Québec, 24 November 2015

Dr. Jorien Vonk, Associate Editor
Biogeosciences (BG)

Re: 'Modern to millennium-old greenhouse gases emitted from ponds and lakes of the Eastern Canadian Arctic (Bylot Island, Nunavut)', by F. Bouchard, I. Laurion, V. Prėskienis, D. Fortier, X. Xu, and M. J. Whiticar

Dear Editor,

We hereby submit the revised version of the above manuscript for publication in BG special issue 'Freshwater ecosystems in changing permafrost landscapes'.

We thank you and both referees for the comments on the originally submitted version of our manuscript (# bg-2015-314). We have carefully considered all the comments and have modified the manuscript accordingly. In the following pages, you will find a point-by-point response to the comments and questions, and when appropriate a reference to the line numbers corresponding to the relevant changes made in the new manuscript (submitted separately as a PDF file). We also attached to this letter a marked-up manuscript version (track changes mode in Word, then saved as PDF) compiling all the changes made.

Please let us know if you have any other question.

Yours sincerely,

Frédéric Bouchard (corresponding author)
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Associate Editor (J. Vonk)
Received and published: 10 November 2015
Decision: «Publish subject to minor revisions».

1. Referee #2 states in his specific comment to p. 11664, l. 6 that - shortly said - the strength of the feedback is not related to age of C released. You answer this comment, but I am not sure if this is clear enough. Emission of OLD carbon is a source of long-term stored carbon that is now re-activated/remobilized and added to the active carbon cycle, whereas YOUNG carbon is already part of the active carbon cycle and will therefore not represent an additional carbon source. In my opinion, the climate feedback for old C is therefore more relevant/stronger than for young C.

We agree with the Editor that the emission of old carbon results in a significant climate feedback, as opposed to modern carbon. We added a paragraph in the answer to the general comment of Referee #2 (see below), and modified the text accordingly (lines 70-72; 80-83; 454-456). ***Note that the following line numbers refer to the 'clean' revised manuscript, submitted separately, and not the marked-up manuscript attached at the end of this letter.

2. I do not find lake or pond depth in any of the tables, only sampling depth - is that correct? I was wondering if you can find a correlation of average age of CO2 or CH4 related to lake and pond depth (or maybe even depth of the thaw bulb?), since sediment depth does to some extent correspond to depth of the deposits that are degrading. This is just an idea.

This is an interesting idea; however the depth *vs*. GHG age data are not sufficient to check for statistical correlations (N = 5 and N = 6 for 2013 and 2014, respectively), and the depth data do not follow a normal distribution anyway. Ponds are mostly clustered around 1-m depth, whereas lakes show either mid-range (4-5 m for thermokarst lake BYL66) or highest depth (11-12 m for kettle lake BYL36). This idea could be further tested in the future, though, with a more robust dataset.

Pond and lake depths can be deduced from Fig. 3 as the maximum sampling depth.

Referee #1 (Anonymous)
Received and published: 4 September 2015
«Needs to expand on some data, otherwise a fine paper.»

Specific Comments:

(1) I want to know more about the C-13 and D stable isotopes in the lakes. Figure 7 only shows 6 lake samples simply labeled as "lake". From my own research I have seen large differences in these isotopes based on where they were collected in an Arctic lake. Samples collected from the perimeter and the surface tended to show more AM influence while samples collected from the surface interior and even more so at depth in the lake showed more HM influence.
In Figure 4 I see 5 lake samples that show large differences in age based on their location, which is interesting. It would be nice if the data of Figure 7 was similarly labeled with the location, allowing for more of a direct comparison between the two figures of these data points.

As suggested by the Referee, we added location labels for lake samples in Figure 7 ('edge', 'center', and 'undiff.' [undifferentiated]). See the new Fig. 7 for details. Indeed, the isotopic signature of the center lies towards the HM region (grey background area) even though it still clearly remains in the AM region. We also added a sentence in the discussion to acknowledge this observation (lines 472-475).

As the Referee remained anonymous, we have to assume that this result has not been yet published (at least not to our knowledge). We also assume that when the Referee says that the signature tended to show more HM "at depth", it means the signature was obtained from diffusive gas samples. Contrastingly, all of our samples for stable isotope analysis were collected from an ebullition funnel.

(2) Despite of this paper's great attributes, it is a dense read. There are a lot of long paragraphs and long sentences, which make all of the information difficult to digest and the paper difficult to read. I think the paper would appeal to a broader biogeochemical group of scientists if the authors broke up some of the long paragraphs and sentences to shorter versions. This is a suggestion, but I think it would benefit the paper in the long run greatly.

We made an effort to shorten the text wherever possible, especially in the Study area section and in the Discussion. As suggested by the Referee, we shortened the text or broke up some of the longest sentences.

(3) The presences of the nutrient data confuses me. There is one table and one paragraph describing the nutrient data. The nutrient data adds nothing to the discussion, yet it is present in the results. Either the nutrient data should be expanded on or cut.
If the authors chose to keep the nutrient data in the paper they should discuss specific nutrient species (TN vs NO3, etc.) rather than all grouped together (how it is currently presented). Also lacking are statistics on the nutrient data. Are there real statistical differences in TN, Fe, Mn, etc., between the lakes or all they the same? A simple ANOVA analysis would answer that question.
I think the nutrient data could help the readers understand the differing biological regimes of the lakes and ponds better (like why there are algal mats in some), but it needs to actually be discussed in the text not just shown in a table.

The nutrient data are presented here as baseline limnological information to help characterizing the studied ponds and lakes for people who want to compare with their systems, considering the large variability in Arctic aquatic systems. On the other hand, we agree that it is relevant to discuss more about them and exploit their value. As suggested by the Referee, we ran an ANOVA analysis for all nutrients. We added new sentences in the Results (lines 254-260). The analysis suggests that only DOC differed significantly between the water bodies during both sampling years.

We further explored the relationships between nutrients and GHG fluxes and found a significant correlation between CO2 and CH4 fluxes, and nutrients related to organic matter (DOC and the chromophoric (coloured) fraction of dissolved organic matter, CDOM) in 2013. This is important and underscores to link between aquatic system limnology and biogeochemistry. We added new sentences in the Results (lines 292-297).

Referee #2 (M. Langer)
Received and published: 9 September 2015
«Some minor issues.»

General Comments:

The study is written very well and understandable. However, a few points concerning the discussion around climate feedback mechanisms require clarification (see specific comments).

We clarified this as suggested (see answers to specific comments below).

In short, we state that emission of old (millennium-aged) carbon is indeed a source of long-term stored carbon (formerly trapped in syngenetic permafrost) that is now re-activated or remobilized in the system, and added to the active carbon cycle. Contrastingly, young (modern) carbon is already part of the active carbon cycle (through short-term processes such as photosynthesis), and will therefore not represent an additional carbon source.

Furthermore, the method section should include a description of the used temperature loggers and sensors (accuracy, location, and so on).

We added a sentence about the temperature data loggers in the Methods section (lines 166-169).

In addition, the calculated CH4 and CO2 fluxes are based on coarse assumptions (average atmospheric concentrations) and a very basic diffusion model. Therefore, it would be recommend to provide realistic uncertainty estimates. Straight forward error propagation methods (e.g. Monte-Carlo simulations) should be applicable. In particular, the available statistics from multiple concentration measurements should be used and could be further investigated. It should be clarified whether the calculated flux magnitudes and directions are significant.

We ran statistical tests on our CO_2 and CH_4 data (including normal distribution, variance, and mean testing). We found that trough ponds (IWT type) were significantly different from the other two types of water bodies (polygonal ponds and lakes), but also from each other. We slightly modified the text accordingly (lines 292-294).

In addition, as mentioned above we tested the statistical relationships between GHG fluxes and nutrients, and found a significant correlation between CO2 and CH4 fluxes, and nutrients related to organic matter (DOC and CDOM) in 2013. We modified the text accordingly (lines 294-297).

It might be also interesting to calculate the vertical C-balance of the different water body types in order to clearly label them as sink or source for atmospheric carbon. For numerous polygon ponds the CO2 gradient seems to be on the order of about -5 μ mol to -10 μ mol which for some ponds is partly balanced by the CH4 gradient (Fig. 5). Taking into account some uncertainties in the atmospheric CO2 and CH4 concentrations and additional CH4 fluxes from ebullition, the C-balance of some ponds might shift from negative to neutral. A short uncertainty analysis is highly recommended as outlined above.

Table 3 could be partly translated into a simple (arrow) diagram in order to illustrate the flux differences.

As suggested by the Referee, we produced an arrow diagram showing the net median fluxes of both CO_2 and CH_4 (diffusion and ebullition merged together) for each water body type. See the new Figure 9 for details. This new figure synthesizes the information from Table 3 without repeating it unnecessarily. As mentioned above (previous comment), we can conclude that trough ponds differ from the other water body types, as they can be considered as a significant GHG source (especially CO_2), whereas the other types are either small net sinks (polygonal ponds) or small net sources (lakes).

Moreover, considering a global warming potential (GWP) of 34 for CH_4 on a 100-year horizon (IPCC Fifth assessment report; see Myhre et al., 2013 in the references), trough ponds still had the highest net carbon balance, higher than the contribution from polygon ponds and lakes taken together. We added a sentence in the discussion (lines 387-390).

The result section appears short in comparison to the discussion. I recommend to embed descriptions of Fig.6 - 8 into the result section.

As suggested by the Referee, we further described these figures in the Results (lines 298-313).

Specific Comments:

Title: Maybe "the eastern Canadian Arctic" is a bit too general since all investigations were carried out on Bylot Island which might be not representative for the entire eastern Canadian Arctic.

We added 'Bylot Island, Nunavut' in the title. To our knowledge however, this work is the first to report on GHG age differences across the Eastern Canadian Arctic (apparently from the whole Canadian Arctic), so we preferred to keep this part. The modified title is now '*Modern to millennium-old greenhouse gases emitted from ponds and lakes of the Eastern Canadian Arctic (Bylot Island, Nunavut)*'.

p.11664; l.6: The statement that the strength of climate feedback is determined by the age of the released carbon requires clarification. Why would carbon that is 5000 years old cause a stronger climate feedback than carbon that is only 500 years old? I completely understand that it makes a difference whether old carbon can be processed or not. However, this would not affect the carbon-climate feedback mechanism, but change the size of the carbon pool that can be activated. I agree that this is an important question which is also reflected in number publications which discuss the permafrost carbon pool every year in very important journals.

We modified the sentence (also in response to the other comment below) to clarify that the strength of climate feedback is not related to the age in years per se (5000 years old is not less efficient than 40 000 years old, as long as it is in excess), but rather that if it's modern it does not generate a significant positive feedback, as opposed to if it's old, or in excess in the system (lines 70-72; 80-83).

Permafrost on Bylot is syngenetic, meaning that carbon has been progressively sequestered (or 'locked') in the frozen ground since the beginning of permafrost inception (i.e. since the mid-

Holocene deglaciation). Thermokarst processes are now doing the opposite, i.e. releasing excess carbon (century to millennium-aged) in the system.

p.11664; l.9-12: I recommend to use the terms "glaciated" or "covered by ice sheets" instead of "ice covered". This might be picky, but it reminds the reader on the extend of the ice cover.

We changed for 'ice sheets' (2nd sentence) as recommended (line 77), but we kept 'ice-covered' (1st sentence) because we mention the last glaciation just after, in the same sentence (line 75).

p.11664; l.15: I would say "contribute to positive climate feedback if released as GHGs".

We modified the sentence as suggested (lines 80-81).

p.11664; l.15-17: Is it possible that water bodies act as carbon sinks under current climate conditions and change to carbon sources or become neutral under warming? Anyways, the atmospheric GHG budget would be affected and, thus, a climate feedback would exist even though relatively modern carbon is processed. However, I agree that the size of the carbon pool that could become available due to the thaw of permafrost is important. In general, I suggest to reduce the argumentation on feedback mechanisms which are not explicitly in the focus of this study. The identification of carbon pools and their pathways and magnitudes of release already make up very good justifications.

We agree that feedback mechanisms are not the focus of this work. We toned down the argumentation as recommended. We think it is nevertheless important to mention this factor in the Introduction and discuss it, as we aimed at identifying aquatic carbon sources of different ages, which is of central importance in the general 'endeavor' of upscaling and modeling GHG emissions at larger spatial scales. As mentioned above, we clarified our climate feedback argumentation, focusing on old (excess) *vs*. modern carbon, and not on a linear age relationship (lines 70-72; 80-83).

p.11666; l.22-23: The information about birds might be not necessary. In general, this section could be condensed a bit. The study site description appears long compared to the result section.

We removed the information about birds, and shortened the Study area section.

p.11672; l 22-23: It should be clearly indicated that the measured temperature and oxygen profiles are not representative for entire July. The measured profiles depict a specific situation. Shallow water bodies such as polygon ponds can change their stratification within a few hours according to wind speed. Furthermore, it is very interesting that the temperature profiles c and d in Fig. 3 show bottom temperatures well below 4°C. Is there any explanation for this?

I guess the Referee refers to p. 11671 (l 22-23), instead of p. 11672? If so, the measured temperature and oxygen profiles (Fig. 3) are indeed representative of the entire month of July, in fact of almost the entire ice-free season. Figure 4 (formerly Figure 6) shows that BYL27 (trough pond) is well stratified during the summer, and we have unpublished data (to be included in a forthcoming paper) showing that polygonal ponds are well mixed (and lakes partly

stratified) through most of the ice-free season. We added a sentence in the Results (lines 266-268).

Regarding the temperature profiles (c-d) in Fig. 3 obtained in two ice-wedge trough ponds (BYL24 and BYL27), the bottom temperature was indeed near 0°C because this layer of water is lying just above the melting ice wedge (as part of the "active layer"), does not mix with surface waters, and is cooled down through sensible heat transfer. This is now briefly underlined in the Discussion (lines 391-393).

p.11674; l.18: I suggest to be more precise with the term feedback mechanism. Landscape features or elements such as a pond or a lake are not mechanisms per se. These landscape features can introduce processes which are relevant for climate feedback mechanisms. I think, a climate feedback mechanism is e.g. increased CH4 emission of an ecosystem due to warming. This could be caused by the formation or extension of lakes due to permafrost degradation and/or by general changes in the biogeochemical processes due to warmer conditions.

We changed the end of the sentence to avoid any confusion (lines 335-337), and did so throughout the manuscript as mentioned above (lines 70-72; 80-83; 454-456). However, as stated above we define the climate feedback effect based on the difference between 'old' carbon (i.e. in excess in the system, regardless of its absolute age) and 'modern' carbon that is constantly used and recycled through short-term biogeochemical processes.

p.11678, l.22-26: This statement is only true under the assumption that only the number of lakes increases with climate warming while all ecosystem processes remain the same. The presented data give no evidence that CO2 emission or uptake of Arctic water bodies will not change with climate warming. This study investigates the current state of an ecosystem from which the response to climate warming is difficult to derive. Nevertheless, it is an important finding that under current conditions the investigated ponds are sources of CH4 but sinks of CO2. However, this state might or might not change under climate warming.

We agree that our work is valid for the current state of these aquatic systems; we also acknowledge at the beginning of the sentence that we are only considering 'ice-free season ebullition'. We added 'under current climate conditions' at the end of the sentence to clarify this aspect (line 454-456).

p.11681; l.19: Does thaw bulb mean talik?

Yes. We modified that as suggested (line 535).

Fig. 4: What indicates the separation line? CH4 and CO2 are already distinguished by filled and unfilled circles.

We added the vertical line to clearly separate CH_4 and CO_2 data, but also to insist on the substantial difference in partial pressure (horizontal axis) for each gas, with at least one order of magnitude difference.

Fig. 5: I suggest to be consistent with units. Does [M] indicate mole? The text uses [m mol] frequently.

The unit [M] (‘molar’) refers to a concentration (= mol L^{-1}), whereas we used the unit ‘mmol’ (as in mmol m^{-2} d^{-1}) when referring to fluxes. We think both units are relevant, standard, and commonly used in the literature (International System).

Modern to millennium-old greenhouse gases emitted from ponds and lakes of the Eastern Canadian Arctic (Bylot Island, Nunavut)

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Keywords: freshwater ecosystems, thermokarst, GHG flux, stable isotopes, radiocarbon dating, continuous permafrost, ice-wedge polygons.

Abstract

Ponds and lakes are widespread across the rapidly changing permafrost environments. Aquatic systems play an important role in global biogeochemical cycles, especially in greenhouse gas (GHG) exchanges between terrestrial systems and the atmosphere. The source, speciation and emission of carbon released from permafrost landscapes are strongly influenced by local conditions, hindering pan-Arctic generalizations. This study reports on GHG ages and emission rates from aquatic systems located on Bylot Island, in the continuous permafrost zone of the Eastern Canadian Arctic. Dissolved and ebullition gas samples were collected during the summer season from different types of water bodies located in a highly dynamic periglacial valley: polygonal ponds, collapsed ice-wedge trough ponds, and larger lakes. The results showed strikingly different ages and fluxes depending on aquatic system types. Polygonal ponds were net sinks of dissolved CO_2, but variable sources of dissolved CH_4. They presented the highest ebullition fluxes, one or two orders of magnitude higher than from other ponds and lakes. Trough ponds appeared as substantial GHG sources, especially when their edges were actively eroding. Both types of ponds produced modern to hundreds of years old (< 550 yr BP) GHG, even if trough ponds could contain much older carbon (> 2000 yr BP) derived from freshly eroded peat. Lakes had small dissolved and ebullition fluxes, however they released much older GHG, including millennium-old CH_4 (up to 3500 yr BP) from lake central areas. Acetoclastic methanogenesis dominated at all study sites and there was minimal, if any, methane oxidation in gas emitted through ebullition. These findings provide new insights on GHG emissions by permafrost aquatic systems and their potential positive feedback effect on climate.

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1 Introduction

Permafrost stores large quantities of carbon compared to the atmosphere, although quantitative estimates are still under discussion (Tarnocai et al., 2009; Hugelius et al., 2014). Climate warming impacts Arctic landscapes through permafrost thawing and erosion (Romanovsky et al., 2010). This results in the release of both old and recent organic carbon to the atmosphere as greenhouse gases (GHG) (Zimov et al., 2006; Schuur et al., 2015). Widespread across permafrost environments, aquatic systems act as biogeochemical hotspots by releasing substantial amounts of carbon dioxide (CO_2) and methane (CH_4) (e.g., Walter et al., 2007; Laurion et al. 2010; Abnizova et al., 2012). It is generally considered that CH_4 ebullition is the main mechanism of GHG emissions from ponds and lakes, a transport mechanism highly heterogeneous in space and time (Wik et al., 2011). However, other processes, such as emissions through diffusion (Bastviken et al., 2008), plant-mediated transport and microbial oxidation (Bastviken et al., 2004; Liebner et al., 2011), also need to be considered in the specific context of the Arctic. Moreover, lateral inputs of CH_4 produced within the active layer or lateral export of permafrost carbon away from thaw sites via streams and rivers were recently demonstrated (Vonk and Gustafsson, 2013; Godin et al., 2014; Paytan et al., 2015). Overall, thermokarst (thaw) ponds and lakes represent a major landscape feature in permafrost-affected regions (Grosse et al., 2013), and there is a growing interest in defining the specific role of various types of freshwater ecosystems in global carbon dynamics associated to permafrost degradation processes, and how they may rapidly respond to environmental changes (see Vonk et al., 2015 and other articles in the present special issue).

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Supprimé: could be underestimated especially in the case of small systems, as flux values are often computed from wind-based empirical models developed for larger lakes located in different climatic regions (

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Supprimé: ; Tedford et al., 2014). Other processes involved in GHG dynamics, such as

Upscaling and modeling GHG emissions is challenging, and oversimplified assumptions can lead to large calculations errors (Stepanenko et al., 2011; van Huissteden et al., 2011; Gao et al., 2013). The gaps that need to be fullfilled to model future GHG emissions with more accuracy include defining the vertical distribution of carbon in permafrost soils accross the Arctic, the interactions between permafrost thaw and surface hydrology, as well as distinguishing CH_4 from CO_2 emissions and gradual warming from abrupt thaw mechanisms (Schuur et al., 2015). Regarding thermokarst systems specifically, aspects that should be further investigated include physical (e.g., heat transfer, diffusive GHG exchange, daily storage flux) and hydrological (e.g., surface and groundwater flows) dynamics, as well as fluxes of particulate (in addition to dissolved) organic carbon from thawing permafrost within these systems (Vonk et al., 2015). Another important yet rarely considered aspect is the age (old *vs.* modern) of the carbon that is processed and released by these biogeosystems, which is linked to their potential to generate a positive feedback on climate (Walter et al., 2006; Vonk et al., 2013; Mann et al., 2015). Large GHG emissions (especially CH_4) from old (late Pleistocene-age) organic ice-rich loess permafrost (*yedoma*) have been reported from thermokarst lakes of Siberia and Alaska in regions that were not ice-covered during the last glaciation (Zimov et al., 1997; Brosius et al., 2012). In Canada, which accounts for a very large portion of circum-Arctic permafrost, these deposits are rare as the territory was almost entirely covered by ice sheets during that period (Dyke and Prest, 1987). The carbon trapped in permafrost is thus younger (Holocene-age) in this part of the northern hemisphere (e.g., Allard, 1996; Burn and Kokelj, 2009; Lauriol et al., 2010; Tremblay et al., 2014). It nevertheless represents an excess carbon stock that can contribute to accelerate climate warming via a positive feedback mechanism if released as GHG, compared to modern

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carbon that is used and recycled through short-term biogeochemical processes (photosynthetic fixation and microbial respiration).

Preliminary data on GHG radiocarbon age from small tundra ponds on Bylot Island (Nunavut) in the Eastern Canadian Arctic showed that the carbon released by these systems was generally modern (Negandhi et al., 2013). The objective of the present study was to further characterize GHG composition, production pathway, age and emission rates in ponds and lakes at this particular site. We analyzed dissolved and ebullition gas samples collected in July from ponds and lakes located within an organic-rich permafrost terrace of Late Holocene age (Fortier et al., 2006).

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2 Study area

Bylot Island (Nunavut) is located in the Eastern Canadian Arctic, within the continuous permafrost zone (Fig. 1). The Byam Martin Mountains run southeast–northwest across the island, and the plains that stretch out on either side of the mountains belong to the Arctic Lowlands physiographic region (Bostock, 1970). The numerous valleys formed in the lowlands were shaped during the successive Pleistocene glaciations (Klassen, 1993). Since the Holocene, these valleys developed highly dynamic biogeosystems rich in permafrost ground ice, peat, and aquatic environments (Fortier and Allard, 2004). The study site (73° 09' N; 79° 58' W) is located in one such valley (glacier C-79) named Qarlikturvik, which has a NE-SW orientation and a surface area of ~ 65 km^2 (~ 15 km-long x 4-5 km-wide). A terminal moraine, located about halfway between the actual glacier front and the seashore and sitting on marine clay, was ^{14}C-dated to ~ 9.8 k yr BP (Allard, 1996). Glacial retreat, accompanied by a marine transgression

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141 phase, ended around 6 k yr BP. The clays were then covered by glacio-fluvial sand and gravels
142 (Fortier and Allard, 2004). Today, a proglacial braided river runs through a glacio-fluvial outwash
143 plain and drains glacier melt waters and sediments towards the Navy Board Inlet, where it forms
144 a delta.
145 The outwash plain is bordered on both sides by a 3 to 5 m-thick terrace, criss-crossed by
146 networks of tundra polygons associated with the formation of syngenetic ice wedges (Figs. 1d
147 and 2a). Along the southern bank of the river, the upper portion of the terrace is composed of
148 alternating organic (peat) and mineral (wind-blown sand and silt) material, which started to
149 accumulate over glacio-fluvial sands and gravels around 3700 years ago (Fortier and Allard,
150 2004). These peaty loess deposits contain excess pore ice (> 100% dry weight) and their
151 gravimetric organic matter content can reach over 50%. The active layer depth in such deposits
152 generally ranges between 40 to 60 cm, and the maximum depth of permafrost on Bylot Island
153 has been estimated to be over 400 m (Smith and Burgess, 2000). The terrace comprises
154 abundant aquatic systems of different sizes and shapes (Fig. 2) that can act as effective
155 biogeochemical hotspots (Laurion et al., 2010; Negandhi et al., 2013). The hydrological network
156 is mainly fed by rain and snowmelt runoff originating from gullies of the valley flanks or large
157 snow banks on the lee side of hills. Most of water loss from ponds and lakes is through
158 evaporation during the ice-free season (Negandhi, 2013).
159 The climate normal (1981-2010) is provided by a meteorological station located near the village
160 of Pond Inlet (Mittimatalik) (72° 41’ N; 77° 58’ W), about 85 km southeast from the study site
161 (Fig. 1c). The region has a polar climate with a slight marine influence, a mean annual air
162 temperature of -14.6°C (average daily temperatures ranging from -33.4°C in January to 6.6°C in

July) and total precipitations of 189 mm, of which 91 mm fall as rain between June and September (Environment Canada, 2015). Thawing and freezing degree-days are around 475 and 5735, respectively. Winter (continuous daily mean air temperature < 0°C) lasts from early September to mid-June, for an average total of 283 days per year. A station from the SILA network, operated since 2004 by the Center for Northern Studies (CEN) in the valley of glacier C-79, provides similar climate data (CEN, 2014).

The southwest plain of Bylot Island is a ~ 1600-km^2 low-lying wetland area of graminoid-moss tundra (Parks Canada, 2014). Local vegetation in the Qarlikturvik valley is dominated by sedges (e.g. *Carex aquatilis* var. *stans*, *Eriophorum scheuchzeri*), grasses (e.g. *Arctagrostis latifolia*, *Dupontia fischeri*, *Pleuropogon sabinei*) and mosses (e.g. *Drepanocladus* spp., *Aulocomnium* spp.) (Duclos, 2002; Ellis et al., 2008).

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Supprimé: . The mean annual air temperature over the last 10 years was -14.5°C (with daily temperatures ranging from -34.7°C in January to 6.2°C in July), and total annual precipitations average 220 mm, of which 94 mm fall as rain (June to September)

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3 Materials and Methods

3.1 Sampling sites

We selected and sampled different types of aquatic systems typical of the tundra polygon terrace of the valley (Fig. 2; Table 1): 1- polygonal ponds over low-centered ice wedge polygons; 2- elongated water channels over melting ice wedges (ponds formed in collapsed ice-wedge troughs, hereafter referred to as trough ponds); 3- lakes with underlying talik (unfrozen soil over permafrost), including a thermokarst (thaw) lake and a kettle (melted buried glacier ice) lake. A total of 23 ponds and lakes were sampled in June-July 2013, including 9 polygonal ponds, 12 trough ponds, and 2 lakes (1 thermokarst and 1 kettle lake). In July 2014, six water bodies (two polygonal ponds, two trough ponds, and two lakes including one thermokarst and one kettle

lake) were selected and studied more intensively, including morphological measurements of ponds (depth, width and length) and lakes (bathymetry with a portable sonar as in Bouchard et al., 2015), and limnological profiles (see below).

3.2 Limnology

We measured a suite of limnological characteristics during both years, including temperature, dissolved oxygen, concentrations of dissolved organic carbon (DOC), chromophoric fraction of dissolved organic matter (CDOM), nutrients (phosphorus, nitrogen) and major ions. Temperature and dissolved oxygen profiles were recorded with a ProODO handheld meter (YSI Inc.). Water samples were filtered through 0.2 µm pre-rinsed cellulose acetate filters (2013) or pre-combusted GF/F filters (2014, nominal porosity 0.7 µm) to analyze DOC and major ions. Cations were fixed with HNO_3 (0.15 % final concentration) while anions and DOC were not fixed but kept in dark and cold. DOC concentrations were measured with a Shimadzu TOC-5000A carbon analyzer calibrated with potassium biphthalate, and CDOM was quantified (in 2013 only) with the absorption coefficient of DOM at 320 nm (a_{320}) obtained on a Cary 300 (Varian; methodological details in Laurion and Mladenov, 2013). Major anions were quantified by ionic chromatography (Dionex ICS-2000), whereas major cations by inductively coupled plasma – optical emission spectrometry (ICP-OES, Varian VISTA AX). Total phosphorus (TP) and total nitrogen (TN) were quantified from unfiltered water samples fixed with H_2SO_4 (0.15% final concentration) as described by Stainton et al. (1977). Finally, the thermal structure of one trough pond (BYL27) was assessed during a full year (July 2013 – July 2014) by recording water temperature at two depths (0 and 50 cm) at a 15-minute interval using two submersible data loggers (Vemco Minilog-II-T, accuracy ± 0.1°C, resolution ± 0.01°C) installed on a mooring line.

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The line was not moored at the deepest point of the pond (~1 m) as found upon its retrieval, but the data still provide a clear picture of the thermal stratification establishing in this type of humic ponds.

3.3 Ebullition flux of greenhouse gases

Ebullition gas samples were collected using submerged funnels (as in Wik et al., 2013) equipped with a 140-mL plastic syringe (Fig. A1 in Appendix) and deployed for a period of 1 hour to 19 days depending on the flux. The samples trapped in the syringe were transferred into 1- 50-mL glass bottles with butyl rubber stoppers (bottles acid-washed, pre-combusted, helium flushed and vacuumed) for ^{14}C dating (see below), and 2- 6-mL glass vials (helium flushed and vacuumed Exetainers) for stable isotope (see below) and gas chromatography analysis (Varian 3800 with a COMBI PAL head space injection system and a CP-Poraplot Q 25 m 3 0.53 mm column, flame ionization detector). Ebullition flux (F_e, in mmol m^{-2} d^{-1}) was calculated as:

$$F_e = (pGas \times V) / (A \times MV \times t)$$

where pGas is the partial pressure of CO_2 or CH_4, V is the collected gas volume, A is the funnel area, MV is the gas molar volume at ambient air temperature, and t is the collecting time.

3.4 Diffusive flux of greenhouse gases

Surface water dissolved GHG concentrations were obtained by equilibrating 2 L of lake or pond water with 20 mL of ambient air during 3 minutes (Hesslein et al., 1991). The resulting gaseous headspace was transferred into 6-mL glass vials and analyzed as above by gas chromatography. Dissolved GHG concentration at the surface (C_{sur}) was calculated using Henry's law, and departure from saturation (sink *vs*. source) was calculated subtracting the gas concentration in the water at equilibrium with the atmosphere (C_{eq}, global values of atmospheric partial

243 pressures from IPCC, 2007 were used). To estimate diffusive flux ($Flux_d$), first the gas transfer
244 coefficient (k_{600}) standardized to a Schmidt number (Sc) of 600 (Wanninkhof, 1992) was
245 calculated with the wind-based model of Cole and Caraco (1998):

246 $$k_{600} = 2.07 + 0.215\ u_{10}^{1.7}$$

247 where u_{10} is the wind speed at 10 m above the ground, and then applying the equation:

248 $$Flux_d = k\ (C_{sur} - C_{eq})$$

249 where k is the gas transfer coefficient for a given gas calculated as:

250 $$k = k_{600}\ (Sc/600)^{-0.5}$$

251 **3.5 Radiocarbon analysis**

252 Ebullition gas samples were analyzed at the Keck Carbon Cycle AMS facility at the University of
253 California, Irvine. First, CH_4 and CO_2 were separated and purified by a zero air carrier gas flow-
254 through line (Pack et al., 2015), and graphitized by the sealed tube Zn reduction method (Xu et
255 al., 2007), then measured for radiocarbon (^{14}C) on a compact accelerator mass spectrometer
256 (AMS) (Southon and Santos, 2007). Data presented here are expressed as $\Delta^{14}C$ (‰), which is
257 normalized to radiocarbon activity of an oxalic acid standard OX1 (decay corrected to 1950) and
258 corrected for isotopic fractionation (Reimer et al., 2004). $\Delta^{14}C$ (‰) > 0 was further used to
259 indicate 'modern' carbon (1950 to present), and $\Delta^{14}C$ (‰) < 0 for "older" carbon (pre-1950).
260 This was particularly helpful for polygonal and trough ponds, which provided modern or very
261 young GHG. The $\Delta^{14}C$ analytical error was ~ 2‰ for modern sample, based on long-term
262 measurements of secondary standards. ^{14}C age (yr BP) is as defined by Stuiver and Polach
263 (1977).

3.6 Stable isotope analysis

Stable carbon and hydrogen isotopic compositions of GHG, $\delta^{13}CO_2$, $\delta^{13}CH_4$, and δDCH_4, were analyzed at the Biogeochemistry Facility School of Earth and Ocean Sciences (BF-SEOS, University of Victoria). Ebullition gas samples were analyzed for $\delta^{13}CH_4$ by introducing the gas onto a GSQ PLOT column (0.32 mm ID, 30 m) using a Valco 6-port valve and sample loop. After chromatographic separation, the CH_4 passes through an oxidation oven (1030 °C), a Nafion water trap, and open-split interface to a Continuous Flow-Isotope Ratio Mass Spectrometer (CF-IRMS). The $\delta^{13}CO_2$ was measured similarly by CF-IRMS, but bypassing the combustion oven. Precision for the $\delta^{13}CH_4$ and $\delta^{13}CO_2$ analyses was ± 0.2‰, relative to Vienna PeeDee Belemnite (VPDB). Hydrogen isotope ratios of CH_4 (δDCH_4) were measured by a TC/EA pyrolysis unit (1450 °C) interfaced to a CF-IRMS. Precision for the δDCH_4 analyses was ± 3‰, relative to Vienna Standard Mean Ocean Water (VSMOW). Carbon and hydrogen isotope ratios are expressed using standard delta (δ) notation as described by deviations from a standard such that:

$$\delta_{sample}\ ‰ = [(R_{sample} / R_{standard}) - 1] \times 1000$$

where R is the $^{13}C/^{12}C$ or $^{2}H/^{1}H$ ratio in the sample or standard. For isotope calibration, methane carbon and hydrogen standards from Isometric Instruments were used. These are traceable back to VPDB for carbon isotope ratios and VSMOW for hydrogen isotope ratios.

4 Results

4.1 Morpho-limnological properties of ponds and lakes

Ponds were generally shallow (~ 0.6-1.0 m and 1.0-1.5 m deep for polygonal and trough ponds, respectively) and thus froze to the bottom during winter, whereas lakes were more variable in

depth depending on their origin and at least a portion of them did not freeze to the bottom in winter. The thermokarst lake was a few meters deep (< 5 m), while the kettle lake was deeper (< 12 m). Polygonal ponds, including different developmental stages and coalesced ponds, generally had flat bottoms covered by cyanobacterial mats (up to 5 cm thick), and stable (non eroding) shores (Fig. 2b,c). Their surface area varied substantially (from 21 to 3350 m^2) with a median of around 160 m^2. Trough ponds were elongated water channels (median width ~ 3 m; median length ~ 10 m), and their shores were either actively eroding with collapsing decimetric peat blocks (Fig. 2f), or stable and colonized by brown mosses (Fig. 2g). The thermokarst lake had sharp edges near the shore, a shallow and gently sloping lake bottom and a deeper central basin. The kettle lake had steeper slopes along its margins, and showed a deep section that was not in the center of the lake (Bouchard et al., 2015).

Ponds and lakes showed contrasting physicochemical conditions during the two sampling years (Table 1). Trough ponds generally had the highest concentrations of DOC, nutrients and ions, followed by polygonal ponds, whereas lakes showed the lowest values. Trough pond BYL27, where shore erosion was active during summer time, had near- or higher-than-average concentrations, whereas trough pond BYL24, with stable shores, showed lower-than average values. Pond DOC, nutrient and ion concentrations were substantially higher in 2014, a particularly dry year (total precipitations from January to June = 27.0 mm in 2014, compared to 50.7 mm in average; Table B1 in Appendix), with resulting low pond water levels as observed in the field. When considering specific solute species separately, all of them except NO_3^- and SO_4^{2-} were statistically different ($p < 0.0001$) among aquatic system types in 2013. Contrastingly, in 2014 only DOC ($p < 0.0001$), total nitrogen (TN, $p < 0.001$) and soluble reactive phosphorus (SRP,

$p < 0.05$) showed significant differences, and only between lakes and ponds (i.e., not between polygonal and trough ponds). Among all the water chemical properties and regardless of the sampling year, DOC showed the highest statistical contrasts between the different types of water bodies.

Polygonal ponds (BYL30, BYL80) had a thermally homogenous and well-oxygenated water column in July, whereas trough ponds (BYL24, BYL27) were notably stratified (Figs. 3 and 4). Thermokarst lake BYL66 was relatively well mixed over most of the water column, except near the sediment-water interface where dissolved oxygen decreased rapidly. Kettle lake BYL36, deeper than the other sampled water bodies, showed a steep gradient between the warmer, well-oxygenated epilimnion and the much colder, anoxic hypolimnion. The thermal profiles on Fig. 3 are representative of the conditions generally prevailing from July to mid August in each type of water bodies.

4.2 Age and concentration of greenhouse gases released through ebullition

Radiocarbon age ($\Delta^{14}C$ signature) and concentration of GHG (CO_2 and CH_4) emitted through ebullition showed strikingly different trends between the various types of aquatic systems (Fig. 5). Polygonal and trough ponds produced modern CH_4 and modern to a few hundred years old (< 550 yr BP) CO_2, whereas lakes generally released older GHG, ranging from 510 to 1425 yr BP for CO_2 and from 125 to 3405 yr BP for CH_4 (Table 2). Moreover, samples from lake edges had younger and less concentrated CH_4 than those coming from lake central area. No such trend was observed for CO_2 in lakes. Considering all ponds and lakes as a whole, CH_4 was generally one to two orders of magnitude more concentrated than CO_2 in emitted bubbles in July,

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4.3 Dissolved and ebullition fluxes of greenhouse gases

Polygonal ponds were generally CO_2 sinks, but they were CH_4 sources with a relatively broad range of saturation levels (~ 0 - 2.4 µM) (Fig. 6). Lakes were near the equilibrium with the atmosphere (all samples clustered near 0 for both gases), being small sinks or sources of CO_2, and small sources of CH_4. Trough ponds were in general supersaturated in both gases, especially when their margins were actively eroding (highest GHG saturation values) (Fig. C1). Trough ponds showed the highest diffusive flux, especially of CO_2 (65.5 mmol m^{-2} d^{-1}; Table 3) with a median diffusive CO_2 flux (21.8 mmol m^{-2} d^{-1}) more than 12 times higher than the median value of all sampled water bodies (1.7 mmol m^{-2} d^{-1}). Polygonal ponds, on the other hand, showed the highest ebullition flux for both CO_2 (16.3 mmol m^{-2} d^{-1}) and CH_4 (534.5 mmol m^{-2} d^{-1}), with a median ebullition CH_4 flux that, although relatively low (~ 1.0 mmol m^{-2} d^{-1}), was ~ 5 times higher than the median value for all ponds and lakes (~ 0.2 mmol m^{-2} d^{-1}). Lakes generally showed the lowest fluxes (both diffusion and ebullition). Globally, diffusion appeared as the dominant mechanism for CO_2 emission, whereas CH_4 was mainly emitted through ebullition. Statistical tests ran on the GHG data showed that trough ponds (BYL24, BYL27) were significantly different ($p < 0.001$) from the other two types of water bodies (polygonal ponds and lakes), but also from each other. Furthermore, dissolved CO_2 and CH_4 fluxes were significantly correlated ($p < 0.006$) with CDOM ($r = 0.79$ and 0.78, respectively; $N = 22$; a_{320} only available in 2013), but only CH_4 fluxes were correlated ($p < 0.003$) with DOC ($R = 0.61$, $N = 28$; data available in both years).

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4.4 Carbon and hydrogen stable isotope ratios in ebullition gas samples

The stable isotope ratios of methane ($\delta^{13}CH_4$, δDCH_4) and carbon dioxide ($\delta^{13}CO_2$) were measured on 18 ebullition samples collected in 2013 and 2014 (Table 2; Figs. 7 and 8). The $\delta^{13}CH_4$ average values were -60.5 ‰ and ranged from -52.1 ‰ to the most ^{13}C-depleted value of -67.6 ‰, both from polygonal ponds. The δDCH_4 values, which averaged -376.80 ‰, were relatively ^{2}H-depleted for naturally occurring methane. The δDCH_4 with the most ^{2}H-enriched value came from the thermokarst lake sample collected at its center (-319.56 ‰; BYL66; Fig. 7). In contrast, the δDCH_4 values from trough ponds (BYL24 and BYL27) were consistently and extremely ^{2}H-depleted, with values from -397.7 ‰ to a very low value of -448.1 ‰. There was no apparent correspondence between the methane concentration and $\delta^{13}CH_4$ or δDCH_4. The CO_2 contents of ebullition samples were sometimes insufficient for carbon isotope measurements. For those with more CO_2, the average $\delta^{13}CO_2$ was -14.3 ‰ and varied from +0.3 (polygonal pond BYL80) to -21.8 ‰ (trough pond BYL24). There was also no apparent correspondence between the CO_2 concentration and $\delta^{13}CO_2$. However, it is worth noting that the sample with the most ^{13}C-enriched CO_2 also corresponded to the one with the most ^{13}C-depleted CH_4 (polygonal pond BYL80; Fig. 8).

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5 Discussion

5.1 The strong heterogeneity in greenhouse gas age and concentration

We observed large variability in the age, composition and emission rate of GHG released by the studied aquatic systems. The GHG escaping through ebullition ranged from modern to a few centuries old for polygonal and trough ponds, and from a few centuries to a few millennia old

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for lakes (Fig. 5). We found that trough ponds emitted slightly but significantly older CH_4 than polygonal ponds ($\Delta^{14}C$ = 10 ± 18 ‰ vs. 43 ± 28 ‰, respectively; $p < 0.05$) such as observed earlier at the same site (Negandhi et al. 2013), although still classified as modern carbon, suggesting a small contribution of peat-derived carbon pool to microbial activity in trough ponds. Surprisingly, trough ponds did not emit millennium-old CH_4, at least in July, despite the fact that they were exposed to eroding peat from down to the base of the active layer in the surroundings (^{14}C dates ranging from ~ 2.2 to 2.5 k yr BP; Table 2) and even older peat strata up-thrusted along ice wedges by cryoturbation and now in contact with surface waters (Fortier and Allard, 2004). Eroding peat was likely leaching old carbon into the water column, but bottom sediment interstitial water, where CH_4 is mostly produced, did not predominantly emit carbon of this age. Permafrost disturbance was indeed shown to deliver millennia-old particulate organic carbon and DOC to arctic streams and rivers (Lamoureux and Lafrenière, 2014; Guo et al., 2007; Vonk et al., 2013), acting as a significant degradable source of bioavailable carbon in Arctic freshwaters (Mann et al., 2015). We speculate that microbes were preferably using young carbon, putatively more labile and more abundant at this time of the year, and may use older carbon stocks later when primary producers are less active. If the CH_4 released from trough ponds is indeed older during the autumn and spring, this could represent a positive climate feedback, but our results now indicate a limited role.

On the other hand, CH_4 ebullition samples collected from lakes provided older dates, up to nearly 3500 yr BP (thermokarst lake BYL66), which is very close to the maximum known age of the permafrost peat layers in the valley (3670 ± 110 yr BP; Fortier and Allard, 2004). It may suggest that permafrost thaw underneath this lake have proceeded through the organic layers

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at this site, which could result in decreased emissions in the future after the microbial exhaustion of the labile fraction of the organic matter pool (Walter et al., 2007). However the timing of this reduction is unknown. We observed a spatial gradient in the age and concentration of CH_4 in bubbles emitted from the thermokarst lake, with younger and less concentrated CH_4 from the lake edge (~3%), and older and more concentrated CH_4 from the center (up to 57%). The development of a talik (unfrozen soil under lake) explains the mobilization of deeper and older CH_4 at the lake center where water remains unfrozen under the ice cover in winter (maximum lake depth > 4 m, ice cover thickness ~ 2 m). Methane emitted from a given location would thus be composed of a mixture of young CH_4 from the edge with older CH_4 from the center (Fig. D1). To our knowledge, the only other studies of thermokarst lakes presenting ^{14}C dates on GHG are in *yedoma* deposits (Alaska, Siberia), which have very different ground ice, sediment and organic carbon contents, and chronostratigraphic history. For these lakes, the release of very old (> 40 k yr BP) and highly concentrated (up to 90%) CH_4 from deep unfrozen lake sediments has been found (Walter et al., 2008). However, this study also reported younger ages for ebullition samples emitted from different parts of the lakes, and generally younger towards the lake center (background ebullition). At our study site, even though older GHG were emitted from lakes compared to ponds, ebullition fluxes remained low during the study period (July). Walter-Anthony and Anthony (2013) concluded that the classic randomized bubble-trap method for estimating mean lake ebullition is highly median-biased toward underestimation of fluxes, and this was possibly also occurring for our data set, although no systematic GHG point source studies have been conducted so far at our study site.

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We also observed strong differences in dissolved GHG flux depending on pond and lake types (Fig. 6; Table 3): polygonal ponds were CO_2 sinks but CH_4 sources while trough ponds were significant sources of both GHG, as previously reported in the valley (Laurion et al., 2010; Negandhi et al., 2013), and lakes were small sources of GHG. This pattern can be explained by the morpho-limnological properties of the water bodies. Polygonal ponds had stabilized shores (no apparent slumping) and more transparent waters compared to other systems, as shown by their lower CDOM content (Laurion et al., 2010). Moreover, they had flat and shallow bottoms covered by abundant cyanobacterial mats actively photosynthesizing and acting as a relatively efficient CO_2 sink (flux reaching -11.8 mmol m^{-2} d^{-1}). This is however one order of magnitude lower than the net ecosystem CO_2 uptake measured over the summer from a wet polygonal tundra site in Siberia (flux reaching -104.7 mmol m^{-2} d^{-1}; Kutzbach et al., 2007). Bottom sediments of the studied polygonal ponds were also colonized by methanotrophic bacteria (Negandhi et al., 2014), which can be a significant control mechanism on CH_4 emissions such as shown in polygonal ponds of the Lena region (Liebner et al., 2011).

Lakes were larger and deeper, thus they were exposed to wind-induced mixing of their epilimnetic waters promoting venting of the GHG from this layer. When the water column is seasonally stratified (like in BYL36), the hypolimnion likely stores a large fraction of the GHG produced by the lake until the autumnal overturn period (Bastviken et al., 2004), allowing more space and time for the oxidation of dissolved CH_4, and for the dissolution of a fraction of ebullition CH_4 (Bastviken et al., 2008). Therefore, it is possible that higher flux of old carbon would be observed later in the season. To fully account GHG emissions from lakes and compare

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them to other aquatic systems, summer and winter storage fluxes will need to be estimated (Boereboom et al., 2012; Langer et al., 2015; Walter Anthony et al., 2010; Wik et al., 2011).

Trough ponds presented the highest combined (CO_2 + CH_4; diffusion + ebullition) GHG fluxes at the time of sampling (Fig. 9). Considering a global warming potential (GWP) of 34 for CH_4 on a 100-year horizon (Myhre et al., 2013), trough ponds presented the highest net carbon efflux (1.5 g CO_2-equivalent m^{-2} d^{-1}, compared to 0.7 and 0.2 g C m^{-2} d^{-1} respectively for polygon ponds and lakes). Despite their shallow depths, trough ponds were strongly stratified with oxygen-depleted and cold bottom waters. The bottom temperature in these ponds was indeed near 0°C (Fig. 3) because this layer of water is lying just above the melting ice wedge (as part of the active layer), does not mix with surface waters, and is cooled down through sensible heat transfer. Moreover, trough ponds were not colonized by photosynthesizing (CO_2 sink) and methanotrophic (CH_4 sink) bacteria such as in polygonal ponds (Negandhi et al., 2014). Stronger water column hypoxia generated anoxia more rapidly in the sediments, and the organic material inputs caused by active erosion likely led to higher CH_4 production, although the young carbon signature of emitted CH_4 is still puzzling (see below). Meanwhile, the eroding conditions and reduced light availability (higher CDOM, TP and turbidity; Table 1) in trough ponds favored net heterotrophy and net CO_2 emissions, such as found in subarctic thermokarst lakes (Roiha et al., 2015). Similar to polygonal ponds, the shallow depth of trough ponds reduces the chances for dissolution of CH_4 bubbles into the water column and its subsequent oxidation before reaching the atmosphere. Moreover, the thermal structure of trough ponds (low transparency, microtopography) can impede mixing for several weeks (Fig. 4), thus favoring GHG summer storage in bottom waters, and likely generating stronger diffusive flux later at the autumnal

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overturn period. Thermal structure might become even stronger in years of low precipitations such as in 2014, when concentrations of solutes (DOC, ions) increase through evaporation, intensifying density gradients thus GHG storage. Diffusive CH_4 fluxes were indeed statistically higher ($p < 0.01$) in 2014 compared to 2013, although no such trend was observed for CO_2. The highest GHG saturation levels observed over the sampling period were measured in a trough pond the day following a major erosion event (peat block collapsing in pond BYL27; Fig. C1). This might result from the disturbance of the thermal structure and transfer of stored GHG to the surface, or from the causal effect of a new input of organic matter to microbial activity. Active shore erosion around tundra ponds, potentially increasing CH_4 production by 2 to 3 orders of magnitude, has been reported from similar systems in Siberia (Langer et al., 2015), suggesting a direct impact of permafrost slumping on GHG emissions. The effect of erosion events on GHG flux must be further evaluated as other factors, such as fluctuating wind and air temperature, can also influence mixing and surface GHG concentrations (Tedford et al., 2014). Interestingly, we also observed substantial differences in GHG concentrations among trough ponds, some presenting much lower values. Trough ponds such as BYL24 (Fig. 2g) had relatively stable (non eroding) shores, and were colonized by abundant vegetation dominated by brown mosses. Methane oxidation by bacteria associated with submerged brown mosses has been reported in Siberian ponds, contributing to smaller CH_4 concentrations in these ecosystems (Liebner et al., 2011). Therefore, there might be cases where the methanotrophic community is also efficient in limiting CH_4 emissions from trough ponds (Negandhi et al., 2014).

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5.2 Production pathways of CO_2 and CH_4

We obtained different radiocarbon ages for CO_2 and CH_4 within the same ebullition samples, as collected from funnels placed at the water surface (Table 2, Fig. 5), suggesting that GHG production was derived by different carbon sources. This divergence in carbon age was even more pronounced for the lakes, where it could reach almost 3000 years. The presence of unfrozen sediment layers (talik) underneath the lakes would explain the older bubbling CH_4 emitted from deeper/older sediments exposed to microbial degradation, such as found in thermokarst lakes of Siberia and Alaska (Walter et al., 2007). Younger CO_2 could then be explained by a larger contribution of younger and shallower surface sediments to bacterial production and respiration. It could also result from lateral inputs of CO_2 produced within younger organic material or from exchanges with atmospheric CO_2.

On the other hand, century-old CO_2 collected from ponds in parallel to modern CH_4 is more difficult to explain. As stated above, emission of young CH_4 suggests the preferential use of modern carbon by methanogens, and also a dominance of background ebullition mode (from surface sediments) in thaw ponds. Meanwhile, emission of older CO_2 could be related to anaerobic CO_2 production in water-saturated and reductive soils and its subsequent lateral transport, as observed in a flooded tundra site in Alaska (Zona et al., 2012). Characterizing organic matter properties and oxidation *versus* reduction (redox) potential of pond and lake sediments at our study sites are required to confirm if such a mechanism can contribute to modern CH_4 emissions from surface layers and, at the same time, older CO_2 emissions from deeper layers. Moreover, a quantification of lateral fluxes of carbon within the active layer (groundwater and streams), an important yet rarely mentioned process driven by the coupling

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between carbon and water cycles (Vonk and Gustafsson, 2013; Paytan et al., 2015), could help to better understand these results.

Notwithstanding the above-mentioned differences, the concentrations of CO_2 and CH_4 emitted through ebullition also need to be taken into account when evaluating the climate feedback potential of these emissions. Even though the age of CO_2 could reach several centuries (> 1000 yr BP for one sample; Fig. 5), it was one to two orders of magnitude less concentrated in the emitted bubbles than CH_4. Hence, such emissions have a much lower potential to generate a positive feedback effect, at least during the ice-free season and under current climate conditions. Similar observations were reported from Siberian lakes, despite notably different geomorphological, geocryological and limnological conditions (Walter et al., 2007).

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Methanogenesis in cold wetland systems typically proceeds via the anaerobic fermentation pathways of acetoclastic methanogenesis (AM) and/or hydrogenotrophic carbonate reduction methanogenesis (HM) (e.g., Kotsyurbenko et al., 2004; Alstad and Whiticar, 2011). AM utilizes the transfer of a CH_3^- group from preformed organic substrates (i.e., acetate, methanol, methylated substrates, etc.), whereas HM utilizes H_2 and CO_2. Numerous studies have demonstrated the ability of using methane C and H isotope signatures to discriminate AM from HM pathways, and to characterize secondarily altered methane (oxidation, mixing, etc.). Polygonal ponds and lakes had combined methane C and H stable isotope signatures that were typical for methanogenesis dominated by AM, as strongly illustrated in the plot of $\delta^{13}CH_4$ versus δDCH_4 (Fig. 7). Trough ponds shared similar $\delta^{13}CH_4$ values with the other water bodies, but had substantially more ^2H-depleted values (δDCH_4 from -398 ‰ to -448 ‰; Table 2, Fig. 7). These values are among the most ^2H-depleted values known for naturally occurring methane (e.g.,

Whiticar, 1999). Although there was some variation between sites, the isotope signatures designate that all CH_4 emitted by ebullition in July is produced by AM, consistent with an earlier study at the same site (Negandhi et al., 2013). There is no indication of HM, which has a very different isotope signature, although the signature of samples collected from the center of lakes tend to lie towards the HM region, suggesting that a small proportion of the CH_4 produced could be through this pathway. This finding of AM dominance is consistent with ombrotrophic bogs with higher pH (ranging from ~ 6.7 to 10.0 in 2014) compared with more acidic minerotrophic wetlands, which can be HM dominated (e.g., Bowes and Hornibrook, 2006; Prater et al., 2007). The dominance of AM is likely related to the carbon precursors; our sites may have more labile organic material present (e.g., organic acids) supporting acetoclastic methanogenesis and recently made available to methanogens. As this labile carbon pool is exhausted, the methanogenic pathway shifts from acetoclastic to more recalcitrant compounds and hence hydrogenotrophic methanogenesis (e.g., Alstad and Whiticar, 2011). It is therefore possible that other periods of the year would show a stronger HM signature, which would also be consistent with the presence of a large fraction of microbes able to perform HM in thaw ponds from this site (Negandhi et al. 2013).

Previous work in this valley indicated a significant relationship between water oxygen concentration and dissolved CH_4 oxidation level (Negandhi et al., 2013). This work also showed evidence that diffusive CH_4 was more susceptible to oxidation in polygonal ponds where a methanotrophic community was favored (Negandhi et al., 2014). This conclusion was supported by the strong shift in $\delta^{13}CH_4$ and δDCH_4 to the heavier isotopes, as expected (Whiticar et al., 1986). In the present study, there was no evidence of methane oxidation in any of the collected

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ebullition samples (Fig. 8), indicating that the conditions did not favor oxidation at the production site (likely in anoxic sediment but also potentially in the water column; Grossart et al., 2011), and that the exchange with a pool of oxidized methane during the transport of bubbles to surface waters was undetectable, possibly linked to the short residence time. This was expected for shallow waters where bubbles can rapidly escape, but it was also the case in larger and deeper stratified lakes such as BYL36.

5.3 Bylot ponds and lakes within the circumpolar North

The general topography and geology of the southwest plain of Bylot, together with the distinct local conditions of the Qarlikturvik valley (e.g., glacier and outwash plain activity, valley orientation in relation to dominant winds, snow cover depth and density), have contributed to the development over thousands of years of what is arguably one of the richest ecosystems in the region. However, taken separately, most of the landscape features in the valley (e.g., tundra polygons, ice-wedges, thermokarst ponds and lakes) are widespread across the Arctic (e.g., Walter Anthony et al., 2010; Abnizova et al., 2012; Langer et al., 2015). When compared with flux values reported in the literature, our results, representing a snapshot of mid-summer conditions, generally appear in the range of what has been observed in other ponds and lakes from northern regions (Table 4). For example, we measured total CO_2 fluxes (diffusion + ebullition) of up to ~ 0.8 g C m^{-2} d^{-1}, which is in the range of those reported from Alaska (0.7 – 2.3 g C m^{-2} d^{-1}; Kling et al., 1992; Sepulveda-Jauregui et al., 2014), Siberia (0.02 – 1.1 g C m^{-2} d^{-1}; Abnizova et al. 2012; Blodau et al., 2008), and Scandinavia (0.9 – 1.6 g C m^{-2} d^{-1}; Huttunen et al., 2003; Kankaala et al., 2013). Methane fluxes (diffusion + ebullition) at our study site varied substantially (0.0005 – 6.4 g C m^{-2} d^{-1}), but could reach values one order of magnitude higher

than those from lakes in Alaska (0.01 – 0.5 g C m^{-2} d^{-1}; Kling et al., 1992; Sepulveda-Jauregui et al., 2014; Walter Anthony and Anthony, 2013) and Scandinavia (0.01 – 0.1 g C m^{-2} d^{-1}; Bastviken et al., 2004; Huttunen et al., 2003; Kankaala et al., 2013). However, median values for polygonal and trough ponds (~ 0.02 and 0.01 g C m^{-2} d^{-1}, respectively) were more similar to published ranges. Yet, these fluxes were lower than those reported from Siberian thermokarst lakes in *yedoma* deposits (nearly 20 g C m^{-2} d^{-1}; Walter Anthony et al., 2010), which however include discrete ebullition seeps and hotspots that were not observed in our study, and most likely do not exist in the case of ponds.

6 Conclusions

Aquatic systems are widespread across permafrost landscapes and play a crucial role in large-scale biogeochemical cycles. Yet, there is still much uncertainty about whether or not the Arctic can globally be considered a carbon source or sink, and how this will change in the coming decades. One element of such uncertainty is the highly heterogeneous distribution of ponds and lakes at the local scale and their different geomorphological and limnological properties, which influence their biogeochemistry and result in highly variable fluxes from these waters, especially for trough ponds. Our study demonstrates that local geomorphology and shoreline erosion around permafrost ponds and lakes can have a strong impact on their GHG concentrations and fluxes. We also report substantially different GHG ages among ponds and lakes of contrasting sizes and depths, and unexpectedly the emission of mainly modern CH_4 from trough ponds despite their exposure to a stock of eroding old carbon. Such results underscore the importance of the combined effects of geomorphology (talik development level, chronostratigraphy),

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limnology (organic matter concentration, CH_4 production and storage in anoxic/hypoxic bottom waters) and hydrology (lateral runoff inputs of organic material or GHG) on GHG emissions by permafrost thaw ponds and lakes. Interestingly, the significant correlation between GHG flux and DOM once more suggests the key role of this limnological characteristic, and calls for a deeper investigation as it could be used as a proxy for upscaling and modeling. The dominance of acetoclastic methanogenesis indicates that the system is presently rich in labile precursor substrates (e.g., acetate, formate, methylated substrates). However, the oldest CH_4 ages (~ 3.5 k yr BP) obtained from a thermokarst lake corresponded to the maximal age of the frozen organic (peat) layers in the valley, suggesting that permafrost thaw might have (or will soon have) proceeded through the organic substrate at this site. The local differences in surface areas, emissions rates, carbon age and sources reported in this study need to be further characterized in other regions of the Arctic in order to properly upscale and model GHG emissions and carbon-climate feedbacks across permafrost lake-rich landscapes.

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Appendix file captions

Figure A1. Picture of the homemade funnels deployed in ponds and lakes (photo taken in July 2014 just after their removal).

Table B1. Temperature and total precipitation data for the six months preceding the sampling period in July 2013 and 2014. The climate normal (1981-2010) is also indicated (Environment Canada, 2015).

Figure C1. Picture of eroding shores (slumping peat) along trough pond BYL27 (photo taken in July 2014). The sampling funnel syringe can be seen just above the water surface.

Figure D1. Keeling plot of lake ebullition CH_4 sampled in 2014, showing a mixing of millennium-old and highly concentrated with near-modern and less concentrated gas. Concentration (x-axis) is expressed as 1000/partial pressure (in ppmv, parts per million volumetric), whereas radiocarbon age is expressed as the normalized radiocarbon activity ($\Delta^{14}C$, in ‰; left y-axis) and in thousands of years before present (k yr BP; right y-axis).

Author contribution

F. B., I. L. and V. P. designed the experiments, and F. B. and V. P. performed them. I. L., D. F., X. X. and M. J. W. contributed materials, instruments and analyses. F. B., I. L., V. P. and D. F. analyzed the data. F. B. prepared the manuscript with contributions from all co-authors.

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Tables

Table 1. Limnological properties of ponds and lakes sampled in July 2013 and July 2014, including sampling depth, dissolved organic carbon (DOC), absorption coefficient of dissolved organic matter at 320 nm (a_{320}), total phosphorus (TP), soluble reactive phosphorus (SRP), total nitrogen (TN), and selected major ions (NO_3, SO_4, Fe). POL = polygonal pond; IWT = ice wedge trough pond; LAK = lake.

Site	Type	Depth m	DOC mg L^{-1}	a_{320} m^{-1}	TP µg L^{-1}	SRP µg L^{-1}	TN mg L^{-1}	NO_3 mg L^{-1}	SO_4 mg L^{-1}	Fe mg L^{-1}
2013										
BYL30	POL	surf	8.7	17.8	14.8	N/A	0.49	0.42	1.3	0.470
BYL80	POL	surf	5.6	9.0	22.9	N/A	0.42	0.09	1.3	0.250
Average POL (n = 9)			*6.7*	*12.7*	*17.6*		*0.47*	*0.17*	*1.3*	*0.282*
BYL24	IWT	surf	6.6	27.0	16.1	N/A	0.29	0.37	4.3	0.270
BYL27	IWT	surf	10.1	42.0	29.0	N/A	0.58	0.07	6.2	1.400
Average IWT (n = 12)			*10.0*	*38.0*	*27.8*		*0.63*	*0.19*	*6.7*	*1.014*
BYL66	LAK	surf	4.2	16.4	20.7	N/A	0.27	0.13	2.9	0.460
BYL36*	LAK	surf	3.9	5.8	16.2	0.33	0.22	0.10	1.7	0.067
2014										
BYL30	POL	surf	12.2	N/A	8.5	1.31	1.32	0.25	2.6	0.648
BYL80	POL	surf	10.6	N/A	22.7	1.75	1.25	< 0.2	1.6	0.266
BYL24	IWT	surf	8.8	N/A	23.7	1.28	1.02	0.30	1.3	1.549
		0.9	9.3	N/A	21.5	1.95	1.16	0.21	1.7	2.169
BYL27	IWT	surf	12.1	N/A	27.4	1.56	1.22	0.29	2.7	0.487
		1.3	14.3	N/A	54.8	1.41	1.70	0.25	2.4	2.979
BYL66	LAK	surf	4.3	N/A	9.8	< 0.5	0.49	< 0.2	2.4	2.949
		2.0	4.2	N/A	10.6	0.74	0.44	< 0.2	2.5	0.627
		4.5	4.1	N/A	28.0	0.75	0.56	0.27	2.6	0.507
BYL36	LAK	surf	4.3	N/A	6.7	1.13	0.45	0.27	2.2	0.023
		2.0	4.2	N/A	-	0.91	0.46	< 0.2	2.2	0.027
		10.0	4.2	N/A	41.2	1.29	0.57	< 0.2	2.3	0.039

* 2011 data

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Table 2. Greenhouse gas radiocarbon and stable isotope results for the six priority ponds and lakes sampled during two consecutive years (2013 and 2014). Active layer samples collected in 2013 near two trough ponds are also included. POL = polygonal pond; IWT = ice wedge trough pond; LAK = lake; UAL = upper active layer (0-5 cm); LAL = lower active layer (50-60 cm); Fm = fraction modern.

Year	Site	Type	Gaseous CO_2 ppmv	Gaseous CH_4 ppmv	Fm CO_2	Fm CH_4	$\Delta^{14}C$ CO_2 ‰	$\Delta^{14}C$ CH_4 ‰	^{14}C age CO_2 BP	^{14}C age CH_4 BP	$\delta^{13}C$ CO_2 vs VPDB	$\delta^{13}C$ CH_4 vs VPDB	δD CH_4 vs VSMOW
2013	BYL30	POL	2580	324066	1.022	1.060	14	52	>Modern	>Modern	-10.6	-63.3	-378
2013	BYL80	POL	29124	784232	1.001	1.027	-7	20	0	>Modern	0.3	-67.6	-347
2013	BYL80	POL	735	234455	0.987	1.006	-21	-1	105	>Modern	-13.7	-65.7	-356
2013	BYL24	IWT	5783	115383	0.987	1.031	-20	23	105	>Modern	-21.8	-61.5	-398
2013	BYL27	IWT	1542	77007	0.934	1.010	-73	2	550	>Modern	-17.4	-60.1	-399
2013	BYL66	LAK	5269	324781	0.837	0.788	-169	-218	1425	1910	-8.4	-63.2	-392
2014	BYL30	POL	1607	18406	1.021	1.073	13	64	Modern	>Modern	-18.1	-57.7	-352
2014	BYL30	POL	2857	15724	N/A	N/A	N/A	N/A	N/A	N/A	-16.2	-52.1	-384
2014	BYL80	POL	< 50	174762	1.010	1.067	3	58	Modern	Modern	N/A	-53.9	-346
2014	BYL80	POL	< 50	232178	0.970	1.076	-38	68	245	>Modern	N/A	-56.5	-372
2014	BYL24	IWT	< 50	330145	1.049	1.043	41	35	Modern	Modern	N/A	-63.0	-426
2014	BYL27	IWT	32383	291005	0.996	1.000	-12	-8	35	5	-16.1	-59.3	-410
2014	BYL27	IWT	< 50	251821	1.009	1.006	1	-2	Modern	Modern	N/A	-59.9	-448
2014	BYL66	LAK	1774	31124	0.935	0.824	-72	-182	540	1555	-17.9	-59.9	-387
2014	BYL66	LAK	< 50	436334	0.909	0.680	-98	-326	765	3105	N/A	-59.2	-344
2014	BYL66	LAK	< 50	330116	0.939	0.655	-69	-350	510	3405	N/A	-57.4	-320
2014	BYL36	LAK	< 50	25187	0.886	0.984	-121	-23	970	125	N/A	-63.1	-379
2014	BYL36	LAK	3845	1761	N/A	N/A	N/A	N/A	N/A	N/A	-17.5	-65.5	-345
2013	BYL27 (UAL)	IWT	N/A	N/A	1.062		62		>Modern		-28.9		N/A
2013	BYL27 (LAL)	IWT	N/A	N/A	0.730		-270		2535		-26.3		N/A
2013	BYL28 (UAL)	IWT	N/A	N/A	1.000		-1		5		N/A		N/A
2013	BYL28 (LAL)	IWT	N/A	N/A	0.759		-241		2210		N/A		N/A

Table 3. Diffusive and ebullition fluxes of CO_2 and CH_4 for the six priority ponds and lakes sampled during two consecutive years (2013 and 2014). POL = polygonal pond; IWT = ice wedge trough pond; LAK = lake; Min = minimum; Med = median; Max = maximum.

			Diffusive fluxes (mmol m^{-2} d^{-1})							Ebullition fluxes (mmol m^{-2} d^{-1})						
			CO_2			CH_4				CO_2			CH_4			
Site	**Type**	**N**	**Min**	**Med**	**Max**	**Min**	**Med**	**Max**	**N**	**Min**	**Med**	**Max**	**Min**	**Med**	**Max**	
BYL30	POL	12	-8.11	-1.04	5.73	0.19	1.07	1.46	12	0.00	0.01	0.26	0.01	0.89	26.57	
BYL80	POL	32	-11.78	-3.14	45.44	0.03	0.53	1.14	9	0.00	0.00	16.32	0.11	0.99	534.54	
BYL24	IWT	18	-5.44	13.27	26.30	0.05	0.17	1.51	8	0.00	0.00	0.02	0.01	0.06	0.29	
BYL27	IWT	26	15.96	25.86	65.50	0.34	1.03	5.82	11	0.00	0.00	5.18	0.00	4.55	32.93	
BYL66	LAK	12	-7.05	1.62	5.13	0.06	0.09	0.27	11	0.00	0.00	0.00	0.00	0.15	5.08	
BYL36	LAK	6	-0.75	1.20	1.37	0.06	0.08	1.13	2	0.00	0.00	0.00	0.00	0.02	0.03	
All water bodies		***106***	***-11.78***	***1.74***	***65.50***	***0.03***	***0.54***	***5.82***	***53***	***0.00***	***0.00***	***16.32***	***0.00***	***0.18***	***534.54***	

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Table 4. Greenhouse gas fluxes of CO_2 and CH_4 from high-latitude sites across the circum-Arctic. D = diffusion; E = ebullition.

				CO_2 mg C m^{-2} d^{-1}		CH_4 mg C m^{-2} d^{-1}		
Reference	**Region**	**Type**	**Mode**	**Min**	**Max**	**Min**	**Max**	**Notes**
Bouchard et al. (this study)	NE Canada	Polygon ponds	D + E	-141.4	741.1	0.5	6432.0	July measurements
		Troughs	D + E	-65.3	848.1	2.6	465.1	
		Lakes	D + E	-84.6	61.6	0.7	74.5	
Laurion et al. 2010	NE Canada	Subarctic ponds	D	27.6	746.4	0.4	5.4	July measurements
		Arctic ponds	D	-246.0	1372.8	0.4	67.4	
		Arctic lakes	D	-63.6	70.8	0.1	0.4	
Buell 2014	NW Canada	Ponds	D + E	-3.5	120.0			Headspace, chamber and flux tower methods
Kling et al. 1992	Alaska	Lakes and rivers	D	-66.0	717.6	1.0	12.2	25 lakes + 4 rivers
Walter Anthony and Anthony 2013	Alaska	Thermokarst lakes	E			0.6	155.7	Strongest emissions = submerged polygons (lake shore)
Sepulveda-Jauregui et al. 2014	Alaska	Lakes	D + E	51.9	2276.9	3.0	455.4	Annual fluxes (ice-free period = 180 days)
Walter Anthony et al. 2010	Alaska, Siberia	Thermokarst lakes	E			0.0	18716.8	Background + seep ebullition
Abnizova et al. 2012	Siberia	Whole landscape	D + E	200.0	1100.0			September measurements, flux tower
Blodau et al. 2008	Siberia	Ponds	D	Average = 20.5		82.3	127.2	
Kankaala et al. 2013	Finland	Lakes	D	140.0	1586.7	0.2	26.7	Annual fluxes (ice-free period = 180 days)
Huttunen et al. 2003	Finland	Lakes and reservoirs	D + E	-21.6	876.0	0.8	99.6	CO_2 = diffusion only
Bastviken et al. 2004	Sweden	Lakes	D			0.6	11.0	Annual fluxes (ice-free period = 180 days)

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Figure captions

Figure 1. Location of the study site in the continuous permafrost zone of the Eastern Canadian Arctic (a), north of Baffin Island (b), within one of the several glacier valleys of Bylot Island, Nunavut (c). The studied valley contains numerous aquatic systems of different sizes (d). Source of the permafrost map (a): Brown et al. (1998). Satellite photo (c): Terra-MODIS, 22 July 2012.

Figure 2. Location of the sampled water bodies (a), including polygonal ponds (b-c), kettle and thermokarst lakes (d-e, respectively) and trough ponds (f-g). Ponds and lakes are located within the limits of a peaty loess permafrost terrace, outlined with the dashed white line. Satellite photo (a): GeoEye-1, 18 July 2010.

Figure 3. Temperature (°C; upper x-axes) and dissolved oxygen (%; lower x-axes) profiles for polygonal ponds BYL30 (a) and BYL80 (b), trough ponds BYL24 (c) and BYL27 (d), and lakes BYL66 (e) and BYL36 (f). Some profiles (a-b-c) were taken in July 2013, whereas the others (d-e-f) were taken in July 2014. Note the different vertical scales (depth).

Figure 4. Water temperature at two depths (surface = 0 cm; mid-depth = 50 cm) in trough pond BYL27 over one year (27 June 2013 to 8 July 2014), showing extended stratification and rare mixing events (lower panels) during the summer.

Figure 5. Concentration and age of ebullition GHG collected from ponds and lakes on Bylot Island, Nunavut. Gas concentration (x-axis) is expressed as partial pressure (in ppmv, parts per million volumetric) of CO_2 (open circles) and CH_4 (full circles). Radiocarbon age is expressed as the normalized radiocarbon activity ($\Delta^{14}C$, in ‰; left y-axis) corrected for isotopic fractionation and decay that took place between sampling and measurement dates, and in thousands of years before present (k yr BP; right y-axis).

Figure 6, Saturation levels of dissolved GHG in pond and lake water. Values are expressed as the departure from saturation (in µM) for CO_2 (x-axis) and CH_4 (y-axis). Values < 0 indicate a sink, whereas values > 0 indicate a source.

Figure 7. Carbon (δ^{13}) and hydrogen (δD) isotope composition of the methane emitted through ebullition by the sampled ponds and lakes, after Whiticar et al. (1986). AM = acetoclastic methanogenesis; HM = hydrogenotrophic methanogenesis; undiff. = undifferentiated lake sample location (edge *vs*. center).

Figure 8. Carbon isotope composition ($\delta^{13}C$) of CH_4 (x-axis) and CO_2 (y-axis) emitted by the sampled ponds and lakes. HM = hydrogenotrophic methanogenesis; AM = acetoclastic methanogenesis; MO = methane oxidation.

Figure 9. Schematic diagram of median fluxes of CO_2 and CH_4 from each type of water body in July. Note that dissolved and ebullition fluxes are combined (see Table 3 for details).

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Commentaire [1]: New figure.