

METHANE AND CARBON DIOXIDE DYNAMICS IN ARCTIC LAKE SEDIMENTS

By

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A handwritten signature in black ink, appearing to read "Joan Curry", is written over a horizontal line.

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## **Abstract**

Rising global temperatures are expected to increase concentrations of greenhouse gases emitted by northern latitudes within the current century. The impact of global warming on Arctic lacustrine systems is generally unknown, although recent studies have examined fluxes of carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>) produced in ebullition events. Few studies have investigated the added impact of atmospheric warming on lake sediments, which produce CO<sub>2</sub> and CH<sub>4</sub> through microbial decomposition and diffusive loss in the water column. To better understand carbon emission scenarios at elevated temperatures, sediment samples from Abisko, Sweden were analyzed for CO<sub>2</sub> and CH<sub>4</sub> production rates through incubation studies, and for concentrations of dissolved inorganic carbon (DIC) and dissolved CH<sub>4</sub> in sediment and porewater. Results showed that room temperature incubations emitted concentrations of CO<sub>2</sub> and CH<sub>4</sub> up to five times greater than those emitted by +5°C incubations. Furthermore, documented peat emissions were one to two orders of magnitude lower than the lake sediment incubation emissions reported in this paper. This study provides some of the first point source microbial emissions by lake sediment depth, and highlights that northern latitude sediments could have unprecedented effects on current spatial and temporal projections of Arctic warming.

## **Introduction**

### *Background*

Since the beginning of the industrial revolution, humans have become a major influence on the global carbon cycle. Anthropogenic activities, such as burning biomass and fossil fuels, rice farming, and raising livestock, contribute billions of tons of carbon to the atmosphere on a yearly basis in the form of greenhouse gases (Walter, 2007). The current concentrations of atmospheric greenhouse gases – specifically carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), and nitrous oxide (N<sub>2</sub>O) – are now higher than they have been in the past 800,000 years, and are causing global warming trends that have been linked with near certainty to anthropogenic activities (IPCC, 2014). The impending risks of global warming are severe and largely irreversible and are expected to adversely impact a number of climate processes, including climate variability, the frequency and intensity of extreme weather events, sea level rise, ocean acidification, and even the socioeconomic situations of low-lying countries, in addition to having a myriad of unknown effects.

As a result of the anticipated rise in global temperatures, the Intergovernmental Panel on Climate Change (the IPCC) estimates that it is very likely – or projected with a 90 to 100 percent probability (Mastrandrea, 2010) – that northern latitudes will undergo a warming effect known as ‘polar amplification’ within the current century (Hezel, 2013). The poles will experience a greater net temperature increase than lower-latitude areas (approximately 5°C to 11+°C as opposed to a global average increase of 2°C to 5°C), and are expected to respond to climatic warming much more rapidly than temperate and equatorial regions (IPCC, 2014). A multitude of research has shown that rising global temperatures that are associated with increases in

atmospheric CO<sub>2</sub> concentrations are also linked to distinct changes in the terrestrial biogeochemistry of the poles. The northern latitudes in particular have natural sinks and sources of greenhouse gases to the atmosphere that will be affected by these changes; sinks are reservoirs that remove greenhouse gases from the atmosphere and sources are reservoirs that contribute them to the atmosphere. Although these systems have been roughly in equilibrium for thousands of years, widespread and documented permafrost thaw has caused previously frozen reservoirs of organic carbon to become available for microbial decomposition. These processes may cause the Arctic to become a net source of carbon to the atmosphere, and yet another positive feedback to the global warming cycle (Algesten, 2003; McGuire, 2009; Walter, 2006).

Some of the largest known pools of organic carbon that will be affected by increasing temperatures reside in the permafrost ecosystems within the Arctic and boreal regions of the Northern Hemisphere (Zhang, 1999). Permafrost, or perennially frozen ground, ranges in thickness from one meter to several hundred meters and is described as being either continuously or discontinuously frozen year around. Permafrost layers are composed of a superficial active layer, an intermediate transition zone, and a deep and fully frozen permafrost zone. The active layer seasonally thaws and refreezes and has a variable thickness that depends on a combination of atmospheric temperatures and local influences (Schuur, 2008). Peatlands, the terrestrial layers that are encompassed by the permafrost, are distinguished from other ecosystems in that they accumulate more organic matter than is decomposed by microbes, and fall somewhere between the classification of mineral soils and waterlogged soils in their degree of saturation (Gilbert, 2006). Due to the fact that permafrost thaw exposes underlying peat to increasing atmospheric temperatures, and because the active layers that are perennially frozen are expected to thaw by

some 37% to 81% during the current century (IPCC, 2014), these finely balanced ecosystems are expected to be early and abrupt indicators of changes in the global carbon cycling process.

### *The significance of lacustrine systems in global warming*

The primary focus of Arctic research is on how terrestrial peatlands will respond to warming temperatures. Although thawing permafrost will be of crucial concern in regard to carbon release and climatic forcing agents, nearly fifty percent of the land area that makes up the Arctic has been disregarded in large-scale climate projections. Few studies have been done to quantify the contribution of greenhouse gases from northern latitude pools, ponds, and lakes to the atmosphere, and yet these bodies of water compose up to 48% of the surface area of the Arctic landscape (Riordan, 2006). Northern peatland lakes are now known to emit globally significant concentrations of CO<sub>2</sub> and CH<sub>4</sub> through the processes of microbial decomposition and ebullition (e.g. methane bubbling) (Cole 1994; Walter, 2007; Wik, 2011). Preliminary studies suggest that CO<sub>2</sub> emissions from inland lakes are approximately 25-54 Tg C/yr (McGuire, 2009), and that CH<sub>4</sub> emissions are in the range of 15-50 Tg CH<sub>4</sub>/yr (McGuire, 2009; Walter, 2007). Exact emission estimates are unknown for both greenhouse gases, and may have uncertainty bounds as low as 32 Tg C/yr and as high as 112 Tg C/yr for CH<sub>4</sub> in particular. Accelerated warming in northern latitudes is projected to increase the efflux of CO<sub>2</sub> and CH<sub>4</sub> to the atmosphere through these diffusive mechanisms and may have unprecedented effects on the spatial and temporal projections of warming in the Arctic. It is therefore imperative that lacustrine systems are factored into global warming models so that conclusions derived about global climate change are representative of real world processes and can provide comprehensive

information about changes in the atmospheric carbon budget (McGuire, 2009; Schuur, 2008; Walter, 2007; Zimov, 2006).

Quantifying emissions from ebullition events has been a priority in the few studies that have examined Arctic lakes. Ebullition sources may account for some 70% of total emissions from these thermokarst systems (i.e. lakes that are created as a direct result of permafrost thaw), whereas microbial decomposition within sediment and the resulting diffusion of carbon through the water column might only account for 5% of emissions (Walter, 2007). It is essential that the system is understood in its entirety though, and to date little to no attention has been given to the microbial sedimentary processes that produce a continual flux of CO<sub>2</sub> and CH<sub>4</sub>.

In order to better understand carbon emissions from northern latitude lakes, and to recognize how flux concentrations may be altered in the presence of rising atmospheric temperatures and thawing permafrost, a baseline study was conducted on two sub-arctic lakes in the discontinuous permafrost zone in Arctic Sweden. The lakes are located in Stordalen Mire, a long-term biogeochemical research site near the town of Abisko. Sediment cores were extracted from each lake that differed in water depth and surface ebullition concentrations, and sediments were analyzed for a variety of characteristics: (1) CO<sub>2</sub> and CH<sub>4</sub> potential production rates through incubation studies; (2) concentrations of dissolved CH<sub>4</sub> in both sediment and sediment porewater; (3) concentrations of dissolved inorganic carbon (DIC) in sediment porewater; (4) microbial community structures by sediment depth; (5) nutrient profiles for total organic carbon (TOC), total organic nitrogen (TON), and calcium carbonate (CaCO<sub>3</sub>); and (6) isotopic signatures by sediment depth ( $\delta^{13}\text{C}$ ) (Freitas, 2012). This research describes how a warming climate will impact the lacustrine systems that cover thawing peatlands, and attempts to quantify concentrations of greenhouse gases that are emitted from sediment under increased temperatures.

## **Site Description**

The study site is located in northern Sweden and is approximately 200 km above the Arctic Circle (About the Research Station, 2012). Lakes Inre Harrsjön and Mellan Harrsjön fall within the Stordalen mire, which is a peatland ecosystem 10 km east of the town of Abisko (68°21'N, 19°02'E) (Wik, 2011). The lakes are connected to one another via an intermediate lake, Yttre Harrsjön, that feeds water into Mellan Harrsjön through a small inlet. Unlike Inre Harrsjön, which only receives runoff from the mire during large precipitation events, Mellan Harrsjön is also supplied with water via a river that runs through the mire (Wik, 2011).

Inre Harrsjön has a larger surface area than Mellan Harrsjön (0.023 km<sup>2</sup> versus 0.011 km<sup>2</sup>) but has a shallower water depth (5 m versus 7 m). These lakes have very small areas and are much shallower than many of the surrounding lacustrine systems, such as the nearby lake Torneträsk that covers approximately 330 km<sup>2</sup> and is considered one of the deepest lakes in Sweden (Coates, 2011).

## **Sampling Protocol**

### *Methodology*

Sediment coring was conducted on two separate days during the summer of 2012. Three cores were taken per depth in each lake, totaling 12 sediment cores for the two lakes. The first set of cores were taken from shallow water (~1 m depth) on July 10, and the second set of cores were taken from deep water (~5.5 m below the surface water in Inre Harrsjön and ~6.5 m below the surface water in Mellan Harrsjön) on July 19. Cores were taken from areas that had produced

more continuous ebullition events over the three-year period, with the hope that incubation studies and microbial activity could eventually be cross-referenced with the ebullition concentrations. Martin Wik provided guidance regarding coring locations and sampling depths. His knowledge and prior research of the two lakes is unparalleled, and was based on seasonally monitored ebullition transects that had been run through each lake over the course of a three year study (Wik, 2011).

The coring device that was used was based on a variation of the Large Bore Sediment Corer (Large Bore Sediment Corer, 2006). The corer was composed of a 1 m long PVC pipe that was approximately 6 cm in diameter. It had 1 cm wide extraction holes that were pre-drilled in 2 cm intervals down one side of the corer – these were then sealed with heavy-duty electrical tape. The ends of the corer were capped with airtight rubber stoppers.

Sediment cores were taken from a rowboat in both Inre Harsjön and Mellan Harsjön; the corer was hand-inserted into the sediment for the shallow depths and was pushed into the sediment with an extension rod for the deep depths. Cores were pulled up using a string attached to the coring device, and were capped from bottom to top so as to prevent sediment loss. Sediments remained sealed in the air-tight corers until all samples could be processed on shore.

Sediment from core 1 was used for CO<sub>2</sub> and CH<sub>4</sub> incubation studies, and was also used to determine the concentration of dissolved CH<sub>4</sub> in sediments and pore water via sodium hydroxide (NaOH) extractions. Core 2 was used to extract dissolved inorganic carbon (DIC) from pore water, and phosphoric acid extractions were used to determine the concentration of CO<sub>2</sub> in the pore water. Core 3 was used to pull samples for DNA and nutrient analysis.

## *Replicate Core 1*

### Sediment/pore water-NaOH additions

Prior to exuding the sediment from each core for the incubation bottles, 2 mL of sediment were extracted every 4 cm down the cores to be analyzed for total CH<sub>4</sub> production by depth. The tips of 5 mL syringes were cut at the 1 cm mark and were used to extract the sediment at each depth. New syringes were used per change in depth in order to minimize cross contamination between samples. Samples were added to 5 mL of 1M NaOH in 120 mL clear glass bottles. The bottles were capped with PTFE/silicone septa (stoppers), clamped with aluminum crimp tops, and were flushed with compressed nitrogen gas (N<sub>2</sub>) for one minute. Each bottle was shaken in the field for two minutes. All bottles were stored on ice in a cooler until they could be transported back to the lab.

Once back in the lab, all NaOH-sediment additions were left at room temperature to degas for 48 hours. The 48-hour concentration of degassed CH<sub>4</sub> was analyzed using the Flame Ionization Gas Chromatograph. Prior to pulling the samples, the bottles were incubated at 60°C for 30 minutes and were shaken for two minutes. A syringe was used to pull 5 mL of headspace from each bottle, and this gas was directly inserted into the Gas Chromatograph.

### Incubations

Four replicate sediment samples (A, B, C, D) were taken at three depths (0-5 cm, 10 cm, 20 cm) down each of the 12 cores. 4 mL of sediment were exuded per depth and inserted into 120 mL clear glass bottles. The bottles were capped with PTFE/silicone septa (stoppers), clamped with aluminum crimp tops, and flushed with compressed nitrogen gas (N<sub>2</sub>) for one minute. All bottles were stored on ice in a cooler until they could be transported back to the lab.

In the lab, A/B replicates from each lake (IH-0-5A, MH-0-5B, IH-10A, etc.) were stored in a +5°C freezer, and C/D replicates were stored at room temperature (~22°C). Incubations were run for five consecutive days, and then headspace gas was re-sampled at days 9 and 13. Every day, 10 mL of N<sub>2</sub> gas was flushed three times into the bottles and then 10 mL of headspace was pulled with a syringe. 5 mL of this diluted headspace were run for CO<sub>2</sub> concentrations on a LI-COR, and the remaining 5 mL were run for CH<sub>4</sub> concentrations on a Flame Ionization Gas Chromatograph.

#### *Replicate Core 2: dissolved inorganic carbon (DIC)*

##### Pore water-H<sub>3</sub>PO<sub>4</sub> additions

Standard Rhizons were used to sample pore water from the sediment cores (Barbier, J., 2015). Rhizons were 2.5 mm in diameter, had a 10 cm porous part, and could withdraw pore water from a mean sediment pore size of 0.15 µm. Rhizons were inserted at 2 cm intervals down each core and extracted water from the sediment into attached syringes. They were filled to 0.5 mL with pore water, purged (in order to rid the samples of the diluted, deionized water within each Rhizon), and were filled to 10 mL. Pore water was added to 0.2 mL 30% phosphoric acid (H<sub>3</sub>PO<sub>4</sub>) in 120 mL clear glass bottles. The bottles were capped with PTFE/silicone septa (stoppers), clamped with aluminum crimp tops, and were flushed with compressed nitrogen gas (N<sub>2</sub>) for one minute. All bottles were stored on ice in a cooler until they could be transported back to the lab.

Samples degassed DIC in the form of CO<sub>2</sub> at room temperature for 24 hours. A syringe was used to pull 0.5 mL of headspace from each bottle, and the gas was directly inserted into the LI-COR for analysis.

### *Replicate Core 3: DNA and nutrient samples*

#### DNA samples

The tips of 5 mL syringes were cut at the 1 cm mark and were used to extract sediment at 6 cm intervals down each core. New syringes were used per change in depth in order to minimize cross contamination between samples. Approximately 1.5 mL of sediment was extracted per sample and was inserted into a 2 mL polypropylene Eppendorf tube. Replicates were taken at every depth. Samples were stored on ice in a cooler until they could be transported back to the lab.

In the lab, all of the DNA plugs were stored in a -80°C freezer in order to prevent microbial transformations until they could be processed.

#### Nutrient samples

The tips of 5 mL syringes were cut at the 1 cm mark and were used to extract sediment at 6 cm intervals down each core. These intervals were offset from the DNA extraction holes by 2 cm to prevent movement of the sediment within the core, and new syringes were used per change in depth in order to minimize cross contamination between samples. Approximately 1.5 mL of sediment was extracted per sample and was inserted into a 2 mL polypropylene Eppendorf tube. Replicates were taken at every depth. Samples were stored on ice in a cooler until they could be transported back to the lab.

In the lab, all of the nutrient plugs were stored in a 2°C freezer in order to prevent nutrient transformations until they could be processed.

## Instrumentation

All CO<sub>2</sub> headspace analysis was run on an LI-6252 CO<sub>2</sub> Analyzer, consistent with LI-COR protocol (Treat, 2012). Twelve CO<sub>2</sub> standards were run prior to the CO<sub>2</sub> headspace samples (three replicates of 1.0% CO<sub>2</sub> standards were tested at the following volumes: 0.5 mL, 1.0 mL, 3.0 mL, 5.0 mL), and four standards were run after the headspace samples (single injections of 1.0% CO<sub>2</sub> standards at the following volumes: 0.5 mL, 1.0 mL, 3.0 mL, 5.0 mL).

Similarly, all CH<sub>4</sub> headspace analysis was run on a Flame Ionization Gas Chromatograph, GC-2014 Shimadzu model (Capillary and Packed Gas Chromatograph GC-2014, 2013). Twelve 200 ppm CH<sub>4</sub> standards were run prior to and after the CH<sub>4</sub> headspace samples were run. The top and bottom values of each set of standards were dropped and the other twenty were kept for comparison with other daily standard runs.

## Calculations

*The following calculations are drawn from a manual constructed by Claire Treat, a researcher at the Abisko Scientific Research Station (Treat, 2012).*

### *CO<sub>2</sub> Standards (for DIC and Incubations)*

A standard curve was constructed for each set of daily CO<sub>2</sub> measurements. The molar concentrations of the standards (umol/mL) were determined by using equation 1: CO<sub>2</sub> concentrations (ppm) were multiplied room pressure (atm); this value was then divided by the temperature of the room (K) and normalized using constants for the LI-COR.

$$1. \quad C_{\text{umol/mL}} = \frac{C_{\text{ppm}} \times (P \times 100 \times 0.98692327)}{(8.2054e^{-5})(T + 273.15)} \times 1e^{-6}$$

A regression curve was then created using equation 2: the volume of injected headspace was multiplied by the molar concentration; the responses were plotted versus concentrations, and slopes and  $R^2$  values were calculated for each curve.

$$2. V \text{ (mL)} * C \text{ (umol/mL)} = N \text{ (umol)}$$

### *CO<sub>2</sub> Samples (for DIC and Incubations)*

Initial flux concentrations were found by dividing the individual responses for the sample injections by the slopes of their associated standard regression curves. These values were then divided by the volume of headspace that was injected into the LI-COR for analysis (equation 3).

$$3. C \text{ (umol/mL)} = \text{response} / \text{slope of calibration curve} / \text{volume injected}$$

Concentrations from equation 3 were corrected ( $C_{\text{corr}}$ ) for daily dilutions of headspace in each incubation vial (equation 4). 10 mL of N<sub>2</sub> gas were injected into the incubation vials every day prior to pulling 10 mL of headspace out of the vials to be run for analysis – 5 mL for CO<sub>2</sub> and 5 mL for CH<sub>4</sub>.

$$4. C_{\text{corr}} = C * (V_{\text{headspace}} + V_{\text{dil}}) / V_{\text{headspace}}$$

The concentration of CO<sub>2</sub> in the headspace was calculated by multiplying the value from equation 4 by the total volume of headspace within each vial (equation 5).

$$5. C_{\text{corr}} \text{ (umol/ml)} * V_{\text{headspace}} \text{ (mL)} = N \text{ (umol)}$$

Overall CO<sub>2</sub> fluxes were determined by plotting the change in total CO<sub>2</sub> (N) by the time that each vial was sampled at (equation 6). The slope of the line was equivalent to the CO<sub>2</sub> flux in each vial per hour. These values were multiplied by the molecular mass of CO<sub>2</sub>, and then normalized by the mass of dry sediment in each vial.

$$6. \text{ CO}_2 \text{ Flux (ugCO}_2 \text{ g}_{\text{ds}}^{-1} \text{ hr}^{-1}) = \text{slope (N vs. time of sampling (hr)) * CO}_2 \text{ (g/mol) / g}_{\text{ds}}$$

*CH<sub>4</sub> Standards (for Dissolved CH<sub>4</sub> and Incubations)*

The response factor (Rf) for each daily run was calculated by plotting known concentrations (ppm) of the standards against the peak area documented for each standard. The Rf values were determined by taking the slope of the standard curves constructed for daily CH<sub>4</sub> measurements (equation 7).

$$7. \text{ Rf} = \text{slope (C (ppm) / response)}$$

*CH<sub>4</sub> Samples (for Dissolved CH<sub>4</sub> and Incubations)*

Initial flux concentrations were found by multiplying the individual response areas (peak areas) for the sample injections by the slopes of their associated standard curves (equation 8).\*\*

$$8. \text{ C (ppm)} = \text{sample response * standard Rf}$$

Equation 2 was used to correct for daily dilutions of headspace in each incubation vial (C<sub>corr</sub>). 10 mL of N<sub>2</sub> gas were injected into the incubation vials every day prior to pulling 10 mL of headspace out of the vials to be run for gas analysis – 5 mL for CO<sub>2</sub> and 5 mL for CH<sub>4</sub>.

$$9. \text{ C}_{\text{corr}} = \text{C (ppm) * (V}_{\text{headspace}} + \text{V}_{\text{dil}}) / \text{V}_{\text{headspace}}$$

Using equation 1,  $C_{\text{corr}}$  (ppm) values were converted to  $C$  ( $\mu\text{mol/mL}$ ). Equation 5 was used to calculate the concentration of  $\text{CH}_4$  in the headspace by multiplying the  $C$  ( $\mu\text{mol/mL}$ ) values by the total volume of headspace within each vial.

$$10. C_{\text{umol/mL}} = \frac{C_{\text{ppm}} \times (P \times 100 \times 0.98692327)}{(8.2054e^{-5})(T + 273.15)} \times 1e^{-6} * V_{\text{headspace}} \text{ (mL)} = N \text{ (umol)}$$

As with  $\text{CO}_2$ , overall  $\text{CH}_4$  fluxes were determined by plotting the change in total  $\text{CH}_4$  ( $N$ ) by the time that each vial was sampled at (equation 6). The slope of each line was equivalent to the  $\text{CH}_4$  flux in each vial per hour. These values were multiplied by the molecular mass of  $\text{CH}_4$ , and then normalized by the mass of dry sediment per vial.

$$11. \text{CH}_4 \text{ Flux (ugCH}_4 \text{ g}_{\text{ds}}^{-1} \text{ hr}^{-1}) = \text{slope (N vs. time of sampling (hr))} * \text{CH}_4 \text{ (g/mol)} / \text{g}_{\text{ds}}$$

*\*\* It is currently unclear, yet plausible, that the  $\text{CH}_4$  values are a vast underestimate of the actual sediment emissions that may have occurred in this study. The procedure that was used in this paper multiplies sample response peak areas by the standard  $R_f$  values (equation 8), when many other procedures call for sample responses to be divided by  $R_f$  values (Fundamentals of Gas Chromatography, 2012). Whether sample peak areas are multiplied by or divided by  $R_f$  values is dependent upon the machinery and analysis software used in experimentation. Because the machinery is located in Abisko, Sweden, it is unclear which method is most appropriate to use in this study, and it was thus decided that the reported results would error on the lower side of the estimates. Although the  $\text{CH}_4$  patterns visible within the lakes remain wholly representative of the relative rates of emissions by depth, a difference in methodology may have increased the overall point source concentrations of  $\text{CH}_4$  by a factor of  $10^6$ - $10^9$ .*

## Results and Discussion

Figure 1a

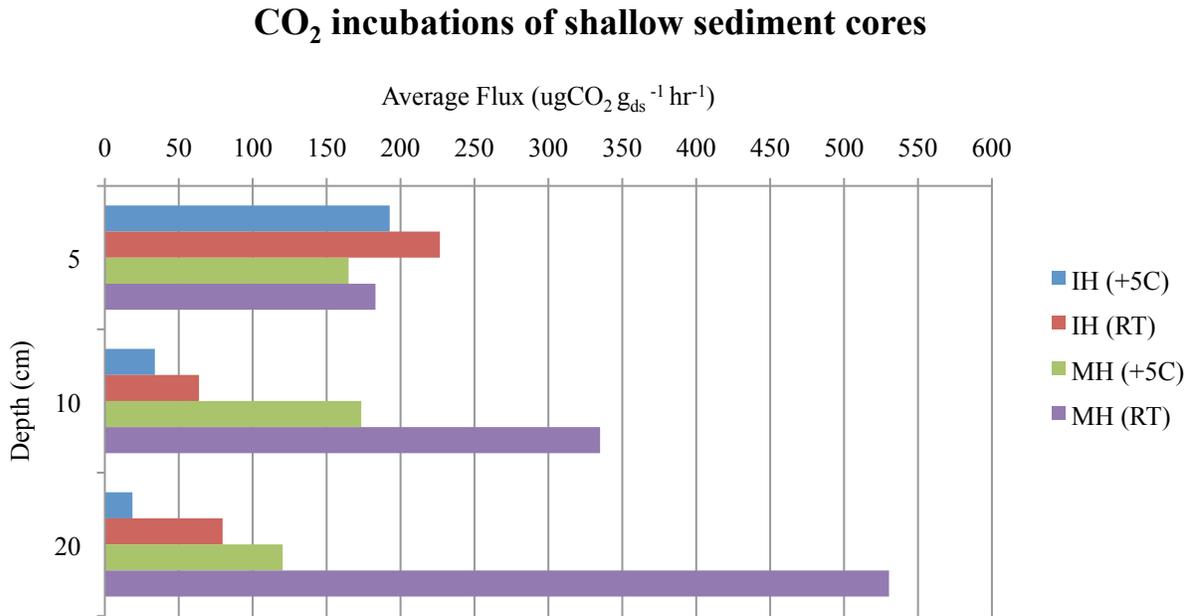


Figure 1b

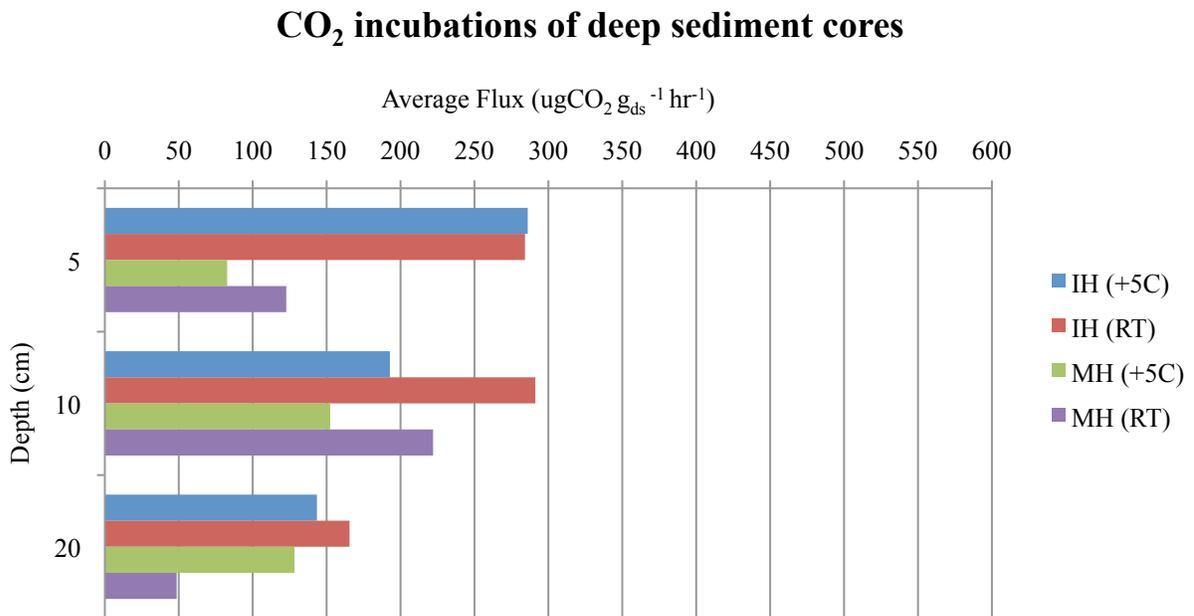


Figure 1. CO<sub>2</sub> emissions from (a) shallow coring depths and (b) deep coring depths, in lakes Inre Harrsjön (blue and red bars) and Mellan Harrsjön (green and purple bars). Bars represent fluxes of CO<sub>2</sub> from sediment that was sampled at three depths down each core and incubated at two temperatures for 13 days. CO<sub>2</sub> concentrations are expressed in  $\mu\text{gCO}_2 \text{ g}_{\text{ds}}^{-1} \text{ hr}^{-1}$ , where flux data is normalized by the mass of incubated dry soil ( $\text{g}_{\text{ds}}$ ).

Figure 2a

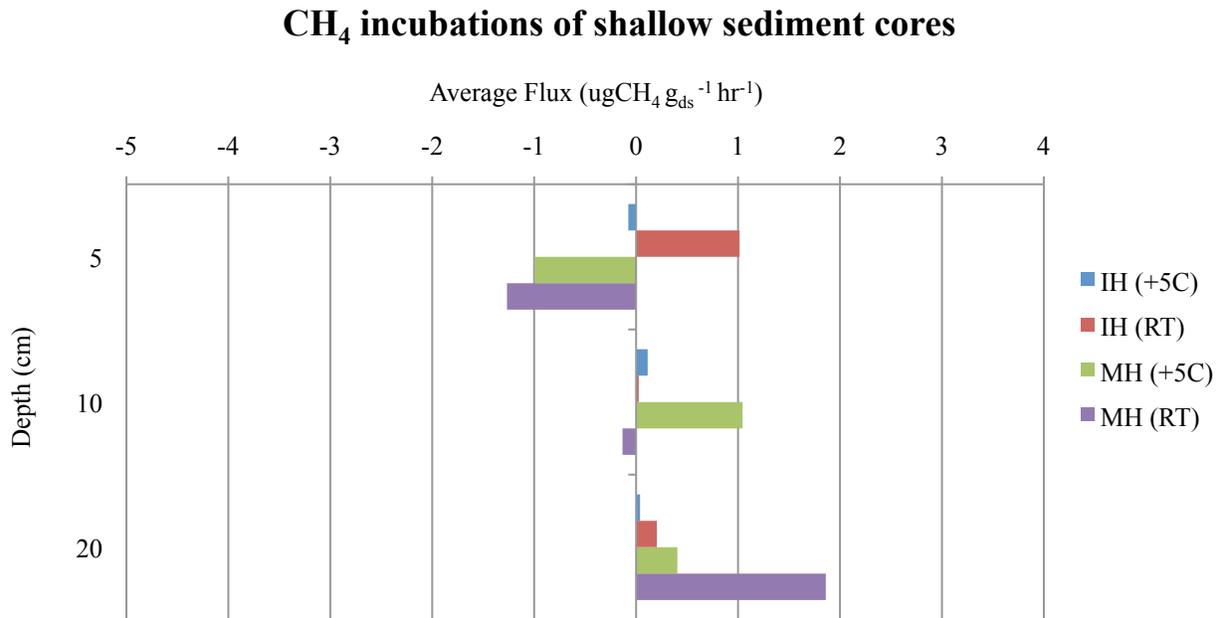


Figure 2b

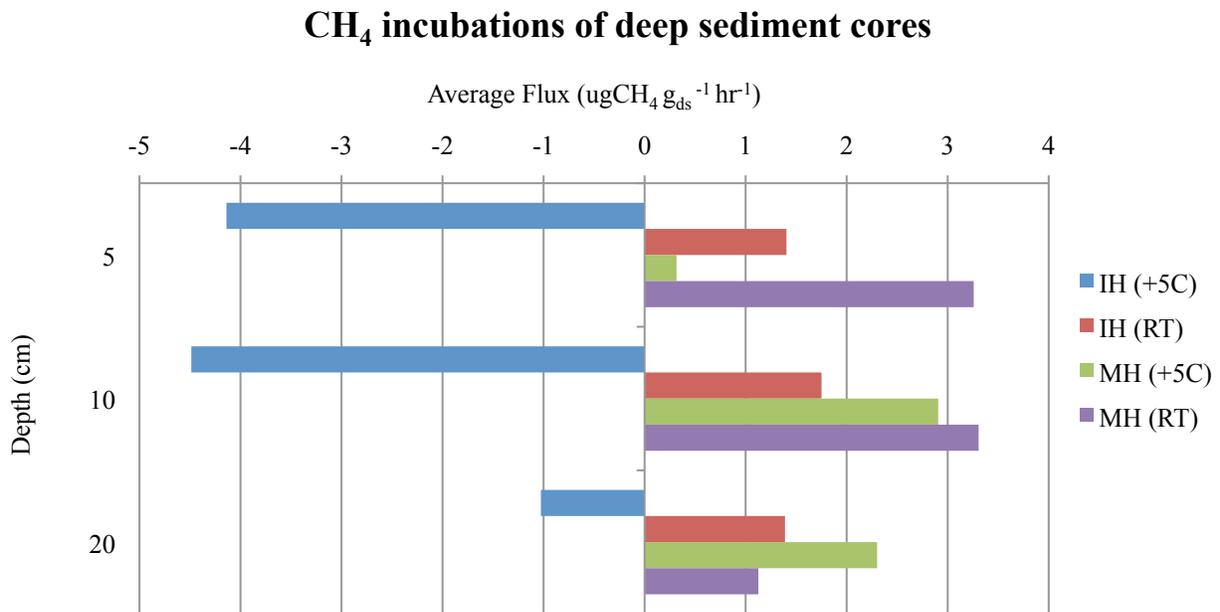


Figure 2. CH<sub>4</sub> fluxes from (a) shallow coring depths and (b) deep coring depths, in lakes Inre Harrsjön (blue and red bars) and Mellan Harrsjön (green and purple bars). Sediment was sampled at three depths down each core and incubated at two temperatures for 13 days. Bars represent emission and consumption of CH<sub>4</sub>, where emissions are positive values and consumptions are negative values. CH<sub>4</sub> concentrations are expressed in  $\mu\text{gCH}_4 \text{g}_{\text{ds}}^{-1} \text{hr}^{-1}$ , and flux data is normalized by the mass of incubated dry soil ( $\text{g}_{\text{ds}}$ ).

### *Sediment Incubations*

Trends in CO<sub>2</sub> and CH<sub>4</sub> emissions from the incubation studies were visible by core, lake, and incubation temperature. Within the cores, room temperature incubations for both Inre Harrsjön and Mellan Harrsjön demonstrated greater CO<sub>2</sub> fluxes than the +5°C incubations at the same depths (Figures 1a, 1b). This trend is also visible in the CH<sub>4</sub> incubations, where room temperature fluxes within each lake were larger than their respective +5°C fluxes (Figures 2a, 2b). In both CO<sub>2</sub> and CH<sub>4</sub> incubation studies, room temperature concentrations were up to five times larger than +5°C concentrations emitted over the two week period. These results indicate clearly that increasing atmospheric temperature (which will warm lakes and their underlying sediment) is correlated with greater effluxes of CO<sub>2</sub> and CH<sub>4</sub> from lake sediment into the atmosphere.

Within each individual lake, the deep cores had greater average CO<sub>2</sub> and CH<sub>4</sub> fluxes than the respective shallow cores (Figures 1a, 1b and 2a, 2b). Only the shallow cores from Mellan Harrsjön had greater CO<sub>2</sub> fluxes than the deep cores. This may be due to the fact that shallow cores were taken in close proximity to inflow from a connecting river, which might carry high concentrations of labile organic carbon that are deposited into the lake.

Comparison between lakes shows that average CO<sub>2</sub> fluxes from shallow cores in Mellan Harrsjön were greater than those from shallow cores in Inre Harrsjön, but deep cores in Inre Harrsjön had greater CO<sub>2</sub> fluxes than deep cores in Mellan Harrsjön (Figures 1a, 1b). In contrast, deep cores taken from Mellan Harrsjön had greater emissions of CH<sub>4</sub> than deep cores from Inre Harrsjön, and the shallow cores were variable in CH<sub>4</sub> emissions (Figures 2a, 2b). Again, some of the greater surface fluxes from Mellan Harrsjön sediment incubations may be explained by riverine deposits of organic carbon. The comparisons both within and between the lakes show

that the deep sediment is generally more productive than the shallow sediment. They also demonstrate that an increased input of organic carbon to one of two very similar lakes is enough to alter potential production rates of CO<sub>2</sub> and CH<sub>4</sub> under elevated temperature conditions.

### *Sediment and Pore Water Degassing*

The sediment profiles of DIC (Figure 3) and dissolved CH<sub>4</sub> (Figure 4) in Inre Harrsjön and Mellan Harrsjön resemble one another closely. Both profiles show increasing concentrations of carbon stores with depth, which are generally consistent with the greater CO<sub>2</sub> and CH<sub>4</sub> fluxes from the deep sediment incubation studies. These kinds of DIC profiles are typically associated with  $\delta^{13}\text{C}$  isotopic analysis, which can provide a range of paleoenvironmental information about the sediment record, including carbon cycling processes relative to sediment stratification, terrestrial and river inputs, and atmospheric carbon sequestration (Myrbo, 2006).

Regrettably, the full biogeological record of the two lakes is not available in this paper because the  $\delta^{13}\text{C}$  signatures associated with the DIC profile have not been completed. Were this study to incorporate  $\delta^{13}\text{C}$  values it might be able to shed light on specific changes that these lacustrine systems have undergone, including changes in organic matter input from surrounding terrestrial environments, variability in microbial mineralization of organic carbon, changes in atmospheric CO<sub>2</sub> concentrations, and large-scale deposition of carbon from rivers and/or precipitation events. Despite the current unavailability of these results, the visible fluctuations in DIC concentration with depth demonstrate that there have been clear changes in carbon storage within the recent sediment record. As climate change is expected to further impact the balance between carbon storage and production, differences like these will be important in documenting the magnitude of environmental change that the Arctic will undergo.

## Dissolved inorganic carbon in porewater

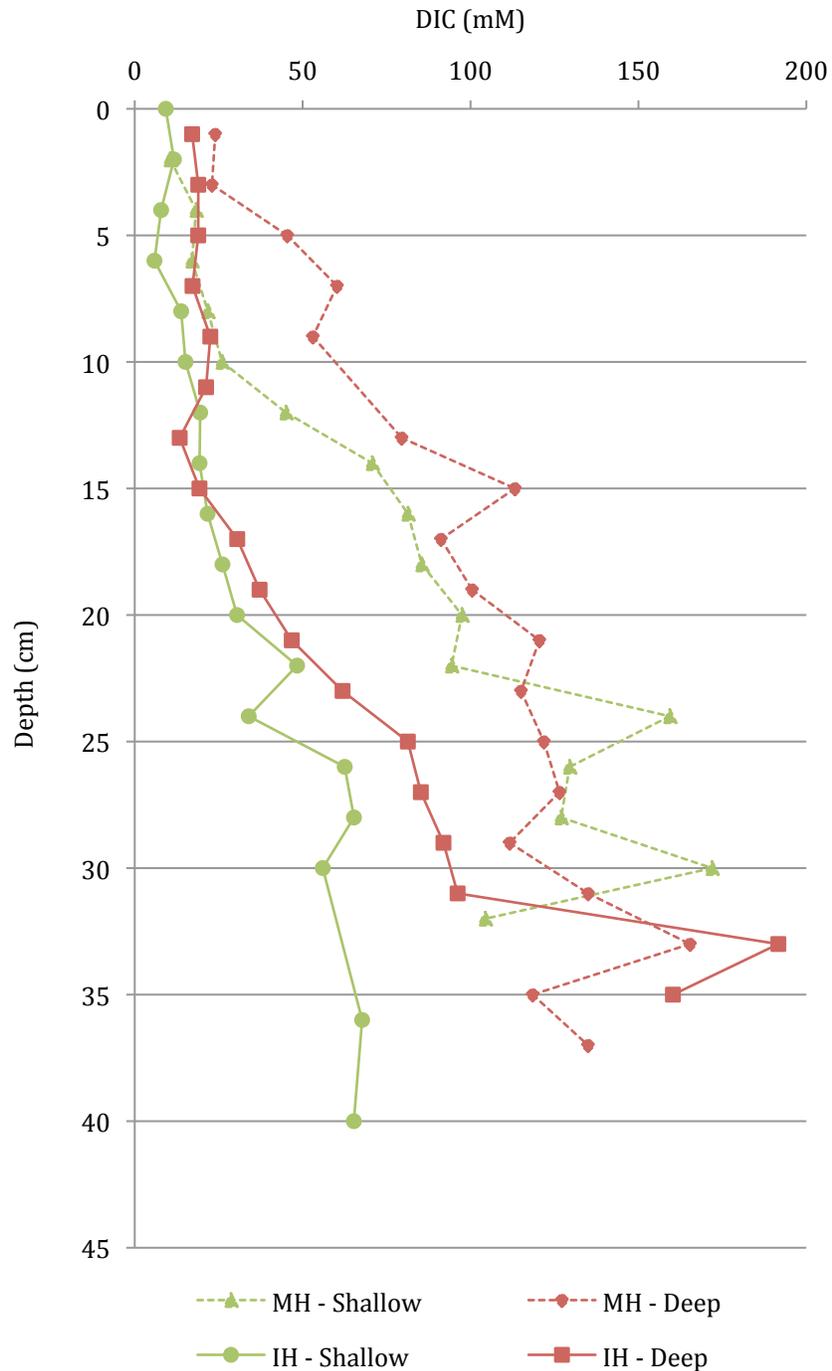


Figure 3. Sediment profiles of dissolved inorganic carbon (DIC) from shallow and deep coring locations in lakes Inre Harrsjön and Mellan Harrsjön. Data show increasing concentrations of DIC in porewater with depth, and parallel the porewater-sediment profiles for dissolved  $\text{CH}_4$  (Figure 4). DIC measurements are a sum of the concentrations of carbonic acid ( $\text{H}_2\text{CO}_3$ ), bicarbonate ( $\text{HCO}_3^-$ ), and carbonate ( $\text{CO}_3^{2-}$ ).

## Dissolved CH<sub>4</sub> in sediments and porewater

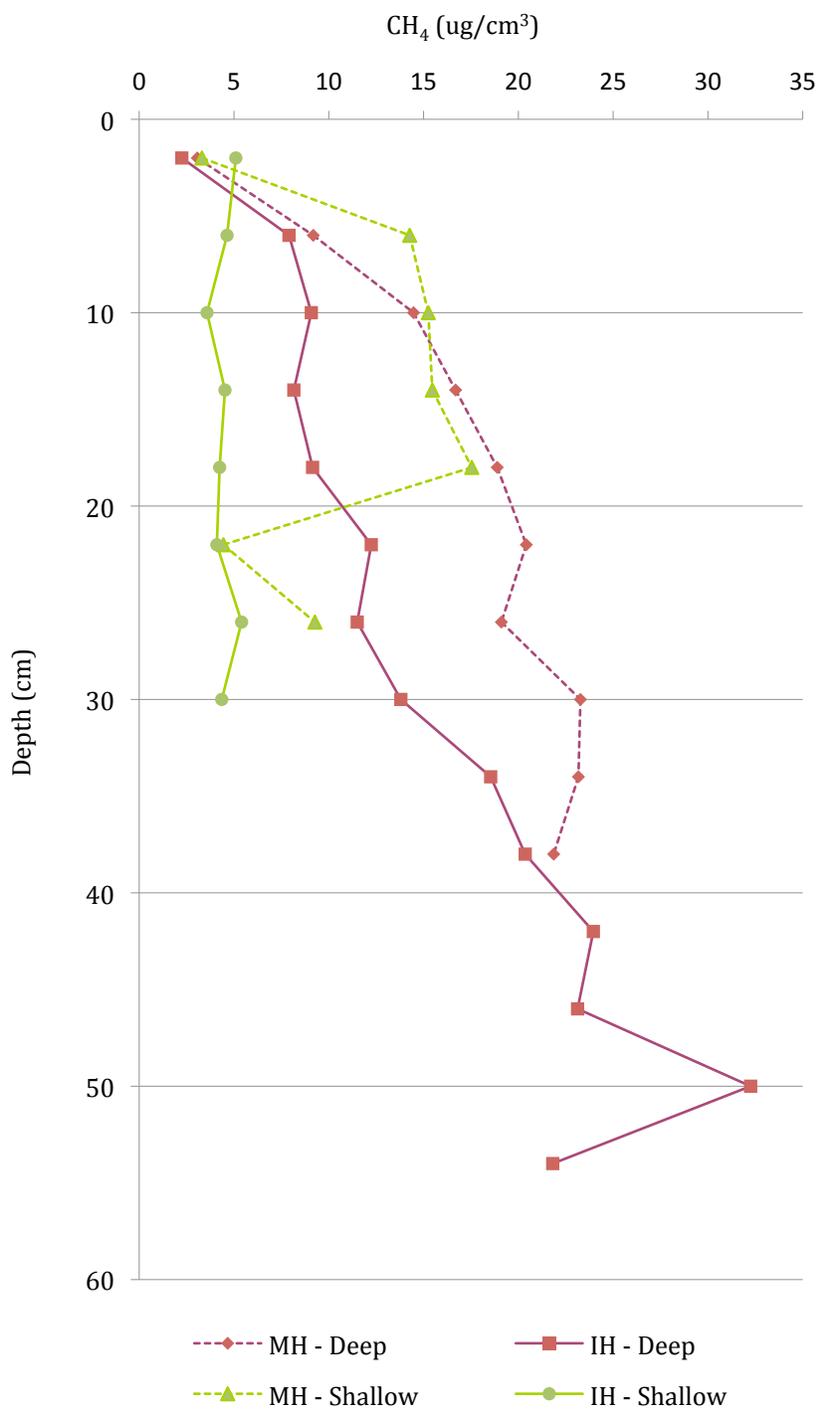


Figure 4. Sediment profiles of dissolved CH<sub>4</sub> from shallow and deep coring locations in lakes Inre Harrsjön and Mellan Harrsjön. Data show increasing concentrations of CH<sub>4</sub> in both porewater and sediment with depth, and parallel the trends visible in the porewater profiles for DIC (Figure 3).

### *The importance of including lake sediments in warming estimates*

Although one of the initial goals for this study was to scale up the lacustrine point source emissions to areal carbon fluxes for the Arctic, the data is limited in various factors that could misrepresent overall Arctic flux patterns unless they are researched in greater depth. For this reason, only point source emissions have been reported in the results. Current limitations of the study are largely due to the fact that a full analysis of the sediment, water, and atmospheric interactions in these lakes requires completion of the following six projects: (1) CO<sub>2</sub> and CH<sub>4</sub> potential production rates through incubation studies; (2) concentrations of dissolved CH<sub>4</sub> in both sediment and sediment porewater; (3) concentrations of dissolved inorganic carbon (DIC) in sediment porewater; (4) microbial community structures by sediment depth; (5) nutrient profiles for total organic carbon (TOC), total organic nitrogen (TON), and calcium carbonate (CaCO<sub>3</sub>); and (6) isotopic signatures by sediment depth ( $\delta^{13}\text{C}$ ) (Freitas, 2012). Due to funding and time constraints, the data has not yet been completed for projects 4-6, which makes it difficult to scale up small subsamples of sediment in each lake to whole Arctic efflux measurements. To attempt to quantify the total concentration of CO<sub>2</sub> and CH<sub>4</sub> that might be effluxed from all Arctic lakes (based solely upon differences in incubation temperatures) would assume that all lacustrine systems in the Arctic have similar biogeochemical composition and atmospheric interactions, and this would not be a true representation of these highly variable ecosystems.

While there are obvious constraints associated with the study until the full range of data can be synthesized, the results that were produced by the sediment incubations are hugely successful in that they provide some of the first point source microbial emissions by lake sediment depth. The data indicate that the difference in microbial decomposition at room temperature versus +5°C can increase CO<sub>2</sub> and CH<sub>4</sub> production by up to five times. It should be

noted that the difference between incubation temperatures (which is approximately 17°C) is 6°C higher than recent projected emission scenarios for the Arctic (IPCC, 2014). Arguably though, the true temperature change that the Arctic will undergo is currently unknown, and the goal of this research was to determine the potential production of greenhouse gases from sediment under a variety of warming scenarios. The data also demonstrates that the productivity of lake sediment may be significantly higher than the microbial productivity that has been documented in the anoxic zones of terrestrial peatlands. Total CO<sub>2</sub> respiration from anoxic peat ranges from 1.8 ugCO<sub>2</sub> g<sup>-1</sup> hr<sup>-1</sup> to 86.1 ugCO<sub>2</sub> g<sup>-1</sup> hr<sup>-1</sup>, and microbial CH<sub>4</sub> emission ranges from 0.2 x 10<sup>-3</sup> ugCH<sub>4</sub> g<sup>-1</sup> hr<sup>-1</sup> to 0.4 ugCH<sub>4</sub> g<sup>-1</sup> hr<sup>-1</sup> (Gilbert, 2006) – these values have been scaled up from the C numbers reported in the Gilbert *et al.* (2006) study (in ugC g<sup>-1</sup> hr<sup>-1</sup>) to their respective CO<sub>2</sub> and CH<sub>4</sub> fluxes using molecular mass ratios. The peat emissions are one to two orders of magnitude lower than those that were documented in the lake sediment incubations within this paper (Figures 1a, 1b, 2a, and 2b): CO<sub>2</sub> respiration ranged from 18.7 ugCO<sub>2</sub> g<sup>-1</sup> hr<sup>-1</sup> to 530.4 ugCO<sub>2</sub> g<sup>-1</sup> hr<sup>-1</sup>, and microbial CH<sub>4</sub> emission ranged from 0.026 ugCH<sub>4</sub> g<sup>-1</sup> hr<sup>-1</sup> to 3.3 ugCH<sub>4</sub> g<sup>-1</sup> hr<sup>-1</sup>. Not only does this data emphasize that northern peatland lakes have equally as much potential as their associated terrestrial systems, if not a greater potential, to be a source of CO<sub>2</sub> and CH<sub>4</sub> to the atmosphere, but also contributes to a relatively small body of research that shows that these overlooked systems have the ability to affect the spatial and temporal projections of warming in the Arctic (McGuire, 2009; Schuur, 2008; Walter, 2007; Zimov, 2006).

### *Future Research*

Continued research involves completing projects 4-6, as listed above, in order to gain a full understanding of how carbon cycling occurs in lakes Inre Harrsjön and Mellan Harrsjön. Until enough data can be compiled that suggests that Arctic lakes function similarly in their carbon sequestration and emission rates, future research should place an emphasis on studying the complexity of lakes and how they react to a variety of environmental factors.

In terms of projecting the impact of lacustrine systems on global warming, it is believed that it will be most important to gain a comprehensive understanding of how nutrient profiles affect the relative abundances of microbes in Arctic sediment. Recommendations include performing a microbial analysis of DNA using 16S rRNA gene amplicon sequencing to obtain a full profile of microbial community structures; these profiles can be associated with both subsurface nutrient data and microbial emission rates in the sediment, and also with surface effluxes of CO<sub>2</sub> and CH<sub>4</sub>. This way, subsurface lacustrine activity can ultimately be linked to lake surface and atmospheric interactions.

Other important areas of research will include expanding upon the sediment sampling techniques that were used in this study. Coring should be done on a monthly basis – from the beginning of the ice-free period until the lakes begin to refreeze. This will provide a higher temporal resolution of the ice-free period, as opposed to the quick snapshot that was gained in this baseline study. In order to obtain areal carbon fluxes for individual lakes, sampling should also occur across a wider area of the sediment and to deeper depths than were accessible within the time frame of this study. These changes will provide a higher spatial resolution of the lakes and, in combination with more frequent sediment sampling, can help create more representative models of atmospheric warming and carbon cycling in the Arctic.

## Conclusion

With global temperatures on the rise, the poles are expected to experience the fastest and largest increases in atmospheric temperatures and are thus anticipated to be early indicators of imminent climate changes. One of the principal concerns of ongoing polar research is how these increased temperatures will affect the immediate and long-term carbon exchange properties of the terrestrial ecosystems in the Arctic. While the land area represents a potential source for major environmental changes, lacustrine systems constitute up to half of the surface area of the Arctic and have been largely overlooked in most climate models. Due to a lack of accessible carbon exchange data, relatively few studies have been able to analyze the potential affects of global warming on Arctic lakes – and of those that have, the majority are focused on quantifying the concentrations of greenhouse gases released by ebullition events.

This paper does not attempt to answer the question of whether the Arctic will ultimately become a net source of carbon to the atmosphere, but rather provides some of the first point source data on Arctic sediment fluxes that explains how individual lakes may respond to a range of polar warming scenarios. Based on the results of the research, it is postulated that microbial productivity can increase the output of CO<sub>2</sub> and CH<sub>4</sub> concentrations from lake sediment by up to five times under elevated temperatures. The data emphasize the need to incorporate the full effects of northern peatland lakes in current and future climate models – not only the ebullition events that release greenhouse gases from lake surfaces, but also the underlying sedimentary processes that drive a more continuous release of CO<sub>2</sub> and CH<sub>4</sub>.

This study laid the groundwork for four continuing years of NSF-funded research, and is a small contribution to a growing body of data about the effects of anthropogenically driven climate change on Arctic lake systems.

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