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Distribution of GDGTs in lake surface sediments on the Tibetan Plateau and its influencing factors

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Abstract Quantitative paleotemperature records are vital not only for verifying and improving the accuracy of climate model simulations, but also for estimating the amplitude of temperature variability under global warming scenarios. The Tibetan Plateau (TP) affects atmospheric circulation patterns due to its unique geographical location and high elevation, and studies of the mechanisms of climate change on the TP are potentially extremely valuable for understanding the relationship of the region with the global climate system. With the development of biomarker-based proxies, it is possible to use lake sediments to quantitatively reconstruct past temperature variability. The source of Glycerol Dialkyl Glycerol Tetraethers (GDGTs) in lake sediments is complex, and their distribution is controlled by both climatic and environmental factors. In this work, we sampled the surface sediments of 27 lakes on the TP and in addition obtained surface soil samples from six of the lake catchments. We analyzed the factors that influence GDGT distribution in the lake sediments, and established quantitative relationship between GDGTs and Mean Annual Air Temperature (MAAT). Our principal findings are as follows: the majority of GDGTs in the lake sediments are bGDGTs, followed by crenarchaeol and GDGT-0. In most of the lakes there were no significant differences between the GDGT distribution within the lake sediments and the soils in the same catchment, which indicates that the contribution of terrestrial material is important. iGDGTs in lake sediments are mainly influenced by water chemistry parameters (pH and salinity), and that in small lakes on the TP, TEX_{86} may act as a potential proxy for lake pH; however, in contrast bGDGTs in the lake sediments are mainly controlled by climatic factors. Based on the GDGT distribution in the lake sediments, we used proxies (MBT, CBT) and the fractional abundance of bGDGTs (f_{abun}) to establish calibrations between GDGTs and MAAT, respectively, which potentially provide the basis for paleoclimatic reconstruction on the TP.

Keywords **Tibetan Plateau, Lake sediment, Soil, GDGT calibration, Quantitative reconstruction**

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1. Introduction

The uplift of the Tibetan Plateau has had a significant influence on the climate of Asia since the Cenozoic (Raymo and Ruddiman, 1992; Kutzbach et al., 1993; An et al., 2001), not only in terms of changing the pattern of atmospheric circulation, but also in relation to the history of monsoon activity and aridification in central Asia (Liu et al., 1998). Consequently, the climatic record of the TP has received increasing attention (Liu and Chen, 2000; Yao et al., 2012). In recent years, improved understanding of mod-

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ern climate change on the TP has been achieved based on precipitation isotope monitoring programs and on climate model simulations (Tian et al., 2001, 2007; Yao et al., 2013). However, the past climatic regime of the TP remains unclear due to the limited number of high-quality paleoclimate records. Therefore, it is important to obtain an increased number of quantitative paleoclimate reconstructions in order to achieve a comprehensive understanding of the climatic effects of changing atmospheric circulation on the TP on different time scales (Benn and Owen, 1998; Schiemann et al., 2009; An et al., 2012; Mölg et al., 2014; Zhu et al., 2015). Moreover, recent studies have revealed significant differences in Holocene temperature variations inferred from paleoclimate records and in the results of different climate simulations (Marcott et al., 2013; Liu Z Y et al., 2014). Therefore additional paleoclimate reconstructions are also likely to improve our understanding of the mechanisms of Holocene temperature changes in the region.

For lake sediments, common climate proxies used to quantitatively reconstruct past climate include pollen assemblages (Herzschuh et al., 2006, 2009; Lu et al., 2011; Leipe et al., 2014), chironomid assemblages (Chen et al., 2009), trace elements (Zhang et al., 2004) and varve thickness (Liu X Q et al., 2014). Over the last decade, biomarker-based proxy indicators have become increasingly important for quantitative climatic reconstruction, and examples include long-chain alkenones (Wang and Zheng, 1997; Liu et al., 2006; He et al., 2013; Zhao et al., 2013; Li et al., 2015) and GDGTs (Wu et al., 2013; Günther et al., 2015; Wang et al., 2015). However, to date few case studies have used GDGTs to reconstruct climate change on the TP and furthermore the existing records are typically beset with problems such as discontinuous and large regression errors. One of the main reasons may be the lack of suitable calibration functions for TP lake sediments. Efforts have been made to establish calibrations between GDGTs and temperature, either based on lake surface sediments (Blaga et al., 2009; Powers et al., 2010; Tierney et al., 2010; Zink et al., 2010; Pearson et al., 2011; Sun et al., 2011; Loomis et al., 2012; Günther et al., 2014) or soils (Weijers et al., 2007; Peterse et al., 2012; Yang et al., 2014a; Ding et al., 2015). However, the reconstructed results are significantly different and in some cases the Root Mean Square Error (RMSE) is quite large. For example, Wu et al. (2013) found that reconstructed MAAT was significantly higher than measured in Kusai Lake; Günther et al. (2015) reconstructed MAAT and Lake Surface Temperature (LST) at Nam Co and concluded that the records could only reflect the trend instead of absolute values; and due to the lack of a regional calibration, Wang et al. (2015) were constrained to use a global calibration to reconstruct July LST (JLST) at Lake Qinghai. These studies demonstrate that it is necessary to establish regional GDGT calibrations that are suitable for TP lakes in order to improve the reliability of the paleoenvironmental reconstructions. This is especially so because the TP has distinctive characteristics such as high elevation, low MAAT and significant spatial differences in precipitation; moreover, the majority of the TP lakes are inland. Therefore, it is necessary to study the modern relationships between GDGTs and climatic/environmental variables of TP lakes before performing quantitative reconstructions.

In this paper, we present the results of analyses of GDGT distributions in lake sediments and soils from sites on the TP; use statistical methods to determine the influences of climatic and environmental factors on the GDGT distributions; and attempt to establish calibrations using GDGT-based proxies (MBT, CBT) and fractional abundance of bGDGTs (f_{abun}), respectively, to provide a basis for quantitative temperature reconstructions for the region.

2. Materials and methods

2.1 Sample collection

2.1.1 Lake sediments

Surface sediments from 27 lakes were collected along a west-east transect across the TP and Qaidam Basin (Figure 1, Table 1). The main vegetation types along the transect included alpine meadow, alpine steppe and temperate steppe (Hou, 2001). In order to investigate the influence of different climatic/environmental factors on GDGTs (GDGT structures see Figure 2), the investigated lakes span a large environmental gradient. The lake areas range from 12 km^2 (Long Co) to 1920 km^2 (Nam Co); the altitudes range from 2798 m (Sugan Lake) to 4718 m (Nam Co); the ranges of MAAT and Mean Annual Precipitation (MAP) are −4–6°C and 41–678 mm, respectively; salinity ranges from freshwater (Ranwu Lake, 0.07 g/L) to saline (Dong Co, 46.25 g/L); and pH ranges from 8.13 to 10.00. In the summer of 2012 and 2013, we collected surface sediment samples (0–1 cm) from the lakes 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 and extraction of the lipids.

2.1.2 Surface soils

In order to assess the source of the GDGTs in the lake sediments, we selected ten surface soil samples from six lake catchments, including Bangong Co, Nam Co, Angrenjin Co, Qiagui Co, Dong Co and Bamu Co (Figure 3, Table 2) and compared the GDGT content with that of lake sediments from the same catchment. In the case of Bangong Co, we collected five soil samples from the main inflowing rivers, Makha and Chiao Ho (Figure 3a). The six lake catchments from which soil samples were obtained represent a large environmental gradient: MAAT ranges from −4.0°C to 4.2°C; lake area ranges from 24 km^2 (Angrenjin Co) to 1920 km^2 (Nam Co); and the types of lake include both freshwater (Bangong Co, salinity 0.47 g/L) and saline

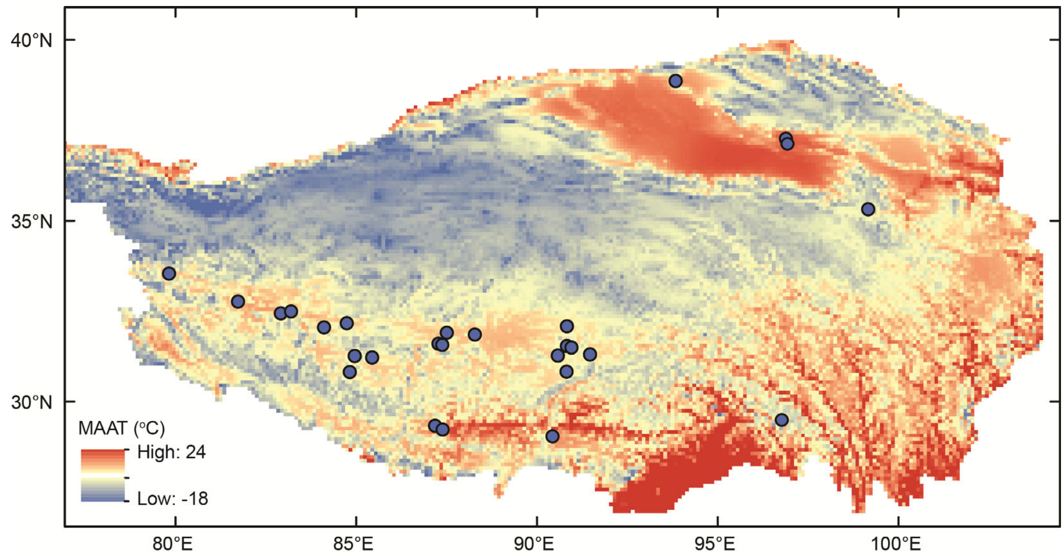

Figure 1 Map of surveyed lake surface sediments on the Tibetan Plateau (base map was generated by China Meteorological Forcing Dataset, 1979–2012).

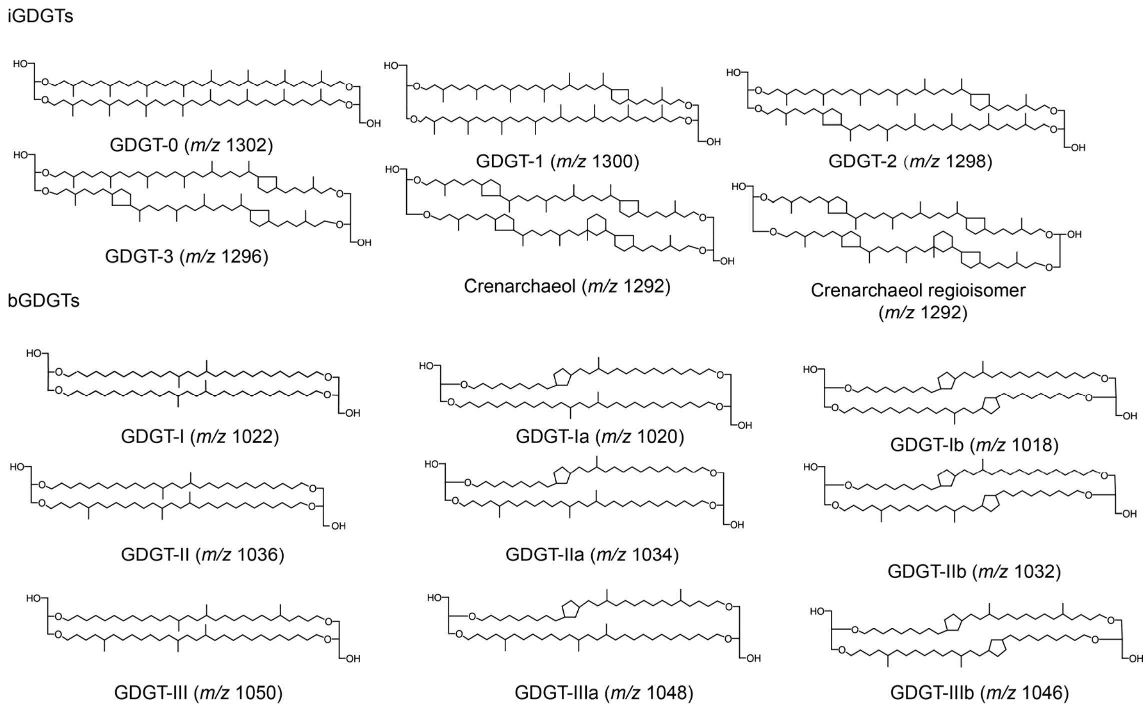

Figure 2 Structure of GDGT compounds (from Schouten et al., 2013).

(Dong Co, salinity 46.25 g/L). The soil samples were processed in a similar manner to the lake sediments. After freeze-drying, the samples were sieved and ground prior to organic pretreatment (Section 2.2).

2.2 Sample preparation and GDGT analysis

2.2.1 Organic pretreatment

In order to obtain the total lipid extract, the freeze-dried sample (5–6 g) were extracted by ultrasonic extraction with dichloromethane/methanol (9∶1, *v*/*v*) (15 minutes, 3×, 30°C). Activated alumina was used as a stationary phase for separating GDGTs and the non-polar and polar fractions (the latter containing GDGTs) were eluted using *n*-hexane/dichloromethane (9∶1, *v*/*v*) and dichloromethane/methanol (1∶1, *v*/*v*), respectively. Blow down the polar fraction by N_2, the sample was dissolved in *n*-hexane/isopropanol (99∶1, *v*/*v*) and a 0.2 μm PTFE filter was used to remove large molecular compounds and particulate matter prior to HPLC-MS analysis.

Table 1 Geographical and limnological data for the 27 TP lakes [a)]

Lake No.	Lake name	Latitude (N)	Longitude (E)	Altitude (m)	Lake area (km^2)	Water depth (m)	SLST (°C)	Salinity (g/L)	pH	Dissolved oxygen (mg/L)	Date or data source	MAAT (°C)	MAP (mm)
1	Aweng Co	32.75°	81.74°	4434	59	6	16.15	27.65	9.20	4.94	2012-08-05	1.20	167
2	Anggu Co	31.20°	85.45°	4665	23	13	15.11	1.89	9.10	5.87	2012-08-13	–2.00	294
3	Angrenjin Co	29.31°	87.19°	4305	24	13	19.50	5.26	9.64	6.56	2012-07-23	4.20	348
4	Bamu Co	31.25°	90.58°	4565	180	48	14.04	7.51	9.70	6.00	2012-08-21	–1.00	430
5	Bangong Co	33.53°	79.83°	4244	604	38	15.60	0.47	8.73	6.40	2012-07-28	–1.00	121
6	Bieruoze Co	32.42°	82.92°	4413	33	2	15.67	27.38	8.97	4.60	2012-08-06	1.30	190
7	Darebu Co	32.47°	83.20°	4441	21	3	16.23	1.39	9.35	6.41	2012-08-06	1.10	186
8	Dawa Co	31.25°	84.97°	4599	114	2	16.08	18.58	9.30	5.73	2012-08-11	0.00	301
9	Dagze Co	31.89°	87.52°	4470	245	34	15.31	14.69	9.80	5.42	2012-08-18	0.95	422
10	Dong Co	32.15°	84.75°	4400	88	2	16.29	46.25	8.82	4.64	2012-08-08	–3.00	259
11	Gemang Co	31.58°	87.28°	4610	52	44	14.30	6.35	9.73	5.92	2012-08-17	–1.00	297
12	Jiang Co	31.52°	90.83°	4603	36	22	12.50	14.10	9.29	5.98	2013-06-30	0.00	488
13	Hurleg Lake	37.27°	96.89°	2840	57	6	17.60	0.66	8.49	7.09	2013-06-15	5.00	212
14	Kongmu Co	29.01°	90.44°	4451	40	17	14.60	0.23	8.55	6.24	2013-07-06	2.00	363
15	Kuhai	35.30°	99.17°	4132	49	10	8.00	16.10	8.82	6.51	2013-06-11	6.00	341
16	Lagor Co	32.03°	84.12°	4471	91	18	16.00	40.27	8.94	4.71	2012-08-08	0.70	182
17	Long Co	29.20°	87.39°	4295	12	25	16.70	1.58	9.44	6.02	2012-07-23	1.00	333
18	Nam Co	30.81°	90.82°	4718	1920	90	11.43	1.17	9.21	8.90	Literature *	–1.00	435
19	Nairiping Co	31.29°	91.47°	4529	70	8	14.20	7.96	9.98	5.71	2013-07-02	0.50	463
20	Peng Co	31.48°	90.96°	4569	136	8	11.70	8.54	9.91	6.29	2013-06-29	0.50	486
21	Qiagui Co	31.84°	88.29°	4645	91	26	14.44	0.22	8.83	6.29	2012-08-19	–4.00	322
22	Ranwu Lake	29.47°	96.78°	4005	22	20	12.03	0.07	8.13	6.44	Literature **	2.00	678
23	Sugan Lake	38.86°	93.85°	2798	120	15	17.00	20.00	8.90	6.46	2013-06-17	2.00	41
24	Tuosu Lake	37.12°	96.94°	2808	240	12	16.00	23.20	8.84	6.38	2013-06-15	5.00	241
25	Youbu Co	30.80°	84.83°	4645	64	33	14.23	16.09	9.62	5.56	2012-08-12	–1.00	359
26	Zhangnai Co	31.55°	87.39°	4611	36	16	14.71	4.06	9.60	5.54	2012-08-13	0.50	302
27	Zigetang Co	32.06°	90.84°	4575	191	15	13.50	13.50	10.00	5.87	2013-06-27	0.00	509

a) * Nam Co: Wang et al. (2009); ** Ranwu Lake: Ju et al. (2015).

Table 2 Geographical data for soil samples

Sample ID	Latitude (N)	Longitude (E)	Altitude (m)	Catchment	Sampling year
BH-2	33.44°	79.77°	4259	Bangong Co	2010
BH-3	33.45°	79.82°	4262	Bangong Co	2010
BH-12	33.56°	79.89°	4256	Bangong Co	2010
BH-24-1	33.63°	79.78°	4279	Bangong Co	2010
BH-24-2	33.62°	79.81°	4279	Bangong Co	2010
TPS14-17	29.24°	87.22°	4513	Angrenjin Co	2014
TPS14-90	32.18°	84.62°	4480	Dong Co	2014
TPS14-97	31.86°	88.32°	4574	Qiagui Co	2014
TPS14-105	31.14°	90.55°	4582	Bamu Co	2014
TPS14-107	30.74°	91.09°	4816	Nam Co	2014

2.2.2 GDGT analysis

GDGT analysis was performed using HPLC-APCI-MS (Agilent 1200 HPLC system with 6100 MS) and the compounds were separated using normal phase chromatography (Grace Prevail Cyano, 150 mm× 2.1 mm, 3 μm). The instrumental setups for HPLC were as follows: column temperature 40°C, injection volume 30 μL, flow rate 0.2 mL/min. For the mobile phase, A is *n*-hexane, D is *n*-hexane/isopropanol (9:1, *v*/*v*), and the elution procedure followed Yang et al. (2014b). APCI-MS conditions were as follows: nebulizer pressure 60 psi, vaporizer temperature 400°C, drying gas flow rate 6 L/min and temperature 200°C, capillary voltage 3500 V, corona 5 μA. The analysis was performed in Single Ion Monitoring (SIM) mode via $[M+H]^+$ of GDGTs (1302, 1300, 1298, 1296, 1292, 1050, 1048, 1046, 1036, 1034, 1032, 1022, 1020, 1018). GDGTs were determined using an external standard and peak areas were manually integrated, assuming an identical response factor for GDGTs. The definition of the structure and number of GDGTs follows Schouten et al. (2013) (Figure 2),

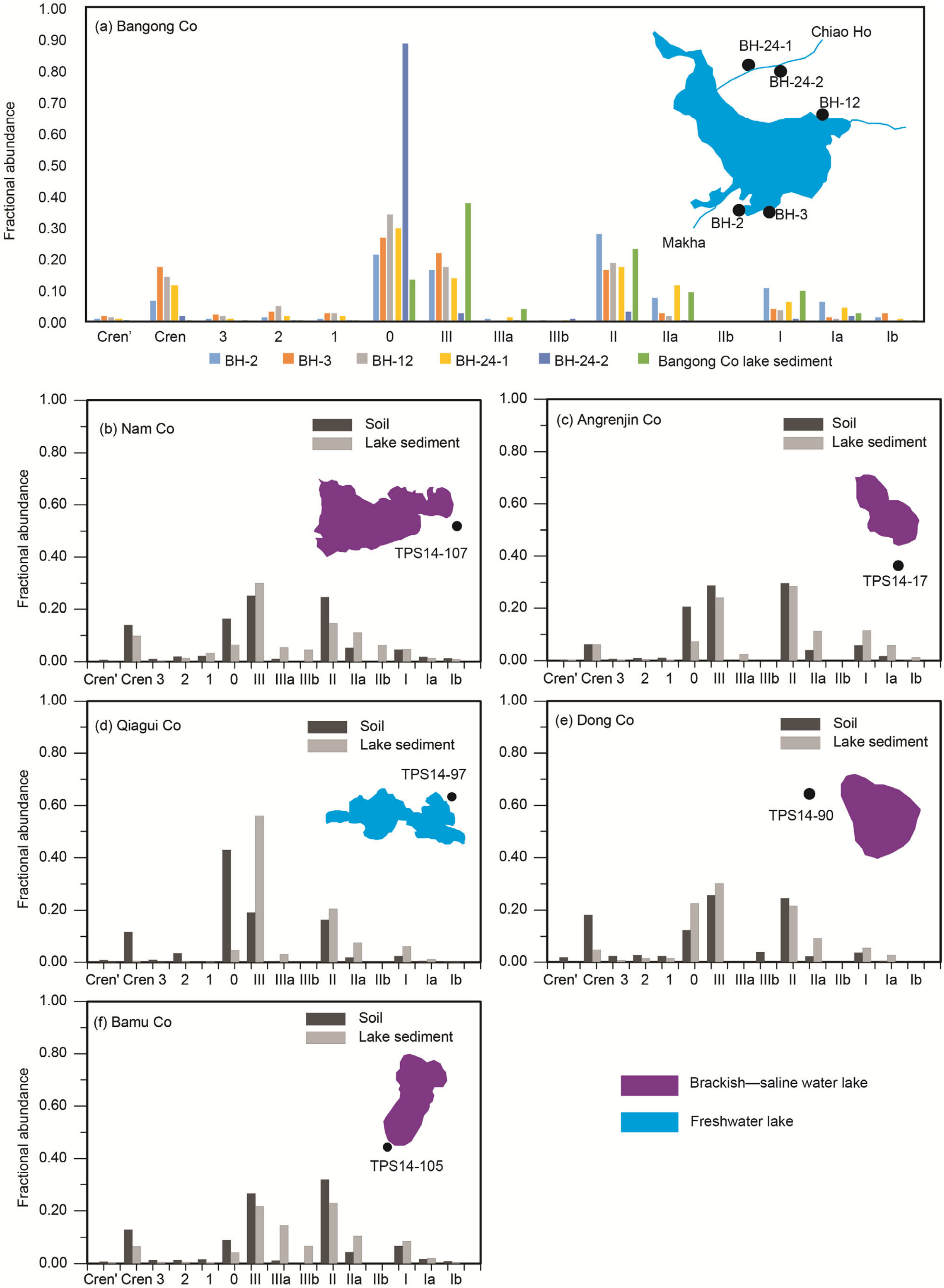

Figure 3 Comparison of GDGT distribution between lake surface sediments and soils from the same catchment.

and the definition of GDGT-based proxies such as TEX_{86}, MBT and CBT follows the original papers (Schouten et al., 2002; Weijers et al., 2007). In the present work, the vast majority of lake sediments contained GDGT-IIIa and GDGT-IIIb and therefore we used MBT instead of MBT' (Peterse et al., 2012).

2.3 Environmental data sources

2.3.1 Meteorological data

Meteorological data were obtained from China Meteorological Forcing Dataset (CMFD) (He and Yang, 2011). The temporal and spatial resolution of data is 3-hr and 0.1°, respectively. We extracted meteorological data corresponding to the locations of the 27 lakes (1979–2012). Temperature data were also obtained from the China Meteorological Data Sharing Service System (http://cdc.nmic.cn) and from the literature (Yang et al., 2013).

2.3.2 Lake water chemistry data

Lake water chemistry data were mainly obtained from *in situ* measurements made during the 2012–2013 field trips. Depth profiles of water temperature, salinity, pH and dissolved oxygen content were obtained by YSI (Table 1). Data for Nam Co and Ranwu Lake were obtained from the literatures (Wang et al., 2009; Ju et al., 2015).

2.4 Statistical methods

Principal components analysis (PCA) and redundancy analysis (RDA) were performed to analyze the relationships between GDGTs and climatic/environmental factors, using R with the Vegan package (Oksanen et al., 2015). All of the data were normalized prior to ordination. For RDA analysis, only five bGDGT compounds (GDGT-III, GDGT-II, GDGT-I, GDGT-Ia and GDGT-Ib) were included due to their high abundance. A Monte Carlo test (999 unrestricted permutations) was performed to determine whether the results were statistically significant (P<0.05). Multiple linear regression was performed using MATLAB (2013b, Mathworks, Natick, MA) to establish the calibrations between GDGTs and MAAT.

3. Results

3.1 GDGT distribution

3.1.1 Lake sediments

bGDGTs dominated all of the lake sediment samples (except for Kuhai where they comprised 10%), followed by GDGT-0 and crenarchaeol (Figure 4). Crenarchaeol and its regio-isomer cren' were detected in 80% of the investigated lakes. By comparison, the GDGT distribution of the TP lakes is similar to that of lakes in polar regions (Shanahan et al., 2013) and Japan (Ajioka et al., 2014), but differs from that of lakes in Africa (Damsté et al., 2009) and Europe (Blaga et al., 2009).

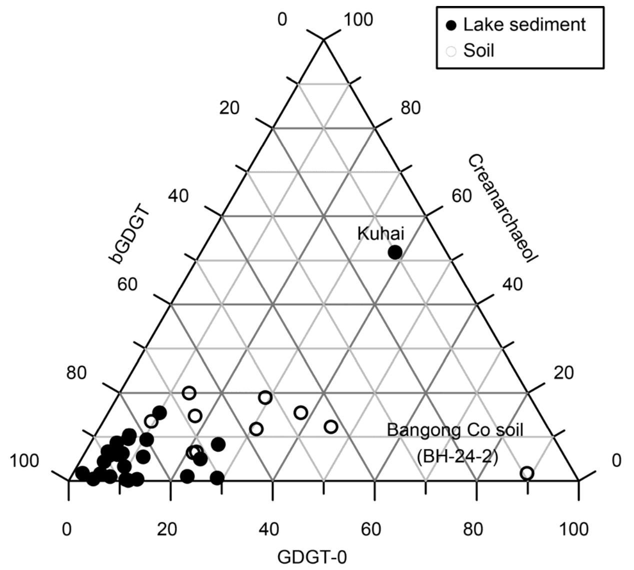

Figure 4 Ternary plot of GDGT distribution in Tibetan Plateau lake sediments.

3.1.2 Soils

In the eastern part of Bangong Co, five soil samples were collected and the abundance of iGDGTs and bGDGTs was similar in four. At all five sites, the GDGT distribution in the soils differed from that of the lake sediments (Figure 3(a)). The distribution for sample BH-24-2 was significantly different from that of the other four samples: the abundance of GDGT-0 was nearly 90%, much higher than for the other GDGTs. The abundance of iGDGTs ranged from 27% to 60% in the five catchments (Figure 3b–f). It is obvious that significantly different GDGT distributions occur in the soil and lake sediment samples for Qiagui Co (Figure 3d).

3.2 GDGT adaptive response to climate and environment change

The PCA results indicate that the first two principal components (PC1 and PC2) account for 29.72% and 26.16% of the total variance, respectively (Figure 5a). The PC1 axis represents MAP, Summer LST (SLST), the salinity of the lake surface water and dissolved oxygen content. The PC2 axis mainly reflects the pH of the lake surface water and MAAT. The PCA ordination diagram effectively distinguishes 27 lakes based on climatic conditions or lake water chemistry parameters. For example, MAAT is relatively high (e.g. at Sugan Lake and Tuosu Lake) in the second quadrant; there is no freshwater lake in the third quadrant; and the climate is relatively humid in the fourth quadrant (Jiang Co and Zigetang Co). In addition, the PCA diagram also clarifies the relationship between GDGT-based proxies and climatic/environmental factors. Since the PC1 axis ex-

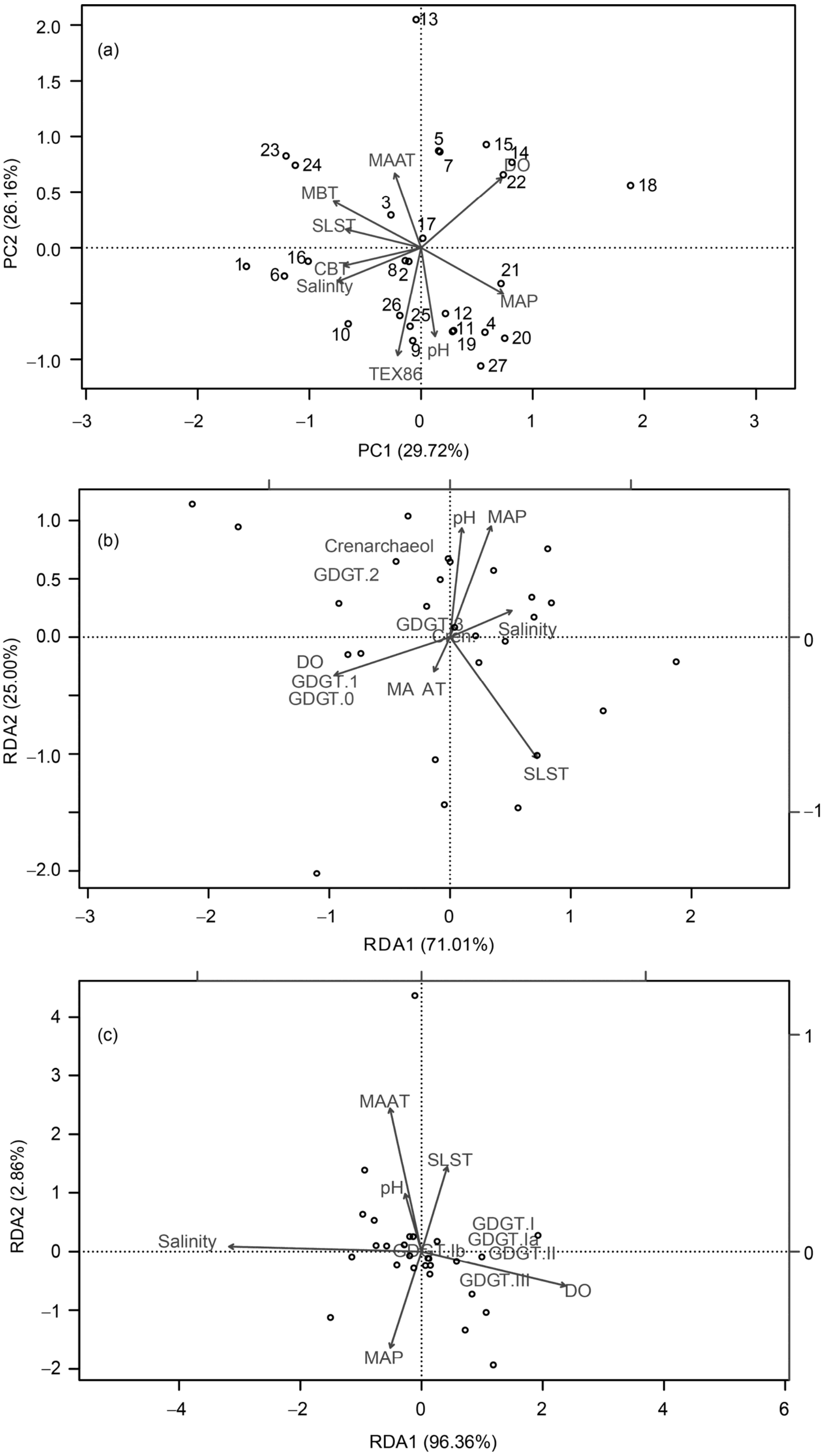

Figure 5 Results of principal components analysis (PCA) and redundancy analysis (RDA) of GDGTs in Tibetan Plateau lake sediments. (a) PCA biplot; (b) RDA triplot of iGDGTs; (c) RDA triplot of bGDGTs.

plains the variations of MBT and CBT, which are influenced by climatic factors, the PC2 axis may represent the variations of TEX_{86} which are affected by lake water pH and MAAT.

In the results of the RDA of the iGDGTs, the RDA1 and RDA2 axes account for 71.01% and 25.00% of the total variance, respectively (Figure 5b). The RDA1 axis reflects dissolved oxygen content of the lake water and SLST, and the RDA2 axis mainly reflects pH of the lake water and MAP. The number of cyclopentyl rings increases at high salinity. Most of the iGDGTs are influenced by SLST except for GDGT-0 and GDGT-1. The results of a Monte Carlo permutation test indicates that the impact of climatic factors on iGDGTs is significant (P=0.002). In the results of the RDA of the bGDGTs, the RDA1 and RDA2 axes account for 96.36% and 2.86% of the total variance, respectively (Figure 5c). The RDA1 axis primarily reflects variations in dissolved oxygen and salinity, and the RDA2 axis mainly reflects temperature and MAP. The abundance of different bGDGTs changes along the RDA2 axis: as temperature increases, the number of methyl groups decreases. The results of a Monte Carlo permutation test indicate that the influence of climatic factors on bGDGTs is also significant (P=0.001).

3.3 Calibrations between GDGTs and MAAT

Two calibrations (eqs. (1) and (2), below) were established based on GDGT-based proxies (MBT, CBT), or on the fractional abundance of bGDGTs (f_{abun}), with MAAT, which would be suitable for paleoclimatic reconstruction using lake sediments of the TP (Figure 6). The results of multiple regression analysis indicated that the correlation coefficient (R) was greater than 0.8 for both calibrations.

$$\text{MAAT}=-3.75+40.92\times\text{MBT}-6.03\times\text{CBT}$$
$$(n=27,\ R=0.80,\ \text{RMSE}=1.47°\text{C},\ P<0.01). \quad (1)$$

$$\text{MAAT}=-0.55-0.17\times f(\text{III})-28.39\times f(\text{II})+43.51\times f(\text{I})+115.50\times f(\text{Ib})$$
$$(n=27,\ R=0.82,\ \text{RMSE}=1.46°\text{C},\ P<0.01). \quad (2)$$

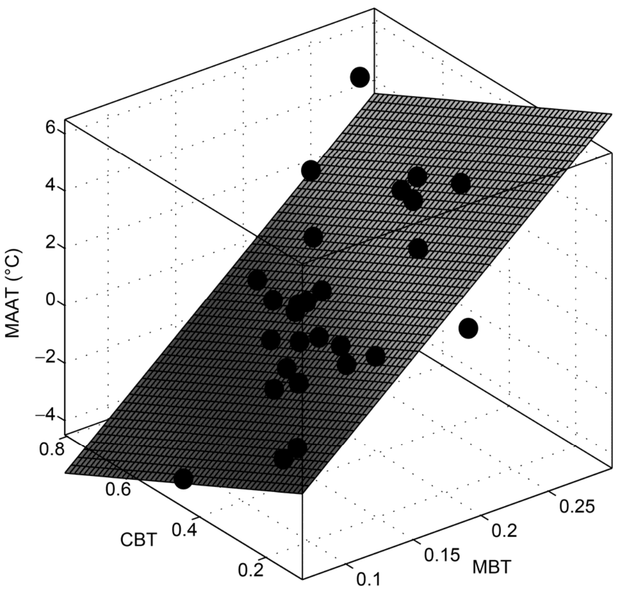

Figure 6 Results of multiple linear regression between MBT, CBT and MAAT.

4. Discussion

4.1 Source of GDGTs

Early studies suggested that iGDGTs were mainly produced by Thaumarchaeota (formerly known as Crenarchaeota) (Powers et al., 2004), while soil bacteria produced bGDGTs (Weijers et al., 2006a, 2007, 2009). However, an increasing number of studies have demonstrated that bGDGTs can also be produced *in situ*, such as in a lake environment (Damsté et al., 2009; Tierney and Russell, 2009; Blaga et al., 2010; Loomis et al., 2011, 2014b; Naeher et al., 2014; Colcord et al., 2015; Huguet et al., 2015); and that the contribution of terrestrial input of iGDGTs should not be ignored (Tierney et al., 2012; Wang et al., 2012). Therefore, it is necessary to investigate the sources of GDGTs before attempting a quantitative paleoclimatic reconstruction.

In the present study, we were unable to estimate the contribution of bGDGTs from the lake environment due to the lack of data for suspended particulate organic matter. However, the GDGT distributions within the soil samples support the likelihood that iGDGTs are in part derived from terrestrial inputs (Figures 3 and 4). Crenarchaeol and its isomer were detected in all of the soil samples and in the majority of the lake sediment samples. In the Bangong Co catchment, the proportion of iGDGTs in five soil samples was higher than in the lake sediments, which indicates that the contribution of terrestrial sources of iGDGTs is potentially significant.

It is noteworthy that the GDGT distribution of two adjacent samples is totally different: in sample BH-24-2 the proportion of GDGT-0 is almost 90%, while in sample BH-24-1 it is 30% (Figure 4a). The reason for this difference is unclear, although it is noteworthy that the location of sample BH-24-2 was closer to a village; however, whether or not this factor is significant requires further investigation. For the five other catchments, although the number of samples was limited, the results still suggest that the contribution of terrestrial sources of iGDGTs is important. For example, the proportion of iGDGTs in soil was greater than that of bGDGTs in the Qiagui Co catchment (Figure 4d). However, except for Bangong Co and Qiagui Co, there are no obvious differences between the lake sediment and soil samples (Figures 4b, 4c, 4e, 4f), which implies that the source of GDGTs in different lakes is complex.

4.2 Influence of climate on GDGT distribution

4.2.1 iGDGTs

The results of ordination and regression analysis indicate that lake water pH, salinity and MAAT are the main factors affecting iGDGT distribution (Figures 5a, 5b, 7b, 7c) and the significance of these factors is considered below.

(1) Lake water pH. The regression analysis results

demonstrate that the iGDGT distribution in the lake sediments is influenced by water pH (R=0.69) (Figure 7b). Previous studies focused on the importance of pH for the distribution of bGDGTs, e.g. soil CBT vs. soil pH (Weijers et al., 2007; Peterse et al., 2010, 2012) and lake sediment CBT vs. lake water pH (Sun et al., 2011; Schoon et al., 2013; Günther et al., 2014; Loomis et al., 2014a). However, there has been little consideration of the influence of lake water pH on iGDGTs. Although Pearson et al. (2008) found that pH could affect iGDGT distribution in extreme environments, such as hot springs, little is known of the effects of pH on iGDGT distribution in low temperature environments. Lake water pH is related to weathering of bedrock, plankton biomass and dissolved oxygen content, while the terrestrial inputs is also influenced by intensity of weathering to some extent. Hence, we speculate there exists inter-relationship between lake water pH and terrestrial inputs. Although the mechanism is unclear, in the present study the influence of pH on iGDGTs is significant (P<0.01), and if Nam Co is excluded the correlation coefficient increases to 0.72. Therefore, we suggest that TEX_{86} may be a potential proxy indicator of lake water pH in the case of small lakes on the TP. We interpret the observed good correlation between TEX_{86} and pH as follows: (1) lake water pH may affect the iGDGT distribution; and (2) soil pH may also influence iGDGTs before they are transported to the lake. The latter factor has been discussed in the literatures (Weijers et al., 2006b; Yang et al., 2012, 2014a).

(2) Salinity. No significant correlation is observed between lake water salinity and TEX_{86} (Figure 7c); however, in the case of salinity values greater than 2 g/L, a negative correlation is evident (Figure 7c, R=0.52, n=18, P<0.05). We speculate that Thaumarchaeota may respond to salinity variations only within a certain range: when salinity <2 g/L, its impact is limited, but if greater than 2 g/L, salinity variations may be important. However, our observations differ from the results of a culture experiment by Wuchter et al. (2004). The latter considered that the effect of salinity on GDGT distribution could be ignored; however, the salinity range used in their experiment was only 27‰–35‰, which is only appropriate for simulating the marine environment. The investigated lakes in the present study exhibit a much greater salinity range (4.06‰–46.25‰), and therefore it may unsurprising that salinity emerges as an important factor.

(3) Lake Surface Temperature (LST). Early studies suggested that TEX_{86} was an effective paleothermometer in lake environments (Powers et al., 2004, 2010; Blaga et al., 2009). However, no significant correlation between TEX_{86} and SLST is observed in the present study (Figure 7a). This may be the result of the fact that the investigated TP lakes are relatively small (<300 km^2), except for Nam Co and Bangong Co (Table 1), since an increased importance of terrestrial organic matter inputs would result in a poor

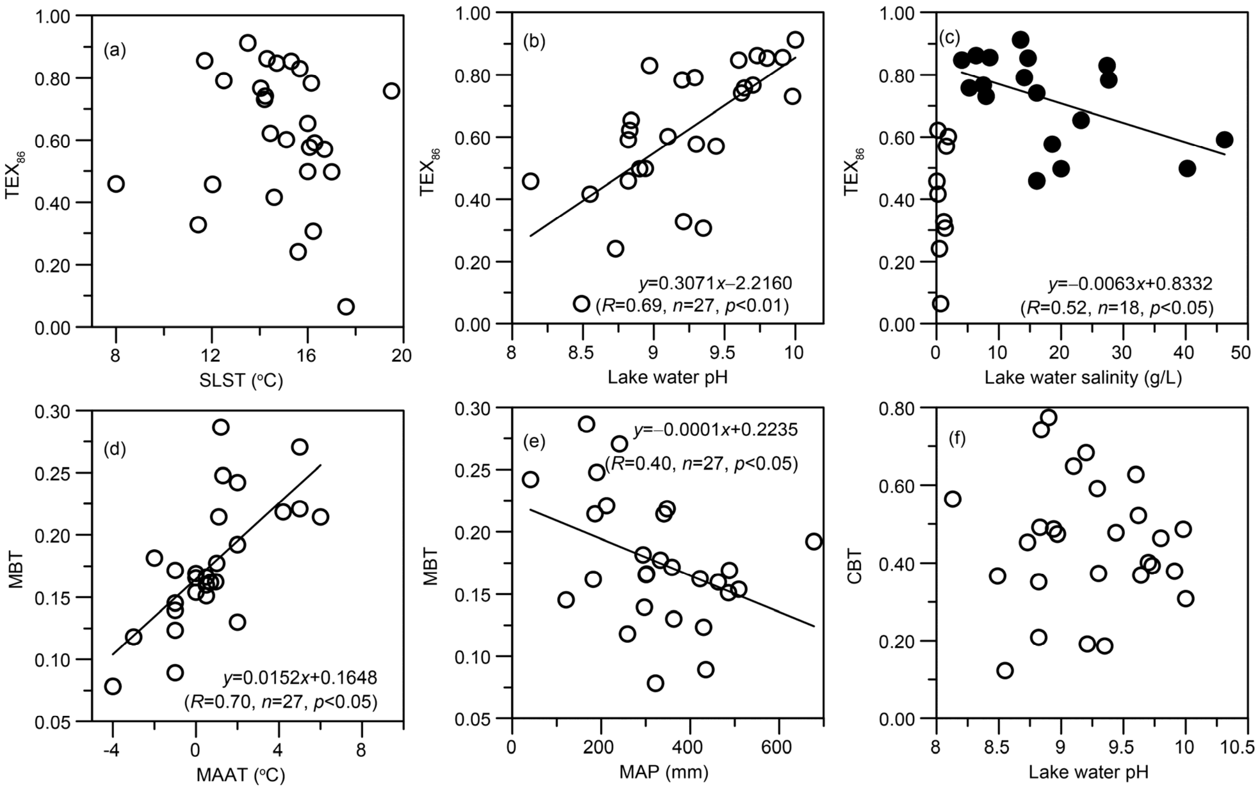

Figure 7 Results of regression analysis between GDGT-based proxies and climatic/environmental factors.

correlation (Weijers et al., 2006b). Therefore, we conclude that that TEX_{86} is not an effective paleothermometer for the small lakes of the TP.

In order to assess the influence of seasonal variations in lake water temperature on iGDGT distribution, Blaga et al. (2009) investigated GDGTs in European lake sediments and found that TEX_{86} correlated well with both Annual LST (ALST) and Winter LST (WLST), especially the latter. This may indicate higher production by iGDGTs producers in winter. In the case of the TP, there is a lack of continuous lake water temperature monitoring for the vast majority of lakes, and therefore we are unable to consider the impact of seasonal variations in lake water temperature on the iGDGT distribution. We suggest that it is important to carry out a comprehensive program of lake water temperature monitoring on the TP in order to improve understanding of the relationship between temperature and iGDGTs (Wang M D et al., 2014).

In addition to the importance of terrestrial inputs and lake water temperature, it is necessary to improve our understanding of the environmental preferences of Thaumarchaeota, such as its preferred water depth. Thus a program of sediment trap monitoring would help us to understand the relationship between GDGT producers and environmental parameters (such as nutrient levels), and to further clarify the environmental significance of TEX_{86} (Woltering et al., 2012).

(4) Mean Annual Air Temperature (MAAT). The PCA results demonstrate a negative correlation between TEX_{86} and MAAT (Figure 5a); however, the mechanisms are unclear due to the lack of related culture experiments. We suggest the following factors may be significant: (i) the influences on iGDGT distribution in terrestrial and aquatic environments may be different; since MAAT was relatively low in the TP (especially the western TP), temperature may have a greater influence on GDGT producers in terrestrial environments in contrast to nutrients or pH in aquatic environments. (ii) the importance of terrestrial inputs in the present study is obvious since most of the surveyed lakes were less than 250 km^2 in area, and this is also supported by the results of the comparison of the soil and lake sediments in the same catchment.

4.2.2 bGDGTs

The ordination and regression analysis results indicate that MAAT and MAP are the major factors which influence the bGDGT distribution in the lake sediments (Figures 5a, 5c, 7d, 7e) and these two factors, together with lake water pH and 6-methyl bGDGTs, are discussed below.

(1) MAAT. MBT is positively correlated with MAAT, which is similar to the results of a previous study (Zink et al., 2010); however, the correlation coefficient is slightly lower in our study (R=0.70) (Figure 7d). We speculate that the possible reasons may include: (i) the fact that the source of GDGTs in TP lakes is complex; and (ii) that in addition to temperature, other factors (such as MAP or water pH) may also affect the bGDGT distribution.

(2) MAP. A weak negative correlation is observed between MBT and MAP (R=0.40) (Figure 7e). Since temperature and precipitation change simultaneously over most of the TP in inter-annual time scale, the possibility that precipitation influences GDGT distribution via temperature (Weijers et al., 2006b) cannot be excluded. Several recent studies have indicated that precipitation may affect bGDGT distribution in certain regions (e.g. the Chinese Loess Plateau (CLP) and the TP) (Xie et al., 2012; Günther et al., 2014; Wang H Y et al., 2014). For example, in a study of surface soils on the CLP, Wang H Y et al. (2014) concluded that precipitation and soil water content were the main factors controlling the bGDGT distribution of alkaline soils in arid and semi-humid regions. Thus, we speculate that in the case of TP, especially in the arid part of the western TP, the bGDGT distribution may also be affected by precipitation.

(3) Lake water pH. Previous studies have found that CBT is well correlated with soil pH (Weijers et al., 2006b) or with lake water pH (Günther et al., 2014; Loomis et al., 2014a); however, the relationship between CBT and lake water pH is debated (Günther et al., 2014; Loomis et al., 2014a). Loomis et al. (2014a) integrated published GDGT data from East African lake sediments and corresponding lake water pH data (Tierney et al., 2010; Loomis et al., 2012) and found a negative correlation between CBT and lake water pH. However, in contrast, Günther et al. (2014) found that that CBT was positively correlated with water pH in a survey of TP lakes. Our results are inconsistent with both of these studies since we observed no correlation (Figure 7f). Previous studies have observed a weak correlation in the case of alkaline soils (pH>7) (Wang H Y et al., 2014; Yang et al., 2014a). In the present study, all 27 lakes were alkaline (pH 8.1~10.0), and therefore the high pH values of the lakes may be one of the reasons.

(4) 6-methyl bGDGTs. Analysis of the GDGT distribution in different geological archives (e.g. loess, peat and marine sediments) reveals a series of bGDGT isomers (Zech et al., 2012; Becker et al., 2013; De Jonge et al., 2013). According to the position of the methyl group, bGDGTs are classified into 5-methyl and 6-methyl bGDGTs. With the development of chromatographic separation techniques, 5-methyl and 6-methyl bGDGTs can be separated effectively to some extent (De Jonge et al., 2014a, 2014b, 2015; Ding et al., 2015; Weber et al., 2015; Yang et al., 2015). This may improve our understanding of the complex relationship between bGDGTs and environmental factors and provide the basis for revision of the calibrations which may in turn improve the accuracy of paleoclimatic reconstructions.

De Jonge et al. (2014b) analyzed bGDGTs in suspended particles from the Yenisei River and found that the abundance of 6-methyl bGDGTs was high. However, there was a large deviation between reconstructed results and *in situ* measurement data, and it was suggested that bGDGTs in particulate organic matter were mainly produced *in situ* and that existing GDGT calibrations need to be recalibrated. Based on this conclusion, De Jonge et al. (2014a) revisited the global soil database by Peterse et al. (2012) and used a modified HPLC method for analysis of bGDGTs. The results revealed that the fractional abundance of 6-methyl bGDGTs correlated well with soil pH while 5-methyl bGDGTs were mainly influenced by MAAT. This finding was also supported by the results of analysis of soil samples collected on a local scale (Ding et al., 2015; Yang et al., 2015). Although it remains unclear how soil pH controls the distribution of 6-methyl bGDGTs, two factors may provide an explanation of the relationship between pH and bGDGTs: (1) bacteria producing bGDGTs adapt to changes in pH by cyclization (Weijers et al., 2007); and (2) bacteria producing bGDGTs adapt to changes in pH by changing the position of the methyl functional group (Ding et al., 2015).

4.3 Applicability of GDGT calibrations for TP lakes

It is necessary to establish calibrations between proxies and climatic/environmental factors before undertaking a paleoclimate reconstruction. The application of a soil-based calibration in a lacustrine environment depends on the assumption that bGDGTs in lake sediments are exclusively derived from terrestrial inputs (Peterse et al., 2014). However, an increasing number of studies have found that bGDGTs are also produced *in situ* (Damsté et al., 2009; Tierney and Russell, 2009; Loomis et al., 2011; Weber et al., 2015) and therefore temperature would be underestimated based on the application of soil-based calibrations (De Jonge et al., 2014b). In the following discussion, all of the selected calibrations were established based on lake sediments.

GDGT calibrations can be classified into two groups according to the type of GDGTs: (1) for iGDGTs, calibrations are established between TEX_{86} and LST (Powers et al., 2005, 2010); and (2) for bGDGTs, calibrations are established using multiple linear regression between MAAT and bGDGT-based proxies (MBT, CBT) or fractional abundance of bGDGTs (Tierney et al., 2010; Sun et al., 2011; Loomis et al., 2012; Günther et al., 2014).

In order to verify the applicability of GDGT calibrations for the TP lakes, including previously published calibrations as well as the new ones developed in the present study, we used Bangong Co as an example. The reasons are as follows: (1) meteorological data are available from China Meteorological Administration (CMA) and Ngari Station for Desert Environment Observation and Research, Chinese Academy of Sciences (NASDE); and (2) continuous lake water temperature monitoring has been carried out (Wang M D et al., 2014), together with *in situ* YSI temperature observations. Based on the bGDGT distribution, we calculated MBT, CBT and the fractional abundance of different bGDGTs in order to reconstruct temperature in the Bangong Co catchment.

Figure 8 demonstrates that MAAT reconstructed by Günther et al. (2014) (B) and by the new calibrations in the present study (I and J)) is close to the local instrumental MAAT (Shiquanhe, 0.69°C, 1961–2013); and that reconstructed SLST (Pearson et al., 2011) (F) is 3–4°C lower than that determined by *in situ* measurement. Meanwhile, MBT-CBT or fractional abundance of bGDGTs derived

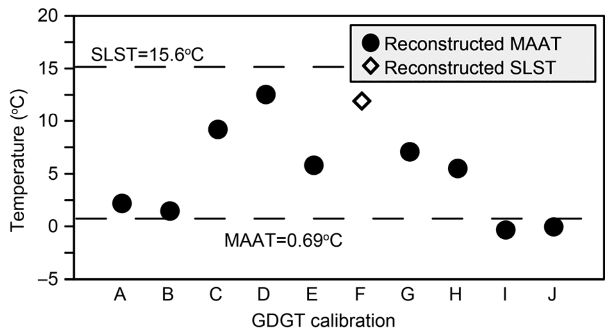

Figure 8 Reconstructed temperature in Bangong Co using different calibrations. A. Zink et al. (2010); B. Günther et al. (2014); C. Sun et al. (2011); D. Tierney et al. (2010) (MBT, CBT); E. Tierney et al. (2010) (f_{abun})); F. Pearson et al. (2011); G. Loomis et al. (2012) (MBT, CBT); H. Loomis et al. (2012) (f_{abun})); I. this study (MBT, CBT); J. this study (f_{abun}).

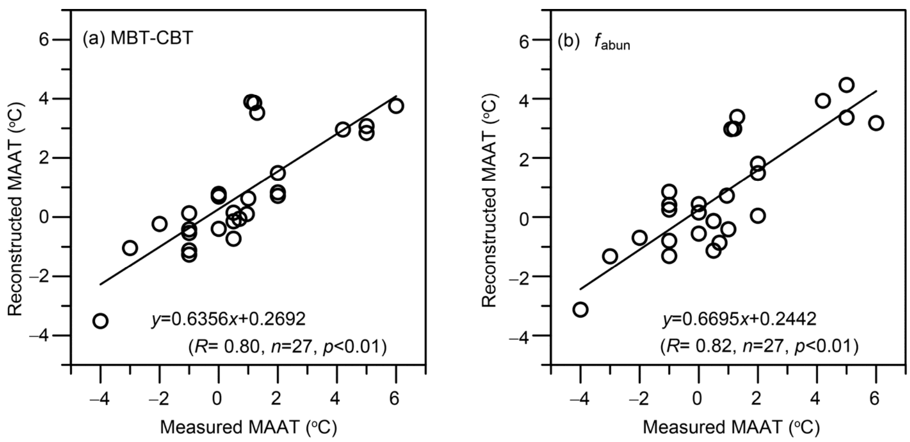

Figure 9 Regression results of reconstructed MAAT versus measured MAAT. (a) MBT-CBT derived MAAT; (b) fractional abundances of bGDGTs derived MAAT).

function for MAAT is well correlated with measured MAAT (Figure 9a, 9b). Based on these results, we conclude that the two calibrations developed in the present paper, together with those of Günther et al. (2014), are applicable to Bangong Co. In contrast to previous work, our new calibrations are not only based on a significantly larger number of TP lakes than used previously, but they also exhibit a smaller RMSE in the same temperature gradient, which should improve the accuracy of the resulting quantitative paleotemperature reconstructions.

5. Conclusions

Based on an investigation of the GDGT distribution in the surface sediments of 27 TP lakes, together with that in soil samples from six of the lake catchments, we conclude that bGDGTs are quantitatively the most significant in the lake sediments, followed by crenarchaeol and GDGT-0. We observed no significant difference between the lake sediments and soils within the same lake catchments, which supports previous conclusions regarding the complexity of GDGT sources. In the case of small lakes on the TP, the iGDGT distribution is mainly influenced by lake water chemistry parameters (pH and salinity); TEX_{86} may be a potential lake pH proxy; and bGDGT distribution is mainly controlled by climatic factors. Finally, we established two calibrations based on MBT-CBT and fractional abundance of different bGDGTs. The reconstructed results using the new calibrations are similar to the results of *in situ* measurements at Bangong Co, and therefore we conclude that they are suitable for quantitative paleotemperature reconstruction for TP lakes.

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