First estimation of the diffusive methane flux and concentrations from Lake Winnipeg, a large, shallow and eutrophic lake

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Abstract

Freshwater lakes are increasingly recognized as significant sources of atmospheric methane (CH_4), potentially offsetting the terrestrial carbon sink. We present the first study of dissolved CH_4 distributions and lake-air flux from Lake Winnipeg, based on two-years of observations collected during all seasons. Methane concentrations across two years had a median of value of 24.6 nmol L^{-1} (mean: 41.6 ± 68.2 nmol L^{-1}) and ranged between 5.0 and 733.8 nmol L^{-1}, with a 2018 annual median of 24.4 nmol L^{-1} (mean: 46.8 ± 99.3 nmol L^{-1}) and 25.1 nmol L^{-1} (mean: 38.8 ± 45.2 nmol L^{-1}) in 2019. The median lake-air flux was 1.1 µmol m^{-2} h^{-1} (range: 0.46 – 70.1 µmol m^{-2} h^{-1}, mean: 2.9 ± 10.2 µmol m^{-2} h^{-1}) in 2018, and 5.5 µmol m^{-2} h^{-1} (range: 0.0 – 78.4 µmol m^{-2} h^{-1}, mean: 2.7 ± 8.5 µmol m^{-2} h^{-1}) in 2019, for a total diffusive emission of 0.001 Tg of CH_4-C yr^{-1}. We found evidence of consistent spatial variability, with higher concentrations near river inflows. Significant seasonal trends in CH_4 concentrations were not observed, though fluxes were highest during the fall season due to strong winds. Our findings suggest Lake Winnipeg is a CH_4 source of similar mean magnitude to Lake Erie, with lower concentrations and fluxes per unit area than smaller mid- to high-latitude lakes. Additional work is needed to understand the factors underlying observed spatial variability in dissolved gas concentration, including estimations of production and consumption rates in the water column and sediments.

Keywords: Methane, Carbon Cycling, Lake Winnipeg, Greenhouse Gases

Introduction

Methane (CH_4) is a potent climate-active gas that accounts for 20 percent of total greenhouse warming (IPCC, 2013), and whose atmospheric concentration has increased 2.5-fold since the pre-industrial period (Wolff and Spahni, 2007). While the largest atmospheric sources of CH_4 are anthropogenic, natural production of this gas is an important carbon loss pathway from terrestrial and aquatic systems, offsetting an estimated 25 percent of the terrestrial carbon sink with lakes and rivers, and accounting for approximately 12 percent of the non-anthropogenic CH_4 flux to the atmosphere (Bastviken et al., 2011, 2004; Huutenen et al., 2003; Kirschke et al., 2013). Ongoing eutrophication is expected to increase the CH_4 flux from lakes over the coming decades, potentially making lakes the largest natural source of CH_4 to the atmosphere (Bastviken et al., 2011; Beaulieu et al., 2019).

The majority of CH_4 in lakes is produced in anoxic sediments by methanogenic archaea during anaerobic metabolism (Bastviken et al., 2004; Borrel et al., 2011). Water column production can occur under anoxic conditions, and oxic CH_4 production has also been recently documented (Bastviken et al., 2004; Borrel et al., 2011; Bogard et al., 2014; Peeters et al., 2019). Much of the CH_4 produced in anoxic environments is oxidized to CO_2 by methanotrophic bacteria in oxygenated waters, but surface water CH_4 super-saturation is still a common feature in lakes, driving net CH_4 evasion to the atmosphere (Bastviken et al., 2004). Previous research has shown that lake CH_4 concentrations and emissions vary with latitude, and are influenced by the physical and biological characteristics of the waterbody, including temperature, nutrient status, algal communities, predominant form of organic matter, and mixing patterns (Bastviken et al., 2004; Emilson et al., 2018; Fernandez et al., 2020; Rasilo et al., 2015). Indeed, lake CH_4 fluxes have shown close relationships with total phosphorus, biological production, and sediment temperature (DelSontro et al., 2016). Accordingly, temperate, subarctic and boreal lakes tend to have relatively lower CH_4 flux per unit area than tropical lakes, but their larger combined surface area makes them a significant CH_4 source to the atmosphere (Bastviken et al., 2011, 2004; Gonzalez-Valencia et al., 2014; Kirschke et al., 2013).

Roughly nine percent of Canada is covered by lakes, and this country contains eight of the twenty largest lakes in the world (Statistics Canada, 2018). To date, there have been

relatively few studies of CH_4 emissions from North American great lakes, with recent exceptions from Lake Erie, Michigan, and Superior (Beeton, 1963; Fernandez et al., 2020; Joung et al., 2019; Lu et al., 2010). Here, we present dissolved CH_4 measurements and estimates of CH_4 flux from Lake Winnipeg, a large, shallow mid-latitude lake in Manitoba. The lake, which is the 11th largest by surface area in the world, has been relatively well-studied in terms of its physical characteristics, nutrient status, and fisheries (Environment and Climate Change Canada [ECCC] and Manitoba Agriculture and Resource Development [MARD], 2020), yet no published measurements of CH_4 concentrations or flux estimates exist. The objectives of this research are to estimate air-water flux during open water season in Lake Winnipeg, characterize the spatial and temporal (seasonal) variability in dissolved methane distributions, and to identify key drivers of spatiotemporal CH_4 variability.

Methods

Study Area - Lake Winnipeg

Lake Winnipeg has a total surface area of 23,750 km^2 (ECCC and ARD, 2020), with a watershed that occupies nearly one million square kilometers. The western half of the watershed is dominated by agriculture and prairie landscape largely overlying carbonate rocks, while the eastern side is dominated by boreal forest, shield lakes, and some hydroelectric development (Levesque and Page, 2011). The basin consists of granitic bedrock to the east and carbonate bedrock to the west, overlain in some places by glacial till and/or fine-grained clay to sandy sediments of varying thicknesses up to 100 m (Levesque and Page, 2011; Todd et al., 1997). The lake is typically ice-covered from late November to May (McCullough, 2020).

The lake consists of a north and south basin, separated by a constricted section referred to as the narrows (Fig.1; Levesque and Page, 2011). The two basins are significantly different from each other in terms of their water chemistry and physical characteristics (Watchorn, 2020). The average depth is 8 m in the south basin and 14 m in the north basin, with the deepest point at just over 60 m located in the narrows (Levesque and Page, 2011). The north basin is cooler than the south basin (Levesque, 2011; McCullough and Levesque, 2011), though both water and air temperatures have increased over the last several decades (Smith, 2020). Total suspended solids (TSS) are generally higher in the south basin than the north basin, which

promotes higher primary productivity in the north basin (McCullough and Levesque, 2011). The lake has a large fetch, approximately 400 km north to south, 111 km west to east in the north basin, and 40 km west to east in the south basin, which allows for seiches to occur (Levesque and Page, 2011). Due to its large fetch and shallow depth Lake Winnipeg is completely polymictic in the south and north basins, stratifying rarely, though increasingly often in the north basin during the open water season, and occasionally beneath the ice in the winter (McCullough and Levesque, 2011; McCullough, 2020). The water column remains oxygenated year-round, with the lowest recorded dissolved oxygen concentration of below 2.5 mg L^{-1} at the bottom of the lake in the north basin in summer of 2003 (McCullough and Levesque, 2011). Less than 3% of observations from 2008 – 2016 had dissolved oxygen concentrations of less than 5 mg L^{-1} (McCullough, 2020), suggesting methanogenesis is largely restricted to sediments.

Lake Winnipeg has been characterized as a hypereutrophic lake that frequently experiences large scale algal blooms, and these are typically most severe along the eastern half of the lake's north basin (Binding 2020). Primary inflows to the lake are the Winnipeg River (43 percent of inflow) and Red River (12 percent of inflow). However, the Red River is by far the largest single nutrient source to the lake, supplying 69 percent of the total phosphorus and 34 percent of the total nitrogen to the lake (Delavau and Lee, 2020; ECCC and ARD, 2020; Wiebe, 2020). The lake drains to the northeast into the Nelson River, which flows to Hudson Bay. The lake level has been regulated through structures along the Nelson River for hydroelectric purposes since the 1970s (Watchorn, 2020), which has reduced the frequency of extreme lake levels by increasing outflow during high water years, and reducing outflow during low water years (Levesque and Page, 2011). The shoreline is predominantly rocky with some sandy beaches. Extensive marshland can be found near the mouths of some rivers, the largest being the Netley-Libau marsh at the south end of the lake near the mouths of the Red River and Netley Creek (Watchorn et al., 2012). Large amounts of commercial agricultural development are in Lake Winnipeg's southern and western portions of its watershed, across three provinces and four American states (Watchorn, 2020; Wiebe, 2020). The eastern and northern regions drain from predominantly forested areas (Watchorn, 2020).

Sampling

Sampling was conducted from the *MV Namao* during the summer (July 31 – August 16 2018; July 23 – August 8, 2019), fall (September 24 – October 22, 2018; September 16 – October 3, 2019), and spring (May 29 – June 13, 2019) cruises, and by helicopter during February 2019. The lake had a complete ice cover by December 2018. Near shore stations (designated by NS in Fig. 2) were sampled by a small boat deployed from the *Namao* in 1 – 3 m deep water (typically ~10 - 100m from shore). A total of 43 and 37 samples were taken in summer 2018 and fall 2018, respectively. In 2019, 35 stations were selected to be sampled. All stations were sampled at one-meter depth and an additional sample was collected one meter above the bottom at "W" stations (Fig. 2).

Water samples were collected using a rosette (SBE 55 ECO Water Sampler) equipped with six 2 L Niskin bottles, an SBE-43 Oxygen sensor, a Turner CYCLOPS-7 Turbidity sensor, a SEACATplus conductivity, temperature, depth sensor, and a Biospherical QSP-2300L photosynthetically active radiation (PAR) sensor providing information on dissolved oxygen, turbidity, transmitted PAR with depth at discrete stations of the cruise. Water samples for CH_4 analysis were collected from the rosette Niskins into 60 mL glass vials using gas-tight tubing and ensuring no air bubbles entered the water sample. The bottle was filled with no headspace after overfilling with three times the vial volume.

Winter samples were collected using a Kemmerer bottle, which was deployed through a hole cut in the ice and sampled in the same manner as described using the Niskin bottles, but instead into a 500 mL glass biological oxygen demand (BOD) bottle. These were flushed with three times the sample volume before capping with a sintered glass stopper. Nearshore station samples were collected by submerging a 500 mL BOD bottle with a piece of tubing to act as a snorkel, which allowed removal of air from the submerged bottle via the tubing without creating bubbles inside the bottle, and capped. Water for CH_4 analysis was then subsampled from the 500 mL glass BOD bottle, which was sealed and stored in the dark until processing, approximately thirty minutes after collection from nearshore sites, and 12-16 hours after sampling in the winter. A 60 mL glass syringe with tubing on the end was rinsed with sample water three times and used to transfer water from the BOD bottle to 60 mL serum bottles

without introducing bubbles. The bottles were overfilled by 50% at minimum. Prior to sealing the vials with a butyl rubber stopper and aluminum crimp seal, 40 µL of saturated mercuric chloride ($HgCl_2$) solution was added for preservation. The samples were stored in the dark at room temperature and were analysed by a purge-and-trap gas-chromatograph mass-spectrometer at the University of British Columbia, following methods outlined by Capelle et al (2015). This method provides precision of <3 % and a detection limit of <0.5 nmol CH_4 L^{-1}.

Instantaneous lake-air CH_4 flux estimation

Lake-air CH_4 flux (F, µmol m^{-2} h^{-1}) was estimated using the bulk flux equation (e.g., Wanninkhof, 2014):

$$F(CH_4) = k_{CH4}(C_w - C_a), \quad (1)$$

where k_{CH4} is the gas transfer velocity (m h^{-1}) for CH_4, C_w is the dissolved CH_4 of the sample and C_a is the equilibrium CH_4 concentration (nmol L^{-1}) that would be expected at the water surface boundary layer in contact with the atmosphere given the temperature and salinity of the water, and the atmospheric CH_4 concentrations. Atmospheric CH_4 concentrations were obtained from the nearest NOAA monthly sampling station in Park Falls, Wisconsin, ranging from 1894.4 to 2006.0 ppb (Dlugonecky et al. 2020). Equilibrium CH_4 concentrations were calculated following Wiesenburg and Guinasso (1979). The difference between measured C_w and calculated C_a (i.e., ΔC_{CH4}) is the excess concentration and was converted from nmol L^{-1} to µmol m^{-3} for input to Equation 1. The transfer velocity is often derived as a function of wind speed, and here we opted for the updated relationship for CO_2 reported by Wanninkhof et al., (2014):

$$k_{CO2\_600} = \left[0.251 \cdot U^2 \cdot \left({Sc_{CO2}}/{600}\right)^{-0.5}\right] \Big/ 100, \quad (2)$$

and scaled to CH_4 using:

$$\frac{k_{CO2\_600}}{k_{CH4\_600}} = \left(\frac{Sc_{CO2}}{Sc_{CH4}}\right)^{-0.5}, \quad (3)$$

where Sc_{CO2} and Sc_{CH4} are the Schmidt Numbers for CO_2 and CH_4 (Jähne et al.,1987; Wanninkhof et al., 2014) for freshwater (Sc=600), and U is the wind speed at 10 m elevation.

Annual Flux Estimation

To obtain an annual open water flux estimate, we used the 2019 dataset only, as no data was available for the spring season of 2018. We derived median daily flux values for each season where spring, summer, and fall were defined as May 1 – June 30; July 1 – August 31; and September 1 – November 30, respectively. We then multiplied median daily fluxes by the number of days during the corresponding season (i.e. spring = 61 days from May 1 – June 30), and then summed the three open-water seasons to obtain an annual flux. In doing so we assumed zero CH_4 flux in winter. The porosity of freshwater ice is very low at near freezing temperatures (Craig and Hayward, 1987; Gosink, 1976) and consequently gas diffusivity rates in freshwater ice are negligible (Hemminsen, 1959). Our assumption is that fluxes during the partial ice cover periods (roughly May and November) would not be reduced by the presence of ice, though this may be an oversimplification (e.g., Manning et al., 2019) which may lead to an overestimation in annual CH_4 loss to the atmosphere. Although we feel this assumption will have a small impact on annual total CH_4 loss, we intend to investigate it further in subsequent studies. We note that our annual lake fluxes derived using median values were ~50% lower than those calculated using means.

Results

Because of logistical constraints, not all stations shown in Figure 2 were sampled during each cruise. Despite these limitations, the distribution of samples in spring and summer provide a balanced representation of the lake north to south and east to west (Fig. 2). Sampling in winter was most constrained given the requirement for helicopter support. Consequently, we are missing winter samples in the northwest of the north basin and in proximity to the east shore of the south basin.

Average wind speed was greatest during the fall cruises (Table 1). In fall 2018, the average wind speed was 5.4 ± 1.6 m s^{-1} (range: 2.1 – 7.7 m s^{-1}) in the north basin, 6.8 ± 3.6 m s^{-1} (range: 3.1 – 11.3 m s^{-1}) in the south basin, and 5.7 ± 2.3 m s^{-1} (range: 2.1 – 11.3 m s^{-1}) across the entire lake. In fall 2019, the north and south basins showed higher wind speed variability and averaged 6.0 ± 4.1 m s^{-1} (range: 0.5 – 14.9 m s^{-1}) and 5.1 ± 2.4 m s^{-1} (range: 1.3 – 12.9 m s^{-1})

respectively in the north and south basins, with an average of 5.8 ± 3.7 m s^{-1} (range: 0.5 – 14.9 m s^{-1}) over the lake (Table 1). The lowest wind speeds were in spring 2019 (3.4 ± 2.1 m s^{-1}, range: 0.6 – 8.5 m s^{-1}) and summer 2019 (2.8 ± 1.5 m s^{-1}, range: 0.3 – 5.8 m s^{-1}). Average water surface temperatures across the lake were highest in the summer cruises, being 19.7 ± 1.3°C (range: 17.2 – 21.9°C) in 2018 and 20.7 ± 1.6°C (range: 17.2 – 23.3°C) in 2019, and lowest in fall of 2018 (8.1 ± 2.6°C, range: 3.2 – 12.2°C) and in the north basin during spring 2019 (8.3 ± 2.7°C, range: 4.9 – 13.1°C). The south basin on average was warmer than the north basin across all seasons (Table 1).

Across all the surveys, the median CH_4 concentration in Lake Winnipeg was 24.10 nmol L^{-1} (mean concentration of 41.6 ± 68.1 nmol L^{-1}) and ranged between 5.0 – 733.8 nmol L^{-1} (Table 2), while the median flux was 0.72 µmol m^{-2} h^{-1}, and ranged between 0 - 78.4 µmol m^{-2} h^{-1} (mean flux: 2.79 µmol m^{-2} h^{-1}). The CH_4 concentration and flux data were right-skewed (Kolmogrov-Smirnov, $\alpha = 0.05$), causing the median concentration to be lower than the mean (Table 2B; Fig.3). For this reason, we report mean and median values throughout, the former to allow for comparison with other studies.

The distribution of CH_4 concentration at the surface is shown in Figure 2. CH_4 concentrations ranged from 5.7 – 733.8 nmol L^{-1} in the south basin and 5.0 – 285.3 nmol L^{-1} in the north basin (Table 2), but the inter-basin CH_4 concentration differences were not statistically significant (Wilcoxon Rank-Sum, $\alpha = 0.05$). Similarly, we observed no significant difference between fluxes from the north and south basins. The highest CH_4 concentrations and fluxes overall were measured near the mouth of the Red and Winnipeg Rivers (stations 4NS and 7, respectively), during summer 2018 (Electronic Supplementary Material (ESM) Table S1). We also observed generally higher CH_4 concentrations in surface samples taken in nearshore areas and near to river outflows in both basins during all seasons (e.g., stations 4NS, 1, 2, 7, 7NS, 12NS identified in Fig. 2). The pattern is particularly strong in summer (2018 and 2019) and fall (2019). Interestingly, the CH_4 concentration in surface samples at the station closest to the Winnipeg River (station 7) was particularly high in the summer of 2018 (733.8 nmol L^{-1}), but not in the summer of 2019 (142.8 nmol L^{-1}). Aside from the high CH_4 concentrations near river mouths and nearshore stations, we also observed consistently high CH_4 concentrations during

all seasons at station W7, which is near the central part of the north basin and far from any river mouth. This station frequently had the highest concentrations of any stations sampled in the north basin (see ESM Table S1). Despite the likelihood of significant CH_4 diffusion from sediments, vertical CH_4 concentration gradients were weak, and differences between surface and bottom CH_4 concentrations were not statistically significant (Wilcoxon Rank-Sum, $\alpha = 0.05$).

We observed no statistically significant differences in annual CH_4 concentrations or fluxes between 2018 and 2019 (Wilcoxon Rank-Sum, $\alpha = 0.05$). The median flux was 1.13 μmol m^{-2} h^{-1} (range: 0.46 – 70.1 μmol m^{-2} h^{-1}, and mean of 3.0 ± 10.1 μmol m^{-2} h^{-1}) in 2018, and 0.55 μmol m^{-2} h^{-1} (range: 0.0 – 78.4 μmol m^{-2} h^{-1}, mean: 2.7 ± 8.5 μmol m^{-2} h^{-1}) in 2019, indicating the lake was consistently emitting CH_4 to the atmosphere during the open water season (Table 2A).

On seasonal time scales, we found no statistically significant differences in CH_4 concentrations (Table 2; Wilcoxon Rank-Sum, $\alpha = 0.05$). Despite the potential for CH_4 accumulation under ice, mean winter 2018 CH_4 concentrations were not statistically different from other seasons (Wilcoxon Rank Sum, $\alpha = 0.05$), but with a much smaller sample size ($n = 8$). In total, seven stations were consistently sampled at the surface depth during each cruise of our study. Of these, two stations exhibited the highest CH_4 concentrations during winter (W12 and W13; see Fig. 2), whereas winter CH_4 concentrations at stations W9 and W10 were lower than during any other cruise.

Maps of CH_4 flux during each survey are shown in ESM Figure S1, while their distribution is shown in Figure 3 for each cruise. Unlike CH_4 concentrations, we observed a clear seasonal trend for diffusive CH_4 fluxes (Table 2, Fig. 3). CH_4 fluxes during fall were significantly higher than during spring (Wilcoxon Rank Sum, $p < 0.0001$) and summer (Wilcoxon Rank Sum, $p < 0.01$). The largest diffusive fluxes were measured in fall 2019 (median: 2.4 μmol m^{-2} h^{-1}, range: 0.0 – 78.4 μmol m^{-2} h^{-1}), and the lowest median flux in spring of 2019 (0.3 μmol m^{-2} h^{-1}, range: 0.0 – 13.4 μmol m^{-2} h^{-1}). This result largely reflects the variability in wind speeds among surveys (Table 1), with higher wind speeds during 2019 driving the largest diffusive fluxes despite lower surface CH_4 concentrations. Fluxes for winter 2019 were not calculated, as ice cover was

present, and flux is assumed to be zero. Our estimated annual CH_4 flux during the open water season (May 01 – Nov 30) was 0.001 Tg CH_4-C y^{-1}.

Dissolved oxygen concentrations during our surveys ranged from 7.0 to 12.6 mg L^{-1}. A strong negative relationship between dissolved oxygen and dissolved CH_4 ($R^2 = 0.93$) was observed in the south basin during the summer of 2018, but no clear relationship in the north basin (Fig. 4A). In fall of 2018, there was no clear relationship between CH_4 and dissolved O_2 in either basin (Fig. 4B). Dissolved O_2 data was not available at the time of this publication for the 2019 surveys.

Discussion

Until very recently, there has been little published research on CH_4 fluxes and dynamics in North America's Great Lakes, with recent exceptions including Lake Erie, Michigan, and Superior (Fernandez et al., 2020; Juong et al., 2019). Concentrations and fluxes reported in these studies appear in Table 3. Results presented here show that Lake Winnipeg is a source of CH_4 to the atmosphere throughout the open water season, and concentrations show a high degree of horizontal spatial variability. The range in concentrations we observed in Lake Winnipeg is larger than that previously reported in other Canadian great lakes. Mean CH_4 concentrations in both Lakes Erie and Winnipeg are higher than reported for Lakes Michigan and Superior (Table 3; Fernandez et al., 2020; Juong et al., 2019).

Lake Winnipeg is of similar area to Lake Erie (~ 23,750 km^2 to 25,700 km^2) and, like Lake Erie in the 1960s (Beeton, 1963; Lu et al., 2010), Lake Winnipeg has undergone intense eutrophication since the late 1990s and early 2000s as the result of non-point source nutrient input (Environment Canada [EC] and Manitoba Water Stewardship [MWS], 2011). Previous work has established relationships between CH_4 and high concentrations of phosphorus and chlorophyll-*a* as a result of increased deposition of organic material to the sediments (Beaulieu et al., 2019). The productivity of a lake has also been linked to frequency of sub-surface anoxia in highly eutrophic lakes, which generally causes productive lakes to produce more CH_4 relative to oligotrophic lakes (Beaulieu et al., 2019; Casper et al., 2000; Gonzalez-Valencia et al., 2014). Average CH_4 concentrations and ranges in Lake Winnipeg were higher than reported for Lake Erie (5.7 – 733.8 nmol L^{-1} compared to 24.3 – 107.1 nmol L^{-1}; Table 3; Fernandez et al., 2020).

Note that while mean hourly fluxes suggest the lakes to be similar in their emission characteristics (Lake Winnipeg: 2.8 $\pm$ 9.2 μmol m^{-2} h^{-1}, and Erie: 2.3 μmol m^{-2} h^{-1}), the median flux from Lake Winnipeg in 2019 (0.55 μmol m^{-2} h^{-1}) is much lower than the mean flux reported for Lake Erie (Table 3). Median values were not reported for Lake Erie. Despite generally higher CH_4 concentrations, we infer that Lake Winnipeg is a lower annual source of CH_4 as Lake Erie (0.001 Tg C yr^{-1} versus 0.008 Tg C yr^{-1}), likely due to the shorter open water season of Lake Winnipeg.

Striking in Table 3 is the large reported range of CH_4 concentration for small boreal lakes (13 - 2,015 nmol L^{-1}; Bastviken et al., 2004) relative to Lake Winnipeg and the other North American Great Lakes. Rasilo et al. (2015) report dissolved CH_4 (as partial pressure - pCH_4) and CH_4 fluxes for boreal lakes over a 6-year study. In their study summertime pCH_4 averaged 191 μatm (ranging between 6 and 3612 μatm) across 224 boreal lakes in Quebec. In that work water temperature was not provided so we cannot convert their partial pressures to CH_4 concentration (nmol L^{-1}) for comparison with this study. We converted our CH_4 concentrations to partial pressures (following Warneck and Williams, 2012) and report that summertime pCH_4 across both basins in Lake Winnipeg averaged 43.1 μatm (ranging between 5.2 μatm and 475.1 μatm) in 2018 and 21.1 μatm (ranging between 3.2 μatm and 92.4 μatm) in 2019, which based on average partial pressure, is respectively 4 and 9 times lower than in the boreal lakes studied in Quebec.

Mean rates of CH_4 evasion from Lake Winnipeg appears to also be lower, and in some cases drastically so, relative to lakes surveyed in temperate, subarctic and boreal environments (Table 3; Bastviken et al., 2004; Fernandez et al., 2020; Juong et al., 2019; Rasilo et al., 2015). For small lakes (< 100 km^2) relationships have been observed (e.g., Rasilo et al., 2015) between summertime lake pCH_4 concentration and flux and lake properties, including lake area, temperature, nutrient concentrations and CDOM. By comparison Lake Winnipeg is a very large water body that drains an enormous area from a regionally diverse catchment. Currently there are not sufficient data to explore which underlying factors drive spatial and temporal variability in both the CH_4 concentration and flux within Lake Winnipeg, nor to explain differences between Lake Winnipeg's CH_4 dynamics relative to studies cited in Table 3. Here we report a

large degree of within lake variability in both the distribution of CH_4 concentration and flux. This is perhaps not unexpected as the two basins that make up Lake Winnipeg can be very different in terms of their biogeochemical, biological, and physical properties (Levesque and Wassenaar, 2011; McCullough and Levesque, 2011; Page, 2011). Observed patterns in CH_4 in Lake Winnipeg may thus reflect local forcing that at this time, with only five surveys of data, are difficult to extrapolate across the entire lake. Although we cannot definitively link CH_4 variability to specific processes, our data may provide some insight into potential drivers of CH_4 variability, and we discuss these below.

Some of the spatial variability in CH_4 concentrations and fluxes we observed may be related to localized CH_4 sources from rivers, and production near river mouths. As shown in Figure 2, generally higher CH_4 concentrations were observed at nearshore stations in the south basin, and in proximity to river and marsh inflow. By comparison, CH_4 concentrations in the north basin tended to be higher along the eastern shore, and at isolated hotspots in central portions of the basin. In 2018, in particular, we observed exceptionally high CH_4 concentrations near the mouths of the Red River and Winnipeg River. Stations 1 and 2 are also downstream of an expansive coastal wetland at the southern edge of the lake. Freshwater marshes are among the largest natural CH_4 sources (Kirschke et al., 2013), and thus may supply CH_4 to the adjacent waters in Lake Winnipeg. Several rivers also drain into the eastern half of the north basin, namely the Poplar, Pigeon, Berens, and Bloodvein Rivers in the north basin, and the Winnipeg and Manigotogan River in the south basin (see Fig.1). These river inputs may explain the higher CH_4 concentrations observed along the eastern half of the lake. Boreal lake systems have shown concentrations of CH_4 to range from 13 – 2,015 nmol L^{-1} (Table 3; Bastviken et al., 2004), and these are similar to aquatic systems which lie to the east of Lake Winnipeg, suggesting rivers draining these watersheds are likely sources of dissolved CH_4 to the lake. We also observed high CH_4 concentrations during summer and fall 2019 near the Saskatchewan River mouth on the western edge of the north basin (Fig. 1; Fig. 2). The Saskatchewan River's catchment extends to the Rockies and thus runs east across the northern great plains underlain by dolomite and limestone, draining largely agricultural lands before entering the lake (Levesque and Page, 2011). The distribution of CH_4 in the lake suggests that rivers import CH_4

rich waters and are important in determining local variation of CH_4 in the lake. These rivers also supply DOC to the lake (McCullough, 2001), which has been shown to have a positive relationship with CH_4 concentrations (Bastviken et al., 2004). Additional work is required to better understand the role of these rivers in moderating the lake's CH_4 dynamics.

A unique feature of Lake Winnipeg relative to other great lakes is that it is a cold polymictic lake, largely due to its shallowness and large fetch that aligns with prevailing winds. The strong mixing and associated ventilation likely contribute to the relatively low CH_4 concentrations observed in our study by 1) providing a constant dissolved O_2 supply to deep waters, which reduces CH_4 production and promotes CH_4 oxidation, and 2) promoting rapid CH_4 outgassing, which reduces CH_4 accumulation in the water column. Water column anoxia has never been observed, but dissolved oxygen concentrations are variable throughout the lake (McCullough, 2020; Wassenaar, 2012). Waters were well-oxygenated during our surveys (O_2 > ~7.0 mg L^{-1}), which may explain the lack of consistent negative correlation between CH_4 and O_2 (Fig. 4). This also suggests that sedimentary methanogenesis is the likely source of autochonous CH_4 production in the lake. Water temperature did not seem to be related to CH_4 concentration outside of the south basin in the summers, where water temperatures were generally higher and where we did see greater CH_4 concentrations on average (Table 1, 2B).

Although mixing and ventilation may reduce CH_4 production and storage, wave-induced sediment disturbance may enhance CH_4 flux from sediments to the water column (Bastviken et al., 2008, 2004; Borrel et al., 2011). Previous studies of Lake Winnipeg have shown that sediment resuspension occurs frequently (Matisoff et al., 2017) and may thus be an important mechanism for CH_4 release in shallow near-shore environments where wave action can disturb sediments. Indeed, we observed an inverse relationship between dissolved CH_4 and water column depth in the south basin during 2018 (R^2 = 0.60), indicating a potential tendency for CH_4 concentration to increase with decreasing water column depth during the open water seasons. Conversely there was no obvious relationship between station depth and CH_4 concentration for the north basin, although the several deep (W1 and W7), and shallow stations (12NS) are conspicuous by their high CH_4 concentration relative to the average of the lake (> 41.6 nmol L^{-1}).

Another factor influencing CH_4 variability may be differences in substrate quantity and quality for CH_4 production. The lake experiences frequent and expansive algal blooms during the open water season, particularly in the north basin (Binding, 2020; Page, 2020; 2011). Algal biomass deposition specifically promotes higher rates of CH_4 production compared to terrestrial plants (West et al., 2012), and limited research on Lake Erie has shown a strong relationship between and CH_4 production and algal blooms (Fernandez et al., 2020). This has important implications for Lake Winnipeg, as a hypereutrophic lake that frequently experiences large scale algal blooms (Page, 2011). The frequency and intensity of these blooms are expected to escalate with higher temperatures and increased watershed nutrient runoff (Beaulieu et al., 2019, Fernandez et al., 2020). Recent work has also shown that marsh vegetation (i.e. *Typha latifolia* aka. Broadleaf Cattail) promotes much higher rates of CH_4 production than the degradation of coniferous needles and deciduous tree leaves (Emilson et al. 2019), which may explain the high CH_4 concentrations near the Netley-Libau Marsh in the southern end of the south basin (Fig. 2).

Given the prolonged ice cover during winter and corresponding suppression of CH_4 evasion, we were surprised to find no evidence of CH_4 accumulation in the water column during the winter sampling. However, our finding is consistent with observations of low CH_4 concentrations under partial ice cover in Lake Erie (Fernandez et al., 2020). The average winter (2019) CH_4 concentration (27.4 ± 20.8 nmol L^{-1}; Table 2) is not significantly different (Wilcoxon Rank-Sum, $\alpha = 0.05$), than observed for the fall of 2018 and spring and summer in 2019 (23.87 ± 14.5, 31.7 ± 41.8 , 32.7 ± 28.4 nmol L^{-1}, respectively; Fig. 3), and the spatial pattern of CH_4 during the winter (Fig. 2) shows distinctive features that have also been identified in the warm season data sets, namely high CH_4 concentration in the southern end of the lake and around W7 in the north basin. These two observations are surprising, given that the complete ice cover has been shown to promote the build-up of CH_4 by preventing direct lake-air gas exchange (Greene et al., 2014; Michmerhuizen et al., 1996). The results suggest that 1) methane is actively oxidized over winter and production rates are reduced, which largely prevents large scale methane evasion during ice breakup, and 2) processes supporting the high CH_4 at the stations identified above are possibly suppressed in the winter season.

Conclusions

Our study revealed that Lake Winnipeg was consistently oversaturated with CH_4 relative to the atmosphere, and a source of CH_4 throughout the open water season. Despite its eutrophic state and frequent algal blooms, Lake Winnipeg does not emit or produce a large amount of dissolved CH_4 per unit area compared to other lakes in the boreal regions of Canada and northern Europe (Bastviken et al., 2011; Casper et al, 2000; Demarty et al., 2010; Huutunen et al, 2003; Kankaala et al., 2013; Table 3). We attribute this to the lake's large fetch and shallow depth, which combine to ventilate the lake, adding O_2 and limiting CH_4 accumulation in the water column. The low emission intensity and large drainage basin area suggest that CH_4 emissions from Lake Winnipeg only modestly offset the terrestrial greenhouse gas sink. However, recent work suggests lake methane emissions could increase in response to eutrophication (Beaulieu et al., 2019), underscoring the importance of continued CH_4 monitoring in Lake Winnipeg.

We identified several sites with persistently high dissolved CH_4 concentrations near the mouths of the Red and Winnipeg Rivers. We found no consistent relationship between dissolved O_2 and CH_4 concentration or flux, and no evidence for significant CH_4 accumulation under ice during winter. Further research should be done on CH_4 in Lake Winnipeg to identify causes of the patterns and hotspots, whether they are related to seeps, riverine input, or produced through in-situ process of methanogenesis, quantify water column oxidation rates, as well as to measure possible ebullitive and plant-mediated fluxes.

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Tables

Table 1: Wind speed (m s^{-1}) and water temperature (°C) in Lake Winnipeg across five surveys, excluding winter 2019. Data is split into north basin (NB) and south basin (SB).

			Wind Speed (m s^{-1})			Water Temp (°C)		
Season	Basin	Year	Mean	Median	Standard Deviation	Mean	Median	Standard Deviation
Spring	NB	2019	3.4	2.7	2.1	8.3	7.4	2.7
	SB	2019	3.4	2.7	2.3	13.4	12.8	2.3
Summer	NB	2018	4.9	4.6	3	19.5	19.6	1.5
		2019	2.6	2.5	1.3	20	19.9	1.6
	SB	2018	4.9	5.1	2.9	20.2	20.2	0.3
		2019	3.4	3.9	1.9	22	22.1	0.7
Fall	NB	2018	5.4	5.7	1.6	8.3	7.9	1.6
		2019	6	5.4	4.1	15.4	15.6	0.3
	SB	2018	6.8	6.2	3.6	7.7	7.6	4.5
		2019	5.1	4.6	2.4	12.6	11	3.3

Table 2: Average, median, standard deviation, maximum, minimum, and sample size (N), for (A) diffusive CH_4 flux (µmol CH_4 m^{-2} h^{-1}) and (B) concentration (nmol L^{-1}) respectively. NB and SB denote north and south basins of Lake Winnipeg. No flux estimate was calculated for winter 2019 as ice cover was present. Data from both surface and bottom samples (when available) were used.

			CH_4 concentration (nmol L^{-1})						CH_4 flux (µmol m^{-2} h^{-1})					
Cruise	Year	Basin	Mean	Median	Standard Deviation	Maximum	Minimum	N	Average	Median	Standard Deviation	Maximum	Minimum	N
Spring	2019	NB	24.0	22.3	7.5	48.7	9.5	33	0.6	0.3	0.7	2.7	0.0	24
	2019	SB	48.6	19.3	73.6	282.5	15.4	15	2.4	0.4	4.4	13.4	0.0	10
	2019	Lake	31.7	22.0	42.2	282.5	9.5	48	1.1	0.3	2.5	3.2	0.0	34
Summer	2018	NB	43.2	32.1	43.4	217.2	8.4	28	1.9	1.3	3.1	13.4	0.0	19
	2018	SB	122.4	32.7	230.3	733.8	8.1	12	11.6	1.4	26.2	70.8	0.4	7
	2018	Lake	67.0	32.1	132.7	733.8	8.1	40	4.5	1.3	13.8	70.8	0.0	26
	2019	NB	29.8	24.2	23.6	126.7	5.0	30	0.4	0.4	0.4	1.2	0.0	22
	2019	SB	39.8	21.1	39.1	142.8	14.7	12	2.5	0.7	4.3	12.2	0.0	11
	2019	Lake	32.7	21.9	28.7	142.8	5.0	42	1.1	0.4	2.6	12.2	0.0	33
Fall	2018	NB	26.2	19.7	15.9	73.7	10.2	26	1.1	1.0	0.8	3.2	0.1	18
	2018	SB	16.7	13.8	7.5	30.7	10.3	9	1.4	1.4	1.0	2.5	0.2	4
	2018	Lake	23.8	18.5	14.7	73.7	10.2	35	1.2	1.1	0.9	3.2	0.1	22
	2019	NB	52.5	43.2	54.1	285.3	12.1	31	7.4	2.3	16.3	78.4	0.0	23
	2019	SB	60.8	41.1	64.3	203.5	11.5	13	2.9	2.4	1.9	6.4	0.8	7
	2019	Lake	54.9	42.1	56.7	285.3	11.5	44	6.4	2.4	14.3	78.4	0.0	30
Winter	2019	NB	28.4	23.0	15.5	50.4	10.2	6						
	2019	SB	25.4	6.5	33.4	63.9	5.7	3						
	2019	Lake	27.4	21.3	20.8	63.9	5.7	9						

Table 3: Comparison of dissolved CH_4 concentration and diffusive flux with other lakes. Concentrations are shown as ranges, fluxes are shown as mean ± 1 standard deviation where data are available. Annual fluxes were derived using mean values, while annual flux for Lake Winnipeg derived using median values is shown in parentheses. Compiled using data from [1]Bastviken et al, (2004); [2]Bastviken et al, (2009); [3]Fernandez et al, (2019); [4]Juong et al, (2019); and [5]Rasilo et al., (2015).

Lake	Surface Area (km^2)	CH_4 ($nmol\ L^{-1}$)	Diffusive CH_4 flux ($\mu mol\ CH_4\ m^{-2}\ h^{-1}$)	($Tg\ CH_4\ y^{-1}$)
Winnipeg[+]	23,750	5.0 - 733.8	2.8 ± 9.2	0.0039 (0.0014)
Erie[3]	25,657	24.3 - 107.1	2.3	0.008
Superior + Michigan[4]	140,103	3.5 - 60.0		
Small Boreal Lakes[1]		13 - 2,015	0.8 - 417.8	
Lakes in Quebec[5++]	35.2 (0.01-5,030)		40.3 ± 91.7	
Lakes between 54-66 deg N[2]	1,533,084		5.1	1.1
Lakes between 25-54 deg N[2]	1,330,264		25.7	4.8

[+] The annual diffusive CH_4 flux calculated using median values for Lake Winnipeg is shown in parentheses

[++] Average surface area of lakes, along with the range in lake area (parentheses) is provided.

Figure Captions

Figure 1: Lake Winnipeg. The location of Lake Winnipeg within Canada is shown on the inset, with major rivers, cities and communities marked on the main map.

Figure 2: Surface concentrations of Lake Winnipeg dissolved CH_4, in nmol L^{-1}. Scale is logarithmic, base 3, to account for the wide range of concentrations. 2019 had a greater sampling density than 2018. The Lake Winnipeg station map located as an inset. Not all stations were sampled each cruise. "NS" denotes "nearshore." All stations sampled were at minimum at the "surface" depth, 1 m below the lake surface, and "W" stations were sampled at both surface depth and "bottom," or 1 m above the lake bottom. Map generated using QGIS (2020).

Figure 3: Lake CH_4 concentration in nmol L-1 (a), and CH_4 flux in µmol m^{-2} h^{-1} (b). Extreme outliers (>400 nmol L^{-1} for concentration and >17 µmol m^{-2} h^{-1} for flux) have been removed to allow better display variability in the data set. No flux estimates were made for winter 2019, as ice cover was present. Data is split into the north basin, and south basin for each cruise sampled.

Figure 4: Summer 2018 (4A) and fall 2018 (4B) dissolved oxygen concentrations in mg L^{-1} and CH_4 in nmol L^{-1}. R^2 values from 2nd order polynomial trendlines for south and north basin respectively in summer 2018 are 0.93 and 0.22, and in fall 2018, 0.16 and 0.12 for north and south basins respectively. More variability is seen for both CH_4 and O_2 in the summer season than fall. Log base 10 is used on the y-axis.

Figure1

Click here to access/download;Figure;JGLR-Figure 1.pdf

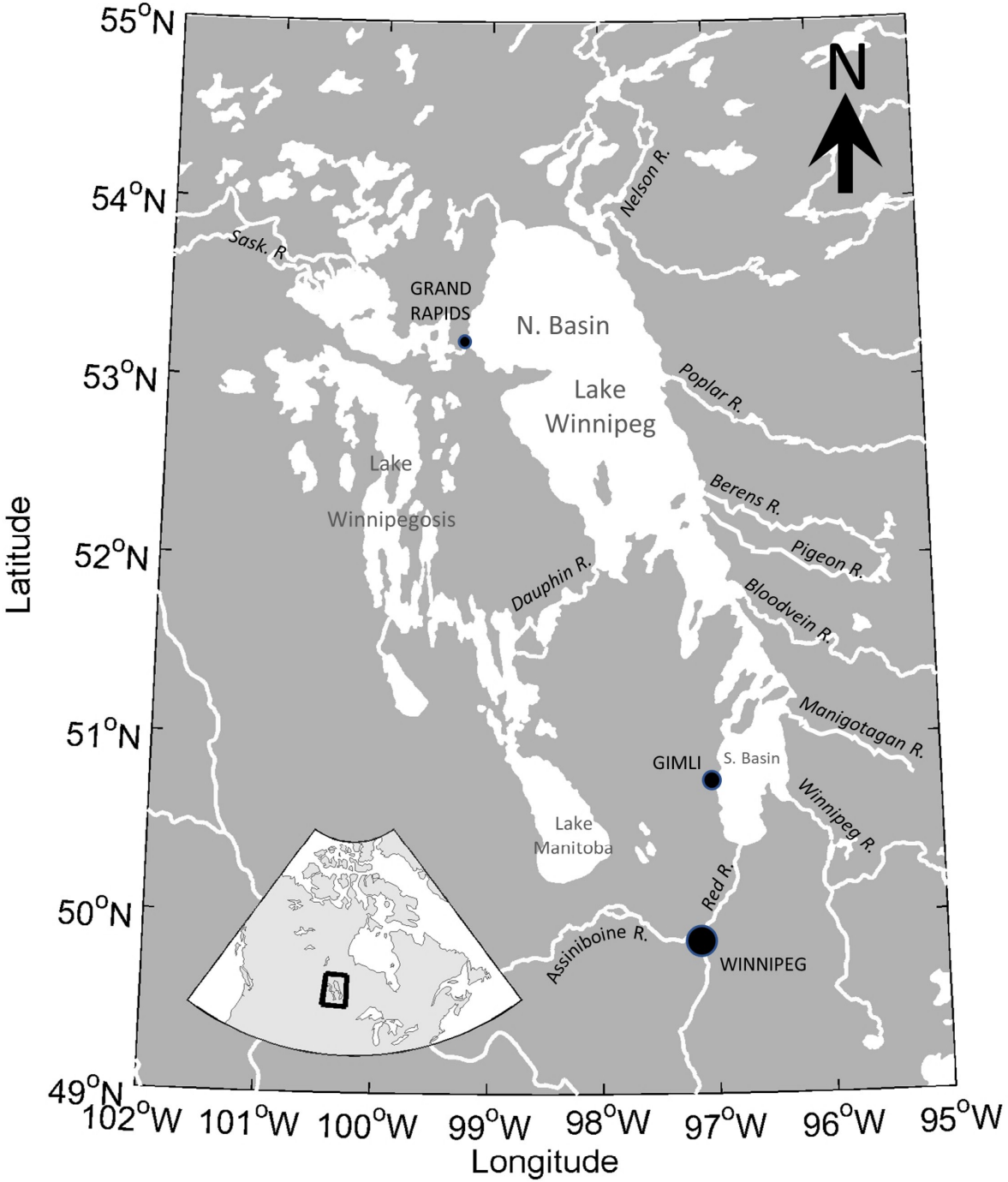


Figure2

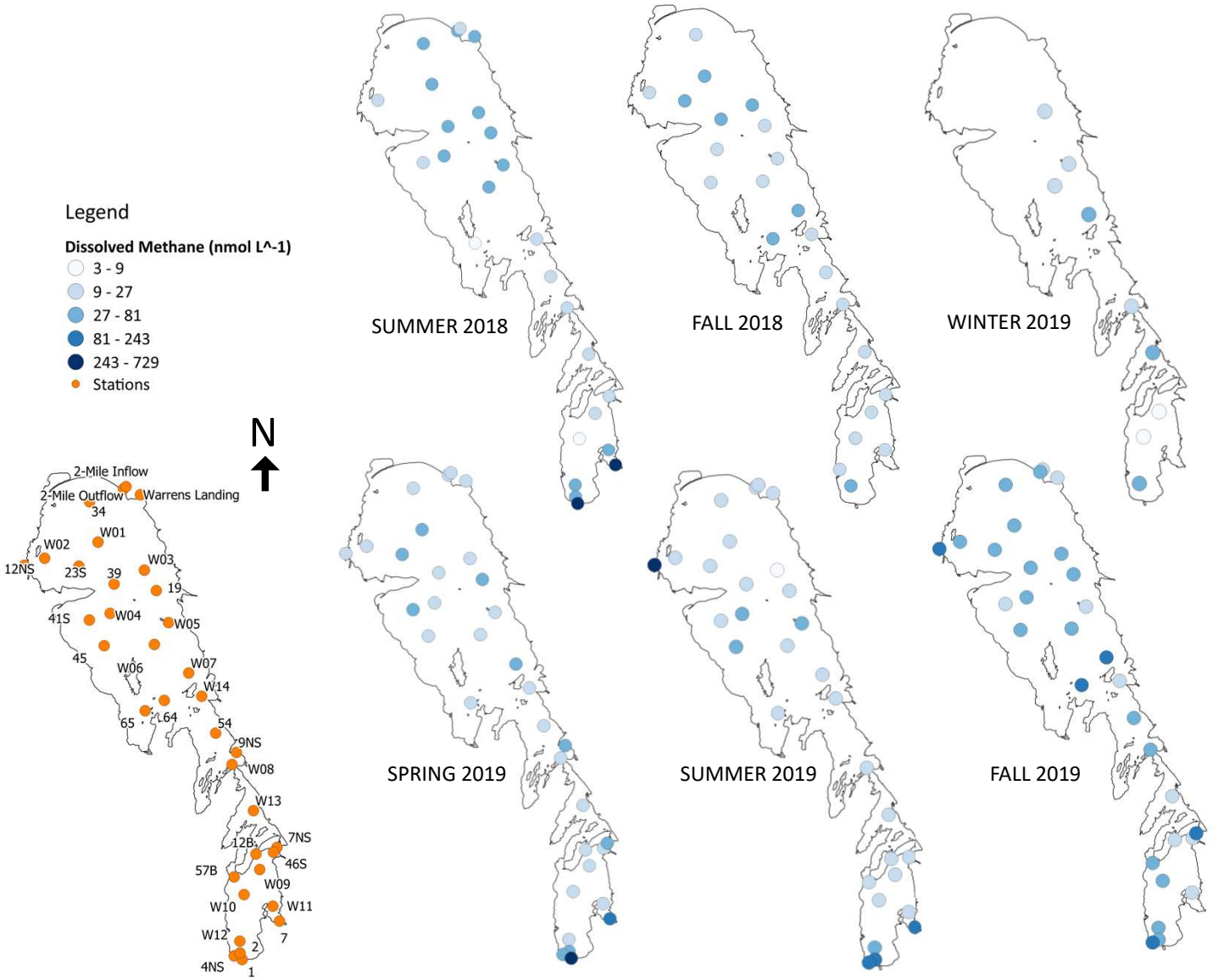


Figure3

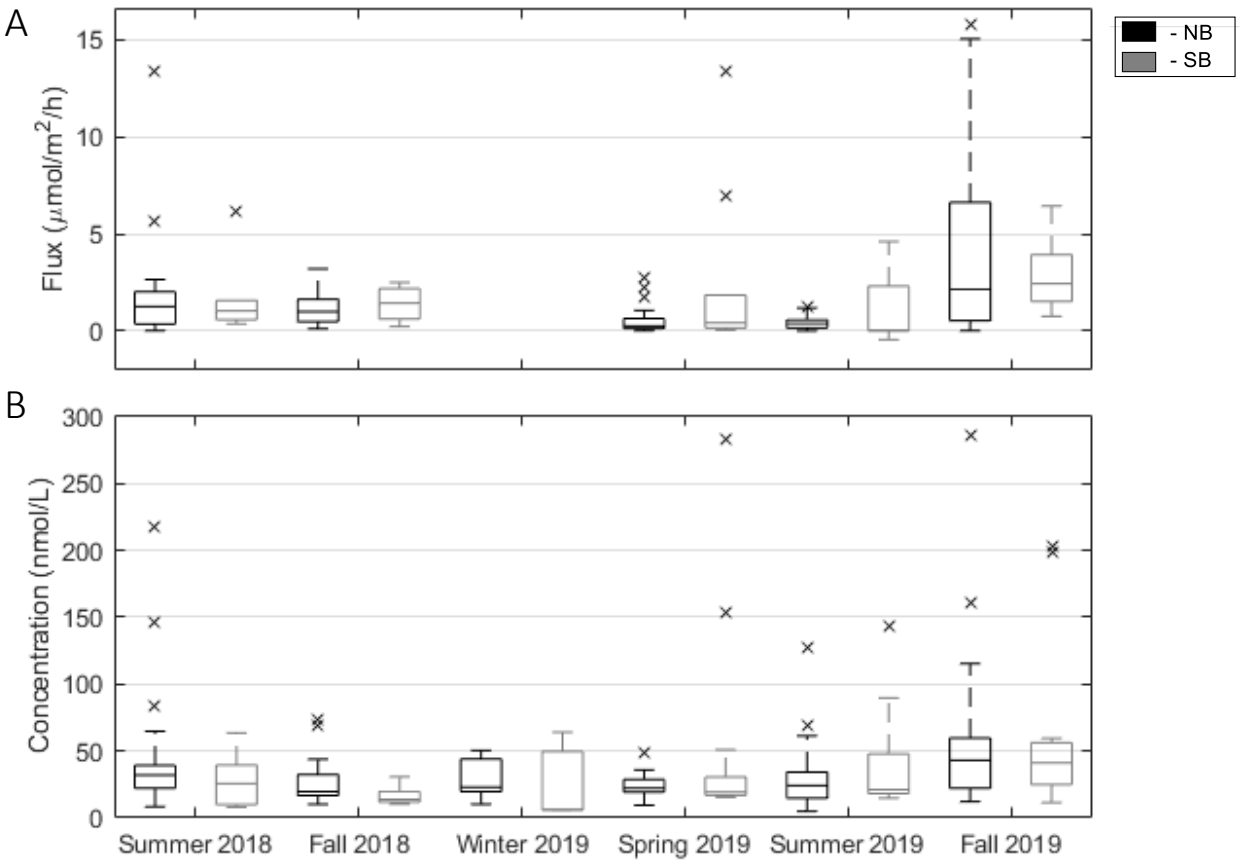
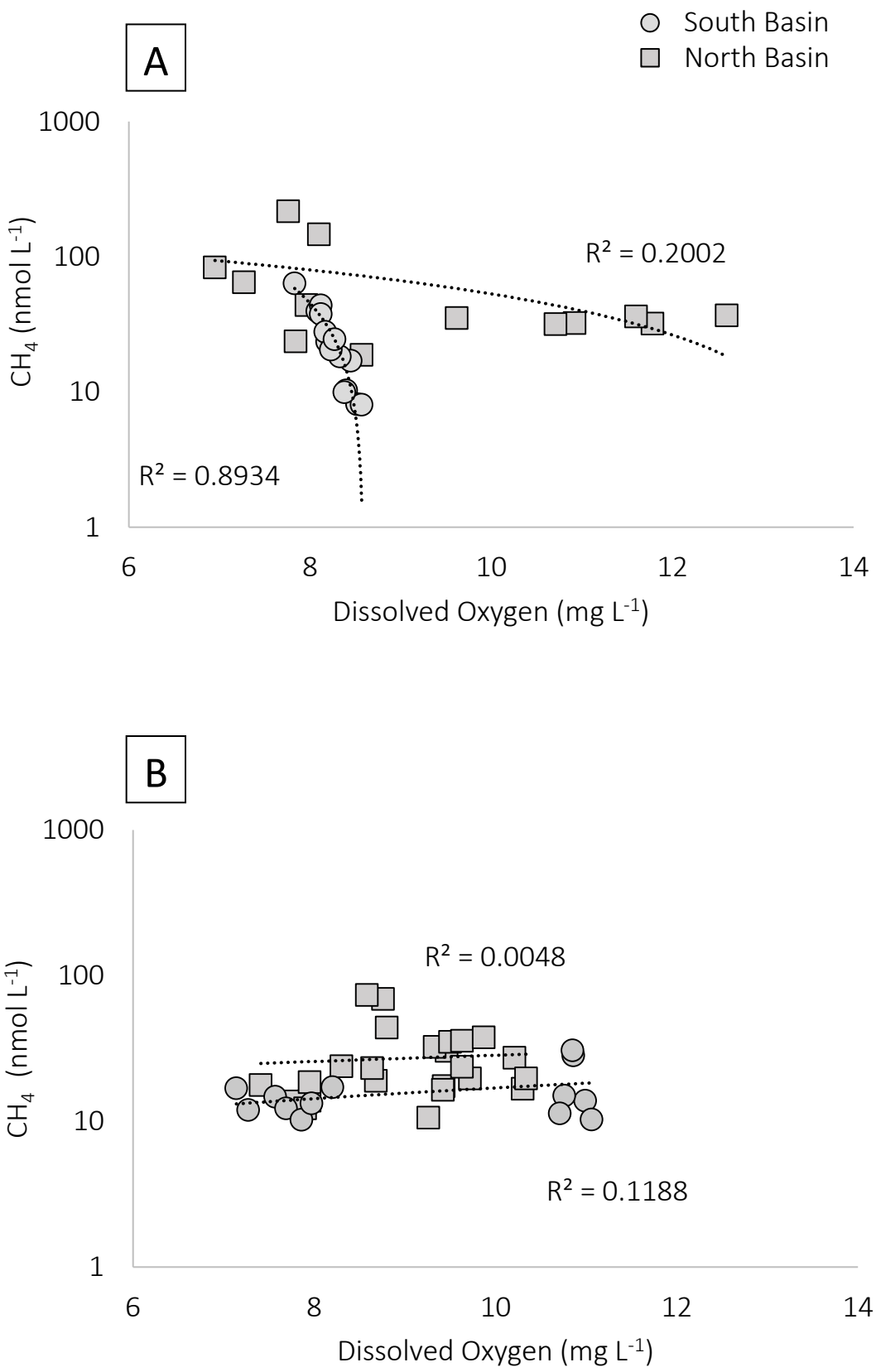


Figure4

Click here to access/download;Figure;JGLR-Figure4.pdf



Electronic Supplementary Material

First estimation of the diffusive methane flux and concentrations from Lake Winnipeg, a large, shallow and eutrophic lake

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Table S1: Summary statistics for Lake Winnipeg CH_4 concentrations in nmol L^{-1}. Samples taken at one meter below surface depth are indicated as "surface" and samples taken from one meter above the lake bottom are marked as "bottom." All stations W01 – 65 are considered "north basin" and all stations W09 – 57B are "south basin" stations.

Station	Summer 2018	Fall 2018	Winter 2019	Spring 2019	Summer 2019	Fall 2019
W01 - Surface	31.7	35.1		27.2	13.2	59.3
W02 -Surface	24.4	16.7		9.5	25.7	27.1
W03 - Surface	32.2	30.4	21.3	26.2	5.0	38.0
W04 - Surface	33.1	23.7		26.4	27.3	43.8
W05 - Surface	36.9	23.8	24.7	21.9	69.2	15.1
W06 - Surface	32.0	18.9	19.7	21.2	25.6	52.0
W07 - Surface		13.8	44.0	30.4	22.4	87.3
W08 -Surface	20.6	17.1	10.2	19.2	26.0	43.5
W13 -Surface	17.0	10.2	50.4	19.7	13.6	14.3
W14 - Surface	18.8	69.1		16.5	26.1	21.6
W01 - Bottom	146.3	35.6		31.9	41.4	70.3
W02 - Bottom	36.2	16.5		28.6	34.3	13.3
W03 - Bottom	83.2	32.4		25.4	61.3	54.4
W04 - Bottom	33.7	23.2		23.8	54.7	70.2
W05 - Bottom	44.0	12.3		35.8	12.2	13.2
W06 - Bottom	64.7	18.5		31.7	19.5	59.9
W07 - Bottom	217.2	13.7		21.6	126.7	285.3
W08 - Bottom	24.6	17.8		20.5	34.8	24.1
W13 - Bottom	18.3	13.3		19.4	22.8	15.3
W14 - Bottom	23.7	73.7		14.2	28.6	24.3
2-M Inflow	37.0			17.4	21.1	28.4
2-M Outflow	24.7			20.0	19.8	24.5
Warrens L.	41.5			17.4	14.7	12.1
9NS				48.8		
12NS				26.1		115.4
19	35.1	19.7		29.1	12.6	70.3
23S		27.3		29.5	26.0	35.2
34	28.3	17.5		25.3	11.3	43.2
39	63.2	43.9		22.3	20.8	44.9
41S	14.8			32.6	20.2	16.5
45		19.6		17.9	43.2	
54	18.7			13.7		43.4
64		37.7				160.9
65	8.4			20.7	13.9	
W09 - Surface	10.3	12.3	5.7	19.3	14.7	
W09 - Bottom	9.9	11.9		15.4	16.6	11.5
W10 - Surface	8.1	10.3	6.5	17.0	21.0	55.1
W10 - Bottom	8.1	11.3		16.9	21.3	13.0

W11 - Surface	43.8	13.8		18.4	18.6	26.6
W11 - Bottom	27.8	15.0		15.9		26.4
W12 - Surface	39.5	28.3	64.0	22.0	48.7	41.1
W12 - Bottom	37.6	30.7		22.4	46.9	50.3
1	462.6			282.5		
2	63.5			50.9	89.7	59.6
4NS				29.0		203.5
7	733.8			153.5	142.8	
7NS				31.0		198.8
12B				18.3	18.0	24.4
46S	23.6	16.9		15.9	21.4	25.1
57B					18.3	54.4
Average	67.0	23.8	27.4	31.7	32.7	54.9
Median	32.1	18.5	21.3	22.0	21.9	42.1
Standard Deviation	131.0	14.5	19.6	41.8	28.4	56.0
Minimum	8.1	10.3	5.7	9.5	5.0	11.5
Maximum	733.8	73.7	64.0	282.5	142.8	285.3
N	40	35	9	48	42	44

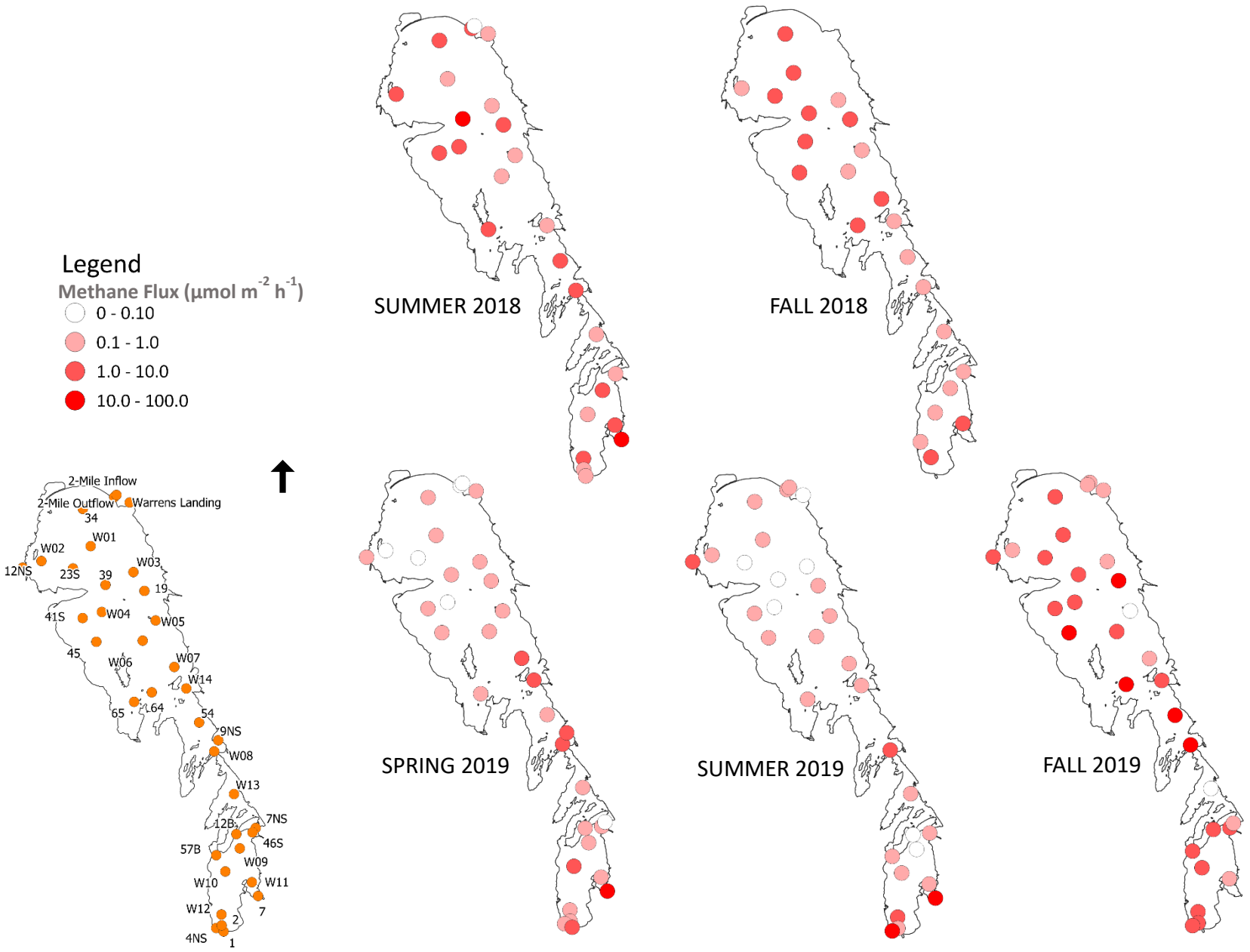

Figure S1: The distribution of the surface flux of CH_4 from Lake Winnipeg (in µmol m^{-2} h^{-1}) for each station sampled during each of the five cruises. The sampling stations for Lake Winnipeg are provided on a separate map in lower left of the figure. The maps were generated using QGIS (2020).