

## RESEARCH ARTICLE

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## Key Points:

- The first annual CH<sub>4</sub> fluxes data from an alpine wetland on the Tibetan Plateau
- The data reveal the importance of CH<sub>4</sub> fluxes during the nongrowing season
- CH<sub>4</sub> fluxes are correlated with temperature, water table, and CO<sub>2</sub> fluxes

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## Methane emissions from an alpine wetland on the Tibetan Plateau: Neglected but vital contribution of the nongrowing season

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**Abstract** The vast wetlands on the Tibetan Plateau are expected to be an important natural source of methane (CH<sub>4</sub>) to the atmosphere. The magnitude, patterns and environmental controls of CH<sub>4</sub> emissions on different timescales, especially during the nongrowing season, remain poorly understood, because of technical limitations and the harsh environments. We conducted the first study on year-round CH<sub>4</sub> fluxes in an alpine wetland using the newly developed LI-COR LI-7700 open-path gas analyzer. We found that the total annual CH<sub>4</sub> emissions were 26.4 and 33.8 g CH<sub>4</sub> m<sup>-2</sup> in 2012 and 2013, respectively, and the nongrowing season CH<sub>4</sub> emissions accounted for 43.2–46.1% of the annual emissions, highlighting an indispensable contribution that was often overlooked by previous studies. A two-peak seasonal variation in CH<sub>4</sub> fluxes was observed, with a small peak in the spring thawing period and a large one in the peak growing season. We detected a significant difference in the diurnal variation of CH<sub>4</sub> fluxes between the two seasons, with two peaks in the growing season and one peak in the nongrowing season. We found that the CH<sub>4</sub> fluxes during the growing season were well correlated with soil temperature, water table depth and gross primary production, whereas the CH<sub>4</sub> fluxes during the nongrowing season were highly correlated with soil temperature. Our results suggested that the CH<sub>4</sub> emission during the nongrowing season cannot be ignored and the vast wetlands on the Tibetan plateau will have the potential to exert a positive feedback on climate considering the increasing warming, particularly in the nongrowing season in this region.

### 1. Introduction

The increase of greenhouse gas (GHG) concentration in the atmosphere is a major cause of the observed rise in land and ocean surface temperature. As one of the most important GHGs exchanged between terrestrial ecosystems and the atmosphere, methane (CH<sub>4</sub>) has a global warming potential 28 times that of CO<sub>2</sub> on a 100 year time horizon [IPCC, 2013]. Thus, CH<sub>4</sub> plays a key role in the greenhouse effects on Earth's atmosphere. A recent study showed that the global atmospheric CH<sub>4</sub> concentration has increased from 700 ppb in preindustrial times to over 1800 ppb in 2012 [Nisbet *et al.*, 2014]. The rapid increase in atmospheric CH<sub>4</sub> concentrations has generated significant interest in quantifying CH<sub>4</sub> emissions from various sources.

Wetlands that occupy only 5–8% of Earth's land surface but contain more than 30% of the world's soil carbon [Mitra *et al.*, 2005; Zedler and Kercher, 2005; Malone *et al.*, 2013; Mitsch *et al.*, 2013] are significant sources of CH<sub>4</sub> emissions [Whalen, 2005; Bridgman *et al.*, 2013]. In general, CH<sub>4</sub> fluxes from wetlands are a balance of CH<sub>4</sub> production in the anaerobic zone, oxidation in the aerobic zone, and transport from soils into the atmosphere by diffusion and bubbling and through vascular plants [Dacey and Klug, 1979; Whiting and Chanton, 1992, 1996; Chanton *et al.*, 1997; Segers, 1998; Joabsson *et al.*, 1999]. A number of studies have examined CH<sub>4</sub> fluxes using various methods and documented that CH<sub>4</sub> fluxes are linked to a number of biological (e.g., plant phenology and production, microbial activities) and physical (e.g., temperature, wind speed and water table) processes at different temporal and spatial scales [Whiting and Chanton, 1996; Segers, 1998; Brumme and Borken, 1999; Öquist and Svensson, 2002; Megonigal and Guenther, 2006; Olefeldt *et al.*, 2013]. Key factors and processes that control the wetland CH<sub>4</sub> emissions often differ among ecosystems, adding more complexities to the estimation of CH<sub>4</sub> emissions at regional and global scales [Bridgman *et al.*, 2013].

It has been estimated that an average of 217 Tg CH<sub>4</sub>/yr, ranging from 177 to 281 Tg CH<sub>4</sub>/yr, is emitted from natural wetlands, accounting for 32% of the total global surface emissions [Fletcher *et al.*, 2004a, 2004b; Zhuang *et al.*, 2004; Evans, 2007; Kirschke *et al.*, 2013]. However, significant uncertainties exist when estimating current and future CH<sub>4</sub> emissions from wetlands for a number of reasons. First, there are limited data on CH<sub>4</sub> fluxes for various wetlands. Recent studies of CH<sub>4</sub> fluxes have focused mainly on high-latitude wetlands because boreal and arctic wetlands store immense amounts of carbon in prevalent waterlogged sediments [Gorham, 1991; Soegaard and Nordstroem, 1999; Bridgham *et al.*, 2006; Peregon *et al.*, 2008; Tarnocai *et al.*, 2009]. However, research on CH<sub>4</sub> fluxes in high-altitude wetland ecosystems, especially in remote areas, remains limited. Second, many previous evaluations on regional or global CH<sub>4</sub> fluxes were based on the results of experiments carried out using the manual static chambers method [Whiting and Chanton, 1993; Nykänen *et al.*, 2003; Chen *et al.*, 2013a, 2013b]. As reported previously, this method may introduce so-called “chamber effects” [Mosier, 1990], potentially modifying the temperature and moisture in the chamber and altering the gas diffusion gradient within the soil profile. Furthermore, the gas fluxes measured with chamber methods cover small patches of soil, which are usually not sufficient to make a full-scale understanding of the carbon exchanges of the heterogeneous ecosystems. In contrast, the eddy covariance method is one of the most useful methods to measure the carbon exchange between the atmosphere and ecosystems over a large area. This micrometeorological approach has been widely used to measure net CO<sub>2</sub> fluxes (net ecosystem exchange). However, studies on CH<sub>4</sub> fluxes using this method are still limited (but see Rinne *et al.* [2007], Gažovič *et al.* [2010], Long *et al.* [2010], Tokida *et al.* [2011], and Baldocchi *et al.* [2012]). Finally, existing studies have mainly focused on the growing season when most of the CH<sub>4</sub> is released. Recent studies reported that during the nongrowing season, particularly in the soil freezing and thawing periods, large CH<sub>4</sub> emissions pulses occurred [Friborg *et al.*, 1997; Huttunen *et al.*, 2003; Tokida *et al.*, 2007; Mastepanov *et al.*, 2008; Song *et al.*, 2012]. Thus, a significant error can occur in evaluating annual CH<sub>4</sub> fluxes if we neglect the fluxes during the nongrowing season, although this statement is open to debate because data reported in the above mentioned studies are often characterized with low time frequency (once per month) and short-term (weeks or months) coverage. To improve our ability to accurately quantify the nongrowing season CH<sub>4</sub> fluxes and their potential responses to climate change, field measurements with high frequency and multiyear coverage are needed.

The Tibetan Plateau, also known as the “Water Tower of Asia,” is the highest and largest plateau on the Earth with an average altitude of ~4000 m above sea level and an area of ~2.5 × 10<sup>6</sup> km<sup>2</sup>, of which nearly 50,000 km<sup>2</sup> is wetlands [Zhao *et al.*, 2010]. According to climate records, the Tibetan Plateau is experiencing substantial climate changes in recent decades with an increase of 0.3 and 0.5°C per decade in annual and winter temperatures, respectively [Piao *et al.*, 2010]. The pronounced climate warming, particularly in the nongrowing season, has been causing numerous alterations to the alpine wetland environments, such as shortening the freezing period, increasing the length of the growing season, and deepening the layer of soil activity [Yang *et al.*, 2010; Piao *et al.*, 2011]. These changes in hydrological and environmental conditions indicate that the role of the alpine wetlands in terrestrial carbon cycling is likely to be affected, especially given that the carbon balance of alpine wetlands on the Tibetan Plateau appears to be sensitive to small environmental changes [Tang *et al.*, 2009].

On the Tibetan Plateau, researchers have conducted a number of studies on CH<sub>4</sub> fluxes of alpine wetlands [Jin *et al.*, 1999; Hirota *et al.*, 2004, 2005; Wang *et al.*, 2004, 2009; Chen *et al.*, 2010, 2013a; Zhu *et al.*, 2011; Deng *et al.*, 2013; Kato *et al.*, 2013]. Some preliminary efforts have been made to estimate the annual or regional emissions from the alpine wetlands on the Tibetan Plateau [Chen *et al.*, 2013a, 2013b]. However, these studies were either carried out using low-resolution measurements with traditional static methods or focused only on growing season CH<sub>4</sub> fluxes. Given that the alpine wetland ecosystems on the Tibetan Plateau are dominated by a long nongrowing season and are typically ice-covered for nearly half of the year, we argue that CH<sub>4</sub> emissions from alpine wetland soils during the nongrowing season is likely an important component of annual CH<sub>4</sub> fluxes. Accurate studies that investigate the magnitude, patterns, and environmental controls of the CH<sub>4</sub> fluxes from Tibetan Plateau wetlands on different timescales, especially during the nongrowing season, are urgently needed. In addition, alpine wetlands of the Tibetan Plateau and high-latitude wetlands share many similar features, such as a long nongrowing season, large soil carbon storage, and high carbon density [Wille *et al.*, 2008; Zona *et al.*, 2009; Bai *et al.*, 2010; Sachs *et al.*, 2010]. Thus, a CH<sub>4</sub> emission study from the Tibetan Plateau wetland can provide a valuable reference to understand the dynamics and budget of CH<sub>4</sub> emissions from the high-latitude regions.

In this study, we presented a field experiment conducted in a typical alpine wetland (*Carex*-dominated peatland) on the Tibetan Plateau during two years (2012 and 2013) and measured CH<sub>4</sub> fluxes using the eddy covariance technique (LI-COR LI-7700). The data presented here are the first year-round CH<sub>4</sub> fluxes from an alpine wetland on the Tibetan Plateau. Our aims of this study were to (1) investigate the patterns and environmental drivers of diurnal and seasonal variations of CH<sub>4</sub> fluxes and (2) analyze the annual budget of CH<sub>4</sub> fluxes and the contribution of nongrowing season CH<sub>4</sub> fluxes to the annual budget. We proposed the following hypotheses: (1) the alpine wetlands on the Tibetan Plateau tend to have high fluxes because the high concentration of substrates favors the production of CH<sub>4</sub> and the low atmospheric oxygen concentration reduces the potential for oxidation; and (2) the cumulative CH<sub>4</sub> fluxes during the nongrowing season are large and their annual contribution is high due to the long duration of the nongrowing season.

## 2. Materials and Methods

### 2.1. Site Description

The study was conducted at the Luanhaizi alpine wetland, adjacent to the Haibei Alpine Grassland Ecosystem Research Station, Chinese Academy of Sciences, in Qinghai Province, China (37°35'N, 101°20'E, 3250 m a.s.l.). The climate is characterized by strong solar radiation with long, cold winters and short, cool summers. According to long-term climate data (1981–2010) from meteorological stations, the mean monthly air temperatures ranged from –18.3°C in January to 12.6°C in July, and the mean annual temperature is –1.1°C. Mean annual precipitation is 490 mm, of which more than 80% occurs during the growing season from May to September. The dominant soil type is a silty clay loam of Mat-Cryic Cambisols. The average C and N contents in the 0–30 cm soil layer is 24.5% and 1.46%, respectively. The wetland is an alpine peatland, characterized by a unique landscape with numerous scattered hummocks, influenced by a seasonal freezing-thawing process. The study site is located in the transitional zone from permafrost to seasonally frozen terrain. The vegetation is dominated by *Carex pamirensis*, with several other species including *Carex alofusca*, *Hippuris vulgaris*, *Triglochin palustre*, and *Heleocharis* spp.

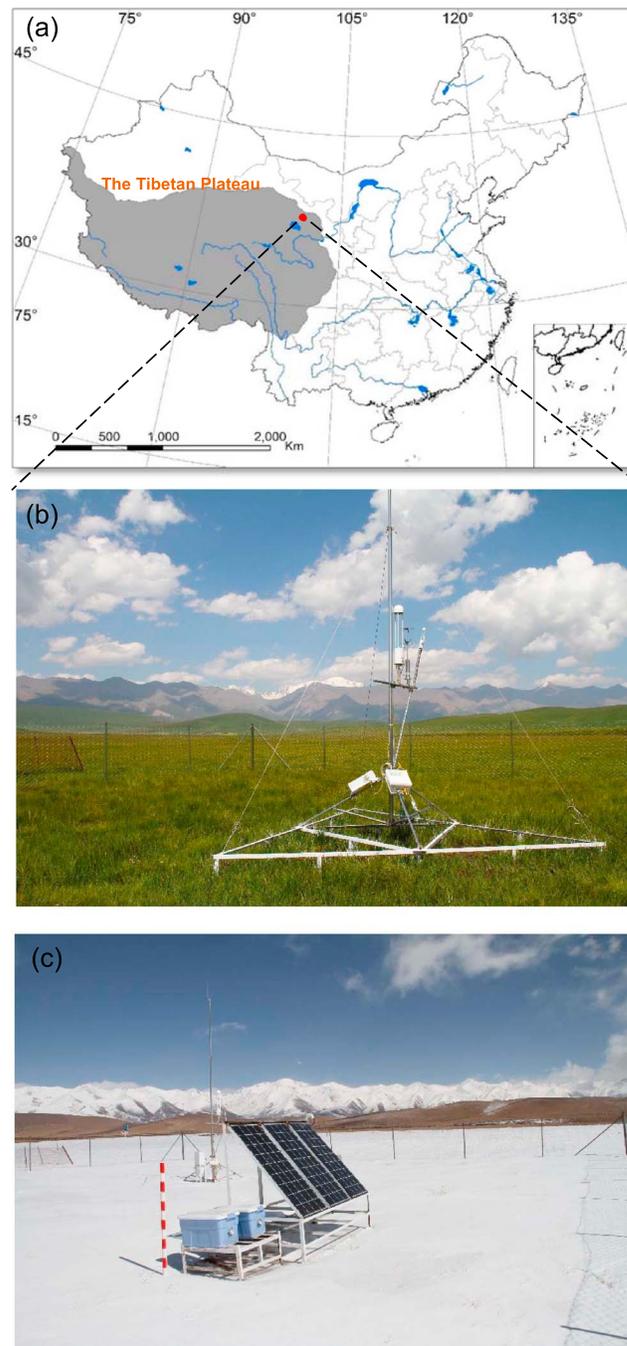
### 2.2. Eddy Covariance and Meteorological and Soil Measurements

CH<sub>4</sub> and CO<sub>2</sub> fluxes were measured from 23 July 2011 to 31 December 2013 using the open-path eddy covariance method at a height of 2.5 m in the center of an open area of radius greater than 1 km (Figure 1). The eddy covariance system consists of a three-axis sonic anemometer (CSAT3; Campbell Scientific Inc. (CSI), Logan, USA), an open-path CO<sub>2</sub>/H<sub>2</sub>O infrared gas analyzer (LI-7500; Li-COR Inc, Lincoln, NE, USA), and an open-path CH<sub>4</sub> infrared gas analyzer (LI-7700; Li-COR). The data from CSAT3, LI-7500 and LI-7700 were recorded at 10 Hz using a datalogger (CR5000, Campbell Scientific, Utah, USA). More information about the measurement procedure was previously described by Yu *et al.* [2013].

Net radiation was measured with a four-component radiometer (CNR-1, Kipp and Zonen, Delft, Netherlands). Photosynthetic photon flux density (PPFD) was measured with a quantum sensor (LI-190SB, LI-COR). Air temperature and relative humidity were measured using HMP45C temperature and relative humidity probes (CSI) with radiation shields. Soil temperature was measured at a depth of 5 cm using a temperature probe (107, CSI). Soil water content reflectometer (CS616, CSI) was installed 10 cm below the soil surface to measure the volumetric soil water content. Wind speed and wind direction were measured at 2.5 m above the ground using a propeller anemometer (CSI), and precipitation (mm) was measured with a tipping bucket rain gauge (TE525MM, CSI). Water table was measured automatically using HOBO dataloggers (Onset Computer, Bourne, MA, USA) in wells made from PVC pipes. Meteorological data were recorded every 30 min by a datalogger (CR1000, Campbell Scientific, Utah, USA) from May 2011 to December 2013. Linear interpolation and redundant measures were used to fill gaps in the meteorological data due to equipment power failures [Falge *et al.*, 2001].

### 2.3. Data Processing

The eddy covariance raw data were postprocessed and quality controlled (tests according to [Foken *et al.*, 2005]) using EddyPro 4.0 software (LI-COR) to compute the CH<sub>4</sub> fluxes over a 30 min interval. The Eddypro software (Express mode) applied the following corrections: Webb-Pearman-Leuning density fluctuations [Webb *et al.*, 1980], spectral corrections [Moncrieff *et al.*, 2005], sonic virtual temperature correction



**Figure 1.** (a) Geographic location (solid circle) of the study site on the Tibetan Plateau: The typical landscapes in (b) the growing season (July 2012) and (c) the nongrowing season (January 2013).

[Van Dijk *et al.*, 2004], and the incorporated frequency response correction [Massman, 2000; Lee *et al.*, 2005]. In addition, in order to filter out half-hourly values from LI-7700 where the instrument was performing poorly, the Eddypro software provides two diagnostic outputs: the received signal strength indicator, which indicates the cleanliness of the mirrors, and a coded value that represents one or more pieces of diagnostic information [McDermitt *et al.*, 2011]. The fluxes footprint was calculated by Eddypro software [Kljun *et al.*, 2004]. Results showed that the source location with maximum contribution to the measured flux was 34 m from the eddy tower, and 90% of the fluxes came from within 101 m of the eddy tower, which confirmed that the fluxes are representative of the wetland area. A data-screening procedure was used to remove anomalous values due to system failures. The data-screening procedure consisted of removing periods when (1) rainfall event occurred; (2) excessive spikes in the infrared gas analyzer data occurred due to precipitation, moisture, frost, and dirt on the sensors; (3) instruments malfunctioned; and (4) friction wind speed ( $u^*$ ) values during nocturnal periods were less than 0.16 [Yu *et al.*, 2013], indicating low turbulence conditions.

Due to missing and discarded data mentioned above, the data gaps of  $\text{CH}_4$  fluxes data during the whole observation period were 34.3%. Currently, there is still no standardized method to gap-fill eddy covariance  $\text{CH}_4$  fluxes data for annual estimations and to improve the analysis of uncertainties associated with annual estimations of  $\text{CH}_4$  fluxes. In addition, methods used to gap-fill fluxes data are often site dependent, including mean replacement (i.e., using mean of observed values to replace

missing data), interpolation and extrapolation (i.e., an estimated value from other observations for the same environmental condition), and nonlinear models such as artificial neural networks [Dengel *et al.*, 2013]. In this study, we filled the data gaps following the method proposed by Falge *et al.* [2001]: (1) gaps of up to 2.5 h were filled by applying a simple linear interpolation method; (2) a mean diurnal variations procedure was used to fill gaps of several hours based on previous and subsequent days; and (3) other data gaps were filled using a semiempirical method (i.e., look-up tables). For look-up tables, the average  $\text{CH}_4$  fluxes were compiled for four periods: growing season, soil freezing, winter, and soil thawing (Table 1). In each period, the average

**Table 1.** The Measurements Were Conducted for Four Different Periods Within a Year in 2012 and 2013

	2012		2013	
	Period (Julian day)	Number of Days	Period (Julian Day)	Number of Days
Growing season	141–271	131	136–273	138
Nongrowing season				
Soil freezing	272–319	48	274–325	52
Winter	1–76 and 320–366	123	1–72 and 326–365	112
Soil thawing	77–140	64	73–135	63

CH<sub>4</sub> fluxes were compiled according to photosynthetic photon flux density (PPFD) and air temperature classes. PPFD classes were defined through 100  $\mu\text{mol m}^{-2} \text{s}^{-1}$  intervals with a separate class for PPFD = 0. Similarly, air temperature classes consisted of 2°C intervals. The gaps were filled using the average CH<sub>4</sub> fluxes with the corresponding PPFD and air temperature class.

#### 2.4. Seasonal and Diurnal Dynamics of CH<sub>4</sub> Fluxes

We divided the CH<sub>4</sub> flux data into four time periods that are related to general stages of physical and biological conditions: (1) growing season, ranging from the first of seven consecutive days with average daily air temperature above 5°C to the first of seven consecutive days with average daily air temperature below 5°C [Lund *et al.*, 2010; Tang and Arnone, 2013]; (2) soil freezing, ranging from the end of the growing season to the first of two consecutive days with average daily soil temperature below 0°C at 5 cm depth; (3) winter, starts at the end of the soil freezing period and ends when the snow melts out; and (4) soil thawing, time period between winter and the growing season. We also analyzed the different seasons for diurnal variations. Only days with more than 70% data coverage were used in the analysis.

#### 2.5. Statistical Analysis

We conducted path analysis to evaluate the relative importance of these environmental variables in affecting the seasonal and annual CH<sub>4</sub> fluxes dynamics using the AMOS 7.0 (Amos Development, Spring House, PA, USA). Path analysis is often used to estimate the magnitude and significance of hypothesized causal connections among variables. Net CH<sub>4</sub> emissions from soils into the atmosphere involve three processes: CH<sub>4</sub> production, oxidation by microbes, and CH<sub>4</sub> transportation. These processes are associated with various environmental variables including air temperature, soil temperature, water table, photosynthetic photon flux density (PPFD), vapor pressure deficit (VPD), and friction wind velocity.

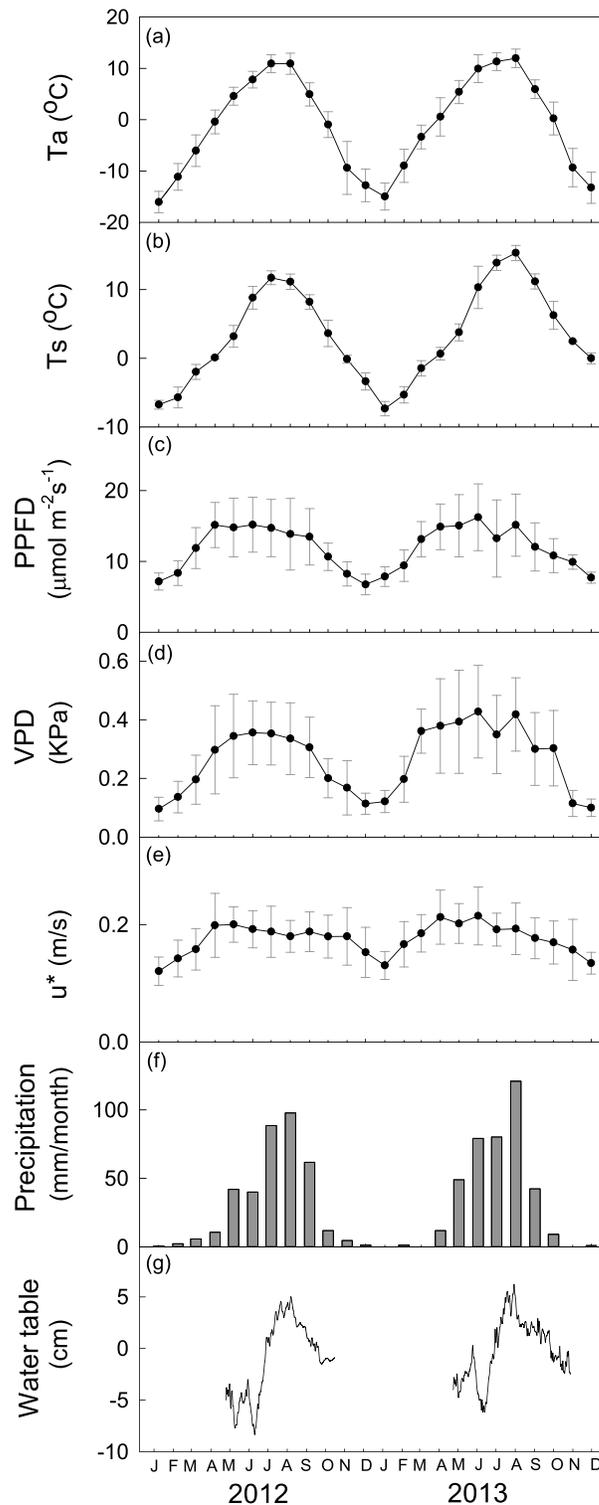
To estimate the path coefficients for the effects of each environmental variable on CH<sub>4</sub> fluxes, we first standardized all variables by subtracting the mean of each variable and dividing by the standard deviation of each variable [Schemske and Horvitz, 1988]. Thus, standardization makes the values of each variable have zero mean and unit variance and allows the strength of linear relationships to be compared.

Linear and exponential regressions were used to test the relationships between soil temperature, water table, gross ecosystem production (GEP), and CH<sub>4</sub> fluxes. Results from the best fitting regression models are presented. All statistical analyses were performed using Statistical Package for the Social Sciences (SPSS) 17.0 (SPSS, Chicago, IL, USA).

### 3. Results

#### 3.1. Environmental Conditions

Strong seasonal dynamics in environmental factors were observed from January 2012 to December 2013 (Figure 2). There was a pronounced seasonal variation in monthly air temperature, ranging from –16.0 to 11.9°C. The highest monthly average temperature was in August and the lowest in January for both 2012 and 2013 (Figure 2a). Soil temperature at 5 cm depth showed a similar seasonal pattern to air temperature with a lower oscillation amplitude from –7.3 to 15.9°C (Figure 2b). The average air temperature and soil temperature at 5 cm depth were lower in 2012 than in 2013 (Table 2). Photosynthetic photon flux density (PPFD) and vapor pressure deficit (VPD) exhibited a clear seasonality with their annual maximum occurring in early July (Figures 2c and 2d). There were no significant differences in PPFD between 2012 and 2013



**Figure 2.** Monthly mean climate parameters: (a) air temperature ( $T_a$ ), (b) soil temperature at a depth of 5 cm ( $T_s$ ), (c) photosynthetic photon flux density (PPFD), (d) vapor pressure deficit (VPD), (e) friction wind velocity ( $u^*$ ), (f) monthly cumulative precipitation, and (g) daily mean water table depth in 2012 and 2013. The positive values of water table mean above the soil surface. Error bars represent standard deviations of the mean.

(Table 2). Daily average friction velocity ( $u^*$ ) showed a slight variation, and the maximum value occurred in April for 2012 and 2013 (Figure 2e). Annual precipitation was 367.5 and 404.7 mm in 2012 and 2013, respectively, and mostly occurred during the growing season (Figure 2f). The seasonal change of water table depth was associated with rainfall amount during the growing season (Figure 2g). The average water table during the growing season was 14.1 and 16.4 cm in 2012 and 2013, respectively (Table 2).

### 3.2. Diurnal Variations in $CH_4$ Fluxes

In order to understand how  $CH_4$  fluxes behave on a diurnal timescale, we collected and averaged data from seven consecutive sunny days in different periods of the year. There were large diurnal variations in  $CH_4$  fluxes and a suite of potential controlling environmental factors for four periods during the two years (Figure 3). These factors include air temperature, photosynthetic photon flux density (PPFD), friction wind velocity ( $u^*$ ) and vapor pressure deficit (VPD). The soil temperature at 5 cm depth and water table are not shown because of their small diurnal changes.

#### 3.2.1. Growing Season

A clear diurnal pattern of  $CH_4$  fluxes was found in 2012 and 2013 (Figure 3a1). The diurnal  $CH_4$  fluxes varied from 0.05 to 0.12  $\mu\text{mol m}^{-2} \text{s}^{-1}$  and from 0.08 to 0.23  $\mu\text{mol m}^{-2} \text{s}^{-1}$ , respectively, in 2012 and 2013. Two flux peaks were observed in this period. The minor peak was about 2 h after sunrise, and the major peak occurred at about 15:00–16:00 local time for each year. We found that the  $CH_4$  fluxes for a given hour were higher in 2013 than for those in 2012 during the daytime. The diurnal patterns of  $CH_4$  fluxes closely followed those of air temperature,  $u^*$ , and VPD.

#### 3.2.2. Nongrowing Season

**Soil freezing:** A diurnal pattern of  $CH_4$  fluxes was also found each year in the soil freezing period (Figure 3a2).  $CH_4$  fluxes increased after sunrise, peaked around 14:00, and gradually decreased to the lowest rate at midnight. The

**Table 2.** Average Daily Air Temperature, Soil Temperature at 5 cm Depth, Photosynthetic Photon Flux Density (PPFD), Total Precipitation, and Water Table for the Four Periods in 2012 and 2013<sup>a</sup>

Period	Year	Air Temperature (°C)	Soil Temperature (°C)	PPFD (mol m <sup>-2</sup> d <sup>-1</sup> )	Precipitation (mm)	Water Table (cm)
Growing season	2012	8.9	13.0	37.05	288.3	0.2
	2013	9.7	13.4	36.74	322.8	0.6
Nongrowing season						
	Soil freezing					
Soil freezing	2012	-6.3	3.3	23.48	16.5	
	2013	-5.9	4.6	25.31	14.2	
Winter	2012	-11.4	-4.1	22.32	9.9	
	2013	-10.0	-4.0	24.07	5.5	
Soil thawing	2012	2.1	2.0	38.80	52.8	
	2013	3.0	2.7	38.80	61.0	
Overall	2012	-1.6	3.5	30.24	367.5	0.2
	2013	-0.7	4.2	30.79	403.4	0.6

<sup>a</sup>The positive values of water table mean above the soil surface.

diurnal variation of  $T_s$ ,  $u^*$ , and VPD showed a clear correlation with that of  $\text{CH}_4$  fluxes in both years. Although the occurrence of peak PPFD was ahead that of the peak  $\text{CH}_4$  fluxes by a few hours, there was a weak relationship between the change in PPFD and that of  $\text{CH}_4$  fluxes during the daylight hours. During the nighttime, however, a strong positive relationship was observed between  $\text{CH}_4$  fluxes and air temperature.

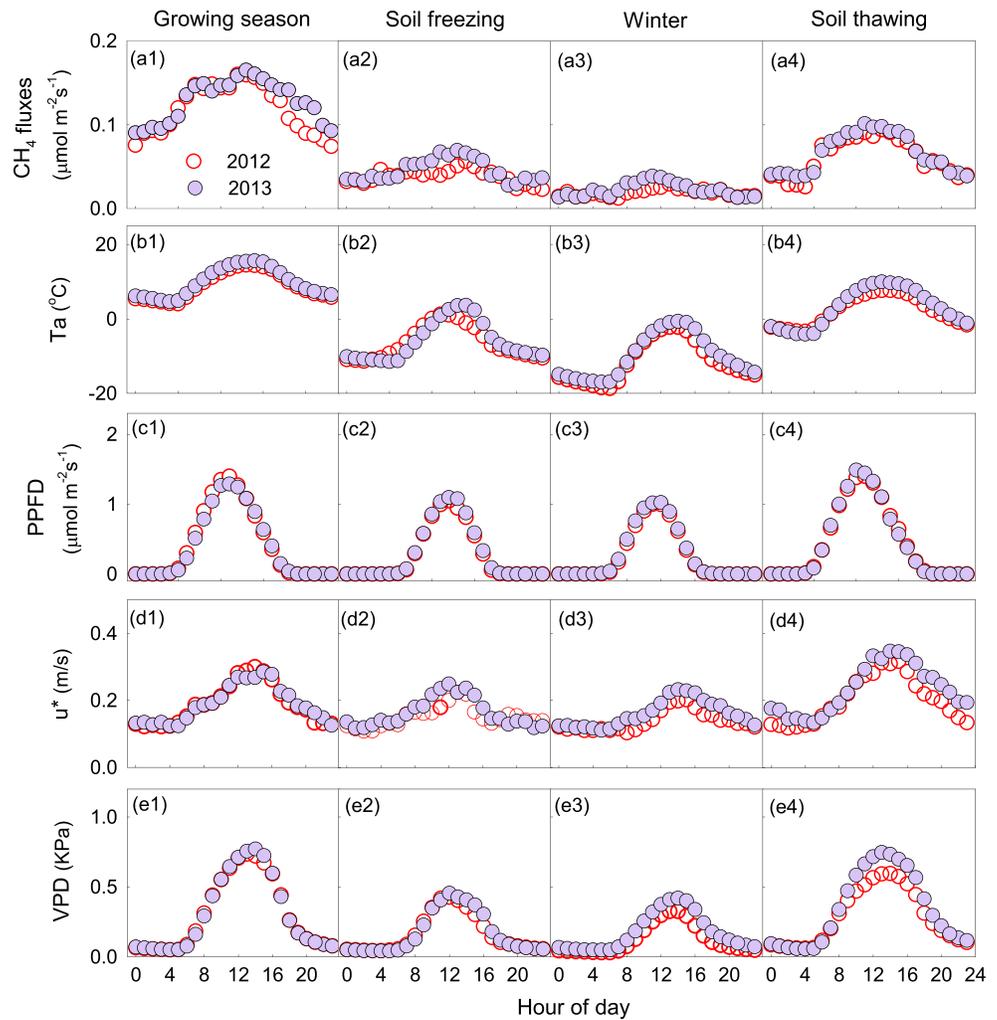
*Winter:* The diurnal change of  $\text{CH}_4$  fluxes was very small in the winters of 2012 and 2013 (Figure 3a3). In general,  $\text{CH}_4$  fluxes increased gradually from morning to midday with a peak of  $0.02 \mu\text{mol m}^{-2} \text{s}^{-1}$ . During winter period, the aboveground vegetation at this site died back, and the site was covered with ice and snow. The diurnal variation of air temperature and friction velocity showed significant correlations with that of  $\text{CH}_4$  fluxes, suggesting that diurnal variation in  $\text{CH}_4$  fluxes may have been associated with variation in turbulence.

*Soil thawing:* Differences in patterns and magnitudes of  $\text{CH}_4$  fluxes between day and night were obvious (Figure 3a4). The highest  $\text{CH}_4$  fluxes occurred at 15:00–16:00 in both years, which coincided with the time of peaks for air temperature and friction velocity. During this period, average daytime flux was  $0.08$  and  $0.09 \mu\text{mol m}^{-2} \text{s}^{-1}$  for 2012 and 2013, respectively. In this period, nighttime flux was nearly stable with a mean rate of  $0.04 \mu\text{mol m}^{-2} \text{s}^{-1}$ .

### 3.3. Seasonal Dynamics of $\text{CH}_4$ Fluxes

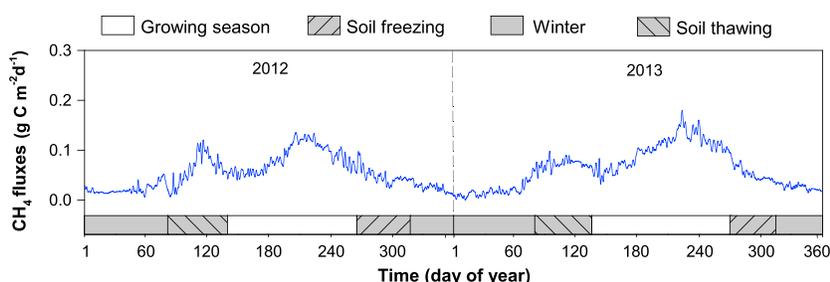
$\text{CH}_4$  fluxes showed typical and similar seasonal patterns in 2012 and 2013 (Figure 4). During the growing season,  $\text{CH}_4$  fluxes gradually increased from a low value in June to a peak in late July and early August and then decreased again to a low value at the end of the growing season (Figure 4). The average growing season  $\text{CH}_4$  fluxes were larger in 2013 than in 2012 with mean values of  $0.11 \text{ g C m}^{-2} \text{ d}^{-1}$  and  $0.09 \text{ g C m}^{-2} \text{ d}^{-1}$ , respectively. Peak daily average  $\text{CH}_4$  fluxes were  $0.13 \text{ g C m}^{-2} \text{ d}^{-1}$  on 28 July 2012 and  $0.16 \text{ g C m}^{-2} \text{ d}^{-1}$  on 15 August 2013. The peak period of  $\text{CH}_4$  fluxes generally coincided with the timing of the peak value of temperature. The total annual  $\text{CH}_4$  fluxes were  $19.8$  ( $26.4 \text{ g CH}_4$ ) and  $25.4$  ( $33.8 \text{ g CH}_4$ )  $\text{g C m}^{-2}$  in 2012 and 2013, respectively. The growing season  $\text{CH}_4$  fluxes accounted for about 53.9% in 2012 and 56.8% in 2013 of the annual emissions (Table 3).

We did not find any significant increase in  $\text{CH}_4$  fluxes during the onset of freezing of the upper soil layer. In this period, the average daily  $\text{CH}_4$  fluxes were  $0.04 \text{ g C m}^{-2} \text{ d}^{-1}$  in 2012 and  $0.05 \text{ g C m}^{-2} \text{ d}^{-1}$  in 2013. The accumulated  $\text{CH}_4$  fluxes during this period accounted for about 12.8% and 13.4% of the annual emissions for 2012 and 2013, respectively (Table 3). In the winter, the rate of  $\text{CH}_4$  fluxes was very low and steady. The average  $\text{CH}_4$  fluxes were similar for both years at  $0.02 \text{ g C m}^{-2} \text{ d}^{-1}$ . The accumulated  $\text{CH}_4$  fluxes during this period accounted for about 13.9% and 11.2% of the annual emissions for 2012 and 2013, respectively (Table 3). During the soil thawing period, we found a significant peak in  $\text{CH}_4$  fluxes, with the average  $\text{CH}_4$  fluxes of  $0.07 \text{ g C m}^{-2} \text{ d}^{-1}$  for 2012 and  $0.08 \text{ g C m}^{-2} \text{ d}^{-1}$  for 2013 (Figure 4). These peak emissions accounted for 19.4% and 18.6% of annual emissions in 2012 and 2013, respectively.



**Figure 3.** Examples of average diurnal variations of (a1–a4) CH<sub>4</sub> fluxes, (b1–b4) air temperature (Ta), (c1–c4) PPFD, (d1–d4) friction velocity (*u*\*), and (e1–e4) VPD on seven consecutive sunny days for the four periods in 2012 and 2013. Growing season, days 194–200 of 2012 and 221–227 of 2013 in Figures 3a1, 3b1, 3c1, 3d1, and 3e1; soil freezing, days 291–297 of 2012 and 293–299 of 2013 in Figures 3a2, 3b2, 3c2, 3d2, and 3e2; winter, days 23–29 of 2012 and 23–29 of 2013 in Figures 3a3, 3b3, 3c3, 3d3, and 3e3; soil thawing, days 116–122 of 2012 and 112–118 of 2013 in Figures 3a4, 3b4, 3c4, 3d4, and 3e4. Error bars represent the standard error of the mean.

Simple path analysis showed that among the six factors directly affecting daily CH<sub>4</sub> fluxes during the growing season, daily changes in both soil temperature and water table had very strong effects on daily CH<sub>4</sub> fluxes in the alpine wetland (Figure 5a). In contrast, temperature was the dominant environmental factor controlling the variation in daily CH<sub>4</sub> fluxes during the nongrowing season (Figure 5b). An exponential function can be used to describe the soil temperature dependence of CH<sub>4</sub> fluxes (Figure 6a), and the temperature sensitivity (*Q*<sub>10</sub>) of CH<sub>4</sub> fluxes (the increase factor in the rate of CH<sub>4</sub> fluxes when soil temperature is increased by 10°C [Lloyd and Taylor, 1994]) was 2.43 in 2012 and 2.55 in 2013. The relationship between CH<sub>4</sub> fluxes and water table can be expressed as an exponential function (Figure 6b). In addition, there was a significant positive relationship between daily GEP and daily CH<sub>4</sub> fluxes (Figure 6c). During the nongrowing season, a significant exponential relationship existed between CH<sub>4</sub> fluxes and soil temperature, and more than 56.2% and 59.1% of the variations in CH<sub>4</sub> fluxes during the nongrowing season could be explained by soil temperature in 2012 and 2013, respectively (Figure 6c). The *Q*<sub>10</sub> for CH<sub>4</sub> fluxes during the nongrowing season was 3.78 in 2012 and 5.92 in 2013.



**Figure 4.** Seasonal patterns of daily CH<sub>4</sub> fluxes in 2012 and 2013 in an alpine wetland on the Tibetan Plateau.

## 4. Discussion

### 4.1. Diurnal Variation in CH<sub>4</sub> Fluxes

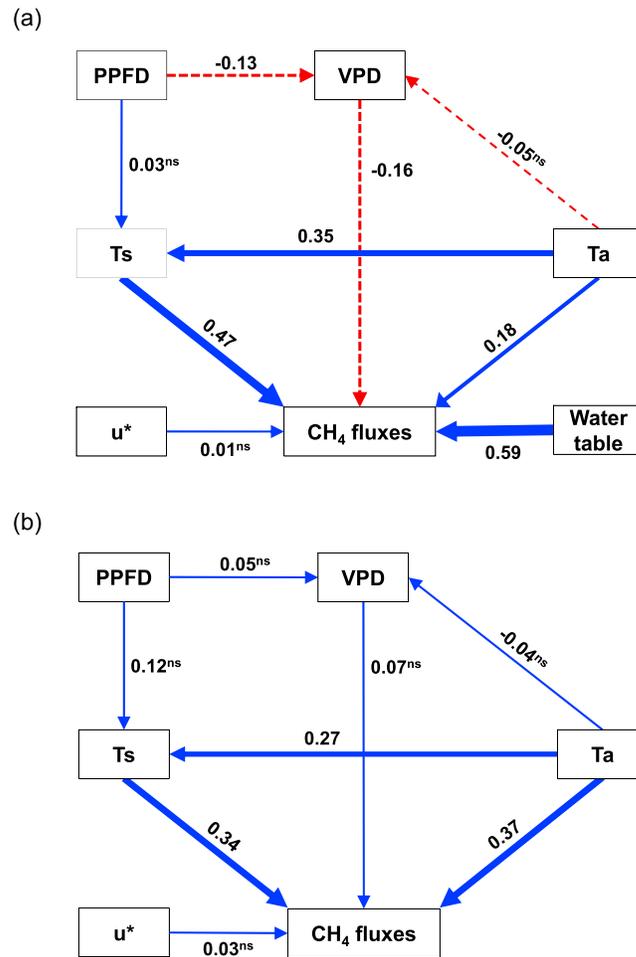
Diurnal variation in CH<sub>4</sub> fluxes is often site-specific due to differences in wetland types, species component, and environmental conditions. In this *Carex*-dominated alpine wetland, we found a clear two-peak diurnal pattern in CH<sub>4</sub> fluxes during the growing season, with a minor peak in the morning after sunrise and a major peak at noon. The observed pattern is consistent with previous results at another site on the Tibetan Plateau [Chen et al., 2010]. However, our results differed from other studies in low-altitude regions with similar vegetation, which reported one unique peak of CH<sub>4</sub> fluxes during the course of 1 day [Ding et al., 2004; Wang and Han, 2005; Long et al., 2010].

Why did this difference in diurnal variation of CH<sub>4</sub> fluxes between the Tibetan Plateau and other regions occur? Two potential explanations can be proposed. First, wetland plants, such as *Carex* species, usually employ a CH<sub>4</sub> transport system based on molecular diffusion [Wang and Han, 2005], and the diffusion of CH<sub>4</sub> gas from leaves to the atmosphere is tightly linked with stomatal conductance. A large-scale investigation reported that the species from the Tibetan Plateau had larger but fewer stomata than species in low-altitude regions [Yang et al., 2014]. These characteristics of the stomata could have directly inhibited gas diffusion, which indirectly favored the accumulation of gas in the aerenchyma tissue at night. Second, the Tibetan Plateau is characterized by low atmospheric oxygen concentration and high diurnal temperature range. At night, the low temperature (often below the frost point) could have inhibited the transportation of oxygen from the atmosphere to the aerenchyma tissues and rhizosphere zones, resulting in a low rate of CH<sub>4</sub> oxidation. Therefore, after sunrise, stomatal conductance rapidly increased in response to the increase in air temperature, and the gas accumulated during the night might have vented to the atmosphere, resulting in the first peak in CH<sub>4</sub> emissions.

The major peak of CH<sub>4</sub> fluxes at noon could be associated with several environmental factors. First, the peak at noon probably arose from an increase in methane production, owing to the higher activities of methanogen in response to warmer temperature [Mikkela et al., 1995; Whiting and Chanton, 1996; Hendriks et al., 2010]. In addition, shifts in VPD associated with increased solar radiation could have enhanced the transport of CH<sub>4</sub> gas from plants into the atmosphere. Finally, gas bubbles adhering to surfaces under water could be released when wind speed increases [Wille et al., 2008]. Increased pressure gradients resulting

**Table 3.** The Total Accumulated CH<sub>4</sub> Fluxes (g C m<sup>-2</sup>) of the Different Periods and Their Contributions to Annual Emissions in 2012 and 2013

Period	2012		2013	
	CH <sub>4</sub> Fluxes	% Contributions	CH <sub>4</sub> Fluxes	% Contributions
Growing season	10.69	53.9%	14.43	56.8%
Nongrowing season				
Soil freezing	2.55	12.8%	3.40	13.4%
Winter	2.76	13.9%	2.84	11.2%
Soil thawing	3.83	19.4%	4.72	18.6%
Total	19.83		25.38	



**Figure 5.** Path diagrams illustrating the effects of the environmental parameters: air temperature (Ta), soil temperature at a depth of 5 cm (Ts), photosynthetic photon flux density (PPFD), friction wind velocity ( $u^*$ ), vapor pressure deficit (VPD), and water table depth on CH<sub>4</sub> fluxes during (a) the growing season and (b) the nongrowing season from 2012 to 2013. Numbers beside the arrows are standardized path coefficients. Width of the arrows indicates the strength of the relationships. Solid arrows represent positive correlations, and dashed ones are negative correlations. Superscript "ns" indicates no significant relationships ( $P > 0.05$ ). All the variables used in the analysis are daily mean values.

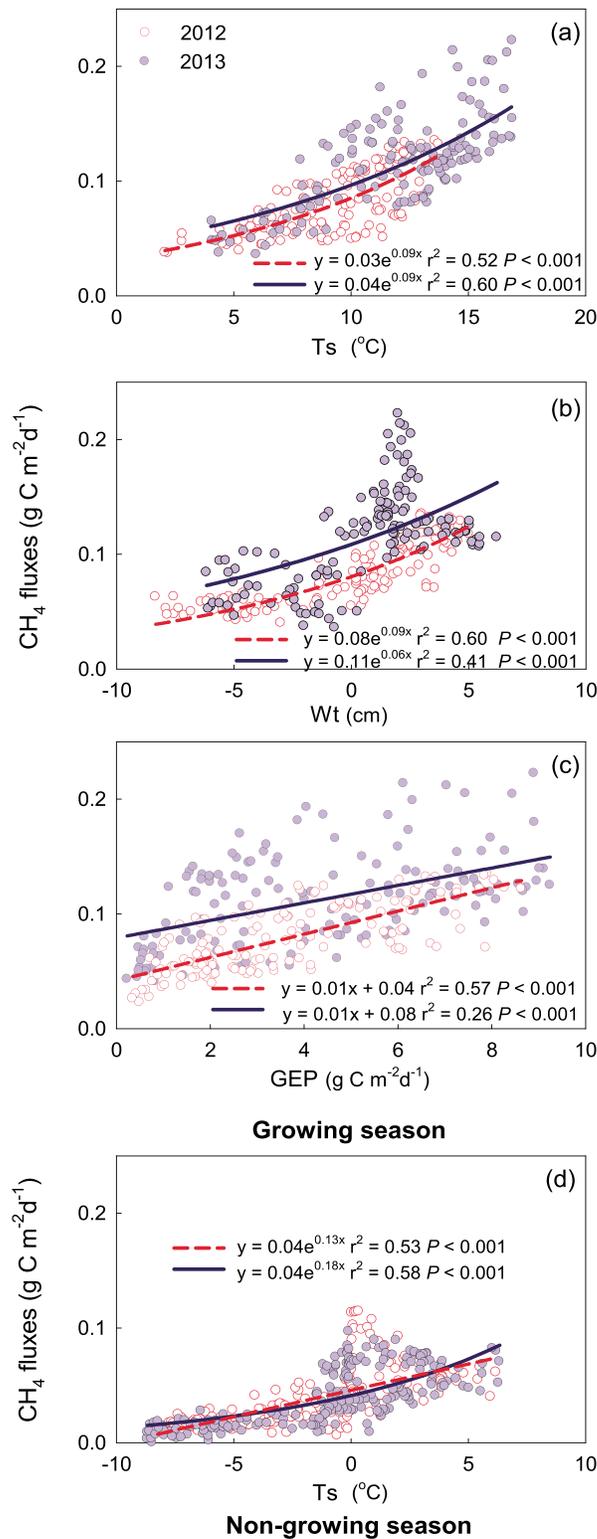
a continuous leaky system in standing litter [Hargreaves et al., 2001; Kutzbach et al., 2004; Wille et al., 2008]. This leaky system has the ability to transport CH<sub>4</sub> gas all day and then reduce the accumulation of CH<sub>4</sub> gas at night and, therefore, is probably responsible for the diurnal pattern in CH<sub>4</sub> fluxes during the soil freezing period.

In addition, we detected a weak diurnal pattern in CH<sub>4</sub> fluxes during the winter when the wetland surface was covered by ice and snow, although the CH<sub>4</sub> flux rates were very low in this period. The explanation for the diurnal variation in CH<sub>4</sub> fluxes is still unclear, but two possible mechanisms may account for it. First, with the increases in the ice thickness, the leaky system in standing litter will be gradually destroyed, while the leaky system can still have the ability to provide a route by which CH<sub>4</sub> can escape to the atmosphere [Hargreaves et al., 2001]. Second, the CH<sub>4</sub> gas accumulated under the ice could be released through cracks and fissures in the frozen surface layers [Dise, 1992], considering the landscape characteristics of this wetland in winter. Nevertheless, the two transport routes could be associated with a pressure gradient, which is regulated by air temperature and turbulence [Wille et al., 2008; Sachs et al., 2010]. Thus, the high-pressure

from changed atmospheric turbulence at noon could have modified the rates of diffusive and turbulent transfer of CH<sub>4</sub> across the water-air interface, leading to higher CH<sub>4</sub> fluxes.

In contrast to the growing season, evident single-peak diurnal patterns in CH<sub>4</sub> fluxes were observed during the nongrowing season (soil freezing, winter, and soil thawing), implying the importance of plants in regulating the diurnal pattern of CH<sub>4</sub> fluxes in different seasons. Even in the nongrowing season, there were clear differences in diurnal patterns among the three periods.

Usually, the upper soil and water layers will gradually freeze after sunset during the soil freezing and thawing periods, which can act as a barrier to CH<sub>4</sub> fluxes [Gažovič et al., 2010]. As a result, CH<sub>4</sub> produced under the ice will accumulate at night. During the daytime when the air and surface temperature increased, the CH<sub>4</sub> trapped under the ice will be expected to release from the melted ice layer, leading to a distinct one-peak diurnal pattern of CH<sub>4</sub> fluxes, with the daytime fluxes significantly higher than those of nighttime. However, we did not detect a significant increase in CH<sub>4</sub> fluxes after sunrise during the soil freezing period, indicating that other factors may cause the diurnal change in this period. During the soil freezing period, wetland plants gradually wilt and die, losing their ability to transport CH<sub>4</sub> gas via their stomata. Meanwhile, the degeneration of plant cortical parenchyma results in the development of a



**Figure 6.** The relationships between daily average CH<sub>4</sub> fluxes and (a) soil temperature, (b) water table depth, and (c) gross ecosystem production (GEP) during the growing season; and (d) between daily average CH<sub>4</sub> fluxes and soil temperature during the nongrowing season for 2012 and 2013. The dotted lines are regressions for the 2012 data series, and the solid lines are regressions for the 2013 data series.

gradient due to the relatively higher air temperature and friction velocity at daytime could have favored the transport of CH<sub>4</sub> gas from the anaerobic layers under the ice to the atmosphere, resulting in the increase of CH<sub>4</sub> fluxes at noon in this study.

Our findings have implications for the evaluation of field CH<sub>4</sub> fluxes under the environments with strong fluctuations. First, daily or seasonal CH<sub>4</sub> fluxes may be overestimated if only daytime flux data are considered because large diurnal variation exists, particularly in the growing season. Second, a significant correlation between CH<sub>4</sub> fluxes and friction velocity was observed. Hence, methods that limit turbulence, such as the static chamber method, may measure turbulence-driven component of the fluxes incorrectly, resulting in underestimated CH<sub>4</sub> fluxes.

#### 4.2. Seasonal Variation in CH<sub>4</sub> Fluxes

Over the two years, the nongrowing season CH<sub>4</sub> emissions accounted for nearly half of the annual emissions, emphasizing the vitally important contribution of the nongrowing season [Dise, 1992; Melloh and Crill, 1996; Mast et al., 1998; Huttunen et al., 2003; Merbold et al., 2013]. Moreover, it is indicated that if one evaluates the annual CH<sub>4</sub> fluxes without considering the nongrowing season fluxes, large errors will most likely occur.

During the growing season, CH<sub>4</sub> fluxes were positively related to soil temperature and water table, which is consistent with some previous studies, confirming the importance of soil hydroclimate [Turetsky et al., 2008; Olefeldt et al., 2013]. The change in soil temperature could have affected not only CH<sub>4</sub> production by stimulating the activities of the methanogens but also CH<sub>4</sub> transportation from soils into the atmosphere [Hosono and Nouchi, 1997]. The temperature sensitivity of CH<sub>4</sub> fluxes during this season was 2.43–2.55, which is well within the range reported earlier [Cao et al., 1996; MacDonald et al., 1998; Whalen, 2005]. At our study site,

we found that the increase of CH<sub>4</sub> fluxes during the early growing season was much faster in 2013 than in 2012, most likely due to the rapid increase in the water table in 2013. The water table operated like an “on-off switch” for CH<sub>4</sub> emissions: a higher water table can limit the oxygen concentration and promote the anaerobic conditions conducive for CH<sub>4</sub> production within the soil, and vice versa [Wang and Han, 2005; Zona et al., 2009; Yrjälä et al., 2011; Bhullar et al., 2013]. On the other hand, the water table depth can also influence soil temperature by controlling thermal conductivity. Climate change with increases in temperature and water table depth caused by increase in precipitation on the Tibetan Plateau [Gao et al., 2012; Zhu et al., 2013] is likely to stimulate CH<sub>4</sub> emissions beyond the effects of soil temperature or water table depth alone. Therefore, modeling the wetland CH<sub>4</sub> budgets under future climate should consider the interactive effects of soil warming and water table depth on CH<sub>4</sub> production and transport. In addition to serving as a channel for transporting CH<sub>4</sub> gas, wetland plants can also provide C substrates for the methanogenic community both through the production of soil organic matter and as fresh exudates and residues [Whiting et al., 1991]. A number of studies suggest that plant productivity is a good predictor of CH<sub>4</sub> fluxes either across a wide range of wetlands [Whiting and Chanton, 1993] or within a single species [Sass et al., 1990; Whiting et al., 1991; Whiting and Chanton, 1992; Lai et al., 2014]. Therefore, an increase in GEP during the growing season in our study could have been favorable for the production of CH<sub>4</sub> by increasing the supplement of organic substrate from belowground tissues for methanogenesis.

In the current study, a large burst of CH<sub>4</sub> fluxes was found during the soil thawing period, which could be attributed to the following two reasons. First, the CH<sub>4</sub> produced beneath ice in winter was stored in the soil porosity and water, and only a small portion was oxidized [Roslev and King, 1996; Le Mer and Roger, 2001]. As ice thawed in the following spring, the stored CH<sub>4</sub> was released to the atmosphere. Second, thawing of previously frozen soils and increases in the thickness of the active layer could have accelerated the decomposition of plant litter and have increased the activities of methanogens [Dörsch et al., 2004]. Thus, the recovery of methanogenic activity was synchronous with increasing substrate availability, which could be partly responsible for the large burst of CH<sub>4</sub> emissions during this period.

During the onset of soil freezing, our study shows no pulse in CH<sub>4</sub> fluxes, in contrast to other studies [Dlugokencky et al., 1994; Mastepanov et al., 2008]. There are few studies addressing the issue of CH<sub>4</sub> fluxes during the soil freezing period, and the causes to the phenomena are still unknown. Mastepanov et al. [2008] hypothesized that downward freezing toward the permafrost table enhanced the concentration of CH<sub>4</sub> gas, thus increasing gas pressure between the frozen active layer, and the permafrost table promoted the accumulated CH<sub>4</sub> burst out through the soil voids or the aerenchyma of senescent plants. Tagesson et al. [2012] suggested that a larger fraction of the CH<sub>4</sub> gas produced during the growing season could be stored in the soil of plots without vascular plants than the soil of plots with vascular plants. This CH<sub>4</sub> gas could then be released during soil freezing due to the increased gas pressure. We suggest that the extensive root system at our site transported the CH<sub>4</sub> gas effectively during the growing season, resulting in less CH<sub>4</sub> stored in the soil, and thus eliminated the pulse at this period.

The observed winter CH<sub>4</sub> fluxes were relatively low (0.02 g C m<sup>-2</sup> d<sup>-1</sup>), which could be related to the decreased CH<sub>4</sub> production due to low soil temperature and related to the resistance in the CH<sub>4</sub> emission paths due to the thick ice and snow cover [Rinne et al., 2007]. In addition, the temperature sensitivity of CH<sub>4</sub> fluxes during the nongrowing season was 3.78 in 2012 and 5.92 in 2013, and the high-temperature dependence of CH<sub>4</sub> fluxes indicates that climate change will potentially result in a larger increase in the nongrowing season CH<sub>4</sub> emissions compared with the growing season because of the more dramatic increase in wintertime temperature (0.5°C per decade [Piao et al., 2010]).

### 4.3. Annual Variation in CH<sub>4</sub> Fluxes

The total annual CH<sub>4</sub> fluxes at our study site were 19.8 g C (26.4 g CH<sub>4</sub>) m<sup>-2</sup> in 2012 and 25.4 g C (33.8 g CH<sub>4</sub>) m<sup>-2</sup> in 2013. The emissions strengths are within the range of emissions from low-altitude boreal wetlands with similar dominant vegetation, using the eddy covariance method [Hargreaves et al., 2001; Rinne et al., 2007; Wille et al., 2008; Hanis et al., 2013]. For example, Rinne et al. [2007] estimated that the annual CH<sub>4</sub> fluxes were 9.5 g C m<sup>-2</sup> yr<sup>-1</sup> on a boreal fen at Siikaneva, Finland. A higher CH<sub>4</sub> emissions intensity was found in the warmer year of 2013, suggesting that there might be a positive feedback between CH<sub>4</sub> emissions and climate in this region. The climate data show a pronounced warming trend,

particularly in the nongrowing season, on the Tibetan Plateau [Liu and Chen, 2000; Piao et al., 2010; Wei and Fang, 2013]. The vast wetlands on the Tibetan Plateau may thus have the potential to exert a positive feedback on climate.

## 5. Conclusions

We conducted the first in situ year-round CH<sub>4</sub> fluxes measurement in an alpine wetland on the Tibetan Plateau from 2012 to 2013, using the newly developed LI-COR LI-7700 open-path methane analyzer. Our study represents a comprehensive assessment of the magnitude, patterns, and environmental drivers of CH<sub>4</sub> fluxes on different timescales. In particular, this study reveals the importance of the nongrowing season CH<sub>4</sub> emissions in the estimation of annual CH<sub>4</sub> budget, which was commonly neglected in the previous studies. In addition, these results provide an important data basis for quantifying and modeling regional and global flux budget of CH<sub>4</sub> from wetlands at high altitudes.

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