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### The importance of freshwater systems to the net atmospheric exchange of carbon dioxide and methane with a rapidly changing high Arctic watershed

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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<sub>2</sub>) and methane (CH<sub>4</sub>) 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<sub>2</sub> and CH<sub>4</sub> 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<sub>2</sub> concentrations (21–28  $\mu$ mol L<sup>-1</sup>) and atmospheric exchange (-0.013 to  $+0.046 \text{ g C}-\text{CO}_2 \text{ m}^{-2} \text{ day}^{-1}$ ) were similar between these systems. Seasonal flooding of ponds bordering Lake Hazen generated considerable CH4 emissions to the atmosphere  $(+0.008 \text{ g C}-\text{CH}_4 \text{ m}^{-2} \text{ day}^{-1})$ , while all other freshwater systems were minimal emitters of this gas (<+0.001 gC- $CH_4 m^{-2} day^{-1}$ ). 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<sub>2</sub> and CH<sub>4</sub> of underproductive but expansive polar semidesert ecosystems.

### 1 Introduction

Freshwater ecosystems cover less than 10% of global icefree 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 belowground stores of organic carbon (Tarnocai et al., 2009). These carbon- and lake-rich northern ecosystems, therefore, have been critically important sinks historically and will potentially be 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 carbon-rich waters can inhibit  $CO_2$  uptake by aquatic primary producers (Karlsson et al., 2009). Conversely, heterotrophic respiration by microbes 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<sub>2</sub> emissions to the atmosphere. Lakes in carbon-rich low Arctic regions ( $\sim 60-70^{\circ}$  N, AMAP, 1998) can account for more than three-quarters of a landscape's net CO<sub>2</sub> emissions to the atmosphere (Abnizova et al., 2012). At the same time, saturated peatlands and shallow ponds and lakes throughout much of the low Arctic can also be robust emitters of the potent GHG methane (CH<sub>4</sub>) to the atmosphere. Anoxic conditions in saturated, shallow, organic-rich soils have strong potential for methanogenic production and release of CH<sub>4</sub> into water (Tagesson et al., 2012). Due to its poor solubility, CH<sub>4</sub> can then be effectively released to the atmosphere from these ecosystems by ebullition and 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 (>  $\sim 70^{\circ}$  N; AMAP, 1998), lake abundance and area are dramatically reduced in 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 low Arctic, with mostly unknown implications for carbon GHG exchange in high Arctic lakes and ponds. Considering these challenging climatic 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 humaninduced 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 are 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., 2005) 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 impact 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<sup>6</sup> km<sup>2</sup>), 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 to 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 (81.8° N, 71.4° W), Canada's most northerly protected area (Fig. 1). Lake Hazen (area: 542 km<sup>2</sup>; max depth: 267 m) is the world's largest high Arctic lake and is surrounded by a substantial watershed (6901 km<sup>2</sup>) composed of carbonate, evaporite, and dolomite rock (Trettin, 1994) and cryosolic 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 in the Supplement). For example, mean July air temperature is typically 8–9°C at the base camp, compared to July 1981-2010 climate normals of 6.1 and 3.4 °C at the coastal Eureka and Alert weather stations on Ellesmere Island, respectively (Environment and Climate Change Canada, 2016a). 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 than surrounding areas (France, 1993), despite receiving only  $\sim$  34 mm of precipitation during the growing season (Table S1). Ultra-oligotrophic Lake Hazen itself dominates the freshwater area of the watershed



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. Inset shows the general locators of the Lake Hazen watershed.

(Keatley et al., 2007) and receives most of its water annually from rivers discharging meltwater 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 and small in area ( $\sim$  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 (Fig. 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 freshwaters, rather than on inter-annual biogeochemical trends within each system. Regardless, all sampling occurred during the summer growing seasons of 2005 to 2012 (except for 2006), between mid-June and early August (Tables 2, S2).

## 2.2 Dissolved CO<sub>2</sub> and CH<sub>4</sub> 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<sub>2</sub>) 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 anal-

Lake or pond (location)	Surface area (ha)	Mean depth (m)	Max depth (m)	Elevation (m a.s.l.)	Primary water sources
Pond 01 (N 81.822 W 71.352)	0.1-0.7	0.2-0.6	0.5-1.3	166	LH, snowmelt
Pond 02 (N 81.811 W 71.453)	0.2-3.4	0.1-0.4	0.3-1.2	165	LH, snowmelt
Pond 03 (N 81.829 W 71.462)	0.04	0.3	0.8	338	Snowmelt
Pond 07 (N 81.835 W 71.305)	0.4	0.1	0.3	184	Snowmelt
Pond 10 (N 81.838 W 71.343)	2.5	1.1	2.4	222	Snowmelt
Pond 11 (N 81.832 W 71.466)	0.2	1.1	2.5	291	Snowmelt, ground ice
Pond 12 (N 81.831 W 71.529)	0.2	0.8	1.9	370	Snowmelt
Pond 16 (N 81.850 W 71.392)	0.7	1.1	2.1	434	Snowmelt, ground ice
Skeleton L. (N 81.829 W 71.480)	1.9	1.9	4.7	299	Snowmelt, ground ice
LH shore (N 81.821 W 71.352)	54 200	95 <sup>a</sup>	267 <sup>a</sup>	158	Glacial, snowmelt

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

<sup>a</sup> 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 2007–2012. All calculated gas fluxes were based on samples collected for concentration analyses.

Water body	2005	2007	2008	2009	2010	2011	2012
CO <sub>2</sub> , CH <sub>4</sub> (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	-

ysed in the accredited Biogeochemical Analytical Service Laboratory (BASL). There, samples were placed in a wristaction shaker for 20 min to equilibrate dissolved CO2 and CH<sub>4</sub> with the N<sub>2</sub> headspace. Headspace CO<sub>2</sub> and CH<sub>4</sub> concentrations were quantified on a Varian 3800 gas chromatograph (GC) using a flame ionization detector at 250 °C with ultra-high-purity helium (He) as a carrier gas passing through a HayeSep D column at 80 °C. A ruthenium methanizer converted CO<sub>2</sub> to CH<sub>4</sub>. Four gas standards (Praxair, Linde-Union Carbide), ranging from 75 to 6000 parts per million for both CO<sub>2</sub> and CH<sub>4</sub>, 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-analysed every 10 samples to reconfirm the calibration, and duplicate injections were performed on all samples. Headspace CO2 and CH4 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  $0.5 \text{ mL H}_3\text{PO}_4$  to convert all DIC to CO<sub>2</sub> and then immediately re-analysed 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; Fig. 1; Table S2). Dissolved  $CO_2$  concentrations were measured every 3 h during several summers. These systems functioned by equilibrating, over a 20 min 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<sup>TM</sup> 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 a CS 014A anemometer (1 m height) and a Kipp & Zonen (Delft, the Netherlands) photosynthetically active radiation (PAR) LITE quantum sensor were mounted. All data were recorded on Campbell Scientific CR10X dataloggers.

# 2.3 Net atmospheric exchange of CO<sub>2</sub> and CH<sub>4</sub> with 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<sub>2</sub> and CH<sub>4</sub> mass fluxes between surface waters and the atmosphere at our remote location. This decision was made because 24 h daylight at our high-latitude location dampened diurnal surface temperature changes to less than 1 °C, and because of the general shallowness of the systems and the steady, sometimes gusty, wind conditions on site. The stagnant film model assumes that 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:

Gas flux 
$$\left(\mu \text{mol } \text{m}^{-2} \text{h}^{-1}\right) = k \left(C_{\text{SUR}} - C_{\text{EQL}}\right),$$
 (1)

where  $C_{SUR}$  (µmol L<sup>-1</sup>) is the concentration of the gas in surface waters,  $C_{\text{EOL}}$  (µmol L<sup>-1</sup>) is the atmospheric equilibrium concentration, and k is the gas exchange coefficient or the depth of water per unit time at which the concentration of the gas equalizes with the atmosphere (i.e. piston velocity). Values of  $k \pmod{h^{-1}}$  were calculated using the automated system's 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 CO2 and CH4 concentrations were quantified using Henry's law, in situ barometric pressure and air temperature, and mean annual CO<sub>2</sub> and CH<sub>4</sub> concentrations in the atmosphere during the year of sampling (Environment and Climate Change Canada, 2016b). If dissolved CO2 and CH<sub>4</sub> 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  $CO_2$  and  $CH_4$  to the atmosphere from two freshwater systems (Skeleton Lake, pond 01) during two growing seasons using manual bubble collection and GC analysis (see the Supplement).

#### 2.4 Supporting measurements

We quantified additional physical and chemical parameters in surface waters at the same sites where we collected our GHG samples, albeit at reduced sampling frequencies (Tables 2, S2). At each site, temperature, pH, specific conductivity, and dissolved  $O_2$  were measured in situ using a YSI (Yellow Springs, OH) 556 Multiprobe System (MPS). Water samples were also collected for general chemical analyses (total dissolved nitrogen (TDN), particulate nitrogen (PN), nitrate + nitrite  $(NO_3^- + NO_2^-)$ , ammonium  $(NH_4^+)$ , total phosphorus (TP), total dissolved phosphorus (TDP), alkalinity, dissolved organic carbon (DOC), total dissolved solids (TDS), 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  $\sim 5 \,^{\circ}$ C 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 chemical analyses (10 sites; n = 62; Table 2). Because sampling was unbalanced in frequency and time between sites due to logistical challenges (Table 2; see Sect. 2.1), potential overlap of chemistries between individual lakes was high, therefore setting a conservative standard for classifying distinct freshwater types. We used between-group linkage and squared Euclidean distances to group similar sites together and delineate distinct high Arctic freshwater classes. 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<sub>2</sub> and CH<sub>4</sub> with 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 2008–2012 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 ex-

**Table 3.** Mean ( $\pm 1$  SD) water temperature and general chemistry of different freshwater types, and other selected locations and periods in the Lake Hazen watershed during the growing seasons (June–August) of 2005 and 2007–2012. All measurements are in  $\mu$ mol L<sup>-1</sup> except for water temperature (°C), total dissolved solids (mg L<sup>-1</sup>), and chlorophyll *a* ( $\mu$ g L<sup>-1</sup>).

	$W_{\mathrm{T}}$	TDS	PC	DIC	DOC	$NO_{3}^{-} + NO_{2}^{-}$	$\mathrm{NH}_4^+$	TDN	TDP	Fe	$SO_{4}^{2-}$	Chl a
Evaporative												
Pond 03	8	485	44	2308	1848	0.01	0.1	113	0.4	0.9	1720	0.9
Pond 07	$12\pm 6$	$1336\pm32$	$62\pm 6$	$2574\pm93$	$3859\pm88$	$0.01\pm0.00$	$1.1\pm1.0$	$125\pm40$	$0.4\pm0.0$	$3.2\pm1.0$	$6628 \pm 186$	$0.5\pm0.2$
Pond 10	$12\pm 6$	$934\pm32$	$47\pm15$	$2248\pm 4$	$1982\pm106$	$0.01\pm0.00$	$0.5\pm0.6$	$121\pm35$	$0.2\pm0.0$	$0.0\pm0.0$	$4676 \pm 113$	$2.4\pm0.8$
Pond 12	$11 \pm 3$	$1060\pm15$	$41 \pm 3$	$1450\pm97$	$1544 \pm 29$	$0.03\pm0.02$	$0.1 \pm 0.1$	$86 \pm 1$	$0.3 \pm 0.0$	$0.2 \pm 0.1$	$6454 \pm 118$	$1.1 \pm 0.1$
Mean $\pm$ SD	$10\pm 2$	$953\pm355$	$49\pm9$	$2145\pm484$	$2308 \pm 1050$	$0.01\pm0.01$	$0.5\pm0.5$	$111\pm18$	$0.3\pm0.1$	$1.1\pm1.5$	$4870\pm2278$	$1.2\pm0.8$
Meltwater												
Pond 11	$12 \pm 2$	$451 \pm 24$	$29 \pm 11$	$1453 \pm 30$	$383 \pm 12$	$0.03\pm0.02$	$0.3 \pm 0.4$	$20 \pm 2$	$0.2 \pm 0.0$	$0.0 \pm 0.0$	$2232\pm52$	$0.6 \pm 0.2$
Pond 16	$11\pm5$	$328 \pm 12$	$18 \pm 3$	$939 \pm 4$	$554 \pm 18$	$0.01\pm0.00$	$0.3 \pm 0.3$	$24 \pm 0$	$0.2\pm0.0$	$0.1 \pm 0.1$	$1885\pm49$	$0.3 \pm 0.1$
Skeleton L.	$11 \pm 4$	$317 \pm 115$	$23 \pm 9$	$1533\pm241$	$447\pm 63$	$0.02\pm0.01$	$2.4 \pm 2.3$	$22 \pm 2$	$0.2\pm0.0$	$0.0 \pm 0.0$	$1669 \pm 392$	$0.5 \pm 0.4$
Mean $\pm$ SD	$11\pm0$	$365\pm75$	$24\pm 6$	$1308\pm323$	$461\pm86$	$0.02\pm0.01$	$1.0 \pm 1.2$	$22\pm 2$	$0.2\pm0.0$	$0.1\pm0.0$	$1928\pm284$	$0.5 \pm 0.1$
Melt. streams	3	653	-	769	67	7.70	0.1	35	0.0	0.6	3318	2.1
Shoreline												
Pond 01	$12\pm3$	$192 \pm 31$	$34\pm17$	$1848 \pm 443$	$409 \pm 124$	$0.11\pm0.18$	$2.8\pm2.8$	$24 \pm 11$	$0.2 \pm 0.1$	$2.1 \pm 1.6$	$407 \pm 129$	$0.5 \pm 1.1$
Pond 02	$10\pm 2$	$131 \pm 26$	$27\pm15$	$1356 \pm 198$	$103 \pm 25$	$0.11\pm0.19$	$0.5 \pm 0.7$	$6 \pm 1$	$0.1 \pm 0.0$	$0.3 \pm 0.3$	$273 \pm 107$	$0.2 \pm 0.1$
Mean $\pm$ SD	$11 \pm 2$	$162\pm43$	$31\pm5$	$1602\pm348$	$256\pm216$	$0.11\pm0.00$	$1.6 \pm 1.6$	$15 \pm 13$	$0.2 \pm 0.1$	$1.2 \pm 1.3$	$340\pm95$	$0.4 \pm 0.3$
Pre-flood	$14\pm3$	$216\pm56$	$34 \pm 4$	$1740\pm243$	$497 \pm 115$	$0.01\pm0.00$	$2.2\pm2.8$	$27\pm4$	$0.3\pm0.0$	$1.7\pm0.7$	$608\pm231$	$0.4 \pm 0.2$
Post-flood	$11\pm 2$	$164\pm40$	$32\pm18$	$1681\pm470$	$270\pm172$	$0.13\pm0.19$	$2.0\pm2.5$	$16\pm13$	$0.2\pm0.1$	$1.5\pm1.7$	$311\pm102$	$0.5\pm1.0$
Lake Hazen sho	oreline											
Mean $\pm$ SD	$5\pm3$	$59\pm68$	$10\pm 5$	$524\pm301$	$51\pm123$	$0.24\pm0.18$	$1.8\pm2.3$	$2\pm 1$	$0.1\pm0.0$	$0.0\pm0.0$	$69\pm42$	$0.1\pm0.1$

 $W_{\rm T}$ : water temperature; TDS: total dissolved solids; PC: particulate carbon; DIC: dissolved inorganic carbon; DOC: dissolved organic carbon; NO<sub>3</sub><sup>-</sup> + NO<sub>2</sub><sup>-</sup>: dissolved nitrate + nitrite; NH<sub>4</sub><sup>+</sup>: dissolved ammonium; TDN: total dissolved nitrogen; TDP: total dissolved phosphorus; Fe: dissolved iron; SO<sub>4</sub><sup>2-</sup>: dissolved sulfate; chl *a*: chlorophyll *a*.

change with terrestrial polar semidesert and meadow wetland landscapes. 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, USA). 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. Ecosystem data were compared using a linear mixed model similar to that used in the freshwater classification (see Section 2.5).

### 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; Fig. S1 in the Supplement; 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-H<sub>2</sub>O) in water taken from each aquatic system in July 2010 (Fig. S2). "Meltwater" systems, including ponds 11 and 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 Sect. 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 (Fig. 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 (Fig. S1). The flooding water from the "Lake Hazen shoreline" was cold and dilute in dissolved ions, organic matter, TDN, and chl *a* but considerably higher in  $NO_3^- + NO_2^-$  than other water bodies.

### 3.2 Dissolved concentrations and net atmospheric exchange of CO<sub>2</sub> and CH<sub>4</sub>

### 3.2.1 CO<sub>2</sub>

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

On average, evaporative ponds had the highest mean  $CO_2$ concentrations (mean  $\pm$  SE; 27.9  $\pm$  4.9  $\mu$ mol L<sup>-1</sup>) compared to other freshwater types (Fig. 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<sub>2</sub> concentrations were above atmospheric equilibrium concentration (Fig. 2), and therefore these ponds were sources of the gas to the atmosphere  $(+177 \pm 66 \,\mu\text{mol CO}_2 \,\text{m}^{-2} \,\text{h}^{-1};$  Fig. 3). The other evaporative ponds (ponds 10, 12) were deeper and had CO<sub>2</sub> concentrations that were typically near those of the atmosphere. This contributed to their near-zero exchange of CO<sub>2</sub> with the atmosphere  $(-5 \pm 17 \,\mu\text{mol CO}_2 \,\text{m}^{-2} \,\text{h}^{-1})$ . Together, dissolved CO<sub>2</sub> 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<sub>2</sub> to the atmosphere  $(+73 \pm 93 \,\mu\text{mol CO}_2 \,\text{m}^{-2} \,\text{h}^{-1};$  Fig. 3).

Meltwater systems had lower, but insignificantly different, CO<sub>2</sub> concentrations  $(26.2 \pm 3.9 \,\mu\text{mol}\,\text{L}^{-1})$  than evaporative ponds (Fig. 3). Meltwater systems showed only gradual declines of CO<sub>2</sub> concentrations through the summer, with strong consistency between sampling times and sites (Fig. 2). However, they emitted higher, though not significantly different, fluxes of CO<sub>2</sub> to the atmosphere overall  $(+160 \pm 66 \,\mu\text{mol CO}_2 \,\text{m}^{-2} \,\text{h}^{-1};$  Fig. 3) than the other freshwater classes. CO2 concentrations of meltwater systems correlated strongly and positively with CH<sub>4</sub> concentrations but negatively with DOC concentrations and measurements that were of high abundances in meltwater streams draining into the systems (e.g.  $SO_4^{2-}$ , TDN; Table 3, S4). Mean diurnal trends in CO<sub>2</sub> concentrations across all sampling years, as measured by the automated system at Skeleton Lake, showed that CO<sub>2</sub> and O<sub>2</sub> concentrations had little association together (Pearson correlation: r = -0.18, df = 7; p = 0.67), but CO<sub>2</sub> concentrations varied strongly and negatively with water temperature (r = -0.97, df = 7, p < 0.001; Fig. 4).

Mean CO<sub>2</sub> concentrations of shoreline ponds  $(22.5 \pm 3.7 \,\mu\text{mol}\,\text{L}^{-1}; \text{ Fig. 3})$  were similar to the other freshwater types, which obscured their considerable sea-

sonal changes within and between growing seasons. In 2005 and 2007, both pond 01 and pond 02 received little floodwater from Lake Hazen due to lower lake water levels (Fig. 2). These conditions resulted in dense wetland vegetation growth surrounding the ponds and low mean daily dissolved CO<sub>2</sub> concentrations  $(6.5 \pm 0.4 \,\mu\text{mol}\,\text{L}^{-1})$  and strong uptake of atmospheric CO<sub>2</sub>  $(-329 \pm 59 \,\mu\text{mol}\,\text{m}^{-2}\,\text{h}^{-1})$ . 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<sub>2</sub> from 2008 to 2011 of each pond together increased substantially  $(30.1 \pm 1.5 \,\mu\text{mol}\,\text{L}^{-1})$ , resulting in strong net emissions of CO<sub>2</sub> to the atmosphere  $(+228 \pm 44 \,\mu\text{mol}\,\text{m}^{-2}\,\text{h}^{-1})$ . Changing dissolved CO<sub>2</sub> concentrations correlated positively with dissolved nutrients and ions (Table S4). Diurnal trends of CO<sub>2</sub> and O<sub>2</sub> concentration measured by the automated system at pond 01 over several growing seasons showed opposite diel patterns of the gases, with greater O<sub>2</sub> concentrations during the warmest and brightest parts of the day (r = -0.98, df = 7, p < 0.001; Fig. 4). However, the net result of strong seasonality in these ponds was slight net emission of CO<sub>2</sub> to the atmosphere  $(+42\pm60\,\mu\text{mol}\,\text{m}^{-2}\,\text{h}^{-1};\text{ Fig. 3})$  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  $CO_2$  ( $21.0 \pm 7.8 \,\mu\text{mol}\,\text{L}^{-1}$ ; Fig. 2) with stable and low  $CO_2$  uptake throughout the season ( $-44 \pm 66 \,\mu\text{mol}\,\text{m}^{-2}\,\text{h}^{-1}$ ; Fig. 3).  $CO_2$  concentrations of this shoreline water correlated positively and most strongly with DIC,  $NO_3^- + NO_2^-$ , major ions, and wind speed (Table S4).

### 3.2.2 CH<sub>4</sub>

Each of the evaporative, meltwater, and Lake Hazen shoreline freshwaters had statistically similar and low CH<sub>4</sub> concentrations (0.06–0.14  $\mu$ mol L<sup>-1</sup>) and fluxes (0 to  $+3 \mu$ mol m<sup>-2</sup> h<sup>-1</sup>) across all growing seasons (Figs. 2, 3, S4, S5). Evaporative ponds had generally flat seasonal CH<sub>4</sub> concentration and flux trends (Figs. 2, S5), except for an outlier sample from pond 10 in mid-July 2011. CH<sub>4</sub> concentrations correlated strongest with  $NO_3^- + NO_2^-$  and alkalinity (Table S4). Meltwater systems were also generally low in CH<sub>4</sub> concentrations and fluxes through the summers and associated positively and closely with CO<sub>2</sub> 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<sub>2</sub> (Fig. S5). However, unlike CO<sub>2</sub>, higher CH<sub>4</sub> concentrations were sustained into July in Skeleton Lake in 2010



**Figure 2.** Dissolved carbon dioxide ( $CO_2$ ) and methane ( $CH_4$ ) concentrations during the 2005 and 2007–2012 growing seasons (June–August) of 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.

(Fig. 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<sub>4</sub> concentrations relative to the other systems  $(1.18 \pm 0.16 \,\mu\text{mol}\,\text{L}^{-1}; \text{Fig. 3})$  and showed a dynamic seasonal pattern dominated by the timing of flooding (Fig. 2). In 2005 and 2007 before substantial seasonal flooding started to occur, CH<sub>4</sub> concentrations  $(0.29 \pm 0.03 \,\mu\text{mol}\,\text{L}^{-1})$  and fluxes to the atmosphere  $(+8 \pm 2 \mu \text{mol m}^{-2} \text{ h}^{-1})$  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 (2008-2011; Table 3), CH<sub>4</sub> concentrations and fluxes increased substantially  $(1.70 \pm 0.13 \,\mu\text{mol}\,\text{L}^{-1})$ ;  $+41 \pm 10 \,\mu\text{mol}\,\text{m}^{-2}\,\text{h}^{-1}$ ) and correlated closely with dissolved organic and inorganic nitrogen (Table S4). This significant increase in CH<sub>4</sub> flux emissions from shoreline ponds during flooding (>5 times higher than during dry periods) was coupled with large increases in pond surface areas, effectively producing even higher total CH<sub>4</sub> 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<sub>4</sub> concentrations, but logistical constraints prevented later summer sampling to investigate if concentrations rebounded thereafter. Overall, aided by poor solubility of CH<sub>4</sub> in water and episodic wind events (Fig. S5), the flooding of shoreline ponds drove significantly larger CH<sub>4</sub> emissions to the atmosphere than other freshwater types  $(+28 \pm 5 \,\mu\text{mol}\,\text{m}^{-2}\,\text{h}^{-1};$  Fig. 3).

# 3.3 Net atmospheric exchange of CO<sub>2</sub> and CH<sub>4</sub> with a large high Arctic watershed

When scaled to total watershed area including Lake Hazen (7443 km<sup>2</sup>), polar semidesert landscapes were inconsequential to total CO<sub>2</sub> exchange (+1253 Mg C–CO<sub>2</sub>; 9% of total exchange) despite comprising a substantial proportion of the catchment (3819 km<sup>2</sup>; 51%; Table 4). All types of standing freshwaters sampled in the watershed from this study showed statistically similar CO<sub>2</sub> 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<sup>2</sup>; 7%) exchanged relatively little CO<sub>2</sub> with the atmosphere ( $-721 Mg C-CO_2$ ; 5%), as did smaller freshwater systems (144 km<sup>2</sup>; 2%) in the watershed (+600 Mg C–CO<sub>2</sub>; 4%). In clear contrast, during the growing season, moist and vegetated meadow wetland ecosystems were found to con-

**Table 4.** Comparison of the daily net exchange of carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>) 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;  $\alpha = 0.05$ ; 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. Measurements that were not available were assumed to be zero and are denoted by NA.

	CC	O <sub>2</sub> flux	CH	Area				
Ecosystem	$gC-CO_2$ $m^{-2} day^{-1}$	Mg C–CO <sub>2</sub> season <sup>-1</sup>	%	$g C-CH_4$ $m^{-2} day^{-1}$	Mg C–CH <sub>4</sub> season <sup>-1</sup>	%	km <sup>2</sup>	%
Aquatic								
Upland	$+0.045 \pm 0.180$	+598	4	$+0.001 \pm 0.001$	+11	2	144	2
Shoreline	$+0.031 \pm 0.218$	+2	0	$\pm 0.008 \pm 0.001$	+0	0	1	0
Lake Hazen	$-0.014 \pm 0.269$	-721	5	$+0.000\pm0.002$	+6	1	542	7
Terrestrial <sup>a</sup>								
Polar semidesert	$+0.004 \pm 0.223$	+1253	9	$-0.001 \pm 0.003$	-412	94	3819	51
Meadow wetland	$-0.955 \pm 0.291$	-11 368	82	$+0.001 \pm 0.002$	+10	2	129	2
Glacial ice	NA	NA	NA	NA	NA	NA	2809	38
Totals	-	-10236	100		-385	100	7443	100

<sup>a</sup> From Emmerton et al. (2014, 2016); slight discrepancies in values exist here compared to original publications due to data handling in the mixed model; see Methods.

sume CO<sub>2</sub> at rates similar to wetlands in the southern Arctic  $(-0.96 \text{ g C}-\text{CO}_2 \text{ m}^{-2} \text{ day}^{-1};$  Emmerton et al., 2016). Consequently, meadow wetlands exchanged an estimated 82 %  $(-11368 \text{ Mg C}-\text{CO}_2)$  of total CO<sub>2</sub> with the atmosphere despite occupying only 2 %  $(129 \text{ km}^2)$  of the area in the Lake Hazen watershed. Total CO<sub>2</sub> exchange of the watershed was  $-10236 \text{ Mg C}-\text{CO}_2 (-1.38 \text{ g C}-\text{CO}_2 \text{ m}^{-2})$  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- $CH_4 m^{-2} day^{-1}$ ; 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 the expansive coverage made the polar semidesert the key landscape controlling net CH<sub>4</sub> exchange throughout the Lake Hazen watershed ( $-412 \text{ Mg C-CH}_4$ ; 94 % of total exchange; Table 4). Surprisingly, a productive meadow wetland in the watershed was a weaker emitter of CH<sub>4</sub> to the atmosphere  $(+0.001 \text{ g C-CH}_4 \text{ m}^{-2} \text{ day}^{-1})$  than other high Arctic wetlands (Emmerton et al., 2014), releasing only 10 Mg C-CH4 (2%) to the atmosphere during the growing season. All upland freshwater systems (evaporative and meltwater systems) had low emissions of CH<sub>4</sub> to the atmosphere (11 Mg C-CH<sub>4</sub>; 2%), as did Lake Hazen itself (+6 Mg C-CH<sub>4</sub>; 1%). All measured ecosystems had statistically similar CH<sub>4</sub> fluxes except for the strong CH<sub>4</sub>-producing shoreline ponds (Table 4). However, poor areal coverage of these dynamic systems in the watershed  $(0.6 \text{ km}^2; < 1 \%)$  resulted in contributions of  $\ll 1 \%$  (+0.4 Mg C–CH<sub>4</sub>) of all CH<sub>4</sub> exchange in the Lake Hazen watershed (-385 Mg C-CH<sub>4</sub>; -0.052 g C- $CH_4 m^{-2}$ ).

### 4 Discussion

# 4.1 Dissolved concentrations and net atmospheric exchange of CO<sub>2</sub> and CH<sub>4</sub>

### 4.1.1 CO<sub>2</sub>

Dissolved CO<sub>2</sub> was likely being produced effectively in all evaporative ponds by ecosystem metabolism because of their high concentrations of DOC. These, and 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, highalkalinity environments  $(2-5 \text{ mEq } \text{L}^{-1})$  may have also contributed towards observed dissolved CO<sub>2</sub> concentrations in evaporative ponds (Trettin, 1994; Marcé et al., 2015). However, differences in pond volumes likely controlled the ultimate concentrations of CO<sub>2</sub> 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 to 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;



**Figure 3.** Mean ( $\pm$ SE) dissolved carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>) 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;  $\alpha = 0.05$ ; see Methods).

Karlsson et al., 2013). However, fluxes of CO<sub>2</sub> to the atmosphere from these systems did not correspond closely to early season venting, but rather to the frequency of episodic releases of  $CO_2$  to the atmosphere (Fig. 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; Fig. S6). Stratification would confine decomposition products (e.g. CO<sub>2</sub>, CH<sub>4</sub>) 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<sub>2</sub> and CH<sub>4</sub> concentrations in surface waters (Table S4). Results from our automated systems supported this argument as mean diurnal CO<sub>2</sub> and O<sub>2</sub> concentrations in surface waters of Skeleton Lake associated poorly and positively together, rather than negatively when metabolic processes (i.e. primary productivity or decomposition of organic matter; see pond 01 below) were dominant drivers in surface waters. 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<sub>2</sub> and DOC concentrations.

Shoreline ponds changed drastically in size and chemistry in response to seasonal flooding by Lake Hazen shoreline water (Tables 1, 3). During pre-flooding conditions,  $CO_2$ concentrations were low, which could be attributed to DIC use by autotrophic plankton (pre-flooding:  $1.2 \,\mu g \, L^{-1}$  chl *a*; post-flooding:  $0.4 \,\mu g \, L^{-1}$  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<sub>2</sub> 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 CO2 and O2 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<sub>2</sub> concentrations.

CO<sub>2</sub> concentrations in Lake Hazen shoreline water were near atmospheric equilibrium and only weakly consumed atmospheric CO<sub>2</sub>. 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<sup>-1</sup>; Keatley et al., 2007; Babaluk et al., 2009). CO<sub>2</sub> 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<sub>2</sub>, as observed elsewhere in glacial environments (Meire et al., 2015), and may explain the slight CO<sub>2</sub> uptake observed by the lake, especially later in summer.

### 4.1.2 CH<sub>4</sub>

Evaporative and meltwater systems were typically weak producers and emitters of CH<sub>4</sub>, 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<sub>2</sub>S 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  $+0.01 \text{ mg m}^{-2} \text{ day}^{-1}$ ; Table S5; see the Supplement). Stratification in meltwater systems likely also limited CH<sub>4</sub> emissions (Table S4). Low production and exchange of CH<sub>4</sub> in Lake Hazen, alternatively, were 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<sub>4</sub> 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 nearshore



**Figure 4.** Mean ( $\pm$ SE) of 3 h diel dissolved carbon dioxide (CO<sub>2</sub>) concentration, oxygen (O<sub>2</sub>) concentration, water temperature, and photosynthetically active radiation (PAR) data measured by automated systems deployed at the shorelines of Skeleton Lake (2008–2010) and pond 01 (2008–2010) during the high Arctic growing season (June–August) in the Lake Hazen watershed.

waters of Lake Hazen release CH<sub>4</sub> to the atmosphere above near-zero values.

Shoreline ponds were regional "hotspots" of CH<sub>4</sub> exchange, which was clearly driven by seasonal flooding (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. Methanogenesis may have been further enhanced by the flushing of shoreline ponds with  $SO_4^{2-}$ -poor Lake Hazen water, thus possibly favouring methanogenesis over  $SO_4^{2-}$  reduction in flooded soils.

## 4.2 Net atmospheric exchange of CO<sub>2</sub> and CH<sub>4</sub> with a large high Arctic watershed

Studies from the southern Arctic have estimated that fluxes of CO<sub>2</sub> (e.g. -1.55 to +1.10 g C–CO<sub>2</sub> m<sup>-2</sup> day<sup>-1</sup>; Tank et al., 2009; Abnizova 2012) and CH<sub>4</sub> (+0.01 to +0.09 g C–

 $CH_4 m^{-2} day^{-1}$ ; Walter, 2006; Sachs, 2010) from ponds and lakes can contribute a strong majority of a region's total exchange of CO<sub>2</sub> and CH<sub>4</sub> 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 been reported prior 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 % of the CO<sub>2</sub> (-0.01 to +0.05 g C-CO<sub>2</sub> m<sup>-2</sup> day<sup>-1</sup>) and 3% of the CH<sub>4</sub> (+0.00 to +0.01 g C-CH<sub>4</sub> m<sup>-2</sup> day<sup>-1</sup>) total watershed exchange of these two GHGs (Table 4). Several reasons may explain the limited role of freshwater 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 h 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 3 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 et al., 2011), exposure to these temperatures is short-lived and likely limits growing season autotrophic and heterotrophic activity and their contributions to freshwater carbon gas exchange. Geochemical production of CO2 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 through 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 lakebeds or submerged benthic mats of weaker productivity.

Despite a harsh climate and poor-quality substrates, our results suggest that the degree of moisture availability in high Arctic ecosystems was an overarching control on CO<sub>2</sub> 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 active layers above permafrost. Productive standing-water environments were rare in the Lake Hazen watershed, except for shoreline ponds during their drier wetland phase. However, the occasional late season flooding of these shoreline ponds with Lake Hazen waters promoted a near balance of net autotrophy and heterotrophy in these systems. For CH<sub>4</sub>, 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 their wetland vegetation, were substantially higher in per-unit CH<sub>4</sub> gas exchange than other ecosystems (Table 4). However, net uptake of CH<sub>4</sub> 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<sub>4</sub> sink (Jorgensen et al., 2015).

Modification of moisture availability in high Arctic regions is likely to occur 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 sitespecific hydrological conditions have important controls on the ultimate sustainability of high Arctic waters (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 to 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. For example, increased glacial melt would 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 and 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 occur would we expect more consistent sources of runoff to these 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 across other high Arctic regions is likely dependent on the trajectory of landscape change of polar semideserts (Sitch et al., 2007). Low CO<sub>2</sub> and CH<sub>4</sub> exchange in upland freshwater 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 meltwater, 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 become less frequent. Regardless, shoreline ponds likely have little role in regional carbon GHG exchange due to their minimal existence. Consequently, changes in the terrestrial ecosystems, over the longer term, should continue to define the direction and in-

tensity of GHG exchanges in the high Arctic. Meadow wetlands are key high Arctic regions due to substantial growing season productivity and CO<sub>2</sub> 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<sub>2</sub> 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 this globally important atmospheric CH<sub>4</sub> 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.

### 5 Data availability

The Polar Data Catalogue is a database of metadata and data that describes, indexes, and provides access to diverse data sets generated by Arctic and Antarctic researchers (http://www.polardata.ca/pdcsearch/ PDCSearchDOI.jsp?doi\_id=12712) (CCIN, 2016).

Third-party data were used in this paper from Environment and Climate Change Canada with references in the reference list of the paper and in the Supplement. This included publicly available climate normals from federal meteorological stations, river flow measurements, and atmospheric  $CO_2$  and  $CH_4$  concentrations.

## The Supplement related to this article is available online at doi:10.5194/bg-13-5849-2016-supplement.

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