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Declining Summertime pCO_2 in Tundra Lakes in a Granitic Landscape

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Key Points:

- Warming lakes feature decreasing carbon dioxide concentrations

Supporting Information:

- Supporting Information S1
- Table S1
- Data Set S1

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Abstract As climate change accelerates, positive feedback loops could establish between atmospheric warming and increasing greenhouse gas (GHG) emissions from natural ecosystems. Of particular interest are high-latitude environments, many of which contain large stores of organic carbon and have experienced decades of accelerated warming. Freshwater ecosystems situated in carbon-rich Arctic landscapes are predicted to respond to warming with higher respiration rates relative to primary production rates, increasing their carbon dioxide partial pressure (pCO_2) and thus CO_2 emissions to the atmosphere. However, large areas of shallow soil/bedrock occur in Arctic and sub-Arctic regions and could respond differently due to the lower availability of stored carbon. Here, we used pH and alkalinity to calculate 23 years (1994–2017) of summertime (July and August) pCO_2 and CO_2 fluxes for 19 sub-Arctic tundra lakes located on the shallow-bedrock granitic Canadian Shield in Nunavut, Canada, and found a significant decline in CO_2 emissions. Regional precipitation did not change over this period, yet dramatic increases in pH, conductivity, and total alkalinity indicated that longer ice-free periods and changes in primary production, both functions of accelerated climate warming at higher latitudes, may be suppressing lacustrine CO_2 emissions. Northern lakes overlying differing geological landscapes may thus be responding in contrasting ways in terms of pCO_2 and CO_2 fluxes, with potential major implications for regional carbon budgets and predicted climate change feedbacks.

Plain Language Summary Carbon dioxide (CO_2) emissions from lakes to the atmosphere are expected to shift with climate change, yet there is no scientific consensus on the overall directionality or timing of these shifts. This study presents 23 years of data from 19 lakes near Rankin Inlet, Nunavut (Canada), representative of a poorly studied yet geographically extensive sub-Arctic region-type (shallow-bedrock catchments). We found declining partial pressure of CO_2 and surface emissions for these lakes, likely due to the direct and indirect effects of shortening ice-cover periods. This is significant as it improves our understanding of regional variation in how high-latitude lakes are responding to climate change, and provides us with mechanistic insights into the potentially variable responses of lake CO_2 emissions to warming.

1. Introduction

Accelerated warming, a high coverage of freshwater systems, and vast stores of frozen organic carbon have put the Arctic at the forefront of research examining feedbacks between climate change, terrestrial-aquatic linkages, and biogeochemical cycling (Wauthy et al., 2018). Given the potential transformation of the north's stores of organic carbon to CO_2 or methane (CH_4), either directly from terrestrial landscapes (McGuire et al., 2009) or via processing in aquatic environments (Tank et al., 2018), research examining the effects of climate change in the north has focused primarily on the impact of permafrost thaw (Oechel et al., 2000; Rouse et al., 1997; Wauthy et al., 2018). Permafrost thaw in peatlands or carbon-rich regions can enable transport of dissolved organic carbon (DOC) and nutrients into nearby surface waters, influencing food webs and net ecosystem metabolism, and potentially enhancing atmospheric GHG emissions (Harms et al., 2014; Hobbie et al., 1999; Tank et al., 2009; Walter et al., 2008). Increasing land-water connectivity increases CO_2 emissions from some northern lakes (Tank et al., 2009), yet evaporative concentration coupled with DOC exudation associated with increasing autochthonous primary production can also result in negative relationships between DOC and lake pCO_2 (Tank et al., 2009). Lakes in various regions have featured

negative (Tank et al., 2009), neutral (Nydahl et al., 2017), and positive (Lapierre & del Giorgio, 2012) relationships between DOC and pCO_2, indicating that multiple biogeochemical drivers of trends in lake CO_2 emissions may be interacting simultaneously.

Emerging is a broader understanding that primary production can play an important role in GHG emissions from northern lakes (Bogard et al., 2019), making many of these systems potential GHG sinks relative to the atmosphere (Bogard et al., 2019; Tank et al., 2009). Paleolimnological studies have shown that even in northern lakes that are not associated with permafrost thaw, biological communities have responded dramatically to climate change (Smol et al., 2005), indicating that within-lake processes may control the CO_2 balance of many northern freshwaters. Broad patterns in responses to climate change are thus difficult to predict, especially in understudied regions where limnological data is scarce (Metcalfe et al., 2018). A diversity of lake carbon cycling responses to climatic warming is not surprising, however, given that the permafrost region is highly heterogeneous, and much of it is underlain by shallow-bedrock zones such as the Canadian Shield, which covers 8,000,000 km^2 of northern North America and roughly half of northern Canada's surface area (above 60°N). Arctic lakes located on such shallow-bedrock often experience low levels of DOC loading (Wauthy et al., 2018), and may thus respond distinctly to climate change.

Regardless of catchment connectivity to thawing permafrost and associated active layers, the water chemistry and biology of northern lakes are changing (Drake et al., 2019; Smol & Douglas, 2007; Smol et al., 2005), with likely consequences to lake pCO_2, and thus CO_2 fluxes between the lake surface and atmosphere (Serikova et al., 2019). Warming increases both gross primary production (GPP) and community respiration (CR) in lakes, but the response of CR to warming water temperatures typically outpaces that of GPP (Yvon-Durocher et al., 2010). This effect is expected to turn lakes into greater CO_2 sources to the atmosphere (Drake et al., 2019; Kraemer et al., 2017; Tanentzap et al., 2019; Yvon-Durocher et al., 2010, 2017). However, long-term analyses of changes in pCO_2 and CO_2 fluxes from lakes are scarce, and provide limited support for a widespread trend of increasing efflux rates (Seekell & Gudasz, 2016). Paleolimnological proxy data have indicated an increase in CO_2 emissions from three large lakes in Europe (Perga et al., 2016), yet increases in the DOC concentrations of Swedish inland waters have generally not been accompanied by increases in pCO_2 (Nydahl et al., 2017). Another study identified high CO_2 emissions from Russian thermokarst lakes, but a space-for-time analysis determined that these emissions may be declining with warming (Serikova et al., 2019). Similarly, CO_2 emissions from alkaline hardwater lakes in central Canada have declined as a function of climate change (Finlay et al., 2015). In this case, longer ice-free periods and subsequent increases in pH drove the chemical enhancement of CO_2 uptake, indicating a critical role for water chemistry and ice-cover (relevant to midlatitude to high-latitude and alpine lakes) rather than solely metabolic responses to temperature (Finlay et al., 2015). Ice-cover duration can be an important factor influencing lake CO_2 emissions (Cohen & Melack, 2019), but it is rarely explicitly accounted for in broad predictions of how lakes will respond to climate change.

Although our mechanistic understanding of the drivers of CO_2 emissions in lakes has improved, much uncertainty remains regarding the net changes of CO_2 fluxes from northern lakes. This is due both to the geological heterogeneity of the region as well as the paucity of data associated with the difficulty of sampling as a function of their remoteness. Much of the current knowledge of northern climate change impacts comes from focused field sites in northern Alaska and Sweden, while regions such as northern Canada remain much more patchily studied (Metcalfe et al., 2018). Here we analyzed 23 years (1994–2017) of summertime (July and August) pCO_2 and lake-atmosphere CO_2 fluxes in 19 tundra lakes near Rankin Inlet, Canada (Figure 1). Although these lakes are within a continuous permafrost zone, they are situated in the shallow granitic bedrock of the Canadian Shield, providing the opportunity to focus on the effects of climate change (warmer summertime waters and longer ice-free periods) on CO_2 dynamics in northern lakes that will likely receive little change in direct allochthonous loading due to permafrost thaw.

2. Materials and Methods

2.1. Lake Sampling and Data

Our study lakes are located in the Meliadine Lake Peninsula region (63°04′ N, 92°19′ W), approximately 15 km northwest of Rankin Inlet, Nunavut, Canada. Early data (1994–2009) were retrieved from an aquatic

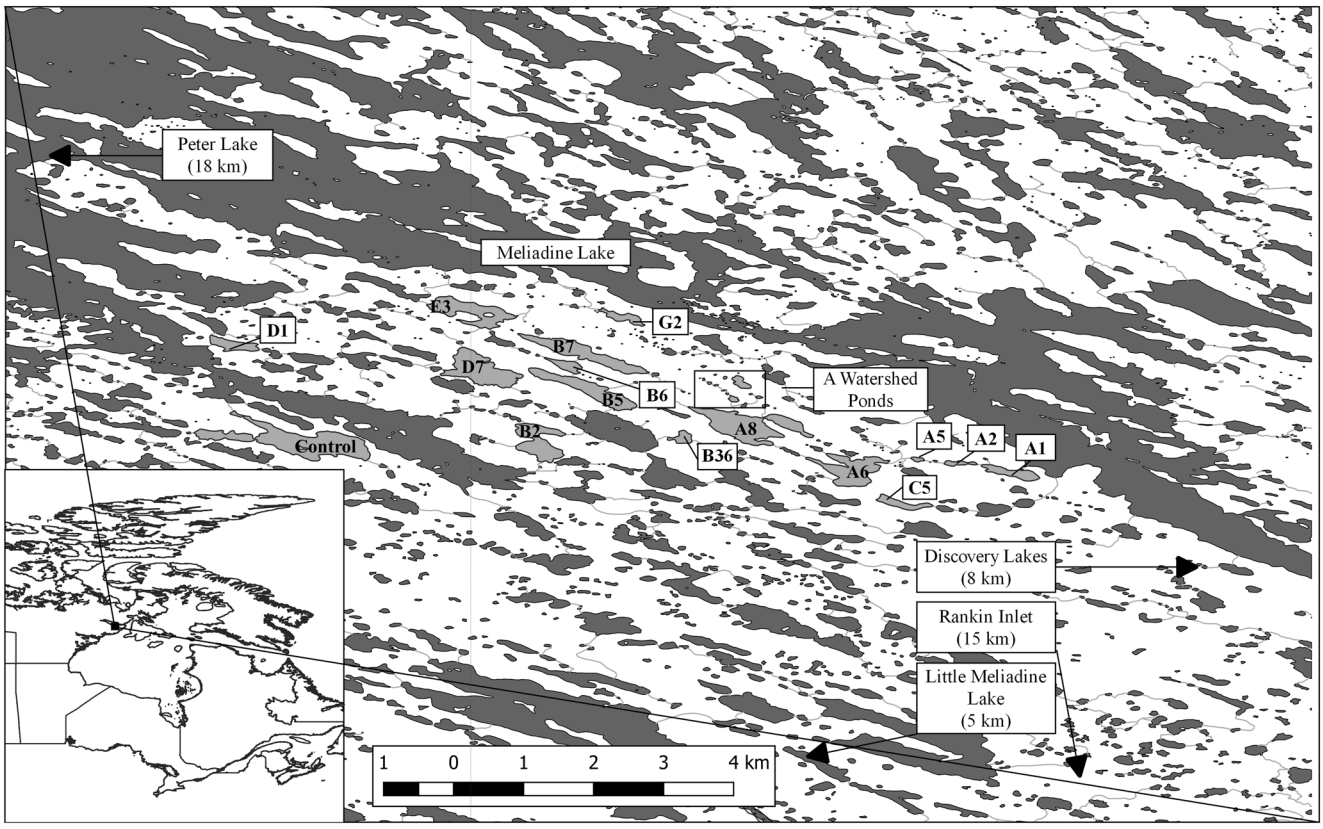

Figure 1. Map of the Meliadine Peninsula region, identifying study lakes included in this analysis. "A Watershed Ponds," along with A5, are included only in supplementary analyses.

synthesis report for a proposed gold mine in the area, which was made available online by the government of Nunavut (Golder Associates, 2009). Additional data (2009–2015) were provided by Agnico-Eagle Mines Limited (hereafter referred to as "Agnico") monitoring campaigns. Recent data (2014–2017) was sampled and/or measured directly by the coauthors. Annual precipitation rates for Rankin Inlet (NU) were available from 1995 to 2009 in the aquatic synthesis report (Golder Associates, 2009) and for more recent years (2010–2017) from historical Weather Network data (Weather Network, 2018). See Extended Data (Table S1) for a complete list of lake names, years of data available, and sources of data. To account for this use of multiple independent sources of data, we applied a Bayesian analysis (see section 2.3 for details).

Field campaigns were carried out in 2014 (July 17–22), 2015 (August 24 to September 5), 2016 (July 29 to August 6), and 2017 (July 25–30). Three sites were randomly chosen on each study lake for vertical profiles and water sampling. Vertical profiles measured water temperature, specific conductivity, and pH using an EXO2 multiparameter sonde (Yellow Springs Instruments, USA), and total nitrogen (TN), total phosphorus (TP), DOC, calcium (Ca), magnesium (Mg), and chlorophyll *a* concentrations were measured from subsurface (i.e., 0.1 m depth) water samples retrieved by hand. The mean value from all three lake sites was applied in analyses. Sonde pH and conductivity probes were calibrated prior to each sampling campaign, and recalibrated weekly during longer sampling campaigns. All pH-linked data (including pCO_2 and CO_2 fluxes) from 2015 were excluded from analyses due to a damaged pH probe. All historical measurements of water chemistry parameters in these lakes followed standard methods, such as those outlined by the United States Environmental Protection Agency (see Tables 5 and 6, p. 73; Tables 5–8, p. 74; Golder Associates, 2009), and parameters were analyzed by accredited laboratories featuring standard quality control procedures (p. 77; Golder Associates, 2009). Water chemistry (Ca, Mg, TN, and TP) from the 2015–2017 field campaigns was analyzed by the Agriculture and Food Laboratory at the University of Guelph from the collected subsurface water samples (described above), which were transported on ice in dark coolers to the field laboratory, where they were frozen until being analyzed following standard procedures and quality control. At the end

of each sampling day, water samples were analyzed immediately for total alkalinity by titration using an alkalinity test kit (Hach), and filtered onto 0.45 μm GF/F filters which were frozen for chlorophyll *a* analysis (carried out at the University of Guelph). Chlorophyll *a* concentrations were measured using a Turner Designs (TD-700) laboratory fluorometer (Sunnyvale, CA) following standard procedures (Bruinsma, 1963).

For DOC concentrations from our monitoring campaign, 2014 and 2017 water samples were filtered in the field with 0.45 μm GF/F filters, stored in glass vials, fixed with hydrochloric acid, and measured within 2 weeks of sampling using a DOC analyzer (Shimadzu). Concentrations for 2015 and 2016 were inferred from 2014 relationships, calibrated to a YSI probe that measured the fluorescence of dissolved organic matter (fDOM, quinine sulphate units; Pearson's product-moment correlation, $p < 0.0001$). In situ fDOM measurements in aquatic systems can be problematic due to turbidity (Downing et al., 2012) and inner-filter effects (Kothawala et al., 2013), but turbidity was low in our oligotrophic study lakes, and the strong relationship between in situ fDOM and separately measured DOC concentrations should provide good estimates of values for the 2 years in question. Total dissolved inorganic carbon (DIC, the sum of CO_2, bicarbonate, and carbonate concentrations), pCO_2, and bicarbonate concentrations were calculated using the computer program CO2SYS (Lewis & Wallace, 1998), using measured total alkalinity, water temperature, and pH values from the available datasets, applying standard equations for freshwater aquatic environments (Dickson, 1990; Lee et al., 2010; Millero, 1979) and an air pressure of 10.1 dbars (assuming an approximate local altitude of 28 m above sea level). Surface pCO_2 is best measured directly in freshwater systems (Abril et al., 2015; Golub et al., 2017), but direct measurements for these lakes were unavailable. Calculations based on water chemistry (as provided by CO2SYS) can greatly overestimate true values when applied to acidic, organic matter-rich lakes (Abril et al., 2015), but are appropriate for well-buffered, oligotrophic lakes. The lakes included in this study tended to feature measured DOC concentrations that were somewhat higher than the global average for lakes (Toming et al., 2020), but another study (Kratz et al., 1997) identified a strong, linear relationship between measured and calculated pCO_2 from a set of lakes in Wisconsin featuring a similar range of DOC as that measured in our study lakes (~2–16 mg L^{-1} versus ~3–8 mg L^{-1} in our study). The pH of our study lakes tended to be greater than 7, indicating that an overestimation of calculated pCO_2, if occurring, was likely minor (~15%; Table 2 in Abril et al., 2015). Regardless, the errors associated with pCO_2 calculations from water chemistry produce uncertainty regarding the specific values for a given lake at a specific time of measurement, but should not in themselves produce or reverse long-term trends in calculated pCO_2. These calculations were thus considered appropriate for this analysis.

2.2. Carbon Dioxide Flux Calculations

Compounding the uncertainties of calculating pCO_2 from water chemistry, surface CO_2 flux calculations derived from pCO_2 must be treated with great caution (Klaus & Vachon, 2020). Recognizing this uncertainty, we calculated CO_2 fluxes to provide a general idea of how changes in fluxes over time might relate to lake surface CO_2 saturation relative to the atmosphere, as the magnitude of the error in calculations should diminish as fluxes approach zero (at atmospheric equilibrium). We believe this calculation also provides a useful comparison with existing literature that focuses on long-term trends in CO_2 fluxes, much of which is also derived from such calculations rather than direct CO_2 flux measurements (e.g., Finlay et al., 2015). As with the uncertainties discussed for pCO_2 calculations, uncertainties associated with air-water gas transfer velocities should not in themselves produce long-term directional trends in CO_2 fluxes independently of lake pCO_2, and should not affect calculations of directional CO_2 fluxes relative to the atmosphere (i.e., whether a lake is a sink or source of atmospheric CO_2). Surface CO_2 fluxes were thus calculated from pCO_2, wind speed, water temperature, lake surface area, and historical atmospheric CO_2 concentrations. Historical water chemistry sources provided only one data point per lake per sampling date, though for recent sampling campaigns the mean pCO_2 for each date/lake was calculated from the three off-shore sites sampled. Briefly, we applied the equation (Cole & Prairie, 2009):

$$F = K_{CO_2} \times \left(\Delta CO_2\left(\text{surface - air}\right)\right) \quad (1)$$

where F is CO_2 flux (mg C m^{-2} d^{-1}), ΔCO_2 (surface-air) is the difference in surface water and atmospheric CO_2 (in mg m^{-3}) and K_{CO_2} is the transfer velocity of CO_2 (in m d^{-1}), calculated (Vachon et al., 2010 and references therein) as:

$$K_{CO_2} = \left(K_{600} / \left(\left(600 / Sc \right)^n \right) \right) \quad (2)$$

where K_{600} is a gas transfer velocity normalized to a Schmidt number of 600, Sc is the CO_2 Schmidt number at a given temperature (Vachon et al., 2010 and references therein) and n is 0.5, or 2/3 when wind speed measured at 10 m height (U_{10}, m s^{-1}) is less than 3.7 m s^{-1} (Guérin et al., 2007 and references therein). Sc is calculated (Wanninkhof, 1992) as:

$$Sc = 1911.1 - \left(118.11 \times T \right) + \left(3.4527 \times T^2 \right) - \left(0.04132 \times T^3 \right), \quad (3)$$

In order to account for differences in fetch between lakes, calculations of K_{600} gas transfer velocities used in our analyses applied both lake surface area (SA, km^2) and wind speed data, following the equation (Vachon & Prairie, 2013):

$$K_{600} = 2.51 + \left(1.48 \times U_{10} \right) + \left(0.39 \times U_{10} \times \log_{10} SA \right) \quad (4)$$

Due to increasing pH values over time, and since another study describing long-term declines in lake CO_2 emissions identified chemical enhancement as an important driver of CO_2 fluxes (Finlay et al., 2015), we furthermore corrected for the effects of chemical enhancement (Bade & Cole, 2006). Lake-specific long-term wind speed measurements were not available, and thus reliable hourly wind-specific CO_2 flux rates could not be calculated. We thus assumed a constant wind speed of 4.8 m s^{-1}, which was the calculated mean of locally available values (measured and provided by Agnico) from June to October in 2015 and 2016. However, long-term changes in wind speeds at northern latitudes have previously been described (Gearheard et al., 2010), which may potentially influence long-term trends in CO_2 fluxes. To provide a comparison of approaches, we thus additionally present CO_2 fluxes (Figure S1) calculated separately using the noontime wind speeds for each sampling date measured at 10 m height at Rankin Inlet (NU), the nearest long-term weather monitoring station for these lakes (Government of Canada, 2019). Given that this is a low-lying, treeless landscape, these wind speed values should be regionally representative while also reflective of any long-term trends in wind speeds.

Combining all data sources, 102 calculated pCO_2 values (and thus CO_2 fluxes) were available for 27 lakes and ponds (Table S1). However, multiple ($n > 1$) years of data were not available for all lakes in the earlier dataset, and due to logistical constraints, we were unable to physically resample all lakes for which earlier data had been available. Some data (16 pCO_2 values) were also available for eight ponds (see Table S1; here distinguished as systems with surface areas < 0.06 km^2), most of which were only sampled once in 2007. Although the inclusion of these ponds in the data analysis did not significantly alter the overall observed downward trend of pCO_2 (Figure S2a) or CO_2 emissions (Figure S2b), these ponds exhibited several water chemistry parameters (such as specific conductivity, Figure S3a, and total alkalinity, Figure S3b) with values well above those of the other local lakes. We therefore excluded these eight ponds from our primary data analysis, and present them only in supporting information. We adopted a Bayesian hierarchical model (described below) that accounts for variation in sampling across lakes through time, as well as how factors such as lake size might affect our analysis. Although we recognized a possible circularity when accounting for the effect of lake size on CO_2 flux through time (Abnizova et al., 2012; Holgerson & Raymond, 2016), ponds were excluded from our primary analysis, and there was no significant increase in surface area of sampled lakes over time in our study (Bayesian $p = 0.499$). There remained no statistical support for surface area as a driver of CO_2 flux, as the model accounts for lake-specific effects (Table S2). This was confirmed by a test of temporal changes in lake pCO_2 (Figure 2; not calculated using lake surface area), which similarly identified lake surface area as a less-important factor than time for driving these observed trends.

Due to the remoteness and extreme climate of these lakes, data for spring, winter, and fall were difficult to obtain. We therefore focused our analyses on summertime data (July and August), as this was the season for which the most data were available, while also providing for the most consistent analysis of long-term trends. The limited data from spring (June, $n = 7$; from 2013 to 2015, CO_2 fluxes ranging from −57.4 to 42.6 mg C m^{-2} h^{-1}) and fall (September and October; $n = 7$; from 2007 to 2015, CO_2 fluxes ranging from −54.3 to 73.2 mg C m^{-2} h^{-1}) fell within the same general range of fluxes calculated for contemporary

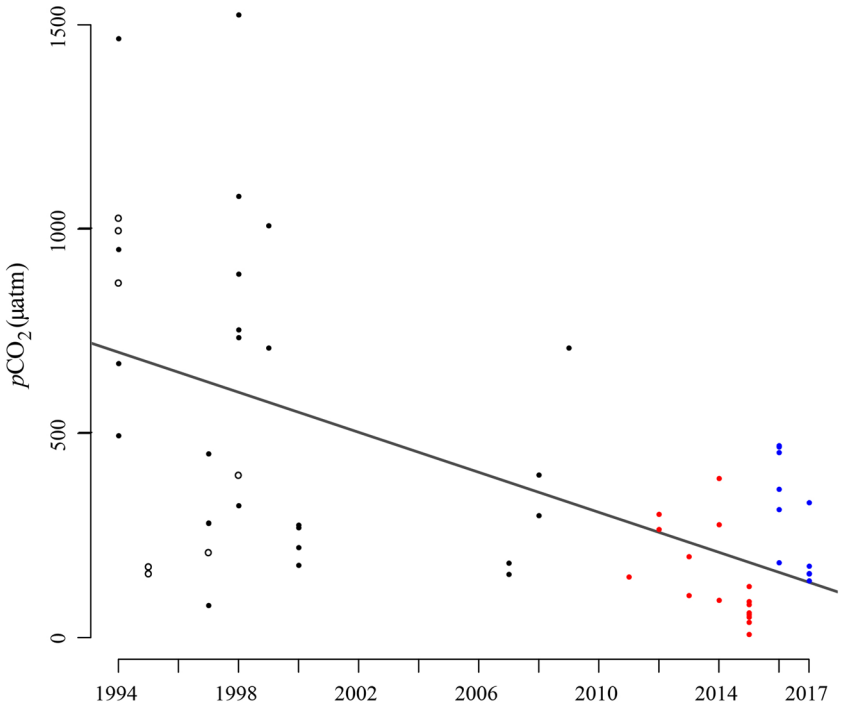

Figure 2. Long-term trends in pCO_2 for the 19 analyzed lakes. Data points are color-coded based on origin of associated data (black = aquatic synthesis report, red = Agnico, blue = authors' field campaigns, as described in section 2). Hollow points indicate lakes for which only one year of data are available.

summertime periods, and did not contraindicate our overall observed trend. These data were nevertheless insufficient to draw any conclusions regarding long-term changes in spring-specific and fall-specific seasonal fluxes. Lakes in this region frequently experience ice-cover from roughly November into June, and thus the period here analyzed represents a substantial portion of the open-water period for these lakes. Furthermore, springtime trends in CO_2 fluxes would be difficult to assess in these lakes, given the late ice-off timing (and thus compressed "spring" period) as well as the fact that spring flux dynamics in lakes can be complex, with peak fluxes typically occurring within days after ice-off (Denfeld et al., 2015).

2.3. Statistical Analysis

All statistical analyses were performed in R (v. 3.6.0) (R Core Team, 2019) and all software packages in *italics* are R packages. We used Bayesian hierarchical models (Gelman et al., 2013), fitted using *rstanarm* and *rstan*, to assess the statistical support for trends in the response variables of interest (CO_2 flux, surface temperature, pH, specific conductivity, total alkalinity, and DIC) through time. These models account for the unequal sampling among, and autocorrelation within, lake measurements and variation in measurement methodology, while also providing robust estimates of explanatory variable importance and uncertainty (Tables S2 and S3). In accounting for this variation, our models coestimate (pool, see Gelman & Pardoe, 2006) estimates at each lake, for each measurement methodology (at a lake), and through time for each of the above. In the supporting information, we report estimates of such pooling for all models (following Gelman & Pardoe, 2006). To further improve the robustness of our analysis outliers were removed in two stages: by visual inspections of residuals in a Gaussian Q-Q plot, and then by fitting a linear model of CO_2 efflux as a function of year, log(lake surface area), annual precipitation, Ca, and Mg, and then removing all data points with a Cook's distance greater than four times the total number of CO_2 efflux measurements (see Bollen & Jackman, 1985 for a discussion of outliner thresholds). To ensure that factors such as outliers, lake size, focusing on summer measurements, and a particularly cool year (2015) did not change our results, we

repeated analyses across various permutations of data subsetting, all of which (and the associated model code) are presented in the supplementary materials.

CO_2 flux was modeled as follows:

$$CO_2 \sim \text{Normal}\left(\alpha_i + \beta_0 \times \text{year} + \beta_{i,j} \times \text{year} + s \times \text{SA} + c \times \text{Ca} + m \times \text{Mg}, \varepsilon\right)$$

where CO_2 is the CO_2 flux, α_i a constant for each lake (i), β_0 the overall rate of change in flux across all lakes, $\beta_{i,j}$ the variation in the rate of change in flux for each lake (i) measured with each method (j), year is the year of measurement, s the coefficient for lake surface area (SA), c the coefficient for lake Ca, m the coefficient for lake Mg, and ε the overall error (which we did not examine). A parallel model was run that additionally accounted for the effect of precipitation and produced nearly identical results regarding overall trends in CO_2 fluxes. However, as regional precipitation data were not available for 1994, precipitation was excluded from the final model so that lake flux data for that year could be included in the final analysis. By nesting the measurement type (j) within each lake (i) in our estimation process, we account for the impact of changes in measurement *before* accounting for variation across lakes. Our analysis is thus robust to changes in sampling method through time within each lake as well as variation in sampling across lakes. All explanatory variables were Z-transformed to provide coefficients that represented relative variable importance in models; in all figures, however, we reverse-transformed the coefficients to make the plots more readily interpretable. Posterior distributions were manually checked for convergence and mixing across all 16 chains (all parameters had potential scale reductions statistics <1.01) and each chain had 2,000 iterations with the first 1,000 discarded as burn-in. Priors were allowed to "auto-scale," were checked to be uninformative, and were proper and so varied across parameter types [and were, specifically: intercepts—N(0,10), coefficients—N(0, 2.5), variance coefficients—Exponential(1), covariances—Decov(1, 1, 1, 1)].

Our analysis of whether other explanatory variables (including annual precipitation, Ca, Mg, DOC, TP, and chlorophyll *a*) exhibited long-term trends was simpler, following the form:

$$\text{response} \sim \text{Normal}\left(\alpha_i + \beta_0 \times \text{year} + s \times \text{SA}, \varepsilon\right)$$

where all definitions and all priors are as above. We also note that Bayesian models do not produce traditional p values, but instead provide quantiles describing the posterior distribution of the coefficients (Tables S2 and S3). If 95% of the distribution of the coefficient's value is greater than 0, then there is a 95% chance that the coefficient is positive. Thus, a "year" effect with a 2.5% quantile of 0 and all higher quantiles being negative would have a 97% chance of being negative. Within the text, we provide the probabilities (labeled as "Bayesian p") that the slope is either positive (e.g., "year > 0") or negative (e.g., "year < 0") in the coefficient file. Within a frequentist framework, a test with an α_{crit} of 5% generally has a statistical power of 80%; thus, we consider our use of 95% as a threshold for significance (and not 80%) to be conservative.

3. Results

We document a substantial reduction in summertime pCO_2 and CO_2 efflux rates from our study lakes over recent decades. Specifically, the available data showed long-term increases in dissolved inorganic carbon (DIC) concentrations (Figure 3a, calculated from water chemistry data), specific conductivity (Figure 3b), pH (Figure 3c), and total alkalinity (Figure 3d; all directly measured). These changes, and particularly the substantial increase in measured pH and total alkalinity (which has been observed elsewhere as well; Drake et al., 2018; Finlay et al., 2015; Raymond & Cole, 2003), resulted in a reduction in calculated lake pCO_2 over the same period (Figure 2; Bayesian $p > 0.999$, representing a >99% probability of a decrease). Calculated surface CO_2 emissions featured the same long-term trend, regardless of whether a constant wind speed (Figure 4a) or regional mean wind speed (Figure S1) was used in these calculations. These calculations both indicated that the lakes were typically net sources of CO_2 to the atmosphere in the mid-late 1990s (specific efflux rates presented are uncertain, as discussed in Section 2), but have been near atmospheric equilibrium since 2010 (Figure 4a, Bayesian $p = 0.999$). These results run counter to the widely hypothesized effect of warming on lake CO_2 fluxes (Kraemer et al., 2017; Tanentzap et al., 2019; Yvon-Durocher et al., 2010), as

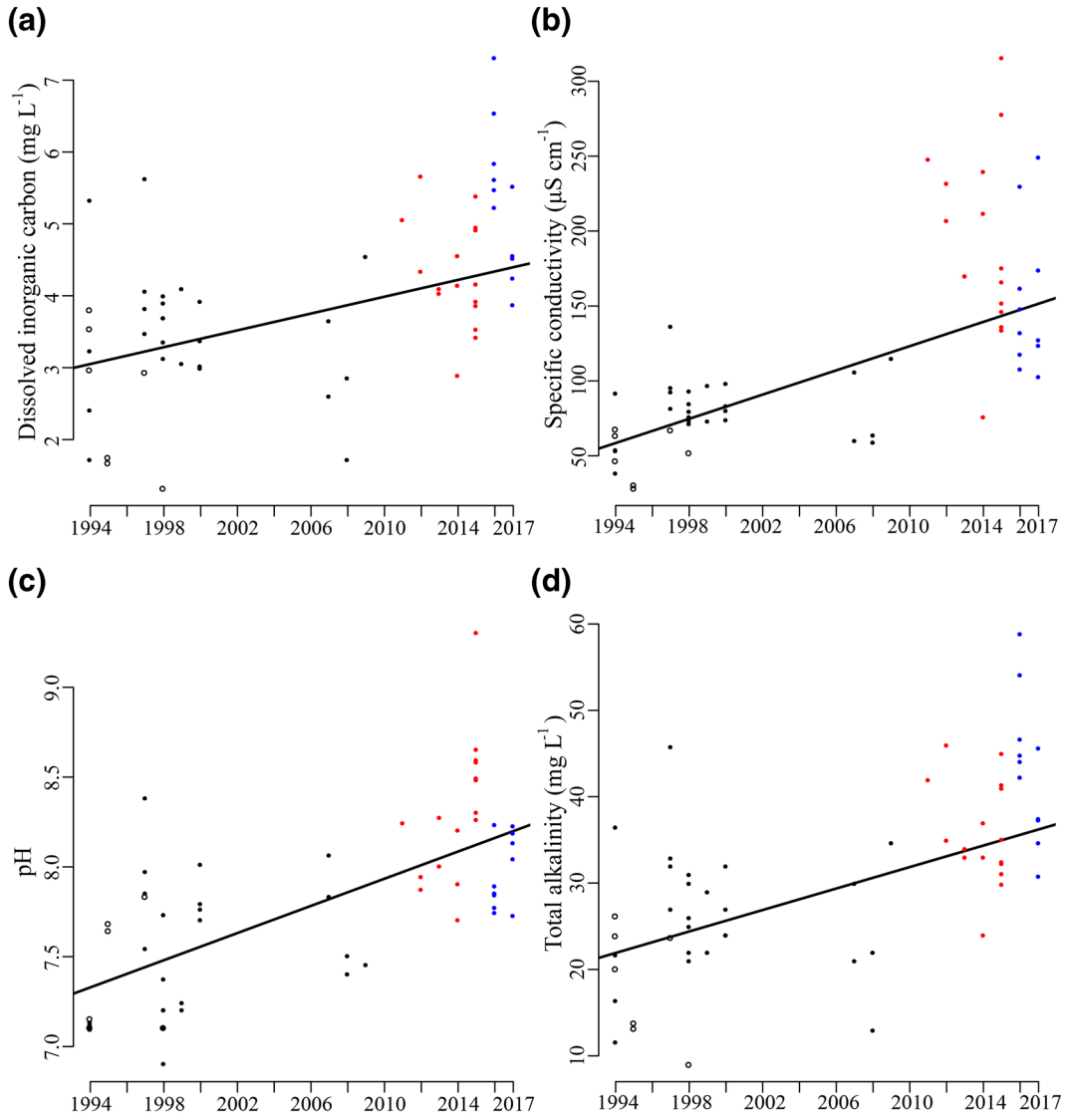

Figure 3. Long-term trends in summertime water chemistry parameters, pooling data from the 19 analyzed lakes. Panels show (a) dissolved inorganic carbon (calculated), (b) specific conductivity, (c) pH, and (d) total alkalinity (measured). Data points are color-coded based on origin of associated data (black = aquatic synthesis report, red = Agnico, blue = authors' field campaigns, as described in section 2). Hollow points indicate data from lakes for which only 1 year of data was available.

well as what has been described for tundra lakes in European Russia (Drake et al., 2019). This indicates that opposing patterns may occur in regions where warming is able to increase the lateral flux of terrestrial organic matter to aquatic systems, relative to those (e.g., underlain by shallow-bedrock) where changes in land-water connectivity are likely to be less impactful.

3.1. Allochthonous Loading Effects on CO_2 Fluxes

A decline in pCO_2 and CO_2 emissions could potentially result from reduced terrestrial (allochthonous) carbon loading (considering organic and inorganic sources), and/or from chemical or metabolic shifts occurring within the lakes. Metabolic processes and pCO_2 in northern and temperate lakes are often strongly linked to their terrestrial catchments (Ask et al., 2009; Lapierre et al., 2017), though terrestrial connectivity, and the effects of organic carbon on lake pCO_2, can vary greatly between regions (Lapierre & del

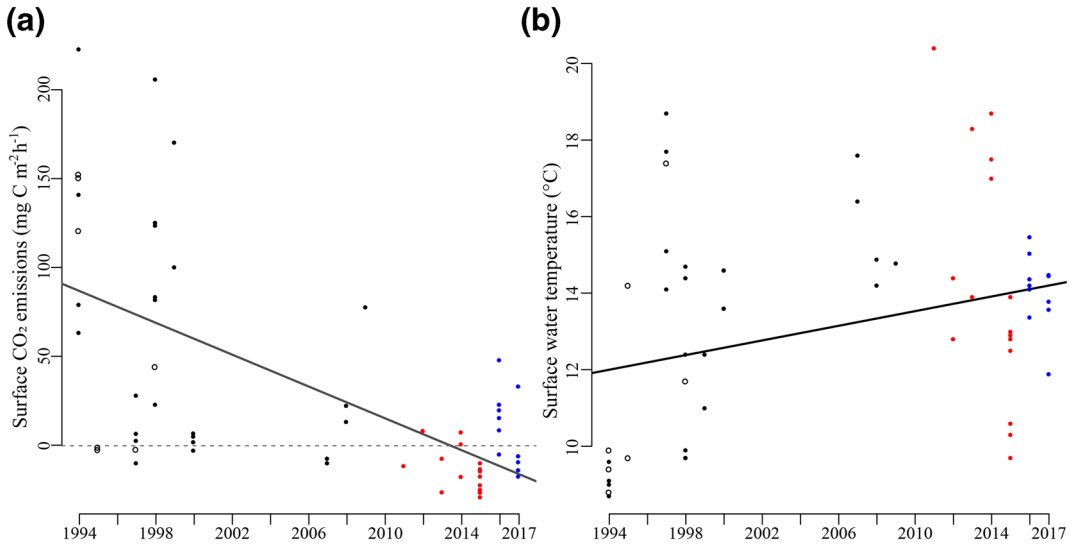

Figure 4. Long-term trends in summertime data pooled from all 19 analyzed lakes. Panels show (a) surface CO_2 emissions and (b) surface water temperatures. A horizontal dotted line demarcates net influx versus outflux of CO_2 relative to the atmosphere. Data points are color-coded based on origin of associated data (black = aquatic synthesis report, red = Agnico, blue = authors' field campaigns, as described in section 2). Hollow points indicate data from lakes for which only 1 year of data was available.

Giorgio, 2012). While variable, annual precipitation rates in this region did not show a significant decline during the study period (Bayesian $p = 0.469$, Figure S4; data available for 15 years from 1995 to 2017), indicating that long-term changes in CO_2 are not due to parallel changes in local precipitation.

Water column DOC concentrations are frequently used as an indicator of terrestrial-aquatic linkages (Dillon & Molot, 1997), and are often positively related with heterotrophic respiration and increased CO_2 fluxes (Brothers et al., 2012; Hanson et al., 2003). A hypothetical long-term decline in DOC concentrations could thus potentially lead to reduced CO_2 emissions in lakes where these factors are positively correlated. Unfortunately, DOC concentrations were unavailable for our study lakes prior to 2008, and early data (2008–2009) were sparse ($n = 3$) making a long-term analysis of directional trends in DOC concentrations impossible. However, recent (2016–2017) DOC concentrations appeared to be generally greater (mean ~5 mg L^{-1}) than early (2008–2009) measured DOC concentrations (mean ~4 mg L^{-1}, Figure S5). Although based on sparse data ($n = 14$), no clear trend linking DOC concentrations and lake CO_2 emissions over this period was apparent (Figure S6). There was thus no evidence from either precipitation or DOC data to indicate that the loading of allochthonous carbon had declined in these lakes, as would be expected if decreasing terrestrial OC was driving the observed decline in CO_2 emissions (due to a positive DOC-CO_2 relationship; e.g., Lapierre & del Giorgio, 2012).

3.2. Autochthonous Effects on CO_2 Fluxes

Many northern lakes have exhibited increases in planktonic diatoms with climate change (Rühland et al., 2008), yet available summertime water column chlorophyll *a* concentrations ($n = 27$, data available for 6 years from 1997 to 2017) indicated an apparent weak downward trend in planktonic primary productivity in our study lakes (Figure S7; Bayesian $p = 0.792$). Mesocosm experiments have indicated that warming may reduce CO_2 emissions by augmenting phytoplankton cell-specific production rates without influencing biomasses (Hanson et al., 2003). However, declining summer phosphorus (as total P) concentrations (Figure S8; Bayesian $p = 0.970$) in our study lakes indicate that daily rates of planktonic GPP are unlikely to have increased, as phosphorus was likely the limiting nutrient for phytoplankton in these lakes, since summertime nitrogen to phosphorus ratios typically ranged from 40 to 60 (Guildford & Hecky, 2000).

However, this does not rule out the possibility of incrementally longer growing seasons increasing planktonic GPP in these lakes on an annual timescale.

Although there is little evidence to support increases in daily phytoplankton GPP rates, it is possible that benthic algae (periphyton) GPP has increased in these lakes. The sediment-water interface (considering both sediment respiration rates and benthic GPP) can be an influential zone for determining CO_2 emissions in northern lakes (Ask et al., 2012; Kortelainen et al., 2006; MacIntyre et al., 2018). Furthermore, oligotrophic lakes around the world are experiencing increases in periphyton biomasses (Hudon et al., 2014; Nozaki, 2001; Timoshkin et al., 2015). Periphyton production frequently dominates primary production in shallow, oligotrophic high-latitude lakes (Ask et al., 2012; Karlsson et al., 2009; Sierszen et al., 2003; Vadeboncoeur et al., 2003), and has been shown to support net autotrophy (implying undersaturation of CO_2 relative to the atmosphere) in northern nutrient-poor lakes, even when they feature high DOC concentrations (Andersson & Brunberg, 2006; Vesterinen et al., 2016). There are no historical periphyton data for these lakes or other lakes in this region, yet periphyton GPP likely represented a high proportion of the total primary production in our oligotrophic study lakes, as they were shallow (median measured water column depth = 3 m) and clear enough that maximum depths were consistently observed within the photic zone. Periphyton GPP is highly responsive to changes in water temperature (DeNicola, 1996), which in our study lakes increased significantly (Figure 4b; Bayesian $p = 0.988$). Increases in conductivity (Figure 3b), and specifically calcium (Figure S9; Bayesian $p > 0.999$) and magnesium (Figure S10; Bayesian $p > 0.999$) concentrations were also observed in our study lakes, which can increase benthic GPP by stimulating enzymatic processes and improving access to nutrients (Drerup & Vadeboncoeur, 2016; Smith et al., 1987; Whitton et al., 1991).

A possible shift to greater autotrophy with warming in our lakes would agree with a recent finding that net ecosystem production (NEP, defined as the difference between gross primary production and community respiration) in boreal lakes in Quebec, Canada is positively linked to temperature (Bogard et al., 2020). However, as warming continues, further metabolic changes (i.e., an increase in net heterotrophy; Yvon-Durocher et al., 2010) due to rising water temperatures or increasing terrestrial productivity (Hutchins et al., 2020) may eventually lead to a return to prior lake CO_2 emission rates. In other words, it is possible that our observed trend in these lakes is only an initial response to climatic warming in lakes featuring seasonal ice-cover, and that the response of lakes to climate change may, as has been observed in terrestrial systems (Melillo et al., 2017), change over longer timescales. Furthermore, DIC would be taken up by planktonic and benthic primary producers (particularly CO_2, but also potentially bicarbonate), yet DIC concentrations increased over the study period (Figure 3a; Bayesian $p > 0.999$). While this does not rule out a possible increase in annual or daily GPP rates, it may indicate that another process is also responsible for the decline in CO_2 emissions.

3.3. Water Chemistry Effects on CO_2 Fluxes

Although not necessarily independent of the metabolic processes described above, chemical changes due to lengthening ice-free periods may also be an important driver of declining CO_2 emissions from these lakes. Long-term ice-cover data are not available for our study lakes, and projections for this particular region have not been previously modeled due to a paucity of data (Sharma et al., 2019). However, it is reasonable to assume that these lakes have experienced declines in ice-cover duration in recent decades given our observed trend in surface water temperatures (Figure 4b), and significant declines in regional sea ice-cover from 1971 to 2016 (Environment Canada, 2016; Welch's Two-Sample t test, $p < 0.0001$). Furthermore, increased ice-free periods have been widely observed in lakes across northern latitudes (Hewitt et al., 2018; Sharma et al., 2019), including those in regions to the north (Mueller et al., 2009) and south (Finlay et al., 2015) of our study region.

Shorter ice-cover periods can result in increased pH, attributed to lengthening growing seasons (i.e., increased annual CO_2 drawdown) and shortening underice periods (i.e., decreased CO_2 accumulation in the water column; Finlay et al., 2015), as well as increases in bicarbonate concentrations (reflected in conductivity, and attributed to increased evaporation/precipitation ratios; Smol & Douglas, 2007). Evaporative losses from lakes are likely increasing globally, with ice-cover loss being the primary driver in medium-latitude

and high-latitude lakes (Wang et al., 2018; Xiao et al., 2018). This effect has led to the complete desiccation of some high Arctic ponds in Canada (Smol & Douglas, 2007). Mean summertime specific conductivity values increased significantly in our study lakes over the study period (Figure 3b; Bayesian $p > 0.999$), which is an indicator of increasing evaporative losses due to decreased ice-cover duration in northern Canadian lakes underlain by shallow granitic bedrock (Smol & Douglas, 2007). The pH of our study lakes increased from roughly 7.3 in 1994 to over 8 by 2017 (Figure 3c; Bayesian $p > 0.999$), likely resulting from a significant reduction in CO_2 concentrations, as well as increased bicarbonate concentrations, reflected in increases in measured total alkalinity (Figure 3d; Bayesian $p > 0.999$). A rise in pH values (from ~8.2 in 1995 to 9.5 in 2010) described previously for hardwater lakes featuring declining CO_2 emissions in Saskatchewan (Finlay et al., 2015) spanned a higher pH range than that measured in our study lakes (Figure 3c), making chemical enhancement of CO_2 uptake a significant driver in those lakes, but not in ours (calculated chemical enhancement augmented flux estimates in our study lakes by ~2% on average). Nevertheless, longer growing seasons and a reduced respiratory winter CO_2 accumulation associated with shorter ice-cover periods could increase pH values over time, independent of increases in daily GPP rates.

4. Conclusions

We observed a significant 23 years decline in calculated pCO_2 (Figure 2) and CO_2 fluxes (Figure 4a) from 19 tundra lakes in northern Canada. Combining the available data for these study lakes with broader trends described for northern lakes, we propose three primary mechanisms that we believe may be working together to explain the observed decline in surface CO_2 emissions. These mechanisms are: (1) an increase in daily benthic GPP rates due to higher temperature and/or cation concentrations, (2) a decreased ice-cover period, leading to longer growing seasons (i.e., annual primary production rates) and shorter underice accumulation of CO_2, and (3) higher summertime evaporative losses (due to longer ice-off periods and warming temperatures) that could increase bicarbonate concentrations, thus shifting carbonate system equilibria. Although we cannot fully exclude the possibility of external drivers (e.g., increased groundwater connectivity increasing DIC concentrations), we consider such mechanisms to be unlikely in these lakes, given the shallow granitic nature of this landscape and the lack of concurrent changes in local precipitation (Figure S4). The data from our study lakes thus emphasize the potential variability of responses of northern lakes to the effects of climate change, allowing us to separate the effects of permafrost thaw and precipitation shifts from in-lake processes.

While the long-term trajectory of GHG emissions from northern aquatic systems is not settled, these results are important in confirming that not all northern lakes are following the predicted (Kraemer et al., 2017; Yvon-Durocher et al., 2010) response of aquatic system CO_2 fluxes to climate change. Specifically, we show that the response of lake biogeochemical fluxes to climate change is likely heavily influenced by the region being considered, emphasizing the necessity for data from such understudied regions (Metcalfe et al., 2018). With regards to lake pCO_2 and CO_2 emissions, empirical time series are extremely rare, yet provide important data that can lend insights into mechanistic shifts, in conjunction with modeled (Kraemer et al., 2017; Yvon-Durocher et al., 2010) and space-for-time (Serikova et al., 2019) approaches. In this study, we argue that a larger role for ice-period duration and primary production, in addition to the direct effects of temperature and permafrost thaw across regions featuring different underlying geologies, appears to be emerging.

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