## Response to Reviewers (bg-2016-286)

#### Reviewer 1

General comments

The manuscript reports on the temporal patterns of methane (CH<sub>4</sub>) efflux in the largest lake in China and the various factors that influence these fluxes over different timescales. CH<sub>4</sub> efflux was slightly greater than other lakes with an area greater than 1km, but was comparable to that found in tropical lakes. The variables best explaining variation on CH<sub>4</sub> efflux was timescale dependent but, overall, temperature was important over seasonal scales and wind speed on a bihourly scale. The paper is well written despite a few grammatical errors. As the authors point out, there is a lack of data explaining CH<sub>4</sub> effluxes in this region and also in larger, nonalpine lakes more generally. As such, the results from this paper will add to the limited understanding of CH<sub>4</sub> dynamics in these lake types. However, I cannot recommend that this paper be published in its current form. I have major issues with a) the premise of the paper, b) some overreaching statements that are made, and c) the statistical approaches used all have major implications for the generalisation of the results. It is difficult for me to assess the results and technical aspects of this study until statistical changes are made. Answer: We thank the reviewer so much for the constructive comments and suggestions. We have considered all the comments and suggestions carefully in revising the manuscript.

1. One major concern is that the study was undertaken in a very small area (three sites with 20km of each other) even though the lake is the largest in China by area (3283sq km). Further, the study sites are situated in a section of the lake that appears to be relatively confined. There is nothing wrong with the site selection. However, the authors cannot make statements about the whole lake because they don't know if the

spatial and temporal patterns of CH<sub>4</sub> vary the same way across the lake. They need to qualify in all statements that the research was undertaken in one small section of the lake. It is not a study of CH<sub>4</sub> effluxes from Poyang Lake, but it is a study of CH<sub>4</sub> effluxes from one section of Poyang Lake.

Answer: We agree with the Reviewer that the CH<sub>4</sub> efflux in the Poyang Lake has a large spatial variation as evidenced in our previous study which examined the spatial variations of greenhouse gas effluxes (including CH<sub>4</sub>) over the lake with 44 sampling locations. The current study focuses on the temporal dynamics of CH<sub>4</sub> efflux. We chose the 3 sites to roughly represent the average CH<sub>4</sub> efflux of the whole lake based on the results of our previous study (Liu et al., 2013). Therefore, our results reflect the general situation of the lake.

2. Another major concern is a statistical one. The authors use average values from three different locations in Poyang Lake for all analyses. The justification for this was to 'minimize the effect of the spatial variation of CH<sub>4</sub> efflux on the temporal dynamics of the efflux'. However I suspect the main motivation for doing this was because the environmental variables were only collected at one location (it is not clear where the environmental variables were collected). Was this the case? Given that CH<sub>4</sub> was only measured in three locations of the lake, surely the degree of variation between them is very important to a) understand and/or b) account for in statistical analyses. The authors should re-analyse their results in one of the following ways: Åâć treat each study site as a random effect in mixed effects models so that variation among the three sites in taken into account when investigating the annual, seasonal, and diurnal variation, as well as the relationships with measured explanatory variables. Including site as a random effect would enable the researchers to make more general statements about CH<sub>4</sub> fluxes from Poyang Lake – this is just common practice these days and

should be incorporated into the study design / statistical analyses. A random effect for site effectively means that these study sites are a random sample of all potential sites in the lake – this is where the generalisability comes in. Please see Section 8.1.1 (Types of predictor variables (factors)) in Quinn & Keough (2002; Experimental Design & Data Analysis for Biologists) or another similar book for information about mixed effects models and random and fixed factors. Ăâć split the analyses into two parts. The first analysis will not average the three study sites prior to the analysis and investigate the spatial and temporal patterns in CH<sub>4</sub> among them. The second analysis could average the study sites (still preferably treat study site as a random factor) and relate this to the measured explanatory variables.

Answer: We actually collected environmental variables at each site except water level which was monitored at the Xingzi Hydrological Station. We appreciate the Reviewers' suggestion (also see Reviewer 2's comments) and re-analyzed the data by treating the site as a random effect. We found that the site effect was not statistically significant including seasonal and diel CH<sub>4</sub> effluxes over the 4-year period (Table S2, S3 in the supplementary material). We also re-analyzed our data for each site and found that the differences among the 3 sites were minor with the 4-year mean of 0.53 mmol m<sup>-2</sup> day<sup>-1</sup>, 0.55 mmol m<sup>-2</sup> day<sup>-1</sup>, and 0.54 mmol m<sup>-2</sup> day<sup>-1</sup> respectively. In addition, we found that the seasonal patterns of CH<sub>4</sub> effluxes at three sites were similar and also in line with the seasonal pattern averaged over the 3 sites. Nevertheless, in the stepwise multiple regressions analyses, the same environmental variables were selected in the final model for each individual site as for the average of the 3 sites with the regression coefficients slightly different (Table S5), but not statistically significant (p > 0.20). So we have focused on presenting the site-averaged CH<sub>4</sub> efflux and its dynamics

due to the length limitation of the paper. But we explained the site effect on CH<sub>4</sub> effluxes in the revised version.

3. One more major concern is the notion that this is a long-term study. 4 years is not long term. Remove all reference to this study being long term, including the second sentence of the Abstract which introduces the idea that this research is filling the knowledge gap around the lack of long term research on CH<sub>4</sub> fluxes. Instead, the authors should frame this 'knowledge gap' around the lack of multi-seasonal investigations into CH<sub>4</sub> effluxes – this is exactly what this paper addresses.

Answer: We agree and thank you so much for the constructive suggestion. We removed the phrase "long term" and changed the tones accordingly in the text during the version. In addition, we have focused on multi-seasonal investigations of  $CH_4$  effluxes as suggested.

Specific comments

1. Line 18. It is stated continuous measurements of  $CH_4$  efflux was measured, but measurements where not continuous. Monthly measurements were made. Change all reference to continuous measurements in the manuscript to monthly measurements.

Answer: Changed as suggested in the revised version.

**2.** Line121-124. Are these parameters an average of the entire lake or for a specific location? Please specify.

Answer: These parameters are averages of the entire lake. We added the information in the revised version (Page 7/lines 135-138).

3. Section 2.3. Environmental variables. Where were the environmental variables collected from? Where samples collected at each of the three study sites and then averaged or from just one site? This information is very important.

Answer: The environmental variables were measured at each of the three study sites and then averaged over the sites except water level which was monitored at a single hydrological station (national class station). We added more details of the environmental variables in the revised version (Page 11/lines 221-222).

4. Line 331-332. This concluding sentence only relates to the first sentence of this paragraph and does not relate or link to the remaining text in the paragraph. This sentence should only be left if a re-working of the paragraph better supports this argument.

Answer: We deleted the sentence because it is not the main point of the paragraph.

5. Line 337-341. An argument is made that this study has lower diurnal variation in CH<sub>4</sub> efflux than other studies and this may be due to differences in sample size in other studies. I would think that more frequent sampling would in-fact lead to more variation. The authors need to report on how much diurnal variability in CH<sub>4</sub> efflux there was among the study sites.

Answer: We agree with the Reviewer that the diurnal range (maximum – minimum) of  $CH_4$  efflux depends on sample size and sampling frequency, which makes the comparison with other lakes less meaningful. Therefore, we deleted the discussion on comparing the ranges of  $CH_4$  effluxes in different lakes, which are not the main focus of the current study (Also see the reviewer 2'comments).

Technical comments

1. Line 163-182. The description of how CH<sub>4</sub> efflux due to ebullition is very confusing and long.

Answer: We rewrote this part to clarify the confusion in the revised version (Page 10/lines 190-200).

2. From Line 125, where the ebullition and diffusive fluxes are introduced, I would

suggest briefly describing how, or how not, the chambers can be used to differentiate

these two fluxes.

Answer: Chambers cannot be used to differentiate ebullitive and diffusive fluxes.

In the current study, the chambers can give the total flux including ebullitive and

diffusive fluxes. We rewrote this section as suggested in the revised version.

3. Line 312. Remove 'obviously'.

Answer: Removed as suggested (Page 18/line 380).

**Reviewer 2** 

This manuscript presents 4 years of CH<sub>4</sub> flux patterns in the largest lake in China

and environmental factors that influence CH4 flux rates. It falls well within the scope

of Biogeosciences, but several aspects need to be improved for publication. Some

suggestions: 1) How do you define "long-term"? To me, 4-year observations can be

short-term. Also, all the statements related to seasonal or inter-annual variability need

to be justified because CH<sub>4</sub> flux rates measured on one day may not represent flux

rates of one month. Furthermore, daily CH<sub>4</sub> flux rates could have been overestimated,

considering that CH<sub>4</sub> flux rates are measured during the day each month, when CH<sub>4</sub>

flux rates were higher than those at night according to diel cycle measurements. 2) All

the assumptions are met for regression models? Did you consider any interactions

among variables? In addition, did you also carry out the analysis before averaging the

flux rates, with replicates as random effects? If so, how did the results differ from

those after averaging? 3) In the discussion section some results were described, which

did not appear in the result section. Results and discussion need to be better separated.

In addition, the interpretation of the results needs to be better supported in the

discussion section, focusing clearly on the core messages, i.e., what the results mean and what we can learn from this study.

Answer: We thank the reviewer so much for the constructive comments and suggestions. We have considered all the comments and suggestions carefully in revising the manuscript. Firstly, we avoided using "long-term" as suggested and focused on multi-seasonal dynamics of CH4 effluxes. We totally agree with the Reviewer that the measured CH<sub>4</sub> effluxes on one day did not represent the mean efflux rate of the month. We used the daily measurements as sampling points to explore the relationships between the CH<sub>4</sub> efflux and environmental variables. We calculated the monthly, seasonal and annual mean CH<sub>4</sub> effluxes using interpolation method (e.g. regression or the random forest model). It is true that most of our measurements were taken during the daytime. However, the daytime and nighttime average CH<sub>4</sub> effluxes were not statistically different (p = 0.19). Moreover, we built our statistical models based on the daytime mean efflux and daytime averages of environmental variables and the nighttime efflux was calculated based on the nighttime averages of the same environmental variables. This avoided the overestimation of daily CH<sub>4</sub> efflux. Secondly, we re-analyzed our data for each site and also treated site as a random effect as suggested. As a result, we found that site had no significant effect on the measured seasonal and diel CH<sub>4</sub> effluxes over the 4-year period (Table S2, S3). In the stepwise multiple regressions analyses, the same environmental variables were selected in the final model for each site as for the 3-site average though the coefficients of each variable were slightly different (Table S5), but not statistically significant (p > 0.20). The seasonal patterns of CH<sub>4</sub> effluxes at individual sites were very similar to the seasonal pattern by averaging CH<sub>4</sub> effluxes over the 3 sites. Therefore, we used average values of the 3 sites in our analyses, but we added those information to the result section. Thirdly, we included the interactions among environmental variables in the revised version as suggested (Table S4). Finally, we rewrote the result and discussion sections as suggested to clarify relevant issues.

1. Line#47-51, there are too few references to represent the minimum and maximum flux rates in lakes, especially given that those references are from lakes in China and Norway only. Also, if such values can be presented with more studies, how would seasonal variations look like in comparison to diurnal ones?

Answer: We agree with the Reviewer that there are too few studies measuring lake  $CH_4$  efflux in the literature and the sampling size and frequency was also different among the limited number of studies (Also see Reviewer #1's comments, specific question 5). Therefore, we deleted the range (maximum and minimum) comparison among lakes and focused on comparing the mean efflux of various lakes in the revised version.

2. Line#75-78, can you add references for each variable? Line#64-72 well covered the references for each variable, but this section lacks it.

Answer: We added related references in the method section as suggested in the revised version (Page 4/lines 81-84).

3. Line#78-82, it sounds like investigating in large lakes is not important. Please rephrase or add some more sentences to justify the importance of this research.

Answer: We added some sentences and references to emphasize the importance of  $CH_4$  emissions from large lakes as suggested (Page 5/lines 88-91).

4. Line#86-87, I suggest adding references that describe the previous studies, e.g. Liu et al. (2013).

Answer: Thanks for your suggestion. We added references to describe the previous studies in the revised version (Page 5/ lines 97-102).

5. Line#109, what are the species names of Carex?

Answer: The species name of Carex in Poyang Lake is Carex cinerascens Kükenth and Carexargyi Levl.etVant. We added the species scientific names in the revised version (Page 7/ lines 123-124).

6. Line#128-146, this section can be written more concisely.

Answer: We rewrote this section as suggested in the revised version.

7. Line#166, can decreases in CH<sub>4</sub> concentrations right after ebullition events be solely explained by diffusion back to lake water? If CH<sub>4</sub> molecules were diffused back to the lake water, partial pressure of CH<sub>4</sub> inside the chamber should be very high, inhibiting further emission from lake water to chamber. Can they be partially from irregular air mixing inside the chamber, which results in errors in CH<sub>4</sub> concentrations? Then, the current method for calculating flux rates needs to be reconsidered.

Answer: We speculate that the short-term decrease or leveling-off of CH<sub>4</sub> concentration inside the chamber after ebullition was mainly caused by the back diffusion of CH<sub>4</sub> to surface water due to the high CH<sub>4</sub> concentration in the bubbles. This back-diffusion phenomenon has been evidenced for CH<sub>4</sub> efflux over water surfaces (Varadharajan et al., 2010; Wik et al., 2013). The ebullition suddenly increased CH<sub>4</sub> concentration, and thus partial pressure of CH<sub>4</sub>, in the chamber headspace, which reversed the normal CH<sub>4</sub> diffusion gradient between surface water and chamber space. We do not think irregular mixing is the main cause in the current study because we had a mixing fan running in each chamber during the whole period of measurement.

**8.** Line#167-182, this section is confusing. It can be written clearly and concisely.

Answer: We rewrote this section more clearly and concisely as suggested.

9. Line#200, were water and sediment samples collected at three sampling points for flux measurements? The paragraph from line#229 can be given in a Table.

Answer: Yes, we collected water and sediment samples at each of the three sampling sites when taking flux measurements. We added a table (Table S1) to the supplementary material section in the revised version as suggested.

10. Line#241, T test  $\rightarrow$  t-test

Answer: Thank you for pointing out the typo. We changed "T test" to t-test as suggested.

11. Line#242, flux rates are measured three times per season and they may not well represent flux rates of one season of the year. Then, can deviation of these three values be used to quantify inter-annual variability?

Answer: We agree that 3 measurements in a season for a given year are not enough to represent the seasonal mean CH<sub>4</sub> efflux due to the high temporal variation of the efflux. In the current study, we used 4-year data to compare the seasonal variations, which means 12 data points for each season. We changed the values in Table 1 accordingly by using 12 data points to calculate the seasonal mean effluxes in the revised version (Page 40). For quantifying inter-annual variability we have to interpolate the measured CH<sub>4</sub> effluxes to annual efflux through modeling approach. The details of the modeling work were presented in another paper (Liu et al. 2016, in revision). We used the model results to compare the inter-annual, seasonal, and diurnal variabilities of CH<sub>4</sub> efflux in the Poyang Lake.

12. Line#247, please write what b represents in the equation.

Answer: Thank you for your suggestion. Here b is the exponent of the

exponential function between  $CH_4$  efflux and sediment temperature. We added it to the text in the revised version (Page 13/lines 275-276).

13. Line#278, what do you mean by "inconsistent and obvious"?

Answer: This is a typo. We fixed it in the revised version (Page 15/line 312).

14. Line#309-331, this part can be written more concisely.

Answer: Rewritten as suggested.

15. Line#331-332, sentences of this paragraph do not support this conclusion.

Answer: We deleted the concluding sentence.

16. Line#335, here again, can the absolute values be compared with a few references, which are probably based on different observation periods?

Answer: We agree that comparing the extreme values (minimum and maximum) among different lakes is not much meaningful. So, we deleted the relevant text and focused on comparing diurnal patterns.

17. Line#338-342, a larger number of data points can produce wider range of values.

Answer: See answers to question #16.

**18.** Line#345-356, possible explanations can be added, such as potential drivers that can affect diel CH<sub>4</sub> flux patterns and their variations (if measured).

Answer: Wind speed strongly influenced diel  $CH_4$  efflux variations in our study. We discussed this point in the 4.3 section.

19. 4.2 CH<sub>4</sub> effluxes in summer, this section contains a lot of new results, which were not presented in the result section. Also, some sentences describe very detailed information from other studies, which hinders the main focus of the paragraph.

Answer: We moved them to the "Results" section and rewrote the discussion by focusing on our own results.

20. 4.3 Timescale dependence of wind, substrate availability, and temperature effects

on CH<sub>4</sub> effluxes, here again, a lot of new results are reported, such as line#410-414, line#436-451, line#457-461 (repetition from result section), and line#462-468.

Answer: Again, we moved the results to the "Results" section and rewrote the discussion accordingly.

21. Line#473-475, considering uncertainties related to infrequent measurements (CH<sub>4</sub> efflux rates measured on one day may not represent the mean rates of that month), this kind of statement needs to be corrected.

Answer: According to our model-based interpolation we found that July had the maximum monthly efflux, while January had the minimum. This conclusion is coincidently in line with the 4-year measurements though we had only 4-day measurements in each month. Therefore, we think that the conclusion still holds.

22. Table 3, can you add the observation period of each study for better comparison? Also, sorting the rows by lake size and climate would make this Table easier to read.

Answer: Great idea! We added the observation period of each study and sorted the rows by lake size in the revised version.

23. Figure 3 and 4, can you add error bars from spatial variability?

Answer: We added errors bars from spatial variability for Figure 3 and 4 as suggested in the revised version.

#### **Reviewer 3**

Specific Comments:

1. Most of the results and discussions were built on the environmental variables and methane flux data. However, there are no data of biogeochemical related environmental variables shown in the figures and tables except Table 2. I would suggest to present the raw data of measured environmental variables in the supplementary material.

Answer: Thank you for the suggestion. We added a table (Table S1) to the Supplementary Material section to present the raw data of measured environmental variables, such as sediment total nitrogen content, water level, DOC content in the water, and pH in the sediment, in the revised version.

2. Substrate availability (Line, 432), biological (e.g., microbial activities) and biochemical (e.g., sediment carbon and nitrogen contents processes) (Lines 454-455) are very important factors to link methane efflux to the biogeochemical cycles and understand methane source and sink. Unfortunately, no comprehensive data or evidence to support the role of substrates and microbial activities on methane efflux in this manuscript which could be an important contribution to this journal.

Answer: We agree with the Reviewer that substrates and microbial activities are important to understanding methane sources and sinks in lakes. In our earlier studies we found that sediment carbon and nitrogen ratio were highly correlated with microbial biomass and community structure (Liu et al., 2015) which was also highly associated with greenhouse gas (CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O) fluxes in the Poyang Lake (Liu and Xu, 2016). In the current study, we focus on examining environmental variables (e.g. climate) that may affect the temporal patterns and variations of CH<sub>4</sub> effluxes in the Poyang Lake. We have added the related information and references to the discussion section in the revised version. Further investigation on the mechanisms of biological and biochemical controls on CH<sub>4</sub> production and oxidation requires lab-based experiments with isotope and microbial DNA sequencing techniques which are beyond the scope of the current study.

3. It might be a risk to use the data from three sampling sites measured from one day (1 hr? Line 148) to represent methane efflux in that month. For example, it appears a

contradiction between high methane efflux measured in July 2011 in Figure 3 and low methane efflux measured in July 2011 in Figure 4a.

Answer: We measured CH<sub>4</sub> effluxes at monthly interval to examine the seasonal dynamics of the efflux and the value does not necessarily represent the monthly average of CH<sub>4</sub> efflux. We measured CH<sub>4</sub> effluxes from early morning to late afternoon with about 6 cycles of measurements during the day (Pages 10-11/lines 213-219). The values of methane efflux measured in July 2011 in Figure 3 and Figure 4a are different because of different units. CH<sub>4</sub> efflux in Figure 3 was measured on a daily scale, but CH<sub>4</sub> efflux in Figure 4a was based on the hourly scale. So we used different units to present seasonal and diel patterns of CH<sub>4</sub> effluxes.

4. How long and what time did the authors deploy the floating chambers in the three sampling sites within a day for the study at the large temporal scales (Fig. 3)? I feel 4-year measurements are not a very large temporal scale especially there are no continues measurements/monitering such as deploying floating chamber within a short interval (every week or every two to three days). Since high methane efflux was shown in the early mornings in Fig. 4a, b and d, were the floating chambers deployed at the same time at three different sites for the data shown in Fig. 3?

Answer: We measured CH<sub>4</sub> fluxes from early morning to late afternoon with about 6 cycles of measurements during the day for the 4-year study. For sampling frequency we measured every monthly. We agree with the Reviewer that 4 year is not "long-term" given the relatively low sampling frequency. So we deleted "long-term" and focused on the multi-seasonal investigations of CH<sub>4</sub> effluxes as suggested by Reviewer 1 in the revised manuscript. We used three boats to monitor CH<sub>4</sub> fluxes at the three sites, so the floating chambers were

## deployed at about the same time at the sites as shown in Fig. 3.

5. The area and water table of Poyang Lake fluctuate dramatically between the wet and dry seasons. The authors only have short but not clear descriptions of the effect of water level on methane efflux, e.g., in Lines 404-405 and Line 432. Methane efflux might be high in dry seasons instead of summer, since methane efflux is expected to be high under lower water level due to decreasing of the hydrostatic pressure (e.g. Chanton et al. 1989). Are there any difference in water level between three sampling sites in different seasons (The mean water depth at three sites should not be always 3m through the whole year; Line 186)? The authors might consider a simple calculation of methane solubility changes due to water level fluctuations to strength the role of water level on methane efflux, e.g., Line 432.

Answer: It is true that the Poyang Lake features a large seasonal variation of water level, high water level in summer and low in winter. However, the water level at the 3 sites was very similar at a given time of the year. We agree that hydrostatic pressure affects CH<sub>4</sub> efflux as reported in Chanton et al. (1989), but our data showed that CH<sub>4</sub> efflux was positively correlated with water level. This is because the water level in the Poyang Lake co-varies with other factors, such as temperature and NH<sub>4</sub><sup>+</sup> content in the water, which also affect the CH<sub>4</sub> efflux throughout the year. For example, we found that the CH<sub>4</sub> efflux was highly correlated with sediment temperature at an annual scale. Our results suggest that the CH<sub>4</sub> efflux in the Poyang Lake was dominated by temperature rather than water level. The high CH<sub>4</sub> efflux in summer was contributed to strong microbial activities induced by warmer temperature and high substrate availability from the flooding water in summer. Therefore, we think the positive correlation between CH<sub>4</sub> efflux and water level in the Poyang Lake is a pseudo

relation which does not reflect the hydrostatic pressure effect on CH<sub>4</sub> efflux as evidenced by Chanton et al. (1989). It is possible to examine the water level effect by calculating CH<sub>4</sub> solubility change due to water level fluctuation. However, given the large seasonal variation of temperature in the study area it is very difficult to separate the water level effect based on the CH<sub>4</sub> efflux measurements on the water surface. In addition, water level induced CH<sub>4</sub> solubility change may affect short-term (minutes to hours) CH<sub>4</sub> diffusion gradient and thus CH<sub>4</sub> efflux and it should have little impact on CH<sub>4</sub> efflux as long as a new diffusion equilibrium has established. Thus, we did not calculate methane solubility changes to further investigate the water level effect on CH<sub>4</sub> efflux in revised the manuscript.

6. As the authors stated in the introduction that methane is driven by three major mechanisms such as molecular diffusion, bubble ebullition and plant-mediated transportation, bubble ebullition is not the only pathway for methane to transport from water to the air. However, data for dissolved methane concentrations in lake water and sediments are lack in this study. No bubble ebullition doesn't mean no methane efflux. I would suggest to include diffusive methane flux to the air for comparison in the future by analyzing surface water methane concentrations and using the equation from the gas-transfer model e.g., Wanninkhof (1992).

## Answer: Great idea! We will take this suggestion in our future study.

7. Since many environmental factors and methane fluxes collected in October 2010 in Poyang Lake have been shown in Liu et al., (2013) for spatial studies, the authors may include Liu et al. (2013) in the introduction and discussions to emphasize why the three sampling sites were chosen in this timescale study and the relations between different environmental factors and methane effluxes in Autumn (October).

Answer: Based on our previous study which examined the spatial variation of  $CH_4$  efflux in the Poyang Lake (Liu et al., 2013), we chose the 3 sites which gave  $CH_4$  effluxes close to the average efflux of the lake. We provided detailed information of Liu et al. (2013) in the introduction and discussion sections as suggested in the revised version.

Minor Comments:

1. Lines 57-59: Please add references for the studies in high-latitude, tropical and subtropical lakes.

Answer: We added references for the studies in high-latitude, tropical and subtropical lakes in the revised version (Page 4/lines 61-63).

2. Line 129: What fluxes did the floating chamber measured while inserting 20 cm above the water surface?

Answer: The chamber measured the total CH<sub>4</sub> efflux including diffusive and ebullitive fluxes as described in the method section. The plant-mediated CH<sub>4</sub> transportation was negligible because no vascular plants grew above water surface at our study sites.

3. Line 150: the air samples ==> the gas samples

Answer: Changed as suggested.

4. Line 159-160; Fig. 4: Since methane efflux was calculated by using a linear regression model to the methane concentration data, should the minimum value be zero instead of a negative value? There should be no negative methane value detected by GC.

Answer: The negative efflux means CH<sub>4</sub>-uptake by the lake water due probably to the short-time change in air pressure.

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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<sub>4</sub> to the atmosphere. However, the long term-multi-seasonal CH<sub>4</sub> efflux in lakes has been rarely studied. In this study, the CH<sub>4</sub> efflux in Poyang Lake, the largest freshwater lake in China, was measured continuous month ly over a 4-year period by using the floating chamber technique. The mean annual CH<sub>4</sub> efflux throughout the 4 years was 0.54 mmol m<sup>-2</sup> day<sup>-1</sup>, ranging from 0.47 to 0.60 mmol m<sup>-2</sup> day<sup>-1</sup>. The CH<sub>4</sub> efflux had a high seasonal variation with an average summer (June to August) efflux of 1.34 mmol m<sup>-2</sup> day<sup>-1</sup> and winter (December to February) efflux of merely 0.18 mmol m<sup>-2</sup> day<sup>-1</sup>. 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<sub>4</sub> efflux. However, the CH<sub>4</sub> 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<sub>4</sub>) 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<sup>-1</sup> in 1999–2006; this rate rapidly increased to 6 ppb year<sup>-1</sup> 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<sub>4</sub> emissions from global lakes account for up to 14.9% of natural CH<sub>4</sub> emissions (IPCC, 2013). However, this estimate has been associated with large uncertainties because of the high spatial and temporal variations of CH<sub>4</sub> emissions and the insufficient multi-seasonal long term measurements of CH<sub>4</sub> 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<sub>4</sub> effluxes in lakes feature high temporal variations (K iki, 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 <a href="mean-minimum and maximum">mean-minimum and maximum</a> CH<sub>4</sub> effluxes over a day were <a href="0.018-1.36">0.018-1.36</a> and <a href="12868.85-27">12868.85-27</a> mmol m<sup>-2</sup> day<sup>-1</sup>, 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<sub>4</sub> effluxes highlight the importance of frequent and <a href="long-term">long-term</a> multi-seasonal measurements (Bastviken et al., 2008; Chen et al., 2013; Bastviken et al., 2015). Unfortunately, most earlier studies on CH<sub>4</sub> 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-seasonal

measurements of CH<sub>4</sub> effluxes have only been conducted in high-latitude lakes (Utsumi et al., 1998a, b; Huttunen et al., 2003; Rõõm et al., 2014; Wik et al., 2014), and few studies on tropical and subtropical lakes (Xing et al., 2005, 2006; Ortiz–Llorente and Alvarez–Cobelas, 2012), especially large ones, had measurement durations longer than one year.

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The magnitude of CH<sub>4</sub> emission mainly depends on the dynamic balance between the microbial processes of CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> 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 (Palma-Silva et al., 2013; Rõm et al., 2014), wind-forced mixing (Wanninkhof, 1992; Palma-Silva et al., 2013), water depth\_(Liu et al., 2013), boundary layer dynamics\_(Poindexter et al., 2015; Anthony and Macintyre, 2016, hydrostatic pressure (Chanton et al., 1989), and

different vascular plants (Juutinen et al., 2009; Zhu et al., 2016). Most studies examined CH<sub>4</sub> emissions and their influencing factors in small lakes because of their large contribution to the global CH<sub>4</sub> budget (Bastviken et al., 2004; Downing et al., 2010; Bartosiewics et al., 2015; Holgerson et al., 2016). Although small lakes are a large source of atmospheric CH<sub>4</sub>, CH<sub>4</sub> emissions from large lakes was not neglected due to their large areas (Bastviken et al., 2010; Rasilo et al., 2015; Townsend-Small et al., 2016). However, few studies reported temporal CH<sub>4</sub> 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<sub>4</sub> effluxes based on multi-seasonal long term measurements in a large lake is also important to estimate lake CH<sub>4</sub> emissions.

Poyang Lake, a subtropical lake, is the largest freshwater lake in China, but its annual\_multi-seasonal\_CH<sub>4</sub> emissions have not been adequately measured. In our previous study, we have explored the spatial variations of CH<sub>4</sub> efflux over the lake with 44 sampling locations (Liu et al., 2013). In addition, we also found that microbial biomass and community structure highly influenced CH<sub>4</sub> efflux in Poyang Lake (Liu et al., 2016). In this study, we measured the CH<sub>4</sub> efflux in three sites which we chose on the basis of our previous result over the course of 4 years in Poyang Lake to (1) examine the annual\_multi-seasonal mean\_CH<sub>4</sub> efflux; (2) explore the CH<sub>4</sub> efflux dynamics, including diel\_and\_seasonal\_and\_inter\_annual\_and\_variations; and (3) quantify the relationships between the CH<sub>4</sub> efflux and environmental factors, and identify the possible factors driving CH<sub>4</sub> effluxes at different temporal scales.

## 2. Materials and methods

#### 2.1. Site description

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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 km² and a total catchment area of 162,000 km², 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 °C and an annual precipitation of 1680 mm (Ye et al., 2011). Vegetation in the lake is composed of macrophytes, including *Carex* sp. (Carex cinerascens K ükenth and Carexargyi Levl.etVant) 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 mean concentrations of total nitrogen (TN), total phosphorous (TP), suspended solids (SS), and chlorophyll *a* (Chl *a*) in the Poyang lake were 3.45, 0.11, 39.98, and 9.04 mg L<sup>-1</sup>, respectively (Yao et al., 2015).\_

## 2.2. CH<sub>4</sub> efflux measurements \_

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The CH<sub>4</sub> 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 2015) have recently conducted a systematic comparison of the effects of chamber shape, dimension, and insertion depth into the water on CH<sub>4</sub> effluxes and found that insertion depth only slightly affects the CH<sub>4</sub> 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, <u>However,</u> the water in Poyang Lake did not have an apparent directional flow except for some waves during the measurement period. So 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 in the current study. A detailed description of the floating chamber system can be found in Liu et al. (2013). So we measured the

total CH<sub>4</sub> efflux including both ebullition and diffusive effluxes and cannot differentiate ebullitive and diffusive fluxes by our chamber.

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 gas-air samples were transported immediately to a laboratory for CH<sub>4</sub> concentration analysis. The CH<sub>4</sub> 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<sub>2</sub>) as the carrier gas, which ran at a flow rate of 30 mL min<sup>-1</sup>. 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 °C and 250 °C, respectively.

Calculation of the CH<sub>4</sub> efflux was based on the CH<sub>4</sub> concentration of the four samples using a linear regression model—which was calculated on the basis of the slope of the concentration change during the whole period when the chamber was closed. 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<sup>2</sup>) greater than 0.95. In case of ebullition, the CH<sub>4</sub> concentration inside the chamber would deviate from the normal trend. Most of the CH<sub>4</sub> concentrations measured immediately after the ebullition point slightly decreased mainly because of the CH<sub>4</sub> diffusion back to water when the CH<sub>4</sub> concentration inside the chamber space increased suddenly from bubbling. To include the ebullition-induced CH<sub>4</sub> emissions, we only used two

measured concentrations, the first measurement (ambient concentration) and an ebullition-adjusted concentration that was obtained by adding the diffusion-induced concentration increment when ebullition occurred (Fig. 2)., in calculating the CH<sub>4</sub> 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. e total CH4 efflux, which includes both ebullition and diffusive effluxes, was culated 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 which was from 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.

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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<sub>4</sub> efflux in the lake (Liu et al., 2013). Therefore, we focused on the long-term multi-seasonal dynamics of CH<sub>4</sub> 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<sub>4</sub> 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

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Various environmental variables were also measured in the lake sediment, surface water, and atmosphere at each individual site and then averaged when we used. 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 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). The values of measured environmental varibles in our study were given in

Considering the different sampling periods, we classified the environmental variables into three groups (Table S1). The first group included sediment temperature, sediment total nitrogen content, water level, DOC content in the water, pH in the sediment, NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> 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

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Table S1.

We averaged the CH<sub>4</sub> effluxes of the three sites to minimize the effect of the

spatial variation of CH<sub>4</sub> 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<sub>4</sub> effluxes. The coefficient of variation (CV) was used to quantify the inter-annual variation of CH4 efflux. We employed stepwise multiple regressions to identify the environmental factors driving the CH<sub>4</sub> effluxes at different temporal scales. We also used regression and correlation analyses to determine the relationships between independent variables and CH<sub>4</sub> effluxes. In addition, we considered each study site as a random effect in linear mixed effects models in order to take into account CH<sub>4</sub> efflux variations among three sites when we investigated seasonal and diurnal variations as well as the relationships between CH<sub>4</sub> efflux and environmental variables. We used the Vant' Hoff equation to calculate the temperature sensitivity ( $Q_{10} = e^{10b}$ , where b is the exponent of the exponential function between CH<sub>4</sub> efflux and sediment temperature) of CH<sub>4</sub> 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 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<sub>4</sub> effluxes in Poyang Lake

## 3.1.1. Annual CH<sub>4</sub> effluxes

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The mean CH<sub>4</sub> efflux was 0.54 year period, with annual mean effluxes of 0.47 and  $0.60 \pm 0.56$  mmol m<sup>-2</sup> day<sup>-1</sup> in 2011, 2012, 2013, and 2014, respectively The inter-annual variation of CH<sub>4</sub> efflux was moderately his 3.1.21. Seasonal CH<sub>4</sub> effluxes

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The seasonal variations of CH<sub>4</sub> effluxes in Poyang Lake were prominent, demonstrating a similar pattern to that of seasonal temperature (Fig. 3). In general, the annual maximum CH<sub>4</sub> effluxes occurred in summers and the minimum in winters. The CH<sub>4</sub> efflux increased slowly in early spring and then rapidly in May, reaching its maximum in July. After reaching the maximum, the CH<sub>4</sub> efflux decreased sharply in August and September and then slowly before reaching its minimum in January (Fig. 3). Significant differences in the mean CH<sub>4</sub> effluxes existed between summers and the other three seasons throughout the 4 years (p < 0.05), whereas the differences in the CH<sub>4</sub> effluxes among the spring, autumn, and winter seasons were not statistically significant (p > 0.05) (Table 1). Additionally, the site effect was not statistically significant over the 4-year period (Table S2). The differences among the three sites were minor with the 4-year mean of 0.53 mmol m<sup>-2</sup> day<sup>-1</sup>, 0.55 mmol m<sup>-2</sup> day<sup>-1</sup>, and 0.54 mmol m<sup>-2</sup> day<sup>-1</sup> respectively. In particular, the seasonal patterns of CH<sub>4</sub> effluxes at the three sites were similar and also in line with the seasonal pattern averaged over the three sites.

## 3.1.32. Diel CH<sub>4</sub> effluxes

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The CH<sub>4</sub> 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<sub>4</sub> efflux was asymmetric, fast increasing in the morning from 8:00 h to 12:00 h and slowly decreasing in the afternoon and during the night, especially in the summer (Fig. 4). However, the diel pattern of the CH<sub>4</sub> 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 14<sup>th</sup>. The CH<sub>4</sub> efflux magnitudes were significantly larger during summer compared to winter. The diel pattern of CH<sub>4</sub> efflux was vague with an average difference between the daily maximum and minimum of only 0.073 mmol m<sup>-2</sup>h<sup>-1</sup>. The CH<sub>4</sub> efflux could also change abruptly throughout a day. For example, the efflux sharply dropped from 0.068 to -0.012 mmol m<sup>-2</sup> h<sup>-1</sup> within barely 2 h, as observed on July 23, 2011, indicating that the lake switched from a CH<sub>4</sub> 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). Furthermore, we compared the differences of diurnal patterns at each sites for the four diel sampling. Our results showed that the diel patterns of CH<sub>4</sub> effluxes were similar in the three sites for each diel investigation (Fig. 5) and the site effect was not statistically significant (Table S3). The diel pattern of the CH<sub>4</sub> efflux was somewhat different during certain hours such as from 22:00 h to 00:00h on July 24<sup>th</sup> in 2011.<del>Further analysis showed that the diel pattern of CH<sub>4</sub> effluxes followed the</del> diel pattern of wind speed (Figs. 5a-5d).

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3.2. Relationships between CH<sub>4</sub> efflux and environmental variables

3.2.1 Simple regression relationships between CH<sub>4</sub> efflux and environmental variables

In our study,  $CH_4$  effluxes increased exponentially with sediment temperature for both in the summer and in other seasons (Fig. 6). The  $CH_4$  effluxes were more sensitive to temperature in the summers than in other seasons. The temperature sensitivity, indicated by the  $Q_{10}$  values, was 2.04 and 1.67 in the summer and other three seasons, respectively (Fig. 6).

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We found that  $CH_4$  effluxes were also highly associated with other climate and environmental variables in both lake water and sediments. We found that  $CH_4$  effluxes were negatively correlated with  $NH_4^+$ , TN and DO concentrations in the lake water, but positively with  $Chl\ a$  content in the water and TN content in the sediment (Table 2). Furthermore, we found that other environmental factors, such as DOC content in the water, pH in the water and in the sediment,  $NO_3^-$  concentrations in the water and in the sediment, COD, and TP in the water, had insignificant (p > 0.05) relationships with  $CH_4$  effluxes in Poyang Lake.

In the current study, we also found that the relationship between  $CH_4$  effluxes and wind speed was scale-dependent. At the diel scale, wind speed was significantly correlated (p < 0.03) with  $CH_4$  effluxes for the average of the 3 sites at the diel scale (Fig. 7), but was weakly correlated with  $CH_4$  effluxes at the diurnal and seasonal scales (Fig. 7). In addition, the relationships between wind speed and  $CH_4$  effluxes for each individual site were similar with the relationships for the average of the 3 sites though the regression coefficients for each individual site were slightly different, but not statistically significant (p > 0.25).

3.2.2 Multiple regression relationships between CH<sub>4</sub> 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<sub>4</sub> effluxes (Table 23). It should be noted that multicollinearity didn't occurred among these significant variables (Table S4). In specific, sediment temperature and sediment TN content explained 65% of the variation in CH<sub>4</sub> effluxes for 4 years when we used the first group of factors. The sediment temperature and TN content explained 73% of the CH<sub>4</sub> 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<sub>4</sub> efflux variation when the three groups of variables were used together. Wind speed was the only significant variable for the CH<sub>4</sub> efflux variations on a diel scale. Wind speed explained 58%, 56%, 84% and 86% of the CH<sub>4</sub> efflux variations in 24–25 July 2011, 5–6 September 2012, 13–14 January 2013 and 14–15 January 2015, respectively (Figs. 547a-547d). In addition, the same environmental variables were selected in the final model for each individual site as for the average of the 3 sites though the regression coefficients were slightly different (Table S5), but not statistically significant (p > 0.20).

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## 4. Discussion

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## 4.1. CH<sub>4</sub> effluxes in Poyang Lake

The mean CH<sub>4</sub> emission in Poyang Lake was moderately higher than those in other large lakes of more than 1 km<sup>2</sup> in the world. The mean CH<sub>4</sub> emission (0.54 mmol m<sup>-2</sup> day<sup>-1</sup>) was within the reported range of approximately 0.022-5.85 mmol m<sup>-2</sup> day<sup>-1</sup> in boreal and temperate lakes over 1 km<sup>2</sup> but was obviously lower than diffusive effluxes in subtropical lakes and total effluxes (including diffusion and ebullition) in tropical lakes (Table  $\frac{34}{2}$ ). In addition, the mean CH<sub>4</sub> emission in Poyang Lake was comparable with the diffusive effluxes in tropical lakes (Table 34). For example, previous studies reported that the diffusive CH<sub>4</sub> efflux was 0.65 mmol CH<sub>4</sub> <sup>1</sup>-in the TR-Lake and 0.50 mmol m<sup>=2</sup>day<sup>-1</sup>-in the BB-Lake in the Pantanal region (Bastviken et al., 2010). However, the mean CH4 efflux in Poyang Lake was only higher than those in other lakes over 100 km<sup>2</sup> (except the V ortsjärv Lake). The low CH<sub>4</sub> efflux in the current study was unlikely caused by our floating chamber system because the CH<sub>4</sub> efflux would have increased if the insertion of chambers considerably disturbed the water profiles. The lower CH<sub>4</sub> 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<sup>-1</sup>, which was much lower than that of the 5.8 mg L<sup>-1</sup> in Biandantang Lake and 7.4 mg L<sup>-</sup> 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<sub>4</sub>

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emissions in large lakes cannot be ignored when estimating the global CH<sub>4</sub> budget because of their area.

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CH<sub>4</sub> effluxes at the air water interface showed high fluctuations in the four eycles, but showed no significant diurnal differences. The diurnal CH<sub>4</sub> efflux ranged from -0.019 to 0.13 mmol m<sup>-2</sup>h<sup>-1</sup>, which was within the reported range of other lakes -0.057 to 5.37 mmol m<sup>-2</sup>h<sup>-1</sup>) 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 CH4 efflux in previous results may be due to differences in sample size in different studies. For example, CH<sub>4</sub> efflux was measured at 2h intervals with 12 data points over a diurnal cycle in this study, but CH<sub>4</sub>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<sub>4</sub> efflux in other studies (K äki 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<sub>4</sub> 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<sub>4</sub>efflux was only measured three times at sunrise, daytime and sunset to represent a diel cycle in Bastyiken et al. (2010). In Keller and Stallard (1994) study, the daytime and nighttime CH<sub>4</sub> 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<sub>4</sub> effluxes than daytime CH<sub>4</sub> effluxes (Poindexter et al. 2015; Anthony and Macintyre 2016). However, we cannot estimate CH<sub>4</sub> effluxes by

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hydrodynamic transport because we did not measure CH<sub>4</sub> concentration in the water

in this study. Further studies are needed to address this issue in the lake.

#### 4.2. CH<sub>4</sub> effluxes in summer

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The CH<sub>4</sub> 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<sub>4</sub> 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.

The high summer CH<sub>4</sub> effluxes may due to high temperature in summer. Poyang

the mean (June–August) air temperature in summer was 28.5 °C, whereas that in winter was only 5.9 °C. The CH<sub>4</sub> effluxes were highly correlated with the sediment temperature through an exponential function in our study. During the study period, the mean (June–August) air temperature in summer was 28.5 °C, whereas that in winter was only 5.9 °C. Our results confirmed the findings of previous studies that lake CH<sub>4</sub> effluxes are driven by temperature (Bastviken et al., 2008; Marinho et al., 2009; Palma–Silva et al., 2013; Rõm 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) and Moreover, recent studies have reported that high temperatures could increase the proportion of hydrogenotrophic methanogenesis, which is an important pathway for CH<sub>4</sub> production (Borrel et al., 2011; Marotta et al., 2014). The

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high summer CH<sub>4</sub> effluxes might also be because of the ample substrate supply in this

season because Tthe decomposition rate of new organic matter was much faster than

that of old organic matter (Davidson and Janssens, 2006; Gudasz et al., 2010). In the

present study, CH<sub>4</sub> efflux positively correlated with the Chl a content ( $\frac{\text{Table } 2r = 0.46}{\text{Table } 2r}$ ) data not shown) that was not correlated with other environmental factors (Table S4) 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<sub>4</sub> 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<sub>4</sub> production (Ferr on et al., 2012; Xiao et al., 2015; Liang et al., 2016). However, we did not find any correlation between the CH<sub>4</sub> efflux and DOC content in the water (p > 0.05). The algal bloom in summer probably masked the DOC effect on stimulating CH<sub>4</sub> 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).

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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<sub>4</sub> 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 dramatically boost CH<sub>4</sub> production and thus the apparent  $Q_{10}$  values during summer. 4.3. Timescale dependence of wind, substrate availability, and temperature effects on CH<sub>4</sub> effluxes

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In this study, the effects of wind, substrate availability, and sediment temperature on CH<sub>4</sub> effluxes were highly timescale dependent. The CH<sub>4</sub> effluxes measured at bihourly intervals positively correlated with wind speed in both simple and multiple regressions (Figs.  $\frac{5a7a}{d}$ , Table 2) but showed no correlation (p > 0.05) when the diurnal or seasonal average CH<sub>4</sub> efflux and wind speed were applied (Figs. 5-67e-f). The effect of wind on CH<sub>4</sub> effluxes was mainly through its effects on the transport, air pressure and storage of CH<sub>4</sub> from the bottom to the surface water (Abril et al., 2005; Hahm et al., 2006; Gu érin 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<sub>4</sub>-rich water from the bottom to the surface in lakes (Wanninkhof, 1992; Palma-Silva et al., 2013; Xiao et al., 2013). The CH<sub>4</sub> efflux rapidly decreases or even becomes negative (indicating CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> effluxes and the post-wind (or between-gusts) negative effluxes (absorptions) were compensated. Our results suggest that wind exerts minor effects on CH<sub>4</sub> 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<sub>4</sub> effluxes over long periods, such as months or years.

Meanwhile, the CH<sub>4</sub> 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<sub>4</sub> 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 °C to 1.85 °C. 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<sub>4</sub> 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<sub>4</sub> 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 °C in winter to 30.8 °C in summer (Fig. 3). The sediment temperature and CH<sub>4</sub> 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<sub>4</sub> efflux in Poyang Lake was regulated by wind speed, but the long term—multi-seasonal CH<sub>4</sub> 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<sub>4</sub> effluxes on lake surfaces require the multi-seasonal long term—measurements of effluxes and 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<sub>4</sub> effluxes on a seasonal scale in the current study (Table 2, 3). 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<sub>4</sub> effluxes in the multivariate regression analysis. In specific, the CH<sub>4</sub> efflux closely correlated with the DO concentration in the water (F= 0.65 Table 2). This close correlation can be explained by the aerobic CH<sub>4</sub> oxidation in the water. Our result was supported by the previous finding that a high DO concentration in the water results in low CH<sub>4</sub> emission (R õõm et al., 2014; McNicol and Silver, 2015; Yang et al., 2015).

#### 5. Conclusion

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The average CH<sub>4</sub> efflux in Poyang Lake during the 4-year study period was 0.54  $\pm$  0.053 mmol m<sup>-2</sup> day<sup>-1</sup>, which was moderately higher than that of the other lakes in the world. The CH<sub>4</sub> efflux in Poyang Lake also featured high multi-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<sub>4</sub> effluxes. Simple and multivariate regression analyses showed that wind speed influenced the diel CH<sub>4</sub> efflux variations. The effects of sediment temperature, substrate availability, and wind speed on CH<sub>4</sub> effluxes were temporal scale dependent. The CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> effluxes has important implications in modeling CH<sub>4</sub> emissions.

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Table 1 Seasonal  $\frac{\text{and annual}}{\text{mean}}$ -mean of  $\text{CH}_4$  effluxes with the chamber measurements in Poyang Lake

CH <sub>4</sub> efflux (mmol m <sup>-2</sup> -day <sup>-1</sup> )	<del>2011</del>	<del>2012</del>	<del>2013</del>	<del>2014</del>
Spring (Mar May)	<del>0.22 ± 0.035b</del>	<del>0.36 ± 0.092 be</del>	<del>0.23 ±0.16b</del>	0.37 ± 0.084
Summer (Jun-Aug)	1.34 ± 0.31a	$\frac{1.21 \pm 0.16a}{1.21 \pm 0.16a}$	<del>1.36 ± 0.44 a</del>	<del>1.44 ± 0.46a</del>
Autumn (Sep. Nov)	<del>0.23 ± 0.12b</del>	<del>0.43 ±0.14b</del>	<del>0.33 ± 0.12b</del>	<del>0.34 ± 0.16 t</del>
Winter (Dec Feb)	0 <del>.11 ± 0.014b</del>	0.23 ± 0.036 b	<del>0.14 ± 0.047 b</del>	<del>0.23 ± 0.10 t</del>
<del>Mean</del>	$\frac{0.47 \pm 0.54a}{0.48}$	$0.56 \pm 0.41ac$	$\frac{0.52 \pm 0.55a}{0.52 \pm 0.55a}$	0.60 ± 0.56b

 Season
 CH<sub>4</sub> efflux (mmol m<sup>-2</sup> day<sup>-1</sup>)

 Spring (Mar–May)
 0.30±0.11bd

 Summer (Jun–Aug)
 1.34±0.32a

 Autumn (Sep–Nov)
 0.33±0.14b

 Winter (Dec–Feb)
 0.18±0.077cd

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**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

Table 2 Correlation relationship between seasonal CH<sub>4</sub> efflux and environmental factors

l	Environmental factors	Correlation coefficient	Environmental factors	Correlation coefficient
1	Dissolved oxygen	<u>-0.74**</u>	Sediment- NO <sub>3</sub>	<u>-0.2</u>
	Sediment nitrogen	<u>0.37*</u>	Sediment-pH	<u>-0.13</u>
l	Sediment carbon	0.24	Water-COD	<u>-0.016</u>
l	pH in the water	<u>-0.29</u>	Water-NO <sub>3</sub> =	<u>-0.24</u>
l	Sediment C/N	<u>-0.064</u>	$\underline{\text{Water-NH}_4}^{\pm}$	<u>-0.36</u> *
l	Conductivity	<u>-0.37</u>	Water-chla	<u>0.46*</u>
	Wind speed	<u>0.008</u>	Water-TN	<u>-0.35*</u>
l	DOC	<u>-0.015</u>	Water-TP	0.11

Note: Asterisks indicate statistically significant differences between CH<sub>4</sub> efflux and environmental

factors (one asterisk, p < 0.05; two asterisks, p < 0.01).

Table  $\frac{23}{2}$  Multivariate regressions between seasonal CH<sub>4</sub> efflux and environment factors

No.	Number of variables	Regression Equation	n	$\mathbb{R}^2$	p
Group 1	12	EffluxCH <sub>4</sub> = -10.48 + 110.57 ST + 65.06SNC	48	0.65	0.004
Group1 + Group 2	16	EffluxCH <sub>4</sub> = -12.66 + 0.57ST + 90.81SNC	43	0.73	0
Group 1 + Group 2 + Group 3	19	EffluxCH <sub>4</sub> = $-3.89 + 0.56$ ST + $102.88$ SNC $-35.56$ TP $-0.74$ DO	19	0.89	0

sediment total nitrogen content (SNC), water level, DOC content in the water, pH in the sediment, NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub> 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

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Note: Nd means that no variable input to the stepwise regression exists. Variables in group 1 included sediment temperature\_

e 	Lake size (km²)	Region	Climate	CH <sub>4</sub> efflux (mmol-m <sup>-2</sup> day <sup>-1</sup> )	References
<del>Hakes</del>	± <b>*</b>	Laurentians, Canada	Boreal-	4.08	Rasilo et al., 2015
5 lakes	47	Chicoutimi, Canada	Boreal	1.08-	Rasilo et al., 2015
7 lakes	41	Abitibi, Canada	Boreal -	1.67	Rasilo et al., 2015
Slakes	<del>171</del>	Chibougamau, Canada	Boreal -	0.17-	Rasilo et al., 2015
<del>)lakes</del>	7	James Bay, Canada	Boreal	1.08-	Rasilo et al., 2015
lakes	5	Côte Nord, Canada	Boreal	1.17	Rasilo et al., 2015
lakes	2	Eastmain, Canada	Boreal	0.58	Rasilo et al., 2015
lakes	242	Seheffervill, Canada	Boreal	0.42-	Rasilo et al., 2015
Mendota	39.4	North America	Boreal	0.50	Fallon et al., 1980
illon	13	North America	Boreal .	0.61	Smith and Lewis, 1992
<del>liolen</del>	#5	Sweden	Boreal	0.02	Bastviken et al., 2004
ev ät ön	4	Finland	Boreal	0.22	Huttunen et al., 2003
iwa	674	J <del>apan</del>	Temperate	0.27	Miyajima et al.,1997
<del>migaura</del>	168	Japan	Temperate	0.26-	<del>Utsuumi et al.,1998a</del>
<del>ojiri</del>	44	Japan	Temperate	0.06-	Utsuumi et al.,1998b
lakes	range 1–11, 3436 <sup>4</sup>	Netherlands	Temperate	5.85	Schrier Uijl et al., 2011
<del>Donghu</del>	<del>27.9</del>	China	Subtropical	1.46-	Xing et al., 2005
R-lake	71.4	Pantanal, South America	Tropical	0.65 <sup>8</sup> /5.74 <sup>C</sup>	Bastviken et al., 2010
B-lake	<del>36.3</del>	Pantanal, South America	Tropical	0.50 <sup>8</sup> /5.63 <sup>C</sup>	Bastviken et al., 2010
dantang	3-3	China	Subtropical	1.32	Xing et al., 2006
<del>ārtsjärv</del>	270	Estonia	Boreal	1.28 <sup>8</sup> /2.09 <sup>6</sup>	R ōcm et al., 2014
3 lakes	range1-10, 782073.8 <sup>4</sup>	worldwide	Mainly.	0.12	Holgerson and Raymond, 2016
			boreal		
lakes	range10-100, 597789.3*	worldwide	Mainly-	0.10	Holgerson and Raymond, 2016
			boreal		
lake	>100, 2024015.8 <sup>A</sup>	worldwide	Mainly	0.06-	Holgerson and Raymond, 2016
			boreal		

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1	<u>Lake</u>	Lake size (km²)	Region	Climate	CH <sub>2</sub> efflux (mmol m <sup>-2</sup> day <sup>-1</sup> )	References	Sampling period
1	11 lakes	1	Laurentians, Canada	Boreal	<u>4.08</u>	Rasilo et al., 2015	11/0 <sup>b</sup>
ı	Fiolen	1.5	Sweden	Boreal	0.02	Bastviken et al., 2004	Once
I	31 lakes	2	Eastmain, Canada	Boreal	0.58	Rasilo et al., 2015	14/17*D
I	Kev ät ön	4	Finland	Boreal	0.22	Huttunen et al., 2003	12 times
l	45 lakes	5	C ôte-Nord, Canada	Boreal	1.17	Rasilo et al., 2015	45/0 <sup>D</sup>
I	20 lakes	2	James Bay, Canada	Boreal	1.08	Rasilo et al., 2015	14/6 <sup>D</sup>
I	Dillon	13	North America	Boreal	0.61	Smith and Lewis,1992	9 times
I	Lake Mendota	<u>39.4</u>	North America	Boreal	0.5	Fallon et al.,1980	6 times
I	27 lakes	41	Abitibi, Canada	Boreal	<u>1.67</u>	Rasilo et al., 2015	21/6 <sup>D</sup>
I	26 lakes	<u>47</u>	Chicoutimi, Canada	Boreal	1.08	Rasilo et al., 2015	19/7 <sup>D</sup>
I	16 lakes	<u>171</u>	Chibougamau, Canada	Boreal	<u>0.17</u>	Rasilo et al., 2015	14/2 <sup>D</sup>
I	48 lakes	<u>242</u>	Scheffervill, Canada	Boreal	0.42	Rasilo et al., 2015	48/0 <sup>D</sup>
I	V õrtsjärv	<u>270</u>	<u>Estonia</u>	Boreal	1.28 <sup>B</sup> /2.09 <sup>C</sup>	R õõm et al., 2014	21 times
I	<u>6 lake</u>	>100, 2024015.8 <sup>A</sup>	worldwide.	Mainly boreal	0.06	Holgerson and Raymond, 2016	Multiple times
ı	18 lakes	range10-100, 597789.3 <sup>A</sup>	<u>worldwide</u>	Mainly boreal	0.1	Holgerson and Raymond, 2016	Multiple times
ı	43 lakes	range1-10, 782073.8 <sup>A</sup>	worldwide	Mainly boreal	0.12	Holgerson and Raymond, 2016	Multiple times
ı	<u>Nojiri</u>	<u>4.4</u>	<u>Japan</u>	<u>Temperate</u>	0.06	Utsuumi et al.,1998b	6 times
I	5 lakes	range 1-11, 3436 <sup>A</sup>	Netherlands	<u>Temperate</u>	<u>5.85</u>	Schrier-Uijl et al., 2011	twice
I	Kasumigaura	168	<u>Japan</u>	<u>Temperate</u>	0.26	Utsuumi et al.,1998a	72 times
I	Biwa	<u>674</u>	Japan	Temperate	0.27	Miyajima et al.,1997	3 times
I	Biandantang	<u>3.3</u>	China	Subtropical	<u>1.32</u>	Xing et al., 2006	12 times
I	Donghu	27.9	China	Subtropical	1.46	Xing et al., 2005	48 times
I	Poyang Lake	3283	<u>China</u>	Subtropical	0.54	Present study	48 times
I	BB lake	<u>36.3</u>	Pantanal, South America	Tropical	0.50 <sup>B</sup> /5.63 <sup>C</sup>	Bastviken et al.,2010	Once
ı	TR lake	71.4	Pantanal, South America	Tropical	0.65 <sup>B</sup> /5.74 <sup>C</sup>	Bastviken et al., 2010	Once

Note: A means total areas in the given lake size. B means diffusive effluxes and C means total effluxes, including diffusion and ebullition. D means number of lakes measured once/twice. \* means 24/0 in 2006, 8/11 in 2007, 0/13 in 2008, 2/10 in 2009, respectively.

### **Figure Captions**

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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<sub>4</sub> 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 ebullition: no ebullition (a), occurrence of ebullition at 20 min (b), 40 min (c), and 60 min (d), respectively.

Figure 3. Seasonal variations of CH<sub>4</sub> effluxes and sediment temperatures in Poyang Lake.

White circles represent the variation of CH<sub>4</sub> effluxes, and black circles describe the variation of sediment temperature in the 4-year period.

Figure 4. Diel variations of CH<sub>4</sub> 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. Diel variations of CH<sub>4</sub> effluxes among three sites.

Different panels present the diel variations of the CH<sub>4</sub> effluxes in 24–25 July 2011 (a), 5–6 September 2012 (b), 13–14 January 2013 (c), and 14–15 January 2015 (d).

Relationships between CH4-effluxes and wind speed in Poyang Lake.

White circles represent the observed values of CH<sub>4</sub> effluxes and wind speed. Different panels mean the variations of CH<sub>4</sub> effluxes at a bihourly interval within a day, including in 24-25 July 2011 (a), 5-6 September 2012 (b), 13-14 January 2013 (e), 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 e in the regression analysis because of a severe cold front.

Figure 6. Relationship between sediment temperature and CH<sub>4</sub> 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  $CH_4$  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.

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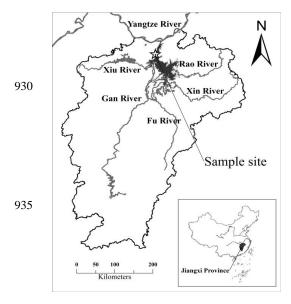
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Figure 7. Relationships between CH<sub>4</sub> effluxes and wind speed in Poyang Lake.

White circles represent the observed values of CH<sub>4</sub> effluxes and wind speed. Different panels mean the variations of CH<sub>4</sub> 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.

Fig. 1



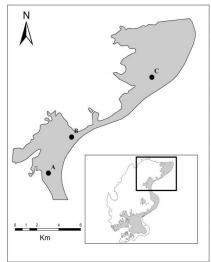
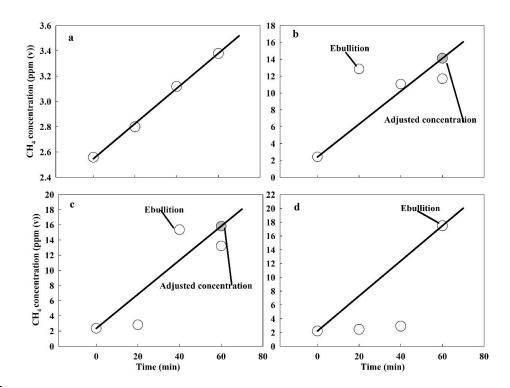
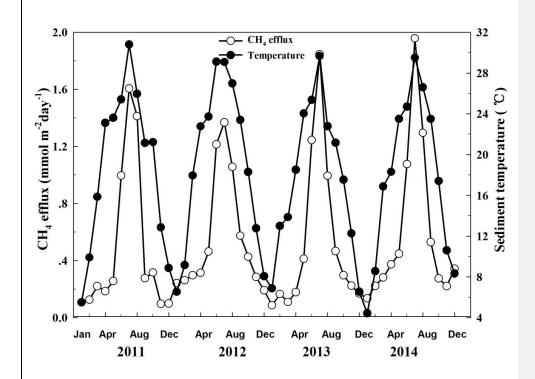
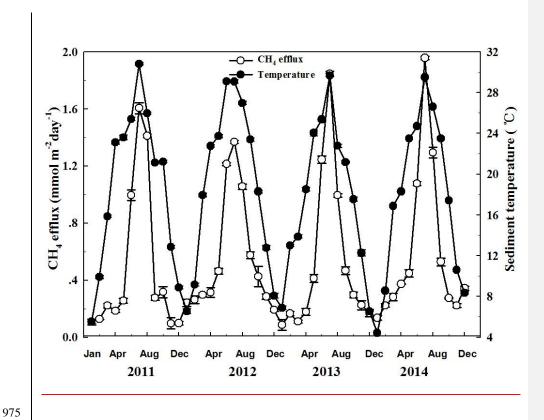


Fig. 2

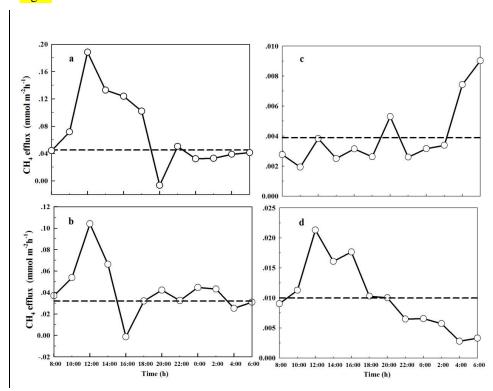


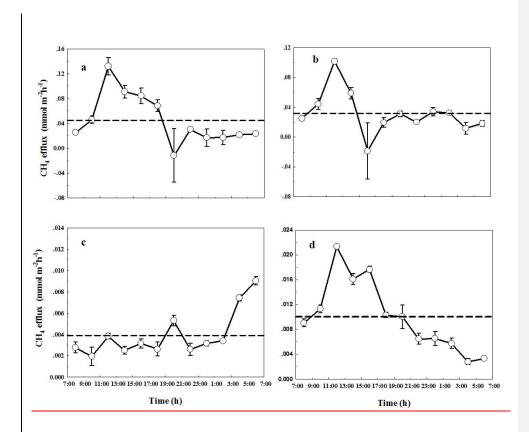
# Fig. 3

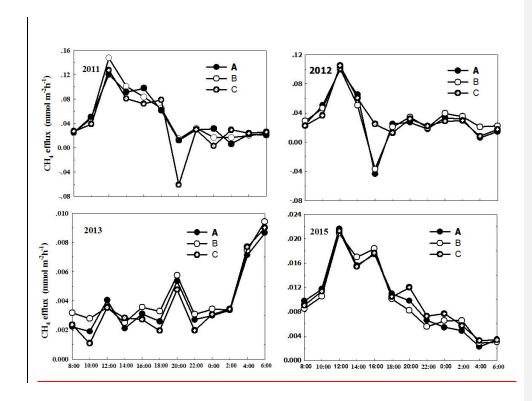




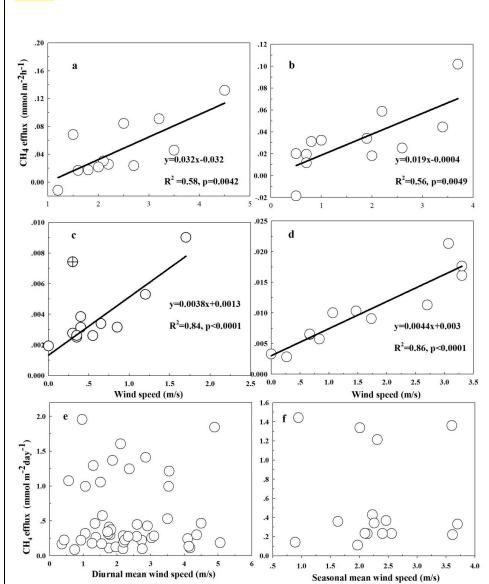


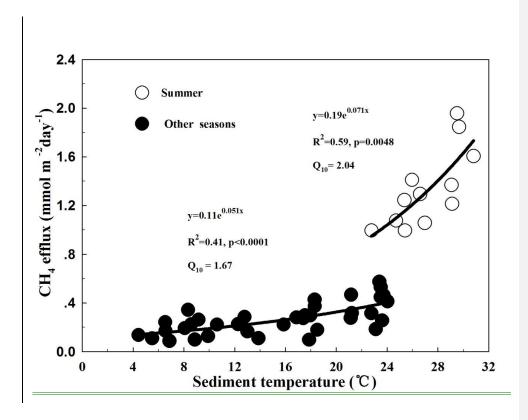












1025 Fig.<del>6</del>7

