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Timescale dependence of environmental controls on methane efflux in Poyang Lake, China

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Abstract

Lakes are an important natural source of CH_4 to the atmosphere. However, the long-term CH_4 efflux in lakes has been rarely studied. In this study, the CH_4 efflux in Poyang Lake, the largest freshwater lake in China, was measured continuously over a 4-year period by using the floating chamber technique. The mean annual CH_4 efflux throughout the 4 years was 0.54 mmol m^{-2} day^{-1}, ranging from 0.47 to 0.60 mmol m^{-2} day^{-1}. The CH_4 efflux had a high seasonal variation with an average summer (June to August) efflux of 1.34 mmol m^{-2} day^{-1} and winter (December to February) efflux of merely 0.18 mmol m^{-2} day^{-1}. The efflux showed no apparent diel pattern, although most of the peak effluxes appeared in the late morning, from 10:00 h to 12:00 h. Multivariate stepwise regression on a seasonal scale showed that environmental factors, such as sediment temperature, sediment total nitrogen content, dissolved oxygen, and total phosphorus content in the water, mainly regulated the CH_4 efflux. However, the CH_4 efflux only showed a strong positive linear correlation with wind speed within a day on a bihourly scale in the multivariate regression analyses but almost no correlation with wind speed on diurnal and seasonal scales.

Keywords: Methane, Sediment temperature, Temperature sensitivity, Substrate availability, Wind speed

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1. Introduction

Methane (CH_4) contributes to about 20% of global warming in terms of radiative forcing, and its concentration in the atmosphere increased at a rate of 0.5 ppb year^{-1} in 1999–2006; this rate rapidly increased to 6 ppb year^{-1} from 2007 to 2011 (IPCC, 2013). Although the total global lake area accounts for approximately 3.7% of the Earth's nonglaciated land area (Verpoorter et al., 2014), CH_4 emissions from global lakes account for up to 14.9% of natural CH_4 emissions (IPCC, 2013). However, this estimate has been associated with large uncertainties because of the high spatial and temporal variations of CH_4 emissions and the insufficient long-term measurements of CH_4 effluxes, especially in tropical and subtropical lakes (Yang et al., 2011; Ortiz–Llorente and Alvarez–Cobelas, 2012; Bastviken et al., 2015; Li and Bush, 2015).

CH_4 effluxes in lakes feature high temporal variations (Käki, 2001; Xing et al., 2004; Duan et al., 2005; Xing et al., 2005, 2006; Palma–Silva et al., 2013). For example, previous studies found that the minimum and maximum CH_4 effluxes over a day were −1.36 and 128.85 mmol m^{-2} day^{-1}, respectively (Xing et al., 2004; Duan et al., 2005; Chen et al., 2007; Podgrajsek et al., 2014a, 2014b); even larger variations were found on a seasonal scale (Xing et al., 2005, 2006; Duan et al., 2005; Ortiz–Llorente and Alvarez–Cobelas, 2012; Wik et al., 2014). These large variations in CH_4 effluxes highlight the importance of frequent and long-term measurements (Bastviken et al., 2008; Chen et al., 2013; Bastviken et al., 2015). Unfortunately, most earlier studies on CH_4 emissions were based on short-term measurements, ranging from daily to seasonal scales, and were conducted during the day time (Xing et al., 2004; Duan et al., 2005; Xing et al., 2005; Schrier–Uijl et al., 2011; Rõõm et al., 2014). To our knowledge, multi-year measurements of CH_4 effluxes have only been conducted in high-latitude lakes, and few studies on tropical and subtropical lakes, especially large

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ones, had measurement durations longer than one year.

The magnitude of CH_4 emission mainly depends on the dynamic balance between the microbial processes of CH_4 production, oxidation, physical transportation from the anaerobic zone to the atmosphere in lakes, and regulation by multiple, interconnected physical, chemical, and biological variables (Sun et al., 2012; Liu et al., 2013; Serrano–Silva et al., 2014; Rasilo et al., 2015). CH_4 production and oxidation are microbial processes regulated by organic carbon loading, dissolved organic matter, lake nutrient status, and N availability (Bridgham et al., 2013; Liu et al., 2013; Hershey et al., 2014; Rasilo et al., 2015); temperature (Liikanen et al., 2003; Marotta et al., 2014; Yvon–Durocher et al., 2014); lake depth and size (Juutinen et al., 2009; Rasilo et al., 2015); pH, O_2, NO_3^{2-}, Fe^{3+}, and SO_4^{2-} in the sediment and water column (van Bodegom and Scholten 2001; Schrier–Uijl et al., 2011; Bridgham et al., 2013); and populations and potential activities of methanogens and methanotrophs (Segers, 1998; van Bodegom and Scholten, 2001; Liu et al., 2015, 2016). CH_4 transportation is driven by three major mechanisms, namely, molecular diffusion, bubble ebullition, and plant-mediated transportation (Bridgham et al., 2013; Chen et al., 2013; Zhu et al., 2016). These mechanisms are affected by water stratification and seasonal overturns of the water mass, which are determined by temperature, wind-forced mixing, water depth, boundary layer dynamics, hydrostatic pressure, and different vascular plants (Juutinen et al., 2009; Zhu et al., 2016). Most studies examined CH_4 emissions and their influencing factors in small lakes because of their large contribution to the global CH_4 budget (Bastviken et al., 2004; Downing et al., 2010; Bartosiewics et al., 2015; Holgerson et al., 2016). However, few studies reported temporal CH_4 emissions and their key regulating factors at different temporal scales in large lakes. Therefore, investigating the impacts of physical and biological factors on temporal CH_4 effluxes based on long-term measurements in a large lake is also important to estimate lake CH_4 emissions.

Poyang Lake, a subtropical lake, is the largest freshwater lake in China, but its annual CH_4 emissions have not been adequately measured. In this study, we measured the CH_4 efflux over the course of 4 years in Poyang Lake to (1) examine the annual CH_4 efflux; (2) explore the CH_4 efflux dynamics, including diel, seasonal, and inter-annual variations; and (3) quantify the relationships between the CH_4 efflux and environmental factors, and identify the possible factors driving CH_4 effluxes at different temporal scales.

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2. Materials and methods

2.1. Site description

Poyang Lake (28°22′–29°45′N, 115°47′–116°45′E) is located in Southern China in Jiangxi Province, with a surface area of 3283 km^2 and a total catchment area of 162,000 km^2, which is separated to the northern and southern parts by the Songmen Mountain. Poyang Lake receives water input from five main tributaries, namely, the Raohe River, Xinjiang River, Fuhe River, Ganjiang River, and Xiushui River. The climate is humid subtropical with a mean annual temperature of 17.5 ℃ and an annual precipitation of 1680 mm (Ye et al., 2011). Vegetation in the lake is composed of macrophytes, including *Carex* sp. and *Artemisia selengensis* in the hydrophyte zone, and the main submerged aquatic macrophytes, including *Ceratophyllum demersum*, *Potamogeton malaianus*, *Potamogeton crispus*, and *Hydrilla verticillata* (Wang et al., 2011).

This study was conducted near the Poyang Lake Laboratory of the Wetland Ecosystem Research Station (operated by the Chinese Academy of Sciences), which is located in the northern sub-basin of Poyang Lake in Xingzi County, Jiangxi Province (Fig. 1). The five tributaries flow into the lake in the southeast of Xingzi County, which then joins with the Yangtze River. The water level fluctuated dramatically from 7.78 m to 18.57 m above sea level (Wu Song) between the wet (April to September) and dry seasons (October to March) during the study period because of rainfall and Three Gorge management. Poyang Lake is not stratified (Zhu and Zhang, 1997), with mean and maximum depths of 8 and 23 m, respectively. The concentrations of total nitrogen (TN), total phosphorous (TP), suspended solids (SS), and chlorophyll *a* (Chl *a*) in the lake were 3.45, 0.11, 39.98, and 9.04 mg L^{-1}, respectively (Yao et al., 2015).

2.2. CH_4 efflux measurements

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The CH_4 efflux was measured using floating chambers, including both ebullition and diffusive fluxes (Bastviken et al., 2004, 2010). The floating chamber was fabricated using a PVC pipe 100 cm in length and 20 cm in diameter with Styrofoam floats attached to the sides. The floating chambers were inserted 80 cm into the water and 20 cm above the water surface to minimize the perturbation of the surface water flow to the pressure inside the chambers. We tested the chamber system with different insertion depths in the laboratory and field, and found that the current depth of about 80 cm could effectively prevent the impacts of the surrounding Styrofoam floats while maintaining the chamber balance in moderate winds. A similar design of floating chambers was used in previous studies (Lorke et al., 2015; Zhao et al., 2015). Zhao et al. (2015) have recently conducted a systematic comparison of the effects of chamber shape, dimension, and insertion depth into the water on CH_4 effluxes and found that insertion depth only slightly affects the CH_4 efflux measured in the Three Gorges Reservoir when wind speed is relatively low. In the current study, the insertion depth was deeper than those of previous studies to avoid the impact of waves in Poyang Lake on the chamber body. Earlier studies also found that floating chambers should be seated at the water surface with minimal insertion into the water in a flowing-water system to minimize the "drag" effect of flowing water on chamber pressure (Bastviken et al., 2010; Vachon et al., 2013; McGinnis et al., 2015). Except for some waves, the water in Poyang Lake did not have an apparent directional flow during the measurement period. A detailed description of the floating chamber system can be found in Liu et al. (2013).

We collected a gas sample (ambient concentration) immediately after the chamber was closed and three other samples at a 20 min interval for 1 h. The gas was extracted into a 12 mL evacuated glass vial by a 2 mL syringe needle with an air pump, which enhanced the pressure in the vial to 3 bars. Subsequently, the air samples were transported immediately to a laboratory for CH_4 concentration analysis. The CH_4 concentration was measured using a gas chromatograph equipped with a flame ionization detector (GC7890A, Agilent Technologies, Inc., Santa Clara, CA, USA). We used nitrogen (N_2) as the carrier gas, which ran at a flow rate of 30 mL min^{-1}. We calibrated the gas chromatograph for every four samples with a calibration gas of 2.03 ppm at 99.92% precision (China National Research Center for Certified Reference Materials, China). The oven and detector temperatures of the GC were set to 55 ℃ and 250 ℃, respectively.

Calculation of the CH_4 efflux was based on the CH_4 concentration of the four samples using a linear regression model. Data quality control was conducted following the method of Rasilo et al. (2015) before the regression models were fitted. As a result, most of the models performed satisfactorily, with a coefficient of determination (R^2) greater than 0.95. In case of ebullition, the CH_4 concentration inside the chamber would deviate from the normal trend. Most of the CH_4 concentrations measured immediately after the ebullition point slightly decreased mainly because of the CH_4 diffusion back to water when the CH_4 concentration inside the chamber space increased suddenly from bubbling. To include the ebullition-induced CH_4 emissions, we only used two measured concentrations, the first measurement (ambient concentration) and an ebullition-adjusted concentration, in calculating the CH_4 efflux when ebullition occurred inside the chamber. The ebullition-adjusted concentration was obtained by adding the diffusion-induced concentration increment, which is a correction term, to the measured concentration when ebullition occurred. The total CH_4 efflux, which includes both ebullition and diffusive effluxes, was calculated on the basis of the slope of the concentration change during the whole period when the chamber was closed (Fig. 2). Specifically, when ebullition occurred during the first 20 min, we obtained the ebullition-adjusted concentration by summing up concentration on 20 min and the 2-fold incremental concentration between the third and fourth sampling times. When the ebullition occurred at the third sampling, we summed up the concentration at 40 min and the incremental concentration between the first and second sampling times. When the ebullition occurred at the fourth sampling, we used the first and fourth sampling concentrations directly to calculate the slope of the total efflux.

Samplings took place at a monthly interval from January 2011 to December 2014 at three sites in Poyang Lake (Fig. 1): site A (Luoxingdun: 29°3′29″N, 116°16′49″E), site B (Mantianxing: 29°34′25″N, 116°13′29″E), and site C (Huoyanshan: 29°39′0″N, 116°16′11″E). The mean water depth in our sampling sites was 3 m. The sampling sites lacked aquatic plants. Our previous study examined the spatial pattern of the CH_4 efflux in the lake (Liu et al., 2013). Therefore, we focused on the long-term dynamics of CH_4 efflux in the current study. At each site, four chambers were placed approximately 10 m away from a small boat to minimize disturbance. Measurements were conducted from early morning to late afternoon with about 6 cycles of measurements for each chamber, except for days when the diel-cycle measurements were taken. We conducted four 24 h measurements at the three sites in 24–25 July 2011, 5-6 September 2012, 13–14 January 2013, and 14–15 January 2015 to examine the diel variations of CH_4 effluxes. These measurements were conducted every 2 h from 8:00 am to 8:00 am the next day, providing 12 cycles of measurements for each chamber per 24 h.

2.3. Environmental variables

Various environmental variables were also measured in the lake sediment,

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surface water, and atmosphere. We collected surface water and sediment samples (0–15 cm) using a plexiglass water grab and a stainless steel sediment sampler (3 cm in diameter) after obtaining gas samples. The water and sediment samples were immediately stored in plastic bottles and bags, respectively. Then, all the samples were stored in ice coolers and transported to a laboratory for analysis within a week. In addition, we measured the wind speed at about 1.5 m above the water surface using a portable anemometer (Testo 410-1, Testo, Germany) and the surface sediment (0–15 cm) temperature using a mercury thermometer. We used a multi-parametric probe (556 MPS, YSI, USA) to measure the water quality factors in situ, such as electrical conductivity and dissolved oxygen (DO) content, at each sampling site from June 2013 to June 2014. The water levels in the lake were obtained from the Xingzi Hydrological Station, about 20 km from our sampling sites.

In the laboratory, the pH values of the water and sediment samples were measured using a pH meter (Delta 320, Mettler–Toledo, Switzerland). Chemical oxygen demand (COD) was measured using the spectrophotometric detection method based on Griess reaction (Jirka and Carter, 1975; Yao et al., 2015). Chl *a* concentration was measured via spectrophotometry (Rasilo et al., 2015; Yao et al., 2015), which was extracted in 90% ethanol and then analyzed spectrophotometrically at 750 and 665 nm in accordance with ISO 10 260 (1992). The SS level in the lake water was measured by a gravimetric procedure, where the solids from the water sample were filtered, dried, and weighed to determine the total non-filterable residue of the sample (Fishman and Friedman, 1989). TP concentration was measured using the molybdenum blue method after persulfate digestion (Karl and Tien, 1992; Yao et al., 2015). In addition, the nitrate (NO_3^-), ammonium (NH_4^+), TN, and dissolved organic carbon (DOC) contents in the water were measured using a total carbon and

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nitrogen analyzer using filtered water (Shimadzu TOC-VCSH + TN module, Shimadzu, Japan). The sediment TN and organic carbon contents after total sediment acidification with HCl 1N were determined using a vario MAX CN element analyzer (NA Series 2, CE Instruments, Germany).

Considering the different sampling periods, we classified the environmental variables into three groups. The first group included sediment temperature, sediment total nitrogen content, water level, DOC content in the water, pH in the sediment, NH_4^+ and NO_3^- concentrations in the water and sediment, sediment organic carbon content, the ratio of carbon and nitrogen, and the mean daily wind speed over a 48-month period. The second group included TN, TP, COD, and Chl *a* contents in the water, which were sampled between June 2011 and December 2014. We sampled the third group variables from June 2013 to June 2014, including DO content, conductivity, and pH in the water.

2.4. Data analysis

We averaged the CH_4 effluxes of the three sites to minimize the effect of the spatial variation of CH_4 efflux on the temporal dynamics of the efflux. One-way ANOVA followed by post-hoc Tukey's test and paired T test were used to analyze the seasonal and inter-annual differences in the CH_4 effluxes. The coefficient of variation (CV) was used to quantify the inter-annual variation of CH_4 efflux. We employed stepwise multiple regressions to identify the environmental factors driving the CH_4 effluxes at different temporal scales. We also used regression and correlation analyses to determine the relationships between independent variables and CH_4 effluxes. We used the Vant′ Hoff equation to calculate the temperature sensitivity ($Q_{10} = e^{10b}$) of CH_4 efflux (Xu and Qi, 2001; Wei et al., 2015). All statistical analyses were performed using the SPSS 17.0 statistical software (SPSS Inc., Chicago, IL, USA), and graphs

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were created using the Sigma Plot 11.0 program (Systat Software Inc., San Jose, CA, USA).

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3. Results

3.1. CH_4 effluxes in Poyang Lake

3.1.1. Annual CH_4 effluxes

The mean CH_4 efflux was 0.54 ± 0.053 mmol m^{-2} day^{-1} in Poyang Lake over the 4-year period, with annual mean effluxes of 0.47 ± 0.54, 0.56 ± 0.41, 0.52 ± 0.55, and 0.60 ± 0.56 mmol m^{-2} day^{-1} in 2011, 2012, 2013, and 2014, respectively (Table 1). The inter-annual variation of CH_4 efflux was moderately high with a CV of 9.8% over the 4 years. The mean CH_4 efflux in 2014 was 25.7% greater than that in 2011, justifying the necessity for long-term measurements.

3.1.2. Seasonal CH_4 effluxes

The seasonal variations of CH_4 effluxes in Poyang Lake were prominent, demonstrating a similar pattern to that of seasonal temperature (Fig. 3). In general, the annual maximum CH_4 effluxes occurred in summers and the minimum in winters. The CH_4 efflux increased slowly in early spring and then rapidly in May, reaching its maximum in July. After reaching the maximum, the CH_4 efflux decreased sharply in August and September and then slowly before reaching its minimum in January (Fig. 3). Significant differences in the mean CH_4 effluxes existed between summers and the other three seasons throughout the 4 years ($p < 0.05$), whereas the differences in the CH_4 effluxes among the spring, autumn, and winter seasons were not statistically significant ($p > 0.05$) (Table 1).

3.1.3. Diel CH_4 effluxes

The CH_4 effluxes in Poyang Lake also exhibited apparent variations within a day because the daily maximum appeared late in the morning (10:00–12:00 h) and the minimum early in the morning the next day (4:00–6:00 h). The diel pattern of the CH_4 efflux was asymmetric, fast increasing in the morning from 8:00 h to 12:00 h and

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slowly decreasing in the afternoon and during the night, especially in the summer (Fig. 4). However, the diel pattern of the CH_4 efflux was inconsistent and obvious. For example, the diel pattern on January 13–14, 2013 was an exception, when the maximum efflux occurred around 6:00 h on January 14th and a severe cold front with heavy fogs enveloped the Poyang Lake area in the early morning of January 14th. The diel pattern of CH_4 efflux was vague with an average difference between the daily maximum and minimum of only 0.073 mmol m^{-2} h^{-1}. The CH_4 efflux could also change abruptly throughout a day. For example, the efflux sharply dropped from 0.068 to −0.012 mmol m^{-2} h^{-1} within barely 2 h, as observed on July 23, 2011, indicating that the lake switched from a CH_4 source to sink within a short period of time (Fig. 4a). This abrupt change was also observed in the afternoon of August 28, 2012 (Fig. 4b). Further analysis showed that the diel pattern of CH_4 effluxes followed the diel pattern of wind speed (Figs. 5a–5d).

3.2. Relationships between CH_4 efflux and environmental variables

In the current study, environmental factors differed in importance depending on the timescale in the stepwise multiple regressions analyses. The results of stepwise multiple regressions on a seasonal scale showed that the sediment temperature, sediment TN content, DO, and TP content in the water were significant predictors of CH_4 effluxes (Table 2). In specific, sediment temperature and sediment TN content explained 65% of the variation in CH_4 effluxes for 4 years when we used the first group of factors. The sediment temperature and TN content explained 73% of the CH_4 efflux variations when the second group of variables was added to the first group. The sediment temperature, sediment TN content, DO, and TP contents in the water explained 89% of the CH_4 efflux variation when the three groups of variables were used together. Wind speed was the only significant variable for the CH_4 efflux

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variations on a diel scale. Wind speed explained 58%, 56%, 84% and 86% of the CH_4 efflux variations in 24–25 July 2011, 5–6 September 2012, 13–14 January 2013 and 14–15 January 2015, respectively (Fig. 5a-5d).

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4. Discussion

4.1. CH_4 effluxes in Poyang Lake

The mean CH_4 emission in Poyang Lake was moderately higher than those in other large lakes of more than 1 km^2 in the world. The mean CH_4 emission (0.54 mmol m^{-2} day^{-1}) was within the reported range of approximately 0.022–5.85 mmol m^{-2} day^{-1} in boreal and temperate lakes over 1 km^2 but was obviously lower than diffusive effluxes in subtropical lakes and total effluxes (including diffusion and ebullition) in tropical lakes (Table 3). In addition, the mean CH_4 emission in Poyang Lake was comparable with the diffusive effluxes in tropical lakes (Table 3). For example, previous studies reported that the diffusive CH_4 efflux was 0.65 mmol CH_4 m^{-2} day^{-1} in the TR Lake and 0.50 mmol m^{-2} day^{-1} in the BB Lake in the Pantanal region (Bastviken et al., 2010). However, the mean CH_4 efflux in Poyang Lake was only higher than those in other lakes over 100 km^2 (except the Võrtsjärv Lake). The low CH_4 efflux in the current study was unlikely caused by our floating chamber system because the CH_4 efflux would have increased if the insertion of chambers considerably disturbed the water profiles. The lower CH_4 emissions in our study may be attributed to the low concentration of carbon substrates in the water and sediments in Poyang Lake. The DOC concentration in Poyang Lake was merely 3.3 mg L^{-1}, which was much lower than that of the 5.8 mg L^{-1} in Biandantang Lake and 7.4 mg L^{-1} in Donghu Lake, which are two subtropical lakes in China (Xing et al., 2005, 2006). Poyang Lake also has a lower organic carbon content in its sediments than most other lakes. The average organic carbon content in the sediments in Poyang Lake was 0.89%, which was much lower than that of 30.76% averaged over five temperate lakes (Schrier–Uijl et al., 2011) and slightly higher than that of nearly 0.75% in tropical lakes in the Pantanal region (Bastviken et al., 2010). Therefore, the CH_4 emissions in large lakes cannot be

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ignored when estimating the global CH_4 budget because of their area.

CH_4 effluxes at the air-water interface showed high fluctuations in the four cycles, but showed no significant diurnal differences. The diurnal CH_4 efflux ranged from -0.019 to 0.13 mmol m^{-2} h^{-1}, which was within the reported range of other lakes (-0.057 to 5.37 mmol m^{-2} h^{-1}) over a diurnal cycle (Xing et al., 2004; Duan et al., 2005; Chen et al., 2007; Podgrajsek et al., 2014a, 2014b). The wide range of diurnal CH_4 efflux in previous results may be due to differences in sample size in different studies. For example, CH_4 efflux was measured at 2h intervals with 12 data points over a diurnal cycle in this study, but CH_4 efflux was measured at 3-6 h intervals with only 4-8 data points in previous studies (Käki et al., 2001; Xing et al., 2004; Duan et al., 2005). Another possible reason for these discrepancies was that vegetation might have played an important role in CH_4 efflux in other studies (Käki et al., 2001; Duan et al., 2005), while there was no vegetation in the water where we sampled in Poyang Lake. In addition, our study showed that there were no significant differences in CH_4 effluxes between the nighttime and the daytime, which was inconsistent with other studies (Keller and Stallard, 1994; Bastviken et al., 2010). This inconsistency may be due to incomplete measurement of the diurnal cycle in other studies. For example, CH_4 efflux was only measured three times at sunrise, daytime and sunset to represent a diel cycle in Bastviken et al. (2010). In Keller and Stallard (1994) study, the daytime and nighttime CH_4 efflux measurements were not conducted on the same day. In particular, two studies reported a new finding that hydrodynamic transport contributed more to nighttime CH_4 effluxes than daytime CH_4 effluxes (Poindexter et al. 2015; Anthony and Macintyre 2016). However, we cannot estimate CH_4 effluxes by hydrodynamic transport because we did not measure CH_4 concentration in the water in this study. Further studies are needed to address this issue in the lake.

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4.2. CH_4 effluxes in summer

The CH_4 effluxes in Poyang Lake were substantially greater in summer than in the other seasons, accounting for more than 63% of the annual total emissions. This finding suggests that summer is the critical season in managing the CH_4 emissions from Poyang Lake. The high effluxes in summer may be attributed to the higher temperature, higher substrate availability, and greater temperature sensitivity during this season than the other seasons.

Poyang Lake features a typical monsoon climate with hot summers. During the study period, the mean (June–August) air temperature in summer was 28.5 ℃, whereas that in winter was only 5.9 ℃. The CH_4 effluxes were highly correlated with the sediment temperature through an exponential function. Our results confirmed the findings of previous studies that lake CH_4 effluxes are driven by temperature (Bastviken et al., 2008; Marinho et al., 2009; Palma–Silva et al., 2013; Rõõm et al., 2014). This is supported by the fact that a warm temperature provides a high optimal temperature for methanogen growth, which increases methane production (Nozhevnikova et al., 2007; Rooney–Varga et al., 2007; Duc et al., 2010). Moreover, recent studies have reported that high temperatures could increase the proportion of hydrogenotrophic methanogenesis, which is an important pathway for CH_4 production (Borrel et al., 2011; Marotta et al., 2014). The high summer CH_4 effluxes might also be because of the ample substrate supply in this season. In the present study, CH_4 efflux positively correlated with the Chl *a* content ($r = 0.46$, data not shown) that was not correlated with other environmental factors and acted as an indicator of primary production. Earlier studies discovered a high amount of labile organic matter, including allochthonous inputs of terrestrial organic matter, during the summer flooding and autochthonous production within-lake by phytoplankton and benthic algae in summer (Crump et al., 2003; Xing et al., 2005, 2006; Bade et al., 2007). The decomposition rate of new organic matter was much faster than that of old organic matter (Davidson and Janssens, 2006; Gudasz et al., 2010). Previous studies showed that fresh organic carbon from dead algae stimulates CH_4 emissions in lakes (Huttunen et al., 2002; Xing et al., 2005) because the degradation of dead alga and algal exudates, such as methylated compounds, are the precursors for CH_4 production (Ferrón et al., 2012; Xiao et al., 2015; Liang et al., 2016). However, we did not find any correlation between the CH_4 efflux and DOC content in the water ($p > 0.05$). The algal bloom in summer probably masked the DOC effect on stimulating CH_4 production. Earlier studies demonstrated that 70%–80% of DOC molecules in lakes are recalcitrant carbon, which are composed of humic substances in the lake from the partial degradation of terrestrial lignin in vegetation (Tranvik and Kokalj, 1998; Wetzel, 2001).

The high summer CH_4 effluxes were also driven by the greater temperature sensitivity during summer. The apparent Q_{10} value in Poyang Lake was 2.04 in summer, which was much greater than the value of 1.67 in the other seasons (Fig. 6). This finding is inconsistent with previous studies in terrestrial and freshwater ecosystems (Davidson and Janssens, 2006; Gudasz et al., 2010; Yvon–Durocher et al., 2014), where the Q_{10} values decreased apparently with the increase in temperature (Xu and Qi 2001a; Chen et al., 2010; Corkrey et al., 2012; Schipper et al., 2014). However, our result was supported by a recent finding that the temperature sensitivities (Q_{10}) of CH_4 effluxes from lake sediments are greater in the tropics than in boreal regions (Marotta et al., 2014). We speculate that the temperature effect on Q_{10} was confounded by other factors, such as water level and substrate availability. The addition of a large amount of fresh carbon from summer floods could

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dramatically boost CH_4 production and thus the apparent Q_{10} values during summer.

4.3. Timescale dependence of wind, substrate availability, and temperature effects on CH_4 effluxes

In this study, the effects of wind, substrate availability, and sediment temperature on CH_4 effluxes were highly timescale dependent. The CH_4 effluxes measured at bihourly intervals positively correlated with wind speed in both simple and multiple regressions (Figs. 5a–d, Table 2) but showed no correlation ($p > 0.05$) when the diurnal or seasonal average CH_4 efflux and wind speed were applied (Figs. 5e–f). The effect of wind on CH_4 effluxes was mainly through its effects on the transport, air pressure and storage of CH_4 from the bottom to the surface water (Abril et al., 2005; Hahm et al., 2006; Guérin et al., 2007). Gas diffusion in water is sensitive to pressure changes at the water–air interface (Paganelli et al., 1975; Massmann and Farrier, 1992; Striegl et al., 2001; Nachshon et al., 2012). High wind speed mechanically induces turbulences through friction in the water and brings CH_4-rich water from the bottom to the surface in lakes (Wanninkhof, 1992; Palma–Silva et al., 2013; Xiao et al., 2013). The CH_4 efflux rapidly decreases or even becomes negative (indicating CH_4 absorption) to compensate for the deficits in the water profile caused by earlier winds when the wind declines or comes to a halt. Our results also confirmed that the CH_4 efflux sharply declined to a negative value after strong wind events (Fig. 4). This wind effect only worked at short timescales, such as bihourly, when temperature only slightly changed and other biological processes, such as microbial community variation, were relatively stable. At a longer temporal scale, such as seasonal scale as observed in the current study, the wind effect disappeared because the wind-stimulated CH_4 effluxes and the post-wind (or between-gusts) negative effluxes (absorptions) were compensated. Our results suggest that wind exerts minor effects on

CH_4 effluxes at large temporal scales when temperature, water level, and substrate availability dominate. Our results also suggest that caution must be taken when one applies the empirical wind speed-driven models developed based on short-term measurements to estimate CH_4 effluxes over long periods, such as months or years.

Meanwhile, the CH_4 effluxes measured at monthly intervals positively correlated with sediment temperature (Fig. 6, Table 2), but the correlation disappeared when applied at bihourly intervals ($p > 0.05$). The lack of correlation between the CH_4 efflux and sediment temperature as measured on a bihourly scale within a day can be explained by the small variation of sediment temperature within a day, ranging from 0.95 ℃ to 1.85 ℃. Other factors, such as wind and atmospheric pressure, might shadow the weak temperature effect within a day. Instead, we found a high correlation between the bihourly measured CH_4 effluxes and sediment temperature during the diel measurement period in January 14 to 15, 2015 ($r = 0.88$, $p < 0.0001$). Further analyses showed that this temperature effect might be apparent and mainly caused by wind speed because the bihourly measured CH_4 effluxes and wind speed were highly correlated only in January 14 to 15, 2015 and not in the other days ($r = 0.90$, $p < 0.0001$). However, sediment temperature became the dominant factor on a seasonal scale when the temperature ranged from about 4.4 ℃ in winter to 30.8 ℃ in summer (Fig. 3). The sediment temperature and CH_4 effluxes averaged over the diurnal period significantly correlated in the 4-year study period (Fig. 6, Table 2). Our results suggest that the short-term CH_4 efflux in Poyang Lake was regulated by wind speed, but the long-term CH_4 efflux was ultimately controlled by sediment temperature and other biological (e.g., microbial activities) and biochemical (e.g., sediment carbon and nitrogen contents) processes. Therefore, understanding and modeling the dynamics of CH_4 effluxes on lake surfaces require the long-term measurements of effluxes and

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related biotic and abiotic factors in lake water and sediments. Finally, substrate availability, such as sediment TN content, TP, and Chl *a* contents in the water, also influenced CH_4 effluxes on a seasonal scale in the current study (Table 2). However, the effects disappeared when applied at bihourly intervals because the substrate did not change significantly within a day.

In addition to the above-mentioned factors, the DO concentration in the water influenced the CH_4 effluxes in the multivariate regression analysis. In specific, the CH_4 efflux closely correlated with the DO concentration in the water (r = –0.65). This close correlation can be explained by the aerobic CH_4 oxidation in the water. Our result was supported by the previous finding that a high DO concentration in the water results in low CH_4 emission (Rõõm et al., 2014; McNicol and Silver, 2015; Yang et al., 2015).

5. Conclusion

The average CH_4 efflux in Poyang Lake during the 4-year study period was 0.54 ± 0.053 mmol m^{-2} day^{-1}, which was moderately higher than that of the other lakes in the world. The CH_4 efflux in Poyang Lake also featured high seasonal variations with the maximum efflux in July and the minimum in January. About 63% of the annual emissions occurred in summer, from June to August. On a seasonal scale, multivariate regression analyses revealed that sediment temperature sediment TN content, TP, and DO contents in the water mainly regulated the CH_4 effluxes. Simple and multivariate regression analyses showed that wind speed influenced the diel CH_4 efflux variations. The effects of sediment temperature, substrate availability, and wind speed on CH_4 effluxes were temporal scale dependent. The CH_4 effluxes increased with the sediment temperature, sediment TN content, Chl *a*, and TP contents in the water on a seasonal scale but were not correlated with sediment temperature on a bihourly scale. In contrast to the temperature and substrate, the CH_4 efflux positively and significantly correlated with wind speed within a day on a bihourly scale but was not correlated with wind speed at larger temporal scales, such as daily and seasonal scales. The timescale dependence of environmental controls on CH_4 effluxes has important implications in modeling CH_4 emissions.

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Table 1 Seasonal and annual means of CH_4 effluxes with the chamber measurements in Poyang Lake

CH_4 efflux (mmol m^{-2} day^{-1})	2011	2012	2013	2014
Spring (Mar–May)	0.22 ±0.035b	0.36 ±0.092 bc	0.23 ±0.16b	0.37 ±0.084 b
Summer (Jun–Aug)	1.34 ±0.31a	1.21 ±0.16a	1.36 ±0.44 a	1.44 ±0.46a
Autumn (Sep–Nov)	0.23 ±0.12b	0.43 ±0.14b	0.33 ±0.12b	0.34 ±0.16 b
Winter (Dec–Feb)	0.11 ±0.014b	0.23 ±0.036 b	0.14 ±0.047 b	0.23 ±0.10 b
Mean	0.47 ±0.54a	0.56 ±0.41ac	0.52 ±0.55a	0.60 ±0.56bc

Note: Means with different letters are significantly different as determined by multiple comparisons on a seasonal scale (one-way ANOVA, post hoc Tukey test, $p < 0.05$) and a pair T test ($p < 0.05$) on an annual scale.

Table 2 Multivariate regressions between seasonal CH_4 efflux and environment factors

No.	Number of variables	Regression Equation	n	R^2	p
Group 1	12	$EffluxCH_4 = -10.48 + 110.57\,ST + 65.06SN$	48	0.65	0.004
Group1 + Group 2	16	$EffluxCH_4 = -12.66 + 0.57ST + 90.81SN$	43	0.73	0
Group 1 + Group 2 + Group 3	19	$EffluxCH_4 = -3.89 + 0.56ST + 102.88SN - 35.56TP - 0.74DO$	19	0.89	0

Note: Nd means that no variable input to the stepwise regression exists. Variables in group 1 included sediment temperature, sediment total nitrogen content, water level, DOC content in the water, pH in the sediment, NH_4^+ and NO_3^- concentrations in the water and in the sediment, sediment organic carbon content, the ratio of carbon and nitrogen, and the mean daily wind speed. Variables in group 2 included TN, TP, COD, and Chl *a* contents in the water. Variables in group 3 included DO content, conductivity, and pH in the water.

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Table 3 Mean CH_4 effluxes in Poyang Lake in comparison with other large lakes

Lake	Lake size (km^2)	Region	Climate	CH_4 efflux (mmol $m^{-2}day^{-1}$)	References
11 lakes	1	Laurentians, Canada	Boreal	4.08	Rasilo et al., 2015
19 lakes	47	Chicoutimi, Canada	Boreal	1.08	Rasilo et al., 2015
21 lakes	41	Abitibi, Canada	Boreal	1.67	Rasilo et al., 2015
14 lakes	171	Chibougamau, Canada	Boreal	0.17	Rasilo et al., 2015
14 lakes	7	James Bay, Canada	Boreal	1.08	Rasilo et al., 2015
45 lakes	5	C^ote-Nord, Canada	Boreal	1.17	Rasilo et al., 2015
14 lakes	2	Eastmain, Canada	Boreal	0.58	Rasilo et al., 2015
48 lakes	242	Scheffervill, Canada	Boreal	0.42	Rasilo et al., 2015
Lake Mendota	39.4	North America	Boreal	0.50	Fallon et al.,1980
Erie	25700	North America	Boreal	0.04	Howard et al.,1971
Dillon	13	North America	Boreal	0.61	Smith and Lewis,1992
Fiolen	1.5	Sweden	Boreal	0.02	Bastviken et al., 2004
Kevätön	4	Finland	Boreal	0.22	Huttunen et al., 2003
Biwa	674	Japan	Temperate	0.27	Miyajima et al.,1997
Constance	540	Europe	Boreal	0.04	Schultz et al., 2001
Kasumigaura	168	Japan	Temperate	0.26	Utsuumi et al.,1998a
Nojiri	4.4	Japan	Temperate	0.06	Utsuumi et al.,1998b
5 lakes	range 1–11, 3436[A]	Netherlands	Temperate	5.85	Schrier–Uijl et al., 2011
Donghu	27.9	China	Subtropical	1.46	Xing et al., 2005
TR lake	71.4	Pantanal, South America	Tropical	0.65[B]/5.74[C]	Bastviken et al., 2010
BB lake	36.3	Pantanal, South America	Tropical	0.50[B]/5.63[C]	Bastviken et al.,2010
Biandantang	3.3	China	Subtropical	1.32	Xing et al., 2006
Võrtsjärv	270	Estonia	Boreal	1.28[B]/2.09[C]	Rõõm et al., 2014

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43 lakes	range1–10, 782073.8[A]	worldwide	Mainly boreal	0.12	Holgerson and Raymond, 2016
18 lakes	range10-100, 597789.3[A]	worldwide	Mainly boreal	0.10	Holgerson and Raymond, 2016
6 lake	>100, 2024015.8[A]	worldwide	Mainly boreal	0.06	Holgerson and Raymond, 2016
Poyang Lake	3283	China	Subtropical	0.54	Present study

Note: A means total areas in the given lake size. B means diffusive effluxes and C means total effluxes, including diffusion and ebullition.

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Figure Captions

Figure 1. Location of sampling sites in Poyang Lake.

Figure 2. Examples of calculating the slope of total effluxes, including diffusive and ebullitive effluxes. All the concentrations are presented in original (volumetric parts per million-units). White circles represent the CH_4 concentrations at different sampling times. Grey circles represent the adjusted concentration. Black trendlines represent the data used for the total efflux calculation. The different letters in the figure panels mean different occurrence times for ebulltion: no ebullition (a), occurrence of ebullition at 20 min (b), 40 min (c), and 60 min (d), respectively.

Figure 3. Seasonal variations of CH_4 effluxes and sediment temperatures in Poyang Lake. White circles represent the variation of CH_4 effluxes, and black circles describe the variation of sediment temperature in the 4-year period.

Figure 4. Diel variations of CH_4 effluxes in Poyang Lake. Different panels present the diel variations of the CH_4 effluxes in 24–25 July 2011 (a), 5–6 September 2012 (b), 13–14 January 2013 (c), and 14–15 January 2015 (d). White circles describe the diel variations of the CH_4 effluxes. Horizontal short dashed lines mean the average value of the diel CH_4 effluxes.

Figure 5. Relationships between CH_4 effluxes and wind speed in Poyang Lake. White circles represent the observed values of CH_4 effluxes and wind speed. Different panels mean the variations of CH_4 effluxes at a bihourly interval within a day, including in 24–25 July 2011 (a), 5–6 September 2012 (b), 13–14 January 2013 (c), and 14–15 January 2015 (d), on a diurnal scale (e), and on a seasonal scale (f). Panels e and f include all the measurements during the observation period. We excluded the white-crossed circle in figure c in the regression analysis because of a severe cold front.

Figure 6. Relationship between sediment temperature and CH_4 effluxes in Poyang Lake. White circles represent the observed values of the diurnal mean CH_4 effluxes and sediment temperature in summer, and black circles represent the observed values of the diurnal mean

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CH_4 effluxes and sediment temperature in the other seasons in the 4-year period. Black lines represent the fitting curves of the relationship between CH_4 effluxes and sediment temperature.

Fig. 1

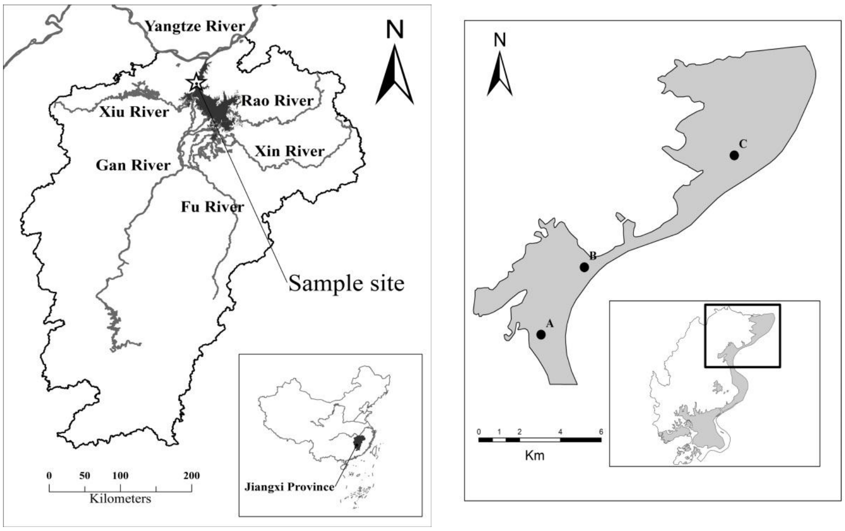
Yangtze River
N
Rao River
Xiu River
Xin River
Gan River
Fu River
Sample site
0 50 100 200
Kilometers
Jiangxi Province
N
C
B
A
0 1 2 4 6
Km

Fig. 2

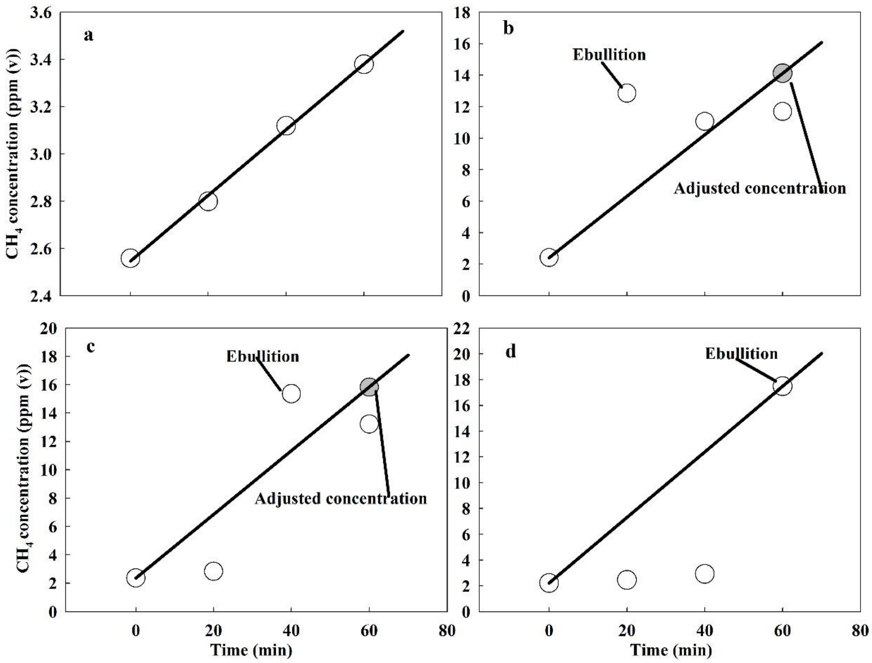
a
b
c
d
Ebullition
Ebullition
Ebullition
Adjusted concentration
Adjusted concentration
CH_4 concentration (ppm (v))
CH_4 concentration (ppm (v))
Time (min)
Time (min)

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Fig. 3

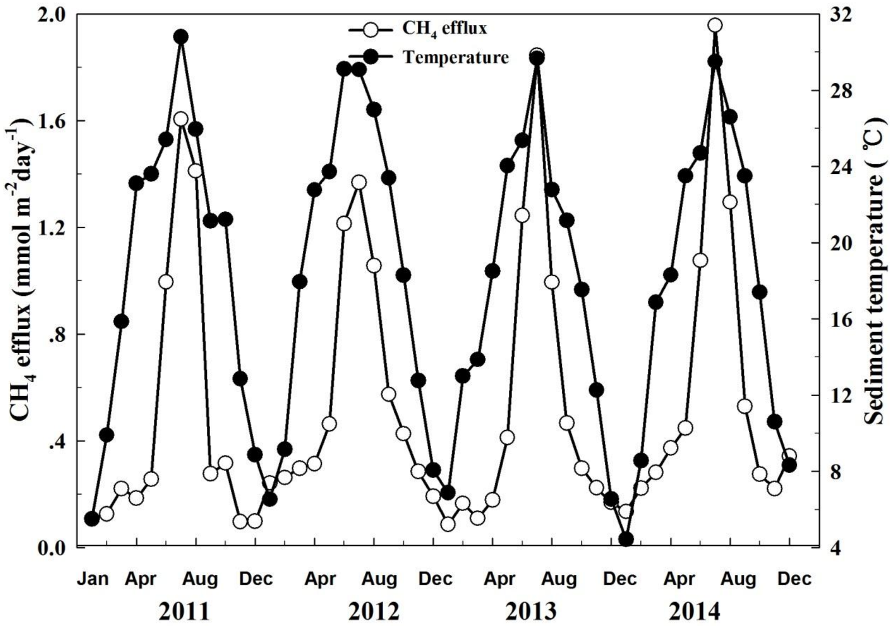

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Fig. 4

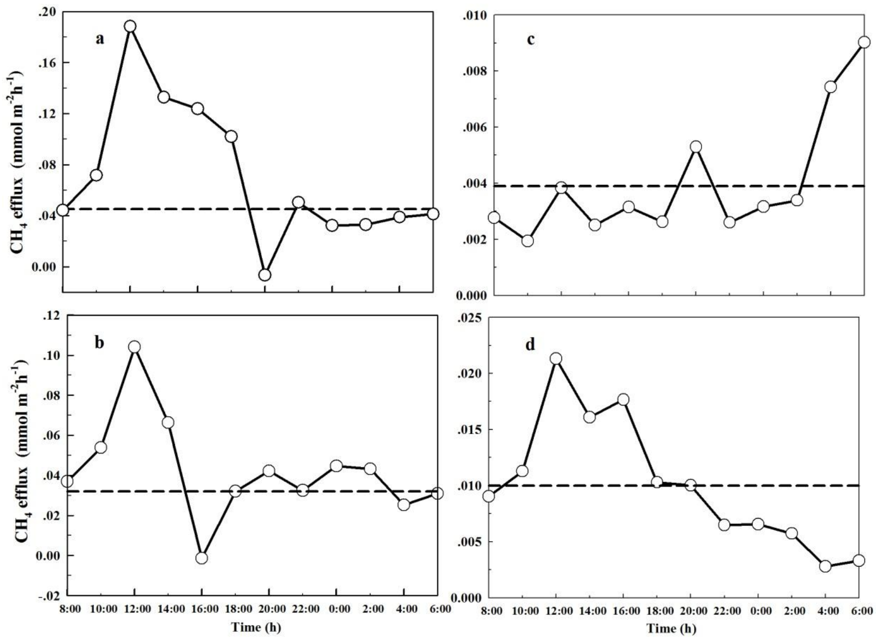

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Fig. 5

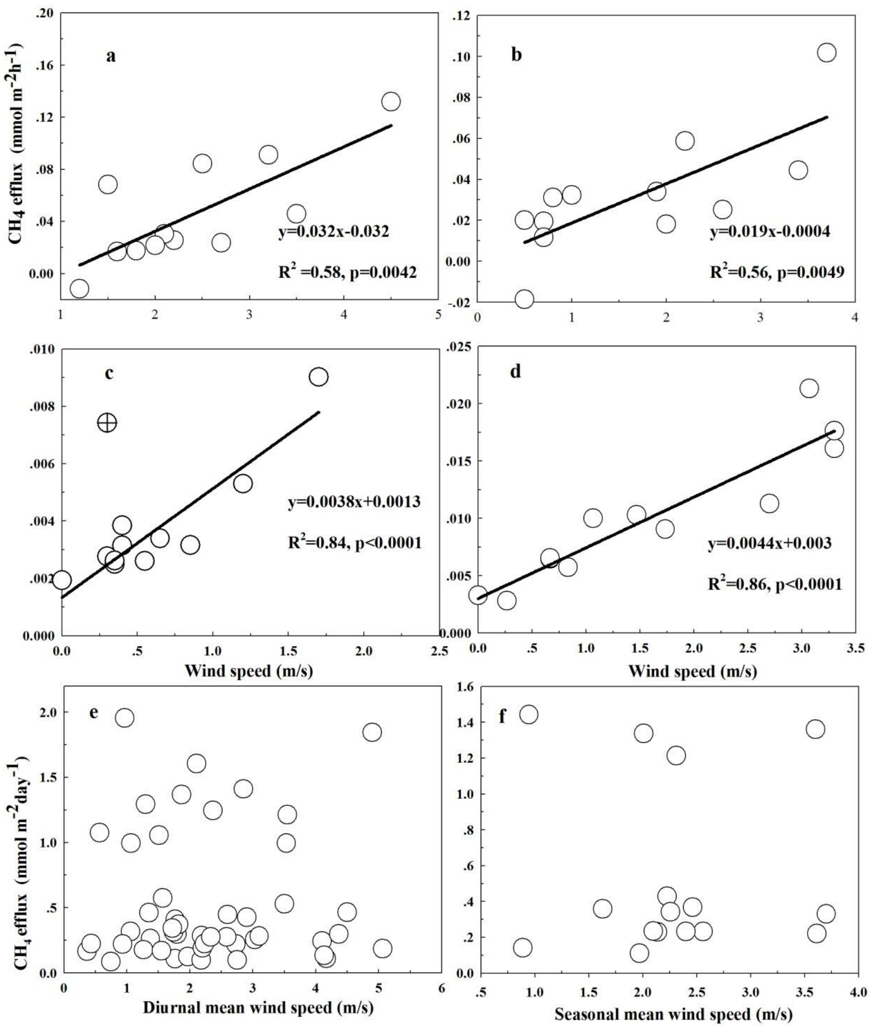

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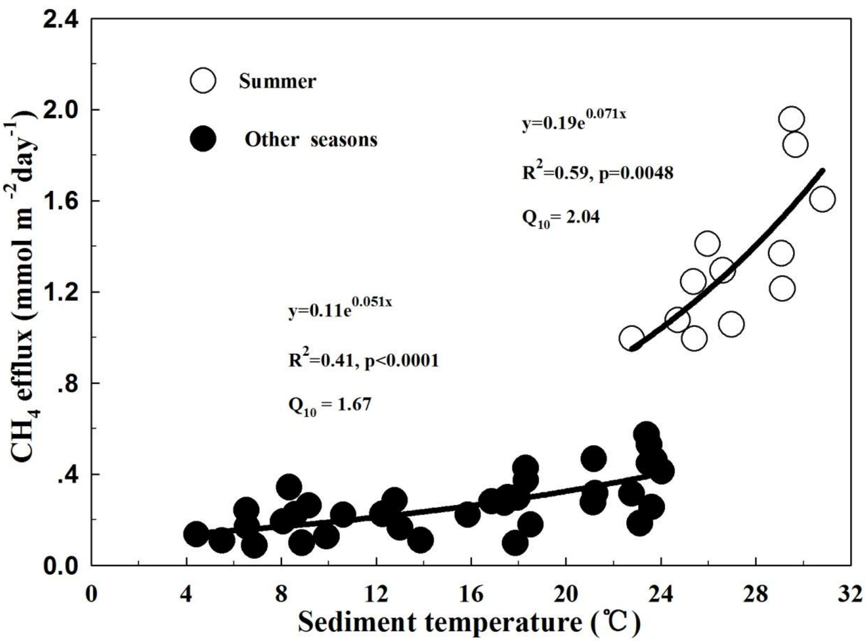
Fig.6



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