



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

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10 Abstract. A warming climate is rapidly changing the distribution and exchanges of carbon within high Arctic 11 ecosystems. Few data exist, however, which quantify exchange of both carbon dioxide (CO₂) and methane (CH₄) 12 between the atmosphere and freshwater systems, or estimate freshwater contributions to total catchment exchange of 13 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 14 15 Hazen, Nunavut, Canada. We identified four types of biogeochemically-distinct freshwater systems in the watershed, however mean CO₂ concentrations (21–28 μ mol L⁻¹) and atmospheric exchange (-0.013–0.046 g C-CO₂ 16 m⁻² d⁻¹) were similar between these systems. Seasonal flooding of ponds bordering Lake Hazen generated 17 18 considerable CH₄ emissions to the atmosphere (0.008 g C-CH₄ m⁻² d⁻¹), while all other freshwater systems were minimal emitters of this gas (<0.001 g C-CH₄ m⁻² d⁻¹). Measurements made on terrestrial landscapes in the same 19 20 watershed between 2008-2012 determined that the near-barren polar semidesert was a very weak consumer of atmospheric CO₂ (-0.004 g C-CO₂ m⁻² d⁻¹), but an important consumer of atmospheric CH₄ (-0.001 g C-CH₄ m⁻² d⁻¹). 21 22 Alternatively, meadow wetlands were very productive consumers of atmospheric CO₂ (-0.96 g C-CO₂ $m^{-2} d^{-1}$) but relatively weak emitters of CH₄ to the atmosphere (0.001 g C-CH₄ m⁻² d⁻¹). When using ecosystem-cover 23 classification mapping, we found that freshwaters were unimportant contributors to total watershed carbon 24 25 exchange, in part because they covered less than 10% of total cover in the watershed. High Arctic watersheds are 26 experiencing warmer and wetter climates than in the past, which may have implications for the net uptake of carbon 27 greenhouse gases by currently underproductive polar semidesert and freshwater systems.

28 Keywords: carbon dioxide, methane, lake, pond, high Arctic, climate change, watershed





29 1 Introduction

30 Freshwater ecosystems cover less than 10% of global ice-free land area (Lehner and Doll, 2004) and have 31 been typically overlooked as substantial contributors to, or sinks of, atmospheric carbon greenhouse gases (GHGs; 32 Bastviken et al., 2011). However, recent studies suggest inland lakes collectively receive and process carbon at 33 magnitudes similar to oceanic uptake and sediment burial, making them important systems within the global carbon 34 cycle (Cole et al., 2007; Battin et al., 2009; Tranvik et al., 2009; Maberly et al., 2013; Raymond et al., 2013). Though these lowland systems efficiently accumulate allochthonous and autochthonous carbon, most natural lakes 35 36 and ponds, on balance, emit carbon GHGs to the atmosphere. For example, in most freshwater ecosystems, 37 decomposition continuously adds carbon dioxide (CO₂) to the water before venting to the atmosphere (Rautio et al., 38 2011), while uptake of CO_2 by autotrophs occurs typically over shorter seasonal periods (Cole et al., 2000; Huttunen 39 et al., 2003; Breton et al., 2009; Bastviken et al., 2011; Rautio et al., 2011; Callaghan et al., 2012). At the same time, 40 lake sediments and even oxic waters can sustain bacterial methanogenesis and the production of the potent GHG 41 methane (CH4; Bogard et al., 2014). Due to the gas's poor solubility in water, ebullition and wind can then 42 efficiently vent CH₄ to the atmosphere from these ecosystems, perhaps contributing up to 12% of global emissions 43 (Lai, 2009; Walter et al., 2006).

44 Lakes, ponds and wetlands are globally most abundant in northern regions, largely due to past periods of 45 glaciation and resulting land deformation. These freshwater environments may cover greater than half the landscape in northern regions, and can account for more than three-quarters of a landscape's net CO₂ exchange with the 46 47 atmosphere (Abnizova et al., 2012). Saturated northern peatlands can also be robust emitters of CH₄ because 48 permafrost impedes drainage of soils, promoting anoxia and methanogensis (Tagesson et al., 2012; Wik et al., 49 2016). However, at the highest northern latitudes (>70°N), polar semidesert landscapes not covered by glaciers 50 generally have cold, relatively well-drained soils (Campbell and Claridge, 1992) and receive little precipitation, 51 resulting in often less than 5% of the landscape being covered by aquatic systems. Though easy to overlook as 52 sparsely-vegetated barrens that exchange few carbon GHGs with the atmosphere (Soegaard et al., 2000; Lloyd, 53 2001; Lund et al., 2012, Lafleur et al., 2012), recent studies have shown that where conditions are ideal, high Arctic 54 ecosystems exchange GHGs at rates similar to ecosystems at more southerly latitudes (Emmerton et al., 2014; 55 Emmerton et al., 2016).





56 High Arctic ecosystem productivity is currently changing rapidly as a warming climate substantially alters 57 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 (>70°N; ACIA, 2004; IPCC, 2007b). Mean 58 59 annual precipitation is projected to increase ~12% for the Arctic as a whole over the same period, and up to 35% in 60 localized regions where the most warming will occur (ACIA, 2004; IPCC, 2007b). Such warming and wetting is 61 already modifying Arctic landscape energy balances (Froese et al., 2008) resulting in glacial melt (Pfeffer et al., 62 2008), permafrost thaw (Peterson et al., 2002), reorganized hydrological regimes (i.e., drying or wetting; Smith et 63 al., 2008) and extended growing seasons (Manabe et al., 1994). These changes are also perturbing watershed carbon 64 cycling through, for example, the liberation of carbon from thawing permafrost, and increases in biological 65 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). Considering the extensive cover of the near-barren polar semidesert in the high 66 67 Arctic $(>10^6 \text{ km}^2)$, these changes may have considerable effects on the future net exchange of carbon GHGs both 68 locally and on a global scale (Anthony et al., 2014). However, climate changes are far better delineated and 69 predicted for high Arctic landscapes in general than specifically for freshwater systems and landscape-scale GHG 70 exchange with the atmosphere. Therefore it is uncertain how rapid climate change will alter the cycling of carbon in 71 these remote regions.

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 recent studies of terrestrial GHG exchange from this remote polar semidesert biome.

75 2 Methods

76 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 (Figure 1). Lake Hazen (area: 542 km²; max. depth: 267 m) is the world's largest high Arctic lake, and is surrounded by a substantial watershed (6,901 km²). About 42% of the Lake Hazen watershed is glaciated with the balance of area covered by a polar semidesert (>90% 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; 92





83 days) 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 °C at the base camp, compared to July 1981-84 2010 climate normals of 6.1 °C and 3.4 °C at the coastal Eureka and Alert weather stations on Ellesmere Island, 85 86 respectively (Environment Canada, 2016). Soils in the region are also atypically warm during the summer because 87 of low moisture content and efficient radiative heating due to an abundance of clear-sky days. These conditions, 88 coupled with continuous daylight during the growing season, have resulted in a greater diversity and abundance of 89 vegetation and wildlife in the Lake Hazen watershed compared to surrounding areas (France, 1993), despite 90 receiving only ~34 mm of precipitation during the growing season (Table S1). Ultra-oligotrophic Lake Hazen itself 91 dominates the freshwater area of the watershed (Keatley et al., 2007) and receives most of its water annually from 92 rivers discharging melt water from glaciers. Water exits Lake Hazen via the Ruggles River. Ice-cover can remain on 93 Lake Hazen throughout the growing season, though in recent years the lake has gone ice-free more frequently, 94 usually by late July. Ponds and a few small lakes are scattered throughout the lower watershed and are mostly 95 shallow, 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 ponds or small lakes to sample within walking distance of base camp in the northwestern portion of the Lake Hazen watershed (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 just offshore in Lake Hazen to obtain water representative of that which interacted with ponds located along its shoreline. All sampling occurred during the summer growing seasons of 2005 to 2012 (except for 2006), between mid-June and early August (Table S2).

103 2.2 Dissolved CO₂ and CH₄ 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 (N₂) 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





111 Alberta, where they were analyzed in the accredited Biogeochemical Analytical Service Laboratory (BASL). There, 112 samples were placed in a wrist-action shaker for 20 minutes to equilibrate dissolved CO₂ and CH₄ with the N₂ 113 headspace. Headspace CO₂ and CH₄ concentrations were quantified on a Varian 3800 gas chromatograph (GC) 114 using a flame ionization detector at 250° C with ultra high purity hydrogen (H₂) as a carrier gas passing through a 115 hayesep D column at 80°C. A ruthenium methanizer converted CO2 to CH4. Four gas standards (Praxair, Linde-116 Union Carbide), ranging from 75 to 6000 ppm for both CO₂ and CH₄, 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. 117 A standard was re-analyzed every 10 samples to reconfirm the calibration, and duplicate injections were performed 118 119 on all samples. Headspace CO₂ and CH₄ concentrations were converted to dissolved molar concentrations using 120 Henry's Law, and corrected for temperature and barometric pressure differences between sample collection and 121 analysis. To quantify dissolved inorganic carbon (DIC) concentrations, samples were acidified with 0.5 mL H_3PO_4 122 to convert all DIC to CO₂, and then immediately reanalyzed on the GC. DIC concentrations were calculated as 123 above.

124 The second approach involved two automated systems to determine detailed diel changes in surface water 125 dissolved CO₂ concentrations at two different sites (Skeleton Lake and Pond 01; Figure 1; Table S2). Dissolved CO₂ 126 concentrations were measured every three hours during several summers. These systems functioned by equilibrating, 127 over a 20-minute period, dissolved CO_2 from pumped surface waters, with a gas cell in a Celgard MiniModule 128 Liqui-Cel. The equilibrated gas was then analysed for CO₂ concentration by a LI-COR (Lincoln, NE) 820 infrared gas analyzer. The systems also measured dissolved oxygen (O_2) concentrations using a QubitTM flow-through 129 130 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 131 132 watertight cases along the shore from which a sample line extended out into the surface waters, and upon which was 133 mounted a CS 014A anemometer (1 m height) and a Kipp & Zonen (Delft, The Netherlands) photosynthetically-134 active radiation (PAR) LITE quantum sensor. All data were recorded on Campbell Scientific CR10X dataloggers.

135 2.3 Dissolved CO₂ and CH₄ concentrations 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 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:

144 Gas flux (
$$\mu$$
mol m⁻² hr⁻¹) = k(C_{SUR} - C_{EOL}) (1)

where C_{SUR} (µmol L⁻¹) is the concentration of the gas in surface waters, C_{EOL} (µmol L⁻¹) is the atmospheric 145 equilibrium concentration, and k is the gas exchange coefficient, or the depth of water per unit time in which the 146 147 concentration of the gas equalizes with the atmosphere (i.e., piston velocity). Values of k (cm h^{-1}) were calculated 148 using automated systems wind measurements and occasionally from nearby (within 2 km) eddy covariance towers 149 (Campbell Scientific CSAT3 Sonic Anemometers; 30 min. means), and published empirical relationships (Table S3; 150 Hamilton et al., 1994). To determine the direction of the flux, atmospheric equilibrium CO₂ and CH₄ concentrations 151 were quantified using Henry's law, in-situ barometric pressure and air temperature, and mean annual CO2 and CH4 152 concentrations in the atmosphere during the year of sampling (Environment Canada, 2015). If dissolved CO₂ and 153 CH_4 concentrations in surface waters were above or below their corresponding calculated atmospheric equilibrium 154 concentrations, the freshwater systems were considered a source (+) or sink (-) relative to the atmosphere, 155 respectively.

We also measured ebullition fluxes of CH₄ 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).

159 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 S2). At each site, temperature, pH, specific conductivity and dissolved O_2 were measured in-situ using a YSI (Yellow Springs, OH) 556 MPS multiprobe. Water samples were also collected for general chemical analyses (total dissolved nitrogen [TDN], particulate N, NO₃⁻⁺+NO₂⁻, NH₄⁺, total phosphorus [P], total dissolved P [TDP], alkalinity, dissolved organic carbon [DOC], total dissolved solids, major cations/anions, chlorophyll-a [chl-a]) into pre-cleaned HDPE bottles. These samples





- 166 were immediately processed in the Lake Hazen/Quttinirpaaq Field Laboratory clean room after water collection, and
- 167 stored in the dark at ~5°C or frozen until analysed at the BASL.

168 2.5 Net atmospheric exchange of CO₂ and CH₄ of a large high Arctic watershed

169 To better understand the role of freshwater ecosystems in regional fluxes of carbon GHGs, freshwater CO₂ 170 and CH₄ fluxes measured in this study were coupled with terrestrial fluxes measured in the watershed during the 171 2008-12 growing seasons (Emmerton et al., 2014; Emmerton et al., 2016). Areal coverage of the different ecosystem 172 types in the watershed was isolated from a previous classification of Quttinirpaaq National Park (Edlund, 1994) 173 using a Geographical Information System (ArcGIS v.10.3; ESRI, Redlands, US). Mean growing season fluxes from 174 each measured ecosystem were then weighted to matching coverage area in the watershed to estimate the total 175 carbon gas exchange with the atmosphere. Glacial ice was assumed to be a net-zero contributor of total watershed 176 gas exchange in this scaling exercise.

177 3 Results

178 **3.1 Biogeochemical classification of high Arctic ponds**

179 Four distinct types of freshwater systems were evident from our sampling in the Lake Hazen watershed 180 (Table 2; Figure 2; hierarchical cluster analysis; see Supporting Information). "Evaporative" ponds (Ponds 07, 10, 181 12) occurred in the upland of the Lake Hazen catchment and were hydrologically-isolated from their surrounding 182 basins post-snowmelt. These ponds were relatively high in concentrations of total dissolved solids, most measured 183 ions, DIC, DOC, organic particles, TDP and chl-a. Pond 03, though not technically clustered with others, was forced 184 to the Evaporative pond category based on lack of consistent inflowing water and high concentrations of most 185 dissolved ions. This delegation was further consistent with isotopic measurements of oxygen ($\delta^{18}O-H_2O$) in water 186 taken from each aquatic system in July 2010 (Figure S2). "Meltwater" systems, including Ponds 11, 16 and Skeleton 187 Lake, also occurred in the upland of the Lake Hazen watershed, but received consistent water supply through the 188 growing season primarily from snowmelt, permafrost thaw water and/or upstream lake drainage. The general 189 chemistry of these systems was therefore consistent and without extremes during the growing season. Typical 190 meltwater streams draining to these ponds were high in TDN and sulfate $(SO_4^{2-}; Table 2)$. "Shoreline" ponds (Ponds 191 01, 02) occurred along the margin of Lake Hazen and were typically physically isolated from the large lake by 192 porous gravel berms, and surrounded by wetland soils and flora during spring low water conditions. As glacial melt





193 accelerated throughout the growing season, though, the water level of Lake Hazen rose and could seep through the 194 berms to incrementally flood the ponds and surrounding wetlands (Figure S3). Shoreline ponds changed chemically 195 during the onset of flooding as indicated, for example, by an increase in the concentration of reduced ions (i.e., NH₄⁺; Table S4). A separate smaller cluster of Pond 01 samples occurred during particularly high-water periods 196 197 when Lake Hazen breached the berms (Figure 2). The flooding water from the "Lake Hazen shoreline" was cold, 198 dilute in dissolved ions, organic matter, TDN, and chl-a, but considerably higher in NO₃⁻ compared to other water 199 bodies. A single sample from a pre-flooded Shoreline pond (Pond 02) grouped within the Lake Hazen shoreline 200 cluster likely because its water was ultimately sourced from Lake Hazen and because of its isolation from its 201 wetland margins during low-water conditions.

202 **3.2** Dissolved concentrations and net atmospheric exchange of CO₂ and CH₄ of high Arctic freshwaters

203 3.2.1 CO₂

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 3, 4; linear mixed-model; α =0.05; see Supporting Information).

On average, Evaporative ponds had the highest mean CO_2 concentrations (mean±SE; 27.9±4.9 µmol L⁻¹) 207 208 compared to other pond types, primarily due to conditions in Pond 03 and Pond 07. These ponds were the shallowest 209 of the four sampled and were rich in reduced ions, DIC, DOC, total P and calcium. CO₂ concentrations were above atmospheric equilibrium concentration and therefore these ponds were sources of the gas to the atmosphere 210 211 (+177 \pm 66 µmol CO₂ m⁻² hr⁻¹; Figure S4). The other Evaporative ponds (Ponds 10, 12) were deeper and had CO₂ 212 concentrations that were typically near those of the atmosphere. This contributed to their near-zero exchange of CO₂ with the atmosphere (-5 \pm 17 µmol CO₂ m⁻² h⁻¹). When combining all Evaporative ponds together, they were net 213 sources of CO₂ to the atmosphere ($+73\pm93 \mu$ mol CO₂ m⁻² hr⁻¹). 214

215 Meltwater systems had lower, but insignificantly different, CO_2 concentrations (26.2±3.9 µmol L⁻¹) than 216 Evaporative ponds. Meltwater systems showed only gradual, venting-related declines of CO_2 concentrations through 217 the summer, with strong consistency in concentrations between sampling times and sites. However, they emitted 218 higher, though not significantly different, fluxes of CO_2 to the atmosphere overall (+160±66 µmol m⁻² hr⁻¹; Figure 4) 219 compared to the other types of systems. CO_2 concentrations of these systems correlated strongly and positively with 220 CH_4 concentrations, but negatively with other measurements that were of high concentrations in meltwater streams





draining into the systems (e.g., SO_4^{2-} , TDN; Table 2, 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 associated positively together, but negatively with water temperature (Figure 5).

Mean CO₂ concentrations of Shoreline ponds (22.5 \pm 3.7 µmol L⁻¹) were similar to the other pond types, 224 225 which obscured their considerable seasonal changes within and between growing seasons. From 2005 to 2007, both 226 Pond 01 and Pond 02 received little floodwater from Lake Hazen due to lower lake water levels. These conditions resulted in dense wetland vegetation growth surrounding the ponds and low mean daily dissolved CO₂ 227 228 concentrations ($6.5\pm0.4 \,\mu\text{mol L}^{-1}$) and strong uptake of atmospheric CO₂ (-329±59 $\mu\text{mol m}^{-2} \,\text{hr}^{-1}$). The drier wetland 229 state of these ponds changed in following summers when Lake Hazen rose substantially upon greater inputs of 230 glacial meltwaters (WSC, 2015), causing the rising waters to seep through porous berms into the ponds through 231 July. In concert with flooding, concentrations of CO₂ from 2008-11 of each pond together increased substantially 232 $(30.1\pm1.5 \text{ }\mu\text{mol } \text{L}^{-1})$ resulting in strong net emissions of CO₂ to the atmosphere (+228±44 $\mu\text{mol } \text{m}^{-2} \text{ } \text{hr}^{-1})$. CO₂ concentrations of the ponds correlated strongly and positively with concentrations of many constituents in the same 233 234 waters (Table S4). Diurnal trends of CO₂ and O₂ concentration measured by the automated system at Pond 01 over 235 several growing seasons showed a primary production signature with opposite temporal patterns of the gases, with 236 greater O_2 during the warmest and lightest parts of the day (Figure 5). However, the net result of strong seasonality 237 in these ponds was slight net emission of CO_2 to the atmosphere (+42±60 µmol m⁻² hr) that was not statistically-238 different from other types of systems.

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) with stable and low CO_2 uptake throughout the season (-44±66 µmol m⁻² hr). CO_2 concentrations of this shoreline water related strongest and positively with DIC, major ions and wind speed (Table S4).

244 3.2.2 CH₄

Each of Evaporative, Meltwater and Lake Hazen shoreline freshwaters had statistically similar and low CH₄ concentrations (0.06-0.14 μ mol L⁻¹) and fluxes (0-3 μ mol m⁻² hr⁻¹) across all growing seasons (Figure 3,4, S4). Evaporative ponds had generally flat seasonal CH₄ concentration and flux trends, except for an outlier sample from Pond 10 in mid July 2011. Meltwater systems were also generally low in CH₄ concentrations and fluxes through the





summers and associated strongly with similar chemical measures as CO_2 (Table S4). Notable flux emissions from these systems only occurred during episodic wind events, also similar to CO_2 (Figure S4). However, unlike CO_2 , higher CH_4 concentrations were sustained into July in Skeleton Lake in 2010. 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 and strongly with particulate carbon concentrations (Table S4).

255 Shoreline ponds, alternatively, had significantly higher CH_4 concentrations relative to the other systems $(1.18\pm0.16 \text{ }\mu\text{mol }L^{-1})$ and showed a dynamic seasonal pattern dominated by the timing of flooding. In 2005 and 256 257 2007 before substantial seasonal flooding started to occur, CH_4 concentrations (0.29±0.03 µmol L⁻¹) and fluxes to the atmosphere ($8\pm2 \mu mol m^{-2} hr^{-1}$) were low. As the Shoreline ponds began to receive NO₃-rich flood water from 258 259 Lake Hazen by mid-summer in subsequent years (Table S4), 2008-11 CH₄ concentrations and fluxes increased substantially (1.70±0.13 μ mol L⁻¹; 41±10 μ mol m⁻² hr⁻¹). This significant increase in CH₄ flux emissions from 260 Shoreline ponds during flooding (>five times higher than during dry periods) was coupled with large increases in 261 262 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 263 264 resulting in rapid dilution of CH₄ concentrations, but logistical constraints prevented later summer sampling to 265 investigate if concentrations rebounded thereafter. Overall, aided by poor solubility of CH4 in water and episodic 266 wind events (Figure S4), the flooding of Shoreline ponds drove significantly larger CH₄ emissions to the atmosphere compared to other pond types (+28 \pm 5 µmol m⁻² hr⁻¹; Figure 4). 267

268 **3.3** Net atmospheric exchange of CO₂ and CH₄ of a large high Arctic watershed

Emmerton et al. (2014; 2016) measured, using eddy covariance flux towers (CO₂, CH₄) and static chambers 269 270 (CH_4) , growing season carbon GHG exchange with terrestrial polar semidesert and meadow wetland landscapes 271 from 2008-12. They found that the dry and mostly barren polar semidesert was among the most unproductive terrestrial ecosystems on Earth, taking up only -0.004 g C-CO₂ m⁻² d⁻¹ during the growing season, similar to findings 272 273 from other studies (Soegaard et al., 2000; Lloyd, 2001; Lund et al., 2012). When scaled to total watershed area 274 including Lake Hazen (7,443 km²), polar semidesert landscapes were inconsequential to total CO₂ exchange (-1,253 Mg C-CO₂; 9% of total exchange) despite comprising a substantial proportion of the catchment (3,819 km²; 51%; 275 276 Figure 6). All types of standing freshwaters sampled in the watershed from this study showed statistically-similar





277 CO_2 fluxes compared to the polar semidesert. When assuming its shoreline waters were representative of the entire 278 lake area, the expansive Lake Hazen (542 km²; 7%) exchanged relatively little CO₂ with the atmosphere (-721 Mg C-CO₂; 5%), as did smaller freshwater systems (145 km²; 2%) in the watershed (+600 Mg C-CO₂; 4%). In clear 279 contrast, during the growing season, moist and vegetated meadow wetland ecosystems were found to consume CO2 280 281 at rates similar to wetlands in the southern Arctic (-0.96 g C-CO₂ m⁻² d⁻¹). Consequently, meadow wetlands 282 exchanged an estimated 82% (-11,368 Mg C-CO₂) of total CO₂ with the atmosphere despite occupying only 2% (129 km²) of the area in the Lake Hazen watershed. Total CO₂ exchange of the watershed was -10,236 Mg C-CO₂ (-283 284 $1.375 \text{ g C-CO}_2 \text{ m}^{-2}$) during the growing season.

285 The high Arctic polar semidesert has recently gained attention as a notable atmospheric sink of CH_4 (-0.001 g C-CH₄ m⁻² d⁻¹; Emmerton et al., 2014), which has since been observed in studies at other high Arctic locations 286 287 (e.g., Jorgensen et al., 2015). These uptake fluxes coupled with its expansive coverage made the polar semidesert the 288 key landscape controlling net CH₄ exchange throughout the Lake Hazen watershed (-412 Mg C-CH₄; 94% of total exchange; Figure 6). Surprisingly, a productive meadow wetland in the watershed was a weaker emitter of CH₄ to 289 the atmosphere (+0.001 g C-CH₄ m⁻² d⁻¹) compared to other high Arctic wetlands (Emmerton et al., 2014), releasing 290 291 only 10 Mg C-CH₄ (2%) to the atmosphere during the growing season. All upland freshwater systems (Evaporative 292 + Meltwater) had low emissions of CH₄ to the atmosphere (11 Mg C-CH₄; 2%), as did Lake Hazen itself (+6 Mg C-293 CH₄; 1%). All measured ecosystems had statistically-similar CH₄ fluxes except for the strong CH₄-producing 294 Shoreline ponds (Figure 6). However, poor areal coverage of these dynamic systems in the watershed (0.6 km²; 295 <1%) resulted in contributions of <<1% (+0.4 Mg C-CH₄) of all CH₄ exchange in the Lake Hazen watershed (-385 296 Mg C-CH₄; -0.052 g C-CH₄ m⁻²).

297 4 Discussion

298 4.1 Dissolved concentrations and net atmospheric exchange of CO₂ and CH₄ of high Arctic freshwaters

299 4.1.1 CO₂

300 Concentrations of CO_2 and other compounds were highest in small and shallow Evaporative ponds (Ponds 301 03, 07) compared with those that were larger and deeper (Ponds 10, 12). Dissolved CO_2 was likely being produced 302 effectively in all Evaporative ponds by considerable ecosystem metabolism and accumulation and dissociation of 303 weathered carbonates and evaporates (Trettin, 1994; Marcé et al., 2015). However, CO_2 was likely more effectively





304 diluted in the larger ponds and therefore less susceptible to wind-related turbulence and gas exchange with the 305 atmosphere. Meltwater systems showed steady biogeochemical conditions but similar CO₂ concentrations as other freshwater types, despite inclusion of early summer sampling at Skeleton Lake (2007, 2010). High CO₂ 306 307 concentrations in Skeleton Lake during that time were typical of post-ice covered waters only beginning to re-308 equilibrate with the atmosphere (Kling et al., 1992; Karlsson et al., 2013). Greater exchange of CO₂ by Meltwater 309 systems, however, was not primarily driven by early season venting or sustained exchanges compared to other 310 ponds, but rather by higher frequency of episodic releases of CO₂ to the atmosphere (Figure S4). This may have 311 been related to their greater mean depths, which promoted stratification in at least one of our sampled Meltwater 312 systems (Skeleton Lake; Figure S5). Stratification would confine decomposition products (e.g., CO₂, CH₄) to near 313 their sites of origin in bottom sediments and extensive benthic mat communities (Rautio et al., 2011), which would 314 then be released most readily during and after wind mixing events. We observed evidence of this process via strong 315 positive associations between CO₂ and CH₄ concentrations in surface waters (Table S4). Further, mean diurnal CO₂ and O₂ concentrations in surface waters trended similarly with temperature- or wind-related solubility changes, 316 317 rather than oppositely if metabolic processes (i.e., primary productivity or decomposition of organic matter) were 318 important drivers in surface waters. Shoreline ponds changed drastically in size and chemistry in response to 319 seasonal flooding by Lake Hazen shoreline water. During pre-flooding conditions, CO₂ concentrations were low 320 which could be attributed to DIC use by autotrophic plankton (pre- to post-flooding mean chl-a concentrations of 1.2 to 0.4 µg L⁻¹), but more likely by observed dense benthic and macrophytic communities along the margins of the 321 322 ponds. When inundated by flood waters, CO₂ concentrations rose sharply which is typically observed in flooded 323 wetlands (Kelly et al., 1997). This occurs because widespread inundation of plants and soils typically prompts rapid decomposition and propagation of reduced compounds (e.g., NH_4^+ ; Table S4). Although diurnal CO₂ and O₂ 324 325 concentrations suggest that primary productivity was consistently occurring in Shoreline pond surface waters, this 326 pattern appeared overwhelmed by acute seasonal adjustments in CO2 exchange driven by flooding.

327 CO_2 concentrations in Lake Hazen shoreline water were near atmospheric equilibrium and only weakly 328 consumed atmospheric CO_2 . These results along the shoreline appear to be similar to other locations offshore 329 (unpublished) and were reflective of most deep lakes with extremely low nutrient, organic matter and chl-a 330 concentrations (0.20 µg L⁻¹; Keatley et al., 2007; Babaluk et al., 2009). CO_2 gas exchange between the lake and the 331 atmosphere correlated well with DIC, alkalinity and other ions, which suggests supply and dissociation of carbonate





332 material from the watershed, as well as wind mixing, were important factors contributing to Lake Hazen surface 333 water CO₂ concentrations, rather than primary productivity or heterotrophic decomposition.

334 4.1.2 CH₄

335 Evaporative and Meltwater systems were typically weak producers and emitters of CH₄, which was possibly sustained by concurrently high SO_4^{2-} concentrations in these systems (Table 2; Trettin, 1994). This may 336 337 have given competitive advantage to SO_4^{2-} -reducing bacterial communities in sediments, which typically 338 outcompete methanogenic bacteria for hydrogen. This hypothesis was supported by the prevalence of H₂S gas in collected sediment cores from Skeleton Lake (unpublished) and by the trivial fluxes of CH₄ in bubbles measured 339 emerging from sediments (0.00-0.01 mg m⁻² d⁻¹; Table S5; see Supporting Information). Low production and 340 exchange of CH₄ in Lake Hazen, alternatively, was most likely associated with the lake's ultra-oligotrophic 341 342 standing, well-oxygenated water, and little accumulation of littoral organic matter where anoxia could prevail and 343 CH₄ be produced. Only during periods of strong wind mixing of surface waters, or when Shoreline ponds breached 344 and released particulate organic matter, did CH_4 release from the shoreline of the lake to the atmosphere increase 345 above near-zero values.

Shoreline ponds were regional "hot-spots" of CH_4 exchange, which was clearly driven by seasonal flooding, similar to that described for CO_2 exchange. 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 $SO_4^{2^2}$ -poor Lake Hazen water, therefore favoring metabolism of methanogens over $SO_4^{2^2}$ -reducers in the flooded soils.

353 4.3 Net atmospheric exchange of CO₂ and CH₄ of a large high Arctic watershed

Most studies of terrestrial and freshwater carbon GHG exchange in the broader Arctic do not occur concurrently at a specific location. Our multiple season program of measuring carbon GHG exchange of both freshwater and terrestrial ecosystems provided a unique opportunity to delineate the relative strengths of freshwater and terrestrial contributions to regional carbon cycling in a high Arctic watershed (Table 3). Polar semideserts are typically dry and barren landscapes with little vegetation growth or organic matter and nutrient accumulation to





359 drive atmospheric exchange of CO₂. Not surprisingly, freshwaters receiving runoff from the polar semidesert also support mostly underproductive ecosystems which exchange little CO₂ with the atmosphere (Figure 6). Lake Hazen, 360 similarly, is sustained by cold and sediment-laden glacial melt water limited in compounds essential for life. 361 362 Together, these oligotrophic freshwater and terrestrial ecosystems largely characterize the current low-production 363 state of the Lake Hazen watershed and much of the high Arctic in general. Meadow wetlands, alternatively, are 364 topographical lowlands with flowing water which are ideal high Arctic environments for vegetation growth and 365 accumulation of soils rich in organic matter and nutrients. Conditions in these spatially-limited meadow wetlands 366 are so profound relative to other ecosystem types in the Lake Hazen watershed that they currently drive a strong 367 majority of atmospheric CO₂ exchange there. Evidently, CO₂ uptake across this, and likely other, high Arctic 368 landscapes is inherently tied to the availability and movement of water.

369 Though polar semidesert landscapes exchanged only limited amounts of CO₂ with the atmosphere, they 370 were extremely important sinks of atmospheric CH₄, such that they dominate high Arctic CH₄ cycling. This strong sink has been attributed to soil conditions that promote efficient atmospheric gas diffusion and temperature 371 372 sensitivity of methanotrophic bacteria at high latitudes (Emmerton et al., 2014; Jorgensen et al., 2015). Any 373 ecosystems where water saturation and anoxia would be expected to prevail during the growing season (i.e., small 374 ponds, lakes and meadow wetlands) were surprisingly weak emitters of CH₄. This may have been due to poor 375 quantities of organic substrates in the soils or beds of these systems, oxidation zones near gas exchange sites, or biogeochemical constraints related to SO_4^{2-} . Regardless, CH_4 exchange in high Arctic ecosystems, at least those 376 377 similar to the landscape composition of the Lake Hazen watershed, cannot be overlooked as substantial sinks of this 378 potent GHG. However, these conditions, which currently define carbon GHG exchange in the Lake Hazen 379 watershed, may be poised to change.

Warming growing season conditions in the Lake Hazen watershed have affected temperature sensitive components of the landscape including deep soils and permafrost (unpublished). Warming has also affected the region's hydrology through the greater delivery of glacial melt water to Lake Hazen and increased frequency and extent of ice-free area across the lake. Further, other studies suggest that changing air masses and evaporation from newly exposed coastal waters due to sea ice loss will deliver increased precipitation to high Arctic landscapes (Bintanja and Selten, 2014). These intensifying temperature and precipitation conditions will likely accelerate landscape changes in the watershed, especially considering the contemporary climate in the region is one at the low





387 global extreme of summer air temperatures and water availability. For example, Evaporative ponds could expand 388 and deepen with greater precipitation and snow runoff, possibly repressing CO₂ emissions similar to Ponds 10 and 12. Greater heating without sustained precipitation may alternatively cause these systems to continue to shallow and 389 possibly strengthen GHG emissions to the atmosphere, similar to Ponds 03 and 07, but would be susceptible to 390 391 ultimate drying (Smol & Douglas, 2007). Meltwater systems, alternatively, may be resistant to climate-related 392 influences because of their already steady water supply and greater volumes, though future water delivery may be 393 affected by diminishing stores of permafrost ice in soils. Shoreline ponds ultimately may endure the most substantial 394 changes in GHG emissions of high Arctic water bodies. Lake Hazen, under warming and wetting conditions, would 395 be expected to receive greater amounts of glacial melt water runoff during the growing seasons. Earlier rises in Lake 396 Hazen water levels would cause earlier flooding of Shoreline ponds and sustain longer periods of organic soil and 397 vegetation inundation, resulting in larger CO₂ and CH₄ emissions to the atmosphere. Eventually, though, extensive 398 flooding may prevent the macrophyte growth currently found in these Shoreline ponds, decreasing the amount of 399 fresh organic available for decomposition once the flooding occurs. However, based on areal coverage in the 400 watershed, future changes in the GHG exchange of the region is likely dependent on changes occurring at the polar 401 semidesert and in Lake Hazen itself. Polar semideserts and meadow wetlands represent a gradient of productivity 402 defined by increasing water availability. If increased heating and precipitation results in greater soil retention of 403 water and support for greater summer vegetation, then the sink strength of CO_2 in the high Arctic may improve 404 drastically, but may at the same time reduce atmospheric CH₄ oxidation in soils. However topographical constraints 405 of water flow may limit the magnitude of vegetation growth in meadow wetlands and across the polar semidesert in 406 the shorter term. For Lake Hazen itself, we may expect that a warming climate will hasten loss of ice from the lake, 407 resulting in greater water column heating, longer growing seasons, and increased river inflow to the lake. This may 408 induce more intense mixing and nutrient availability in the water column and perhaps higher productivity and CO₂ 409 sequestration. Loss of glacial ice also presents new, but uncertain, opportunities of gas exchange on newly exposed 410 landscapes.

411 Ultimately, freshwater systems in the Lake Hazen watershed did not produce considerable enough fluxes of 412 GHGs relative to the atmosphere to dominate regional GHG exchange, as observed in other studies to the south. 413 However, potential does exist in the watershed for hot-spots of GHG exchange to emerge from greater water 414 availability, which is a distinct possibility in the near-future with ongoing rapid climate warming in the region.





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595 Tables

596 Table 1 Morphometry and hydrology of ponds and lakes sampled for greenhouse gases concentrations and general chemistry in the Lake Hazen (LH) watershed during the growing seasons of 2005, and 2007-2012. 597

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)	0.2-0.7	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, permafrost
Pond 12 (N81.831W71.529)	0.2	0.8	1.9	370	Snowmelt
Pond 16 (N81.850W71.392)	0.8	1.1	2.1	434	Snowmelt, permafrost
Skeleton L. (N81.829W71.480)	1.9	1.9	4.7	299	Snowmelt, permafrost
LH-shore (N81.821 W71.352)	54,200	95 ^a	267 ^a	158	Glacial, snowmelt

^aKock et al., 2012

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Table 2 Mean water temperature and general chemistry of different freshwater types in the Lake Hazen (LH) watershed during the growing seasons of 2005, 2007-2012. All measurements are in μ mol L⁻¹ except for W_T (°C), TDS (mg L⁻¹) and chl-*a* (μ g L⁻¹). 601 602 603

Lake Type	WT	pН	TDS	PC	DIC	DOC	NO ₃	$\mathbf{NH_4}^+$	TDP	Fe	SO4 ²⁻	Chl-a
Evaporative	10.8	8.3	1,020	49	2,122	2,374	0.01	0.5	0.32	1.13	5,320	1.3
Meltwater	11.0	8.2	333	23	1,458	452	0.02	1.9	0.17	0.05	1,755	0.5
Shoreline	11.6	8.2	173	32	1,691	311	0.11	2.0	0.18	1.56	365	0.4
LH-shoreline	5.4	7.9	59	10	524	51	0.24	1.8	0.08	0.04	69	0.1
Meltwater source	2.6	7.6	653	-	769	67	7.70	0.1	0.05	0.6	3318	-

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605 W_T: water temperature; TDS: total dissolved solids; PC: particulate carbon; DIC: dissolved inorganic carbon; DOC: dissolved

606 organic carbon; NO_3 : dissolved nitrate + nitrite; NH_4^+ : dissolved ammonium; TDP: total dissolved phosphorus; Fe: dissolved iron; SO42: dissolved sulfate; chl-a: chlorophyll-a 607

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608	Table 3 Ranges or means of CO ₂ and CH ₄ fluxes (g C m ² d ⁻¹) from selected studies investigating terrestrial and
609	freshwater greenhouse gases exchange during the growing season of high, low and subarctic regions. Positive values
610	represent emission to the atmosphere.

Location*	Ecosystem	Period	CO ₂ flux	CH ₄ flux	Study	
High Arctic						
Lake Hazen, CA	Dry tundra	JunAug.	-0.00	-0.00	a, b	
Zackenberg, GR	Heath	May-Sep.	-0.39,0.03	0.04,0.06	c, d	
Lake Hazen, CA	Ponds	JunAug.	-0.01,0.05	0.00,0.01	This study	
Pond Inlet, CA	Ponds	Jul.	-0.22,0.72 0.00,0.07		e	
Low Arctic						
Lena delta, RU	Wet tundra	May-Aug.	-0.35	0.01	f, g	
Barrow, US	Moist tundra	JunSep.	-0.02,0.66	0.02	h, i	
Lena delta, RU	Ponds	AugSep.	0.38,1.10		j	
Toolik Lake, US	Lakes	JulAug.	-0.07,0.72	0.96,12.25	k	
Yukon delta, US	Lakes	JunAug.		0.04	1	
Subarctic						
Chokurdakh, RU	Tundra	Jul-Aug		0.05	m	
Abisco, SE	Shrub tundra	Jul-Aug.	-0.95,-0.83		n	
Cherskii,, RU	Tussock tundra	JulAug.	-0.15,0.50	0.32	0	
James Bay, CA	Mixed tundra	Jun-Oct.		0.04	р	
Narvik, SE	Lakes	May-Oct.	-0.03,0.13	0.00,0.02	q	
Inuvik, CA	Lakes	JunAug.	-1.55,-0.65	-	r	
Cherskii, RU	Ponds	JunSep.		0.09	S	

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a Emmerton et al., 2016; b Emmerton et al., 2014; c Lloyd, 2001; dTagesson et al., 2012; e Laurion et al., 2010; f Kutzbach et al., 2007; g Sachs et al., 2010; h Kwon et al., 2006; i Sturtevant and Oechel, 2013; j Abnizova et al., 2012; k Kling et al., 1992; l 612

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614 Fan et al., 1992; m Parmentier et al., 2011; n Fox et al., 2008; o Merbold et al., 2009; p Roulet et al., 1994; q Karlsson et al.,

2013; r Tank et al., 2009; s Walter et al., 2006; *as delineated by AMAP, 1998. 615





616 Figures



617 618

619 Figure 1 Map of the Lake Hazen base camp in Quttinirpaaq National Park, Nunavut, Canada. Ponds and lakes 620 investigated in this study are indicated on the map and shown in photographs. Shown inset of the map are the general 621 locators of the Lake Hazen watershed.







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623 Figure 2 Dendogram of sampled high Arctic freshwater systems in the Lake Hazen watershed between 2005, and 2007-2012 (hierarchical cluster analysis; see Supporting Information). Water chemistry (see Methods) and carbon greenhouse

624 625 gases concentrations measured periodically from 10 locations (Figure 1) were used as inputs to the analysis.







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Figure 4 Mean (±SE) dissolved carbon dioxide (CO₂) and methane (CH₄) concentrations and fluxes during the 2005, and 2007-2012 growing seasons from four different freshwater system types in the Lake Hazen watershed. Letters denote statistical differences between ecosystem types for each gas (linear mixed model; α =0.05; see Supporting Information).







Figure 5 Three-hour diurnal dissolved carbon dioxide (CO2) concentration, oxygen (O2) 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 in the Lake Hazen watershed.

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Figure 6 (a.) Comparison of the 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 of 2005, and 2007-2012. Letters denote statistical differences between ecosystem types for each gas (linear mixed model; α =0.05; see Supporting Information). (b.) Total growing season (June-August) watershed exchange of CO₂ and CH₄ by terrestrial and freshwater types in

the total Lake Hazen watershed.

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