River inflow dominates methane emissions in an Arctic coastal system

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Key Points:

- Methane concentrations in an Arctic estuary show strong seasonality; elevated concentrations are associated with early freshet river inflow
- Observations with a novel robotic kayak demonstrate that methane and carbon dioxide in the estuary are rapidly ventilated following ice melt
- River discharge is estimated to account for >95% of annual methane emissions from the estuary
Abstract
Measurements of greenhouse gases in Arctic waters are strongly biased toward low-ice summer conditions, with few observations during periods of seasonal ice retreat. We present a year-round time series of dissolved methane (CH\(_4\)) and nitrous oxide (N\(_2\)O), along with targeted observations during ice melt of CH\(_4\) and carbon dioxide (CO\(_2\)) in a river and estuary adjacent to Cambridge Bay, Nunavut, Canada. CH\(_4\) displayed dramatic seasonality, in contrast to limited seasonal changes in N\(_2\)O. During the river freshet, CH\(_4\) concentrations in the river and ice-covered estuary were up to 240,000% saturation and 19,000% saturation, respectively, but quickly dropped by >100-fold following ice melt. Observations with a robotic kayak revealed that river-derived CH\(_4\) and CO\(_2\) were transported to the estuary and rapidly ventilated to the atmosphere once ice cover retreated. We estimate that river discharge accounts for >95% of annual CH\(_4\) sea-to-air emissions from the estuary.

Plain Language Summary
The primary cause of recent global climate change is increasing concentrations of heat-trapping greenhouse gases in the atmosphere. Ongoing rapid Arctic climate change is affecting the annual cycle of sea ice formation and retreat, however most published studies of greenhouse gases in Arctic waters have been conducted during ice-free, summertime conditions. In order to characterize seasonal variability in greenhouse gas distributions, we collected year-round measurements of the greenhouse gases methane (CH\(_4\)) and nitrous oxide (N\(_2\)O) in a coastal Arctic system near Cambridge Bay, Nunavut, Canada. We found that during the ice melt season, river water contains methane concentrations up to 2000 times higher than the wintertime methane concentrations in the coastal ocean. We utilized a novel robotic kayak to conduct high-resolution mapping of greenhouse gas distributions during ice melt. From these data, we demonstrate that the river water containing elevated levels of methane and carbon dioxide (CO\(_2\)) flowed into the coastal ocean, and when ice cover melted, these greenhouse gases were rapidly emitted into the atmosphere. We estimate that in this system more than 95% of all annual methane emissions from the coastal ocean are driven by river inflow.

1 Introduction
Methane (CH\(_4\)) emissions from Arctic waters and sediments may accelerate in the future as part of positive feedback from ongoing climate change (Biastoch et al., 2011; James et al., 2016; Shakhova et al., 2010). Landscapes that were once permanently frozen are now seasonally thawing, and the ice-free season is lengthening in freshwater and marine systems (Magnuson, 2000; Stroeve et al., 2012; Zona et al., 2016). Thawing can result in the mobilization of labile organic matter and emissions of greenhouse gases such as carbon dioxide (CO\(_2\)), methane (CH\(_4\)), and nitrous oxide (N\(_2\)O) (Karlsson et al., 2013; Kvenvolden et al., 1993; Lamarche-Gagnon et al., 2019; Voigt et al., 2017; Zona et al., 2016). Studies of terrestrial and freshwater Arctic systems have demonstrated strong temporal variability in greenhouse gas emissions in these environments (Denfeld et al., 2018; Karlsson et al., 2013; Lamarche-Gagnon et al., 2019; Phelps et al., 1998; Voigt et al., 2017; Zona et al., 2016), yet published measurements in Arctic marine and estuarine waters are strongly biased toward summertime, low-ice conditions (Fenwick et al., 2017; Shakhova et al., 2010).
Ice acts as a barrier to gas exchange, sustaining strong disequilibria in gas concentrations between the atmosphere and ice-covered waters (Butterworth & Miller, 2016; Denfeld et al., 2018; Karlsson et al., 2013; Wand et al., 2006). Rapid re-equilibration of the mixed layer can occur following ice melt. Quantifying the impacts of sea ice loss on Arctic greenhouse gas emissions requires seasonally-resolved measurements; yet few measurements of dissolved CH\textsubscript{4} or other greenhouse gases are available in ice-covered or recently ice- liberated Arctic Ocean waters and connected estuaries. Here we present new observations that address this critical observational gap, demonstrating that the vast majority of annual CH\textsubscript{4} release in an Arctic estuary occurs during the ice melt period.

2 Observations, Results, and Discussion

2.1 Field observations

To quantify the annual sea-air emissions of greenhouse gases in a coastal Arctic system, we collected measurements in a well-sheltered bay with two inlets (west arm and east arm, Figure 1a) adjacent to the town of Cambridge Bay (Iqaluktuttiaq), Nunavut, Canada. Surface waters are seasonally ice covered, and the dominant freshwater source is Freshwater Creek, which discharges water into the east arm of Cambridge Bay from Greiner Lake and the associated watershed. Terrestrial snowmelt in this region typically begins in late May (Tedesco et al., 2009), and as a result, Freshwater Creek begins to flow before significant sea ice melt has occurred. This freshwater discharge causes the rapid melt of sea ice along the east arm, creating open water in June (Figure 1b), with the rest of the bay typically becoming ice-free 2–3 weeks later, in late June to early July. During 2017 and 2018, we collected a time-series of dissolved CH\textsubscript{4} and nitrous oxide (N\textsubscript{2}O) measurements in the estuary and river (Figures 1 and S1). Additionally, in 2018, we used a remotely operated robotic kayak, the ChemYak (Kimball et al., 2014; Nicholson et al., 2018), to characterize fine-scale spatiotemporal changes in dissolved CH\textsubscript{4} and carbon dioxide (CO\textsubscript{2}) in the estuary during peak river inflow (Figures 2–3), and collected water samples from Greiner Lake. Methodological details for bottle samples and the ChemYak are provided in the Supporting Information.

During winter and spring (January–May) in 2017 and 2018, CH\textsubscript{4} concentrations throughout the estuary water column (station B1 in Fig. 1b) were closely distributed around the atmospheric equilibrium of 4 nM (range 3–10 nM). In early June, river discharge from the spring thaw began to enter the estuary, and elevated CH\textsubscript{4} concentrations up to 860 nM (19,000% saturation) were measured in near-surface waters of Cambridge Bay (2 m below the surface of the ice). A water mass analysis using salinity and water isotope data from station B1 confirmed that the elevated CH\textsubscript{4} concentrations were associated with river runoff rather than ice melt (Figure S2 and Text S1). Ice-free summer surface waters sampled in July 2017 and 2018 had much lower CH\textsubscript{4} concentrations, ranging from 4–65 nM.

From 28 June to 2 July 2018, we used the ChemYak for high-resolution spatial mapping and vertical profiling of CH\textsubscript{4}, CO\textsubscript{2}, salinity, and temperature distributions in a ~1 km\textsuperscript{2} open water area between the river mouth and the ice edge during the dynamic melt period (Figures 2, 3 and S3–S6). These ChemYak measurements confirmed elevated greenhouse gas concentrations in river-derived estuary water. The river-derived water occurred throughout the study area as a
shallow, fresh surface mixed layer (<2 m depth), separated from deeper waters by a sharp pycnocline (Figure 2 f-g). During the ChemYak measurement period, CH₄ concentrations in the surface water decreased as the water flowed from the river toward the coastal ocean, suggesting a rapid ventilation within the estuary. For example, the CH₄ concentration in Freshwater Creek decreased from 560±10 nM on 27 June to 290±20 nM by 3 July, whereas CH₄ at station B1 was 130±10 nM on 3 July (Figure 4b). In the ChemYak sampling area (between Freshwater Creek and station B1), on 28 June, CH₄ and CO₂ concentrations in the upper 1 m of the water column were up to 470 nM and 1470 μatm, respectively (mean 410±20 nM and 1340±40 μatm). Concentrations in the upper 1 m decreased over the campaign to 150±70 nM CH₄ and 600±150 μatm CO₂ by 2 July (Figures 3 and S6).
**Figure 1.** Map of study site and time-series data.  

**a** Map of eastern Canadian Arctic showing the location of Cambridge Bay.  

**b** Satellite image of study area on 21 June 2017 (obtained by Google, DigitalGlobe). Pink circles indicate the locations of the main sampling stations FWC (Freshwater Creek) and B1, as well as ONC (Ocean Networks Canada observatory with ice profiler). The approximate region where the ChemYak was deployed is shown with a green outline. Time-series of CH$_4$ concentrations in Cambridge Bay estuary and Freshwater Creek in **c** 2017 and **d** 2018. Surface samples in Cambridge Bay were collected at 2 m depth below the ice surface, or 0.75 m below the open water surface. The dashed horizontal line represents atmospheric equilibrium and the dashed vertical line indicates when sampling station B1 became ice-free. Error bars reflect the standard deviation of duplicate measurements.
Figure 2. Spatial observations made by the ChemYak vehicle. a The study site with ChemYak trajectories from each day overlaid. The mouth of Freshwater Creek (69.1257 °N, 105.0042 °W) is marked with a star, and concentric rings at increments of 500 m centered at the mouth are provided for scale. Northeast of the red dashed line lies Freshwater Creek (red arrow and box) and a small embayment (blue label and box) which receives input from a much smaller river. b-g Observations made by the ChemYak for two representative days, 29 June and 2 July, are plotted by depth versus distance from the Freshwater Creek mouth. Negative distances (to the right of the axis) represent points northeast of the mouth (a small embayment) and positive distance (to the left of the axis) represent points southwest of the mouth (downstream). As indicated by the salinity plots (f-g), the mixed layer depth is <2 m throughout the study area, and the fresh surface layer was generally higher in both CH₄ and CO₂ concentration than layers deeper than 2 m. The gas concentrations decreased over the multi-day measurement campaign. Equivalent plots and temperature data for the other measurement days are shown in Figure S5.
Figure 3. Spatial and temporal trends observed by the ChemYak. a-b Each day of the measurement campaign is marked with a unique color, and samples collected are binned into 0.25 m increments from the surface to 6 m. Both CH$_4$ (a) and CO$_2$ (b) exhibit decreasing trends for each subsequent day, and there is strong stratification between the surface layer and water below 2 m. Error bars represent the standard deviation of measurements for each depth bin. c-j Temperature-salinity plots showing changes in CH$_4$ (c-f) and CO$_2$ (g-j) concentrations and water mass distributions over the time-series.
In the estuary, at depths below the mixed layer (>2 m depth), CH$_4$ and CO$_2$ concentrations decreased from 29 June to 2 July. Elevated wind speeds (up to 10 m s$^{-1}$) appear to have enhanced mixing across the sharp pycnocline on 30 June (Figures 3d, 3h, S5, and S6). On 30 June, the depth of the pycnocline shoaled and CH$_4$ and CO$_2$ concentrations below the mixed layer increased near the ice edge and the river mouth. Over the following days, lower wind speeds (3.7±1.1 m s$^{-1}$), coupled with decreasing river inflow concentrations and restratification of the water column, led to decreased gas concentrations throughout the water column by 2 July. Changes in the observed temperature-salinity properties of the water suggest that mixing reduced the vertical salinity gradient over the measurement period. The mixed layer near the river mouth showed significant warming between 28 June and 2 July (Figures 3c-j, S5, and S6).

To evaluate the importance of atmospheric ventilation to the CH$_4$ budget in the estuary, we performed sea-air flux calculations (Wanninkhof, 2014) using observed wind speeds. In the absence of lateral transport and river discharge, CH$_4$ and CO$_2$ concentrations in the estuary over our sampling period would be expected to decrease to ~70 nM and ~570 μatm, respectively. In actuality, we observed a mean surface CH$_4$ concentration of ~150 nM at the end of the sampling period, suggesting that the continued inflow of high-CH$_4$ river water into the ChemYak sampling region contributed to maintaining elevated CH$_4$ concentrations following ice melt (Figure 4a). Based on the observed river discharge of ~40 m$^3$ s$^{-1}$, we estimate that the residence time of water in the ChemYak measurement region was ~0.6 d.

In addition to measuring the estuary downstream of the Freshwater Creek mouth, the ChemYak was also used to collect observations in a small embayment at the outlet of a much smaller river on 29 June, 1 July, and 2 July (Figures 2, 3, S4, and S5). This embayment generally exhibited higher temperatures and lower CH$_4$ and CO$_2$ concentrations relative to adjacent waters. For example, on 29 June, the mean CH$_4$ concentration in the upper 2 m was 242±41 nM in the embayment, in contrast to 417±31 nM within 100 m downstream of the river mouth. For CO$_2$, the mean concentration was 790±140 μatm in the embayment and 1400±110 μatm downstream of the river mouth. In late June to early July 2018, we collected bottle samples at the head of the embayment in this smaller river and found that CH$_4$ concentrations in Freshwater Creek were two times higher than in the smaller river. The CH$_4$ and CO$_2$ levels in the embayment may therefore reflect lower inflowing CH$_4$ and CO$_2$ from the smaller river, and/or a longer residence time for river-derived surface water to exchange with the atmosphere in the embayment.

Overall, we conclude that the declining CH$_4$ and CO$_2$ concentrations throughout the water column between 28 June and 2 July, and along the spatial gradient from Freshwater Creek to station B1, primarily reflect a combination of decreasing gas concentrations in the river water (Figure 1c), loss due to gas exchange within the ice-liberated area, and lateral/vertical mixing. Below, we present a more complex physical model for the estuarine mixed layer CH$_4$ budget, and discuss potential impacts of microbial processes on the CH$_4$ budget. The observed differences between the smaller river and embayment compared to Freshwater Creek and the rest of the estuary demonstrate the need to conduct studies in a diverse range of Arctic coastal systems to better understand the complex hydrological controls on the magnitude and location of greenhouse gas emissions.
Figure 4. Observations and model-derived output of CH$_4$ delivery to the estuary mixed layer. a Historical river discharge data from Freshwater Creek. b Modeled and measured CH$_4$ concentration in the river (Freshwater Creek) and the Cambridge Bay estuary mixed layer (Model Regions 1 and 2), based on data and the model. The range of modeled values across the estuary is shown with grey shading and the range of values measured with the ChemYak is shown with blue symbols (error bars represent the standard deviation of daily measurements). The red symbols represent bottle measurements at station B1, and the black outlined symbols represent measurements at Freshwater Creek. The grey dotted line shows the equilibrium concentration in the estuary. c-d Modeled daily (c) and cumulative (d) CH$_4$ delivery to the estuary, from river inflow, ice melt, and gas exchange (positive delivery represents an input to the mixed layer). The CH$_4$ delivery caused by ice melt is negligible relative to the other terms.
2.2 Mixed layer model

To quantify the fate of river-derived CH$_4$ over the entire river inflow season, we developed a mixed layer model for the Cambridge Bay estuary (Figures 4 and S7). The model was constrained with measured CH$_4$ concentrations in Freshwater Creek (bottle samples), river discharge and wind speed measurements, and ice thickness records from the Ocean Networks Canada (ONC) cabled observatory (Figure S8 and Text S1). The results of this analysis suggest that the annual CH$_4$ cycle in the estuary is driven by river inflow, with sea ice melt contributing a comparatively negligible amount of CH$_4$ to the mixed layer (Figure 4d). This conclusion is consistent with a water mass property (salinity/water isotope) analysis showing an insignificant impact of sea ice melt on the CH$_4$ budget (Figure S2 and Text S1). The model was able to simulate the observed changes in estuarine CH$_4$ concentrations (measured at station B1 and with the ChemYak) without including an additional sink for microbial oxidation of CH$_4$ within the bay.

Using the model, we estimate that ~800 kg CH$_4$ was released from Freshwater Creek into the estuary in 2018 (volume-weighted mean concentration in river water of 360 nM), with 20% (160 kg) ventilated to the atmosphere from the estuary following ice cover retreat (Figure 4d). The remaining 80% of the river-derived CH$_4$ was laterally transported across the estuary beneath sea ice to the coastal ocean, where it was likely ventilated to the atmosphere following ice melt in mid-late July. Indeed, a model run with a larger spatial footprint (including 60 km$^2$ of coastal ocean surrounding the Cambridge Bay estuary) yielded a cumulative annual sea-air emission of 640 kg CH$_4$ from the coastal ocean derived from river discharge, which occurs rapidly following ice melt (Figure 4d). To estimate the CH$_4$ emissions from the estuary in the absence of river inflow, we prescribe a fixed CH$_4$ concentration of 6 nM (~150% saturation), the typical surface concentration before and after the river inflow period. Under this scenario, with no river discharge, we derived annual estuarine CH$_4$ emissions of 7.4 kg, 22 times lower than the emissions derived from the full model including riverine CH$_4$ inputs. Given that our model predicts that 80% of the riverine CH$_4$ is ventilated beyond the estuary in the adjacent coastal ocean, small river systems such as Freshwater Creek may be of primary importance to annual methane budgets through much of the coastal Arctic Ocean. Accurate calculation of such short-lived, high magnitude CH$_4$ emissions following ice melt, requires a more extensive under-ice sampling program, including melt-season measurements in multiple river-influenced Arctic estuaries and coastal systems.

2.3 Comparison to other systems

The CH$_4$ concentrations we measured in Freshwater Creek (ranging from 10–11000 nM, volume-weighted mean 360 nM) are similar to other river systems in the Arctic and worldwide. For example, mean CH$_4$ concentrations observed in the Yukon River, Lena Delta, and Leverett Glacier runoff range from 70–750 nM (Bussmann, 2013; Lamarche-Gagnon et al., 2019; Striegl et al., 2012). Furthermore, a data compilation of over 900 rivers and streams worldwide reported a mean CH$_4$ concentration of 1400 ± 5200 nM and median of 250 nM (Stanley et al., 2016). The peak CH$_4$ concentrations observed in Cambridge Bay estuary (up to 900 nM during the freshet, prior to ice melt) are similar to maximum values measured in other Arctic coastal waters (Bussmann et al., 2017; Shakhova et al., 2010).
In contrast to CH$_4$, N$_2$O concentrations throughout 2017-2018 measured at station B1 and Freshwater Creek displayed limited seasonal variability (Figure S1). During open water conditions in Cambridge Bay, N$_2$O was typically undersaturated or close to equilibrium in near-surface waters. Anoxic conditions in the sediments and water column that promote CH$_4$ accumulation may simultaneously drive N$_2$O consumption via denitrification (Naqvi et al., 2010). These conditions contrast with the lower- and mid-latitudes where rivers and estuaries are typically considered significant N$_2$O sources (Seitzinger & Kroeze, 1998; Seitzinger S.P. et al., 2000).

2.4 Future work

This research motivates two directions for future work in Cambridge Bay: characterization of natural sinks of CH$_4$ (e.g., microbial oxidation) and identification of CH$_4$ sources. The extent of microbial CH$_4$ oxidation in river-derived water transported from Freshwater Creek to the coastal ocean is currently unknown. The early-season river discharge containing >1000 nM CH$_4$ may remain under the ice for ~1 month before being exposed to the atmosphere, during which time microbial oxidation could potentially decrease concentrations. Recent studies of CH$_4$ oxidation rates in Arctic estuarine and marine waters have found rates ≤2 nM CH$_4$ d$^{-1}$ (Bussmann et al., 2017; Gentz et al., 2014; Mau et al., 2013; Uhlig & Loose, 2017). Oxidation rates ≤2 nM CH$_4$ d$^{-1}$ would have a small impact on under-ice CH$_4$ levels in Cambridge Bay estuary (considering that we measured up to 860 nM CH$_4$ under ice in the estuary). However, some studies in ice-covered lakes have reported significantly higher oxidation rates (Bastviken et al., 2002; Ricão Canelhas et al., 2016). Additionally, our model reproduces the seasonally observed progression of CH$_4$ levels in the estuary without invoking CH$_4$ oxidation. To further investigate the extent and dynamics of under-ice CH$_4$ plumes, robotic underwater vehicles and moored sensors could be developed and deployed to enable high resolution under-ice measurements. Tracer-based incubation experiments could be performed to measure oxidation rates (Uhlig & Loose, 2017).

Another area of future research is determining CH$_4$ sources and characterizing the relative emissions of CH$_4$ and CO$_2$ from the lake versus the river and coastal ocean. Our preliminary measurements in Greiner Lake suggest that anoxic zones in the lake are a significant source of CH$_4$. However, the formation mechanism is not clear. For example, CH$_4$ could be generated in situ each winter within anoxic sediments and/or overlying anoxic waters, or, alternatively, could be liberated from existing pools in methane seeps or thawing permafrost. More extensive measurements of CH$_4$ concentration and isotopic composition in the watershed surrounding Freshwater Creek would assist in determining the source of the CH$_4$. Additionally, radiocarbon measurements would demonstrate whether ancient CH$_4$ sources such as thawing permafrost are significant (Sparrow et al., 2018).

This study demonstrates the importance of fully resolving seasonal processes in inter-connected marine and freshwater environments to accurately quantify CH$_4$ emissions from Arctic systems. The results motivate future coastal Arctic field campaigns at other sites with measurement technologies capable of high spatial and temporal resolution mapping immediately before and during ice melt. Such studies will provide critical information to characterize current and future Arctic greenhouse gas emission, improving quantitative estimates of changes in CH$_4$ and CO$_2$ emissions across the rapidly changing Arctic environment.
3 Conclusions and implications

Our results, derived from a year-round times series of CH$_4$ measurements and dense spatio-temporal observations from a remotely-operated robotic kayak, show that CH$_4$ discharge via Freshwater Creek drives intense CH$_4$ emissions immediately following ice melt in the Cambridge Bay estuary and surrounding waters. River discharge also acts as a significant seasonal source of CO$_2$ to the estuary, whereas N$_2$O shows limited seasonality. Samples collected exclusively during low-ice summer conditions would likely miss the primary driver of the annual CH$_4$ budget, and underestimate the sea-air fluxes. Similar seasonal variability in CH$_4$ emissions likely occurs in other river-influenced, seasonally ice-covered Arctic estuaries, which receive ~10% of global river discharge (Dai & Trenberth, 2002). Our results suggest that the ongoing and projected future increases in Arctic river discharge (Macdonald et al., 2015) could drive increased CH$_4$ and CO$_2$ emissions from Arctic estuaries. The low and stable CH$_4$ concentrations observed below the mixed layer in the estuary indicate that sedimentary CH$_4$ sources within the estuary are negligible relative to river-derived inputs, in contrast to published studies in some other Arctic coastal and shelf systems where significant sedimentary sources are observed (Gentz et al., 2014; Shakhova et al., 2010).

Acknowledgments, Samples, and Data

All data generated by the authors that were used in this article have been submitted to PANGAEA, where publication is pending, and a PANGAEA DOI will be included in the final published manuscript. Code for the biogeochemical model used to estimate CH$_4$ transport is available on GitHub (https://github.com/caramanning/cambridge-bay-model). We acknowledge the use of imagery from the NASA Worldview application (https://worldview.earthdata.nasa.gov), part of the NASA Earth Observing System Data and Information System (EOSDIS), and data from Ocean Networks Canada, and Environment Canada. We thank everyone involved in the fieldwork including C. Amegainik, Y. Bernard, A. Cranch, F. Emingak, S. Marriott, and A. Pedersen. Laboratory analysis and experiments were performed by A. Cranch, R. McCulloch, A. Morrison, and Z. Zheng. We thank J. Brinckerhoff, the Arctic Research Foundation, and the staff of the Canadian High Arctic Research Station for support with field logistics. Funding for the work was provided by MEOPAR NCE funding to B. Else, a WHOI Interdisciplinary Award to A. Michel., D. Nicholson. and S. Wankel, and Canadian NSERC grants to P. Tortell. and B. Else. Authors received fellowships, scholarships and travel grants including an NSERC postdoctoral fellowship to C. Manning., an NDSEG fellowship to V. Preston., NSERC PGS-D and Izaak Walton Killam Pre-Doctoral scholarships to S. Jones, Northern Scientific Training Program funds (Polar Knowledge Canada) to S. Jones. and P. Duke.. We also thank Polar Knowledge Canada (POLAR) and Nunavut Arctic College for laboratory space and field logistics support.

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