

Geographic and seasonal variation of dissolved CH₄ and aerobic CH₄ oxidation

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Geographic and seasonal variation of dissolved methane and aerobic methane oxidation in Alaskan lakes

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Abstract

Methanotrophic bacteria play an important role oxidizing a significant fraction of methane (CH_4) produced in lakes. Aerobic CH_4 oxidation depends on lake CH_4 and oxygen (O_2) concentrations, temperature, and organic carbon input to lakes, including from thawing permafrost in thermokarst (thaw)-affected lakes. Given the large variability in these environmental factors, CH_4 oxidation is expected to be subject to large seasonal and geographic variations, which have been scarcely reported in the literature. In the present study, we measured CH_4 oxidation rates in 30 Alaskan lakes along a north–south latitudinal transect during winter and summer with a new field laser spectroscopy method. Additionally, we measured dissolved CH_4 and O_2 concentrations. We found that in the winter, aerobic CH_4 oxidation was mainly controlled by the dissolved O_2 concentration, while in the summer it was controlled primarily by the CH_4 concentration, which was in deficit compared to dissolved O_2 . The permafrost environment of the lakes was identified as another key factor. Thermokarst (thaw) lakes formed in yedoma-type permafrost had significantly higher CH_4 oxidation rates compared to other thermokarst and non-thermokarst lakes formed in non-yedoma permafrost environments. These results confirm that landscape processes play an important role in lake CH_4 cycling.

1 Introduction

Freshwater ecosystems are an important source of atmospheric CH_4 , responsible for 6–16 % of global emission to the atmosphere (Bastviken et al., 2011). Northern lakes are responsible for as much as 6 % of these global CH_4 emissions (Walter et al., 2007). Methane emission from aquatic ecosystems is significantly mitigated by CH_4 oxidation (MO) by aerobic methanotrophs, a group of gram-negative bacteria that use CH_4 as a carbon and energy source (Murrell et al., 1993; Trotsenko and Murrell, 2008). It has been estimated that globally, 30–99 % of total CH_4 produced in freshwater

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ecosystems is microbiologically oxidized in the water column rather than being released to the atmosphere (Bastviken et al., 2002; Thauer et al., 2008). Likewise, MO plays an important role in northern lakes specifically by oxidizing up to 88 % of the CH₄ production (Kankaala et al., 2006, 2007; Bastviken et al., 2008; Bellido et al., 2011). MO is therefore a pathway that reincorporates a significant fraction of the CH₄-C produced into the biogeochemical carbon cycle within lakes. As recently demonstrated using stable isotopes, after assimilating CH₄, methanotrophs are incorporated into the lake food web by zooplankton (Kankaala et al., 2006; Jones and Grey, 2011), *Daphnia magna* (Taipale et al., 2012), *Odonata* spp. (Seifert and Scheu, 2012), and *Chironomus larvae* (Gentzel et al., 2012; Wooller et al., 2012), among others.

Several environmental factors directly affect aerobic MO in freshwater ecosystems. First, methanotrophy depends on the availability of both CH₄ and O₂. Higher MO rates are usually found at the oxic/anoxic interface, where both CH₄ and O₂ are present (Utsumi et al., 1998a, b; Bastviken et al., 2002; Liikanen et al., 2002; Carini et al., 2005; Schubert et al., 2010). In turn, CH₄ and O₂ concentrations depend on numerous other processes involved in biogeochemical carbon cycling (Fig. 1). Among these, the most important are methanogenesis producing CH₄, primary production and atmospheric diffusion supplying O₂, and several aerobic metabolic processes that compete with MO for available O₂ (Dzyuban, 2010).

In addition to autochthonous and allochthonous carbon inputs to lakes, permafrost thaw can provide an additional source of labile organic carbon to fuel methanogenesis and carbon mineralization in thermokarst (thaw) lakes (Zimov et al., 1997; Walter et al., 2006). MO in northern regions is therefore directly and indirectly linked to permafrost type and landscape processes that are highly variable. Permafrost ranges from sporadic to continuous and is also variable in composition (Jorgenson et al., 2008). For instance, yedoma-type permafrost (organic-rich, ice-supersaturated, Pleistocene-aged loess deposits) is unevenly distributed in Siberia and Alaska (Czudek and Demek, 1970; Walter et al., 2007; Kanevskiy et al., 2011; Grosse et al., 2013). Non-yedoma permafrost soils are more common, but the organic-rich horizons of

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non-yedoma permafrost and permafrost-free soils are typically thinner than those of yedoma deposits (Ping et al., 2008; Tarnocai et al., 2009; Hugelius et al., 2014).

Many northern lakes are located in continental climate zones, subject to contrasting seasonal conditions with long, cold winters followed by relatively short, but warm summers. Although psychrotolerant and psychrophilic methanotrophs have been reported (Omelchenko et al., 1993, 1996; Bowman et al., 1997; Trotsenko and Khmelenina, 2002), MO occurs more efficiently at mesophilic temperatures, from 20 to 35 °C (Semrau et al., 2010). During winter, northern lakes are covered by a thick ice layer for seven to nine months. Surface lake ice impedes oxygen transfer from the atmosphere to the lake and, when snow-covered, substantially reduces light penetration and oxygen production by photosynthesis (White et al., 2008; Clilverd et al., 2009). Thus, the combination of low temperature and limited oxygen availability suggests lower MO rates in northern lakes in winter than in summer.

Given the number of parameters having a potential effect on MO, as well as the patchwork of seasonal and geographic conditions found among northern lakes, MO is expected to exhibit large geographic and seasonal variations that still remain to be characterized. The goal of our study was to determine these variations through measurement of dissolved CH₄ and O₂ as well as MO rates in the winter and summer in 30 lakes along a south–north transect in Alaska.

2 Materials and methods

2.1 Site description

We sampled 30 Alaskan lakes during two field campaigns, one in late winter (March–April 2011) and one in summer (June–July 2011). To evaluate the effects of latitudinal variation and permafrost type on MO, lakes were selected along a transect from the southcentral Alaskan coast on the Kenai Peninsula to the Arctic Ocean near Prudhoe Bay (Fig. 2). The transect crossed through glaciated mountain ranges and

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discontinuous, sporadic, or no permafrost in south-central Alaska; discontinuous to isolated yedoma permafrost in the interior of Alaska; and continuous permafrost in northern Alaska. In this work, for simplicity, lakes located in yedoma-type permafrost areas will be referred to as “yedoma lakes” and all others as “non-yedoma lakes”.

5 Geographic variability along the north–south Alaska transect has been previously described for ecosystems, climate, geology, and permafrost type (Gregory-Eaves et al., 2000; Jorgenson et al., 2008; Smith et al., 2010). Additionally, Sepulveda-Jauregui et al. (2014) quantified the surface area of the selected lakes (0.002–1.45 km²), their trophic states (ultraoligotrophic to eutrophic), and their annual CH₄ fluxes (0.5–
10 317 g CH₄ m⁻² yr⁻¹). Table 1 shows the location and permafrost type of the selected lakes.

2.2 Sampling and field measurements

We sampled lake water offshore and usually near the center of each lake. In the winter, the ice cover was drilled through with a motorized auger (0.3 m in diameter).
15 Using a Hydrolab DataSonde (Hach Hydromet, Loveland, CO, USA), we measured temperature, pH, chlorophyll *a*, and dissolved oxygen (DO). The Hydrolab was calibrated regularly, before and after each section of the latitudinal lake transect (four sections per transect, approximately one calibration per week). All parameters were measured at 0.5 or 1 m depth intervals throughout the water column, except in Dolly
20 Varden L. where measurement intervals were increased to every 5 m from 15 to 25 m depth. In lakes shallower than 1 m, we measured Hydrolab parameters at three distributed depths throughout the lake water column.

Water samples for MO rates and dissolved CH₄ concentration were taken at a depth of within 1 m of the ice–water interface in winter and usually at 0.75–1 m water depth in
25 summer. Due to differences in lake depths and thicknesses of the ice sheets, in some lakes, lake-bottom water was sampled, while in other lakes near-surface or mid water column water was sampled. Samples were taken with a horizontal Van Dorn bottle (Wildco, Yulee, FL, USA).

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Water density derived from surface and bottom water temperatures were used to determine the relative water column stability (RWCS; Padisak et al., 2003). Lakes with RWCS > 56.5 were considered fully stratified, lakes with RWCS < 16.3 were considered fully mixed, and lakes with intermediate RWCS were considered partially stratified (Branco et al., 2009). Similarly, we determined whether an oxycline was present in each lake based on a sharp DO gradient or presence of an oxic/anoxic interface. Detailed temperature and DO profiles are available as a Supplement in Sepulveda-Jauregui et al. (2014). We report all results in mean \pm standard deviation (SD).

2.3 Dissolved CH₄ concentration and MO rate

To avoid long delays in sample transfer from remote locations to the laboratory, we used a previously described method for determination of dissolved CH₄ concentrations (HE-TDLAS; Sepulveda-Jauregui et al., 2012). This method consisted of determining the CH₄ concentration in the headspace of an equilibration vial containing a known volume of lake water and in which gas/liquid equilibrium has been reached by 10 s of vigorous shaking. The CH₄ concentration in the headspace was determined using a laser beam crossing the headspace of the equilibration vial. This measurement was conducted with a modified open-field CH₄ analyzer (GasFinder 2; Boreal Laser, Edmonton, Canada). The CH₄ concentration in the water sample was calculated from the measured headspace concentration according to Henry's law (see Sepulveda-Jauregui et al., 2012 for details).

We determined duplicate MO rates in one water sample from each lake as described above, using a modified HE-TDLAS method to allow for measurement of MO in the field. This new method was based on a previous development using the HE-TDLAS method for the determination of methanogenic activity (Martinez-Cruz et al., 2012). Two 60 mL lake water subsamples from a single Van Dorn bottle sample were gently transferred to two 100 mL equilibration vials (duplicates). Equilibration vials were immediately closed with rubber stoppers and vigorously shaken for 10 s to transfer

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most of the dissolved CH₄ contained in the water sample to the headspace. Next, the headspace was vented, the vial was closed, and the sample was shaken again to evacuate the residual CH₄ content of the water sample. Using this procedure, more than 99.5% of the original CH₄ content of the sample was evacuated. The equilibration vials were then closed with rubber stoppers and aluminum crimp caps, spiked with 0.6 mL CH₄ (99.0% purity; Air Liquide, Houston, TX, USA) injected with a disposable syringe, and vigorously shaken for 10 s. This approach allowed MO tests to be conducted with an initial standard CH₄ concentration in the liquid phase ($\sim 0.6 \text{ mg L}^{-1}$). It also provided an initial CH₄ to O₂ molar ratio of 0.062, significantly below the stoichiometric ratio (0.5), ensuring no O₂ limitation.

Equilibration vials were incubated for 10–12 days in a water bath inside insulated boxes placed in our vehicle. In the winter, the vials were maintained at $2 \pm 2^\circ\text{C}$ in a water bath with ice supplements; in the summer, the vials were maintained at $15 \pm 2^\circ\text{C}$. The temperature of the water bath was measured daily. We measured the CH₄ concentration in the equilibration vials daily using the HE-TDLAS method described in detail by Sepulveda-Jauregui et al. (2012). Briefly, dry control MO test vials containing only CH₄ standards were read by the TDLAS for calibration. Each experimental equilibration vial was vigorously shaken for 10 s to reach phase equilibrium and then immediately placed in the laser beam path, after which a stable HE-TDLAS reading was typically observed within 5 s. Five readings were taken for each MO test vial and recalibration was conducted after measuring each set of test vials to ensure instrument stability. The field HE-TDLAS method allowed measurement of dissolved CH₄ and MO rates. This technique was simple, rapid (about 60 s per sample measurement), non-invasive, and avoided complications and long delays in sample transfer from remote locations to the laboratory.

We calculated the total CH₄ concentration (C_{CH_4} = total CH₄ mass present in the gas and liquid phases divided by the sample liquid volume) in each vial during the MO tests. MO rates were determined from the decrease in C_{CH_4} in the equilibration vials with time. MO rates determined by this method represent the MO rate after

aeration and CH₄ addition (vials spiked with CH₄ and vigorously shaken). Thus, these MO rates do not correspond to actual observations of in situ DO and dissolved CH₄ concentrations in the lakes. The measured CH₄ oxidation rate was considered the potential MO (r_{\max} ; mg CH₄ L⁻¹ d⁻¹) under non-limiting CH₄ and DO concentrations.

To estimate the actual rate (r ; mg CH₄ L⁻¹ d⁻¹) from r_{\max} , a double Monod model was used (Bae and Rittmann, 1996; Segers, 1998) in which C_{CH_4} and C_{O_2} represent the actual dissolved CH₄ and DO concentrations measured in the lake, respectively, and $K_{\text{S-CH}_4}$ and $K_{\text{S-O}_2}$ are the apparent affinity constants of the methanotrophic community, for CH₄ and DO, respectively:

$$r = r_{\max} \cdot \frac{C_{\text{CH}_4}}{K_{\text{S-CH}_4} + C_{\text{CH}_4}} \cdot \frac{C_{\text{O}_2}}{K_{\text{S-O}_2} + C_{\text{O}_2}} \quad (1)$$

Average $K_{\text{S-CH}_4}$ and $K_{\text{S-O}_2}$ values for lakes have been determined by previous studies: $K_{\text{S-CH}_4} = 0.110 \pm 0.053 \text{ mg L}^{-1}$ (mean \pm SD; Liikanen et al., 2002; Lofton et al., 2013) and $K_{\text{S-O}_2} = 0.624 \pm 0.064 \text{ mg L}^{-1}$ (mean \pm SD; Lidstrom and Somers, 1984; Frenzel et al., 1990). To the best of our knowledge, the highest $K_{\text{S-CH}_4}$ reported in lakes is 0.704 mg L^{-1} (Liikanen et al., 2002). It should be noted that these reported K_{S} values refer to the apparent affinity constants for the methanotrophic community, rather than the half-saturation constant for the CH₄ monooxygenase enzyme that catalyzes CH₄ oxidation. The potential error caused by using previously reported K_{S} , instead of experimentally determined values will be considered in the discussion section.

To establish the extent of potential MO limitation by CH₄ or DO, two limitation factors were defined, where β is the limitation factor for CH₄ (%) and γ is the limitation factor

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for DO (%):

$$0\% \leq \beta = \left(1 - \frac{C_{\text{CH}_4}}{K_{\text{S-CH}_4} + C_{\text{CH}_4}} \right) \cdot 100 \leq 100\% \quad (2)$$

$$0\% \leq \gamma = \left(1 - \frac{C_{\text{O}_2}}{K_{\text{S-O}_2} + C_{\text{O}_2}} \right) \cdot 100 \leq 100\% \quad (3)$$

A limitation factor of 100 % means that 100 % of a process ceases to occur due to the absence of the limiting substrate, while a limitation factor of 0 % indicates a process occurring at maximum rate ($r = r_{\text{max}}$). When $\beta > \gamma$, CH_4 was considered to be the limiting factor; conversely, when $\gamma > \beta$, DO was considered to be the limiting factor.

2.4 Statistical analyses

Significant differences between all parameters except r and r_{max} were determined using the Tukey–Kramer (TK) multiple comparison test performed after analyses of variance (ANOVA; $p < 0.05$). Because of the non-normal distributions of r and r_{max} , significant differences were determined using the Kruskal–Wallis (KW) test for non-parametric data ($p < 0.05$). Statistical analyses were conducted using the NCSS 2000 Statistical Analysis System software (Number Cruncher Statistical Systems, Kaysville, UT, USA). Linear regressions were also conducted to determinate MO rates using Wolfram Mathematica 7.0 (Wolfram, Minneapolis, MN, USA).

3 Results

3.1 Physicochemical parameters

The sampled lakes were shallow; other than four atypical lakes with a maximum known depth > 20 m (lakes #4, #24, #26 and #30), the average lake depth in summer was

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4.5 ± 2.6 m (mean ± SD). During winter, none of the lakes was completely frozen at the sampling stations. Liquid water was always present underneath the ice cover, which ranged in thickness from 0.60 to 1.25 m (mean ± SD, 0.81 ± 0.14 m). The mean temperature throughout the lake water columns was 2.4 ± 0.6 °C (mean ± SD, $n = 103$) in the winter and 13.9 ± 2.4 °C (mean ± SD, $n = 235$) in the summer. According to RWCS, during the summer, 15 lakes of the 28 for which a complete temperature profile was determined were fully thermally stratified. Six lakes were partially stratified and seven lakes were mixed. During the winter, 16 of 18 lakes were fully mixed, while two lakes were partially stratified and none was fully stratified. Overall, only one third of the temperature profiles indicated clear stratification.

Lake water pH ranged from 5.9 to 8.2 in winter and from 6.3 to 9.2 in summer among the study lakes. Chlorophyll *a* was only detected during the summer, ranging from 1.0 to 45.9 µg L⁻¹ (detection limit, 0.03 µg L⁻¹). The concentration of dissolved CH₄ in the 30 lakes ranged from 0.01 to 14.77 mg L⁻¹ during the winter and from 0.02 to 1.51 mg L⁻¹ during the summer (Table 2). The DO concentration at the same depths ranged from 0.10 to 13.63 mg L⁻¹ during the winter and from 0.22 to 11.07 mg L⁻¹ during the summer (Table 2). During summer, a clear oxycline was observed in all yedoma lakes, but only in six of 20 non-yedoma lakes. In contrast, during winter, an oxycline was not observed in any of the yedoma lakes, which were largely anaerobic throughout the whole water column. We observed an oxycline in winter in four of 13 non-yedoma lakes. Overall, an oxycline was observed in 30% of the DO profiles. Temperature-oxygen profiles for all 30 studied lakes are shown in Sepulveda-Jauregui et al. (2014).

Figure 3 shows the statistical distributions of the dissolved CH₄ and DO concentrations, as well as the TK comparisons. Significant differences were observed between yedoma and non-yedoma lakes ($p < 0.05$). In yedoma lakes, the CH₄ and DO concentrations were significantly higher and lower, respectively, than in non-yedoma lakes during both seasons (TK test, $p < 0.05$). In addition to differences related to permafrost type, higher CH₄ concentrations and lower DO concentrations were

observed during the winter than in the summer (Fig. 3) and an apparent geographic trend was observed. Higher dissolved CH_4 and lower DO concentrations were found in lakes from central Alaska than in those from southern and northern Alaska (Sepulveda-Jauregui et al., 2014).

3.2 Methane oxidation rates

The HE-TDLAS method allowed us to determine the potential MO potential in the field in all studied lakes. Figure 4 shows three representative C_{CH_4} trends observed in the MO vials. In some cases, MO began on the first day of incubation (Fig. 4a) and the initial slope of the change in C_{CH_4} was taken into account in determining the MO rate. In about 60% of the cases during the summer and 80% during the winter, a lag phase was observed (Fig. 4b) before C_{CH_4} began to decrease. This behavior, termed “induction of MO”, has previously been reported for various soils (Bender and Conrad, 1995; Dunfield et al., 1999). In lakes in which this pattern was observed, the lag phase was not taken into account and the MO rate was instead determined from the slope of C_{CH_4} after the lag phase. When no significant decrease in C_{CH_4} was observed during the first seven days (Fig. 4c; TK test, $p < 0.05$), we assumed an MO rate of zero, consistent with previous reports for various soils (Whalen et al., 1990; Bender and Conrad, 1995; Dunfield et al., 1999). We observed MO rates of zero in only three non-yedoma lakes during winter. Otherwise, no correlation with lake morphology, season, or permafrost type was observed in regard to the existence of a lag phase or its duration.

The potential MO rate r_{max} ranged from 0.000 to $0.488 \text{ mg L}^{-1} \text{ d}^{-1}$ during the winter and from 0.073 to $1.339 \text{ mg L}^{-1} \text{ d}^{-1}$ during the summer (Fig. 5a). Seasonal variation of r_{max} was significant, with summer r_{max} up to 47 times higher than winter rates. Permafrost type was also an important determining factor, because during the summer, yedoma lakes had higher r_{max} than non-yedoma lakes (KW test, $p < 0.05$); specifically, r_{max} was 0.71 ± 0.36 and $0.29 \pm 0.16 \text{ mg L}^{-1} \text{ d}^{-1}$ (mean \pm SD) for yedoma and non-yedoma lakes, respectively. However, during the winter, no significant differences were

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observed between yedoma and non-yedoma lakes. In addition to differences related to permafrost type, an apparent latitudinal pattern was also observed, with higher r_{\max} for lakes from central Alaska compared to those from southern and northern Alaska (Fig. 5a).

4 Discussion

4.1 Geographic and seasonal variations in physicochemical parameters

In yedoma lakes, the CH_4 and DO concentrations were significantly higher and lower, respectively, than in non-yedoma lakes during both seasons. This observation is most likely due to higher organic carbon and nutrient inputs associated with thawing permafrost in yedoma-type lakes. Walter Anthony et al. (2014) and Sepulveda-Jauregui et al. (2014) showed that thawing yedoma permafrost not only provides ancient (Pleistocene-aged) organic carbon stimulating CH_4 production but also phosphate and nitrogen (ammonium), which promotes contemporary plant growth in and around lakes. Since terrestrial plant matter surrounding lakes gets deposited in thermokarst-lake sediments as lakes laterally expand, both enhanced allochthonous and autochthonous productivity of yedoma-type lake ecosystems results in higher rates of contemporary organic matter loading to sediments of yedoma-type lakes compared to non-yedoma lakes (Walter Anthony et al., 2014). Contemporary plant matter decomposes in part to form CH_4 in surface lake sediments, whereas ancient yedoma carbon is largely introduced to lakes at greater depths in the thaw bulb sediments beneath lakes. Thus higher organic carbon and nutrient inputs in yedoma-type lakes promote higher anaerobic and aerobic metabolism and accordingly, lower DO concentrations. Conversely, higher organic carbon inputs promote higher rates of methanogenesis in the sediments (Huttunen et al., 2003), leading to higher dissolved CH_4 concentrations in the lake water column.

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In both yedoma and non-yedoma lakes, higher CH₄ concentrations and lower DO concentrations were observed during the winter than in the summer (Fig. 3). This seasonal variation can be attributed to thick ice covering the lakes in winter. Ice cover impedes gas exchange between the water and the atmosphere, promoting CH₄ build-up in the water column (Phelps et al., 1998; Bastviken et al., 2004; Juutinen et al., 2009) and hindering oxygen transfer from the atmosphere, except in some locations where high-flux ebullition seeps allow gas exchange through local holes in lake ice (Greene et al., 2014). Ice and snow also reduce light penetration and oxygen production by photosynthesis beneath the ice (White et al., 2008; Clilverd et al., 2009). The absence of detectable levels of chlorophyll *a* in ice-covered lakes during March and April (see results section) despite the longer springtime photoperiod was supportive evidence of reduced photosynthesis under the ice. In summer, although CH₄ production was higher due to warmer sediments, ice was not a physical barrier to CH₄ exchange between the lake water and the atmosphere (Fig. 1).

Geographic variations were also observed with higher dissolved CH₄ and lower DO concentrations being found in lakes from central Alaska than in those from southern and northern Alaska. However, this apparent latitudinal pattern was related to the higher concentration of yedoma lakes in central Alaska. No significant latitudinal trend was observed when yedoma and non-yedoma lakes were analyzed separately (TK test, $p < 0.05$).

Figure 3 shows that when relatively high CH₄ concentrations were found, relatively low DO concentrations were observed and conversely, when low dissolved CH₄ concentrations were found, higher DO concentrations were observed. This pattern was particularly clear in yedoma lakes: in winter, a CH₄ concentration of $7.32 \pm 5.86 \text{ mgL}^{-1}$ (mean \pm SD) was found, while the DO concentration was $0.13 \pm 0.03 \text{ mgL}^{-1}$ (mean \pm SD). In the same yedoma lakes, the summer CH₄ concentration was $0.49 \pm 0.52 \text{ mgL}^{-1}$ (mean \pm SD), while the DO concentration was $3.19 \pm 3.24 \text{ mgL}^{-1}$ (mean \pm SD). This observation suggests that MO was actively controlling O₂ and CH₄ concentrations by oxidizing CH₄ when O₂ was present.

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The trend toward higher CH₄ concentrations and lower DO concentrations in winter than in summer was not as strong in non-yedoma lakes as in yedoma lakes (Fig. 3). These results provide additional evidence that organic carbon inputs to yedoma lakes fuel methanogenesis and MO more strongly than in non-yedoma lakes. Another reason is that yedoma lakes have a significantly higher ebullition year round (Walter et al., 2007; Sepulveda-Jauregui et al., 2014). Even during winter, Greene et al. (2014) found that 80 % of CH₄ in ebullition bubbles trapped by lake ice dissolves into the lake water column, leading to elevated dissolved CH₄ beneath the ice. Another possible explanation for higher MO in yedoma lakes compared to non-yedoma lakes may be related to microbial community composition, but this was beyond the scope of our study.

4.2 Limiting elements of MO rates

The actual MO rates r estimated from r_{\max} , reduced the magnitude of the MO, with r ranging from 0.000 to 0.124 mgL⁻¹ d⁻¹ during the winter and from 0.017 to 0.538 mgL⁻¹ d⁻¹ during the summer (Fig. 5b). These values are within the range reported for arctic lakes of 0.001 to 1.000 mgL⁻¹ d⁻¹ (Liikanen et al., 2002; Kankaala et al., 2006; Lofton et al., 2014). Similarly, r values were 1 to 50-fold higher in the summer than in the winter. We attribute this finding to the temperature dependence of methanotrophy (Semrau et al., 2008; Borrel et al., 2011), but also to the limited DO concentration under the ice cover during the winter.

In addition to seasonal variations, permafrost type was also a determining factor of r . As observed with r_{\max} , during summer yedoma lakes showed 2–3 times higher r than non-yedoma lakes (KW test, $p < 0.05$; $r = 0.28 \pm 0.17$ mgL⁻¹ d⁻¹, mean \pm SD, yedoma lakes; $r = 0.09 \pm 0.08$ mgL⁻¹ d⁻¹, mean \pm SD, non-yedoma lakes). Higher r values for yedoma lakes in summer is most likely linked to the higher dissolved CH₄ concentration in presence of a relatively high DO concentration above the oxycline (Fig. 3). An apparent latitudinal trend was observed, with higher r and r_{\max} for lakes from central Alaska compared to those from southern and northern Alaska (Fig. 5).

This apparent trend was associated with a higher number of yedoma lakes in central Alaska. No significant latitudinal trend in MO was observed when yedoma and non-yedoma lakes were analyzed separately.

The actual MO rates; r , were determined from r_{\max} and CH_4 and DO concentrations using two affinity constants, $K_{\text{S-CH}_4}$ and $K_{\text{S-O}_2}$. These affinity constants are highly variable, because their determination is challenging and subject to relatively high determination error (Segers et al., 1998) and because the methanotrophic community is sensitive to numerous factors and changes over time and space (Carini et al., 2005; He et al., 2012). For instance, Lofton et al. (2014) reported a variation of 150 % in $K_{\text{S-CH}_4}$ within the hypolimnetic water column of two lakes with similar characteristics. The determination of MO rates may, therefore, be subject to large error if reported values are used instead of experimental parameters or if an error occurs in experimental K_{S} determinations. To quantify these potential errors, a sensitivity analysis was conducted. We arbitrarily modified $K_{\text{S-CH}_4}$ and $K_{\text{S-O}_2}$ and calculated the resulting r (Eq. 1) using the experimental r_{\max} , C_{CH_4} , and C_{O_2} measured in the 30 lakes.

Figure 6 shows the impact of varying $K_{\text{S-O}_2}$ (Fig. 6a) and $K_{\text{S-CH}_4}$ (Fig. 6b) on the sensitivity coefficient ($\Delta r/\Delta K_{\text{S}}$), where Δr is the variation in r (%) for a given variation in K_{S} (%). In Fig. 6, $\Delta r/\Delta K_{\text{S}}$ is expressed as a function of K'_{S} , which is K_{S} normalized over the range reported in the literature, with 1.0 being the average K'_{S} reported (e.g. 0.110 mg L^{-1} for $K_{\text{S-CH}_4}$). Figure 6a shows that $\Delta r/\Delta K_{\text{S-O}_2}$ ranged from 6 to 12 % for all lakes and seasons, except in yedoma lakes during winter, where an error of up to 200 % was estimated. This relatively high sensitivity to error in $K_{\text{S-O}_2}$ in yedoma lakes during winter is likely due to DO concentrations close to $K_{\text{S-O}_2}$. Likewise, Fig. 6b shows that $\Delta r/\Delta K_{\text{S-CH}_4}$ ranged from 2 to 9 % for all lakes and seasons, except in non-yedoma lakes during summer where an error of up to 100 % was estimated. As above, the latter is likely due to CH_4 concentrations close to $K_{\text{S-CH}_4}$. This sensitivity analysis shows that, other than for $K_{\text{S-O}_2}$ in yedoma lakes during winter and $K_{\text{S-CH}_4}$ in non-yedoma lakes during summer, errors or variations in K_{S} would have relatively little

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impact on determination of methanotrophic rates. Overall, under the conditions of the present study, taking into account yedoma and non-yedoma lakes in each season, the weighted average of $\Delta r/\Delta K_{S-O_2}$ and $\Delta r/\Delta K_{S-CH_4}$ ranged from 8 to 40%. This error is significantly lower than the standard deviation of r , which averaged 119%.

From Eqs. (2) and (3), we estimated that, during the summer CH_4 was the main limiting element in 25 out of 30 lakes. In contrast, during winter, CH_4 was the main limiting element in 10 of 26 lakes (Table 2). Notably, during the winter, DO was the limiting element for all seven yedoma lakes, while during the summer, MO was limited by CH_4 for all non-yedoma lakes.

A similar sensitivity analysis was done on β and γ , as done with r , to estimate if the estimated limiting element would change for variable K_S . The results showed no impact on the limiting element in the 30 lakes and for both seasons. These results confirm that MO was mainly controlled by DO and CH_4 availability, which in turn, depended on the season and landscape processes.

A potential bias in our r estimates may have arisen from taking duplicate samples at a single depth in each lake. The literature has clearly shown that a higher MO rate is often found at the oxic/anoxic interface in stratified lakes (Utsumi et al., 1998a, 1998b; Bastviken et al., 2002; Carini et al., 2005; Pimenov et al., 2010; Schubert et al., 2010). Estimation of MO rates consistently measured at a single depth that was not necessarily located at the oxic/anoxic interface may have neglected potentially higher rates occurring at the oxic/anoxic interface in stratified lakes. However, in the present study, the sampled lakes were in many cases shallow, relatively well mixed, and without a clear oxycline (see Results section), suggesting a relatively homogeneous water column. Utsumi et al. (1998b) observed homogeneous MO rates at all depths of a shallow and mixed temperate lake, while Rudd and Hamilton (1978) also reported homogeneous MO rates during overturn of a dimictic lake. Determination of MO rates at the oxic/anoxic interface, in the few cases in which such an interface was observed, would likely have indicated higher MO rates. Thus, the results of r presented here, may be underestimated to an unknown extent.

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We developed a new method based on a TDLAS for the determination of MO rates together with dissolved CH₄ concentration in lakes in the field. This method was successfully applied to 30 lakes along a north–south transect and allowed for the determination of MO potentials ranging from 0.000 to 1.339 mgL⁻¹d⁻¹ in winter and summer. MO rates showed high seasonal and geographic variability. In addition to temperature effects, the main factors controlling MO were: (1) CH₄ availability during the summer, limited both by exchange with the atmosphere and by MO itself, (2) DO availability during the winter, mainly due to ice cover impeding gas exchange with the atmosphere and primary production; and (3) inputs of organic substrates to lakes, mainly related to the presence or absence of yedoma permafrost as an additional source of carbon and nutrients. These results indicate that MO may substantially mitigate the increase in CH₄ emission predicted by permafrost thawing (Khvorostyanov et al., 2008; Walter Anthony et al., 2014).

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Table 1. Identification, location, and permafrost soil type for lakes included in the study.
*Indicates informal lake name.

#	Name	Lat.	Long.	Permafrost type
1	Big Sky* A31	69.581	-148.639	Non-Yedoma
2	GTH 112	68.672	-149.249	Non-yedoma
3	NE2	68.647	-149.582	Non-yedoma
4	Toolik A28	68.632	-149.605	Non-yedoma
5	E1	68.626	-149.555	Non-yedoma
6	Julieta* A27	68.447	-149.369	Non-yedoma
7	El Fuego* A36	67.666	-149.716	Non-yedoma
8	Jonas* A26	67.647	-149.722	Non-yedoma
9	Augine Zoli* A25	67.138	-150.349	Non-yedoma
10	Ping*	67.136	-150.370	Non-yedoma
11	Grayling* A24	66.954	-150.393	Non-yedoma
12	Eugenia*	65.834	-149.631	Yedoma
13	Goldstream*	64.916	-147.847	Yedoma
14	Killarney*	64.870	-147.901	Yedoma
15	Smith A13	64.865	-147.868	Non-yedoma
16	Stevens Pond*	64.863	-147.871	Yedoma
17	Duece A2	64.863	-147.942	Yedoma
18	Ace A1	64.862	-147.937	Yedoma
19	Rosie Creek*	64.770	-148.079	Yedoma
20	Otto	63.842	-149.037	Non-yedoma
21	Floatplane* A16	63.394	-148.670	Non-yedoma
22	Montana A40	62.143	-150.048	Non-yedoma
23	Rainbow Shore* A41	61.694	-150.089	Non-yedoma
24	Big Merganser A49	60.726	-150.644	Non-yedoma
25	Rainbow A48	60.719	-150.808	Non-yedoma
26	Dolly Varden A47	60.704	-150.787	Non-yedoma
27	Abandoned Cabin* A50	60.696	-151.315	Non-yedoma
28	Scout A46	60.533	-150.843	Non-yedoma
29	Engineer A45	60.478	-150.323	Non-yedoma
30	Lower Ohmer A44	60.456	-150.317	Non-yedoma

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Table 2. Methane oxidation parameters for 30 Alaskan lakes. * Indicates median; ND – Not determined; BDL – Below detection limit.

#	CH ₄ (mgL ⁻¹)		O ₂ (mgL ⁻¹)		Potential MO (mgL ⁻¹ d ⁻¹)		Actual MO (mgL ⁻¹ d ⁻¹)		Limiting element	
	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer
1	6.43	0.05	0.13	10.53	0.23	0.11	0.08	0.03	O ₂	CH ₄
2	0.58	0.09	0.43	8.28	0.14	0.36	0.05	0.15	O ₂	CH ₄
3	0.02	0.05	1.73	9.80	BDL	0.07	0.02	0.02	CH ₄	CH ₄
4	0.11	0.03	10.09	9.46	BDL	0.28	0.00	0.06	CH ₄	CH ₄
5	0.05	0.04	9.59	9.52	0.10	0.19	0.03	0.05	CH ₄	CH ₄
6	0.08	0.07	ND	9.81	0.12	0.20	ND	0.09	ND	CH ₄
7	ND	0.06	ND	9.65	ND	0.18	ND	0.04	ND	CH ₄
8	3.68	0.03	13.63	10.30	0.03	0.18	0.03	0.04	O ₂	CH ₄
9	8.83	0.11	3.64	9.87	0.05	0.39	0.04	0.19	O ₂	CH ₄
10	3.00	0.06	0.25	6.94	0.11	0.11	0.03	0.03	O ₂	CH ₄
11	8.43	0.88	0.19	9.31	BDL	0.28	0.00	0.23	O ₂	CH ₄
12	0.79	0.07	0.15	6.90	0.09	0.29	0.02	0.11	O ₂	CH ₄
13	8.43	0.19	0.11	6.23	0.49	0.54	0.07	0.48	O ₂	O ₂
14	12.59	0.31	0.09	0.31	0.20	0.92	0.03	0.30	O ₂	O ₂
15	1.30	0.02	0.23	3.93	0.05	0.31	0.01	0.05	O ₂	CH ₄
16	ND	0.72	0.11	1.36	ND	0.34	ND	0.20	O ₂	O ₂
17	6.60	0.59	0.19	0.57	0.06	1.34	0.01	0.54	ND	O ₂
18	0.70	0.03	0.14	6.74	0.02	0.77	0.00	0.15	O ₂	CH ₄
19	14.77	1.51	0.13	0.22	0.20	0.74	0.04	0.19	O ₂	O ₂
20	1.24	0.03	0.31	9.47	0.05	0.67	0.01	0.15	O ₂	CH ₄
21	ND	0.04	ND	9.52	ND	0.33	ND	0.08	ND	CH ₄
22	0.08	0.05	2.79	11.07	0.05	0.20	0.01	0.06	CH ₄	CH ₄
23	0.30	0.08	5.84	9.59	0.02	0.68	0.01	0.33	CH ₄	CH ₄
24	0.04	0.02	12.40	9.66	0.06	0.34	0.02	0.08	CH ₄	CH ₄
25	0.08	0.03	11.91	10.20	0.32	0.25	0.12	0.04	CH ₄	CH ₄
26	0.01	0.03	10.00	10.24	0.04	0.08	0.00	0.02	CH ₄	CH ₄
27	0.03	0.02	7.90	9.67	0.15	0.41	0.03	0.07	CH ₄	CH ₄
28	0.07	0.04	0.20	9.01	BDL	0.38	0.00	0.09	O ₂	CH ₄
29	0.04	0.04	9.13	10.19	0.02	0.28	0.00	0.05	CH ₄	CH ₄
30	ND	0.03	ND	10.25	ND	0.38	ND	0.11	ND	CH ₄
	Mean									
Global	3.29	0.47	3.91	7.95	0.10	0.39	0.03	0.13	O ₂ *	CH ₄ *
Yedoma	7.53	1.73	0.14	3.19	0.18	0.71	0.03	0.28	O ₂ *	CH ₄ *
Non-yedoma	2.02	0.09	5.30	9.40	0.08	0.29	0.03	0.09	CH ₄ *	CH ₄ *

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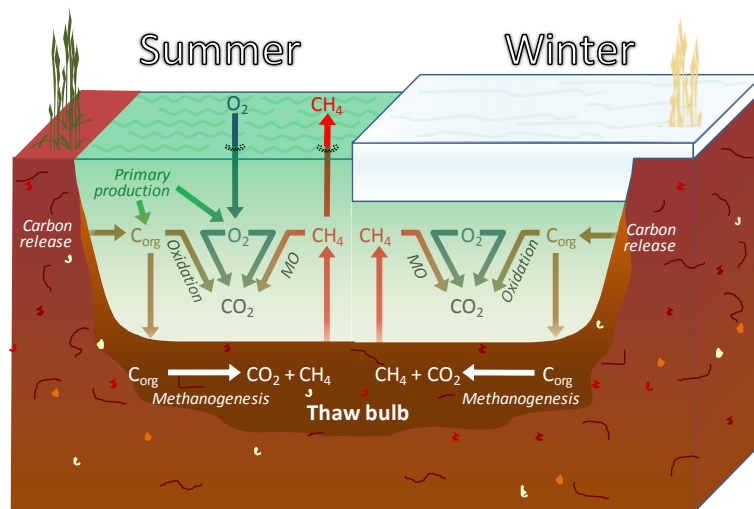


Figure 1. Carbon cycling in northern high-latitude lakes during the summer and winter. Carbon (C_{org}) release from primary production and landscape processes promotes CH_4 production and competes with MO for O_2 .

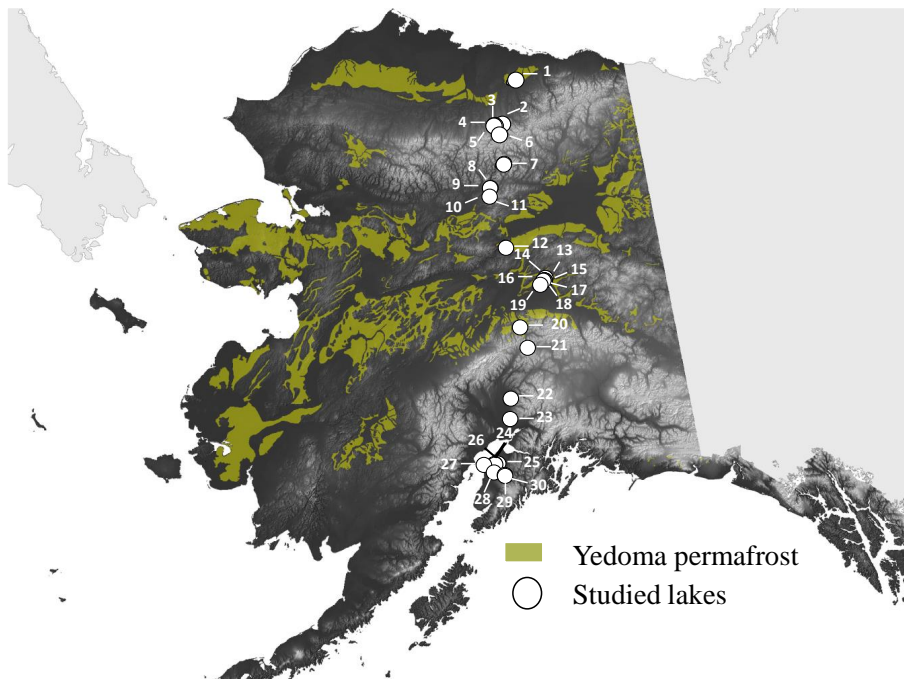


Figure 2. Locations of studied Alaskan lakes (white circles) plotted on the Alaska DEM hillshade raster. Information about the distribution of yedoma-type deposits (Pleistocene-aged, ice-rich silt containing deep thermokarst lakes) was from Jorgenson et al. (2008) and Kanevskiy et al. (2011). The Alaska map is the National Elevation Data Set 30 m hillshade raster.

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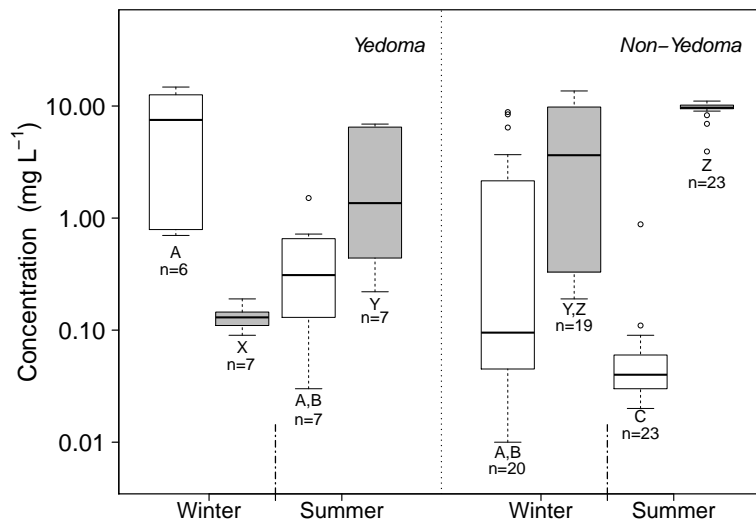


Figure 3. Statistical distributions of CH₄ (white boxes) and DO (grey boxes) water concentrations in yedoma and non-yedoma lakes during the winter and summer. The boxes include the median (Q2) and the quartile range (Q1 and Q3). The whiskers show minimum and maximum data. The open circles show outlier data. Capital letters are Tukey–Kramer test results; values with the same capital letter are not significantly different ($p < 0.05$). n represents the number of lakes measured.

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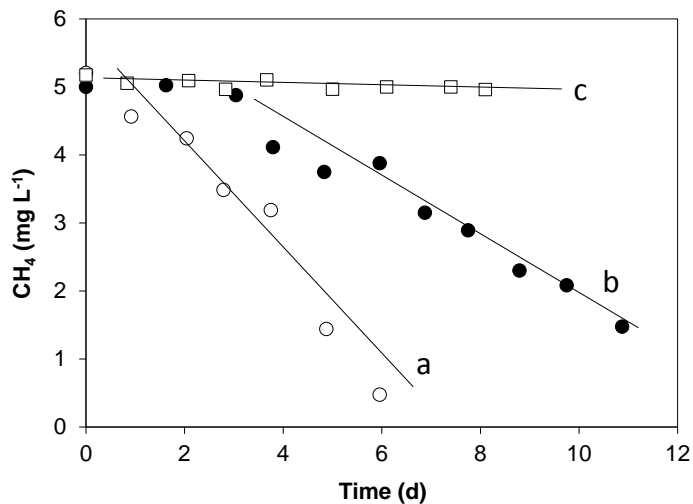


Figure 4. Examples of CH₄ oxidation patterns observed during the MO assays: **(a)** assay with no lag-phase, **(b)** assay with a 3-d lag-phase, and **(c)** assay with no detected activity. Straight lines are linear correlations.

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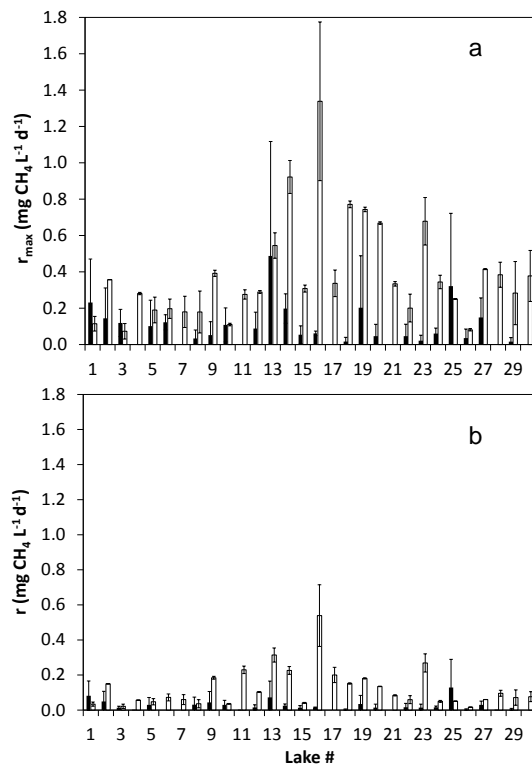


Figure 5. (a) CH₄ oxidation potential (r_{max}) and (b) CH₄ oxidation rates observed in 30 lakes along a north–south transect (left–right) during the summer (white bars) and the winter (black bars).

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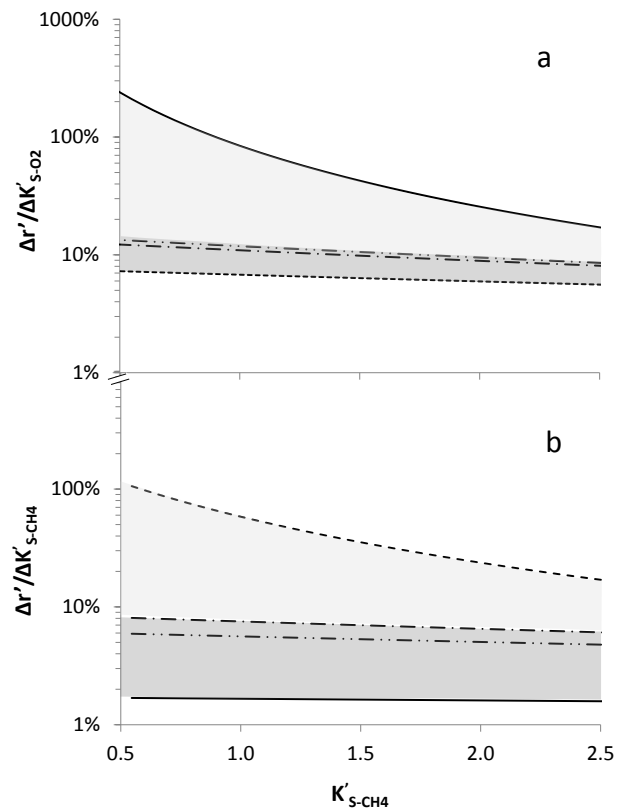


Figure 6. Sensitivity analysis of the impact of an error or variation in K_{S-O_2} **(a)** and K_{S-CH_4} **(b)** on r ; yedoma lakes in winter (—), yedoma lakes in summer (---), non-yedoma lakes in winter (-·-·-) and non-yedoma lakes in summer (----).

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