RESEARCH ARTICLE

Recent environmental changes in the Yunnan–Guizhou Plateau inferred from organic geochemical records from the sediments of Fuxian Lake

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During the past century, many lacustrine environments have changed substantially at the ecosystem level as a result of anthropogenic activities. In this study, the distributions of n-alkane homologues, carbon isotopes ($\delta^{13}C_{org}$), organic carbon, and the C/N atomic ratio in two sediment cores from Fuxian Lake (Yunnan, southwest China) are used to elucidate the anthropogenic impacts on this deep, oligotrophic, freshwater lake. The carbon preference index (CPI) of long-chain components, average chain length (ACL), proportion of aquatic macrophytes (Paq), and terrigenous/aquatic ratios (TAR) show different temporal patterns that reflect variations in biological production. Notably, the n-alkane homologues are shown to be more sensitive to environmental changes than $\delta^{13}C_{org}$ and the C/N ratio. Prior to the 1950s, minor variations in the sedimentary geochemical record were likely caused by climate changes, and they represent a natural stage of lake evolution. The onset of cultural eutrophication in Fuxian Lake occurred in the 1950s, when the n-alkane proxies collectively exhibited high-amplitude fluctuations but overall decreasing trends that coincided with population growth and related increases in land-use pressure. In the 21st century, Fuxian Lake has become even more eutrophic in response to human activities, as indicated by sharp increases in C/N ratio, Paq, $\delta^{13}C_{org}$, ACL, CPI, and TAR. Our findings provide robust molecular sedimentary evidence confirming that the environmental evolution of lakes in the Yunnan–Guizhou Plateau over the past century was closely associated with enhanced anthropogenic activities.

Keywords: Lake sediments, Environmental change, Organic carbon, Biomarker, Fuxian Lake

1. Introduction

Lacustrine sediments are valuable archives of paleoclimatic and related paleoenvironmental changes, as well as human impacts. Organic carbon (OC) burial in lacustrine sediments is mainly characterized by rapid accumulation (Dean and Gorham, 1998; Cole et al., 2007; Mendonça et al., 2017) and a high preservation factor that on average is approximately 50 times greater than that in the ocean (Einsele, 2001). Hence, the short-term processes that affect OC delivery and burial are amplified in sedimentary records from lakes. The stable isotopes of carbon and oxygen, inorganic geochemistry, and palynology can be used to reconstruct natural and human-induced changes in local and regional ecosystems. In addition to these paleolimnological proxies, biological marker (biomarker) molecules are valuable (Meyers, 2003; Sikes et al., 2009; Ortiz et al., 2011; Silva et al., 2012; Wang and Liu, 2012; Zhang et al., 2015; Wang et al., 2019) because they can characterize specific biotic sources and they retain source information after burial in sediments, even after minor alteration (Blumer et al., 1971; Meyers, 2003; Fokin et al., 2012).

Fuxian Lake is located in the Yunnan–Guizhou Plateau, in a region strongly influenced by the Indian summer monsoon (ISM; **Figure 1**). It is the second-deepest plateau oligotrophic freshwater lake in China. Fuxian Lake is a semi-closed lake with a small catchment. Notably, the region has experienced a series of major socioeconomic transformations during the 20th century, including World War Two, the Chinese Civil War, the foundation of the People's Republic of China in 1949, the Great Proletarian Cultural Revolution during the 1960s and

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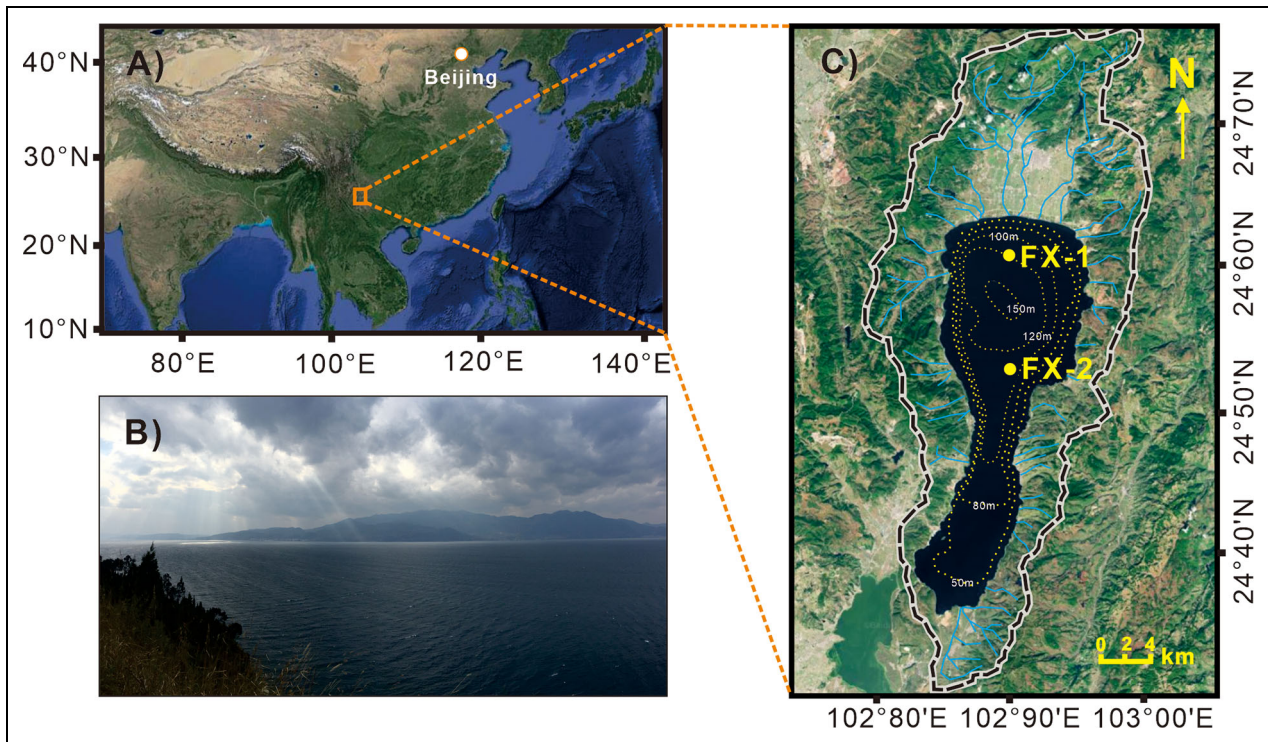Emails: hehaibo@ynu.edu.cn; liuzaihua@vip.gyig.ac.cn

Figure 1. Study site: (A) Location of the Fuxian Lake catchment. (B) Locations of cores FX-1 and FX-2 in the northern and central sectors of the lake, respectively. The basin boundary (black dashed line) and lake depth contours (dotted lines) are from Liu et al. (2008). (C) Photograph of Fuxian Lake. DOI: https://doi.org/10.1525/elementa.2021.00068.f1

1970s, and economic liberalization after the 1980s, all of which influenced the catchment and ecology of the lake. Currently, a large number of tourists visit the area each year and they have a substantial impact on the lake ecosystem. Although the lake is classified as oligotrophic, it has become increasingly biologically productive (Zhang et al., 2015) as a consequence of recent human activities. During the past few decades, numerous paleoenvironmental studies have been conducted at Fuxian Lake, focused mainly on sediment accumulation, trophic status, trace metal pollution, aquatic organisms, and aquaculture (Liu et al., 2009; Zeng and Wu, 2009; Liu et al., 2014; Zhang et al., 2015; Wang et al., 2018; He et al., 2019, 2020). However, there have been few detailed sediment-based studies of climate change and anthropogenic impacts at the site.

Here we report the results of a study of two sediment cores from Fuxian Lake that span the last approximately 100 years. They were sampled at a high temporal resolution (1-cm intervals corresponding to approximately 5 yr per interval). The parameters measured include (1) C and N content of the sedimentary organic matter (OM), (2) stable carbon isotopes ($\delta^{13}C$), and (3) biomarkers (mainly n-alkanes compound). The results provide new insights into local and regional paleoclimatic and paleoenvironmental changes during the past century, and on human impacts.

2. Site description
Fuxian Lake (24°17′–24°37′ N, 102°49′–102°57′ E) is located at 1,723 m above sea level on a plateau in the

central Yunnan Province in southwest China (**Figure 1**). The catchment area is 675 km^2, the lake surface area is 212 km^2, and the maximum water depth is 158 m. The lake basin is wide and deep in the north but narrow and comparatively shallow in the south. The Liangwang River in the north is the main inflow, although there are also numerous small streams and springs (e.g., Luchong spring). The Haikou River is the single outlet. The lake water has a long residence time (167 years; Wang and Dou, 1998), and a thermocline generally develops between March and December in this dimictic lake (He et al., 2019). The modern climatic conditions in the region are characterized by seasonality and they are dominated by the ISM. Typical average summer and winter temperatures are 20.2 °C and 9.2 °C, respectively, and the average annual precipitation is 951 mm, with 83% of the total occurring between May and October.

3. Materials and methods
Two sediment cores from the northern (FX-1) and central (FX-2) sectors of the lake were collected in January 2017 using a gravity corer fitted with 58-mm internal diameter Perspex tubes (**Figure 1**). The water depths at the two coring sites were 115 m (FX-1) and 120 m (FX-2). The cores were approximately 20-cm long and were sectioned into 1-cm intervals and then immediately transported to the laboratory in precleaned polyethylene bags where they were freeze-dried. Here we focus on the organic geochemical record of the past century.

Table 1. Summary of the indices and their calculations used in this study to describe the changes in the alkane composition of the sediments of Fuxian Lake. DOI: https://doi.org/10.1525/elementa.2021.00068.t1

Index	Calculation	Reference
ACL	$[(25(C_{25}) + 27(C_{27}) + 29(C_{29}) + 31(C_{31}) + 33(C_{33})]/$ $(C_{25} + C_{27} + C_{29} + C_{31} + C_{33})$	Poynter and Eglinton (1990)
CPI	$1/2[(C_{25} + C_{27} + C_{29} + C_{31} + C_{33})/(C_{24} + C_{26} + C_{28} + C_{30} + C_{32})$ $+ (C_{25} + C_{27} + C_{29} + C_{31} + C_{33})/(C_{26} + C_{28} + C_{30} + C_{32} + C_{34})]$	Bray and Evans (1961); Shen et al. (2019)
TAR	$(C_{27} + C_{29} + C_{31})/(C_{15} + C_{17} + C_{19})$	Meyers (2003)
Paq	$(C_{23} + C_{25})/(C_{23} + C_{25} + C_{29} + C_{31})$	Ficken et al. (2000)

ACL = average chain length; CPI = carbon preference index; TAR = terrigenous/aquatic ratios; Paq = proportion of aquatic macrophytes.

3.1. Sediment chronology

The freeze-dried and homogenized core sediments were sliced into 1-cm intervals for radiometric dating. The activities of ^{210}Pb and ^{226}Ra were determined using a large-volume coaxial reversed electrode high-purity germanium detector (Canberra, >60% relative efficiency) following the methods of Kirchner and Ehlers (1998). Weighed sediment samples (10 g each) were sealed in capped plastic test tubes for 3 weeks to ensure their decay to radioactive equilibrium. The activity of excess ^{210}Pb (^{210}Pb$_{ex}$) was obtained by subtracting the activity of ^{226}Ra from the total ^{210}Pb (Appleby, 2008). Using the constant rate of supply (CRS) model, ^{210}Pb$_{ex}$ was used to date the sediment cores according to Equation (1) (Appleby, 2008; Sanchez-Cabeza and Ruiz-Fernández, 2012):

$$t = \frac{1}{\lambda}\ln\left(\frac{A_n}{A_x}\right) \qquad (1)$$

where t is the age in years, λ is the decay constant of ^{210}Pb (0.03114 yr^{-1}), A_n is the content of ^{210}Pb$_{ex}$ at each sediment depth, and A_x is the inventory of ^{210}Pb$_{ex}$ below depth x in the sediment core.

3.2. Carbon and nitrogen concentrations and C/N ratios

Carbonates were removed by leaving the samples overnight in 50 ml of 1.5 mol/L HCl. The contents of total organic carbon (TOC) and total nitrogen (TN) content were determined using an elemental analyzer (Elementar-vario MACRO cube), which was calibrated using sediment standard materials (B2150 and AR2026) with an analytical precision of >0.2%. The TOC/TN (atomic) ratio was then calculated.

3.3. Measurement of stable carbon isotope ratios

Stable carbon isotope analysis (^{13}C/^{12}C) of the OC component of the samples was performed using a MAT-252/253 mass spectrometer. The δ^{13}C values are reported relative to the international Vienna Peedee Belemnite standard. The analytical precision was based on replicate measurements of an internal laboratory standard (Sun et al., 2011) and was typically better than $\pm 0.03\%$. The δ^{13}C value was calculated using Equation (2):

$$\delta^{13}C_{sample} = (R_{sample}/R_{standard} - 1) \times 1000 \qquad (2)$$

where R is the ^{13}C/^{12}C ratio of the sample or standard.

3.4. n-alkanes extraction and analysis

Sediment samples for lipid determination were first Soxhlet extracted (3-g samples) using a dichloromethane–methanol mixture (93:7 v: v) to obtain the soluble fraction. The samples were re-extracted with n-hexane and the combined organic phase was left over anhydrous Na$_2$SO$_4$ until the following day to remove any water. A portion of the extract was saponified (70 °C for 2 hr) using a KOH–MeOH solution mixture (3 ml). Then, n-alkanes were extracted into n-hexane from the saponified samples and concentrated to 1 ml using rote-evaporation (Waterson and Canuel, 2008). The n-alkanes were quantified by measuring the concentrated eluents via gas chromatography (GC) (Agilent 7890A). The GC conditions for n-alkanes were as follows: the temperature was raised from 70 °C (held for 1 min) to 140 °C at 10 °C/min and then at 3 °C/min to 310°C (held for 15 min; Mortillaro et al., 2011).

Changes in alkane composition of the sediment core samples were evaluated along with the following n-alkane indices: carbon preference index (CPI) of the long-chain components, average chain length (ACL), proportion of aquatic macrophytes (Paq), and terrigenous/aquatic ratios (TAR), which was calculated as the ratio between summed peak areas of certain alkane groups (**Table 1**).

4. Results

4.1. ^{210}Pb dating

Depth profiles of the activity of ^{210}Pb$_{ex}$ in cores FX-1 and FX-2 exhibit an approximately exponential decrease with depth when plotted against the cumulative mass (**Figure 2**). The values range from 32.78 $\pm$ 12.99 Bq kg^{-1} to 397.62 $\pm$ 40.08 Bq kg^{-1} (mean of 106.39 $\pm$ 19.11 Bq kg^{-1}). This exponential decrease is similar to the profiles observed in previous studies (Liu et al., 2009; Li et al., 2011; Liu et al., 2013; Wang et al., 2018), which indicates that the ^{210}Pb$_{ex}$ CRS model (Appleby, 2008) is appropriate for dating the recent sediments of Fuxian Lake. The period covered by each sediment core, as calculated by the ^{210}Pb$_{ex}$ CRS model, exceeds 100 years. The geochronology from 1910 to 2017 is shown in **Figure 2**.

4.2. Carbon and nitrogen content

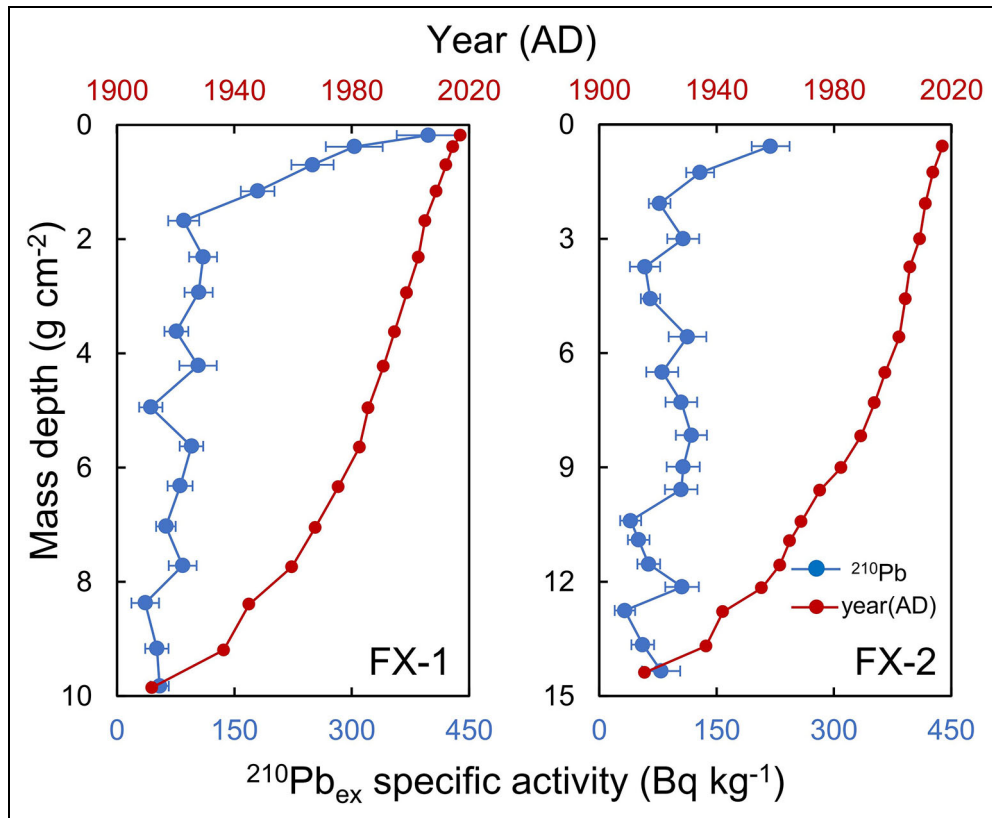Both cores exhibit exponential increases in TOC with decreasing depth. As shown in **Table 2**, the TOC content

Figure 2. Depth profiles of $^{210}Pb_{ex}$ for cores FX-1 and FX-2 Fuxian Lake. The geochronology of the sediment cores is based on the stratigraphies of $^{210}Pb_{ex}$ (CRS model). DOI: https://doi.org/10.1525/elementa.2021.00068.f2

Table 2. Total organic carbon (TOC) and total nitrogen (TN) contents, C/N ratios, $\delta^{13}C_{carb}$, $\delta^{13}C_{org}$, and proxies of n-alkanes of the sediment cores from Fuxian Lake. DOI: https://doi.org/10.1525/elementa.2021.00068.t2

Site	TOC (mg g^{-1})	TN (mg g^{-1})	C/N	$\delta^{13}C_{org}$ (‰)	ACL	CPI	TAR	Paq
FX-1	8.75–31.22[a]	1.10–3.44	7.54–10.71	−28.07−24.77	29.17–29.94	0.92–1.44	0.58–2.19	0.27–0.48
	(13.78)[b] [1.83][c]	(1.78) [0.18]	(8.76) [0.24]	(−26.13) [0.26]	(29.57) [0.08]	(1.10) [0.04]	(1.25) [0.17]	(0.39) [0.02]
FX-2	6.23–18.22	0.99–2.15	7.53–9.89	−27.71−24.87	29.08–29.77	1.02–1.12	0.23–1.16	0.41–0.48
	(7.92) [0.62]	(1.12) [0.06]	(8.14) [0.14]	(−25.60) [0.15]	(29.52) [0.08]	(1.07) [0.01]	(0.85) [0.12]	(0.44) [0.01]

ACL = average chain length; CPI = carbon preference index; TAR = terrigenous/aquatic ratios; Paq = proportion of aquatic macrophytes.

[a]Minimum-maximum.

[b]Mean value.

[c]Standard error.

in cores FX-1 and FX-2 ranges from 8.8 to 31.2 mg g^{-1} and from 6.2 to 18.2 mg g^{-1}, respectively. The mean TOC content of core FX-1 (23.42 mg g^{-1}) is considerably higher than that of core FX-2 (15.02 mg g^{-1}). The C/N atomic ratio of both cores is generally <10 and exhibits low amplitudes variations during 1910–2000 and a rapid increase after approximately 2000 (**Figure 3**, Tables S1 and S2).

4.3. Carbon isotope ratios of organic carbon
The $\delta^{13}C_{org}$ values in cores FX-1 and FX-2 decrease exponentially from the bottom to the top, with the values ranging from −28.07 to −24.77 ‰ and from −27.71 to −24.87 ‰, respectively (**Table 2**, Tables S1 and S2). The values are relatively stable below 7 cm with mean values of 25.46‰ and 25.22‰ in cores FX-1 and FX-2, respectively. The values decrease rapidly above 7 cm and are the most negative at the top (**Figure 3**).

4.4. Distribution of n-alkanes and the proportions of autochthonous carbon
In all of the samples, the n-alkane distributions range from C_{12} to C_{34}. The total concentration of n-alkanes ($\sum(C_{12}$ to C_{34})) ranges from 8,929 to 37,973 ng g^{-1} (mean of 14,131 ng g^{-1}). The n-alkanes of the sediment cores
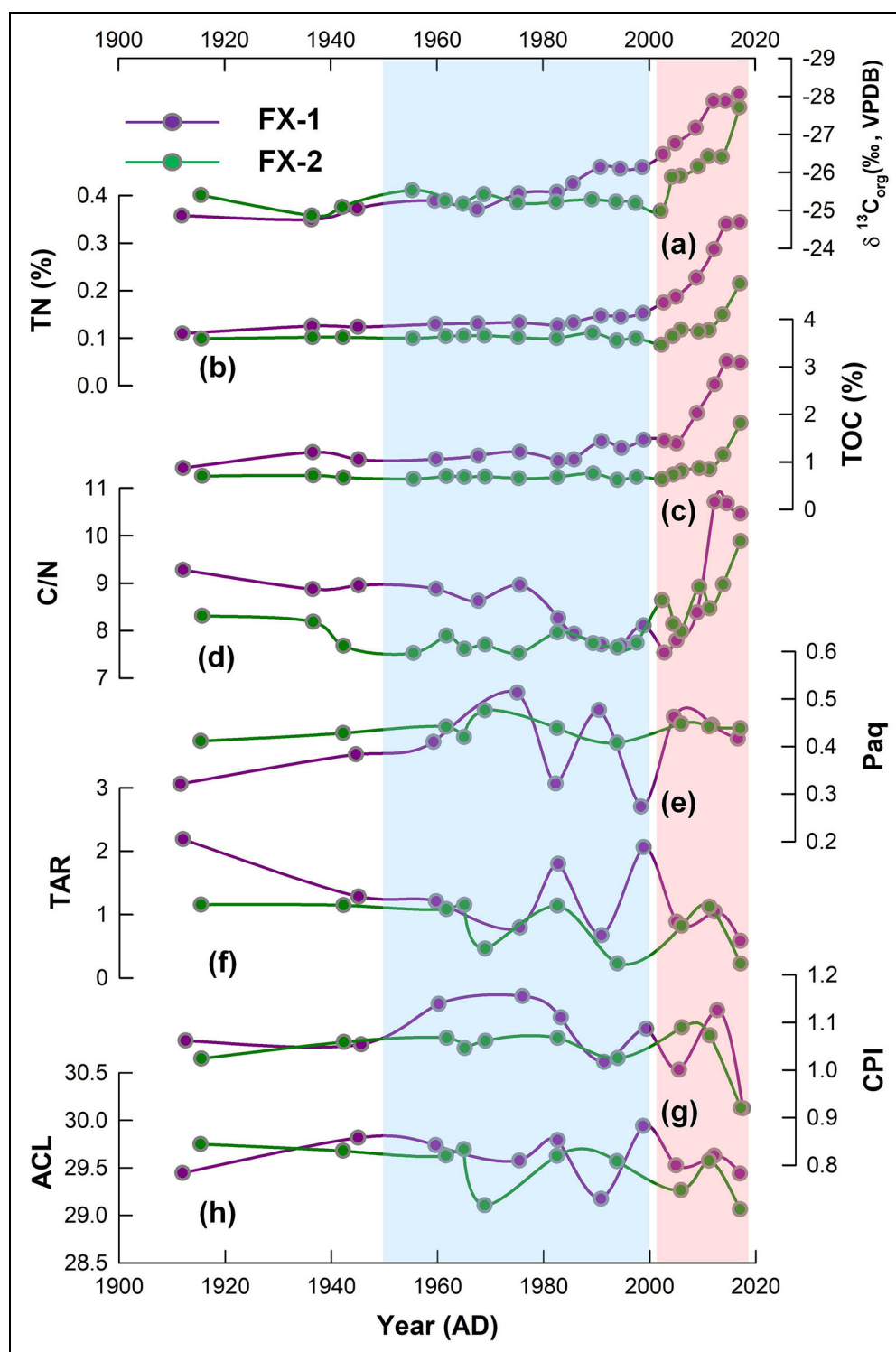
Figure 3. Summary plot of the organic geochemical parameters derived from the Fuxian Lake sedimentary sequence over the past century. (A) $\delta^{13}C_{org}$, (B) TOC, (C) TN, (D) C/N, (E) Paq, (F) TAR, (G) CPI, (H) ACL. DOI: https://doi.org/10.1525/elementa.2021.00068.f3

exhibit a bimodal carbon number distribution with an odd–even pattern in the range of C_{25}–C_{33}. The abundance of short-chain n-alkanes below C_{20} in the samples is relatively high: C_{17} (range of 348.70–8,845.98 ng g^{-1}, mean of 1,419.01 ng g^{-1}) and C_{18} (range of 312.01–7,230.11 ng g^{-1}, mean of 1,178.74 ng g^{-1}) are the main peak carbon numbers, and there is no obvious odd–even pattern. The values of the various indices are listed in **Table 2** and Tables S1 and S2. The n-alkane ACL values range from 29.08 to 29.94 (mean of 29.54), the CPI values range from 0.92 to 1.44 (mean of 1.07), and the Paq values range from 0.27 to 0.48 (mean of 0.42). The TAR reflects the terrigenous/aquatic ratio of the hydrocarbons in the samples, with the values ranging from 0.23 to 2.19 (mean of 1.05). The variations in Paq during the past approximately 100 years are similar to those of the conventional geochemical
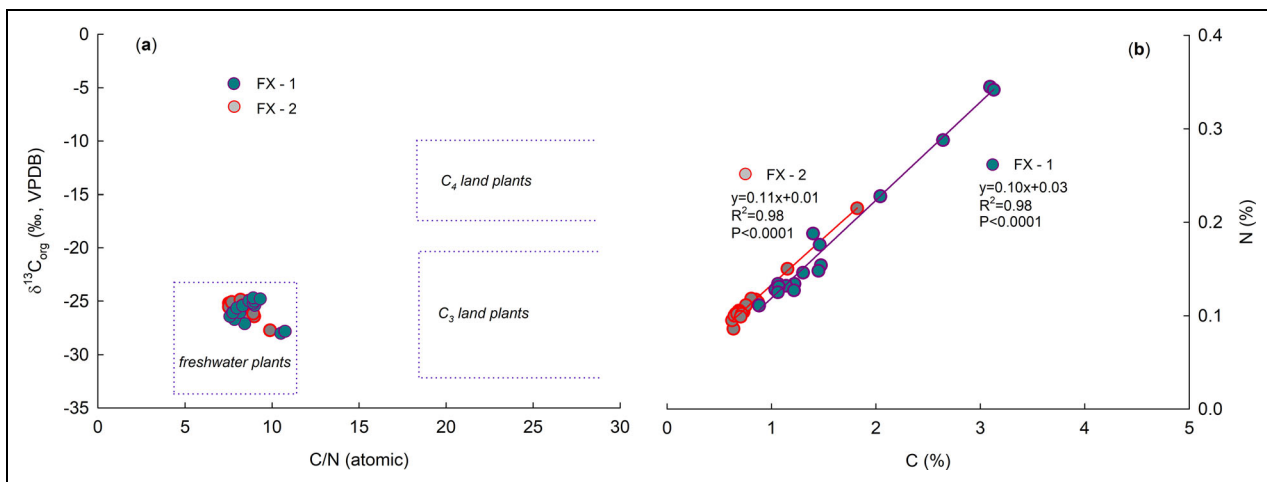
Figure 4. Major sources of the organic material in the recent sediments of Fuxian Lake. (A) Distinctive source combinations of the C/N ratio and $\delta^{13}C_{org}$ values, reflecting allochthonous and autochthonous sources (based on Meyers, 1994, and Tyson, 2012). (B) Biplot of N and C carbon concentrations in the sediments of Fuxian Lake. DOI: https://doi.org/10.1525/elementa.2021.00068.f4

proxies (TOC, C/N, and $\delta^{13}C_{org}$). ACL, CPI, and TAR show similar trends, which are in contrast with that of Paq (**Figure 3**).

5. Discussion

5.1. Effects of diagenesis on the geochemical records

The geochemical data obtained in this study provide a basis for reconstructing the paleoenvironment of Lake Fuxian based on the temporal distribution of n-alkanes, C/N ratio, and $\delta^{13}C_{org}$ values. However, the possible impacts of diagenetic processes on the OM composition of the sediments must first be considered. OM degradation is strongly controlled by the oxygen level, with lipids being more rapidly decomposed in an aerobic environment than in an anaerobic environment (Khan et al., 2015). Fuxian Lake is a deep oligotrophic freshwater lake, with a low dissolved oxygen content (≤ 4) below water depths of 100–120 m (He et al., 2019). The coring sites were located in a water depth of 120 m and are believed to have been continuously hypoxic. Furthermore, the enhanced preservation of OM in an aerobic combined with the rapid accumulation rate (the mean OC accumulation rate is 16.83 g C m^{-2}yr^{-1}) suggests that the impact of OM degradation is limited.

The differential degradation of OM during diagenesis is another factor that should be considered when evaluating the sedimentary record. The C/N atomic ratios are much higher at the top of the cores than at the bottom (**Figure 3**), which suggests the limited impact of early diagenesis. As nitrogen compounds are usually preferentially remineralized, the C/N atomic ratio is expected to increase with depth and the degree of diagenesis within a core (Meyers, 1997). Additionally, the TOC and TN contents are highly correlated both within and between the two cores (**Figure 4**), which suggests the absence of significant preferential degradation of carbon or nitrogen during the past century. Notably, compared with other organic components in sediments, n-alkanes contain carbon bonds with a high

bond energy (i.e., they are relatively stable; Blumer et al., 1971; Fokin et al., 2012) and are readily preserved in sediments as they are resistant to degradation (Eglinton and Hamilton, 1967; Meyers, 2003). For these reasons, we assume that the Fuxian Lake sediment record mainly reflects changes in the sources of the OM, the signals of which dominate any effects of diagenesis.

5.2. Organic geochemical evidence for changes in the source of sedimentary OC

The relative abundances of autochthonous and allochthonous OC in lake sediments depend closely on variations in the environmental conditions within and around the lake (Choudhary et al., 2009). In terrestrial aquatic ecosystems, photosynthetic uptake of dissolved inorganic carbon or CO_2 transforms these carbon sources into autochthonous OM (Einsele, 2001; Lerman and Mackenzie, 2005; Nõges et al., 2016; Yang et al., 2016; Chen et al., 2017; Maavara et al., 2017; He et al., 2019). Allochthonous OC, on the other hand, represents terrigenous carbon sources such as vascular plant tissues, detritus from marsh vegetation, and upland sources (Bianchi, 2007; O'Reilly et al., 2014).

n-alkanes in lake sediments are widely used as tracers (or proxies) to characterize the sources of OM due to their source specificity and relatively high resistance to bacterial degradation (Eglinton and Hamilton, 1967; Blumer et al., 1971; Meyers, 2003; Fokin et al., 2012; He et al., 2020). The n-alkane proxies for each core site in this study are shown in **Figure 3**. The n-alkane ACL value is greater in higher plants than in lower plants and aquatic algae (Poynter and Eglinton, 1990). The low variation and values of the ACL for the Fuxian Lake sediment cores are due to the low input of terrestrial plant materials. The CPI of the long-chain n-alkanes can be used to differentiate biogenic sources of the OM in lake sediments. In this study, the decrease in the CPI of the long-chain n-alkanes during the past century can be explained by increases in aquatic macrophyte inputs (Shen et al., 2019). Similarly, the TAR values
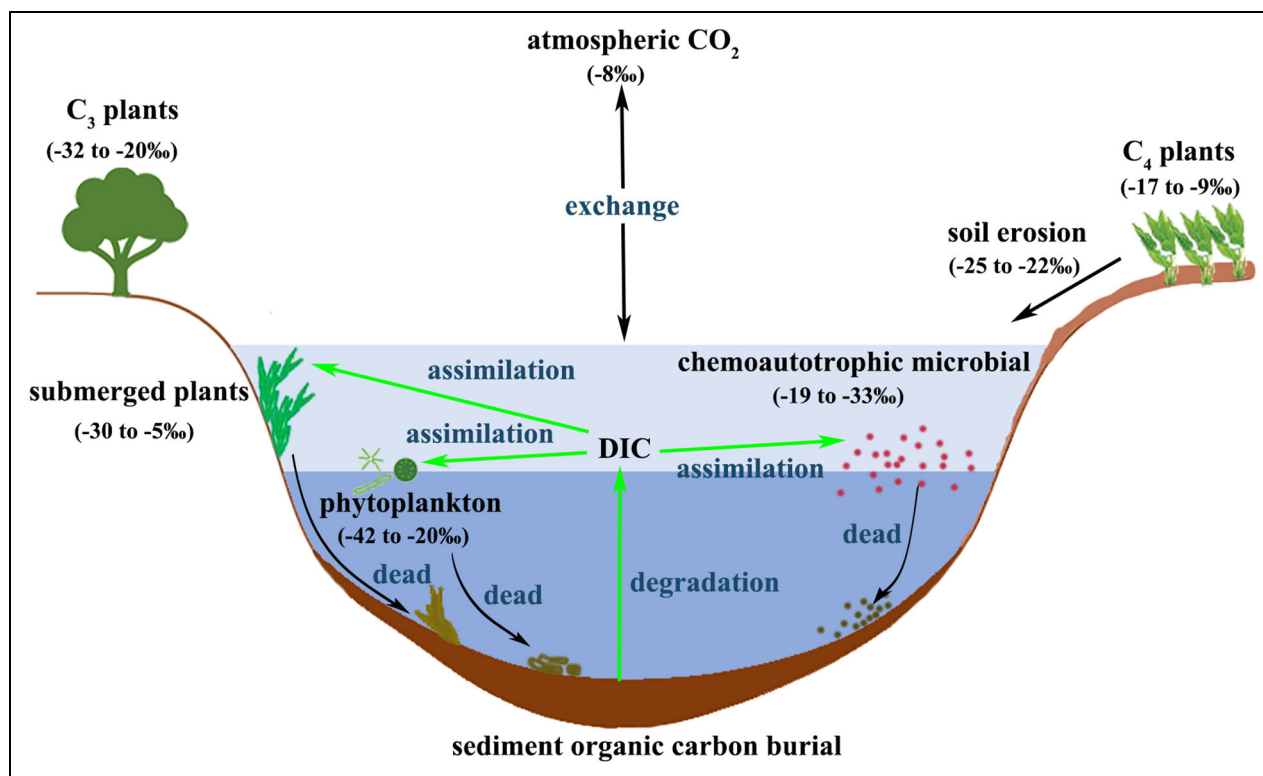
Figure 5. Schematic diagram showing potential factors influencing the organic carbon isotopes of lake sediments. The direction indicated by the green arrow indicates the enrichment of ^{12}C. DOI: https://doi.org/10.1525/elementa.2021.00068.f5

in the sediment cores represent an increasing algal contribution to the sedimentary n-alkane composition during the past century (Meyers, 2003). In contrast to the above n-alkane proxies, the Paq values in the sediment cores show an increasing trend, and the lower values suggest an interval that was relatively enriched with OM derived from submerged and floating-leaved plants and a reduction in that from emergent and terrestrial plants (Ficken et al., 2000). These proxy data, combined with the fact that the most abundant n-alkane is C_{17} in both cores, indicate that aquatic plants have made an increasing contribution to the sedimentary OM.

C/N ratios and $\delta^{13}C_{org}$ values have also been widely used to identify variations in the relative inputs of autochthonous and allochthonous OM in aquatic environments (Meyers, 2003; Zhong et al., 2018). The C/N ratios of terrigenous plants, bacteria, phytoplankton, macrophytes, and soil are suggested to fall within the following respective ranges: 20–100 (Meyers, 1994), 4–6 (Vane et al., 2013), 4–10 (Meyers, 1994, 2003), ≥12 (Tyson, 2012), and 10–13 (Goñi et al., 2003). Terrestrial C_3 plants have a mean $\delta^{13}C_{org}$ value of approximately −27‰ (range of −32 to −20‰), whereas C_4 plants have a mean value of approximately −13‰ (range of −17‰ to −9‰; Meyers and Ishiwatar, 1993; Meyers, 2003). It has been found that algae and plankton have more negative $\delta^{13}C_{org}$ values (−42‰ to −20‰) due to C_3-like photosynthesis, while the small degree of carbon isotopic fractionation through C_4-like photosynthesis results in the enriched $\delta^{13}C_{org}$ values (−30‰ to −5‰) of submerged plants (Farquhar et al.,

1989; Liu et al., 2013; Li et al., 2019). Thus, the ranges of the C/N atomic ratio (5.80–10.71) and $\delta^{13}C_{org}$ values (−28.07‰ to −24.24‰) of the organic component of the sediments from Fuxian Lake originate mainly from lacustrine algae (**Figure 4**).

These observations therefore confirm that the source of the sedimentary OC in Fuxian Lake is mainly autochthonous. Furthermore, the multidecadal variations evident in the characteristics of the sedimentary OM (n-alkane distribution and elemental and isotopic parameters) are significant for understanding the origins of the organic inputs to the sedimentary sequence.

5.3. Human-induced environmental changes recorded by the sediments of Fuxian Lake
Ideally, the variations of different proxies in lacustrine sediments should be combined with historical records to produce an integrated record of the impacts of anthropogenic activity over time. The relative contributions of aquatic macrophytes, phytoplankton, terrestrial plants, soil, and bacteria to lake sediments can be inferred from organic geochemical parameters, and they can then be used to elucidate the nature of environmental changes associated with human activities in the catchment. As noted previously, autochthonous sources were the primary contributors to the sediments of Fuxian Lake during the past century. However, at different stages, variations in the patterns of the organic geochemical record in lake sediment cores suggest the occurrence of environmental changes. Correspondingly, the evolution of the lake has

been divided into three stages (**Figure 3**), which are discussed below.

5.3.1. Natural evolution of the lake environment
The establishment of human settlements and the development of agriculture in the Lake Fuxian watershed have a long history. Up until the 1950s, agricultural practices were relatively primitive with a low productivity and they can be assumed to have had a relatively minor influence on the lake environment. This interpretation is in accord with the sedimentary record: during the period of approximately 1910–1950, corresponding to the lower part of the sediment cores, the geochemical indicators indicate relatively stable conditions. The ACL and TAR indices are comparatively high, whereas the Paq values are low during this period (**Figure 3**), suggesting that aquatic macrophytes and phytoplankton had a relatively low productivity. These indices indicate that the source of the sedimentary OC was mainly autochthonous due to the uniform and relatively low C/N ratios and higher $\delta^{13}C_{org}$ values, compared with the younger sediments (**Figure 3**). In addition, all of the proxy data point to occurrence of oligotrophic conditions at this time, which explains the observed low sedimentary contents of TN and TOC. Liu et al. (2014) found that the content of trace metals (Cr, Pb, and Zn) of the Lake Fuxian sediments predating approximately 1950 were low and constant. Therefore, the influence of human activities was probably a minor factor in the lake watershed environment prior to 1950, and the measured parameters therefore represent the natural evolution of Fuxian Lake.

5.3.2. Anthropogenic transformation of the lake environment
Changes in land-use patterns and the associated soil erosion are one of the most significant human influences on lake environments, especially over recent decades (Ning et al., 2018). The period from approximately 1950 to 2000 recorded in the sedimentary records from Lake Fuxian coincided with population growth and related increases in land-use pressure (e.g., agricultural activity, deforestation, urbanization, and industrialization) within the Fuxian Lake catchment after the Chinese Civil War (1945–1949). The sedimentary organic geochemical record indicates major changes in the lake watershed system at this time: The ACL, CPI, and TAR values exhibit high-amplitude fluctuations but overall decreasing trends during this interval (**Figure 3**), clearly differentiating it from the earlier stage of predominantly natural evolution. The data suggest that during this later phase, there was the increased input of autochthonous OM derived mainly from algal and bacterial communities, and/or aquatic plants. The Paq values show the opposite trend during this period (**Figure 3**), suggesting a relative enrichment from submerged/floating plants and a reduction in emergent/terrestrial plants. n-alkanes are more sensitive to environmental changes than the conventional geochemical parameters. The C/N atomic ratio and $\delta^{13}C_{org}$ values in the sediment cores both show little variation during 1950–2000 (**Figure 3**). This suggests that the source of

sedimentary OM in Fuxian Lake was mainly autochthonous because of the dominantly low TAR values. This was likely due to the increased input of nutrients to the lake, which would have increased aquatic photosynthesis and hence primary productivity of the lake water, thus leading to the flourishing of aquatic plants (Schelske et al., 1988).

During this period, the TOC and TN contents of core FX-2 were relatively uniform, whereas they increased slightly in core FX-1 core, which was retrieved from a location close to a densely populated area. An increase is also evident in the n-alkane proxies, and these changes suggest that an increased intensity of human activity resulted in a rise in the influx of nutrients to the lake basin, causing the increased accumulation of sedimentary OC. This is consistent with the findings of Liu et al. (2014), who reported the increased input of Cr in the northern part of Fuxian Lake during the same period, while the Cr concentration in the sediments in the center of the basin were relatively stable. It should be noted that the lake ecosystem, including the water quality, was adversely affected by economic development. Although it is an oligotrophic lake, accelerated trace metals deposition (e.g., Pb, Zn, and Cd), as early as the 1950s and especially from the mid-1980s (Liu et al., 2014), has accompanied the transformation of the trophic status of the lake toward eutrophication.

5.3.3. Recent intensive exploitation of the lake environment
The third stage of lake environmental evolution occurred in the 21st century, when the Fuxian Lake watershed experienced more intensive exploitation, including cultivation, industrialization, and tourism. The $\delta^{13}C_{org}$ values in the sediment cores show a markedly decreasing trend from 2000 to the present (Figure 3). Although the amount and composition of OM deposited in lake sediments can be greatly affected by metabolism and remineralization both during and after sedimentation, carbon isotopic fractionation can be negligible during postphotosynthetic processes (Khan et al., 2015). For Fuxian Lake, the negative shift induced by the Suess effect (i.e., a shift to more negative $\delta^{13}C$ values of atmospheric CO_2 caused by fossil fuel combustion and deforestation) is insufficient to explain the very large variations in $\delta^{13}C_{org}$ values in the cores. Studies of OC in lake sediments have demonstrated that the carbon isotope ratio can vary considerably (Figure 5), which can be the consequence of the following factors: (1) the increased contribution of phytoplankton, which assimilate proportionally larger amounts of $^{12}CO_2$ (Falkowski and Raven, 2013); (2) changes in the structure of the lake biota with different elemental ratio and isotope values (Chen et al., 2015); (3) an expansion of the chemoautotrophic microbial communities with intensified eutrophication, thereby causing greater fractionation effects than those associated with photosynthesis (Kelleyl et al., 1998); and (4) the increased degradation of OM, which produces more $^{12}CO_2$ that is gradually consumed by algae, thus resulting in more negative $\delta^{13}C_{org}$ values in OM (Chen et al., 2018). Hence, the markedly negative $\delta^{13}C$ values indicate that the OM input mainly originated from mixed sources, which were easily influenced by other

environmental factors, producing a mixed signal which is difficult to interpret.

The C/N atomic ratio in the Fuxian Lake sediments shows a rapid increase (Figure 3) that coincides with an increase in the TOC content. This suggests that the C/N ratio was influenced by contributions from terrestrial sources (e.g., soil and C_3 plants) and autochthonous OM (e.g., submerged/floating-leaved macrophytes). However, traditional geochemical indicators ($\delta^{13}C_{org}$, C/N) used to distinguish autochthonous OM from allochthonous OM are based on the comparison between characteristic $\delta^{13}C_{org}$ and C/N values that can only provide a general indication of the OC source. These parameters are influenced by various environmental processes which makes them difficult to interpret. However, biomarker compounds such as n-alkanes are a sensitive recorder of environmental changes (Figure 3). The ACL and CPI values show decreasing trends, and Paq an increasing trend, which are consistent with a predominance of submerged/floating-leaved macrophytes in the aquatic macrophyte community. Moreover, the TAR values decrease over time in the two cores, which suggests the increasing input of OM from algal and bacterial communities, and/or aquatic plants.

In summary, the C-enriched sedimentary OM in the upper sediment layer (i.e., after 2000) may result from an increase in the abundance of planktonic algae accompanied by a large increase in submerged plants, although the former may have been more important. However, an increasing OM contribution from terrestrial sources is also possible. Periods of widespread trace metal pollution since approximately 2000 are reflected in peaks in the Pb–Zn–Cr profiles in the sediments of Lake Fuxian (Liu et al., 2014), which are correlative with increases in the TOC and TN contents. These changes may be the result of increases in anthropogenic nutrient inputs. The results of modern hydrochemical monitoring have revealed that the TN and total P contents have increased rapidly over the last two decades in Fuxian Lake (Li et al., 2012), which have promoted the growth of aquatic plants, especially phytoplankton. Notably, changes in the trophic status of Fuxian Lake indicate an increasing trend toward eutrophication despite the fact that it has never been considered to be a eutrophic lake.

6. Conclusions

We have used measurements of $\delta^{13}C_{org}$, C/N ratios, and n-alkane proxies (ACL, CPI, TAR, and Paq) from the recent (last approximately 100 years) sediments of Lake Fuxian in the Yunnan–Guizhou Plateau to reconstruct the effects of human activities on the lake watershed. The stratigraphic records of these parameters indicate variations in biological production, with the n-alkanes proxies shown to be more sensitive than $\delta^{13}C_{org}$ and C/N ratio. The results indicate increasing human impacts from the 1950s onward, with pronounced eutrophication occurring in the 21st century. Significantly, the differences in the stratigraphic records of the various organic geochemical parameters emphasize the value of a multi-proxy approach in paleolimnological investigations. Overall, the molecular

sedimentary evidence from Lake Fuxian confirms that the environmental evolution of lakes in the Yunnan–Guizhou Plateau over the past century was closely associated with the intensity of human activity.

Supplemental files
The supplemental file for this article can be found as follows:

Tables S1–S2. Supplemental Materials. Docx

Competing interests
The authors do not have any competing interests that might influence the interpretation of this manuscript.

Author contributions

- Concept and design: HH, ZL.
- Acquisition of data: HH, CC, QB, YW.
- Analysis and interpretation of data: HH, DL, HB, JX, HS, HY.
- Drafting the article or revising it for important intellectual content: HH.
- Final approval of the version to be published: HH, ZL.

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