-basin lakes (e.g. Hurleg Lake), and then to closed-basin lakes (e.g. Gahai Lake and Toson Lake, Fig. 2, Supplemental Table S1). Generally, water samples from springs, wells, rain and puddles scatter close to the lower end of the global meteoric water line (Rozanski et al., 1993), while those from Hurleg Lake, Gahai Lake and Toson Lake show a clear local evaporation line ($\delta D = 5.5 * \delta^{18}O - 12.2$, Fig. 2). Water samples from wells and springs have the most negative mean $\delta^{18}O$ value of -10.2% ($n = 11$, standard deviation of 0.81%) and δD value of -68% ($n = 11$, standard deviation of 8.3%). Water samples from rivers also have similar negative mean $\delta^{18}O$ value of -9.6% ($n = 9$, standard deviation of 0.36%) and δD value of -61% ($n = 9$, standard deviation of 3.9%). In Hurleg Lake, water samples have an average $\delta^{18}O$ value of -4.9% ($n = 15$, standard deviation of 0.96%) and δD value of -39% ($n = 15$, standard deviation of 5.0%). At Gahai Lake, a closed basin lake fed by ground water, the average $\delta^{18}O$ value of water samples is 2.0% ($n = 4$, standard deviation of 0.37%), whereas the average δD value is -4.8% ($n = 4$, standard deviation of 1.3%). At the terminal Toson Lake which receives water from Hurleg lake, the average isotopic values of water samples are highest, about 7.7% for $\delta^{18}O$ ($n = 5$, standard deviation of 1.15%) and 30.6% for δD ($n = 5$, standard deviation of 7.6%).

n -C₂₄ and n -C₂₈ FA δD values show almost identical variations with n -C₂₆ FA δD values, in terms of both absolute values and direction of changes (Fig. 3C, Supplemental Table S4). Due to relatively low data resolution of n -C₂₄ and n -C₂₈ FA δD values (Fig. 3C), we chose n -C₂₆ FA δD values to represent isotopic signal of long-chain n -FA. The n -C₂₆

and n -C₁₆ FA δD records both show large amplitude of variability, especially after 6 cal ka BP. The n -C₂₆ FA δD values range from -111% to -192% over the past 10,500 years. The n -C₂₆ FA δD values were about -170% on average before 6 cal ka BP. After 6 cal ka BP, n -C₂₆ FA δD values show increased millennial- to centennial-scale variability. The n -C₁₆ FA δD values show an 80% change from -125% to -205% over the last 10,500 years (Fig. 3B). Before 6 cal ka BP, the n -C₁₆ FA δD shows similar trends and values with n -C₂₆ FA δD . After 6 cal ka BP, the n -C₁₆ FA δD shows a roughly opposite pattern with n -C₂₆ FA δD .

$\%C_{37:4}$ values range from 5% to 58% (Fig. 3D, Supplemental Table S2). After decreasing from $\sim 30\%$ to 10% at 9 cal ka BP, $\%C_{37:4}$ values remained relatively low ($\sim 10\%$) except during the periods of 5–4, 3–2 and 1–0 cal ka BP. The ACL_{n-FA} values (Fig. 3A, Supplemental Table S3) range from 20.0 to 24.4 over the past 10,500 years. The ACL_{n-FA} was at its lowest value of about 20.0 before ~ 8 cal ka BP. After 8 cal ka BP, it increased to the highest value of 24.4 at 6.2 cal ka BP. After gradually decreasing to 22.0 at 4 cal ka BP, the ACL_{n-FA} was relatively stable for the last 4000 years with small fluctuations between 21.0 and 23.0.

4. Discussion

4.1. Contribution of snowmelt water and evaporative enrichment on modern water isotopes

The water isotope results from rivers, groundwater, and lakes (Fig. 2, Supplemental Table S1) indicate that the cumulative evaporation effect becomes stronger from source water (e.g. river and well/spring), to hydrologically open lake systems (e.g. Hurleg Lake), and then to closed-basin lake systems (e.g. Gahai Lake and Toson Lake). The stable isotopes of river and groundwater samples are more negative than those of summer rain samples, suggesting that rivers and groundwater might be mainly fed by sources other than direct summer precipitation. Since the $\delta^{18}O$ values of about -10% in river and groundwater samples are close to those from the surface ice sample in the nearby Dunde ice cap at about -10.7% (Thompson et al., 1993), considerable part of river water and groundwater is probably derived from the snowmelt water. Isotopic results from lake water samples are systematically more enriched in both 2H and ^{18}O than those from rivers and groundwater. The lake water isotopic data fall on a local evaporation line with a slope of 5.5 (Fig. 2), while rain, rivers, and groundwater roughly fall on the global meteoric water line with a slope of ~ 8 (Fig. 2; Rozanski et al., 1993). This indicates substantial evaporative enrichment on δD and $\delta^{18}O$ values of lake water in the arid Qaidam Basin. Such evaporative enrichment on lake water isotopes can be further supported by higher δD and $\delta^{18}O$ values in saline lakes (e.g. Gahai Lake and Toson Lake) than freshwater lakes (e.g. Hurleg Lake).

4.2. Hydroclimatic characteristics of the Hurleg Lake region

Before discussing down-core data, we acknowledge there might exist quite large dating uncertainties for core HL06-1, especially considering the quite large 2758 ^{14}C -year “old carbon” effect (Zhao et al., 2010). In reality, this old carbon correction may not even be constant through time. We made some efforts trying to minimize such effect, including using the same dating material (*Ruppia*), testing reproducibility of age models of cores collected from different part of the lake, and selecting dating samples with a narrow range of carbonate contents (30% to 40%, Zhao et al., 2013). Also, many records discussed in this study are derived from the same sediment core. Thus the age model uncertainty will not impact our inference of the internal relationships between proxies, which our main points are built on.

The $C_{37:4}$ alkenone has been widely found in Chinese lakes (Chu et al., 2005; Liu et al., 2006a, 2008b, 2011). Recent studies around Lake Qinghai and the Qaidam Basin indicate that $\%C_{37:4}$ could be used as a proxy for changes in lake salinity, with lower $\%C_{37:4}$ corresponding to higher salinity, and vice versa (Liu et al., 2011; He et al., 2013). In Hurleg

Fig. 2. Water isotope results from rain, river and lakes around the Lake Hurleg region. The global meteoric water line (blue, Rozanski et al., 1993) and regional evaporation line (red) are also shown for comparison.

Fig. 3. Biomarker results from core HL06-1 at Hurler Lake. (A) *n*-fatty acid ACL. (B) *n*-C₁₆ fatty acid δ D. (C) *n*-C₂₄, *n*-C₂₆ and *n*-C₂₈ fatty acid δ D. (D) Alkenone-based %C_{37:4}. Analytical error of the *n*-fatty acid δ D records is also shown.

Lake, only %C_{37:4} values of samples with unknown contamination at C_{37:3} component (4.9–4.3, ~2, and 1.1–0.6 cal ka BP, see details in Zhao et al., 2013) might contain relatively large uncertainty. The samples are mainly from the intervals when temperature appears low (Zhao et al., 2013), lake level is high (Zhao et al., 2010) and the %C_{37:4} values are generally high. Thus if we take the unknown contamination at C_{37:3} into consideration, the real %C_{37:4} values in these samples can only be higher than the measured high values. Therefore, the unknown contamination at C_{37:3} doesn't impact much on the variation of %C_{37:4} in Hurler Lake as a salinity indicator.

Our %C_{37:4}-based salinity record shows consistent pattern with carbonate-content based lake-level reconstruction from the same core, with lower %C_{37:4} values (higher salinity) corresponding to lower lake levels (Fig. 4B, C, Zhao et al., 2010). As shown in our previous studies, change in carbonate content under different water depths at Hurler Lake is controlled by the dilution of endogenic carbonate by fine-grained silicates delivered from surrounding deserts with sparse vegetation cover (Zhao et al., 2010). With the chronological uncertainty, the lake-level record is further supported by the quartz optically stimulated luminescence ages for beach deposits (Fan et al., 2014), which suggests higher lake level at 6.8–6.4, 5.0–4.7 and 2.2–1.4 cal ka BP. Thus, the anti-correlation of salinity and lake level has been consistent throughout the Holocene at Hurler Lake. Together with U^K₃₇-based temperature reconstruction (Fig. 4A), where the “predicted warm (or cold)” based on multiple evidence comes from unreported data due to either very low alkenone concentrations or contamination by unknown compounds (Zhao et al., 2013), our data reveal a warm-dry/cold-wet climate association at millennial-scale. Also, these data support our

previous finding of intensified millennial-scale climate variability after the mid-Holocene, caused by an increase in climate sensitivity of the arid Qaidam Basin which followed an abrupt decrease in vegetation (forest) cover in the monsoonal region surrounding the eastern part of the basin at that time (Zhao et al., 2013).

4.3. Long-chain *n*-FA δ D as a proxy for terrestrial vegetation source water δ D

Long-chain leaf wax δ D values are mainly controlled by the original isotopic compositions of the source water used by higher terrestrial plants and the isotopic fractionation between leaf wax and water due to the evapotranspiration (Sachse et al., 2004, 2012; Liu et al., 2006b, 2008b; Hou et al., 2008; Feakins and Sessions, 2010; Polissar and Freeman, 2010; McInerney et al., 2011; Nelson et al., 2013). The consistent variations of *n*-C₂₄, *n*-C₂₆, and *n*-C₂₈ FA δ D values (Fig. 3C) probably indicate that these δ D proxies may all faithfully record changes of δ D in terrestrial plants. δ D fractionation in leaf wax may also vary with different terrestrial vegetation types (e.g. trees, shrubs and grass; Sessions et al., 1999; Liu et al., 2006b; Smith and Freeman, 2006; Chikaraishi and Naraoka, 2007; Hou et al., 2007; Zhang and Sachs, 2007; Feakins and Sessions, 2010; McInerney et al., 2011; Sachse et al., 2012). For example, some studies showed that the mean C₄ grass leaf wax δ D values are ~15‰ higher than C₃ grass but ~20‰ lower than those of C₃ trees and shrubs (Liu et al., 2006b; Smith and Freeman, 2006; McInerney et al., 2011; Sachse et al., 2012). Therefore, in an extreme situation, the absolute change in leaf wax δ D from trees and shrubs to C₃ grass can account for 35‰ variation in leaf wax δ D. At Hurler Lake, ACL_{*n*-FA} values range

Fig. 4. Comparison of isotopic records (D–F) with temperature (A) and moisture (B–C) records core HL06-1 at Hurleg Lake. (A) Alkenone-based $U_{K'37}$. Predicted warm and cold conditions based on alkenones results were also indicated (Zhao et al., 2013). (B) Alkenone-based $\%C_{37:4}$ salinity records. (C) Lake level record indicated by carbonate contents (Zhao et al., 2010). (D) n - C_{26} fatty acid δD . (E) n - C_{16} fatty acid δD and $\delta^{18}O$ values of endogenic calcite from core HL05-2 (Zhao et al., 2010). (F) $\Delta DC_{16:2:6}$. Light green shadings indicate periods of weaker evaporation at Hurleg Lake over the past 6000 years.

between 21.5 and 24 (Fig. 3A), probably indicating limited vegetation changes during the last 6000 years, when n - C_{26} FA δD values show large variations between -120% and -200% (Fig. 3C). This is supported by fossil pollen data from Hurleg Lake indicating that the surrounding region has been covered by desert steppe and desert vegetation dominated by pollen from *Nitraria*, *Artemisia* and *Chenopodiaceae* during the entire Holocene (Zhao et al., 2007). In view of that, the estimated isotopic difference induced by the vegetation types might be much less than 35‰, failing to explain the $\sim 80\%$ variation in the n - C_{26} FA δD record (Fig. 3C). Therefore, the isotopic composition in source water used by terrestrial plants plays a major control on n - C_{26} FA δD , while vegetation

type only exerts minor influence. This idea is also supported by studies of lacustrine sediment transects across the mid-latitude regions (Sachse et al., 2004; Hou et al., 2008; Douglas et al., 2012).

In order to find climatic controls of the down-core n - C_{26} FA δD variation, we compare the n - C_{26} FA δD record with the temperature (Fig. 4A; Zhao et al., 2013), lake-level (Fig. 4C; Zhao et al., 2010) and salinity (Fig. 4B, this study) records from the same sediment core. The n - C_{26} FA δD values show an enrichment trend from more negative value around 7.5 cal ka BP to more positive value around 6 cal ka BP, while the temperature-moisture records show a cool-wet condition during this interval (Zhao et al., 2010, 2013). Over the last 6000 years, the n - C_{26} FA δD

data and temperature-moisture records both show increased millennial-scale variability. Five periods of high n -C₂₆ FA δ D values centered at ~0.8, 2.2, 3.2, 4.6 and 5.8 cal ka BP (Fig. 4D) coincide with the cold-wet periods. Such relationship is opposite to what would be expected, if the water isotopic values were mainly controlled by temperature and moisture availability, as shown by the long-chain n -alkane δ D record from Sugan Lake in the western Qaidam Basin (Wang et al., 2013).

Analysis of modern precipitation water from the nearby Delingha Weather Station indicates a strong temperature influence on precipitation isotopic composition in the Qaidam Basin, with higher temperature corresponding to higher isotopic value in precipitation, and vice versa (Tian et al., 2003). Also direct evaporation caused by temperature increase would further enrich the precipitation isotopic values. Therefore, the temperature and the temperature-induced evaporation changes cannot explain changes in n -C₂₆ FA δ D values, and the major source water for terrestrial vegetations around Hurleg Lake may not come from direct precipitation over the entire Holocene. We argue that the water source for terrestrial vegetations around Hurleg Lake mainly comes from rivers and groundwater originating from the snowmelt water in the high-elevation Qilian Mountain to the north, as is the case for source water of Hurleg Lake. This is supported by nearly identical $\delta^{18}\text{O}$ values from modern river/groundwater samples (-9.3% to -10.5%) and from the nearby Dundee ice core over the past 12,000 years (mostly from -10.5% to -12% , with the average values of -11.0% , Thompson et al., 1993). We can also make a simple calculation about the resulted lipid δ D values, taking the modern source waters (rivers and groundwater) δ D values of about -60% and the apparent isotopic fraction between terrestrial plants and source water of about -95% in arid environments similar to the Hurleg Lake region (Gunther et al., 2013; Feakins et al., 2014; Aichner et al., 2015). Although we do not have lipid δ D values from modern sediments, the resulted -155% is quite close to the average of down-core n -C₂₆ FA δ D value of -147% , further supporting that the source water for terrestrial vegetations mainly coming from snowmelt water. In this sense, variations in n -C₂₆ FA δ D values are likely influenced by the snowmelt water. During warm periods, more water melts down from the nearby glaciated mountains, and isotopic values of water used by terrestrial plants get lower. In cold periods when melt water decreased, direct precipitation probably contributes more to the source water of terrestrial vegetation and thus increases δ D and $\delta^{18}\text{O}$ values. In an extreme situation, $\delta^{18}\text{O}$ values of the Dundee ice core samples are up to 13‰ lower than that of a modern rain sample (equivalent to ~100‰ in δ D, Fig. 2), even larger than the up to 80‰ variation in n -C₂₆ FA δ D record (Fig. 4D). Therefore, it is likely that variations in n -C₂₆ FA δ D values during the Holocene were mainly controlled by changes of the contribution from snowmelt water from high-elevated mountains around the region, probably with minor modification by vegetation type. As for the case at Sugan Lake, more negative long-chain n -alkane δ D values occurred at cold-wet periods during the late Holocene (Wang et al., 2013). Possibly, the long-chain leaf wax δ D values in Sugan Lake are mainly controlled by vegetation type, which depends on moisture variation (Wang et al., 2013). Or more probably, evaporation has controlled terrestrial water isotopic signal just like what happened on the hydrology variation of Sugan Lake (He et al., 2013). The different isotopic variation models between lakes on the Tibetan Plateau thus highlight the complexity of using lipid δ D as regional moisture indicator and the necessity of systematic analysis on specific lake hydrology before interpreting the δ D results.

4.4. $\Delta D_{C16-C26}$ as a proxy for evaporation changes in lake

As suggested by Wang and Liu (2012), n -C₁₆ FA δ D is mainly controlled by isotopic compositions of lake water used by aquatic plants/algae in Qinghai Lake, which is close to Hurleg Lake. This notion is also correct for Hurleg Lake, considering the fact that the n -C₁₆ FA δ D record

is in general consistent with the endogenic calcite $\delta^{18}\text{O}$ data from core HL05-2 from shallower part of Hurleg Lake (Fig. 4E; Zhao et al., 2010), which has similar lithologic pattern with core HL06-1. To the first order, the n -C₁₆ FA δ D record resembles the endogenic calcite $\delta^{18}\text{O}$ record, especially when considering the different sampling resolution and chronological uncertainty between two cores (Zhao et al., 2010, 2013). Therefore, n -C₁₆ FA δ D values from the Hurleg Lake core largely reflect the isotopic composition of lake water as $\delta^{18}\text{O}$ values of endogenic calcite do.

Since the ultimate water source is comparable for terrestrial plants around Hurleg Lake and aquatic organisms within the lake, the δ D difference between aquatic and terrestrial source water allows us to cancel out the moisture source and biosynthetic fractionation effect on isotopes and focus only on the regional evaporation in the lake surface (Sachse et al., 2004; Mügler et al., 2008). As discussed earlier, the isotopic composition of source snowmelt water for Hurleg Lake and surrounding soil is indicated by n -C₂₆ FA δ D values, while the isotopic composition of lake water is recorded by n -C₁₆ FA δ D values. The difference between n -C₁₆ FA δ D and n -C₂₆ FA δ D (defined as $\Delta D_{C16-C26}$) can be an effective way to track changes in evaporation at Hurleg Lake. We understand that using $\Delta D_{C16-C26}$ as an evaporation proxy is a little simplistic, considering that factors other than evaporation might also have minor to moderate impact on the isotopic fractionation. However, we believe that it is still much better than δ D values from individual n -FAs alone, either C₁₆ or C₂₆, since both of them are affected by the complicated isotopic signal in water sources, which is largely cancelled out in the calculated difference. In cold and humid conditions, aquatic n -FAs isotopes are less affected by lacustrine evaporation enrichment, making terrestrial n -FA δ D values slightly higher than aquatic n -FA δ D values due to the terrestrial evapotranspiration effect (Sachse et al., 2004, 2006). In warm and arid conditions, extensive enrichment of aquatic n -FA δ D occurs due to the intensive evaporation, overwhelming the terrestrial evapotranspiration effect (Sachse et al., 2004, 2006) and leading to opposite directions on the difference between aquatic and terrestrial n -FAs isotopes (Mügler et al., 2008).

During the Holocene Climate Optimum before 6 cal ka BP, the $\Delta D_{C16-C26}$ seem to show a moderate and stable evaporation effect on lake water, while the isotopic signals of both lake (n -C₁₆ FA) and surrounding soil water (n -C₂₆ FA) remain in negative values (Fig. 4). Other data including temperature, salinity, and lake-level proxies indicate relatively warm, slightly enriched water body and high salinity conditions, although a short period around 9 cal ka BP showing anomalous cooling and low salinity condition. This could be interpreted by the melting of snow water under warm conditions. During the Holocene Climate Optimum, the higher summer air temperature would considerably increase the snowmelt from the high-elevation mountains to the north of the Qaidam Basin. The large contribution of melt water input with very low isotope values into Hurleg Lake via rivers and groundwater would possibly increase the lake volume and lead to low n -C₁₆ and n -C₂₆ FA δ D values in the relatively high evaporation under a warm condition. Meanwhile, increased river and groundwater input might bring more mineral solutes from the catchment basin, which could account for the increase of lake salinity in a slightly enriched lake water body condition. Thus, temperature induced snowmelt water input was the dominate control for hydrology changes in this region. However, since there are limited data points during the early Holocene in our $\Delta D_{C16-C26}$ record and chronological uncertainty before 7.4 cal ka BP, our view on the early Holocene status is largely tentative and needs further confirmation.

After 6 cal ka BP, our $\Delta D_{C16-C26}$ data show increased millennial-scale variability from ~-50‰ to 40‰, in terms of both frequency and amplitude (Fig. 4F). These variations coincide with intensified millennial-scale changes in established temperature and hydrology records since an abrupt decrease in vegetation (forest) cover in the monsoonal region surrounding the eastern part of the Qaidam Basin around 6 ka, probably reflecting an increased climate sensitivity within the arid basin (Zhao et

al., 2013). When climate getting warmer, more snowmelt water (as indicated by the lower $n\text{-C}_{26}$ FA δD values) would discharge into the lake. Meanwhile, the even stronger evaporation effect caused by temperature rise would strongly enrich the lake water isotopes. On the other hand, when climate getting colder, less snowmelt water (as reflected by the higher $n\text{-C}_{26}$ FA δD values) can discharge into the lake, but the weaker evaporation effect would decrease the water loss and induce higher lake levels. Therefore, evaporation became the main control on the regional hydrology change during the mid- and late Holocene.

Overall, using modern water and down-core long-chain $n\text{-FA}$ isotopic data from the Tibetan Plateau, we provide evidence that the later is likely influenced by snowmelt water from nearby higher mountains. This fact is often neglected but may complicate the interpretation of stable isotope values in lake sediments in comparable environments. Further, the difference between long-chain and short-chain $n\text{-FA}$ δD derived from an open lake could still be used to better infer evaporation effect in an arid region.

5. Conclusions

- 1) The stable isotope data of modern water samples in the Qaidam Basin indicate that snowmelt water originated from high-elevation mountains to the north is the main water source for lakes in the eastern Qaidam Basin via river and groundwater discharge, and that evaporation is the main control for lake hydrology in this region.
- 2) Analysis of $n\text{-C}_{26}$ FA δD in down-core sediment samples of Hurler Lake show that changes in soil water isotopic values are mainly controlled by the snowmelt water around this open lake system during the Holocene. This result is different with the previous finding from Sugan Lake in the western Qaidam Basin where evaporation induced vegetation might be the major control. Meanwhile, changes in $n\text{-C}_{16}$ FA δD values at Hurler Lake reflect the evolution of isotopic compositions of lake water.
- 3) The $\Delta\text{D}_{\text{C}_{16}\text{-C}_{26}}$ values at Hurler Lake can be used to evaluate the evaporation effect and to infer the controlling factor for changes in lake hydrology in this climatic complex region. Before 6 cal ka BP, the moderate $\Delta\text{D}_{\text{C}_{16}\text{-C}_{26}}$ values with low $n\text{-C}_{16}$ and $n\text{-C}_{26}$ FA δD values probably indicate large contribution of snowmelt water even under the relatively high evaporation condition during the Holocene Climate Optimum. After 6 cal ka BP, the increased millennial-scale variability in $\Delta\text{D}_{\text{C}_{16}\text{-C}_{26}}$ values suggest that evaporation became the major control on lake hydrology changes, probably resulting from the increased climate sensitivity after an abrupt decrease in vegetation (forest) cover in the monsoonal region surrounding the eastern part of the Qaidam Basin.
- 4) Our study highlights the importance of systematic analysis on modern processes of regional hydrology before using δD or/and $\delta^{18}\text{O}$ values for paleoclimate reconstructions in a specific lake. Also, our data indicate that the comparison of long-chain and short-chain $n\text{-FA}$ δD might be an effective way to better infer hydroclimate variations in the climatic complex region like the northeastern Tibetan Plateau.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.chemgeo.2016.07.024>.

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