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Methane and carbon dioxide emissions from 40 lakes along a north–south latitudinal transect in Alaska

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Abstract

Uncertainties in the magnitude and seasonality of various gas emission modes, particularly among different lake types, limit our ability to estimate methane (CH_4) and carbon dioxide (CO_2) emissions from northern lakes. Here we assessed the relationship between CH_4 and CO_2 emission modes in 40 lakes along a latitudinal transect in Alaska to physicochemical limnology and geographic characteristics, including permafrost soil type surrounding lakes. Emission modes included Direct Ebullition, Diffusion, Storage flux, and a newly identified Ice-Bubble Storage (IBS) flux. We found that all lakes were net sources of atmospheric CH_4 and CO_2, but the climate warming impact of lake CH_4 emissions was two times higher than that of CO_2. Ebullition and Diffusion were the dominant modes of CH_4 and CO_2 emissions respectively. IBS, ~ 10 % of total annual CH_4 emissions, is the release to the atmosphere of seasonally ice-trapped bubbles when lake ice confining bubbles begins to melt in spring. IBS, which has not been explicitly accounted for in regional studies, increased the estimate of springtime emissions from our study lakes by 320 %. Geographically, CH_4 emissions from stratified, dystrophic interior Alaska thermokarst (thaw) lakes formed in icy, organic-rich yedoma permafrost soils were 6-fold higher than from non-yedoma lakes throughout the rest of Alaska. Total CH_4 emission was correlated with concentrations of phosphate and total nitrogen in lake water, Secchi depth and lake area, with yedoma lakes having higher nutrient concentrations, shallower Secchi depth, and smaller lake areas. Our findings suggest that permafrost type plays important roles in determining CH_4 emissions from lakes by both supplying organic matter to methanogenesis directly from thawing permafrost and by enhancing nutrient availability to primary production, which can also fuel decomposition and methanogenesis.

1 Introduction

Lakes are an important source of atmospheric greenhouse gases, methane (CH_4) and carbon dioxide (CO_2) (Battin et al., 2009; Tranvik et al., 2009; Bastviken et al., 2011). In lakes CH_4 and CO_2 have contrasting patterns of production, consumption, and exchange with the atmosphere. CH_4 is produced in anaerobic environments (mainly in sediments), while CO_2 in lakes originates from respiration throughout the water column and sediments, inflow of terrestrially derived dissolved inorganic carbon from surrounding watersheds, and photooxidation of dissolved organic carbon (DOC) (Graneli et al., 1996; Battin et al., 2009; Tranvik et al., 2009). CO_2 is also formed in lakes by aerobic oxidation of CH_4, a process that oxidizes 30–99 % of CH_4 produced in lakes (Bastviken et al., 2008; Thauer et al., 2008). Meanwhile, CO_2 is consumed by photosynthesis and other autotrophic or chemical processes that depend on pH and/or the availability of light (Madigan et al., 2009).

Despite recycling of CH_4 and CO_2 internally in lakes, a significant quantity of these greenhouse gases is released from lakes to the atmosphere (Cole et al., 2007). Most of Earth's lakes are located in northern high latitudes, overlapping the permafrost-dominated region (Downing et al., 2006; Smith et al., 2007; Grosse et al., 2013). It is estimated that CH_4 emission from lakes globally comprises about 16 % (71.6 Tg) of all human and natural atmospheric sources (Bastviken et al., 2011), and that northern lakes (> 55° N) contribute about 20 % of these emissions (13.6 Tg; Bastviken et al., 2011). Constraining CO_2 emissions is challenged by variability in patterns of CO_2 partial pressure mainly due to photosynthesis, inputs from terrestrial ecosystems, and mineralization of the organic matter (Kling et al., 1991; Battin et al., 2009; Tranvik et al., 2009). CO_2 emissions from northern lakes constitute approximately 43 % (1.2 Pg CO_2) of global emissions from lakes (Battin et al., 2009; Tranvik et al., 2009; Maberly et al., 2013). Due to a disproportionately low number of northern high latitude lakes represented in previous studies of global CH_4 emissions (Bastviken et al., 2011), and

a paucity of studies that considered various modes of emission together, CH_4 and CO_2 emissions from northern high latitude lakes are still poorly constrained.

Geographic diversity in Alaska provides a valuable opportunity to study CH_4 and CO_2 emission patterns from lakes exhibiting wide variability in lake origin, climate, ecology, geology, and permafrost coverage. Across Arctic, Continental, and Transitional climate zones in Alaska, ecological habitats include arctic, alpine and forest tundra, and northern and southern boreal forests (Gregory-Eaves et al., 2000). The surficial geology in which Alaskan lakes are found varies primarily from fine-grain aeolian deposits; to coarser-grain coastal, glacial, fluvial and volcanic deposits; to rubble and bedrock (Karlstrom et al., 1964; Arp and Jones, 2009). Alaska is also characterized by a variety of permafrost types (Fig. 1) ranging from isolated permafrost in south-central Alaska to continuous permafrost in northern Alaska (Jorgenson et al., 2008).

Within the context of permafrost soil carbon content, Alaskan lakes can be classified depending on whether they are surrounded by yedoma-type permafrost or non-yedoma substrates (Walter Anthony et al., 2012). Yedoma is typically thick (tens of meters), organic-rich (~1 % C by mass), Pleistocene-aged loess permafrost with ice content of 50–90 % by volume which deep thermokarst (thaw) lakes with high CH_4 production potentials form when ground ice melts (Zimov et al., 1997; Kanevskiy et al., 2011; Walter Anthony and Anthony, 2013). Non-yedoma permafrost can also have high organic carbon and excess ice concentrations within several meters of the ground surface; however, these organic-rich, permafrost horizons are typically thinner than yedoma deposits (Ping et al., 2008; Tarnocai et al., 2009). As a result, thermokarst lakes formed in non-yedoma permafrost soils are commonly shallower than yedoma lakes and may produce less CH_4 (West and Plug, 2008; Walter Anthony and Anthony, 2013; Grosse et al., 2013).

Estimating CH_4 and CO_2 emissions from northern high latitude lakes, which are seasonally covered by ice, represents a difficult task because there are at least four emission pathways, all of which have not been consistently and simultaneously measured

1
contrasting with what?

2
this is a commentary paper, so not the best citation to support this sentence. Check all other citations of Battin et al. 2009 below as well, if relevant.

3
Bastviken et al. 2008 estimated the importance of MOx in 3 lakes, and Thauer et al. 2008 is a paper on methanogeny (maybe this paper suggest oxidation rates? if so, on how many lakes was this done??). I suggest you tone down your sentence as this cannot be generalised! It depends on so many things and it could be anything from 0 to 100%.

4
can you provide examples of these "chemical processes"?

5
no need to cite this paper 2 times within one sentence

6
would be interesting to discuss why northern emissions represent 20% of global lake CH4 emissions, but 43% of global lake CO2 emissions.

7
a better link between these 2 sentences is missing

8
what do you mean by geographic diversity?? what does geography include? Might be useful to
define early in the paper cause it's been used extensively.

9
in this sentence structure, it looks like these characteristics all pertains to "lake" but a few does not fit (rather to the landscape)

Nombre : 10
are you talking specifically about organic carbon?

11
is this really organic-rich? Aren't organic soils defined when they have >20% of organic C?

12
this is a rather long sentence...

13
are you putting all other pmf soils in non-yedoma type or only the "organic-rich" ones as it seems to be qualified in the second part of the sentence? Because non-yedoma soils could include for ex. bare rock, right? This non-yedoma classification looks rather vague to me. Specifically after
reading non-yedoma definition at section 2.1: would this include oligotrophic rocky lakes as well for ex.?

14
icy or ice-rich?

15
they may or they have been shown to produce less CH4?

in the past: (1) Direct Ebullition, (2) Diffusion, (3) Storage flux, and a newly identified (4) Ice-Bubble Storage (IBS) flux (Greene et al., 2014).

Ebullition (bubbling) has been observed as the dominant pathway of CH_4 emissions from many lakes (Casper et al., 2000; Bastviken et al., 2004; Walter et al., 2006). Since CH_4 is relatively soluble, high concentrations of sediments lead to bubble formation and emission to the atmosphere by ebullition. In contrast, CH_4 diffusion flux to the atmosphere is usually relatively low and occurs mainly in summer when ice cover is absent. Due to much higher solubility, CO_2 tends to occur in low concentrations in ebullition bubbles, and instead escapes lakes predominately by Diffusion (Abril et al., 2005).

During winter, ice formation on most northern lakes impedes gas emissions to the atmosphere. Dissolved CH_4 and CO_2 accumulate in the lake water column beneath the ice, resulting in gas "storage." Storage emissions occur when dissolved CH_4 and CO_2 are emitted by diffusion when the ice melts in spring, often enhanced by full or partial lake overturn (Michmerhuizen et al., 1996; Phelps et al., 1998; Bellido et al., 2009). Storage emissions also occur in some lakes in autumn, if lake overturn caused by falling temperature brings high concentrations of dissolved gases from deeper water depths to the surface, resulting in rapid CH_4 and CO_2 emission by diffusion from the water column. Bastviken et al. (2004) coined the term "Storage flux" when they considered it in regional lake emission estimates as a function of differences in water column CH_4 stocks before and after lake ice-out, CH_4 production rates, and loss by CH_4 oxidation.

The fourth potential emission component involves CH_4 release to the atmosphere from ice-trapped ebullition bubbles in spring before the ice disappears. This mode of emissions occurs when lake ice begins to degrade and thin ice lenses that previously sealed CH_4 bubbles in and under ice during winter melt, releasing bubble gas to the atmosphere (Greene et al., 2014). Gas in small, tubular bubbles formed in lake ice by exclusion of dissolved gases as ice freezes (Gow and Langston, 1977; Langer et al., 2014) is presumably released to the atmosphere when ice degrades as well; however,

given the substantially lower concentration of CH_4 in these non-ebullition, freeze-out bubbles (usually < 0.01 % by volume; Boereboom et al., 2012), this mode of emission is relatively insignificant in comparison to the larger ebullition-sourced bubbles, in which CH_4 concentrations typically range from 40–90 % by volume (Martens et al., 1992; Semiletov et al., 1996; Walter Anthony et al., 2010). Ponded water on the lake-ice surface can enhance the release of ice-trapped bubbles to the atmosphere and also provides the opportunity for visual observation of gas release from bubbles trapped by degrading ice (K. M. W. A. unpublished data, 2014). In this study, we investigated this emission pathway, which Greene et al. (2014) called "Ice-Bubble Storage" (IBS).

Finally, it is important to understand how changes in nutrient availability and temperature influence CO_2 and CH_4 cycling in lakes. Increasing nutrients and temperature stimulates primary production and microbial decomposition of organic matter, which in turn consumes oxygen (O_2) and enhances anaerobic decay processes, particularly in sediments, where CH_4 and CO_2 are produced (Conrad et al., 2010). The presence of O_2 facilitates aerobic CH_4 oxidation, but the efficiency of this process is controlled directly by O_2 and CH_4 concentrations and temperature (Utsumi et al., 1998; Bastviken et al., 2002; Borrel et al., 2011) and indirectly by nutrient availability (Dzyuban et al., 2010). Measurement of O_2 and CH_4 concentrations in lakes are essential for assessing global carbon cycling, and in this framework, correlating both parameters in situ has recently been promoted as an indirect means of assessing CH_4 oxidation by methanotrophs (Bastviken et al., 2004; Guerin and Abril, 2007; Sepulveda-Jauregui et al., 2012).

In this study we assessed the relationships between measured CH_4 and CO_2 emission modes in 40 lakes along a north–south Alaska transect to the lakes' physicochemical limnology and geographic characteristics. Our goal was to assess the magnitude, variability and seasonality of individual modes of emission, particularly among the wide range of geographic lake settings in Alaska.

Page : 3

1
less soluble, not INsoluble

2
CH4 is produced in interstitial water

3
why using capital letter for these processes (above and below)?

4
rewrite: "and their emission to the atmosphere."

5
not any type of deeper water, but especially when hypolimnion is formed

6
check structure of sentence

7
this is hard to understand, can you clarify? and what is winter melt?

8
can you explain why?

9
not only this one

10
I think you may not want to mention O2 here because of the begginning of the sentence (problem of logic): if O2 facilitates methane oxidation, it's redundant to say it increases the efficiency, especially after a "but"

11
recently? (citations are from 2004)

12
physicochemical properties or characteristics (check throughout the ms)

2 Materials and methods

2.1 Study lakes and permafrost zones

We sampled water from 40 Alaskan lakes during open-water conditions in June–July 2011 and 2012 (Fig. 1) and from 26 of the lakes toward the end of the winter ice-cover period in March–April 2011. Our study lakes were located near the road system along a north–south transect in Alaska that spans a variety of geographic and limnological settings, described previously by Gregory-Eaves (2000), Jorgenson et al. (2008), and Walter Anthony et al. (2012). Our study lakes occupied three general climatic/permafrost zones: (1) the northern study area (66–70° N, Arctic climate/continuous permafrost), (2) the interior study area (64–66° N, Continental climate/discontinuous permafrost), and the southern study area (60–64° N, transitional climate/sporadic and isolated permafrost) (Gregory-Eaves et al., 2000, Jorgenson et al., 2008). Additionally, we distinguished yedoma-type thermokarst lakes as those formed in yedoma permafrost with active, ongoing thermokarst activity from non-yedoma type lakes, which were lakes occurring in all other non-yedoma deposits in permafrost and non-permafrost soils (Fig. 1). Lake names, sizes, geographical and limnological characteristics are shown in Table 1.

2.2 Water-dissolved CH_4, CO_2 and O_2

Offshore and usually near the center of each lake, we sampled lake water at one to nine distributed depths throughout the water column for dissolved CH_4 and CO_2 concentrations and at 0.5 m depth intervals for O_2 concentrations during winter and summer. In lakes shallower than 1 m we sampled only one depth within 25 cm of the lake bottom. In the field we measured CH_4 concentration by the Headspace Equilibration-Tunable Diode Laser Spectroscopy (HE-TDLAS) method (Sepulveda-Jauregui et al., 2012) using a GasFinder 2.0 (Boreal Laser Inc., Edmonton, Canada; Appendix A). Additionally, we determined concentrations of headspace CH_4 and CO_2 in bottles of lake water in

the laboratory following Kling (2010) using a GC-2014 gas chromatograph (Shimadzu, Addison, Illinois, USA) equipped with a flame ionization detector and a PLOT alumina column (detector temperature 250 °C, oven 40 °C, high purity Helium as carrier gas). Dissolved O_2 concentrations were measured in the field with a luminescence sensor connected to a calibrated multiparametric probe Hydrolab DataSonde (Hach LDO, Loveland, Colorado, USA).

2.3 CH_4 and CO_2 diffusion flux

We estimated the Diffusion flux of CH_4 and CO_2 ($g\,m^{-2}\,yr^{-1}$) during summer by applying Fick's Law to our measurements of dissolved CO_2 and CH_4 in surface water following the boundary layer method of Kling et al. (1992):

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

where T is the conversion factor from seconds to years (31 536 000); D is the molecular diffusivity of CH_4 or CO_2 ($m^2\,s^{-1}$) following Kling et al. (1992); z (m) is the thickness of the surface boundary layer, assumed to be 200 µm as an average for Alaskan lakes following Kling et al. (1992); C_w is the measured gas concentration at the bottom of the boundary layer ($g\,m^{-3}$); C_{eq} is the equilibrium gas concentration in surface lake water ($g\,m^{-3}$) exposed to the atmosphere at the top of the boundary layer, calculated using Henry's Law constants at 22 °C of 1.47×10^{-3} and 3.61×10^{-2} $mol\,L^{-1}\,bar^{-1}$ for CH_4 and CO_2, respectively (NIST, 2011).

2.4 Storage flux

To estimate Storage flux, dissolved CH_4 and CO_2 profiles were measured in spring before the ice began to melt and in summer during ice-free conditions. We multiplied the average concentration of dissolved CH_4 and CO_2 measured in samples collected from distributed depths in the water column by the height of the unfrozen water column. Storage flux ($g\,m^{-2}\,yr^{-1}$) was calculated as the difference between total mass

1
It would be interesting to see the correlation between summer and winter limno data (is summer representative of winter? is summer sampling sufficient to characterize the lake limno?)

2
does it only involve thermokarst lakes or any other types of lakes? if any other, is the sampling representative of all types of lakes found in such a climate gradient?

3
at what time of the day did you sample lake water? (diurnal variations, especially of CO2)

4
did you find a good correlation between GasFinder measurements and bottle headspace? (if they were taken simultaneously)
Interesting result to provide.

5
why converting to year flux already here if diffusion is stopped under the ice cover?
Are you using the one summer measurement per lake to extrapolate over the year (but for the open water season I guess??).
This needs to be clarified.

6
Basically you assume there is NO change in gas echange coefficient over the day/weeks/year… This is an assumption that needs to be discussed and acknowledged (or maybe you do so lower? if so, please ignore this comment). I am convinced there are large variations in turbulence; the wind is certainly not that constant and heat exchange (at least over a day cycle) is likely quite variable (heat exchange also affects gas exchange, see for ex. Tedford et al. 2014).
One thing to note is that Kling et al. 1992 were using monthly wind speed averages calculated from daily averages! this is generating constancy artificially.
Another thing is that Kling et al. mentioned that this value of 200 Õm is likely overestimated... (thus flux would be conservative). This will influence the relative importance of diffusion.
Wind speed changing by a factor of 2 (regularly observed) can generate very large changes in flux (when using gas exchange coefficients computed following Cole & Caraco).

7
did you consider ambiant water temperature to calculate Cw and Ceq or you used 22degC??
For a water at 8deg, it can generate 6% difference in flux.

8
this simple computation is considering a square lake morphology, but if they are rather like a bowl (deepest layer has a smaller volume), there is likely a bias toward the deepest gas concentrations (overestimation of total mass)

9
was this measured at the end of summer when storage is maximal? (see below comment)

10
so I understand that you do not consider the autumnal storage flux, even though your lakes seem stratified as described below (p. 13269).
Also, calculation of winter storage would be more accurate when comparing late autumn water column mass to late winter mass; I understand there are field logistic constraints, but the consequences of your assumptions needs to be acknowledged at some point. If summer mass is indeed overestimated, the difference (storage) would be underestimated (?)

of dissolved gas in spring before ice break up and the total mass of dissolved gas in summer.

2.5 CH_4 and CO_2 ebullition from sediments

We estimated CH_4 and CO_2 ebullition from sediments associated with discrete seeps following the lake-ice ebullition survey method of Walter Anthony et al. (2010). Seeps are defined as point-source locations of repeated bubbling and identified as A, B, C, and Hotspot classes according to distinct patterns of bubbles trapped in lake ice (Appendix A). To quantify seep ebullition, we removed snow from early winter lake ice to expose ebullition bubble clusters trapped in ice for seep classification, GPS mapping, flux measurements and gas collection using submerged bubble traps. On foot, we surveyed 9355 individual seeps within 161 plots (30–300 m^2 per plot) positioned randomly within both littoral and profundal zones of lakes. In some lakes, ice was opened above the seeps for placement of submerged bubble traps ~~over the seeps.~~ Semi-automated bubble traps remained in place over individual seeps year-round (Walter Anthony et al., 2010), providing daily and seasonal flux from sediment data ~~for individual seeps.~~ Seep class-specific flux rates and bubble CH_4 and CO_2 concentrations measured on a subset of seeps were applied to all mapped seeps to estimate whole-lake ebullition rates, indexed by Julian Day of the year (Appendix A). These fluxes represent bubbling rates from sediments as measured at the lake surface, not necessarily Direct Ebullition to the atmosphere. The following two section describe the fate of ebullition bubbles during the ice-cover and ice-free seasons.

2.6 Ice-bubble storage (IBS) flux

During the open-water (ice-free) summer season, ebullition bubbles reaching the lake surface release CH_4 directly to the atmosphere (Direct Ebullition). In winter, lake ice impedes Direct Ebullition emissions. Many ebullition bubbles reaching the top of the water column hit the underside of lake ice, come to rest, and exchange gases with the

water column until the downward-growing ice encapsulates the bubbles. Since ebullition bubbles typically have a high CH_4 concentration (40–90 %) in comparison to most lake water, which is generally far below saturation with respect to CH_4 (Wilhelm et al., 1977; Sepulveda-Jauregui et al., 2012), CH_4 readily diffuses out of bubbles into the lake water column.

We collected 37 samples of ebullition bubbles trapped as pockets in lake ice from five Alaskan lakes, expanding upon the lake ice-bubble data set of Walter et al. (2008). Additionally, we opened the lake ice and placed bubble traps beneath ice, above seeps, to sample "fresh" ebullition bubbles at the lake surface before they are impeded by ice (n = 2–41 seeps per lake; total of 560 samples). This allowed us to compare concentrations of CH_4 in ice-trapped bubbles (n = 2–8 seeps per lake) to gas concentrations in "fresh" bubbles prior to ice entrapment.

Numerical modeling informed by detailed field studies of CH_4 diffusion from ice-trapped bubbles in one of our study lakes, Goldstream L. (#18) revealed that 80 % of CH_4 in bubbles trapped by ice dissolves into the lake water column in winter (Greene et al., 2014). The remaining 20 % of CH_4 ebullition trapped by ice is released to the atmosphere, either from Hotspot seep sites that open periodically throughout the winter, or from A, B, and C seep sites as ice melts in spring (i.e. IBS emissions). With input of observed ice-growth rates on a subset of lakes in each of the three study regions and mean monthly atmospheric temperatures during 2003–2013 (US National Weather Service), we employed this model to calculate a first-order estimate of IBS in 34 of the 40 study lakes in which we had measurements of both seep ebullition and water-column dissolved CH_4 concentrations, which affect the CH_4 dissolution rate from bubbles. We linearly interpolated between measured surface CH_4 concentrations in the summer and spring to estimate water-column CH_4 concentrations during the ice-cover period.

Page : 5

1

2

something is missing in this sentence to understand these other steps.
Were ebullition fluxes only measured in early winter??

3

suggested text to delete

4

suggested text to delete

5

check structure

6

do you mean far below maximum solubility? i.e. far below the point where it starts to form bubbles?
Because surface waters (especially of thaw lakes) are often found above saturation relative to the atmosphere (=supersaturated), but still below max solubility, and still offer the potential to dissolve ebullition bubbles.

7

does this mean trapped bubbles are impoverished in CH4? could this mislead a proper distinction between dissolved CH4 trapped in ice (non-ebullition, freeze-out bubbles) and ebullition bubbles trapped in ice?

2.7 Direct ebullition in winter and summer

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

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

2.8 Seasonal and mean annual emissions

We estimated mean annual emissions from lakes as the sum of various modes of emissions seasonally: (1) Direct Ebullition from all seeps and Diffusion from the water column in summer (ice-free period); (2) winter (ice-cover period) Direct Ebullition emissions through ice-free Hotspot seeps during shoulder seasons and from all open seeps during the final month of the spring ice-melt season; and (3) spring emissions as the sum of first the release of IBS (ebullition seep gases trapped by lake ice) before lake ice disappears, and second, the release of lake water column Storage of dissolved gases, previously described by Michmerhuizen et al. (1996), Phelps et al. (1998), and Bastviken et al. (2004), when ice melts.

Because lakes were classified according to three geographic zones based on climate and permafrost, the average timing of ice cover was used to estimate the seasonal differences between CH_4 and CO_2 emissions for all lakes within each zone. Mean annual ice-on and ice-off dates from were compiled for years 2000–2012 for study lakes near Toolik Field Station in the northern region (1 October–18 June), our own observations of interior Alaska study lakes near Fairbanks from years 2008–2012 (8 October–9 May), and from Arp et al. (2013) and the National Park Service Inventory and Monitoring Program during years 2000–2013 for southern region lakes near Denali National Park (1 October–23 May) and southcentral Alaska, south of the Alaska Range (15 November–7 May).

2.9 Physical and chemical limnology

We measured the physicochemical limnology of lakes during winter and summer field campaigns at the same locations where dissolved gases were measured. Measurements of in situ parameters along vertical depth profiles in lakes included temperature, pH, oxidation reduction potential (ORP), and chlorophyll *a* (Chl *a*) obtained using a calibrated multiparametric probe Hydrolab DataSonde (Hach, Loveland, CO, USA). For a subset of lakes in each region, we used temperature data loggers (UA-001-08, Onset HOBO, Bourne, MA, USA) to record water temperature year-round in five-minute intervals at two depths (1 m water depth and lake bottom). Secchi disk depth (SecD) was measured with a 0.2 m Secchi disk. We collected water samples for ex situ analyses using a horizontal 2.2 L Van Dorn Bottle (WILDCO, Yulee, FL, USA). The con-

1

does it mean you do not consider background ebullition? (although presented as 25% of total emissions in Walter et al. 2007 pie chart). I can see you acknoledged this in the discussion.

2

see comment above

3

water properties? The term 'parameter' is rather used for modelling.

centrations of dissolved nitrate (NO_3^-), phosphate (PO_4^{3-}) and sulfate (SO_4^{2-}) in lake water were measured with a high-performance liquid chromatograph equipped with an electrochemical detector (ED40 Dionex, Dionex, USA). We determined total organic carbon (TOC) and total nitrogen (TN) with a total carbon and nitrogen analyzer (Shimadzu TOC-Vcsn equipped with TNM1 module, Shimadzu, Japan).

Trophic state indexes (TSI), calculated from SecD, Chl *a*, and TPO_4^{3-}, were used to estimate the trophic states of the lakes (Carlson, 1977). Based on field and laboratory observations we classified some lakes as "dystrophic" (Wetzel, 2001). In these lakes, water had a dark brown color resulting from high concentrations of dissolved organic carbon (DOC), including humic substances and organic acids presumably leached from litter and soils in their watersheds.

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

2.10 Statistical analysis

Since data were not normally distributed and did not meet the assumption of homoscedasticity, we tested relationships between CH_4 and CO_2 emissions vs. geography and limnological characteristics for the different lakes using the non-parametric two-tailed Mann–Whitney U test for comparison of two groups and Kruskal–Wallis One Way Analysis of Variance for comparison of several groups. We followed the Kruskal–

Wallis analysis with the Multiple-Comparison *Z* value test; differences were significant when the *Z* value was > 1.96.

We used single linear regression analysis to quantify relationships between CH_4 and CO_2 emissions and geographical and limnological characteristics. For these analyses, data normalization was obtained using logarithm base 10 (Log) transformation. Before and after data transformation, normality was assessed by the Shapiro–Wilk test. Regression models were accepted when the *p* value was < 0.01.

Relationships between lake-bottom water dissolved CH_4, lake-bottom water dissolved O_2, and ebullition were evaluated graphically and by Spearman Product-Moment Correlation Coefficients (ρ).

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

3 Results

3.1 Geographical and limnological patterns of CH_4 and CO_2 emissions

Total annual CH_4 and CO_2 emissions were highly variable, ranging two orders of magnitude among lakes (2.0 to > 300 g CH_4 m^{-2} yr^{-1} and 34.2 to > 1500 g CO_2 m^{-2} yr^{-1}; Table 2, Fig. 2). Among the geographical parameters presented in Table 1 and CH_4 and CO_2 emissions presented in Table 2, we found that the type of permafrost soil (yedoma vs. non-yedoma) was the geographical parameter most closely related to CH_4 and CO_2 emissions (Table 3). Total annual CH_4 emissions from yedoma lakes (43.8 ± 17.3 g m^{-2} yr^{-1}, mean ± SD, $n = 7$ lakes, excluding outlier lake #25) was significantly higher than from non-yedoma lakes (8.0 ± 4.1 g m^{-2} yr^{-1}, $n = 32$ lakes) (Table 2). Total annual CO_2 emissions appeared higher in yedoma (784 ± 757 g m^{-2} yr^{-1}, mean ± SD, $n = 8$ lakes, excluding outlier lake #25) than non-

Page : 7

1

specify here that this is used to approximate DOC??

2

It's rather total phosphorus that is used in trophic index, not solely soluble reactive phosphorus.

3

instead of using brakets you may define it as low production associated to high humic color.
And what do you mean by 'based on field observations'?
See below comment on dystrophy definition.
One thing I realised reading below is that you do not provide DOC but only TOC (including particles); do you think the particle contribution to TOC is negligible and TOC is a good surrogate to DOC? It is acknowledged but only too late in the text.
And how did you classify dystrophic lakes that had no TOC value measured?

4

?

5

this still remain obscure: what do these geographic characteristics include? It needs to be briefly clarified earlier in ms.

yedoma lakes ($137 \pm 129\,g\,m^{-2}\,yr^{-1}$, n = 32 lakes) (Table 2); however, due to high variability among lakes, the difference was not significant. Rosie Creek beaver pond (#25), an outlier lake with particularly high CH_4 and CO_2 emissions ($317\,g\,CH_4\,m^{-2}\,yr^{-1}$; $1138\,g\,CO_2\,m^{-2}\,yr^{-1}$; Fig. 2), was formed by beaver activity in an active stream system that drains into the Tanana River. The pond was subsequently influenced by thermokarst expansion into yedoma-type deposits, which further enhances carbon cycling in the fluvial system.

The relationship between CH_4 and CO_2 emissions and other geographic parameters followed the same pattern to the extent that they were related to characteristics of yedoma and non-yedoma permafrost soils (Table 3). For instance, yedoma is characterized by eolian deposits, which among the surface geologic deposit types was also most strongly related to CH_4 and CO_2 emissions. Among our study lakes, yedoma lakes occurred in the interior Alaska region (Fig. 1) and tended to have a dystrophic state, parameters that were both related to CH_4 and CO_2 emissions. Since the particular yedoma lakes in our study were relatively small lakes ($\leq 0.1\,km^2$), lake area was a morphologic parameter closely related to CH_4 and CO_2 emissions.

Regressions models showed that physical and chemical limnological parameters (Table 1) explained 19–63 % of deviation in the different flux pathways of CH_4 emissions (Table 4). Total CH_4 emission was correlated with Area, SecD, PO_4^{3-}, and TN (Table 4). We did not find any relationships between total CO_2 and the lakes' physicochemical characteristics, probably due to chemical equilibrium in water.

3.2 Modes of CH_4 and CO_2 emission

Total annual ebullition, consisting of Direct Ebullition in summer and winter as well as springtime release from IBS, was the dominant mode of CH_4 emission in lakes, comprising 86 % of total annual emissions from yedoma lakes and 65 % from non-yedoma lakes (Table 2). Summer Direct Ebullition was higher in yedoma-type lakes ($25.9 \pm 16.1\,g\,CH_4\,m^{-2}\,yr^{-1}$, $n = 6$ lakes, excluding lake # 25) than non-yedoma lakes

($4.0 \pm 3.7\,g\,CH_4\,m^{-2}\,yr^{-1}$, $n = 28$ lakes). This contrast drove other significant relationships in the data set: since yedoma lakes were primarily located in the interior discontinuous permafrost zone, and they dominated the dystrophic and northern boreal forest lakes category, we found that summer ebullition was higher in interior lakes than in northern and southern lakes; summer ebullition was higher in dystrophic lakes than in lakes of other trophic states; and northern boreal forest lakes had higher summer Direct Ebullition than lakes from other ecozonal categories (Tables 2 and 3). Direct Ebullition of CH_4 in winter and summer was correlated with lake Area; smaller lakes had higher Direct Ebullition (Table 4). Yedoma lakes were the only lakes in which we observed Hotspot ebullition and seep densities of all seep classes were higher in yedoma lakes (mean ± SD: 2.11 ± 2.51 A seeps m^{-2}, 0.27 ± 0.20 B seeps m^{-2}, 0.06 ± 0.06 C seeps m^{-2}, 0.01 ± 0.01 Hotspot seeps m^{-2}) compared 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 seeps m^{-2}, 0 Hotspot seeps m^{-2}). It follows that Direct Ebullition during the winter ice-cover period was also much higher from yedoma lakes ($5.9 \pm 3.6\,g\,CH_4\,m^{-2}\,yr^{-1}$, $n = 6$ lakes; excluding lake #25) than non-yedoma lakes ($0.6 \pm 0.6\,g\,CH_4\,m^{-2}\,yr^{-1}$, $n = 28$ lakes) (Table 2). In contrast, ebullition was not an important mode of CO_2 emission from any lakes. Total ebullition, including summer and winter Direct Ebullition, contributed 0.1 % of the total annual CO_2 emissions among all lakes (Table 2).

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

The IBS model, which accounts for bubble volume loss as gas exchanges between bubbles and the lake water column under ice (Greene et al., 2014), revealed that IBS was on average 13 % of total annual CH_4 emissions from yedoma lakes ($5.7 \pm 4.7\,g\,m^{-2}\,yr^{-1}$, $n = 6$) and 9 % for non-yedoma lakes ($0.7 \pm 0.7\,g\,m^{-2}\,yr^{-1}$, $n = 28$) (Table 2, Fig. 2). The CH_4 IBS flux from lakes was negatively correlated with Area and

Page : 8

1
was this occurring over the sampling period 2000-2012?

2
past tense?

3
could you get rid of the covariance instead? (in stat tests)

4
related in which way? dystrophic having higher emission rates compared to other trophic states?

Dystrophy needs to be better defined (low productivity/high nutrients) since it turns to be a 'controlling' factor.

And I think lakes should be classified as dystrophic OR UO, O, M or E, but not as both. Dystrophy is defined by low productivity (low chla) despite high nutrients, because of high DOC that is limiting light to primary producers. As it is, you seem to define dystrophy solely by the richness in DOC.

5
Is this relationship holding within each categories (Y and NY)? i.e. is it only related to the fact that Y lakes are smaller?

6
some of these numbers are already given in Table 2

7
it's not clear why you have to use the model to calculate this percentage, if IBS is calculated from what is measured in the ice bubbles (gas composition, bubble density)
"We collected 37 samples of ebullition bubbles trapped as pockets in lake ice from five Alaskan lakes,..."

8
is it a volume loss or a CH4 impoverishment in the bubble?

9
is it in the surface water under the ice or throughout the whole water column?

SecD (Table 4). Given the minor role of CO_2 Direct Ebullition in the annual emission budget (< 0.1 %), and the lesser role of springtime IBS, we considered IBS an insignificant mode of CO_2 emission.

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

CH_4 Diffusion emissions were statistically different between yedoma (5.0 ± 1.4 g CH_4 m^{-2} yr^{-1}, $n = 5$; excluding lake #25) and non-yedoma lakes (2.4 ± 1.3 g CH_4 m^{-2} yr^{-1}, $n = 26$). Rosie Creek beaver pond (#25) had the highest diffusive flux (160.3 g CH_4 m^{-2} yr^{-1}). Diffusion comprised 11 % and 30 % of total annual CH_4 emissions from yedoma and non-yedoma lakes respectively. We found significant a positive correlation between CH_4 diffusive flux and PO_4^{3-} (Table 4). In contrast, Diffusion was the dominant CO_2 mode of emission among all of our study lakes. Diffusion constituted 100 % and 92 % of CO_2 emissions from yedoma and non-yedoma lakes respectively. Diffusion from yedoma lakes (784 ± 757 g CO_2 m^{-2} yr^{-1}, $n = 4$ lakes) was significantly higher than Diffusion from non-yedoma lakes (127 ± 127 g CO_2 m^{-2} yr^{-1}, $n = 23$ lakes). It was not possible to normalize CO_2 Diffusion data, so we were unable to determine potential correlations between this mode of emission and limnological parameters.

3.3 Seasonal emissions

Figure 4 illustrates the contribution of different gas emissions pathways to annual emissions by season. Approximately three quarters of annual CH_4 emissions were released from lakes during the open water summer season: 71 % and 79 % of total annual CH_4 emissions in yedoma lakes and non-yedoma lakes respectively were the sum of summer Direct Ebullition and Diffusion. Spring and winter CH_4 emissions were also important. From yedoma lakes, first 13 % of total annual emissions occurred via IBS in spring when the ice started to degrade; subsequently, water column Storage release of dissolved gases was 3 % of total annual emissions. From non-yedoma lakes, total springtime emissions were 14 % of annual, consisting first of IBS (9 %) followed by Storage (5 %). Wintertime emissions via Direct Ebullition from ice-free holes above seeps were 13 % of total annual emissions from yedoma lakes and 7 % from non-yedoma lakes. It is of interest to note that accounting for IBS, a newly recognized mode of emission, increased the estimate of springtime CH_4 emissions based on the more commonly reported Storage emission by 320 %.

Seasonally, ~ 100 % and 92 % of total annual CO_2 emissions from yedoma and non-yedoma lakes respectively occurred in summer by Diffusion from the open water surface. The remaining 8 % of annual emissions in non-yedoma lakes occurred in spring from water column Storage flux (7 %) and winter Direct Ebullition (less than 1 %) (Table 2 and Fig. 2).

3.4 Physical and chemical patterns

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

1
lesser than minor?

2
make sure this is discussed; could it be that winter CH4 production in Y lakes is greatly supressed?

3
a significant (reverse words)

4
do you think dissolved CH4 is produced in the water column or CH4 is diffusing from sediment?

5
make sure you discuss why Y lakes do not store CO2; water column CO2 reduction by methanogens? is there O2 left in the water column during the winter?

6
does it mean they were morphologically 'in between'?

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

In winter, most lakes demonstrated inverse stratification. The coldest temperatures were at the surface of the water column beneath ice and water temperature tended to increase with depth. We found that winter bottom temperature was significantly different between northern lakes ($1.3 \pm 1.5\,^{\circ}C$) and southern lakes ($2.6 \pm 1.1\,^{\circ}C$), but none of these were significantly different from lake bottom temperature in Interior Alaska ($1.4 \pm 1.0\,^{\circ}C$), which is mainly due to the contrasting climatic conditions and the relatively shallow depths of northern lakes compared to southern lakes.

In most lakes, if there was a dissolved O_2 (DO) gradient, then DO was highest near the lake surface and decreased with depth in winter and summer. Three exceptions were El Fuego L. (#11), 91 L. (#27) and Dolly Varden L. (#36), where we observed an increase in DO with depth in summer, likely due to benthic photosynthesis. Among yedoma lakes, lake-bottom dissolved oxygen (DO) concentrations were $< 0.1\,mg\,L^{-1}$ in both winter and summer. In contrast, 81 % of the 32 non-yedoma lakes had well-aerated lake bottoms in summer; the lake-bottom water DO concentration in the other 19 % of lakes was $< 0.1\,mg\,L^{-1}$. In winter, we observed the reverse pattern among non-yedoma lakes: 76 % of 17 non-yedoma lakes measured had lake-bottom DO $< 0.1\,mg\,L^{-1}$ while 24 % of non-yedoma lakes, all which were southern lakes, had well-aerated lake bottoms in winter. All temperature and DO profiles measured on the study lakes are shown in Supplement Fig. B.

DO concentrations were inversely related to dissolved CH_4 concentrations in the lake bottom water during winter and summer (Fig. 5). This relationship suggests a strong

influence by biological processes, particularly in the dystrophic, yedoma lakes of interior Alaska (Table 5). Additionally, we found significant statistical relationships between lake area and dissolved gas concentrations (CH_4 and O_2) among our yedoma (small lakes) and non-yedoma study lakes (generally larger lakes) (Table 5).

Five additional limnological parameters also showed significant differences between yedoma and non-yedoma lakes (Table 1). The TOC, PO_4^{3-}, TN, Chl *a*, and SecD indicated higher nutrient availability and higher primary production in the dystrophic, yedoma lakes and/or their watersheds (Table 1). ORP values were significantly different between winter and summer in all lakes (Table 1), but were more than 2.5 and 1.5 times lower in yedoma lakes compared to non-yedoma lakes in winter and summer respectively, indicating more reducing conditions in yedoma-lake water columns. Together, these findings of higher primary production and lower ORP are consistent with the observations of high CH_4 and low O_2 concentrations in yedoma lakes compared to non-yedoma lakes (Fig. 5).

4 Discussion

4.1 Emission modes

The relative magnitude of different emission modes in this study followed the same general pattern observed previously (Casper et al., 2000; Bastviken et al., 2004; Abril et al., 2005; Repo et al., 2007), with ebullition dominating lake CH_4 emissions and diffusion dominating CO_2 emissions. Most studies of ebullition are conducted by distributing bubble traps in lakes without prior knowledge of discrete seep locations. Since seep locations are identified in winter as vertical stacks of bubbles in lake ice that represent repeated ebullition from discrete point-sources, surveys of lake-ice bubbles reveal the densities of seeps on lakes and the relative proportion of bubble-free black ice, which in nearly all northern lakes dominates on an area basis. Walter et al. (2006) identified non-point source bubbling from the seep-free fraction of the lake as "Background Ebul-

Page : 10

1
Wind protection by topography or vegetation?

2
thus, how come summer storage in hypolimnia is not considered?

3
was

4
obvious

5
showed? presented?

6
It is necessary to specify the profile shape for Dolly Varden, i.e. an increase from 10 to 12 mg/L from surface to 10m (deep chla maximum? do you know the chla at this depth?) and then it lowers again to approx 9 mg/L. The way you present this here makes us think there is an increase in DO toward the bottom waters, but this would seem strange to have a large contribution of benthic photosynthesis at depth for such a deep lake. We assume (ND) that DOC (TOC) is low for this lake as it was not classified as dystrophic... I wonder how you classified it as dystrophic without TOC however, with the eye? (brown color)

7
oxygenated? (and correct below if appropriate)

8
make sure we can read the lake name in final figure version

9
microbial?

10
I am not sure what I should look at in this table; can you be more explicit?

11
relationship between CH4 and area was already presented above, no?

12
I am lost; higher nutrients and higher PP (approximated with chla) in dystrophic Yedoma vs NY, while dystrophy should be defined as high nutrients but low PP because of light limitation caused by high DOC… Are you considering other primary producers than plankton (chla) here? If you consider macrophytes (floating Sphagnum?) in your characterization of primary production, it needs to be clarified.

13
I look forward to read the discussion to get clarifications

14
I think it needs to be rewritten
(and I don't think this is exclusive of northern lake)

lition". Background Ebullition is thought to originate primarily from methanogenesis in surface lake sediments in summer; in contrast, ebullition seeps consist of bubble tubes that allow CH_4 produced at depth in sediments to migrate efficiently as bubbles to the sediment surface in summer and winter by the repeated release from point-source locations. Bubble traps placed in seep and non-seep locations and monitored year-round in two Siberian lakes showed that seep ebullition dominated total annual CH_4 emissions. Background Ebullition was high in summer, nearly absent in winter, and altogether comprised ~25 % of total annual CH_4 emissions in the Siberian lakes. Preliminary results from bubble-traps placed in some of our Alaskan study lakes in locations where no seep ebullition bubbles were observed in winter also showed high summertime bubbling (K. M. W. A. unpublished data, 2014). This suggests that Background Ebullition occurs in Alaska too. [1] Since our estimate of lake ebullition in the Alaskan lakes is based solely on discrete seeps and does not include non-seep Background Ebullition, we consider that our estimate of total lake ebullition is below the total actual ebullition flux. Given that methanogenesis is highly temperature dependent (Dunfield et al., 1993; Schulz et al., 1997; Nguyen et al., 2010; Marotta et al., 2014; Yvon-Durocher et al., 2014) and surface lake sediments heat up in summer, accounting for Background Ebullition would likely increase the total ebullition emissions from all of the Alaskan study lakes.

The Ice-Bubble Storage (IBS) mode of emission described here is a newly recognized CH_4 flux component in lakes (Greene et al., 2014), [2] that has not previously been included in regional studies. Given the coarse temporal resolution of temperature and dissolved gas data used as input to the IBS model, we acknowledge that our estimate of IBS is a first-order approximation. However, strong agreement in the relative importance of IBS in the annual CH_4 budget of Goldstream Lake (#18) in this study using coarse resolution data (IBS 6 % of total annual CH_4 emission) vs. the estimate from Greene et al. (2014) using highly detailed field data allowing detailed modeling (IBS was 5 % and 9 % of total annual emissions in two different years), suggests that our first-order approximations of IBS may be valid. Since IBS was an important mode of CH_4 emissions among our study lakes (13 % and 9 % of total annual emissions

in yedoma and non-yedoma lakes, respectively), it is likely that past estimates of the magnitude and seasonality of CH_4 emissions from lakes with ebullition seeps were incomplete. Greene et al. (2014) found that a large fraction (~80 %) of CH_4 diffused from ebullition bubbles trapped under lake ice into the lake water in Goldstream L. [3] Coarser-resolution modeling of the IBS process for our study lakes also suggested that approximately 80 % of CH_4 dissolved out of ice-trapped bubbles. Detailed measurements and modeling in Goldstream L. showed that about half of this re-dissolved CH_4 was ultimately oxidized (Greene et al., 2014). Due to a paucity of field data, we did not model CH_4 oxidation; however, given the observed CH_4 oxidation potentials in our study lakes through incubation studies (Martinez-Cruz et al., 2014), it is likely that some fraction of the re-dissolved ebullition bubbles is oxidized. The un-oxidized fraction of dissolved CH_4 is subject to release to the atmosphere via water column convection and diffusion as Storage emissions in spring when ice more completely disintegrates and as Diffusion during summer (Greene et al., 2014). [4] Thus the Storage and Diffusion modes of emission may involve not only dissolved CH_4 that diffused out of lake sediments, but also dissolved CH_4 that first originated as ebullition bubbles prior to ice entrapment. Since ebullition seeps were important components of whole-lake CH_4 emissions in all of our study lakes, as well as in tens of other lakes previously reported in Alaska (Walter Anthony et al., 2012) and Siberia (Walter et al., 2006; Walter Anthony et al., 2010), IBS should be studied and accounted for in global lake CH_4 emission budgets.

Lake CH_4 Storage emission estimates for our Alaska study lakes ($0.5 \pm 0.7\,g\,CH_4\,m^{-2}\,yr^{-1}$; Table 2), which comprised ~4 % of total annual emissions, were highly variable and on the same order of magnitude as the mean estimate for other northern lakes reported by Bastviken et al. (2004) ($2.4\,g\,CH_4\,m^{-2}\,yr^{-1}$) and Bastviken et al. (2011) ($0.8\,g\,CH_4\,m^{-2}\,yr^{-1}$; pan-Arctic). Storage emission from global lakes ranged from < 0.1 to $37\,g\,CH_4\,m^{-2}\,yr^{-1}$, comprising 0.5 % to 81 % of the total annual emissions (Bastviken et al., 2011). This also suggests high variability in this emission mode among global lakes. The large relative error for Storage flux measured

1
I think this should be clarified higher in the ms (in method section)

2
maybe cite Langer et al. 2014 in BGS Discus.?

3
where can we appreciate how you did this?
you got the exact same 80% than Greene et al.?
Is the range of variation tight?
A little more details is needed so readers do not have to read Greene et al. to understand well what it means/involve.

4
how could we resolve that fact? with 14C?

among our Alaska study lakes (140 %; mean ± SD, 0.5 ± 0.7 g CH_4 m^{-2} yr^{-1}) confirms that there is large variability associated with this mode of emission; however, CH_4 Storage emissions in our Alaska study lakes were < 2.7 g CH_4 m^{-2} yr^{-1}, except in Rosie Creek beaver pond (#25, 39 g CH_4 m^{-2} yr^{-1}). In other northern lakes, full or partial mixing of the water column leads to the release of stored gas during a 10–15 day period associated with ice-out (Phelps et al., 1998; Striegl and Michmerhuizen, 1998; Bellido et al., 2009). Additionally, full or partial turnover of the lake water column in fall can release additional stored CH_4 (Bastviken et al., 2004; Bellido et al., 2009). Since we estimated only spring Storage emission and did not take into account potential additional emissions associated with fall turnover, our Storage values are likely an underestimate.

4.2 Geographic patterns of lake CH_4 and CO_2 emissions in Alaska

Previous regional analyses of northern lake emissions found a relationship between CH_4 emissions from lakes and latitude that was explained by temperature (Sarotta et al., 2014; Yvon-Durocher et al., 2014). Primary production in warmer climates supplies more organic substrate for methanogenesis, and methanogenesis is sensitive to temperature. However, the lakes in these studies were not permafrost-affected. In our N-S Alaska transect we did not find a relationship between lake CH_4 emissions and latitude or temperature. We attribute this finding to the presence and geographic diversity of permafrost types (yedoma vs. non-yedoma) (Jorgenson et al., 2008; Kanevskiy et al., 2011), which is more a function of periglacial history and topography in Alaska than it is of latitude or recent climate. While methanogenesis in surface sediments of lakes globally is fueled by contemporary autochthonous primary production and allochthonous organic matter supply (processes typically controlled by latitude and climate in undisturbed systems), thermokarst-influenced lakes have an additional, deeper source of organic matter that fuels methanogenesis: thawing permafrost in the thaw bulbs beneath lakes and along thermally eroding shorelines. Organic matter supplied by thawing permafrost, particularly in lakes formed in thick, organic-rich yedoma-type deposits, can overwhelm the more contemporary organic carbon substrates supplied to surface lake sediments (Kessler et al., 2012).

The interior Alaska yedoma lakes, which had the highest CH_4 and CO_2 emissions, are largely thermokarst lakes formed by thaw of organic-rich yedoma permafrost. Radiocarbon ages (18–33 kyr BP) and δD-depleted values of CH_4 in ebullition bubbles collected from the interior Alaskan thermokarst lakes suggested that thaw of late Pleistocene yedoma organic matter fuels methanogenesis in these lakes (Walter et al., 2008; Brosius et al., 2012). The 6-fold difference in CH_4 emissions between yedoma lakes and non-yedoma lakes throughout the rest of Alaska is likely explained by the variability in the availability of recently thawed permafrost organic matter, which provides a larger additional substrate for methanogenesis in the yedoma lakes owing to the thickness (usually tens of meters) of organic-rich yedoma deposits (Kanevskiy et al., 2011; Walter Anthony et al., 2012).

Previous research using stable isotopes and radiocarbon dating of CH_4 in ebullition bubbles in yedoma lakes demonstrated that stronger ebullition seeps originate from greater depths beneath the sediment-interface and are characterized by older ^{14}C ages and more depleted δD values associated with thaw of Pleistocene-aged yedoma permafrost (Walter et al., 2008). The disproportionately large contribution of strong Hotspot ebullition seeps to emissions from yedoma lakes (mean ± SD: 17 ± 12 % of total annual emissions) in this study suggests microbial production of CH_4 at greater depths in sediments beneath yedoma lakes. In contrast, the absence of Hotspot ebullition seeps in non-yedoma lakes, which we observed to also have dense sediments, suggests that CH_4 formation by microbial decomposition of organic matter is more restricted to shallower sediment depths in the non-yedoma lakes.

The relationship between ebullition, dissolved CH_4 concentration and lake type (Fig. 6) also indicates that ebullition seeps releasing CH_4 produced deep in the thaw bulb enhance CH_4 cycling in yedoma lakes more than in non-yedoma lakes. Yedoma lakes, which had a higher density of ebullition seeps than non-yedoma lakes (Sect. 3.2),

1

In addition to the fact that you did not consider summer storage in deep waters released in autumn (as explained below), could this range of values for winter storage be underestimated if the starting point to calculate storage is summer (when concentrations are higher) instead of late autumn prior to ice formation (true strating point for storage; when concentrations could be lower after venting part of the summer production)? Does this make any sense?

2

what is the point of this sentence?

3

this paper is on tropics so not supporting your sentence, or you may want to modify your sentence

4

you mean 'directly' (physiologically) sensitive (as oposed to the indirect effect of the first part of the sentence)?

5

Are your temperature measurements appropriate to explore such dependency? What did you use in your statistical analyses: bottom or surface or an average water column T? Did you only use your sub data set where 2 thermistors were placed year-round or you used all lake data set with only 2 profiles over a complete year? This could also be considered to acknowledge the absence of relationship.

6

It is not clear what you mean by "can overwhelm"; can you be more explicit?

And for the stat analyses, would the T in sediments (where methanogens are located) be more appropriate than water column?

7

maybe one ending sentence is missing to make a link to previous discussion?

8

Enhance CH4 cycling: what does it mean?

had both higher volumes of CH_4-rich bubbles impeded by lake ice and higher concentrations of dissolved CH_4 in the lake water in winter (Fig. 6a, $p = 0.72$). Based on Greene et al. (2014), in which 27 % of dissolved CH_4 in the water column in winter originated from CH_4 dissolution from ebullition bubbles trapped by lake ice, we attribute the higher concentrations of dissolved CH_4 in the yedoma study lakes to the process of CH_4 dissolution from ice-trapped bubbles. Modeling results, which showed that approximately 80 % of CH_4 in bubbles trapped by lake ice in our study lakes dissolved into the water column, support this conclusion. Other important processes that would also control dissolved CH_4 concentrations in lake water are diffusion from sediments and CH_4 oxidation. Given the thicker CH_4-producing sediment package beneath yedoma lakes, we would expect diffusion of dissolved CH_4 from yedoma lakes to be higher than that of non-yedoma lakes. Ex situ incubations by Martinez-Cruz et al. (2014) on a subset of our Alaska study lakes also showed that yedoma lakes had higher CH_4 oxidation potentials, owing in large part to higher concentrations of the dissolved CH_4 substrate in these lakes. Compared to winter, the weaker correlation between dissolved CH_4 and Direct Ebullition in summer (Fig. 6b, $p = 0.42$) has several potential explanations. First, in summer, ebullition bubbles escape directly to the atmosphere, so the dissolved CH_4 stock of the water column is not supplied from ice-trapped bubble dissolution like it is in winter unless residual winter-dissolved bubble CH_4 remains in the water column in summer. Second, dissolved CH_4 diffusing from lake sediments in summer may be more immediately oxidized by aerobic CH_4 consumption since O_2 is more available in lake water from atmospheric diffusion and autochthonous primary production. Finally, higher PO_4^{-3}, TN and Chl *a* concentrations in yedoma lakes (Table 1) suggests primary production in yedoma lakes may contribute relatively more substrate to methanogenesis in surface sediments. CH_4 produced in surface sediments more readily escapes to the water column via diffusion than CH_4 produced in thaw bulbs, which preferentially escapes by ebullition (Tan et al., 2014). Higher diffusion from surface sediments would support higher concentrations of dissolved CH_4 in lake water, a process that can be independent of ebullition from thaw bulbs in summer. This explanation is supported by

two times higher summer Diffusion emissions from yedoma lakes compared to non-yedoma lakes (Table 2), despite higher observed CH_4 oxidation potentials in yedoma lakes vs. non-yedoma lakes (Martinez-Cruz et al., 2014).

CO_2 Diffusion, which was ~100 % and 92 % of total annual CO_2 emissions from yedoma and non-yedoma lakes respectively, was 6 times higher on average in yedoma lakes than in non-yedoma lakes. Potential explanations include enhanced CO_2 production associated with yedoma organic matter decomposition, photooxidation of the large DOC pool observed in the dystrophic yedoma lakes, and potentially higher rates of CH_4 oxidation in yedoma lakes (Martinez-Cruz et al., 2014) generating more CO_2 in the lake water columns. Possible differences in watershed sizes draining into lakes could also influence CO_2 concentrations in lakes and Diffusion emissions since terrestrial dissolved inorganic carbon often dominates lake CO_2 pools (Kling et al., 1992; Battin et al., 2009; Tranvik et al., 2009). Since we did not study these processes, and since our calculations contain uncertainty associated with the assumption that single-day measurements of dissolved CO_2 and CH_4 in lakes represent the mean flux for the entire open water period, further research is needed to validate these hypotheses in the Alaskan lakes.

3 Dissolved CH_4 and O_2 dynamics

Dissolved O_2 concentration is a useful parameter for predicting the CH_4 concentrations in Alaskan lakes. The inverse relationship between CH_4 and O_2 concentration in lake water (Fig. 5) suggests that low dissolved O_2 levels in lakes are due to high microbial activity, which in turn leads to optimal anoxic conditions for methanogenesis (Boon and Mitchell, 1995).

There are several possible explanations for the pattern of seasonally higher dissolved CH_4 and lower O_2 concentrations in winter among lakes (Fig. 5): (1) Ice cover inhibits O_2 transfer from the atmosphere into the water column (White et al., 2008); (2) Primary production in lakes declines as day length shortens (White et al., 2008; Clilverd et al., 2009); (3) Snow cover impedes light transfer, further extinguishing photosynthe-

1
there is a little redundancy (present and next sentence) with above p. 13272

2
wasn't it 80% above?

3
this could be especially true if spring mixing is short: did you observe this from your 2 thermistor chain data set?

4
any loss processes that could account for this difference?

5
this is also to consider for CH4 flux estimations, but its placed in CO2 paragraph

6
Be careful with redundancy, especially in this section.

7
Fig. 5: I think it would be clearer to use r instead of p as the symbol for spearman coefficient (figure legend)
The CH4 bars are black, not grey
We can barely see the words in this graph.

8
but methanogenesis occurs in sediments, and having O2 in bottom water does not preclude methanogenesis in sediment right?

sis beneath the ice (Clilverd et al., 2009; Welch et al., 1987); and (4) Finally, aerobic microorganisms consume residual O_2 in the water beneath the ice (Bellido et al., 2009, Clilverd et al., 2009). The resulting anoxic conditions facilitate anaerobic processes like methanogenesis and decrease methanotrophy (Dunfield et al., 1993). All the while, CH_4 is emitted from lake sediments throughout winter via diffusion and seep ebullition. Many ebullition bubbles are impeded by lake ice, leading to dissolution of CH_4 from bubbles and an increase in dissolved CH_4 concentration. In summer, the lack of ice cover allows CH_4 in bubbles to be released directly to the atmosphere without partially dissolving in the lake water column. This explains in part the lower CH_4 concentrations in lake water in summer (Greene et al., 2014). Furthermore, the O_2 concentration in lake water increases in summer by gas exchange with the atmosphere and by primary production in lakes (Fig. 5b). As a result, a fraction of dissolved CH_4 in lake water is emitted to the atmosphere, while methanotrophic activity, supported by elevated O_2 concentration, oxidizes another fraction (Martinez-Cruz et al., 2014).

In addition to the seasonal variations described above, a permafrost type effect on dissolved CH_4 and O_2 patterns was also observed. While during summer, most of the non-yedoma lakes were well oxygenated, yedoma lakes in interior Alaska had contrastingly lower O_2 concentrations and higher dissolved CH_4 concentrations beneath the thermocline. This suggests high methanogenic activity in sediments that fuels CH_4 oxidation, reducing the O_2 concentration under the thermocline, where stratification limits O_2 ingress from superficial water layers.

4.4 Limnological and morphological patterns

Single linear regression analysis showed that the most useful limnological variables to predict CH_4 emissions in the Alaskan lakes were Area, MecD, PO_4^{3-}, and TN, all which are indicators of lake metabolism and morphology (Table 4). These findings are consistent with the patterns that explain CH_4 emissions in Swedish and Michigan lakes (Bastviken et al., 2004) and Finnish lakes (Juutinen et al., 2009). The association between high CH_4 emissions and high nutrients and Chl *a* concentrations among yedoma

lakes compared to non-yedoma lakes is consistent with the geographic patterns previously observed in Siberian lakes. Higher aquatic production observed in Siberian yedoma lakes compared to non-yedoma lakes in the same climate zone was attributed to fertilization of the yedoma lakes by nitrogen- and phosphorus-rich thawing yedoma permafrost (Walter Anthony et al., 2014).

The negative correlation between CH_4 emissions and lake area indicates that small lakes had higher total annual CH_4 emissions. This finding is driven by yedoma lakes, which were on average much smaller than non-yedoma lakes (Table 1). It is also consistent with lake CH_4 emission patterns in other regions whereby smaller lakes have higher CH_4 emissions due to a stronger relative contribution of littoral organic matter to whole-lake methanogenesis (Bastviken et al., 2004).

Significant differences between other limnological and geographical parameters also distinguished yedoma from non-yedoma lakes. Dystrophy (i.e. brown-water lakes due to high DOC concentrations; Wetzel, 2001), characterized yedoma lakes. Due to the absorption of incoming solar radiation by DOC in the surface water (indicated by TOC, which corresponds approximately to DOC in natural lake ecosystems (Tranvik et al., 2009, Table 1), the yedoma lakes were thermally stratified in summer, and had relatively cold, anoxic hypolimnia nearly all year round. Since non-yedoma lakes have a variety of different origins, lake depth was highly variable among them; however, lower concentrations of DOC in non-yedoma lakes and, in many cases, large surface area to volume ratios that facilitated water column mixing by wind contributed to less intense stratification or the absence of stratification among many of the non-yedoma lakes.

4.5 Climate warming impacts of Alaskan lake emissions

Previously, Kling et al. (1992) showed that tundra lakes near Toolik Field station emit CH_4 and CO_2 via Diffusion. More recently, Walter Anthony et al. (2012) recognized the importance of CH_4 ebullition from biological seeps in Alaskan lakes (0.75 Tg CH_4 yr^{-1}); however, this represented the quantity of ebullition seep CH_4 released from sedi-

1
this is the obvious reversal of above sentence for winter

2
not only methanotrophy is consuming O2

3
Indeed, a multivariate stat analysis, eliminating covariance, would be more appropriate

4
at most 0.23; is this really useful as a single predictor?

5
at most 0.32

6
which means the system would be nutrient-limited, not C limited? that deserves a discussion I think

7
Have you tested the 2 classes separately? That deserves discussion.

8
but would this hold true considering the same argument as above, that Y lakes have a thaw bulb and that most emissions come from talik thus lake size does not really matter? The relationship should hold for Y category to make this argument stronger: do size really matter or it's only a question of Y vs NY?

9
Is this paragraph bringing something to the overall goal of the paper? It would if a link with GHG emissions is made

10
see above comment on dystrophy definition (Y lakes have higher chla)

11
this would rather appear earlier in paper (cf my above comment)

12
I think the feeling of redundancy as we read the discussion comes from the fact that you gave too much info in the result section

13
maybe needs a definition?

ments rather than the magnitude of atmospheric emissions. Since ebullition emission is partially impeded by lake ice in winter, and a fraction of CH_4 dissolved out of bubbles beneath ice is oxidized by microbes (Greene et al., 2014), ebullition emissions to the atmosphere are lower than what is released annually from sediments. This study is the first to consider multiple modes of emissions for CO_2 and CH_4 together, including the ice-bubble storage process, for a large number of Alaskan lakes spanning large geographic gradients. Scaling total annual CH_4 and CO_2 emissions observed among yedoma and non-yedoma lakes to the extent of these lake types in Alaska (Walter Anthony et al., 2012) ($44 \pm 17\,g\,CH_4\,m^{-2}\,yr^{-1} \times \sim 8800\,km^2$ yedoma lakes; $8 \pm 4\,g\,CH_4\,m^{-2}\,yr^{-1}$ x $\sim 41\,700\,km^2$, non-yedoma lakes), we estimate that yedoma and non-yedoma lakes emit a total of $0.72\,Tg\,CH_4\,yr^{-1}$ ($\sim 0.39\,Tg\,CH_4\,yr^{-1}$ from yedoma lakes, $0.33\,Tg\,CH_4\,yr^{-1}$ from non-yedoma lakes). This estimate of Alaska lake emissions increases the previous estimate of Alaska's wetland ecosystem emissions ($3\,Tg\,CH_4\,yr^{-1}$, Zhuang et al., 2007), in which lakes were not included, by 24 %. Our estimate of lake CH_4 emission is conservative because it does not include Background (non-seep) Ebullition or Storage emissions associated with fall lake turnover events.

If we assume that our study lakes represent the CH_4 and CO_2 emission dynamics of all lakes in Alaska and account for the 34-fold stronger global warming potential of CH_4 vs. CO_2 over 100 years (GWP_{100}; Myhre et al., 2013), the impact to the climate based on CO_2 equivalent (CO_2-eq) emissions from yedoma lakes is $\sim 20\,Tg\,CO_2$-eq yr^{-1} ($13\,Tg\,CO_2$-eq yr^{-1} from CH_4 and $7\,Tg\,CO_2\,yr^{-1}$ from CO_2). For non-yedoma lakes, the total climate impact is $\sim 17\,Tg\,CO_2$-eq yr^{-1} ($11\,Tg$ CO_2-eq yr^{-1} from CH_4 and $6\,Tg\,CO_2\,yr^{-1}$ from CO_2). These results have several important implications. First, CH_4 emissions have nearly twice the impact on climate as CO_2 emissions among all Alaskan lakes. Second, the climate impact of yedoma and non-yedoma lakes in Alaska due to carbon greenhouse gas emissions are approximately equal, despite yedoma lakes comprising less than 1/5 of the total lake area in Alaska. The disproportionately large climate impact of CH_4 emissions from yedoma lakes is due in large part to thaw of

deep, organic-rich yedoma permafrost beneath these lakes; however, higher concentrations of nitrogen, phosphorus and chlorophyll *a* in these lakes suggests enhanced primary production in the lakes, which can also fuel decomposition and methanogenesis, as recently demonstrated in Siberia (Walter Anthony et al., 2014).

5 Conclusions

We have shown that Alaskan lakes are net sources of atmospheric CH_4 and CO_2. Total annual CH_4 and CO_2 emissions were dominated by ebullition and diffusion, respectively; however, the climate warming impact of CH_4 emissions was twice that of CO_2. Our 40 study lakes spanned large gradients of physicochemical limnology and geography in Alaska. We attribute the 6-fold higher CH_4 and CO_2 emissions observed in thermokarst lakes formed in icy, organic-rich yedoma permafrost in interior Alaska compared to non-yedoma lakes throughout the rest of Alaska to enhanced organic matter supplied from thawing yedoma permafrost, which is typically thicker than the organic-rich strata of non-yedoma soils. Higher total nitrogen, PO_4^{3-}, and Chl *a* concentrations in yedoma lakes suggest that higher primary production may also enhance organic substrate supply to decomposition and greenhouse gas production in these lakes. Consideration of multiple modes and seasonality of CH_4 and CO_2 emissions revealed that summer emissions were largest. However, winter and spring emissions of CH_4, including Direct Ebullition through holes in lake ice and the ice-bubble storage and release process, were also significant components of the annual CH_4 budget. Our results imply that regional assessments of lake CH_4 and CO_2 emissions in other parts of the pan-Arctic should take into account the myriad of emission modes, lake type and geographic characteristics, such as permafrost type.

1

So it seems other emission processes than seeps cancel out with CH4 consumption (oxidation) to yield the same total previously estimated in Walter Anthony et al. 2012?

2

Is water column primary production truly an OC contribution or a priming effect? (if the system is not C-limited)
Can you estimate the C-stock provided by planktonic growth and compare it to thaw bulb C-stock (on a m-2 basis)?

Appendix A: Methods

A1 Dissolved gas measurements

We used the Headspace Equilibration-Tunable Diode Laser Spectroscopy (HE-TDLAS) technique, described in detail by Sepulveda-Jauregui et al. (2012), to measure the concentration of CH_4 dissolved in lake water. Briefly, we collected water samples using a Van Dorn Bottle (WILDCO, Yulee, FL, USA) and gently transferred 60 mL into three borosilicate vials (100 mL volume) using disposable polypropylene syringes for triplicate measurements. Vials were immediately sealed with butyl rubber stoppers and aluminum crimp caps. The vials containing the water samples were shaken vigorously for 10 s to transfer CH_4 from the water into the vials' headspace for subsequent measurement with the GasFinder 2.0.

In addition to HE-TDLAS, we also measured dissolved CH_4 and CO_2 in a subset of samples using the traditional headspace equilibration method by gas chromatography (Kling et al., 1992). Water samples (10 mL) collected with the Van Dorn Bottle were transferred into 25 mL glass serum bottles and immediately sealed with butyl rubber stoppers and aluminum crimp caps. Serum bottles were stored upside down and frozen until laboratory analysis. In the laboratory, we thawed the samples to room temperature, shook bottles for 10 s to equilibrate headspace and water samples, and then measured CH_4 and CO_2 of the headspace by gas chromatography (Shimadzu GC-2014).

A2 Seep ebullition

GPS-mapped ebullition seeps were classified as A, B, C and Hotspot types, based on ice-bubble morphologies. This classification system has been described in detail, with example photographs and bubble morphology classification criteria presented in multiple previous publications (Walter et al., 2006, 2008; Walter Anthony et al., 2010, 2013). Briefly, A-type ebullition seeps are relatively small clusters of ebullition bubbles in which individual bubbles stack on top of each other in the winter ice sheet without

merging laterally. Due to progressively higher ebullition rates, individual bubbles of B-type seeps laterally merge into larger bubbles under the ice prior to freezing in ice. Types A and B seeps produce low gas-volume clusters of bubbles in lake ice with cluster diameters typically < 40 cm. The larger C seeps result in large (usually > 40 cm diameter) pockets of gas in ice separated vertically by ice layers containing few or no bubbles. Bubble-trap measurements showed that the solid ice layers in between the large gas pockets of C-type seeps represent periods of relative quiescence in between large ebullition events (Walter et al., 2006; Walter Anthony et al., 2010). Hotspot seeps have the greatest mean daily bubbling rates. The frequency of ebullition release from Hotspot seeps and the associated convection in the water column created by rising bubble plumes can be strong enough to maintain ice-free holes in winter lake ice or ice-free cavities covered by thin layers of ice during cold periods.

Thirty-day averages of bubbling rates (mL gas $seep^{-1}$ d^{-1}) were determined through bubble-trap measurements of seep fluxes and associated with seep classes for each Julian day of the year (Walter Anthony et al., 2010). This data set consists of ~ 210 000 individual flux measurements made using submerged bubble traps placed over ebullition seeps year-round. These class-specific fluxes were applied to the whole-lake mean densities of seeps on lakes to derive estimates of bubble-release rates from lake bottom sediments indexed by Julian Day. To determine mass-based estimates of CH_4 and CO_2 in ebullition bubbles, we applied lake specific measurements of CH_4 and CO_2 bubble concentrations to the individual lakes where seep-bubble gases were collected and measured. Methods of bubble-trap gas collection and measurements were described in detail by Walter et al. (2008). We sampled with bubble traps and measured by gas chromatography the CH_4 and CO_2 compositions of seep ebullition bubbles collected from up to 246 individual ebullition events per lake. In lakes where few or no seep-bubble gas concentrations were determined, we applied mean values of CH_4 and CO_2 by seep class (Walter Anthony et al., 2010): A, 73 % CH_4, 0.51 % CO_2; B, 75 % CH_4, 0.40 % CO_2; C, 76 % CH_4, 0.55 % CO_2; Hotspot, 78 % CH_4, 0.84 % CO_2. Whole-lake mean ebullition was the sum of seep fluxes observed along an average of five 50 m

long transects per lake (median 4 transects per lake), divided by the total area surveyed. In a recent comparison of methods for quantifying ebullition, Walter Anthony and Anthony (2013) showed that when at least three 50 m transects per lake are used to quantify seep ebullition, the estimate of mean whole-lake ebullition is 4–5 times more accurate than the mean flux determined by placement of seventeen 0.2 m^2 bubble traps randomly distributed across lake surfaces.

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Table 1. Lake physical and chemical properties from 40 Alaskan lakes. *N* – Lake number; * indicates informal lake names, the A-number refers to lake identification numbers used by Gregory Eaves et al. (2000) for study of the same lakes; Y/NY – permafrost soil type as Yedoma or non-Yedoma[a,b]; TSI – Trophic State Index[c]; EC – Ecozonal Categories[d]; Lat – Latitude; Long – Longitude; DN[e] – Sedimentary Deposit Name[e]; MD – Maximum known depth; A – Area; SecD – Secchi Depth; *T* (Win) Winter temperature; *T* (Sum) Summer temperature; pH (Win) – Winter; pH (Sum) – Summer; ORP (Win) – Winter redox potential average; ORP (Sum) – Summer redox potential; Chl *a*-Summer surface chlorophyll *a*; PO_4^{3-} – Dissolved phosphate; NO_3^- – Nitrate; SO_4^{2-} – Sulfate; TOC – Total Organic Carbon; TN – Total Nitrogen; TOCS-Total organic carbon in surface sediments; TNS-Total nitrogen in surface sediments. Water column-averages are reported for temperature, pH and ORP. Water-column carbon and nutrient values are from 1 m depth, except data summarized from other investigators[g,h]. Error terms are the standard deviation. ND indicates not determined; CF indicates lake completely frozen; "<" indicates below detection limit, "–" indicates no standard deviation due a sample size of one.

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	D, M	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	Julieta* 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	D, M	FoT	65.834	149.631	ES	3.3	0.027	0.70	0.5 ± 0.7	17.0 ± 4.0
17	Vault*	Y	D, O	NBF	65.029	147.699	MAC	3.7	0.003	1.00	0.3 ± 0.3	9.5 ± 7.7
18	Goldstream*	Y	D, M	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	0.035	1.59	0.7 ± 0.8	22.2 ± 2.2

Table 1. Continued.

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)
20	Killarney*	Y	D, M	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	D, E	NBF	64.865	147.868	E	4.4	0.094	0.50	0.5 ± 0.7	19.0 ± 1.7
22	Stevens Pond*	Y	D, M	NBF	64.863	147.871	E	1.1	0.002	0.50	CF	17.6 ± 1.6
23	Duece A2	Y	D, UO	NBF	64.863	147.942	E	6.0	0.023	0.79	0.9 ± 0.6	11.4 ± 7.0
24	Ace A1	Y	D, M	NBF	64.862	147.937	E	9.0	0.077	1.26	2.9 ± 0.9	11.6 ± 6.3
25	Rosie Creek*	Y	D, O	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	D, UO	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	AlT	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]	–	–	–	–	–	–	4.1[k]	0.022[k]	0.82[k]	1.1[k] ± 1.0	11.3[k] ± 4.5
	Non-Yedoma[j]	–	–	–	–	–	–	7.6[k]	0.267[l]	2.39[l]	1.6[k] ± 1.3	14.9[l] ± 3.0

Table 1. Continued.

N	Name	pH (Win)	pH (Sum)	ORP (Win) (mV)	ORP (Sum) (mV)	Chl *a* ($\mu g\,L^{-1}$)	PO_4^{3-} ($\mu g\,L^{-1}$)	NO_3^- ($mg\,L^{-1}$)
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[h]	ND
5	E6	ND	7.7 ± 0.7	ND	272 ± 80	5.9 ± 6.2	1.1[h]	ND
6	E5 Oil Spill A30	ND	7.1 ± 0.8	ND	322 ± 64	13.5 ± 2.9	1.8[h]	ND
7	Toolik A28	6.9 ± 0.1	7.9 ± 0.8	303 ± 32	308 ± 75	1.5 ± 0.4	1.6[h]	< 0.01
8	E1	7.0 ± 0.1	9.1 ± 0.4	283 ± 58	231 ± 71	1.3[g] ± –	1.1[h]	< 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 ± 0.1	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
35	Rainbow A48	7.0 ± 0.0	7.7 ± 0.6	241 ± 62	289 ± 85	12.6 ± 0.4	4.8[g]	< 0.01
36	Dolly Varden A47	ND	7.1 ± 0.3	ND	282 ± 22	3.7 ± 0.5	2.1[g]	< 0.01
37	Abandoned Cabin* A50	6.0 ± 0.5	6.3 ± 0.2	299 ± 113	338 ± 33	10.2 ± 1.1	2.3[g]	0.04
38	Scout A46	6.3 ± 0.4	7.0 ± 0.4	290 ± 36	347 ± 25	10.9 ± 0.4	4.7[g]	0.01
39	Engineer A45	6.7 ± 0.3	7.8 ± 0.4	273 ± 31	267 ± 43	7.0 ± 0.2	7.5[g]	< 0.01
40	Lower Ohmer A44	ND	7.5 ± 0.5	ND	379 ± 50	9.9 ± 0.5	1.8[g]	< 0.01
	Yedoma[i]	7.1[k] ± 0.5	8.2[k] ± 0.9	84[k] ± 27	187[k] ± 118	34.5[k] ± 18.0	27.9[k]	0.09[k]
	Non-Yedoma[j]	6.7[l] ± 0.5	7.7[k] ± 0.7	222[l] ± 95	295[l] ± 51	14.5[l] ± 21.8	5.3[l]	0.02[k]

Table 1. Continued.

N	Name	SO_4^{2-} ($mg\,L^{-1}$)	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	Julieta* 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

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Table 1. Continued.

N	Name	SO_4^{2-} (mg L^{-1})	TOC (mg L^{-1})	TN (mg L^{-1})	TOCS (%)	TNS (%)
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[j]	5.39[k]	10.1[l]	0.6[l]	10.0[k] ± 10.6	1.0[k] ± 0.6

[a] Doughnut L., a partially-drained lake (uncalibrated ^{14}C age 1190 ± 20 yr BP, measured on outer wood of an in situ, dead tree near the lake center), Smith L., and Monasta L. were included in the non-yedoma lake classification. While Doughnut and Monasta lakes likely formed in yedoma permafrost originally, following partial drainage events, they no longer appear to be influenced by active yedoma thaw along the margin. Smith Lake is thought to have formed as part of a previous river drainage network (V. Alexander, personal communication, 2011).

[b] Permafrost soil type: Y – Yedoma, NY – Non yedoma.

[c] Trophic State Index: UO – Ultraoligotrophic, O – Oligotrophic, M – Mesotrophic, E – Eutrophic, D – Dystrophic.

[d] Ecozonal categories according to Gregory Eaves et al. (2000): ArT – Arctic tundra, AlT – Alpine tundra, FoT – Forest tundra, NBF – Northern boreal forest, SBF – Southern boreal forest.

[e] Deposit Name: ES – Eolian silt, GF – Glaciofluvial, GMD – old Glacial moraines and drift, F – Fluvial, MAC – Mountain alluvium and colluvium, E – Eolian, GL – Glacio lacustrine (Jorgenson et al., 2008).

[f] Winter (October–April) temperature average from Hobo measurements.

[g] Data from Gregory Eaves et al. (2000).

[h] Data from Giblin et al. (2009); water-column average.

[i] Average from yedoma lakes (Lake #25 excluded).

[j] Average from non-yedoma lakes.

[k,l] Different letters indicate a significant difference between yedoma and non-yedoma means.

Table 2. Total annual CH_4 and CO_2 emissions by mode from 40 lakes along a north–south latitudinal transect in Alaska. * indicates informal lake names. Eb. Sum. – Direct Ebullition emission to the atmosphere from seeps during the ice-free summer season; Eb. Win. – Direct Ebullition emission to the atmosphere from seeps during the ice-cover winter season; IBS – Ice-bubble storage during spring ice melt; Stor. – Storage emission following ice-out; Diff. – Diffusive emission in summer, Total – Total annual emissions. If there was ND (no determination) for one or more modes in a lake, then total annual emission for the lake is likely an underestimate. Average emissions are summarized at the bottom of the table as is the percent of total annual emissions contributed by each mode as well as statistical results for differences in means among yedoma and non-yedoma lakes (Mann–Whitney test). Error terms represent standard deviation; *N* number of lakes analyzed; CF – Indicates impossible determination due to lake ice completely freezing to the lake bed in winter. CO_2 diffusive flux from lakes #17 and #18 were estimated from samples taken on multiple dates in June and July 2013 since no data were available in 2011–2012. Different letters[a,b] indicate a significant difference between yedoma and non-yedoma means.

N	Lake name	CH_4 (g m^{-2} yr^{-1})					
		Eb. Sum.	Eb. Win.	IBS	Diff.	Stor.	Total
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	Julieta* 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*	11.8	6.5	1.7	6.0	1.9	28.0
19	Doughnut *	ND	ND	ND	3.1	ND	3.1

1

(ignore this note)

Table 2. Continued.

N	Lake name	CH_4 ($g\,m^{-2}\,yr^{-1}$)					
		Eb. Sum.	Eb. Win.	IBS	Diff.	Stor.	Total
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)	25.9 ± 16.1^a	5.9 ± 3.6^a	5.7 ± 4.7^a	5.0 ± 1.4^a	1.2 ± 0.9^a	43.8 ± 17.3^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	

Table 2. Continued.

N	Lake name	CO_2 ($g\,m^{-2}\,yr^{-1}$)				
		Eb. Sum.	Eb. Win.	Diff.	Stor.	Total
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	1278	0	1279
18	Goldstream*	0.236	0.161	1582	0	1583
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	1136	ND	1138
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

Table 3. The Mann–Whitney and Kruskal–Wallis test results of the limnological and geographical characteristics of lakes using CH_4 or CO_2 emission mode as the factor. (≠) indicates a significant difference between limnological or geographical characteristic vs. flux; (=) indicates no significant difference at Z value < 1.96. IBS – Ice-Bubble Storage; Latitude: I – interior, N – northern, S – southern according to Sect. 2.1; Permafrost Soil Type (Y – yedoma/YN – non-yedoma); Trophic State Index (TSI), Ecozonal Categories (EC), Deposit type (DN), according to descriptions in Table 1; Maximum depth known (MD) and Area (A). In the MD analysis we considered two categories: shallow lakes ≤ 2.5 m and deeper lakes > 2.5 m. In the A analysis we considered two categories: small lakes ≤ 0.1 km^2 and large lakes > 0.1 km^2.

Emission mode	Latitude	Y/NY	TS	EC	DN	MD	A
CH_4							
Direct Ebullition (Summer)	I ≠ N-S	≠	O ≠ 2 UO	NBF ≠ ArT-SBF	=	=	≠
Direct Ebullition (Winter)	S ≠ I-N	≠	O ≠ D-UO	SBF ≠ FoT-NBF	E ≠ GMD-GL	=	≠
IBS	S ≠ I-N	≠	O ≠ D-UO	SBF ≠ FoT-NBF	E ≠ GL	=	≠
Diffusion	I ≠ N	≠	D ≠ O-UO	ArT ≠ NBF-SBF	=	=	=
Storage	=	=	=	=	=	=	=
Total	I ≠ S	≠	O ≠ D-UO	=	GL ≠ E-GMD	=	≠
CO_2							
Direct Ebullition (Summer)	I ≠ N-S	≠	O ≠ D-UO	NBF ≠ ArT-SBF	E ≠ GMD-GL	=	≠
Direct Ebullition (Winter)	S ≠ I-N	≠	O ≠ D-UO	SBF ≠ FoT-NBF	E ≠ GMD-GL	=	≠
Diffusion	I ≠ N	≠	=	NBF ≠ ArT-FoT-SBF	=	=	≠
Storage	=	=	=	=	=	=	=
Total	=	=	=	=	=	=	=

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-CH4) = −0.50Log(Area)	32	0.30	14.4919	0.0006
Direct Ebullition (winter)	Log(EW-CH4) = −0.93 – 0.68Log(Area)	28	0.60	43.6036	0.0000
	Log(EW-CH4) = 0.10 – 1.12Log(SecD)	28	0.23	9.3352	0.0050
	Log(ES-CH4) = −2.63 + 0.81Log(TN)	24	0.32	12.4092	0.0018
IBS	Log(IBS-CH4) = −0.83 – 0.64Log(Area)	29	0.58	50.705	0.0001
	Log(IBS-CH4) = 0.10 – 1.00Log(SecD)	29	0.19	7.9309	0.0088
Diffusion	Log(DF-CH4) = 0.55Log(PO_4^{-3})	24	0.40	16.7767	0.0004
Total	Log(Tot-CH4) = 0.43 – 0.37Log(Area)	38	0.27	15.0877	0.0004
	Log(Tot-CH4) = 1.01 – 0.77(SecD)	38	0.21	11.1414	0.0019
	Log(Tot-CH4) = 0.42 + 0.55Log(PO_4-3)	30	0.22	9.4969	0.0045
	Log(Tot-CH4) = 0.98 – 0.61Log(TN)	32	0.29	13.7928	0.0008
CO_2					
Direct Ebullition (summer)	Log(ES-CO2) = −1.72 – 0.50Log(Area)	32	0.30	14.6253	0.0006
Direct Ebullition (winter)	Log(EW-CO2) = −2.78 – 0.76Log(Area)	30	0.63	52.0960	0.0000
	Log(EW-CO2) = −1.83 – 0.76Log(TN)	26	0.24	9.0882	0.0058

1
TS is used in table

2
(ignore this note)

3
I think this line belongs to above summer section?

Table 5. Mann–Whitney and Kruskal–Wallis test results of the limnological and geographical characteristics of lakes using mode of dissolved gases concentrations (CH_4, O_2) during winter and summer. (≠) indicates a significant difference between a geographic characteristic and flux when $Z > 1.96$; (=) indicates no significant difference. Latitude: I – interior, N – northern, S – southern according to Sect. 2.1; Permafrost Soil Type (Y – yedoma/NY – non-yedoma); Trophic State Index (TSI), Ecozonal Categories (EC), Deposit type (DN) according to descriptions in Table 1; Maximum depth known (MD) and Area (A). In the MD analysis we considered two categories: shallow lakes ≤ 2.5 m and deeper lakes > 2.5 m. In the A analysis we considered two categories: small lakes ≤ 0.1 km^2 and large lakes > 0.1 km^2.

Dissolved Gas (Season)	Latitude	Y/NY	TS	EC	DN	MD	A
CH_4 (Winter)	I ≠ S	≠	D ≠ O	=	E ≠ GL, GMD	≠	≠
CH_4 (Summer)	I ≠ N, S	≠	D ≠ O, UO	NBF ≠ ArT, SBF, FoT	E ≠ GMD	=	≠
O_2 (Winter)	I ≠ S	≠	D ≠ O	=	E ≠ GL, GMD	=	≠
O_2 (Summer)	I ≠ N, S	≠	D ≠ O, UO	NBF ≠ ArT, SBF, FoT	E ≠ GL, GMD	=	≠

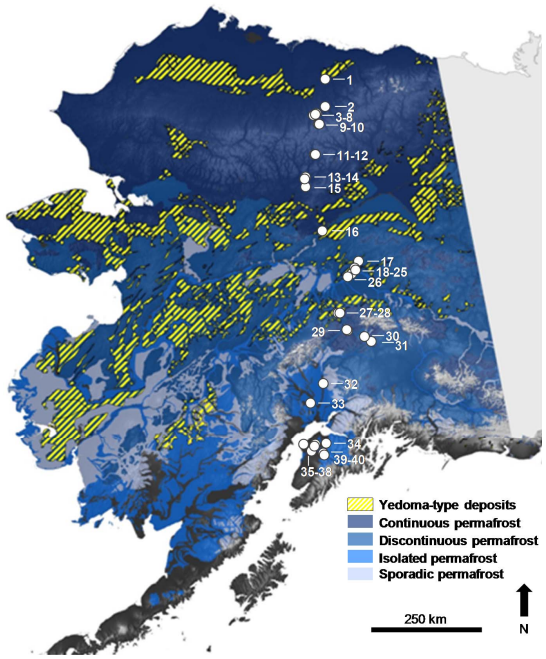
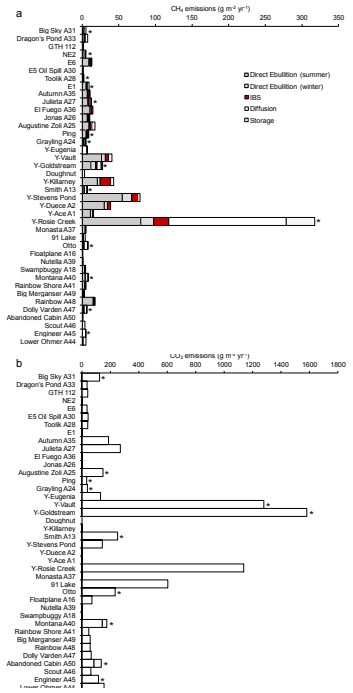


Figure 1. Locations of study lakes in Alaska (circles) plotted on the Alaska DEM hillshade raster. Information about the distribution of yedoma-type deposits (ice-rich silt containing deep thermokarst lakes) and permafrost was from Jorgenson et al. (2008) and Kanevskiy et al. (2011). The Alaska map is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



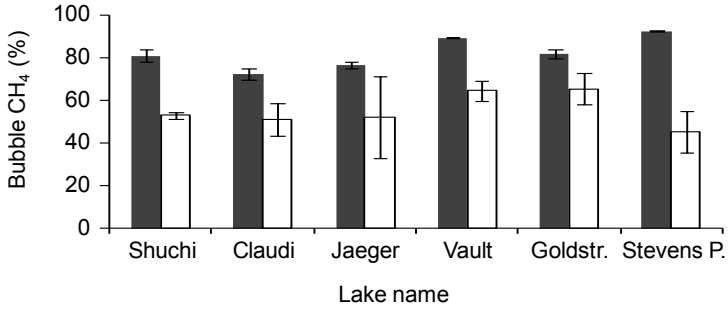


Figure 3. Average CH_4 concentrations in ebullition bubbles collected at the lake surface before interaction with lake ice ("fresh bubbles", grey bars) and in ebullition bubbles trapped by the lake ice (white bars). Error bars represent standard error for $n = 2$ to 41 seeps per lake. Among lakes, CH_4 concentrations in ice-trapped bubbles were 33 ± 12 % lower than in fresh bubbles (Mann–Whitney U test, $Z > 1.96$, $p < 0.05$).

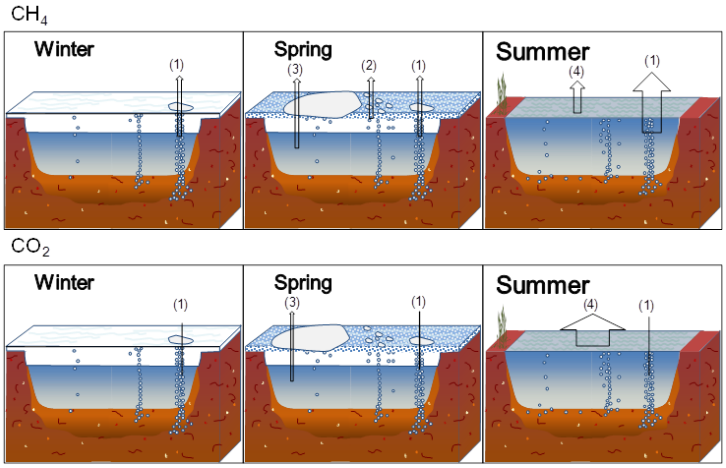


Figure 4. Illustration of CH_4 and CO_2 emissions pathways during different seasons in Alaskan lakes. The thickness of arrows indicates the relative magnitude of contribution from each pathway according to Table 2: (1) Direct Ebullition through ice-free Hotspot seeps in winter and from all seep classes during the last month of ice cover in spring and in summer; (2) Ice-Bubble Storage (IBS) emission during spring ice melt; (3) Storage emission of dissolved gases accumulated under lake ice when ice melts in spring; (4) Diffusion emission from open water in summer.

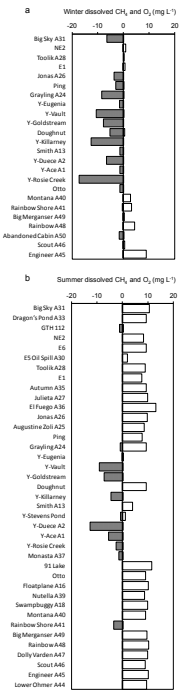


Figure 5. Average dissolved CH_4 (gray bars) and O_2 (white bars) concentrations in lake bottom water during winter **(a)** and summer **(b)**. Yedoma lakes are indicated by "Y". In winter, Spearman coefficient $p = 0.58$ indicates a moderate positive correlation between dissolved CH_4 and O_2; in summer $p = 0.70$ indicates a strong positive correlation.

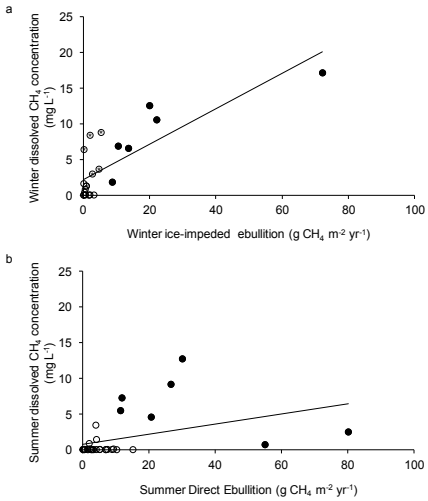


Figure 6. Dissolved CH_4 concentrations measured in lake bottom water vs. winter ice-impeded ebullition in winter **(a)** and Direct Ebullition in summer **(b)**. The Spearman coefficients, $p = 0.72$ and $p = 0.42$ indicate a strong positive correlation and a weak positive correlation in winter and summer, respectively. All lakes were considered a single population; however, yedoma lakes (closed circles) had higher concentrations of lake-bottom dissolved CH_4 (mean ± SD: 9.3 ± 5.4 mg L^{-1} winter, 6.7 ± 4.1 mg L^{-1} summer) and a higher density of ebullition seeps (Sect. 3.2) than non-yedoma lakes (open circles; 2.1 ± 3.0 mg L^{-1} winter, 0.3 ± 0.7 mg L^{-1} summer). We observed relatively high concentrations of dissolved CH_4 in some non-yedoma lakes in winter due to dissolved gas exclusion during ice formation in shallow lakes that nearly froze to the lake bed, indicated by *. Excluding lakes that nearly froze to the lake bed, the mean dissolved CH_4 in the remaining non-yedoma lakes was 0.3 ± 0.5 mg L^{-1} in winter.