Lakes on the Tibetan Plateau as Conduits of Greenhouse Gases to the Atmosphere

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Key Points:
- Littoral zones of lakes on the Tibetan Plateau are sources of carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O)
- Dissolved organic carbon, dissolved organic nitrogen, water salinity, and water temperature are indicators of CO_2 fluxes
- CO_2 exchange with the atmosphere is enhanced 2.5 times due to the high salinity and pH of lakes on the Tibetan Plateau

Supporting Information:
- Supporting Information S1
- Data Set S1

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Lakes on the Tibetan Plateau as Conduits of Greenhouse Gases to the Atmosphere

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Abstract Lakes play an important role in the global carbon cycle, and littoral zones of lakes are potential hotspots of greenhouse gas production. In this study, we measured the partial pressures of carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) in the littoral zones of 17 lakes on the Tibetan Plateau. The littoral zones of lakes on the Tibetan Plateau were supersaturated and acted as sources of CO_2, CH_4, and N_2O to the atmosphere. The average partial pressures of CO_2, CH_4, and N_2O in the surface lake water were 664.8 ± 182.5, 139.8 ± 335.6, and 0.3 ± 0.1 μatm, respectively. The average diffusive fluxes (and uncentainty intervals) of these three gases were 73.7 (0.9–295.3) mmol · m^{-2} · day^{-1}, 5.2 (0.0008–45.9) mmol · m^{-2} · day^{-1}, and 6.5 (0.07–20.9) μmol · m^{-2} · day^{-1}, respectively. The diffusive fluxes of CO_2 in lakes were significantly correlated with dissolved organic carbon, dissolved organic nitrogen, salinity, and water temperature. The diffusive fluxes of N_2O were significantly correlated with lake water depth. However, no relationships were found between environmental factors and the CH_4 diffusive flux at the scale of this study. CO_2 exchange with the atmosphere from saline lakes was found to be higher than from freshwater lakes with equivalent CO_2 concentrations by a factor of 2.5 due to chemical enhancement of the gas transfer velocity. Therefore, further study with enhanced spatiotemporal resolution and breadth is needed to better understand the important role played by lakes on the Tibetan Plateau in both regional and global carbon cycles.

1. Introduction

Inland waters (particularly lakes, rivers, and reservoirs) affect not only regional climate by exchanging water and heat with the atmosphere (Tranvik et al., 2009) but also global climate by playing an important role in the carbon cycle (Cole et al., 2007; Tranvik et al., 2009). The production and consumption of carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) by inland waters alter atmospheric greenhouse gas levels and thus the heat exchange between the atmosphere and other ecosystems (Tranvik et al., 2009). CO_2, CH_4, and N_2O are three important greenhouse gases (GHGs) that account for 80% of the total radiative forcing of well-mixed GHGs (Ciais et al., 2014). Although inland waters cover a small fraction of the Earth's surface, they play a spatially disproportionate role in the global carbon cycle (Cole et al., 2007; Raymond et al., 2013; Tranvik et al., 2009). For example, the global annual CO_2 emissions from inland waters was estimated to be 2.1 Pg C/year (1 Pg = 10^{15} g) from a surface area of approximately 3,620,000 km^2 (Raymond et al., 2013), while the annual CO_2 emissions from soils was estimated at 98 Pg C/year from a surface area of 134,038,710 km^2 (Bond-Lamberty & Thomson, 2010; Oertel et al., 2016). Lakes are an important component of the inland water system regulating the carbon cycle and climate by storing, transporting, and transforming carbon (Tranvik et al., 2009). They are also important sources of CO_2, CH_4, and N_2O (Bastviken et al., 2004; Battin et al., 2009; Butman & Raymond, 2011; Ran et al., 2017; Tranvik et al., 2009). For example, approximately 93% of 4,902 lakes distributed in different climatic regions were supersaturated with CO_2 (Sobek et al., 2005). A study of 207 boreal lakes in Finland showed that all were supersaturated with CH_4 (Juutinen et al., 2009). Globally, the annual emissions of CO_2 and CH_4 from lakes and ponds were estimated to be 0.571 and 0.012 Pg C/year, respectively, with small ponds playing a disproportionately important role in relation to their surface area (Holgerson & Raymond, 2016). In addition, the annual emissions of CO_2 from global saline lakes were 0.11–0.15 Pg C/year (Duarte et al., 2008), accounting for approximately 39–47% of CO_2 emissions from worldwide lakes.

Because the environment on the Tibetan Plateau is pristine, lakes on this remote plateau are vulnerable to the effects of anthropogenic activities. Thus, lakes on the Tibetan Plateau are considered to be sensitive indicators of global climate change (Fang et al., 2016; Pan & Li, 1996). Due to climate change, the ice-free period for lakes on the Tibetan Plateau will become longer (Song et al., 2016), and the surface areas of these lakes will become more variable (Fang et al., 2016). Additionally, with increasing temperatures (Yao et al., 2012) and the deposition of black carbon (Li et al., 2016), glaciers on the Tibetan Plateau are rapidly melting, which may affect the surface areas of lakes in the region. Furthermore, glacier melt water is high in bioavailable dissolved organic carbon (DOC); therefore, organic carbon supplies to lakes in the area may increase (Hood et al., 2015; Yan et al., 2016). Because climate warming on the Tibetan Plateau is expected to continue at a more accelerated rate than global climate warming (Kang et al., 2010), it is necessary to investigate how these lakes will function in the future.

Several studies on CO_2, CH_4, and N_2O emissions from lakes on the Tibetan Plateau have been conducted. A study of thermokarst lakes on the Tibetan Plateau indicated that CO_2 and CH_4 concentrations were higher in July than in September because higher temperatures promote DOC decomposition (Mu et al., 2016). The littoral zones of Huahu Lake in this region are sources of CH_4 and N_2O (Chen et al., 2009, 2011). Multiple studies have proven that littoral zones, an interface between terrestrial and aquatic ecosystems, are potential hotspots of CO_2, CH_4 (Chen et al., 2009; Larmola et al., 2004), and N_2O production (Chen et al., 2011; Diem et al., 2012; Huttunen et al., 2003). Nevertheless, additional studies of littoral zones of lakes on the Tibetan Plateau are needed because of the large spatial extent and number of lakes on this plateau, in addition to their utility as global change indicators and potential for saline-enhanced CO_2 production. Therefore, it is necessary to conduct large-scale research to evaluate CO_2, CH_4, and N_2O emissions from lakes on the Tibetan Plateau to understand their accumulative contribution to atmospheric GHGs.

GHG variations in lake water can be predicted by different environmental factors and lake characteristics. For example, DOC (Hope et al., 1996), phytoplankton chlorophyll α, and pH (Holgerson, 2015) are predictors of CO_2 concentration, and dissolved oxygen is predictor of CO_2 flux (Kortelainen et al., 2006). Lake area, depth (Bastviken et al., 2004), water temperature (Borrel et al., 2011) and precipitation (Holgerson, 2015) are used to predict CH_4 concentration and flux. Nitrate content, water temperature, and dissolved oxygen are predictors of N_2O flux (Mengis et al., 1997; Zhu et al., 2015). However, the environmental predictors of CO_2, CH_4, and N_2O concentration and flux differ by lake due to the variable physical and chemical characteristics of lakes.

In this study, we analyzed the partial pressures and diffusive fluxes of CO_2, CH_4, and N_2O in the littoral zones of lakes on the Tibetan Plateau to provide a preliminary mapping data set of GHG emissions in this high-altitude vulnerable region, and to investigate the environmental factors influencing GHG emissions.

2. Materials and Methods

2.1. Site Description and the Study Lakes

The Tibetan Plateau has an area of approximately 2.5×10^6 km^2, with an average elevation of over 4,000 m above sea level (asl). This region is one of the most sensitive areas to climate change in the world and manifests as both a *driving force* and an *amplifier* of global climate change (Pan & Li, 1996). From 2000 to 2015, the annual average temperature of the Tibetan Plateau was 3.39°C, and it ranged from −19.5°C (early February) to 25.1°C (late July; Song et al., 2016). During the last half century, the Tibetan Plateau experienced significant warming at a mean annual rate of 0.17°C/decade (Song et al., 2016). The average annual precipitation ranges from 100 to 800 mm on the Tibetan Plateau, and the amount decreases from the southeast to the northwest (Fang et al., 2016; Xu et al., 2008). The majority of precipitation on the Tibetan Plateau occurs between June and September (60%–90%), and the region experiences humid, hot summers and dry, cold winters (Xu et al., 2008). The length of the growing season increased from 150 days in 1961 to 167 days in 2003 over the eastern and central regions of the Tibetan Plateau (Liu et al., 2006).

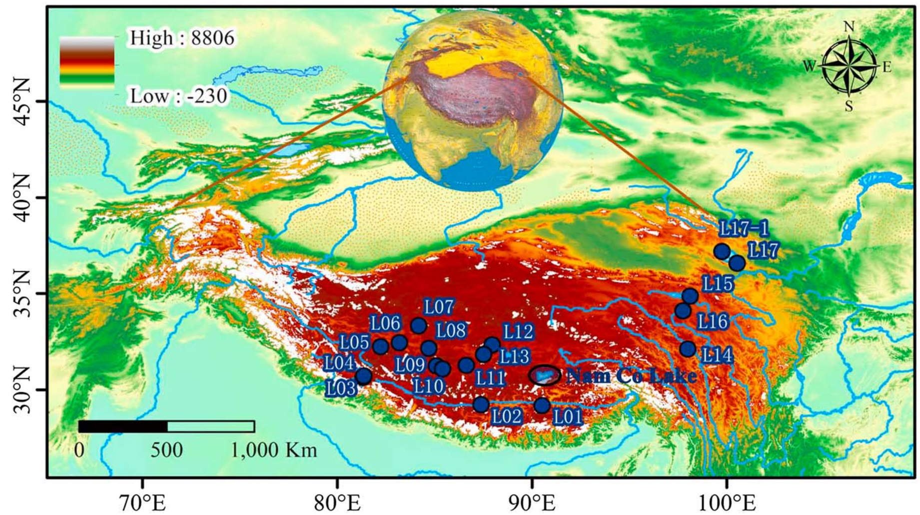

Figure 1. Sampling lake sites on the Tibetan Plateau.

The largest number of lakes in China are distributed on this high plateau. It is estimated that there are 1,091 lakes larger than 1 km^2 on the Tibetan Plateau, covering an area of 45,000 km^2, accounting for approximately 50% of the lake area in China. Among these lakes, 346 are larger than 10.0 km^2, making up almost 95% of the lake area in this region (Wang & Dou, 1998). Lakes on the Tibetan Plateau are primarily formed by tectonic movement, river erosion, landslides, and glacial activities (Wang & Dou, 1998). Most of these lakes are saline inland lakes, with the exception of fresh outflow lakes in the headwater regions of the Yangtze and the Yellow Rivers (Wang & Dou, 1998). The surface temperature of lakes on the Tibetan Plateau decreases rapidly from late autumn to winter. Ice cover begins in late November or December, while ice off usually starts in the following April or May (Song et al., 2016). The ice-free period for lakes on the Tibetan Plateau ranges from 144 to 365 days, with a shorter duration in northwestern lakes and a longer duration in southern lakes. Longer durations can also be found in lakes with large water volumes or high salinity (Song et al., 2016).

In the present study, GHG and water samples were collected from a single point in the littoral zone of 17 lakes on the Tibetan Plateau (Figure 1 and Table 1). These lakes were chosen because they were relatively easy to access and representative of large lakes on the southwestern and northeastern Tibetan Plateau. For example, Yamzho Yumco is the largest lake on the southern Tibetan Plateau, Serling Co and Dangreyong Co are large lakes characteristic of the area between the Tanggula and Gangdisi-Nyainqentanglha Ranges, and Ngoring Lake is one of the largest fresh outflow lakes in the region north of the Bayan Har Mountains (Wang & Dou, 1998).

2.2. Sample Collection and Measurement

Field expeditions were conducted in August 2014 and May 2015. The headspace equilibration method was adopted for CO_2, CH_4, and N_2O sample collection (Hope et al., 2001; Kling et al., 1991). In brief, 40 mL of lake water was collected with a 60 mL plastic syringe at approximately a 10 cm water depth. Twenty milliliters of N_2 was added to the syringe. The syringe was then shaken (underwater to maintain constant temperature) for 2 min to equilibrate the headspace with the water sample (Kling et al., 1991; Qu, Aho, et al., 2017). A 15 mL of headspace sample was then injected into a vaccumed airtight Exetainer vial (Labco Exetainer®, UK). All samples were collected in duplicate or triplicate. GHG concentrations were analyzed using a Shimadzu gas chromatograph (GC-2014, Kyoto, Japan) with a methanizer and flame ionization and electron capture detectors (Holgerson, 2015). The precisions of measurement were ±2.5%, ±2.9%, and ± 3.0% for CO_2, CH_4, and N_2O, respectively. Water sample gas concentrations were calculated from headspace concentrations according to the ideal gas law and Henry's law. Henry's law constants were from Weiss (1974) for CO_2, Wiesenburg and Guinasso (1979) for CH_4, and Weiss and Price (1980) for N_2O.

Water samples for measurements of DOC, dissolved inorganic carbon (DIC), dissolved organic nitrogen (DON) and major ions were collected at an approximately 10 cm depth below the surface water. Samples for DOC and DON measurement were collected in 250 mL acid-washed (10% HCl and ultrapure water) polycarbonate bottles after being filtered directly from the surface water through precombusted (450°C for 6 hr) glass fiber

Table 1
Sampling Information and Physical Parameters of the Studied Lakes

No.	Name	Sampling date (yy/mm/dd)	Sampling time	Latitude (N)	Longitude (E)	Elevation (m)	Lake area (km^2)	Maximum depth (m)	Water temperature (°C)	Type
L01	Yamzho Yumco	2015/5/8	14:20–14:35	29°10′08.52″	90°31′16.59″	4,445	650.5	40	16.5	Saline lake
L02	Angrenjin Co	2015/5/9	16:25–16:40	29°12′39.56″	87°22′59.36″	4,304	24.3	13	9.9	Saline lake
L03	Lhanag Tso	2015/5/11	13:30–13:45	30°37′58.78″	81°19′39.53″	4,476	268.5	49	9.4	Saline lake
L04	Mapam Yumco	2015/5/11	14:50–15:05	30°42′36.98″	81°22′05.02″	4,600	412	46	16.5	Freshwater lake
L05	Nieer Co	2015/5/16	16:00–16:15	32°13′58.64″	82°14′07.50″	4,442	33	-	16.7	Saline lake
L06	Cangmu Co	2015/5/16	20:10–20:25	32°26′49.86″	83°11′41.62″	4,439	87.5	6.3	9	Saline lake
L07	Chabo Co	2015/5/17	14:40–14:55	33°18′58.41″	84°11′10.04″	4,553	32	-	13	Saline lake
L08	Dong Co	2015/5/18	15:25–15:40	32°09′16.40″	84°42′05.67″	4,418	87.7	2	17.2	Saline lake
L09	Dawa Co	2015/5/18	19:25–19:39	31°13′52.77″	85°05′38.94″	4,639	114.4	-	17.1	Saline lake
L10	Zhari Nam Co	2015/5/19	11:15–11:30	31°04′46.35″	85°24′29.65″	4,791	996.9	71.6	11.0	Saline lake
L11	Dangreyong Co	2015/5/19	19:30–19:50	31°15′43.82″	86°37′54.43″	4,630	835.3	223	7.6	Saline lake
L12	Norma Tso	2015/5/20	15:20–15:40	32°19′37.97″	87°57′41.73″	4,867	68.1	-	9.2	Saline lake
L13	Dogze Co	2015/5/21	11:05–11:20	31°49′57.23″	87°31′04.40″	4,560	244.7	34	9.1	Saline lake
L14	Serling Co	2015/5/21	15:29–15:40	32°06′37.95″	89°00′13.02″	4,545	2391	-	12.2	Saline lake
L15	Xingxinghai	2014/8/15	17:35–18:00	36°34′34.32″	100°32′15.9″	4,212	26.28	-	17.6	Freshwater lake
L16	Ngoring Lake	2014/8/13	11:00–11:20	34°04′59.94″	97°45′8.58″	4,274	610	30.7	12.5	Freshwater lake
L17	Qinghai Lake-1	2014/8/14	9:00–9:30	36°34′34.32″	100°32′15.9″	3,199	4236.6	26	14.1	Saline lake
L17–1	Qinghai Lake-2	2014/8/14	15:31–15:50	37°10′32.88″	99°45′46.56″	3,199	4236.6	26	18	Saline lake

filters (GF-F; 47 mm in diameter; 0.7 μm nominal pore size) and stored at −20°C until laboratory analysis (Raymond et al., 2004, 2007). Samples for DIC measurement were collected in 125-mL brown, gastight, glass bottles (precleaned using ultrapure water and then combusted at 450°C for 5 hr). To prevent any biological and photodegradation, DIC samples were poisoned with a 0.2‰ saturated $HgCl_2$ solution and stored in the dark at 4°C (Raymond et al., 2004). DOC, DIC, and DON were analyzed with a TOC-500A analyzer (Shimadzu Corp, Kyoto, Japan), and major ions were measured with a Dionex–6000 Ion Chromatograph and a Dionex–3000 Ion Chromatograph (Dionex, USA). Water salinity, pH, and temperature were measured in situ using portable water testing kits (Wagtech CP1000). Air temperature and wind velocity were recorded with a portable anemometer (LZ836).

2.3. Calculation of Diffusive Fluxes of CO_2, CH_4, and N_2O

CO_2, CH_4, and N_2O diffusive fluxes were calculated based on the gas concentration gradients between the water and the atmosphere and the gas transfer velocity (Raymond et al., 2012):

$$F = k \times \left(C_{sur} - C_{eq}\right) \quad (1)$$

where k is the gas transfer velocity (cm/hr), C_{sur} is the gas concentration in surface water (μmol/L), C_{eq} is the gas concentration in water equilibrated with the atmosphere using Henry's law with the constants corrected by in situ water temperature and salinity, and F is the gas diffusive flux across the water-air interface (mmol · m^{-2} · day^{-1}). In this study, the partial pressure of CO_2, CH_4, and N_2O in the atmosphere were from Waliguan Baseline Observatory (36°17′N, 100°54′E, 3,816 m asl), located near the top of the Mount Waliguan on the edge of the northeastern Tibetan Plateau.

The k value for a given gas can be converted to that of any other gases based on their Schmidt numbers (Jähne, Heinz, et al., 1987; Jähne, Münnich, et al., 1987):

$$\frac{k_{gas1}}{k_{gas2}} = \left(\frac{Sc_{gas1}}{Sc_{gas2}}\right)^{-n} \quad (2)$$

where the Schmidt number (Sc) is the ratio of kinematic viscosity/gas diffusion coefficient. The exponent n is considered to be 0.5 (Jähne, Münnich, et al., 1987). Sc values for CO_2, CH_4, and N_2O were calculated based on the constants and formulas from Wanninkhof (1992):

$$Sc_{CO_2} = 2073.1 - 125.62t + 3.6276t^2 - 0.043219t^3 \quad (3)$$

$$Sc_{CH_4} = 2039.2 - 120.31t + 3.4209t^2 - 0.040437t^3 \quad (4)$$

$$Sc_{N_2O} = 2301.1 - 151.1t + 4.7364t^2 - 0.059431t^3 \quad (5)$$

where t is water temperature (in °C).

In this study, k_{CO_2}, k_{CH_4}, and k_{N_2O} were calculated from k_{600}. To reduce the uncertainties of k_{600}, the following three models from Cole and Caraco (1998), Crusius and Wanninkhof (2003), and Vachon et al. (2010), respectively, were used, and the average k_{600} was applied in the flux estimations:

$$k_{600} = 2.07 + 0.215U^{1.7} \quad (6)$$

$$k_{600} = 0.168 + 0.228U^{2.2} \quad (7)$$

$$k_{600} = 2.51 + (1.48\times U) + (0.39\times U\times \log_{10}LA)\times 0.228U^{2.2} \quad (8)$$

where U is wind speed and LA is lake area.

Additionally, k_{CO_2} was corrected for chemical enhancement, which is usually operative in saline lakes and calculated from water temperature, salinity, and pH (Duarte et al., 2008) using the formula given by Hoover and Berkshire (1969). The constants needed were from Johnson (1982), Dickson and Millero (1987) and Jähne, Heinz, et al. (1987). Moreover, Monte Carlo simulations were performed to estimate uncertainties in applying different k models in GHG flux estimations. This was achieved by randomly picking values from uncertainty intervals of pCO_2, pCH_4, pN_2O, and k. Totally, 10,000 iterative calculations were performed to achieve CO_2, CH_4, and N_2O fluxes (Raymond et al., 2013). The upper and lower limits are respectively the 5th and 95th confidence interval percentiles.

3. Results and Discussion

3.1. Partial Pressures of CO_2, CH_4, and N_2O in Surface Water

The partial pressure of CO_2 (pCO_2) in the littoral zones of the sampled lakes varied from 295 to 923 μatm (Figure 2). This variation fell within the ranges of global lakes studied in four seasons (1–7,845 μatm; Cole et al., 1994), saline lakes around the world (0.2–40,700 μatm; Duarte et al., 2008), and arctic Alaskan lakes (93–3,578 μatm; Kling et al., 1991). However, there are large discrepancies in pCO_2 between the littoral and pelagic zones of a lake. For example, the spatial distribution of pCO_2 increase during nighttime in a small embayment of Lake Memphremagog (Québec) suggested that pCO_2 increase in littoral zones reached as much as 140 μatm and emitted more CO_2 than pelagic area (Roehm, 2005). In addition, our pCO_2 in littoral zones of the Tibetan Plateau was comparable to the pCO_2 in littoral zone of Lake Baikal (620 to 1,070 μatm; Domysheva et al., 2013).

The partial pressures of CH_4 (pCH_4) varied greatly from 1.2 μatm in Dangreyong Co to 1,364.4 μatm in Lhanag Tso, with an average of 139.8 ± 335.6 μatm (Figure 2). This large divergence of pCH_4 was likely due to the various production mechanisms of CH_4 in lakes with diverse physical and chemical characteristics, especially during the summer stratification, when anoxic conditions developed in the hypolimnion (Bastviken et al., 2008). Compared to that in other studies, a range this wide is not unexpected. For example, the pCH_4 of a catchment in subarctic Sweden exhibited large variation from 150 to 2,980 μatm with an average of 580 ± 710 μatm, which was higher than our average and could be attributed to a comparatively organic-rich sediment, extensive distribution of carbon-rich permafrost, and intensified regional climate warming (Lundin et al., 2013). Large variation of the pCH_4 was also found among the lakes in arctic Alaska (range from 37 to 70,590 μatm, with an average of 272 ± 50 μatm) (Kling et al., 1992). However, the average pCH_4 in the present study was higher than those of a boreal humic oligotrophic lake (22 ± 0.76 μatm) and clear-water productive lake (37 ± 0.56 μatm) during the whole growing season in Finland (Rantakari et al., 2015).

The partial pressures of N_2O (pN_2O) in this study varied from 0.14 to 0.5 μatm, with an average of 0.3 ± 0.1 μatm (Figure 2). A direct comparison of pN_2O was not available because relatively few studies have addressed pN_2O in the lakes. However, the littoral zones of a boreal lake in Finland and the littoral zones of a lake on the Qinghai-Tibetan Plateau were N_2O hotspots relative to the lakes in our study (Chen et al., 2011; Huttunen et al., 2003).

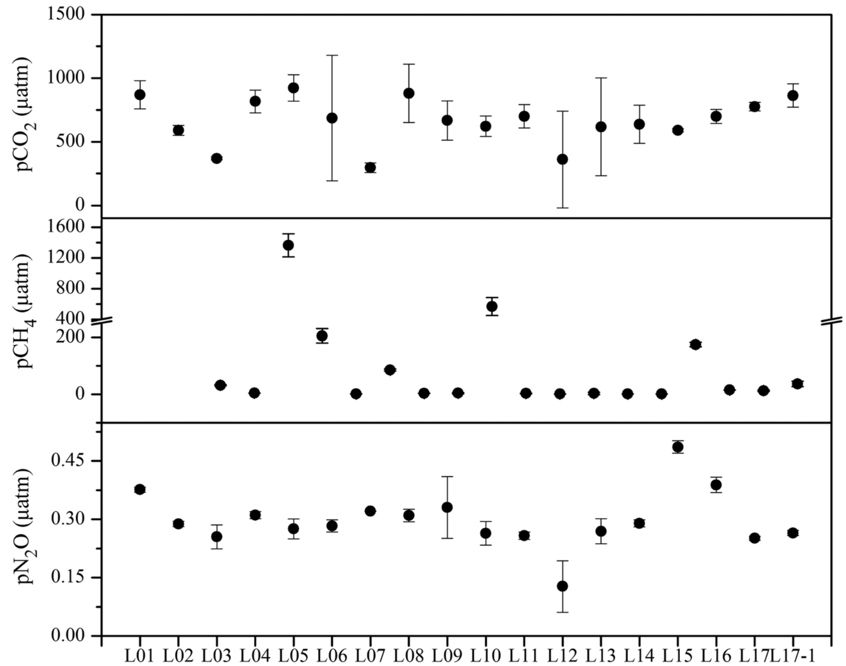

Figure 2. Partial pressures of CO_2, CH_4, and N_2O in the studied lakes on the Tibetan Plateau. Some bars are small and hidden by symbols.

3.2. Diffusive Fluxes of CO_2, CH_4, and N_2O and Potential Influencing Factors

3.2.1. Diffusive Fluxes of CO_2 and Potential Influencing Factors

The peak value of the CO_2 diffusive flux was discovered in Serling Co (245.3 mmol · m^{-2} · day^{-1}), the largest lake in Tibet, with relatively high DOC and DIC concentrations, while the smallest value was found in Chabo Co (3.1 mmol · m^{-2} · day^{-1}), a small saline lake located on the southwestern Tibetan Plateau (Table 2). This flux variation was smaller than that of global saline lakes (−286–9,860 mmol · m^{-2} · day^{-1}) despite comparable average values (Duarte et al., 2008; Table 3). However, the variation and average value of lakes in this study were larger than those of lakes across arctic Alaska (range from −5.5 to 59.8 mmol · m^{-2} · day^{-1}) (Kling et al., 1991), lakes in subarctic discontinuous and arctic continuous permafrost regions (range from −20.5 to 114.4 mmol · m^{-2} · day^{-1}) (Laurion et al., 2010) and reservoirs in subalpine and alpine regions (range from 3.0 to 57.1 mmol · m^{-2} · day^{-1} (Diem et al., 2012; Table 3). The average CO_2 flux we calculated was also comparable to that of lakes in the semihumid and semiarid regions of northeastern China (−16.0–505.9 mmol · m^{-2} · day^{-1}), where most of the lakes were eutrophic (Wen et al., 2016). Evidently, multiple factors such as lake size, chemistry, trophic status, and local climate all influence relative rates of CO_2 diffusive flux, though further studies are needed to elucidate the role of each of these factors in regulating the lake CO_2 diffusive flux.

CO_2 in lakes is derived from multiple sources, such as respiration in the sediment and water column, decomposition and photooxidation of organic carbon, and oxidation of CH_4 in deep water column (Bastviken et al., 2004, 2008; Tranvik et al., 2009). A previous study found that CO_2 supersaturation in lakes could be explained very well by climatically induced organic carbon export from other systems to lakes (Sobek et al., 2005). DIC, which is generally composed of dissolved CO_2, CO_3^{2-}, and HCO_3^-, has been shown to be an important determinant of the CO_2 exchange across the air-water interface of certain lakes (Weyhenmeyer et al., 2015). Other influencing factors, such as lake morphology, hydrology, and climate characteristics, including evaporation and the supply of lake water, may also affect the CO_2 fluxes among lakes (Song et al., 2013). Based on the available data in this study, we proposed that CO_2 diffusive fluxes were partly influenced by DOC, DON, water salinity, and water temperature because of the significantly positive relationships between these variables and CO_2 diffusive fluxes (Table 4). These trends suggest that the relatively high DOC concentration (average 10.0 mg/L) and warm water temperature (average 12.8°C) among

Table 2
Chemical Parameters and Diffusive Fluxes of CO_2, CH_4 and N_2O in the Studied Lakes

No.	Name	DOC (mg/L)	DIC (mg/L)	DON (mg/L)	pH	Salinity	CO_2 flux ($mmol \cdot m^{-2} \cdot day^{-1}$)	CH_4 flux ($mmol \cdot m^{-2} \cdot day^{-1}$)	N_2O flux ($\times 10^{-2}$ $mmol \cdot m^{-2} \cdot day^{-1}$)
L01	Yamzho Yumco	6.3	114.1	0.57	9.19	1.34	75.0 ± 12.9	1.2 ± 0.07	0.6
L02	Angrenjin Co	1.9	28.3	0.35	9.75	1.49	45.2 ± 4.9	0.1 ± 0.01	0.34 ± 0.02
L03	Lhanag Tso	1.2	9.2	0.12	9.24	0.96	8.2 ± 0.7	24.8 ± 2.7	0.11 ± 0.05
L04	Mapam Yumco	2.8	39.4	0.31	9.07	0.26	76.3 ± 11.6	8.6 ± 1.1	0.45 ± 0.03
L05	Nieer Co	56.5	99.1	5.69	8.59	35.55	156.4 ± 23.6	0.09 ± 0.08	0.53 ± 0.16
L06	Cangmu Co	8.7	168.3	0.76	10.2	1.59	23.1 ± 25.0	1.39 ± 0.06	0.13 ± 0.02
L07	Chabo Co	2.4	110.6	0.89	9.64	1.04	3.1 ± 1.7	0.03 ± 0.006	0.17 ± 0.007
L08	Dong Co	18.3	242.6	2.30	8.82	3.72	62.1 ± 21.9	0.11 ± 0.07	0.32 ± 0.04
L09	Dawa Co	17.4	144.5	1.67	9.88	2.77	106.8 ± 37.3	49.7 ± 10.2	0.97 ± 0.53
L10	Zhari Nam Co	6.7	378.3	0.74	9.83	8.16	83.1 ± 16.7	0.16 ± 0.04	0.48 ± 0.18
L11	Dangreyong Co	6.4	313.2	0.63	10.1	6.43	175.2 ± 34.2	0.04 ± 0.007	0.75 ± 0.10
L12	Norma Tso	8.2	816.5	1.12	10.2	1.05	47.0 ± 13.6	0.19 ± 0.05	0.26 ± 0.01
L13	Dogze Co	11.0	1266	1.29	10.2	15.65	41.0 ± 40.5	0.02 ± 0.006	0.24 ± 0.09
L14	Serling Co	5.9	239.9	0.53	9.9	18.21	245.3 ± 90.2	0.04 ± 0.009	1.71 ± 0.15
L15	Xingxinghai	5.8	41.0	—	8.53	0.35	31.6 ± 1.2	5.2 ± 0.2	0.53 ± 0.03
L16	Ngoring Lake	3.7	40.17	—	7.95	0.28	36.8 ± 4.4	0.4 ± 0.005	0.44 ± 0.04
L17	Qinghai Lake-1	8.3	240.5	—	9.01	9.89	44.7 ± 3.0	0.45 ± 0.1	0.08 ± 0.01
L17–1	Qinghai Lake-2	9.8	247.1	—	9.03	9.39	65.3 ± 10.0	1.8 ± 0.5	0.14 ± 0.02

"—" = no data.

study lakes on the Tibetan Plateau promote DOC decomposition. In addition, the significant relationship between CO_2 diffusive fluxes and the DOC concentration may also be an indicator of heterotrophic activities in these lakes. There was no relationship between DIC concentrations and CO_2 diffusive fluxes despite the high average DIC concentration (252.2 mg/L; Table 2), probably because most of the lakes in this study are saline lakes with high pH; Therefore, DIC in these lakes is present mostly in the form of CO_3^{2-} or HCO_3^- rather than CO_2 (Duarte et al., 2008). However, DIC in saline lakes is used as the primary carbon source for photosynthesis and the generation of organic matter, supporting active biologic communities (Song et al., 2013). Therefore, DIC may indirectly contribute to the lake CO_2 flux.

Table 3
CO_2, CH_4, and N_2O Diffusive Fluxes From Lakes on the Tibetan Plateau and Other Studied Lakes

	Site	No. or area (km^2) of lake	Flux ($mmol \cdot m^{-2} \cdot day^{-1}$)	Reference
CO_2	Lakes on the Tibetan Plateau	*N* = 17	73.7 (0.9–295.3)	This study
	Global lakes	*N* = 10	20.9 ± 4.08	Holgerson and Raymond (2016)
	Global saline lakes	*N* = 196	80	Duarte et al. (2008)
	Lakes across arctic Alaska	*N* = 25	20.9 ± 3.3	Kling et al. (1991)
	Lakes in a subarctic catchment, northern Sweden	*N* = 27	15 ± 9.2	Lundin et al. (2013)
	Alpine reservoirs in the Alps	*N* = 5	22 ± 7.7	Diem et al. (2012)
	Lakes in semi-humid/arid regions, northeastern China	*N* = 95	67.6	Wen et al. (2016)
CH_4	Lakes on the Tibetan Plateau	N = 17	5.2 (0.0008–45.9)	This study
	Lakes in Alaska	A = 3.0155×10^4	6.9	Tan and Zhuang (2015)
	Lakes in northern Siberia	A = 1.3976×10^5	6.1	Tan and Zhuang (2015)
	Lakes in northern Europe	A = 1.2917×10^5	0.8	Tan and Zhuang (2015)
	Alpine reservoirs in the Alps	*N* = 11	0.013 ± 0.01	Diem et al. (2012)
	Global lakes	*N* = 18	0.1 ± 0.05	Holgerson and Raymond (2016)
	Littoral zones of a lake on the Tibetan Plateau	N = 1	22.7	Chen et al. (2009)
	Lakes in semi-humid/arid regions, northeastern China	N = 95	2.77	Wen et al. (2016)
N_2O	Lakes on the Tibetan Plateau	N = 17	0.0065 (0.00007–0.0209)	This study
	Littoral zones of a boreal lake	N = 1	0.002–0.012	Huttunen et al. (2003)
	Littoral zones of a lake on the Tibetan Plateau	N = 1	0.044	Chen et al. (2011)
	A hyper-eutrophic lake in China	N = 1	−0.15–1.2	Wang et al. (2006)
	Alpine reservoirs in the Alps	*N* = 4	0.0012 ± 0.0003	Diem et al. (2012)

Table 4
Pearson Correlation Between Diffusive Fluxes (mmol · m^{-2} · day^{-1}) of Three Greenhouse Gases and DIC, DOC, DON, pH, Salinity, Water Temperature (WT), Lake Area, and Depth

	DIC (mg/L)	DOC (mg/L)	DON (mg/L)	pH	Salinity (mg/L)	WT (°C)	Area (m^2)	Depth (m)	Elevation (m a.s.l)
CO_2 flux (*N* = 16)	0.228	0.788[a]	0.731[a]	−0.236	0.670[a]	0.515[b]	0.009	0.320	0.182
CH_4 flux (N = 18)	−0.242	0.027	−0.051	0.159	−0.226	0.210	−0.184	−0.019	0.156
N_2O flux (N = 18)	0.196	0.066	0.002	0.090	0.279	0.107	0.019	0.664[b]	0.337

Note. L11 and L14 were excluded in CO_2 relationship analyses due to the high instantaneous wind velocities.
[a]Correlation is significant at the 0.05 level (two-tailed). [b]Correlation is significant at the 0.01 level (two-tailed).

3.2.2. Diffusive Fluxes of CH_4 and Potential Influencing Factors

The average CH_4 diffusive flux of the study lakes was 5.2 mmol · m^{-2} · day^{-1}, with variation from 0.02 mmol · m^{-2} · day^{-1} in Dogze Co, a deep lake, to 49.7 mmol · m^{-2} · day^{-1} in Dawa Co, a shallow lake with an average water depth of 2 m (Table 2). In general, the CH_4 diffusive flux we observed was comparable to those of the lakes in Alaska and northern Siberia, despite the extensive distribution of yedoma thermokarst lakes in these two regions (Tan & Zhuang, 2015), but higher than those of lakes in northern Europe (Tan & Zhuang, 2015), lakes in the semihumid and semiarid regions of northeastern China (Wen et al., 2016) and global lakes and Alpine reservoirs in the Alps (Table 3). The variation in the CH_4 flux of lakes in the present study was larger than those of the lakes in arctic Alaska (0.08–1.02 mmol · m^{-2} · day^{-1}; Kling et al., 1992), Alpine reservoirs in the Alps (0–0.13 mmol · m^{-2} · day^{-1}) and global lakes (varied from 0.06 mmol · m^{-2} · day^{-1} in lakes larger than 100 km^2 to 2.28 mmol · m^{-2} · day^{-1} in lakes smaller than 0.001 km^2; Holgerson & Raymond, 2016). This difference probably arose because lakes in the present study were stratified during the summer sampling period, creating anoxic conditions, and a large volume of CH_4 was generated as the final product of carbon metabolism (Kirillin et al., 2017; Li et al., 2005). Large divergences of the average CH_4 flux and flux variation were found between the present study and the littoral zones of Huahu Lake in the Zoige National Wetland Reserve on the Qinghai-Tibetan Plateau because the latter has high DOC input from nearby areas and emergent plant vegetation as a substrate for methanogens to produce CH_4 (Chen et al., 2009; Table 3).

CH_4 is a major product of carbon metabolism by methanogens in anaerobic water conditions (Bastviken et al., 2008). CH_4 in lakes has at least four different emission pathways: bubble flux (ebullition), diffusive flux, storage flux, and flux through aquatic plants (Bastviken et al., 2004). Generally, shallow and epilimnetic lake sediments largely contributed to CH_4 emissions. During the summer stratification, epilimnetic sediments even accounted for 100% of CH_4 emissions, as stratification limited the potential for CH_4 oxidation by methanotrophic bacteria (Bastviken et al., 2004, 2008). Compared to CO_2, CH_4 is a less soluble gas and more prone to ebullition (Sander, 2015). For instance, Bastviken et al. (2008) found that the CH_4 diffusive flux accounted for approximately 35%–92% of bubble emissions from three lakes of the United State during the summer period. Therefore, CH_4 fluxes were likely underestimated in this study because CH_4 bubble emissions were not considered. In addition, concentrations of organic matter and the activity of methanogens are two other important factors in CH_4 formation. Although DOC concentrations in this study were high, no relationship was discovered between the CH_4 fluxes and DOC concentrations, possibly because the high pH in these saline lakes inhibited methanogen activity (Wen et al., 2016). Another plausible interpretation of this lack of environmental predictors was that the relationships between environmental factors and CH_4 emissions may not be valid at very large scales but instead suitable only for the individual study region with measurement duplications due to regionally variable factors influencing CH_4 emissions (Ortiz-Llorente & Alvarez-Cobelas, 2012). Furthermore, potential CH_4 oxidation rates may also regulate the final CH_4 diffusive flux (Bastviken et al., 2004) because CH_4 can be oxidized under aerobic conditions into CO_2 (Bastviken et al., 2004, 2008) and by SO_4^{2-} into HCO_3^- simultaneously (Avrahamov et al., 2014), and most lakes in the present study are of the sulfate type (Wang & Dou, 1998).

3.2.3. Diffusive Fluxes of N_2O and Potential Influencing Factors

The average N_2O diffusive flux of the studied lakes was 0.0065 mmol · m^{-2} · day^{-1}, with the highest diffusive flux in Serling Co (0.017 mmol · m^{-2} · day^{-1}) and the lowest value in Qinghai Lake (0.0008 mmol · m^{-2} · day^{-1}) (Table 2). The N_2O diffusive flux we observed was lower than those observed previously across eutrophic or hyper-eutrophic lakes (varied from −0.15 to 1.2 mmol · m^{-2} · day^{-1}; Wang et al., 2007), within the littoral

Table 5
Uncertainties of Applying Different K Models in GHG Flux Estimations

Approach	CO_2 (mmol · m^{-2} · day^{-1})	CH_4 (mmol · m^{-2} · day^{-1})	N_2O (*10^{-3} mmol · m^{-2} · day^{-1})
$K_{600} = 2.07 + 0.215U^{1.7}$	3.1–139.1	0.0036–35.0	0.5–10.9
$K_{600} = 0.168 + 0.228U^{2.2}$	0.4–320	0.0053–34.6	0.5–24.6
$K_{600} = 2.51 + (1.48 \times U) + (0.39 \times U \times \log_{10}LA) \times 0.228U^{2.2}$	7.7–329.5	0.0078–84.2	0.9–25.8

zones of a boreal lake (0.002–0.012 mmol · m^{-2} · day^{-1}) and within the littoral zones of a Tibetan lake (−0.038–0.19 mmol · m^{-2} · day^{-1}), but greater than that of Alpine reservoirs (0.0009–0.00163 mmol · m^{-2} · day^{-1}; Chen et al., 2011; Diem et al., 2012; Huttunen et al., 2003; Table 3). This distinction was likely due to the remoteness and the pristine nature of the lakes on the Tibetan Plateau as well as the lack of measurement duplication.

As a potent greenhouse gas, N_2O is produced via aerobic nitrification and anaerobic denitrification in the lake sediments (Ciais et al., 2014). Several factors, including the nitrification rate (limited by oxygen content), nitrate supply, and activity of denitrifying bacteria may influence N_2O emissions (Huttunen et al., 2003). Generally, incomplete denitrification of nitrate is considered the most important source of N_2O efflux (Sasaki et al., 2011; Zhu et al., 2015). Under completely anoxic conditions, denitrifiers will reduce N_2O to N_2, providing a sink for N_2O (Zhu et al., 2015). Although the nitrate content and temperature are positive regulators of denitrification, oxygen constrains the conversion of N_2O to N_2 more strongly than the conversion of NO_3^- to N_2O (Mengis et al., 1997), which is probably the reason that maximum N_2O concentrations occur within the oxic-anoxic layer (Mengis et al., 1997; Zhu et al., 2015). A positive relationship was found between N_2O diffusive fluxes and maximum lake water depths in this study (Table 4). The relationship may exist because some of these saline lakes (e.g., Dogze Co, Dangreyong Co, and Zhari Nam Co) are meromictic lakes with both a thermocline and a halocline (Li et al., 2005; Wang et al., 2014) or have the characteristics of meromictic lakes (i.e., increased conductivity and decreased dissolved oxygen with increasing water depth; Wang et al., 2010; Wang et al., 2014). Therefore, large salinity gradients combined with relatively deep water in the saline lakes we studied facilitated stratification regimes that created relatively stable anoxic conditions for denitrification (Li et al., 2005; Wang et al., 2014). However, the concentrations of NO_3^-, NH_4^+, and NO_2^- in most of the studied lakes were very low.

3.3. The Uncertainties of GHG Estimations and Importance of Lakes on the Tibetan Plateau as GHG Sources

This is a preliminary study of the GHGs of lakes on the Tibetan Plateau, in which the direct measurement of GHGs was adopted. All the samples were collected in the littoral zone of each lake, an interface between the catchment, the lake, and the atmosphere. Littoral zones have been proven to be hot spots of GHG emissions (Chen et al., 2011; Huttunen et al., 2003; Wang et al., 2006), which is likely the main reason that the results in this study were relatively high. For example, the average CH_4 efflux from the littoral zones of these lakes was comparable to that from a swamp on the Tibetan Plateau (Wei et al., 2015). However, with the limited data on GHG fluxes, we could not estimate the total GHG effluxes from littoral zones of lakes or all lakes on the Tibetan Plateau. Therefore, this preliminary study provides a foundation for future research of lake GHG fluxes on the Tibetan Plateau.

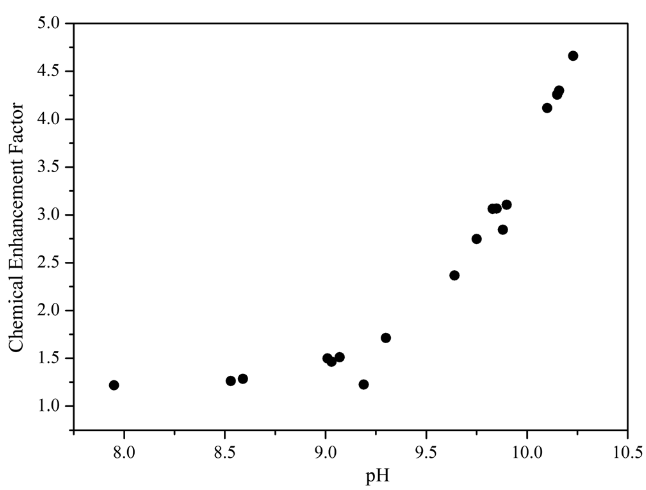

Figure 3. The relationship between the chemical enhancement and pH in saline lakes on the Tibetan Plateau.

Three different *k* models from Cole and Caraco (1998), Crusius and Wanninkhof (2003), and Vachon et al. (2010) were used to calculate the diffusive fluxes of CO_2, CH_4, and N_2O. For large lakes, the model from Cole and Caraco (1998) normally generated the lowest *k* values, while the model from Crusius and Wanninkhof (2003) generally produced the largest range of *k* value, and the model from Vachon et al. (2010) produced a range of *k* values intermediate to the other two models (Dugan et al., 2016). The uncertainty intervals in simultaneously applying three models among studied lakes were 0.9–295.3 mmol · m^{-2} · day^{-1}, 0.0008–45.9 mmol · m^{-2} · day^{-1}, and 0.07–20.9 μmol · m^{-2} · day^{-1} for flux of CO_2, CH_4, and N_2O, respectively, from Monte Carlo simulations (Tables 3 and 5), which suggests the need for more spatiotemporally resolved techniques, such as eddy-covariance towers and automated floating chambers (Ojala et al., 2011; Vesala et al., 2006). In this study, only one sampling site at each lake was studied, and the sampling times of each site were different, both of which can cause uncertainty (Demarty et al., 2009). Further study that focused on Nam Co showed that the spatial and diurnal variations of GHG for a given lake on the Tibetan Plateau were nonsignificant over a 2-day period in 2017 (see supporting information, Figures S1 to S3), suggesting that the results were likely representative in this study period. However, the remote nature of the lakes in this study prevented spatially and temporally representative sampling using our manual technique. Further study should be performed continually at an hourly frequency across seasons using autonomous techniques, with three or more sampling sites within the zone of each lake to be assessed.

As most lakes in this study have high salinity and pH, chemical enhancement could increase the CO_2 exchange with the atmosphere by an average of 2.5-fold (ranging from 1.2 to 4.7) above that of freshwater lakes with equivalent CO_2 concentrations (Figure 3). The enhancement factor we discovered was comparable with the value provided in Duarte et al. (2008; average of 2.3). Accordingly, in this study, higher-average CO_2 fluxes were found in saline lakes than in freshwater lakes. It is predicted that climate warming due to elevated GHG emissions will be strongly enhanced on the Tibetan Plateau (Chen et al., 2002) and cause a series of feedbacks. For example, the glaciers on the Tibetan Plateau are experiencing rapid melting (Yao et al., 2012), which may result in the increased contribution of downstream lakes and wetlands to GHG emissions and greater exports of bioavailable DOC (Hood et al., 2015; Wei & Wang, 2017; Yan et al., 2016). Moreover, there is approximately 1.73×10^6 km^2 of permafrost distributed on the Tibetan Plateau, most of which is predicted to experience degradation under climate warming scenarios (Yang et al., 2010). Widespread permafrost degradation may trigger further deterioration locally and globally, with a loss of carbon from permafrost to the lotic and lentic water system (Qu, Sillanpaa, et al., 2017) or a direct carbon loss to the atmosphere, ultimately leading to increased atmospheric carbon stocks, accelerated warming, and a positive feedback to climate change (Yang et al., 2010).

4. Conclusion

We investigated the partial pressures, diffusive fluxes, and environmental factors influencing CO_2, CH_4, and N_2O in littoral zones of lakes on the Tibetan Plateau in May 2014 and August 2015. We found littoral zones of lakes on the Tibetan Plateau to be sources of CO_2, CH_4, and N_2O with average efflux rates of 73.7, 5.2, and 0.0065 mmol · m^{-2} · day^{-1}, respectively. The average CO_2 efflux rate observed in this study was comparable with that of saline lakes globally and was likely influenced by DOC concentations, DON concentrations, salinity, and water temperature. CO_2 exchange with the atmosphere was chemically enhanced in saline lakes, indicating the important role of these lakes in terms of global CO_2 emissions. The CH_4 efflux rate varied largely across the Tibetan Plateau due to various factors, including lake stratification, water depth, oxic layers, and salinity. N_2O efflux rates were related to the maximum lake water depths. The *k* models, single-point sampling and sampling locations (littoral zones) may have contributed to uncertainties in the estimate of GHG emissions from these saline lakes. In order to make a more accurate estimates of GHG exchange across the air-atmosphere boundary, future studies should include parameters optimized to *k* models and sampling programs with greater spatiotemporal resolution. The Tibetan Plateau is vulnerable to ongoing and projected climate change. GHG emissions from lakes on the Tibetan Plateau will propagate positive feedback cycles, which will promote glacier melting and permafrost degradation. Therefore, the biogeochemical cycles of saline lakes on the Tibetan Plateau should be the focus of future study to inform both global climate change and regional environmental predictions.

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