

1 **Methane and carbon dioxide emissions from 40 lakes along a North-South latitudinal**
2 **transect in Alaska**

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1 **Abstract**

2 Uncertainties in the magnitude and seasonality of various gas emission modes,
3 particularly among different lake types, limit our ability to estimate methane (CH₄) and
4 carbon dioxide (CO₂) emissions from northern lakes. Here we assessed the relationship
5 between CH₄ and CO₂ emission modes in 40 lakes along a latitudinal transect in Alaska to
6 lakes' physicochemical properties and geographic characteristics, including permafrost soil
7 type surrounding lakes. Emission modes included Direct Ebullition, Diffusion, Storage flux,
8 and a newly identified Ice-Bubble Storage (IBS) flux. We found that all lakes were net
9 sources of atmospheric CH₄ and CO₂, but the climate warming impact of lake CH₄ emissions
10 was two times higher than that of CO₂. Ebullition and Diffusion were the dominant modes of
11 CH₄ and CO₂ emissions respectively. IBS, ~10% of total annual CH₄ emissions, is the release
12 to the atmosphere of seasonally ice-trapped bubbles when lake ice confining bubbles begins
13 to melt in spring. IBS, which has not been explicitly accounted for in regional studies,
14 increased the estimate of springtime emissions from our study lakes by 320%.
15 Geographically, CH₄ emissions from stratified, mixotrophic interior Alaska thermokarst
16 (thaw) lakes formed in icy, organic-rich yedoma permafrost soils were 6-fold higher than
17 from non-yedoma lakes throughout the rest of Alaska. The relationship between CO₂
18 emissions and geographic parameters was weak, suggesting high variability among sources
19 and sinks that regulate CO₂ emissions (e.g. catchment waters, pH equilibrium). Total CH₄
20 emission was correlated with concentrations of soluble reactive phosphorus and total nitrogen
21 in lake water, Secchi depth and lake area, with yedoma lakes having higher nutrient
22 concentrations, shallower Secchi depth, and smaller lake areas. Our findings suggest that
23 permafrost type plays important roles in determining CH₄ emissions from lakes by both
24 supplying organic matter to methanogenesis directly from thawing permafrost and by
25 enhancing nutrient availability to primary production, which can also fuel decomposition and
26 methanogenesis.

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1 **1 Introduction**

2 Lakes are an important source of atmospheric greenhouse gases, methane (CH₄) and
3 carbon dioxide (CO₂) (Battin et al., 2009; Tranvik et al., 2009; Bastviken et al., 2011). In
4 lakes, CH₄ is produced, consumed, and exchanged with the atmosphere in a different manner
5 than CO₂. CH₄ is produced in anaerobic environments (mainly in sediments), while CO₂ in
6 lakes originates from respiration throughout the water column and sediments, inflow of
7 terrestrially derived dissolved inorganic carbon from surrounding watersheds, and
8 photooxidation of dissolved organic carbon (DOC) (Graneli et al., 1996; Tranvik et al., 2009;
9 Weyhenmeyer et al., 2012; Maberly et al., 2013). CO₂ is also formed in lakes by aerobic
10 oxidation of CH₄, a process that can consume a significant fraction of CH₄ produced in lakes
11 (Kankaala et al., 2006; Bastviken et al., 2008; Lofton et al., 2013). The ratio of CO₂
12 emissions versus carbon sequestration in northern lakes was found to be controlled by nitrate
13 concentrations in lake water (Kortelainen et al., 2013). Meanwhile, CO₂ is consumed by
14 photosynthesis and other autotrophic or chemical processes (e.g. increasing alkalinity,
15 photooxidation) that depend on pH and/or the availability of light (Madigan et al., 2009).

16 Despite recycling of CH₄ and CO₂ internally in lakes, a significant quantity of these
17 greenhouse gases is released from lakes to the atmosphere (Cole et al., 2007). Most of Earth's
18 lakes are located in northern high latitudes, overlapping the permafrost-dominated region
19 (Downing et al., 2006; Smith et al., 2007; Grosse et al., 2013). It is estimated that CH₄
20 emission from lakes globally comprises about 16% (71.6 Tg) of all human and natural
21 atmospheric sources, and that northern lakes (> 55 °N) contribute about 20% of these
22 emissions (13.6 Tg; Bastviken et al., 2011). In contrast, CO₂ emissions from northern lakes
23 constitute approximately 43% (1.2 Pg CO₂) of global emissions from lakes (Battin et al.,
24 2009; Tranvik et al., 2009; Maberly et al., 2013). This disproportionality between the
25 contribution of CH₄ and CO₂ emissions from northern lakes is not well understood, and may
26 be due to numerous factors, including sensitivity of methanogenesis to temperature and lake
27 trophic status (Tranvik et al., 2009; Ortiz-Llorente and Alvarez-Cobelas, 2012; Marotta et al.,
28 2014) versus processes that control CO₂ availability (e. g. photosynthesis, inputs from
29 terrestrial ecosystems, and organic matter mineralization) (Kling et al., 1991; Battin et al.,

1 2009; Tranvik et al., 2009). Furthermore, lake CH₄ emission data is scarce relative to CO₂
2 data, particularly at high northern latitudes (Tranvik et al., 2009; Bastviken et al., 2011). Due
3 to a disproportionately low number of northern high latitude lakes represented in previous
4 studies of global CH₄ emissions (Bastviken et al., 2011), and a paucity of studies that
5 considered various modes of emission together, CH₄ and CO₂ emissions from northern high
6 latitude lakes are still poorly constrained.

7 Landscape diversity in Alaska provides a valuable opportunity to study CH₄ and CO₂
8 emission patterns from lakes as they relate to origin, climate, ecology, geology, and
9 permafrost coverage. Across Arctic, Continental, and Transitional climate zones in Alaska,
10 ecological habitats include arctic, alpine and forest tundra, and northern and southern boreal
11 forests (Gregory-Eaves et al., 2000). The surficial geology in which Alaskan lakes are found
12 varies primarily from fine-grain aeolian deposits; to coarser-grain coastal, glacial, fluvial and
13 volcanic deposits; to rubble and bedrock (Karlstrom et al., 1964; Arp and Jones, 2009).
14 Alaska is also characterized by a variety of permafrost types (Fig. 1) ranging from isolated
15 permafrost in south-central Alaska to continuous permafrost in northern Alaska (Jorgenson et
16 al., 2008).

17 Within the context of permafrost soil organic carbon content, Alaskan lakes can be
18 classified depending on whether they are surrounded by yedoma-type permafrost or non-
19 yedoma substrates (Walter Anthony et al., 2012). Yedoma is typically thick (tens of meters),
20 Pleistocene-aged loess-dominated permafrost sediment with high organic carbon (~2% by
21 mass) and ice (50-90% by volume) contents (Zimov et al., 2006). When yedoma thaws and
22 ground ice melts, deep thermokarst (thaw) lakes with high CH₄ production potentials form
23 (Zimov et al., 1997; Kanevskiy et al., 2011; Walter Anthony and Anthony, 2013). Some non-
24 yedoma permafrost soils can also have high organic carbon and excess ice concentrations
25 within several meters of the ground surface; however, these organic- and ice-rich permafrost
26 horizons are typically thinner than yedoma deposits (Ping et al., 2008; Tarnocai et al., 2009).
27 As a result, thermokarst lakes formed in non-yedoma permafrost soils are commonly
28 shallower than yedoma lakes and have been shown to emit less CH₄ (West and Plug, 2008;
29 Grosse et al., 2013; Walter Anthony and Anthony, 2013).

1 Estimating CH₄ and CO₂ emissions from northern high latitude lakes, which are
2 seasonally covered by ice, represents a difficult task because there are at least four emission
3 pathways, all of which have not been consistently and simultaneously measured in the past:
4 (1) Direct Ebullition, (2) Diffusion, (3) Storage flux, and a newly identified (4) Ice-Bubble
5 Storage (IBS) flux (Greene et al., 2014).

6 Ebullition (bubbling) has been observed as the dominant pathway of CH₄ emissions
7 from many lakes (Casper et al., 2000; Bastviken et al., 2004; Walter et al., 2006). Since CH₄
8 is less soluble, high concentrations in interstitial sediment water lead to bubble formation and
9 their emission to the atmosphere. In contrast, CH₄ Diffusion flux to the atmosphere is usually
10 relatively low and occurs mainly in summer when ice cover is absent. Due to much higher
11 solubility, CO₂ tends to occur in low concentrations in ebullition bubbles, and instead escapes
12 lakes predominately by Diffusion (Abril et al., 2005).

13 During winter, ice formation on most northern lakes impedes gas emissions to the
14 atmosphere. Dissolved CH₄ and CO₂ accumulate in the lake water column beneath the ice,
15 resulting in gas “storage.” Storage emissions occur when dissolved CH₄ and CO₂ are emitted
16 by diffusion when the ice melts in spring, often enhanced by full or partial lake overturn
17 (Michmerhuizen et al., 1996; Phelps et al., 1998; Bellido et al., 2009). Storage emissions also
18 occur in some lakes in autumn, if lake overturn caused by falling temperature brings high
19 concentrations of dissolved gases from the hypolimnion to the surface, resulting in rapid CH₄
20 and CO₂ emission by diffusion from the water column. Bastviken et al. (2004) coined the
21 term “Storage flux” when they considered it in regional lake emission estimates as a function
22 of differences in water column CH₄ stocks before and after lake ice-out, CH₄ production, and
23 CH₄ oxidation.

24 The fourth potential emission component involves CH₄ release to the atmosphere
25 from seasonally ice-trapped ebullition bubbles in spring before the ice disappears. During
26 winter, emission to the atmosphere of many bubbles rising from sediments is impeded by
27 seasonal lake ice. When bubbles come to rest under the ice, they exchange gases with the
28 water column (Greene et al., 2014). Some bubbles become sealed in ice as ice thickens
29 downward. Due to the insulation property of gas bubbles, ice is locally thinner where bubbles

1 are trapped, and bubbles usually stack in vertical columns separated by ice lenses of various
2 thicknesses. As a result, when lake ice begins to melt in spring, bubble-rich patches of ice
3 begin to locally degrade before the rest of the ice sheet. These ebullition bubbles previously
4 sealed in and under ice are released to the atmosphere by an emission mode termed “Ice-
5 Bubble Storage” (IBS) (Greene et al., 2014). Ponded water on the lake-ice surface can
6 accelerate the release of ice-trapped bubbles to the atmosphere and also provides the
7 opportunity for visual observation of gas release from bubbles trapped by degrading ice
8 (K.M.W.A. unpublished data, 2014). It should be noted that gas in small, tubular bubbles
9 formed in lake ice by the exclusion of dissolved gases as ice freezes (Gow and Langston,
10 1977; Langer et al., 2014) is presumably released to the atmosphere when ice degrades as
11 well; however, given the substantially lower concentration of CH₄ in these non-ebullition,
12 freeze-out bubbles (usually < 0.01% by volume; Boereboom et al., 2012), this mode of
13 emission is relatively insignificant in comparison to the larger ebullition-sourced bubbles, in
14 which CH₄ concentrations typically range from 40-90% by volume (Martens et al., 1992;
15 Semiletov et al., 1996; Walter Anthony et al., 2010).

16 Finally, it is important to understand how changes in nutrient availability and
17 temperature influence CO₂ and CH₄ cycling in lakes. Increasing nutrients and temperature
18 stimulates primary production and microbial decomposition of organic matter, which in turn
19 consumes oxygen (O₂) and enhances anaerobic decay processes, particularly in sediments,
20 where CH₄ and CO₂ are produced (Conrad et al., 2010). Aerobic CH₄ oxidation is controlled
21 directly by O₂ and CH₄ concentrations and temperature (Utsumi et al., 1998; Bastviken et al.,
22 2002; Borrel et al., 2011) and indirectly by nutrient availability (Dzyuban et al., 2010).
23 Measurement of O₂ and CH₄ concentrations in lakes are essential for assessing global carbon
24 cycling, and in this framework, correlating both parameters in situ has been promoted as an
25 indirect means of assessing CH₄ oxidation by methanotrophs (Bastviken et al., 2004; Guerin
26 and Abril, 2007; Sepulveda-Jauregui et al., 2012).

27 In this study we assessed the relationships between measured CH₄ and CO₂ emission
28 modes in 40 lakes along a North-South Alaska transect to the lakes' physicochemical
29 properties and geographic characteristics. Our goal was to assess the magnitude, variability

1 and seasonality of individual modes of emission, particularly among the wide range of
2 geographic lake settings in Alaska.

3

4 **2 Materials and Methods**

5 **2.1 Study lakes and permafrost zones**

6 We sampled water from 40 Alaskan lakes during open-water conditions in June-July
7 2011 and 2012 (Fig. 1) and from 26 of the lakes toward the end of the winter ice-cover period
8 in March-April 2011. [Measurements were usually made during daylight hours between 10:00
9 and 18:00.](#) Our study lakes were located near the road system along a North-South transect in
10 Alaska that spans a variety of geographic and limnological settings, described previously by
11 Gregory-Eaves et al. (2000), Jorgenson et al. (2008), and Walter Anthony et al. (2012). Our
12 study lakes occupied three general climatic/permafrost zones: (1) The northern study area
13 (66-70 °N, Arctic climate/continuous permafrost), (2) the interior study area (64-66 °N,
14 Continental climate/discontinuous permafrost), and the southern study area (60-64 °N,
15 Transitional climate/sporadic and isolated permafrost) (Gregory-Eaves et al., 2000; Jorgenson
16 et al., 2008). Additionally, we distinguished yedoma-type thermokarst lakes as those formed
17 in yedoma permafrost with active, ongoing thermokarst activity from non-yedoma type lakes,
18 which were lakes occurring in other non-yedoma deposits in permafrost and non-permafrost
19 soils (Fig. 1). Lake names, sizes, geographic characteristics and limnological properties are
20 shown in Table 1.

21 **2.2 Water-dissolved CH₄, CO₂ and O₂**

22 Offshore and usually near the center of each lake, we sampled lake water at one to
23 nine distributed depths throughout the water column for dissolved CH₄ and CO₂
24 concentrations and at 0.5-m depth intervals for O₂ concentrations during winter and summer.
25 In lakes shallower than 1 m we sampled only one depth within 25 cm of the lake bottom. In
26 the field we measured CH₄ concentration by the Headspace Equilibration-Tunable Diode
27 Laser Spectroscopy (HE-TDLAS) method (Sepulveda-Jauregui et al., 2012) using a
28 GasFinder 2.0 (Boreal Laser Inc., Edmonton, Canada; Appendix A). Additionally, we
29 determined concentrations of headspace CH₄ and CO₂ in bottles of lake water in the

1 laboratory following Kling (2010) using a GC-2014 gas chromatograph (Shimadzu, Addison,
2 Illinois, USA) equipped with a flame ionization detector and a PLOT alumina column
3 (detector temperature 250 °C, oven 40 °C, high purity Helium as carrier gas). Strong
4 correlation between the GasFinder and bottle headspace methods was reported previously by
5 Sepulveda-Jauregui et al. (2012). Dissolved O₂ concentrations were measured in the field
6 with a luminescence sensor connected to a calibrated multiparametric probe Hydrolab
7 DataSonde (Hach LDO, Loveland, Colorado, USA).

8 **2.3 CH₄ and CO₂ Diffusion Flux**

9 We estimated the Diffusion flux of CH₄ and CO₂ (g m⁻² yr⁻¹) based on the once per
10 summer measurement of dissolved CO₂ and CH₄ in surface water from each lake and
11 extrapolating results to the summer time open water period. We applied Fick's Law to our
12 measurements of dissolved CO₂ and CH₄ in surface water following the boundary layer
13 method of Kling et al. (1992):

$$14 \text{ Diffusion flux} = T \times D \times z^{-1} \times (C_w - C_{eq}) \quad (1)$$

15 where T is the conversion factor from seconds to years (31,536,000); D is the molecular
16 diffusivity of CH₄ or CO₂ (m² s⁻¹) following Kling et al. (1992); z (m) is the thickness of the
17 surface boundary layer, assumed to be 200 μm as an average for Alaskan lakes following
18 Kling et al. (1992); C_w is the measured gas concentration at the bottom of the boundary layer
19 (g m⁻³); C_{eq} is the equilibrium gas concentration in surface lake water (g m⁻³) exposed to the
20 atmosphere at the top of the boundary layer. We calculated C_w and C_{eq} using measured
21 surface water temperatures, Henry's Law constants, and temperature dependence constants
22 for CH₄ and CO₂, respectively (NIST, 2011). We acknowledge that wind speed and heat
23 exchange vary over different time scales and that they have a large effect on the gas exchange
24 coefficient (Cole and Caraco, 1998; Tedford et al., 2014) and thus on the relative importance
25 of diffusion emission from lakes. However, lacking wind speed and heat exchange data for
26 our study lakes, our calculations are based on the assumption of a constant gas exchange
27 coefficient derived from averaged wind speed values from lakes in our northern tundra study
28 region (Kling et al., 1992). Because many of our study lakes are surrounded by trees, the
29 average wind speed at these lakes during the open-water periods is likely more similar to that

1 of the low-wind Mirror Lake, studied by Cole and Caraco (1998). On one lake, Goldstream
2 L. (forested, Interior Alaska), where we had higher temporal resolution data for surface water
3 dissolved CH₄ concentrations (Greene et al., 2014) during the open water summer period, we
4 explored the effect of using the average value of the exchange coefficient from Cole and
5 Caraco (1998) instead of Kling et al. (1992) and found that the exchange coefficient
6 calculated from the boundary layer thickness of Kling et al. (1992) differed by 2% from that
7 from Cole and Caraco (1998).

8 **2.4 Storage flux**

9 To estimate Storage flux, dissolved CH₄ and CO₂ profiles were measured in spring
10 before the ice began to melt and in summer during ice-free conditions. We multiplied the
11 average concentration of dissolved CH₄ and CO₂ measured in samples collected from
12 distributed depths in the water column by the height of the unfrozen water column. Storage
13 flux (g m⁻² yr⁻¹) was calculated as the difference between total mass of dissolved gas in spring
14 before ice break up and the total mass of dissolved gas in summer.

15 **2.5 CH₄ and CO₂ Ebullition from Sediments**

16 We estimated CH₄ and CO₂ ebullition from sediments associated with discrete seeps
17 following the lake-ice ebullition survey method of Walter Anthony et al. (2010). Seeps are
18 defined as point-source locations of repeated bubbling and identified as A, B, C, and Hotspot
19 classes according to distinct patterns of bubbles trapped in lake ice (Appendix A). To
20 quantify seep ebullition, we removed snow from early winter lake ice to expose ebullition
21 bubble clusters trapped in ice for seep classification, GPS mapping, flux measurements and
22 gas collection using submerged bubble traps. On foot, we surveyed 9,355 individual seeps
23 within 161 plots (30-300 m² per plot) positioned randomly within both littoral and profundal
24 zones of lakes. In some lakes, ice was opened above the seeps for placement of submerged
25 bubble traps. We retained semi-automated bubble traps placed over individual seeps year-
26 round (Walter Anthony et al., 2010) to provide daily and seasonal ebullition flux data from
27 sediments. Seep class-specific flux rates and bubble CH₄ and CO₂ concentrations measured
28 on a subset of seeps were applied to all mapped seeps to estimate whole-lake ebullition rates,
29 indexed by Julian Day of the year (Appendix A). These fluxes represent bubbling rates from

1 sediments as measured at the lake surface, not necessarily Direct Ebullition to the
2 atmosphere. The following two section describe the fate of ebullition bubbles during the ice-
3 cover and ice-free seasons.

4 **2.6 Ice-Bubble Storage (IBS) flux**

5 During the open-water (ice-free) summer season, ebullition bubbles reaching the lake
6 surface release CH₄ directly to the atmosphere (Direct Ebullition). In winter, lake ice impedes
7 Direct Ebullition emissions. Many ebullition bubbles reaching the top of the water column hit
8 the underside of lake ice, come to rest, and exchange gases with the water column until the
9 downward-growing ice encapsulates the bubbles. Since lake water is typically undersaturated
10 in CH₄ with respect to the CH₄ concentration (40-90%) of most ebullition bubbles
11 (Sepulveda-Jauregui et al., 2012), CH₄ readily diffuses out of bubbles into the lake water
12 column.

13 We collected 37 samples of ebullition bubbles trapped as pockets in lake ice from five
14 Alaskan lakes, expanding upon the lake ice-bubble data set of Walter et al. (2008).
15 Additionally, we opened the lake ice and placed bubble traps beneath ice, above seeps, to
16 sample 'fresh' ebullition bubbles at the lake surface before they are impeded by ice (n = 2-41
17 seeps per lake; total of 560 samples). This allowed us to compare concentrations of CH₄ in
18 ice-trapped bubbles (n = 2-8 seeps per lake) to gas concentrations in 'fresh' bubbles prior to
19 ice entrapment.

20 Numerical modeling informed by detailed field studies of CH₄ diffusion from ice-
21 trapped bubbles in one of our study lakes, Goldstream L. (#18) revealed that 80% of CH₄ in
22 bubbles trapped by ice dissolves into the lake water column in winter (Greene et al., 2014).
23 The remaining 20% of CH₄ ebullition trapped by ice is released to the atmosphere, either
24 from Hotspot seep sites that open periodically throughout the winter, or from A, B, and C
25 seep sites as ice melts in spring (i.e. IBS emissions). With input of observed ice-growth rates
26 on a subset of lakes in each of the three study regions and mean monthly atmospheric
27 temperatures during 2003-2013 (U.S. National Weather Service), we employed this model to
28 calculate a first-order estimate of IBS in 34 of the 40 study lakes in which we had
29 measurements of both seep ebullition and water-column dissolved CH₄ concentrations, which

1 affect the CH₄ dissolution rate from bubbles. We linearly interpolated between measured
2 surface CH₄ concentrations in the summer and spring to estimate water-column CH₄
3 concentrations during the ice-cover period. The decrease in the volume of ice-trapped
4 bubbles in each lake, as calculated by this model, was used together with the decrease in their
5 CH₄ concentration, calculated from our measurements of fresh vs. ice-trapped bubbles, to
6 determine the IBS flux for each lake.

7 **2.7 Direct Ebullition in Winter and Summer**

8 Since ice-bubble pockets above A-, B-, and C-type seeps open approximately one
9 month prior to complete disappearance of lake ice in spring (K.M.W.A. unpublished data,
10 2014; Greene et al., 2014), we assume in our calculations that subsequent ebullition by seeps
11 releases fresh bubbles directly to the atmosphere through open holes during this spring melt
12 period. Particularly high bubbling rates from 'Hotspot' seeps maintain ice-free conditions
13 above these point-sources of bubbling, allowing for Direct Ebullition to the atmosphere when
14 air temperature is higher than -15 °C (Zimov et al., 2001; Greene et al., 2014). In interior
15 Alaska, the only region where Hotspot seeps were observed, mean monthly temperatures
16 from 2003-2013 indicated that on average, wintertime Direct Ebullition from hotspots occurs
17 for several weeks post-freeze up in October and in spring from February until ice melt in
18 May. These shoulder seasons of bubble emissions through open holes in lake ice are
19 consistent with our field observations. However, warm temperature anomalies or heavy
20 snowfall events can also open hotspots at other times (on the scale of days) during winter
21 (K.M.W.A. personal observation, 2014; Zimov et al., 2001; Greene et al., 2014), but these
22 were not included in our calculations. In this study, ebullition from all seep classes during the
23 final month of ice cover and from Hotspots during fall and spring shoulder seasons when
24 mean monthly atmospheric temperatures were higher than -15 °C (U.S. National Weather
25 Service) together comprised Direct Ebullition in winter.

26 Direct Ebullition in summer was estimated as the product of average seep densities on
27 each lake and the sum of daily ebullition measured in bubble traps placed on representative
28 seeps of each class in a subset of lakes during the open-water summer period (Sect. 2.5).

29 **2.8 Seasonal and mean annual emissions**

1 We estimated mean annual emissions from lakes as the sum of various modes of
2 emissions seasonally: 1) Direct Ebullition from all seeps and Diffusion from the water
3 column in summer (ice-free period); 2) winter (ice-cover period) Direct Ebullition emissions
4 through ice-free Hotspot seeps during shoulder seasons and from all open seeps during the
5 final month of the spring ice-melt season; and 3) spring emissions as the sum of first the
6 release of IBS (ebullition seep gases trapped by lake ice) before lake ice disappears, and
7 second, the release of lake water column Storage of dissolved gases, previously described by
8 Michmerhuizen et al. (1996), Phelps et al. (1998), and Bastviken et al. (2004), when ice
9 melts. We acknowledge that our calculations contain uncertainty associated with the
10 assumption that single-day measurements of dissolved CO₂ and CH₄ in lakes represent the
11 mean for calculating Diffusion flux for the entire open water period; however, these were the
12 best available data at the time of this study, and a similar approach has been used in
13 numerous other studies reviewed by Bastviken et al. (2011). Due to a paucity of field
14 measurements on the Alaskan lakes, annual emissions estimates do not include background
15 (non-seep) ebullition, which was found to be 25% of annual emission in Siberian lakes
16 (Walter et al., 2006).

17 Because lakes were classified according to three geographic zones based on climate
18 and permafrost, the average timing of ice cover was used to estimate the seasonal differences
19 between CH₄ and CO₂ emissions for all lakes within each zone. Mean annual ice-on and ice-
20 off dates from were compiled for years 2000-2012 for study lakes near Toolik Field Station
21 in the northern region (1 Oct. – 18 Jun.), our own observations of interior Alaska study lakes
22 near Fairbanks from years 2008-2012 (8 Oct. - 9 May), and from Arp et al. (2013) and the
23 National Park Service Inventory and Monitoring Program during years 2000-2013 for
24 southern region lakes near Denali National Park (1 Oct. – 23 May) and southcentral Alaska,
25 south of the Alaska Range (15 Nov. - 7 May).

26 **2.9 Physical and chemical limnology**

27 We measured the physicochemical properties of lakes during winter and summer field
28 campaigns at the same locations where dissolved gases were measured. Measurements of in
29 situ water properties along vertical depth profiles in lakes included temperature, pH,

1 oxidation reduction potential (ORP), and chlorophyll-a (Chl-a) obtained using a calibrated
2 multiparametric probe Hydrolab DataSonde (Hach, Loveland, CO, USA). For a subset of
3 lakes in each region, we used temperature data loggers (UA-001-08, Onset HOBO, Bourne,
4 MA, USA) to record water temperature year-round in five-minute intervals at two depths (1
5 m water depth and lake bottom). Secchi disk depth (SecD) was measured with a 0.2 m Secchi
6 disk. We collected water samples for ex situ analyses using a horizontal 2.2 L Van Dorn
7 Bottle (WILDCO, Yulee, FL, USA). The concentrations of dissolved nitrate (NO_3^-), soluble
8 reactive phosphorus, (SRP) and sulfate (SO_4^{2-}) in lake water were measured with a high-
9 performance liquid chromatograph equipped with an electrochemical detector (ED40 Dionex,
10 Dionex, USA). We determined total organic carbon [TOC; used to approximate DOC
11 following Wetzel (2001) and Weyhenmeyer and Karlsson (2009)] and total nitrogen (TN)
12 with a total carbon and nitrogen analyzer (Shimadzu TOC-Vcsn equipped with TNM1
13 module, Shimadzu, Japan).

14 Trophic state indexes (TSI), calculated from Chl-a, SecD, and SRP, were used to
15 estimate the trophic states of the lakes (Carlson, 1977). Since total phosphorus (TP) is
16 typically used in TSI calculations, our calculation is an approximation of trophic state.
17 However, we do not expect the use of SRP instead of TP has a large effect on our results,
18 since Chl-a is the primary index for trophic state classification (Carlson and Simpson, 1996).
19 Furthermore, SRP is the more biologically reactive form of phosphorous in lake water lake,
20 and has been shown to be a good predictor of trophic status (Stendick and Hall, 2003;
21 Haberman and Haldna, 2014).

22 We classified some lakes as mixotrophic since our field and laboratory observations
23 of brown water color (high DOC), low SecD, high nutrients, high epilimnetic Chl-a
24 concentrations, abundant macrophytes, and anoxic hypolimnion matched the definition of
25 mixotrophic, provided by Williamson et al. (1999). In these lakes, water had a dark brown
26 color resulting from high concentrations of DOC, presumably from humic substances and
27 organic acids leached from litter and soils in their watersheds. Rather than recognizing two
28 separate classes of high DOC lakes (mixotrophy with high nutrient concentrations vs.
29 dystrophy with low nutrient concentrations), Wetzel (2001) considered all high DOC lakes as

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1 | dystrophic. Wetzel (2001) explained that the productivity of most dystrophic lakes has
2 classically been described as low; however, more detailed examinations indicated that
3 chlorophyll concentration (phytoplankton biomass) was significantly higher in the more
4 shallow photic zone of brown-water lakes than in clear lakes when expressed per volume of
5 epilimnion. We did not quantify macrophyte biomass, but our qualitative observation of a
6 higher abundance of submerged and emergent plants growing in the brown-water lakes is
7 also consistent with Wetzel's description of littoral plants often contributing significantly to
8 lake ecosystem metabolism in dystrophic(mixotrophic) lakes.

9 Surface sediment samples (1-5 cm depth) were collected in summer 2008 from a
10 subset of lakes using a 6.6 cm diameter piston hammer corer at multiple locations within
11 individual lakes. Samples were stored under refrigeration and then dried (105 °C), acidified
12 (5-15 mL 2N HCl) and the top 1-cm was analyzed for TOC and TN on a Costech ESC 4010
13 elemental analyzer (Alaska Stable Isotope Facility at the University of Alaska Water and
14 Environmental Research Center). Additional surface lake sediment samples were collected in
15 2012 from a central lake location using the hammer corer. These sediments were analyzed for
16 moisture content by weighing and drying to 105 °C. We determined organic matter content
17 on a dry weight basis via loss-on-ignition at 550 °C (Dean, 1974).

18 **2.10 Statistical Analysis**

19 Since data were not normally distributed and did not meet the assumption of
20 homoscedasticity, we tested relationships between CH₄ and CO₂ emissions vs. geographic
21 characteristics and limnological properties for the different lakes using the non-parametric
22 Two-tailed Mann-Whitney U test for comparison of two groups and Kruskal-Wallis One Way
23 Analysis of Variance for comparison of several groups. We followed the Kruskal-Wallis
24 analysis with the Multiple-Comparison Z-value test; differences were significant when the Z
25 value was > 1.96.

26 We used single linear regression analysis to quantify relationships between CH₄ and
27 CO₂ emissions and geographic and limnological properties. For these analyses, data
28 normalization was obtained using logarithm base 10 (Log) transformation. Before and after
29 data transformation, normality was assessed by the Shapiro-Wilk test. Regression models

1 were accepted when the p-value was < 0.01 . Mean values from full vertical depth profiles of
2 temperature, pH, ORP and from epilimnion measurements for Chl-a are shown in Table 1 and
3 were used in these single linear regression analyses. We used the mean winter temperature
4 measured with Hobo data loggers (1 m water depth and lake bottom) to fill data gaps in some
5 northern lakes (Table 1).

6 Relationships between separately permafrost type CH_4 ebullition and lake area, lake-
7 bottom water dissolved CH_4 , lake-bottom water dissolved O_2 , and ebullition were evaluated
8 graphically and by Spearman Product-Moment Correlation Coefficients (r_s).

9 Statistical analyses were performed with NCSS 2000 Statistical Analysis 193 System
10 software (Number Cruncher Statistical Systems, USA). To fill data gaps, we added additional
11 limnological, geographic and ecological zone information from the literature to our own
12 measurements (Table 1).

13

14 **3 Results**

15 **3.1 Geographic and limnological patterns of CH_4 and CO_2 emissions**

16 Total annual CH_4 and CO_2 emissions were highly variable, ranging two orders of
17 magnitude among lakes (2.0 to $> 300 \text{ g CH}_4 \text{ m}^{-2} \text{ yr}^{-1}$ and 34.2 to $> 1,500 \text{ g CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$;
18 Table 2, Fig. 2). Among the geographic characteristics presented in Table 1 and CH_4 and CO_2
19 emissions presented in Table 2, we found that the type of permafrost soil (yedoma vs. non-
20 yedoma) was the geographic characteristic most closely related to CH_4 and CO_2 emissions
21 (Table 3). Total annual CH_4 emissions from yedoma lakes ($44.2 \pm 17.0 \text{ g m}^{-2} \text{ yr}^{-1}$, mean \pm
22 SD, $n = 7$ lakes, excluding outlier lake #25) was significantly higher than from non-yedoma
23 lakes ($8.0 \pm 4.1 \text{ g m}^{-2} \text{ yr}^{-1}$, $n = 32$ lakes) (Table 2). Total annual CO_2 emissions appeared
24 higher in yedoma ($784 \pm 757 \text{ g m}^{-2} \text{ yr}^{-1}$, mean \pm SD, $n = 8$ lakes, excluding outlier lake #25)
25 than non-yedoma lakes ($137 \pm 129 \text{ g m}^{-2} \text{ yr}^{-1}$, $n = 32$ lakes) (Table 2); however, due to high
26 variability among lakes, the difference was not significant. Rosie Creek beaver pond (#25),
27 an outlier lake with particularly high CH_4 and CO_2 emissions ($317 \text{ g CH}_4 \text{ m}^{-2} \text{ yr}^{-1}$; 1138 g
28 $\text{CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$; Fig. 2), was formed prior to our study by beaver activity in an active stream
29 system that drains into the Tanana River. The pond was subsequently influenced by

1 thermokarst expansion (Walter Anthony, personal observation) into yedoma-type deposits,
2 which further enhanced carbon cycling in the fluvial system.

3 The relationship between CH₄ and CO₂ emissions and other geographic parameters
4 followed the same pattern to the extent that they were related to characteristics of yedoma
5 and non-yedoma permafrost soils (Table 3). For instance, yedoma is characterized by eolian
6 deposits, which among the surface geologic deposit types was also most strongly related to
7 CH₄ and CO₂ emissions. Among our study lakes, yedoma lakes occurred in the interior
8 Alaska region (Fig. 1) and tended to have a mixotrophic state, parameters that were both
9 related to CH₄ and CO₂ emissions. Since the particular yedoma lakes in our study were
10 relatively small lakes ($\leq 0.1 \text{ km}^2$), lake area was a morphologic parameter closely related to
11 CH₄ and CO₂ emissions.

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12 Regressions models showed that physical and chemical limnological parameters
13 (Table 1) explained 19-63% of deviation in the different flux pathways of CH₄ emissions
14 (Table 4). Total CH₄ emission was correlated with Area, SecD, SRP, and TN (Table 4). We
15 did not find any relationships between total CO₂ and the lakes' physicochemical properties,
16 probably due to chemical equilibrium in water.

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17 **3.2 Modes of CH₄ and CO₂ emission**

18 Total annual ebullition, consisting of Direct Ebullition in summer and winter as well
19 as springtime release from IBS, was the dominant mode of CH₄ emission in lakes, comprising
20 86% of total annual emissions from yedoma lakes and 65% from non-yedoma lakes (Table
21 2). Summer Direct Ebullition was higher in yedoma-type lakes ($26.2 \pm 15.9 \text{ g CH}_4 \text{ m}^{-2} \text{ yr}^{-1}$, n
22 $= 6$ lakes, excluding lake # 25) than non-yedoma lakes ($4.0 \pm 3.7 \text{ g CH}_4 \text{ m}^{-2} \text{ yr}^{-1}$, $n = 28$
23 lakes). This contrast drove other significant relationships in the data set: since yedoma lakes
24 were primarily located in the interior discontinuous permafrost zone, and they dominated the
25 mixotrophic and northern boreal forest lakes category, we found that summer ebullition was
26 higher in interior lakes than in northern and southern lakes; summer ebullition was higher in
27 mixotrophic lakes than in lakes of other trophic states; and northern boreal forest lakes had
28 higher summer Direct Ebullition than lakes from other ecozonal categories (Tables 2 and 3).
29 Direct Ebullition of CH₄ in winter and summer was correlated with lake Area. Smaller lakes

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1 had higher Direct Ebullition (Table 4); since our yedoma study lakes were smaller than non-
2 yedoma lakes, this factor is strongly influenced by permafrost type. The regression analysis
3 with permafrost type categories separately (yedoma and non-yedoma lake type) creates
4 scarce data in yedoma lakes ($n = 5$) to do this analysis. However Spearman coefficients
5 supports this tendency, since it indicates a negative correlation with lake area among yedoma
6 lakes (summer $r_s = -0.66$, winter $r_s = -0.71$) and in non-yedoma lakes (summer $r_s = -0.45$,
7 winter $r_s = -0.63$).

8 Yedoma lakes were the only lakes in which we observed Hotspot ebullition and seep
9 densities of all seep classes were higher in yedoma lakes (mean \pm SD: 2.12 \pm 2.50 A seeps m^{-2} ,
10 0.28 ± 0.19 B seeps m^{-2} , 0.06 ± 0.06 C seeps m^{-2} , 0.01 ± 0.01 Hotspot seeps m^{-2}) compared
11 to non-yedoma lakes (0.70 ± 0.68 A seeps m^{-2} , 0.05 ± 0.06 B seeps m^{-2} , 0.001 ± 0.003 C
12 seeps m^{-2} , 0 Hotspot seeps m^{-2}). It follows that Direct Ebullition during the winter ice-cover
13 period was also much higher from yedoma lakes (5.9 ± 3.6 g CH_4 m^{-2} yr^{-1} , $n = 6$ lakes;
14 excluding lake #25) than non-yedoma lakes (0.6 ± 0.6 g CH_4 m^{-2} yr^{-1} , $n = 28$ lakes) (Table 2).
15 In contrast, ebullition was not an important mode of CO_2 emission from any lakes. Total
16 ebullition, including summer and winter Direct Ebullition, contributed 0.1% of the total
17 annual CO_2 emissions among all lakes (Table 2).

18 A comparison of CH_4 composition in fresh ebullition bubbles vs. bubbles trapped by
19 lake ice revealed that the CH_4 concentration in ebullition bubbles trapped by ice was $33 \pm$
20 12% (mean \pm SD, $n = 6$ lakes) lower than in ebullition bubbles escaping to the atmosphere at
21 the lake surface unimpeded by ice (Fig. 3; Mann-Whitney U Test, $Z > 1.96$, $p < 0.05$).

22 The IBS model, which accounts for decreases in the volume and CH_4 concentration of
23 ice-trapped bubbles as their CH_4 dissolves into the water column (Greene et al., 2014),
24 revealed that IBS was on average 13% of total annual CH_4 emissions from yedoma lakes (5.8
25 ± 4.6 g m^{-2} yr^{-1} , $n = 6$) and 9% for non-yedoma lakes (0.7 ± 0.7 g m^{-2} yr^{-1} , $n = 28$) (Table 2,
26 Fig. 2). The CH_4 IBS flux from lakes was negatively correlated with Area and SecD (Table
27 4). Given the minor role of CO_2 Direct Ebullition in the annual emission budget ($< 0.1\%$),
28 and the even smaller role of springtime IBS, we considered IBS an insignificant mode of CO_2
29 emission.

1 Storage emissions were highly variable among all lakes ($0.5 \pm 0.7 \text{ g CH}_4 \text{ m}^{-2} \text{ yr}^{-1}$, n =
2 20 lakes; $7 \pm 17 \text{ g CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$, n = 18 lakes; excluding lake #25). We did not find a
3 significant difference in Storage flux between yedoma vs. non-yedoma lakes. As with all
4 modes of emission, lake #25 had the highest Storage CH_4 flux ($39.0 \text{ g m}^{-2} \text{ yr}^{-1}$). We did not
5 find a correlation between CH_4 Storage flux and limnological parameters ($p < 0.01$). Since we
6 were unable to normalize the CO_2 Storage flux data, it was not possible to assess potential
7 correlations between this mode of emission and limnological parameters. Comparing
8 emission modes, Storage flux contributed 3% and 0% of total annual CH_4 and CO_2 emissions,
9 respectively, from yedoma lakes and 5% and 7% of total annual CH_4 and CO_2 emissions,
10 respectively, from non-yedoma lakes (Table 2).

11 CH_4 Diffusion emissions were statistically different between yedoma ($5.0 \pm 1.4 \text{ g CH}_4$
12 $\text{m}^{-2} \text{ yr}^{-1}$, n = 5; excluding lake #25) and non-yedoma lakes ($2.4 \pm 1.3 \text{ g CH}_4 \text{ m}^{-2} \text{ yr}^{-1}$, n = 26).
13 Rosie Creek beaver pond (#25) had the highest diffusive flux ($160.3 \text{ g CH}_4 \text{ m}^{-2} \text{ yr}^{-1}$).
14 Diffusion comprised 11% and 30% of total annual CH_4 emissions from yedoma and non-
15 yedoma lakes respectively. We found a significant positive correlation between CH_4 diffusive
16 flux and [SRP](#) (Table 4). In contrast, Diffusion was the dominant CO_2 mode of emission
17 among all of our study lakes. Diffusion constituted 100% and 92% of CO_2 emissions from
18 yedoma and non-yedoma lakes respectively. Diffusion from yedoma lakes ($784 \pm 757 \text{ g CO}_2$
19 $\text{m}^{-2} \text{ yr}^{-1}$, n = 4 lakes) was significantly higher than Diffusion from non-yedoma lakes ($127 \pm$
20 $127 \text{ g CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$, n = 23 lakes). It was not possible to normalize CO_2 Diffusion data, so we
21 were unable to determine potential correlations between this mode of emission and
22 limnological parameters.

23 3.3 Seasonal emissions

24 Figure 4 illustrates the contribution of different gas emissions pathways to annual
25 emissions by season. Approximately three quarters of annual CH_4 emissions were released
26 from lakes during the open water summer season: 71% and 79% of total annual CH_4
27 emissions in yedoma lakes and non-yedoma lakes respectively were the sum of summer
28 Direct Ebullition and Diffusion. Spring and winter CH_4 emissions were also important. From
29 yedoma lakes, first 13% of total annual emissions occurred via IBS in spring when the ice

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1 started to degrade; subsequently, water column Storage release of dissolved gases was 3% of
2 total annual emissions. From non-yedoma lakes, total springtime emissions were 14% of
3 annual, consisting first of IBS (9%) followed by Storage (5%). Wintertime emissions via
4 Direct Ebullition from ice-free holes above seeps were 13% of total annual emissions from
5 yedoma lakes and 7% from non-yedoma lakes. It is of interest to note that accounting for
6 IBS, a newly recognized mode of emission, increased the estimate of springtime CH₄
7 emissions based on the more commonly reported Storage emission by 320%.

8 Seasonally, ~100% and 92% of total annual CO₂ emissions from yedoma and non-
9 yedoma lakes respectively occurred in summer by Diffusion from the open water surface.
10 The remaining 8% of annual emissions in non-yedoma lakes occurred in spring from water
11 column Storage flux (7%) and winter Direct Ebullition (less than 1%) (Table 2 and Fig. 2).

12 **3.4 Physical and chemical patterns**

13 The difference between yedoma and non-yedoma lakes was observed in several
14 physical and chemical parameters (Tables 1, 3, and 5). Southern lakes (non-yedoma lakes)
15 are deeper and larger than Interior lakes (mostly yedoma lakes), while northern lakes (non-
16 yedoma lakes) were not statistically different from lakes in the other regions.

17 Deep lakes (> 20 m), moderately deep lakes (usually > 6 m) with adequate wind
18 protection from topography and/or vegetation, and all yedoma lakes, owing to their small
19 surface area to volume ratios and high TOC concentrations were thermally stratified in
20 summer. Exceptions were two yedoma-type lakes with creeks flowing through them
21 (Killarney L. #20 and Rosie Creek beaver pond #25) and a small, shallow, yedoma
22 thermokarst pond (Stevens Pond #22, 1.1 m) that was semi-stratified. In contrast, shallow,
23 non-yedoma lakes (usually < 3m) and non-yedoma lakes located in mountain regions with
24 large surface area to volume ratios and high wind conditions were well mixed.

25 In winter, most lakes showed inverse stratification. We found that winter bottom
26 temperature was significantly different between northern lakes (1.3 ± 1.5 °C) and southern
27 lakes (2.6 ± 1.1 °C), but none of these were significantly different from lake bottom
28 temperature in Interior Alaska (1.4 ± 1.0 °C), which is mainly due to the contrasting climatic
29 conditions and the relatively shallow depths of northern lakes compared to southern lakes.

1 In most lakes, if there was a dissolved O₂ (DO) gradient, then DO was highest near
2 the lake surface and decreased with depth in winter and summer. Three exceptions were El
3 Fuego L. (#11), 91 L. (#27) and Dolly Varden L. (#36), where we observed an increase in
4 DO with depth in summer, likely due to benthic photosynthesis in the shallow lakes (#11 and
5 #27) and a deep chlorophyll maximum (DCM) in the deep lake (#36). In #36 we observed
6 Chl-a concentrations near the surface of ~ 3.7 µg L⁻¹; Chl-a concentrations increased with
7 depth to a maximum (23.0 µg L⁻¹) just below 20 m. DCM is a common trend in deep, clear-
8 water lakes with low trophic state (Gervais et al., 1997; Camacho, 2006). Among yedoma
9 lakes, lake-bottom dissolved oxygen (DO) concentrations were < 0.1 mg L⁻¹ in both winter
10 and summer. In contrast, 81% of the 32 non-yedoma lakes had well-oxygenated lake bottoms
11 in summer; the lake-bottom water DO concentration in the other 19% of lakes was < 0.1 mg
12 L⁻¹. In winter, we observed the reverse pattern among non-yedoma lakes: 76% of 17 non-
13 yedoma lakes measured had lake-bottom DO < 0.1 mg L⁻¹ while 24% of non-yedoma lakes,
14 all which were southern lakes, had well-oxygenated lake bottoms in winter. All temperature
15 and DO profiles measured on the study lakes are shown in Supplement Fig. B.

16 DO concentrations were inversely related to dissolved CH₄ concentrations in the lake
17 bottom water during winter and summer (Fig. 5). This relationship suggests a strong
18 influence by microbial processes that consume O₂, consequently reducing aerobic oxidation
19 of dissolved CH₄, particularly in the organic-rich, yedoma lakes of interior Alaska (Table 5
20 and sec. 4.3). Additionally, we found significant statistical relationships between lake area
21 and dissolved gas concentrations (CH₄ and O₂) among our yedoma (small lakes) and non-
22 yedoma study lakes (generally larger lakes) (Table 5).

23 Five additional limnological parameters also showed significant differences between
24 yedoma and non-yedoma lakes (Table 1). The TOC, ~~SRP~~, TN, Chl-a, and SecD indicated
25 higher nutrient availability and higher primary production in the ~~mixotrophic~~ yedoma lakes
26 and/or their watersheds (Table 1). ORP values were significantly different between winter
27 and summer in all lakes (Table 1), but were more than 2.5 and 1.5 times lower in yedoma
28 lakes compared to non-yedoma lakes in winter and summer respectively, indicating more
29 reducing conditions in yedoma-lake water columns. Temperature and pH were significantly

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1 different between summer and winter in non-yedoma lakes, while only temperature differed
2 seasonally in yedoma lakes. Altogether, these findings of higher primary production and
3 lower ORP are consistent with the observations of high CH₄ and low O₂ concentrations in
4 yedoma lakes compared to non-yedoma lakes (Fig. 5).

5 **4 Discussion**

6 **4.1 Emission modes**

7 The relative magnitude of different emission modes in this study followed the same general
8 pattern observed previously (Casper et al., 2000; Bastviken et al., 2004; Abril et al., 2005;
9 Repo et al., 2007), with ebullition dominating lake CH₄ emissions and diffusion dominating
10 CO₂ emissions. Most studies of ebullition are conducted by distributing bubble traps in lakes
11 without prior knowledge of discrete seep locations. Since seep locations are identified in
12 winter as vertical stacks of bubbles in lake ice that represent repeated ebullition from discrete
13 point-sources, surveys of lake-ice bubbles reveal the locations and densities of ebullition
14 seeps on lakes. Surveys also show the relative proportion of (ebullition) bubble-free black
15 ice, which in nearly all ice-covered lakes dominates on an area basis. Walter et al. (2006)
16 identified non-point source bubbling from the seep-free fraction of the lake as "Background
17 Ebullition". Background Ebullition is thought to originate primarily from methanogenesis in
18 surface lake sediments in summer; in contrast, ebullition seeps consist of bubble tubes that
19 allow CH₄ produced at depth in sediments to migrate efficiently as bubbles to the sediment
20 surface in summer and winter by the repeated release from point-source locations. Bubble
21 traps placed in seep and non-seep locations and monitored year-round in two Siberian lakes
22 showed that seep ebullition dominated total annual CH₄ emissions. Background Ebullition
23 was high in summer, nearly absent in winter, and altogether comprised ~25% of total annual
24 CH₄ emissions in the Siberian lakes. Preliminary results from bubble-traps placed in some of
25 our Alaskan study lakes in locations where no seep ebullition bubbles were observed in
26 winter also showed high summertime bubbling (K.M.W.A. unpublished data, 2014). This
27 suggests that Background Ebullition occurs in Alaska too. Since our estimate of lake
28 ebullition in the Alaskan lakes is based solely on discrete seeps and does not include non-
29 seep Background Ebullition, we consider that our estimate of total lake ebullition is below the

1 total actual ebullition flux. Given that methanogenesis is highly temperature dependent
2 (Dunfield et al., 1993; Schulz et al., 1997; Duc et al., 2010; Marotta et al. 2014; Yvon-
3 Durocher et al. 2014) and surface lake sediments heat up in summer, accounting for
4 Background Ebullition would likely increase the total ebullition emissions from all of the
5 Alaskan study lakes.

6 The Ice-Bubble Storage (IBS) mode of emission described here is a newly recognized
7 CH₄ ebullition flux component in lakes (Greene et al., 2014) that has not previously been
8 included in regional studies. Given the coarse temporal resolution of temperature and
9 dissolved gas data used as input to the IBS model, we acknowledge that our estimate of IBS
10 is a first-order approximation. However, strong agreement in the relative importance of IBS
11 in the annual CH₄ budget of Goldstream Lake (#18) in this study using coarse resolution data
12 (IBS 6% of total annual CH₄ emission) vs. the estimate from Greene et al. (2014) using
13 highly detailed field data allowing detailed modeling (IBS was 6% and 9% of total annual
14 emissions in two different years), suggests that our first-order approximations of IBS may be
15 valid. Since IBS was an important mode of CH₄ emissions among our study lakes (13% and
16 9% of total annual emissions in yedoma and non-yedoma lakes, respectively), it is likely that
17 past estimates of the magnitude and seasonality of CH₄ emissions from lakes with ebullition
18 seeps were incomplete. Greene et al. (2014) found that a large fraction (~80%) of CH₄
19 diffused from ebullition bubbles trapped under lake ice into the lake water in Goldstream L.
20 Coarser-resolution modeling of the IBS process for our study lakes also suggested that
21 approximately 80% of CH₄ dissolved out of ice-trapped bubbles. The mean and standard
22 deviation of the CH₄ fraction dissolving out of ice-trapped bubbles was $83 \pm 0.9\%$ for 34
23 lakes (range 65-89% for 33 lakes, excluding Killarney L. with anomalously low CH₄ content
24 in bubbles freshly released from sediments). Detailed measurements and modeling in
25 Goldstream L. showed that about half of this re-dissolved CH₄ was ultimately oxidized
26 (Greene et al., 2014). Due to a paucity of field data, we did not model CH₄ oxidation;
27 however, given the observed CH₄ oxidation potentials in our study lakes through incubation
28 studies (Martinez-Cruz et al., 2015), it is likely that some fraction of the re-dissolved
29 ebullition bubbles is oxidized. The un-oxidized fraction of dissolved CH₄ is subject to release

1 to the atmosphere via water column convection and diffusion as Storage emissions in spring
2 when ice more completely disintegrates and as Diffusion during summer (Greene et al.,
3 2014). Thus the Storage and Diffusion modes of emission may involve not only dissolved
4 CH₄ that diffused out of lake sediments, but also dissolved CH₄ that first originated as
5 ebullition bubbles prior to ice entrapment. Since ebullition seeps were important components
6 of whole-lake CH₄ emissions in all of our study lakes, as well as in tens of other lakes
7 previously reported in Alaska (Walter Anthony et al., 2012) and Siberia (Walter et al., 2006;
8 Walter Anthony et al., 2010), IBS should be studied and accounted for in global lake CH₄
9 emission budgets.

10 Lake CH₄ Storage emission estimates for our Alaska study lakes (0.5 ± 0.7 g CH₄ m⁻²
11 yr⁻¹; Table 2), which comprised ~4% of total annual emissions, were highly variable and on
12 the same order of magnitude as the mean estimate for other northern lakes reported by
13 Bastviken et al. (2004) (2.4 g CH₄ m⁻² yr⁻¹) and Bastviken et al. (2011) (0.8 g CH₄ m⁻² yr⁻¹;
14 pan-Arctic). Storage emission from global lakes ranged from < 0.1 to 37 g CH₄ m⁻² y⁻¹,
15 comprising 0.5% to 81% of the total annual emissions (Bastviken et al., 2011). This also
16 suggests high variability in this emission mode among global lakes. The large relative error
17 for Storage flux measured among our Alaska study lakes (140%; mean \pm SD, 0.5 ± 0.7 g CH₄
18 m⁻² yr⁻¹) confirms that there is large variability associated with this mode of emission;
19 however, CH₄ Storage emissions in our Alaska study lakes were < 2.7 g CH₄ m⁻² yr⁻¹, except
20 in Rosie Creek beaver pond (#25, 39 g CH₄ m⁻² yr⁻¹). The small sample size (n = 2 yedoma
21 lakes) might lead to potential bias in the Storage emissions for yedoma vs. non-yedoma lakes.
22 Further analyses are required to address the differences in Storage emissions between these
23 lake types. Additionally, full or partial turnover of the lake water column in fall can release
24 additional stored CH₄ (Bastviken et al., 2004; Bellido et al., 2009). We acknowledge that our
25 Storage values for CH₄ and CO₂ are gross estimations since we estimated only spring Storage
26 emission and did not take into account potential additional emissions associated with fall
27 turnover or the impacts of lake morphology. Low spatiotemporal resolution sampling to
28 calculate storage emissions also introduces imprecision in our estimates. A better method

1 would involve continuous measurements of dissolved CH₄ and CO₂, temperature and pH in
2 lake water column at multiple locations in the lake throughout the full ice-melt period.

3 **4.2 Geographic patterns of lake CH₄ and CO₂ emissions in Alaska**

4 Previous regional analyses of northern lake emissions found a relationship between
5 CH₄ emissions from lakes and latitude that was explained by temperature (Marotta et al.,
6 2014; Rasilo et al., 2014; Yvon-Durocher et al., 2014). Primary production in warmer
7 climates may supplies more organic substrate for methanogenesis (Duc et al., 2010; Ortiz-
8 Llorente and Alvarez-Cobelas, 2012; Marotta et al., 2014), and methanogenesis is
9 physiologically sensitive to temperature (Schulz et al., 1997; Yvon-Durocher et al., 2014).
10 However, the lakes in these studies were not permafrost-affected. In our N-S Alaska transect
11 we did not find a relationship between any pathway of lake CH₄ emissions and latitude or
12 temperature. We attribute this finding to the presence and geographic diversity of permafrost
13 types (yedoma vs. non-yedoma) (Jorgenson et al., 2008; Kanevskiy et al., 2011), which is
14 more a function of periglacial history and topography in Alaska than it is of latitude or recent
15 climate. While methanogenesis in surface sediments of lakes globally is fueled by
16 contemporary autochthonous primary production and allochthonous organic matter supply
17 (processes typically controlled by latitude and climate in undisturbed systems), thermokarst-
18 influenced lakes have an additional, deeper source of organic matter that fuels
19 methanogenesis: thawing permafrost in the thaw bulbs beneath lakes and along thermally
20 eroding shorelines. Organic matter supplied by thawing permafrost, particularly in lakes
21 formed in thick, organic-rich yedoma-type deposits, can supply more substrate to
22 methanogenesis than the more contemporary organic carbon substrates supplied to surface
23 lake sediments (Kessler et al., 2012).

24 The interior Alaska yedoma lakes, which had the highest CH₄ and CO₂ emissions, are
25 largely thermokarst lakes formed by thaw of organic-rich yedoma permafrost. Radiocarbon
26 ages (18-33 kyr BP) and δD-depleted values of CH₄ in ebullition bubbles collected from the
27 interior Alaskan thermokarst lakes suggested that thaw of late Pleistocene yedoma organic
28 matter fuels methanogenesis in these lakes (Walter et al., 2008; Brosius et al., 2012). The 6-
29 fold difference in CH₄ emissions between yedoma lakes and non-yedoma lakes throughout

1 the rest of Alaska is likely explained by the variability in the availability of recently thawed
2 permafrost organic matter, which provides a larger additional substrate for methanogenesis in
3 the yedoma lakes owing to the thickness (usually tens of meters) of organic-rich yedoma
4 deposits (Kanevskiy et al. 2011; Walter Anthony et al. 2012).

5 Previous research using stable isotopes and radiocarbon dating of CH₄ in ebullition
6 bubbles in yedoma lakes demonstrated that stronger ebullition seeps originate from greater
7 depths beneath the sediment-interface and are characterized by older ¹⁴C ages and more
8 depleted δD values associated with thaw of Pleistocene-aged yedoma permafrost (Walter et
9 al., 2008). The disproportionately large contribution of strong Hotspot ebullition seeps to
10 emissions from yedoma lakes (mean ± SD: 17 ± 12% of total annual emissions) in this study
11 suggests microbial production of CH₄ at greater depths in sediments beneath yedoma lakes.
12 In contrast, the absence of Hotspot ebullition seeps in non-yedoma lakes, which we observed
13 to also have dense sediments, suggests that CH₄ formation by microbial decomposition of
14 organic matter is more restricted to shallower sediment depths in the non-yedoma lakes. This
15 is consistent with maps of permafrost soil organic carbon distributions, whereby the organic-
16 horizons of non-yedoma permafrost soils are typically thinner than yedoma deposits (Ping et
17 al., 2008; Tarnocai et al., 2009; Kanevskiy et al., 2011).

18 The relationship between ebullition, dissolved CH₄ concentration and lake type (Fig.
19 6) also indicates that ebullition seeps releasing CH₄ produced deep in thaw bulbs contribute
20 more to CH₄ cycling in yedoma lakes than in non-yedoma lakes. Yedoma lakes, which had a
21 higher density of ebullition seeps than non-yedoma lakes (Sect. 3.2), had both higher volumes
22 of CH₄-rich bubbles impeded by lake ice and higher concentrations of dissolved CH₄ in the
23 lake water in winter (Fig. 6a, $r_s = 0.72$). Based on Greene et al. (2014), in which 93% of
24 dissolved CH₄ in the water column in winter originated from CH₄ dissolution from ebullition
25 bubbles trapped by lake ice, we attribute the higher concentrations of dissolved CH₄ in the
26 yedoma study lakes to the process of CH₄ dissolution from ice-trapped bubbles. Modeling
27 results, which showed that approximately 80% of CH₄ in bubbles trapped by lake ice in our
28 study lakes dissolved into the water column, support this conclusion. Other important
29 processes that would also control dissolved CH₄ concentrations in lake water are diffusion

1 from sediments and CH₄ oxidation. Given the thicker CH₄-producing sediment package
2 beneath yedoma lakes, we would expect diffusion of dissolved CH₄ from yedoma lakes to be
3 higher than that of non-yedoma lakes. Ex situ incubations by Martinez-Cruz et al. (2015) on a
4 subset of our Alaska study lakes also showed that yedoma lakes had higher CH₄ oxidation
5 potentials, owing in large part to higher concentrations of the dissolved CH₄ substrate in these
6 lakes. Compared to winter, the weaker correlation between dissolved CH₄ and Direct
7 Ebullition in summer (Fig. 6b, $r_s = 0.42$) has several potential explanations. First, in summer,
8 ebullition bubbles escape directly to the atmosphere, so the dissolved CH₄ stock of the water
9 column is not supplied from ice-trapped bubble dissolution like it is in winter unless residual
10 winter-dissolved bubble CH₄ remains in the water column in summer. Second, dissolved CH₄
11 diffusing from lake sediments in summer may be more immediately oxidized by aerobic CH₄
12 consumption since O₂ is more available in lake water from atmospheric diffusion and
13 autochthonous primary production. Finally, higher [SRP](#), TN and Chl-a concentrations in
14 yedoma lakes (Table 1) suggests primary production in yedoma lakes may contribute
15 relatively more substrate to methanogenesis in surface sediments. CH₄ produced in surface
16 sediments more readily escapes to the water column via diffusion than CH₄ produced in thaw
17 bulbs, which preferentially escapes by ebullition (Tan et al., 2014). Higher diffusion from
18 surface sediments would support higher concentrations of dissolved CH₄ in lake water, a
19 process that can be independent of ebullition from thaw bulbs in summer. This explanation is
20 supported by two times higher summer Diffusion emissions from yedoma lakes compared to
21 non-yedoma lakes (Table 2), despite higher observed CH₄ oxidation potentials in yedoma
22 lakes vs. non-yedoma lakes (Martinez-Cruz et al., 2015).

23 CO₂ Diffusion, which was ~100% and 92% of total annual CO₂ emissions from
24 yedoma and non-yedoma lakes respectively, was 6 times higher on average in yedoma lakes
25 than in non-yedoma lakes. Potential explanations include enhanced CO₂ production
26 associated with yedoma organic matter decomposition, photooxidation of the large DOC pool
27 observed in the [mixotrophic](#) yedoma lakes, and potentially higher rates of CH₄ oxidation in
28 yedoma lakes (Martinez-Cruz et al., 2015) generating more CO₂ in the lake water columns.
29 The higher DOC content of yedoma lakes would favor CO₂ production; however, DOC

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1 quality has also been observed to be an important control over CO₂ emissions from northern
2 lakes (Kortelainen et al., 2006). Vonk et al. (2013) recently showed that Pleistocene-aged
3 DOC mobilized in stream water draining yedoma outcrops is exceptionally biolabile among
4 contemporary fluvial systems in the Arctic. This suggests that yedoma-derived DOC in lakes
5 may be more easily decomposed than non-yedoma DOC. Finally, possible differences in
6 watershed sizes draining into lakes could also influence CO₂ concentrations in lakes and
7 Diffusion emissions since terrestrial dissolved inorganic carbon often dominates lake CO₂
8 pools (Kling et al., 1992; Battin et al., 2009; Tranvik et al., 2009). While Kortelainen et al.
9 (2013) found lake water NO₃⁻ concentrations in Finnish lakes to control the ratio of
10 terrestrially-derived CO₂ emissions from lakes versus long-term carbon sequestration in lake
11 sediments, we found no relationship between CO₂ emissions and NO₃⁻ concentrations. Since
12 we did not study long-term carbon sequestration or the other aforementioned processes, and
13 since our calculations contain uncertainty associated with the assumption that single-day
14 measurements of dissolved CO₂ and CH₄ in lakes represent the mean flux for the entire open
15 water period, further research is needed to validate these hypotheses in the Alaskan lakes.

16 **4.3 Dissolved CH₄ and O₂ dynamics**

17 Dissolved O₂ concentration is a useful parameter for predicting the CH₄
18 concentrations in Alaskan lakes. The inverse relationship observed between CH₄ and O₂
19 concentration in lake water (Fig. 5) suggests physical and biological processes govern the
20 availability of these compounds to different degrees in various lakes.

21 There are several possible explanations for the pattern of seasonally higher dissolved
22 CH₄ and lower O₂ concentrations in winter among lakes (Fig. 5): (1) Ice cover inhibits O₂
23 transfer from the atmosphere into the water column (White et al., 2008); (2) Primary
24 production in lakes declines as day length shortens (White et al., 2008; Clilverd et al., 2009);
25 (3) Snow cover impedes light transfer, further extinguishing photosynthesis beneath the ice
26 (Welch et al., 1987; Clilverd et al., 2009); and (4) Finally, aerobic microorganisms consume
27 residual O₂ in the water beneath the ice (Bellido et al., 2009; Clilverd et al., 2009). The
28 resulting anoxic conditions facilitate anaerobic processes like methanogenesis and decrease
29 methanotrophy (Dunfield et al., 1993). All the while, CH₄ is emitted from lake sediments

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1 throughout winter via diffusion and seep ebullition. Many ebullition bubbles are impeded by
2 lake ice, leading to dissolution of CH₄ from bubbles and an increase in dissolved CH₄
3 concentration. In summer, the lack of ice cover allows CH₄ in bubbles to be released directly
4 to the atmosphere without partially dissolving in the lake water column. This explains in part
5 the lower CH₄ concentrations in lake water in summer (Greene et al., 2014). Furthermore, the
6 O₂ concentration in lake water increases in summer by gas exchange with the atmosphere and
7 by primary production in lakes (Fig. 5b). As a result, a fraction of dissolved CH₄ in lake
8 water is emitted to the atmosphere, while methanotrophic activity, supported by elevated O₂
9 concentration, oxidizes another fraction (Martinez-Cruz et al., 2015).

10 In addition to the seasonal variations described above, a permafrost-type effect on
11 dissolved CH₄ and O₂ patterns was also observed. While during summer, most of the non-
12 yedoma lakes were well oxygenated, yedoma lakes in interior Alaska had contrastingly lower
13 O₂ concentrations and higher dissolved CH₄ concentrations beneath the thermocline. This
14 suggests high methanogenic activity in sediments that fuels CH₄ oxidation in the water
15 column. Aerobic methane oxidation together with other aerobic processes reduce O₂
16 concentration under the thermocline, where stratification limits O₂ ingress from superficial
17 water layers.

18 Understanding the dynamics of dissolved CH₄ and O₂ in northern lakes also has
19 relevance to the distribution of lake biota. Ohman et al. (2006) showed that CH₄ concentration
20 in the water column is correlated with fish community composition in lakes, which is easily
21 understood since CH₄ can be used as an indicator of anoxia and therefore, correlated with the
22 fish O₂ requirements.

23 **4.4 Limnological and morphological patterns**

24 Single linear regression analysis indicated that the best limnological predictors of CH₄
25 emissions in the Alaskan lakes were Area, SecD, ~~SRP~~, and TN, all which are indicators of
26 lake metabolism and morphology (Table 4). These findings are consistent with the patterns
27 that explain lake CH₄ emissions in Michigan, Canada, Sweden, and Finland (Bastviken et al.,
28 2004; Juutinen et al., 2009; Rasilo et al., 2014), suggesting that lake trophic state and organic
29 matter quality, rather than carbon concentration alone, might play prevailing roles in CH₄ and

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1 CO₂ production and fluxes. The association between high CH₄ emissions and high nutrients
2 and Chl-a concentrations among yedoma lakes compared to non-yedoma lakes is consistent
3 with the geographic patterns previously observed in Siberian lakes. Higher aquatic production
4 observed in Siberian yedoma lakes compared to non-yedoma lakes in the same climate zone
5 was attributed to fertilization of the yedoma lakes by nitrogen- and phosphorus-rich thawing
6 yedoma permafrost (Walter Anthony et al., 2014). Positive relationships between lake
7 nutrient status and CH₄ fluxes together with low or negative CO₂ fluxes observed in other
8 northern lakes also suggested that lake trophic plays diverging roles in CH₄ and CO₂ fluxes
9 (Del Giorgio et al., 1999; Lapierre and Del Giorgio, 2012). Nutrients can increase primary
10 productivity that simultaneously fuels methanogenesis and draws down dissolved CO₂.

11 The negative correlation between CH₄ emissions and lake area indicates that small
12 lakes had higher total annual CH₄ emissions. This finding is driven by yedoma lakes, which
13 were on average much smaller and tended to develop more noticeable anaerobic hypolimnia
14 than non-yedoma lakes (Table 1, Fig. 5; Supplement Fig. B). This finding is also consistent
15 with lake CH₄ emission patterns in other regions whereby smaller lakes have higher CH₄
16 emissions due to a stronger relative contribution of littoral organic matter to whole-lake
17 methanogenesis (Bastviken et al., 2004; Juutinen et al., 2009; Rasilo et al. 2014).

18 **4.5 Climate warming impacts of Alaskan lake emissions**

19 Previously, Kling et al. (1992) showed that tundra lakes near Toolik Field station emit
20 CH₄ and CO₂ via Diffusion. More recently, Walter Anthony et al. (2012) recognized the
21 importance of CH₄ ebullition from ecological seeps (formed from recent microbial
22 decomposition vs. geologic seeps releasing fossil CH₄) in Alaskan lakes (0.75 Tg CH₄ yr⁻¹);
23 however, this represented the quantity of ebullition seep CH₄ released from sediments rather
24 than the magnitude of atmospheric emissions. Since ebullition emission is partially impeded
25 by lake ice in winter, and a fraction of CH₄ dissolved out of bubbles beneath ice is oxidized
26 by microbes (Greene et al., 2014), ebullition emissions to the atmosphere are lower than what
27 is released annually from sediments. This study is the first to consider multiple modes of
28 emissions for CO₂ and CH₄ together, including the ice-bubble storage process, for a large
29 number of Alaskan lakes spanning large geographic gradients. Scaling total annual CH₄ and

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1 CO₂ emissions observed among yedoma and non-yedoma lakes to the extent of these lake
2 types in Alaska (Walter Anthony et al., 2012) (44 ± 17 g CH₄ m⁻² y⁻¹ x ~8,800 km² yedoma
3 lakes; 8 ± 4 g CH₄ m⁻² y⁻¹ x ~41,700 km², non-yedoma lakes), we estimate that yedoma and
4 non-yedoma lakes emit a total of 0.72 Tg CH₄ yr⁻¹ (~0.39 Tg CH₄ yr⁻¹ from yedoma lakes,
5 0.33 Tg CH₄ yr⁻¹ from non-yedoma lakes). This estimate of Alaska lake emissions increases
6 the previous estimate of Alaska's wetland ecosystem emissions (3 Tg CH₄ yr⁻¹, Zhuang et al.,
7 2007), in which lakes were not included, by 24%. Our estimate of lake CH₄ emission is
8 conservative because it does not include Background (non-seep) Ebullition or Storage
9 emissions associated with fall lake turnover events.

10 If we assume that our study lakes represent the CH₄ and CO₂ emission dynamics of all
11 lakes in Alaska and account for the 34-fold stronger global warming potential of CH₄ vs. CO₂
12 over 100 years (GWP₁₀₀; Myhre et al., 2013), the impact to the climate based on CO₂
13 equivalent (CO₂-eq) emissions from yedoma lakes is ~20 Tg CO₂-eq yr⁻¹ (13 Tg CO₂-eq yr⁻¹
14 from CH₄ and 7 Tg CO₂ yr⁻¹ from CO₂). For non-yedoma lakes, the total climate impact is
15 ~17 Tg CO₂-eq yr⁻¹ (11 Tg CO₂-eq yr⁻¹ from CH₄ and 6 Tg CO₂ yr⁻¹ from CO₂). These results
16 have several important implications. First, CH₄ emissions have nearly twice the impact on
17 climate as CO₂ emissions among all Alaskan lakes. Second, the climate impact of yedoma
18 and non-yedoma lakes in Alaska due to carbon greenhouse gas emissions are approximately
19 equal, despite yedoma lakes comprising less than 1/5 of the total lake area in Alaska. The
20 disproportionately large climate impact of CH₄ emissions from yedoma lakes is due in large
21 part to thaw of deep, organic-rich yedoma permafrost beneath these lakes; however, higher
22 concentrations of total nitrogen, soluble reactive phosphorus and chlorophyll-a in these lakes
23 suggests enhanced primary production in the lakes, which can also fuel decomposition and
24 methanogenesis, as recently demonstrated in Siberia (Walter Anthony et al., 2014). Based on
25 relationships observed in Finnish lakes, it is possible that shifts in nitrate availability could
26 also control the long-term patterns of terrestrially-derived CO₂ emission versus carbon
27 sequestration by our study lakes as well.

28

29 **5 Conclusions**

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1 Total annual CH₄ and CO₂ emissions were dominated by ebullition and diffusion,
2 respectively; however, the climate warming impact of CH₄ emissions was twice that of CO₂.
3 Our 40 study lakes spanned large gradients of physicochemical properties and geography in
4 Alaska. We attribute the 6-fold higher CH₄ and CO₂ emissions observed in thermokarst lakes
5 formed in icy, organic-rich yedoma permafrost in interior Alaska compared to non-yedoma
6 lakes throughout the rest of Alaska to enhanced organic matter supplied from thawing
7 yedoma permafrost, which is typically thicker than the organic-rich strata of non-yedoma
8 soils. Higher total nitrogen, [SRP](#), and Chl-a concentrations in yedoma lakes suggest that
9 higher primary production may also enhance organic substrate supply to decomposition and
10 greenhouse gas production in these lakes. Consideration of multiple modes and seasonality of
11 CH₄ and CO₂ emissions revealed that summer emissions were largest. However, winter and
12 spring emissions of CH₄, including Direct Ebullition through holes in lake ice and the ice-
13 bubble storage and release process, were also significant components of the annual CH₄
14 budget. Our results imply that regional assessments of lake CH₄ and CO₂ emissions in other
15 parts of the pan-Arctic should take into account the myriad of emission modes, lake type and
16 geographic characteristics, such as permafrost type.

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18 **Appendix A: Methods**

19 **A1. Dissolved gas measurements**

20 We used the Headspace Equilibration-Tunable Diode Laser Spectroscopy (HE-
21 TDLAS) technique, described in detail by Sepulveda-Jauregui et al. (2012), to measure the
22 concentration of CH₄ dissolved in lake water. Briefly, we collected water samples using a
23 Van Dorn Bottle (WILDSCO, Yulee, FL, USA) and gently transferred 60 mL into three
24 borosilicate vials (100 mL volume) using disposable polypropylene syringes for triplicate
25 measurements. Vials were immediately sealed with butyl rubber stoppers and aluminum
26 crimp caps. The vials containing the water samples were shaken vigorously for 10 seconds to
27 transfer CH₄ from the water into the vials' headspace for subsequent measurement with the
28 GasFinder 2.0.

1 In addition to HE-TDLAS, we also measured dissolved CH₄ and CO₂ in a subset of
2 samples using the traditional headspace equilibration method by gas chromatography (Kling
3 et al., 1992). Water samples (10 mL) collected with the Van Dorn Bottle were transferred into
4 25 mL glass serum bottles and immediately sealed with butyl rubber stoppers and aluminum
5 crimp caps. Serum bottles were stored upside down and frozen until laboratory analysis. In
6 the laboratory, we thawed the samples to room temperature, shook bottles for 10 seconds to
7 equilibrate headspace and water samples, and then measured CH₄ and CO₂ of the headspace
8 by gas chromatography (Shimadzu GC-2014).

9

10 **A2 Seep ebullition**

11 GPS-mapped ebullition seeps were classified as A, B, C and Hotspot types, based on
12 ice-bubble morphologies. This classification system has been described in detail, with
13 example photographs and bubble morphology classification criteria presented in multiple
14 previous publications (Walter et al., 2006, 2008; Walter Anthony et al., 2010, 2013). Briefly,
15 A-type ebullition seeps are relatively small clusters of ebullition bubbles in which individual
16 bubbles stack on top of each other in the winter ice sheet without merging laterally. Due to
17 progressively higher ebullition rates, individual bubbles of B-type seeps laterally merge into
18 larger bubbles under the ice prior to freezing in ice. Types A and B seeps produce low gas-
19 volume clusters of bubbles in lake ice with cluster diameters typically < 40 cm. The larger C
20 seeps result in large (usually > 40 cm diameter) pockets of gas in ice separated vertically by
21 ice layers containing few or no bubbles. Bubble-trap measurements showed that the solid ice
22 layers in between the large gas pockets of C-type seeps represent periods of relative
23 quiescence in between large ebullition events (Walter et al., 2006; Walter Anthony et al.,
24 2010). Hotspot seeps have the greatest mean daily bubbling rates. The frequency of ebullition
25 release from Hotspot seeps and the associated convection in the water column created by
26 rising bubble plumes can be strong enough to maintain ice-free holes in winter lake ice or ice-
27 free cavities covered by thin layers of ice during cold periods.

28 Thirty-day averages of bubbling rates (mL gas seep⁻¹ d⁻¹) were determined through
29 bubble-trap measurements of seep fluxes and associated with seep classes for each Julian day

1 of the year (Walter Anthony et al., 2010). This data set consists of ~210,000 individual flux
2 measurements made using submerged bubble traps placed over ebullition seeps year-round.
3 These class-specific fluxes were applied to the whole-lake mean densities of seeps on lakes to
4 derive estimates of bubble-release rates from lake bottom sediments indexed by Julian Day.
5 To determine mass-based estimates of CH₄ and CO₂ in ebullition bubbles, we applied lake
6 specific measurements of CH₄ and CO₂ bubble concentrations to the individual lakes where
7 seep-bubble gases were collected and measured. Methods of bubble-trap gas collection and
8 measurements were described in detail by Walter et al. (2008). We sampled with bubble traps
9 and measured by gas chromatography the CH₄ and CO₂ compositions of seep ebullition
10 bubbles collected from up to 246 individual ebullition events per lake. In lakes where few or
11 no seep-bubble gas concentrations were determined, we applied mean values of CH₄ and CO₂
12 by seep class (Walter Anthony et al., 2010): A, 73% CH₄, 0.51% CO₂; B, 75% CH₄, 0.40%
13 CO₂; C, 76% CH₄, 0.55% CO₂; Hotspot, 78% CH₄, 0.84% CO₂. Whole-lake mean ebullition
14 was the sum of seep fluxes observed along an average of five 50-m long transects per lake
15 (median 4 transects per lake), divided by the total area surveyed. In a recent comparison of
16 methods for quantifying ebullition, Walter Anthony and Anthony (2013) showed that when at
17 least three 50-m transects per lake are used to quantify seep ebullition, the estimate of mean
18 whole-lake ebullition is 4-5 times more accurate than the mean flux determined by placement
19 of seventeen 0.2-m² bubble traps randomly distributed across lake surfaces.

20

21 **The Supplement related to this article is available online at [doi:10.5194/bgd-11-1-2014-](https://doi.org/10.5194/bgd-11-1-2014-supplement)**
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23

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3

4 **Author Contributions**

5 K. M. Walter Anthony and A. Sepulveda-Jauregui conceived of the study. A. Sepulveda-
6 Jauregui and K. M. Walter Anthony wrote the manuscript. K. M. Walter Anthony, A.
7 Sepulveda-Jauregui, K. Martinez Cruz and F. Thalasso were responsible for field and lab
8 work. A. Sepulveda-Jauregui conducted statistical analyses. S. Greene modeled ice-bubble
9 storage emissions. All authors commented on the composition of the manuscript.

10

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5

1 **Tables**

2

3 Table 1. Lake physical and chemical properties from 40 Alaskan lakes. N-Lake number; *
4 indicates informal lake names, the A-number refers to lake identification numbers used by
5 Gregory Eaves et al. (2000) for study of the same lakes; Y/NY- permafrost soil type as
6 Yedoma or non-Yedoma^{a,b}; TSI- Trophic State Index^c; EC-Ecozonal Categories^d; Lat-
7 Latitude; Long-Longitude; DN^e-Sedimentary Deposit Name^e; and MD-Maximum known
8 depth; A-Area; SecD-Secchi Depth. Winter and summer temperature [T (Win) and T (Sum)],
9 pH [pH (Win) and pH (Sum)], and oxidation reduction potential [ORP (Win) and ORP
10 (Sum)] are the mean values measured along the full vertical profiles. Summer chlorophyll-a
11 concentrations (Chl-a) are the mean of epilimnion measurements. Water-column nutrient and
12 carbon values (SRP- soluble reactive phosphorus; NO₃⁻-Nitrate; SO₄²⁻-Sulfate; TOC-Total
13 Organic Carbon; TN-Total Nitrogen) are from 1-m depth, except data summarized from other
14 investigators^{g,h}. TOCS-Total organic carbon in surface sediments; TNS-Total nitrogen in
15 surface sediments. Error terms are the standard deviation. ND indicates not determined; CF
16 indicates lake completely frozen; '<' indicates below detection limit, '-' indicates no standard
17 deviation due a sample size of one.

18

Deleted: PO₄³⁻

Deleted: Dissolved phosphate

N	Name	Y/nY ^b	TSI ^c	EC ^d	Lat (°N)	Long (°W)	DN ^e	MD (m)	A (Km ²)	SecD (m)	T (Win) (°C)	T (Sum) (°C)
1	Big Sky* A31	NY	O	ArT	69.581	148.639	ES	2.2	0.349	1.30	0.7 ± 0.2	15.7 ± 0.9
2	Dragon's Pond* A33	NY	O	ArT	68.795	148.843	GF	1.5	0.010	1.30	2.4 ^f ± 2.2	18.4 ± 0.9
3	GTH 112	NY	Mx	ArT	68.672	149.249	GF	4.8	0.025	0.80	2.6 ^f ± 1.1	11.7 ± 3.8
4	NE2	NY	O	ArT	68.647	149.582	GMD	2.7	0.067	2.70	0.4 ± 0.6	15.3 ± 0.6
5	E6	NY	O	ArT	68.643	149.440	GMD	2.6	0.027	2.60	3.3 ^f ± 1.5	15.8 ± 1.0
6	E5 Oil Spill A30	NY	O	ArT	68.642	149.458	GMD	11.9	0.116	3.10	2.8 ^f ± 1.3	10.8 ± 4.2
7	Toolik A28	NY	UO	ArT	68.632	149.605	GMD	24.1	1.449	3.31	2.2 ± 1.1	10.3 ± 4.1
8	E1	NY	UO	ArT	68.626	149.555	GMD	6.4	0.026	2.55	2.4 ± 0.8	12.4 ± 3.7
9	Autumn* A35	NY	UO	ArT	68.462	149.393	GMD	7.5	0.057	4.51	0.45 ^f ± 4.4	13.5 ± 1.9
10	Julietta* A27	NY	UO	ArT	68.447	149.369	GMD	7.0	0.051	3.40	-1.4 ^f ± 2.0	14.3 ± 1.2
11	El Fuego* A36	NY	UO	FoT	67.666	149.716	GMD	2.5	0.057	2.71	2.9 ^f ± 4.5	15.7 ± 1.2
12	Jonas* A26	NY	UO	FoT	67.647	149.722	GMD	4.2	0.170	0.95	-0.2 ± 0.0	14.2 ± 4.8
13	Augustine Zoli* A25	NY	O	FoT	67.138	150.349	F	3.0	0.069	1.12	ND	17.3 ± 1.7
14	Ping*	NY	UO	FoT	67.136	150.370	F	1.4	0.102	1.08	0.1 ± 0.3	18.5 ± 1.7
15	Grayling A24	NY	O	FoT	66.954	150.393	MAC	1.8	0.401	1.80	0.4 ± 0.1	17.0 ± 0.8
16	Eugenia*	Y	Mx	FoT	65.834	149.631	ES	3.3	0.027	0.70	0.5 ± 0.7	17.0 ± 4.0
17	Vault*	Y	Mx	NBF	65.029	147.699	MAC	4.6	0.003	1.00	0.3 ± 0.3	9.5 ± 7.7
18	Goldstream*	Y	Mx	NBF	64.916	147.847	E	3.3	0.010	1.00	1.5 ± 1.5	9.3 ± 6.9
19	Doughnut* ^a	NY	O	NBF	64.899	147.908	E	3.8	0.034	1.59	0.7 ± 0.8	22.2 ± 2.2
20	Killarney*	Y	Mx	NBF	64.870	147.901	E	2.1	0.008	0.50	0.6 ± 0.7	7.8 ± 4.5
21	Smith A13 ^a	NY	Mx	NBF	64.865	147.868	E	4.4	0.094	0.50	0.5 ± 0.7	19.0 ± 1.7
22	Stevens Pond*	Y	Mx	NBF	64.863	147.871	E	1.1	0.002	0.50	CF	17.6 ± 1.6
23	Duece A2	Y	Mx	NBF	64.863	147.942	E	6.0	0.023	0.79	0.9 ± 0.6	11.4 ± 7.0
24	Ace A1	Y	Mx	NBF	64.862	147.937	E	9.0	0.077	1.26	2.9 ± 0.9	11.6 ± 6.3
25	Rosie Creek*	Y	Mx	NBF	64.770	148.079	E	3.7	0.004	1.46	0.0 ± 0.3	11.9 ± 2.4
26	Monasta A37 ^a	NY	Mx	NBF	64.741	148.276	MAC	5.6	0.005	0.43	ND	8.8 ± 5.6
27	91 Lake*	NY	O	NBF	63.848	148.973	F	0.5	0.066	1.40	ND	15.3 ± 0.7
28	Otto	NY	O	FoT	63.842	149.037	GMD	3.1	0.515	1.60	1.6 ± 1.3	12.0 ± 6.4
29	Floatplane* A16	NY	O	FoT	63.394	148.670	GL	5.0	0.103	1.20	3.9 ^f ± 1.5	13.1 ± 1.3
30	Nutella* A39	NY	O	AIT	63.215	147.678	I	9.4	0.020	3.10	3.4 ^f ± 1.1	10.2 ± 3.4
31	Swampbuggy A18	NY	O	FoT	63.055	147.421	GL	4.9	0.142	1.20	3.2 ^f ± 2.3	13.7 ± 0.4
32	Montana A40	NY	O	SBF	62.143	150.048	F	9.0	0.300	2.80	0.8 ± 0.7	16.2 ± 2.4
33	Rainbow Shore* A41	NY	M	SBF	61.694	150.089	GL	11.5	0.575	2.00	0.9 ± 1.0	17.2 ± 1.8
34	Big Merganser A49	NY	O	SBF	60.726	150.644	GL	24.2	0.210	2.00	2.9 ± 1.3	14.4 ± 4.7
35	Rainbow A48	NY	UO	SBF	60.719	150.808	GMD	5.5	0.630	3.00	1.7 ± 1.6	14.8 ± 5.6
36	Dolly Varden A47	NY	UO	SBF	60.704	150.787	GL	30.0	1.074	11.00	2.5 ± 0.2	17.1 ± 0.6
37	Abandoned Cabin* A50	NY	O	SBF	60.696	151.315	GL	3.0	0.031	3.00	1.9 ^f ± 1.6	17.4 ± 1.7
38	Scout A46	NY	O	SBF	60.533	150.843	GL	6.3	0.384	4.00	0.7 ± 0.7	16.4 ± 1.7
39	Engineer A45	NY	O	SBF	60.478	150.323	GMD	3.9	0.909	1.60	0.4 ± 0.6	16.4 ± 1.2
40	Lower Ohmer A44	NY	O	SBF	60.456	150.317	GMD	28.0	0.471	2.70	3.6 ^f ± 0.5	11.6 ± 3.7
	Yedoma ⁱ	-	-	-	-	-	-	4.2 ^k	0.022 ^k	0.82 ^k	1.1 ^{k,m} ± 1.0	11.3 _{k,n} ± 4.5
	Non-Yedoma ^j	-	-	-	-	-	-	7.6 ^k	0.267 ^l	2.39 ^l	1.6 ^{k,o} ± 1.3	14.9 ^l _p ± 3.0

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1 Table 1. cont.

N	Name	pH (Win)		pH (Sum)		ORP (Win) (mV)		ORP (Sum) (mV)		Chl-a ($\mu\text{g L}^{-1}$)	SRP ($\mu\text{g P L}^{-1}$)	NO ₃ ⁻ (mg NO ₃ -N L ⁻¹)
1	Big Sky* A31	7.0	± 0.0	8.8	± 0.7	102	± 18	254	± 78	2.6 ± 3.3	4.2 ^g	< 0.01
2	Dragon's Pond* A33		ND	7.7	± 0.5		ND	304	± 78	4.7 ± 4.2	5.9 ^g	ND
3	GTH 112		ND	7.2	± 0.7		ND	264	± 69	45.9 ± 7.4	ND	< 0.01
4	NE2	6.6	± 0.1	7.9	± 0.6	322	± 17	299	± 66	3.7 ± 4.6	1.3 ^b	ND
5	E6		ND	7.7	± 0.7		ND	272	± 80	5.9 ± 6.2	1.1 ^b	ND
6	E5 Oil Spill A30		ND	7.1	± 0.8		ND	322	± 64	13.5 ± 2.9	1.8 ^b	ND
7	Toolik A28	6.9	± 0.1	7.9	± 0.8	303	± 32	308	± 75	1.5 ± 0.4	1.6 ^b	< 0.01
8	E1	7.0	± 0.1	9.1	± 0.4	283	± 58	231	± 71	1.3 ^g ± -	1.1 ^b	< 0.01
9	Autumn* A35		ND	8.2	± 0.6		ND	303	± 45	2.9 ± 2.4	2.8 ^g	ND
10	Julieta* A27		ND	8.5	± 0.6		ND	318	± 34	3.4 ± 3.8	3.6 ^g	< 0.01
11	El Fuego* A36		ND	8.8	± 0.4		ND	271	± 50	1.2 ± 0.1	ND	ND
12	Jonas* A26	8.2	± 0.0	8.5	± 0.6	23	± 4	250	± 119	1.0 ± 0.0	6.6 ^g	0.02
13	Augustine Zoli* A25		ND	8.7	± 0.6		ND	259	± 80	10.1 ± 11.4	9.8 ^g	< 0.01
14	Ping*	5.9	± 0.0	6.9	± 0.2	211	± 6	303	± 21	22.4 ± 0.0	ND	< 0.01
15	Grayling A24	6.3	± 0.0	7.6	± 0.5	119	± 4	323	± 66	20.7 ± 20.5	5.3	< 0.01
16	Eugenia*	6.3	± 0.0	7.0	± 0.3	118	± 9	314	± 45	41.9 ± 2.4	ND	< 0.01
17	Vault*	7.7	± 0.7	8.6	± 0.8	75	± 62	156	± 87	35.0 ± 15.0	ND	ND
18	Goldstream*	7.4	± 0.6	7.9	± 0.7	117	± 118	216	± 134	31.0 ± 14.5	9.7	0.01
19	Doughnut* ^a	6.8	± 0.1	7.7	± 0.6	189	± 56	254	± 77	113.4 ± 0.0	ND	ND
20	Killarney*	7.0	± 0.1	7.6	± 0.7	66	± 45	316	± 99	ND	10.2	0.01
21	Smith A13 ^a	6.5	± 0.0	8.3	± 1.1	98	± 16	187	± 99	44.7 ± 0.6	16.2 ^g	< 0.01
22	Stevens Pond*		CF	8.4	± 1.7		CF	212	± 136	43.7 ± 13.4	CF	CF
23	Duece A2	7.2	± 0.0	9.2	± 0.4	58	± 10	-20 ± 94	1.5 ^g ± -	60.2 ^g	0.32	
24	Ace A1	7.1	± 0.0	8.1	± 1.0	68	± 15	116	± 161	54.0 ^g ± -	31.5 ^g	0.02
25	Rosie Creek*	7.1	± 0.0	8.1	± 1.0	33	± 19	245	± 127	45.3 ± 1.9	ND	ND
26	Monasta A37 ^a		ND	6.3	± 1.1		ND	160	± 119	ND	24.9 ^g	ND
27	91 Lake*		ND	8.2	± 0.0		ND	351	± 25	ND	ND	ND
28	Otto	7.1	± 0.1	7.8	± 0.5	120	± 141	260	± 59	8.2 ± 11.6	9.8	0.01
29	Floatplane* A16		ND	8.1	± 0.5		ND	349	± 25	27.1 ± 1.3	4.3 ^g	ND
30	Nutella* A39		ND	7.2	± 0.3		ND	384	± 20	13.6 ± 1.4	3.3 ^g	ND
31	Swampbuggy A18		ND	7.3	± 0.0		ND	362	± 1	7.9 ± 0.9	4.7 ^g	ND
32	Montana A40	6.1	± 0.0	7.1	± 0.4	290	± 31	329	± 61	9.5 ± 0.4	2.2 ^g	< 0.01
33	Rainbow Shore* A41	6.5	± 0.3	7.9	± 0.4	289	± 12	305	± 49	7.2 ± 0.9	4.7 ^g	0.02
34	Big Merganser A49	6.4	± 0.4	7.1	± 0.3	321	± 38	325	± 49	7.4 ± 1.1	4.4 ^g	< 0.01

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35	Rainbow A48	7.0	±	0.0	7.7	±	0.6	241	±	62	289	±	85	12.6	±	0.4	4.8 ^e	< 0.01
36	Dolly Varden A47			N D	7.1	±	0.3			N D	282	±	22	3.7	±	0.5	2.1 ^e	< 0.01
37	Abandoned Cabin* A50	6.0	±	0.5	6.3	±	0.2	299	±	113	338	±	33	10.2	±	1.1	2.3 ^e	0.04
38	Scout A46	6.3	±	0.4	7.0	±	0.4	290	±	36	347	±	25	10.9	±	0.4	4.7 ^e	0.01
39	Engineer A45	6.7	±	0.3	7.8	±	0.4	273	±	31	267	±	43	7.0	±	0.2	7.5 ^e	< 0.01
40	Lower Ohmer A44			N D	7.5	±	0.5			N D	379	±	50	9.9	±	0.5	1.8 ^e	< 0.01
	Yedoma ⁱ	7.1 ^k _m	±	0.5	8.2 ^k _m	±	0.9	84 ^{k,m}	±	27	187 ^k _m	±	118	34.5 ^k	±	18.0	27.9 ^k	0.09 ^k
	Non-Yedoma ^j	6.7 ^{lo}	±	0.5	7.7 ^{k,p}	±	0.7	222 ^l _o	±	95	295 ^{lp}	±	51	14.5 ^l	±	21.8	5.3 ^l	0.02 ^k

1

2

1 Table 1. cont.

N	Name	SO ₄ ²⁻ (mg SO ₄ ²⁻ -S L ⁻¹)	TOC (mg L ⁻¹)	TN (mg L ⁻¹)	TOCS (%)		TNS (%)	
1	Big Sky* A31	<0.04	16.48	1.3	1.8	± 0.0	1.5	± 0.3
2	Dragon's Pond* A33	6.20 ^g	16.98	3.2	6.2	± 0.8	2.2	± 0.3
3	GTH 112	0.51	ND	ND	ND		ND	
4	NE2	ND	0.93	0.2	2.9	± 0.5	1.1	± 0.2
5	E6	ND	ND	ND	3.5	± 0.5	1.4	± 0.1
6	E5 Oil Spill A30	<0.04	ND	0.2 ^g	8.1	± 0.1	0.7	± 0.0
7	Toolik A28	<0.04	0.70	0.2	7.8	± 1.3	0.8	± 0.2
8	E1	<0.04	0.18	0.2	ND		ND	
9	Autumn* A35	5.30 ^g	3.66	0.4	ND		ND	
10	Julietta* A27	<0.04	0.71	0.3 ^g	0.8	± 0.8	0.4	± 0.2
11	El Fuego* A36	40.40 ^g	ND	0.4	1.1	± 0.2	0.5	± 0.1
12	Jonas* A26	0.25	0.89	0.7	2.9	± 2.2	1.1	± 0.8
13	Augustine Zoli* A25	<0.04	4.42	0.9	3.0	± 0.4	1.1	± 0.1
14	Ping*	0.18	12.38	0.9	ND		ND	
15	Grayling A24	0.86	8.34	1.0	7.3	± 1.8	0.3	± 0.1
16	Eugenia*	<0.04	16.51	0.8	22.0	± 0.3	ND	
17	Vault*	ND	ND	ND	8.0	± 1.2	ND	
18	Goldstream*	0.30	45.30	3.0	4.2	± 0.6	ND	
19	Doughnut ^a	ND	ND	ND	24.0	± 2.2	ND	
20	Killarney*	0.01	18.12	2.3	3.5	± 2.5	0.2	± 0.1
21	Smith A13 ^a	11.60	ND	1.3 ^g	ND		ND	
22	Stevens Pond*	CF	CF	CF	CF		CF	
23	Duece A2	1.10	ND	2.4 ^g	5.0	± 0.7	1.8	± 0.7
24	Ace A1	0.34	ND	1.3 ^g	2.6	± 2.5	1.0	± 0.9
25	Rosie Creek*	ND	ND	ND	ND		ND	
26	Monasta A37 ^a	ND	58.80 ^g	2.2 ^g	ND		ND	
27	91 Lake*	ND	ND	ND	ND		ND	
28	Otto	0.20	3.63	0.8	8.8	± 1.3	ND	
29	Floatplane* A16	ND	ND	0.5 ^g	ND		ND	
30	Nutella* A39	ND	ND	0.3 ^g	ND		ND	
31	Swampbuggy A18	ND	ND	0.3 ^g	ND		ND	
32	Montana A40	<0.04	0.16	0.3	ND		ND	
33	Rainbow Shore* A41	0.33	52.20	0.1	38.8	± 15.2	ND	
34	Big Merganser A49	12.32	2.38	0.3	ND		ND	
35	Rainbow A48	2.30	1.05	0.2	ND		ND	
36	Dolly Varden A47	1.70	ND	0.2 ^g	6.2	± 0.7	ND	
37	Abandoned Cabin* A50	0.76	ND	0.3 ^g	25.7	± 0.4	ND	
38	Scout A46	0.78	2.58	0.4	23.0	± 0.1	ND	
39	Engineer A45	<0.04	5.71	0.6	7.6	± 1.2	ND	
40	Lower Ohmer A44	2.50	ND	0.3 ^g	ND		ND	
	Yedoma ⁱ	0.44 ^k	26.6 ^k	2.0 ^k	7.6 ^k	± 7.3	1.0 ^k	± 0.8
	Non-Yedoma ⁱ	5.39 ^k	10.1 ^l	0.6 ^l	10.0 ^k	± 10.6	1.0 ^k	± 0.6

2 ^aDoughnut L., a partially-drained lake (uncalibrated ¹⁴C age 1,190 ± 20 yr BP, measured on
3 outer wood of an in situ, dead tree near the lake center), Smith L., and Monasta L. were

1 included in the non-yedoma lake classification. While Doughnut and Monasta lakes likely
2 formed in yedoma permafrost originally, following partial drainage events, they no longer
3 appear to be influenced by active yedoma thaw along the margin. Smith Lake is thought to
4 have formed as part of a previous river drainage network (V. Alexander, pers. com, Aug.
5 2011).

6 ^bPermafrost soil type: Y-Yedoma, NY-Non yedoma.

7 ^cTrophic State Index: UO-Ultraoligotrophic, O-Oligotrophic, M-Mesotrophic, E-Eutrophic,

8 [Mx-Mixotrophic](#),

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9 ^dEcozonal categories according to Gregory Eaves et al. (2000): ArT-Arctic tundra, AIT-
10 Alpine tundra, FoT-Forest tundra, NBF-Northern boreal forest, SBF-Southern boreal forest.

11 ^eDeposit Name: ES-Eolian silt, GF-Glaciofluvial, GMD-old Glacial moraines and drift, F-
12 Fluvial, MAC-Mountain alluvium and colluvium, E-Eolian, GL-Glacio lacustrine (Jorgenson
13 et al., 2008).

14 ^fWinter (October-April) temperature average from Hobo measurements.

15 ^gData from Gregory Eaves et al. (2000)

16 ^hData from Giblin et al. (2009); water-column average.

17 ⁱAverage from yedoma lakes (Lake # 25 excluded).

18 ^jAverage from non-yedoma lakes.

19 ^{k, l}Different letters indicate a significant difference between yedoma and non-yedoma means

20 ^{m, n}Different letters indicate a significant difference between summer and winter means in
21 yedoma lakes for temperature, pH and ORP (Mann-Whitney U test).

22 ^{o, p}Different letters indicate a significant difference between summer and winter means in
23 non-yedoma lakes for temperature, pH and ORP (Mann-Whitney U test).

1 Table 2. Total annual CH₄ and CO₂ emissions by mode from 40 lakes along a North-South
2 latitudinal transect in Alaska. * indicates informal lake names. Eb. Sum.-Direct Ebullition
3 emission to the atmosphere from seeps during the ice-free summer season; Eb. Win.-Direct
4 Ebullition emission to the atmosphere from seeps during the ice-cover winter season; IBS-
5 Ice-bubble storage during spring ice melt; Stor.-Storage emission following ice-out; Diff.-
6 Diffusive emission in summer, Total-Total annual emissions. If there was ND (no
7 determination) for one or more modes in a lake, then total annual emission for the lake is
8 likely an underestimate. Average emissions are summarized at the bottom of the table as is
9 the percent of total annual emissions contributed by each mode as well as statistical results
10 for differences in means among yedoma and non-yedoma lakes (Mann-Whitney test). Error
11 terms represent standard deviation; n number of lakes analyzed; CF-Indicates impossible
12 determination due to lake ice completely freezing to the lake bed in winter. CO₂ diffusive
13 flux from lakes #17 and #18 were estimated from samples taken on multiple dates in June and
14 July 2013 since no data were available in 2011-2012. Different letters^{a, b} indicate a significant
15 difference between yedoma and non-yedoma means.

N	Lake name	CH ₄ (g m ⁻² yr ⁻¹)					Total
		Eb. Sum.	Eb. Win.	IBS	Diff.	Stor.	
1	Big Sky* A31	0.2	0.0	0.1	2.0	2.7	5.0
2	Dragon's Pond* A33	3.0	0.6	0.6	3.2	ND	7.4
3	GTH 112	ND	ND	ND	2.0	0.0	2.0
4	NE2	2.8	0.5	0.5	1.3	0.0	5.1
5	E6	8.8	1.6	1.9	1.0	ND	13.3
6	E5 Oil Spill A30	0.4	0.1	0.1	0.9	ND	1.4
7	Toolik A28	0.6	0.1	0.1	0.9	0.2	2.0
8	E1	5.1	0.9	0.9	2.5	0.0	9.4
9	Autumn* A35	6.9	1.3	1.5	1.0	ND	10.7
10	Julietta* A27	7.5	1.3	1.6	1.9	0.0	12.3
11	El Fuego* A36	10.2	2.0	2.2	ND	ND	14.5
12	Jonas* A26	7.0	1.3	1.4	ND	0.7	10.4
13	Augustine Zoli* A25	9.3	1.7	2.3	4.5	ND	17.7
14	Ping*	5.1	1.0	1.0	1.0	0.9	9.0
15	Grayling A24	1.9	0.4	0.6	2.1	0.0	5.0
16	Eugenia*	ND	ND	ND	6.6	0.6	7.2
17	Vault*	26.6	4.9	4.5	4.8	ND	40.9
18	Goldstream*	13.4	6.7	2.3	6.0	1.9	30.3
19	Doughnut *	ND	ND	ND	3.1	ND	3.1
20	Killarney*	20.7	4.1	14.0	4.4	ND	43.3
21	Smith A13	2.7	0.3	0.4	3.2	0.2	6.7
22	Stevens Pond*	55.0	12.8	8.1	3.1	CF	79.0
23	Duece A2	30.1	4.2	4.6	ND	ND	38.9
24	Ace A1	11.4	2.7	1.5	ND	ND	15.6
25	Rosie Creek*	80.1	17.4	20.5	160.3	39.0	317.4
26	Monasta A37	4.1	0.3	0.7	ND	ND	5.1
27	91 Lake*	1.5	0.2	0.2	2.3	ND	4.2
28	Otto	2.1	0.2	0.3	4.9	0.6	8.1
29	Floatplane* A16	ND	ND	ND	1.1	ND	1.1
30	Nutella* A39	0.1	0.0	0.0	1.1	ND	1.3
31	Swampbuggy A18	3.2	0.3	0.4	0.8	ND	4.8
32	Montana A40	4.1	0.2	0.3	3.5	0.0	8.1
33	Rainbow Shore* A41	3.9	0.2	0.3	ND	0.9	5.4
34	Big Merganser A49	0.5	0.0	0.0	1.8	0.1	2.5
35	Rainbow A48	15.1	0.8	1.3	ND	0.0	17.2
36	Dolly Varden A47	2.4	0.1	0.2	3.2	0.9	6.8
37	Abandoned Cabin* A50	0.4	0.0	0.0	ND	ND	0.5
38	Scout A46	ND	ND	ND	3.6	0.0	3.6
39	Engineer A45	0.0	0.0	0.0	4.9	0.0	4.9
40	Lower Ohmer A44	1.4	0.1	0.1	3.6	ND	5.3
Yedoma (mean ± SD)		26.2 ± 15.9 ^a	5.9 ± 3.6 ^a	5.8 ± 4.6 ^a	5.0 ± 1.4 ^a	1.2 ± 0.9 ^a	44.2 ± 17.0 ^a
Percent		59%	13%	13%	11%	3%	100%
Non-yedoma (mean ± SD)		4.0 ± 3.7 ^b	0.6 ± 0.6 ^b	0.7 ± 0.7 ^b	2.4 ± 1.3 ^b	0.4 ± 0.7 ^a	8.0 ± 4.1 ^b
Percent		50%	7%	9%	30%	5%	100%
All lakes (mean ± SD)						0.5 ± 0.7	

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1 Table 2. cont.

N	Lake name	CO ₂ (g m ⁻² yr ⁻¹)				Total
		Eb. Sum.	Eb. Win.	Diff.	Stor.	
1	Big Sky* A31	0.005	0.001	124	0	124.4
2	Dragon's Pond* A33	0.056	0.010	37	ND	37.1
3	GTH 112	ND	ND	42	ND	41.8
4	NE2	0.048	0.009	ND	ND	0.1
5	E6	0.153	0.028	36	ND	36.2
6	E5 Oil Spill A30	0.006	0.002	44	ND	44.3
7	Toolik A28	0.011	0.002	40	ND	40.5
8	E1	0.088	0.016	ND	ND	0.1
9	Autumn* A35	0.157	0.030	186	ND	186.5
10	Julieta* A27	0.128	0.023	270	ND	269.8
11	El Fuego* A36	0.181	0.036	ND	ND	0.2
12	Jonas* A26	0.122	0.023	ND	0	0.1
13	Augustine Zoli* A25	0.172	0.032	148	0	148.5
14	Ping*	0.097	0.018	34	0	34.2
15	Grayling A24	0.033	0.007	40	0	39.7
16	Eugenia*	ND	ND	131	ND	131.0
17	Vault*	0.445	0.099	1,278	0	1,279
18	Goldstream*	0.261	0.164	1,582	0	1,583
19	Doughnut *	ND	ND	ND	0	0.0
20	Killarney*	0.723	0.070	ND	0	0.8
21	Smith A13	0.052	0.006	251	0	250.9
22	Stevens Pond*	0.991	0.292	144	CF	144.9
23	Duece A2	0.477	0.087	ND	0	0.6
24	Ace A1	0.196	0.059	ND	0	0.3
25	Rosie Creek*	1.462	0.404	1,136	ND	1,138
26	Monasta A37	0.076	0.005	ND	ND	0.1
27	91 Lake*	0.029	0.003	604	ND	604.2
28	Otto	0.040	0.004	234	0	233.9
29	Floatplane* A16	ND	ND	69	ND	69.5
30	Nutella* A39	0.002	0.000	ND	ND	0.0
31	Swampbuggy A18	0.056	0.006	ND	ND	0.1
32	Montana A40	0.076	0.004	143	33	176.4
33	Rainbow Shore* A41	0.075	0.004	ND	48	47.6
34	Big Merganser A49	0.010	0.001	59	ND	58.9
35	Rainbow A48	0.289	0.016	59	ND	59.4
36	Dolly Varden A47	0.047	0.003	65	ND	64.7
37	Abandoned Cabin* A50	0.008	0.000	85	52	137.5
38	Scout A46	ND	ND	64	0	63.9
39	Engineer A45	0.000	0.000	118	0	117.8
40	Lower Ohmer A44	0.027	0.001	157	ND	156.6
Yedoma (mean ± SD)		0.5 ± 0.3 ^a	0.13 ± 0.09 ^a	784 ± 757 ^a	0 ^a	784 ± 757 ^a
Percent		0.07%	0.02%	100%	0%	100%
Non-yedoma (mean ± SD)		0.07 ± 0.07 ^b	0.01 ± 0.01 ^b	127 ± 127 ^b	10 ± 20 ^a	137 ± 129 ^a
Percent		0.05%	0.01%	92%	7%	100%
All lakes (mean ± SD)					7 ± 17	159 ± 322

1 Table 3. The Mann-Whitney and Kruskal-Wallis test results of the limnological and
 2 geographic characteristics of lakes using CH₄ or CO₂ emission mode as the factor. (≠)
 3 indicates a significant difference between limnological property or geographic characteristic
 4 vs. flux; (=) indicates no significant difference at Z value < 1.96. IBS-Ice-Bubble Storage;
 5 Latitude: I-interior, N-northern, S-southern according to Sect. 2.1; Permafrost Soil Type (Y-
 6 yedoma/YN-non-yedoma); Trophic State Index (TSI), Ecozonal Categories (EC), Deposit
 7 type (DN), according to descriptions in Table 1; Maximum depth known (MD) and Area (A).
 8 In the MD analysis we considered two categories: shallow lakes ≤ 2.5 m and deeper lakes >
 9 2.5 m. In the A analysis we considered two categories: small lakes ≤ 0.1 km² and large lakes
 10 > 0.1 km².

Emission mode	Latitude	Y/NY	TSI	EC	DN	MD	A
CH ₄							
Direct Ebullition (Summer)	I ≠ N-S	≠	O ≠ Mx- UO	NBF ≠ ArT-SBF	=	=	≠
Direct Ebullition (Winter)	S ≠ I-N	≠	O ≠ Mx- UO	SBF ≠ FoT-NBF	E ≠ GMD-GL	=	≠
IBS	S ≠ I-N	≠	O ≠ Mx- UO	SBF ≠ FoT-NBF	E ≠ GL	=	≠
Diffusion	I ≠ N	≠	Mx ≠ O- UO	ArT ≠ NBF-SBF	=	=	=
Storage	=	=	=	=	=	=	=
Total	I ≠ S	≠	O ≠ Mx- UO	=	GL ≠ E-GMD	=	≠
CO ₂							
Direct Ebullition (Summer)	I ≠ N-S	≠	O ≠ Mx- UO	NBF ≠ ArT-SBF	E ≠ GMD-GL	=	≠
Direct Ebullition (Winter)	S ≠ I-N	≠	O ≠ Mx- UO	SBF ≠ FoT-NBF	E ≠ GMD-GL	=	≠
Diffusion	I ≠ N	≠	=	NBF ≠ ArT-FoT-SBF	=	=	≠
Storage	=	=	=	=	=	=	=
Total	=	=	=	=	=	=	=

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1 Table 4. Single regression equations for emission modes based on data from Table 1.

Flux/Characteristic	Regression Equation	n	Adjusted r ²	F	p
CH ₄					
Direct Ebullition (summer)	Log(ES-CH ₄)= -0.50Log(Area)	32	0.30	14.4919	0.0006
Direct Ebullition (winter)	Log(EW-CH ₄)= -0.93 - 0.68Log(Area)	28	0.60	43.6036	0.0000
	Log(EW-CH ₄)= 0.10 - 1.12Log(SecD)	28	0.23	9.3352	0.0050
	Log(EW-CH ₄)= -2.63 + 0.81Log(TN)	24	0.32	12.4092	0.0018
IBS	Log(IBS-CH ₄)= -0.83 - 0.64Log(Area)	29	0.58	50.705	0.0001
	Log(IBS-CH ₄)= 0.10 - 1.00Log(SecD)	29	0.19	7.9309	0.0088
Diffusion	Log(DF-CH ₄)= 0.55Log(SRP)	24	0.40	16.7767	0.0004
Total	Log(Tot-CH ₄)= 0.43 - 0.37Log(Area)	38	0.27	15.0877	0.0004
	Log(Tot-CH ₄)= 1.01 - 0.77(SecD)	38	0.21	11.1414	0.0019
	Log(Tot-CH ₄)= 0.42 + 0.55Log(SRP)	30	0.22	9.4969	0.0045
	Log(Tot-CH ₄)= 0.98 - 0.61Log(TN)	32	0.29	13.7928	0.0008
CO ₂					
Direct Ebullition (summer)	Log(ES-CO ₂)= -1.72 - 0.50Log(Area)	32	0.30	14.6253	0.0006
Direct Ebullition (winter)	Log(EW-CO ₂)= -2.78 - 0.76Log(Area)	30	0.63	52.0960	0.0000
	Log(EW-CO ₂)= -1.83 - 0.76Log(TN)	26	0.24	9.0882	0.0058

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1 Table 5. Mann-Whitney and Kruskal-Wallis test results of the limnological and geographic
 2 characteristics of lakes using mode of dissolved gases concentrations (CH₄, O₂) during winter
 3 and summer. (≠) indicates a significant difference between a geographic characteristic and
 4 flux when Z > 1.96; (=) indicates no significant difference. Latitude: I-interior, N-northern, S-
 5 southern according to Sect. 2.1; Permafrost Soil Type (Y-yedoma/NY-non-yedoma); Trophic
 6 State Index (TSI), Ecozonal Categories (EC), Deposit type (DN) according to descriptions in
 7 Table 1; Maximum depth known (MD) and Area (A). In the MD analysis we considered two
 8 categories: shallow lakes ≤ 2.5 m and deeper lakes > 2.5 m. In the A analysis we considered
 9 two categories: small lakes ≤ 0.1 km² and large lakes > 0.1 km².

10

Dissolved Gas (Season)	Latitude	Y/NY	TS	EC	DN	MD	A
CH ₄ (Winter)	I ≠ S	≠	Mx ≠ O	=	E ≠ GL, GMD	≠	≠
CH ₄ (Summer)	I ≠ N, S	≠	Mx ≠ O, UO	NBF ≠ ArT, SBF, FoT	E ≠ GMD	=	≠
O ₂ (Winter)	I ≠ S	≠	Mx ≠ O	=	E ≠ GL, GMD	=	≠
O ₂ (Summer)	I ≠ N, S	≠	Mx ≠ O, UO	NBF ≠ ArT, SBF, FoT	E ≠ GL, GMD	=	≠

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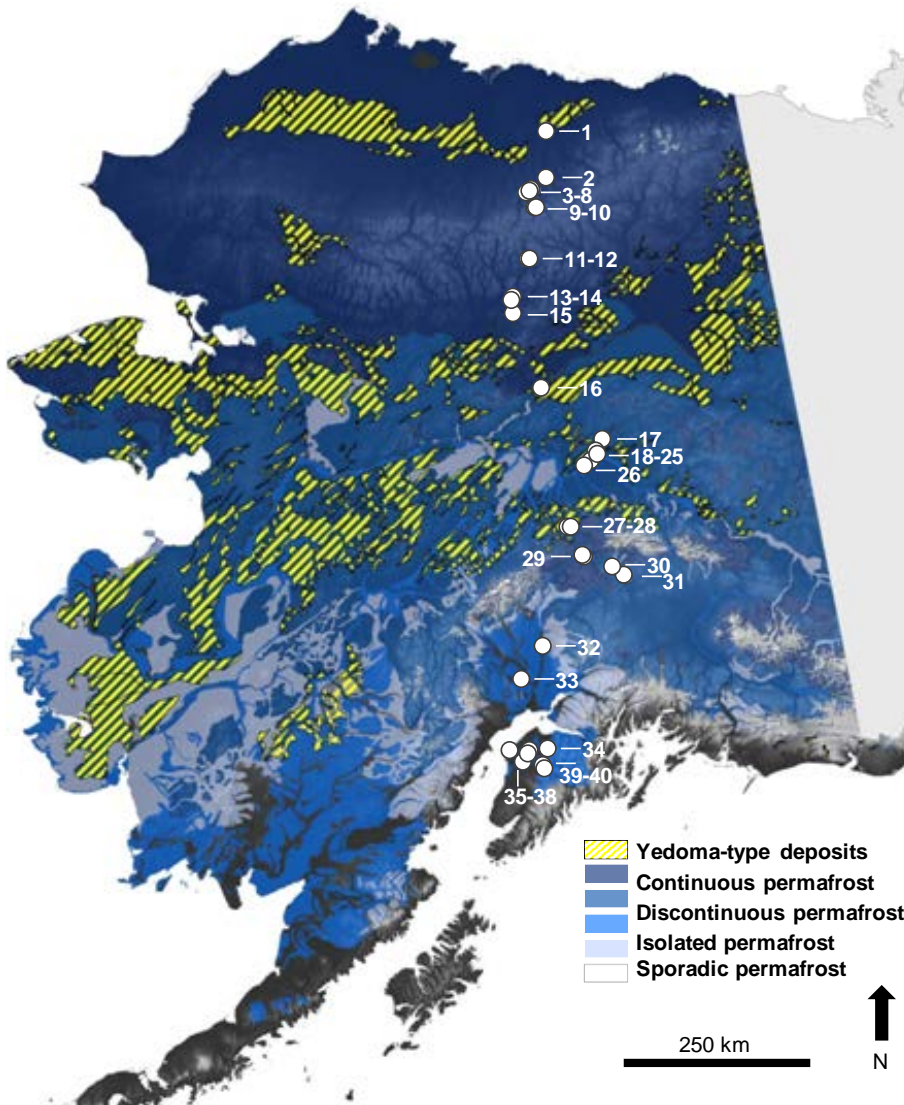
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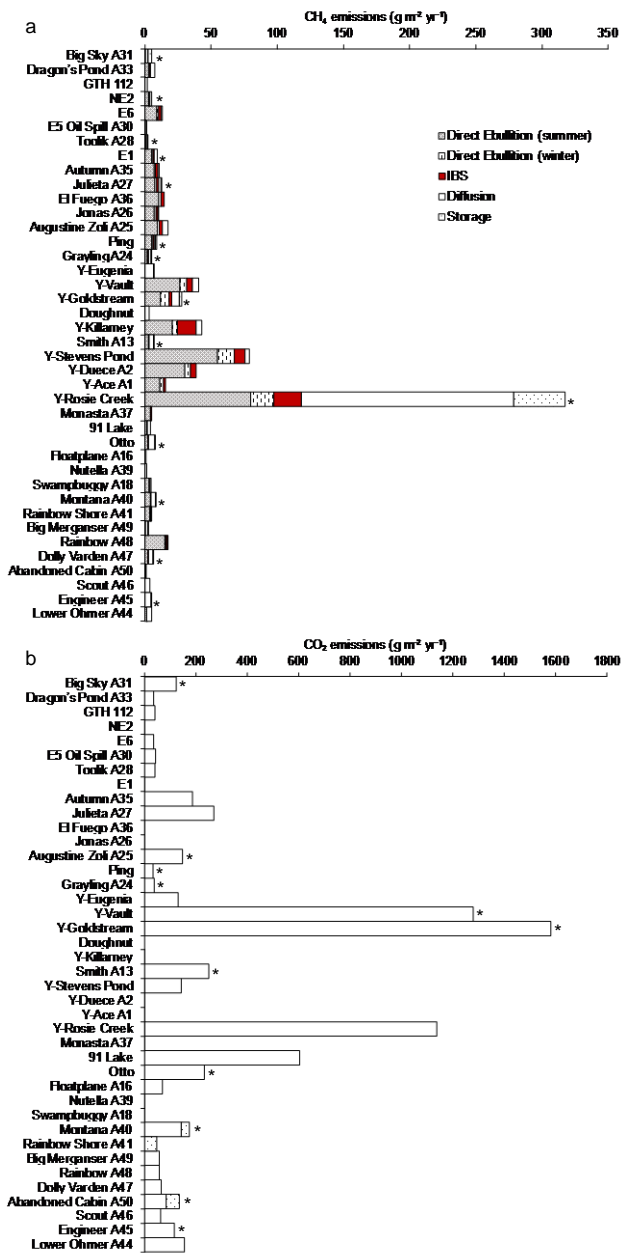
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1 **Figures**

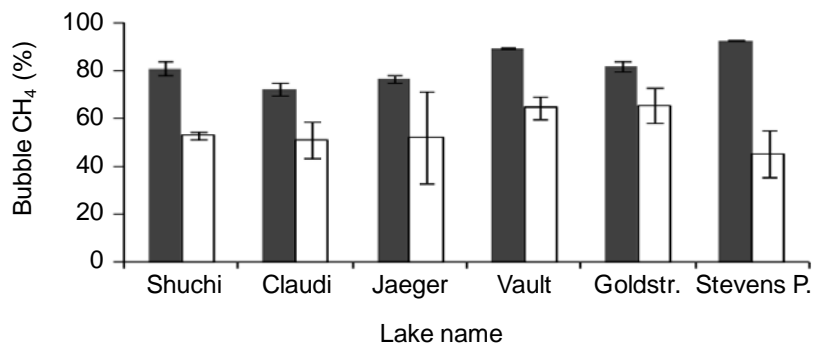


2
3 Figure 1. Locations of study lakes in Alaska (circles) plotted on the Alaska DEM hillshade
4 raster. Information about the distribution of yedoma-type deposits (ice-rich silt containing
5 deep thermokarst lakes) and permafrost was from Jorgenson et al. (2008) and Kanevskiy et
6 al. (2011). The Alaska map is the National Elevation Data Set 30 m hillshade raster.

7



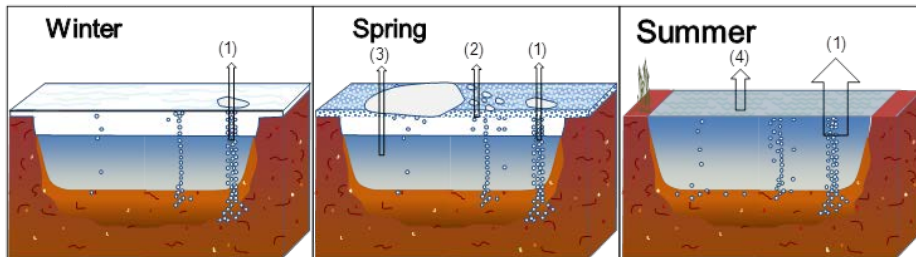
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 2 Figure 2. Total annual CH₄ (a) and CO₂ (b) emissions by mode from 40 lakes along a North-
 3 South latitudinal transect in Alaska. Yedoma lakes are indicated by 'Y'. Lakes for which all
 4 emission modes were measured are indicated by '*' (see Table 2). Panels a and b follow the
 5 legend shown in 'a'.



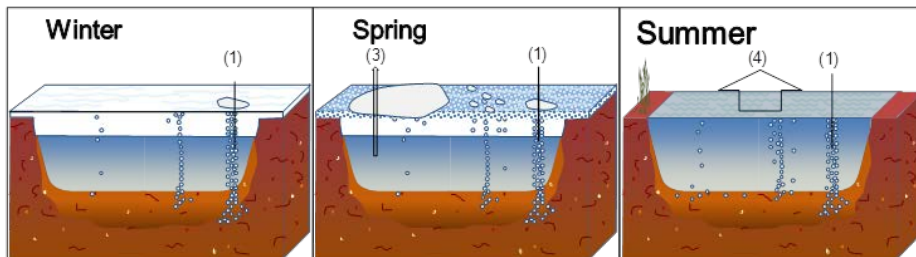
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 2 Figure 3. Average CH₄ concentrations in ebullition bubbles collected at the lake surface
 3 before interaction with lake ice ('fresh bubbles', grey bars) and in ebullition bubbles trapped
 4 by the lake ice (white bars). Error bars represent standard error for n = 2 to 41 seeps per lake.
 5 Among lakes, CH₄ concentrations in ice-trapped bubbles were 33 ± 12% lower than in fresh
 6 bubbles (Mann-Whitney U test, Z > 1.96, p < 0.05).

7
 8

CH₄

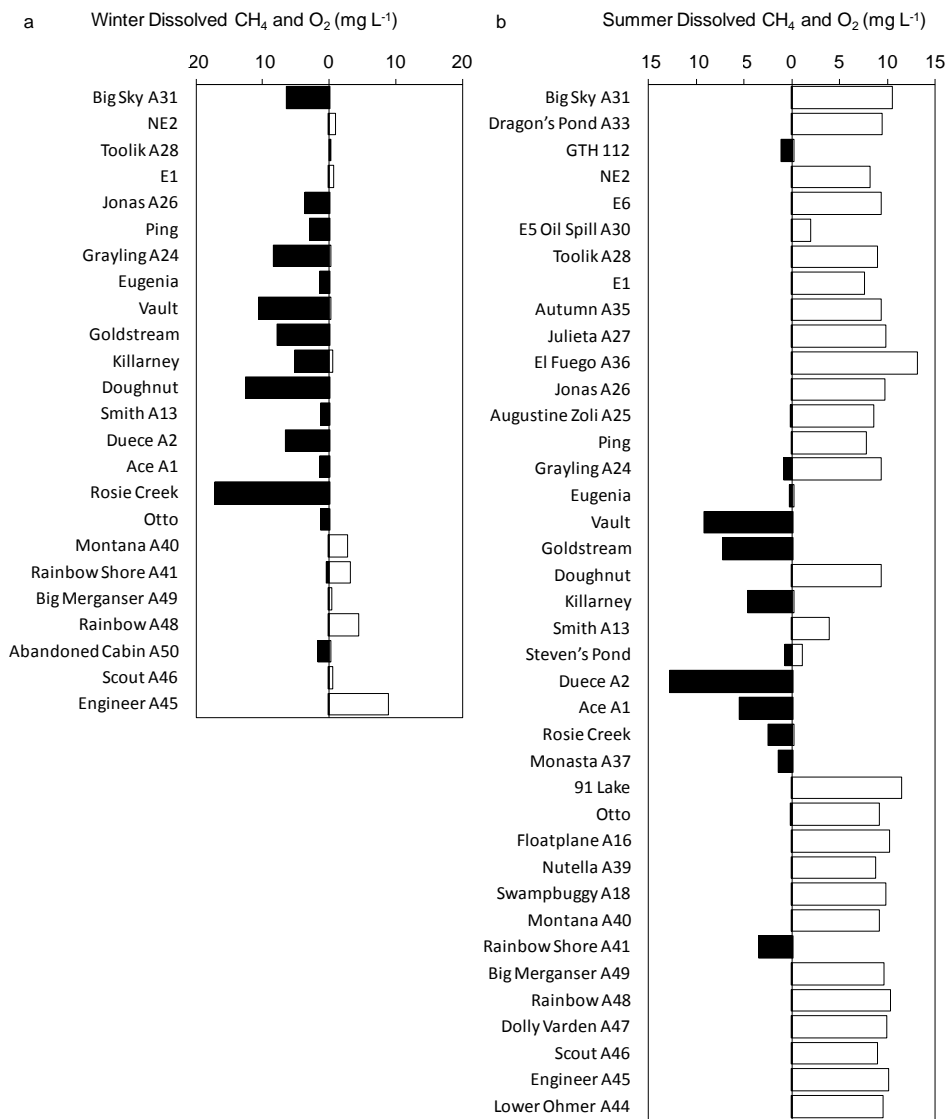


CO₂



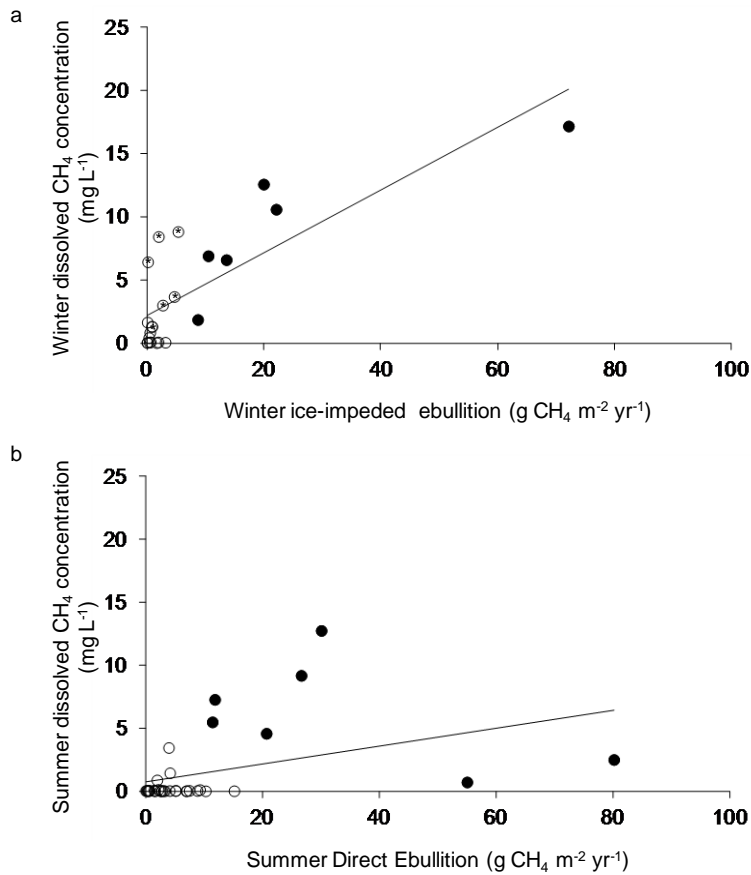
1
2 Figure 4. Illustration of CH₄ and CO₂ emissions pathways during different seasons in Alaskan
3 lakes. The thickness of arrows indicates the relative magnitude of contribution from each
4 pathway according to Table 2: (1) Direct Ebullition through ice-free Hotspot seeps in winter
5 and from all seep classes during the last month of ice cover in spring and in summer; (2) Ice-
6 Bubble Storage (IBS) emission during spring ice melt; (3) Storage emission of dissolved
7 gases accumulated under lake ice when ice melts in spring; (4) Diffusion emission from open
8 water in summer.

9



1
 2 Figure 5. Average dissolved CH₄ (black bars) and O₂ (white bars) concentrations in lake
 3 bottom water during winter (a) and summer (b). Yedoma lakes are indicated by 'Y'. In
 4 winter, Spearman coefficient $r_s = 0.58$ indicates a moderate positive correlation between
 5 dissolved CH₄ and O₂; in summer $r_s = 0.70$ indicates a strong positive correlation.

6



1

2 Figure 6. Dissolved CH₄ concentrations measured in lake bottom water vs. winter ice-
 3 impeded ebullition in winter (a) and Direct Ebullition in summer (b). The Spearman
 4 coefficients, $r_s = 0.72$ and $r_s = 0.42$ indicate a strong positive correlation and a weak positive
 5 correlation in winter and summer, respectively. All lakes were considered a single
 6 population; however, yedoma lakes (closed circles) had higher concentrations of lake-bottom
 7 dissolved CH₄ (mean \pm SD: 9.3 ± 5.4 mg L⁻¹ winter, 6.7 ± 4.1 mg L⁻¹ summer) and a higher
 8 density of ebullition seeps (Sect. 3.2) than non-yedoma lakes (open circles; 2.1 ± 3.0 mg L⁻¹
 9 winter, 0.3 ± 0.7 mg L⁻¹ summer). We observed relatively high concentrations of dissolved
 10 CH₄ in some non-yedoma lakes in winter due to dissolved gas exclusion during ice formation
 11 in shallow lakes that nearly froze to the lake bed, indicated by *. Excluding lakes that nearly
 12 froze to the lake bed, the mean dissolved CH₄ in the remaining non-yedoma lakes was $0.3 \pm$
 13 0.5 mg L⁻¹ in winter.