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Carbon isotope and environmental changes in lakes in arid Northwest China

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Abstract The stable carbon isotope composition ($\delta^{13}C$) of lacustrine sediments, which can record changes in past environmental conditions such as the C_4/C_3 terrestrial vegetation composition, has been widely used for the reconstruction of terrestrial ecosystems and global climate changes. It has also been widely used in paleolimnological studies in arid northwestern China. In recent years, however, an increasing number of studies have enriched the environmental significance reflected by the stable carbon isotope geochemistry of lakes; therefore, the interpretation of lake $\delta^{13}C$ variations in paleoenvironmental reconstructions should be carefully evaluated from various aspects. In this paper, previous studies from the past several decades on the lacustrine $\delta^{13}C$ variations in arid northwestern China were reviewed, and the possible mechanisms of the $\delta^{13}C$ variations were discussed. Our study will provide a reference for the application of carbon isotope geochemistry in paleoenvironmental reconstructions.

Keywords Carbon isotope, Lake, Arid region of northwestern China, Total organic carbon, Organic biomarkers

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1. Introduction

Lake sediments record the changes in natural ecosystems such as the terrestrial hydrology and vegetation in the lake drainage areas and the information of related climate and environmental changes. Because of its continuity and high-resolution characteristics, lake sediments can serve as one of the ideal archives for studying regional and global climate change (Wang and Zhang, 1999). As continuous deposits of soils, ocean sediments and peat bogs are rare in the inland arid regions of China, lake sediments become the main object for paleoenvironmental studies. Due to the arid climate, lakes in Qinghai and Xinjiang located in northwest China have relatively rapid depositional rates and high resolutions and are therefore ideal for paleoclimatic studies.

The extraction method of reliable past environmental information from lake sediments is a concern in lake geochemical studies (Shen, 2009). The abundant organic matter in lakes provides rich materials for environmental studies, while the stable carbon isotope composition ($\delta^{13}C$) of lake sediments can trace the history of past environmental changes. In the past few decades, the $\delta^{13}C$ of total organic carbon (TOC) has substantially contributed to the geochemical basis for the lacustrine carbon cycle and environmental change. In particular, since the $\delta^{13}C$ values are quite different for terrestrial C_3 and C_4 plants, with mean values of −26.7‰ and −12.6‰, respectively, for the two types of plants, the information on vegetation changes can be preserved in the $\delta^{13}C$ of TOC in lake sediments. Therefore, the $\delta^{13}C$ of lacustrine TOC has attracted considerable attention from lake geochemists and paleoclimatologists around the world.

Organic biomarkers in lake sediments, which are con-

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sidered an important media recording the regional ecology and corresponding environmental change, have been widely used in reconstructing past environmental changes (Castañeda and Schouten, 2011). In the past decades, along with the development of analytical methods, our knowledge on the compound-specific $\delta^{13}C$ values of biomarkers in geological records has developed rapidly, and the compound-specific $\delta^{13}C$ has potentially become a new method for the reconstruction of past global environmental changes (Hayes et al., 1990; Freeman et al., 1990; Rieley et al., 1991; Xie et al., 2000, 2003; Sauer et al., 2001; Zhang and Jia, 2009). It has now been widely used in paleolimnological studies in different regions of the world (Meyers, 2003; Castañeda and Schouten, 2011). Notably, long-chain *n*-alkanes and *n*-fatty acids, which are primarily derived from plant leaf wax, are relatively abundant in geological bodies and resistant to degradation (Eglinton and Hamilton, 1967; Yang and Huang, 2003; Peters et al., 2005; Castañeda and Schouten, 2011). Thus, the $\delta^{13}C$ of *n*-alkanes or *n*-fatty acids extracted from lacustrine sediments can faithfully record changes in the paleoclimatic conditions (Sauer et al., 2001; Yang and Huang, 2003).

Over the past several decades, a large amount of research has been carried out on the modern geochemical processes and paleoclimate applications of sedimentary organic matter $\delta^{13}C$ in lakes in arid northwest China. Here, we systematically review the research progress of lacustrine organic matter $\delta^{13}C$ in arid northwest China and discuss the mechanisms of changes in lacustrine organic matter $\delta^{13}C$ in an effort to provide a reference for the application of carbon isotope geochemistry in paleoclimatic studies.

2. The source of total organic carbon in lake sediments and its geochemical implications

The organic matter in lacustrine sediments is the complex mixture of carbohydrates, lipids, proteins, etc., which are mainly derived from lake aquatic plants and terrestrial plants around lakes. Therefore, the source of lacustrine organic matter includes two parts: aquatic plant inputs and terrestrial plant inputs. Endogenous sources of lake sediments, including planktonic or benthic algae, submerged macrophytes and emergent plants, respond to the environmental information in lakes. In contrast, exogenous inputs in lake sediments, which are transported by river or wind (Meyers and Ishiwatari, 1993), represent the change in the growing environmental information of terrestrial plants from the surrounding region. The endogenous and exogenous sources have significantly different carbon isotopic compositions. For example, the carbon isotopes of dissolved inorganic carbon (DIC) in water and atmospheric CO_2 show significant differences. In addition, the changes in the lake water temperature and air temperature also substantially differ. Therefore, distinguishing the source of organic matter in lake sediments is the basis of lacustrine isotope geochemistry. The previous studies suggest that the C/N ratio of organic matter in the sediment could determine whether the organic matter in lake sediments originates from the lake endogenous or exogenous sources.

The organic matter in lake sediments is generally represented as total organic carbon (TOC), which depends on the productivity in lakes or lake drainage areas, and their preservation in sediments (Meyers and Lallier-vergés, 1999). As plant growth is affected by factors such as temperature, precipitation and other lake conditions, TOC could be used as a proxy for paleoclimatic and paleoenvironmental changes. For instance, a cold and dry climate could shorten the growth period of plants, decrease photosynthesis, extend the lake icebound season and then reduce the primary productivity, leading to decreased TOC. Conversely, a warm and humid climate is conducive to the higher biomass of terrestrial and aquatic plants and increased TOC in lake sediments (Chen et al., 2002). Therefore, the TOC indicator has been widely used in regional and global climate and environmental change studies (Lücke et al., 2003).

The interpretation of the lacustrine TOC indicator differed in the various regions. In the lakes of the Tibet Plateau and western China, for example, the TOC content in Nam Co is considered to be related to either increased endogenous productivity or increased exogenous input (Zhu et al., 2008), and TOC indicates the air and water temperature changes since the Holocene (Doberschütz et al., 2014). In Beston Lake (Wünnemann et al., 2006) and Hurleg Lake (Zhao et al., 2010), TOC records are considered to represent endogenous biomass or organic matter productivity and reflect changes in the lake productivity under different climatic conditions. In Lake Qinghai, TOC fluxes recorded the increases in terrestrial plant biomass and lake productivity under warm and humid climates, revealing the strength changes in the Asian summer monsoon (An et al., 2012). The TOC content in Lake Daihai is thought to be mainly from terrestrial plant inputs and used to indicate the regional precipitation intensity (Xiao et al., 2006). In Lake Yanhaizi of Inner Mongolia, the TOC content is also thought to represent the effective precipitation changes, because the humid climate promotes plant biomass and increases the TOC content (Chen et al., 2003). In previous studies, most of the lacustrine TOC records are used to indicate changes in the biomass but also emphasize one or more climate/environmental controlling factor. The trend of biomass changes in different lakes during the Holocene could reveal the regional climate and environment characteristics in western China. However, the comparison of TOC records from different lakes shows that the biomass in lakes is quite different, and even the opposite trends occur during some time periods

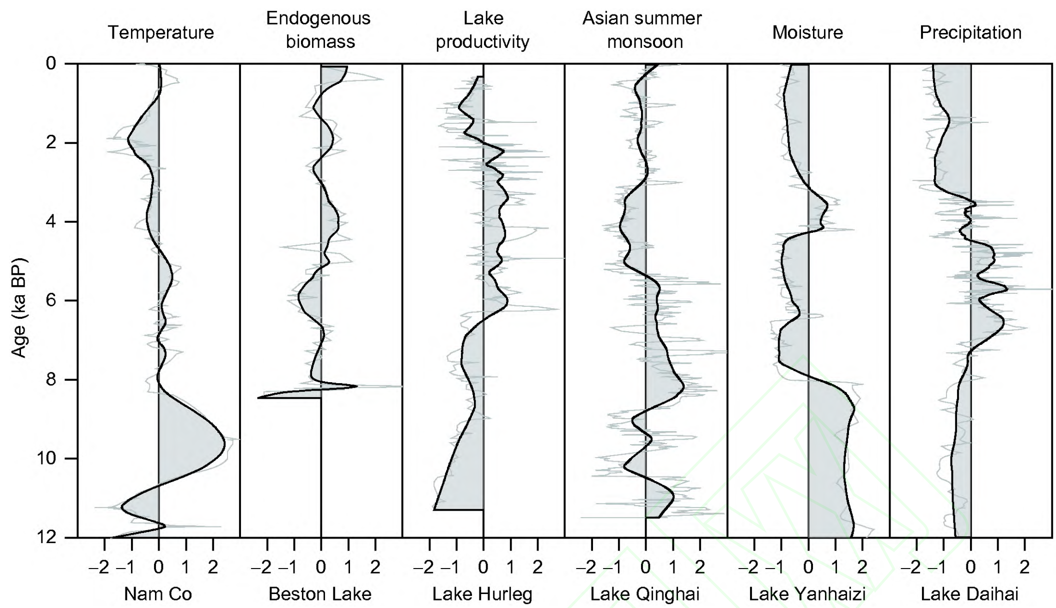

Figure 1 The Holocene lake TOC records in western China. Nam Co: Doberschütz et al. (2014); Beston Lake: Wünnemann et al. (2006); Lake Hurleg: Zhao et al. (2010); Lake Qinghai: An et al. (2012); Lake Yanhaizi: Chen et al. (2003); Lake Daihai: Xiao et al. (2006). All data are normalized to z-scores and the shading areas indicate the millennium trends.

(Figure 1). Therefore, the biomass changes cannot be completely explained by the regional climate or environmental changes.

To discuss TOC sources in lacustrine sediments, for the characteristics of internal and external input mixing, we first need to answer a few questions: Is the sediment TOC primarily from terrestrial or lake endogenous sources? If the input source is mixed, is the mixing ratio stable or not? Are the response mechanisms of terrestrial and aquatic plant biomass to climate change consistent? If the response of terrestrial plants and aquatic plants to climate change is different, the possible unsynchronized trend between air temperature and lake temperature (Schneider et al., 2009), the different carbon sources utilized by terrestrial plants (from CO_2) and aquatic plants (from DIC) or the differences in synthesis pathways (Touchette and Burkholder, 2000; Maberly and Madsen, 2002) are all associated with differences in the source of organic matter. Therefore, distinguishing the source of organic matter in lacustrine sediments is the basis of lacustrine organic geochemistry and is very important for the use of TOC records to discuss regional climatic characteristics and even climate model changes.

C/N ratio is a simple and effective indicator to distinguish the TOC source in lacustrine environments. Microalgae in lakes have higher pro-protein content than terrestrial plants, and the C/N ratio of aquatic phytoplankton is usually between 4 and 10, which is well below the C/N ratio of terrestrial plants (Hedges and Oades, 1997). Therefore, a high C/N ratio in the lake sediment would indicate that the organic matter is mainly from terrestrial plants, while a low C/N ratio indicates that the lake endogenous organism is the major contributor to organic matter (Meyers and Lallier-vergés, 1999). For example, the total C/N ratio in the Nam Co is less than 10, and the sediment is mainly derived from aquatic plants (Doberschütz et al., 2014). The total C/N ratios in Daihai and Yanhaizi are greater than 10, and the TOC in sediments is considered to be mainly from terrestrial plants (Chen et al., 2003; Xiao et al., 2006).

However, the C/N ratio also has some defects in distinguishing the source of lacustrine TOC. Earlier studies suggested that the distinction of the boundary between the C/N ratio of phytoplankton and terrestrial higher plants is clear (Meyers and Lallier-vergés, 1999). However, subsequent studies noted that the C/N ratio of macrophytes overlapped with terrestrial plants (Finlay and Kendall, 2007), leading to many uncertainties in using the C/N ratio to distinguish terrestrial or aquatic sources (Figure 2). Therefore, more indicators are still needed to comprehensively analyze the source of sediments in lakes, e.g., using the $\delta^{13}C$ differences between the organic molecular compounds of terrestrial and aquatic plants.

3. Source and geochemical significance of organic biomarkers in lakes

The organic matter in lacustrine sediments has complex sources, which are mainly divided into internal and external inputs. The exploration of organic biomarkers originating from specific sources in lacustrine sediments has become one of the key issues in the study of lake geochemistry. Investigations over the past few decades have shown that *n*-

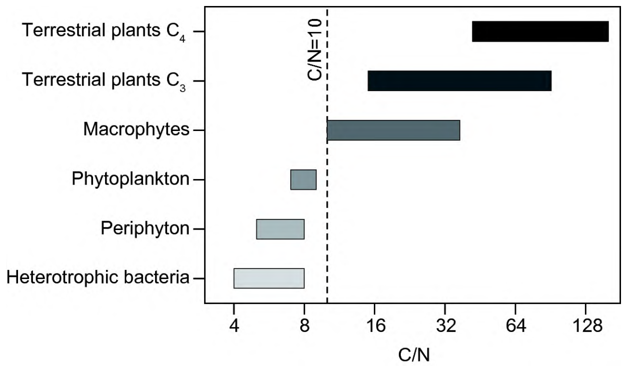

Figure 2 C/N differences among various biological sources, modified from Meyers and Lallier-vergés (1999), Finlay and Kendall (2007).

alkane and *n*-alkanoic acids in lacustrine sediments could indicate different input sources. For example, long-chain *n*-alkanes (C_{27}, C_{29} and C_{31}) were thought to be derived from terrestrial plants and short- or mid-chain *n*-alkanes (C_{17}, C_{19} and C_{21}) were thought to be produced by algae and aquatic plants. Therefore, the relative contribution of organic matter in lakes between internal and external inputs can be achieved by analyzing the variations in *n*-alkane abundances.

n-Alkanes, a class of organic biomarkers with simple molecular structures, are biosynthesized from long-chain *n*-alkanoic acids via the decarboxylation pathway (Kunst and Samuels, 2003). They belong to long-lived compounds that can be not only well preserved for millions of years but also easily extracted and analyzed, causing them to attract particular interest for paleoclimatic reconstructions (Schimmelmann et al., 1999; Sessions et al., 2004; Sachse et al., 2006; Eglinton and Eglinton, 2008). However, *n*-alkanes in lacustrine sediments often have various source pools, including terrestrial plants, aquatic plants and lower organisms (Rao et al., 2014), and their distributions are generally used to determine different source inputs. Long-chain *n*-alkanes (C_{27}–C_{33}) were believed to typically originate from terrestrial and emersed plants (Cranwell et al., 1987; Street-Perrott et al., 1997; Ficken et al., 2000; Hu et al., 2014), mid-chain *n*-alkanes (C_{21}–C_{25}) are mainly produced by floating/submerged plants (Ficken et al., 2000; Duan and Xu, 2012; Duan et al., 2014) and sphagnum (Nott et al., 2000; Pancost et al., 2002), and short-chain *n*-alkanes are derived from algae or bacteria (Han and Calvin, 1969; Cranwell et al., 1987). Ficken et al. (2000) defined a proxy named P_{aq}[P_{aq}=(C_{23}+C_{25})/(C_{23}+C_{25}+C_{29}+C_{31})] for distinguishing the *n*-alkane contribution from submerged/floating plants relative to emergent and terrestrial plants, where P_{aq}<0.1 corresponds to terrestrial plants, 0.1–0.4 to emergent plants and 0.4–1 to submerged/floating plants (Figure 3a).

Investigations of aquatic plants from the Tibetan Plateau found that emersed plants were dominated by C_{27} and C_{29}, submerged plants by C_{23} and C_{25}, and mid-chain *n*-alkanes were effective indicators of paleohydrological conditions (Aichner et al., 2010a, 2010b). By analyzing the *n*-alkanes in terrestrial and aquatic plants in Qinghai Lake and Gannan Gahai Lake, Duan and Xu (2012) and Duan et al. (2014) demonstrated that terrestrial and emersed plants were dominated by C_{27}, C_{29} and C_{31}, while submerged plants were dominated by C_{23} and C_{25}. Similar results were reported for other lakes on the Qinghai-Tibetan Plateau (Mügler et al., 2008; Guenther et al., 2013), as well as in Japan and Thailand (Chikaraishi and Naraoka, 2003), Europe (Sachse et al., 2004; Mügler et al., 2008), America (Mead et al., 2005; Gao et al., 2011) and Africa (Ficken et al., 2000).

However, some recent studies have found that submerged plants may have large contributions to the long-chain *n*-alkane pools in lacustrine sediments. For example, Aichner et al. (2010a) found that parts of the submerged plants displayed higher proportions of long-chain *n*-alkanes in some shallower lakes, which may be due to their adaptation to the partial exposure to air (Figure 3b). Liu et al. (2015, 2016a) analyzed *n*-alkane distributions in submerged and terrestrial plants in the Qinghai Lake area, suggesting that submerged plants could produce considerable amounts of long-chain homologues (Figure 3c). Therefore, the extent to which their accumulation is influenced by *n*-alkanes in lacustrine sediments originating from aquatic plants needs further study.

Similarly, the distributions of *n*-alkanoic acids are also used to determine organic matter inputs to lacustrine sediments. The short-chain *n*-alkanes (C_{14}–C_{18}) are thought to be mixed by algae, submerged plants and terrestrial plants; mid-chain *n*-alkanes (C_{20}–C_{24}) mainly originate from submerged/floating plants, and long-chain *n*-alkanes (C_{26}–C_{32}) are produced by terrestrial plants (Simoneit, 1977; Street-Perrott et al., 1997; Tuo et al., 2011). Silliman et al. (1996) proposed that *TAR*[*TAR*=(C_{24}+C_{26}+C_{28})/(C_{12}+C_{14}+C_{16})] can be used to evaluate the *n*-alkanoic acid contribution of aquatic plants and terrestrial plants to lacustrine sediments, with high *TAR* values corresponding to a high contribution of terrestrial plants and vice versa. Ishiwatari et al. (2006) used the ACL of *n*-alkanoic acids to evaluate organic matter inputs to the sediments of Lake Baikal, with high ACL values for terrestrial plants and low ACL values for aquatic plants. Gao et al. (2011) analyzed the *n*-alkanoic acid distributions in terrestrial and aquatic plants in Blood Pond, finding that *n*-alkanoic acids in submerged/floating plants were dominated by C_{20}–C_{24}, and terrestrial plants were dominated by C_{26}–C_{32}. Wang and Liu (2012) investigated the *n*-alkanoic acids in terrestrial and aquatic plants in Lake Qinghai and found that the $ACL_{16\text{-}32}$ values of *n*-alkanoic acids between terrestrial (avg. 23.3) and aquatic plants (avg. 18.6) has significant differences (Figure 4), suggesting that the $ACL_{16–32}$ values of *n*-alkanoic acids could be a potential proxy in distinguishing the organic matter input sources in lacustrine sediments. The results of Fang et al. (2014) conducted in Lake Dianchi indicated that short-chain *n*-alkanoic acids were mainly pro-

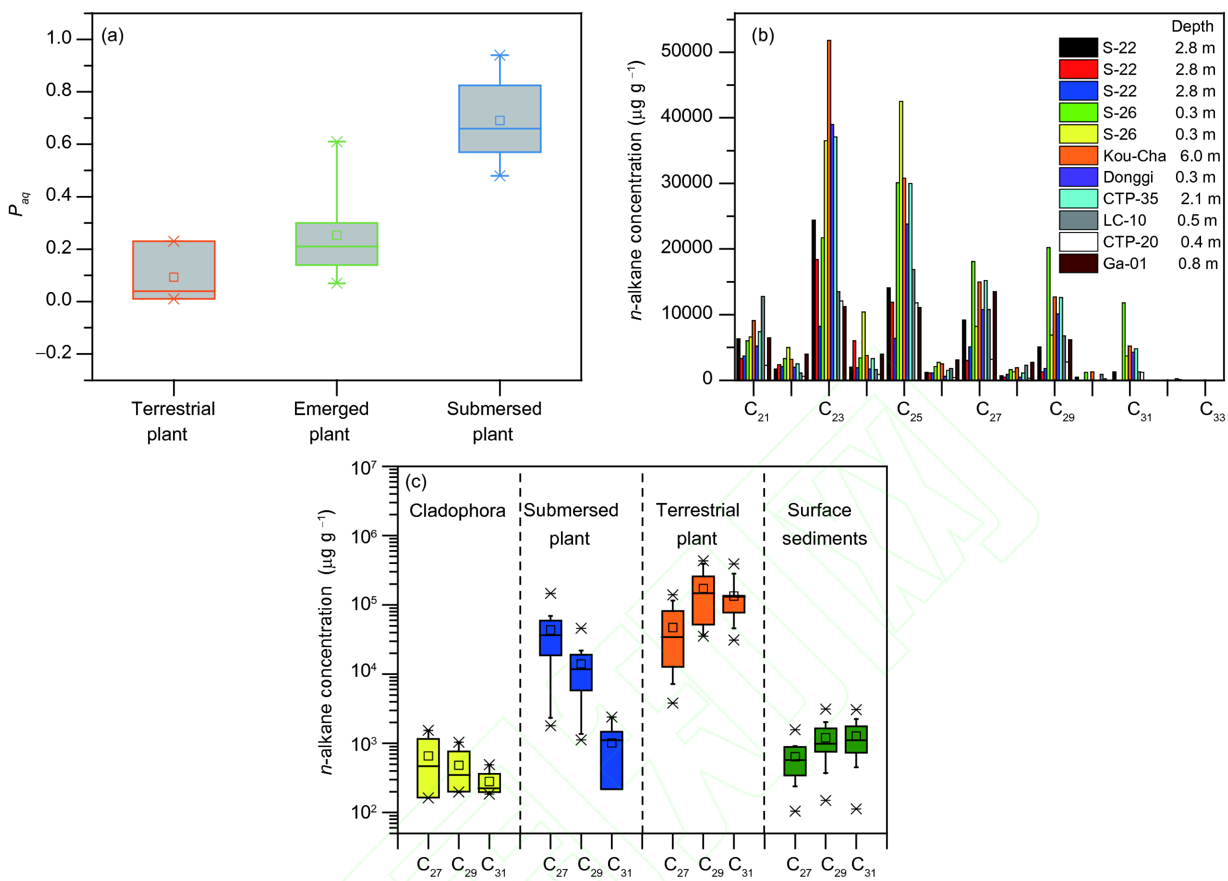

Figure 3 *n*-Alkane concentrations and proxy in terrestrial and aquatic plants. (a) P_{aq} values in different plants reported by Ficken et al. (2000); (b) *n*-Alkane concentrations in submerged plants displayed by Aichner et al. (2010a); (c) *n*-Alkane concentrations in different plant sources in Lake Qinghai catchment reported by Liu et al. (2015).

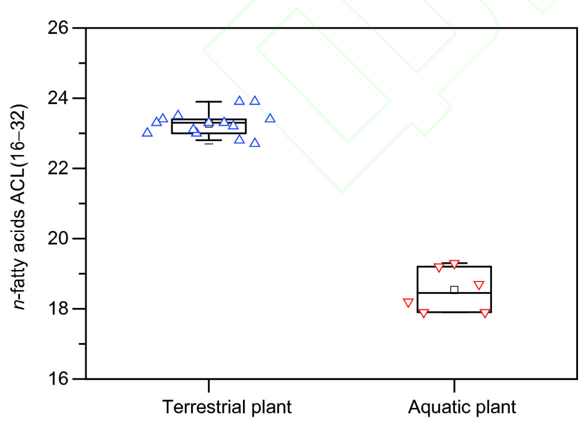

Figure 4 *n*-Alkanoic acids $ACL_{16\text{–}32}$ values in terrestrial and aquatic plants in Lake Qinghai catchment (Wang and Liu, 2012).

duced by aquatic plants, and long-chain free *n*-alkanoic acids are mainly derived from terrestrial sources.

In a recent report, Liu and Liu (2017) investigated the concentrations and distributions of *n*-alkanoic acids from 18 lakes on the northeastern Tibetan Plateau and found that the concentrations of C_{26}–C_{30} *n*-alkanoic acids in submerged plants (avg. 216 μg g^{-1} for *Potamogeton*, 52 μg g^{-1} for *Myriophyllum* and 134 μg g^{-1} for *Ruppia*) were close to those for terrestrial plants (avg. 161 μg g^{-1}), indicating that the contributions of submerged plants to lake sedimentary C_{26}–C_{30} pools should be considered, whereas the influence of algae on C_{26}–C_{30} *n*-alkanoic acids in lake sediments may be minor because of their low concentrations (avg. 7 μg g^{-1} for *Chara*, 8 μg g^{-1} for *Cladophora*, and 18 μg g^{-1} for *Spirogyra*).

Therefore, the molecular distributions of *n*-alkanes and *n*-alkanoic acids are important methods to distinguish the organic matter input sources in lake sediments, but the combination with the analysis of $\delta^{13}C$ or other proxies (e.g., P_{aq} and ACL) can provide a more accurate understanding of climatic changes when using *n*-alkane or *n*-alkanoic acids in lacustrine sediments in paleoenvironmental reconstructions.

4. Climate changes indicated by sedimentary total organic matter $\delta^{13}C$ in lakes in northwestern China

The carbon isotope of total organic matter from lake sediments can be used to indicate the variations in terrestrial vegetation and the regional climatic changes if the carbon source of sedimentary organic matter is mainly from terrestrial plants. In the past several decades, the carbon isotopes of lacustrine sediments have been frequently used to indicate paleoenvironmental changes and have been an important index of the lake environmental changes in northwest China (Liu et al., 2003; Zhang et al., 2004; Luo et al., 2008; Mischke et al., 2008; Sun et al., 2014, 2016). In general, the total organic carbon isotopes of sedimentary cores from lakes have been widely used to study C_3/C_4 plant changes, variable humidity conditions, the sources of organic matter, lake productivity and carbon burial translation in lakes (Liu et al., 2013, 201b6).

The carbon isotope analysis of total organic matter from lake sediments is simple, dependable and efficient, so the carbon isotopes of lacustrine sedimentary organic matter play a very important role in studying the biogeochemical cycle in lakes, including the sources of organic matter, lake productivity, carbon burial translation and the changes in integrated bulk biomass, although more attention has been paid to the geochemical significance of compound-specific carbon isotopes in lakes (Liu et al., 2013, 2016b). In the past several decades, the carbon isotopes of total organic matter have been widely used to reconstruct paleoenvironmental changes in lakes and are an important index of the lake environmental changes in northwest China (Liu et al., 2003; Zhang et al., 2004; Luo et al., 2008; Mischke et al., 2008; Sun et al., 2014, 2016).

Usually, a variation in $\delta^{13}C_{org}$ indicates terrestrial vegetation changes in the catchments of lakes if the source is from land; thus, $\delta^{13}C_{org}$ can be used to reconstruct varied C_3/C_4 plant histories and related climate/environment changes (Figure 5b). However, the environmental/climatic implication of $\delta^{13}C_{org}$ becomes more complicated if an aquatic plant such as algae is the main contributor of organic matter in lake sediments. In general, the carbon isotopic composition of organic matter mainly from aquatic plants is controlled by lake water $[HCO_3^-]$ and primary productivity; thus, the variation in $\delta^{13}C_{org}$ is explained as primary productivity changes (Figure 5a) or biogeochemical processes in lakes (Figure 5d). However, the TOC $\delta^{13}C_{org}$ values in lake sediments are usually interpreted in multiple ways, and there are still some debates and even contrary explanations of $\delta^{13}C_{org}$ in common lakes. For instance, the carbon isotopic composition of TOC was used as a proxy for precipitation or temperature based on the assumption that the organic carbon matter of lake sediments was mainly from the terrestrial C_3 plants in Lake Qinghai (Zhang et al., 2004; Xu et al., 2006). However, Shen et al. (2005) suggested that changes in $\delta^{13}C_{org}$ mainly indicated a variation in the primary productivity in Lake Qinghai and the relative contribution of terrestrial C_3 plants on the assumption that the total organic matter in sediments is from aquatic plants and terrestrial C_3 plants in the Lake Qinghai region.

In recent decades, many researchers have known the effect of carbon sources on the carbon isotopes of lake sediments, and the distribution of aquatic plants and their effect on carbon isotopes of lake sediments have been systematically investigated by some studies on the Qinghai-Tibet Plateau (Aichner et al., 2010c; Liu et al., 2013). The result of a recent study showed that the aquatic plants in the deep water areas (>10 m) were primarily dominated by *Cladophora* with low $\delta^{13}C_{org}$ values, but submerged plants such as *Potamogeton* with enriched $\delta^{13}C_{org}$ values are the dominant species in shallow water (<10 m) (Liu et al., 2013). As shown in Figure 6, the $\delta^{13}C_{org}$ values of surface sediments become lower with increased water depth in Lake Qinghai, but the $\delta^{13}C_{org}$ values of surface soil and terrestrial plants vary in a range from −27.7‰ to −24.5‰ in the catchment of Lake Qinghai (Liu et al., 2013). As shown, the carbon isotopic variation in surface sediments cannot be fully explained by the contribution of terrestrial sources in Lake Qinghai, but it can be reasonably explained by the contribution of aquatic plants in different water depths. In general, the $\delta^{13}C_{org}$ values of surface sediments vary between the $\delta^{13}C_{org}$ values of submerged plants and *Cladophora* in shallow water, and then the $\delta^{13}C_{org}$ values abruptly decrease caused by the much higher contribution of *Cladophora* with negative $\delta^{13}C_{org}$ when the water depth exceeds 10 m in Lake Qinghai (Figure 6). Thus, the isotopic results from Lake Qinghai indicate that the carbon isotopes of the organic material in the surface sediments are primarily controlled by the types of aquatic plants and that the TOC $\delta^{13}C_{org}$ can be used to indicate the variation in the water depth/lake level (Liu et al., 2013).

According to the above results of the modern process in Lake Qinghai, the paleoenvironmental significance of sedimentary $\delta^{13}C_{org}$ can be explained reasonably in the core during the Holocene. In the core, the enriched $\delta^{13}C_{org}$ is mainly from the contributions of submerged plants (Figure 5c), indicating a small area and shallow lake during the early Holocene, but not caused by C_4 expansion and the increased contribution of C_4 plants (Thomas et al., 2014). In addition, the presence of *Ruppia maritima* seeds with enriched $\delta^{13}C_{org}$ values from −14.6‰ to −9.7‰, which prefers living in shallow water, further proves that a large number of submerged plants flourished in Lake Qinghai during the early Holocene (Liu et al., 2013). Otherwise, the low $\delta^{13}C_{org}$ value in the core suggests that the size of Lake Qinghai increased and that the lake level was high during the late Holocene (Figure 5c).

Consequently, the environmental changes cannot be reasonably reconstructed using total organic carbon isotopes of sediments from lakes when the sedimentary $\delta^{13}C_{org}$ changes were simply explained as variations in the terrestrial plants

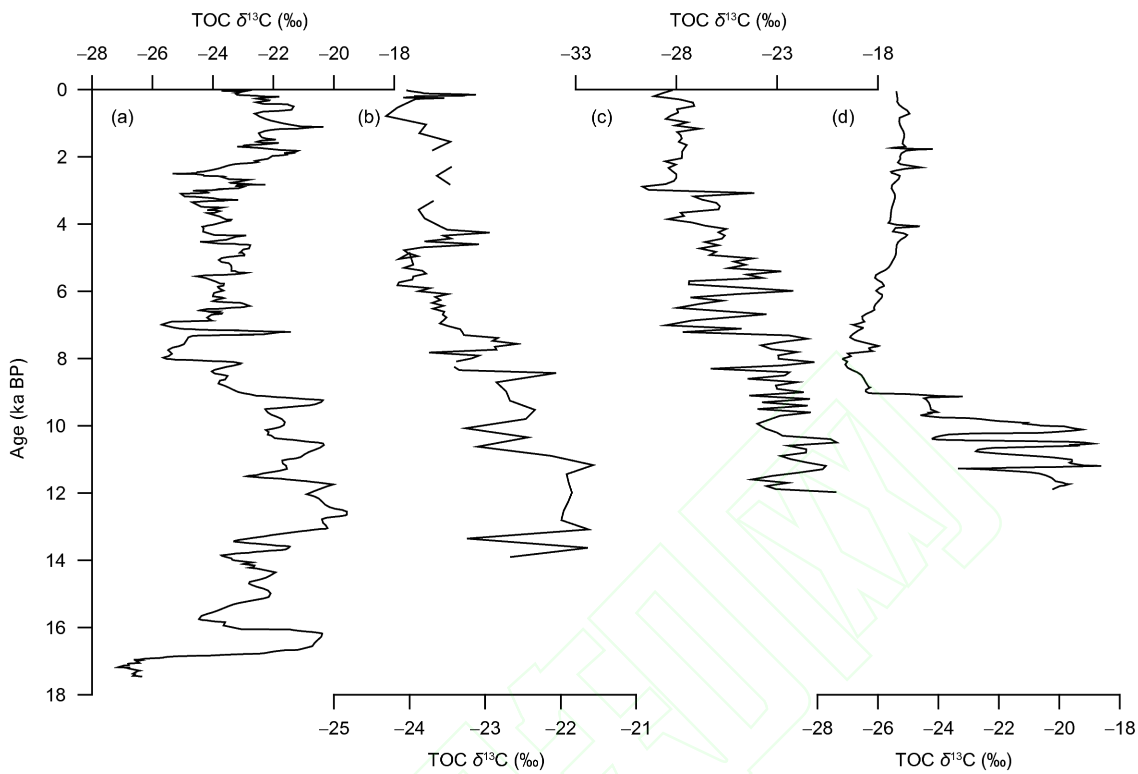

Figure 5 Environmental changes recorded by carbon isotopes of lacustrine sedimentary organic matter for the past 18 ka in lakes on the northwestern China. (a) Lake Koucha (Mischke et al., 2008); (b) Lake Balikun (Sun et al., 2014); (c) Lake Qinghai (Liu et al., 2013); (d) Lake Muge co (Sun et al., 2016).

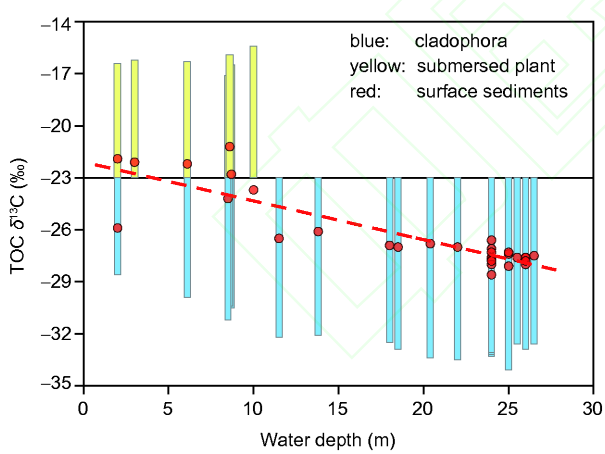

Figure 6 Variation in total organic carbon isotopes of surface sediments, submerged plants and *Cladophora* with water depth in modern Lake Qinghai (amended from Liu et al., 2013).

and related climatic/environmental changes but not considered the contribution of aquatic plants to sedimentary organic matters in the past studies. Currently, the results from an increasing amount of studies suggest that the contribution of aquatic plants to sedimentary organic matter is very common in lakes in west-arid China, so we should fully consider the contribution of aquatic plants when we study climatic/environmental changes using sedimentary $\delta^{13}C_{org}$ in these lakes. In other words, the sedimentary $\delta^{13}C_{org}$ can be considered as an ideal proxy of environmental changes when the organic carbon source of sediments is known in the lakes.

5. Stable carbon isotopes of organic molecular compounds in lakes and implications for paleoclimatic changes in Northwestern China

As has been mentioned above, the total organic matter in lakes may have multiple sources, and consequently, there are many factors affecting the $\delta^{13}C$ value of total organic matter. On the other hand, some large molecular compounds are source specific and relatively resistant to degradation over geological time, and consequently, the separate analysis of the $\delta^{13}C$ values of lipid biomarkers specifically from terrestrial sources and lacustrine sources would allow a better reconstruction of past ecological changes on land and in lakes. Currently, the compound-specific $\delta^{13}C$ values of lacustrine long-chain *n*-alkanes and fatty acids, which originate mainly from the cuticle surface of terrestrial higher plants, have been widely used in paleolimnological studies, of which the most striking achievement is the reconstruction of past C_4/C_3 vegetation shifts over geological time (Street-Perrott et al., 1997; Huang et al., 2001; Sinninghe Damsté et al., 2011; Sun et al., 2013; Chu et al., 2014; Thomas et al., 2014; Jia et al., 2015). On the other hand, sedimentary short-chain *n*-alkanes and some other biomarkers for algae are generally considered to be produced within the lake, and their $\delta^{13}C$ values can be used to trace changes in the lake primary productivity and carbon sources of aquatic organisms (Ostrom et al., 1998; Huang et al., 1999; Filley et al., 2001; Castañeda et al., 2009; Aichner et al., 2010a).

In the last several decades, our knowledge on the modern process of biomarker $\delta^{13}C$ has grown for lakes in northwestern China. After investigating the $\delta^{13}C$ values of total organic matter and *n*-alkanes in a set of surface sediment samples and corresponding macrophytes in 40 lakes on the Qinghai-Tibetan Plateau, Aichner et al. (2010b) found that submerged plants can significantly affect the distribution and $\delta^{13}C$ values of *n*-alkanes, while the contribution from submerged plants can be up to 60% (mean 40%) to TOC and up to 100% (mean 66%) to mid-chain *n*-alkanes (C_{23} and C_{25}). Consequently, in regions where C_4 vegetation is absent, the relative contribution of submerged plants to sediments can be assessed quantitatively from sedimentary $\delta^{13}C$ values (Aichner et al., 2010b). By studying the $\delta^{13}C$ values of GDGTs in the Lake Qinghai region, Lu et al. (2013) found that the $\delta^{13}C$ values of GDGT-derived biphytanes are different in lake sediments and their surrounding soils; therefore, the $\delta^{13}C$ values of BP-0 can be used as indicators for distinguishing the depositional environments between terrestrial and lacustrine systems. Additionally, this study also supports that the $\delta^{13}C$ values of BP-cren may record the DIC $\delta^{13}C$ values well in lacustrine systems (Lu et al., 2013).

Compound-specific $\delta^{13}C$ values of lipid biomarkers have been applied to reconstruct the paleoenvironmental changes in lakes in northwestern China. Aichner et al. (2010a) investigated organic biomarkers in a 4.23 m-long sediment core from Lake Koucha (eastern Qinghai-Tibetan Plateau) and found that the $\delta^{13}C$ values of aquatic plant-derived *n*-C_{23}, which ranged from −23.5% to −12.6%, are an indicator of carbon-limiting conditions in this lake since 16 ka. Since the $\delta^{13}C$ value of the inorganic carbon source may vary, Aichner et al. (2010a) further suggested that the offset between the $\delta^{13}C$ values of *n*-C_{23} and TIC could serve as a more precise proxy for carbon-limiting conditions or paleo-productivity in lacustrine environments. Aichner et al. (2015) also reported the $\delta^{13}C$ values of *n*-acids from a late Holocene sediment core from Lake Karakuli, Xinjiang Province, China. The results showed that the $\delta^{13}C$ value of *n*-C_{28} acid has been stable since 4.2 ka, indicating that the regional vegetation is dominated by C_3 plants in eastern Pamir. Moreover, Thomas et al. (2014) reconstructed a 32-kyr history of the relative abundance of C_4 plants on the northeastern Qinghai-Tibetan Plateau based on the $\delta^{13}C$ values of *n*-C_{28} acids in Lake Qinghai sediments. They found an expansion of C_4 plants (accounting for 50% of terrestrial primary productivity) on the Qinghai-Tibetan Plateau during the Late-glacial and early Holocene (Figure 7a). This might be an important finding that reveals ancient ecosystem and environmental changes on the Qinghai-Tibetan Plateau, and it may also provide further insight into the mechanism of global C_4 plant evolution.

In recent years, however, people began to notice that there are some uncertainties in the interpretation of the compound-specific $\delta^{13}C$ values of lipid biomarkers, which have been widely used to reconstruct paleoenvironmental conditions in lake and terrestrial systems (Castañeda and Schouten, 2011). Hence, it is quite necessary to further investigate the $\delta^{13}C$ signature of biomarkers from different sources in order to explore more valid proxies based on the $\delta^{13}C$ values of lipid biomarkers. Recently, Liu et al. (2015) systematically analyzed the distribution and $\delta^{13}C$ values of long-chain *n*-alkanes derived from terrigenous sources (including surrounding plants and nearby surface soils), aquatic plants at various water depths, and surface lake sediments from nearshore and offshore settings in Lake Qinghai and small lakes nearby. They found that aquatic plants, particularly the C_4-like submerged plants, may contribute a certain amount of long-chain *n*-alkanes (C_{27}, C_{29} and C_{31}) to lake sediments, although these long-chain *n*-alkanes are generally considered to originate from terrestrial higher plants. Since individual long-chain *n*-alkanes from a single plant have similar $\delta^{13}C$ values, whereas the $\delta^{13}C$ values of long-chain *n*-alkanes from different sources might be quite different, systematic deviations of the $\delta^{13}C$ values among long-chain *n*-alkanes (C_{27}, C_{29} and C_{31}) in a sediment sample likely suggest source heterogeneity. That is, sedimentary C_{27} *n*-alkane might be more enriched in ^{13}C than C_{31} *n*-alkane due to the contribution from C_4-like submerged plants with more positive *n*-alkane $\delta^{13}C$ values. Hence, the $\delta^{13}C$ values of long-chain *n*-alkanes in Lake Qinghai sediments may have recorded $\delta^{13}C$ signatures for both terrestrial plants and aquatic plants. The authors noted that while the $\delta^{13}C$ values of C_{31} *n*-alkanes might have relatively faithfully recorded the stable carbon isotope composition of terrestrial vegetation, the contribution from aquatic plants increases as the chain length decreases.

According to the abovementioned modern results in the Lake Qinghai region, Liu et al. (2015) further studied compound-specific $\delta^{13}C$ values of long-chain *n*-alkanes in a Holocene sediment core retrieved from Lake Qinghai. Overall, there are significant changes in the long-chain *n*-alkane $\delta^{13}C$ records, particularly in the early Holocene, where the $\delta^{13}C$ values of C_{27} *n*-alkanes were more positive, which fell within the range of *n*-alkane $\delta^{13}C$ values for terrestrial C_4 plants (Figure 7b). This is in agreement with the $\delta^{13}C$ record of C_{28} *n*-fatty acids reported by Thomas et al. (2014) (Figure 7a). However, according to the relatively constant $\delta^{13}C$ values of C_{31} *n*-alkanes (−31.4±0.5‰), the regional terrestrial vegetation of Lake Qinghai might be dominated by C_3 plants, and there is no evidence for the expansion of C_4 plants (Figure 7b). Thus, the compound-

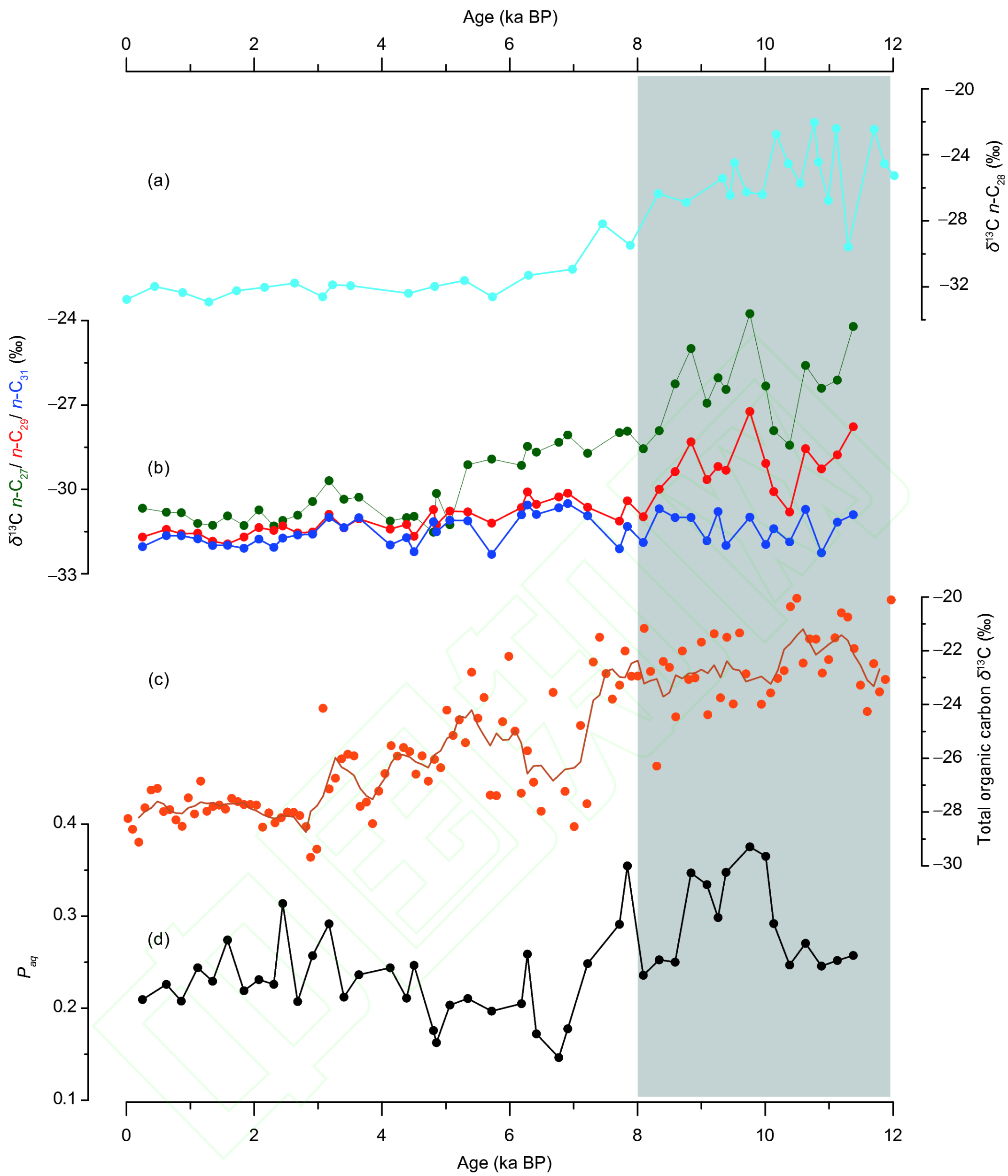

Figure 7 Holocene $\delta^{13}C$ and P_{aq} records in Lake Qinghai sediments. (a) Compound-specific $\delta^{13}C$ of C_{28} *n*-fatty acid (Thomas et al., 2014). (b) Compound-specific $\delta^{13}C$ of long-chain *n*-alkanes (C_{27} in green, C_{29} in red and C_{31} in blue) in core 1F (Liu et al., 2015). (c) The $\delta^{13}C$ of total organic carbon in core 1F (Liu et al., 2013). (d) The variation in P_{aq} index representing relative input from aquatic plants (Liu et al., 2015).

specific $\delta^{13}C$ values of some sedimentary long-chain *n*-alkanes might have been affected by both terrestrial plants and aquatic plants and cannot be simply explained by terrestrial C_4 vs. C_3 vegetation changes (Liu et al., 2015).

During the early Holocene, the relatively positive $\delta^{13}C$ values of C_{27} and C_{31} *n*-alkanes is in agreement with the relatively positive $\delta^{13}C$ values of total organic carbon compared with those during the mid- and late Holocene (Figure 7c), possibly due to the contribution of *n*-alkanes and total organic matter from aquatic plants (especially submerged plants). Moreover, the abundant seeds of *Ruppia* (a submerged flowering plant) (Liu et al., 2013) and the high P_{aq} (indicating a substantial input from submerged plant) (Figure 7d) further suggest that the positive $\delta^{13}C$ values in the early Holocene sediment are caused by the enhanced contribution from submerged plants, in accord with the low lake level for Lake Qinghai at that time (Liu et al., 2013). Therefore, it is quite necessary to carefully examine the sources and $\delta^{13}C$ values of sedimentary long-chain alkanes in paleolimnological studies. Otherwise, past terrestrial C_4/C_3 ratios might be overestimated/underestimated, potentially hindering our understanding of the important global ecological issues, i.e., the terrestrial C_4 vs. C_3 vegetation change and its corresponding environmental/climatic mechanisms.

6. The relationship between the carbon isotope of lake sediments and variations in the global C_3/C_4 vegetation

Currently, the studies on global change are mainly focused on reconstructing paleoenvironmental changes using different proxies from different geological objects such as marine sediments, stalagmites, ice cores, tree rings, loess deposits, and lake sediments. Few of these geological objects can be used to reconstruct the ecological history on land, especially for tracing variations in terrestrial C_3 and C_4 vegetation. In general, the changing history of C_3/C_4 vegetation was commonly reconstructed through lacustrine records, in addition to some researchers who studied terrestrial C_3/C_4 vegetation changes using the compound-specific carbon stable isotopic composition of a marine sedimentary core from a marginal sea. Thus, the lake is favored by most researchers for studying the C_3/C_4 vegetation history on land. Over the past several decades, most of the publications on understanding the varied history of global C_3/C_4 vegetation are mainly based on leaf-wax compound-specific carbon isotopic records in lakes, except for some loess records. Next, we re-evaluate the environmental significance of sedimentary carbon isotopes in the lakes of Africa and North America (Street-Perrott et al., 1997; Huang et al., 2001) based on the knowledge from the geochemical studies of the lakes in arid northwest China.

Lake Sacred in Kenya Mount (0°03′N, 37°32′E), a crater lake, is located in a humid mountain rainforest at an altitude of 2350 m. The lake is very oligotrophic, small (0.51 km^2) and shallow (≤5 m). The lake was surrounded by a floating mat of emergent macrophytes. In a past study, the *n*-alkanes C_{27}–C_{31} in the drilling core from Sacred Lake were suggested to be from leaf waxes of terrestrial plants; their $\delta^{13}C$ values were approximately −23‰ during the glacial period, and these values varied from −37‰ to −30‰ during the Holocene (Street-Perrott et al., 1997). Molecular-level isotopic data confirm that the large carbon isotope variations in total organic carbon reflect both terrestrial and aquatic signals. The $\delta^{13}C$ values of leaf wax components of higher plants display an average glacial-to-interglacial difference exceeding 15‰. Enriched values for *n*-alkyl lipids (weighted mean: −23‰ to −17‰) were present in sediments before 34000 and from 24000 to 13000 ^{14}C years BP, indicating that the total organic matter in sediments was mainly contributed by C_4 plants in Lake Sacred during the early- and full-glacial times (Figure 8). In the intervening interstadial, the contribution of C_3 plants increased (mean $\delta^{13}C$ values varied from −28‰ to −26‰). At the end of the glacial, the leaf wax $\delta^{13}C$ values sharply decreased, except for a minor positive excursion in the Younger Dryas. The leaf wax $\delta^{13}C$ values reached their lowest values (−36‰ to −28‰) at the beginning of the interglacial, ~10300 ^{14}C years BP. This result suggests a

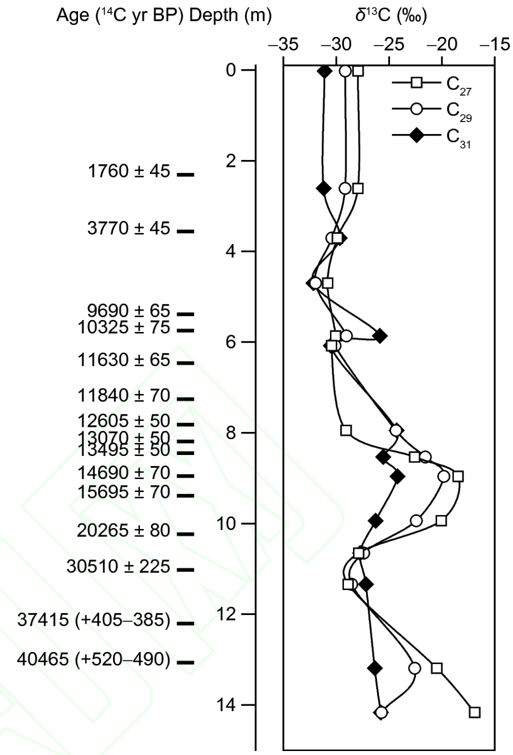

Figure 8 Carbon-isotope values of leaf wax components(C_{27}, C_{29} and C_{31}) from Lake Sacred (amended from Street-Perrott et al., 1997).

dominantly C_3 plant source, compatible with the re-establishment of dense moist montane forest. After 4000 years BP, the relative contribution of C_4 plants significantly increased in this region.

In addition, Lake Kimilili of Elgin Mountain (1°06′N, 34°34′E) is located in a glacial basin that is 4150 m above sea level and surrounded by sparse Afroalpine vegetation. The lake has an area of approximately 5000 m^2 and is shallow. Although the Lake Limilili sedimentary sequence did not extend back to the LGM, the leaf wax $\delta^{13}C$ values in both cores averaged between −20‰ and −17‰ during the Younger Dryas period (11000–10000 ^{14}C years BP), suggesting the larger contribution of C_4 plants relative to present. During the Holocene, the weighted mean leaf wax $\delta^{13}C$ values decreased to a modern level of −27‰ to −24 ‰, consistent with the observed expansion of C_3 shrubs.

In summary, the carbon isotope values of leaf wax components and TOC in the two cores from Lake Sacred and Lake Kimilili both indicated the changing history and controlling factors of terrestrial C_3/C_4 plants in this region since the last glacial stage. The important conclusion of this study is that the C_4 vegetation expansion was evident in this region when atmospheric pCO_2 decreased globally at the LGM. In other words, the results of this study prove that the low global atmospheric pCO_2 is one of the key factors for the global C_4 expansion. This study provides an understanding of the mechanism of the evolution of terrestrial C_3/C_4 vegetation.

Huang et al. (2001) studied the C_3/C_4 vegetation changes since the LGM based on the analysis TOC and *n*-alkane (C_{27}, C_{29} and C_{31}) $\delta^{13}C$ values in cores from Lake Alta Babicora and Lake Quexil in Central America. These two lakes have different hydrological conditions. The average annual precipitation of Lake Alta Babicora (29°N, 108°W) in Mexico is 500 mm (main occurrence times are in July and August) with an annual average temperature of 20°C, and the surrounding modern vegetation is dominated by C_4 plants. Conversely, the annual mean precipitation and annual average temperature of Lake Quexil are 1600 mm (frequently occurs in June to December) and 25°C, respectively, and the lake is mainly surrounded by evergreen forest vegetation.

The varied trends of TOC and *n*-alkane (C_{27}, C_{29} and C_{31}) $\delta^{13}C$ values in the sedimentary core from Lake Alta Babicora in Mexico are almost common (Figure 9a). The TOC $\delta^{13}C_{org}$ values varied from −24‰ to −22‰, and the *n*-alkane (C_{27}, C_{29} and C_{31}) $\delta^{13}C$ values ranged from −30‰ to −26‰ from 20700 to 12700 ^{14}C years BP, indicating that the terrestrial plants were dominated by C_3 plants in this region at that time. In the early Holocene, the sharply enriched $\delta^{13}C$ values of C_{27} and C_{29} from −30‰ to −24‰ indicate an evident C_4 expansion during this period, and the enriched *n*-alkane $\delta^{13}C$ values also suggest that C_4 plants were the dominant species at this region throughout the Holocene.

The varied $\delta^{13}C$ trends of TOC and *n*-alkanes (C_{27}, C_{29} and C_{31}) in sedimentary core from Lake Quexil in Guatemala are different (Figure 9b), suggesting that their $\delta^{13}C_{org}$ values were influenced by both aquatic and terrestrial plants. Therefore, only the $\delta^{13}C$ values of individual leaf wax *n*-alkanes (C_{27}, C_{29} and C_{31}) are used to investigate changes in the proportion of C_3 and C_4 plants. In this study, the leaf wax $\delta^{13}C$ values in the core reached a maximum (from −28‰ to −27‰) around

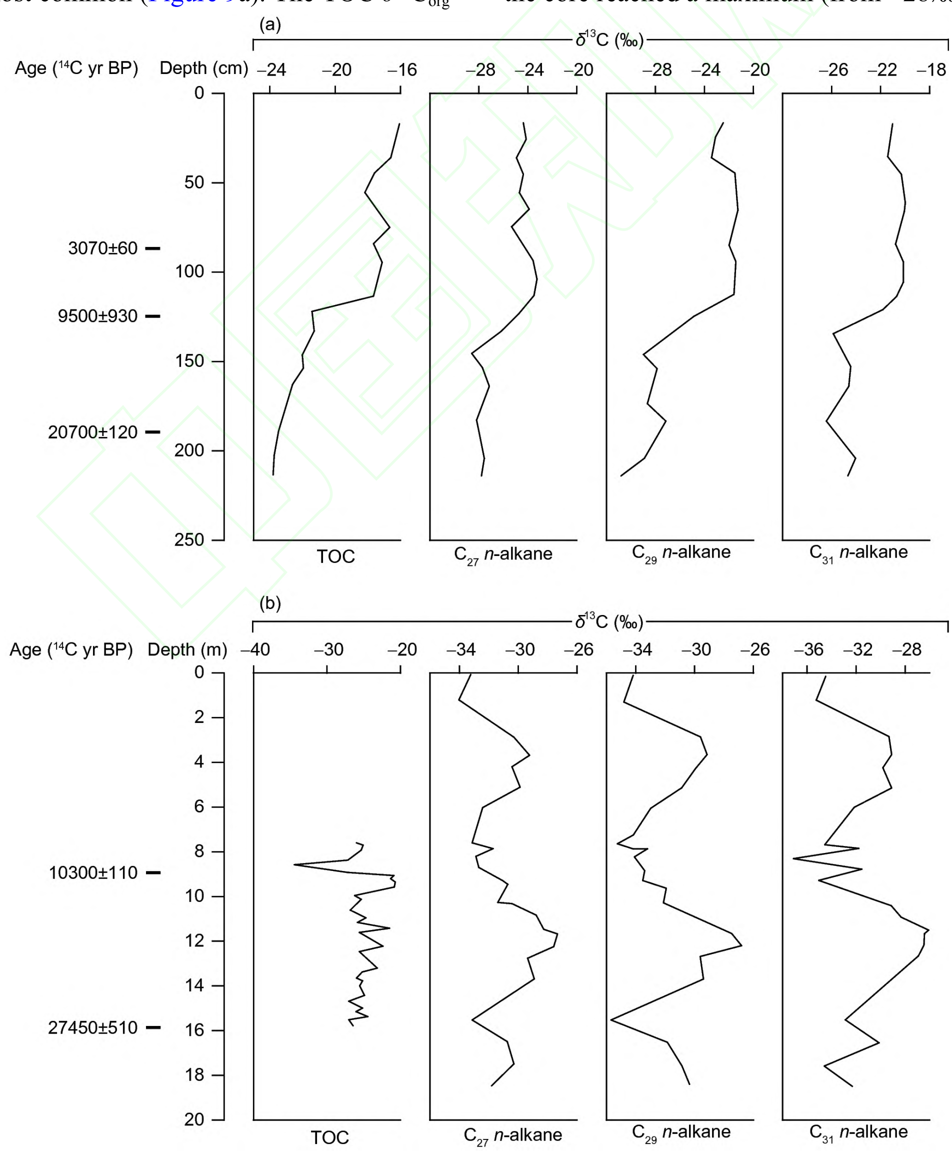

Figure 9 The carbon isotopes of *n*-alkanes (C_{27}, C_{29} and C_{31}) from leaf wax in (a) Lake Alta Babicora and (b) Lake Quexil (amended from Huang et al., 2001).

the LGM (18000 ^{14}C yr BP), implying the expansion of C_4 plants. The calculated relative contribution from C_4 plants at the LGM was almost 40% based on a binary model with endpoints of −19‰ for C_4 plant wax and −34‰ for C_3 plant wax. During the early Holocene (10000 to 8000 ^{14}C yr BP), a 6‰ to 8‰ decrease in $\delta^{13}C$ values reflects a shift to an overwhelming predominance of C_3 plants in the Lake Quexil region. However, leaf wax *n*-alkane $\delta^{13}C$ values increased by up to 5‰ in the mid- and late Holocene, possibly caused by a decrease in trees and an increase in herbs.

In addition, the relationship between the expansion of C_3 or C_4 vegetation and atmospheric pCO_2 was evaluated in this study. The results show that the relative abundance of C_3 or C_4 vegetation is mainly controlled by multiple factors, including precipitation, seasonal precipitation, temperature and atmospheric CO_2 concentration. More importantly, the authors suggested that low pCO_2 alone is insufficient to trigger the expansion of C_4 plants in the absence of favorable climatic conditions.

However, the conclusions from the above studies are questionable because the possible contribution of aquatic plants to the leaf wax *n*-alkanes in sediments is not considered in these studies. As shown in Figure 8, the $\delta^{13}C$ values of individual leaf wax *n*-alkanes (C_{27}, C_{29} and C_{31}) in Sacred Lake have very similar change characteristics to those in Lake Qinghai. In general, the $\delta^{13}C$ values of individual leaf wax *n*-alkanes (C_{27} and C_{29}) varied on a large scale, but the $\delta^{13}C$ values of individual leaf wax *n*-alkane C_{31} had few changes (Figure 8), which suggests that the large $\delta^{13}C$ changes in the individual leaf wax *n*-alkanes (C_{27} and C_{29}) may be caused by contributions from aquatic plants. Consequently, the findings from the lakes on the northeastern Qinghai-Tibet Plateau can be used for other lakes in the world, and the aquatic plant contribution should be clear and definite when we use leaf wax *n*-alkane $\delta^{13}C$ values from lakes to reconstruct the history of terrestrial C_3/C_4 changes.

7. Summary

Isotopic geochemical tracing for the source(s) of organic matter is one of the major applications of carbon isotope geochemistry in limnological studies. Due to the complex source(s) of lake organic matter, it is necessary to analyze both the distribution of organic molecular compounds and their compound-specific $\delta^{13}C$ characteristics to resolve this issue. The source(s) of lake organic matter can be roughly determined by the distribution of biomarkers and their individual $\delta^{13}C$ values from lake sediments. At the same time, only after constraining the source(s) of lake organic matter, the paleoclimatic implication of lacustrine $\delta^{13}C$ can be reasonably explained.

It is essential to combine sedimentary TOC with lipid biomarkers to trace historical changes in vegetation. The $\delta^{13}C$ values of TOC in sediments can be easily prepared and analyzed, allowing the generation of high-resolution records. On the other hand, as the source(s) of organic matter in lacustrine sediments is complicated, it is necessary to indicate the characteristics of the organic matter source(s) based on the analysis of biomarker distribution and the compound-specific $\delta^{13}C$ values. However, it should be noted that the analysis of compound-specific $\delta^{13}C$ is costly and time consuming. Therefore, the comprehensive analysis of TOC $\delta^{13}C$ values and compound-specific $\delta^{13}C$ values of lipid biomarkers can help to obtain more effective research results.

It should be pointed out that different lakes have different climates and sedimentary environments, so the sources of organic matter and biomarkers may have some differences. Therefore, we suggest that the specific problems should be concretely analyzed when applying lacustrine $\delta^{13}C$ analysis in paleoclimate studies; that is, we should first systematically study the modern process of the study lake or at least have a certain understanding of the regional modern process and identify the sources of organic matter and biomarkers.

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