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ORIGINAL ARTICLE

Dissolved organic carbon, CO_2, and CH_4 concentrations and their stable isotope ratios in thermokarst lakes on the Qinghai-Tibetan Plateau

Cuicui MU,[1,2] Tingjun ZHANG,[2*] Qingbai WU,[1*] Xiaoqing PENG,[2] Peng ZHANG,[1] Yuzhong YANG,[1] Yandong HOU,[1] Xiankai ZHANG,[2] Guodong CHENG[1]

[1]State Key Laboratory of Frozen Soil Engineering, Cold and Arid Regions Environmental and Engineering Research Institute (CAREERI), Chinese Academy of Sciences, Lanzhou, Gansu 730000, China; [2]Key Laboratory of Western China's Environmental Systems (MOE), College of Earth and Environmental Sciences, Lanzhou University, Lanzhou, Gansu 730000, China
*Corresponding authors: tjzhang@lzu.edu.cn ; qbwu@lzb.ac.cn

ABSTRACT
Thermokarst lakes are widely distributed on the Qinghai-Tibetan Plateau (QTP), which accounts for 8% of the global permafrost area. These lakes probably promote organic matter biodegradation and thus accelerate the emission of carbon-based greenhouse gases. However, little is known about greenhouse gas concentrations and their stable isotopes characteristics of these lakes. In this study, we measured the concentrations of dissolved organic carbon (DOC), dissolved CO_2 and CH_4, as well as the distribution of $\delta^{13}C_{CO2}$, $\delta^{13}C_{CH4}$, and $\delta^{13}C_{OM}$ (organic matter) of lake sediments in thermokarst lakes on the QTP. Results showed that the OM of the lake sediments was highly decomposed. The concentrations of DOC, CO_2 and CH_4 in the lake water on the QTP were 1.2-49.6 mg L^{-1}, 3.6-45.0 µmol L^{-1} and 0.28-3.0 µmol L^{-1}, respectively. The highest CO_2 and CH_4 concentrations were recorded in July while the lowest values in September, which suggested that temperature had an effect on greenhouse gas production, although this pattern may also relate to thermal stratification of the water column. The results implied that thermokast lakes should be paid more attention to regarding carbon cycle and greenhouse gas emissions on the QTP.

Key words: Qinghai-Tibetan Plateau; thermokarst lakes; greenhouse gas; dissolved organic carbon; stable carbon isotope.

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INTRODUCTION

Thermokarst lakes are the most abundant and easily recognizable forms of thermokarst terrains, and are widespread in tundra regions with ice-rich permafrost. Thermokarst lakes form as ice-rich permafrost thaws, and expand as lake water surface increase into the adjacent permafrost. The lakes usually start with some shallow soil depressions that transform into small ponds, grow into thermokarst lakes, and eventually form large lakes (Ling *et al.,* 2012). Thermokarst lakes are widely distributed throughout most permafrost regions, including northern Canada, Russia, Mongolia, China, and Alaska (Hinkel *et al.,* 2005; Veremeeva and Gubin, 2009; Ling *et al.,* 2012). Currently, newly formed thermokarst lakes in Western Siberia make up ~48% of the surface area in the region (Zimov *et al.,* 1997). Total thermokarst lake area is up to 6% of the continuous permafrost on Earth (Kirpotin *et al.,* 2011). Recently, carbon cycle in thermokarst lakes has been received increased attention worldwide because the emissions of greenhouse gases into the atmosphere may play an important role in a warming climate (Walter *et al.,* 2006; Walter *et al.,* 2007; Shirokova *et al.,* 2013; Langer *et al.,* 2015; Sepulveda-Jauregui *et al.,* 2015). The Qinghai-Tibetan Plateau (QTP) in China has the largest extent of permafrost in the low-middle latitudes of the world, with permafrost underlying ~67% of the land area (Wu and Zhang, 2010). Although their total numbers and surface areas remain unknown, thermokarst lakes on the QTP are commonly distributed. It has been revealed that there were about 250 thermokarst lakes with an average size of 5580 m^2 and a total size of 1.39×10^6 m^2 located along the Qinghai-Tibetan Railway (Niu *et al.,* 2011).

The thermokarst lakes are developing rapidly due to the degradation of the permafrost on the QTP (Nan *et al.,* 2005; Wu *et al.,* 2014a). A survey from August 2007 to October 2008 showed that ~80% of thermokarst lake shores collapsed every year, with a maximum rate of ~1.8 m y^{-1} (Lin *et al.,* 2010). Similar to the sub-Arctic regions, a large amount of carbon-based greenhouse gas emissions were observed from the thermokarst lakes on the QTP (Wu *et al.,* 2014a). So far, little is known about the concentrations of these carbon-based greenhouse gases, and it also remains unknown about the seasonal changes of gas concentrations during summer periods in thermokarst lakes on the QTP. The stable carbon isotope ($\delta^{13}C$‰) is a potentially valuable tool for studying carbon dynamics in ecosystems. Isotopes can be used as evidences of CH_4 production and oxidation (Conrad, 2005), organic matter accumulation (Andersson *et al.,* 2012), and environmental conditions affecting fractionation (Manasypov *et al.,* 2014). The ′$^{13}C_{CH4}$ is also a powerful tool to uncover the mechanism of CH_4 formation because the fractionation during methanogenesis would cause $\delta^{13}C_{CH4}$ values to be considerably depleted compared to that of either the precursor organic compound or CO_2 (Hershey *et al.,* 2014).

Since permafrost regions on the QTP may also play an important role in greenhouse gas emissions (Mu *et al.,* 2015), it is important to understand the concentration of these gases and their dynamics. We hypothesized that gas concentrations in thermokarst lakes may be comparable with those of circum-Arctic regions with considerable variation among different lakes and may also have temporal variations, which may be affected by temperature. To test this hypothesis, we measured the organic carbon contents of the lake sediments, concentrations of DOC, dissolved CO_2 and CH_4 in the lake water, as well as the stable carbon isotopes of these components. The objectives of the present study were to i) examine the concentrations of dissolved organic carbon, CO_2 and CH_4 in thermokarst lakes on the QTP; ii) reveal the possible sources of these dissolved carbon and greenhouse gases; iii) discuss the possible effects of temperature on these dissolved carbon and greenhouse gases. The results will offer some insight into the organic carbon biogeochemical characteristics in thermokarst lakes on the QTP, and therefore improve our knowledge of carbon cycle processes in permafrost regions.

METHODS

Study area

The sampling sites on the QTP are located on a high plain landform, marked by alluvial and aeolian soil, where thermokarst ponds and lakes are found over much of the permafrost regions. Nine thermokarst lakes along the Qinghai-Tibetan Highway were selected: *Qingshuihe* (QSH), *Xieshuihe1* (XSH1), *Chumaerhe* (CMEH), *Beiluhe1* (BLH1), *Beiluhe2* (BLH2), *Wuli* (WL), *Wudaoliang* (WDL), *Xiushuihe2* (XSH2), and *Beiluhe3* (BLH3) (Fig. 1). These lakes have formed 900 years ago (Niu *et al.,* 2011), and belong to closed-basin lakes. These lakes are brackish due to the high evaporation in the plateau and the absence of lake outlets. All the thermokarst lakes are isolated water bodies, with well-defined edges.

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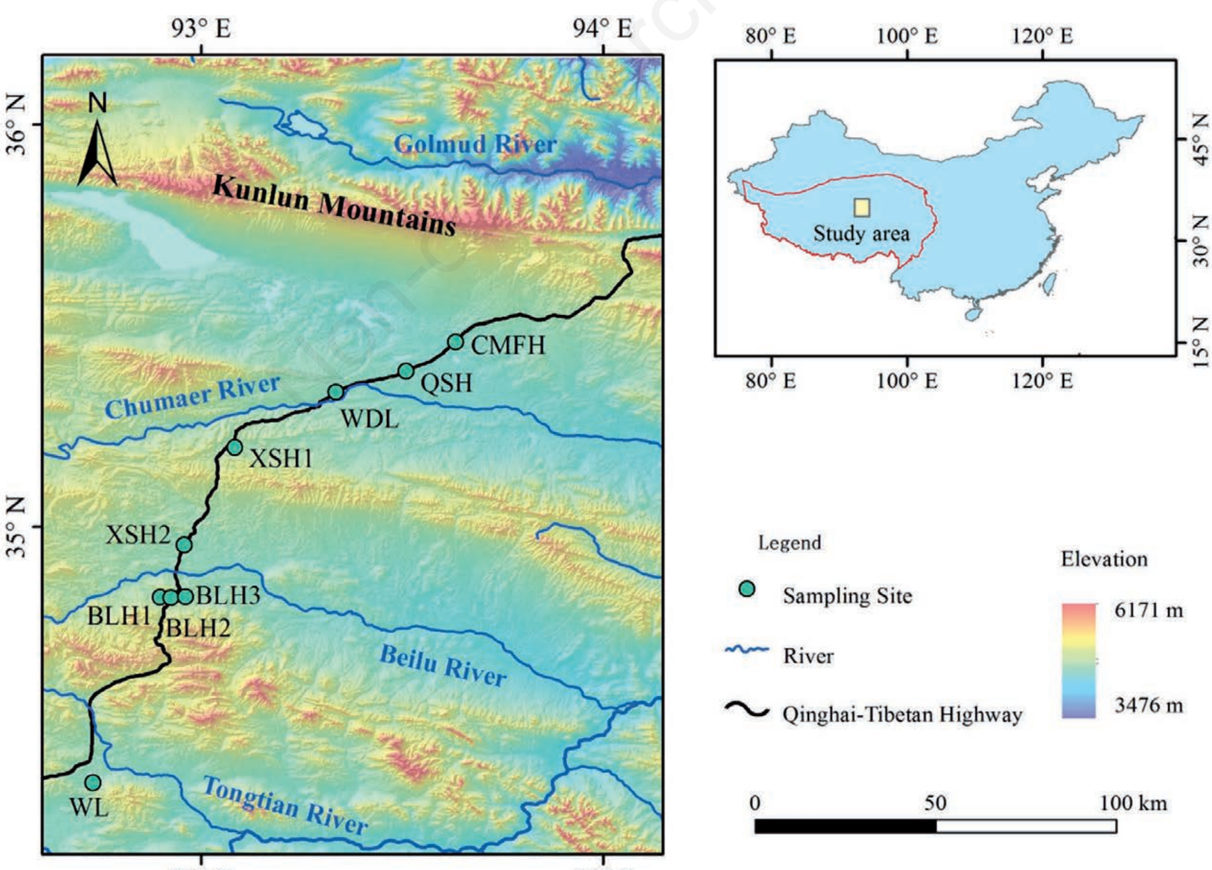

Fig. 1. Geographic location of the study area and the distribution of sampling sites.

The vegetation types are mainly alpine steppe, alpine meadow, and desert steppe (Tab. 1). Surficial geology is characterized by mainly Quaternary alluvial sand, silt, and silty clay, underlain by Tertiary mudstone and sandstone. As for the soil taxonomy, it is difficult to list the soil group or soil family in the watersheds of the lakes since several soil types usually co-existed in some areas. The dominant soil orders are presented in Tab. 1.

Currently, the sampling areas are characterized as an alpine semiarid climate, with annual mean precipitation of ~300 mm, mean annual air temperature of -5.0°C to -3.8°C, and mean annual ground temperature ranging from -2.0°C to -0.5°C (Zhu *et al.,* 2011). The thermokarst lakes lie in the ice-rich continuous permafrost (Niu *et al.,* 2011), with an active layer thickness of 1.8-3.0 m and a permafrost thickness of ~20-80 m (Lin *et al.,* 2010).

Sampling and analysis

Field sampling was carried out using a boat in July, August, and September 2013. The perimeter of lake was measured by ground-based surveying using GPS, and then its area was calculated. Ten sites were sampled in each lake, at ~3-4 m increments in a cross pattern, with double sampling at the lake center. The lake depth was measured at the same site and provided a mean value. Dissolved CO_2 and CH_4 concentrations in lake water were determined on a total of 90 open water samples. During sampling, water samples were collected from the upper 10 cm of water using a 60 mL polypro syringe. 30 mL water samples were injected from the syringe through a 0.45 mm syringe filter into 50 mL glass serum bottles (Striegl *et al.,* 2001). The bottles were previously flushed with N_2 at atmospheric pressure and sealed with rubber stoppers at State Key Laboratory of Frozen Soil Engineering, China, to exclude the effects of the gases in the air. The aluminum caps were fixed in field after the injection of water samples to ensure the sealing performance of the bottles. Then the bottles were placed upside down. The serum bottles also contained 2 g KCl to inhibit microbial activity (Striegl *et al.,* 2001). The released gas samples by diffusive flux were sampled through a plastic chamber and vacuum pump. To avoid the interference of atmosphere, the plastic chamber was covered at the depth of 5 cm to collect the samples to analyze $\delta^{13}C_{CO2}$ and $\delta^{13}C_{CH4}$ (Walter *et al.,* 2007). Three sediment samples were collected at each lake (except at QSH and XSH1) using a gravity sediment corer to analyze the $\delta^{13}C_{OC}$ (organic carbon). All lake water in the upper 10 cm was sampled and kept at 5°C to analyze the DOC concentrations. Meanwhile, surface water temperature, air temperature, atmospheric pressure, specific conductivity, and pH values were measured when the samples were collected.

Organic carbon (OC) of lake sediments and DOC concentrations were analyzed by using the solid and liquid modules of OI Analytical Analyzer (OI-Picarro, CA, USA) respectively, with precision of 0.5‰. The analysis

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Tab. 1. Physical and chemical parameters of thermokarst lakes on the Qinghai-Tibet Plateau.

Lakes		Qingshuihe	Xieshuihe1	Chumaerhe	Beiluhe1	Beiluhe2	Wuli	Wudaoliang	Xiushuihe2	Beiluhe3
Latitude		35°23.23’	35°27.53’	35°20.10’	34°49.49’	34°49.46’	34°21.86’	35°11.87’	34°57.31’	34°49.61’
Longitude		93°30.56’	93°37.97’	93°20.04’	92°53.82’	92°55.42’	92°43.76’	92°05.01’	92°57.39’	92°05.01’
Elevation (m)		4490	4457	4536	4660	4643	4572	4627	4562	4615
Vegetation type in the catchment		Alpine steppe	Desert steppe	Alpine steppe	Alpine meadow	Alpine meadow	Desert steppe	Alpine meadow	Desert steppe	Desert steppe
Soil orders		Entisol	Entisol	Entisol	Mollisol/ Gelisol	Mollisol/ Gelisol	Inceptisol	Mollisol	Inceptisol/ Entisol	Inceptisol
Vegetation cover		40-60%	20-30%	30%	80%	80-90%	20%	80%	10%	5%
Lake area (m^2)		19500	34000	17000	4200	15000	8200	7700	19000	3600
Maximum depth (m)		1.5	2.8	0.85	0.65	1.25	1.25	1.4	0.65	0.88
Perimeter (m)		595	710	515	277	538	340	393	560	249
pH	July	8.90	8.57	9.22	9.23	9.66	8.65	8.78	9.23	8.61
	Aug	9.09	8.55	9.52	9.85	10.0	8.57	8.57	9.53	8.67
	Sep	8.67	8.36	8.94	10.3	8.57	8.02	6.36	5.62	9.87
EC (ms/cm)	July	46.2	14.3	32.7	23.1	28.3	28.4	38.0	22.7	37.8
	Aug	42.7	12.6	32.4	20.0	27.9	24.3	48.9	26.3	45.3
	Sep	33.1	10.9	20.3	22.2	25.0	21.3	49.5	28.8	41.9
Temperature (°C)	July	15.8	14.2	18.6	17.3	18.1	20.7	17.8	19.4	18.1
	Aug	17.1	17.1	14.9	21.7	19.3	19.5	16.2	13.7	19.3
	Sep	8.7	8.4	8.9	10.3	5.6	8.0	6.4	5.6	9.9
OC%		__	__	2.4%	1.5%	7.6%	4.2%	1.4%	1.0%	2.0%
$\delta^{13}C_{OC}$		__	__	-8.7‰	-6.9‰	-18.9‰	-7.6‰	-7.2‰	-10.0‰	-9.8‰

was performed in triplicate with an uncertainty of 2%, and DOC detection limit of 0.3 mg L^{-1}. CO_2 concentrations (volume fraction) in serum bottle headspace were determined for three times by using a Li-Cor 7000 infrared CO_2 analyzer fitted with a sample injection port and N_2 carrier gas (Striegl *et al.,* 2012) at the Laboratory of Snow and Frozen Ground at Lanzhou University, China. CH_4 concentrations were determined for three times using a GC-7890A gas chromatograph with a flame ionization detector (Striegl and Michmerhuizen, 1998). After every 15 samples, a calibration of the detectors was performed using national gas standards (100 ppmv CH_4, 1000 ppmv CO_2). Duplicate injection of the samples showed that the results were reproducible within the accuracy of ±5%.

An isotope mass spectrometer (Thermo Finnigan, NJ, USA) was used to measure $\delta^{13}C_{CO2}$ and $\delta^{13}C_{CH4}$, with the estimated measurement uncertainty of ±0.2‰, and the disagreement between the measured and certified values was less than 0.3‰. The $\delta^{13}C_{OC}$ values in lake sediments were measured by OI Anlytical Analyzer (OI-Picarro) using the samples collected in September. Results were based on the mean of three replicates of each sample, and expressed as δ values relative to the Vienna Peedee belemnite (VPDB) standard for $\delta^{13}C$. The δ values are defined as:

$$\delta^{13}C = [(R_{sample} / R_{standard}) - 1] \times 1000 \qquad \text{(eq. 1)}$$

where R_{sample} and $R_{standard}$ are $^{13}C/^{12}C$ ratios of the samples and standards, respectively.

Data in the present study were presented as mean±SD. Statistical analysis (*t*-tests) was performed using SPSS 19.0.

RESULTS

Physical and chemical characteristics of thermokarst lake water

The physical and chemical parameters of thermokarst lakes are summarized in Tab. 1. The surface water temperatures of thermokarst lakes on the QTP exhibited little variations among the lakes, and decreased from July and August (average of 17.7°C) to September (average of 8.0°C). The average pH value of lake water was 9.15±0.03. The conductivity of lake water ranged from 10.9 to 49.5 ms cm^{-1}.

The organic carbon contents of the lake sediments ranged from 1.0% to 7.6%. The highest OC content was recorded in BLH2 thermokarst lake (with alpine meadow vegetation), followed by WL, which belonged to the desert steppe area, with the content of 4.2%.

The DOC, CO_2 and CH_4 showed different pattern within the three months (Fig. 2). The coefficients of variations (CVs) for DOC among the sampling periods ranged from 9.6% to 46.0%, and the DOC concentrations in July were significantly lower than those of August (*t*-test, P=0.02) and September (*t*-test, P=0.03). In contrast, CO_2 and CH_4 concentrations in July were significantly higher than those of August (*t*-test, $P<0.01$) and September (*t*-test, P=0.02).

Stable carbon isotopes

The $\delta^{13}C_{OC}$ in lake sediments ranged from -18.9‰ to -6.9‰, with a mean value of -8.4‰ (Tab. 1). The $\delta^{13}C_{CO2}$ values varied with less amplitude among the thermokarst lakes and ranged from -18.4‰ to -15.1‰. The mean value of $\delta^{13}C_{CO2}$ was -16.3‰ during the sampling period (Fig. 3). The $\delta^{13}C_{CH4}$ values were much lower than those of OC of lake sediments and CO_2. The $\delta^{13}C_{CH4}$ values showed a decreasing trend from July to September from -27.3‰ to -48.8‰. The $\delta^{13}C_{CH4}$ values in September were significantly lower than those of July and August (Fig. 3).

DISCUSSION

DOC, and dissolved concentrations of CO_2 and CH_4

Soil organic carbon (SOC) content is closely related to the vegetation types in permafrost regions on the QTP (Wu *et al.,* 2012; Wu *et al.,* 2014b). Since the lake sediments may receive allochthonous carbon from the watershed

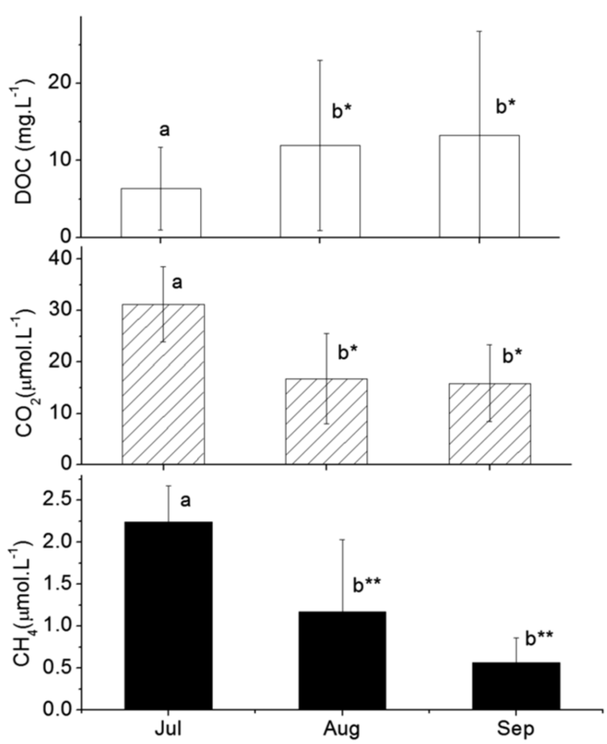

Fig. 2. Dissolved DOC, CO_2 and CH_4 concentrations during sampling periods. The letters a,b show the significant difference among different groups; $^*P<0.05$; $^{**}P<0.01$.

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(Hershey *et al.,* 2014), the organic carbon of lake sediments may also relate to the vegetation types of the terrestrial ecosystems. In the present study, it could be found that there was the highest OC content in the lake of alpine meadow, the second highest OC was in the lake of alpine desert and there are small variations of OC in other lakes (Tab. 1). This suggests that the OC in thermokarst lake sediments did not strictly follow the patterns of SOC in the terrestrial ecosystems in the permafrost regions on the QTP.

The DOC concentrations varied greatly in the present study. The highest DOC concentration (49.6 mg L^{-1} in WDL) was similar with that (37.5 mg L^{-1} of DOC) in thermokarst lakes of Alaska. Other DOC concentrations on the QTP (1.2-15.7 mg L^{-1}) are comparable with those in thermokarst lakes of western Siberia (5-6 mg L^{-1}) (Shirokova *et al.,* 2013), in subarctic region of northern Sweden (7.2 mg L^{-1}) (Karlsson *et al.,* 2010), and in northern Canada (~1-25 mg L^{-1}) (Kokelj *et al.,* 2005; Prairie *et al.,* 2009; Laurion *et al.,* 2010). The increasing trends of DOC from July to August probably indicated the dissolved organic matter input from the watershed with the ground water in summer (Manasypov *et al.,* 2015).

In circum-Arctic regions, the CO_2 concentrations in lakes varied from 25 to 200 μmol L^{-1}, and most thermokarst lakes show values from 30 to 70 μmol. L^{-1} (Sobek *et al.,* 2003; Striegl *et al.,* 2012; Shirokova *et al.,* 2013). The CH_4 concentrations in circum-Arctic regions ranged from 0.01 to 5 μmol L^{-1}, and most of them were lower than 1 μmol L^{-1} (Striegl *et al.,* 2012; Shirokova *et al.,* 2013). In comparison with those studies, it could be seen that the CO_2 concentrations in the present study (mean value of 21.3 μmol L^{-1}) were relatively lower, while the CH_4 concentrations (mean value of 1.33 μmol L^{-1}) were higher.

During the sampling periods, dissolved CO_2 and CH_4 showed higher values in July, while DOC showed higher contents in August and September (Fig. 3). Higher concentrations of dissolved CO_2 and CH_4 occurred in July were reasonable since temperatures in July and August (mean 17.7°C) are more favorable for microbial activity (Tab. 1), which causes DOC to be decomposed into CO_2 and CH_4 (Zimov *et al.,* 1997). Correspondingly, relative lower temperatures in September (mean 8.0°C) mean lower decomposition of DOC. In addition, higher DOC concentrations at the end of summer may also be a product of allochthonous input (such as supply of DOC from the soil) and the accumulation of autochthonous carbon (Andersson *et al.,* 2012; Shirokova *et al.,* 2013; Hershey *et al.,* 2014). In the present study, we did not measure the vertical temperature regimes in the thermokarst lakes and could not know that if there were thermal stratifications in the lake during summer (Boike *et al.,* 2015). Therefore, further studies are needed to address the possible effects of stratification on the concentration of these greenhouse gases in these lakes.

It has been pointed out that the thermokarst lakes in circum-Arctic regions played important roles in the greenhouse gas emissions in permafrost regions (Walter *et al.,* 2006; Walter *et al.,* 2007; Shirokova *et al.,* 2013). Since mountain permafrost constitutes a significant part of the global permafrost area and QTP accounts for about 8% of the global permafrost area (Wu *et al.,* 2016), the investigation of greenhouse gases in thermokarst lakes on QTP provides important data for climate change study. The concentrations of the carbon based greenhouse gases showed that more attention should be paid on these thermokarst lakes in the future.

Sources of CO_2 and CH_4

The $\delta^{13}C_{OC}$ values ranged from -10‰ to -6.85‰, with an exception of relative lower values (-18.4‰) in our study. These values are much higher than boreal (-21‰ to -24‰) (Peters *et al.,* 1978) and subtropical lakes (-25‰ to -26‰) (Torres *et al.,* 2012). This result suggests that the OC in the thermokarst lake sediments was highly decomposed. The lowest value of $\delta^{13}C_{OC}$ appeared in the lake with the highest OC contents (7.6%), which suggests the preservation of organic carbon associated with a lower decomposition rate.

The mean $\delta^{13}C$ value of atmospheric CO_2 from remote areas is about -7.8‰ (Boutton, 1991) with a wide range from -30‰ to almost 0‰ (Affek and Yakir, 2014), and the CO_2 from vegetation decomposition with a $\delta^{13}C$ value of approximately -27‰ (Boutton, 1991), and the $\delta^{13}C$ val-

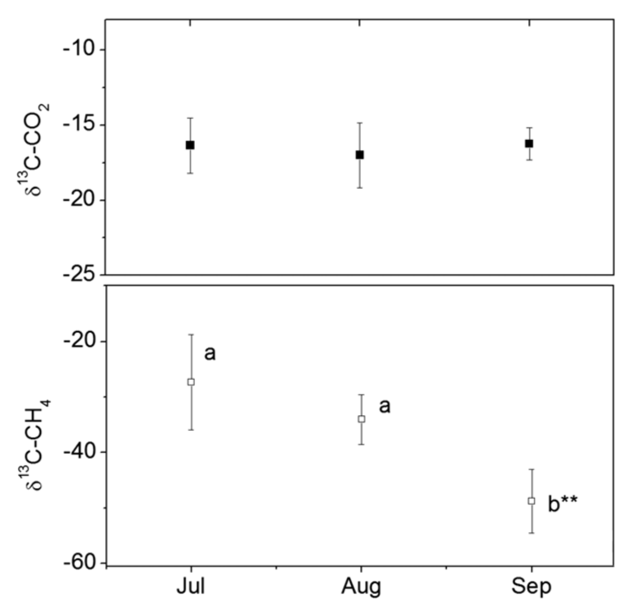

Fig. 3. $\delta^{13}C$ values of DOC, CO_2 and CH_4. The letters a,b show the significant difference among different groups; *P<0.05; **P<0.01.

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ues of CH_4 are usually around -60‰ (Chasar *et al.,* 2000). The $\delta^{13}C$ values of CO_2 showed relatively small changes over thermokarst lakes in the present study (-18.4‰ to -15.1‰). The relative higher $\delta^{13}C$ values of both CO_2 and CH_4 in the present study can also be explained by the highly decomposed OC in the sediments. These small changes of $\delta^{13}C_{CO2}$ indicated that CO_2 in lake water on the QTP may originate from the same source.

The $\delta^{13}C_{CH4}$ values in thermokarst lakes in September (-58.2‰ to -42.9‰) were significantly lower (more depleted) than those in July and August (-41.1‰ to -16.5‰). It has been demonstrated that the CH_4 came from microbial decomposition of organic matter under anaerobic conditions (Knorr *et al.,* 2008). The lower $\delta^{13}C_{CH4}$ values indicated that the lower activities of Achaea populations constrain their decomposition of the organic matter with higher $\delta^{13}C$ values (Wachinger *et al.,* 2000). This could be confirmed by the lower concentrations of CH_4 in September, which probably suggested that anaerobic respiration was less intense at the end of summer due to the decreasing temperature (Martinez-Cruz *et al.,* 2015). It is worth mentioning that it is difficult to draw more robust conclusions from the isotope fractions of CO_2 and CH_4 in the present study due to the lack of the data of dissolved inorganic carbon (DIC). However, the isotope of the carbon based greenhouse gases still merits further study since it could be a powerful indictor of the processes of the production of these greenhouse gases (Corbett *et al.,* 2013; Throckmorton *et al.,* 2015).

CONCLUSIONS

Our study investigated the concentrations of DOC, CO_2 and CH_4, as well as the characteristics of stable carbon isotopes in thermokarst lakes in permafrost regions on the QTP. The results suggested that the CO_2 concentrations in these lakes were lower than those of sub-Arctic regions while the CH_4 were higher. The evidence from $\delta^{13}C$ showed that organic carbon in the lake sediments was highly decomposed. The production of CH_4 showed a seasonal variation, which was probably affected by the temperature. The results highlighted that the carbon-based greenhouse gases in the thermokarst lakes on the QTP should be paid more attention to in the future since they may also contribute a part of greenhouse gas emissions in permafrost regions.

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