Isabelle Laurion

Associate Editor

Biogeosciences Discussion

RE: Comments C1087 y C1149

May 29 2015

Dear Dr. Isabelle Laurion,

Thank you for providing constructive comments on our manuscript, "Geographic and seasonal variation of dissolved methane and aerobic methane oxidation in Alaskan lakes (bg-2015-49)". We have re-written the manuscript, taking into account the concerns and recommendations of the two referees.

Please find below our detailed response to the comments received, as well as an indication of how we addressed the comments in the revised manuscript. Thanks to the comments received, we consider that the revised manuscript has been substantially improved and we hope that you will now consider it suitable for publication at *Biogeoscience*.

Sincerely,

Frédéric Thalasso

Comments received and our answer

Anonymous Referee #1

Thank you for providing constructive comments on our manuscript, "Geographic and seasonal variation of dissolved methane and aerobic methane oxidation in Alaskan lakes (bg-2015-49)". We have re-written the manuscript, taking into account your concerns and recommendations.

Comment: Martinez-Cruz and co-authors performed surveys along a transect through Alaska, incorporating 30 lakes during a summer and a winter season. By doing this, they accumulated a huge data-set on methane - and oxygen concentrations in these lakes. Additionally they analysed aerobic methane oxidation (MO) rates. This became possible due to a laser spectroscopy method for the field, newly developed by this group. The authors are well known specialists for studies on greenhouse gases (GHG), mainly on methane in arctic environments. Global changes in climate lead to thawing of permafrost in the arctic regions, with related increase in organic carbon supply to aquatic systems. The study is highly important because of the amount of data from arctic lakes, as well as because of the differences in the lakes, chosen. Methane oxidation within the environment of production is one of the most important pathways to mitigate GHG emission to the atmosphere. It is a merit of this study to summarize such a data-set from the arctic. The ms is well written. The authors have chosen a fixed pattern of sampling depths (same depths in all lakes, except very shallow or very deep ones). To overcome the problem of sampling outside the oxycline, the place of maximum MO, they used a double monod model. A sensitivity analysis was conducted to calculate the MO rate when affinities (Ks-CH4 and Ks-O2) would change. It is an important paper addressing relevant scientific (and social) question on the basis of a well developed sampling design.

Our answer: We are grateful for this encouraging comment.

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Specific comments:

1. A clear description of yedoma and non-yedoma types is needed on a prominent position.

Our answer: We have modified the section located on Page 4215 L.26, as follows;

"Yedoma-type permafrost is an organic-rich (about 2% carbon by mass) Pleistocene-age permafrost with ice content of 50–90% by volume (Zimov et al., 2006), which occurs mainly in the previously unglaciated regions of Siberia, Alaska, and NW Canada (Czudek and

Demek, 1970; Walter et al., 2007; Kanevskiy et al., 2011; Grosse et al., 2013). Non-yedoma permafrost is characterized by thinner ice-rich horizons and have a more widespread distribution (Ping et al., 2008; Tarnocai et al., 2009; Hugelius et al. 2014)".

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1.a. In both tables I recommend to underlay the yedoma lakes rows by a light grey bar

Our answer: We agree and we have modified both Tables.

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2. page2/line 4: dependance of MO on CH4 and O2 concentrations is to general; I would prefer to read about 'relation at the interface'

2.a. page2/line 4: MO depends only indirectly on OC supply –via methanogenesis and see 2. on relation at interface

Our answer: We have modified that section of the Abstract, as follows;

"Aerobic CH_4 oxidation depends mainly on lake CH_4 and oxygen (O_2) concentrations, in such manner that higher MO rates are usually found at the oxic/anoxic interface, where both molecules are present. MO also depends on temperature, and via methanogenesis, on organic carbon input to lakes, including from thawing permafrost in thermokarst (thaw)-affected lakes."

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3. page2/line 13: CH4 concentration may be lower at a given depth in summer due to the better oxygen supply compared to winter; it should not be called 'deficit'

Our answer: We agree and we removed the term "deficit", as follows;

"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 scarce compared to dissolved O_2".

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4. page2/line 17: the meaning of 'landscape processes' could be more clearly described as 'coupling of terrestrial and aquatic habitats'

4.a. page2/line 17/Fig 1 etc: the thawing permafrost needs to be more clearly described: when thawing impacts terrestrial plants in the catchment of the lakes (p12/l13), a direct surface input/inflow of thawed material into the lake seems to be likely; perhaps this can be shown in Fig 1 also

Our answer: Thank you for this comment. We agree and we modified the abstract section, by adding a new sentence on coupling of terrestrial and aquatic habitats, as follows;

"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. As thermokarst lakes formed in yedoma-type permafrost have been identified to receive large quantities of terrestrial organic carbon from thaw and subsidence of the surrounding landscape into the lake (Walter Anthony et al. 2014), these results confirm that coupling of terrestrial and aquatic habitats play an important role in lake CH_4 cycling".

We also modified Figure 1, as suggested.

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5. page2/line 22: references should be clear (in text, in list) if Walter, Walter Anthony or Anthony, KMW (p21/l30) are different persons

Our answer: All of these are the same person. Walter is a maiden name, and Walter Anthony is a married name. We revised the manuscript to more consistently refer to this person and clearly follow the reference database.

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6. page5/line 13: please delete 'offshore and'

Our answer: Thank you for this observation. Changed accordingly.

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7. page6/line 13: please, give full name of HE-TDLAS –I guess it is an abbreviation

Our answer: This is correct, we included now a full reference;

"To avoid long delays in sample transfer from remote locations to the laboratory, we determined dissolved CH_4 concentrations with a previously described method based on Headspace Equilibration using Infrared Tunable Diode Laser Absorption Spectroscopy (HE-TDLAS; Sepulveda-Jauregui et al., 2012)".

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8. page10/line 25: see 7., here TK

Our answer: The first reference on TK was presented on Page 9, L10 (P4221, L10 of the PDF file). Please note that according to the first general comment from reviewer 2 (Dr. Stepanenko), we modified the statistical test from Tukey-Kramer to Kruskal-Wallis multiple comparison test. However, based on this comment, we decided to write out Kruskal-Wallis every time it appears in the text.

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9. page12/line 13: nutrient supply (P, N) can force bacterial and algal growth also, with similar effect on sedimentation etc.

Our answer: Thank you for this important clarification. We modified this sentence, as follows;

"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 bacterial, algal and contemporary plant growth in and around lakes".

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10. page12/line 20: see 4a: please, describe clearly, where thawed C reaches the lake; deep under beneath the sediment or from the catchment or in medium depth directly from thawed surface sediments (as in Fig. 1)

Our answer: This is an important comment that we took into account, trying to describe concisely the carbon release, as follows;

"Contemporary organic matter decomposes in part to form CH_4 in surface lake sediments, whereas ancient yedoma carbon is progressively released from thaw bulb beneath lakes to surface sediments (Heslop et al., 2015). Hence, organic carbon is made available to microbial decomposition in both shallow and deep sedimentary environments (Fig. 1)".

Please note that we modified Fig. 1 accordingly.

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11. page13/line 13: "CH4 production was higher due to warmer sediments" as there is no measurement for this statement, it should be rewritten

Our answer: We agree, we modified that sentence accordingly;

"In summer, although CH_4 production was probably higher due to warmer sediments, ice was not a physical barrier to CH_4 exchange between the lake water and the atmosphere (Fig. 1).

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12. page13/line 15ff: The title of ms "Geographic and seasonal variation. . ." has to be followed by a clear statement about geographical variation;

12.a. please replace 'concentration..of lakes' by 'number of lakes' or 'portion of ..'

Our answer: We are not sure we understand clearly the first part of this comment. Reviewer 1 will probably agree that "geographic" refers to "all of the natural features of a region or regions" and as such, our manuscript discuss in detail the effect of the landscape, permafrost type and latitude. If Reviewer 1 could be more specific we would be happy to further attend to

this comment in our manuscript. We agree with the second part of the comment and we changed “concentration of yedoma lakes” to “proportion of yedoma lakes”

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Comment: The references are on the state of the art. All figures are necessary and well prepared.

Our answer: Thanks for your supporting review.

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Reviewer 2. V. Stepanenko (Referee)

Thank you for providing constructive comments on our manuscript, “Geographic and seasonal variation of dissolved methane and aerobic methane oxidation in Alaskan lakes (bg-2015-49)”. We have re-written the manuscript, taking into account your concerns and recommendations.

General comment

This manuscript presents results of estimate of methane oxidation in over 30 Alaskan lakes. This is the first such study for this region, and, to my knowledge, is the methane oxidation study involving data from the largest lake set so far. The study is significant for the area of greenhouse gas dynamics in lacustrine ecosystems since it presents the new field method for determining methane oxidation potential, and achieves clear conclusions on key factors controlling methane oxidation in Arctic tundra. The manuscript is well written, and the main conclusions are unambiguously stated. These are the strong points.

Our answer: We are grateful for this encouraging comment.

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Comment: The weak points are two in my view.

1) The authors sampled 7 yedoma lakes and 23 non-yedoma lakes. So, the reliability of statistics on these two sets is different. Are there any estimates on the sufficient n for the statistical estimates accuracy needed? This is especially relevant for yedoma lakes.

Our answer: Dr. Stepanenko underlined a very important point. In the previous version of our manuscript, we used the Tukey-Kramer test to compare dissolved CH_4 and DO concentration, during winter and summer, for yedoma and non-yedoma lakes. This test was performed using the actual sample size (unequal number of yedoma and non yedoma lakes). However, after

revision, Dr. Stepanenko is correct and Tukey-Kramer test was not the most appropriate test to make comparisons with different samples size. To avoid confusion, we decided to re-analyze the data using the Kruskal-Wallis multiple comparison test, which is a more appropriate method for comparisons with different sample size and non-normally distributed data. Please note that we obtained the same test results than with Tukey-Kramer test. Thank you for this important observation. We modified the Results section accordingly and we changed the Material and Method section as follows;

"Normality was assessed by the Shapiro-Wilk test. Since most of the data was non-normally distributed and with unequal samples number, significant differences among all parameters were determined using Kruskal-Wallis multiple comparison test (differences were considered significant at $p < 0.05$, $Z > 1.96$). To assess whether CH_4 was oxidized during the MO incubation tests, significant differences between C_{CH4} were determined by an analysis of variance (ANOVA; $p < 0.05$), after normality was assessed by the Shapiro-Wilk test ".

Comment: 2) The authors admit (p.4228, str.15-30) that measuring methane and oxygen concentrations at 0.5 and 1 m depths they likely underestimate the maximal methane oxidation rate in a lake that is typically located in the thermocline. Thus, the authors should precise that they assess methane oxidation rates in the surface layer, that is not a good proxy for larger depths and lake as a whole. Therefore, I strongly recommend to look through the text and modify it accordingly, replacing "MO in a lake" by "MO in a surface lake layer" etc. The title of the manuscript should be rewritten as well, e.g.: "Geographic and seasonal variation of dissolved methane and aerobic methane oxidation in the surface layer of Alaskan lakes". Otherwise the authors would have to exclude deep lakes from their analysis where the thermocline is well-developed in summer.

Our answer: We partially agree with this comment. As clearly stated in our manuscript, all water samples were taken at a depth of one meter, except in some cases, when the ice layer was thicker than 1 m. We also agree that in several cases, 1-m depth can be considered as surface water. However, and in many cases, lakes were shallow and our sampling regime of near 1-m can reflects mid-column or lake bottom water environments. We clarified that point in the Materials and Methods section, as follows;

"Water samples for MO rates and dissolved CH_4 concentration were taken at a depth of within 1 m of the ice-water interface in winter and usually at 0.75 to 1 m water depth in summer. Due to differences in lake depth and thickness of the ice sheets, samples reflected surface water in deep lakes, but mid water column or even lake bottom water environment in shallow lakes".

Specific comments

Comment: p. 4222, 5-10, It would be useful to indicate if there was a connection between lakes' depths and RWCS. E.g., were deeper lakes more stratified in general?

Our answer: As suggested, we checked for a correlation between lake depth and RWCS. We did not find any correlation and we indicated in the manuscript;

"Overall, only one third of the temperature profiles indicated clear stratification. In both seasons, no correlation between RWCS and lake depth was found, probably due to the fact that lakes were shallow and with an uneven depth distribution".

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Comment: p. 4223, 6, Remove one "potential"

Our answer: Changed accordingly.

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Comment: p. 4223, 15-20, An interpretation of lag phase is relevant

Our answer: We modified that section to make clearer the interpretation of the lag phase. This section is now;

"In about 60% of the cases during the summer and 80% during the winter, a lag phase was observed; i.e. period of time with no apparent MO (Fig. 4b). This behavior, termed "induction of MO", has previously been reported for various soils (Bender and Conrad, 1995; Dunfield et al., 1999) and can be interpreted as an adaptation period of the CH_4 oxidizers to the culture conditions".

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Comment: p. 4225, 29-30, This requires more quantitative estimates.

Our answer: We agree this is important but the impact of MO on oxygen uptake is difficult to estimate quantitatively, from our results. Therefore, we opted for a suggestion as follows;

"This observation suggests that MO was actively controlling O_2 and CH_4 concentrations by oxidizing CH_4 when O_2 was present. To confirm the latter, it would be necessary to measure experimentally the O_2 uptake rate by methanotrophs and by the other aerobic processes that compete with MO".

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Comment: p. 4226, Title, May be, "Limiting factors of MO rates" is better?

Our answer: We agree and we changed the title of that section as well as the all text, Tables and Figures, to include “limiting factor” instead of “limiting element”.

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Comment: p. 4226, 23-26, "is most likely linked to the higher dissolved CH4 concentration" sounds strangely, since due to eq. (1) it is straightforward to check the contribution of both CH4 and DO into reduction of potential MO.

Our answer: This is correct, we were being too cautious. As we indeed have clear evidences, we modified that section as follows;

“Higher r values for yedoma lakes in summer is explained by the higher dissolved CH_4 concentration in presence of a relatively high DO concentration above the oxycline (Fig. 3).”

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Comment: p. 4227, Any discussion on maximal MO potential (rmax) for yedoma lakes is missing

Our answer: We agree and we included a new discussion section, as follows;

“In addition to seasonal variations, permafrost type was also a determining factor of *r and* r_{max}. As mentioned before, although no difference in r_{max} was observed during winter between yedoma and non-yedoma lakes, r_{max} in yedoma lakes was about twice higher than in non-yedoma lakes during summer. We attribute that difference to a more active MO methanotrophic community in yedoma lakes, as all r_{max} tests were conducted in aerated vials with a fixed initial standard CH_4 concentration in the liquid phase (~0.6 mg L^{-1}), thus ensuring optimal conditions”.

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Comment: p. 4243, Fig.6, a, Horizontal axis should have a label kS−O2 p. 4227, 16-25, Δr/ΔKS – is that a ratio of two values or just a notation for Δr for a given ΔKS? In the former case this ratio cannot be expressed in %, and in the latter please denote it as Δr(ΔKS), i.e. Δr as a function of ΔKS.

Our answer: That was certainly unclear. Please check our answer to the last comment, which was on the same point.

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Comment: p. 4241, Caption, Replace "3-d" by "3-day"

Our answer: Yes, indeed, changed accordingly.

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Comment: p. 4242, Figure 5a consists of two small plots. Please enlarge them

Our answer: We enlarged the plots

Comment: p. 4243, Fig.6, I'm totally confused with this Figure. If Δr is a deviation of r from its value at a mean K'_{S-CH4} , Δr must be 0 when $K'_{S-CH4} = 1$. Moreover, increasing K'_{S-CH4} above 1 we must get negative Δr (decrease below r corresponding to mean K'_{S-CH4}). Please clarify what are the values Δr and how they are calculated. Moreover, the authors use Δr' notation, whereas I can't find it in the text.

Our answer: We agree this sensitivity analysis was probably not easy to understand. Based on that comment, we decided to simplify substantially the analysis and we present now a simple error estimation on *r*, for a given error on K_S. With that new presentation, we believe we reached the same conclusion in a simpler way, more understandable for reader unfamiliar with sensitivity analysis. Thanks for this comment. This section has been modified as follows;

"To quantify these potential errors, we conducted a sensitivity analysis. We arbitrarily modified K_{S-CH4} and K_{S-O2} and calculated the resulting *r* (Eq. 1) using the experimental r_{max}, C_{CH4}, and C_{O2} measured in the 30 lakes. Fig. 6 shows the error on *r* caused by a given error on K_{S-O2} (Fig. 6a) and K_{S-CH4} (Fig. 6b), for yedoma and non yedoma lakes, in winter and in summer. According to this analysis, an underestimation of K_{S-O2} or K_{S-CH4} would lead to an overestimation of the actual MO rate (positive error), while an overestimation of these affinity constants would produce an underestimation of r (negative error). Fig. 6a shows that, an error on K_{S-O2} ranging from -50% to 200%, would cause from 10% to -6% error on r, for all lakes and all seasons, except in yedoma lakes during winter, where an error from 75% to -50% would be generated. This relatively high sensitivity of *r* to error in K_{S-O2} in yedoma lakes during winter is due to DO concentrations close to K_{S-O2}. Likewise, Fig. 6b shows that, from an error on K_{S-CH4} ranging from -50% to 200%, a resulting error on r from 6% to -4% would be done, for all lakes and all seasons, except in non yedoma lakes during summer, where an error from 50% to -34% would be generated. As above, the latter is due to CH_4 concentrations close to K_{S-CH4} in non yedoma lakes during summer. This sensitivity analysis shows that, other than for K_{S-O2} in yedoma lakes during winter and K_{S-CH4} in non-yedoma lakes during summer, errors on K_S would have relatively little impact on determination of methanotrophic rates".

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 mainly on lake CH_4 and oxygen (O_2) concentrations, in such manner that higher MO rates are usually found at the oxic/anoxic interface, where both molecules are present. MO also depends on temperature, and via methanogenesis, on ~~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~~scarce 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. As thermokarst lakes formed in yedoma-type permafrost have been identified to receive large quantities of

terrestrial organic carbon from thaw and subsidence of the surrounding landscape into the lake, these results confirm that coupling of terrestrial and aquatic habitats. ~~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 to 99% of total CH_4 produced in freshwater 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 CH_4 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 CH_4-C produced into the biogeochemical carbon cycle within lakes. As recently demonstrated using stable isotopes, after assimilating CH_4, 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 CH_4 and O_2. Higher MO rates are usually found at the oxic/anoxic interface, where both CH_4 and O_2 are present (Utsumi et al., 1998a, 1998b; Bastviken et al., 2002; Liikanen et al., 2002; Carini et al., 2005; Schubert et al., 2010). In turn, CH_4 and O_2 concentrations depend on numerous other processes involved in biogeochemical carbon cycling (Fig. 1). Among these, the most important are methanogenesis producing CH_4, primary production and atmospheric diffusion supplying O_2, and several aerobic metabolic processes that compete with MO for available O_2 (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, y~~Yedoma-type permafrost ~~is an organic-rich (about 2% carbon by mass) Pleistocene-age permafrost with ice content of 50–90% by volume (Walter et al., 2006), (organic-rich, ice-supersaturated, Pleistocene-aged loess deposits) which 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, are characterized by but the a thinner organic-rich horizons of non-yedoma permafrost and permafrost free soils are typically thinner than those of yedoma deposits, and are more evenly distributed than yedoma permafrost (Ping et al., 2008; Tarnocai et al., 2009; Hugelius et al. 2014)~~ is an organic-rich (about 2% carbon by mass) Pleistocene-age permafrost with ice content of 50–90% by volume (Zimov et al., 2006), which occurs mainly in the previously unglaciated regions of Siberia, Alaska, and NW Canada (Czudek and Demek, 1970; Walter et al., 2007; Kanevskiy et al., 2011; Grosse et al., 2013). Non-yedoma permafrost is characterized by thinner ice-rich horizons and have a more widespread distribution (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 CH_4 and O_2 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 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". 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 km^2), their trophic states (ultraoligotrophic to eutrophic), and their annual CH_4 fluxes (0.5–317 g CH_4 m^{-2} y^{-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). 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 Varden L. where measurement intervals were increased to every five meters from 15 m to 25m 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 CH_4 concentration were taken at a depth of within 1 m of the ice-water interface in winter and usually at 0.75 to 1 m water depth in 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.~~ Due to differences in lake depth and thickness of the ice sheets, samples reflected surface water in deep lakes, but mid water column or even lake bottom water environment, in shallow lakes. Samples were taken with a horizontal Van Dorn bottle (Wildco, Yulee, FL, USA).

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 ± standard deviation (SD).

2.3 Dissolved CH_4 concentration and MO rate

To avoid long delays in sample transfer from remote locations to the laboratory, we determined dissolved CH_4 concentrations ~~used~~ with a previously described method ~~for determination of dissolved CH_4 concentrations (~~based on Headspace Equilibration using Infrared Tunable Diode Laser Absorption Spectroscopy (HE-TDLAS; Sepulveda-Jauregui et al., 2012). This method consisted of determining the CH_4 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 CH_4 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 CH_4 analyzer (GasFinder 2; Boreal Laser, Edmonton, Canada). The CH_4 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 taken 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 most of the dissolved CH_4 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 CH_4 content of the water sample. Using this procedure, more than 99.5% of the original CH_4 content of the sample was evacuated. The equilibration vials were then closed with rubber stoppers and aluminum crimp caps, spiked with 0.6 mL CH_4 (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 CH_4 concentration in the liquid phase (~0.6 mg L^{-1}). It also provided an initial CH_4 to O_2 molar ratio of 0.062, significantly below the stoichiometric ratio (0.5), ensuring no O_2 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 ± 2 °C in a water bath with ice supplements; in the summer, the vials were maintained at 15 ± 2 °C. The temperature of the water bath was measured daily. We measured the CH_4 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 CH_4 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 CH_4 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 CH_4 concentration (C_{CH4} = total CH_4 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_{CH4} in the equilibration vials with time. MO rates determined by this method represent the MO rate after aeration and CH_4 addition (vials spiked with CH_4 and vigorously shaken). Thus, these MO rates do not correspond to actual observations of in situ DO and dissolved CH_4 concentrations in the lakes. The measured CH_4 oxidation rate was considered the potential MO (r_{max}; mg CH_4 L^{-1} d^{-1}) under non-limiting CH_4 and DO concentrations. To estimate the actual rate (r; mg CH_4 L^{-1} d^{-1}) from r_{max}, a double

Monod model was used (Bae and Rittmann, 1996; Segers, 1998) in which C_{CH4} and C_{O2} represent the actual dissolved CH_4 and DO concentrations measured in the lake, respectively, and $K_{S\text{-}CH4}$ and $K_{S\text{-}O2}$ are the apparent affinity constants of the methanotrophic community, for CH_4 and DO, respectively:

$$r = r_{\max} \cdot \frac{C_{CH4}}{K_{S-CH4} + C_{CH4}} \cdot \frac{C_{O2}}{K_{S-O2} + C_{O2}} \quad (1)$$

Average $K_{S\text{-}CH4}$ and $K_{S\text{-}O2}$ values for lakes have been determined by previous studies: $K_{S\text{-}CH4}$ = 0.110 ± 0.053 mg L^{-1} (mean ± SD; Liikanen et al., 2002; Lofton et al., 2013) and $K_{S\text{-}O2}$ = 0.624 ± 0.064 mg L^{-1} (mean ± SD; Lidstrom and Somers, 1984; Frenzel et al., 1990). To the best of our knowledge, the highest $K_{S\text{-}CH4}$ 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 CH_4 monooxygenase enzyme that catalyzes CH_4 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 CH_4 or DO, two limitation factors were defined, where β is the limitation factor for CH_4 (%) and γ is the limitation factor for DO (%):

$$0\% \le \beta = \left(1 - \frac{C_{CH4}}{K_{S-CH4} + C_{CH4}}\right) \cdot 100 \le 100\% \quad (2)$$

$$0\% \le \gamma = \left(1 - \frac{C_{O2}}{K_{S-O2} + C_{O2}}\right) \cdot 100 \le 100\% \quad (3)$$

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 ($r = r_{max}$). 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

Normality was assessed by the Shapiro-Wilk test. Since most of the data was non-normally distributed and with unequal samples number, significant differences among all parameters were determined using Kruskal-Wallis multiple comparison test (differences were considered significant at $p < 0.05$, $Z > 1.96$). To assess whether CH_4 was oxidized during the MO incubation tests, significant differences between C_{CH4} were determined by an analysis of variance (ANOVA; $p < 0.05$), after normality was assessed by the Shapiro-Wilk test.

~~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 ~~correlation between~~ the 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 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. In both seasons, no correlation between RWCS and lake depth was found, probably due to the fact that lakes were shallow and with an uneven depth distribution.

Lake water pH ranged from 5.9 to 8.2 in winter and 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 CH_4 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).

Fig. 3 shows the statistical distributions of the dissolved CH_4 and DO concentrations, as well as the ~~TK~~ Kruskal-Wallis comparisons. Significant differences were observed between yedoma and non-yedoma lakes ($p < 0.05$). In yedoma lakes, the CH_4 and DO concentrations were significantly higher and lower, respectively, than in non-yedoma lakes during both seasons (Kruskal-Wallis ~~TK~~ test, $p < 0.05$). In addition to differences related to permafrost type, higher CH_4 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. Fig. 4 shows three representative C_{CH4} 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_{CH4} 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; i.e. period of time with no apparent MO ~~was observed~~ (Fig. 4b) ~~before C_{CH4} began to decrease~~. This behavior, termed "induction of MO", has previously been reported for various soils (Bender and Conrad, 1995; Dunfield et al., 1999) and can be interpreted as an adaptation period of the CH_4 oxidizers to the culture conditions. 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_{CH4} after the lag phase. When no significant decrease in C_{CH4} was observed during the first seven days (Fig. 4c; ~~TK~~ ANOVA ~~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 mg L^{-1} d^{-1} during the winter and from 0.073 to 1.339 mg L^{-1} 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 (~~TK test or~~ Kruskal-Wallis ~~K-W~~ test, $p < 0.05$); specifically, r_{max} was 0.71 ± 0.36 and 0.29 ± 0.16 mg L^{-1} d^{-1} (mean ± SD) for yedoma and non-yedoma lakes, respectively. However, during the winter, no significant differences were 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 bacterial, algal and 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 organic matter decomposes in part to form CH_4 in surface lake sediments, whereas ancient yedoma carbon is progressively released largely introduced to lakes at greater depths to in the thaw bulb sediments beneath lakes and are made available to biological processes (Fig. 1)~~ Contemporary organic matter decomposes in part to form CH_4 in surface lake sediments, whereas ancient yedoma carbon is progressively released from thaw bulb beneath lakes to surface sediments (Heslop et al., 2015). Hence, organic carbon is made available to microbial decomposition in both shallow and deep sedimentary environments

(Fig. 1). 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.

In both yedoma and non-yedoma lakes, higher CH_4 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 CH_4 build-up in the water column (Phelps et al., 1998; Bastviken et al., 2004; Juutinen et al., 2009) and hindering ~~oxygen~~ O_2 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 CH_4 production was probably higher due to warmer sediments, ice was not a physical barrier to CH_4 exchange between the lake water and the atmosphere (Fig. 1).

Geographic variations were also observed with higher dissolved CH_4 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~~ proportion of yedoma lakes in central Alaska. No significant latitudinal trend was observed when yedoma and non-yedoma lakes were analyzed separately (~~TK~~ Kruskal-Wallis test, $p < 0.05$).

Fig. 3 shows that when relatively high CH_4 concentrations were found, relatively low DO concentrations were observed and conversely, when low dissolved CH_4 concentrations were found, higher DO concentrations were observed. This pattern was particularly clear in yedoma lakes: in winter, a CH_4 concentration of 7.32 ± 5.86 mg L^{-1} (mean ± SD) was found, while the DO concentration was 0.13 ± 0.03 mg L^{-1} (mean ± SD). In the same yedoma lakes, the summer CH_4 concentration was 0.49 ± 0.52 mg L^{-1} (mean ± SD), while the DO concentration was 3.19 ± 3.24 mg L^{-1} (mean ± SD). This observation suggests that MO was actively controlling O_2 and CH_4 concentrations by oxidizing CH_4 when O_2 was present. To

confirm the latter, it would be necessary to measure experimentally the O_2 uptake rate by methanotrophs and by other aerobic processes that compete with MO (Dzyuban, 2010).

The trend toward higher CH_4 concentrations and lower DO concentrations in winter than in summer was not as strong in non-yedoma lakes as in yedoma lakes (Fig. 4). 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 CH_4 in ebullition bubbles trapped by lake ice dissolves into the lake water column, leading to elevated dissolved CH_4 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~~ factors 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 mg L^{-1} d^{-1} during the winter and from 0.017 to 0.538 mg L^{-1} d^{-1} during the summer (Fig 6b). These values are within the range reported for arctic lakes of 0.001 to 1.000 mg L^{-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 and r_{max}. As mentioned before, although no difference in r_{max} was observed during winter between yedoma and non-yedoma lakes, r_{max} in yedoma lakes was about twice higher than in non-yedoma lakes during summer. We attribute that difference to a more active MO methanotrophic community in yedoma lakes, as all r_{max} tests were conducted in aerated vials with an initial standard CH_4 concentration in the liquid phase (~0.6 mg L^{-1}), thus ensuring optimal conditions. As observed with r_{max}, during summer yedoma lakes showed 2–3 times higher r than non-yedoma lakes (~~TK test or K-W~~Kruskal-Wallis test, $p < 0.05$; $r = 0.28 \pm 0.17$, mean ± SD, yedoma lakes; r = 0.09 ± 0.08 mg L^{-1} d^{-1}, mean ± SD, ~~-~~non-yedoma lakes). Higher r values for yedoma lakes in summer is ~~most likely linked to~~ explained by the higher dissolved CH_4 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~~ proportion 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, $K_{S\text{-}CH4}$ and $K_{S\text{-}O2}$. 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 $K_{S\text{-}CH4}$ 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 $K_{S\text{-}CH4}$ and $K_{S\text{-}O2}$ and calculated the resulting r (Eq. 1) using the experimental r_{max}, C_{CH4}, and C_{O2} measured in the 30 lakes. Fig. 6 shows the error on r caused by a given error on $K_{S\text{-}O2}$ (Fig. 6a) and $K_{S\text{-}CH4}$ (Fig. 6b), for yedoma and non yedoma lakes, in winter and in summer. According to this analysis, an underestimation of $K_{S\text{-}O2}$ or $K_{S\text{-}CH4}$ would lead to an overestimation of the actual MO rate (positive error), while an overestimation of these affinity constants would produce an underestimation of r (negative error). Fig. 6a shows that, an error on $K_{S\text{-}O2}$ ranging from -50% to 200%, would cause from 10% to -6% error on r, for all lakes and all seasons, except in yedoma lakes during winter, where an error from 75% to -50% would be generated. This relatively high sensitivity of r to error in $K_{S\text{-}O2}$ in yedoma lakes during winter is due to DO concentrations close to $K_{S\text{-}O2}$. Likewise, Fig. 6b shows that, from an error on $K_{S\text{-}CH4}$ ranging from -50% to 200%, a resulting error on r from 6% to -4% would be done, for all lakes and all seasons, except in non yedoma lakes during summer, where an error from 50% to -34% would be generated. As above, the latter is due to CH_4 concentrations close to $K_{S\text{-}CH4}$ in non yedoma lakes during summer. This sensitivity analysis shows that, other than for $K_{S\text{-}O2}$ in yedoma lakes during winter and $K_{S\text{-}CH4}$ in non-yedoma lakes during summer, errors on K_S would have relatively little impact on determination of methanotrophic rates

From Eq. 2 and 3, we estimated that, during the summer CH_4 was the main limiting ~~element~~ factor in 25 out of 30 lakes. In contrast, during winter, CH_4 was the main limiting ~~element~~ factor in 10 of 26 lakes (Table 2). Notably, during the winter, DO was the limiting ~~element~~ factor for all seven yedoma lakes, while during the summer, MO was limited by CH_4 for all non-yedoma lakes. A similar error analysis was done on β and γ, as done with r, to estimate if the estimated limiting factor would change as a result of error on K_S ranging from -50% to 200%. The results showed no impact on the limiting ~~element~~ factor 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 water 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.

5 Conclusions

We developed a new method based on a TDLAS for the determination of MO rates together with dissolved CH_4 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 mg L^{-1} d^{-1} in winter and summer. MO rates in water of Alaskan lakes showed high seasonal and geographic variability. In addition to temperature effects, the main factors controlling MO were: 1) CH_4 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 CH_4 emission predicted by permafrost thawing (Khvorostyanov, et al. 2008; Walter Anthony et al., 2014).

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Tables

Table 1. Identification, location, and permafrost soil type for lakes included in the study. *Indicates informal lake name, yedoma lakes are marked on light grey..

#	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	Augustine 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 - Bellow detection limit, yedoma lakes are marked on light grey..

#	CH_4 (mg L^{-1})		O_2 (mg L^{-1})		Potential MO (mg L^{-1} d^{-1})		Actual MO (mg L^{-1} d^{-1})		Limiting ~~element~~factor	
	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	0.12	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^*

Figure captions

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.

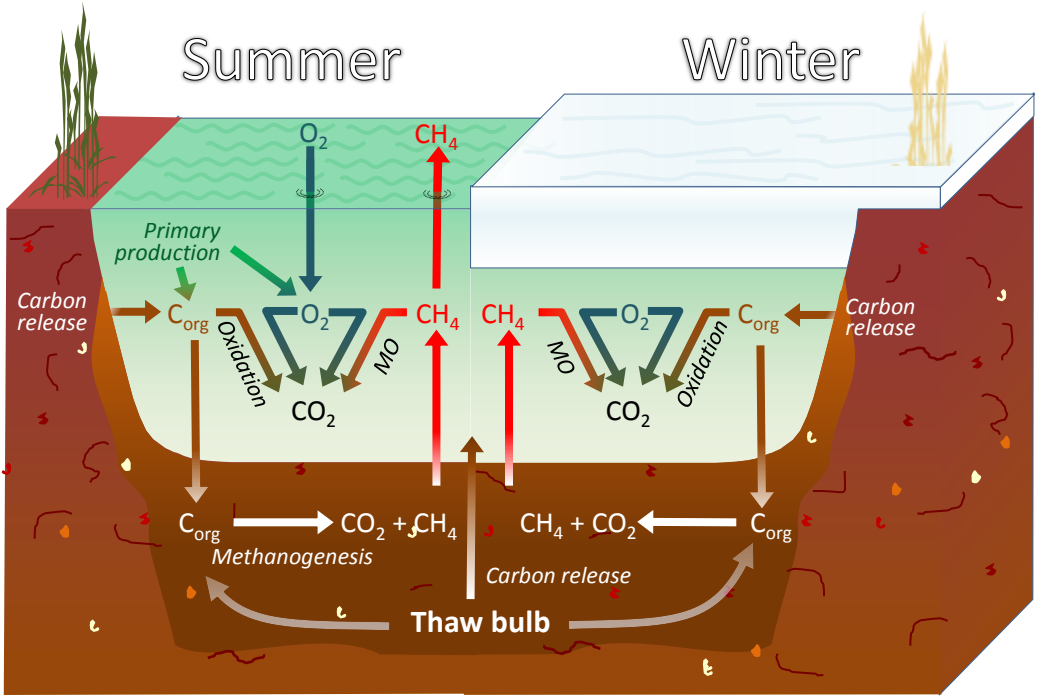
Figure 2. Locations of studied Alaskan lakes (white circles) plotted on the Alaska DEM hillshade raster. Information about the distribution of yedoma-type deposits (Pleistocene-aged, ice-rich silt containing deep thermokarst lakes) was from Jorgenson et al. (2008) and Kanevskiy et al. (2011). The Alaska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

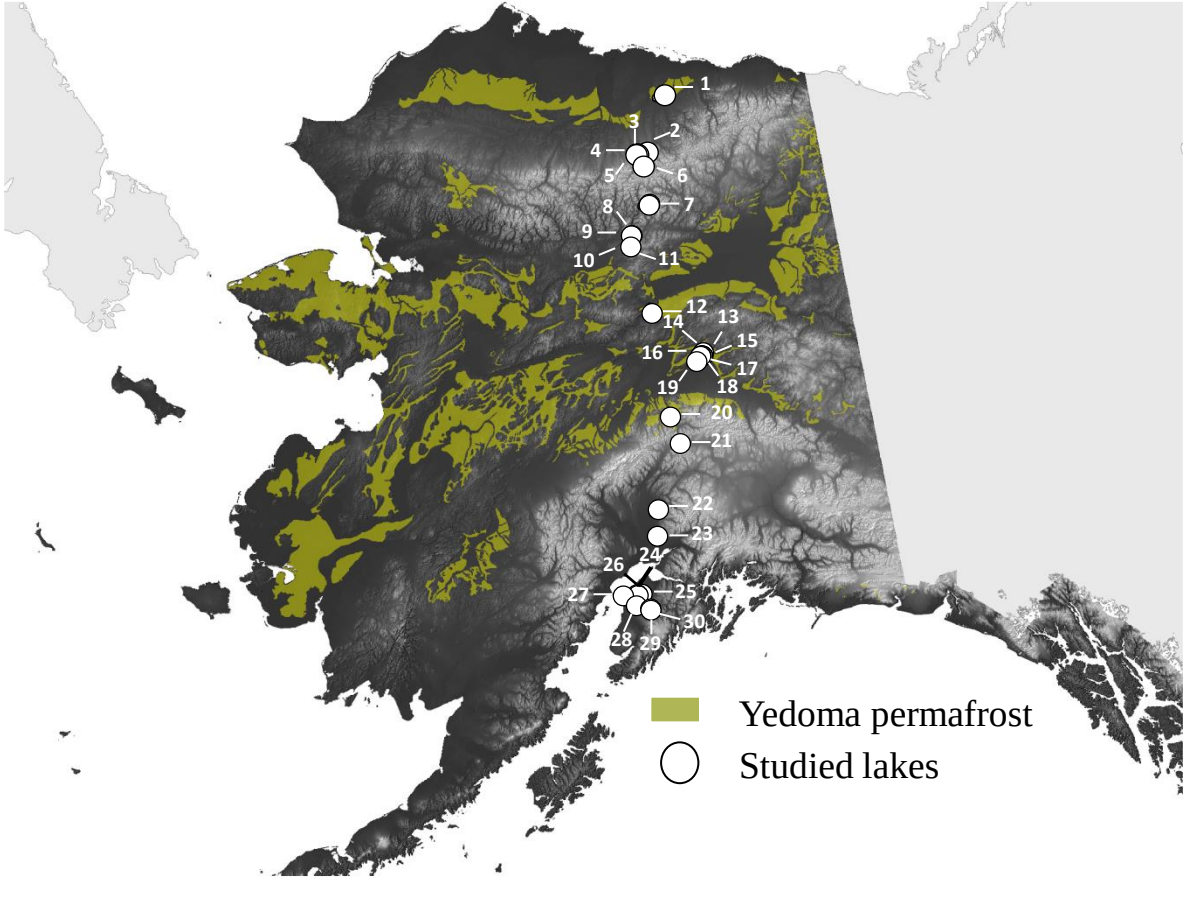
Figure 3. Statistical distributions of CH_4 (white boxes) and DO (grey boxes) water concentrations in yedoma and non-yedoma lakes during the winter and summer. The boxes include the median (Q2) and the quartile range (Q1 and Q3). The whiskers show minimum and maximum data. The open circles show outlier data. Capital letters are Kruskal-Wallis multiple comparison test~~Tukey–Kramer test results~~; values with the same capital letter are not significantly different ($p < 0.05$, $Z > 1.96$~~$p < 0.05$~~). *n* represents the number of lakes measured.

Figure 4. Examples of CH_4 oxidation patterns observed during the MO assays: (a) assay with no lag-phase. (b) assay with a 3-day lag-phase. and (c) assay with no detected activity. Straight lines are linear correlations.

Figure 5. (a) CH_4 oxidation potential (r_{max}) and (b) CH_4 oxidation rates observed in 30 lakes along a north-south transect (left-right) in Alaska during the summer (white bars) and the winter (black bars).

Figure 6. Sensitivity analysis of the impact of an error or variation in $K_{S\text{-}O2}$ (a.) and $K_{S\text{-}CH4}$ (b.) on r; yedoma lakes in winter (—), yedoma lakes in summer (— · — ·), non-yedoma lakes in winter (— ·· — ··) and non-yedoma lakes in summer (-----).


Summer
Winter
O_2
CH_4
Primary production
Carbon release
C_{org}
Oxidation
O_2
MO
CH_4
CO_2
CH_4
MO
O_2
Oxidation
C_{org}
Carbon release
C_{org}
$CO_2 + CH_4$
Methanogenesis
$CH_4 + CO_2$
C_{org}
Carbon release
Thaw bulb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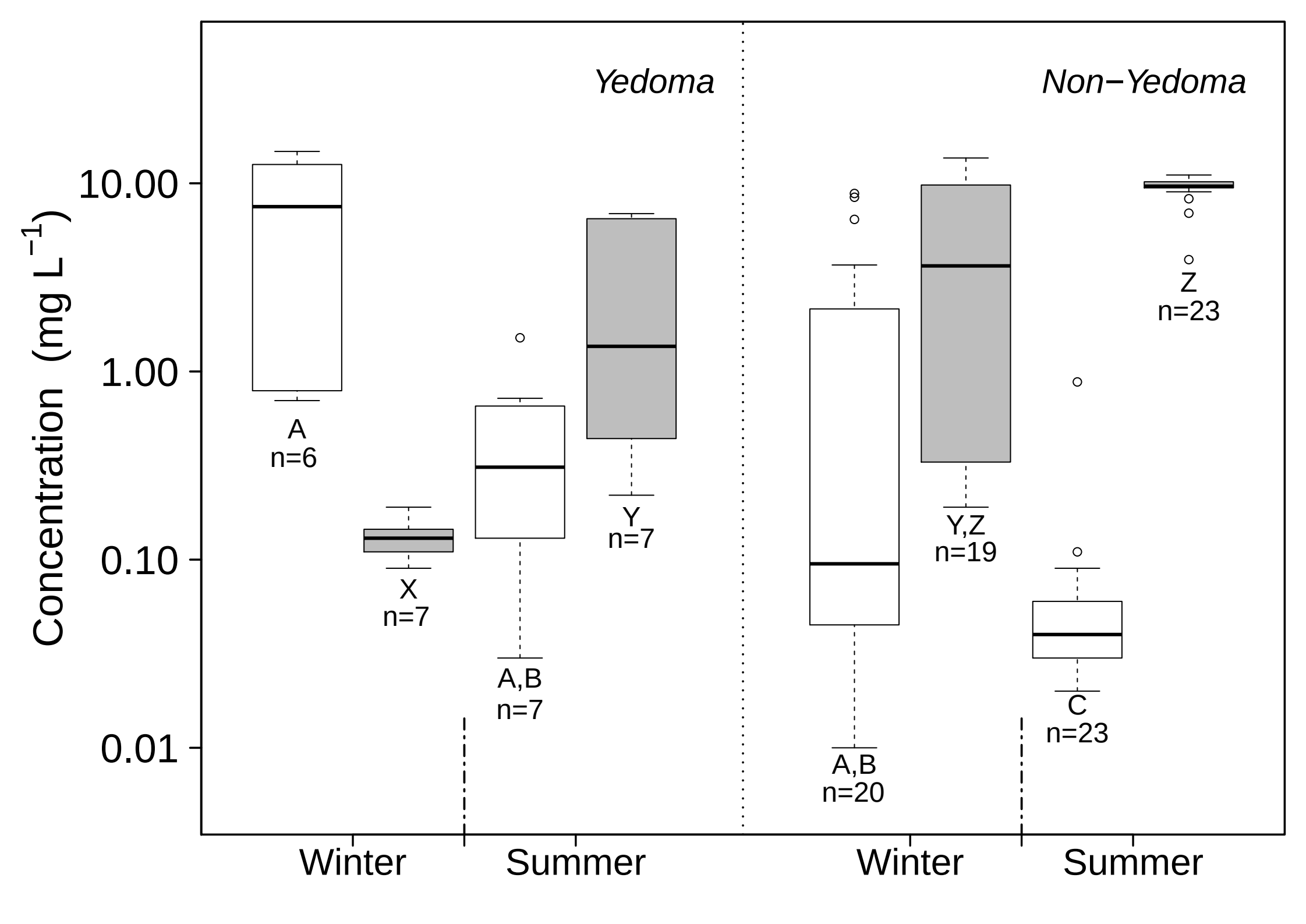
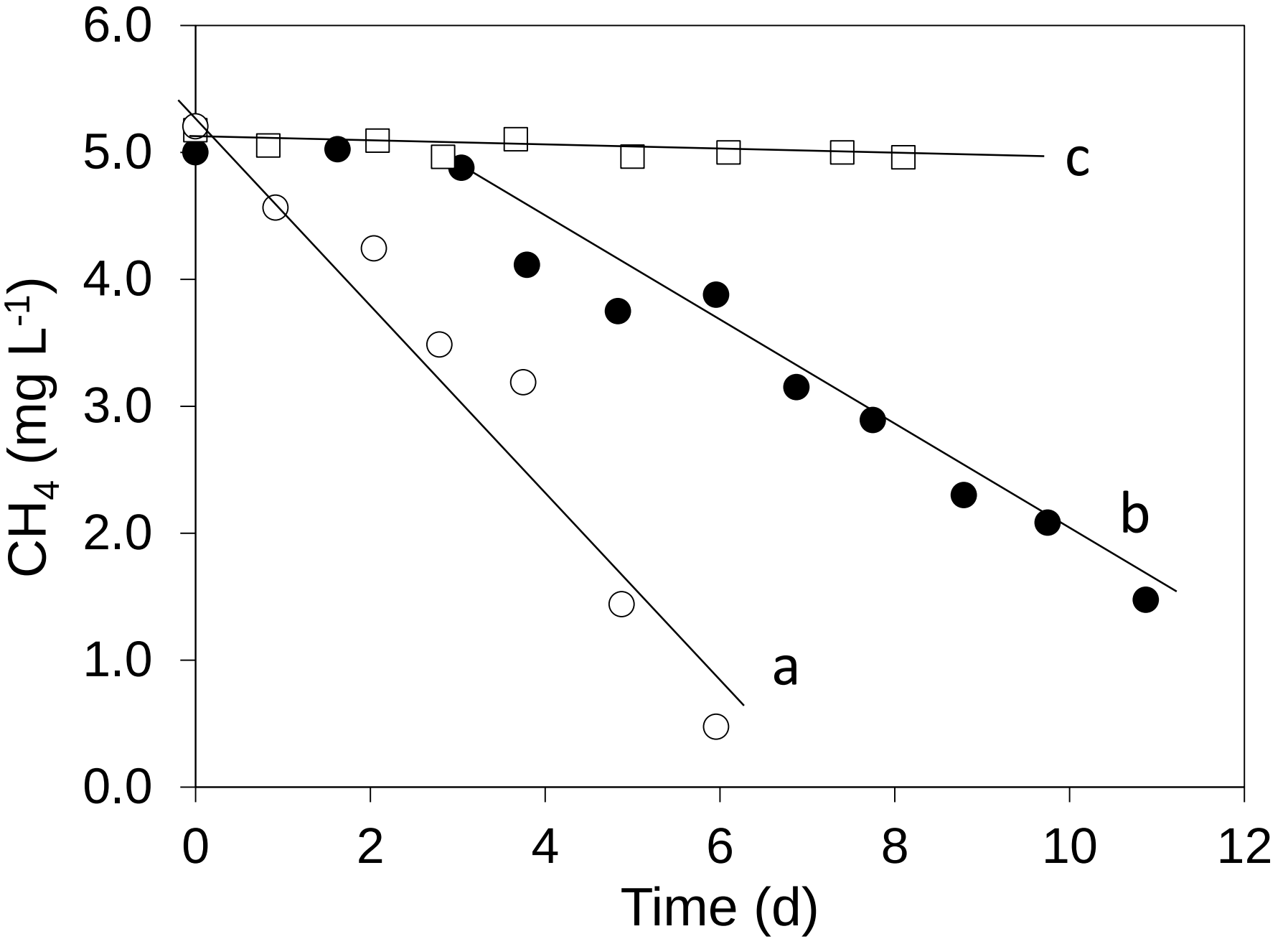
Yedoma permafrost
Studied lakes


Yedoma
Non−Yedoma
Concentration (mg L−1)
10.00
1.00
0.10
0.01
A
n=6
X
n=7
A,B
n=7
Y
n=7
A,B
n=20
Y,Z
n=19
C
n=23
Z
n=23
Winter
Summer
Winter
Summer


6.0
5.0
4.0
3.0
2.0
1.0
0.0
CH_4 (mg L^{-1})
0
2
4
6
8
10
12
Time (d)
a
b
c

a

r_{max} (mg CH_4 L^{-1} d^{-1})

1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0

1 3 5 7 9 11 13 15 17 19 21 23 25 27 29

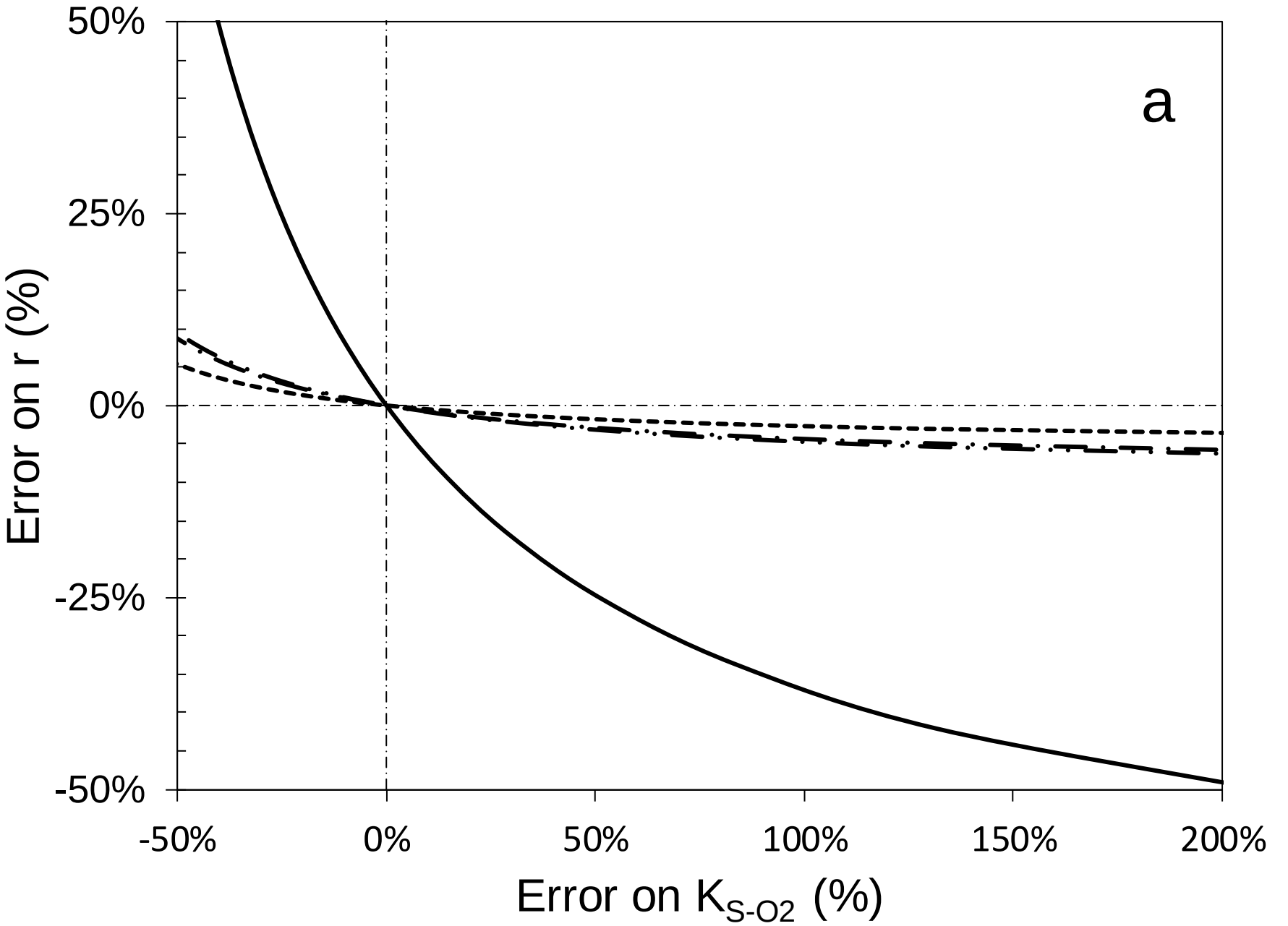
b

r (mg CH_4 L^{-1} d^{-1})

1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0

1 3 5 7 9 11 13 15 17 19 21 23 25 27 29

Lake #


a
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Error on r (%)
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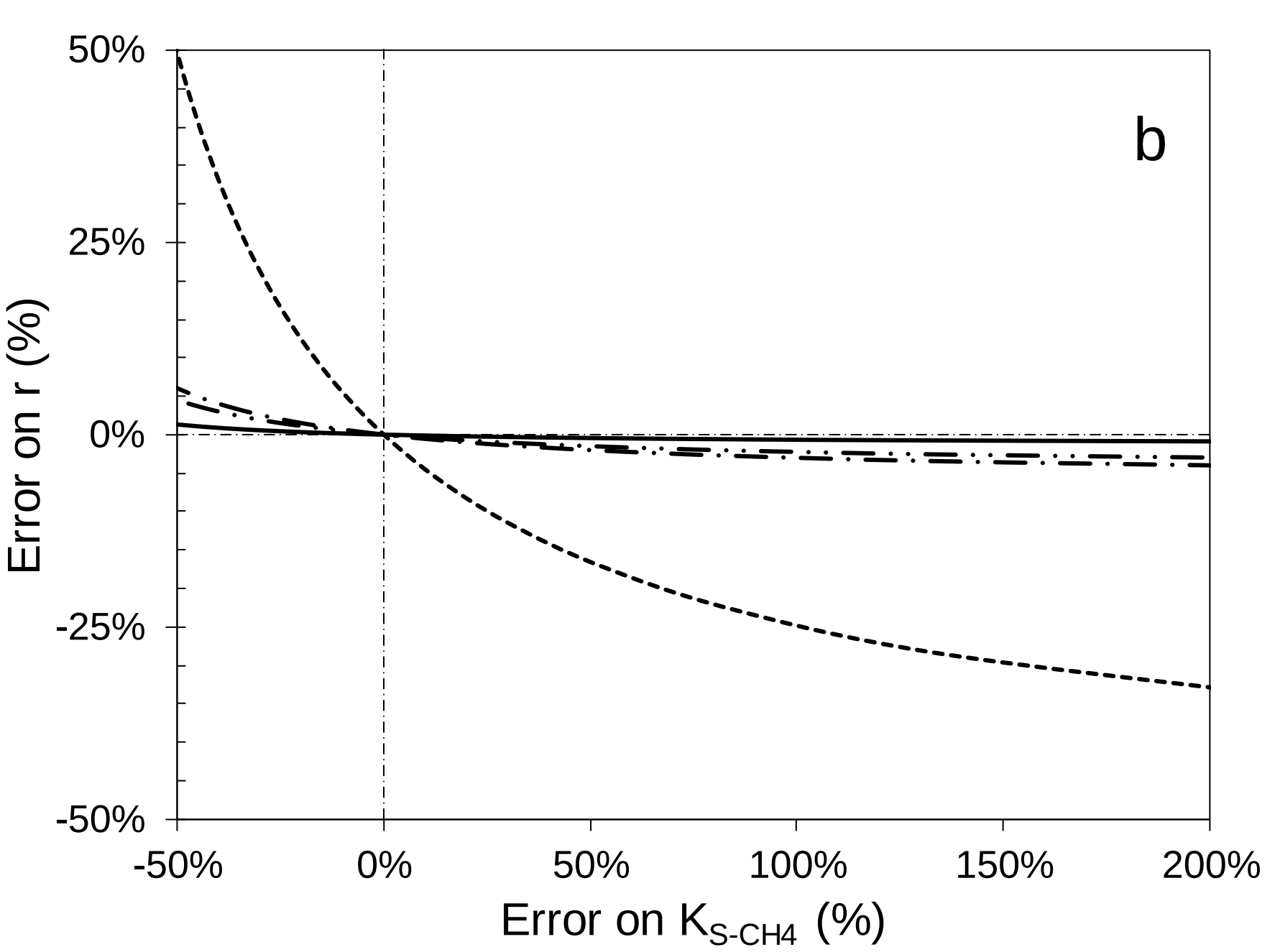
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100%
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200%
Error on K_{S-CH4} (%)