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## Diffusion of $\text{CH}_4$ and $\text{CO}_2$ from subarctic lakes in Stordalen, Abisko

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## Abstract

Extensive amounts of carbon are stored in the Arctic region, and high latitudes lakes play a large role in the dynamics of methane ( $\text{CH}_4$ ) and carbon dioxide ( $\text{CO}_2$ ).  $\text{CH}_4$  is a very important greenhouse gas and one of the major emission pathways is via diffusion. During the ice-free seasons in 2016 and 2017, diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes were measured in three subarctic lakes in northern Sweden. 21 equilibration samplers were deployed in the lakes and 1050 measurements were taken, with 19 respectively 18 calculations of diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes. The diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes ranged between 2.57-14.41 respectively 3.82-61.64  $\text{mg m}^{-2} \text{d}^{-1}$ . Almost all flux and concentration ratios of  $\text{CO}_2:\text{CH}_4$  exceeded the 1:1 ratio, which is important in regards to estimations of global warming potential of northern lakes. With lake chambers were diffusive turbulence-driven water surface  $\text{CH}_4$  fluxes estimated and 298 measurements were taken. During 2016 (June to October) the fluxes ranged between 0.77-14.01  $\text{mg m}^{-2} \text{d}^{-1}$  and during 2017 (June to September) the fluxes ranged between 1.62-10.19  $\text{mg m}^{-2} \text{d}^{-1}$ . In general was the diffusive sedimentary  $\text{CH}_4$  fluxes higher than the diffusive turbulence-driven water surface  $\text{CH}_4$  fluxes, when compared during the same temporal period and spatial condition.

A systematic temporal variability was presented for the diffusive turbulence-driven water surface  $\text{CH}_4$  fluxes, with high fluxes during the warmest months July and August. Due to limited data of the diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes, no clear temporal pattern was presented. Distinct spatial trends could be distinguished in one out of the three lakes, with higher fluxes by the shore border which could be related to easily accessible labile carbon from mire deposits. How atmospheric pressure, air temperature, lake temperature and wind speed control the variability of the diffusive turbulence-driven water surface  $\text{CH}_4$  fluxes were analyzed. Increasing diffusive turbulence-driven water surface  $\text{CH}_4$  fluxes was presented with higher air- and water temperatures, as well as decreasing atmospheric pressure. The environmental parameters, including depth, was as well analyzed in relation to the diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes, were no significant correlations were shown.

With amplifying temperature rises in the Arctic region, it is of great importance to understand the controlling mechanisms of diffusive sedimentary and turbulence-driven water surface fluxes. Quantification of sedimentary diffusive fluxes from northern lakes are limited, and measurements are critical in order to further assess the global warming potential of high latitude lakes.

# Contents

<b>1</b>	<b>Introduction</b>	<b>2</b>
1.1	Background . . . . .	2
1.1.1	Effect of climate change in Arctic regions . . . . .	2
1.1.2	Arctic and the dynamics of CH <sub>4</sub> and CO <sub>2</sub> . . . . .	3
1.1.3	The role of northern lakes . . . . .	4
1.1.4	Estimations of diffusive fluxes via pore water sampling . . . . .	6
<b>2</b>	<b>Method</b>	<b>7</b>
2.1	Study site . . . . .	7
2.2	Measurements . . . . .	9
2.2.1	Equilibration samplers . . . . .	9
2.2.2	Lake chambers . . . . .	11
2.2.3	Water samples . . . . .	12
2.3	Calculation of diffusive fluxes . . . . .	13
2.4	Environmental parameters . . . . .	13
2.5	Statistical analysis . . . . .	14
2.6	Purpose and study aim . . . . .	15
<b>3</b>	<b>Results</b>	<b>17</b>
3.1	Environmental parameters during the study seasons 2016 and 2017 . . . . .	17
3.2	Equilibration samplers . . . . .	17
3.2.1	Comparison of equilibration samplers and water samples . . . . .	17
3.2.2	Overall sedimentary diffusive CH <sub>4</sub> and CO <sub>2</sub> fluxes . . . . .	20
3.2.3	Villasjön . . . . .	23
3.2.4	Inre Harrsjön . . . . .	30
3.2.5	Mellersta Harrsjön . . . . .	35
3.3	Lake chambers . . . . .	38
3.3.1	Overall water surface diffusive CH <sub>4</sub> fluxes . . . . .	38
3.4	Comparison of equilibration sampler and lake chamber fluxes . . . . .	40
<b>4</b>	<b>Discussion</b>	<b>43</b>
4.1	Environmental parameters . . . . .	43
4.2	Comparison of equilibration samplers and water samples . . . . .	44
4.3	Diffusive fluxes measured with equilibration samplers and lake chambers . . . . .	45
<b>5</b>	<b>Conclusion</b>	<b>49</b>
<b>6</b>	<b>Acknowledgements</b>	<b>50</b>
	<b>References</b>	<b>51</b>

# 1 Introduction

The Arctic has a great impact on the atmospheric carbon cycle. Cold and anaerobic environments inhibit decomposition, resulting in large amounts of carbon stored in the region. Extensive waterlogged areas in the Arctic create suitable habitats for methane ( $\text{CH}_4$ ) production, and it is one of the major natural sources of  $\text{CH}_4$  emissions. Carbon dioxide ( $\text{CO}_2$ ) and water vapor are more abundant in the atmosphere, but  $\text{CH}_4$  has a large impact on the climate system.  $\text{CH}_4$  have 28 times the global warming potential (GWP) compared to  $\text{CO}_2$  over a 100 year period, and is thereby the most important greenhouse gas after  $\text{CO}_2$  (Saunio et al., 2016). Estimated global emissions of  $\text{CH}_4$  ranges from about 542-852 Tg  $\text{CH}_4 \text{ yr}^{-1}$  but substantial uncertainties exist due to removal of  $\text{CH}_4$  by hydroxyl (OH) radicals and geographical overlapping sources of  $\text{CH}_4$  (Saunio et al., 2016).  $\text{CH}_4$  production is projected to increase with warming temperatures, and the Arctic is especially sensitive due to temperatures close to the melting point (McGuire et al., 2006). Large areas are underlain with permafrost in the northern hemisphere and during warming temperatures the frozen soil thaws, mobilizing carbon which are decomposed by microbes and converted into  $\text{CH}_4$  and  $\text{CO}_2$ . Temperatures have risen twice as much in the Arctic compared to low latitude regions due to polar amplification (McGuire et al., 2009). The effect induce warming in high latitudes due to changes in the cryosphere and transferring heat from the tropics (McGuire et al., 2006).

Northern lakes have a large role in  $\text{CH}_4$  and  $\text{CO}_2$  dynamics and approximate 40 % of the total global lake area is located north of  $50^\circ$  (Verpoorter et al., 2014). Lakes generally act as a net source for  $\text{CO}_2$  and a source for  $\text{CH}_4$  but emissions from lakes are not well understood or quantified. The primary reason for this is the great spatial and temporal variability of emissions (Wik et al., 2013). Diffusion is one of the main emission pathways from lakes and it is of great concern to understand how a warming climate will affect the emission rates of  $\text{CH}_4$  and  $\text{CO}_2$ . This study will measure diffusive sedimentary and turbulence-driven water surface fluxes from three subarctic lakes in Sweden, and analyze how environmental parameters might have affected the fluxes.

## 1.1 Background

### 1.1.1 Effect of climate change in Arctic regions

Anthropogenic climate forcing has since the industrial revolution disturbed the natural carbon cycle (IPCC, 2013). Between 1880-2012 has land and ocean temperatures increased by approximately  $0.86^\circ\text{C}$  [0.65-1.06] as a result of an increase in greenhouse gases (IPCC, 2013). Air surface temperatures in high northern latitudes are projected to amplify with significant warming in the Arctic region (Serreze & Francis, 2006). Between 1971 – 2000 has mean annual temperatures risen to  $1.5^\circ\text{C}$  (McGuire et al., 2009), and some climate models estimate an increase in mean annual temperatures of up to  $9^\circ\text{C}$  in Arctic by 2100 (Collins et al., 2013). The region is especially sensitive due to large areas underlain by permafrost and temperatures close to a mean annual isotherm of  $0^\circ\text{C}$ . Permafrost underlies about 25% of the landmass in the northern hemisphere (Brown et al., 1997), and the area has the highest amount of organic carbon on Earth (Vonk et al., 2012). Permafrost is perennially frozen soil and consists of an active layer on top that thaws every year. Thaw mobilize carbon in the permafrost, resulting in microbial decomposition that effects the balance of  $\text{CH}_4$  and  $\text{CO}_2$  emissions (Vonk et al., 2015). Increasing temperatures enhances thaw and mobilization of carbon resulting in permafrost-carbon feedbacks with impact on the atmospheric carbon cycle. Recent global climate models (GCMs) has not included all processes needed for projections of permafrost

changes hence future estimations are relatively uncertain (Collins et al., 2013).

The Arctic is prone to polar amplification which refers to processes that induce the amount of heat in the area (McGuire et al., 2006). The northern hemisphere has deficient radiative energy in comparison to tropical regions that contain a surplus, which drives the hemispheric atmospheric circulation. Water vapor content plays an important role which is transported from lower latitudes into the Arctic and increases the amount of heat by additional supply of moisture. A warming climate may enhance evapotranspiration and increase the amount of moisture content which induces the greenhouse effect, resulting in further temperature rise. Arctic regions contain limited amounts of water vapor and are thereby especially susceptible to concentration changes. Reduction of ice and snow content is another important process that increases the amount of absorbed solar radiation taken up by the surface due to lower albedo, and causes further decrease of the cryosphere (McGuire et al., 2006). Drastic changes have occurred during the last decades in the Arctic and there have been observations of a 75 % decrease of sea ice volume since 1980, thawing permafrost, vegetation shifts and an increase in soil temperature and precipitation, with some changes that may be irreversible on a century time scale (Hodgson et al., 2014; McGuire et al., 2009; Serreze & Francis, 2006; McGuire et al., 2006). The Arctic region has been, and will likely continue to be, dominated by positive feedbacks for the next 50 to 100 years, which would further induce warming in the region (McGuire et al., 2006).

### 1.1.2 Arctic and the dynamics of CH<sub>4</sub> and CO<sub>2</sub>

The Arctic contains large amounts of stored carbon and in the top 3 m of Arctic's land surface is about  $1035 \pm 150$  Pg carbon stored in perennially frozen soils (Schuur et al., 2015). Cold and wet areas in the Arctic inhibits decomposition, resulting in large repositories of carbon (Jahn et al., 2010). The amount of carbon that accumulates in the northern hemisphere is though small, and most carbon input occurred during Holocene (Smith et al., 2004) or Pleistocene (Zimov et al., 2006). A limited amount of the stored carbon is metabolized by microbes in the soil and emitted as CO<sub>2</sub> during aerobic conditions and CH<sub>4</sub> during anaerobic conditions. Large wetland areas in the Arctic regions promote CH<sub>4</sub> production and emissions of CH<sub>4</sub> is estimated to about 32-112 Tg each year (Overland et al., 2013).

Methanogenic Archea produces about two thirds by fermentation of acetate (CH<sub>3</sub>COO<sup>-</sup>) and one third by reduction of CO<sub>2</sub> (J. Ferry, 2010; Costa & Leigh, 2014). The reduction process occurs either with electrons from hydrogen (H<sub>2</sub>) or formate (HCOO<sup>-</sup>) (J. G. Ferry, 2012). Produced ratios of CO<sub>2</sub>:CH<sub>4</sub> present normally a ratio of 1:1, hence the same amount of moles of CO<sub>2</sub> and CH<sub>4</sub> are produced during methanogenesis (Wilson et al., 2017). Some field and incubation studies present results of exceeding 1:1 ratios, and Wilson et al., (2017) states that this is related to the molecules different oxidation states. The electron imbalance affect the hydrogenation processes and generates CO<sub>2</sub> without impacting the CH<sub>4</sub> production, resulting in elevated CO<sub>2</sub>:CH<sub>4</sub> ratios during certain spatial conditions.

Production of CH<sub>4</sub> is governed by different environmental parameters such as water table depth, soil temperature as well as substrate availability and quality (Ström et al., 2012). Temperature is a key driver of methanogenesis (Blake et al., 2015; Nozhevnikova et al., 1997; Yvon-Durocher et al., 2014). A study by Nozhevnikova et al. (1997) compared rates of methanogenesis at 5 °C with rates at 15 °C and 50 °C with observed increases by 5 respectively 20 times. Respiration and photosynthesis have much lower average temperature dependency than CH<sub>4</sub> production which results in a relatively large contribution to the global carbon emissions during warmer temperatures

(Yvon-Durocher et al., 2014). Optimum for CH<sub>4</sub> production is at 30 °C but methanogenesis can occur at much lower and higher temperatures (Blake et al., 2015).

Emissions of both CH<sub>4</sub> and CO<sub>2</sub> are estimated to increase in Arctic under a warming climate (Tranvik et al., 2009). Mainly due to increases in precipitation and mobilization of organic carbon stored in peatlands which could accelerate fluxes in the region.

### 1.1.3 The role of northern lakes

About 5 million m<sup>2</sup> of the Earth's surface is covered by lakes and about 40 % of this area is located by latitudes north of 50° (Verpoorter et al., 2014). Lakes are generally categorized in four different type of water bodies, with basis on how the lakes were formed. There categorizes are beaver ponds, glacial/post-glacial lakes, peatland ponds and thermokarst water bodies (Wik et al., 2016). The highest density of lakes occurs in previously glaciated areas where a large quantity has underlying permafrost that stores extensive amounts of soil organic carbon (Smith et al., 2007). Warming could induce permafrost thaw which would mobilize carbon that becomes available for microbial decomposition (Hugelius et al., 2013). Biological transitions have already occurred due to Arctic warming which has increased the primary production in the lake systems (Michelutti et al., 2005; Smith et al., 2007). Previously major shifts in the lake hydrological balance were seen during the Holocene. In Eurasia thawing permafrost changed the water storage and impacted all aspects of the biophysical systems (Smith et al., 2007). Today has indications of a shift been seen in Russia with an increase in groundwater mobilization and if larger changes would occur lake and wetlands could potentially disappear on a longer time scale (Smith et al., 2007).

About 16 % of the permafrost areas in the Arctic are covered by freshwater systems (Vonk et al., 2015). The distribution of northern wetland, lakes and ponds are largely affected by topographic relief, substrate geology, drainage patterns, periglacial and climatic processes (Smith et al., 2007). Large land surface areas in the Arctic region are relatively flat with low water flows, resulting in saturated soils with suitable conditions for wetlands to form (Gutowski et al., 2007). Even though the region has low amounts of precipitation the wetland areas are consistently wet during late spring and early fall due to thawing permafrost. High latitude areas are as well exposed to long summer days which increases the solar radiation and induces evapotranspiration processes (Gutowski et al., 2007). The magnitude of impact on northern freshwater ecosystems has increased during recent years due extensive permafrost thaw (Vonk et al., 2015).

Most lakes are net sources of CO<sub>2</sub> (Algesten et al., 2005) and carbon emitted as CO<sub>2</sub> from lakes are estimated globally at about 0.14 Pg yr<sup>-1</sup> (Cole et al., 1994). Global CH<sub>4</sub> emissions from lakes are estimated to 4-48 Tg yr<sup>-1</sup> and accounts for higher CH<sub>4</sub> emissions than oceanic sources (Bastviken et al., 2004). Northern lakes emit about 16.5 Tg CH<sub>4</sub> yr<sup>-1</sup> (Wik et al., 2016b).

Emissions of CH<sub>4</sub> from lakes are poorly understood due to large spatiotemporal uncertainties and few studies (Bastviken et al., 2004). A study by Wik et al., (2016) state that if the ice-free season increases with 20 days by the end of 2100 there is a risk that emissions from water bodies may increase with 20 to 54 %. Lakes contain favorable conditions for methanogenesis due to anoxic sediments and produced CH<sub>4</sub> in lake sediments are emitted via ebullition (bubble flux), diffusion and vascular plants (Tranvik et al., 2009; Bastviken et al., 2004; Ström et al., 2003). Diffusion and ebullition are considered the main emission paths from lake sediments (Wik et al., 2013).

### 1.1.3.1 Emission pathways

*Ebullition*, CH<sub>4</sub> bubble flux, can occur due to that CH<sub>4</sub> is a relatively insoluble gas. CH<sub>4</sub> bubbles are released from the lake sediment when the hydrostatic pressure has been overcome hence ebullition increases during low hydrostatic pressure (Martens & Val Klump, 1980; Bastviken et al., 2004). Rapid ascents inhibit oxidation from the sediment to the water surface and the net production has direct correlations to the CH<sub>4</sub> flux (Bastviken et al., 2004). Measurements conducted by Wik et al. (2013) showed that ebullition increased with warmer temperatures correlating to higher production rates of CH<sub>4</sub>. Ebullition has large temporal and spatial variations which causes difficulties in estimations (Wik et al., 2013).

*Diffusion* of CH<sub>4</sub> is an extremely slow process that occurs due to differences in molecular concentration between either the sediment-water column interface (diffusive sedimentary CH<sub>4</sub> fluxes) or the water surface-air interface (diffusive turbulence-driven water surface CH<sub>4</sub> fluxes). As an example; the diffusion coefficient for CH<sub>4</sub> in water is  $1.48 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$  at 15°C (Jähne et al., 1987), and by estimating the Brownian motion (MörTERS et al., 2010), it would take the molecules almost 11 years to diffuse through 1 m of water. This is a very robust estimation, and the time scale is as well affected by the initial concentration gradient. The slow rate of dissolved CH<sub>4</sub> results in large amounts being oxidized by methanotrophic bacteria if transport occurs in aerobic sediments or water columns (Bastviken et al., 2008). In contrary to the diffusive sedimentary CH<sub>4</sub> flux is the diffusive turbulence-driven water surface CH<sub>4</sub> flux not only driven by diffusion. The dissolved CH<sub>4</sub> in the water column is affected by turbulence and advection, whereas diffusion only occurs through the thin boundary layer by the water surface-air interface (Liss & Slater, 1974). The amount of dissolved CH<sub>4</sub> emitted from the water surface is mainly affected by turbulence caused by wind mixing patterns. Hence, the diffusive turbulence-driven water surface CH<sub>4</sub> flux is driven by differences in molecular concentration, as well as turbulence and advection. Hereafter is diffusive turbulence-driven water surface CH<sub>4</sub> fluxes shortened to diffusive water surface CH<sub>4</sub> fluxes in the thesis.

Diffusive CH<sub>4</sub> fluxes during the night have been shown to be 50 % lower due to decreasing winds that reduces the surface turbulence and limits the diffusive CH<sub>4</sub> flux (Walter Anthony & Macintyre, 2016). The diffusion rate at the water surface-air interface depends on Equation (1).

$$F = k(C_{aq} - C_{eq}) \quad (1)$$

$F$  is the diffusive flux [ $\text{mmol m}^{-2} \text{ d}^{-1}$ ],  $k$  is the gas exchange coefficient [ $\text{m d}^{-1}$ ], also called piston velocity,  $C_{aq}$  is the surface water gas concentration [ $\mu\text{M}$ ] and  $C_{eq}$  is the theoretical surface water concentration [ $\mu\text{M}$ ] during equilibrium with the air partial pressure (Cole & Caraco, 1998). Estimations of  $k$  is often related to wind speed based on empirical correlations (Schilder et al., 2013).

How heat loss from water surface affects the diffusive fluxes has been quantified by Poindexter et al., (2016), which observed an increase of diffusive fluxes during cooling conditions. Radiation emitted from the water surface during the night cools the water surface and due to higher density the cool water sinks. This results in the formation of eddies which create turbulence that accelerate the diffusive fluxes. This process occurs mainly during clear nights with no wind. The effect may also be induced by evaporation and conduction which cools down the water surface and can occur even when there is some wind. Poindexter et al., (2016) differ between large eddies in the water column which disperse the dissolved gas, and eddies by the water surface-air interface which increases the diffusive fluxes to the atmosphere. Hydrodynamic transport is used as a summarizing

term by Poindexter et al., (2016) for mixing processes driven by cooling but Walter Anthony and Macintyre (2016) state that the term includes several processes that influence dissolved gases and mention both internal- and surface waves. Previously measurements of diffusive fluxes have mainly been performed during day-time which has disregarded the diurnal patterns and a reassessment of the partitioning of the methane pathways may therefore be necessary. These cooling mixing patterns mainly occur in water bodies located in warmer regions but with projections of up to 9 °C in the Arctic the process may have a great impact on wetlands and lakes in the region (Poindexter et al., 2016; Walter Anthony & Macintyre, 2016; Bastviken et al., 2011; Collins et al., 2013).

Several environmental parameters affect diffusion of CH<sub>4</sub>. In a study by Natchimuthu et al. (2016) was diffusive CH<sub>4</sub> fluxes shown to be larger at shallow lake depths and warmer temperatures resulted in an exponential increase. It has also been observed that stratified lakes can build up CH<sub>4</sub> storages which can be drastically released via diffusion during seasonal turnovers (Bastviken et al., 2004). Rainfall affects the stratification in lakes and during periods with much precipitation stratification increases (Bartosiewicz et al., 2015). Bartosiewicz et al. (2015) observed larger quantities of dissolved CH<sub>4</sub> after rainfall when the stratification had been amplified.

*Vascular plants* has an important role in CH<sub>4</sub> dynamics. Certain species have been shown to supply substrates that are easily available to methanogenic microbes through processes in the root systems (Ström et al., 2003). Plant-mediated transport of CH<sub>4</sub> from the soil to the atmosphere occurs in vascular plants through their aerenchymatous tissue. An increase in biomass of specific vascular plants has shown to increase emissions of CH<sub>4</sub> (Ström et al., 2003).

#### 1.1.4 Estimations of diffusive fluxes via pore water sampling

Comprehensive studies of CH<sub>4</sub> diffusion fluxes from lakes are essential due to the significant role the process has in Arctic freshwater ecosystems. By measuring the concentration gradient by the sediment-water column interface, diffusive fluxes can be calculated and partitioned (Harper et al., 1997). Produced and oxidized CH<sub>4</sub> can be quantified and analyzed in relation to environmental parameters to gain insight into the controlling mechanisms of diffusive CH<sub>4</sub> fluxes.

One way to measure diffusive sedimentary CH<sub>4</sub> fluxes is by using EQ samplers (also called peeper or diffusion dialysis samplers) and analyzing pore water samples via headspace EQ. EQ samplers are used for in situ measurements and consist of a multichambered EQ dialyzes which collects interstitial water from sediment pores (Webster et al., 1998). The technique produces concentration profiles of chemical compounds which can be further analyzed. The flux calculations are entirely dependent on accurate measurements of the concentration profile (Harper et al., 1997). That produced concentration gradients have systematic measurement inaccuracies are well understood but to what degree the inaccuracies affect the flux estimations are mainly unquantified (Harper et al., 1997).

Hesslein (1976) first developed the EQ sampler and proposed an EQ time of about one week. Reported EQ times vary from 3 to 20 days. After the development of the EQ technique did Carignan (1984) identify parameters that limit EQ such as sediment porosity, adsorption to the solid phase, temperature and specie-diffusivity. Besides mentioned parameters, the EQ time are also affected by the ratio of the area to volume compartments and dissolution to the solid phase (Van Oploo et al., 2008). Different forms of pore water sampling exist in the form of pressure filtration, sediment cores, centrifugation, ceramic-cup suction, and in situ dialysis sampling such as the EQ sampler (Van Oploo et al., 2008).

The previous standard has been to obtain pore water concentration from sediment cores, but

these techniques have caused erroneous concentration gradients due to e.g. oxidation, low resolution and temperature changes (Van Oploo et al., 2008). EQ samplers used *in situ* eliminate the risk of manipulation of the concentration gradient due to sediment removal. Disadvantages with EQ samplers are e.g. averaging of the produced profiles and membrane breakdown. EQ samplers are otherwise a relatively simple technique to use and produce high-resolution profiles (Van Oploo et al., 2008), making it an alternative for estimating sedimentary diffusive CH<sub>4</sub> fluxes.

## 2 Method

### 2.1 Study site

The studied lakes Villasjön, Mellersta Harrsjön and Inre Harrsjön are located in the northmost part of Sweden. The lakes are situated in the mire Stordalen, Abisko (68°21'N, 19°02'N), see Figure 1. The mire consists of a large plateau (palsa) with permafrost, fens and bogs, with ombrotrophic and minerotrophic areas of varied wetness (Kokfelt et al., 2009; Olefeldt & Roulet, 2012). The area around Abisko is affected by a rainshadow resulting in low amounts of precipitation (Callaghan et al., 2010). In between 1913 to 2006 the precipitation increased from 306 mm yr<sup>-1</sup> to 336 yr<sup>-1</sup> (Olefeldt & Roulet, 2012). Measurements from Abisko Scientific Research Station (ANS) has observed an increase of the mean annual temperature by 2.5 °C between 1913 to 2006, which are now above zero at 0.7 °C (Callaghan et al., 2010). Thawing permafrost is turning palsas into fens and an increase of wet areas has been observed in the area, likely due to permafrost degradation (Kokfelt et al., 2010; Malmer et al., 2005). A vegetation shift has occurred which may be a result of an increase in spring temperatures with higher rates of snow melt and induced risk of frost drought. The vegetation shifts have resulted in higher litter input and an increase in carbon loss due to erosion (Malmer et al., 2005). An increase of nutrient may occur in the lakes due to the mobilization of mire deposits (Kokfelt et al., 2010). Erosion around the lakes has though been limited and the lakes are mainly enclosed by birch forest and fens (Wik et al., 2013).

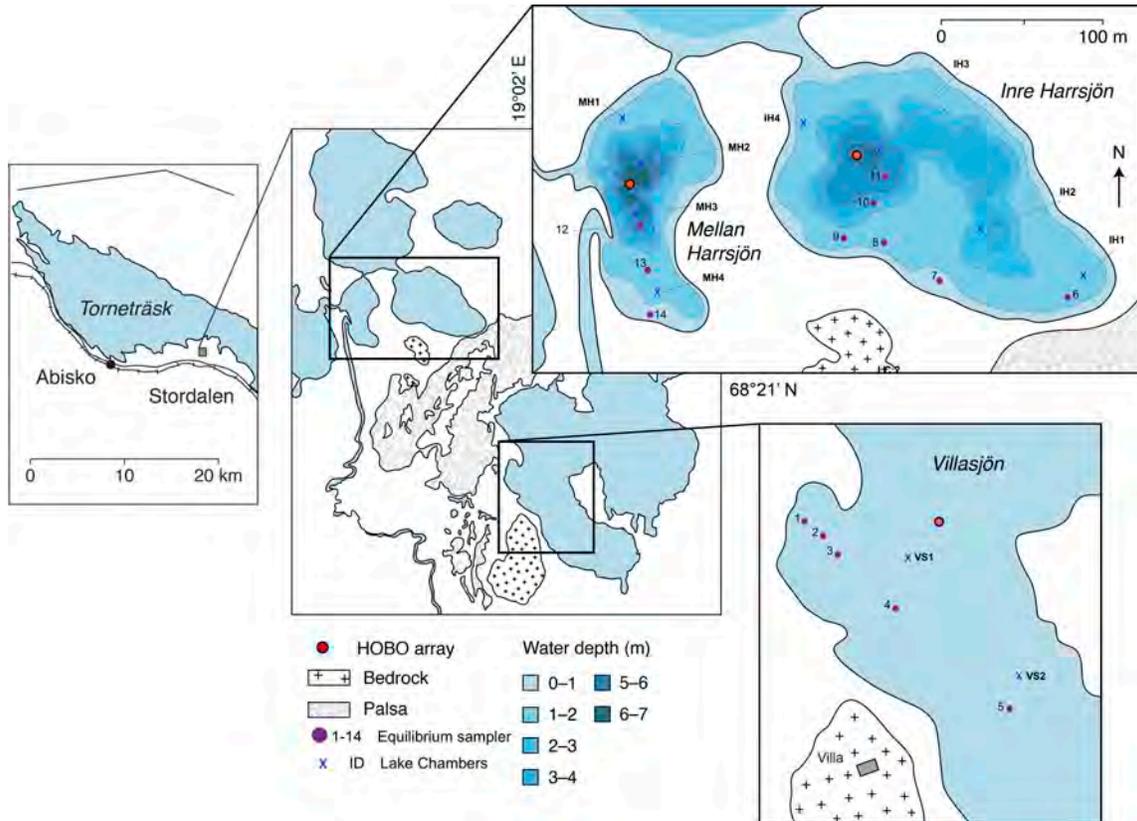


Figure 1: Studied lakes Villasjön, Inre Harrsjön and Mellersta Harrsjön in the northmost part of Sweden. Modified from Wik et al. (2013).

The studied lakes are categorized as postglacial lakes and contain no underlying permafrost. Villasjön has the largest area of about 17 ha but is relatively shallow in comparison to the other lakes Inre Harrsjön and Mellersta Harrsjön, see Table 1.

Table 1: Area, depth, and average zone-specific lake temperatures for each of the study lakes

Lake	Area (ha)	Water depth (m)		Mean lake temperature* (°C)		
		Mean	Maximum	Shallow (0-2 m)	Intermediate (2-4 m)	Deep (4-7 m)
Inre Harrsjön	2.2	2.0	5.0	12.6	12.2	10.0
Mellersta Harrsjön	1.1	1.8	7.0	11.4	10.3	8.0
Villasjön	17.0	0.7	1.3	12.5		

\*Mean lake temperatures were estimated by Wik et al. (2013) with temperatures measured in the lakes throughout the years 2009-2012. Modified from Wik et al. (2013).

Mellersta Harrsjön and Inre Harrsjön have the largest depth of maximum 7.0 m respectively 5 m whereas Villasjön only has a maximum depth of 1.3 m, resulting in bottom freeze during the winter period (Olefeldt et al., 2013). The lakes are generally ice-free from the middle of May to the middle of October (Wik et al., 2013). The pH in the lakes varies from approximately 6.5 to 7, with indications that the bedrock contains carbonate in the drainage areas (Kokfelt et al., 2010). The amount of dissolved organic carbon (DOC) in the lakes are estimated to about 10 mg L<sup>-1</sup> (Olefeldt

& Roulet, 2012).

All lakes partly receive water from aquifers (Nilsson, 2006). Mellersta Harrsjön and Inre Harrsjön are linked via the lake Yttre Harrsjön. Mellersta Harrsjön receives most surface water from a north flowing stream close by while Inre Harrsjön does not have a surface water source (Wik et al., 2013). Villasjön receive surface water from the same north-flowing stream as Mellersta Harrsjön and a small inflow east of the lake. The lake is connected to Mellersta Harrsjön and Inre Harrsjön when high water flows seep through the surrounded fen (Olefelt et al., 2013).

There is relatively limited information about the lake sediments (Wik et al., 2013). Measurements of the sediment in Inre Harrsjön and Villasjön has been performed by Kokfelt et al. (2010) at a depth of 3.5 m respectively 1.3 m. The first 275 cm layer in Inre Harrsjön contains of gyttja with laminated silt located below. The first 320 cm layer in Villasjön contain detritus gyttja and lower parts organic-rich silty clay. Villasjön has also been shown to be more stratified than Inre Harrsjön with 7 different stratigraphic zones compared to 3, based on geochemical and lithological changes (Kokfelt et al., 2010). A similar measurement for Mellersta Harrsjön has not been published.

## 2.2 Measurements

### 2.2.1 Equilibration samplers

EQ samplers modelled by the technique of Hesslein (1976) were used to retrieve high-resolution pore water concentration profiles of  $\text{CH}_4$  and  $\text{CO}_2$ , see Figure 2 and Figure 3. With the  $\text{CH}_4$  and  $\text{CO}_2$  concentration profiles were diffusive sedimentary fluxes calculated using Fick's first law (further described in section 2.3). The EQ sampler was made out of acrylic, with wells drilled into it every 0.9 cm. The EQ sampler had the dimensions 75 x 15 x 2 cm (length x width x depth). Each well was filled with about 12 mL MilliQ; deionized and distilled water. A 0.1 mm PTFE skiving tape was used to cover the wells in 2016 and a 0.05 mm PTFE skiving tape was used to cover the wells in 2017, which functioned as a membrane. The samplers were placed in the sediment-water column interface directly after each EQ sampler had been prepared. The samplers were deployed by hand in shallow parts or by the aid of a pole-like device in deeper parts. One measurement occurred with a diver when samplers were placed at certain deep parts of Inre Harrsjön. 3 samplers were deployed each time and the samplers were deployed in different lakes, depths and time periods, see Table 2.

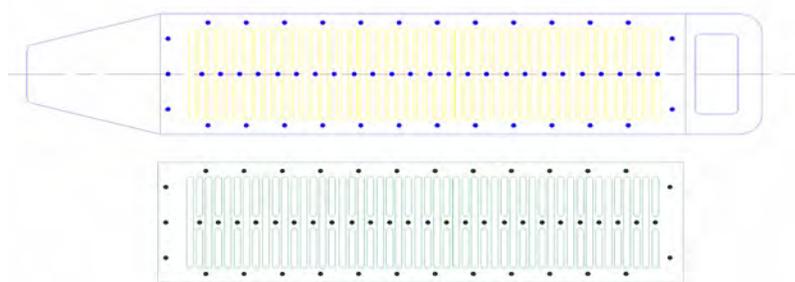


Figure 2: Schematic illustration of equilibration samplers used in study lakes. CAD drawing by Draupnir Einarsson, Department of Geological Science, Stockholm University.

After 9 to 14 days the samplers were retrieved. Transfer of the pore water from the wells occurred

in the field with 10 mL polypropylene syringes and needles. The process took between 18 to 52 minutes for each sampler. A rubber butyl stopper used to seal the needles on the syringes while a stopcock was used in combination with a needle in some measurements. If analysis did not occur the same day as sampling, the samples were stored and cooled down in a freezer to 2°C.

Table 2: Information about equilibration samplers

ID	Lake	Deployed	Deployment depth [cm]	Time deployed [Days]	Map*
VSI160801 (I)	Villasjön	2016-07-19	100	14	1
VSI160801 (II)	Villasjön	2016-07-19	100	14	2
VSI160801 (III)	Villasjön	2016-07-19	100	14	3
IHI160819 (I)	Inre Harrsjön	2016-08-11	100	9	9
IHI160819 (II)	Inre Harrsjön	2016-08-11	250	9	10
IHI160819 (III)	Inre Harrsjön	2016-08-11	400	9	11
VSI170629 (I)	Villasjön	2017-06-21	100	9	1
VSI170629 (II)	Villasjön	2017-06-21	100	9	4
VSI170629 (III)	Villasjön	2017-06-21	100	9	5
MHI170716 (I)	Mellersta Harrsjön	2017-07-04	150	13	14
MHI170716 (II)	Mellersta Harrsjön	2017-07-04	150	13	12
MHI170716 (III)	Mellersta Harrsjön	2017-07-04	150	13	13
IHI170731 (I)	Inre Harrsjön	2017-07-19	150	13	6
IHI170731 (II)	Inre Harrsjön	2017-07-19	150	13	7
IHI170731 (III)	Inre Harrsjön	2017-07-19	150	13	8
VSI170814 (I)	Villasjön	2017-08-03	100	12	1
VSI170814 (II)	Villasjön	2017-08-03	100	12	4
VSI170814 (III)	Villasjön	2017-08-03	100	12	5
VSI170924 (I)	Villasjön	2017-09-14	100	11	1
VSI170924 (II)	Villasjön	2017-09-14	100	11	4
VSI170924 (III)	Villasjön	2017-09-14	100	11	5

\*Locations marked in Figure 1.

flame ionization detector (FID). Injection loops of 25  $\mu\text{L}$  and 250  $\mu\text{L}$  were used for low respectively high concentration levels. A 2.059 ppm (Air Liquide) and a 2010 ppm (AGA)  $\text{CH}_4$  was used as a standard for low respectively high concentrations. The coefficient of variance for the low and high calibration standard measurements used during analyze of the equilibration samplers had an average of 1.22 % for low concentrations and 0.69 % for high concentrations, summarized in Table 1 in Appendices.



Figure 3: Equilibration sampler used in study lakes. Photo taken by Johannes West.

Gas samples were also run on a LI-COR 6262 infrared gas analyzer (IRGA) to assess the amount of CO<sub>2</sub>. A standard of 2000 ppm CO<sub>2</sub> (Air Liquide) was used. Before gas analysis on the IRGA occurred, a calibration was performed by using triplicate measurements of 5 different volumes (5.0, 3.0, 1.5, 0.5 and 0.2 mL) of the 2000 ppm standard to produce a response curve. Hence the peak area range of the calibration curve was used to bracket the peak areas of the samples. Gas samples were injected twice, in volumes of 0.5 ml and 1 ml during high respectively low concentrations. The coefficient of variance was normally below 2%.

The CH<sub>4</sub> and CO<sub>2</sub> concentration were calculated using the following Equations (2), (3) and (4).

$$n_{aq} = CX_{ppm} \times P_{air} \times \beta \times V_{water} \quad (2)$$

$n_{aq}$  is the amount of molar CH<sub>4</sub> or CO<sub>2</sub> in the water volume [mol],  $CX_{ppm}$  is the partition of CH<sub>4</sub> or CO<sub>2</sub> [ppm] in the gas volume,  $P_{air}$  is the atmospheric pressure 0.952 [atm],  $V_{water}$  is the volume of water [ml] and  $\beta$  is the Bunsen coefficient for CH<sub>4</sub> or CO<sub>2</sub> [mol L<sup>-1</sup> atm<sup>-1</sup>] at an ambient temperature of 15°C.

$$n_g = \frac{CX_{ppm} \times P_{air} \times V_{head}}{R \times (T + 273.15)} \quad (3)$$

$n_g$  is the amount of molar CH<sub>4</sub> or CO<sub>2</sub> in the gas volume [mol],  $CX_{ppm}$  is the partition of CH<sub>4</sub> or CO<sub>2</sub> [ppm] in the gas volume,  $P_{air}$  is the atmospheric pressure 0.952 [atm],  $V_{head}$  is the volume of gas [ml], R is the gas constant 8.31 [KPa L mol<sup>-1</sup> K<sup>-1</sup>] and T is the ambient temperature of 15°C.

$$CX_x = \frac{n_{aq} + n_g}{V_{water}} \quad (4)$$

$CX_x$  is the CH<sub>4</sub> or CO<sub>2</sub> concentration [ $\mu$ M],  $n_{aq}$  is the amount of molar CH<sub>4</sub> or CO<sub>2</sub> in the water volume [mol],  $n_g$  is the amount of molar CH<sub>4</sub> or CO<sub>2</sub> in the gas volume [mol] and  $V_{water}$  is the volume of water [ml].

For 21 deployments was a total of 1050 measurements performed in the study lakes at 14 different locations and time periods, see Figure 1 and Table 2. In Villasjön the deployments occurred at the same depth gradient, but noticeable was the rockier bottom at location 5. In Inre Harrsjön the deployments were placed at a depth gradient from 1 to 4 m. Deployment location 7, 8 and 9 was placed close to the fen border by the shore. Deployment location 8 was placed in a notable vegetated area and location 9 was placed in a thin organic layer in Inre Harrsjön. In Mellersta Harrsjön the deployments occurred at the same depth gradient, and deployment location 12 were placed close to the stream inlet.

No diffusive fluxes were calculated for IHI170731 since the majority of the sampler were placed in the water column, hence no appropriate sediment-water column interface could be distinguished in the concentration profile. A similar concentration profile was obtained with VSI170814 but with the difference that the sampler was placed almost entirely in the sediment. No CO<sub>2</sub> analysis was performed for sampler VSIII160801.

### 2.2.2 Lake chambers

Measurements of diffusive water surface CH<sub>4</sub> fluxes were performed using floating chambers, Figure 4. The lake chambers were placed over different depths in the lakes, see Figure 1. The chambers

were placed at 4 locations in Inre Harrsjön respectively Mellersta Harrsjön, while at 2 locations in Villasjön. At every location two chambers were placed, one with a semi-closed shield at the opening and one without. The shield was located about 40 cm below the surface and hindered CH<sub>4</sub> bubbles to enter, hence only flux values from the shielded chambers have been analyzed in this study. The opening area of the chamber was 0.061 to 0.066 m<sup>2</sup> and the headspace volume about 0.004 to 0.005 m<sup>3</sup> depending on the amount of infiltrated water. The chamber volume was noted down in the field and adjusted in the calculations. At each measurement, samples were taken with a 60 mL polypropylene syringe at the time of deployment, after 10 minutes and after 24 hours. The syringe was flushed with the gas inside the chamber three times before sampling. After each sampling, the lake chambers was taken out of the lakes to air. Samples were taken to ANS where the analysis of CH<sub>4</sub> was performed on the GC. A standard for low concentrations was normally used, and the coefficient of variance ranged between 0.04-4.29% during the two study years, and was in general below 1%.

About 14 to 16 samples were taken at each sampling location during the months June to October in 2016 and June to September in 2017. A total of 298 measurements were performed with the shielded chambers during the two study seasons. Measurements were generally taken once a week during the summer months. No CO<sub>2</sub> analysis were performed for the lake chamber measurements.

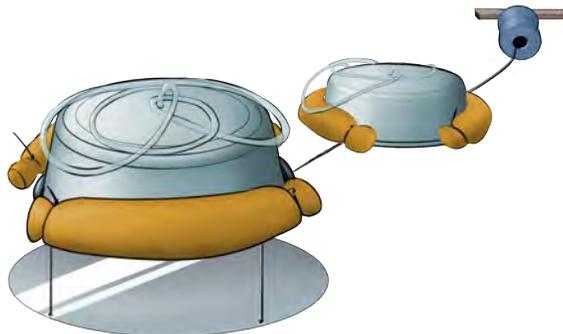


Figure 4: Schematic figure of lake chambers used in study lakes. Drawing by Noah Jansen.

### 2.2.3 Water samples

Water samples were collected approximately once a week in the study lakes, see locations in Figure 1. A 100 cm hose was used for depths up to 40 cm and a 750 cm hose, with an attached weight, was used for depths to approximate 650 cm. A 60 mL polypropylene syringe was used for cleaning the hose and taking water samples. The hose was cleaned by filling the syringe with water 5 or 7 times depending on the length of the hose. In Villasjön was water samples taken at a depth of 40 cm, in Inre Harrsjön and Mellersta Harrsjön was water samples taken at different depths from 40 cm down to 450 cm respectively 650 cm. Water samples were taken at 2 locations in Villasjön, 3 locations i Inre Harrsjön and 4 locations in Mellersta Harrsjön. Samples were prepared at ANS with the same procedure as for the EQ samplers and analyzed on the GC and IRGA, and the CH<sub>4</sub> and CO<sub>2</sub> concentrations were calculated using the following Equations (2), (3) and (4). Calibration standard measurements were used to estimate the uncertainty of the measurements and the coefficient of variance for selected water samples ranged between 0.26-0.83%. 21 water samples were taken approximately once a week during the months June to October in 2016 and 2017. A total of 861 measurements were performed during the two study seasons and 36 water samples,

which coincided with the deployment times of the EQ samplers, have been analyzed in this study.

### 2.3 Calculation of diffusive fluxes

All data has been processed using the software RStudio (R version 3.4.1). With the EQ samplers diffusive CH<sub>4</sub> and CO<sub>2</sub> fluxes were estimated with Fick's first law which has been modified by Berner (1971), see Equation (5).

$$J = -D'_s(\partial c/\partial z)_{pw}\Phi \quad (5)$$

$J$  is the sedimentary diffusive flux,  $\Phi$  is the porosity and  $(\partial c/\partial z)_{pw}$  is the concentration gradient.  $D'_s$  is the diffusion coefficients estimated by  $D_s = \Phi^2 D_0$  were  $D_0$  is the free solution diffusion coefficient for specific specie (Scranton et al., 1993). The porosity of the sediments in the study lakes was set to 0.9. Specific information about the porosity of the lakes was not available but measurements performed by Kokfelt et al. (2010) showed that the surface sediment layer in Villasjön and Inre Harrsjön contained gyttja, which normally has high porosity. The  $(\partial c/\partial z)_{pw}$  was calculated in [ $\mu\text{M dm}^{-1}$ ], diffusive fluxes  $J$  in [ $\text{mg m}^{-2} \text{d}^{-1}$ ] and  $D'_s$  in [ $\text{cm}^2 \text{s}^{-1}$ ]. The concentration profile was plotted and the  $(\partial c/\partial z)_{pw}$  was estimated by visually choosing linear measurement points, about 6 to 11, below the sediment-water column interface in order to estimate the diffusive flux using Fick's first law.

To estimate the value  $D_0$  in our study lakes Equation (6) were used.

$$D_0 = Ae^{-Ea/RT} \quad (6)$$

$D_0$  was first derived by Eyring (1936) using an empirical formulation.  $Ea$  is the diffusion activation energy in water. For CH<sub>4</sub> and CO<sub>2</sub> the  $Ea$  are 18.36 [ $\text{kJ Mol}^{-1}$ ] respectively 19.51 [ $\text{kJ Mol}^{-1}$ ] (Jähne et al., 1987).  $R$  is the gas constant, 8.31 [ $\text{kPa L mol}^{-1} \text{K}^{-1}$ ] and  $T$  is temperature [ $\text{K}$ ].  $D_0$  has been measured in [ $\text{dm}^2 \text{s}^{-1}$ ]. Mean temperature were estimated during the sampling period and used in Equation (6) to estimate  $D_0$  during the specific sampling period.

With the lake chambers were diffusive CH<sub>4</sub> fluxes estimated using Equation (7).

$$F = C \times \frac{1}{RT} \times M_{\text{CH}_4} \times \frac{V}{A} \times \frac{1}{1000} \quad (7)$$

$F$  is the turbulence-driven diffusive CH<sub>4</sub> flux [ $\text{mg m}^{-2} \text{d}^{-1}$ ] at the water surface,  $C$  is the CH<sub>4</sub> changes in concentration over time [ $\text{ppmv d}^{-1}$ ],  $R$  is the gas constant 8.31 [ $\text{kPa L mol}^{-1} \text{K}^{-1}$ ],  $T$  is the temperature [ $\text{K}$ ],  $M_{\text{CH}_4}$  is CH<sub>4</sub> molar mass 16.01 [ $\text{g mol}^{-1}$ ],  $V$  and  $A$  is the volume [ $\text{m}^3$ ] respectively the area [ $\text{m}^2$ ] of the lake chamber (Wik et al., 2016a).

### 2.4 Environmental parameters

Environmental parameters (see Table 3) used in the study has been monitored by Stockholm University (SU) and by the observation network Integrated Carbon Observation System (ICOS), Sweden. All environmental parameters have been measured on the Stordalen mire. Lake temperatures have been monitored by SU, for locations see Figure 1. HOBO Water Temp Pro v2, model u22, Onset Computer, sensors have been used, and lake temperatures have been measured with a 5 minutes

interval. The temperature has been measured at depths of 0.1 m, 0.3 m, 0.5 m and 1 m in all study lakes, whereas the deepest sensor in the lakes has been placed in the surface sediment. Since Inre Harrsjön and Mellersta Harrsjön is deeper than Villasjön, additional sensors have been used. Quality control has been performed via an inter-calibration of each sensor.

Table 3: Environmental parameters

Parameter	Equipment
Wind [m s <sup>-1</sup> ]	Weatherhawk
Atmospheric pressure [hPa]	Vaisala PTB210
Lake temperature [°C]	HOBO
Air temperature [°C]	Rotronic MP102H

Wind, atmospheric pressure and air temperature have been obtained from ICOS which perform long-term monitoring. Wind measurements have been logged every 30 minutes, atmospheric pressure every 30 minutes (at the height of 1.65 m) and air temperature every 30 minutes (at the height of 2.5 m). Quality control has been performed by ICOS by logging potential disturbances in the field and noting outliers

for elimination of erroneous values. No quality control has been performed on the 2017 data by ICOS. Outliers of presented environmental parameters have been removed in this study.

Monthly averaged environmental parameters were analyzed in relation to the monthly diffusive water surface CH<sub>4</sub> fluxes. The monthly averaged environmental parameters were measured June to October during 2016, and June to September during 2017. Measurements for water temperature varies with a few days in comparison to the other environmental parameters. The water temperature used for the monthly averages was measured at a depth of 0.1 m in the study lakes, in order to estimate general trends.

Environmental parameters measured during the deployment period for each EQ sampler were analyzed in relation to the diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes, measured with the EQ samplers.

## 2.5 Statistical analysis

The sediment-water column interface was visually chosen during take out of the EQ samplers and later confirmed or adjusted during analysis of the CH<sub>4</sub> concentration. When the CH<sub>4</sub> concentration increased an order of magnitude in the concentration profile, the sediment-water column interface was chosen at this location. The concentration profile was plotted and linear measurement points (6 to 11) were chosen directly below the sediment-water column interface in order to determine the concentration gradient  $(\partial c/\partial z)_{pw}$  for calculations of diffusive sedimentary CH<sub>4</sub> fluxes using Fick's first law. The equivalent measurement points for the CO<sub>2</sub> concentrations were chosen for calculation of diffusive sedimentary CO<sub>2</sub> fluxes using Fick's first law. For the concentration gradients  $(\partial c/\partial z)_{pw}$ , a linear regression analysis was performed. Additional information and statistics for the CH<sub>4</sub> and CO<sub>2</sub> concentrations, and  $(\partial c/\partial z)_{pw}$ , are summarized in Tables 5-10 in Appendices.

The uncertainty (flux error) of the diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes for individual samplers was calculated via an uncertainty propagation, see Equation (8).

$$\sigma f = \sqrt{(a^2\sigma^2)_A + (b^2\sigma^2)_B + (2ab\sigma)_{AB}} \quad (8)$$

The first term (A) was estimated using the concentration gradient  $(\partial c/\partial z)_{pw}$  and the coefficient of variance for the concentration gradient  $(\partial c/\partial z)_{pw}$ . The second term (B), was set to zero due to the numerous measurements (>thousands) of the lake temperatures, hence the mean lake temperature was set as a constant. The third term (AB) was set to zero due to no cross-correlation between  $D_s$

and the concentration gradient  $(\partial c/\partial z)_{pw}$ . The uncertainty of the sedimentary diffusive fluxes are presented in Figure 5, 6 and 7, as well as Tables 8, 10 and 12.

A statistical comparison of the CH<sub>4</sub> and CO<sub>2</sub> concentrations means, measured with the EQ samplers and the water samples, was intended to be performed with the purpose of evaluating how well the interstitial water in the EQ samplers had equilibrated with the lake water. Due to few measurements, temporal and spatial differences, sample groups with both non normal and normal distribution a statistical test of the significance would not be useful. An unpaired t-test and a Wilcoxon-Mann-Whitney u-test were evaluated as an option (Agresti & Franklin, 2007), but after expert consultation with J. Persson, Department of Mathematics, Stockholm University; personal communication, a comparison of the means and standard deviations (SDs) was chosen as a method. Means and SDs for the CH<sub>4</sub> and CO<sub>2</sub> concentrations, for EQ samplers and water samples, are presented in Tables 6 and 7. Additional information about the comparisons can be seen in Table 2 in Appendices.

A statistical comparison of the diffusive sedimentary and water surface CH<sub>4</sub> fluxes, measured with the EQ samplers and the lake chambers, was intended to be performed with the purpose of analyzing the differences in fluxes. But due to similar circumstances as mentioned above, a comparison of the means and SDs was chosen as a method. Means and SDs for the diffusive sedimentary and water surface CH<sub>4</sub> fluxes are presented in Table 17, 18 and 19. Additional information about the comparisons can be seen in Table 3 in Appendices.

A statistical comparison of the diffusive water surface CH<sub>4</sub> fluxes was performed after consultation with J. Persson, Department of Mathematics, Stockholm University; personal communication. The purpose was to compare how the monthly fluxes within the lakes differed between the study seasons 2016 and 2017. Due to static sampling locations (with averaged monthly fluxes per sampling location) a paired t-test was used for comparisons of the monthly fluxes. Since the number of measurements was few (n:2-4), a non numeric statistical test was excluded. The months July, August and September were chosen for comparisons while June and October were excluded due to few measurements. Since Villasjön only had two sampling locations no comparisons were performed on Villasjön due to the risk of biases, hence comparisons were only performed on the lakes Inre Harrsjön and Mellersta Harrsjön. The comparison of the diffusive water surface CH<sub>4</sub> fluxes are presented in section 3.3.1, and in Table 4 in Appendices.

Multivariate analysis and linear regressions have been performed between environmental parameters and diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes as well as diffusive water surface CH<sub>4</sub> fluxes in order to analyze how the environmental parameters control the variability of the diffusive fluxes.

No further statistical comparisons have been performed due to few measurements, temporal and spatial differences, which increases the risk for biases and false significance's.

## 2.6 Purpose and study aim

The aim of this study is to estimate diffusive CH<sub>4</sub> and CO<sub>2</sub> fluxes from three subarctic lakes by using equilibration (EQ) samplers (Hesslein., 1976) and lake chambers. Analysis of what impact specific environmental parameters might have on the fluxes will also be performed. Wind speed, atmospheric pressure, lake temperature and air temperature will be evaluated in relation to the diffusive fluxes. Further comparisons of in-lake and between-lake differences of diffusive fluxes will be done to asses spatial differences, as well as temporal variations throughout the season of the two study years 2016 and 2017. The purpose of this study is to gain further insight about sedimentary and turbulence-driven water surface diffusive fluxes from northern lakes. To following two research

questions have been proposed:

- *What is the net emission rate of methane and carbon dioxide from the study lakes?*
- *What environmental parameters control the variability of the methane and carbon dioxide diffusion-limited fluxes from the sediment and water surface?*

Data collection took place in the mire Stordalen, Abisko. The study period occurred between June to October in 2016, and between June and September in 2017. Since 2002 has a research group run by Patrick Crill, Stockholm University, managed a long-term monitoring program of carbon fluxes in the mire. In 2009 the research group started conducting measurements on the three lakes in the area; Villasjön, Inre Harrsjön and Mellersta Harrsjön. All measurements in this study have been performed on these three lakes in cooperation with SU and analysis have been done at ANS.

## 3 Results

### 3.1 Environmental parameters during the study seasons 2016 and 2017

Monthly averaged environmental parameters measured during the two study years are presented in Tables 4 and 5.

Atmospheric pressure shows relatively similar patterns during July and August in both 2016 and 2017. July and August were warmer during 2016 than 2017, while the opposite occurred for June and September when comparing the air temperature. July was the warmest month in both years. The wind speed patterns were relatively similar during 2016 and 2017. Notable is the higher wind speed observed in September 2017 in comparison to September 2016. The water temperatures were overall warmer at the beginning of the study season 2017 than in 2016 when comparing June month. All study lakes in 2016 show the warmest water temperatures in July with notable warmer temperatures in comparison to 2017. 2017 shows a dissimilar trend with the warmest water temperature observed in July for Villasjön, in June for Inre Harrsjön and August for Mellersta Harrsjön.

Table 4: Environmental parameters during 2016 with mean and standard error of the mean

	Atmospheric pressure [hPa]	Air temperature [°C]	Wind speed [m s <sup>-2</sup> ]	Villasjön Water temperature [°C]	Inre Harrsjön Water temperature [°C]	Mellersta Harrsjön Water temperature [°C]
June	971.49 ± 0.20	7.99 ± 0.20	4.83 ± 0.20	11.57 ± 0.04	11.42 ± 0.03	10.47 ± 0.03
July	967.55 ± 0.32	12.50 ± 0.30	4.78 ± 0.30	15.23 ± 0.03	15.36 ± 0.02	14.02 ± 0.02
August	968.54 ± 0.17	9.95 ± 0.17	4.50 ± 0.17	12.39 ± 0.03	13.05 ± 0.02	11.42 ± 0.02
September	969.06 ± 0.11	8.32 ± 0.11	4.87 ± 0.11	8.62 ± 0.01	8.80 ± 0.01	8.28 ± 0.01
October	983.73 ± 0.07	1.51 ± 0.07	4.46 ± 0.07	4.00 ± 0.02	4.70 ± 0.01	4.09 ± 0.02

*Water temperature have been averaged at a depth of 0.1 m in each lake.*

Table 5: Environmental parameters during 2017 with mean and standard error of the mean

	Atmospheric pressure [hPa]	Air temperature [°C]	Wind speed [m s <sup>-2</sup> ]	Villasjön Water temperature [°C]	Inre Harrsjön Water temperature [°C]	Mellersta Harrsjön Water temperature [°C]
June	966.40 ± 0.12	8.82 ± 0.12	4.42 ± 0.12	12.32 ± 0.03	14.48 ± 0.01	12.33 ± 0.02
July	968.06 ± 0.06	11.31 ± 0.07	4.27 ± 0.06	14.27 ± 0.03	12.92 ± 0.01	11.34 ± 0.02
August	966.46 ± 0.07	9.84 ± 0.07	4.50 ± 0.07	11.95 ± 0.03	14.87 ± 0.02	13.23 ± 0.02
September	976.46 ± 0.06	6.26 ± 0.06	5.22 ± 0.06	7.58 ± 0.02	9.06 ± 0.01	8.21 ± 0.01

*Water temperature have been averaged at a depth of 0.1 m in each lake.*

### 3.2 Equilibration samplers

A comparison of CH<sub>4</sub> and CO<sub>2</sub> concentration from the EQ samplers and water samples are presented in section 3.2.1, with the purpose of evaluating how well the equilibration has been performed with the EQ samplers. Overall diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes and ratios are presented in section 3.2.2. In section 3.2.3 to 3.2.5 the result from the EQ samplers is presented for each study lake: Villasjön, Inre Harrsjön and Mellersta Harrsjön.

#### 3.2.1 Comparison of equilibration samplers and water samples

A comparison of the CH<sub>4</sub> and CO<sub>2</sub> concentrations from the EQ samplers and water samples can indicate how well the interstitial water in the EQ samplers have equilibrated with the lake water. Due to few measurements at different locations and dates, a descriptive analyze will be performed by

comparing means and SDs. Except the potential for differences in concentrations due to temporal and spatial variations, the samples have been taken at different depths hence a potential variation could occur in the concentration due to the depth gradient. Other impacts are the oxidation of  $\text{CH}_4$  to  $\text{CO}_2$ . The lakes are though well mixed and small differences in the concentration occur during the summer months. A comparison of concentration mean, for EQ samplers and water samples, can therefore be used to evaluate how well the equilibration has been performed with the EQ samplers. Means and SDs of  $\text{CH}_4$  and  $\text{CO}_2$  concentrations, from EQ samplers and water samples, can be seen in Table 6 and 7.

Table 6: Comparison of  $\text{CH}_4$  concentration from equilibration samplers and water samples

ID	Equilibration samplers				Water samples			
	Deployment depth [cm]	n	Mean [ $\mu\text{M}$ ]	SD	Sampling depth [cm]	n	Mean [ $\mu\text{M}$ ]	SD
VSI160801	100	11	1.10	2.72	50	4	0.37	0.10
VSI160801	100	13	0.53	0.40	50	4	0.37	0.10
VSI170629	100	10	0.25	0.03	50	4	0.23	0.11
VSI170629	100	15	0.28	0.13	50	4	0.23	0.11
VSI170629	100	28	3.56	2.60	50	4	0.23	0.11
VSI170814	100	12	0.54	0.15	50	4	0.81	0.33
VSI170814	100	13	1.78	0.58	50	4	0.81	0.33
VSI170924	100	17	0.54	0.05	50	4	0.19	0.07
VSI170924	100	4	0.17	0.03	50	4	0.19	0.07
VSI170924	100	24	8.73	14.27	50	4	0.19	0.07
IHI160819	100	17	1.19	2.07	100	3	0.82	0.28
IHI160819	250	4	0.33	0.21	200 & 300	6	0.87	0.32
IHI160819	400	4	0.39	0.31	400	3	1.07	0.68
IHI170731	150	10	0.34	0.09	100 & 200	4	0.56	0.09
IHI170731	150	29	0.63	0.26	100 & 200	4	0.56	0.09
MHI170716	150	31	1.25	0.74	100 & 200	4	1.35	0.51
MHI170716	150	31	1.55	3.30	100 & 200	4	1.35	0.51
MHI170716	150	29	0.87	0.58	100 & 200	4	1.35	0.51

*Water samples that were taken during to the equilibration deployment period were chosen for comparison. The water samples were taken in the same lake as the deployed equilibration sampler. Mean values for the equilibration samplers were calculated by averaging the measurement points in the water column above the sediment-water interface. Additional information is presented in Table 2 in Appendices. VSI160801 was placed entirely in the sediment.*

Two EQ samplers in Villasjön, VSI170814 and VSI170924, has lower means compared to the water sample means, with a difference of -0.27 respectively -0.02  $\Delta\mu\text{M}$ , see Table 6. The last mentioned EQ sampler presents though a mean very close to the water sample mean. The EQ samplers VSI170629 and VSI170629 display means very close to water sample means with a difference of 0.02 respectively 0.05 $\Delta\mu\text{M}$ . The EQ sampler VSI170924 shows a notable higher mean than the water sample mean with a difference of 8.54 $\Delta\mu\text{M}$ . The EQ sampler present as well a large SD, hence the  $\text{CH}_4$  concentration varies greatly. Similar circumstances occur for the EQ sampler VSI170629 with a difference in mean of 3.33 $\Delta\mu\text{M}$  and a large SD. The remaining EQ samplers in Villasjön has a difference in mean of 0.16-0.97 $\Delta\mu\text{M}$ .

Three EQ samplers in Inre Harrsjön, IHI160819, IHI160819 and IHI170731, has lower means compared to the water sample means, with a difference of -0.54, -0.68 respectively -0.22 $\Delta\mu\text{M}$ . The EQ sampler IHI170731 has a mean very close to the water sample mean with a difference of

0.07 $\Delta\mu\text{M}$ . The remaining EQ sampler IHI160819 has a difference in mean of 0.37 $\Delta\mu\text{M}$

One EQ sampler in Mellersta Harrsjön, MHI170716, has a lower mean compared to the water sample mean, with a difference of -0.10, while the remaining two EQ samplers, MHII170716 and MHIII170716, has a difference in mean of 0.20 respectively 0.52 $\Delta\mu\text{M}$ .

This robust analysis could indicate that the EQ samplers VSI170629, VSII170629 and IHIII170731 have equilibrated well due to the small difference in means compared to the water sample means. If the EQ sampler mean is lower than the water sample mean, it indicates that the equilibration has not been completed. This is the case for the EQ samplers VSII170814, VSII170924 (though a small difference), IHII160819, IHIII160819, IHII170731 and MHI170716.

Table 7: Comparison of CO<sub>2</sub> concentration from equilibration and water samples

ID	Equilibration samplers				Water samples			
	Deployment depth [cm]	n	Mean [ $\mu\text{M}$ ]	SD	Sampling depth [cm]	n	Mean [ $\mu\text{M}$ ]	SD
VSI160801	100	11	46.72	36.41	50	4	63.32	29.46
VSI170629	100	10	108.43	41.41	50	4	39.30	9.10
VSII170629	100	15	110.63	48.32	50	4	39.30	9.10
VSIII170629	100	28	73.02	11.57	50	4	39.30	9.10
VSII170814	100	12	104.23	34.26	50	4	51.23	5.01
VSIII170814	100	13	109.13	20.83	50	4	51.23	5.01
VSII170924	100	17	84.53	23.51	50	4	41.19	7.86
VSIII170924	100	4	78.60	23.75	50	4	41.19	7.86
VSIII170924	100	24	101.45	27.62	50	4	41.19	7.86
IHI160819	100	17	141.13	50.77	100	3	67.45	18.50
IHI160819	250	4	135.06	36.96	200 & 300	6	75.52	26.86
IHIII160819	400	4	144.15	43.52	400	3	90.59	29.43
IHII170731	150	10	117.20	35.9	100 & 200	4	59.09	14.30
IHIII170731	150	29	256.46	36.56	100 & 200	4	59.09	14.30
MHI170716	150	31	198.25	23.25	100 & 200	4	114.33	13.79
MHII170716	150	31	309.77	224.42	100 & 200	4	114.33	13.79
MHIII170716	150	29	175.41	40.05	100 & 200	4	114.33	13.79

*Water samples that were taken during to the equilibration deployment period were chosen for comparison. The water samples were taken in the same lake as the deployed equilibration sampler. Mean values for the equilibration samplers were calculated by averaging the measurement points in the water column above the sediment-water interface.*

*Additional information are presented in Table 2 in Appendices.*

All EQ samplers show higher CO<sub>2</sub> concentrations than CH<sub>4</sub> concentrations, see Table 6 and 7. Almost all EQ samplers show a relatively large differences in CO<sub>2</sub> concentrations when comparing the means from the EQ samplers with the means from the water samples. Also noted is the general high SDs for the EQ samplers, hence large variations occur in the CO<sub>2</sub> concentrations. The SDs are a bit smaller for the water samples but the variation in CO<sub>2</sub> concentrations for the water samples are also relatively large. The EQ samplers with the highest and lowest CH<sub>4</sub> concentrations do not present the highest and lowest CO<sub>2</sub> concentrations. VSI160801 is the only EQ sampler that has a lower mean compared to the water sample mean, with a difference of -16.6 $\Delta\mu\text{M}$ .

### 3.2.2 Overall sedimentary diffusive CH<sub>4</sub> and CO<sub>2</sub> fluxes

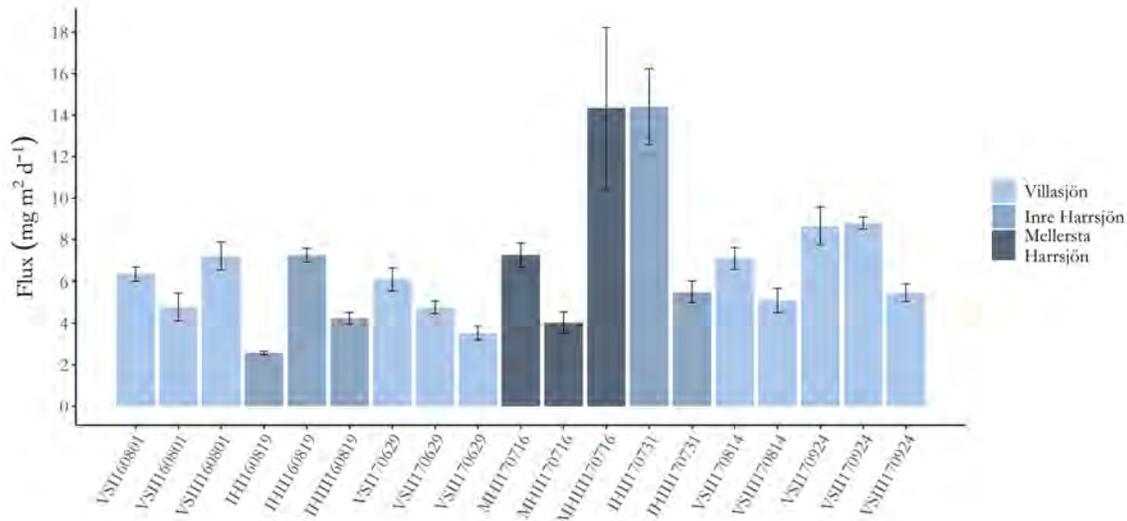


Figure 5: Diffusive sedimentary CH<sub>4</sub> fluxes measured with the equilibration samplers. Error bars represent the flux error.

The diffusive sedimentary CH<sub>4</sub> fluxes were calculated for 19 out of 21 EQ samplers. The diffusive CH<sub>4</sub> fluxes varied consistently with study year and lake, but no systematic variation was presented with measured environmental parameters. The diffusive CH<sub>4</sub> fluxes varied from 2.56 to 14.41 mg m<sup>-2</sup> d<sup>-1</sup> during the study seasons 2016 and 2017, see Figure 5.

The diffusive sedimentary CH<sub>4</sub> fluxes measured each study year were averaged. Annual mean and SEM of the diffusive CH<sub>4</sub> fluxes was estimated to 5.39 ± 0.75 in 2016 (n:6), and 7.31 ± 0.97 mg m<sup>-2</sup> d<sup>-1</sup> in 2017 (n:13).

An overall monthly division with mean and SEM presents July (n:5) with the highest diffusive CH<sub>4</sub> flux of 9.11 ± 2.21 and August with the lowest (n:8) diffusive CH<sub>4</sub> flux of 5.27 ± 0.60 mg m<sup>-2</sup> d<sup>-1</sup>, over the two study seasons.

An overall division by lake with mean and SEM presents Mellersta Harrsjön (n:3) with the highest diffusive sedimentary CH<sub>4</sub> flux of 8.55 ± 3.04, and Villasjön (n:11) with the lowest diffusive CH<sub>4</sub> flux of 5.27 ± 0.60 mg m<sup>-2</sup> d<sup>-1</sup>, over the two study seasons. Inre Harrsjön (n:5) ranges in between with a diffusive CH<sub>4</sub> flux of 6.79 ± 2.05 mg m<sup>-2</sup> d<sup>-1</sup>.

The deployment-period averaged environmental parameters (see Table 9, 11 and 13), were compared to the overall diffusive CH<sub>4</sub> fluxes. A multivariate analysis and linear regressions were performed for all diffusive sedimentary CH<sub>4</sub> fluxes and environmental parameters, including depth. No significant correlations were shown (All: R<sup>2</sup>:<0.02, p:>0.05) for the analyzed parameters.

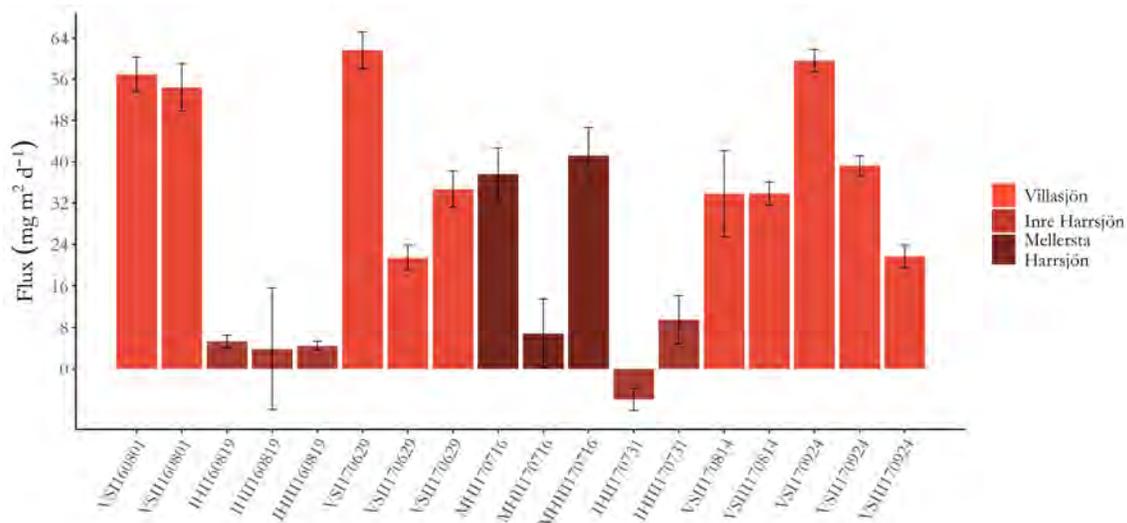


Figure 6: Diffusive sedimentary CO<sub>2</sub> fluxes measured with the equilibration samplers. Error bars represent the flux error.

The diffusive sedimentary CO<sub>2</sub> fluxes were calculated for 18 out of 21 EQ samplers. The diffusive CO<sub>2</sub> fluxes varied consistently with study year and lake, but no systematic variation was presented with measured environmental parameters. With the exceptions for depth and wind speed with very low, but significant correlation coefficients. The diffusive CO<sub>2</sub> fluxes varied from 3.82 to 61.63 mg m<sup>-2</sup> d<sup>-1</sup> (without extrema IHII170731) during the study seasons 2016 and 2017, see Figure 6.

The diffusive sedimentary CO<sub>2</sub> fluxes measured each study year were averaged. Annual mean and SEM of the diffusive sedimentary CO<sub>2</sub> fluxes was estimated to  $24.98 \pm 12.53$  in 2016 (n:5), and  $33.42 \pm 4.88$  mg m<sup>-2</sup> d<sup>-1</sup> in 2017 (n:13).

An overall monthly division with mean and SEM presents September (n:3) with the highest diffusive sedimentary CO<sub>2</sub> flux of  $40.17 \pm 7.18$ , and July (n:5) with the lowest diffusive CO<sub>2</sub> flux of  $23.79 \pm 9.06$  mg m<sup>-2</sup> d<sup>-1</sup>, over the two study seasons.

An overall division by lake with mean and SEM presents Villasjön (n:10) with with the highest diffusive CO<sub>2</sub> flux of  $41.72 \pm 4.83$ , and Inre Harrsjön (n:4) with the lowest diffusive CO<sub>2</sub> flux of  $5.78 \pm 1.28$  mg m<sup>-2</sup> d<sup>-1</sup>, over the two study seasons. Mellersta Harrsjön (n:3) ranges in between with a diffusive CO<sub>2</sub> flux of  $28.56 \pm 10.09$  mg m<sup>-2</sup> d<sup>-1</sup>.

The deployment-period averaged environmental parameters (see Table 9, 11 and 13), were compared to the overall diffusive CO<sub>2</sub> fluxes. A multivariate analysis and linear regressions were performed for all diffusive sedimentary CO<sub>2</sub> fluxes and environmental parameters, including depth. No significant correlations were shown (All: R<sup>2</sup>:<0.03, p:>0.05) for the analyzed parameters. A very low significant correlation was noted for depth (R<sup>2</sup>:0.25, p:0.03) and wind speed (R<sup>2</sup>:0.33, p:0.01), with increasing diffusive sedimentary CO<sub>2</sub> fluxes at shallower depths and higher wind speeds.

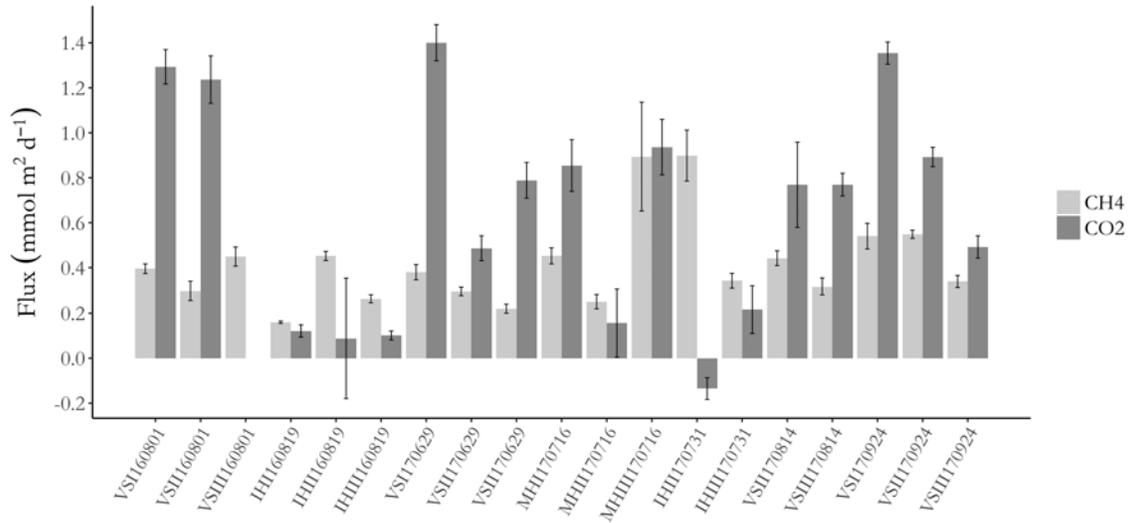


Figure 7: Diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes measured with the equilibration samplers. Error bars represent the flux error. Note mmol instead of mg on y-axis.

The diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes ranged between 0.16-0.89 respectively 0.08-1.40 mmol m<sup>-2</sup> d<sup>-1</sup> (without extrema IH1170731), see Figure 7. Highest flux ratio of CO<sub>2</sub>:CH<sub>4</sub> was seen in Villasjön (VSI170629, 5.29:1.00) and the lowest ratio in Inre Harrsjön (IH1160819, 0.22:1.00).

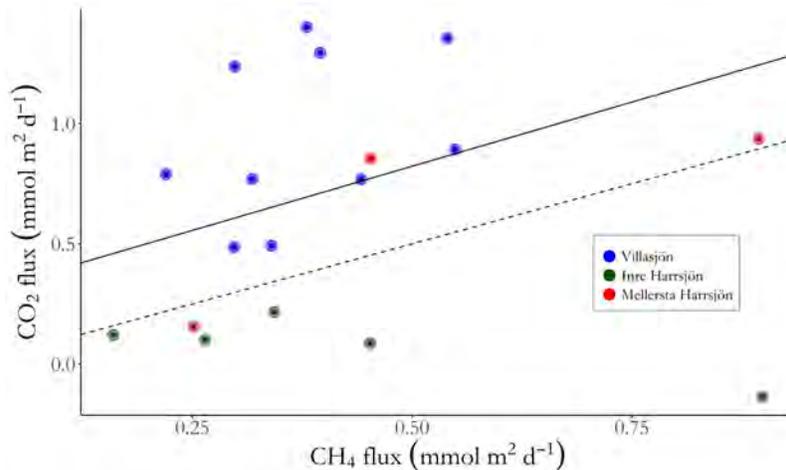


Figure 8: Diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes measured with the equilibration samplers. Solid line: overall linear regression (without extrema IH1170731), R<sup>2</sup>: 0.15. Dashed line: 1:1 line. Note mmol instead of mg on y- and x-axis.

The diffusive sedimentary CO<sub>2</sub> flux increases with the CH<sub>4</sub> overall for the three study lakes, see Figure 8. The CO<sub>2</sub> increases with 1.06 mmol m<sup>-2</sup> d<sup>-1</sup> per every CH<sub>4</sub> mmol m<sup>-2</sup> d<sup>-1</sup> but due to a low insignificant correlation no conclusions about the flux ratio can be made (R<sup>2</sup>:0.15, p:0.11). In Villasjön and Mellersta Harrsjön do the CO<sub>2</sub> increase with 1.16 (R<sup>2</sup>:0.12, p:0.31) respectively 1.04

( $R^2:0.64$ ,  $p:0.40$ )  $\text{mmol m}^2 \text{d}^{-1}$  per every  $\text{CH}_4 \text{ mmol m}^2 \text{d}^{-1}$ . Villasjön show a low insignificant correlation coefficient while Mellersta Harrsjön show a relatively high insignificant correlation coefficient that the  $\text{CO}_2$  flux increases with the  $\text{CH}_4$  flux. Inre Harrsjön show the opposite pattern of a decrease of  $\text{CO}_2$  with  $0.01 \text{ mmol m}^2 \text{d}^{-1}$  per every  $\text{CH}_4 \text{ mmol m}^2 \text{d}^{-1}$  ( $R^2:<0.00$ ,  $p:0.90$ ). But the correlation coefficient is insignificant and extremely low, hence no conclusions about how the  $\text{CO}_2$  flux vary with  $\text{CH}_4$  can be performed for Inre Harrsjön.

### 3.2.3 Villasjön

Three EQ samplers were placed in Villasjön (1 deployment) during 2016, and nine EQ samplers were placed in Villasjön (3 deployments) during 2017. One EQ sampler (VSI170814) were excluded in 2017. The diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes with flux errors and  $\text{CO}_2:\text{CH}_4$  ratios can be seen in Table 8.

Table 8: Diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes and  $\text{CO}_2:\text{CH}_4$  ratios from equilibration samplers

Date	ID	$\text{CH}_4$ flux and error [ $\text{mg m}^{-2} \text{d}^{-1}$ ]	$\text{CO}_2$ flux and error [ $\text{mg m}^{-2} \text{d}^{-1}$ ]	$\text{CO}_2:\text{CH}_4$ [ $\mu\text{M}$ ]	$R^2$	$p$
July 2016	VSI160801	$6.34 \pm 0.34$	$56.92 \pm 3.36$	4.70:1.00	0.92	<0.01
	VSI160801	$4.78 \pm 0.67$	$54.42 \pm 4.64$	5.91:1.00	0.84	<0.01
	VSI160801	$7.21 \pm 0.68$				
	Mean $\pm$ SEM	$6.11 \pm 0.71$	$55.67 \pm 1.25$			
June 2017	VSI170629	$6.09 \pm 0.54$	$61.64 \pm 3.54$	5.66:1.00	0.91	<0.01
	VSI170629	$4.77 \pm 0.30$	$21.40 \pm 2.44$	2.75:1.00	0.97	<0.01
	VSI170629	$3.53 \pm 0.32$	$34.71 \pm 3.49$	3.83:1.00	0.96	<0.01
	Mean $\pm$ SEM	$4.80 \pm 0.74$	$39.25 \pm 11.83$			
August 2017	VSI170814	$7.09 \pm 0.53$	$33.82 \pm 8.35$	2.40:1.00	0.94	<0.01
	VSI170814	$5.10 \pm 0.58$	$33.86 \pm 2.22$	4.00:1.00	0.94	<0.01
	Mean $\pm$ SEM	$6.10 \pm 0.81$	$33.84 \pm 0.02$			
September 2017	VSI170924	$8.66 \pm 0.92$	$59.60 \pm 2.17$	3.52:1.00	0.96	<0.01
	VSI170924	$8.80 \pm 0.29$	$39.26 \pm 1.89$	2.94:1.00	0.89	<0.01
	VSI170924	$5.45 \pm 0.41$	$21.65 \pm 2.18$	2.60:1.00	0.71	<0.01
	Mean $\pm$ SEM	$7.64 \pm 1.09$	$40.17 \pm 10.96$			

*Mean and SEM only calculated for fluxes, not flux errors.*

The diffusive sedimentary  $\text{CH}_4$  fluxes in Villasjön varied from  $3.53$  to  $8.80 \text{ mg m}^{-2} \text{d}^{-1}$ . The diffusive sedimentary  $\text{CO}_2$  fluxes varied from  $21.40$  to  $61.64 \text{ mg m}^{-2} \text{d}^{-1}$ . Out of the four deployments the EQ sampler VS170924 had the highest diffusive sedimentary  $\text{CH}_4$  flux mean while VS170629 had the lowest. The EQ sampler VS160819 had the highest diffusive sedimentary  $\text{CO}_2$  flux mean while VS170814 had the lowest.

The EQ samplers VS170629 (I-III), VS170814 (I-III) and VS170924 (I-III) were all placed at the locations 1, 4 and 5, see Figure 1 and Table 2. The EQ samplers VS160801 (I-III) were placed at locations 1, 2 and 3, all in the northern part of the lake. The highest diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes have been measured in the northern part of the lake, at location 1 and 3. The lowest diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes have been measured at the southern part of the

lake, at location 4 and 5.

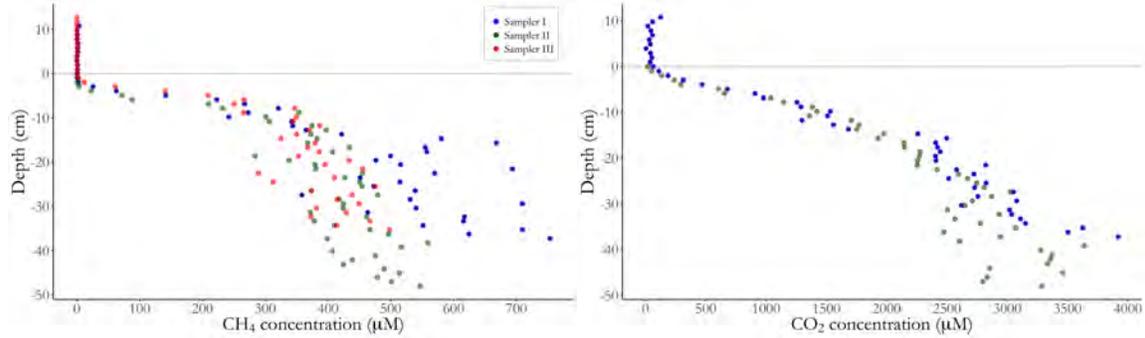
All estimated ratios of  $\text{CO}_2:\text{CH}_4$  exceeds the 1:1 ratio. The  $\text{CO}_2$  and  $\text{CH}_4$  concentrations measured for each deployment can be seen in Figures 9c, 11c, 13c and 15c.

Table 9: Mean and standard error of mean for environmental parameters and diffusive fluxes

ID	$\text{CH}_4$ flux [ $\text{mg m}^{-2} \text{d}^{-1}$ ]	$\text{CO}_2$ flux [ $\text{mg m}^{-2} \text{d}^{-1}$ ]	Atm. pressure [hPa]	Lake temp. <sup>1</sup> [°C]	Air temp. [°C]	Wind speed [ $\text{m s}^{-2}$ ]	<sup>2</sup> Depth [cm]
VS160801	$6.11 \pm 0.71$	$55.67 \pm 1.25$	$971.50 \pm 0.23$	$16.75 \pm 0.03$	$14.05 \pm 0.16$	$4.24 \pm 0.08$	100
VS170629	$4.80 \pm 0.74$	$39.25 \pm 11.83$	$962.00 \pm 0.33$	$11.92 \pm 0.02$	$7.88 \pm 0.12$	$4.53 \pm 0.08$	100
VS170814	$6.10 \pm 0.81$	$33.84 \pm 0.02$	$965.18 \pm 0.23$	$14.45 \pm 0.01$	$11.31 \pm 0.09$	$4.32 \pm 0.08$	100
VS170924	$7.64 \pm 1.09$	$40.17 \pm 10.96$	$975.80 \pm 0.53$	$6.89 \pm 0.02$	$5.81 \pm 0.09$	$4.61 \pm 0.11$	100

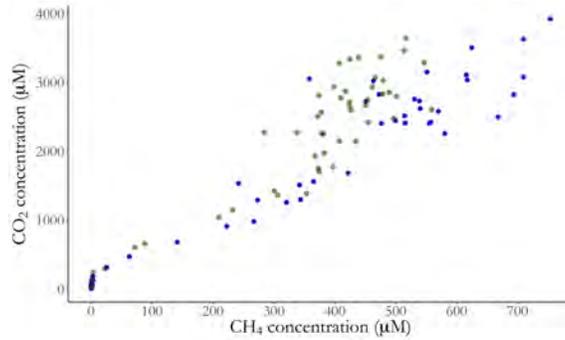
<sup>1</sup>Lake temperature measured at deployment depth. <sup>2</sup>Depth: deployment depth.

The environmental parameters measured during the deployment period for each EQ sampler in Villasjön are seen in Table 9, with mean and SEM. Multivariate analysis and linear regressions were performed for the diffusive sedimentary  $\text{CH}_4$  fluxes and the environmental parameters, excluding depth since all deployments occurred at a depth of 1 m. No significant linear relationships were shown ( $R^2 < 0.35$ ,  $p > 0.05$ ) for the analyzed parameters. The same result occurred for diffusive sedimentary  $\text{CO}_2$  fluxes, but with even lower insignificant correlations ( $R^2 < 0.08$ ,  $p > 0.05$ ).



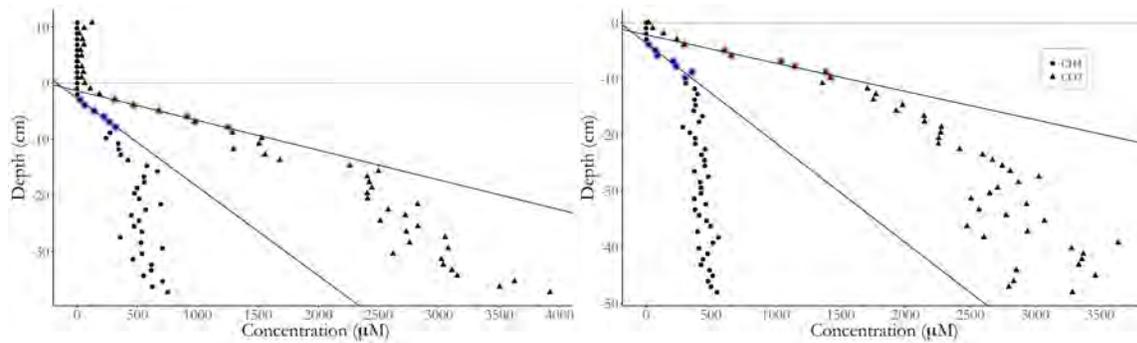
(a)  $\text{CH}_4$  concentration for VS160801.

(b)  $\text{CO}_2$  concentration for VS160801.



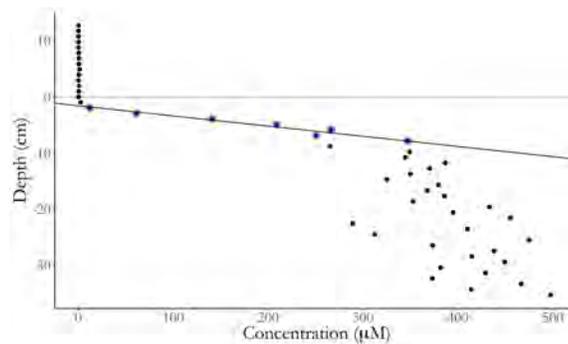
(c)  $\text{CH}_4$  and  $\text{CO}_2$  concentration for VS160801.

Figure 9:  $\text{CH}_4$  and  $\text{CO}_2$  concentrations for VS160801. No  $\text{CO}_2$  values for VS160801 (III).



(a) CH<sub>4</sub> and CO<sub>2</sub> concentration for VS160801.

(b) CH<sub>4</sub> and CO<sub>2</sub> concentration for VSII160801.

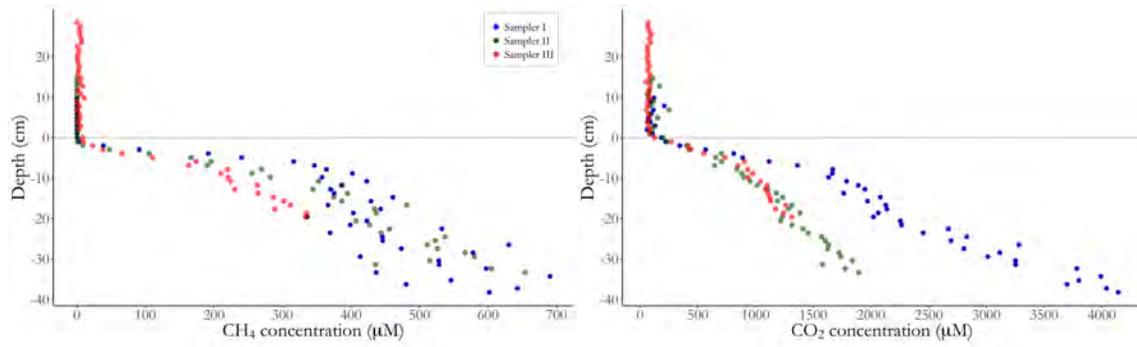


(c) CH<sub>4</sub> concentration for VSIII160801.

Figure 10: VS160801 with linear regression for CH<sub>4</sub> (blue) and CO<sub>2</sub> (red) measurement points which have been used in flux calculations. No CO<sub>2</sub> values for VS160801 (III).

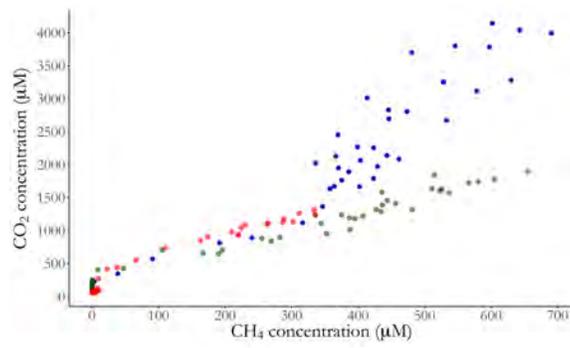
The EQ samplers VS160801 (I-III) were all placed relatively close to each other in Villasjön, see Figure 1. Sampler I had higher concentrations of CH<sub>4</sub> (maximum of approximate 800 $\mu$ M) than sampler II and III (maximum of approximate 500 to 600 $\mu$ M), see Figure 9a.

The selected measurement points for CH<sub>4</sub> and CO<sub>2</sub> has high correlation coefficients ( $R^2$ :0.91-0.99), see Table 5 and 6 in Appendices.



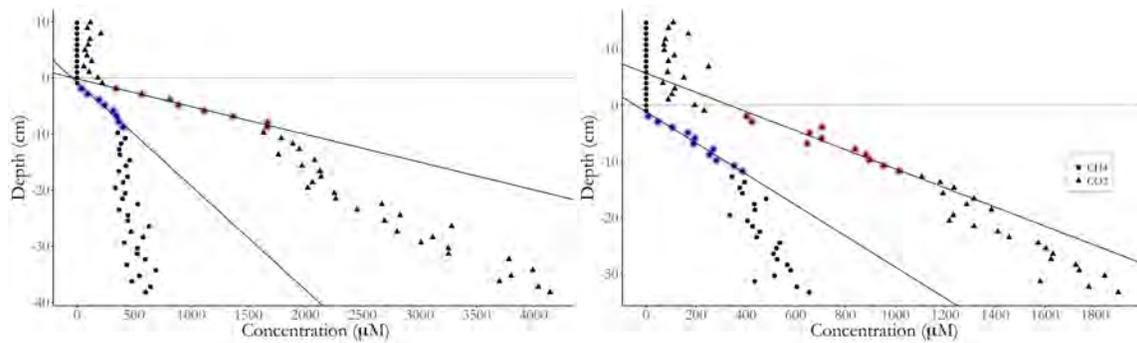
(a) CH<sub>4</sub> concentration for VS170629.

(b) CO<sub>2</sub> concentration for VS170629.



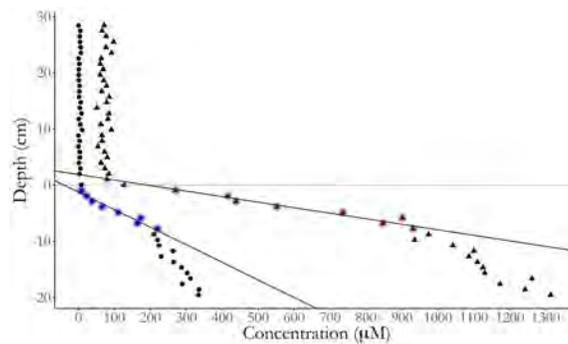
(c) CH<sub>4</sub> and CO<sub>2</sub> concentration for VS170629.

Figure 11: CH<sub>4</sub> and CO<sub>2</sub> concentrations for VS170629.



(a) CH<sub>4</sub> and CO<sub>2</sub> concentration for VSII170629

(b) CH<sub>4</sub> and CO<sub>2</sub> concentration for VSII170629.

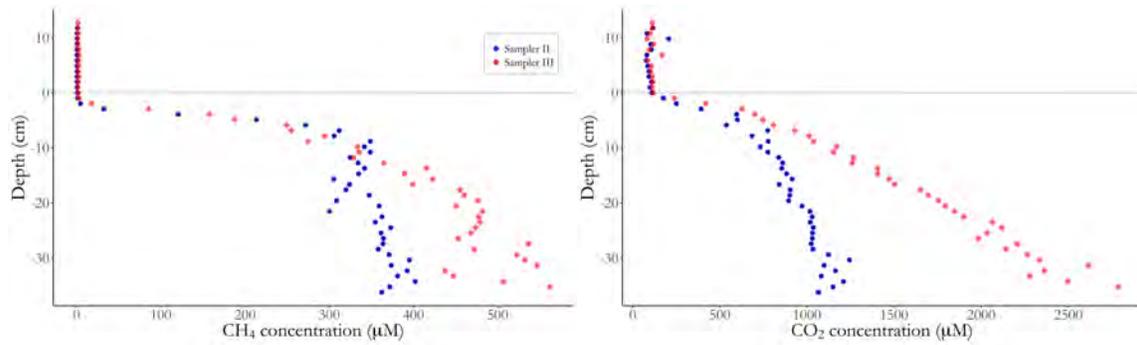


(c) CH<sub>4</sub> and CO<sub>2</sub> concentration for VSIII170629.

Figure 12: VS170629 with linear regression for CH<sub>4</sub> (blue) and CO<sub>2</sub> (red) measurement points which have been used in flux calculations.

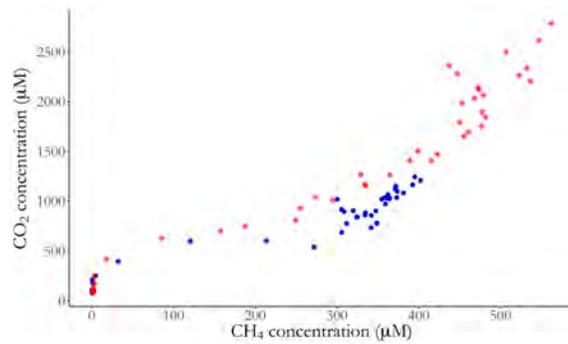
The EQ samplers VS170629 (I-III) were relatively spread out in Villasjön, see Figure 1. All samplers show relatively similar concentrations of CH<sub>4</sub>, see Figure 11a. Sampler I shows a higher concentration of CO<sub>2</sub> (maximum of approximate 4000  $\mu$ M) compared to sampler I and III (maximum of approximate 1500 to 2000  $\mu$ M), see Figure 11b.

The selected measurement points for CH<sub>4</sub> and CO<sub>2</sub> has high correlation coefficients ( $R^2$ :0.90-0.97), see Table 5 and 6 in Appendices.



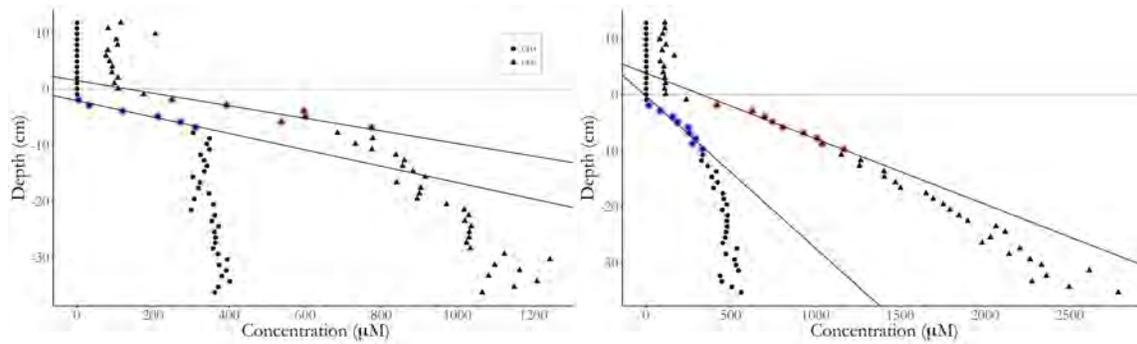
(a) CH<sub>4</sub> concentration for VS170814.

(b) CO<sub>2</sub> concentration for VS170814.



(c) CH<sub>4</sub> and CO<sub>2</sub> concentration for VS170814.

Figure 13: CH<sub>4</sub> and CO<sub>2</sub> concentrations for VS170814. No values for VSI170814 (I).



(a) CH<sub>4</sub> and CO<sub>2</sub> concentration for VSIII170814.

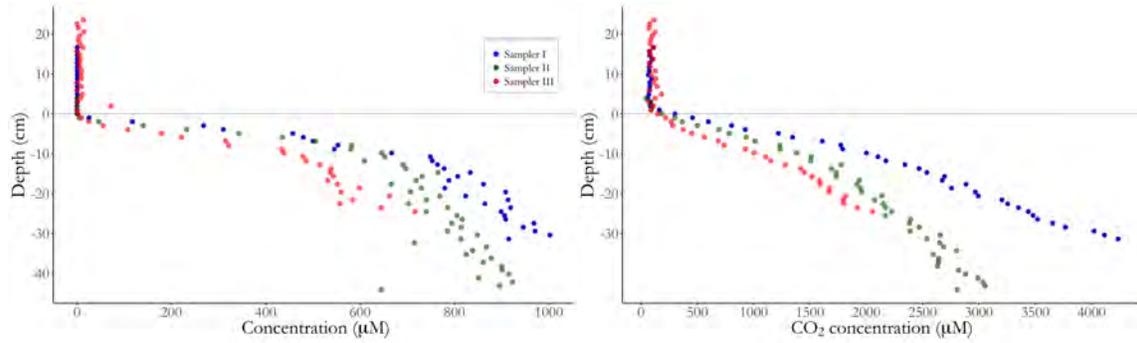
(b) CH<sub>4</sub> and CO<sub>2</sub> concentration for VSIII170814.

Figure 14: VS170814 with linear regression for CH<sub>4</sub> (blue) and CO<sub>2</sub> (red) measurement points which have been used in flux calculations. No values for VSI170814.

The EQ samplers VS170814 (II-III) were relatively spread out in Villasjön, see Figure 1. Sampler III has higher CH<sub>4</sub> and CO<sub>2</sub> concentration than sampler II, see Figure 13. Sampler II had a CH<sub>4</sub> and CO<sub>2</sub> concentration maximum of approximate 400 respectively 1200 µM, while sampler III had

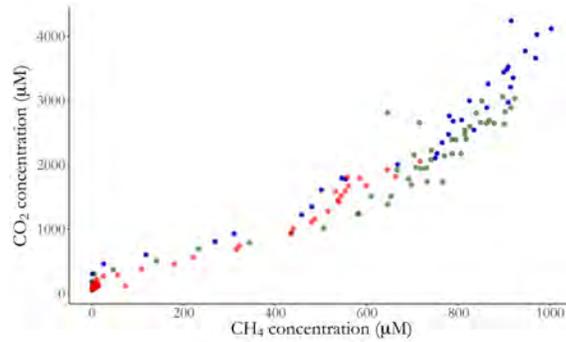
a  $\text{CH}_4$  and  $\text{CO}_2$  concentration maximum of approximate 600 respectively 3000 $\mu\text{M}$ .

The selected measurement points for  $\text{CH}_4$  and  $\text{CO}_2$  has high correlation coefficients ( $R^2$ :0.92-0.97), with sampler II as an exception ( $R^2$ :0.80), see Table 5 and 6 in Appendices.



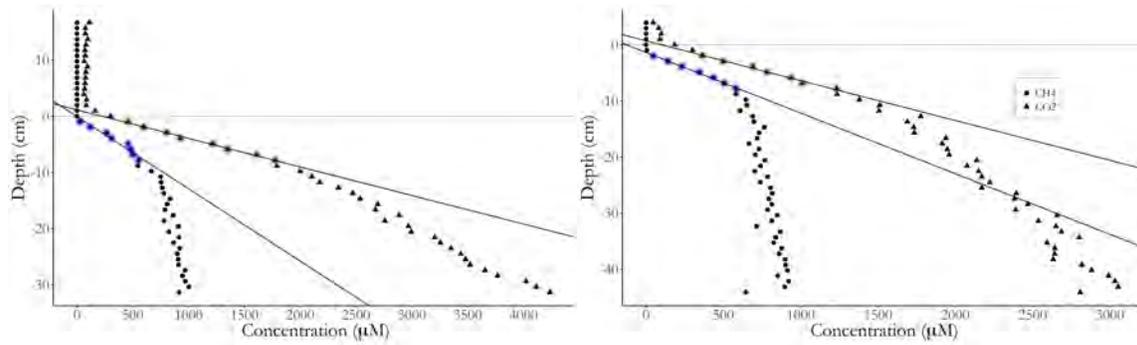
(a)  $\text{CH}_4$  concentration for sampler VS170924.

(b)  $\text{CO}_2$  concentration for sampler VS170924.



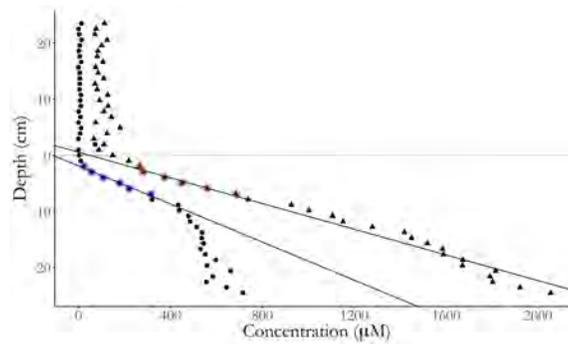
(c)  $\text{CH}_4$  and  $\text{CO}_2$  concentration for sampler VS170924.

Figure 15:  $\text{CH}_4$  and  $\text{CO}_2$  concentrations for sampler VS170924.



(a) CH<sub>4</sub> and CO<sub>2</sub> concentration for VSII170924.

(b) CH<sub>4</sub> and CO<sub>2</sub> concentration for VSII170924.



(c) CH<sub>4</sub> and CO<sub>2</sub> concentration for VSIII170924.

Figure 16: VS170924 with linear regression for CH<sub>4</sub> (blue) and CO<sub>2</sub> (red) measurement points which have been used in flux calculations.

The EQ samplers VS170924 (I-III) were relatively spread out in Villasjön, see Figure 1. Sampler I have slightly higher CH<sub>4</sub> and CO<sub>2</sub> concentration (maximum of approximate 1000 respectively 4500 μM) than sampler II (maximum of approximate 900 respectively 3000 μM), and III (maximum of approximate 700 respectively 2000 μM), see Figure 15b and 15a.

The selected measurement points for CH<sub>4</sub> and CO<sub>2</sub> has high correlation coefficients ( $R^2$ :0.94-1.00), see Table 5 and 6 in Appendices.

### 3.2.4 Inre Harsjön

Three EQ samplers were placed in Inre Harsjön (1 deployment) during 2016, and three EQ samplers were placed in Inre Harsjön (1 deployment) during 2017. One EQ sampler (IHI170731) were excluded in 2017. The diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes with flux error and CO<sub>2</sub>:CH<sub>4</sub> ratios can be seen in Table 10.

Table 10: Diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes and CO<sub>2</sub>:CH<sub>4</sub> ratios from equilibration samplers

Date	ID	CH <sub>4</sub> flux and error [mg m <sup>-2</sup> d <sup>-1</sup> ]	CO <sub>2</sub> flux and error [mg m <sup>-2</sup> d <sup>-1</sup> ]	CO <sub>2</sub> :CH <sub>4</sub> [μM]	R <sup>2</sup>	<i>p</i>
August 2016	IHI160819	2.57 ± 0.08	5.33 ± 1.20	1.58:1.00	0.71	<0.01
	IHII160819	7.25 ± 0.32	3.82 ± 11.72	2.06:1.00	0.61	<0.01
	IHIII160819	4.24 ± 0.29	4.45 ± 0.87	0.91:1.00	0.83	<0.01
	Mean ± SEM	4.69 ± 1.68	4.53 ± 0.44			
July 2017	IHII170731	14.41 ± 1.82	-5.95 ± -2.13	0.91:1.00	0.29	<0.01
	IHIII170731	5.50 ± 0.50	9.52 ± 4.66	1.78:1.00	0.76	<0.01
	Mean ± SEM	9.96 ± 4.45	1.78 ± 7.74			

Mean and SEM only calculated for fluxes, not flux errors.

The diffusive sedimentary CH<sub>4</sub> fluxes in Inre Harrsjön varied from 2.57 to 14.41 mg m<sup>-2</sup> d<sup>-1</sup>. The diffusive sedimentary CO<sub>2</sub> fluxes in Inre Harrsjön varied from 3.82 to 9.52 mg m<sup>-2</sup> d<sup>-1</sup> (without extrema IHII170731). Out of the two deployments the EQ IH170731 had the highest diffusive sedimentary CH<sub>4</sub> flux mean while IH160819 had the lowest. IH160819 had the highest diffusive sedimentary CO<sub>2</sub> flux while IH170731 had the lowest.

The analyzed EQ samplers were all placed at different locations in Inre Harrsjön, see Figure 1 and Table 2. IH160819 (I-III) was placed on a depth gradient, while IH170731 (II-III) was placed on a line on the west side of the lake, by the shore. The highest diffusive sedimentary CH<sub>4</sub> flux was measured at a depth of 1.5 m, with EQ sampler IHIII170731, close to the shore, at location 7. The lowest diffusive sedimentary CH<sub>4</sub> flux was measured at a depth of 1 m, with EQ sampler IHI160819, close to the shore as well, at location 9. The highest diffusive sedimentary CO<sub>2</sub> flux was measured at a depth of 1.5 m, with EQ sampler IHIII170731, close to the shore, at location 8. The lowest diffusive sedimentary CH<sub>4</sub> flux was measured at a depth of 2.5 m, with EQ sampler IHI160819, which were deployed between the shore and the deepest point, at location 10.

The majority of estimated ratios of CO<sub>2</sub>:CH<sub>4</sub> exceeds the 1:1 ratio. The two EQ samplers, IHIII160819 and IHII170731, measured ratios of 0.91:1.00. Notable is that EQ sampler IHIII170731 has a very low correlation coefficient. The CO<sub>2</sub> and CH<sub>4</sub> concentrations measured for each deployment can be seen in Figures 17c and 19c.

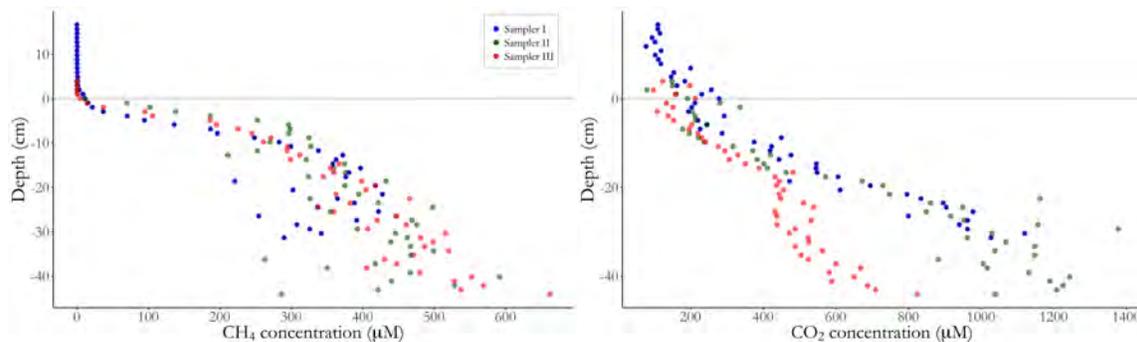
Table 11: Mean and standard error of mean for environmental parameters and diffusive fluxes

ID	CH <sub>4</sub> flux [mg m <sup>-2</sup> d <sup>-1</sup> ]	CO <sub>2</sub> flux [mg m <sup>-2</sup> d <sup>-1</sup> ]	Atm. pressure [hPa]	Lake temp. <sup>1</sup> [°C]	Air temp. [°C]	Wind speed [m s <sup>-2</sup> ]	<sup>2</sup> Depth [cm]
IHI160819	4.69 ± 1.68	4.53 ± 0.44	973.40 ± 0.37	11.29 ± 0.01	9.94 ± 0.18	4.11 ± 0.11	100
IHII160819	4.69 ± 1.68	4.53 ± 0.44	973.40 ± 0.37	12.29 ± 0.02	9.94 ± 0.18	4.11 ± 0.11	250
IHIII160819	4.69 ± 1.68	4.53 ± 0.44	973.40 ± 0.37	12.52 ± 0.01	9.94 ± 0.18	4.11 ± 0.11	400
IH170731	9.96 ± 4.45	1.78 ± 7.74	971.33 ± 0.32	14.51 ± 0.04	13.15 ± 0.18	3.94 ± 0.08	150

<sup>1</sup>Lake temperature measured at deployment depth. <sup>2</sup>Depth: deployment depth. Deployment IH160819 was placed at different depths, hence different averaged lake temperatures. CH<sub>4</sub> and CO<sub>2</sub> fluxes present mean and SEM for the EQ samplers IH160819 (I-III), and not the fluxes for the individual EQ samplers.

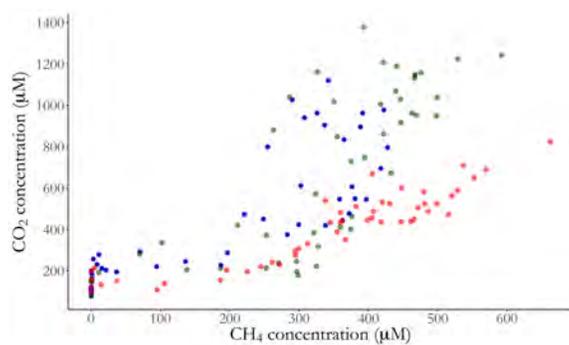
The environmental parameters measured during the deployment period for each EQ sampler in Inre Harrsjön are seen in Table 11, with mean and SEM. Multivariate analysis and linear regressions were performed for the diffusive sedimentary CH<sub>4</sub> fluxes and the environmental parameters, in-

cluding depth. No significant linear relationships were shown ( $R^2 < 0.40$ ,  $p > 0.05$ ) for the analyzed parameters. The same result occurred for diffusive sedimentary  $\text{CO}_2$  fluxes, but with even lower insignificant correlations ( $R^2 < 0.06$ ,  $p > 0.05$ ). Significant correlations for diffusive sedimentary  $\text{CO}_2$  fluxes was presented when extrema were removed (IH1170731), but due to a degree of freedom of only 2, no further conclusions could be made based on so few measurements.



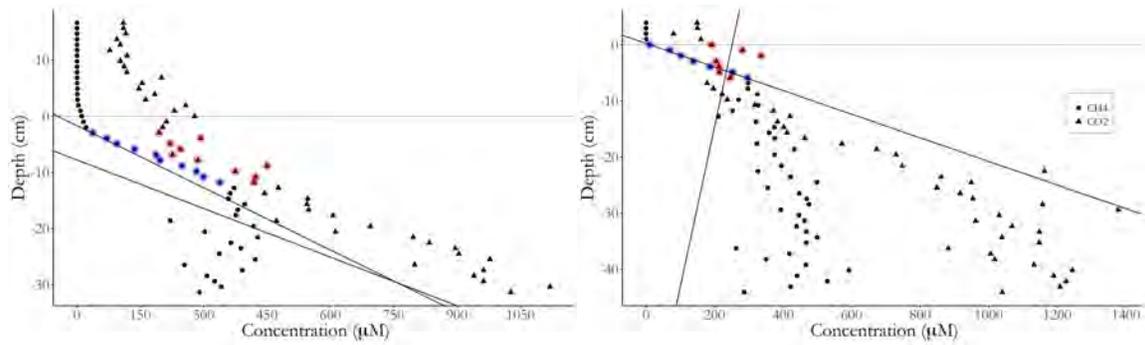
(a)  $\text{CH}_4$  concentration for IH160819.

(b)  $\text{CO}_2$  concentration for IH160819.



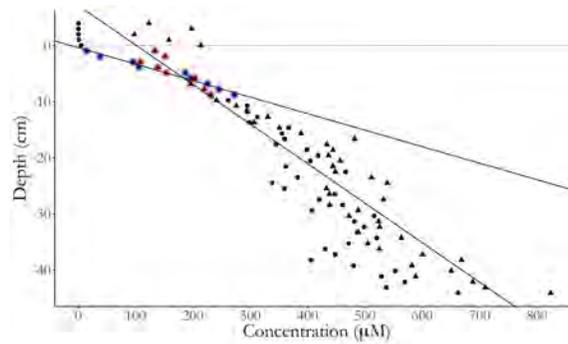
(c)  $\text{CH}_4$  and  $\text{CO}_2$  concentration for IH160819.

Figure 17:  $\text{CH}_4$  and  $\text{CO}_2$  concentrations for IH160801.



(a) CH<sub>4</sub> and CO<sub>2</sub> concentration for IH160819.

(b) CH<sub>4</sub> and CO<sub>2</sub> concentration for IHIII160819.

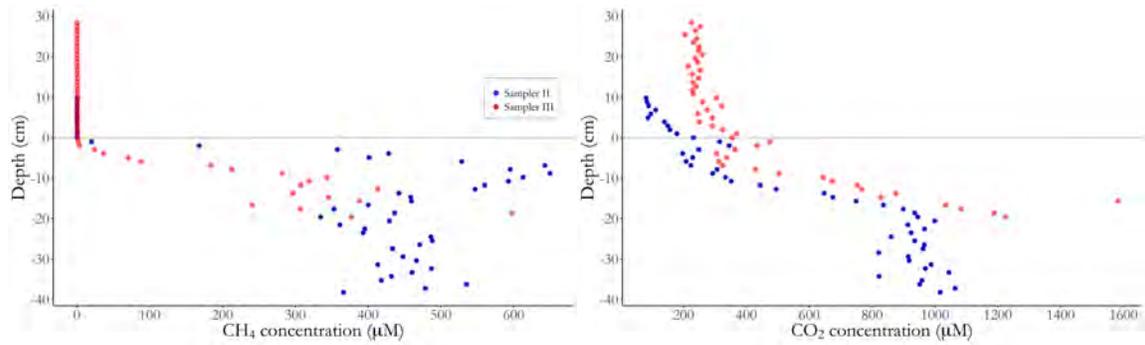


(c) CH<sub>4</sub> and CO<sub>2</sub> concentration for IHIII160819.

Figure 18: IH160819 with linear regression for CH<sub>4</sub> (blue) and CO<sub>2</sub> (red) measurement points which have been used in flux calculations.

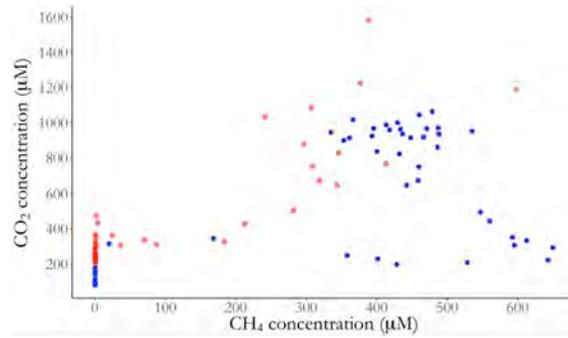
The EQ samplers IH160801 (I-III) were placed on a depth gradient in Inre Harsjön, see Figure 1. Notable is the lower CO<sub>2</sub> concentration for sampler III with a difference of approximate 300 to 600  $\Delta\mu\text{M}$  compared to sampler I and II, see Figure 17b. Similar circumstances occur for sampler I, with lower CH<sub>4</sub> concentrations of approximate 200 to 300  $\Delta\mu\text{M}$  compared to sampler II and III, see Figure 17a. Sampler I portray as well an s-shape in the CH<sub>4</sub> concentration profile has, see Figure 18a. The negative correlation for the CO<sub>2</sub> concentration for sampler IHIII160819 should also be noted, see Figure 18b.

The selected measurement points for CH<sub>4</sub> has high correlation coefficients ( $R^2$ :0.97-0.99), while the measurement points for CO<sub>2</sub> has lower correlation coefficients ( $R^2$ :0.02-0.79), see Table 7 and 8 in Appendices.



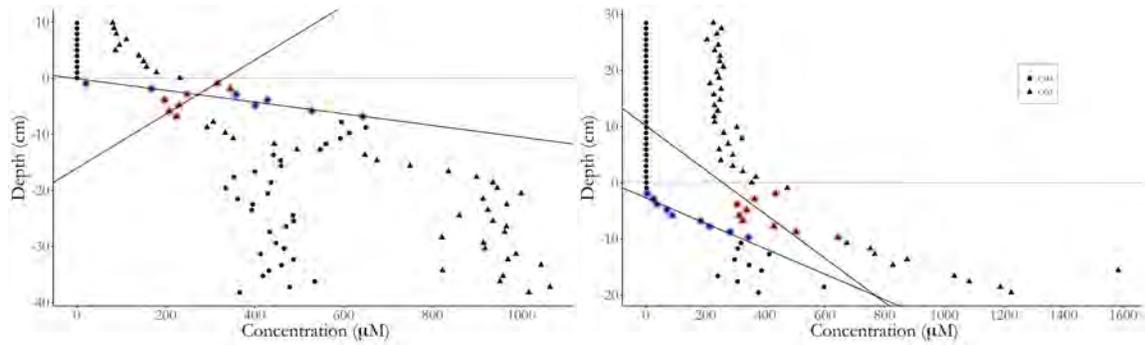
(a) CH<sub>4</sub> concentration for IH170731.

(b) CO<sub>2</sub> concentration for IH170731.



(c) CH<sub>4</sub> and CO<sub>2</sub> concentration for IH170731.

Figure 19: CH<sub>4</sub> and CO<sub>2</sub> concentrations for IH170731. No values for IHI170731 (I).



(a) CH<sub>4</sub> and CO<sub>2</sub> concentration IHIII170731.

(b) CH<sub>4</sub> and CO<sub>2</sub> concentration for IHIII170731.

Figure 20: IH170731 with linear regression for CH<sub>4</sub> (blue) and CO<sub>2</sub> (red) measurement points which have been used in flux calculations. No values for IHI170731.

The EQ samplers IH170731 (II-III) were placed close to the west shore in Inre Harsjön, see Figure 1. Sampler II has a CH<sub>4</sub> and CO<sub>2</sub> concentration maximum of approximate 700 respectively 1100µM, while sampler III has a maximum of approximate 600 respectively 1200µM, see Figure 19a. Sampler II portrays as well an s-shape for the CH<sub>4</sub> concentration, seen in Figure 20a.

The selected measurement points for CH<sub>4</sub> has high correlation coefficients (R<sup>2</sup>:0.93-0.95) while the measurement points for CO<sub>2</sub> has much lower correlation coefficients (R<sup>2</sup>:0.37-0.61), see Table 7 and 8 in Appendices.

### 3.2.5 Mellersta Harrsjön

No EQ samplers were placed in Mellersta Harrsjön during 2016, and three EQ samplers were placed in Mellersta Harrsjön (1 deployment) during 2017. The diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes with flux error and CO<sub>2</sub>:CH<sub>4</sub> ratios can be seen in Table 12.

Table 12: Diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes and CO<sub>2</sub>:CH<sub>4</sub> ratios from equilibration samplers

Date	ID	CH <sub>4</sub> flux and error [mg m <sup>-2</sup> d <sup>-1</sup> ]	CO <sub>2</sub> flux and error [mg m <sup>-2</sup> d <sup>-1</sup> ]	CO <sub>2</sub> :CH <sub>4</sub> [μM]	R <sup>2</sup>	<i>p</i>
July 2017	MHI170716	7.26 ± 0.57	37.60 ± 5.06	2.83:1.00	0.94	<0.01
	MHII170716	4.03 ± 0.51	6.86 ± 6.68	1.73:1.00	0.69	<0.01
	MHIII170716	14.34 ± 3.88	41.21 ± 5.43	2.11:1.00	0.85	<0.01
	Mean ± SEM	8.55 ± 3.04	28.56 ± 10.90			

*Mean and SEM only calculated for fluxes, not flux errors.*

The sedimentary diffusive CH<sub>4</sub> fluxes in Mellersta Harrsjön varied from 4.03 to 14.34 mg m<sup>-2</sup> d<sup>-1</sup>. The sedimentary diffusive CO<sub>2</sub> fluxes varied from 6.86 to 41.21 mg m<sup>-2</sup> d<sup>-1</sup>. The mean sedimentary diffusive CH<sub>4</sub> flux was 8.55 while the mean sedimentary diffusive CO<sub>2</sub> flux was 28.56 mg m<sup>-2</sup> d<sup>-1</sup>.

The analyzed EQ samplers MH170716 (I-III) were all placed at different locations, at a depth of 1.5 m, in Mellersta Harrsjön, see Figure 1 and Table 2. The highest diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes were measured with the EQ sampler MHIII170716, at the middle part of the lake, location 13. The lowest diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes were measured with the EQ sampler MHII170716, at the most northern part of the sampling locations, location 12.

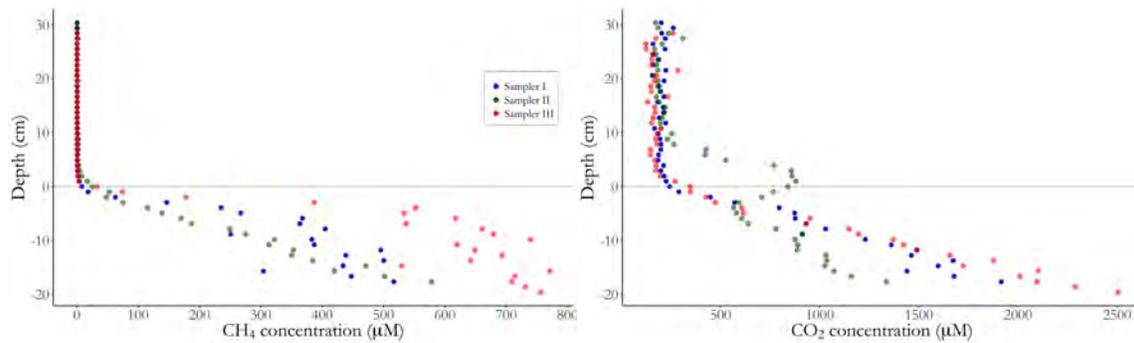
All estimated ratios of CO<sub>2</sub>:CH<sub>4</sub> exceeds the 1:1 ratio. The CO<sub>2</sub> and CH<sub>4</sub> concentrations measured for each deployment can be seen in Figures 21c.

Table 13: Mean and standard error of mean for environmental parameters and diffusive fluxes

ID	CH <sub>4</sub> flux [mg m <sup>-2</sup> d <sup>-1</sup> ]	CO <sub>2</sub> flux [mg m <sup>-2</sup> d <sup>-1</sup> ]	Atm. pressure [hPa]	Lake temp. <sup>1</sup> [°C]	Air temp. [°C]	Wind speed [m s <sup>-2</sup> ]	Depth <sup>2</sup> [cm]
MH170716	8.55 ± 3.04	28.56 ± 10.90	966.59 ± 0.15	11.01 ± 0.02	9.63 ± 0.14	4.47 ± 0.10	150

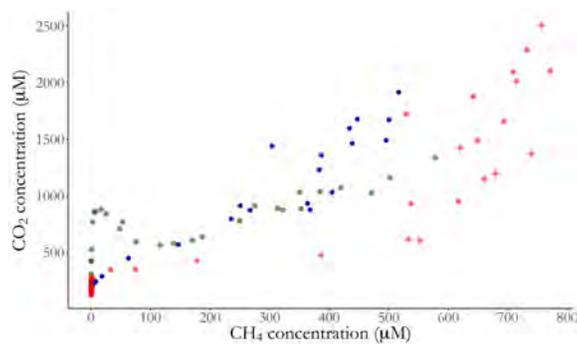
<sup>1</sup>Lake temperature measured at deployment depth. <sup>2</sup>Depth: deployment depth.

The environmental parameters measured during the deployment period for each EQ sampler in Mellersta Harrsjön are seen in Table 13, with mean and SEM. Due to the low number of measurements, no multivariate analysis or linear regressions was performed for the diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes and the environmental parameters, including depth.



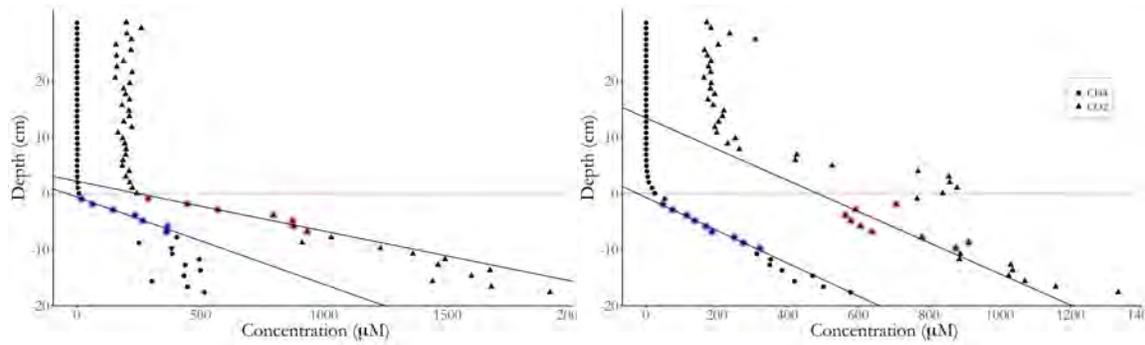
(a) CH<sub>4</sub> concentration for MH170716.

(b) CO<sub>2</sub> concentration for MH170716.



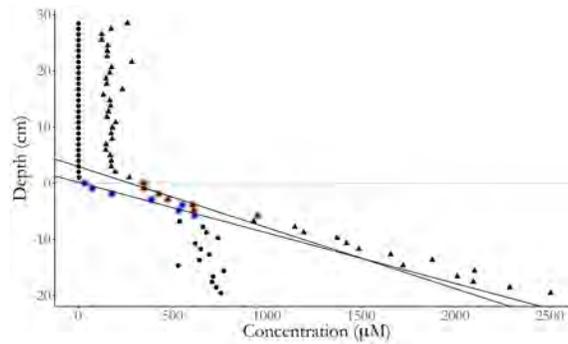
(c) CH<sub>4</sub> and CO<sub>2</sub> concentration for MH170716.

Figure 21: CH<sub>4</sub> and CO<sub>2</sub> concentrations for MH170716.



(a) CH<sub>4</sub> and CO<sub>2</sub> concentration for MHI170716.

(b) CH<sub>4</sub> and CO<sub>2</sub> concentration for MHIII170716.



(c) CH<sub>4</sub> and CO<sub>2</sub> concentration for MHIII170716.

Figure 22: MH170716 with linear regression for CH<sub>4</sub> (blue) and CO<sub>2</sub> (red) measurement points which have been used in flux calculations.

The EQ samplers MH170716 (I-III) were placed from the middle to the most southern part of Mellersta Harrsjön, see Figure 1. The CH<sub>4</sub> concentration for sampler III is slightly higher (maximum of approximate 800 $\mu$ M), compared to sampler I and II (maximum of approximate 500 to 600 $\mu$ M), see Figure 21a. The CO<sub>2</sub> concentration for sampler II has an s-shape and is slightly lower (maximum of approximate 1500 $\mu$ M), compared to sampler I and III (maximum of approximate 2000 respectively 2500 $\mu$ M), see Figure 21b.

The selected measurement points for CH<sub>4</sub> has high correlation coefficients ( $R^2$ :0.94-0.99). The CO<sub>2</sub> measurement points for the samplers MHI170716 and MHIII170716 has high correlation coefficients ( $R^2$ :0.84-0.92), while MHII170716 has a lower correlation coefficient ( $R^2$ : 0.54), see Table 9 and 10 in Appendices.

### 3.3 Lake chambers

#### 3.3.1 Overall water surface diffusive CH<sub>4</sub> fluxes

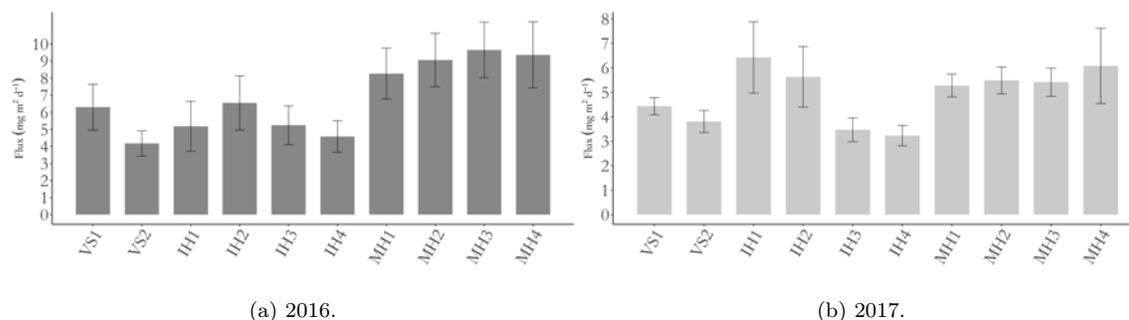


Figure 23: Diffusive water surface CH<sub>4</sub> fluxes measured with the lake chambers. Annual mean calculated for each lake chamber. Error bars represent the standard error of the mean.

The diffusive water surface CH<sub>4</sub> fluxes were calculated for 10 sampling locations for the lake chambers in the study lakes, see placements in Figure 1. The fluxes were averaged for each sampling location and year, see Figure 23. In 2016 the diffusive water surface CH<sub>4</sub> fluxes ranged from 4.17 to 9.65 mg m<sup>-2</sup> d<sup>-1</sup> (n:284) and in 2017 the diffusive water surface CH<sub>4</sub> fluxes ranged from 3.23 to 6.45 mg m<sup>-2</sup> d<sup>-1</sup> (n:158). All sampling locations had higher fluxes in 2016 (exception IH1) than in 2017.

In Villasjön had the sampling location VS1 higher fluxes in both 2016 and 2017, in comparison to VS2. VS1 is located north-west of VS2 at a depth of 0-1 m. In Inre Harrsjön had the sampling location IH2 the highest flux in 2016, while sampling location IH1 had the highest flux in 2017. Both are located in the south-east part of the lake at a depth of 2-3 m (IH2) and 1-2 m (IH1). The lowest flux both in 2016 and 2017 had the sampling location IH4, at the north-west part of the lake at a depth of 1-2 m. In Mellersta Harrsjön had the sampling location MH3 the highest flux in 2016, while sampling location MH4 had the highest flux in 2017. MH3 are located close to a stream inlet, south of the middle part of the lake, at a depth of 3-4 m. MH4 are located furthest south in the lake, at a depth of 1-2 m. The lowest flux, both in 2016 and 2017, had the sampling location MH1, at the north-west part of the lake at a depth of 1-2 m.

Table 14: Diffusive water surface CH<sub>4</sub> fluxes divided by lake, year and month, measured with the lake chambers

Diffusive CH <sub>4</sub> flux [mg m <sup>-2</sup> d <sup>-1</sup> ]						
Month	2016			2017		
	Villasjön Flux ± SEM	Inre Harrsjön Flux ± SEM	Mellersta Harrsjön Flux ± SEM	Villasjön Flux ± SEM	Inre Harrsjön Flux ± SEM	Mellersta Harrsjön Flux ± SEM
June	4.36 ± 1.15	4.75 ± 0.50	14.01 ± 1.23	3.99 ± 0.38	10.19 ± 1.97	9.60 ± 1.68
July	8.12 ± 2.32	9.83 ± 2.06	13.68 ± 2.27	5.06 ± 0.58	4.66 ± 0.62	6.25 ± 0.51
August	7.01 ± 0.98	6.91 ± 1.02	11.30 ± 1.20	4.79 ± 0.52	4.21 ± 0.46	4.32 ± 0.25
September	2.76 ± 0.53	2.49 ± 0.38	4.29 ± 0.58	2.56 ± 0.17	1.62 ± 0.10	3.40 ± 0.24
October	0.77 ± 0.31	0.90 ± 0.25	1.94 ± 0.29			
Mean ± SEM	4.66 ± 0.79	4.98 ± 0.65	9.04 ± 0.84	4.10 ± 0.28	5.17 ± 0.52	5.89 ± 0.43

Diffusive water surface CH<sub>4</sub> fluxes have been averaged monthly and divided by year and lake, see Table 14. The monthly fluxes were all higher in 2016 than in 2017 (exception Inre Harrsjön in June and Mellersta Harrsjön in September). The highest flux was observed in June 2016 in Mellersta Harrsjön while the lowest flux was observed in October 2016 in Villasjön (no values for October 2017 was obtained).

In 2016 had Villasjön the highest fluxes in July, and thereafter August and June. The same pattern was shown for 2017. Inre Harrsjön had the same pattern as Villasjön for 2016, but in 2017 were the highest fluxes observed in June, and thereafter July and August. In 2016 had Mellersta Harrsjön the highest fluxes in June, and thereafter July and August, with the same pattern in 2017.

A paired t-test was performed for Inre Harrsjön and Mellersta Harrsjön, with the purpose of comparing if significant differences occurred during the months July, August and September in 2016 compared to 2017 (for additional information see Table 4 in Appendices). In Inre Harrsjön had August and September significantly similar monthly means during 2016 and 2017, while July presented a significant difference. Mellersta Harrsjön presented a dissimilar pattern, with all months showing a significant difference in the monthly means during the two study years, see Table 14.

Table 15: Annual diffusive CH<sub>4</sub> fluxes for all study lakes

Diffusive CH <sub>4</sub> flux [mg m <sup>-2</sup> d <sup>-1</sup> ]				
Year	June	July	August	September
2016	8.38 ± 1.62	10.89 ± 1.11	8.74 ± 0.73	3.23 ± 0.34
2017	8.66 ± 1.09	5.38 ± 0.35	4.36 ± 0.23	2.52 ± 0.17
Total	8.59 ± 0.90	7.95 ± 0.63	6.48 ± 0.43	2.85 ± 0.18

The three study lakes presented overall higher fluxes during 2016 than 2017, see Table 15. August and July showed notable higher fluxes in 2016 than 2017.

Environmental parameters have been averaged monthly and divided by year, seen in Tables 4 and 5. Linear regressions of the diffusive water surface CH<sub>4</sub> fluxes have been performed with the environmental parameters, exclusive depth. Significant, but low correlations coefficients, were found between diffusive water surface CH<sub>4</sub> fluxes and lake temperature (R<sup>2</sup>:0.37, p:<0.01), atmospheric pressure (R<sup>2</sup>:0.29, p:<0.01) and air temperature (R<sup>2</sup>:0.36, p:<0.01) for the three study lakes. No

significant correlation coefficients were presented for wind speed. In Villasjön there was a significant correlation between the diffusive water surface CH<sub>4</sub> fluxes and lake temperature (R<sup>2</sup>:0.79, p:<0.01) as well as air temperature (R<sup>2</sup>:0.77, p:<0.01). A low significant correlation coefficient existed for atmospheric pressure (R<sup>2</sup>:0.51, p:0.03). No significant linear relationship for wind speed was presented. In Inre Harrsjön there was a significant and relatively high correlation coefficient between the diffusive water surface CH<sub>4</sub> fluxes and lake temperature (R<sup>2</sup>:0.66, p:0.00). For air temperature and atmospheric pressure, there were a significant but low correlation coefficient (R<sup>2</sup><0.50, p:<0.04). No significant linear relationship existed for wind speed. In Mellersta Harrsjön there was no significant correlation for any of the environmental parameters (R<sup>2</sup>:<0.40, p:>0.06).

For all linear regressions were the diffusive water surface CH<sub>4</sub> fluxes seen to increase during higher air and lake temperatures, and lower atmospheric pressure.

### 3.4 Comparison of equilibration sampler and lake chamber fluxes

Due to few measurements with the EQ samplers, temporal and spatial differences, only a robust analysis of the diffusive fluxes can be performed on an annual time scale for the three study lakes, see Table 16.

Both Villasjön and Inre Harrsjön presents the highest diffusive sedimentary CH<sub>4</sub> fluxes in 2017 compared to 2016. A comparison of diffusive sedimentary CO<sub>2</sub> fluxes can only be performed in Villasjön which presents higher fluxes in 2016 compared to 2017. Diffusive water surface CH<sub>4</sub> fluxes are higher in 2016 compared to 2017 for the lakes Villasjön and Mellersta Harrsjön, while Inre Harrsjön presents the opposite pattern.

Table 16: Measured interlake diffusive CH<sub>4</sub> and CO<sub>2</sub> fluxes, divided by year and method

Villasjön								
Year	Method	Measuring period	CH <sub>4</sub> range [mg m <sup>-2</sup> d <sup>-1</sup> ]	Mean ± SEM	n	CO <sub>2</sub> range [mg m <sup>-2</sup> d <sup>-1</sup> ]	Mean ± SEM	n
2016	Equilibration samplers	August	4.78 - 7.21	6.11 ± 0.71	3	54.42 - 56.92	55.67 ± 1.25	2
	Lake Chambers	June to October	0.46 - 20.18	4.66 ± 0.79	29			
2017	Equilibration samplers	June, August, September	3.53 - 8.80	6.18 ± 0.66	8	21.40 - 61.64	38.24 ± 5.36	8
	Lake Chambers	June to September	2.07 - 8.33	4.10 ± 0.28	31			
Inre Harrsjön								
2016	Equilibration samplers	August	2.57 - 7.25	4.68 ± 1.36	3	3.82 - 5.35	4.53 ± 0.43	3
	Lake Chambers	June to October	0.84 - 22.26	4.98 ± 0.65	57			
2017	Equilibration samplers	July	5.50 - 14.31	9.95 ± 4.45	2			1*
	Lake Chambers	June to September	0.95 - 21.89	5.17 ± 0.52	63			
Mellersta Harrsjön								
2016	Equilibration samplers							
	Lake Chambers	June to October	0.44 - 26.57	9.04 ± 0.84	56			
2017	Equilibration samplers	July	4.03 - 14.34	8.55 ± 3.04	3	6.86 - 41.21	28.56 ± 10.90	3
	Lake Chambers	June to September	1.78 - 27.11	5.89 ± 0.43	64			

\*Without extrema (IHIII170731), hence diffusive sedimentary CO<sub>2</sub> flux was only measured with one sampler (IHIII170731: 9.52 mg m<sup>-2</sup> d<sup>-1</sup>). No measurement conducted with equilibration samplers in Mellersta Harrsjön during 2016.

A comparison of the diffusive sedimentary CH<sub>4</sub> and the water surface CH<sub>4</sub> fluxes have been performed, see Tables 17, 18 and 19. Due to few measurements, sampling at different locations and dates, a comparison of the means and SDs was performed to evaluate the differences in means from the two fluxes. The comparison has been performed by averaging the EQ fluxes measured during one deployment, as an indicator of the diffusive sedimentary CH<sub>4</sub> flux in the chosen study lake during the month of deployment. Lake chamber fluxes, which coincide with the month and lake of

the EQ deployment, have been averaged in order to compare the lake chamber fluxes with the EQ fluxes. Additional information about the comparison can be seen in Table 3 in Appendices.

Out of 7 comparisons, seen in Tables 17, 18 and 19, the water surface diffusive fluxes were higher in 2 out of 7 comparisons. The SDs were in general relatively high for all measurements, portraying a large variation in the diffusive CH<sub>4</sub> fluxes.

Table 17: Comparison of CH<sub>4</sub> fluxes from equilibration samplers and lake chambers

Diffusive CH <sub>4</sub> flux [mg m <sup>-2</sup> d <sup>-1</sup> ]					
Villasjön					
Date	Sampler	ID	n	Mean	SD
July 2016	Equilibration sampler	VS160801	3	6.11	1.23
	Lake chamber		7	8.12	6.13
June 2017	Equilibration sampler	VSI170629	3	4.80	1.28
	Lake chamber		6	3.99	0.92
August 2017	Equilibration sampler	VS170814	2	6.10	1.41
	Lake chamber		9	4.79	0.55
September 2017	Equilibration sampler	VS170924	3	7.64	1.89
	Lake chamber		8	2.56	0.49

In Villasjön were the diffusive sedimentary CH<sub>4</sub> fluxes higher than the diffusive water surface CH<sub>4</sub> fluxes in all comparisons except in July 2016, see Table 17. Notable is the high SD for the lake chamber measurements in July 2016 which portrays a large variation in the measured diffusive CH<sub>4</sub> flux. The highest diffusive CH<sub>4</sub> flux was measured with the lake chambers in July 2016, and the lowest flux was measured with the lake chambers in September 2017.

Table 18: Comparison of CH<sub>4</sub> fluxes from equilibration samplers and lake chambers

Diffusive CH <sub>4</sub> flux [mg m <sup>-2</sup> d <sup>-1</sup> ]					
Inre Harrsjön					
Date	Sampler	ID	n	Mean	SD
July 2016	Equilibration sampler	IH160819	3	4.69	2.37
	Lake chamber		18	6.91	4.32
July 2017	Equilibration sampler	IH170731	2	9.96	6.30
	Lake chamber		16	4.66	2.48

In Inre Harrsjön were the diffusive sedimentary CH<sub>4</sub> fluxes higher than the water surface CH<sub>4</sub> in 1 out of 2 comparisons, see Table 17. The highest diffusive CH<sub>4</sub> flux was measured with the EQ samplers in July 2017. There is though large variations in the diffusive CH<sub>4</sub> flux measured with the EQ samplers in July 2017, noted by the large SD. The lowest diffusive CH<sub>4</sub> flux was measured with the lake chambers in July 2017.

Table 19: Comparison of CH<sub>4</sub> fluxes from equilibration samplers and lake chambers

Diffusive CH <sub>4</sub> flux [mg m <sup>-2</sup> d <sup>-1</sup> ]					
Mellersta Harrsjön					
Date	Sampler	ID	n	Mean	SD
July 2017	Equilibration sampler	MH170716	3	8.55	5.27
	Lake chamber		16	6.25	2.06

In Mellersta Harrsjön was the highest diffusive CH<sub>4</sub> flux measured with the EQ samplers in July 2017, see Table 19. Large variations exist for the diffusive CH<sub>4</sub> flux measured with the EQ samplers, noted by the large SD. The lowest diffusive CH<sub>4</sub> flux was measured with the lake chambers in July 2017.

## 4 Discussion

### 4.1 Environmental parameters

No significant correlation was presented for the environmental parameters and the diffusive sedimentary CH<sub>4</sub> fluxes for the three study lakes. A large factor is the limited data set with great temporal and spatial variability. A similar pattern was seen for the environmental parameters and the diffusive sedimentary CO<sub>2</sub> fluxes, with the exceptions of low significant linear relationships for depth and wind speed. The linear relationship between diffusive sedimentary CO<sub>2</sub> fluxes and wind speed as well as depth, could potentially be related to the mobilization of labile organic carbon with increasing turbulence during higher wind speeds, which might be more accessible at shallower lake depths. The correlation coefficients were though relatively low ( $R^2$ :0.25-0.33), hence no general conclusions can be made, and causation between the parameters cannot be stated.

In Inre Harrsjön were measurements taken at a depth gradient but no significant correlation was presented in relation to diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes. In previous studies has the diffusive CH<sub>4</sub> fluxes been seen to increase at shallower depths (Bastviken et al., 2004), but no linear relationship was shown in Inre Harrsjön. Likely due to too few measurements, with only one measurement per depth gradient during 2016.

In conclusion, more measurements are needed for further analysis of how the environmental parameters control the variability of the diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes in the lakes Villasjön, Inre Harrsjön and Mellersta Harrsjön.

Some significant correlations were presented for the environmental parameters and the diffusive water surface CH<sub>4</sub> fluxes. Low but significant correlations were found for lake temperature, air temperature and atmospheric pressure.

In Villasjön there was relatively high significant correlation coefficients for lake temperature and air temperature. Hence, the diffusive water surface CH<sub>4</sub> fluxes increases with air and lake temperatures, which is in line with previous studies related to increasing CH<sub>4</sub> production during warmer lake temperatures (Blake et al., 2015; Nozhevnikova et al., 1997; Yvon-Durocher et al., 2014). A lower significant correlation was found for atmospheric pressure. The diffusive water surface CH<sub>4</sub> increases with lower atmospheric pressure, and that is the same trend which has been presented for ebullition of CH<sub>4</sub> (Martens & Val Klump, 1980; Bastviken et al., 2004). During lower atmospheric pressure the hydrostatic pressure is overcome and ebullition of CH<sub>4</sub> increases. Further research is needed on the controlling mechanisms for increasing diffusive water surface CH<sub>4</sub> fluxes during decreasing atmospheric pressure, and if a causation is present. Same significant patterns presented for Villasjön exist for Inre Harrsjön, with increasing diffusive water surface CH<sub>4</sub> fluxes during warmer air and water temperatures, and increasing fluxes with decreasing atmospheric pressure. In Mellersta Harrsjön there were no significant correlations shown between the environmental parameters and the diffusive water surface CH<sub>4</sub> fluxes, and none of the lakes presented any linear relationship for wind speed.

Increasing wind speed induces the turbulence by the water surface, hence studies have shown larger diffusive water surface CH<sub>4</sub> fluxes during higher wind speeds (Walter Anthony & Macintyre, 2016; Schilder et al., 2013). A correlation between wind speed and diffusive water surface CH<sub>4</sub> was expected, but no correlation was presented with the measurements from the lake chambers. Due to the monthly averaging of wind speed and diffusive water surface CH<sub>4</sub> fluxes, the rapid fluctuations of wind speed and the potential impact on the diffusive water surface CH<sub>4</sub> fluxes might have been dismissed during the analysis.

Diffusive water surface  $\text{CH}_4$  fluxes have been seen to increase during cooling conditions (such as during e.g. the night) due to the formation of small eddies by the water surface-air interface (Poindexter et al., 2016). As with wind speed, the monthly averaging may dismiss diurnal patterns, such as the impact of hydrodynamic transports, hence only a significant correlation is seen for increasing diffusive water surface  $\text{CH}_4$  fluxes during warmer air temperatures.

A lot of environmental parameters affect the amount of dissolved  $\text{CH}_4$  and  $\text{CO}_2$ . Due to stratification in lakes after rainfall, larger quantities of  $\text{CH}_4$  have been measured during these temporal periods (Bartosiewicz et al., 2015). Stordalen is though located in an area with a rainshadow (Callaghan et al., 2010), and less affected by the impact of rainfalls. Other important factors that govern the diffusive fluxes are the lake sediment and substrate availability as well as quality (Ström et al., 2012), which there is limited information about in regards to the three study lakes. Hence, more information about these spatial circumstances is needed for further analysis.

## 4.2 Comparison of equilibration samplers and water samples

A comparison of the  $\text{CH}_4$  concentration was performed for the EQ samplers and the water samples, see Table 6. A complete equilibration was presented for the EQ samplers VSII170629, VSIII170629 and IHIII170731 with a difference in mean of only  $0.02\text{-}0.05\Delta\mu\text{M}$  in comparison to the water samples. Concluding that the EQ samplers have been deployed at sufficient length for the distilled water in the EQ sampler to equilibrate with the outside lake water.

The EQ samplers with a lower mean than the water samples might not have equilibrated properly and could potentially affect the accuracy of the calculation of the diffusive fluxes. The EQ samplers with lower means in comparison to the water samples were VSII170814, VSII170924 (though a small difference), IHIII160819, IHIII160819, IHIII170731 and MHI170716. The  $\text{CO}_2$  and  $\text{CH}_4$  concentrations depth profiles seen in Figures 14a, 16c, 18b, 18c, 20a and 22a, present in general no irregularities. The EQ samplers IHIII160819 and IHIII160819, seen in Figures 18b and 18c, have though some non normal  $\text{CO}_2$  concentration profiles, while EQ sampler IHIII170731, seen in Figure 20a, show a distinct s-shape of the  $\text{CH}_4$  concentration profile, which can be seen as an outlier in comparison to the other EQ samplers. With the exception of the last mentioned EQ sampler, the EQ samplers which have not equilibrated properly have relatively normal distributed concentration profiles. When comparing mean and ranges of  $\text{CH}_4$  concentrations, the EQ sampler VSIII170924 presented a lower  $\text{CH}_4$  concentration than the EQ samplers VSI170924 and VSII170924, seen in Table 5 in Appendices. The opposite pattern was presented for the EQ samplers IHIII160819, IHIII160819 and IHIII170731, which had the highest  $\text{CH}_4$  concentration in comparison the other EQ samplers deployed during the same temporal period in Inre Harsjön, seen in Table 7 in Appendices. The remaining EQ samplers presented no distinct pattern when comparing the  $\text{CH}_4$  concentration with the EQ samplers deployed during the same period. Concluding, an incomplete equilibration will likely affect the  $\text{CH}_4$  concentrations and the calculation of the diffusive sedimentary  $\text{CH}_4$  fluxes, but no systematic pattern was though presented for the EQ samplers which had not been equilibrated properly.

The EQ samplers which presented distinctively higher means than the water samples were VSIII170629 ( $3.33\Delta\mu\text{M}$ ) and VSIII170924 ( $8.54\Delta\mu\text{M}$ ), which both display large variations in the  $\text{CH}_4$  concentrations noted by the high SDs. Both EQ samplers were placed at location 5 in Villasjön, see Figure 1 and Table 2. The EQ sampler VSIII170814 was deployed at the same placement, location 5, and show a similar  $\text{CH}_4$  concentration mean to the water sample with a difference of only  $0.16\Delta\mu\text{M}$ . Hence the EQ sampler present an opposite pattern compared to the two previously

mentioned EQ samplers. Location 5 has a rocker bottom then the other deployment locations in Villasjön, but with the limited number of measurement no pattern can be distinguished in regards of potential spatial impacts on the equilibration.

Most notable is the EQ samplers deployed during the same temporal period which presents EQ samplers with both incomplete and complete equilibrations. This is the case for the deployment VS170629 (I-III), where two EQ samplers have a difference in means of only 0.02-0.05 $\Delta\mu\text{M}$ , while one has a difference in mean of 3.33 $\Delta\mu\text{M}$ . All EQ deployments during the two study seasons, with the exception of VS160801 (I-III), present EQ sampler means which are both higher and lower than the water sample means.

While a lower EQ sampler mean in comparison to the water sample mean, indicate an incomplete equilibration, the opposite pattern have no clear explanation. Same applies for the EQ samplers deployed during the same temporal periods and presents means which are both higher and lower than the water sample means. This could demonstrate that the comparison of the EQ sampler and water sample means are too robust, that there are spatial differences in the study lakes or that the samples have been impacted during analysis.

The comparison of the  $\text{CO}_2$  concentrations for the EQ samplers and the water samples was presented in Table 7. All EQ sampler presented higher  $\text{CO}_2$  concentrations than  $\text{CH}_4$  concentrations, which will be further discussed in section 4.3. Large differences in mean occurred for all EQ samplers with large variability in the  $\text{CO}_2$  concentration presented by high SDs. The difference in mean was large both in 2016 and in 2017. The tapwater used in the EQ samplers could have impacted the  $\text{CO}_2$  concentrations. The tapwater was though boiled and cooled down under a nitrogen atmosphere, and since it was degassed the tapwater should not have impacted the  $\text{CO}_2$  concentrations. There might though have been a risk that the tapwater could have had elevated  $\text{CO}_2$  concentrations. No conclusions about the elevated  $\text{CO}_2$  concentrations measured with the EQ samplers can be made, and further research is needed.

### 4.3 Diffusive fluxes measured with equilibration samplers and lake chambers

The measured diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes in *Villasjön* have been presented in Table 8. The lowest diffusive sedimentary  $\text{CH}_4$  fluxes were measured at the southern part of Villasjön and ranged between 3.53-5.45  $\text{mg m}^{-2} \text{d}^{-1}$ . The highest diffusive sedimentary  $\text{CH}_4$  fluxes were measured in the northern part, close to the shore, and ranged between 6.09-8.66  $\text{mg m}^{-2} \text{d}^{-1}$ . The notable higher fluxes by the shoreline could have been impacted by mobilization of mire deposits, hence more mobilized carbon could have been available for  $\text{CH}_4$  production in the lake sediments (Vonk et al., 2015). There is a possibility that the northern part of Villasjön has been affected by nutrients transported from the mire (Kokfelt et al., 2010).

The highest diffusive sedimentary  $\text{CO}_2$  fluxes ranged between 56.92-61.54  $\text{mg m}^{-2} \text{d}^{-1}$ , and was measured in the northern part of Villasjön. In the southern part of Villasjön, the diffusive sedimentary  $\text{CO}_2$  fluxes ranged between 21.65-34.71  $\text{mg m}^{-2} \text{d}^{-1}$ , which are notable lower. This may be due, as mentioned above, to the close distance to the shore, hence possible erosion and permafrost thaw could have induced mobilizing of organic carbon into the lake.

As mentioned above, no linear relationship between the deployment-period averaged environmental parameters, and the diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes were presented. Notable is though that the highest diffusive sedimentary  $\text{CH}_4$  flux of 7.64  $\text{mg m}^{-2} \text{d}^{-1}$ , was measured in September 2017 in Villasjön, during a temporal period with relatively cold lake and air temper-

atures of 6.89 respectively 5.81°C. As mentioned, the diffusive sedimentary CH<sub>4</sub> flux is expected to be at optimum during warmer temperatures (Blake et al., 2015; Nozhevnikova et al., 1997; Yvon-Durocher et al., 2014), but here is the opposite pattern presented.

The diffusive water surface CH<sub>4</sub> fluxes in Villasjön were presented in Figure 23 and Table 14. The lake chambers fluxes followed the same trend as the EQ samplers fluxes, with lower diffusive water surface CH<sub>4</sub> fluxes in the most southern part of the lake and highest in the most northern part of the lake. The same pattern occurred for ebullitive CH<sub>4</sub> fluxes, with the highest fluxes in the northern part of the lake (Wik et al., 2013). The same systematic variation appeared for the diffusive sedimentary CO<sub>2</sub> fluxes which were in general higher in the northern part of the lake.

The overall measured CH<sub>4</sub> fluxes in Villasjön present a relatively distinct spatial pattern with the highest diffusive sedimentary and water surface CH<sub>4</sub> fluxes, as well as ebullitive CH<sub>4</sub> fluxes, in the northern part of the lake. The ebullitive CH<sub>4</sub> fluxes measured by Wik et al., (2013), were though measured and averaged for the study seasons 2009 to 2012, and the temporal variability needs to be considered. The measurements could though be analyzed in regards to spatial trends in Villasjön, and remaining lakes.

The diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes in *Inre Harrsjön* have been presented in Table 10. No clear trend was distinguished for the depth gradient with the EQ samplers IH160819 (I-III), where the deepest point measured a diffusive sedimentary CH<sub>4</sub> fluxes of 4.24 while the flux at 2.5 m and 1 m, was measured to 7.25 respectively 2.57 mg m<sup>-2</sup> d<sup>-1</sup>. The EQ sampler deployed at a depth of 1 m were placed at a thin organic layer, which could relate to the low diffusive sedimentary CH<sub>4</sub> flux measured at the location. The locations 7, 8 and 9, were all placed close to the west shoreline, by the fen border, and presented relatively different diffusive sedimentary CH<sub>4</sub> fluxes of 14.41, 5.50 respectively 2.57 mg m<sup>-2</sup> d<sup>-1</sup>. The ebullitive CH<sub>4</sub> fluxes was very low at the deepest point, approximate <5 mg m<sup>-2</sup> d<sup>-1</sup> (Wik et al., 2013), which is relatively similar to the diffusive sedimentary CH<sub>4</sub> flux. Ebullitive CH<sub>4</sub> fluxes measured close to locations 7, 8 and 9, presented relatively similar ebullitive CH<sub>4</sub> fluxes of approximate 10 mg m<sup>-2</sup> d<sup>-1</sup> (Wik et al., 2013).

The diffusive sedimentary CO<sub>2</sub> fluxes in *Inre Harrsjön* were in generally very low, compared to Villasjön and Mellersta Harrsjön, see Tables 8, 10 and 12, and ranged between 3.82-9.52 mg m<sup>-2</sup> d<sup>-1</sup> (without extrema IH1170731). The CO<sub>2</sub> concentration measurement points, which coincided with the selected CH<sub>4</sub> concentration measurements points, presented low correlations coefficients which might have affected the flux calculations, see Figure 20a and 18b. Two EQ samplers with very low correlation coefficients were noted, EQ sampler IH1160819 (R<sup>2</sup>:0.02) and IH1170731 (R<sup>2</sup>:0.37), with measured fluxes of 3.82 respectively 9.52 mg m<sup>-2</sup> d<sup>-1</sup>. EQ sampler IH1170731 presented a negative flux of -5.95 mg m<sup>-2</sup> d<sup>-1</sup>, which is not realistic, and seen as an outlier in the study. The low correlation coefficients for the CO<sub>2</sub> concentration may indicate that the diffusive sedimentary CO<sub>2</sub> fluxes are less representable, which is also noted by the large flux error, seen in Table 10.

The diffusive water surface CH<sub>4</sub> fluxes in *Inre Harrsjön* were presented in Figure 23 and Table 14. All sampling locations had higher fluxes in 2016 than in 2017, with the exception of IH1, placed in the south-east part of the lake, see Figure 1. Sampling location IH4 had slightly lower fluxes than remaining sampling locations, but in general could no other trends between the sampling locations be distinguished during the two study years. IH4 was placed in the northwest part of *Inre Harrsjön*, where the ebullitive CH<sub>4</sub> fluxes were very low (Wik et al., 2013).

All 5 EQ samplers deployed in *Inre Harrsjön* presented a broad range of diffusive sedimentary and water surface CH<sub>4</sub> fluxes with no systematic spatial variation, but presented in general relatively similar magnitudes of fluxes as the ebullitive CH<sub>4</sub> fluxes. As for the diffusive sedimentary CO<sub>2</sub> fluxes, no spatial trends were distinguished and a few outliers were noted.

The diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes in *Mellersta Harrsjön* have been presented in Table 12. The highest diffusive sedimentary CH<sub>4</sub> flux of 14.34 mg m<sup>-2</sup> d<sup>-1</sup> was presented at location 13, centered between the sampling locations 14 and 12 in the lake. The lowest diffusive sedimentary CH<sub>4</sub> flux of 4.03 mg m<sup>-2</sup> d<sup>-1</sup> was presented at location 12, by the stream inlet. At location 14, by the shore at the southern part of Mellersta Harrsjön, was the diffusive sedimentary CH<sub>4</sub> flux measured to 7.26 mg m<sup>-2</sup> d<sup>-1</sup>. The ebullitive CH<sub>4</sub> fluxes measured at the same locations (Wik et al., 2013), presented a different pattern. At the highest measured diffusive sedimentary CH<sub>4</sub> flux the lowest ebullitive CH<sub>4</sub> fluxes was measured, location 13, and at the lowest measured diffusive sedimentary CH<sub>4</sub> flux the highest ebullitive CH<sub>4</sub> fluxes was measured, location 12.

The diffusive sedimentary CO<sub>2</sub> fluxes followed the same trend as the diffusive sedimentary CH<sub>4</sub> fluxes with the highest flux at location 13, with a measured flux of 41.21 mg m<sup>-2</sup> d<sup>-1</sup>, while location 14 and 12 presented diffusive sedimentary CO<sub>2</sub> fluxes of 37.60 respectively 6.86 mg m<sup>-2</sup> d<sup>-1</sup>.

The diffusive water surface CH<sub>4</sub> fluxes for Mellersta Harrsjön were presented in Figure 23 and Table 14. All sampling locations presented higher diffusive water surface CH<sub>4</sub> fluxes in 2016 than in 2017, see Figure 23. Sampling location MH1, in the northern part of the lake, had the lowest diffusive water surface CH<sub>4</sub> during both study years. No other general trends could be distinguished among the sampling locations for the diffusive water surface CH<sub>4</sub> fluxes.

No general pattern of the diffusive sedimentary CH<sub>4</sub> fluxes, the diffusive water surface CH<sub>4</sub> fluxes and the ebullitive CH<sub>4</sub> fluxes could be distinguished. The diffusive sedimentary CO<sub>2</sub> fluxes did though follow the same pattern as the diffusive sedimentary CH<sub>4</sub> fluxes, with the highest and lowest fluxes at the same locations.

A clear temporal trend was displayed with the *monthly water surface CH<sub>4</sub> fluxes* in the three study lakes. In Villasjön were all monthly diffusive water surface CH<sub>4</sub> fluxes higher in 2016 than in 2017 in Villasjön, with September as an exception, see Table 14. Inre Harrsjön presented the same pattern but with June as an exception. In Mellersta Harrsjön were all water surface CH<sub>4</sub> higher in 2016 than in 2017. In Villasjön presented July and August much higher fluxes in 2016 compared to 2017. The fluxes in July and August were consistent with the air and water temperatures, which were higher in 2016 compared to 2017, see Tables 4 and 5. The same systematic variability occurred for Inre Harrsjön, which presented relatively high fluxes in July and August during 2016, coherent with the high air and water temperatures measured during the same time period. Mellersta Harrsjön presented the same pattern, with the exception of August which demonstrated high fluxes in 2016, but had warmer water temperatures in August 2017 than in 2016. Diffusive water surface CH<sub>4</sub> fluxes were only measured in October during 2016 and not in 2017, but present the lowest fluxes overall. The temporal trend for June and September are less distinct, but overall the fluxes vary with air and water temperature in the three lakes. The systematic temporal variability was expected due to the increase of CH<sub>4</sub> production during higher temperatures (Blake et al., 2015; Nozhevnikova et al., 1997; Yvon-Durocher et al., 2014).

The robust annual analysis of the *interlake diffusive fluxes*, seen in Table 16, presents Mellersta Harrsjön with the highest diffusive water surface CH<sub>4</sub> fluxes and Villasjön with the lowest. This could be due to several spatial differences such as e.g. lake sediments, water surface inflow and mire deposits. Further research is needed to conclude the interlake variabilities. Due to the few measurements with the EQ samplers, and the great spatial and temporal variability, a further comparison of the diffusive sedimentary CH<sub>4</sub> and CO<sub>2</sub> fluxes would not be suitable.

A comparison of the diffusive sedimentary and water surface CH<sub>4</sub> fluxes were presented in Tables 17, 18 and 19. 5 out of 7 comparisons presented higher fluxes with the EQ samplers than with the lake chambers. Villasjön and Inre Harrsjön had both higher lake chamber fluxes in July 2016,

but the diffusive water surface  $\text{CH}_4$  fluxes portrayed high SDs, hence large variability occurred and the mean value may be less representable. Due to oxidation of  $\text{CH}_4$  to  $\text{CO}_2$  in the water column (Bastviken et al., 2002), it is expected that the lake chamber fluxes are lower than the EQ sampler fluxes, which is the overall trend when comparing the diffusive sedimentary  $\text{CH}_4$  fluxes with the water surface  $\text{CH}_4$  fluxes.

Flux ratios of  $\text{CO}_2:\text{CH}_4$  were presented in Figure 8, and concentration ratios of  $\text{CO}_2:\text{CH}_4$  were presented in Tables 8, 10 and 12, for the three study lakes. Almost all flux and concentration ratios exceeded the 1:1 ratio.  $\text{CO}_2:\text{CH}_4$  produced during methanogenesis is general equimolar, hence with a 1:1 ratio but Wilson et al., (2017) has presented evidence that the ratio of  $\text{CO}_2:\text{CH}_4$  in some cases exceeds the 1:1 ratio due to different oxidation states for the molecules involved in methanogenesis. The global warming potential will be modified with ratios exceeding the 1:1 ratio (Wilson et al., 2017), which affects the global warming potential assessments of northern lakes. Further research is needed, as well on the relationship between concentration ratios and flux ratios of  $\text{CO}_2:\text{CH}_4$ .

For the concentration ratios presented Villasjön the largest ratios, ranging between 3.52:1.00-5.66:1.00  $\mu\text{M}$ , at the northern part of the lake, which is consistent with the high diffusive sedimentary  $\text{CO}_2$  fluxes measured at the same location. On the contrary, presented Inre Harrsjön the highest ratio of  $\text{CO}_2:\text{CH}_4$ , 2.60:1.00 $\mu\text{M}$ , at the deployment location with the lowest measured diffusive sedimentary  $\text{CO}_2$  flux. In Mellersta Harrsjön has two out of three locations relatively high  $\text{CO}_2:\text{CH}_4$  ratios, ranging between 2.83:1.00-2.11:1.00 $\mu\text{M}$ , and the locations present as well the highest diffusive sedimentary  $\text{CO}_2$  fluxes. More research is needed on  $\text{CO}_2:\text{CH}_4$  ratios for further analysis of the global warming potential of high latitude lakes.

## 5 Conclusion

Net emission rates of  $\text{CH}_4$  and  $\text{CO}_2$  have been quantified for the three subarctic lakes Villasjön, Inre Harrsjön and Mellersta Harrsjön, during the ice-free seasons 2016 and 2017.

A systematic temporal variability for the monthly diffusive water surface  $\text{CH}_4$  fluxes has been presented for the study lakes. High fluxes were measured during warm months, with especially high fluxes during July and August. No distinct temporal trend was presented for the limited data set of the diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes.

A distinct spatial variability was presented for Villasjön, with high fluxes in the northern part of the lake, both in regards to ebullitive  $\text{CH}_4$  fluxes and diffusive  $\text{CH}_4$  and  $\text{CO}_2$  fluxes. This is likely due to eroded peatland close to the shoreline, hence more labile carbon is accessible (Kokfelt et al., 2010). Inre Harrsjön and Mellersta Harrsjön demonstrated a less systematic spatial variability than Villasjön. The highest diffusive water surface  $\text{CH}_4$  fluxes were measured in Mellersta Harrsjön and the lowest in Villasjön, likely due to spatial differences. Further research is needed to conclude the interlake variations of the diffusive water surface  $\text{CH}_4$  fluxes. An interlake comparison of the diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes would not be representable due to few measurements, and the impact of the spatial and temporal differences. The diffusive sedimentary  $\text{CH}_4$  were in general higher than the diffusive water surface  $\text{CH}_4$  fluxes, which is likely related to the oxidation of  $\text{CH}_4$  to  $\text{CO}_2$  in the water column (Bastviken et al., 2002).

The concentration ratios of  $\text{CO}_2:\text{CH}_4$ , measured with the EQ sampler, all exceeded the 1:1 ratio, which impacts the estimation of the global warming potential (Wilson et al., 2017), for northern lakes. The mole flux ratios of  $\text{CO}_2:\text{CH}_4$  exceeded in general the 1:1 ratios, but due to low correlation coefficients no conclusion could be made about the flux ratios. The relationship between the concentration ratios and the flux ratios of  $\text{CH}_4:\text{CO}_2$ , needs further research in order to fully understand what impact high latitude lakes have on the dynamics of  $\text{CH}_4$  and  $\text{CO}_2$ .

A significant correlation with air temperature, water temperature and atmospheric pressure was presented for the diffusive water surface  $\text{CH}_4$  fluxes in the study lakes. Villasjön and Inre Harrsjön demonstrated higher diffusive water surface  $\text{CH}_4$  fluxes during warmer air and water temperatures, presenting similar patterns as previous studies with increasing  $\text{CH}_4$  production during higher temperatures (Blake et al., 2015; Nozhevnikova et al., 1997; Yvon-Durocher et al., 2014). Increasing diffusive water surface  $\text{CH}_4$  fluxes were measured during decreasing atmospheric pressure for the lakes Villasjön and Inre Harrsjön. A similar pattern occurs for the ebullitive  $\text{CH}_4$  fluxes, which increases when the hydrostatic pressure is overcome (Martens & Val Klump, 1980; Bastviken et al., 2004). No significant correlation was displayed for Mellersta Harrsjön, and none of the northern lakes portrayed any linear relationship with wind speed, which is likely due to the monthly averaging which may dismiss fluctuations on a shorter time span. For the environmental parameters and the diffusive sedimentary  $\text{CH}_4$  and  $\text{CO}_2$  fluxes, no significant correlations were presented.

Further research is needed in order to understand the mechanisms of the diffusive sedimentary and water surface fluxes from northern lakes, and how the environmental parameters control the variability of the fluxes.

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# Appendices

## A CH<sub>4</sub> and CO<sub>2</sub> concentrations and statistical measures

### A.1 Overall

Table 1: Coefficient of variance for the low and high calibration standard measurements used during analyze of the equilibration samplers

ID	CV [%]		CV [%]	
	Low concentration	n	High concentration	n
VSI160801	0.36	8	1.39	15
VSII160801	3.96	18	0.21	10
VSIII160801	0.30	8	0.12	16
IHI160819	0.24	8	0.14	8
IHII160819	0.43	8	0.43	16
IHIII160819	0.30	8	0.13	16
VSI170629	3.99	16	0.16	16
VSII170629	0.49	20	0.62	20
VSIII170629	0.31	10	0.34	20
MHI170716	0.83	16	0.62	16
MHII170716	0.75	16	3.56	16
MHIII170716	0.70	16	0.18	16
IHI170731	0.65	16	0.62	19
IHII170731	0.53	16	0.25	16
IHIII17073	0.57	16	0.29	16
VSI170814		0	0.38	19
VSII170814	0.47	16	0.21	20
VSIII170814	0.92	15	0.24	19
VSI170924	2.93	16	0.50	16
VSII170924	2.86	16	0.35	16
VSIII170924	2.90	16	3.71	8
Mean	1.22		0.69	

Table 2: Additional information for comparison of CO<sub>2</sub> and CH<sub>4</sub> concentrations from equilibration samplers and water samples

ID	Date deployed	Equilibration samplers				Water samples				
		CO <sub>2</sub> <sub>n</sub>	CO <sub>2</sub> Range [ $\mu$ M]	CH <sub>4</sub> <sub>n</sub>	CH <sub>4</sub> Range [ $\mu$ M]	Sampling days	CO <sub>2</sub> <sub>n</sub>	CO <sub>2</sub> Range [ $\mu$ M]	CH <sub>4</sub> <sub>n</sub>	CH <sub>4</sub> Range [ $\mu$ M]
VSI160801	2016-07-19 - 2016-08-01	11	4.80 - 125.32	11	0.18 - 3.31	2016-07-22, 31	4	29.04 - 95.55	4	0.27 - 0.52
VSI160801	2016-07-19 - 2016-08-01					2016-07-22, 31	4	29.04 - 95.55	4	0.27 - 0.52
VSI170629	2017-06-21 - 2017-06-29	10	59.96 - 209.49	10	0.22 - 0.33	2017-06-22, 28	4	30.87 - 48.76	4	0.12 - 0.37
VSI170629	2017-06-21 - 2017-06-29	15	67.71 - 250.98	15	0.18 - 0.59	2017-06-22, 28	4	30.87 - 48.76	4	0.12 - 0.37
VSI170629	2017-06-21 - 2017-06-29	28	51.46 - 97.86	28	0.17 - 10.55	2017-06-22, 28	4	30.87 - 48.76	4	0.12 - 0.37
VSI170814	2017-08-03 - 2017-08-14					2017-08-03, 09	4	46.59 - 58.22	4	0.39 - 1.18
VSI170814	2017-08-03 - 2017-08-14	12	76.57 - 205.97	12	0.40 - 0.97	2017-08-03, 09	4	46.59 - 58.22	4	0.39 - 1.18
VSI170814	2017-08-03 - 2017-08-14	13	82.20 - 167.47	13	1.08 - 3.02	2017-08-03, 09	4	46.59 - 58.22	4	0.39 - 1.18
VSI170924	2017-09-14 - 2017-09-24	17	60.78 - 162.04	17	0.42 - 0.61	2017-09-13, 21	4	37.02 - 52.98	4	0.14 - 0.29
VSI170924	2017-09-14 - 2017-09-24	4	44.92 - 98.97	4	0.13 - 0.20	2017-09-13, 21	4	37.02 - 52.98	4	0.14 - 0.29
VSI170924	2017-09-14 - 2017-09-24	24	67.58 - 180.97	24	0.16 - 72.43	2017-09-13, 21	4	37.02 - 52.98	4	0.14 - 0.29
IHI160819	2016-08-11 - 2016-08-19	17	77.70 - 257.28	17	0.18 - 8.65	2016-08-07, 14, 23	3	51.00 - 87.47	3	0.59 - 1.13
IHI160819	2016-08-11 - 2016-08-19	4	80.15 - 160.49	4	0.19 - 0.64	2016-08-07, 14, 23	6	48.16 - 120.40	6	0.65 - 1.49
IHI160819	2016-08-11 - 2016-08-19	4	97.56 - 197.76	4	0.23 - 0.86	2016-08-07, 14, 23	3	59.34 - 117.79	3	0.65 - 1.85
IHI170731	2017-07-19 - 2017-07-31					2017-07-20, 26	4	31.66 - 65.77	4	0.55 - 0.75
IHI170731	2017-07-19 - 2017-07-31	10	80.71 - 178.85	10	0.23 - 0.49	2017-07-20, 26	4	31.66 - 65.77	4	0.55 - 0.75
IHI170731	2017-07-19 - 2017-07-31	29	204.47 - 368.64	29	0.34 - 1.50	2017-07-20, 26	4	31.66 - 65.77	4	0.55 - 0.75
MHI170716	2017-07-04 - 2017-07-16	31	155.34 - 260.51	31	0.60 - 4.05	2017-07-05, 12	4	100.82 - 133.57	4	0.89 - 1.92
MHI170716	2017-07-04 - 2017-07-16	31	164.11 - 879.04	31	0.39 - 17.09	2017-07-05, 12	4	100.82 - 133.57	4	0.89 - 1.92
MHI170716	2017-07-04 - 2017-07-16	29	123.8 - 283.61	29	0.54 - 3.67	2017-07-05, 12	4	100.82 - 133.57	4	0.89 - 1.92

Table 3: Additional information for comparison of CH<sub>4</sub> fluxes from equilibration samplers and lake chambers

Lake	Date	Equilibration samplers				Lake chambers			
		ID	Date deployed	n	CH <sub>4</sub> Range [mg m <sup>-2</sup> d <sup>-1</sup> ]	ID	Sampling days	n	CH <sub>4</sub> Range [mg m <sup>-2</sup> d <sup>-1</sup> ]
Villasjön	2016 July	VSI160801	2016-07-19 - 2016-08-01	3	4.78 - 7.21	VS1 - VS2	2016-07-06, 12, 21, 27	7	2.91 - 20.18
Villasjön	2017 June	VSI170629	2017-06-21 - 2017-06-29	3	3.53 - 6.09	VS1 - VS2	2017-06-13, 21, 28	6	2.84 - 5.35
Villasjön	2017 August	VSI170814	2017-08-03 - 2017-08-14	3	5.10 - 7.09	VS1 - VS2	2017-08-02, 09, 16, 23, 29	9	2.78 - 8.11
Villasjön	2017 September	VSI170924	2017-09-14 - 2017-09-24	3	5.45 - 8.80	VS1 - VS2	2017-09-06, 13, 20, 28	8	2.21 - 20.36
Inre Harrsjön	2016 August	IHI160819	2016-08-11 - 2016-08-19	2	2.57 - 7.25	IHI - IH4	2016-08-03, 09, 17, 25, 31	18	2.21 - 20.36
Inre Harrsjön	2017 July	IHI170731	2017-07-19 - 2017-07-31	2	5.50 - 14.41	IHI - IH4	2017-07-05, 12, 19, 26	16	1.99 - 10.47
Mellersta Harrsjön	2017 July	MHI170716	2017-07-04 - 2017-07-16	3	4.03 - 14.34	MHI - MH4	2017-07-05, 12, 19, 26	16	3.22 - 9.86

Table 4: Paired t-test for the lake chambers in Inre and Mellersta Harrsjön: comparisons of the months July, August and September, during the two study years 2016 and 2017

Inre Harrsjön				
Month	t(df)	<i>p</i>	CH <sub>4</sub> Mean 2016 [mg m <sup>-2</sup> d <sup>-1</sup> ]	CH <sub>4</sub> Mean 2017 [mg m <sup>-2</sup> d <sup>-1</sup> ]
July	t(3)=8.59	0.00	9.75	4.66
August	t(3)=3.07	0.05	6.76	4.20
September	t(3)=2.16	0.11	2.48	1.62

Mellersta Harrsjön				
Month	t(df)	<i>p</i>	CH <sub>4</sub> Mean 2016 [mg m <sup>-2</sup> d <sup>-1</sup> ]	CH <sub>4</sub> Mean 2017 [mg m <sup>-2</sup> d <sup>-1</sup> ]
July	t(3)=8.91	0.00	13.32	6.25
August	t(3)=10.26	0.00	11.21	4.32
September	t(3)=3.70	0.03	4.17	3.40

## A.2 Villasjön

Table 5: CH<sub>4</sub> concentrations and statistics for equilibration samplers, Villasjön

ID	CH <sub>4</sub> Mean [μM]	CH <sub>4</sub> Range [μM]	<i>D<sub>s</sub></i> [10 <sup>-7</sup> dm <sup>2</sup> s <sup>-1</sup> ]	<i>D<sub>s</sub></i> SD [%]	$(\partial c/\partial z)_{pw}$ [mol L <sup>-1</sup> dm <sup>-1</sup> ]	$(\partial c/\partial z)_{pw}$ CV [%]	n	$(\partial c/\partial z)_{pw}$ R <sup>2</sup>
VSI160801	332.22	0.18 - 753.54	1.50	5.31	-63.27	-5.30	6	0.99
VSII160801	358.94	0.42 - 558.96	1.50	5.31	-56.23	-13.96	7	0.91
VSIII160801	243.65	0.20 - 497.75	1.50	5.31	-55.06	-9.35	7	0.96
VSI170629	322.29	0.22 - 690.43	1.32	2.60	-54.07	-8.77	8	0.96
VSII170629	252.91	0.18 - 654.56	1.32	2.60	-36.17	-6.20	11	0.97
VSIII170629	85.36	0.17 - 335.27	1.32	2.60	-31.94	-9.05	8	0.95
VSII170814	229.90	0.40 - 402.03	1.41	2.13	-68.44	-7.33	6	0.98
VSIII170814	275.54	1.08 - 562.07	1.41	2.13	-36.92	-11.39	9	0.92
VSI170924	459.86	0.42 - 1002.44	1.15	3.30	-77.62	-10.46	8	0.94
VSII170924	618.24	0.04 - 922.52	1.15	3.30	-92.60	-2.92	7	1.00
VSIII170924	216.45	0.16 - 715.95	1.15	3.30	-58.94	-7.33	6	0.98

Table 6: CO<sub>2</sub> concentrations and statistics for equilibration samplers, Villasjön

ID	CO <sub>2</sub> Mean [μM]	CO <sub>2</sub> Range [μM]	$D_s$ [10 <sup>-7</sup> dm <sup>2</sup> s <sup>-1</sup> ]	$D_s$ SD [%]	$(\partial c/\partial z)_{pw}$ [mol L <sup>-1</sup> dm <sup>-1</sup> ]	$(\partial c/\partial z)_{pw}$ CV [%]	n	$(\partial c/\partial z)_{pw}$ R <sup>2</sup>
VSI160801	1645.33	4.80 - 3920.80	1.53	5.64	-188.69	-5.81	6	0.99
VSI160801	2188.55	20.13 - 3639.96	1.53	5.64	-198.10	-8.45	7	0.97
VSI160801								
VSI170629	1809.84	59.96 - 4142.12	1.34	2.76	-202.05	-5.54	8	0.98
VSI170629	850.50	67.71 - 1893.80	1.34	2.76	-58.73	-11.28	11	0.90
VSI170629	416.65	51.46 - 1313.31	1.34	2.76	-101.53	-9.95	8	0.94
VSI170814	677.61	76.57 - 1243.38	1.44	2.26	-89.18	-24.65	6	0.80
VSI170814	1180.40	82.20 - 2786.34	1.44	2.26	-85.67	-6.45	9	0.97
VSI170924	1658.17	60.78 - 4236.77	1.15	3.50	-196.62	-3.30	8	0.99
VSI170924	1812.78	44.92 - 3053.68	1.15	3.50	-140.96	-4.56	7	0.99
VSI170924	638.29	67.58 - 2054.79	1.15	3.50	-87.44	-9.93	6	0.96

### A.3 Inre Harrsjön

Table 7: CH<sub>4</sub> concentrations and statistics for equilibration samplers, Inre Harrsjön

ID	CH <sub>4</sub> Mean [μM]	CH <sub>4</sub> Range [μM]	$D_s$ [10 <sup>-7</sup> dm <sup>2</sup> s <sup>-1</sup> ]	$D_s$ SD [%]	$(\partial c/\partial z)_{pw}$ [mol L <sup>-1</sup> dm <sup>-1</sup> ]	$(\partial c/\partial z)_{pw}$ CV [%]	n	$(\partial c/\partial z)_{pw}$ R <sup>2</sup>
IHI160819	180.05	0.18 - 427.72	1.34	2.36	-34.65	-2.93	10	0.99
IHI160819	325.23	0.19 - 591.99	1.33	2.32	-47.57	-4.22	7	0.99
IHI160819	334.56	0.23 - 662.39	1.30	1.04	-34.02	-6.66	9	0.97
IHI170731	346.06	0.23 - 649.82	1.41	5.66	-95.93	-12.56	7	0.93
IHI170731	97.19	0.34 - 597.61	1.41	5.66	-44.16	-9.09	9	0.95

Table 8: CO<sub>2</sub> concentrations and statistics for equilibration samplers, Inre Harrsjön

ID	CO <sub>2</sub> Mean [μM]	CO <sub>2</sub> Range [μM]	$D_s$ [10 <sup>-7</sup> dm <sup>2</sup> s <sup>-1</sup> ]	$D_s$ SD [%]	$(\partial c/\partial z)_{pw}$ [mol L <sup>-1</sup> dm <sup>-1</sup> ]	$(\partial c/\partial z)_{pw}$ CV [%]	n	$(\partial c/\partial z)_{pw}$ R <sup>2</sup>
IHI160819	427.95	77.70 - 1120.22	1.36	2.51	-27.11	-22.51	10	0.71
IHI160819	675.87	80.15 - 1377.84	1.35	2.46	3.50	306.97	7	0.02
IHI160819	399.10	97.56 - 823.92	1.31	1.11	-14.20	-19.61	9	0.79
IHI170731	581.57	80.71 - 1064.13	1.44	6.02	20.75	35.81	7	0.61
IHI170731	438.63	204.47 - 1583.23	1.44	6.02	-25.44	-48.98	9	0.37

### A.4 Mellersta Harrsjön

Table 9: CH<sub>4</sub> concentrations and statistics for equilibration samplers, Mellersta Harrsjön

ID	CH <sub>4</sub> Mean [μM]	CH <sub>4</sub> Range [μM]	$D_s$ [10 <sup>-7</sup> dm <sup>2</sup> s <sup>-1</sup> ]	$D_s$ SD [%]	$(\partial c/\partial z)_{pw}$ [mol L <sup>-1</sup> dm <sup>-1</sup> ]	$(\partial c/\partial z)_{pw}$ CV [%]	n	$(\partial c/\partial z)_{pw}$ R <sup>2</sup>
MHI170716	121.45	0.60 - 516.72	1.29	3.40	-64.35	-7.80	7	0.97
MHI170716	101.62	0.39 - 577.99	1.29	3.40	-34.20	-4.20	9	0.99
MHI170716	236.68	0.54 - 771.34	1.29	3.40	-110.96	-11.37	7	0.94

Table 10: CO<sub>2</sub> concentrations and statistics for equilibration samplers, Mellersta Harrsjön

ID	CO <sub>2</sub> Mean [ $\mu\text{M}$ ]	CO <sub>2</sub> Range [ $\mu\text{M}$ ]	$D_s$ [ $10^{-7} \text{ dm}^2 \text{ s}^{-1}$ ]	$D_s$ SD [%]	$(\partial c / \partial z)_{pw}$ [ $\text{mol L}^{-1} \text{ dm}^{-1}$ ]	$(\partial c / \partial z)_{pw}$ CV [%]	n	$(\partial c / \partial z)_{pw}$ R <sup>2</sup>
MHI170716	539.17	155.34 - 1915.14	1.30	3.61	-112.56	-13.40	7	0.92
MHII170716	517.90	164.11 - 1334.90	1.30	3.61	-36.01	-35.03	9	0.54
MHIII170716	653.26	123.80 - 2505.20	1.30	3.61	-91.97	-19.32	7	0.84

## B Environmental parameters

### B.1 Overall environmental parameters during the study seasons 2016 and 2017

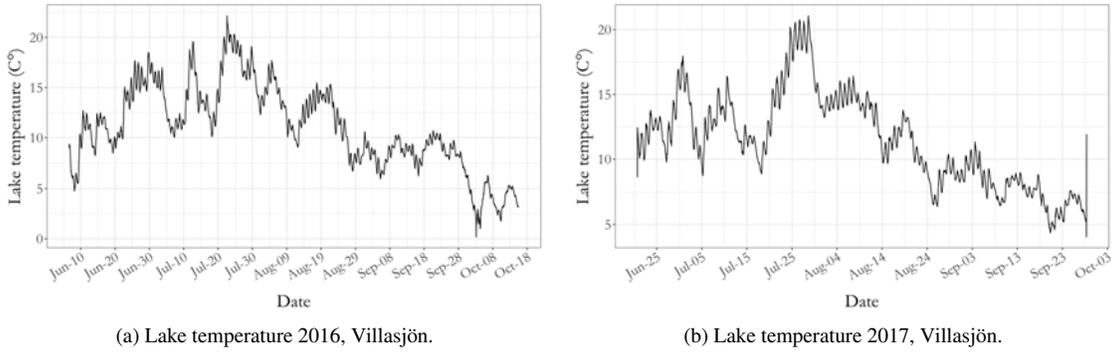


Figure 1: Villasjön.

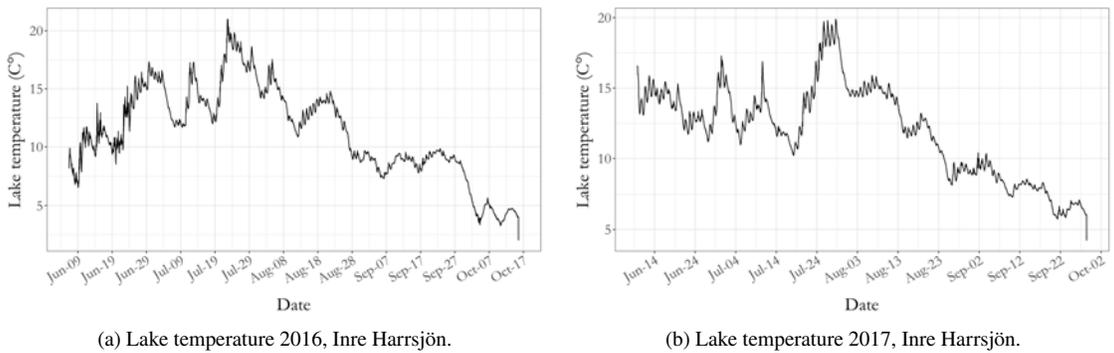
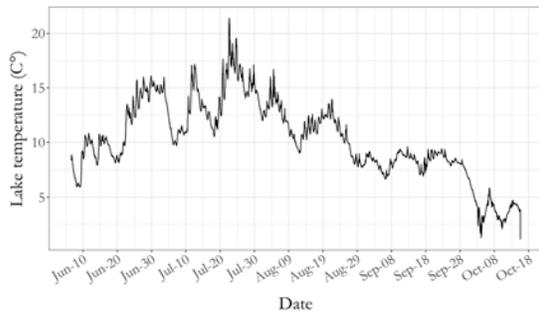
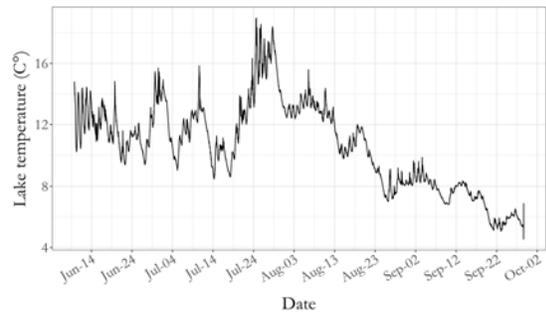


Figure 2: Inre Harrsjön.

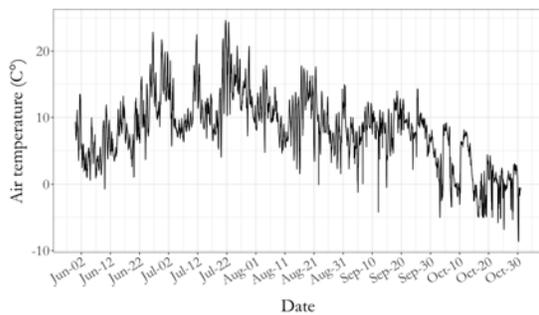


(a) Lake temperature 2016, Mellersta Harrsjön.

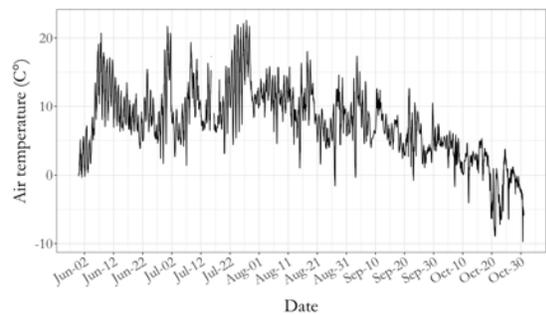


(b) Lake temperature 2017, Mellersta Harrsjön.

Figure 3: Mellersta Harrsjön.

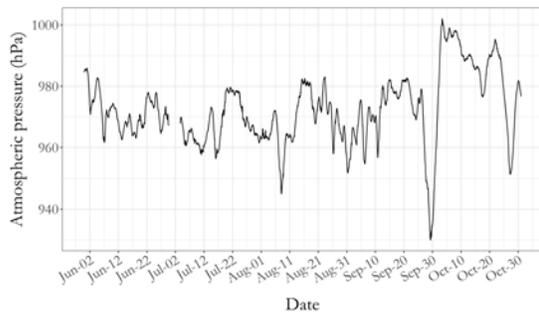


(a) Air temperature 2016, Stordalen.

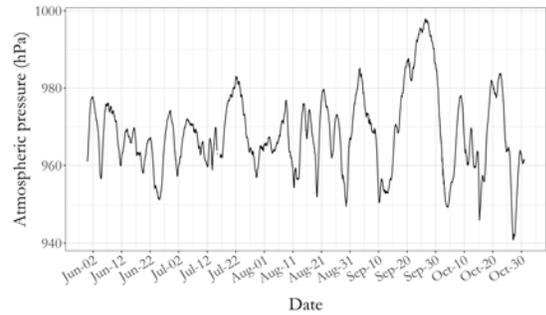


(b) Air temperature 2017, Stordalen.

Figure 4: Air temperature.

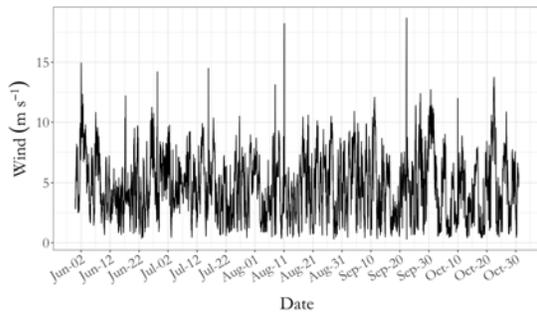


(a) Atmospheric pressure 2016, Stordalen.

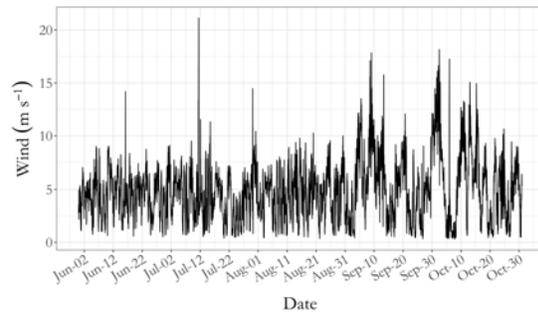


(b) Atmospheric pressure 2017, Stordalen.

Figure 5: Atmospheric Pressure.



(a) Wind 2016, Stordalen.

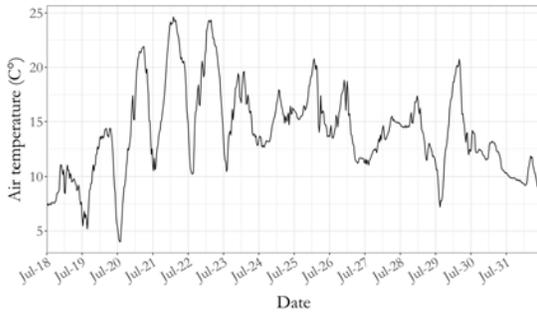


(b) Wind 2017, Stordalen.

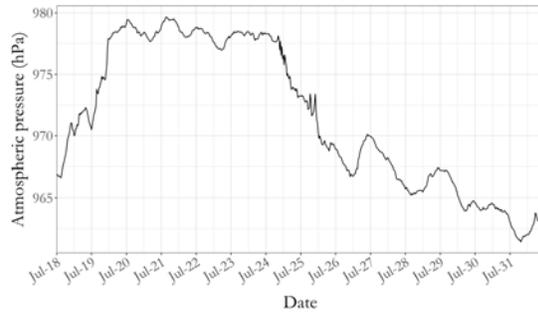
Figure 6: Wind.

## B.2 Villasjön

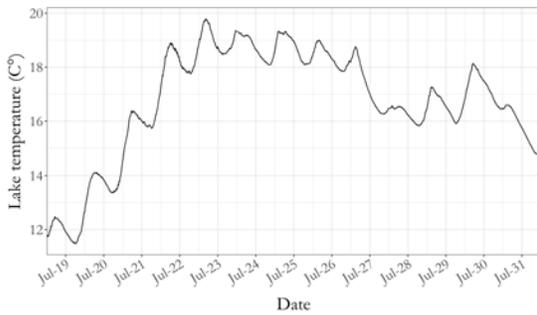
Environmental parameters measured during the deployment periods in Villasjön are presented. Lake temperature has been measured at a depth of 1 m.



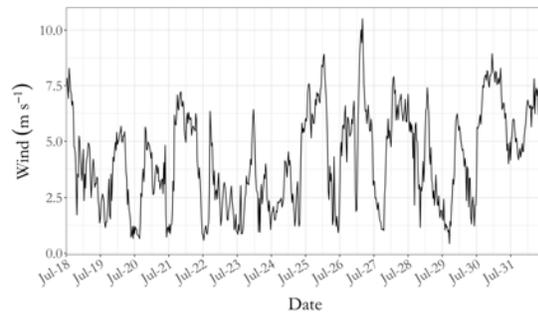
(a) Air temperature, Stordalen.



(b) Atmospheric pressure, Stordalen.

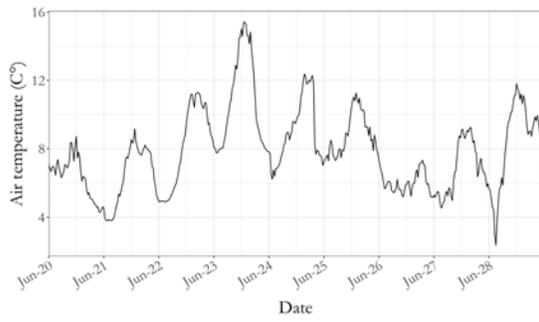


(c) Lake temperature at a depth of 1 m.

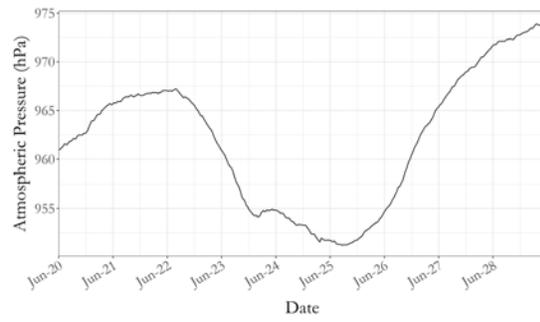


(d) Wind speed, Stordalen.

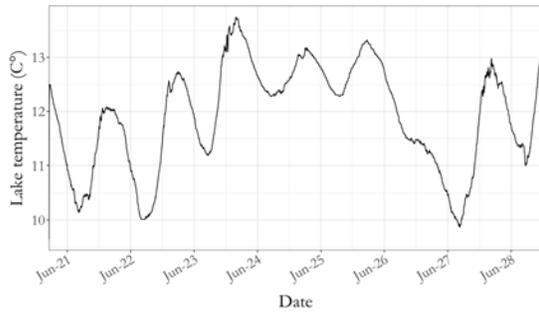
Figure 7: Environmental parameters for VS160801.



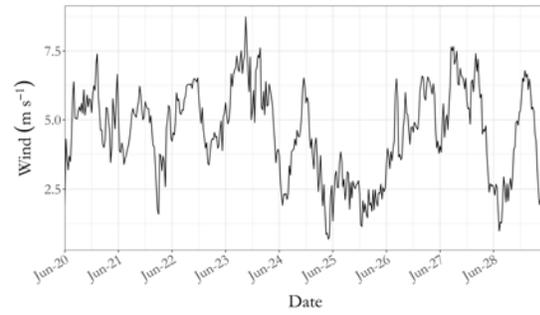
(a) Air temperature, Stordalen.



(b) Atmospheric pressure, Stordalen.

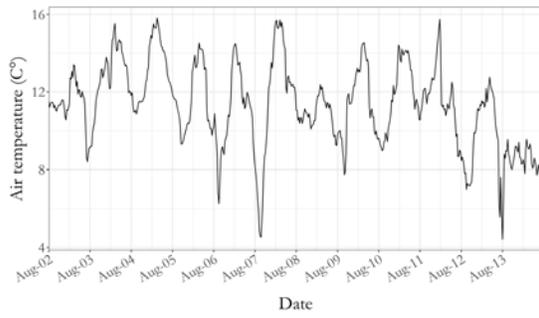


(c) Lake temperature at a depth of 1 m.

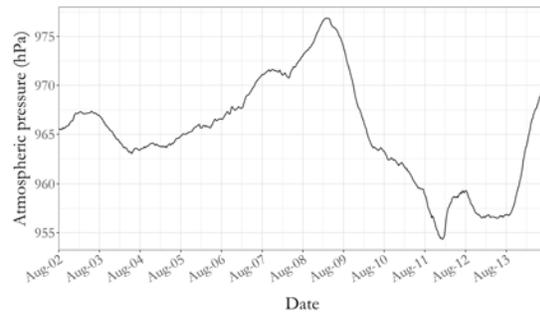


(d) Wind speed, Stordalen.

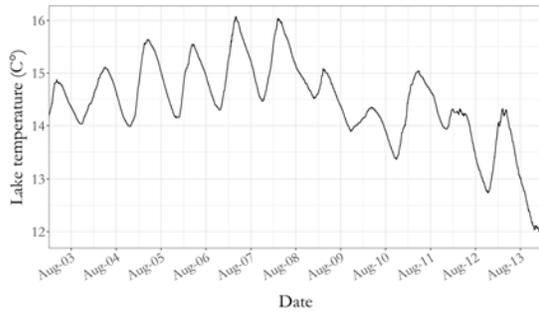
Figure 8: Environmental parameters for VS170629.



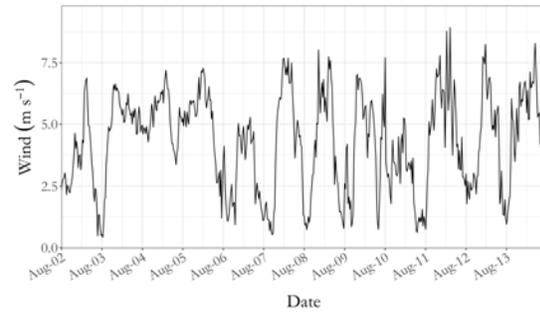
(a) Air temperature, Stordalen.



(b) Atmospheric pressure, Stordalen.

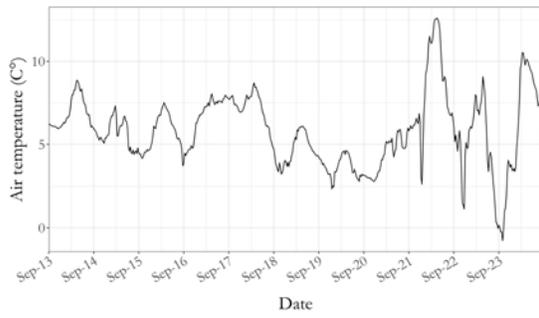


(c) Lake temperature at a depth of 1 m.

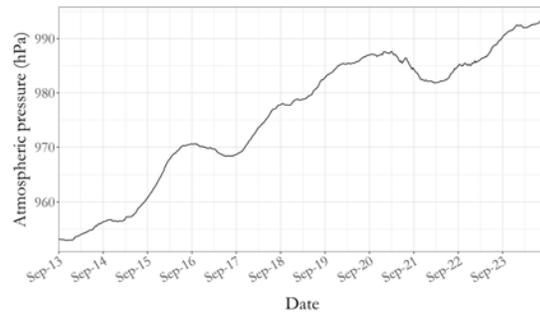


(d) Wind speed, Stordalen.

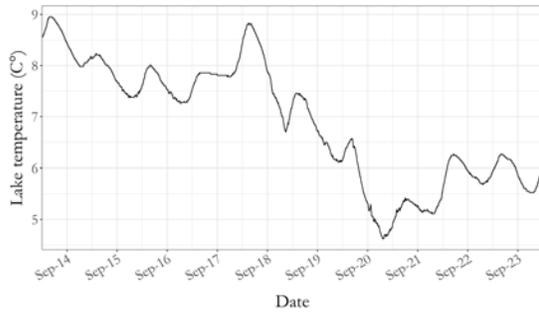
Figure 9: Environmental parameters for VS170814.



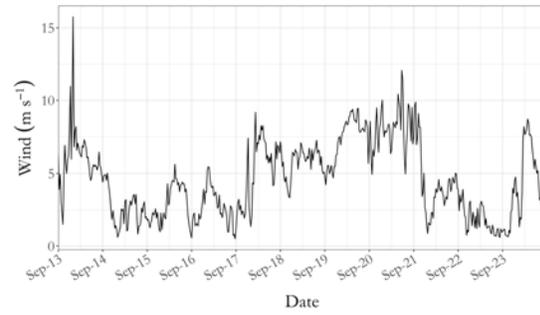
(a) Air temperature, Stordalen.



(b) Atmospheric pressure, Stordalen.



(c) Lake temperature at a depth of 1 m.

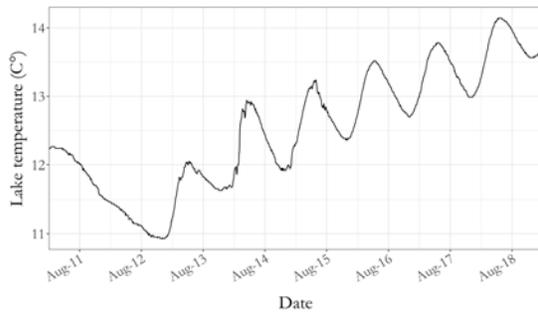


(d) Wind speed, Stordalen.

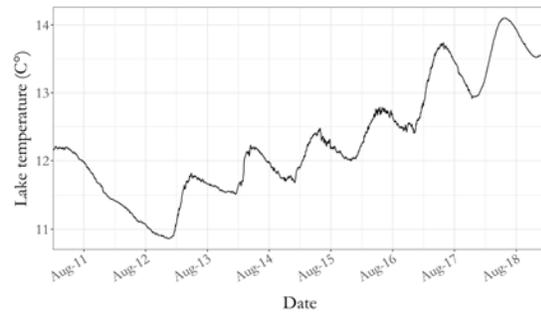
Figure 10: Environmental parameters for VS170924.

### B.3 Inre Harrsjön

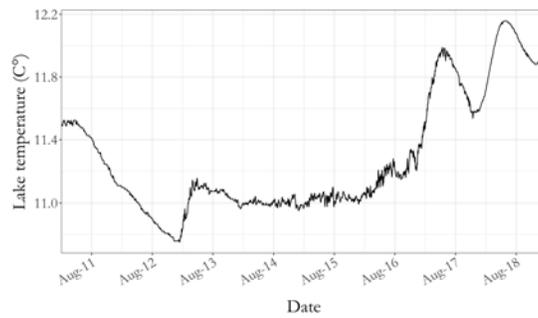
Environmental parameters measured during the deployment periods in Inre Harrsjön are presented. Lake temperature has been measured at a depth of 1 to 4 m.



(a) Lake temperature at a depth of 1 m, IHI160819.

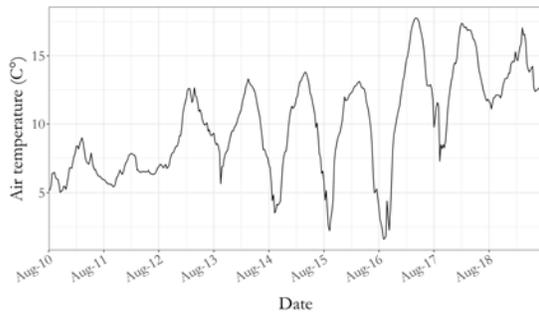


(b) Lake temperature at a depth of 2.5 m, IHII160819.

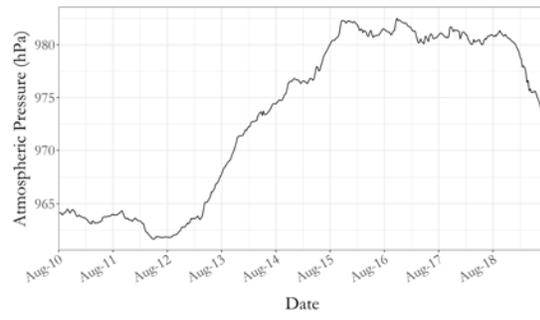


(c) Lake temperature at a depth of 4 m, IHIII160819.

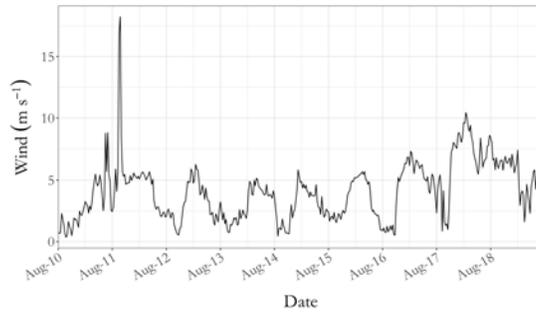
Figure 11: Lake temperature for IH160819.



(a) Air temperature, Stordalen.

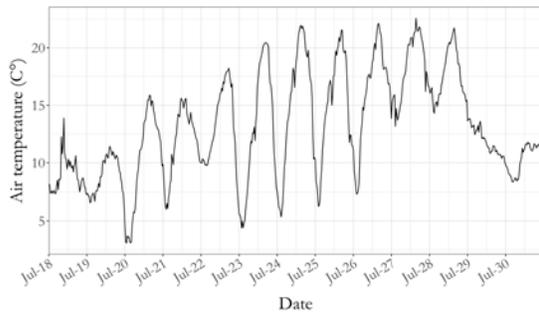


(b) Atmospheric pressure, Stordalen.



(c) Wind speed, Stordalen.

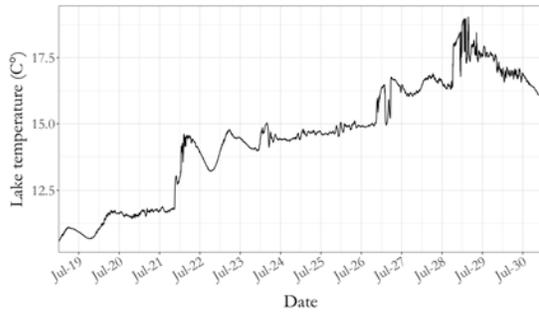
Figure 12: Environmental parameters for IH160819.



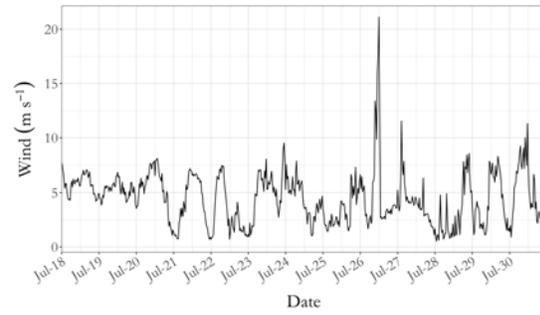
(a) Air temperature, Stordalen.



(b) Atmospheric pressure, Stordalen.



(c) Lake temperature at a depth of 1.5 m.

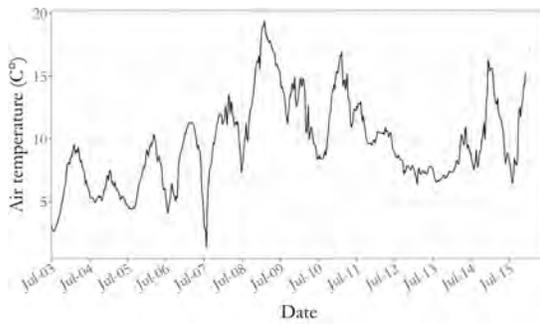


(d) Wind speed, Stordalen.

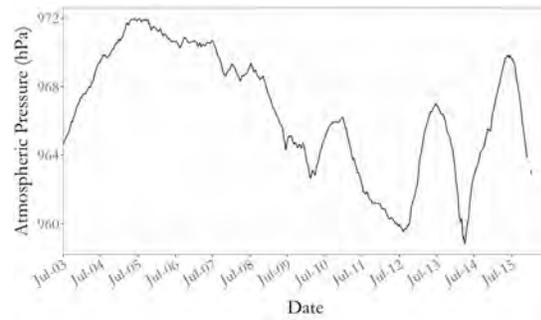
Figure 13: Environmental parameters for IH170731.

## B.4 Mellersta Harrsjön

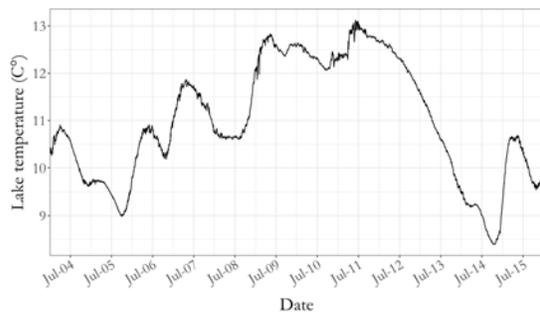
Environmental parameters measured during the deployment periods in Mellersta Harrsjön are presented. Lake temperature has been measured at a depth of 1.5 m.



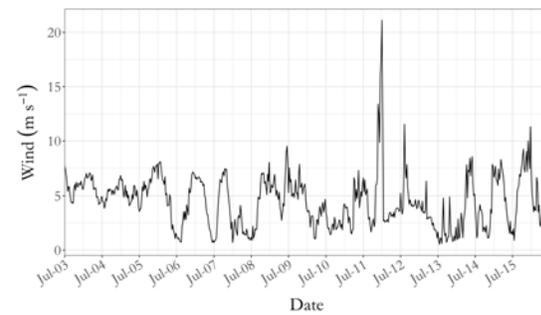
(a) Air temperature, Stordalen.



(b) Atmospheric pressure, Stordalen.



(c) Lake temperature at a depth of 1.5 m.



(d) Wind speed, Stordalen.

Figure 14: Environmental parameters for MH170716.