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Spatial heterogeneity and lake morphology affect diffusive greenhouse gas emission estimates of lakes

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[1] Most estimates of diffusive flux (F) of methane (CH_4) and carbon dioxide (CO_2) from lakes are based on single-point flux chamber measurements or on piston velocity (k) modeled from wind speed and single-point measurements of surface water gas concentrations (C_{aq}). We analyzed spatial variability of F of CH_4 and CO_2, as well as C_{aq} and k in 22 European lakes during late summer. F and k were higher in the lake centers, leading to considerable bias when extrapolating single-point chamber measurements to whole-lake estimates. The ratio of our empirical k estimates to wind speed-modeled k was related to lake size and shape, suggesting a lake morphology effect on the relationship between wind speed and k. This indicates that the error inherent to established wind speed models can be reduced by determining k and C_{aq} at multiple sites on lakes to calibrate wind speed-modeled k to the local system. **Citation:** Schilder, J., D. Bastviken, M. van Hardenbroek, P. Kankaala, P. Rinta, T. Stötter, and O. Heiri (2013), Spatial heterogeneity and lake morphology affect diffusive greenhouse gas emission estimates of lakes, *Geophys. Res. Lett.*, *40*, 5752–5756, doi:10.1002/2013GL057669.

1. Introduction

[2] Large amounts of methane (CH_4) and carbon dioxide (CO_2), two important greenhouse gases, are released to the atmosphere from inland waters [*Bastviken et al.*, 2004; *Cole et al.*, 2007]. Estimates show that these fluxes correspond to a large share of the terrestrial carbon sink [*Bastviken et al.*, 2011; *Tranvik et al.*, 2009]. Therefore, accurate assessments of CO_2 and CH_4 fluxes from inland waters are vital for reliable estimates of the terrestrial greenhouse gas balance [*Battin et al.*, 2009]. From open water (i.e., the lake area free of emerging vegetation), lakes emit CO_2 and CH_4 in several ways, including ebullition (bubbling of nondissolved gases from the sediments) and by diffusive exchange (flux of dissolved gases across the air-water interface). Diffusive flux (F) dominates for highly soluble gases such as CO_2, and F may account for up to 50% of the total CH_4 flux [*Bastviken et al.*, 2004]. Most estimates of diffusive flux of CO_2 (F_{CO2}) and CH_4 (F_{CH4}) from aquatic ecosystems rely on measurements of surface water concentrations (C_{aq}) and the following equation:

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

where F is the diffusive flux (mmol m^{-2} day), k is the gas exchange coefficient or piston velocity ($m\,d^{-1}$ in equation (1); frequently and here expressed in $cm\,h^{-1}$), and C_{eq} is the theoretical surface water concentration (μM) when in equilibrium with air partial pressure (typically calculated from Henry's Law) [*Cole and Caraco*, 1998]. C_{aq} is generally measured at one single location in a lake, usually the center [e.g., *Bastviken et al.*, 2004; *Sobek et al.*, 2005; *Juutinen et al.*, 2009; *Marotta et al.*, 2009]. In turn, k is frequently estimated from wind speed at 10 m height (U_{10}) based on empirical relationships. Unfortunately, empirical relationships between k and U_{10} are only available from a few systems [*Bade*, 2009; *Wanninkhof et al.*, 2009; *Vachon et al.*, 2010], and it is unclear to what extent the general use of these models is valid. Gas accumulation measurements with floating chambers can also provide estimates of F in lakes [*Cole et al.*, 2010]. However, such estimates are again often based on measurements in a single location within the lake.

[3] Available equations for calculating k from U_{10} [e.g., *Crusius and Wanninkhof*, 2003; *Cole and Caraco*, 1998] lead to differing estimates of k at a given U_{10}. For many values of U_{10} in the range 0–10 $m\,s^{-1}$, one model returns a k value which is twice as high as the other. Further, a comparison of the model by *Cole and Caraco* [1998] with the underlying data set shows that for different sites, k at a given U_{10} can differ twofold from the model prediction. Hence, the error inherent to wind speed models, combined with variation caused by the choice of model, results in an uncertainty in whole-lake estimates of F that can be substantial.

[4] Only few studies provide information on how F_{CH4} and F_{CO2} vary within lakes [e.g., *Hofmann*, 2013], and we know of none which specifically addresses the within-lake variability of k. Such studies are needed for evaluating the reliability of F estimated from single-spot measurements. Furthermore, they might reveal factors responsible for the variability of k at a given U_{10} as documented by SF_6 tracer experiments [e.g., *Cole and Caraco*, 1998]. We measured within-lake spatial patterns in C_{aq} of CH_4 and of accumulation of CH_4 and CO_2 in floating chambers for 32 lakes in Europe. At each site, chambers were deployed at four locations across the surface of the lake. Based on these data, we estimated F_{CH4} for chambers which were not affected by ebullition, resulting in a data set of 12 lakes with reliable F_{CH4} estimates from four zones within the lake and a second data set of 22 lakes with at least one estimate in the zones nearest to shore and one in the more central zones. This in turn allowed an assessment of the

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variation in k within and between lakes. We estimated the bias in whole-lake F_{CH4} and F_{CO2} when scaling up from single-point estimates. Finally, we discuss mechanisms that can explain the within- and between-lake patterns of k and the discrepancies between k estimates from wind speed-based models and our observations.

2. Methods

2.1. Sample Collection and Gas Measurements

[5] In August and September 2010 and 2011 we measured CH_4 and CO_2 accumulation into floating chambers, C_{aq} of CH_4, and surface water temperatures at multiple locations on 32 small- to intermediate-sized lakes in Europe. At each lake, four groups of three replicate chambers were deployed for 6 h (~ 10:00–16:00) on a transect from the nearshore zone to the central zone of each lake (zones A, B, C, and D), with increasing distance between chamber zones (see Figure S2 in the supporting information). The chamber group in zone A was always close to the shore line, just beyond emerging macrophytes (if any). Chambers representing zone D were always in the center of the lake. The group representing zone C was placed approximately halfway between A and D and the group in zone B halfway between A and C. Chambers that had captured ebullition as well as diffusive emissions were identified using our replicate flux measurements in each zone and the method described by *Bastviken et al*. [2004] (see supporting information). Chambers identified as having received diffusive emissions only were used to provide estimates of F_{CH4}. F_{CH4}, C_{aq} of CH_4, and surface water temperatures were then used to calculate k according to equation (1).

[6] We adopted the floating chamber design and deployment methodology described by *Bastviken et al*. [2004] and *Cole et al*. [2010], which were shown to correspond well with multiple independent methods to determine k, implying negligible bias in chamber measurements [*Cole et al*., 2010; *Gålfalk et al*., 2013]. Samples for determining C_{aq} were collected at each group of chambers following *Bastviken et al*. [2010]: 40 mL of water was sampled 5 cm below the surface with 60 mL syringes (Becton-Dickinson) and exposed to 20 mL of ambient air in the syringes by shaking for 60 s. The headspace, now equilibrated with the water in terms of gases, was injected into a glass vial (10 mm thick butyl rubber stopper; Apodan) prefilled with saturated brine solution. These samples were stored in vials until analysis. Ambient air was collected to correct for the background air concentration in the water sample extractions and in the chamber measurements. After approximately 6 h, 30 mL gas was taken from each chamber with a syringe. These samples were again transferred to and stored in glass vials. Within 60 days of sampling, CH_4 and CO_2 concentrations were analyzed in the laboratory by gas chromatography using a flame ionization detector with a methanizer (GC-FID; Shimadzu GC-8, PoropackN column). Tests initiated in 2006 confirmed that samples collected and stored as described here are stable for years. Samples from the Finnish lakes were measured on a gas chromatograph with an autosampler (Agilent 6890 N, PlotQ capillary column, with FID for CH_4 and Thermal Conductivity Detector for CO_2).

2.2. Data Analysis and Upscaling

[7] For chambers not affected by ebullition, F_{CH4} can be determined from CH_4 accumulation in the chambers, accounting for chamber area, volume, and deployment duration and correcting for the decreasing concentration gradient due to increasing CH_4 concentration in the chambers [*Cole et al*., 2010]. The chamber deployment, sample handling, and storage were optimized for CH_4. Deployment was too long for providing estimates of F_{CO2} because CO_2 equilibrates much faster with the chamber headspace (typically within a few hours). Therefore, after 6 h concentrations of CO_2 in the chamber headspace will have reached values close to equilibrium with pCO_2 in the surface water. This also implies that final CO_2 concentrations in the chambers can be used to estimate C_{aq} of CO_2 following Henry's Law, which allowed us to estimate F_{CO2} (using k of CH_4 transformed to k of CO_2; *Bade* [2009]). We report k as k_{600}, the value corresponding to a gas with a Schmidt number of 600 (CO_2 at 20°C).

[8] For analysis of within-lake patterns of k, F, and C_{aq}, and for scaling up, we partitioned the original data set into different subsets (Tables S1 and S2): A data set of 13 lakes that consists of lakes that yielded at least one estimate of k for each sampling zone (subset 1). This subset was used to identify spatial patterns of k, F_{CH4}, and F_{CO2} in the study lakes. The second subset consists of 24 lakes with at least one estimate of k each in the central part of the lake (zones C + D) and the nearshore area (zones A + B) (subset 2). For these lakes, surface areas were digitized from national topographic maps. Using ArcMap 9.3 (Esri), the area belonging to the nearshore and the central zones was quantified. Nearshore area was defined as lying within a distance to shore corresponding to the mean distance of chamber groups B and C to shoreline. The central area consisted of the remaining surface. Estimates of k and F for nearshore and central areas from subset 2 were then used to scale up to whole-lake averages weighted by area. U_{10} was provided by the respective national meteorological services (see supporting information).

[9] One lake that fulfilled the selection criteria for subset 1 and two lakes that fulfilled the criteria for subset 2 were not included in further analysis. At Valkea-Kotinen, CH_4 accumulation rates were so low they resulted in k values lower than commonly found in literature. Therefore, this lake was eliminated from both subsets. The wind speed data for lake Kisasjön (3.9 m s^{-1}) deviated strongly from our observations in the field (the lake was sampled on the windiest day of the campaign). Therefore, we concluded that the wind speed data did not reflect the local conditions, and this lake was eliminated from subset 2. Measurements from these lakes can be found in the supporting information (Tables S1 and S2) with those of the other lakes not incorporated in either subset (Table S3).

3. Spatial Patterns and Whole-Lake Estimates of k

[10] Values of k_{600} were clearly higher in the central zone of the lake in 11 of the 12 lakes remaining in subset 1 (Figure 1). C_{aq} of CH_4 was usually lower in the center, in agreement with recent findings by *Hofmann* [2013]. F_{CH4} showed the opposite pattern, however, with highest values in the central zones, which is in contrast with the conclusions by *Hofmann* [2013] that are based on wind speed-derived k values. C_{aq} of CO_2 and F_{CO2} were lowest nearshore and more elevated in the central zones (Figure 1).

[11] The range of k_{600} was similar in the two nearshore sampling zones (A + B), with values typically below the lake average (Figure 1). Estimates from the central zones (C + D)

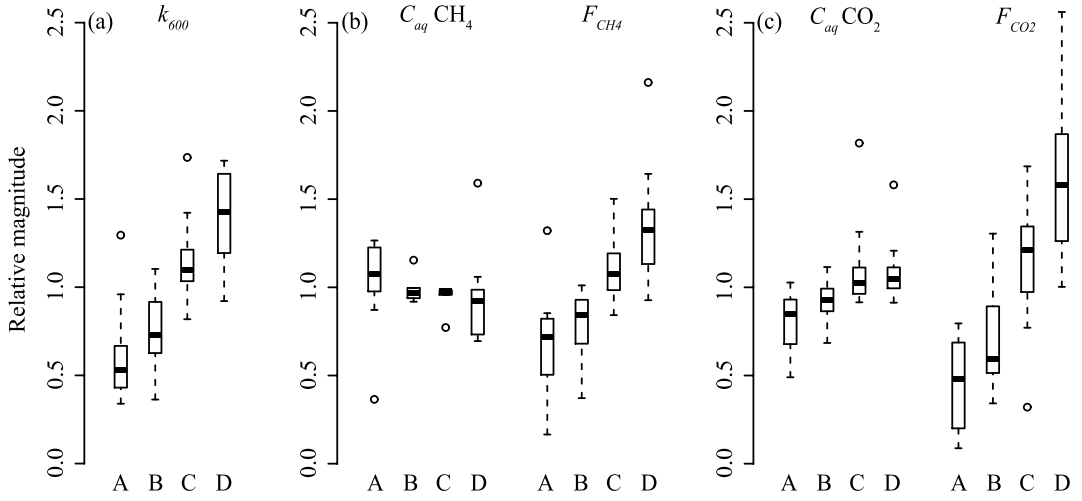

Figure 1. Summary of spatial patterns in (a) k_{600}, (b) C_{aq} of CH_4 and F_{CH4}, and (c) C_{aq} of CO_2 and F_{CO2} in the 12 lakes that yielded estimates of F for all four zones. Values are grouped per zone (A, nearshore, to D, central). Estimates were divided by the average of the respective variable in each lake, removing the lake-dependent variability in the data set. Whiskers of the boxplots encompass data points no more than 1.5 times the interquartile range from the box; circles indicate more extreme points.

were usually higher than average, again with a large overlap in range. Grouping chamber measurements into two groups instead of four allowed spatially resolved estimates of k_{600} and F to be calculated for 22 of our 32 study lakes. Whole-lake estimates of k_{600} based on this 22-lake data set are in the range of 1.2–6.3 cm h^{-1}. The relationship between U_{10} and our estimates of k_{600} is in agreement with the model by *Cole and Caraco* [1998] (Figure 2). Residuals of k_{600} to modeled values for our data set (range −1.9 to 1.7 cm h^{-1}, standard deviation (SD) 1.1 cm h^{-1}) are similar to the residuals recalculated for the SF_6 tracer studies that were originally used to develop this model (range −2.8 to 1.7 cm h^{-1}, SD 1.3 cm h^{-1}) [*Cole and Caraco*, 1998, Figure 8a]. We therefore conclude that the chamber method we used for assessing k provides comparable results to tracer studies.

4. Implications for Whole-Lake Estimates of *F*

[12] We compared F derived from single-point chamber measurements to whole-lake estimates based on the spatially resolved 22-lake data set (the values are provided in Table S2). Values based on single locations ranged from 38 to 222% of whole-lake F_{CH4}. On average, inferences from nearshore measurements slightly underestimated F_{CH4} (88 %), whereas those from the center provided overestimates (115%). Measuring C_{aq} in the lake center and then applying the model by *Cole and Caraco* [1998] yields F_{CH4} amounting to 55–300% of our whole-lake estimates (average 110%) and 33–320% for F_{CO2} (average 148%). Hence, this common way of estimating fluxes results in F_{CH4} and F_{CO2} being overestimated on average by 10% and 48%, respectively, in our data set. For individual lakes, the bias is highly variable, reflecting the large variability in the data behind the k versus U_{10} relationship.

5. Relationship of *k* With Lake Morphometry

[13] The relationship between whole-lake k_{600} and U_{10} is strong and well studied [e.g., *Cole and Caraco*, 1998; *Crusius and Wanninkhof*, 2003; *Wanninkhof et al*., 2009; *Vachon et al*., 2010]. Available wind speed-based models provide realistic estimates of the average whole-lake k_{600} expected at a given wind speed. However, estimates for individual lakes can still deviate considerably from this value (Figure 2) [*Cole and Caraco*, 1998]. We used the ratio between k_{600} modeled following *Cole and Caraco* [1998] (kCC) and k_{600} inferred from our chamber measurements (kS) to explore where the two methods lead to the most pronounced differences in k_{600}. This ratio (kCC/kS) was highest for lakes in which k_{600} was higher in the center than nearshore (Figure 3a), suggesting that the spatial gradient in k_{600} may be a source of bias in wind speed-based estimates of k_{600}.

[14] We consider the most likely explanation for the observed within-lake variability of k to be the proximity to shoreline and sheltering shoreline structures and vegetation. This potentially reduces the direct impact of wind at the lake

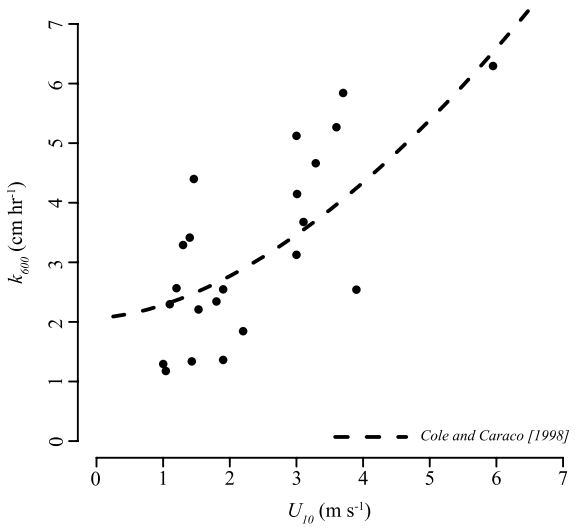

Figure 2. Whole-lake estimates of k_{600} plotted against U_{10} in the 22-lake data set. The dashed line indicates the relationship between k_{600} and U_{10} described by *Cole and Caraco* [1998].

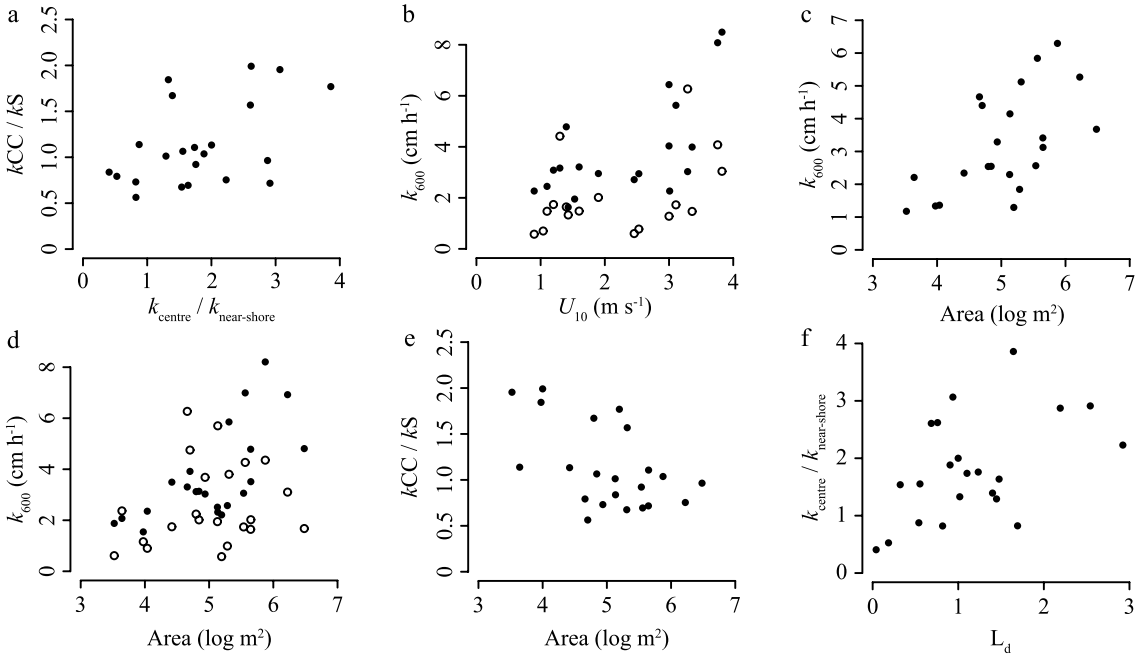

Figure 3. (a) Ratio of k_{600} inferred from *Cole and Caraco* [1998] and our whole-lake k_{600} estimates (kCC/kS) plotted versus the strength of spatial variability in k_{600} ($k_{central}/k_{nearshore}$) ($r = 0.48$, $p < 0.05$). (b) The relationship between U_{10} and k_{600} for nearshore chamber group A (open circles; $r = 0.41$, $p = 0.1$) and central chamber group D (solid circles; $r = 0.67$, $p < 0.005$). (c) Whole-lake k_{600} plotted against area ($r = 0.59$, $p < 0.005$). (d) Relationship between area and k_{600} determined from the nearshore chamber groups A and B (open circles; $r = 0.19$, $p = 0.39$) and from the central chamber groups C and D (solid circles; $r = 0.68$, $p < 0.001$). (e) The kCC/kS plotted versus area ($r = -0.53$, $p < 0.05$). (f) Lake shape complexity, here presented as shore development (L_d), and its relationship to $k_{central}/k_{nearshore}$ ($r = 0.49$, $p < 0.05$). L_d is defined as the circumference of a lake divided by the circumference of a circle with the same area [*Håkanson*, 2004].

surface and can explain low k_{600} and F recorded in nearshore zones (Figure 1). If this hypothesis is true, we would expect this sheltering to lead to a less pronounced relationship of U_{10} with k_{600} in nearshore areas than in the lake center, which is confirmed by our data (Figure 3b). Also, the proportion of the lake influenced by sheltering effects of the shoreline is expected to be larger on small lakes. Therefore, we also expect higher k_{600} in larger lakes. In our data, this is indeed the case (Figure 3c). Unfortunately, lake size is also correlated with U_{10} in our data ($r = 0.49$, $p < 0.05$), so we cannot separate the relationship between k_{600} and area from its relationship with U_{10}. However, the relationship of k_{600} with lake area (Figure 3d) is absent or weak in nearshore areas ($r = 0.19$, $p = 0.39$), whereas it is strong in central zones ($r = 0.68$, $p < 0.001$). This again supports our interpretation that under similar wind conditions, nearshore areas are more protected.

[15] If distance to shoreline influences k_{600}, both lake size and shape have the potential to affect the relationship of k_{600} with U_{10}. However, both of these variables are presently rarely taken into account when producing wind speed-based estimates of k_{600}. That lake area can influence modeled k_{600} is supported by its significant relationship with kCC/kS in our data set (Figure 3e). This indicates that k_{600} modeled following *Cole and Caraco* [1998] tends to provide overestimated values for small lakes. Similarly, a first analysis of our data in respect to the potential effects of lake shape reveals that more complex lake basins, which in our campaign were typically elongated in shape, were characterized by higher within-lake variability in k_{600} (Figure 3f). Lakes with high variability in k_{600} were also the ones in which the approach of *Cole and Caraco* [1998] returns higher values of k_{600} than we observe (Figure 3a). Hence, lake shape also appears to influence whole-lake k_{600}.

[16] Our interpretations suggest that the effect of U_{10} on k_{600} should be greater on larger lakes and lakes with simple shapes, in which sheltering effects of shoreline are limited. Large lakes provide the potential for a larger fetch, and lakes with more complex shorelines have shorter average distance to sheltering shoreline vegetation. We therefore expect the influence of U_{10} on k_{600} to be reduced in small, complex, and very elongated lakes. This also implies that wind speed-based models may have to be adapted for these kinds of systems.

[17] *Wanninkhof* [1992] suggested lake size as an explanation for the discrepancy between existing SF_6 tracer studies, and *Guérin et al*. [2007] identified a relationship between basin size and k for rivers and estuaries. *Read et al*. [2012] also stressed the importance of lake size for the convection component of k. Our data were collected at daytime, which means that the convection driven by heat loss during nights is less likely to have contributed to the observed spatial patterns. Our data support *Wanninkhof*'s [1992] suggestion and imply that wind speed models are only applicable to lakes with a size and shape similar to the lakes the model was developed on.

6. Conclusions

[18] Our study shows distinct and profound spatial patterns in F of greenhouse gases from lakes as well as in the driving parameters, k and C_{aq}. It also reveals lake size and shape as

plausible explanations for the high prediction uncertainty observed in wind speed models. We found that for individual lakes, models commonly used to determine whole-lake F yield results that are between 55 and 300% (CH_4) or 33 and 320% (CO_2) of our in situ F measurements. Given the importance of lakes in the terrestrial greenhouse gas balance, this error range is unsatisfying.

[19] Whole-lake estimates of F, depending on in situ measurements of F, k, or C_{aq}, should preferably be based on spatial transects. There is presently no model that takes wind, lake size, lake shape, presence and height of vegetation, and convection into account. However, k values for individual lakes can be determined in situ, and the results can be used to calibrate established wind speed models to local conditions. Our data show that this can be done with floating chambers if properly designed [*Cole et al*., 2010] and with appropriate consideration of confounding factors like ebullition. The method is inexpensive, swift, and uncomplicated. CH_4 appears to be more suitable than CO_2 for determining k using chambers because CH_4 is nearly always supersaturated and less affected by chemical processes or variables with diurnal variations such as primary production, respiration, and pH [*Bastviken*, 2009]. Furthermore, the equilibration of CH_4 between the water and chamber headspace is often slow enough to allow chamber deployments over 24 h, yielding a daily average k accounting for different wind- and water-mixing conditions during day and night [*Bastviken et al*., 2010].

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