

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

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11

12 **Abstract**

13 Methanotrophic bacteria play an important role oxidizing a significant fraction of methane
14 (CH₄) produced in lakes. Aerobic CH₄ oxidation depends mainly on lake CH₄ and oxygen
15 (O₂) concentrations, in such manner that higher MO rates are usually found at the oxic/anoxic
16 interface, where both molecules are present. MO also depends on temperature, and via
17 methanogenesis, on organic carbon input to lakes, including from thawing permafrost in
18 thermokarst (thaw)-affected lakes.

19 Given the large variability in these environmental factors, CH₄ oxidation is expected to be
20 subject to large seasonal and geographic variations, which have been scarcely reported in the
21 literature. In the present study, we measured CH₄ oxidation rates in 30 Alaskan lakes along a
22 north-south latitudinal transect during winter and summer with a new field laser spectroscopy
23 method. Additionally, we measured dissolved CH₄ and O₂ concentrations. We found that in
24 the winter, aerobic CH₄ oxidation was mainly controlled by the dissolved O₂ concentration,
25 while in the summer it was controlled primarily by the CH₄ concentration, which was scarce
26 compared to dissolved O₂. The permafrost environment of the lakes was identified as another
27 key factor. Thermokarst (thaw) lakes formed in yedoma-type permafrost had significantly
28 higher CH₄ oxidation rates compared to other thermokarst and non-thermokarst lakes formed
29 in non-yedoma permafrost environments. As thermokarst lakes formed in yedoma-type

1 permafrost have been identified to receive large quantities of terrestrial organic carbon from
2 thaw and subsidence of the surrounding landscape into the lake, confirming the strong
3 coupling between terrestrial and aquatic habitats and its influence on CH₄ cycling.

4

5 **1 Introduction**

6 Northern lakes are an important source of atmospheric CH₄ (Bastviken et al. 2011), and it has
7 been estimated that they are responsible for as much as 6% of global emission to the
8 atmosphere (Walter et al., 2007). Methane emission from aquatic ecosystems is significantly
9 mitigated by CH₄ oxidation (MO) by aerobic methanotrophs, a group of gram-negative
10 bacteria that use CH₄ as a carbon and energy source (Murrell et al., 1993; Trotsenko and
11 Murrell, 2008). It has been estimated that globally, 30 to 99% of total CH₄ produced in
12 freshwater ecosystems is microbiologically oxidized in the water column rather than being
13 released to the atmosphere (Bastviken et al., 2002; Thauer et al., 2008). Likewise, MO plays
14 an important role in northern lakes specifically by oxidizing up to 88% of the CH₄ diffusing
15 through the water column (Kankaala et al., 2006, 2007; Bellido et al., 2011). As recently
16 demonstrated using stable isotopes, after assimilating CH₄, methanotrophs are incorporated
17 into the lake food web by zooplankton (Kankaala et al., 2006; Jones and Grey, 2011),
18 *Daphnia magna* (Taipale et al., 2012), *Odonata spp.* (Seifert and Scheu, 2012), and
19 *Chironomus larvae* (Gentzel et al., 2012; Wooller et al., 2012), among others. In addition to
20 CH₄ respiration and conversion to CO₂, MO is therefore a pathway that reincorporates a
21 fraction of the CH₄-C produced into the biogeochemical carbon cycle within lakes.

22 Several environmental factors directly affect aerobic MO in freshwater ecosystems. First,
23 methanotrophy depends on the availability of both CH₄ and O₂. Higher MO rates are usually
24 found at the oxic/anoxic interface, where both CH₄ and O₂ are present (Utsumi et al., 1998a,
25 1998b; Bastviken et al., 2002; Liikanen et al., 2002; Carini et al., 2005; Schubert et al., 2010).
26 In turn, CH₄ and O₂ concentrations depend on numerous other processes involved in
27 biogeochemical carbon cycling (Fig. 1). Among these, the most important are methanogenesis
28 producing CH₄, primary production and atmospheric diffusion supplying O₂, and several
29 aerobic metabolic processes that compete with MO for available O₂ (Dzyuban, 2010).

30 In addition to autochthonous and allochthonous carbon inputs to lakes, permafrost thaw can
31 provide an additional source of labile organic carbon to fuel methanogenesis and carbon
32 mineralization in thermokarst (thaw) lakes (Zimov et al., 1997; Walter et al., 2006). MO in

1 northern regions is therefore directly and indirectly linked to permafrost type and landscape
2 processes that are highly variable. Permafrost ranges from sporadic to continuous and is also
3 variable in composition (Jorgenson et al., 2008). Yedoma-type permafrost is an organic-rich
4 (about 2% carbon by mass) Pleistocene-age permafrost with ice content of 50–90% by
5 volume (Zimov et al., 2006), which occurs mainly in the previously unglaciated regions of
6 Siberia, Alaska, and NW Canada (Czudek and Demek, 1970; Walter et al., 2007; Kanevskiy
7 et al., 2011; Grosse et al., 2013). Non-yedoma permafrost has a more widespread distribution
8 than yedoma permafrost and is also characterized by a much thinner carbon-rich horizon,
9 variable in composition (Ping et al., 2008; Tarnocai et al., 2009; Hugelius et al. 2014).

10 Many northern lakes are located in continental climate zones, subject to contrasting seasonal
11 conditions with long, cold winters followed by relatively short, but warm summers. Although
12 psychrotolerant and psychrophilic methanotrophs have been reported (Omelchenko et al.,
13 1993, 1996; Bowman et al., 1997; Trotsenko and Khmelenina, 2002), MO was reported to
14 occur more efficiently at mesophilic temperature (Semrau et al., 2010). During winter,
15 northern lakes are covered by a thick ice layer for seven to nine months. Surface lake ice
16 impedes oxygen transfer from the atmosphere to the lake and, when snow-covered,
17 substantially reduces light penetration and oxygen production by photosynthesis (White et al.,
18 2008; Clilverd et al., 2009). Thus, the combination of low temperature and limited oxygen
19 availability suggests lower MO rates in northern lakes in winter than in summer.

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

25

26 **2 Materials and Methods**

27 **2.1 Site description**

28 We sampled 30 Alaskan lakes during two field campaigns, one in late winter (March–April
29 2011) and one in summer (June–July 2011). To evaluate the effects of latitudinal variation
30 and permafrost type on MO, lakes were selected along a transect from the southcentral
31 Alaskan coast on the Kenai Peninsula to the Arctic Ocean near Prudhoe Bay (Fig. 2). The

1 transect crossed through glaciated mountain ranges and discontinuous, sporadic, or no
2 permafrost in south-central Alaska; discontinuous to isolated yedoma permafrost in the
3 interior of Alaska; and continuous permafrost in northern Alaska. In this work, for simplicity,
4 lakes located in yedoma-type permafrost areas will be referred to as “yedoma lakes” and all
5 others as “non-yedoma lakes”. Geographic variability along the north-south Alaska transect
6 has been previously described for ecosystems, climate, geology, and permafrost type
7 (Gregory-Eaves et al., 2000; Jorgenson et al., 2008; Smith et al., 2010). Additionally,
8 Sepulveda-Jauregui et al. (2014) quantified the surface area of the selected lakes (0.002–1.45
9 km²), their trophic states (ultraoligotrophic to eutrophic), and their annual CH₄ fluxes (0.5–
10 317 g CH₄ m⁻² y⁻¹). Table 1 shows the location and permafrost type of the selected lakes.

11 **2.2 Sampling and field measurements**

12 We sampled lake water usually near the center of each lake. In the winter, the ice cover was
13 drilled through with a motorized auger (0.3 m in diameter). Using a Hydrolab DataSonde
14 (Hach Hydromet, Loveland, CO, USA), we measured temperature, pH, chlorophyll *a*, and
15 dissolved oxygen (DO). The pH and DO sensors of the Hydrolab were calibrated regularly,
16 before and after each section of the latitudinal lake transect (four sections per transect,
17 approximately one calibration per week). Temperature and chlorophyll *a* sensors were not
18 regularly calibrated. All parameters were measured at 0.5 or 1-m depth intervals throughout
19 the water column, except in Dolly Varden L. where measurement intervals were increased to
20 every five meters from 15 m to 25m depth. In lakes shallower than 1 m, we measured
21 Hydrolab parameters at three distributed depths throughout the lake water column.

22 Water samples for MO rates and dissolved CH₄ concentration were taken at a depth of within
23 1 m of the ice-water interface in winter and usually at 0.75 to 1 m water depth in summer.
24 Due to differences in lake depth and thickness of the ice sheets, samples reflected surface
25 water in deep lakes, but mid water column or even lake bottom water environment, in shallow
26 lakes. Samples were taken with a horizontal Van Dorn bottle (Wildco, Yulee, FL, USA).

27 Water density derived from surface and bottom water temperatures were used to determine
28 the relative water column stability (RWCS; Padisak et al., 2003). Lakes with RWCS >56.5
29 were considered fully stratified, lakes with RWCS <16.3 were considered fully mixed, and
30 lakes with intermediate RWCS were considered partially stratified (Branco et al., 2009).
31 Similarly, we determined whether an oxycline was present in each lake based on a sharp DO

1 gradient or presence of an oxic/anoxic interface. Detailed temperature and DO profiles are
2 available as a Supplement in Sepulveda-Jauregui et al. (2014). We report all results in mean \pm
3 standard deviation (SD).

4 **2.3 Dissolved CH₄ concentration and MO rate**

5 To avoid long delays in sample transfer from remote locations to the laboratory, we
6 determined dissolved CH₄ concentrations with a previously described method based on
7 Headspace Equilibration using Infrared Tunable Diode Laser Absorption Spectroscopy (HE-
8 TDLAS; Sepulveda-Jauregui et al., 2012). This method consisted of creating a gas/liquid
9 equilibrium in a 100 mL equilibration vial containing 60 mL of lake water sample and 40 mL
10 of headspace (air) by vigorous shaking for 10 s. As previously reported (Sepulveda-Jauregui
11 et al., 2012), this shaking time was enough to reach equilibrium and allowed the non-invasive
12 determination of CH₄ concentration in the water sample by measuring headspace
13 concentration with a laser beam crossing the headspace of the equilibration vial, through the
14 glass walls of the vial. This measurement was conducted with a modified open-field CH₄
15 analyzer (GasFinder 2; Boreal Laser, Edmonton, Canada). The CH₄ concentration in the water
16 sample was calculated from the measured headspace concentration according to Henry's law.
17 In the present work, all CH₄ concentrations are expressed in mg CH₄ L⁻¹.

18 We determined duplicate MO rates in one water sample from each lake taken as described
19 above, using a modified HE-TDLAS method to allow for measurement of MO in the field.
20 This new method was based on a previous development using the HE-TDLAS method for the
21 determination of methanogenic activity (Martinez-Cruz et al., 2012). Two 60-mL lake water
22 subsamples from a single Van Dorn bottle sample were gently transferred to two 100-mL
23 equilibration vials (duplicates). Equilibration vials were immediately closed with rubber
24 stoppers and vigorously shaken for 10 s to transfer most of the dissolved CH₄ contained in the
25 water sample to the headspace. Next, the headspace was vented, the vial was closed, and the
26 sample was shaken again to evacuate the residual CH₄ content of the water sample. Using this
27 procedure, more than 99.5% of the original CH₄ content of the sample was evacuated. The
28 equilibration vials were then closed with rubber stoppers and aluminum crimp caps, spiked
29 with 0.6 mL CH₄ (99.0% purity; Air Liquide, Houston, TX, USA) injected with a disposable
30 syringe, and vigorously shaken for 10 s. This approach allowed MO tests to be conducted
31 with an initial standard CH₄ concentration in the liquid phase (\sim 0.6 mg L⁻¹). It also provided

1 an initial CH₄ to O₂ molar ratio of 0.062, significantly below the stoichiometric ratio (0.5),
2 ensuring no O₂ limitation.

3 Equilibration vials were incubated for 10–12 days in a water bath inside insulated boxes
4 placed in our vehicle. In the winter, the vials were maintained at 2 ± 2 °C in a water bath with
5 ice supplements; in the summer, the vials were maintained at 15 ± 2 °C. The temperature of
6 the water bath was measured daily. We measured the CH₄ concentration in the equilibration
7 vials daily using the HE-TDLAS method described in detail by Sepulveda-Jauregui et al.
8 (2012). Briefly, dry control MO test vials containing only CH₄ standards were read by the
9 TDLAS for calibration. Each experimental equilibration vial was vigorously shaken for 10 s
10 to reach phase equilibrium and then immediately placed in the laser beam path, after which a
11 stable HE-TDLAS reading was typically observed within 5 s. Five readings were taken for
12 each MO test vial and recalibration was conducted after measuring each set of test vials to
13 ensure instrument stability. The field HE-TDLAS method allowed measurement of dissolved
14 CH₄ and MO rates. This technique was simple, rapid (about 60 s per sample measurement),
15 non-invasive, and avoided complications and long delays in sample transfer from remote
16 locations to the laboratory.

17 We calculated the total CH₄ concentration (C_{CH_4} = total CH₄ mass present in the gas and
18 liquid phases divided by the sample liquid volume) in each vial during the MO tests. MO rates
19 were determined from the decrease in C_{CH_4} in the equilibration vials with time. MO rates
20 determined by this method represent the MO rate after aeration and CH₄ addition (vials spiked
21 with CH₄ and vigorously shaken). Thus, these MO rates do not correspond to actual
22 observations of in situ DO and dissolved CH₄ concentrations in the lakes. The measured CH₄
23 oxidation rate was considered the potential MO (r_{max} ; mg CH₄ L⁻¹ d⁻¹) under non-limiting CH₄
24 and DO concentrations. To estimate the actual rate (r ; mg CH₄ L⁻¹ d⁻¹) from r_{max} , a double
25 Monod model was used (Bae and Rittmann, 1996; Segers, 1998) in which C_{CH_4} and C_{O_2}
26 represent the actual dissolved CH₄ and DO concentrations measured in the lake, respectively,
27 and K_{S-CH_4} and K_{S-O_2} are the apparent affinity constants of the methanotrophic community, for
28 CH₄ and DO, respectively:

$$29 \quad r = r_{max} \cdot \frac{C_{CH_4}}{K_{S-CH_4} + C_{CH_4}} \cdot \frac{C_{O_2}}{K_{S-O_2} + C_{O_2}} \quad (1)$$

30 Average K_{S-CH_4} and K_{S-O_2} values for lakes have been determined by previous studies: K_{S-CH_4} =
31 0.110 ± 0.053 mg L⁻¹ (mean \pm SD; Liikanen et al., 2002; Lofton et al., 2013) and K_{S-O_2} =

1 0.624 ± 0.064 mg L⁻¹ (mean ± SD; Lidstrom and Somers, 1984; Frenzel et al., 1990). To the
2 best of our knowledge, the highest K_{S-CH₄} reported in lakes is 0.704 mg L⁻¹ (Liikanen et al.,
3 2002). It should be noted that these reported K_S values refer to the apparent affinity constants
4 for the methanotrophic community, rather than the half-saturation constant for the CH₄
5 monooxygenase enzyme that catalyzes CH₄ oxidation. The potential error caused by using
6 previously reported K_S, instead of experimentally determined values will be considered in the
7 discussion section.

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

$$10 \quad 0\% \leq \beta = \left(1 - \frac{C_{CH_4}}{K_{S-CH_4} + C_{CH_4}}\right) \cdot 100 \leq 100\% \quad (2)$$

$$11 \quad 0\% \leq \gamma = \left(1 - \frac{C_{O_2}}{K_{S-O_2} + C_{O_2}}\right) \cdot 100 \leq 100\% \quad (3)$$

12 A limitation factor of 100% means that 100% of a process ceases to occur due to the absence
13 of the limiting substrate, while a limitation factor of 0% indicates a process occurring at
14 maximum rate ($r = r_{max}$). When $\beta > \gamma$, CH₄ was considered to be the limiting factor;
15 conversely, when $\gamma > \beta$, DO was considered to be the limiting factor.

16 **2.4 Statistical analyses**

17 Normality was assessed by the Shapiro-Wilk test. Since most of the data was non-normally
18 distributed and with unequal samples number, significant differences among all parameters
19 were determined using Kruskal-Wallis multiple comparison test (differences were considered
20 significant at $p < 0.05$, $Z > 1.96$). To assess whether CH₄ was oxidized during the MO
21 incubation tests, significant differences between C_{CH_4} were determined by an analysis of
22 variance (ANOVA; $p < 0.05$), after normality was assessed by the Shapiro-Wilk test.
23 Statistical analyses were conducted using the NCSS 2000 Statistical Analysis System
24 software (Number Cruncher Statistical Systems, Kaysville, UT, USA). Linear regressions
25 were also conducted to determinate the MO rates using Wolfram Mathematica 7.0 (Wolfram,
26 Minneapolis, MN, USA).

27

1 **3 Results**

2 **3.1 Physicochemical parameters**

3 The sampled lakes were shallow; other than four atypical lakes with a maximum known depth
4 >20 m (lakes #4, #24, #26 and #30), the average lake depth in summer was 4.5 ± 2.6 m (mean
5 \pm SD). During winter, none of the lakes was completely frozen at the sampling stations.
6 Liquid water was always present underneath the ice cover, which ranged in thickness from
7 0.60 to 1.25 m (mean \pm SD, 0.81 ± 0.14 m). The mean temperature, measured at all depth
8 throughout the lake water columns with the Hydrolab probe was 2.4 ± 0.6 °C (mean \pm SD, n =
9 103) in the winter and 13.9 ± 2.4 °C (mean \pm SD, n = 235) in the summer. According to
10 RWCS, during the summer, 15 lakes of the 28 for which a complete temperature profile was
11 determined were fully thermally stratified. Six lakes were partially stratified and seven lakes
12 were mixed. During the winter, 16 of 18 lakes for which a complete temperature profile was
13 determined were fully mixed, while two lakes were partially stratified and none was fully
14 stratified. We observed temperature inversion in 15 of the 18 lakes, with temperature
15 gradients ranging from -0.1 to -3.4 °C, with an average of -1.4 °C (top minus bottom
16 temperature). Overall, only one third of the temperature profiles indicated clear stratification.
17 In both seasons, no correlation between RWCS and lake depth was found, probably due to the
18 fact that lakes were shallow and with an uneven depth distribution.

19 Lake water pH ranged from 5.9 to 8.2 in winter and 6.3 to 9.2 in summer among the study
20 lakes. Chlorophyll *a* was only detected during the summer, ranging from 1.0 to 45.9 $\mu\text{g L}^{-1}$
21 (manufacturer detection limit, 0.03 $\mu\text{g L}^{-1}$). The concentration of dissolved CH₄ in the 30
22 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
23 the summer (Table 2). The DO concentration at the same depths ranged from 0.10 to 13.63
24 mg L^{-1} during the winter and from 0.22 to 11.07 mg L^{-1} during the summer (Table 2). During
25 summer, a clear oxycline was observed in all yedoma lakes, but only in six of 20 non-yedoma
26 lakes. In contrast, during winter, an oxycline was not observed in any of the yedoma lakes,
27 which were largely anaerobic throughout the whole water column. We observed an oxycline
28 in winter in four of 13 non-yedoma lakes. Overall, an oxycline was observed in 30% of the
29 DO profiles. Temperature-oxygen profiles for all 30 studied lakes are shown in Sepulveda-
30 Jauregui et al. (2014).

1 Fig. 3 shows the statistical distributions of the dissolved CH₄ and DO concentrations, as well
2 as the Kruskal-Wallis comparisons. Significant differences were observed between yedoma
3 and non-yedoma lakes ($p < 0.05$). In yedoma lakes, the CH₄ and DO concentrations were
4 significantly higher and lower, respectively, than in non-yedoma lakes during both seasons
5 (Kruskal-Wallis test, $p < 0.05$). In addition to differences related to permafrost type, higher
6 CH₄ concentrations and lower DO concentrations were observed during the winter than in the
7 summer (Fig. 3) and an apparent geographic trend was observed. Higher dissolved CH₄ and
8 lower DO concentrations were found in lakes from central Alaska than in those from southern
9 and northern Alaska (Sepulveda-Jauregui et al., 2014).

10 **3.2 Methane oxidation rates**

11 The HE-TDLAS method allowed us to determine the MO potential in the field in all studied
12 lakes. Fig. 4 shows three representative C_{CH_4} trends observed in the MO vials. In some cases,
13 MO began on the first day of incubation (Fig. 4a) and the initial slope of the change in C_{CH_4}
14 was taken into account in determining the MO rate. In about 60% of the cases during the
15 summer and 80% during the winter, a lag phase was observed; i.e. period of time with no
16 apparent MO (Fig. 4b). This behavior, termed “induction of MO”, has previously been
17 reported for various soils (Bender and Conrad, 1995; Dunfield et al., 1999) and can be
18 interpreted as an adaptation period of the CH₄ oxidizers to the culture conditions. In lakes in
19 which this pattern was observed, the lag phase was not taken into account and the MO rate
20 was instead determined from the slope of C_{CH_4} after the lag phase. When no significant
21 decrease in C_{CH_4} was observed during the first seven days (Fig. 4c; ANOVA, $p < 0.05$), we
22 assumed an MO rate of zero, consistent with previous reports for various soils (Whalen et al.,
23 1990; Bender and Conrad, 1995; Dunfield et al., 1999). We observed MO rates of zero in only
24 three non-yedoma lakes during winter. Otherwise, no correlation with lake morphology,
25 season, or permafrost type was observed in regard to the existence of a lag phase or its
26 duration.

27 The potential MO rate r_{max} ranged from 0.000 to 0.488 mg L⁻¹ d⁻¹ during the winter and from
28 0.073 to 1.339 mg L⁻¹ d⁻¹ during the summer (Fig. 5a). Seasonal variation of r_{max} was
29 significant, with summer r_{max} up to 47 times higher than winter rates. Permafrost type was
30 also an important determining factor, because during the summer, yedoma lakes had higher
31 r_{max} than non-yedoma lakes (Kruskal-Wallis test, $p < 0.05$); specifically, r_{max} was 0.71 ± 0.36
32 and 0.29 ± 0.16 mg L⁻¹ d⁻¹ (mean \pm SD) for yedoma and non-yedoma lakes, respectively.

1 However, during the winter, no significant differences were observed between yedoma and
2 non-yedoma lakes. In addition to differences related to permafrost type, an apparent
3 latitudinal pattern was also observed, with higher r_{\max} for lakes from central Alaska compared
4 to those from southern and northern Alaska (Fig. 5a).

5

6 **4 Discussion**

7 **4.1 Geographic and seasonal variations in physicochemical parameters**

8 In yedoma lakes, the CH₄ and DO concentrations were significantly higher and lower,
9 respectively, than in non-yedoma lakes during both seasons. This observation is most likely
10 due to higher organic carbon and nutrient inputs associated with thawing permafrost in
11 yedoma-type lakes. Walter Anthony et al. (2014) and Sepulveda-Jauregui et al. (2014)
12 showed that thawing yedoma permafrost not only provides ancient (Pleistocene-aged) organic
13 carbon stimulating CH₄ production but also phosphate and nitrogen (ammonium), which
14 promotes bacterial, algal and contemporary plant growth in and around lakes. Since terrestrial
15 plant matter surrounding lakes gets deposited in thermokarst-lake sediments as lakes laterally
16 expand, both enhanced allochthonous and autochthonous productivity of yedoma-type lake
17 ecosystems results in higher rates of contemporary organic matter loading to sediments of
18 yedoma-type lakes compared to non-yedoma lakes (Walter Anthony et al., 2014).
19 Contemporary organic matter decomposes in part to form CH₄ in surface lake sediments. In
20 contrast, ancient yedoma carbon is decomposed throughout the sediment profile (Fig. 1), with
21 particularly high rates of methanogenesis occurring along the permafrost thaw front, located
22 deep in the thaw bulb beneath the lake (Heslop et al., 2015). Methane produced at depth in the
23 thaw bulb subsequently migrates, primarily as free-phase bubbles through bubble tubes in
24 sediments, to the surface sediments where it escapes the lake via ebullition (Walter Anthony
25 and Anthony 2013; Tan et al. 2015). Thus higher organic carbon and nutrient inputs in
26 yedoma-type lakes promote higher anaerobic metabolism in the sediments together with
27 aerobic metabolism in the water column, leading to higher CH₄ and lower DO concentrations
28 in the water column.

29 In both yedoma and non-yedoma lakes, higher CH₄ concentrations and lower DO
30 concentrations were observed during the winter than in the summer (Fig. 3). This seasonal
31 variation can be attributed to thick ice covering the lakes in winter. Ice cover impedes gas

1 exchange between the water and the atmosphere, promoting CH₄ build-up in the water
2 column (Phelps et al., 1998; Bastviken et al., 2004; Juutinen et al., 2009) and hindering O₂
3 transfer from the atmosphere, except in some locations where high-flux ebullition seeps allow
4 gas exchange through local holes in lake ice (Greene et al., 2014). Ice and snow also reduce
5 light penetration and oxygen production by photosynthesis beneath the ice (White et al., 2008;
6 Clilverd et al., 2009). The absence of detectable levels of chlorophyll *a* in ice-covered lakes
7 during March and April (see results section) despite the longer springtime photoperiod was
8 supportive evidence of reduced photosynthesis under the ice. In summer, although CH₄
9 production was probably higher due to warmer sediments, it did not cumulate in the water
10 column and was released to the atmosphere (Fig. 1).

11 Geographic variations were also observed with higher dissolved CH₄ and lower DO
12 concentrations being found in lakes from central Alaska than in those from southern and
13 northern Alaska. However, this apparent latitudinal pattern was related to the higher
14 proportion of yedoma lakes in central Alaska. No significant latitudinal trend was observed
15 when yedoma and non-yedoma lakes were analyzed separately (Kruskal-Wallis test, *p* <
16 0.05).

17 Fig. 3 shows that when relatively high CH₄ concentrations were found, relatively low DO
18 concentrations were observed and conversely. This pattern was particularly clear in yedoma
19 lakes: in winter, a CH₄ concentration of 7.32 ± 5.86 mg L⁻¹ (mean \pm SD) was found, while the
20 DO concentration was 0.13 ± 0.03 mg L⁻¹ (mean \pm SD). In the same yedoma lakes, the
21 summer CH₄ concentration was 0.49 ± 0.52 mg L⁻¹ (mean \pm SD), while the DO concentration
22 was 3.19 ± 3.24 mg L⁻¹ (mean \pm SD). This observation suggests that MO was a significant
23 contributor of the biogeochemical processes, actively controlling O₂ and CH₄ concentrations
24 by oxidizing CH₄ when O₂ was present. To confirm the latter, it would be necessary to
25 measure experimentally the O₂ uptake rate by methanotrophs and by other aerobic processes
26 that compete with MO (Dzyuban, 2010).

27 The trend toward higher CH₄ concentrations and lower DO concentrations in winter than in
28 summer was not as strong in non-yedoma lakes as in yedoma lakes (Fig. 4). These results
29 provide additional evidence that in non-yedoma lakes, the lower organic carbon inputs fuels
30 more weakly methanogenesis and aerobic processes including MO than in yedoma lakes,
31 resulting in a lower seasonal variation of CH₄ and DO concentration. Another reason is that
32 yedoma lakes have a significantly higher ebullition year round (Walter et al., 2007;

1 Sepulveda-Jauregui et al., 2014). Even during winter, Greene et al. (2014) found that 80% of
2 CH₄ in ebullition bubbles trapped under the ice cover dissolves into the lake water column
3 before being confined within the growing ice sheet, leading to elevated dissolved CH₄ beneath
4 the ice.

5 **4.2 Limiting factors of MO rates**

6 The actual MO rates r ranged from 0.000 to 0.124 mg L⁻¹ d⁻¹ during the winter and from 0.017
7 to 0.538 mg L⁻¹ d⁻¹ during the summer (Fig 6b), which as expected were significantly lower
8 than the potential MO rates. These r values are within the range reported for arctic lakes,
9 which ranges over 3 magnitude order, from 0.001 to 1 mg L⁻¹ d⁻¹ (Liikanen et al., 2002;
10 Kankaala et al., 2006; Lofton et al., 2014). Similarly, r values were 1 to 50-fold higher in the
11 summer than in the winter. We attribute this finding to the temperature dependence of
12 methanotrophy (Semrau et al., 2008; Borrel et al., 2011), but also to the limited DO
13 concentration under the ice cover during the winter.

14 In addition to seasonal variations, permafrost type was also a determining factor of r and r_{max} .
15 As mentioned before, although no difference in r_{max} was observed during winter between
16 yedoma and non-yedoma lakes, r_{max} in yedoma lakes was about twice higher than in non-
17 yedoma lakes during summer. We attribute that difference to a more active MO
18 methanotrophic community in yedoma lakes, as all r_{max} tests were conducted in aerated vials
19 with an initial standard CH₄ concentration in the liquid phase (~0.6 mg L⁻¹), thus ensuring
20 similar and non-limiting conditions. As observed with r_{max} , during summer yedoma lakes
21 showed 2–3 times higher r than non-yedoma lakes (Kruskal-Wallis test, $p < 0.05$; $r = 0.28 \pm$
22 0.17 , mean \pm SD, yedoma lakes; $r = 0.09 \pm 0.08$ mg L⁻¹ d⁻¹, mean \pm SD, non-yedoma lakes).
23 Higher r values for yedoma lakes in summer is explained by the higher dissolved CH₄
24 concentration in presence of a relatively high DO concentration above the oxycline (Fig. 3).
25 As observed with CH₄ and DO concentrations, no significant latitudinal trend in MO was
26 observed, when yedoma and non-yedoma lakes were analyzed separately.

27 The actual MO rates; r , were determined from r_{max} and CH₄ and DO concentrations using two
28 affinity constants, K_{S-CH_4} and K_{S-O_2} . These affinity constants are highly variable, because their
29 determination is challenging and subject to relatively high determination error (Segers et al.,
30 1998) and because the methanotrophic community is sensitive to numerous factors and
31 changes over time and space (Carini et al., 2005; He et al., 2012). For instance, Lofton et al.

1 (2014) reported a variation of 150% in K_{S-CH_4} within the hypolimnetic water column of two
2 lakes with similar characteristics. The determination of MO rates may, therefore, be subject to
3 large error if reported values are used instead of experimental parameters or because of
4 standard error associated to experimental K_S determination (Kovarova-Kovar and Egli, 1998).
5 To quantify these potential errors, a sensitivity analysis was conducted. We arbitrarily
6 modified K_{S-CH_4} and K_{S-O_2} and calculated the resulting r (Eq. 1) using the experimental r_{max} ,
7 C_{CH_4} , and C_{O_2} measured in the 30 lakes. Fig. 6 shows the error on r caused by a given error on
8 K_{S-O_2} (Fig. 6a) and K_{S-CH_4} (Fig. 6b), for yedoma and non-yedoma lakes, in winter and in
9 summer. According to this analysis, an underestimation of K_{S-O_2} or K_{S-CH_4} would lead to an
10 overestimation of the actual MO rate (positive error), while an overestimation of these affinity
11 constants would produce an underestimation of r (negative error). Fig. 6a shows that, an error
12 on K_{S-O_2} ranging from -50% to 200%, would cause from 10% to -6% error on r , for all lakes
13 and all seasons, except in yedoma lakes during winter, where an error from 75% to -50%
14 would be generated. This relatively high sensitivity of r to error in K_{S-O_2} in yedoma lakes
15 during winter is due to DO concentrations close to K_{S-O_2} . Likewise, Fig. 6b shows that, from
16 an error on K_{S-CH_4} ranging from -50% to 200%, a resulting error on r from 6% to -4% would
17 be done, for all lakes and all seasons, except in non-yedoma lakes during summer, where an
18 error from 50% to -34% would be generated. As above, the latter is due to CH_4 concentrations
19 close to K_{S-CH_4} in non-yedoma lakes during summer. This sensitivity analysis shows that,
20 other than for K_{S-O_2} in yedoma lakes during winter and K_{S-CH_4} in non-yedoma lakes during
21 summer, errors on K_S would have relatively little impact on determination of methanotrophic
22 rates. The potentially significant error on methanotrophic rates in yedoma lakes during winter
23 and in non-yedoma lakes during summer encourages further studies including experimental
24 K_S determination.

25 From Eq. 2 and 3, we estimated that, during the summer CH_4 was the main limiting factor in
26 25 out of 30 lakes. In contrast, during winter, CH_4 was the main limiting factor in 10 of 26
27 lakes (Table 2). Notably, during the winter, DO was the limiting factor of MO for all seven
28 yedoma lakes, while during the summer, CH_4 was the limiting factor for all non-yedoma
29 lakes. A similar error analysis was done on β and γ , as done with r , to estimate if the estimated
30 limiting factor would change as a result of error on K_S ranging from -50% to 200%. The
31 results showed no impact on the limiting factor in the 30 lakes and for both seasons. These
32 results confirm that MO was mainly controlled by DO and CH_4 availability, which in turn,
33 depended on the season and landscape processes.

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

15

16 **5 Conclusions**

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

29

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6

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1 **Tables**

2

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

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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**

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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-O_2} (a.) and K_{S-CH_4}
24 (b.) on r ; yedoma lakes in winter (—), yedoma lakes in summer (— · — ·), non-yedoma lakes
25 in winter (— · · — · ·) and non-yedoma lakes in summer (-----).

Summer

Winter











