bg-2016-79-RC1, 2016 Response to Reviewers Document

Authors note (August 2016): We appreciate the close attention that Anonymous Referee #1 provided on their review of this paper. Their suggestions, we believe, substantially improve this manuscript, especially the Discussion section. We have incorporated nearly all of the suggested edits that the Reviewer provided and discuss why we propose not to incorporate others. We would like to take this opportunity to thank Reviewer #1. Our comments are posted below Reviewer comments starting with a "--".

Anonymous Referee #1
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General comments on the overall quality of the paper

In this study, authors examine CO2 and CH4 exchange between four common freshwater systems located in the high Arctic watershed of Lake Hazen, Elsmere Island, Nunavut, Canada. This study aims to measure net exchange of CO2 and CH4 between atmosphere and common high Arctic freshwater ecosystems. In previous studies, authors investigated CO2 and CH4 net exchanges from terrestrial ecosystems (e.g. polar semi-desert, meadow wetlands, uplands) in the watershed of Lake Hazen. In this study, authors also aim to contextualize their new findings about freshwater systems in Lake Hazen watershed with both their previous results and literature. The scientific context and questions are well defined but specific issues about High Arctic landscapes could be clarified. What are the conditions and issues in High Arctic environments that differ from Low Arctic and Sub Arctic regions?

--***We have attempted to address this in a revised Introduction section (see below).***

The objectives are well exposed and correspond to data showed by authors. However, because of too general sentences and a lack of references, the answer of the second objective (contextualization) is not well developed and not accurate enough.

--***This has been addressed in a new section 4.2 in the Discussion (see below).***

The authors present interesting data about CO2 and CH4 concentrations and exchange with atmosphere in freshwater systems in a high Arctic watershed. Measurements have been performed during summers from 2005 to 2012 in four freshwater ecosystems: evaporative ponds, meltwater ponds, shoreline ponds and Lake Hazen shoreline. These four freshwater systems were identified using a hierarchical clustering analysis based on gas concentrations and biogeochemistry data. The main findings from this study are: Mean CO2 concentrations and atmospheric exchanges were statistically similar between freshwater systems. The three types of ponds were weak CO2 emitters. Shoreline ponds exhibited the highest dissolved CH4 concentrations and fluxes to the atmosphere. However, because shoreline ponds cover a small area, their contribution to overall CH4 emissions was weak. Other ponds and the Lake Hazen shoreline showed similar CH4 concentrations and fluxes, which were weak. The same authors evidenced in previous studies that polar semi-desert ecosystems were weak sink of atmospheric CO2 but they significantly consumed CH4. Alternatively, meadow wetlands were important sink of CO2 and weak emitter of CH4. Considering their cover surface, all freshwater systems did not significantly contribute to total net C exchange from Lake Hazen watershed. This study showed interesting results about the weak CO2 and CH4 emissions and uptake from freshwater systems in High Arctic region. These results are crucial to better constrain the assessment of future

carbon feedback from permafrost environments with climate change. Furthermore, high values of CH4 concentrations and fluxes in shoreline ponds are important considering likely evolution of both the water level and the biogeochemistry of Arctic lakes. From my point of view, the main result is the seasonal flooding of Lake Hazen that led to strong increase in CO2 and CH4 emissions from ponds bordering the lake while other ecosystems were weak CH4 emitter. The highlighted processes are interesting and important although more evidences of the impact of biogeochemistry change on CH4 emissions would be necessary.

--Yes, we agree that the flooding story is interesting and may warrant further investigation with more targeted studies. However, an important goal of this work was to weight the relative importance of different ecosystems within watershed-scale exchange of carbon GHGs. We found that shoreline ponds were insignificant contributors to regional GHG exchange because of their low abundances in the watershed. We suggest in the new Discussion section 4.2 that future changes in the regional climate will likely not considerably affect the net exchange of carbon GHGs from Shoreline systems. Therefore, we have not focused on particular results from the Shoreline systems relative to other ponds and lakes.

The second part of the discussion infers about the evolution of carbon exchange from freshwater systems in warmer and wetter conditions due to climate change. This section (paragraph 4.3) should be strongly modified. The current discussion section is too general and not adequately based on the findings from this study. There is a strong lack of references in this section (only four, an one auto-citation). Authors should develop a more specific and accurate discussion using more references. Despite the importance of data, authors are strongly recommended to do a major revision before acceptation by 1) better investigating the biogeochemical processes responsible of contrasted CO2 and CH4 concentrations and fluxes among ponds and 2) strongly improving the discussion.

--Both points have been addressed below.

<u>Scientific questions and issues</u>

- I would recommend changing the title that does not reflect the main findings

--We have altered the manuscript title to more appropriately focus on the approach and findings of our study: "Carbon dioxide and methane fluxes of freshwater systems in the rapidly changing high Arctic".

- The number of samples should be indicated. The standard deviation of the mean should be indicated.

--The number of samples issue has been addressed with the new Table 2, and revised Table 3 (formerly Table 2) which now includes means and standard deviations.

How are representative the different measurements considering the differences in quantity of samples? How evolved the number of samples during time from 2005 to 2012? How many samples per site did you use to build the dendrogram?

--In the methods section "Numerical analysis" we discuss the advantages of using both hierarchical cluster analysis and linear mixed models for unbalanced sampling programs. We

have now included a new Table 2 to show the number of samples taken for both greenhouse gases measurements and general chemistry, which were used in the cluster analysis.

- -Are the ponds permanent throughout the year? Do you consider these freshwater systems as ponds of small lakes?

--We have included the word "permanent" after "several" in the first sentence of the second paragraph of section 2.1. We have named our sites based on the type of water body they are (pond or lake).

- What is the geologic substrate and the soil nature in the watershed? It may help to discuss your interesting results.

--We have modified the second sentence of section 2.1 to include after "(6,901 km^2),", "composed of carbonate, evaporite and dolomite rock (Trettin, 1994) and crysolic soils."

- Place the section 'numerical analysis' currently located in the supplementary data in the main manuscript.

--We have now placed the "numerical analysis" section into the main manuscript at the end of the Methods section.

- Authors studied both spatial and temporal variability. The two perspectives are not clearly exposed. I would recommend separating results about spatial variability from temporal evolution of gas concentrations and fluxes. The robustness of the spatial variability should be better explained by improving Figure 2 and Table 2. The available samples/data and the significance of differences in biogeochemical composition should be added. Some temporal trends should be better illustrated and explained (Figure 3).

--This has been addressed below.

- This manuscript reproduces and repeats some results already presented in Emmerton et al. (2014). Results from previous studies should be removed from the abstract and from the result section.

--We have modified the results section (see below). We have also removed two sentences from the abstract which present terrestrial gas exchange data, and have added "and data from previous studies" after "When using ecosystem-cover classification mapping".

- Most of the figures should be modified in order to clarify the main information. Concentrations and fluxes bar plots should be separated; vertical scales should be changed. Some figures in supplementary data could be placed in the main manuscript such as Figure S2.

--See responses below. Figure S2 is solid support for our lake classification, but we feel it is not a significant part of our results or discussion, especially considering that we moved Figure 2 to the SI.

- Authors highlighted interesting biogeochemical processes, which could be better evidenced

--We have attempted to expand explanations of our CO2 and CH4 concentrations and fluxes using biogeochemical processes inferred by correlations in Table S4. However, we were cognizant of limiting these statements because of their speculative and correlative nature. We

have added references and interpretations of CO2 and CH4 concentrations and fluxes within Results sections 3.2.1 and 3.2.2, and Discussion sections 4.1.1 and 4.1.2.

- This manuscript requires a substantial improvement of the **section 4.3**. Scientific arguments should be more specific and based on the findings from this study.
--See below.

Technical corrections

In the introduction:
From 35-36: The paragraph seems to be general and does not provide precise information about the weight of each process or where do they mainly occur? Is there a latitudinal gradient from Sub-Arctic to Low-Arctic and High-Arctic.
--We have attempted to increase the focus of the introduction on the differences between low Arctic and high Arctic greenhouse gases exchange. Most of the section has been re-written and re-organized with additions of new information.

41-42: Check sentence structure
--This sentence has been modified to, "Due to its poor solubility in water, CH_4 can then be effectively released to the atmosphere from these ecosystems by ebullition and wind turbulence, perhaps contributing up to 12% of global emissions (Lai, 2009; Walter et al., 2006)."

45-46 and 51: The freshwater systems cover more than 50% of area in northern regions but less than 5% in polar semi-desert landscapes? Authors may better explain this important difference between the general point of view and specific semi-desert landscapes, and could describe the latitudinal/landscape gradient?
--In the third sentence of the Introduction, we now specify that, "Northern latitudes, between approximately 45 and 75 °N, contain the highest abundance of lakes, ponds and wetlands on the planet (Lehner and Doll, 2004) due to historical glaciations and moderate annual precipitation." In the third paragraph of the Introduction, we now specify that, "In the high Arctic (>~70°N; AMAP), lake abundance and area are dramatically reduced on the landscape. The prevalence of cold and dry high pressure air masses results in a semi-arid climate with relatively well-drained and unproductive inorganic soils (Campbell and Claridge, 1992). This environment, therefore, discourages surface water retention with often less than 5% of the landscape being covered by aquatic systems" After each of these statements, we now provide more detailed information about the landscapes with respect to carbon GHG exchange.

61-62: Control the relevance of chosen references, particularly Peterson et al. (2002) about permafrost thaw and Manabe et al., (1994) about growing seasons.
--These references have been replaced with Froese et al. 2008 (for Peterson et al.) and Myneni et al. 1997 (for Manabe 1994). Euskirchen 2007 replaced Froese et al. 2008.

68: the reference (Antony et al., 2014) does not correspond to the sentence about polar semidesert.

--This sentence has been replaced, including the original reference.

68-70: Sentence is not clear, check its structure
70: although it is uncertain how rapid climate change will alter the C cycle in northern landscapes, this study does not provide strong information about its evolution.
--These two sentences have been replaced with the following: "However, the net result of these processes on high-latitude freshwater carbon GHG exchange is not well delineated, nor is the relative contribution of freshwater systems to total landscape carbon GHG exchange. This information, from a rapidly changing and extensive biome (>10^6 km) is critical for improved global carbon models and budgeting."

In Methods

Figure 1: the figure should be modified. The general maps are too small and thus not useful, the north arrow and the scale are also too small and not visible. Even on a half page, pond and lake pictures are small and don not provide any information. Authors may choose some of the pictures to illustrate the differences among landscapes/freshwater systems.
--Figure 1 has now been modified as per the reviewer's suggestions.

100: Sentence structure
--This sentence has been modified to, "We also sampled shoreline water of Lake Hazen which potentially interacted with ponds located adjacent to its shoreline."

102: how many samples were collected each year and what would be there contribution to mean values? If there is temporal heterogeneity in sampling, mean values may differ with both spatial and temporal evolution.
--We have now included a new Table 2 which is similar to Table S2, but shows only individual sample numbers of greenhouse gas collections, and full chemistry collections. We also bring more attention to the spatial imbalance of sampling. We include a reference to the new Table 2 by re-writing the last few sentences of section 2.1 to read, "Due to logistical issues related to accessing this remote area over consistent time periods each year, and due to the distances of some ponds from base camp, we completed an overall unbalanced sampling program in space and time. As a result, we focused on delineating biogeochemical differences between different types of high Arctic lakes, rather than on inter-annual biogeochemical trends within lakes. Regardless, all sampling occurred during the summer growing seasons of 2005 to 2012 (except for 2006), between mid-June and early August (Table 2, S2).

From 103: how many samples did you analyse for dissolved CO2 and CH4 and how many did you use to calculate fluxes?
--We have now included a new Table 2 showing the frequency and year of CO2 and CH4 sampling, as well as chemistry sampling. The caption states that fluxes were calculated based on concentration sampling.

135: same title for 2.3 and 2.2
--This has been fixed, 2.3 should have stated "fluxes" rather than "concentrations".

In general how are analysis and calculation representative?

--There were two main analyses using results from multiple lake sites within different lake types. The hierarchical cluster analysis used each full biogeochemical sampling effort from each lake. Some lakes were sampled more intensively than others, so this would work as a conservative approach against finding defined groupings (e.g., there is potential for high biogeochemical variability between lakes sampled fewer times, so these should not separate from each other in the HCA in a consistent manner, unless differences were large). However, because the lake types represent such strongly different chemistries, our HCA results were well-defined, despite the higher chances of having hard to interpret results. We have added a sentence in the methods describing how our unbalanced approach should be conservative against finding well-separated groupings. The linear mixed model internally adjusts to unbalanced designs by using means of individual sites to find a representative group mean. Therefore this approach, as explained in the methods in Section 2.5, should be a solid technique for quantifying differences in CO2 and CH4 concentrations and fluxes between the lake types.

From 160: do differences in sampling frequencies have consequences to compare dissolved gas concentrations and fluxes to biogeochemical functioning? For example, for the ponds 10, 11, 16, chemical analyses were only performed on samples collected from 2010. In 5 years, pond conditions may have significantly changed with the important climate change in this region.

--Because of logistical constraints which resulted in an unbalanced sampling design in time and space, we have focused on comparing lake types, rather than changes over time, or changes between individual lakes. The reviewer is correct that interannual changes in climate and possibly lake chemistry could have been considerable over a several year time scale, however by focusing on the large differences between lake types and their general time series patterns in our results and interpretations, we circumvent many of the difficulties associated with the unbalanced approach. Further, when comparing dissolved gases and chemistry between lake types, we only used concurrent samples when gases and chemistry were taken together.

<u>In Results</u>

Figure 2: Not useful, could be put in supplementary data. Moreover, what represent the numbers between brackets? If they represent the number of samples, how authors can compare some sites with 15 samples and some sites with only 1? Noteworthy is the close relationship between Shoreline and Meltwater ponds, closer than Lake Hazen shoreline.

--This figure has been moved to SI. We have updated the caption to explain the numbers in the brackets, which are the number of nodes (samples) compressed by site for ease of display. Please see responses above and below about comparing samples of different sizes. We agree that the closer relationship between Shoreline and Meltwater ponds is interesting, and likely driven by the fact they are each small systems. However, we are having difficulties finding a suitable place within the paper to discuss these more resolved results and setting them within the much larger context of watershed-scale gas exchange.

Table 2: How many samples for each pond type (not lake type)? Standard deviation should also be added. Authors my also provide mean and SD of the different physical and chemical parameters for each pond in order to compare with group values (in supplementary data for example). TDN could be added.

--Please see the new Table 2 for number of samples taken for each water body. Standard deviations have been added to the former Table 2 (now Table 3). Table 3 now has mean and SD for each pond and TDN has been added to the table.

189: Illustrate the sentence 'without extremes during the growing season' with a figure;

--The statement, "without extremes during the growing season" has been supplemented with, "(see section 3.2)." at the end of the sentence. This section introduces the time series figure.

195: Ammonia is not only produced in anoxic conditions, 'reduced ions' could be rephrased as ammonia or nutrient or inorganic ions, mineralization products: : :

--This sentence has been removed from the manuscript.

196: How the Table S4 shows the increase in concentration of NH4 with chemical change during the onset of flooding?

--We have now added pre- and post-flooding chemistry into the former Table 2 (now Table 3). We have updated the reference to Table S4 to Table 3. We now focus on NO_3^-+NO_2^- as an indicator of chemical change during flooding.

196: Both spatial and temporal aspects are used in cluster analysis. This may not be representative due to the discrepancy in sampling.

--By using the cluster analysis with multiple sites over multiple years, we are maximizing variation within and between sites, and increasing the potential for spurious organization of the freshwater systems. However, based on the results and the chemical differences between groups of lakes (new Table 3), it was evident that between-lake chemistry differences outweighed within-lake differences between years. We have modified the "Numerical analysis" section by adding after the first sentence, "Because sampling was unbalanced in frequency and time between sites due to logistical challenges (Table 2; see section 2.1), potential overlap of chemistries between individual lakes was high, therefore setting a conservative standard for classifying distinct lake types."

200-201: Inference from results that may be placed in discussion

--This result or interpretation is not central to this paper, so the sentence has been removed.

Figure 3: The figure is not clear, seasonal trends are not clear, differences among sites and years are difficult to see. Scales of vertical axes could be modified according to maxima and minima values, especially for CH4 in meltwater ponds and lake Hazen shoreline. Lines between dots for Evaporative ponds should be removed; authors do not know what occur between their measurements. Evaporative ponds exhibit significantly less measurements than other sites.

--At the outset of this study, our aim was to group and describe different types of high Arctic aquatic systems. Each year, the timing and extent of our visits changed due to logistical

difficulties and the timing of other studies on site, so interannual differences within systems were difficult to delineate. We felt, therefore, that by standardizing the axes of the graph, we would better highlight differences between the lake types. We believe this also allows the reader to more easily see the general intra-annual trend for each lake type. For CH4, this meant that only the Shoreline ponds showed important trends, which was much of our point in our discussion anyways. We have removed lines between the dots in all graphs. Unbalanced sampling design has been discussed in other responses above.

Results from this figure are not well explained and explored. Only cited twice at the beginning of 3.2.1 and 3.2.2, but not any arguments are based on this figure. Authors do not develop the seasonal trend of dissolved CO2 ad CH4. Comparison between years would be better highlighted using bars plots or a simple table.

--We realize that the beginning sentence of section 3.2.1, where Figures 3 and 4 are referenced, may de-emphasize the role the figures play in presenting the results through the balance of the section. We have strategically placed more references to Figures 3 and 4 throughout the balance of section 3.2.1 so as to clarify the importance of these figures. We do structure arguments later on based on the general trends in GHG exchange and differences between systems shown in Figure 3. However our sampling logistics would not allow for meaningful inter-annual comparisons within lake types, though that was not the aim of this study. We would prefer to keep Figure 3 as is, because bar plots, tables or individually-scaled axes would over-emphasize interannual differences, rather than between-type differences which were the focus of this study. However, we have added this same figure, fully-scaled for each site, in the SI (Figure S4). We have also removed connecting lines in the plots.

Figure 4: Unclear, concentrations and fluxes should not be placed together in the same graph. Comparison between concentrations and fluxes in ponds are difficult. I advise to place dissolved gas concentrations in a graph and fluxes in another.

--This figure has been modified so that the upper panel shows only concentrations of CO2 and CH4, while the lower panel shows only fluxes of CO2 and CH4.

205-206: Although dissolved CO2 concentrations showed non-significant differences, authors compared these values between system types.

--We have stated at the beginning of the section that the differences were not statistically significant. However, we feel it is still a useful exercise to compare the systems because, for example, the relatively fewer samples from the Evaporative ponds could have possibly influenced their high variability and therefore non-significant differences with the other systems, which were sampled more consistently.

209: same comment as line 195.

--This sentence has been modified to, "These ponds were the shallowest of the four sampled and were rich in dissolved iron, DIC, and , TDP."

221-222: CO2 and O2 correlation and relationship with water temperature not well showed in the Figure 5. Correlation coefficients may be placed in the main manuscript.

--We have updated this sentence to reflect the weak relationship between CO2 and O2, but strong association between CO2 and water temperature, "Mean diurnal trends in CO_2

concentrations across all sampling years, as measured by the automated system at Skeleton Lake, showed that CO_2 and O_2 concentrations had little association together (Pearson correlation: r= -0.18, df=7, p=0.67), but CO_2 associated strongly and negatively with water temperature (r=-0.97, df=7, p<0.001; Figure 5)." We have also updated section 4.1.1, to reflect the poor association between CO2 and O2, "Further, mean diurnal CO_2 and O_2 concentrations in surface waters associated poorly together, rather than oppositely if metabolic processes (i.e., primary productivity or decomposition of organic matter; see Pond 01 below) were dominant drivers in surface waters." We have updated the same results for Pond1 in the following results paragraph to, "Diurnal trends of CO_2 and O_2 concentration measured by the automated system at Pond 01 over several growing seasons showed opposite diel patterns of the gases, with greater O_2 during the warmest and lightest parts of the day (r=-0.98, df=7, p<0.001; Figure 4)."

233-234: The sentence is not clear. Clarify in text

--We have decided that this sentence should be removed for clarity purposes.

245-246: Still not any significant differences among pond types, but authors compared shoreline ponds values to other systems (255).

--There are statistical differences between Shoreline Ponds (discussed in paragraph following this one) and the others with respect to CH4 concentrations (Figure 4). Therefore, it is prudent to discuss why Shoreline Ponds were different from the other systems.

269-273: These are not results from this study, should be placed in discussion.

--The first sentence of this group has been placed in section 2.5 of the Methods section. The second sentence has been removed from the manuscript.

277: Is the assumption of generalization relevant and representative of the mean lake composition?

--There are two reasons that this assumption may be valid. First, only a thin moat along the shoreline of Lake Hazen is exposed during many years if summer temperatures remain cool and wind storms are infrequent. During these years, lake-scale gas exchange would only occur in the shoreline areas. Second, more targeted work is ongoing at Lake Hazen that has uncovered evidence that shoreline gas exchange is comparable to pelagic regions of this ultra-oligotrophic lake. We have modified this sentence to include reference to unpublished data from 2015, "When assuming its shoreline waters were representative of the entire lake area as recent evidence suggests (unpublished data, 2015), the expansive Lake Hazen…".

Figure 6: CO2 and CH4 fluxes should be separated. Vertical scales should be modified, for most ecosystems CO2 flux values cannot be read. Figure 6b may be change to a table. Although units were different, CO2 and CH4 fluxes have been already shown in Figure 4. This figure should be modified.

--This figure has now been replaced with a table of values and indications of statistical significance between ecosystem types.

In the discussion

300: 'other compounds' is not clear

--We have updated this sentence to read, "Concentrations of CO_2 and other water chemistry measurements were highest in small…".

302: 'considerable' is a bit excessive considering dissolved CO2 concentrations

--We have removed the word "considerable".

303: Are there evaporates in Lake Hazen watershed? Do you think weathering of carbonates is higher in Evaporative lake than in other systems (pH almost similar in all ponds)? Can DIC be released from surface water exhibiting pH around 8.3? This sentence is too general, higher CO2 concentration originates from higher microbial decomposition or as you write after due to concentration effects.

--We have included in the methods (Section 2.1) a sentence that states evaporitic and carbonitic geology is prevalent in the watershed. Marce et al. 2015 (Nature Geoscience v8) demonstrate the potentially large contribution to CO2 supersaturation in lakes by carbonate in high alkalinity environments, such as those at our sites, though its magnitude is affected by water temperature. Evaporation-concentration of shallow evaporative lakes have helped push their alkalinities above 2 mEq L-1, unlike most other systems we studied, which were less than 2 mEq L-1 (except Pond 1: 2.3 mEq L-1). This may have amplified the contribution of carbonates to CO2 supersaturation in these lakes, but yes, overall we may not expect substantial weathering differences between lake types. We have added to this sentence to strengthen the argument that carbonate weathering played a role in CO2 concentrations in our lakes: "Dissolved CO_2 was likely being produced effectively in all Evaporative ponds by ecosystem metabolism because of their high concentrations of DOC. These, another other, isolated systems concentrate many solutes in their waters including degraded allochthonous and fresh autochthonous DOC (Tank et al., 2009), which would be available as a source of energy to heterotrophs. Accumulation and dissociation of weathered carbonates and evaporates in these moderately warm, high alkalinity environments (2-5 mEq L^{-1}) may have also been important (Trettin, 1994; Marcé et al., 2015)."

311: Do you have evidence of pond stratification other than correlation between CO2 and CH4 concentrations?

--Yes, in the previous sentence we refer to Figure S6, which shows stratification of Skeleton Lake, a Meltwater system.

315: Associations may be replaced by correlations.

--"Associations" has been replaced by "correlations".

316-318: How do you evidence that productivity of microbial decomposition where not the main drivers? Both primary productivity and microbial activity could increase with temperature during the day and lead to diurnal O2 and CO2 concentration trends following temperature.

--This is a good point and not one that our data could definitively solve. Our statement, rather, is supported by the strong, opposite diurnal patterns of CO2 and O2 observed in the visibly more productive Pond 01 compared to Skeleton Lake. Pond 01 supported a widespread

emergent vegetation community compared to Skeleton Lake's benthic mat communities in the near-shore area. We have attempted to downplay our statement by modifying the sentence in question to, "Results from our automated systems supported this argument as mean diurnal CO_2 and O_2 concentrations in surface waters of Skeleton Lake associated poorly together, rather than oppositely if metabolic processes (i.e., primary productivity or decomposition of organic matter; see Pond 01 below) were dominant drivers in surface waters."

320-321: rephrase 'pre to post-flooding mean chl-a concentrations of 1.2 to 0.4 _g l-1)

--The bracketed item has been rephrased to, "(pre-flooding: 1.2 µg L^{-1} chl-a; post-flooding: 0.4 µg L^{-1} chl-a)"

324: 'reduce compounds' could be rephrase as nutrients or ammonia/nitrates, ammonia is not only produced in reduce conditions.

--The second half of this sentence has been removed from the manuscript.

325-326: The sentence is not clear. Moreover, how diurnal O2 and CO2 concentration trends suggest that primary productivity was consistently occurring in Shoreline while you seem to suggest the opposite l 316-318?

--Discussion of diurnal gas concentrations on lines 316-318 refers to trends observed in Skeleton Lake only. We have modified the unclear sentence to, "Although negatively correlated diurnal CO_2 and O_2 concentrations suggest that primary productivity was consistently occurring in Shoreline pond surface waters, seasonal flooding of the ponds was ultimately the more important process controlling seasonal CO_2 concentrations."

336-340: How can you evidence that SO42- production outcompeted CH4 production? Maybe the locations of SO42- and CH4 productions were different or the anoxia could not sustain methanogenic bacteria activity. Do you have measurements of dissolved O2 or redox potential in the ponds?

--This statement is indeed somewhat speculative based on the lack of site-specific data from the sediments. We use anecdotal evidence to suggest that sulfate-reduction was the important anoxic process occurring in the sediments based on low CH4 concentrations in the lake's waters, a strong H2S smell from sediments in extracted cores (results unpublished to date), and the relative lack of ebullition fluxes from the lake. We would expect at least some evidence of higher CH4 fluxes during or after wind events over the extensive sampling record of Skeleton if location of methanogenic CH4 production was simply distributed in space according to micro-conditions in the sediments. Our water boxes did measure dissolved O2 in surface waters, which were relatively high, but unfortunately we do not have such measurements from the sediments. Since we qualify our statement in the proceeding sentence, we believe that the statement, though somewhat speculative, still provides some value to the interpretation of the results by at least highlighting the high concentrations of SO4 in these upland systems. The new sentences read, "Evaporative and Meltwater systems were typically weak producers and emitters of CH_4, which was possibly related to concurrently high SO_4^{2-} concentrations in these systems due to additions of water draining evaporite geologies (Table 3; Trettin, 1994). This may have given competitive advantage to SO_4^{2-}-reducing bacterial communities in sediments, which typically outcompete methanogenic bacteria for hydrogen."

344-345: The sentence is not clear, rephrase.

--This sentence has been modified to, "Only during periods of strong wind mixing of surface waters, or when Shoreline ponds breached and mixed organic particles (Table S4) across its shoreline, did the near shore waters of Lake Hazen release CH_4 to the atmosphere above near-zero values."

354-355: Are you sure (to your knowledge)?

--We have reviewed several studies of terrestrial and aquatic systems, especially from the high Arctic, and most sites and programs do not investigate fluxes from aquatic and terrestrial systems concurrently. However, we will put the caveat "To our knowledge," at the start of the third sentence in Section 4.2.

Table 3 (l 357): Considering the intense Arctic change these last 25 years, how the compilation of data of CO2 and CH4 fluxes throughout more than twenty years can be relevant? Moreover, CO2 and CH4 fluxes may mostly differ according to soil nature, moisture, vegetation, microtopography or local climate conditions and not as a function of large latitudinal regions. I do not think this table provide useful and accurate information. Few words about the comparison between the measurements from this research and other studies would be enough. The main information provided by the table is also not clear.

--We agree with the reviewer that Table 3 should be removed, and this has been done. We have added elements of our findings from this table into other portions of the Discussion, including in the first paragraph of section 4.2.

Paragraph 4.3: This paragraph is too general; no specific point from your study is developed. Only few references are used to support your discussion (4 references, of which one is an article from authors). This entire paragraph should be modified: the discussion should be more based on your results, a specific and original point of view should be developed and your findings better compared with more articles.

--Section 4.2 (formerly incorrectly numbered 4.3) has been completely re-written with improved focus on our results, with more specific original discussion points and with more complete referencing.

Reviewer #2

…better investigating of the biogeochemical processes responsible of contrasted CO2 and CH4 concentrations and fluxes and for a strongly improved discussion.

--We have attempted to improve some of the biogeochemical interpretations, as outlined above. Discussion section 4.2 has been re-written and improved (see reviewer response above).

…Lack of references in certain sections and calls for improved figures and provides several suggestions how to improve the figures and the text.

--Referencing in the last discussion section (4.2) has been improved with the re-write. Figures have been improved based on Reviewer #1 comments. We prefer not to make the suggested changes in the old Figure 3 (now Figure 2), please see reviewer response above. Text-based suggestions by Reviewer #1 have been made (see above).

Please also check the numbering sections and subsections in the revised manuscript (section 4.2 seems missing).
--This has been fixed appropriately.

Carbon dioxide and methane fluxes of freshwater systems in the rapidly changing high Arctic

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Abstract. A warming climate is rapidly changing the distribution and exchanges of carbon within high Arctic ecosystems. Few data exist, however, which quantify exchange of both carbon dioxide (CO_2) and methane (CH_4) between the atmosphere and freshwater systems, or estimate freshwater contributions to total catchment exchange of these gases, in the high Arctic. During the summers of 2005 and 2007-2012, we quantified CO_2 and CH_4 concentrations in, and atmospheric exchange with, common freshwater systems in the high Arctic watershed of Lake Hazen, Nunavut, Canada. We identified four types of biogeochemically-distinct freshwater systems in the watershed, however mean CO_2 concentrations (21–28 µmol L^{-1}) and atmospheric exchange (-0.013 to +0.046 g C-CO_2 m^{-2} d^{-1}) were similar between these systems. Seasonal flooding of ponds bordering Lake Hazen generated considerable CH_4 emissions to the atmosphere (+0.008 g C-CH_4 m^{-2} d^{-1}), while all other freshwater systems were minimal emitters of this gas (<+0.001 g C-CH_4 m^{-2} d^{-1}). When using ecosystem-cover classification mapping and data from previous studies, we found that freshwaters were unimportant contributors to total watershed carbon exchange, in part because they covered less than 10% of total area in the watershed. High Arctic watersheds are experiencing warmer and wetter climates than in the past, which may have implications for moisture availability, landscape cover, and the exchange of CO_2 and CH_4 of underproductive, but expansive, polar semidesert ecosystems.

Keywords: carbon dioxide, methane, high Arctic, ponds, lakes, climate change, watershed

1 Introduction

Freshwater ecosystems cover less than 10% of global ice-free land area (Lehner and Doll, 2004) and have been typically overlooked as substantial contributors to, or sinks of, atmospheric carbon greenhouse gases (GHGs; Bastviken et al., 2011). However, recent studies suggest inland lakes collectively receive and process carbon at magnitudes similar to oceanic uptake and sediment burial, making them important systems within the global carbon

cycle (Cole et al., 2007; Battin et al., 2009; Tranvik et al., 2009; Maberly et al., 2013; Raymond et al., 2013). Northern latitudes, between approximately 45 and 75 °N, contain the highest abundance of lakes, ponds and wetlands on the planet (Lehner and Doll, 2004) due to historical glaciations and moderate annual precipitation. These regions also contain the world's largest below-ground stores of organic carbon (Tarnocai et al., 2009). These carbon and lake-rich northern ecosystems, therefore, have been critically-important sinks historically, and potentially strong emitters of this legacy carbon moving forward (ACIA, 2004).

Most northern lakes are net sources of the GHG carbon dioxide (CO_2) to the atmosphere (Jonsson et al., 2003; Tranvik et al., 2009, Laurion et al., 2010). Cold climates, short growing seasons, and light limitation in stained, carbon-rich waters can inhibit activities of aquatic primary producers (Karlsson et al., 2009), and therefore the uptake of atmospheric CO_2 by the lake ecosystem. Conversely, heterotrophic respiration by microbes, amplified under favourable biogeochemical conditions, continues perennially in most lake waters and sediments, therefore continuously releasing CO_2 to the water column. Turbulence, water temperature, degree of ice-cover and other factors may then influence the intensity of CO_2 emissions to the atmosphere. Lakes in carbon-rich lower Arctic regions (~60-70 °N, AMAP, 1998) can account for more than three-quarters of a landscape's net CO_2 emissions to the atmosphere (Abnizova et al., 2012). At the same time, saturated peatlands and shallow ponds and lakes throughout much of the lower Arctic can also be robust emitters of the potent GHG methane (CH_4) to the atmosphere. Permafrost in northern soils is an effective barrier to drainage of soils, and combined with generally low-elevation topography, means shallow standing water is prevalent throughout northern latitudes. Anoxic conditions in saturated, shallow, organic-rich soils have strong potential for methanogenic production and release of CH_4 into water (Tagesson et al., 2012). Due to its poor solubility, CH_4 can then be effectively released to the atmosphere from these ecosystems by ebullition and even minor wind turbulence, perhaps contributing up to 12% of all global emissions (Lai, 2009; Walter et al., 2006). These dynamic and carbon-rich environments, though, are not ubiquitous across the North, particularly towards the highest latitude regions.

In the high Arctic (>~70°N; AMAP, 1998), lake abundance and area are dramatically reduced on the landscape. The prevalence of cold and dry high pressure air masses results in a semi-arid climate with relatively well-drained and unproductive inorganic soils (Campbell and Claridge, 1992). This environment, therefore, discourages surface water retention with often less than 5% of the landscape being covered by aquatic systems. These conditions, in most cases, restrict primary production and accumulation of organic matter across these

landscapes compared to the lower Arctic, with mostly unknown implications for carbon GHG exchange in high Arctic lakes and ponds. Considering these challenging conditions, it may be easy to overlook the high Arctic, and its freshwater systems, as important contributors to global carbon cycling (Soegaard et al., 2000; Lloyd, 2001; Lund et al., 2012, Lafleur et al., 2012). However, recent studies have shown that where conditions are favourable (e.g., moist, organic-rich lowlands), high Arctic ecosystems exchange GHGs at rates similar to ecosystems at more southerly latitudes (Emmerton et al., 2016). Lack of a broad understanding of carbon cycling in high Arctic freshwater systems is further complicated by rapidly changing climate and landscapes across these latitudes due to human-induced warming.

High Arctic ecosystem productivity is currently changing as a warming climate substantially alters polar watersheds (IPCC, 2007a). Some climate models predict that in the Canadian Arctic, autumn and winter temperatures may rise 3-5°C by 2100, and up to 9°C in the high Arctic (ACIA, 2004; IPCC, 2007b). Mean annual precipitation is projected to increase ~12% for the Arctic as a whole over the same period, and up to 35% in localized regions where the most warming will occur (ACIA, 2004; IPCC, 2007b). Such warming and wetting is already modifying Arctic landscape energy balances (Euskirchen et al., 2007) resulting in glacial melt (Pfeffer et al., 2008), permafrost thaw (Froese et al., 2008), reorganized hydrological regimes (i.e., drying or wetting; Smith et al., 2008) and extended growing seasons (Myneni et al., 1997). These changes are also perturbing watershed carbon cycling through, for example, the liberation of carbon from thawing permafrost, and increases in biological productivity on landscapes and in lakes, ponds and wetlands (Mack et al., 2004; Smol et al., 2005; Walker et al., 2006; Smol and Douglas, 2007). However, the net result of these processes on high-latitude freshwater carbon GHG exchange is not well delineated, nor is the relative contribution of freshwater systems to total landscape CO_2 and CH_4 exchange. This information, from a rapidly changing and extensive biome ($>10^6$ km^2) is critical for improved global carbon models and budgeting.

The primary objective of this study was to measure the net atmospheric exchange of CO_2 and CH_4 with common high Arctic freshwater ecosystems, and place these findings in context with similar terrestrial studies from the same high Arctic location. Using these sources together, we aim to delineate a clearer, watershed-scale understanding of high Arctic exchange of CO_2 and CH_4.

2 Methods

2.1 Location and sampling overview

We conducted our research at the Lake Hazen base camp in central Quttinirpaaq National Park, Ellesmere Island, Nunavut (81.8º N, 71.4º W), Canada's most northerly protected area (Figure 1). Lake Hazen (area: 542 km^2; max. depth: 267 m) is the world's largest high Arctic lake, and is surrounded by a substantial watershed (6,901 km^2) composed of carbonate, evaporite and dolomite rock (Trettin, 1994) and crysolic soils. About 38% of the Lake Hazen watershed is glaciated with the balance of area covered by a polar semidesert (>80% of ice–free area; Edlund, 1994), small lakes, ponds and meadow wetlands. The lower Lake Hazen watershed is a high Arctic thermal oasis (France, 1993) as it experiences anomalously warm growing season (June–August) conditions because it is protected from cold coastal weather by the Grant Land Mountains and Hazen Plateau (Table S1). For example, mean July air temperature is typically 8-9 ºC at the base camp, compared to July 1981-2010 climate normals of 6.1 ºC and 3.4 ºC at the coastal Eureka and Alert weather stations on Ellesmere Island, respectively (Environment Canada, 2016). Soils in the region are also atypically warm during the summer because of low moisture content and efficient radiative heating due to an abundance of clear-sky days. These conditions, coupled with continuous daylight during the growing season, have resulted in a greater diversity and abundance of vegetation and wildlife in the Lake Hazen watershed compared to surrounding areas (France, 1993), despite receiving only ~34 mm of precipitation during the growing season (Table S1). Ultra-oligotrophic Lake Hazen itself dominates the freshwater area of the watershed (Keatley et al., 2007) and receives most of its water annually from rivers discharging melt water from glaciers. Water exits Lake Hazen via the Ruggles River. Ice-cover can remain on Lake Hazen throughout the growing season, though in recent years the lake has gone ice-free more frequently, usually by late July. Ponds and a few small lakes are scattered throughout the lower watershed and are mostly shallow, small in area (~70% are <1 ha) and typically go ice-free by mid- to late-June each year.

To quantify net GHG exchange of typical high Arctic freshwater bodies, we identified several permanent ponds or small lakes to sample within walking distance of base camp to the northwest of Lake Hazen (Figure 1). These systems were chosen systematically to incorporate a gradient of watershed position, surface area, mean depth, emergent vegetation productivity, and hydrological connectivity (Table 1). We also sampled shoreline water of Lake Hazen which potentially interacted with ponds located adjacent to its shoreline. Due to logistical issues related to accessing this remote area over consistent time periods each year, and due to the distances of some ponds from base

camp, we completed an overall unbalanced sampling program in space and time. As a result, we focused on delineating biogeochemical differences between different types of high Arctic lakes, rather than on inter-annual biogeochemical trends within lakes. Regardless, all sampling occurred during the summer growing seasons of 2005 to 2012 (except for 2006), between mid-June and early August (Table 2, S2).

2.2 Dissolved CO_2 and CH_4 concentrations of high Arctic freshwaters

Two approaches were used to quantify concentrations of dissolved CO_2 and CH_4 in surface waters. The first approach was employed at all sites and used the common method of collecting water directly into evacuated 160-mL Wheaton glass serum bottles capped with butyl rubber stoppers (Hamilton et al., 1994; Kelly et al., 1997). Each bottle contained 8.9 g of potassium chloride (KCl) preservative to kill all microbial communities (Kelly et al., 2001), and 10 mL of ultra high purity dinitrogen (N_2) as a gas headspace. To collect a sample, a bottle was submersed ~5 cm below the water surface and punctured with an 18-gauge needle. Barometric pressure and water temperature were recorded. Dissolved gas samples were stored in the dark at ~5˚C until return to the University of Alberta, where they were analyzed in the accredited Biogeochemical Analytical Service Laboratory (BASL). There, samples were placed in a wrist-action shaker for 20 minutes to equilibrate dissolved CO_2 and CH_4 with the N_2 headspace. Headspace CO_2 and CH_4 concentrations were quantified on a Varian 3800 gas chromatograph (GC) using a flame ionization detector at 250°C with ultra high purity hydrogen (H_2) as a carrier gas passing through a hayesep D column at 80°C. A ruthenium methanizer converted CO_2 to CH_4. Four gas standards (Praxair, Linde-Union Carbide), ranging from 75 to 6000 parts-per-million for both CO_2 and CH_4, were used to calibrate the GC. A Varian Star Workstation program integrated peak areas and only calibration curves with an r^2 >0.99 were accepted for analyses. A standard was re-analyzed every 10 samples to reconfirm the calibration, and duplicate injections were performed on all samples. Headspace CO_2 and CH_4 concentrations were converted to dissolved molar concentrations using Henry's Law, and corrected for temperature and barometric pressure differences between sample collection and analysis. To quantify dissolved inorganic carbon (DIC) concentrations, samples were acidified with 0.5 mL H_3PO_4 to convert all DIC to CO_2, and then immediately reanalyzed on the GC. DIC concentrations were calculated as above.

The second approach involved two automated systems to determine detailed diel changes in surface water dissolved CO_2 concentrations at two different sites (Skeleton Lake and Pond 01; Figure 1; Table S2). Dissolved CO_2 concentrations were measured every three hours during several summers. These systems functioned by equilibrating,

over a 20-minute period, dissolved CO_2 from pumped surface waters, with a gas cell in a Celgard MiniModule Liqui-Cel. The equilibrated gas was then analysed for CO_2 concentration by a LI-COR (Lincoln, NE) 820 infrared gas analyzer. The systems also measured dissolved oxygen (O_2) concentrations using a Qubit™ flow-through sensor. Concentrations were then converted to aqueous molar concentrations using Henry's Law and water temperature quantified with a Campbell Scientific (Logan, UT) 107-L thermistor. The systems were housed in watertight cases along the shore from which a sample line extended out into the surface waters, and upon which was mounted a CS 014A anemometer (1 m height) and a Kipp & Zonen (Delft, The Netherlands) photosynthetically-active radiation (PAR) LITE quantum sensor. All data were recorded on Campbell Scientific CR10X dataloggers.

2.3 Dissolved CO_2 and CH_4 fluxes of high Arctic freshwaters

Though several models exist for quantifying turbulent gas fluxes of lakes (e.g., MacIntyre et al., 2010), we decided to use the stagnant film model described by Liss and Slater (1974) to quantify net CO_2 and CH_4 mass fluxes between surface waters and the atmosphere at our remote location. This decision was made because of 24-hour daylight at our high-latitude location dampened diurnal surface temperature changes to less than 1°C, the general shallowness of the systems, and the steady, sometimes gusty, wind conditions on site. The stagnant film model assumes gas concentrations in both surface waters and the atmosphere are well-mixed, and that gas transfer between the phases occurs via diffusion across a diminutive stagnant boundary layer. Diffusive gas transfer across the boundary layer is assumed to follow Fick's First Law:

$$\text{Gas flux } (\mu\text{mol m}^{-2}\text{ hr}^{-1}) = k(C_{SUR} - C_{EQL}) \quad (1)$$

where C_{SUR} (μmol L^{-1}) is the concentration of the gas in surface waters, C_{EQL} (μmol L^{-1}) is the atmospheric equilibrium concentration, and k is the gas exchange coefficient, or the depth of water per unit time in which the concentration of the gas equalizes with the atmosphere (i.e., piston velocity). Values of k (cm hr^{-1}) were calculated using automated systems wind measurements and occasionally from nearby (within 2 km) eddy covariance towers (Campbell Scientific CSAT3 Sonic Anemometers; 30 min. means), and published empirical relationships (Table S3; Hamilton et al., 1994). To determine the direction of the flux, atmospheric equilibrium CO_2 and CH_4 concentrations were quantified using Henry's law, in-situ barometric pressure and air temperature, and mean annual CO_2 and CH_4 concentrations in the atmosphere during the year of sampling (Environment Canada, 2015). If dissolved CO_2 and CH_4 concentrations in surface waters were above or below their corresponding calculated atmospheric equilibrium

concentrations, the freshwater systems were considered a source (+) or sink (-) relative to the atmosphere, respectively.

We also measured ebullition fluxes of CH_4 to the atmosphere from two freshwater systems (Skeleton Lake, Pond 01) during two growing seasons using manual bubble collection and GC analysis (see Supporting Information).

2.4 Supporting measurements

We quantified additional physical and chemical parameters in surface waters at the same sites as we collected our GHG samples, although at reduced sampling frequencies (Table 2, S2). At each site, temperature, pH, specific conductivity and dissolved O_2 were measured in-situ using a YSI (Yellow Springs, OH) 556 MPS multi-probe. Water samples were also collected for general chemical analyses (total dissolved nitrogen [TDN], particulate N, NO_3^-+NO_2^-, NH_4^+, total phosphorus [TP], total dissolved phosphorus [TDP], alkalinity, dissolved organic carbon [DOC], total dissolved solids, major cations/anions, dissolved iron, chlorophyll-*a* [chl-a]) into pre-cleaned HDPE bottles. These samples were immediately processed in the Lake Hazen/Quttinirpaaq Field Laboratory clean room after water collection, and stored in the dark at ~5ºC or frozen until analysed at the BASL.

2.5 Numerical analysis

We used hierarchical clustering analysis (IBM SPSS Statistics 23) to organize ponds and lakes into type categories based on concurrent GHG and chemistry analyses (10 sites; n=62; Table 2). Because sampling was unbalanced in frequency and time between sites due to logistical challenges (Table 2; see section 2.1), potential overlap of chemistries between individual lakes was high, therefore setting a conservative standard for classifying distinct lake types. We used between-group linkage and squared Euclidean distances to group similar sites together and delineate distinct high Arctic freshwater types. We then used linear-mixed models (SPSS) to quantify differences in GHG concentrations and fluxes between these different high Arctic freshwater types. Linear-mixed models are ideal for analysing non-independent and repeated measures data as they integrate inherent errors in repeated sampling designs to more clearly distinguish statistical differences between groups. These models also can efficiently handle unbalanced designs by standardizing results from each site within groups. Linear mixed model details included: use of an auto-regressive moving average (1,1) repeated covariance model; use of a Maximum Likelihood estimation method; and variables organized by freshwater type (fixed) and year (random).

2.6 Net atmospheric exchange of CO_2 and CH_4 of a large high Arctic watershed

To better understand the role of freshwater ecosystems in regional fluxes of carbon GHGs, freshwater CO_2 and CH_4 fluxes measured in this study were coupled with terrestrial fluxes measured in the watershed during the 2008-12 growing seasons (Emmerton et al., 2014, 2016). The authors measured, using eddy covariance flux towers (CO_2, CH_4) and static chambers (CH_4), growing season carbon GHG exchange with terrestrial polar semidesert and meadow wetland landscapes from 2008-12. Areal coverage of the different ecosystem types in the watershed was isolated from a previous classification of Quttinirpaaq National Park (Edlund, 1994) using a Geographical Information System (ArcGIS v.10.3; ESRI, Redlands, US). Mean growing season fluxes from each measured ecosystem were then weighted to matching coverage area in the watershed to estimate the total carbon gas exchange with the atmosphere. Glacial ice was assumed to be a net-zero contributor of total watershed gas exchange in this scaling exercise.

3 Results

3.1 Biogeochemical classification of high Arctic freshwaters

Four distinct types of freshwater systems were evident from our sampling in the Lake Hazen watershed (Table 3; **Error! Reference source not found.**; hierarchical cluster analysis; see Methods). "Evaporative" ponds (Ponds 07, 10, 12) occurred in the upland of the Lake Hazen catchment and were hydrologically-isolated from their surrounding basins post-snowmelt. These ponds were relatively high in concentrations of total dissolved solids, most measured ions, DIC, DOC, organic particles, TDP and chl-*a*. Pond 03, though not technically clustered with others, was forced to the Evaporative pond category based on lack of consistent inflowing water and high concentrations of most dissolved ions. This delegation was further consistent with isotopic measurements of oxygen ($\delta^{18}O\text{-}H_2O$) in water taken from each aquatic system in July 2010 (Figure S2). "Meltwater" systems, including Ponds 11, 16 and Skeleton Lake, also occurred in the upland of the Lake Hazen watershed, but received consistent water supply through the growing season primarily from snowmelt, permafrost/ground ice thaw water or upstream lake drainage. The general chemistry of these systems was therefore consistent and without extremes during the growing season (see section 3.2). Typical meltwater streams draining to these ponds were high in TDN and sulfate (SO_4^{2-}), but low in DOC (Table 3), though streams drained through marginal wetlands surrounding the lakes and ponds downstream of our sampling sites. "Shoreline" ponds (Ponds 01, 02) occurred along the margin of Lake Hazen and were

typically physically isolated from the large lake by porous gravel berms, and surrounded by wetland soils and flora during spring low water conditions. As glacial melt accelerated throughout the growing season, though, the water level of Lake Hazen rose and could seep through the berms to incrementally flood the ponds and surrounding wetlands (Figure S3). Shoreline ponds changed chemically during the onset of flooding as indicated, for example, by an increase in the concentration of NO_3^-+NO_2^- (Table 3). A separate smaller cluster of Pond 01 samples occurred during particularly high-water periods when Lake Hazen breached the berms (**Error! Reference source not found.**). The flooding water from the "Lake Hazen shoreline" was cold, dilute in dissolved ions, organic matter, TDN, and chl-*a*, but considerably higher in NO_3^-+NO_2^- compared to other water bodies.

3.2 Dissolved concentrations and net atmospheric exchange of CO_2 and CH_4 of high Arctic freshwaters

3.2.1 CO_2

Growing season concentrations of dissolved CO_2 in sampled high Arctic freshwaters from 2005 to 2012 varied substantially within and between the system types, and therefore overall resulted in non-significant differences between them (Figure 2, 3, S4, S5).

On average, Evaporative ponds had the highest mean CO_2 concentrations (mean±SE; 27.9±4.9 μmol L^{-1}) compared to other pond types (Figure 3), primarily due to conditions in Pond 03 and Pond 07. These ponds were the shallowest of the four sampled and were rich in dissolved iron, DIC, and TDP. CO_2 concentrations were above atmospheric equilibrium concentration (Figure 2) and therefore these ponds were sources of the gas to the atmosphere (+177±66 μmol CO_2 m^{-2} hr^{-1}; Figure 3). The other Evaporative ponds (Ponds 10, 12) were deeper and had CO_2 concentrations that were typically near those of the atmosphere. This contributed to their near-zero exchange of CO_2 with the atmosphere (-5±17 μmol CO_2 m^{-2} hr^{-1}). Together, dissolved CO_2 concentrations correlated closely and positively with DOC and dissolved iron concentrations in Evaporative ponds (Table S4). When combining all Evaporative ponds together, they were net sources of CO_2 to the atmosphere (+73±93 μmol CO_2 m^{-2} hr^{-1}; Figure 3).

Meltwater systems had lower, but insignificantly different, CO_2 concentrations (26.2±3.9 μmol L^{-1}) than Evaporative ponds (Figure 3). Meltwater systems showed only gradual, venting-related declines of CO_2 concentrations through the summer, with strong consistency in concentrations between sampling times and sites (Figure 2). However, they emitted higher, though not significantly different, fluxes of CO_2 to the atmosphere overall (+160±66 μmol m^{-2} hr^{-1}; Figure 3) compared to the other types of systems. CO_2 concentrations of these systems

correlated strongly and positively with CH_4 concentrations, but negatively with DOC concentrations and measurements that were of high concentrations in Meltwater streams draining into the systems (e.g., SO_4^{2-}, TDN; Table 3, S4). Mean diurnal trends in CO_2 concentrations across all sampling years, as measured by the automated system at Skeleton Lake, showed that CO_2 and O_2 concentrations had little association together (Pearson correlation: r= -0.18, df=7; p=0.67), but CO_2 associated strongly and negatively with water temperature (r=-0.97, df=7, p<0.001; Figure 4).

Mean CO_2 concentrations of Shoreline ponds (22.5±3.7 µmol L^{-1}; Figure 3) were similar to the other pond types, which obscured their considerable seasonal changes within and between growing seasons. From 2005 to 2007, both Pond 01 and Pond 02 received little floodwater from Lake Hazen due to lower lake water levels (Figure 2). These conditions resulted in dense wetland vegetation growth surrounding the ponds and low mean daily dissolved CO_2 concentrations (6.5±0.4 µmol L^{-1}) and strong uptake of atmospheric CO_2 (-329±59 µmol m^{-2} hr^{-1}). The drier wetland state of these ponds changed in following summers when Lake Hazen rose substantially upon greater inputs of glacial meltwaters (WSC, 2015), causing the rising waters to seep through porous berms into the ponds through July. In concert with flooding, concentrations of CO_2 from 2008-11 of each pond together increased substantially (30.1±1.5 µmol L^{-1}) resulting in strong net emissions of CO_2 to the atmosphere (+228±44 µmol m^{-2} hr^{-1}). Changing dissolved CO_2 concentrations correlated positively with dissolved nutrients and ions (Table S4). Diurnal trends of CO_2 and O_2 concentration measured by the automated system at Pond 01 over several growing seasons showed opposite diel patterns of the gases, with greater O_2 during the warmest and lightest parts of the day (r=-0.98, df=7, p<0.001; Figure 4). However, the net result of strong seasonality in these ponds was slight net emission of CO_2 to the atmosphere (+42±60 µmol m^{-2} hr; Figure 3) that was not statistically-different from other types of freshwaters.

Lake Hazen shoreline water, though not necessarily representative of the entire lake itself, was characteristic of its moat occurring early each growing season, and of water that intruded Shoreline ponds in July. This water was generally near atmospheric equilibrium concentrations of CO_2 (21.0±7.8 µmol L^{-1}; Figure 2) with stable and low CO_2 uptake throughout the season (-44±66 µmol m^{-2} hr; Figure 3). CO_2 concentrations of this shoreline water related strongest and positively with DIC, NO_3^-+NO_2^-, major ions and wind speed (Table S4).

3.2.2 CH_4

Each of Evaporative, Meltwater and Lake Hazen shoreline freshwaters had statistically similar and low CH_4 concentrations (0.06-0.14 μmol L^{-1}) and fluxes (+0 to +3 μmol m^{-2} hr^{-1}) across all growing seasons (Figure 2,3, S4, S5). Evaporative ponds had generally flat seasonal CH_4 concentration and flux trends (Figure 2), except for an outlier sample from Pond 10 in mid July 2011. CH_4 concentrations correlated strongest with NO_3^-+NO_2^- and alkalinity (Table S4). Meltwater systems were also generally low in CH_4 concentrations and fluxes through the summers and associated positively and closely with CO_2 concentrations, and strongly but negatively with SO_4^{2-}, alkalinity and other ions (Table S4). Notable flux emissions from these systems only occurred during episodic wind events, also similar to CO_2 (Figure S5). However, unlike CO_2, higher CH_4 concentrations were sustained into July in Skeleton Lake in 2010 (Figure 2). Lake Hazen shoreline water showed low and stable CH_4 concentrations and fluxes each growing season with infrequent and small releases of the gas to the atmosphere. CH_4 concentrations in this water correlated positively only with particulate carbon concentrations (Table S4).

Shoreline ponds, alternatively, had significantly higher CH_4 concentrations relative to the other systems (1.18±0.16 μmol L^{-1}; Figure 3) and showed a dynamic seasonal pattern dominated by the timing of flooding (Figure 2). In 2005 and 2007 before substantial seasonal flooding started to occur, CH_4 concentrations (0.29±0.03 μmol L^{-1}) and fluxes to the atmosphere (+8±2 μmol m^{-2} hr^{-1}) were low. As the Shoreline ponds began to receive NO_3^-+NO_2^--rich flood water from Lake Hazen by mid-summer in subsequent years (Table 3), 2008-11 CH_4 concentrations and fluxes increased substantially (1.70±0.13 μmol L^{-1}; +41±10 μmol m^{-2} hr^{-1}) and correlated closely with dissolved organic and inorganic nitrogen (Table S4). This significant increase in CH_4 flux emissions from Shoreline ponds during flooding (>five times higher than during dry periods) was coupled with large increases in pond surface areas, effectively producing even higher total CH_4 emissions to the atmosphere. Towards the end of July during flooding conditions, full berm breach of the Shoreline ponds by rising Lake Hazen waters occurred resulting in rapid dilution of CH_4 concentrations, but logistical constraints prevented later summer sampling to investigate if concentrations rebounded thereafter. Overall, aided by poor solubility of CH_4 in water and episodic wind events (Figure S5), the flooding of Shoreline ponds drove significantly larger CH_4 emissions to the atmosphere compared to other pond types (+28±5 μmol m^{-2} hr^{-1}; Figure 3).

3.3 Net atmospheric exchange of CO_2 and CH_4 of a large high Arctic watershed

When scaled to total watershed area including Lake Hazen (7,443 km^2), polar semidesert landscapes were inconsequential to total CO_2 exchange (-1,253 Mg C-CO_2; 9% of total exchange) despite comprising a substantial proportion of the catchment (3,819 km^2; 51%; Table 4). All types of standing freshwaters sampled in the watershed from this study showed statistically-similar CO_2 fluxes compared to the polar semidesert. When assuming its shoreline waters were representative of the entire lake area as recent evidence suggests (unpublished data, 2015), the expansive Lake Hazen (542 km^2; 7%) exchanged relatively little CO_2 with the atmosphere (-721 Mg C-CO_2; 5%), as did smaller freshwater systems (144 km^2; 2%) in the watershed (600 Mg C-CO_2; 4%). In clear contrast, during the growing season, moist and vegetated meadow wetland ecosystems were found to consume CO_2 at rates similar to wetlands in the southern Arctic (-0.96 g C-CO_2 m^{-2} d^{-1}; Emmerton et al., 2016). Consequently, meadow wetlands exchanged an estimated 82% (-11,368 Mg C-CO_2) of total CO_2 with the atmosphere despite occupying only 2% (129 km^2) of the area in the Lake Hazen watershed. Total CO_2 exchange of the watershed was -10,236 Mg C-CO_2 (-1.38 g C-CO_2 m^{-2}) during the growing season.

The high Arctic polar semidesert has recently gained attention as a notable atmospheric sink of CH_4 (-0.001 g C-CH_4 m^{-2} d^{-1}; Emmerton et al., 2014), which has since been observed in studies at other high Arctic locations (e.g., Jorgensen et al., 2015). These uptake fluxes coupled with its expansive coverage made the polar semidesert the key landscape controlling net CH_4 exchange throughout the Lake Hazen watershed (-412 Mg C-CH_4; 94% of total exchange; Table 4). Surprisingly, a productive meadow wetland in the watershed was a weaker emitter of CH_4 to the atmosphere (+0.001 g C-CH_4 m^{-2} d^{-1}) compared to other high Arctic wetlands (Emmerton et al., 2014), releasing only 10 Mg C-CH_4 (2%) to the atmosphere during the growing season. All upland freshwater systems (Evaporative and Meltwater systems) had low emissions of CH_4 to the atmosphere (11 Mg C-CH_4; 2%), as did Lake Hazen itself (+6 Mg C-CH_4; 1%). All measured ecosystems had statistically-similar CH_4 fluxes except for the strong CH_4-producing Shoreline ponds (Table 4). However, poor areal coverage of these dynamic systems in the watershed (0.6 km^2; <1%) resulted in contributions of <<1% (+0.4 Mg C-CH_4) of all CH_4 exchange in the Lake Hazen watershed (-385 Mg C-CH_4; -0.052 g C-CH_4 m^{-2}).

4 Discussion

4.1 Dissolved concentrations and net atmospheric exchange of CO_2 and CH_4 of high Arctic freshwaters

4.1.1 CO_2

Dissolved CO_2 was likely being produced effectively in all Evaporative ponds by ecosystem metabolism because of their high concentrations of DOC. These, another other, isolated systems concentrate many solutes in their waters including degraded allochthonous and fresh autochthonous DOC (Tank et al., 2009), which would be available as a source of energy to heterotrophs. Accumulation and dissociation of weathered carbonates and evaporates in these moderately warm, high alkalinity environments (2-5 mEq L^{-1}) may have also been important (Trettin, 1994; Marcé et al., 2015). However, differences in pond volumes likely controlled the ultimate concentrations of CO_2 found in Evaporative ponds. Small and shallow Evaporative ponds (Ponds 03, 07) showed much higher concentrations compared with those that were larger and deeper (Ponds 10, 12) and were therefore more susceptible to wind-related turbulence and gas exchange with the atmosphere.

The biogeochemistry of Meltwater systems was steady and similar between sites, possibly related to stream flushing, but they ultimately had similar CO_2 concentrations and fluxes as other freshwater types. This occurred despite inclusion of early summer sampling at Skeleton Lake (2007, 2010) when CO_2 concentrations were higher as post-ice-covered waters were re-equilibrating with the atmosphere (Kling et al., 1992; Karlsson et al., 2013). However, fluxes of CO_2 to the atmosphere from these systems did not correspond closely with early season venting, but rather to the frequency of episodic releases of CO_2 to the atmosphere (Figure S5). This may have been related to their greater mean depths, which promoted stratification in at least one of our sampled Meltwater systems (Skeleton Lake; Figure S6). Stratification would confine decomposition products (e.g., CO_2, CH_4) to near their sites of origin in bottom sediments and extensive benthic mat communities, which would then be released most readily during and just after wind mixing events. We observed evidence of this process via strong positive correlations between CO_2 and CH_4 concentrations in surface waters (Table S4). Results from our automated systems supported this argument as mean diurnal CO_2 and O_2 concentrations in surface waters of Skeleton Lake associated poorly together, rather than oppositely if metabolic processes (i.e., primary productivity or decomposition of organic matter; see Pond 01 below) were dominant drivers in surface waters. Mixing-related releases of CO_2 would be offset by calmer, lower-flux conditions when DOC concentrations may have been higher in upper layers. Meltwater streams flushing

through marginal wetlands before entry into the Meltwater systems, but then not mixing with the entire lake, may explain the negative correlation observed between CO_2 and DOC concentrations.

Shoreline ponds changed drastically in size and chemistry in response to seasonal flooding by Lake Hazen shoreline water (Table 1, 3). During pre-flooding conditions, CO_2 concentrations were low which could be attributed to DIC use by autotrophic plankton (pre-flooding: 1.2 μg L^{-1} chl-*a*; post-flooding:~~to~~ 0.4 μg L^{-1} chl-*a*), but more likely by observed dense benthic and macrophytic communities along the margins of the ponds (Tank et al, 2009). When inundated by flood waters, CO_2 concentrations rose sharply which is typically observed in flooded wetlands (Kelly et al., 1997). This occurs because widespread inundation of plants and soils typically prompts rapid decomposition (Table S4). Although negatively correlated diurnal CO_2 and O_2 concentrations suggest that primary productivity was consistently occurring in Shoreline pond surface waters, flooding of the ponds was ultimately the more important process controlling seasonal CO_2 concentrations.

CO_2 concentrations in Lake Hazen shoreline water were near atmospheric equilibrium and only weakly consumed atmospheric CO_2. These results along the shoreline appear to be similar to other locations offshore (unpublished, 2015) and were reflective of most deep lakes with extremely low nutrient, organic matter and chl-*a* concentrations (0.20 μg L^{-1}; Keatley et al., 2007; Babaluk et al., 2009). CO_2 gas exchange between the lake and the atmosphere correlated well with DIC, alkalinity and other ions, which are considerable in glacial rivers draining to the lake (Babaluk et al., 2009). These rivers were also strongly undersaturated in CO_2, as observed elsewhere in glacial environments (Meire et al., 2015), and may explain the slight CO_2 uptake observed by the lake, especially later in summer.

4.1.2 CH_4

Evaporative and Meltwater systems were typically weak producers and emitters of CH_4, which was possibly related to concurrently high SO_4^{2-} concentrations in these systems due to additions of water draining evaporite geologies (Table 3; Trettin, 1994). This may have given competitive advantage to SO_4^{2-}-reducing bacterial communities in sediments, which typically outcompete methanogenic bacteria for hydrogen. This hypothesis was supported by the prevalence of H_2S gas in collected sediment cores from Skeleton Lake (unpublished, 2013) and by the trivial fluxes of CH_4 in bubbles measured emerging from sediments (+0.00 to +0.01 mg m^{-2} d^{-1}; Table S5; see Supporting Information). Stratification in Meltwater systems and the only periodic wind-related releases of CH_4, similar to CO_2, likely also limited CH_4 emissions (Table S4). Low production and exchange of CH_4 in Lake Hazen,

alternatively, was most likely associated with the lake's ultra-oligotrophic standing (Keatley et al., 2007), well-oxygenated water, and little accumulation of littoral organic matter where anoxia could prevail and CH_4 be produced. Only during periods of strong wind mixing of surface waters, or when Shoreline ponds breached and mixed organic particles (Table S4) across its shoreline, did the near shore waters of Lake Hazen release CH_4 to the atmosphere above near-zero values.

Shoreline ponds were regional "hot-spots" of CH_4 exchange, which was clearly driven by seasonal flooding, and releases of organic matter and nutrients (Table S4). Pre-flooding conditions in the ponds were characterized by dry and oxygenated wetland soils which were exposed to the atmosphere and not connected to the central pond where we sampled. Flooding induced saturation of organic soils surrounding the wetland and perhaps provided advantageous conditions for anaerobic metabolism, including methanogenesis. This may have been further supported by the flushing of the ponds with SO_4^{2-}-poor Lake Hazen water, therefore potentially favouring metabolism of methanogens over SO_4^{2-}-reducers in the flooded soils.

4.2 Net atmospheric exchange of CO_2 and CH_4 of a large high Arctic watershed

Studies from the southern Arctic have estimated that fluxes of CO_2 (e.g., -1.55 to +1.10 g C-CO_2 m^{-2} d^{-1}, Tank et al., 2009, Abnizova 2012) and CH_4 (+0.01 to +0.09 g C-CH_4 m^{-2} d^{-1}, Walter 2006, Sachs 2010) from ponds and lakes can contribute a strong majority of a region's total exchange of CO_2 and CH_4 with the atmosphere (Sachs et al., 2010; Abnizova et al., 2012). Carbon and nutrient-rich soils, longer growing seasons, and high densities of aquatic and wetland ecosystems are likely key characteristics responsible for these strong signals. To our knowledge, concurrent measurement of freshwater and terrestrial carbon GHG exchange at a high Arctic location has not occurred previous to this study. We found that in a large high Arctic watershed, a size range from small ponds up to one of the world's largest high-latitude lakes, together contributed only an estimated 9% (CO_2; -0.01 to +0.05 g C-CO_2 m^{-2} d^{-1} and 3% (CH_4: +0.00 to +0.01 g C-CH_4 m^{-2} d^{-1} of all carbon GHG exchanges (Table 4). Several reasons may explain the limited role of aquatic systems there. First, pond and lake coverage in the high Arctic is typically very low (<10% of Lake Hazen watershed; Table 4) compared to the southern Arctic (Lehner and Doll, 2004). Well-drained soils, a semi-arid climate and continuous evaporation throughout a 24-hour daylight growing season all contribute to negative pond and lake water balances often observed across the high Arctic (Woo and Guan, 2006). Second, growing seasons of high Arctic freshwaters are very short as ice-cover can remain perennially on some lakes, or may vacate for only three months (Rautio et al., 2011). Though ponds in the Lake

Hazen watershed can warm to moderate levels compared to other Arctic locations (Table 3, Rautio 2011), time exposure to these temperatures is short and likely limits growing season autotrophic and heterotrophic activity and their contributions to freshwater carbon gas exchange. Geochemical production of CO_2 in high-alkalinity ponds and lakes is also lessened in only moderately warm environments (Marcé et al., 2015). Third, runoff delivered to high Arctic freshwaters is typically dilute, nutrient-poor and low in quality organic matter because it drains among the most unproductive and desiccated soils anywhere on Earth (ACIA, 2004). Therefore, neither important nutrients key for aquatic photosynthesis (Markager et al., 1999), nor labile carbon for heterotrophic activities are supplied to many high Arctic lakes in great quantities, thus limiting potential biological carbon GHG uptake or emission. These constraints on aquatic productivity were visible at our sites as few were dominated by productive emergent plants, but rather by barren lake beds or submerged benthic mats of weaker productivity.

Despite a challenging climate and poor-quality substrates, our results suggest that the degree of moisture availability in high Arctic ecosystems was an overarching control on CO_2 exchanges. Running water environments are the most productive landscapes in the Lake Hazen watershed (Table 4) because they are consistently wet, but not starved of (e.g., polar semidesert) or inundated by (e.g., ponds, lakes) water. These ideal conditions support productive emergent plant communities, which typically outgrow other vegetation types along the terrestrial-aquatic watershed gradient (Wetzel, 2001). This occurred despite low soil temperatures in these wetlands because of shallow permafrost tables. Productive standing water environments were rare in the Lake Hazen watershed, except for Shoreline ponds during their drier wetland phase. However, the flooding hydrology of Lake Hazen promoted a near balance of net autotrophy and heterotrophy in these systems. For CH_4, the spatial coverage of ecosystem types was the most important factor controlling its exchange at the watershed scale. Only Shoreline ponds, due to the flooding of its wetland vegetation, were substantially higher in per-unit CH_4 gas exchange than other ecosystems (Table 4). However, net uptake of CH_4 by methanotrophs in polar semidesert soils was ultimately of greatest importance at the watershed scale because of the landscape's extensive spatial coverage relative to other ecosystem types. This finding supports other recent studies which highlight the potential global importance of this substantial high Arctic CH_4 sink (Jorgensen et al., 2015).

Modification of moisture availability in high Arctic regions is likely in a changing climate. High Arctic latitudes are expected to endure considerable warming and increased precipitation, resulting in shifting snow and ice phonologies, greater contributions to runoff from subsurface ice and glaciers, and greater evaporation rates (ACIA,

2004). These changes will affect the distribution and sustainability of water across high Arctic landscapes. Smol and Douglas (2007) have suggested that negative water balances and the drying of small and shallow aquatic systems will become a more frequent response to rapidly increasing temperatures and enhanced evaporation. Others have suggested that site-specific hydrological conditions have important controls on the ultimate sustainability of high Arctic waters, including substrate characteristics, snowpack accumulation, and connection to water sources (Abnizova and Young, 2010). In the Lake Hazen watershed, expected increases in nearby coastal evaporation and landward precipitation (Bintanja and Selten, 2014) may deliver larger snowpacks, recharges of subsurface ice or water storage, and increases in summertime runoff to aquatic systems. Increased temperatures, however, should also work to sustain wet areas in the watershed. Increased glacial melt would continue to deliver more water to Lake Hazen and flood Shoreline Lakes for longer periods. Higher temperatures should also improve water delivery to Meltwater systems and meadow wetlands supplied by thawing subsurface ice. Only shallow Evaporative ponds, which endure a precarious existence based on net balances in snowmelt and evaporation, have a less certain future. We suspect that these Evaporative systems may be susceptible to drying over the shorter term as air temperatures increase, but the weak water storage capacity of well-drained polar semidesert soils continues. Only until long-term improvements in productivity and organic matter content in soils occurs, would we expect more consistent sources of runoff to shallow systems. Well-drained polar semideserts, similarly, may also be expected to remain relatively dry until water holding capacity of the soils improves (Emmerton et al., 2016).

With expected sustainability of water delivery to most wet systems in the Lake Hazen watershed over the longer term, future carbon GHG exchange there and other high Arctic regions, is likely dependant on the trajectory of landscape change of polar semideserts (Sitch et al., 2007). Low CO_2 and CH_4 exchange in upland systems and Lake Hazen will likely continue until water and nutrient conditions in polar semidesert soils draining to them improves over the longer term. Shoreline ponds may be flooded earlier and for longer periods as Lake Hazen receives increased glacial melt water, possibly amplifying carbon GHG emissions over the short term. However, supply of decomposable organic carbon may decrease as periods when these systems are in a productive wetland-state becomes less frequent. Regardless, Shoreline ponds likely have little role in regional carbon GHG exchange due to minimal abundances. Consequently, changes in the terrestrial ecosystems, over the longer term, should continue to define the direction and intensity of GHG exchanges in the high Arctic. Meadow wetlands are key high Arctic regions due to substantial growing season productivity and CO_2 consumption, despite their low abundance.

Notable spatial expansion of these very productive systems, though, is unlikely due to topographical constraints. The potential of dry polar semideserts to change, however, is great over the long term (ACIA, 2004). As plant growth, organic matter production and soil water retention improve as expected in the polar semidesert, its CO_2 sink strength during the growing season should also improve. However, this may also work to perturb atmospheric oxygen and methane infiltration into polar semidesert soils and perhaps decrease the magnitude of its globally-important atmospheric CH_4 sink (Jorgensen et al., 2015). Ultimately, terrestrial ecosystems and their future climate-related changes, rather than those in lakes and ponds, will likely control future carbon cycling at high Arctic latitudes.

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Tables

Table 1 Morphometry and hydrology of ponds and lakes sampled for dissolved greenhouse gases concentrations and general chemistry in the Lake Hazen (LH) watershed during the growing seasons (June-August) of 2005, and 2007-2012.

Lake or Pond (location)	Surface area (ha)	Mean depth (m)	Max. depth (m)	Elevation (m asl)	Primary water sources
Pond 01 (N81.822 W71.352)	0.1-0.7	0.2-0.6	0.5-1.3	166	LH, snowmelt
Pond 02 (N81.811W71.453)	0.2-3.4	0.1-0.4	0.3-1.2	165	LH, snowmelt
Pond 03 (N81.829 W71.462)	0.04	0.3	0.8	338	Snowmelt
Pond 07 (N81.835 W71.305)	0.4	0.1	0.3	184	Snowmelt
Pond 10 (N81.838 W71.343)	2.5	1.1	2.4	222	Snowmelt
Pond 11 (N81.832W71.466)	0.2	1.1	2.5	291	Snowmelt, ground ice
Pond 12 (N81.831W71.529)	0.2	0.8	1.9	370	Snowmelt
Pond 16 (N81.850W71.392)	0.7	1.1	2.1	434	Snowmelt, ground ice
Skeleton L. (N81.829W71.480)	1.9	1.9	4.7	299	Snowmelt, ground ice
LH-shore (N81.821 W71.352)	54,200	95[a]	267[a]	158	Glacial, snowmelt

[a]*Kock et al., 2012*

Table 2 Number of samples collected for both dissolved greenhouse gases and general chemical analyses within freshwater systems of the Lake Hazen watershed during the growing seasons (June-August) of 2005, and 2007 to 2012. All calculated gas fluxes were based on samples collected for concentration analyses.

Water body	2005	2007	2008	2009	2010	2011	2012
CO_2, CH_4 (*chemistry*)							
Pond 01	16	25	30*(5)*	24*(5)*	35*(5)*	3	-
Pond 02	16	1	5*(5)*	-	2*(2)*	3	-
Pond 03	1	1	1	-	1*(1)*	3	-
Pond 07	1	1	1	-	2*(2)*	3	-
Pond 10	1	-	-	-	2*(2)*	3	-
Pond 11	1	-	-	-	2*(2)*	6	3
Pond 12	1	1	1	-	2*(2)*	-	-
Pond 16	-	-	-	-	2*(2)*	3	-
Skeleton Lake	-	19	16*(5)*	23*(5)*	29*(4)*	6	3
Lake Hazen shoreline	17	27	30*(6)*	24*(5)*	29*(4)*	3	-

Table 3 Mean (±1SD) water temperature and general chemistry of different freshwater types, and other selected locations and periodsb in the Lake Hazen watershed during the growing seasons (June-Aguust) of 2005, 2007-2012. All measurements are in µmol L^{-1} except for water temperature (°C), total dissolved solids (mg L^{-1}) and chlorophyll-*a* (µg L^{-1}).

	W_T	TDS	PC	DIC	DOC	NO_3^-+NO_2^-	NH_4^+	TDN	TDP	Fe	SO_4^{2-}	Chl-a
Evaporative												
Pond 03	8	485	44	2,308	1,848	0.01	0.1	113	0.4	0.9	1,720	0.9
Pond 07	12±6	1,336±32	62±6	2,574±93	3,859±88	0.01±0.00	1.1±1.0	125±40	0.4±0.0	3.2±1.0	6,628±186	0.5±0.2
Pond 10	12±6	934±32	47±15	2,248±4	1,982±106	0.01±0.00	0.5±0.6	121±35	0.2±0.0	0.0±0.0	4,676±113	2.4±0.8
Pond 12	11±3	1,060±15	41±3	1,450±97	1,544±29	0.03±0.02	0.1±0.1	86±1	0.3±0.0	0.2±0.1	6,454±118	1.1±0.1
Mean±SD	**10±2**	**953±355**	**49±9**	**2,145±484**	**2,308±1,050**	**0.01±0.01**	**0.5±0.5**	**111±18**	**0.3±0.1**	**1.1±1.5**	**4,870±2278**	**1.2±0.8**
Meltwater												
Pond 11	12±2	451±24	29±11	1,453±30	383±12	0.03±0.02	0.3±0.4	20±2	0.2±0.0	0.0±0.0	2,232±52	0.6±0.2
Pond 16	11±5	328±12	18±3	939±4	554±18	0.01±0.00	0.3±0.3	24±0	0.2±0.0	0.1±0.1	1,885±49	0.3±0.1
Skeleton L.	11±4	317±115	23±9	1,533±241	447±63	0.02±0.01	2.4±2.3	22±2	0.2±0.0	0.0±0.0	1,669±392	0.5±0.4
Mean±SD	**11±0**	**365±75**	**24±6**	**1,308±323**	**461±86**	**0.02±0.01**	**1.0±1.2**	**22±2**	**0.2±0.0**	**0.1±0.0**	**1,928±284**	**0.5±0.1**
Melt. streams	*3*	*653*	*-*	*769*	*67*	*7.70*	*0.1*	*35*	*0.0*	*0.6*	*3,318*	*2.1*
Shoreline												
Pond 01	12±3	192±31	34±17	1,848±443	409±124	0.11±0.18	2.8±2.8	24±11	0.2±0.1	2.1±1.6	407±129	0.5±1.1
Pond 02	10±2	131±26	27±15	1,356±198	103±25	0.11±0.19	0.5±0.7	6±1	0.1±0.0	0.3±0.3	273±107	0.2±0.1
Mean±SD	**11±2**	**162±43**	**31±5**	**1,602±348**	**256±216**	**0.11±0.00**	**1.6±1.6**	**15±13**	**0.2±0.1**	**1.2±1.3**	**340±95**	**0.4±0.3**
Pre-flood	*14±3*	*216±56*	*34±4*	*1,740±243*	*497±115*	*0.01±0.00*	*2.2±2.8*	*27±4*	*0.3±0.0*	*1.7±0.7*	*608±231*	*0.4±0.2*
Post-flood	*11±2*	*164±40*	*32±18*	*1,681±470*	*270±172*	*0.13±0.19*	*2.0±2.5*	*16±13*	*0.2±0.1*	*1.5±1.7*	*311±102*	*0.5±1.0*
Lake Hazen shoreline												
Mean±SD	**5±3**	**59±68**	**10±5**	**524±301**	**51±123**	**0.24±0.18**	**1.8±2.3**	**2±1**	**0.1±0.0**	**0.0±0.0**	**69±42**	**0.1±0.1**

W_T: water temperature; TDS: total dissolved solids; PC: particulate carbon; DIC: dissolved inorganic carbon; DOC: dissolved organic carbon; NO_3^-+NO_2^-: dissolved nitrate + nitrite; NH_4^+: dissolved ammonium; TDN: total dissolved nitrogen; TDP: total dissolved phosphorus; Fe: dissolved iron; SO_4^{2-}: dissolved sulfate; chl-a: chlorophyll-a

Table 4 Comparison of the daily net exchange of carbon dioxide (CO_2) and methane (CH_4) between high Arctic terrestrial and freshwater ecosystems and the atmosphere in the Lake Hazen watershed during the growing seasons (June-August) of 2005 and 2007-2012. Positive values represent net emission of a gas to the atmosphere. Underlined values denote statistical differences of daily fluxes from other ecosystem types for each gas (linear mixed model; α=0.05; see Methods). The total and percent growing season exchange of each gas and ecosystem is also shown, as is the surface area of each ecosystem.

Ecosystem	CO_2 flux			CH_4 flux			Area	
	g C-CO_2 m^{-2} d^{-1}	Mg C-CO_2 season^{-1}	%	g C-CH_4 m^{-2} d^{-1}	Mg C-CH_4 season^{-1}	%	km^2	%
Aquatic								
Upland	+0.045±0.180	+598	4	+0.001±0.001	+11	2	144	2
Shoreline	+0.031±0.218	+2	0	+0.008±0.001	+0	0	1	0
Lake Hazen	-0.014±0.269	-721	5	+0.000±0.002	+6	1	542	7
Terrestrial[a]								
P. semidesert	+0.004+0.223	+1,253	9	-0.001±0.003	-412	94	3,819	51
M. wetland	-0.955±0.291	-11,368	82	+0.001±0.002	+10	2	129	2
Glacial ice	n/a	n/a	n/a	n/a	n/a	n/a	2,809	38
Totals	**-**	**-10,236**	**100**	**-**	**-385**	**100**	**7,443**	**100**

[a] *from Emmerton et al. 2014, 2016*

Figures

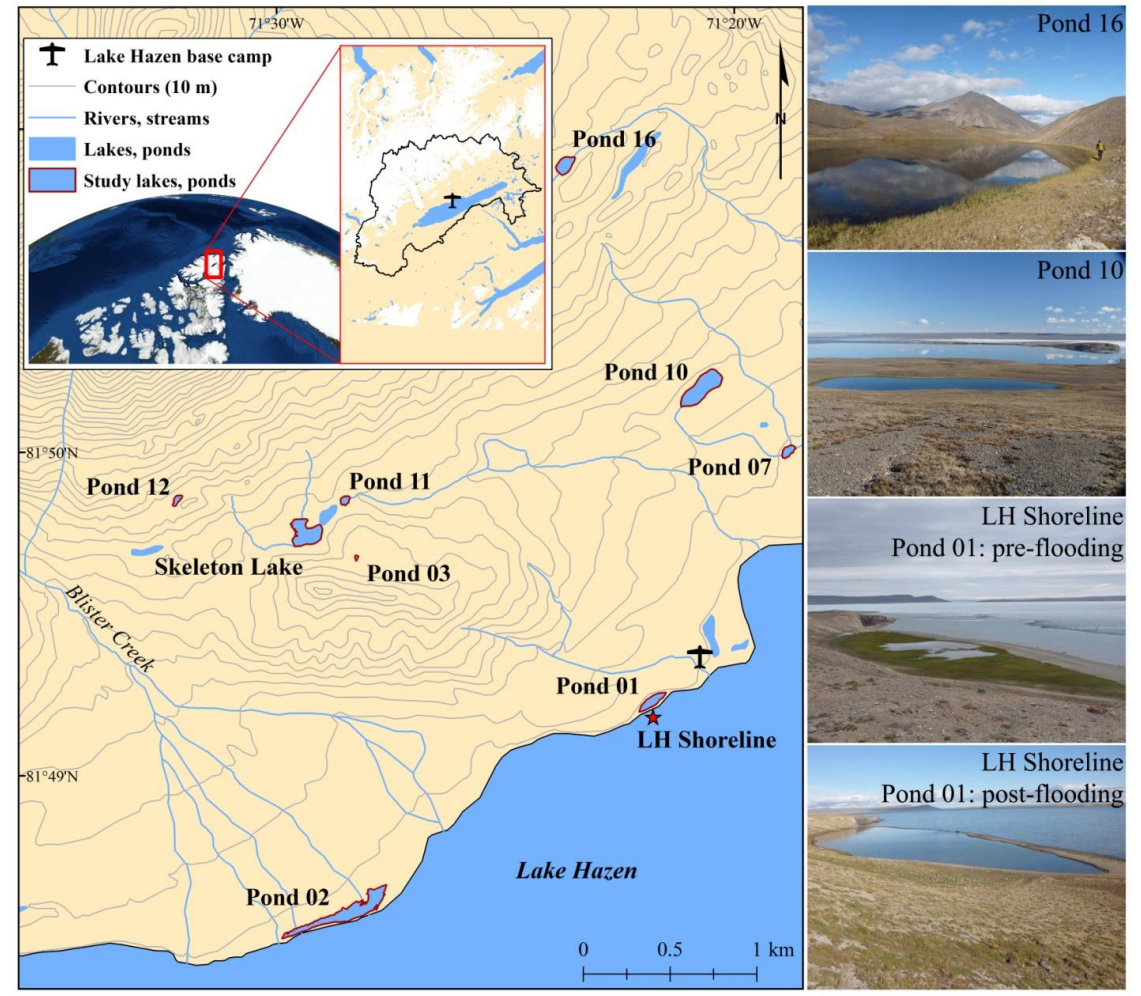

Figure 1 Map of the Lake Hazen base camp in Quttinirpaaq National Park, Nunavut, Canada. Ponds and lakes investigated in this study are indicated on the map and selected sites are shown in photographs. Shown inset are the general locators of the Lake Hazen watershed.

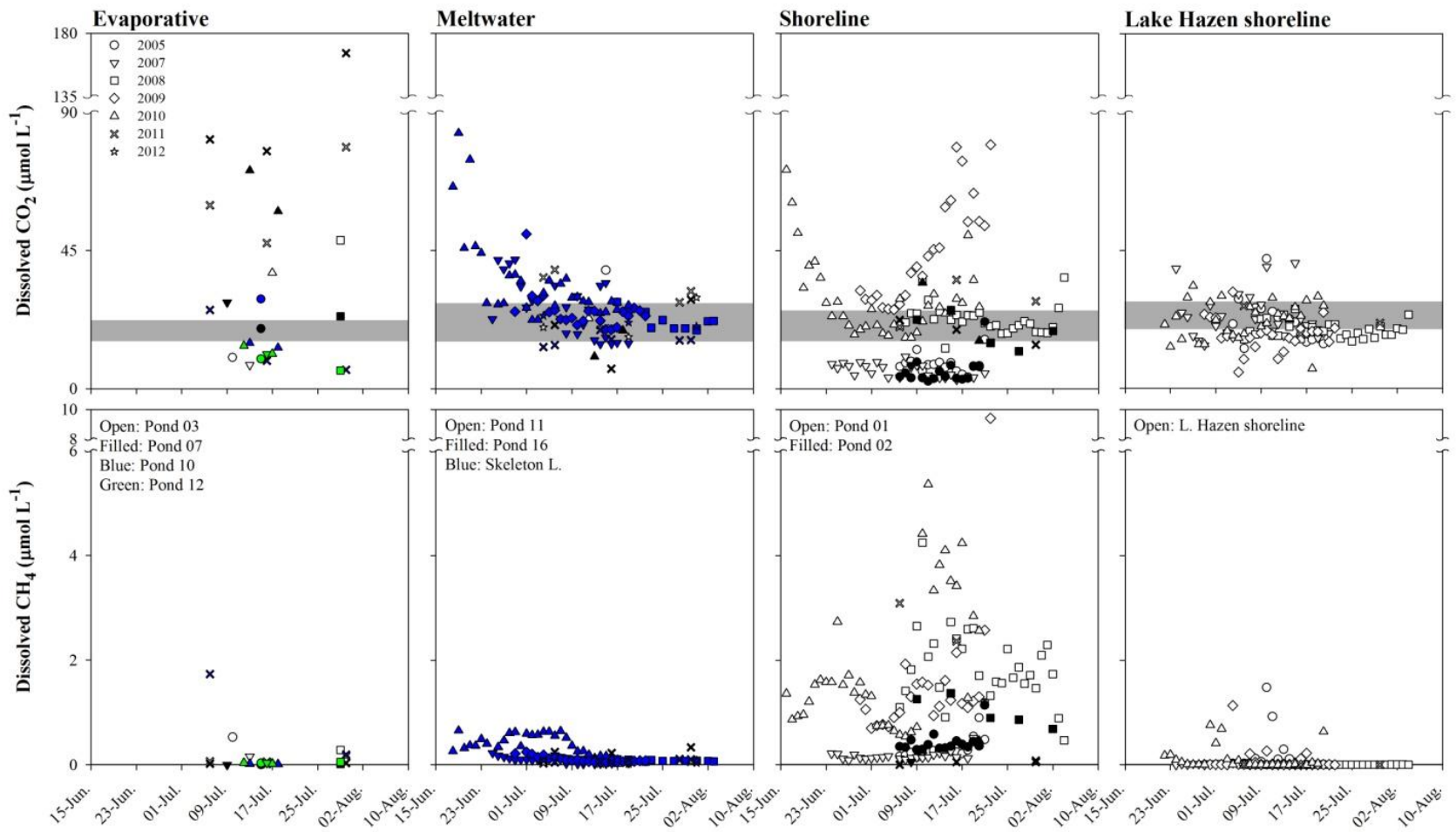

Figure 2 **Dissolved carbon dioxide (CO_2) and methane (CH_4) concentrations during the 2005, and 2007-2012 growing seasons (June-August) from different types of high Arctic freshwater systems in the Lake Hazen watershed. Inset text shows site names within each freshwater type. Grey areas indicate the range of atmospheric equilibrium concentrations CO_2 and CH_4 during the sampling period.**

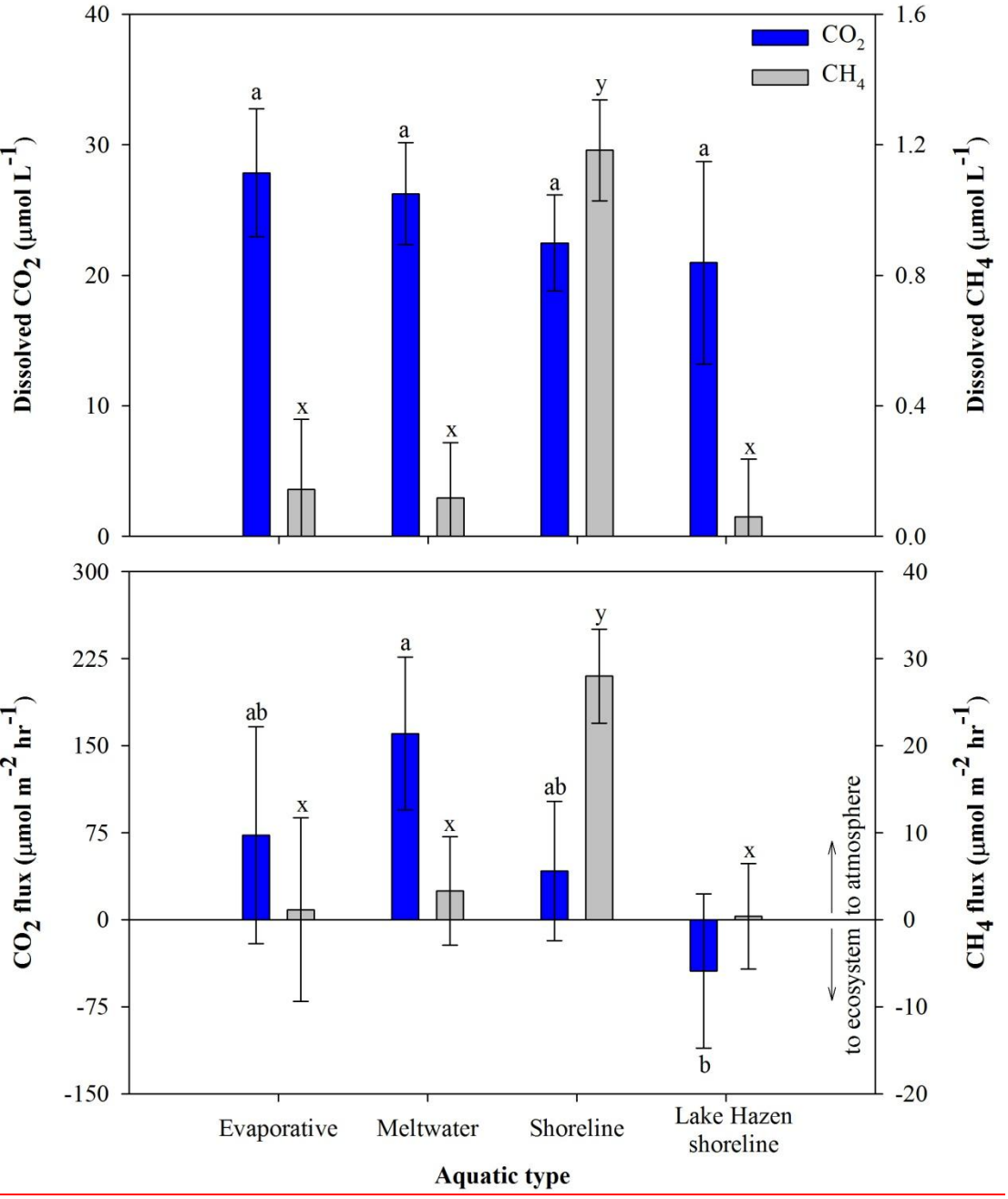

Figure 3 Mean (±SE) dissolved carbon dioxide (CO_2) and methane (CH_4) concentrations and fluxes during the 2005, and 2007-2012 growing seasons (June-August) from four different freshwater types in the Lake Hazen watershed. Letters denote statistical differences between ecosystem types for each gas (linear mixed model; α=0.05; see Methods).

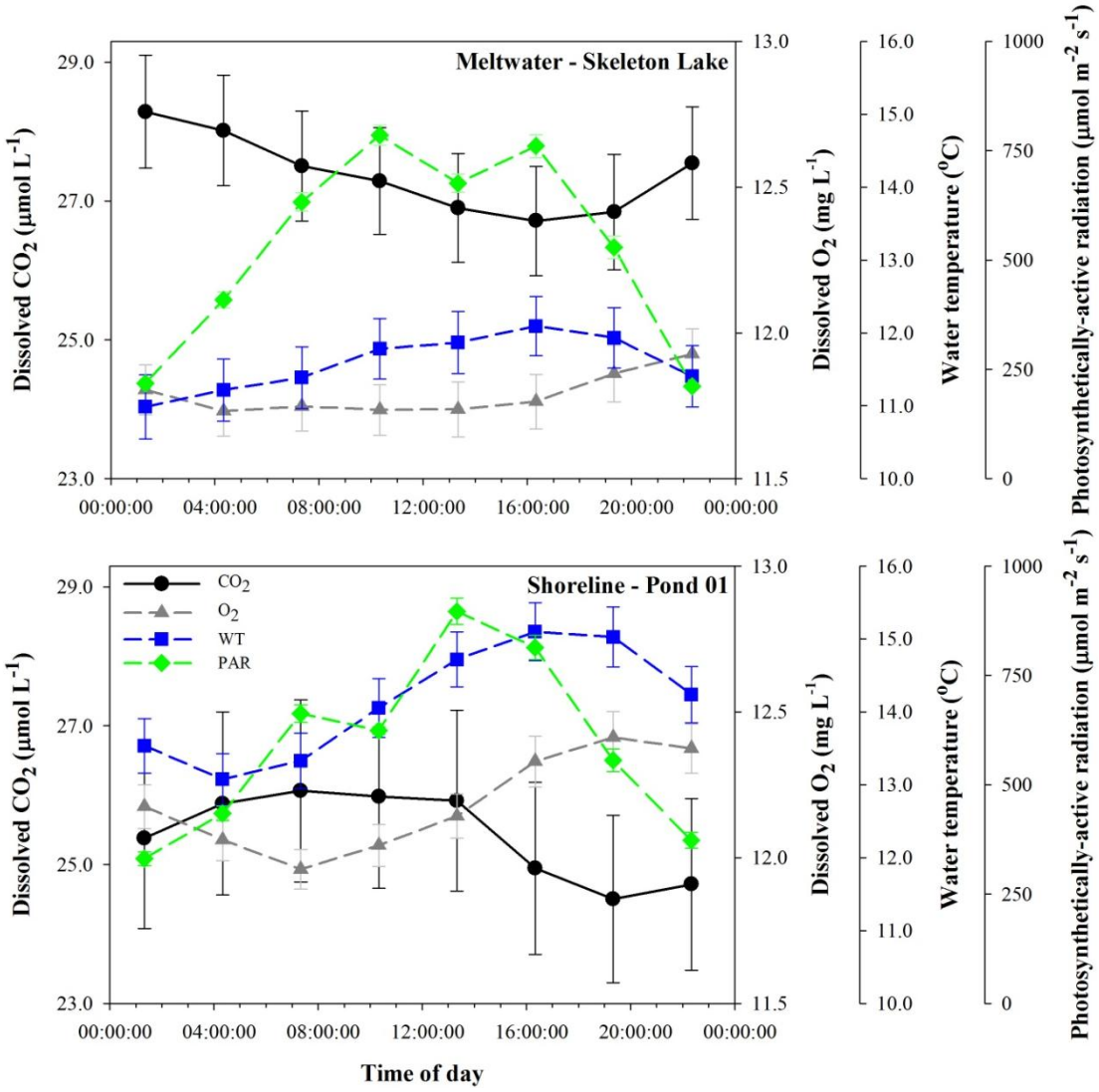

Figure 4 Three-hour diel dissolved carbon dioxide (CO_2) concentration, oxygen (O_2) concentration, water temperature and photosynthetically-active radiation (PAR) data measured by automated systems deployed at the shorelines of Skeleton Lake (2008-10) and Pond 01 (2008-10) during the high Arctic growing season (June-August) in the Lake Hazen watershed.