Dr. Isabelle Laurion
Editor
Biogeosciences

RE: Comments on bg-2015-49

July 15, 2015

Dear Dr. 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 revised the manuscript, taking into account all your concerns and recommendations.

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 *Biogeosciences*.

Sincerely,

Frédéric Thalasso

Comment 1 (P2, L1-3): revise english;
I suggest something like: "… into the lake, confirming the strong coupling between terrestrial and aquatic habitats and its influence on CH4 cycling."

Our answer: We agree and we have modified that section accordingly; "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, confirming the strong coupling between terrestrial and aquatic habitats and its influence on CH$_4$ cycling.

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Comment 2 (P2, L5-7): Sounds unrealistic to have 6% from northern lakes while global is 6-16%. These global estimates are approximations. Considering the variability you observed, and the complexity of methane cycling, do you really consider that providing these numbers are useful?

Original text related; 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).

Our answer: This sentence was probably unclear and it is correct that methane emission data are approximations. However, we would like to introduce our manuscript with one general sentence on the magnitude of CH$_4$ emission. We substituted the first two sentences by the following one: "Northern lakes are an important source of atmospheric CH$_4$ (Bastviken et al. 2011), and it has been estimated that they are responsible for as much as 6% of global emissions to the atmosphere (Walter et al., 2007)".

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Comment 3 (P2, L10-15): Is this really considering ebullition?!

Original text related; "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; Bellido et al., 2011)."

Our answer: Thanks for this useful comment. Yes, ebullition was considered in the first sentence. Please note that, as indicated in the references and in that sentence, this percentage is "of total CH$_4$ produced in freshwater ecosystems". However, after revisiting the cited literature here, the works of Kankaala *et al.* and Bellido *et al.*, did not consider ebullition. We modified that sentence as follows; "Likewise, MO plays an important role in northern lakes specifically by oxidizing up to 88% of the CH$_4$ diffusing through the water column (Kankaala et al., 2006, 2007; Bellido et al., 2011)".

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Comment 4 (P2, L16-20): what fraction of oxidized CH4 is converted to uper food web C compared to the fraction converted to CO2 and released? I think we need to read about some semi-quantitative estimations otherwise it's difficult to state that it's "significant".

Original text related; "MO is therefore a pathway that reincorporates a 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".

Our answer: Yes indeed, we agree that this section was unclear and that it was potentially misleading. We have modified that section as follows; "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. In addition to CH_4 respiration and conversion to CO_2, MO is also a mechanism for reincorporation of a fraction of the CH_4-C produced into the biogeochemical carbon cycle within lakes.

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Comment 5 (P2, L25-26): thermal structure is not mentioned nor expressed in Fig. 1, but I think it's fundamental to these processes.

Our answer: The objective of Figure 1 was to show the flowchart of CH_4 cycling in northern lakes, with contrasting seasons. On our point of view, the thermal structure does not modify that chart while it certainly does modify the relative importance of each process. Beside, arctic lakes are usually dimictic, with mixing and stratification episodes that would be hard to represent in the Figure. We request to keep that Figure in its present form.

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Comment 6 (P3, L6-7): what about organic content? too variable to underline here?

Text related: Non-yedoma permafrost is characterized by thinner carbon- and ice-rich horizons and have a more widespread distribution (Ping et al., 2008; Tarnocai et al., 2009; Hugelius et al. 2014).

Our answer: The organic carbon-rich horizons of non-yedoma soils are typically much thinner than in yedoma soils. The cited literature explains that in detail. To improve clarity, we revised the sentence to include mention that carbon-rich layers are also thinner in non-yedoma soils; "Non-yedoma permafrost has a more widespread distribution than yedoma permafrost and is also characterized by a much thinner carbon-rich horizon, variable in composition (Ping et al., 2008; Tarnocai et al., 2009; Hugelius et al. 2014)".

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Comment 7 (P3, L12-13): was this study comparing arctic communities under a range of T including low T (4degC and below), otherwise the sentence could be toned down ("… MO was reported to occur more efficiently at mesophilic T…")

Text related: "MO occurs more efficiently at mesophilic temperatures, from 20 to 35 °C (Semrau et al., 2010)".

Our answer: Semrau et al. (2010) is a general review, in such manner that we accept the suggestion to tone down that sentence as follows; "==MO was reported to occur more efficiently at mesophilic temperature==".

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Comment 8 (P4, L14): which sensors? you mean pH and DO were calibrated weakly or T and Chla also? (I doubt Chla; was it ever calibrated?)

Our answer: Thank you, this was a mistake, we did calibrate pH and DO, only. We modified the sentence accordingly: "The ==pH and DO sensors of the Hydrolab were calibrated== regularly, before and after each section of the latitudinal lake transect (four sections per transect, approximately one calibration per week). ==Temperature and chlorophyll a sensors were not regularly calibrated=="

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Comment 9 (P5, L9): I assume this 10 sec has been tested as sufficient to reach equilibrium? And can you also mention the ratio of water to headspace volumes? (same as below for MO?)

Text related; "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)".

Our answer: Yes indeed, equilibration time has been tested and reported before. We agree with both comments and we modified that section as follows; "This method consisted ==of creating a gas/liquid equilibrium in a 100 mL equilibration vial containing 60 mL of lake water sample and 40 mL of headspace (air) by vigorous shaking for 10 s. As previously reported (Sepulveda-Jauregui et al., 2012), this shaking time was enough to reach equilibrium and allowed the non-invasive determination of CH_4 concentration in the water sample by measuring headspace concentration with a laser beam crossing the headspace of the equilibration vial, through the glass walls of the 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",

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Comment 10 (P6, L8-9): does it mean it reads through the glass wall of the vial? (the non-invasive indicated below?)

Text related; "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.".

Our answer: Yes indeed, this non-invasive measurement is based on the laser beam crossing the headspace, through the glass walls of the vial. In response to this comment, we revised the manuscript to include a more detailed description the first time we mentioned the method; i.e. P5, L9 (see previous comment).

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Comment 11 (P8, L3-5): does it mean thermistor chains were installed? (not mentionned in Method section if I recall well). Otherwise it means this average is obtained from discrete profiles. Readers can wonder where do the 103 or 235 data come from.

Text related; "The mean temperature throughout the lake water columns was 2.4 ± 0.6 °C (mean $\pm$ SD, n = 103) in the winter and 13.9 ± 2.4 °C (mean $\pm$ SD, n = 235) in the summer".

Our answer: No, as indicated in the Material and Methods section (P4, L14), temperature profiles were determined from discrete measurements at all depths, with the Hydrolab probe. We modified that sentence as follows; "The mean temperature, measured at all depth throughout the lake water columns with the Hydrolab probe was 2.4 ± 0.6 °C (mean $\pm$ SD, n = 103) in the winter and 13.9 ± 2.4 °C (mean $\pm$ SD, n = 235) in the summer"

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Comment 12 (P8, L7-8): no winter temperature inversion?

Text related: During the winter, 16 of 18 lakes were fully mixed, while two lakes were partially stratified and none was fully stratified.

Our answer: Yes indeed, we observed temperature inversion in 15 of 18 lakes. We modified this sentences as follows; "During the winter, 16 of 18 lakes for which a complete temperature profile was determined were fully mixed, while two lakes were partially stratified and none was fully stratified. We observed temperature inversion in 15 of the 18 lakes, with temperature gradients ranging from -0.1 to -3.4 °C, with an average of -1.4 °C (top minus bottom temperature)".

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Comment 13 (P8, L13-14): if this was not calibrated and come from the manufacturer calibration, companies are usually very optimistic about their detection limit…

Text related: 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}).

Our answer: This is correct, we didn't calibrate the chlorophyll a sensor. We modified this sentence as follows; "Chlorophyll *a* was only detected during the summer, ranging from 1.0 to 45.9 µg L^{-1} (manufacturer detection limit, 0.03 µg L^{-1}).

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Comment 14 (P8, L15): precise if in mgC or mg CH4; I assume of CH4 but doubt remains as often people use mgC to express GHG concentrations

Text related: 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).

Our answer: This is correct; all CH_4 concentrations are expressed in CH_4 units, not carbon units. To avoid doubt without tedious repetitions, we included a new sentence in the Material and Methods section (P5, L15); "In the present work, all CH_4 concentrations are expressed in mg CH_4 L^{-1}.

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Comment 15 (P10, L15): thaw bulb is beneath the lakes but I do not understand "to surface sediments"; do you talk about the transfer? or the location where CH4 is produced?

Test related; "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)".

Our answer: We agree that this sentence was unclear. To improve clarity, we modified the sentence as follows; "Contemporary organic matter decomposes in part to form CH_4 in surface lake sediments. In contrast, ancient yedoma carbon is decomposed throughout the sediment profile (Fig. 1), with particularly high rates of methanogenesis occurring along the permafrost thaw front, located deep in the thaw bulb beneath the lake (Heslop et al., 2015). Methane produced at depth in the thaw bulb subsequently migrates, primarily as free-phase bubbles through bubble tubes in sediments, to the surface sediments where it escapes the lake via ebullition (Walter Anthony and Anthony 2013; Tan et al. 2015). ".

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Comment 16 (P10, L17-20): redundant with previous sentence above (underlined in green above), and with the following sentence.

Text related; "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".

Our answer: We agree this section was improvable in a more direct form. We modified it as follows; "Thus higher organic carbon and nutrient inputs in yedoma-type lakes promote higher anaerobic metabolism in the sediments together with aerobic metabolism in the water column, leading to higher CH_4 and lower DO concentrations in the water column".

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Comment 17 (P10, L32 – P11, L2): It sounds strange; maybe something like "it did not cumulate in the water column as much as in winter and was released to the atmosphere"

Text related: "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)".

Our answer: We modified this sentences accordingly; "In summer, although CH_4 production in surface sediments was probably higher due to warmer conditions, CH_4 did not cumulate in the water column and was released to the atmosphere (Fig. 1)".

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Comment 18 (P11, L10-11): superfluous

Our answer: We removed the marked text and the sentence is now as follows; "Fig. 3 shows that when relatively high CH_4 concentrations were found, relatively low DO concentrations were observed and conversely".

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Comment 19 (P11, L15-16): is this the reverse: O2 controls microbial assemblages?

Text related: "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)".

Our answer: In a biological process, the presence of a substrate shapes the microbial community while the microbial community depletes the available substrate. We would prefer to keep the description at a "process level" rather than in terms of microbial assemblage. Please note that the microbial assemblage is briefly mentioned (see answer to comment 22). To keep more neutral, we modified that sentence as follows; "This observation suggests that MO was a significant contributor to the biogeochemical processes, 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)".

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Comment 20 (P11, L19-22): is this really a causal link? Can you be more explicit?

Text related: "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".

Our answer: We modified that sentence to avoid unclear linkage. "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 in non-yedoma lakes, the lower organic carbon inputs fuels more weakly methanogenesis and aerobic processes, including MO than in yedoma lakes, resulting in a lower seasonal variation of CH_4 and DO concentration".

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Comment 21 (P11, L25): in spring? otherwise, if you talk about winter CH4 produced, it will not be trapped by lake ice if dissolving fraction

Text related: "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".

Our answer: To improve clarity, we modified the sentence as follows; "Even during winter, Greene et al. (2014) found that 80% of CH_4 in ebullition bubbles trapped under the ice cover dissolves into the lake water column before being confined within the growing ice sheet, leading to elevated dissolved CH_4 beneath the ice".

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Comment 22 (P11, L26-28): microbial communities are shaped by limnological conditions...

Text related: "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".

Our answer: We agree that this sentence was misleading because it gave the idea that the microbial community is an independent variable, which it is certainly not. We removed the sentence.

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Comment 23 (P11, 30): this is not clear cf the beginning of sentence; rewrite.

Text related: "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)".

Our answer: To improve clarity, we modified the sentence as follows; "The actual MO rates r ranged 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). As expected, these actual MO rates were significantly lower than the potential MO rates".

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Comment 24 (P12, L1): is this number really that precise?

Our answer: No, we modified the sentence as follows; "These r values are within the range reported for arctic lakes, which ranges over 3 orders of magnitude, from 0.001 to 1 mg L^{-1} d^{-1} (Liikanen et al., 2002; Kankaala et al., 2006; Lofton et al., 2014)".

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Comment 25 (P12, L9-12): equal, even, equivalent, similar?

Test related: "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".

Our answer: As we want to indicate that all conditions were similar and that there was no DO and CH_4 limiting conditions, we changed that sentence as follow. "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 similar and non-limiting conditions".

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Comment 26 (P12, L18-19): this is becoming redundant...

Related text: "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 proportion of yedoma lakes in central Alaska. No significant latitudinal trend in MO was observed when yedoma and non-yedoma lakes were analyzed separately".

Our answer: We agree and we removed the two first sentences, keeping the last one, indicating no latitudinal trend in MO, as follows; "As observed with CH_4 and DO concentrations, no significant latitudinal trend in MO was observed, when yedoma and non-yedoma lakes were analyzed separately".

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Comment 27 (P12, L27-29): what does it mean??

Related text: "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".

Our answer: In this section, we are referring to the standard error associated to experimental determination. We rephrased it as follows; "The determination of MO rates may, therefore, be subject to large error if reported values are used instead of experimental

parameters <mark>or because of standard error associated to experimental K_S determination (Kovarova-Kovar and Egli, 1998)</mark>".

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Comment 28 (P13, L11-14): explicitely discuss the consequence of making larger errors in the case of Yedoma-winter and non-Yedoma-summer

Text related "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"

Our answer: In our point of view, the consequence of an error would appear clearly only if a quantification of MO in the context of global CH_4 cycling is presented. This is a step we decided not to take, for several reasons (please see our answer to comment 31). This is the reason why we decided not to go further than a sensitivity analysis, the objective of which is to estimate the uncertainty of our estimations for an unknown error on K_S. We addresses very briefly this comment as follows: "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. <mark>The potentially significant error on methanotrophic rates in yedoma lakes during winter and in non-yedoma lakes during summer encourages further studies including experimental K_S determination.</mark>"

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Comment 29 (P13, L18): this reversing of subject compared to the rest of sentence brings confusion.

Text related: "Notably, during the winter, DO was the limiting factor for all seven yedoma lakes, while during the summer, MO was limited by CH_4 for all non-yedoma lakes".

Our answer: This sentence was changed accordingly; "Notably, during the winter, DO was the limiting factor <mark>of MO for all seven yedoma lakes, while during the summer, CH_4 was the limiting factor for all non-yedoma lakes</mark>".

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Comment 30 (P13, L24): this word not necessary as your point is about the fact that you sampled a single depth, not that you took duplicates.

Our answer: We removed the word "duplicate".

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Comment 31 (P14, L17-19): I think this sentence would be stronger if above you demonstrate above in discussion how the estimated MO rates can deplete CH4 and to what degree

Text related: "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)".

Our answer: This is an important comment. During the preparation of our manuscript, we submitted to several colleagues the important question if the MO rates determined in the present work could be used to quantify MO in the context of global CH_4 cycling. Specifically our question was to quantify the percentage of CH_4 produced in lakes that is oxidized. That suggestion raised several criticisms, mostly because, as mentioned in our manuscript, samples were taken at a single depth. The reviewer is requesting that we use our data to go further in our description of methane cycling, a direction we feel would not be justified based on the limitations of sampling at a single depth.

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 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 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~~ confirming the strong coupling between terrestrial and aquatic habitats and its influence on 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).~~ Northern lakes are an important source of atmospheric CH_4 (Bastviken et al. 2011), and it has been estimated that they are responsible for as much as 6% of global emission to the atmosphere (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 diffusing through the water column (Kankaala et al., 2006, 2007; Bellido et al., 2011). ~~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. In addition to CH_4 respiration and conversion to CO_2, MO is therefore a pathway that reincorporates a fraction of the CH_4-C produced into the biogeochemical carbon cycle within lakes.

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,

1 1998b; Bastviken et al., 2002; Liikanen et al., 2002; Carini et al., 2005; Schubert et al., 2010).

2 In turn, CH_4 and O_2 concentrations depend on numerous other processes involved in

3 biogeochemical carbon cycling (Fig. 1). Among these, the most important are methanogenesis

4 producing CH_4, primary production and atmospheric diffusion supplying O_2, and several

5 aerobic metabolic processes that compete with MO for available O_2 (Dzyuban, 2010).

6 In addition to autochthonous and allochthonous carbon inputs to lakes, permafrost thaw can

7 provide an additional source of labile organic carbon to fuel methanogenesis and carbon

8 mineralization in thermokarst (thaw) lakes (Zimov et al., 1997; Walter et al., 2006). MO in

9 northern regions is therefore directly and indirectly linked to permafrost type and landscape

10 processes that are highly variable. Permafrost ranges from sporadic to continuous and is also

11 variable in composition (Jorgenson et al., 2008). Yedoma-type permafrost is an organic-rich

12 (about 2% carbon by mass) Pleistocene-age permafrost with ice content of 50–90% by

13 volume (Zimov et al., 2006), which occurs mainly in the previously unglaciated regions of

14 Siberia, Alaska, and NW Canada (Czudek and Demek, 1970; Walter et al., 2007; Kanevskiy

15 et al., 2011; Grosse et al., 2013). ~~Non-yedoma permafrost is characterized by thinner ice-rich~~

16 ~~horizons and have a more widespread distribution~~ Non-yedoma permafrost has a more

17 widespread distribution than yedoma permafrost and is also characterized by a much thinner

18 carbon-rich horizon, variable in composition (Ping et al., 2008; Tarnocai et al., 2009;

19 Hugelius et al. 2014).

20 Many northern lakes are located in continental climate zones, subject to contrasting seasonal

21 conditions with long, cold winters followed by relatively short, but warm summers. Although

22 psychrotolerant and psychrophilic methanotrophs have been reported (Omelchenko et al.,

23 1993, 1996; Bowman et al., 1997; Trotsenko and Khmelenina, 2002), MO was reported to

24 occur more efficiently at mesophilic temperature ~~MO occurs more efficiently at mesophilic~~

25 ~~temperatures, from 20 to 35 °C~~ (Semrau et al., 2010). During winter, northern lakes are

26 covered by a thick ice layer for seven to nine months. Surface lake ice impedes oxygen

27 transfer from the atmosphere to the lake and, when snow-covered, substantially reduces light

28 penetration and oxygen production by photosynthesis (White et al., 2008; Clilverd et al.,

29 2009). Thus, the combination of low temperature and limited oxygen availability suggests

30 lower MO rates in northern lakes in winter than in summer.

31 Given the number of parameters having a potential effect on MO, as well as the patchwork of

32 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 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 pH and DO sensors of the Hydrolab were calibrated The Hydrolab was calibrated regularly, before and after each section of the latitudinal lake transect (four sections per transect, approximately one calibration per week). Temperature and chlorophyll *a* sensors were not regularly calibrated. 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

1 shallower than 1 m, we measured Hydrolab parameters at three distributed depths throughout

2 the lake water column.

3 Water samples for MO rates and dissolved CH_4 concentration were taken at a depth of within

4 1 m of the ice-water interface in winter and usually at 0.75 to 1 m water depth in summer.

5 Due to differences in lake depth and thickness of the ice sheets, samples reflected surface

6 water in deep lakes, but mid water column or even lake bottom water environment, in shallow

7 lakes. Samples were taken with a horizontal Van Dorn bottle (Wildco, Yulee, FL, USA).

8 Water density derived from surface and bottom water temperatures were used to determine

9 the relative water column stability (RWCS; Padisak et al., 2003). Lakes with RWCS >56.5

10 were considered fully stratified, lakes with RWCS <16.3 were considered fully mixed, and

11 lakes with intermediate RWCS were considered partially stratified (Branco et al., 2009).

12 Similarly, we determined whether an oxycline was present in each lake based on a sharp DO

13 gradient or presence of an oxic/anoxic interface. Detailed temperature and DO profiles are

14 available as a Supplement in Sepulveda-Jauregui et al. (2014). We report all results in mean ±

15 standard deviation (SD).

16 **2.3   Dissolved CH_4 concentration and MO rate**

17 To avoid long delays in sample transfer from remote locations to the laboratory, we

18 determined dissolved CH_4 concentrations with a previously described method based on

19 Headspace Equilibration using Infrared Tunable Diode Laser Absorption Spectroscopy (HE-

20 TDLAS; Sepulveda-Jauregui et al., 2012). This method consisted of creating a gas/liquid

21 equilibrium in a 100 mL equilibration vial containing 60 mL of lake water sample and 40 mL

22 of headspace (air) by vigorous shaking for 10 s. As previously reported (Sepulveda-Jauregui

23 et al., 2012), this shaking time was enough to reach equilibrium and allowed the non-invasive

24 determination of CH_4 concentration in the water sample by measuring headspace

25 concentration with a laser beam crossing the headspace of the equilibration vial, through the

26 glass walls of the vial. This measurement was conducted with a modified open-field CH_4

27 analyzer (GasFinder 2; Boreal Laser, Edmonton, Canada). The CH_4 concentration in the water

28 sample was calculated from the measured headspace concentration according to Henry's law.

29 In the present work, all CH_4 concentrations are expressed in mg CH_4 L^{-1}.

30 This method consisted of determining the CH_4 concentration in the headspace of an

31 equilibration vial containing a known volume of lake water and in which gas/liquid

7   We determined duplicate MO rates in one water sample from each lake taken as described

8   above, using a modified HE-TDLAS method to allow for measurement of MO in the field.

9   This new method was based on a previous development using the HE-TDLAS method for the

10 determination of methanogenic activity (Martinez-Cruz et al., 2012). Two 60-mL lake water

11 subsamples from a single Van Dorn bottle sample were gently transferred to two 100-mL

12 equilibration vials (duplicates). Equilibration vials were immediately closed with rubber

13 stoppers and vigorously shaken for 10 s to transfer most of the dissolved CH$_4$ contained in the

14 water sample to the headspace. Next, the headspace was vented, the vial was closed, and the

15 sample was shaken again to evacuate the residual CH$_4$ content of the water sample. Using this

16 procedure, more than 99.5% of the original CH$_4$ content of the sample was evacuated. The

17 equilibration vials were then closed with rubber stoppers and aluminum crimp caps, spiked

18 with 0.6 mL CH$_4$ (99.0% purity; Air Liquide, Houston, TX, USA) injected with a disposable

19 syringe, and vigorously shaken for 10 s. This approach allowed MO tests to be conducted

20 with an initial standard CH$_4$ concentration in the liquid phase (~0.6 mg L^{-1}). It also provided

21 an initial CH$_4$ to O$_2$ molar ratio of 0.062, significantly below the stoichiometric ratio (0.5),

22 ensuring no O$_2$ limitation.

23 Equilibration vials were incubated for 10–12 days in a water bath inside insulated boxes

24 placed in our vehicle. In the winter, the vials were maintained at 2 ± 2 °C in a water bath with

25 ice supplements; in the summer, the vials were maintained at 15 ± 2 °C. The temperature of

26 the water bath was measured daily. We measured the CH$_4$ concentration in the equilibration

27 vials daily using the HE-TDLAS method described in detail by Sepulveda-Jauregui et al.

28 (2012). Briefly, dry control MO test vials containing only CH$_4$ standards were read by the

29 TDLAS for calibration. Each experimental equilibration vial was vigorously shaken for 10 s

30 to reach phase equilibrium and then immediately placed in the laser beam path, after which a

31 stable HE-TDLAS reading was typically observed within 5 s. Five readings were taken for

32 each MO test vial and recalibration was conducted after measuring each set of test vials to

1 ensure instrument stability. The field HE-TDLAS method allowed measurement of dissolved

2 CH4 and MO rates. This technique was simple, rapid (about 60 s per sample measurement),

3 non-invasive, and avoided complications and long delays in sample transfer from remote

4 locations to the laboratory.

5 We calculated the total CH4 concentration (C_{CH4} = total CH4 mass present in the gas and

6 liquid phases divided by the sample liquid volume) in each vial during the MO tests. MO rates

7 were determined from the decrease in C_{CH4} in the equilibration vials with time. MO rates

8 determined by this method represent the MO rate after aeration and CH4 addition (vials spiked

9 with CH4 and vigorously shaken). Thus, these MO rates do not correspond to actual

10 observations of in situ DO and dissolved CH4 concentrations in the lakes. The measured CH4

11 oxidation rate was considered the potential MO (r_{max}; mg CH4 L^{-1} d^{-1}) under non-limiting CH4

12 and DO concentrations. To estimate the actual rate (r; mg CH4 L^{-1} d^{-1}) from r_{max}, a double

13 Monod model was used (Bae and Rittmann, 1996; Segers, 1998) in which C_{CH4} and C_{O2}

14 represent the actual dissolved CH4 and DO concentrations measured in the lake, respectively,

15 and K_{S-CH4} and K_{S-O2} are the apparent affinity constants of the methanotrophic community, for

16 CH4 and DO, respectively:

17 $$r = r_{max} \cdot \frac{C_{CH4}}{K_{S-CH4} + C_{CH4}} \cdot \frac{C_{O2}}{K_{S-O2} + C_{O2}} \qquad (1)$$

18 Average K_{S-CH4} and K_{S-O2} values for lakes have been determined by previous studies: K_{S-CH4} =

19 0.110 ± 0.053 mg L^{-1} (mean ± SD; Liikanen et al., 2002; Lofton et al., 2013) and K_{S-O2} =

20 0.624 ± 0.064 mg L^{-1} (mean ± SD; Lidstrom and Somers, 1984; Frenzel et al., 1990). To the

21 best of our knowledge, the highest K_{S-CH4} reported in lakes is 0.704 mg L^{-1} (Liikanen et al.,

22 2002). It should be noted that these reported K_S values refer to the apparent affinity constants

23 for the methanotrophic community, rather than the half-saturation constant for the CH4

24 monooxygenase enzyme that catalyzes CH4 oxidation. The potential error caused by using

25 previously reported K_S, instead of experimentally determined values will be considered in the

26 discussion section.

27 To establish the extent of potential MO limitation by CH4 or DO, two limitation factors were

28 defined, where β is the limitation factor for CH4 (%) and γ is the limitation factor for DO (%):

29 $$0\% \leq \beta = (1 - \frac{C_{CH4}}{K_{S-CH4} + C_{CH4}}) \cdot 100 \leq 100\% \qquad (2)$$

30 $$0\% \leq \gamma = (1 - \frac{C_{O2}}{K_{S-O2} + C_{O2}}) \cdot 100 \leq 100\% \qquad (3)$$

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. 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 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, measured at all depth throughout the lake water columns with the Hydrolab probe 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. 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 for which a complete temperature profile was determined

were fully mixed, while two lakes were partially stratified and none was fully stratified. We observed temperature inversion in 15 of the 18 lakes, with temperature gradients ranging from -0.1 to -3.4 °C, with an average of -1.4 °C (top minus– bottom temperature)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} (manufacturer 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 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 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 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 (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. 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; ANOVA , $p < 0.05$), 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 (Kruskal-Wallis test, $p < 0.05$); specifically, r_{max} was 0.71 ± 0.36 and 0.29 ± 0.16 mg L^{-1} d^{-1} (mean $\pm$ 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 organic matter decomposes in part to form CH_4 in surface lake sediments. In contrast, ancient yedoma carbon is decomposed throughout the sediment profile (Fig. 1), with particularly high rates of methanogenesis occurring along the permafrost thaw front, located deep in the thaw bulb beneath the lake (Heslop et al., 2015). Methane produced at depth in the thaw bulb subsequently migrates, primarily as free-phase bubbles through bubble tubes in sediments, to the surface sediments where it escapes the lake via ebullition (Walter Anthony and Anthony 2013; Tan et al. 2015). ~~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 metabolism in the sediments together with aerobic metabolism in the water column, leading to higher CH_4 and lower DO concentrations in the water column. ~~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 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 CH4 production was probably higher due to warmer sediments,~~ In summer, although CH4 production was probably higher due to warmer sediments, it did not cumulate in the water column and was released to the atmosphere ~~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 proportion of yedoma lakes in central Alaska. No significant latitudinal trend was observed when yedoma and non-yedoma lakes were analyzed separately (Kruskal-Wallis test, $p <$ 0.05).

Fig. 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 ± 5.86 mg L^{-1} (mean $\pm$ SD) was found, while the DO concentration was 0.13 ± 0.03 mg L^{-1} (mean $\pm$ SD). In the same yedoma lakes, the summer CH4 concentration was 0.49 ± 0.52 mg L^{-1} (mean $\pm$ SD), while the DO concentration was 3.19 ± 3.24 mg L^{-1} (mean $\pm$ SD). This observation suggests that MO was a significant contributor of the biogeochemical processes, actively controlling O2 and CH4 concentrations by oxidizing CH4 when O2 was present. To confirm the latter, it would be necessary to measure experimentally the O2 uptake rate by methanotrophs and by other aerobic processes that compete with MO (Dzyuban, 2010). ~~This observation suggests that MO was actively controlling O2 and CH4 concentrations by oxidizing CH4 when O2 was present. To confirm the latter, it would be necessary to measure experimentally the O2 uptake rate by methanotrophs and by other aerobic processes that compete with MO (Dzyuban, 2010).~~

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. 4). These results provide additional evidence that in non-yedoma lakes, the lower organic carbon inputs fuels more weakly methanogenesis and aerobic processes including MO than in yedoma lakes, resulting in a lower seasonal variation of CH4 and DO concentration~~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 under the ice cover dissolves into the lake water column before being confined within the growing ice sheet, leading to elevated dissolved CH$_4$ beneath the ice~~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 factors of MO rates

The actual MO rates r ranged 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), which as expected were significantly lower than the potential 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~~ r values are within the range reported for arctic lakes, which ranges over 3 magnitude order, from ~~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 similar and non-limiting conditions~~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~~

1. ~~optimal conditions~~. As observed with r_{max}, during summer yedoma lakes showed 2–3 times

2. higher r than non-yedoma lakes (Kruskal-Wallis test, $p < 0.05$; $r = 0.28 \pm 0.17$, mean $\pm$ SD,

3. yedoma lakes; $r = 0.09 \pm 0.08$ mg L^{-1} d^{-1}, mean $\pm$ SD, non-yedoma lakes). Higher r values for

4. yedoma lakes in summer is explained by the higher dissolved CH_4 concentration in presence

5. of a relatively high DO concentration above the oxycline (Fig. 3). ~~An apparent latitudinal~~

6. ~~trend was observed, with higher r and r_{max} for lakes from central Alaska compared to those~~

7. ~~from southern and northern Alaska (Fig. 5). This apparent trend was associated with a higher~~

8. ~~proportion of yedoma lakes in central Alaska.~~ As observed with CH_4 and DO concentrations,

9. ~~n~~No significant latitudinal trend in MO was observed, when yedoma and non-yedoma lakes

10. were analyzed separately.

11. The actual MO rates; r, were determined from r_{max} and CH_4 and DO concentrations using two

12. affinity constants, K_{S-CH4} and K_{S-O2}. These affinity constants are highly variable, because their

13. determination is challenging and subject to relatively high determination error (Segers et al.,

14. 1998) and because the methanotrophic community is sensitive to numerous factors and

15. changes over time and space (Carini et al., 2005; He et al., 2012). For instance, Lofton et al.

16. (2014) reported a variation of 150% in K_{S-CH4} within the hypolimnetic water column of two

17. lakes with similar characteristics. The determination of MO rates may, therefore, be subject to

18. large error if reported values are used instead of experimental parameters or because of

19. standard error associated to experimental K_S determination (Kovarova-Kovar and Egli,

20. 1998)~~The determination of MO rates may, therefore, be subject to large error if reported~~

21. ~~values are used instead of experimental parameters or if an error occurs in experimental K_S~~

22. ~~determinations~~. To quantify these potential errors, a sensitivity analysis was conducted. We

23. arbitrarily modified K_{S-CH4} and K_{S-O2} and calculated the resulting r (Eq. 1) using the

24. experimental r_{max}, C_{CH4}, and C_{O2} measured in the 30 lakes. Fig. 6 shows the error on r caused

25. by a given error on K_{S-O2} (Fig. 6a) and K_{S-CH4} (Fig. 6b), for yedoma and ~~non-yedoma~~non-

26. yedoma lakes, in winter and in summer. According to this analysis, an underestimation of K_{S-}

27. $_{O2}$ or K_{S-CH4} would lead to an overestimation of the actual MO rate (positive error), while an

28. overestimation of these affinity constants would produce an underestimation of r (negative

29. error). Fig. 6a shows that, an error on K_{S-O2} ranging from -50% to 200%, would cause from

30. 10% to -6% error on r, for all lakes and all seasons, except in yedoma lakes during winter,

31. where an error from 75% to -50% would be generated. This relatively high sensitivity of r to

32. error in K_{S-O2} in yedoma lakes during winter is due to DO concentrations close to K_{S-O2}.

33. Likewise, Fig. 6b shows that, from an error on K_{S-CH4} ranging from -50% to 200%, a resulting

1 error on r from 6% to -4% would be done, for all lakes and all seasons, except in ~~non~~

2 ~~yedoma~~non-yedoma lakes during summer, where an error from 50% to -34% would be

3 generated. As above, the latter is due to CH_4 concentrations close to K_{S-CH4} in ~~non~~

4 ~~yedoma~~non-yedoma lakes during summer. This sensitivity analysis shows that, other than for

5 K_{S-O2} in yedoma lakes during winter and K_{S-CH4} in non-yedoma lakes during summer, errors

6 on K_S would have relatively little impact on determination of methanotrophic rates. The

7 potentially significant error on methanotrophic rates in yedoma lakes during winter and in

8 non-yedoma lakes during summer encourages further studies including experimental K_S

9 determination. ~~This sensitivity analysis shows that, other than for K_{S-O2} in yedoma lakes~~

10 ~~during winter and K_{S-CH4} in non-yedoma lakes during summer, errors on K_S would have~~

11 ~~relatively little impact on determination of methanotrophic rates~~

12 From Eq. 2 and 3, we estimated that, during the summer CH_4 was the main limiting factor in

13 25 out of 30 lakes. In contrast, during winter, CH_4 was the main limiting factor in 10 of 26

14 lakes (Table 2). Notably, during the winter, DO was the limiting factor of MO for all seven

15 yedoma lakes, while during the summer, CH_4 was the limiting factor for all ~~non-yedoma~~non-

16 yedoma lakes ~~Notably, during the winter, DO was the limiting factor for all seven yedoma~~

17 ~~lakes, while during the summer, MO was limited by CH_4 for all non-yedoma lakes~~. A similar

18 error analysis was done on β and γ, as done with r, to estimate if the estimated limiting factor

19 would change as a result of error on K_S ranging from -50% to 200%. The results showed no

20 impact on the limiting factor in the 30 lakes and for both seasons. These results confirm that

21 MO was mainly controlled by DO and CH_4 availability, which in turn, depended on the

22 season and landscape processes.

23 A potential bias in our *r* estimates may have arisen from taking ~~duplicate~~ water samples at a

24 single depth in each lake. The literature has clearly shown that a higher MO rate is often

25 found at the oxic/anoxic interface in stratified lakes (Utsumi et al., 1998a, 1998b; Bastviken

26 al. 2002; Carini et al., 2005; Pimenov et al., 2010; Schubert et al., 2010). Estimation of MO

27 rates consistently measured at a single depth that was not necessarily located at the

28 oxic/anoxic interface may have neglected potentially higher rates occurring at the oxic/anoxic

29 interface in stratified lakes. However, in the present study, the sampled lakes were in many

30 cases shallow, relatively well mixed, and without a clear oxycline (see Results section),

31 suggesting a relatively homogeneous water column. Utsumi et al. (1998b) observed

32 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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3 **Table 1.** Identification, location, and permafrost soil type for lakes included in the study.

4 *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

5

1   **Table 2.** Methane oxidation parameters for 30 Alaskan lakes. * indicates median; ND - Not

2   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 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	ND	O_2
17	6.60	0.59	0.19	0.57	0.06	1.34	0.01	0.54	O_2	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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1 **Figure captions**

2

3 **Figure 1.** Carbon cycling in northern high-latitude lakes during the summer and winter.

4 Carbon (C_{org}) release from primary production and landscape processes promotes CH_4

5 production and competes with MO for O_2.

6 **Figure 2.** Locations of studied Alaskan lakes (white circles) plotted on the Alaska DEM

7 hillshade raster. Information about the distribution of yedoma-type deposits (Pleistocene-

8 aged, ice-rich silt containing deep thermokarst lakes) was from Jorgenson et al. (2008) and

9 Kanevskiy et al. (2011). The Alaska map is the National Elevation Data Set 30 m hillshade

10 raster.

11 **Figure 3.** Statistical distributions of CH_4 (white boxes) and DO (grey boxes) water

12 concentrations in yedoma and non-yedoma lakes during the winter and summer. The boxes

13 include the median (Q2) and the quartile range (Q1 and Q3). The whiskers show minimum

14 and maximum data. The open circles show outlier data. Capital letters are Kruskal-Wallis

15 multiple comparison test; values with the same capital letter are not significantly different (*p*

16 < 0.05, $Z > 1.96$). *n* represents the number of lakes measured.

17 **Figure 4.** Examples of CH_4 oxidation patterns observed during the MO assays: (a) assay with

18 no lag-phase. (b) assay with a 3-day lag-phase. and (c) assay with no detected activity.

19 Straight lines are linear correlations.

20 **Figure 5.** (a) CH_4 oxidation potential (r_{max}) and (b) CH_4 oxidation rates observed in 30 lakes

21 along a north-south transect (left-right) in Alaska during the summer (white bars) and the

22 winter (black bars).

23 **Figure 6.** Sensitivity analysis of the impact of an error or variation in K_{S-O2} (a.) and K_{S-CH4}

24 (b.) on r; yedoma lakes in winter (——), yedoma lakes in summer (— · — ·), non-yedoma lakes

25 in winter (— ·· — ··) and non-yedoma lakes in summer (-----).

Yedoma permafrost

◯ Studied lakes