

A re-evaluation of the average chain length of lacustrine sedimentary *n*-alkanes as a paleoproxy on the Qinghai-Tibet Plateau

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Abstract Long-chain *n*-alkanes are one of the most common organic compounds in terrestrial plants and they are well-preserved in various geological archives. *n*-alkanes are relatively resistant to degradation and thus they can provide high-fidelity records of past vegetation and climate changes. Nevertheless, previous studies have shown that the interpretation of *n*-alkane proxies, such as the average chain length (ACL), is often ambiguous since this proxy depends on more than one variable. Both vegetation and climate could exert controls on the *n*-alkane ACL, and hence its interpretation requires careful consideration, especially in regions like the Qinghai-Tibet Plateau (QTP) where topography, biome type and moisture source are highly variable. To further evaluate the influences of vegetation and climate on the ACL in high-elevation lakes, we examined the *n*-alkane distributions of the surface sediments of 55 lakes across the QTP. Our results show that the ACL across a climatic gradient is significantly affected by precipitation, rather than by temperature. The positive correlation between ACL and precipitation may be because of the effect of microbial degradation during deposition. Finally, we suggest that more caution is needed in the interpretation of ACL data in different regions.

Keywords ACL, average chain length, *n*-alkanes, leaf wax, lake sediments, Qinghai-Tibet Plateau

1 Introduction

Rapid climate changes are especially evident in high mountain regions (Liu et al., 2009; Mountain Research Initiative EDW Working Group, 2015), such as the Qinghai-Tibet Plateau (QTP). As the largest and most important “water tower” in Asia, the QTP is experiencing rapid and unprecedented warming (Immerzeel et al., 2010; Immerzeel et al., 2020). Consequently, the water availability and the regional ecosystems are likely to be increasingly affected by this warming (Yao et al., 2019). Therefore, it is important to achieve a comprehensive understanding of past climate changes in such high-elevation regions and their potential effects on different types of water resource, which necessitates an improved understanding of the natural climate variability of these regions and the assessment of anthropogenic effects.

Sedimentary long-chain *n*-alkanes are often regarded as terrestrial higher plant biomarkers and their distributions and isotopic signatures are commonly used to reconstruct changes in continental climatic conditions and vegetation (Brincat et al., 2000; Zhou et al., 2005; Castañeda et al., 2009b; Sachse et al., 2012), for the following reasons. 1) As one of the major components of leaf waxes (Eglinton and Hamilton, 1967), *n*-alkanes are widespread and well-preserved in different sedimentary deposits (Diefendorf et al., 2011); and 2) plants can adapt to environmental stresses via the modification of the acyl chain, such as the biosynthesis of longer chain-length waxes to limit water loss (Shepherd and Wynne Griffiths, 2006, Tipple and Pagan, 2013). Due to their high relative abundance and additional characteristic source information, the *n*-alkane average chain length (ACL) has been used to infer past climatic and vegetation changes (Chen et al., 2020; Toney

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et al., 2020; Xie et al., 2020). ACL can be interpreted in different ways, but it typically characterizes the relative contribution of different organic matters in the sediments when targeting a wide carbon range of ACL values (Norström et al., 2014; Jin et al., 2016; Günther et al., 2016; Witt et al., 2016; Saini et al., 2017; Callegaro et al., 2018; Yan et al., 2020). It is also regarded as a paleoproxy if the long-chain homologs are selected (Pu et al., 2013; Ling et al., 2017b; Liu et al., 2021).

During the past decade, the long-chain *n*-alkane ACL has been used for continental vegetation reconstruction (Zhou et al., 2012; Castañeda et al., 2016; Norström et al., 2017; Norström et al., 2018; Häggi et al., 2019). With the development of new proxies, various biomarker approaches have been used to reveal vegetation histories, such as lignin phenols ratios (Castañeda et al., 2009b), and the stable carbon isotope ratios of *n*-alkanes and lignin phenols (Huang et al., 1999a, 1999b; Feakins et al., 2005; Castañeda et al., 2009a; Sinninghe Damsté et al., 2011; Kuechler et al., 2013; Garcin et al., 2014); however, the analysis of *n*-alkane distributions is relatively straightforward. In some cases, the ACL proxy may complement other approaches used to reconstruct vegetation changes, such as the ratio of grasses to woody plants (Norström et al., 2018). The ACL can also be used to infer past climatic variability in natural archives, including marine sediments (Poynter et al., 1989; Li et al., 2015), lake sediments (Castañeda et al., 2009b; Pu et al., 2013; Sun et al., 2018; Tian et al., 2019), peat sediments (Nichols et al., 2006; Zhou et al., 2010; Zhang et al., 2014; Naafs et al., 2019), and loess-paleosol sequences (Zhang et al., 2006; Bai et al., 2009; Zech et al., 2013). However, several studies have revealed that the factors determining *n*-alkane distributions are not uniform (Schefuß et al., 2003; Bush and McInerney, 2015; Eley and Hren, 2018; Wang et al., 2018a).

For the Qinghai-Tibet Plateau, much effort has been expended to evaluate the impact of climatic variables on *n*-alkane average chain lengths; nevertheless, there is a debate about the interpretation of *n*-alkane ACL across the QTP. ACL was shown to respond to precipitation rather than to temperature, based on the analysis of lake surface sediments in the south-western QTP (Hu et al., 2014). Guo et al. (2015) investigated plants and surface soils in alpine meadow ecosystems across the eastern QTP and observed the effect of temperature on ACL. However, a study of soil samples by Jia et al. (2016) showed that both climatic variables (precipitation and temperature) were positively correlated with ACL. Although these studies have reported the relationships between ACL and climatic variables, a recent field experiment conducted in central Tibet concluded that neither temperature nor precipitation had a strong impact on the chain length of *n*-alkanes (Bai et al., 2019). Hence it is imperative to clarify whether ACL is an effective paleoproxy for lake sediments in this region.

To this end, we investigated the environmental factors determining the ACL in a series of surface sediments from 55 lakes across the QTP that were obtained during the past decade. Our specific objectives were to disentangle the influences of vegetation and climate on the long-chain *n*-alkane distributions, and to identify the main climatic variables that control the chain length of sedimentary *n*-alkanes in these lakes.

2 Materials and methods

2.1 Field sampling

Lake surface sediments from 55 lakes were collected during fieldwork between 2010 and 2017; 46 lakes are located along a west-east transect in Tibet, and 9 lakes are in the Qaidam Basin, an intermontane basin in the north-eastern QTP (Fig. 1(a), Table 1). The lake areas range from 6.3 km² (Zongxiong Co) to 2300.4 km² (Selin Co), and the altitudes range from 2689 m (Xitaijinaier) to 5086 m (Guozha Co) (Table 1). The 55 lakes span a relatively large climatic gradient, with the mean annual air temperature (MAAT) range from −6.9°C to 5.4°C and the mean annual precipitation (MAP) ranges from 44 mm to 635 mm (Figs. 1(b) and 1(c)). The main vegetation types around these lakes are meadow, steppe and desert (Hou, 2001). We collected the surface lake sediment samples (0–2 cm) using an Ekman-Birge grab, and immediately placed them in Nasco Whirl-Pak bags. In the laboratory, the samples were kept at −20°C prior to freeze-drying.

2.2 Lipid extraction and measurement

Approximately 2–4 g of freeze-dried lake surface sediments were ground and homogenized before lipid extraction. Samples were extracted using an accelerated solvent extractor system (Dionex ASE350) with dichloromethane (DCM):methanol (MeOH) (9:1, v:v), at the temperature and pressure of 120°C and 1500 psi, respectively. The total lipid extracts (TLEs) were quantified by comparing the weights of lipid extracts before and after drying under a stream of nitrogen gas using a TurboVap system. TLE were separated into neutral and acid fractions by elution through a LC-NH₂ SPE column using DCM:isopropyl alcohol (1:1, v:v), followed by ether with 4% acetic acid (v:v) as eluents, respectively. The neutral fractions were further separated into four fractions of increasing polarity via chromatography over a silica gel column using hexane, DCM, ethyl acetate:hexane (1:3, v:v) and MeOH as eluents.

n-Alkanes from the hexane fraction were detected and quantified using gas chromatography with a flame ionization detector (GC-FID). Samples were passed through the GC-FID (model Agilent 7890B) using GC

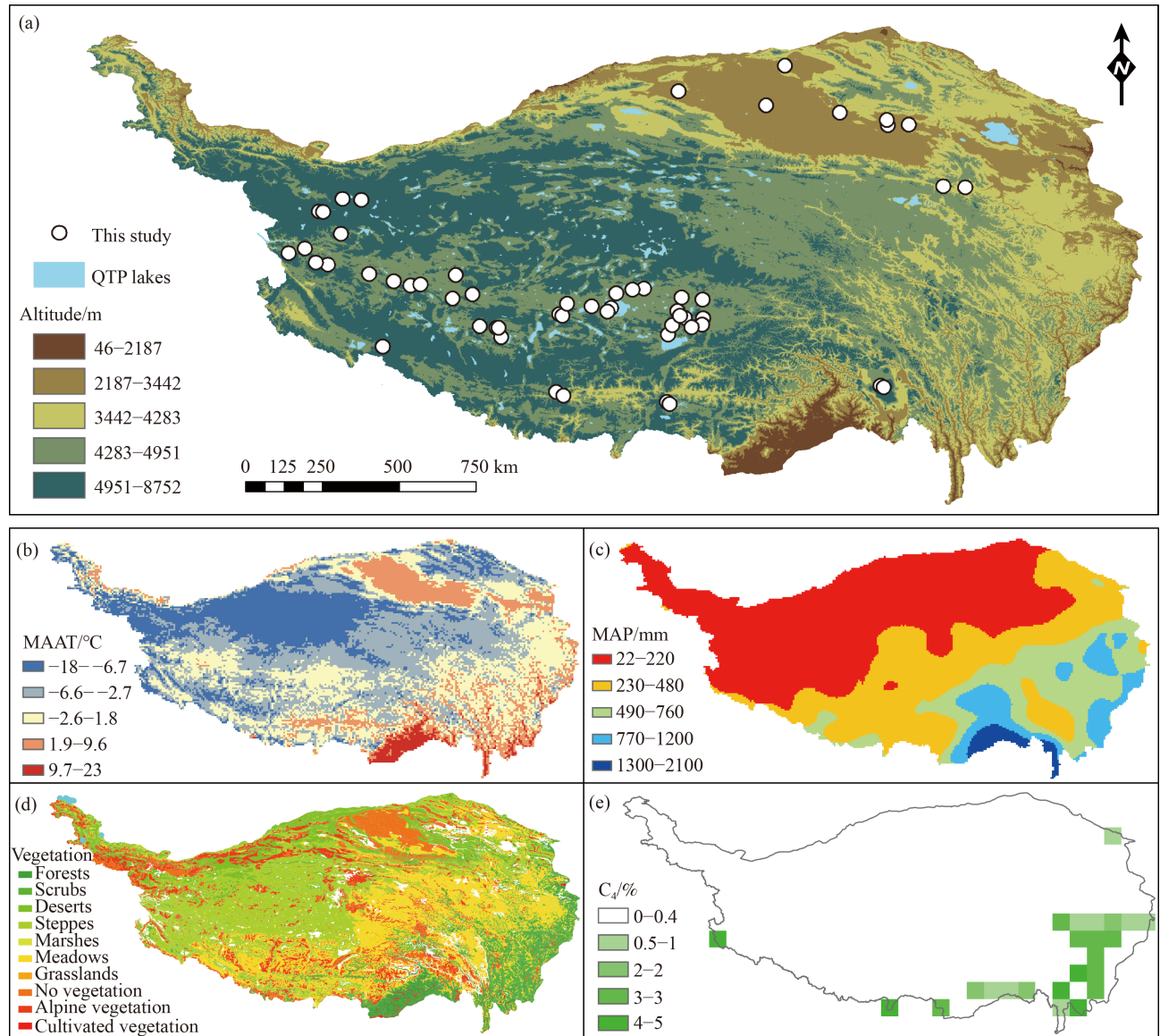


Fig. 1 The study area and sampling locations along the two transects on the Qinghai-Tibet Plateau (QTP) (a) Gridded climate data are presented for both Mean Annual Air Temperature (MAAT). (b) and Mean Annual Precipitation (MAP). (c) Maps of vegetation information including biomes and the percentage of C₄ plants, respectively (d, e).

Table 1 Sediment sampling site information

No.	Lake	Latitude/°N	Longitude/°E	Elevation/m	Lake area/ km ²	Salinity/(g·L ⁻¹)	pH	MAAT/°C	MAP/mm	Vegetation zone
1	Shen Co	30.9983	90.4792	4734	52	9.26	9.73	-1.45	350	steppe
2	Guozha Co	34.9791	80.9386	5086	245	2.98	9.20	-6.93	44	desert
3	Bamu Co	31.2634	90.5895	4565	255	7.51	9.70	-0.87	350	steppe
4	Gahai	37.1404	97.5304	2852	35	76–94*	8.40	4.73	197	desert
5	Zigetang Co	32.0729	90.8625	4573	238	13.5	10.00	-1.38	372	steppe
6	Rena Co	32.7328	84.2545	4599	21	15.2	9.25	-0.83	131	steppe
7	Xiaochaidan	37.4964	95.5019	3177	88	50*	8.30	2.29	88	desert
8	Bangong Co	33.5171	79.8252	4244	671	0.47	8.73	-0.59	88	desert
9	Yangnapeng Co	32.3409	89.7726	4634	17	30	9.92	-0.57	285	steppe
10	Cuoe-2	31.4606	91.4999	4528	85	3.63	9.51	-1.30	394	meadow
11	Bieruoze Co	32.4304	82.9228	4407	36	27.38	8.97	1.47	131	steppe

(continued)

No.	Lake	Latitude/°N	Longitude/°E	Elevation/m	Lake area/ km ²	Salinity/(g·L ⁻¹)	pH	MAAT/°C	MAP/mm	Vegetation zone
12	Gemang Co	31.5825	87.2822	4610	62	6.35	9.73	-1.25	219	steppe
13	Dasugan	38.8740	93.9036	2796	108	20	8.90	1.77	44	desert
14	Bangda Co	34.9420	81.4888	4909	143	35.11	8.50	-5.53	44	desert
15	Anggu Co	31.1997	85.4482	4665	33	1.89	9.10	-2.35	197	steppe
16	Jieze Chaka	33.9417	80.8814	4530	114	146*	9*	-1.64	66	desert-steppe
17	Nairiping Co	31.2821	91.4713	4529	93	7.96	9.98	-1.22	394	steppe
18	Lagor Co	32.0506	84.1696	4472	96	40.27	8.94	-0.36	110	steppe
19	Pusaier Co	32.3147	89.4297	4593	34	5.3	9.66	-0.06	263	steppe
20	Aweng Co	32.7695	81.7193	4430	70	27.65	9.20	1.63	131	steppe
21	Dong Co	32.1752	84.7392	4399	105	46.25	8.82	-0.35	110	steppe
22	Angrenjin Co	29.3104	87.1997	4303	21	5.26	9.64	3.90	350	steppe
23	Cuona	32.0215	91.4819	4590	191	0.27	8.70	-1.54	372	meadow
24	Lang Co	29.2068	87.4050	4296	10	1.58	9.44	4.57	329	steppe
25	Zhangnai Co	31.5467	87.3852	4611	44	4.06	9.60	-1.13	241	steppe
26	Peng Co	31.4843	90.9576	4534	176	8.54	9.91	-1.29	372	steppe
27	Dawa Co	31.2371	84.9667	4628	118	18.58	9.30	-2.14	197	steppe
28	Selin Co	31.7675	88.7988	4544	2300	8.36	9.33	0.22	241	steppe
29	Qiagui Co	31.8153	88.2320	4558	89	0.22	8.83	-0.11	241	steppe
30	Kongmu Co	29.0128	90.4394	4450	37	0.23	8.55	2.02	416	steppe
31	Dongji Cona	35.3334	98.5351	4086	241	0.35	8.76	-2.30	219	steppe
32	Daru Co	31.6838	90.7440	4688	70	5.05	9.23	-2.13	350	steppe
33	Kuhai	35.3054	99.1753	4132	47	16.1	8.82	-2.89	263	steppe
34	Beng Co	31.2149	91.1654	4671	144	0.16	8.74	-2.14	394	steppe
35	Zhari Namco	30.9136	85.5945	4617	1001	10.6	9.49	-1.63	219	steppe
36	Ga'a Co	32.2090	88.9592	4620	13	3.38	9.68	-0.44	241	steppe
37	Xiada Co	33.3888	79.3618	4358	8	0.15	8.60	-2.29	110	desert
38	Gasikule	38.1209	90.7732	2857	116	333*	7.50	2.31	66	desert
39	Rebang Co	33.0377	80.5138	4326	46	53.6	9.19	-0.31	66	desert
40	Xitaijinaier	37.7175	93.3465	2689	NIA	336*	7.7*	3.99	44	desert
41	Ranwu (AMC)	29.4880	96.7010	3920	7	0.07*	8.13*	1.03	635	Conifer forest
42	Ranwu (AC)	29.4489	96.7921	3920	9	0.07*	8.13*	1.03	635	Conifer forest
43	Zongxiong Co	33.1032	80.1610	4351	6	0.15	9.34	-1.49	66	desert
44	Sumxi Co	34.5997	80.2496	5057	31	0.26	8.53	-5.86	44	desert
45	Longmu Co	34.5882	80.3677	5010	106	174*	7.8*	-5.61	44	desert
46	Zhacang Chaka	32.5494	82.4297	4354	19	211*	8*	2.16	153	steppe
47	Darebu Co	32.4625	83.2161	4438	26	1.39	9.35	1.16	110	steppe
48	Qige Co	31.1957	85.5173	4667	20	0.18	10.46	-2.38	175	steppe
49	Cuo-e-1	31.6655	88.7011	4568	268	0.21	8.84	0.11	241	steppe
50	Chen Co	28.9580	90.5174	4436	39	0.75	8.62	2.22	394	steppe
51	Tuosu Lake	37.1265	96.9176	2808	151	23.2	8.84	5.27	131	desert
52	Hurleg Lake	37.2868	96.8864	2817	55	0.66	8.49	5.39	131	desert
53	Dagze Co	31.8917	87.5235	4470	311	14.69	9.80	0.07	219	steppe
54	Jiang Co	31.5433	90.8179	4603	40	14.1	9.29	-1.71	372	steppe
55	Gongzhu Co	30.6436	82.1090	4789	56	4.95	9.20	-3.06	197	steppe

Notes: Source of data: 1) elevation: Google Earth Pro; 2) lake area: GaoFen-1 satellite, more details in Wan et al. (2016); 3) salinity and pH, in situ measurements; 4) temperature and precipitation: China meteorological forcing dataset (1979–2018); 5) vegetation zone: Hou (2001), vegetation zone in italic (Dr. Feng Qin, personal communication). An asterisk(*) is used to indicate the data from published papers, including Wang and Dou, (1998), Zhao et al. (2018), Liu et al. (2011), Ju et al. (2015), NIA=No Information Available.

column (Restek Rtx-1, 60 m × 0.25 mm id, 0.25 μm film thickness) for separation and then compared to an external standard (11 homologous series of *n*-alkanes from C₁₆ to C₃₇, CPAchem). The concentration of *n*-alkanes was calculated using the calibration curves of an external standard. The GC oven temperature program was as follows: initial temperature at 60°C, hold for 2 min, then ramp at 30°C/min to 120°C, and then ramp at 5°C/min to 330°C, hold at 330°C for 15 min.

In the present study, the ACL was calculated following Poynter et al. (1989), who first defined the metric ACL for the range of *n*-alkanes from C₂₃ to C₃₃. Later, this was refined by Poynter and Eglinton (1990) to target higher plants by employing only long-chain, odd *n*-alkanes. In the equation, C_{*i*} = the peak area of *n*-alkanes containing *i* carbon atoms. We calculated the *n*-alkane ACL with different carbon ranges. Freeman and Pancost (2014) proposed that it was necessary to define the ACL with the molecule range in subscripts to avoid confusion; hence, it is important to carefully choose the carbon numbers for the calculations, which is necessary for assessing the impact of vegetation and climate on the *n*-alkane chain length. The other well-established *n*-alkane proxy, the Carbon Preference Index (CPI), was also calculated. In this study, we calculated the CPI values using the formula in Cooper and Bray (1963). ACL and CPI calculations with *n*-alkanes range from C₂₇ to C₃₃ are given below as examples:

$$ACL_{27-33} = \frac{(27 \times C_{27} + 29 \times C_{29} + 31 \times C_{31} + 33 \times C_{33})}{C_{27} + C_{29} + C_{31} + C_{33}}, \quad (1)$$

$$CPI_{27-33} = \frac{1}{2} \left(\frac{C_{27} + C_{29} + C_{31} + C_{33}}{C_{26} + C_{28} + C_{30} + C_{32}} + \frac{C_{27} + C_{29} + C_{31} + C_{33}}{C_{28} + C_{30} + C_{32} + C_{34}} \right). \quad (2)$$

2.3 Climate estimates

Due to the sparse and inhomogeneous distribution of weather stations across the QTP, the meteorological observation data are limited, particularly for western Tibet. In this study, climate variables, including annual precipitation and temperature, were obtained from gridded climate data sets, namely the China Meteorological Forcing Data set (CMFD) (1979–2018) (He et al., 2020). The extracted temperature data represent the average value of a grid cell (with the spatial resolution of 0.1°), which requires calibration since there is an elevation difference between the location of the lake and the grid cell (Dr. Jie He, personal communication). The CMFD was developed by the Institute of Tibetan Plateau Research, Chinese Academy of Sciences, and the database has been widely utilized for climate studies throughout the QTP (Qiao et al., 2019; Wang et al.,

2020). Recently, Wu et al. (2019b) compared three precipitation products for the QTP and concluded that the CMFD performed better than the others; hence, the CMFD product is the preferred gridded product for assessing climate data in this region.

2.4 Vegetation types

The dominant vegetation zones (biomes) were extracted from the Vegetation Atlas of China (Hou, 2001). In some cases, it is difficult to characterize the relatively complex vegetation cover in this region, which comprises diverse vegetation zones. Hence, GIS-based automated extractions and the literatures were both considered. The typical vegetation types in the catchments of the surveyed lakes include steppe, meadow and desert (Fig. 1(d)). We also extracted vegetation information in terms of photosynthetic composition, and the distribution of C₄ vegetation was provided from the ISLSCP II (International Satellite Land Surface Climatology Project) global data sets (Still et al., 2003, 2009) (Fig. 1(e)).

3 Results

3.1 Concentration of total lipid extracts

The total lipid extracts of the lake surface sediments, normalized to dry weight, generally vary between 1 mg/g and 15 mg/g with the average of 3.96 mg/g (Fig. 2). No significant differences were observed among lakes with contrasting salinity levels. The two highest TLE concentrations of the investigated lakes are Yanapeng Co (a mesosaline lake) and Darebu Co (a subsaline lake), with the values of 14.66 mg/g and 14.93 mg/g, respectively. Two lakes situated in western Kunlun (Sumxi Co and Guozha Co) and one proglacial lake (Ranwu Lake with three sub-basins, two of them are RW-AY and RW-AMC) in south-east Tibet had much lower TLE concentration (< 1 mg/g).

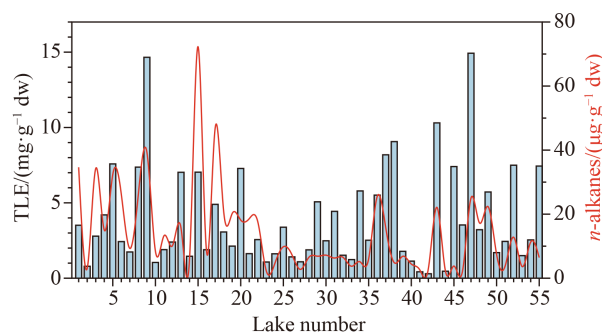


Fig. 2 The concentrations of Total Lipid Extracts (TLEs) and *n*-alkanes from C₂₁ to C₃₅ (TLE in mg/g d.w., ΣALK₂₁₋₃₅ in μg/g d.w.). The x-axis labels are consistent with the numbers in Table 1.

3.2 Distribution and concentration of *n*-alkanes

The surface sediments from the 55 lakes contain homologous series of *n*-alkanes between C₂₁ and C₃₇, albeit the homologs with the longest chain lengths (C₃₆ and C₃₇) are present at very low concentrations. Most of the samples showed strong odd/even predominance (CPI₂₇₋₃₃ values of these samples were generally > 5), except for the sample from Guozha Co (CPI₂₇₋₃₃ = 3.06). Marked differences in the *n*-alkanes distributions are evident, and four representative GC-FID chromatograms (Type I, Type II, Type III, and Type IV) are displayed in Fig. 3. Type I and Type II have a unimodal distribution with the dominant peaks C₂₉/C₃₁ and C₂₃/C₂₅, respectively. Type III is characterized by a bimodal distribution with maxima at C₂₃/C₂₅ and C₂₉/C₃₁. A distinctive *n*-alkane distribution pattern was observed for Dasugan Lake (Type IV), where the peak heights of the C₂₃, C₂₅, C₂₇, and C₂₉ *n*-alkanes were almost identical. The sum of the *n*-alkane concentrations (from C₂₁ to C₃₅) of the lake surface sediments varied significantly among the lakes, ranging from 2.11 μg/g to 72.26 μg/g (normalized to dry weight) with the average of 14.10 μg/g (Fig. 2). The summed concentrations of *n*-alkanes generally show similar spatial variations with the TLEs (Fig. 2). We calculated the *n*-alkane ACL values based on long-chain *n*-alkanes, but with different carbon ranges. For instance, ACL₂₇₋₃₁, ACL₂₇₋₃₃, and ACL₂₉₋₃₃ yielded the average values of 29.38 ± 0.37, 29.87 ± 0.38, and 30.57 ± 0.20, respectively.

4 Discussion

4.1 Sources of the long-chain *n*-alkanes

It is imperative to determine the specific sources of *n*-

alkanes in sediments before using them as proxies for the reconstruction of paleovegetation and paleoclimate. There are two primary factors to be considered to elucidate the sources of recent sedimentary *n*-alkanes. One is the relative importance of aeolian and fluvial transport processes (McDuffee et al., 2004), and the other is the different biological sources of long-chain *n*-alkanes and their contributions.

4.1.1 Aeolian versus fluvial transport

The leaf wax lipids are easily abraded from the plant leaf surface, and these lipids could be transported in air and deposited on the lake surface (Feakins et al., 2005). In addition, the aeolian transport of leaf wax lipids could occur via the erosion and redeposition of soil (Poynter et al., 1989; Kusch et al., 2010). Previous studies have demonstrated that aeolian dusts are the principal source of biological lipids of terrigenous materials in open ocean settings (Simoneit et al., 1977; Gagosian and Peltzer, 1986; Schefuß et al., 2003) and marginal seas (Wakeham, 1996; Yokoyama et al., 2006; Kusch et al., 2010). However, unlike the marine environment, the contribution of windblown *n*-alkanes to sediments could be quite different among lakes. Although several reviews have shown that the aeolian component was typically a small fraction of the total organic matter in lake sediments (Meyers and Ishiwatari, 1995; Meyers, 1997), several case studies have demonstrated that atmospheric deposition was an important mechanism for transporting waxes (Doskey, 2000; Hockun et al., 2016). Currently, there are few published data to evaluate the contribution of sedimentary *n*-alkanes for the lakes on the QTP. Here, we infer that the transport mechanism of the long-chain *n*-alkanes of the surveyed lakes is mainly associated with fluvial processes, via two mechanisms. 1) Lake ice prevents the atmospheric input of *n*-alkanes to the lake

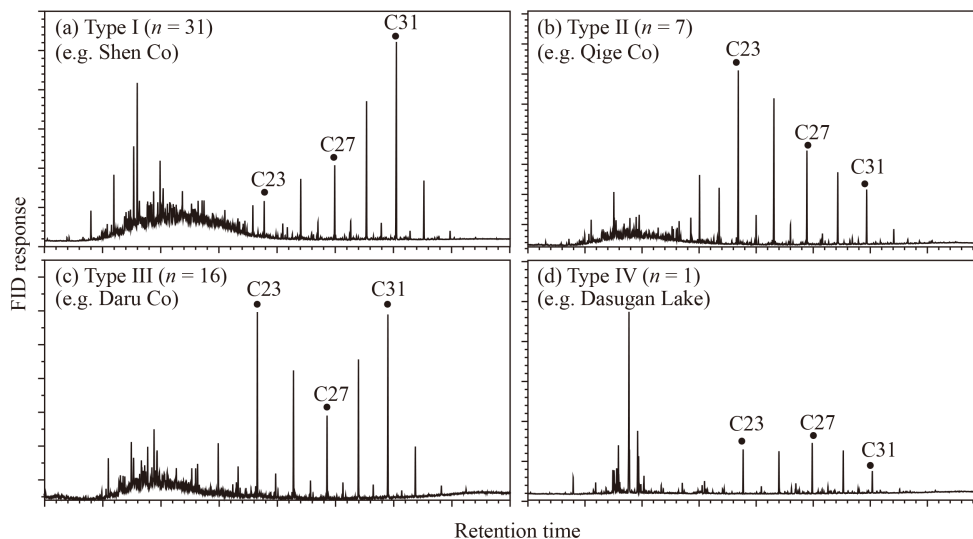


Fig. 3 The four types of *n*-alkane distribution among the studied QTP lakes.

sediments during the windiest period of the year. Modern observations have shown strong winds prevail over the QTP from December to April, and the concentration of *n*-alkanes in total suspended particles is the highest at this time (Chen et al., 2014). However, ice phenology data revealed that a period of lake ice cover for most QTP lakes is from winter to spring (Wang et al., 2020); hence, the wind-blown *n*-alkane contribution to these lakes is probably minor. 2) Material sources available for transport during the ice-free season are not abundant. Atmospheric total suspended particle data in central Tibet and the south-eastern QTP showed that the lowest concentration of *n*-alkanes was in summer (Gong et al., 2011; Chen et al., 2014; Zhu et al., 2020). This evidence supports the assumption that contribution of airborne material to sedimentary *n*-alkanes is generally minor. However, we note that aeolian transport processes differ between lakes (Nelson et al., 2018), and further investigations are needed to quantitatively disentangle the organic matter contributions from different sources and transport mechanisms.

4.1.2 Biological sources of long-chain *n*-alkanes

The chain length of the *n*-alkane homologs is usually from C₂₁ to C₃₇ in leaf waxes (Eglinton and Hamilton, 1967). ACL was first defined by Poynter et al. (1989), with the range from C₂₃ to C₃₃ *n*-alkanes, and it was later revised to C₂₇ to C₃₁ since this range is characteristic of terrestrial higher plants (Poynter and Eglinton, 1990). Recent studies have shown that submerged aquatic plants can also contribute to the sedimentary long chain *n*-alkanes (e.g., C₂₇ and C₂₉) of QTP lakes, particularly in the littoral zone (Aichner et al., 2010; Liu et al., 2015; Liu and Liu, 2016). We conducted a literature survey to determine if this was commonly the case for other lakes across the QTP. We addressed this issue by comparing the trends of variation in leaf-wax *n*-alkane δD values (*n*-C₂₇, *n*-C₂₉, and *n*-C₃₁) from a paleolimnological perspective. Although we found that the isotopic trends in *n*-C₂₇ were basically the same as in *n*-C₂₉ and *n*-C₃₁ over time (Bird et al., 2014; Rao et al., 2014; Günther et al., 2016), the correlation coefficients were only moderately high (*n*-C₂₇ vs. *n*-C₃₁, $R^2 = 0.49\text{--}0.58$), reflecting the mixed source of the C₂₇ *n*-alkane. In addition, we examined the potential sources of the C₂₇ *n*-alkane by applying principal component analysis to our data set. From Fig. 4 it is evident that the compound classes of *n*-alkanes are generally grouped into three clusters: Cluster 1 (C₂₁, C₂₃, C₂₅), Cluster 2 (C₂₇), and Cluster 3 (C₂₉, C₃₁, C₃₃, C₃₅). Therefore, the C₂₇ *n*-alkane likely has a mixed signal from terrestrial and aquatic plants, in agreement with previous studies.

4.2 Evaluation of vegetation influences on ACL

Previous studies have demonstrated that ACL responds to

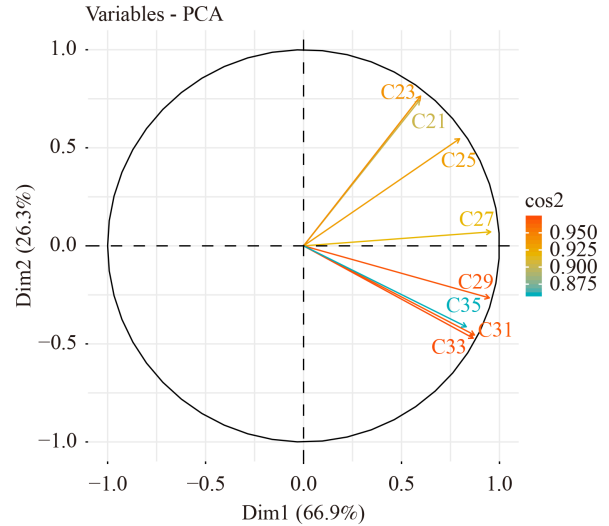


Fig. 4 Results of principal components analysis (PCA). High \cos^2 indicates the strong loading of that variable on a given principal component, and vice versa.

variations in plant type (Vogts et al., 2009; Badewien et al., 2015) or climatic conditions (Castañeda et al., 2009b) in different environments; hence, caution is needed before interpreting the ACL data. Here, we aim to evaluate the influence of vegetation on the chain length of long-chain *n*-alkanes, and two types of information on plants were retrieved for reference: 1) life-forms such as trees, shrubs and grasses, and 2) different photosynthetic types such as C₃ and C₄ plants.

4.2.1 Evaluation of vegetation influences on ACL from a plant life-form perspective

Many studies have shown that the ACL of *n*-alkanes varies between different plant types; for example, samples from grassland sites show systematically higher ACL values than those from deciduous trees/shrubs (Rommerskirchen et al., 2006; Vogts et al., 2009; Duan and He, 2011; Bliedtner et al., 2018). However, a meta-analysis of modern plants revealed no significant differences in chain length between grasses and woody plants (Bush and McInerney, 2013). Our previous study supported the conclusion of Bush and McInerney (2013) and concluded that the ACL could not distinguish graminoids from shrubs in some cases (Wang et al., 2015). For example, there were large overlaps between shrubs and grasses around Ranwu Lake in the south-eastern QTP, and similar observations are reported for other regions, like Australia (Howard et al., 2018) and Canada (Hollister et al., 2022).

For the sites in this study, the biome distribution was extracted from Vegetation Map of China (Hou, 2001). In addition, to extract plant life-form information, we also referred to the literature for verification (Wang and Dou, 1998; Qin, 2021). The dominant vegetation types for all

our sites are steppe, meadow and desert (Table 1). Trees are very sparsely distributed on the QTP due to the low temperatures and semi-dry to dry environment at high altitudes, resulting in a prevailing graminoid community with a few small shrubs in the lake catchments. Considering the large amount of overlap of the *n*-alkane chain length between grasses and shrubs, while grasses and shrubs were dominant compared to trees at the studied sites, the changes in ACL are unlikely to be caused by a shift in vegetation type, for example, from grasses to shrubs.

4.2.2 Evaluation of vegetation influences on ACL from a photosynthetic perspective

Chain length, as well as carbon isotope composition, could distinguish C_3 from C_4 leaf wax *n*-alkanes, supported by the results obtained from modern plants (Rommerskirchen et al., 2006; Vogts et al., 2009), and hence ACL is deemed to be a good indicator of the spatiotemporal evolution of continental vegetation between the C_3 and C_4 photosynthetic pathways. Accordingly, it is widely used for paleovegetation reconstruction combined with compound specific stable carbon isotope analysis (Sarkar et al., 2015; Jalali et al., 2017).

Model simulations with a global distribution of C_4 mapping have shown that the modern C_4 plant species are very scarce at the study sites (Fig. 1(e)) (Still et al., 2003). Only a few field studies have reported that C_4 plants can survive in high-elevation environments (Wang, 2003; Wang et al., 2004). The scarcity of C_4 plants could be explained by the low temperature conditions in this high-elevation environment, regardless of precipitation (Rao et al., 2010). Therefore, the negligible distribution of C_4 plants at our study sites simplifies the interpretation of ACL from a photosynthetic perspective.

4.3 Evaluation of climatic influences on ACL

Plants can adapt to environmental stress by means of various physiologic mechanisms (Shepherd and Wynne Griffiths, 2006). Although ACL has been widely used as a paleoclimate proxy, its interpretations are ambiguous and region-specific. Transect studies have demonstrated the response of *n*-alkane ACL values to changes in hydrology (Hoffmann et al., 2013; Eley and Hren, 2018) and/or temperature (Tipple and Pagani, 2013; Bush and McInerney, 2015; Wang et al., 2018a). Accordingly, there are different interpretations of long-chain *n*-alkane ACL records. In paleolimnological studies in the QTP, ACL has been used to reconstruct past variations in effective humidity (Ling et al., 2017b; Zhang et al., 2017) or temperature (Pu et al., 2013; Ling et al., 2017a). Therefore, proxy calibrations in this region are needed to clarify its climatic implications and to reconcile the

discrepancies between existing *n*-alkane ACL records.

The lakes surveyed in this study span a wide range of precipitation and temperature (Fig. 1), enabling us to assess the impact of these two climatic variables. Previous studies have suggested that precipitation and temperature usually exert opposing influences on the chain length of *n*-alkanes. A shift to longer chain homologs was found to be accompanied by lower precipitation (Carr et al., 2014; Eley and Hren, 2018) or higher temperatures (Leider et al., 2013; Tipple and Pagani, 2013; Bush and McInerney, 2015; Wang et al., 2018a). Our *n*-alkane data show that ACL is positively correlated with precipitation but it shows no relationship with temperature (Fig. 5), which is consistent with previous research on the QTP (Hu et al., 2014; Ling et al., 2021). However, such a positive correlation is contrary to the generally accepted understanding that land plants tend to synthesize longer *n*-alkane homologs in drier climatic conditions (Dodd and Afzal-Raffi, 2000; Dodd and Poveda, 2003). Hence, we now attempt to examine each of these two factors and to explain the observed relationships.

4.3.1 Relationship between ACL and temperature

The proposed physiologic mechanism of the response of chain length to temperature is that plants increase the chain length to maintain their crystalline structure (Carr et al., 2014), or the so-called “hardness” of the leaf surface (Kawamura et al., 2003). The rationale behind is that compounds with longer carbon chains have slightly higher melting points, and their relative abundance in plants may be sufficient to maintain molecular alignment in the wax crystals under warmer condition (Gaines et al., 2009). Although numerous soil- and plant-based approaches have observed a close relationship between temperature and ACL (Tipple and Pagani, 2013; Bush and McInerney, 2015; Wang et al., 2018a, 2018b), no significant correlations were found for our data set (Fig. 5). To date, few studies have reported a positive response of *n*-alkane ACL values to temperature changes across the QTP, as evidenced by studies of modern plants and soils (Guo et al., 2015; Jia et al., 2016). Since all the samples in the present study are lake sediments, we ascribe this lack of a relationship between ACL and temperature to the complex processes of *n*-alkanes deposition in lakes. We also examined the temperature values extracted from CMFD and compared them with those of other studies. We found that the temperatures in our study were much lower and the gradient much smaller. For example, the temperature range was 20.2°C in Bush and McInerney (2015), which is almost twice as large as in our study. Hence the observed results could be partially explained by the temperature gradient, since the regression model showed a much greater scatter over a narrow temperature range (Bush and McInerney, 2015).

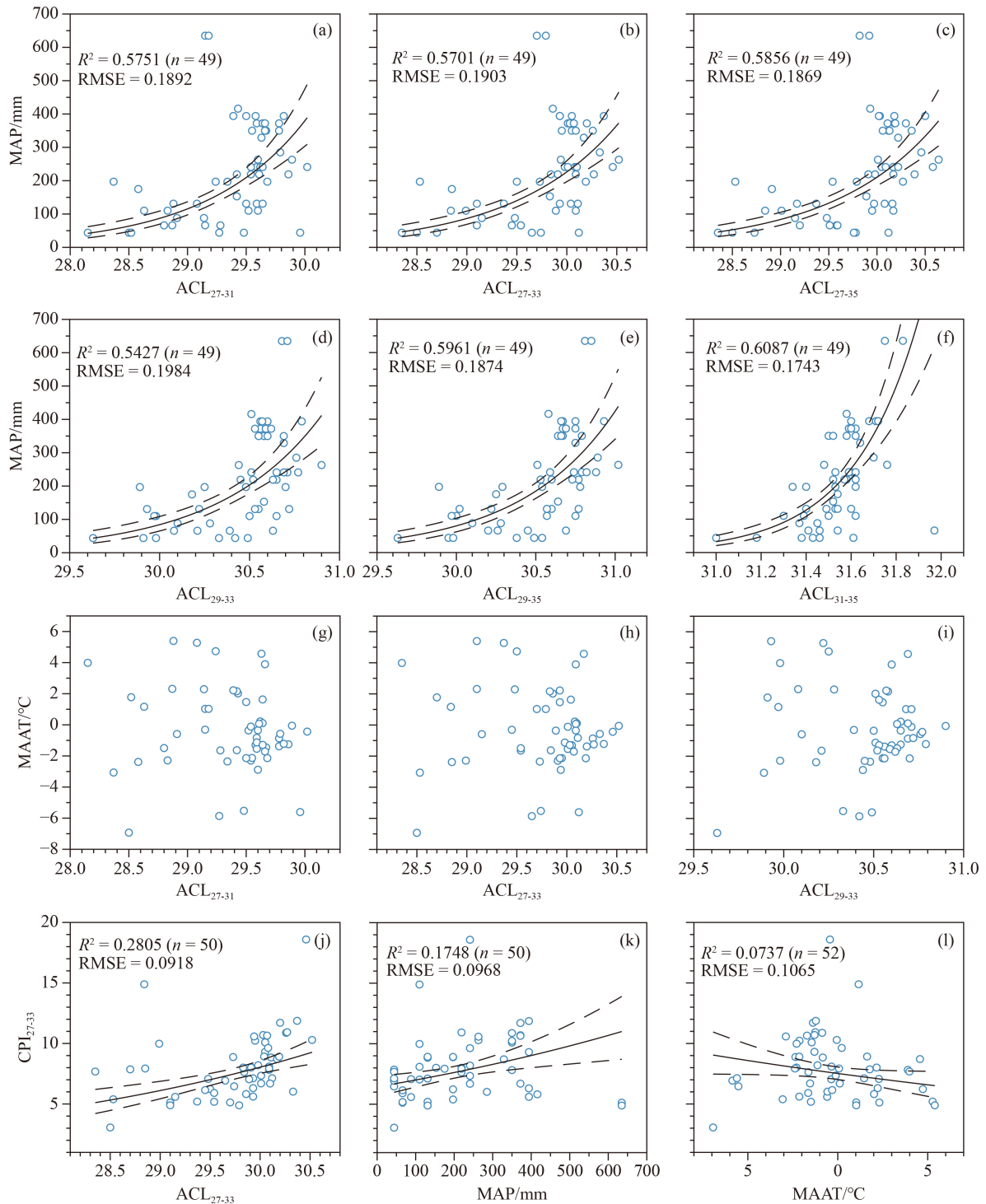


Fig. 5 Results of exponential regression analysis, showing significant relationships between ACL, CPI, and climatic variables. Outliers were removed before the statistical analyses were performed.

On the other hand, precipitation at our surveyed sites ranges from 44 to 635 mm, while the temperature range is relatively small, reducing the effects of temperature on the long-chain *n*-alkane distributions. A similar interpretation of other biomarkers, including alkenones, was made by Liu et al. (2011) for the lakes across the QTP.

4.3.2 Relationship between ACL and precipitation

Water stress has been shown to induce the production of alkanes with longer chain lengths in leaf wax (Bondada et al., 1996), and that the chain length decreases with increasing precipitation. In our database, ACL is positively correlated with precipitation (Fig. 5), which

contradicts the physiologic stress hypothesis. We speculate that such a relationship is the result of the distinct response of the plants to hydrological changes. Our hypothesis is that plants on the QTP produce higher ACLs in wetter environments. However, in a few cases the data from different plant species have shown a positive but statistically insignificant relationship (Guo et al., 2015). Additionally, to the best of our knowledge, only one case study observed a similar correlation: the plant, *Eucalyptus*, in northern Australia, showed an increase in ACL under wetter conditions (Hoffmann et al., 2013). Clearly, there is a substantial difference in vegetation types between the QTP and Australia, and thus the plants at our sites are unlikely to show a similar response. The cause of the relationship between ACL and the hydrological conditions remains unclear, and next we attempt to examine the potential causes of the observed relationship between ACL and precipitation.

First, we speculate that the cause of a positive relationship between ACL and precipitation is due to the direct impact of hydrological changes on the ACL. A relatively high proportion of long-chain *n*-alkanes from terrestrial higher plants transported to lakes may be supplied by surface runoff. As precipitation increases, the enhanced runoff would cause a greater contribution from land plants to the sedimentary long-chain *n*-alkane pools. Liu and Liu (2016) considered that the contribution of long chain *n*-alkanes to lake sediments from certain aquatic plants may be significant. Aquatic plants have longer than expected chain lengths; for example, some submerged plants contain high concentrations of C₂₇ and C₂₉ *n*-alkanes. Our PCA results support the previous finding that the C₂₇ and C₂₉ *n*-alkanes are of mixed origin (Fig. 4). Nevertheless, *n*-alkane chain length distributions are generally different between terrestrial plants and aquatic plants, and therefore higher ACL values reflect a greater contribution from terrestrial plants over aquatic plants. To test this assumption, we compared the correlations between precipitation and ACL with different carbon number ranges, and we observed a similar robust regression relationship for the longer *n*-alkanes (e.g., ACL₃₁₋₃₅) (Fig. 5). Therefore, it is unlikely that the cause of the positive correlation between ACL and precipitation is due to the relative contributions of terrestrial and aquatic plants driven by hydrological changes.

Second, we infer that microbial degradation of long-chain *n*-alkanes could account for the unexpected response. Previous investigations found that alkane-degrading microorganisms are widespread in natural environments, and Rojo (2009) pointed out that a typical soil contained significant amounts of hydrocarbon-degrading microorganisms. Alkane-degrading bacteria were also abundant for lake sediments, such as those across the QTP (Wang et al., 2019). Previous findings demonstrated that leaf waxes could potentially undergo

microbial degradation during deposition (Zech et al., 2011; Li et al., 2017; Wu et al., 2019a); hence, selective degradation could modify the distributions of long-chain *n*-alkanes during deposition in soils and sediments. We compared the relationships between ACL and precipitation in different types of samples from the early research on the QTP, including for modern plants (Guo et al., 2015), soils (Guo et al., 2015; Jia et al., 2016), and lake sediments (Hu et al., 2014; Ling et al., 2021). We found that the responses of ACL to precipitation were similar in these samples, all showing positive correlations. However, Guo et al. (2015) found that relationship was statistically insignificant for plants and soils, which can be ascribed to the specific habitats for plant growth, such as alpine meadow on the QTP (Dr. Yanjun Guo, personal communication). We infer that the positive correlation between ACL and precipitation may be the result of soil microbial activity. The moisture level is one of the factors that determine the rate of microbial decomposition of organic compounds in soils (Šepič et al., 1995). A higher soil water content caused by higher precipitation may greatly affect soil chemical processes and soil microbial activity, degrading shorter *n*-alkanes in long-chain homologs and leading to higher ACLs. This assumption is supported by the CPI data, which is positively correlated with precipitation (Fig. 5). Overall, we conclude that microbial degradation may play a major role in determining this relationship, but further investigations are needed to elucidate this process.

4.4 Implications for lake sediment-based paleoclimate reconstruction across the QTP

ACL changes are considered to reflect climate variables only if the vegetation types remain unchanged during the study period (Hoffmann et al., 2013; Pu et al., 2013). A substantial body of research demonstrates that the vegetation is a major determinant of the chain length of leaf waxes (Vogts et al., 2009); hence, it is imperative to consider the vegetation history over the interval of interest before using the long-chain *n*-alkane ACL as a paleoclimate indicator. Our results reveal that ACL mainly reflects regional climate signals rather than vegetation dynamics. On the other hand, paleovegetation reconstructions have shown that a forest biome was almost absent in most parts of the QTP since the Last Glacial Maximum (Ni et al., 2014; Li et al., 2019; Qin et al., 2022). A spatiotemporal reconstruction of C₄ plants also showed that their distribution was less than 10% on the QTP over the past 21000 years (Jiang et al., 2019). Considering the results of the present study and past vegetation reconstructions, we regard the ACL as a proxy for precipitation across most of the QTP, where trees and C₄ plants are largely absent.

5 Conclusions

We report the distribution of *n*-alkanes extracted from the surface sediments of 55 lakes across the Qinghai-Tibet Plateau. The results show the ACL of the long-chain *n*-alkanes is more responsive to precipitation rather than to temperature in this region. We also found a significant positive relationship between ACL and precipitation, and thus our data support the conclusion that precipitation is the main driver of the variations in ACL. Precipitation changes may affect soil moisture levels and thereby affect microbial degradation. Our results provide new insights into the interpretation of the chain length variability of *n*-alkanes in the lake sediments of the QTP, and they have implications for the use of the *n*-alkane ACL in paleolimnological studies in other high-elevation environments.

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