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Dissolved Methane Dynamics in a Subarctic Peatland



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Master degree thesis in Physical Geography and Ecosystem Analysis

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ABSTRACT

The dissolved methane (CH₄) dynamics in pore water and nearby stream water were investigated in a subarctic peatland in West Greenland during summer and autumn in 2011. The results from the field sampling indicate that the average pore water concentration in early autumn (the onset of freezing) was generally higher than that in summer, in particular in the deep peat layer (around 30 cm beneath soil surface). According to the results of a correlation analysis, the average dissolved CH₄ magnitude was negatively correlated with air temperature (R = -0.961, p = 0.019) and surface CH_4 flux (R= -0.880, P = 0.060). The higher concentration in early autumn suggests that the freezing ground and air temperature play an important role in the dissolved CH₄ dynamics. The frozen ground may restrict both CH₄ emissions to the atmosphere and oxygen (O₂) diffusion into the peat, and temperature influences the CH₄ solubility in water. Meanwhile, a significant inverse correlation was found between the pore water concentration in the deep layer and the surface CH_4 flux, (R = -0.977, p = 0.012). Increased stream water CH₄ concentration near the fen site, corresponding with higher dissolved CH₄ concentration accumulated in the deep peat layer during the early autumn, further indicated that the freezing ground not only affected the CH₄ transport mechanisms, but also may have a physical effect on the dissolved CH₄ export. The results presented may bring contribute to a better understanding of the autumn CH₄ dynamics in northern peatlands.

Keywords: Dissolved CH_4 , CH_4 flux, CH_4 solubility, Peatland, Pore water, Permafrost, Subarctic.

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INTRODUCTION

Climate change

Global warming is already a hot topic in climate research, as well as human daily life. Since the early 20th century, global average surface temperatures have risen by about 0.74 °C and with an increasing rate over the last 50 years (IPCC, 2007). A warmer climate may sound attractive to the people who live at high latitudes, as these then become more habitable. However, increasing global temperature will lead to global climate changes, i.e. sea level rise, precipitation pattern change and more extreme events will occur (Houghton, 2009). Scientists are confident that global warming is most likely due to rising emissions of greenhouse gases, mainly from human activities (IPCC, 2007; Houghton, 2009). The three important greenhouse gases are carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O). Ignoring the effects of other greenhouse gases (such as water vapor, CFCs and ozone), they have contributed to approximately 72%, 21% and 7% respectively of the enhanced greenhouse effect since the pre-industrial (1750) (Houghton, 2009).

The rate and scale of warming is not equally distributed around over the world. The rate of warming in the arctic region almost twice the global average rate (0.07 \C \pm 0.02 \C per decade) over the last 100 years (IPCC, 2007). Arctic ecosystem is widely considered as most vulnerable to climate warming, which is evident from observations of permafrost degradation (Christensen et al., 2004; Åkerman and Johansson, 2008), widespread melting of ice and snow (Serreze *et al.*, 2000; Tarnocai, 2006), increased emission of CO₂ and CH₄ (Johansson *et al.*, 2006; Svensson *et al.*, 1999), expanding of vegetation distribution and extending of the growing season (Malmer *et al.*, 2005).

Methane in northern peatlands

Northern peatland ecosystems play a significant role in the global terrestrial carbon cycle. Although the northern permafrost regions cover only about 16% of the global land area these regions store approximately 50% of the global soil organic carbon at present (McGuire et al., 2009, Tarnocai et al., 2009). Owing to the presence of waterlogged, anoxic and cool conditions, decomposition rates in northern peatlands are generally low, which is suitable for peat formation and carbon accumulation (Gorham, 1991; Moore and Basiliko, 2006). Meanwhile, Northern peatland is a dominating natural source of CH₄ with an estimated range between 32 and 112 Tg CH₄ emitted from these waterlogged soils to the atmosphere each year (McGuire et al., 2009), which contribute with 5% - 20% of the total average annual emissions, approximately 582 Tg (CH₄) yr⁻¹.the average over 2000 to 2005 (IPCC, 2007).

 CH_4 is believed to be a powerful greenhouse gas, on a mass basis CH_4 having 25 times the warming effect compared with the same mass of CO_2 over a 100-year time horizon (IPCC, 2007). CH_4 is contained in the Arctic in a number of forms, and once

possibly unlocked by adequately warmer temperatures, large amounts of dissolved CH_4 may be released into atmosphere sharply rather than gradually. Considering the current warming and thawing in the permafrost of the Arctic region, i.e. Greenland, Russia, Canada and northern America (Alaska) (IPCC, 2007; Pavlov, 1996), it is particularly important to understand the processes controlling the land-atmosphere exchange of CH_4 in peatlands.

Late-season CH₄ burst in the Arctic

In 2007, CH₄ emission monitoring in a wetland at Zackenberg (northeast Greenland, 74 30' N, 21 00' W), representing a high arctic peatland, showed a surprising second peak in early autumn (October) after the usual summer optimum (Mastepanov *et al.*, 2008). *Mastepanov et al.* (2008) suggested that physical factors were the main reason for this late-season (the period of the end of growing season, usually in early autumn) CH₄ burst in Zackenberg. That is, when the soil at the surface started freezing, high pressure would build up between the deepening frost front and the permafrost bottom, so that methane-saturated soil between was sandwiched and gas bubbles squeezed out via plant stems or tiny cracks among ice. This hypothesis is supported by the fact that such large amounts of CH₄ emitted during the late-season have not been observed in subarctic peatlands without permafrost.

Objective and hypotheses

The study site in this project was located in the low Arctic region, named Kobbefjord in west Greenland, where any late-season CH_4 burst had not been observed in the previous and ongoing CH4 flux monitoring. The aim of this study is to investigate the seasonal changes of dissolved CH_4 in a subarctic peatland. Through studying the dissolved CH_4 dynamics we may better understand the processes affecting late-season CH_4 emission in northern peatland.

The hypotheses are, 1) Increased CH_4 solubility at lower temperature and reduced CH_4 transportation at colder condition lead to pore water CH_4 concentration increase as the ice cover is forming and the frozen front progressing. 2) Under high pressure, CH_4 beneath the freezing ground tend to accumulate at the deep peat layer. Further, we hypothesize that peatland with thicker active layer may have better capacity of storing CH_4 during onset of freezing making any late-season burst unlikely. 3) A peatland with good mobility of water, such as one located nearby stream, river and lake, may reduce the magnitude of CH_4 stored in the fen and potentially leads to lower possibility of the late-season burst.

BACKGROUND

Methane is a moderately reactive trace gas, having a relatively short lifetime in the atmosphere of about 7.9 years (Lelieved *et al.*, 1998). It plays important roles in atmospheric chemistry and in the global heat balance. The concentration of CH_4 in the atmosphere is much lower than that of CO_2 , but it is more effectively trapping infrared radiation and therefore contributes significantly to the "greenhouse effect" of

the Earth. Major sources of CH₄ includes biomass burning, gas release during oil production and coal mining, ruminant animal population, rice paddies and natural wetlands, which contribute about 10% 15%, 14%, 20% and 24% respectively of the estimated total annual CH₄ emissions globally (Lelieved *et al.*, 1998; Whalen, 2005; Backman, 2009). Most CH₄ is removed from the atmosphere via oxidation with hydroxyl radicals (OH) in the troposphere and a minor proportion by microbial oxidation in soils (Levy, 1971; Lelieved *et al.*, 1998; Whalen, 2005; Backman, 2009). An assessment of the dissolved CH₄ dynamic in subarctic peatland would conduce towards a better understanding of the major mechanisms of peatland CH₄ production, consumption and transport, as well as the important factors controlling variations in CH₄ exchange and CH₄ solubility.

Methane production

Methane production takes place in waterlogged and substrate abundant peat, owing to much organic matter can be broken down by methanogenic bacteria under the necessary anaerobic conditions (Whalen, 2005; Christensen, 2010). Methanogens, a special group of microorganisims, are the major producers of methane in the anaerobic soils of wetlands. These obligate anaerobes mainly use acetate as a carbon substrate to produce CH_4 and CO_2 (Whalen, 2005):

$$CH_3COOH \rightarrow CH_4 + CO_2, \tag{a}$$

while other substrates, such as H_2 and CO_2 , can also contribute to methanogenesis (Whalen, 2005):

$$4H_2 + CO_2 \rightarrow CH_4 + 2H_2O,$$
 (b)

Hornibrook et al. (1997) noted that in general acetoclastic methanogenesis (Eq. a) was favored in the upper peat layer, and H_2 and CO_2 dependent methanogenesis (Eq. b) would take place in the deeper layer.

Methane consumption

Some of the methane produced through the bacterial methanogenesis is consumed in the aerobic zones by methanotrophs, a subgroup of eubacteria that use CH₄ to gain energy and carbon for their growth and maintenance (Hanson and Hanson, 1996). Highest methanotrophic activity in peatlands generally occurs surround the average water table, normally considered as the boundary between aerobic and anaerobic zones, where the optimal ratio of substrates to oxygen for functioning is present (Dedysh, 2002). The reasons are, below the water table level, inadequate supply of oxygen results in limitation of oxidation reactions, and above, less methane is available for supporting the methanotrophic activities (Dedysh, 2002). Although methanotrophic activity was regarded as the dominant consumer of CH₄ in wetlands and estimated to remove about 30 Tg of CH₄ from the atmosphere each year, at the large scale, the largest sink for atmosphere CH₄ is reaction with tropospheric OH⁻, this process removes approximately 510 Tg of CH₄ per year (Lelieved *et al.*, 1998; Whalen, 2005; Backman, 2009).

Methane transport

Once produced, CH_4 can be transported into the atmosphere by three main pathways: diffusion, ebullition and plant-mediated transport (Chanton, 2005; Lai, 2009).

Diffusive CH_4 emission is the random movement along a concentration gradient, from higher to lower concentration (Chanton, 2005). The rate of diffusion is regulated by the concentration gradient or the change in concentration (Berner, 1980). In general, diffusion is the slowest one of the three transport mechanisms, though it plays an important role in methanotrophic activities at the upper aerobic layer (Whalen, 2005).

Ebullition, release CH₄ in the form of gas bubbles, contributes to a significant fraction of total CH₄ emission in northern peatland (Christensen *et al.*, 2003a; Rosenberry *et al.*, 2003; Tokida *et al.*, 2007). Because of the low solubility of CH₄ and very high rates of methanogenesis, supersaturated CH₄ appearing as gas bubbles can be found in the pore water of deep anaerobic peat layers (Yamamoto *et al.*, 1976; Chanton and Whiting, 1995; Lai, 2009). As the size of bubbles grow close to the pore diameter and partial pressure reaches the threshold pressure level, these bubbles would be suddenly ejected into atmosphere (Kellner *et al.*, 2004; Kellner *et al.*, 2005). At the same time, most of them episodically escape to the atmosphere without being oxidized by methanotrophs due to the low solubility and rapid transportation (Boone, 2000). The ebullition can be triggered by falling atmospheric pressure, reducing hydrostatic pressure and rising temperature (Tokida *et al.*, 2007; Strack *et al.*, 2005; Fechner-Levy and Hemond, 1996).

Plant-mediated transport mechanism is the process where some emergent plants or vascular plants conduct the CH₄ produced in anaerobic zones passing through their roots and aerenchymatous tissues, and finally emit to the atmosphere, the whole process bypassing methane-oxidizing the peat layer (Chanton, 2005; Tokida, 2005; Whalen, 2005). The main drivers of this mechanism are the concentration gradient, the pressure gradient and the temperature gradient between the plant interior and exterior (Whalen, 2005; Lai, 2009). Therefore, these plants can serve as conduits not only for CH₄ transport from the sediment, but also for transporting O_2 to the root and rhizomes (Chanton, 2005; Whalen, 2005). This plant-mediated transport enables efficient CH₄ emission in reference to previous experiments and studies, presenting that vegetated areas generally emit 10 times the amount of CH₄ relative to the non-vegetated areas of peatlands (Whiting *et al.*, 1991; Whiting and Chanton, 1992). Certainly, the magnitude of the emission via this pathway is highly also species-dependent (Bubier, 1995).

Factors affecting CH₄ emission

Important controlling factors of CH_4 emission from wetland include substrate, water table position, soil temperature, vegetation types and net ecosystem exchange (NEE) (Christensen *et al.*, 2003b; Whalen, 2005; Lai, 2009). These environmental variables are strongly correlated and the affects from each are generally hard to distinguish from the other.

Available organic substrate is a prerequisite for methanogenic activity taking place when anaerobic conditions are established in a wet soil (Segers, 1988; Coles and Yavitt, 2002). These organic substrates, especially organic acid, provide carbon (C) for CH_4 production and have a significant correlation with seasonal averaged CH_4 emissions (Christensen *et al.*, 2003b).

Water table position plays a critical role for CH_4 emission from peatlands (Blodau and Moore, 2003; Whalen, 2005; Lai, 2009). The main effect of the water table is determining the boundary between aerobic and anaerobic zones in waterlogged soils, which regulate the soil volume available for methanogenic and methanotrophic activity respectively. *Christensen et al.* (2003b) noted that water table most likely acted as "on-off switch" for large-scale CH_4 emissions, when water table within 10 cm depth relative to soil surface, it has significant correlation with CH_4 flux. Once water table is around or beyond the 10 cm depth, CH_4 flux will be dominated by other environmental controls.

Soil temperature is one of the key environmental controls of CH₄ emission (Whalen, 2005; Lai, 2009; Christensen, 2010). The temperature has direct influence on the activity of methanogens and methanotrophs, and further on the microbial-mediated CH₄ production and oxidation rates and ultimately on the total CH₄ emission (Dunfield *et al.*, 1993; Bubier and Moore, 1994). Dunfield *et al.* (1993) showed the optimum temperature for both microbial production and consumption were around 20 and 25 $^{\circ}$ C in subarctic peatlands with CH₄ production being much more sensitive to temperature changes.

Vegetation type is another important controlling factor of CH₄ emissions. On one hand, vegetation can fix carbon from the atmosphere via photosynthesis, and provide labile C substrate for CH₄ production (Christensen *et al.*, 2003; Ström *et al.*, 2003; Ström *et al.*, 2005; Lai, 2009). On the other hand, some species, like vascular plants, can transport CH₄ from peat to the atmosphere while transfer O₂ in the atmosphere to the root and rhizomes (Chanton and Whiting, 1995; Chanton, 2005; Whalen, 2005), and further influence net CH₄ exchange in the peatlands. Therefore, changes in vegetation species and coverage in a given wetland would lead to CH₄ flux changes.

NEE is also considered as an important factor influencing CH_4 flux although the relationship is considered uncertain. *Whiting* and *Chanton* (1993) found variability of NEE was correlated to the changes of CH_4 flux, based on measurements in 9 wetland sites, with the span of 25 latitudinal degrees. A later study by *Christensen et al.* (2003), showed no correlation between mean seasonal NEE and CH_4 flux based on the measurements in 5 wetland sites, with the span of 17 latitudinal degrees.

Factors affecting CH₄ solubility

The solubility of CH_4 in water is rather low. According to the nature of CH_4 , the solubility increases with a rise in pressure, and as temperature drops. The dissolved CH_4 will always follow Henry's law, stating that the solubility of a gas in a liquid is determined by temperature, the partial pressure of the gas over the liquid, the nature of both the solvent and the gas (Volland, 2011).

METHODS

Site description

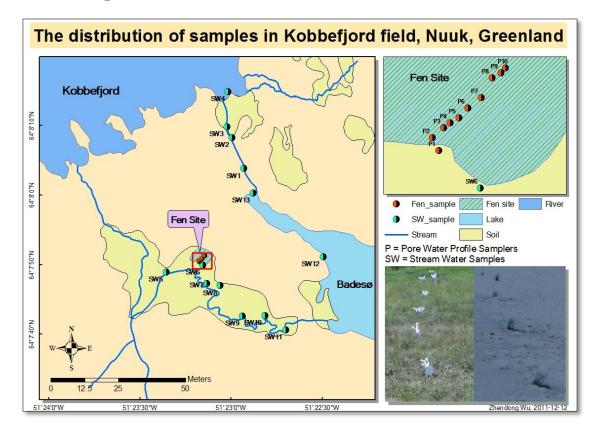


Figure 1 The map gives overview of the Kobbefjord region. SW1-SW13 (green-black circle) represents location of sampling in nearby stream. The magnified map in top right corner shows location of pore water samplers (red-black circle), P2-P10 in fen site, P1 in the stream (see Figure 3). The base map is created from DEM (Digital Elevation Model) data and feature data. Projection system refers to WGS_1984_Complex_UTM_Zone_22N. The photo in lower right shows the real distribution of the samplers in summer and autumn.

This project studying dissolved CH₄ measurements was carried out in a subarctic fen in Kobbefjord research area (64 07'51.31" N, 51 23'08.71"W). This study site located in West Greenland, approximately 20 km from Nuuk (the Capital of Greenland), was established as part of the GeoBasis (Greenland Environmental Monitoring program, Nuuk Basic) in 2007. The fen here is permanently wet and there are some places with more than 1 m of peat, which promote anaerobic decomposition (Iversen, 2010). These conditions were in favor of methane production. Figure 1 shows the overview of Kobbefjord area, the feature with wetlands-color is the fen site named SoilFen, which is one of three automatic soil stations in this area. According to the meteorological data from August 2007 to October 2010, mean monthly air temperature at SoilFen during summer ranged from +4.1 $\$ to +11.3 $\$, and between -5.5 $\$ and -13.5 $\$ during winter (Jensen and Rasch, 2011). The dominant wind direction came from NE, except during summer where it changed slightly to a westerly wind (Jensen and Rasch, 2011). The average relative humidity was 72.9% for 2007 - 2010, with a standard deviation of 4.6% (Jensen and Rasch, 2011). The vascular plant community at SoilFen was dominated by *Carex rariflora*, *Scirpus caespitosus*, *Eriophorum angustifolium* (Bay *et al.*, 2008). Especially, the *Eriophorum* species had a highly significant correlation with CH₄ emissions (Ström *et al.*, 2012).

Measurements

Pore water concentration

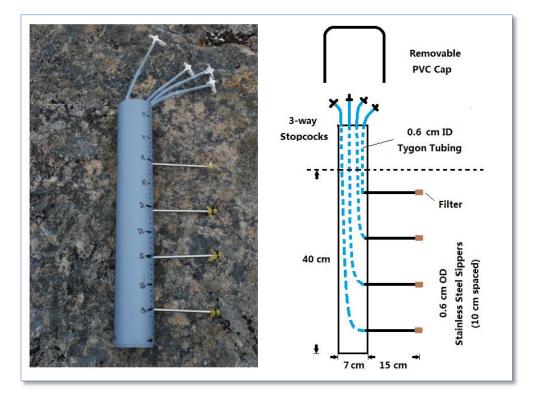


Figure 2 (left) Pore water sampler used in field work and (right) pore water sampler model shows the principle of construction.

A pore water sampling method (Melloh and Crill, 1996) was used to obtain water samples in the fen site, the principle of construction of the sampler illustrated in Figure 2. Nine samplers were deployed in a row from the outer boundary of the fen to its central part with 4-6 m interval, as showed in Figure 1 (P2-P10). Each sampler was vertically installed into the peat on August 3, 2011. Two steel sheets were used to gently spread peat, meanwhile take out some peat so that PCV pipes can be perpendicularly inserted into the peat, and then slipped the horizontal sippers into place, at last carefully covered the peat back. The samplers allowed repeat sampling in the unfrozen peat layers from 5-, 15-, 25- and 35-cm depth. At every sampling occasion, pore water was drawn up by 50-ml plastic syringe until the pore water flow was bubble-free, and then switching 3-way stopcocks to block the water flow in the tygon tubing. After that 2-ml sample was collected from the water flow using 10-ml plastic syringe, and injected into a 12-ml glass vial that had been prepared in advance marked and with 12-ml fresh ambient air. After sampling, a measured amount of

ambient air was pushed back into the tygon tubing to empty the tubes for water, in order to avoid freeze-up between sampling dates.

Stream water samples were collected by 10-ml plastic syringe from the stream, which surround the fen site and connected it with Badesødrainage basin Kobbefjord, as showed in Figure and (SW1-SW13). One of the samplers was used to collect stream water sample as well, it was located in the stream next to the fen (3 m away from P2), as showed in Figure 3 (P1). The quantity of stream water samples was the same as pore water samples the summer, 2011. (2 ml), and using the same previously prepared vials to store the samples.



Figure 3 Pore water sampler (P1) was used to collect stream water sample. The photo was taken in

A gas chromatographic method, the most common and powerful technique for trace gas analysis, was used to analyze the water samples in this experiment (Matson et al., 1995). There were 329 samples collected in this project, 211 in summer and 118 in autumn (during the onset of freeze-in). The autumn samples were diluted with 5 ml fresh ambient air (inject 5 ml ambient air into each sample vial prior to analysis). Samples in summer were without dilutions. Before processing began, samples were allowed to warm 1.5 hour at room temperature. In summer, SRI 8610C Gas Chromatograph (GC) equipped with Thermal Conductivity Detector (TCD) was used to analysis. Injection, detection and column oven temperatures were 180 $^{\circ}$ C, 180 $^{\circ}$ C and 40 °C, respectively, and inert gas helium (He) was used as carrier gas. Prior to analysis, the vial was shaken for 15 seconds to extract dissolved CH₄ and promote the gas mixing inside vial. Then 1 ml of the headspace from vial was sampled by syringe and directly injected into the column of GC (without a sample loop). In autumn, samples were processed on a Shimadzu (Kyoto, Japan) GC-2014 gas chromatography with a Flame Ionizing Detector (FID). Injection, detection and column oven temperatures were 140 °C, 140 °C and 40 °C, respectively, and inert gas helium (He) was used as carrier gas. 4 ml of the headspace (injected through a sample loop) was analyzed by the GC after 15 seconds shaking treatment.

CH₄ fluxes

An automatic chamber method was used to monitor carbon gas (CH₄ and CO₂) fluxes at the Kobbefjord SoilFen site. The system consisted of six flux chambers, and was designed for automatic round-the-clock monitoring of the exchange of CH₄ and CO₂. The chambers were connected to a set of analytical instruments (CH₄ and CO₂ analyzers) by couples of tubes of the same length. Each chamber had a moveable lid that could be automatically opened and closed by a motor. When the lid was open, the gas mixture inside the chamber was equal to the ambient; when closed, the concentrations of some gases can decrease due to consumption, for others it can increase due to emission. The rate of variation is proportional to the gas flux. Using certain clock and a set of valves, chambers worked one after the other in an agreed order. For more detail see Mastepanov (2010).

Air temperature, soil temperature and water table

The air temperature and soil temperature data were obtained from Kobbefjord SoilFen micrometeorological station located about 80 m away from the site. Air temperature was measured by thermometer at 2 m height relative to surface, and soil temperature was taken by sensors at four depths. The visual water level relative to soil surface was monitored manually by a piezometer located between chamber 5 and 6.

Data handling

Pore water concentration

The dissolved CH_4 concentrations in pore water samples were calculated using the ideal gas law (Dickerson *et al.*, 1979), and assuming almost 100% of dissolved CH_4 was extracted to the vial headspace after 15 seconds of shaking. Since the GC provided the concentration data in ppm, in order to convert them into traditional unit (µmol/L), the data of ambient pressure and air temperature were used.

Parts per million (ppm) is equal to a certain amount of volumes of a given gas mixed in a million volumes of air (Thompson and Taylor, 2008). That is, 1 ppm would therefore be equal to 1 micro liter volume of gas in 1 liter of air:

$$1ppm = \frac{1 \,\mu\,L\,gas}{1 \,L\,air} \,\,, \tag{1}$$

To convert ppm to a metric expression, at the different temperature and pressure conditions, the density of the concerning gas is needed. The density of gas can be calculated by the Avogadro's Law (Dickerson *et al.*, 1979), and specific molar volume of ideal gas at pressure P and temperature T can be calculated by the ideal gas Law:

$$1ppm = \frac{V_n}{M} \frac{1 \,\mu g \,\text{gas}}{1 \,\text{L} \,\text{air}}, \qquad (2)$$

$$V_n = V/n = R \frac{T}{P} , \qquad (3)$$

Where:

V_n = specific molar volume of ideal gas (at pressure P and temperature	e T) [L/mol]
M = molecular weight of gas	[g/mol]
V = volume of the gas	[m ³]
n = amount of molecules	[mol]
R = universal gas law constant	$[8.314510 \text{ m}^3 \text{ Pa K}^{-1} \text{ mol}^{-1}]$
T = temperature	[K]
P = pressure	[Pa]

To sum up, conversation from ppm to μ mol/L was calculated by the following expression:

$$C_{\mu mol/L} = \frac{100*C_{ppm}*P_{hPa}}{1000*R*(273.15+T_C)} , \qquad (4)$$

Where:

$C_{\mu mol/L} = CH_4$ concentration	[µmol/L]
$C_{ppm} = CH_4$ concentration	[ppm]
R = universal gas law constant	[8.314510 m ³ Pa K ⁻¹ mol ⁻¹]
T _c = air temperature	[°C]
P _{hPa} = air pressure	[hPa]

However, the CH₄ concentrations measured by the GC just represented the concentration of the vial headspace. The quantity of CH₄ in 2 ml Pore water sample should be equal to the measured concentration by the GC (C_{GC} , ppm) minus the CH₄ concentration in ambient air (C_{amb} , 1.8 ppm), and then multiplied by the total air volume in the glass vial ($V_{vialair}$, ml). Therefore, the pore water concentration (C_{sample} , µmol/L) can be calculated using equation 4:

$$C_{\text{sample}} = \frac{0.5*100*(C_{GC} - C_{amb})*V_{vialair}*P_{hPa}}{1000*R*(273.15+T_{C})} , \qquad (5)$$

CH₄ solubility

In reality, dissolved CH_4 cannot be totally extracted to the vial headspace after 15 seconds of shaking, therefore this experiment just measured the extracted section of CH_4 to the vial headspace (the partitioning of CH_4 in gas phase), without including the section remaining in the sample (the partitioning of CH_4 in liquid phase). To get high accuracy on the pore water concentration numbers a study of CH_4 solubility was need. Therefore, the dissolved CH_4 concentrations discussed in this paper were the overall amount of CH_4 in the sample, including the portions of CH_4 in gas and liquid phases.

CH₄ solubility prediction using Henry's law, a common method for quantifying the solubility of a gas in a liquid, which asserted that the solubility of a solute (CH₄) in a solvent (H₂O) is directly proportional to the solute's gas phase partial pressure (Cichowski *et al.*, 2005; Prausnitz *et al.*, 1998). Based on Henry's Law (Sander, 1999), the expression for solubility of dissolved methane can be updated as:

$$Sol_{CH_4} = \frac{P_g}{k_H(T)} = \frac{P * C_{gc} * 10^{-6}}{k_H(T)}$$
, (6)

Where,

Sol_{CH_4} = solubility of dissolved CH_4	[mol/L]
$k_{ m H}(T)$ = Henry's Law constant at temperature T (K)	[L* bar / mol]
P_{g} = partial pressure of the gas	[bar]
P = Total pressure of a gas mixture in the vial	[bar]
$C_{gc}\text{=}$ the concentration in gas phase which is measured by GC	[ppm]

And Henry's Law constant for a solute in water solution was given by (Sander, 1999):

$$k_{\rm H}({\rm T}) = k_{\rm H}^0 * \exp\left[-C * \left(\frac{1}{{\rm T}} - \frac{1}{{\rm T}^0}\right)\right] ,$$
 (7)

Where,

 k_{H}^{0} = Henry's law constant at 298.15 K (Lide and Frederikse, 1995) [714.28 L*bar/mol] T⁰ = The standard temperature [298.15 K]

CH₄ flux

The raw CH₄ flux data (in ppm / min), collected from the automatic chamber system, was calculated from the slope of concentration changes during 5 minutes in which the lip of chamber was closed. Meanwhile, a program was applied to find the best linear fit, in case the variation was not linear (Mastepanov, 2010). The conversion from ppm/min to traditional unit (mg CH₄ m⁻² h⁻¹) utilizes the ideal gas law, taking chamber volume, barometric pressure and air temperature into account.

[K]

[K]

Data selection

For the data of pore water concentration, some extreme low or high values were found in the results, which was most likely due to leakage of the vial or many bubbles were collected in the sample. The extreme values thus were excluded. Besides, the pore water concentrations measured at certain depth in one sampler, which was much higher or much lower than the concentrations at the same depth in other samplers, were excluded as well. The probable reason can be the steel sipper was not situated at right depth. For the data of stream water concentration, all of the data were used for analysis.

RESULTS

CH₄ flux

The automatic chamber system consisting of six chambers was used to monitor the exchange of CH₄. In 2011, the system was running from 26 July to 18 October. The CH₄ flux pattern in 2011 was similar to the pattern observed during the same period in the previous three years (Jensen and Rasch, 2009; Jensen and Rasch, 2010; Jensen and Rasch, 2011). The emissions in August and at the end of July were around 3 mg CH₄ m⁻² h⁻¹, and then gradually reduced to approximately 0.4 mg CH₄ m⁻² h⁻¹ (Figure 4A). During October there was tiny fluctuation that can be found at the lower right corner of Figure 4A. The temporal and spatial variation in CH₄ flux was primarily related to temperature, water table depth, net ecosystem exchange (NEE) and vegetation species (Christensen *et al.*, 2003b; Ström *et al.*, 2005; Lai, 2009).

Pore water concentration

The varying pattern of daily average concentration of dissolved CH₄ in pore water (mean of 5-35 cm depth) can be seen Figure 4B and 4C, representing summer and autumn season, respectively. According to the results of correlation analysis (Table 1), the daily mean pore water concentration of all depths (PWconAll) was significant inversely correlated with the daily mean air temperature (R = -0.961, p = 0.019) and the seasonal mean CH₄ emissions (R = -0.880, p = 0.060). An increasing trend of PWconAll during summer sampling date, corresponding with a decreasing trend of air

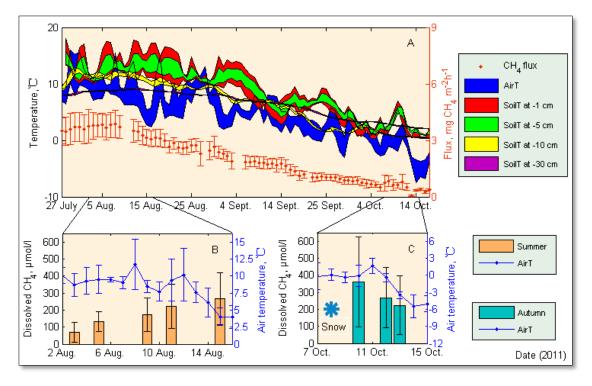


Figure 4 A. CH₄ flux, soil temperature and air temperature during sampling date. The colored area represents the range of standard deviation of the daily mean value. Soil temperatures at 1, 5, 10 and 30 cm depth are shown as red, green, yellow and violet, respectively. Air temperature at 2 m height in the study area is shown as blue. The brown dot with error bar shows daily mean fluxes over six individual chambers and the standard error of mean among the chambers. **B**. Pore water concentration and air temperature in summer. Orange bars show daily mean concentration of dissolved CH₄ over four depths (5, 15, 25 and 35 cm depth). The error bars show the range of variations among the four depths. **C**. Pore water concentration and air temperature in autumn. The features of the chart are the same as described in B. Snow mark represents the first day (8 Oct.) of snowing at study site in 2011.

temperature can be seen in Figure 4B. The reducing trend of PWconAll in early autumn is shown in Figure 4C. In 2011, the first snow in the SoilFen started on 8 Oct. and it was discontinuously snowing until 10 Oct. On 11 Oct., the air temperature was positive all the time and the mean air temperature with standard deviation measured as 1.68 ± 1.32 °C leading to snow melt. Comparing the three diagrams in Figure 4, CH₄ flux was in general showing a negative relation to PWconAll.

Depth profiles of pore water concentration from the SoilFen show that dissolved CH₄ concentration was increasing with soil depth (Figure 5). The concentrations in the study area ranged from 0 to 858 μ mol/L, while the peat depth ranged between 0 and 37.5 cm. By observing the distribution of pore water concentration with the peat depth and the curve fitting (Figure 5), we found the concentration was increasing rapidly from 0 to 15 cm depth and then stabilized in summer season. However, in autumn season, the concentration kept growing with the peat depth and had no sign of stopping or to slow down this tendency. The orange and light blue dash lines showing the estimated maximum CH₄ solubility in summer and autumn season, they were 182.64 and 239.99 μ mol/L, respectively (Figure 5). Bunsen solubility coefficients and

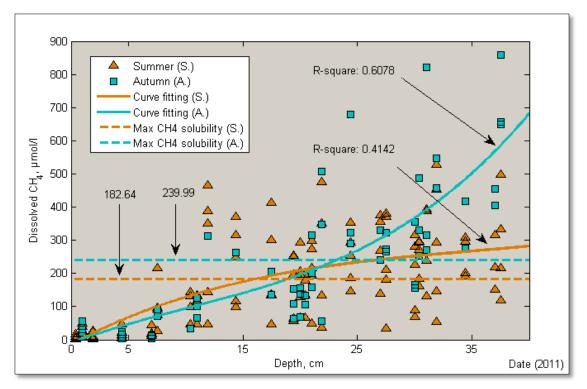


Figure 5 Dissolved CH₄ in pore water plotted against depth relative to the soil surface. Orange triangles and light blue squares represent summer and autumn samples, respectively. Curve fitting for summer and autumn data are orange and light blue solid line, respectively. Dash line shows the estimated maximum CH₄ solubility in the SoilFen, orange and light blue respectively represent summer and autumn season.

soil temperature were used to estimate maximum CH_4 solubility within the SoilFen environmental conditions. The detail of this calculation can be found in Appendix 2. The results of maximum CH_4 solubility revealed that the pore water concentrations of early autumn in general were higher than the concentrations in summer.

Stream water concentration

Dissolved CH₄ concentrations measured in the stream water around the SoilFen site can be seen in Figure 6. The bar chart in Figure 6A shows the mean dissolved CH₄ concentration in the stream water at 2 cm depth (relative to stream surface) in the summer and autumn season. The distribution of sampling plots, SW1 – SW13 and P1, can be found in Figure 1 and Figure 3. P1 was the nearest plot to the fen site, following by SW6, SW7 and so on. The results of measurement presented that high concentration of dissolved CH₄ was found in the plots close to the fen site rather than the ones that were furthest away from the fen site, the phenomena appeared in both summer and autumn season. Further, the concentrations of dissolved CH₄ in stream water close to the fen in autumn were much higher than the ones in summer (Figure 6A), i.e. summer vs autumn: 4.34 vs 14.32 (SW6), 2.42 vs 5.19 (SW7) and 0.69 vs 1.40 (SW8) µmol/L respectively. The autumn data in plot P1 was missing due to freezing of the stream surface. Figure 6B represent the daily mean stream water concentration at P1 (mean of 2-22 cm depth) in summer. Figure 6C show depth profiles of dissolved CH₄ in stream water at P1. The concentrations of dissolved CH₄

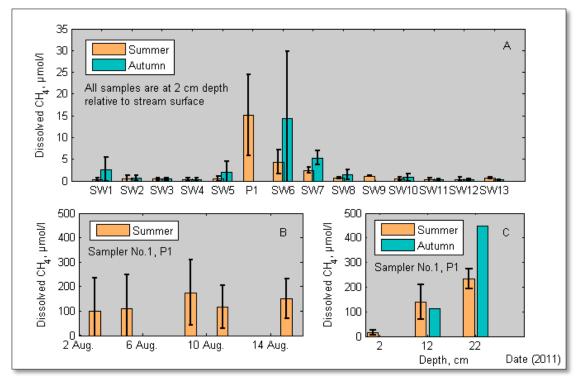


Figure 6 Stream water concentration. Orange and light blue represent summer and autumn respectively. **A.** Mean concentration of dissolved CH_4 at 2 cm depth relative to the stream water surface and standard error of mean during sampling periods (5 days in summer and 3 days in autumn). SW1-13 and P1 represent location of sampling. **B.** Daily mean concentration of dissolved CH_4 over three depths (2, 12 and 22 cm) and range in pore water sampler No.1 (P1). **C.** Mean concentration of dissolved CH_4 in stream water plotted against depth in P1. The error bars show standard error of mean between sampling periods.

in the stream water raised during the early autumn. There is no autumn data in Figure 6B and no standard deviation on autumn data in Figure 6C, due to fewer samples collected in autumn (the water in some sampling plots was frozen).

Controls of CH₄ flux and pore water concentration

In order to understand the observed CH_4 emission and pore water concentration dynamics better, a correlation analysis was performed. It seeks to explore whether there is a variable or a small group of independent variables that can explain the observed dependent variables. This correlation analysis was done using SPSS 19.0 and the results are shown in Table 1 with a complete version in Appendix 2.

The mean seasonal CH₄ flux is positively correlated to soil temperature (Table 1), and there is even significant correlations with soil temperature in all layers, i.e. with SoilT_1 (R = 0.870, p = 0.065), SoilT_5 (R = 0.897, p = 0.052), SoilT_10 (R = 0.993, p = 0.004), and SoilT_30 (R = 0.975, p = 0.013). The mean seasonal soil temperature at 10 cm depth has a similar pattern as the mean seasonal CH₄ emissions can be seen in Figure 4A. However, no correlation between CH₄ flux and water table depth were found in this project.

Table 1 The correlation matrix between the seasonal mean CH₄ flux, air temperature (AT), pore water concentrations of dissolved CH₄ (PWcon5, PWcon15, PWcon25, PWcon35 and PWconAll represent 5-, 15-, 25-, 35-cm and all depth), soil temperature (SoilT_1, SoilT_5, SoilT_10 and SoilT_30 represent 1-, 5-, 10- and 30-cm depth) and water-table (WtD). Bold numbers indicate significant correlations, ** correlation significant at the 0.01 level and * correlation significant at the 0.05 level.

		CH ₄ Flux	AT	PWcon5	PWcon15	PWcon25	PWcon35	PWconAll	SoilT_1	SoilT_5	SoilT_10	SoilT_30	WtD
CH ₄ Flux	[mg CH ₄ m ⁻² h ⁻¹]	1.000											
AT	[deg C]	0.931	1.000										
PWcon5	[µmol/L]	0.121	-0.247	1.000									
PWcon15	[µmol/L]	-0.252	-0.518	0.682	1.000								
PWcon25	[µmol/L]	-0.798	-0.912*	0.325	0.783	1.000							
PWcon35	[µmol/L]	-0.977**	-0.987**	0.090	0.427	0.888*	1.000						
PWconAll	[µmol/L]	-0.880*	-0.961**	0.251	0.682	0.988**	0.949**	1.000					
SoilT_1	[deg C]	0.870*	0.706	0.349	0.234	-0.407	-0.778	-0.544	1.000				
SoilT_5	[deg C]	0.897*	0.705	0.436	0.202	-0.449	-0.793	-0.578	0.987**	1.000			
SoilT_10	[deg C]	0.993**	0.882*	0.235	-0.148	-0.731	-0.944**	-0.823	0.905*	0.937*	1.000		
SoilT_30	[deg C]	0.975**	0.827	0.332	-0.051	-0.660	-0.904*	-0.761	0.927*	0.963**	0.994**	1.000	
WtD	[cm]	-0.412	-0.071	-0.864*	-0.743	-0.189	0.212	-0.057	-0.729	-0.757	-0.515	-0.602	1.000

High inverse correlations between pore water concentration and CH₄ flux, air temperature and soil temperature were found in this statistic analysis as well (Table 1). Further, dissolved CH₄ concentration at deep layer had significant correlations with them, i.e. PWcon35 – CH₄ flux (R = -0.977, p = 0.012), PWcon35 – AT (R = -0.987, p = 0.006), PWcon25 – AT (R = -0.912, p = 0.044), PWcon35 – SoilT_10 (R = -0.944, p = 0.028) and PWcon35 – SoilT_30 (R = -0.904, p = 0.048). Beside, other measured interdependent environmental parameters (like AT, SoilT) had expected correlations to each other as well, i.e. AT – SoilT_10 (R = 0.882, p = 0.059).

DISCUSSION

Pore water concentration

Results of the correlation analysis show that average pore water CH₄ concentration was negatively correlated with CH₄ flux and air temperature (Table 1), i.e. CH₄ flux (R = -0.880, p = 0.060) and AT (R = -0.961, p = 0.019). In summer, the low concentration was measured on the first day of sampling and then rebound during the following sampling date (Figure 4B). It was raining in the SoilFen around the first day of sampling, rainfall may flush or oxygenate water in the peat and enhance the consumption of stored CH₄, at the same time rainfall can also dilute pore water concentration and accelerate CH₄ transportation with water from the fen site to the stream. The following recovery of the concentration was likely due to increased CH_4 production under the warm soil and air conditions (Figure 4A). The increasing trend of the dissolved CH₄ concentration estimated to have been maintained reaching a peak in September, because the CH_4 production still remained at a high level while the CH_4 transport due to plant senescence was reduced during this period. For instance, the ebullition of CH₄ was reduced as falling temperature (Fechner-Levy and Hemond, 1996), and plants start wilting may result in lower plant-mediated transport (Whiting et al., 1991; Whiting and Chanton, 1992; Bubier, 1995; Chanton, 2005; Whalen, 2005).

The measured average pore water concentration in early autumn, when the ice cover started forming and the freezing front began progressing, was generally higher than in summer as seen in Figure 4C. Increased CH₄ solubility at lower temperatures and seasonal changes in gas transport mechanisms were considered as the main contribution to the seasonal variation in pore water concentrations in an earlier comparable study (Melloh and Crill, 1996). Depending on the estimation of solubility of CH₄ (Appendix 1), the solubility increased from 182.64 µmol/L at 11 °C to 239.99 μ mol/L at 2 °C representing summer and autumn soil temperatures, respectively. CH₄ produced at low rates under cold conditions most likely remain dissolved in pore water, whereas CH₄ produced at high rates at warm would more likely lead to outgassing (Melloh and Crill, 1996). On the other hand, growing surface frost lowered directly methanotrophic activities through these being closest to the temperature changes as well as through efficiently insulating oxygen from the anaerobic zone. The lower temperature would lower CH₄ ebullition (Strack et al., 2005), as well as freeze-in situations may block or reduce a number of pathways of CH₄ transportation (Chanton, 2005; Whalen, 2005). The declining CH₄ transport and reducing methanotrophic activities probably resulted in more CH₄ trapped and accumulated within the pore water, although the CH₄ production in early autumn was slowing down.

In addition, seasonal variability of subsurface pressure can be considered as an extra contribution to the higher dissolved CH_4 found during early autumn. Although the hydrostatic pressure and the partial pressure of the dissolved gases in pore water were not measured in this project, both of these pressures could be expected to have a risen as the surface started freezing. The higher hydrostatic pressure will mainly be due to the process of freezing from the surface and the higher partial pressure most likely due to the requirement of the gas bubbles forming under higher hydrostatic pressure (Chanton and Whiting, 1995). As a result, the increased hydrostatic pressure can lower the CH_4 ebullition (Fechner-Levy and Hemond, 1996), and the raised partial pressure of the dissolved gases lead to higher CH_4 solubility.

The decreasing trend of pore water concentration in early autumn (Figure 4C) is probably caused by two reasons: 1) further decreased CH_4 production as the cold climate progressed and 2) discontinuous snowmelt diluted pore water concentration that at the same time flushed stored CH_4 into the stream.

Late-season CH₄ flux and deep layer pore water concentration

In accordance with the distribution and trend of dissolved CH_4 varying with peat depth (Figure 5), the revealed pore water CH_4 concentration at deep soil layer during autumn was higher than that during summer. *Melloh* and *Crill* (1996) also observed a quite similar phenomena in a temperate peatland, where dissolved CH_4 at deep peat layer showed seasonal trends of high concentrations during winter and low concentrations during summer. The factors that contributed to the seasonal variation in deep layer pore water concentration may include increased CH_4 solubility at lower temperatures and seasonal changes in gas transport mechanisms, as discussed above.

Besides, physical factors were also considered as having an important contribution. The notable increased CH_4 concentrations in deep pore water during late-season was not only observed in this project (Figure 5), but also occurred at temperate peatland (Melloh and Crill, 1996), the freeze-in condition was a common feature of these two locations. It may imply the growing frost front causing the produced CH_4 accumulated at deep peat layer.

The correlation analysis result also presented a significant negative correlation (R = -0.977, p = 0.012) between CH₄ flux and deep layer pore water concentration (Table 1). The significant correlation was most clearly expressed during early autumn, when the lowest CH₄ emissions were measured (Figure 4A), corresponding with the highest pore water CH₄ concentration at the deepest peat layer (Figure 5). Under freeze-in conditions, low temperature and net ecosystem production (NEP) would result in a low methanogenesis rate (Joabsson and Christensen, 2001; Lai, 2009). Additionally considering the reduced CH₄ transport capacity under such conditions, a low CH₄ flux would logically be expected. However, recent studies found that large amounts of CH₄ may be released from the freezing ground of Arctic tundra during onset of soil freezing (Mastepanov *et al.*, 2008; Sturtevant *et al.*, 2011).

Although the measured soil temperature during the autumn sampling period reveal the soil had not frozen yet (Figure 4A), freezing snowmelt on the cold peatland surface had already deposited a 1-2 cm layer of ice companying with a continuously accumulated snowpack. Therefore, to a certain extent we can consider the peat surface had already frozen and the freezing front was going to propagate. The observed pattern of CH₄ flux during late-season from the Kobbefjord site this year (Figure 4A), as well as the previous three years (Jensen and Rasch, 2009; Jensen and Rasch, 2010; Jensen and Rasch, 2011), did not perform unexpected burst as occurred in Zackenberg in 2007 (Appendix 3). The thickness of active layer in Zackenberg fen site ranges between 50 and 56 cm (Mastepanov, 2010). It is most likely owing to the lack of permafrost in this subarctic peatland, which limited the pressured CH₄ and prevented it from sudden emission. The increased hydrostatic pressure at the freezing front growing would restrict CH₄ bubbles form and bubbles grow in size (Chanton and Whiting, 1995), thus the formed gas bubbles may be forced to stay and accumulate in the rhizosphere (where they were mainly produced) due to the low partial pressure of the gases compared with the raised hydrostatic pressure. In such scenario, the CH₄ transport would be dominated by molecular diffusion, the produced CH₄ could diffuse along the hydrostatic concentration gradient (Berner, 1980; Chanton, 2005; Whalen, 2005). This mechanism may be another way of understanding of increased pore water concentration at deep layer (around 30 cm depth) during the frost front propagating, and also need future study and validation.

During the late-season high arctic peatlands, which with thin active layer, thus have less space for CH_4 diffusion, the continuous accumulation of gas bubbles may lead to the rapid development of overpressure zones in the waterlogged peat. Once the threshold pressure level is reached, the accumulated gas bubbles will be suddenly ejected into the atmosphere.

Certainly, CH_4 burst in low arctic peatland during late autumn or winter may be possible in theory. Recently, because of technical limitation, it was difficult to keep CH_4 measuring instruments working under harsh arctic winter conditions. Therefore, the CH_4 flux pattern in both low and high arctic during the later autumn and winter remain uncertain.

Measured pore water concentration higher than estimated CH₄

solubility

The original intention of estimating the maximum CH₄ solubility was to examine the seasonal variation of CH₄ solubility at the study site and verify the accuracy of dissolved CH₄ measurements in this project. The results of the dissolved CH₄ measurements in SoilFen showed that, the measured pore water CH₄ concentrations beyond the estimated maximum CH₄ solubility were quite common and they appear in both summer and autumn samples (Figure 5). It is widely known that the solubility of CH₄ is very low, so that it was easily saturated in the water and exited as bubbles among water before emitted to the atmosphere. The most likely reason for the measured high concentrations was some tiny bubbles hidden in the samples and impossible to avoid during the procedure of sampling. This was in particular a problem during the autumn sampling period. In order to prevent the water in the tygon tubing and syringe from freezing, the speed of operation of sampling was faster in autumn, and more likely lead to tiny bubbles remaining in the samples. On the other hand, the accuracy of calculating maximum CH₄ solubility was considered as the second factor contributing to the measured high concentrations. Owing to time and technical limitation, the average salinity and partial pressure (mean of 0-35 cm depth) in the SoilFen were assumed as 0 ppt and 1.013 bar, respectively, though both of them may be underrated comparing with the reality, especially the partial pressure. Therefore, the estimated maximum CH₄ solubility can be underestimated in this project.

Stream water concentration

The measured dissolved CH₄ concentration in the stream was gradually falling with distance away from the SoilFen (Figure 6A). This indicated strongly the main source of dissolved CH₄ in stream water was derived from the fen site, and progressively consumed (or oxidized) and emitted into the atmosphere during transporting with stream. The results also show the higher stream water concentration in early autumn, notably in the plots nearby the fen. It implied that CH₄ in the soil most likely would be squeezed into the closed stream under high pressure as frost front growing. This further exposes that a peatland with high water mobility (such as located next to stream, river and lake), regardless of situating in high or low arctic, will see a high export through stream in the autumn. The measured concentration in the pore water (Figure 5) and stream water of P1 which was closed to fen site (Figure 6C), might be able to act as an indicator of seasonal dissolved CH₄ dynamics and export from in the peatland.

Limitations, accuracy and future work

This study has some limitations. One constraint was the limited amount of samples that can be collected in autumn due to the cold climate, thus large deviations were found in the autumn measurements. Secondly, leakage testing of the vials storing water samples was involved in this project, however, it was failed at last due to an inappropriate choice of the standard CH_4 in the procedure of preparation. Therefore, all of the results might be underestimating the real concentrations in the event the quality of the vial is not good. Third, the correlation analysis performed in this project, was an excellent method to predict the relationship among factors. However, at the same time it required a number of data to support this kind of statistical analysis, eight-days sampling data might not be enough to get reasonable results. During the process of sampler installation, some peat was taken out and environment might be destroyed a little bit thus, first two days data of pore water concentration might remain underestimated. In addition, human manual operations throughout all of the sample collection analysis brought an element of inaccuracy.

In future work, extra parameters might be considered as well, i.e. pressure, salinity, acidity, vegetation species and snow depth. The hydrostatic pressure beneath the surface and the partial pressure of gas bubble at different peat layers can influence the dissolved CH_4 concentration in the different peat layers. Salinity and acidity of pore water affect the solubility of CH_4 , and further dissolved CH_4 concentration. Different vegetation species in peatland carry a different capacity of CH_4 in the peat. For snow depth, on one hand snowmelt would flush CH_4 in peatland into the nearby stream, on the other hand snow accelerate ice cover and snowpack form, which will lead to high pressure beneath the ground.

CONCLUSIONS

The focus of this study was on the dissolved CH_4 dynamics in a subarctic peatland ecosystem in Kobbefjord, West Greenland. A pore water sampling method was used to collect samples at four depths (5-, 15-, 25- and 35-cm depth) in the fen site. Stream water samples were collected from 13 plots around the study site. The dissolved CH_4 concentration was calculated from the extraction derived from 2-ml samples and the solubility of CH_4 , corrected by air temperature and pressure.

The seasonal variation of dissolved CH_4 in this peatland, was most likely a result of seasonal changes in temperature and CH_4 transport mechanisms. Pore water CH_4 concentration increased as the ground started freezing, suggesting that the frozen ground and low temperature may play an important role in dissolved CH_4 rising in pore water during early autumn. The increased stream water concentration near the fen site and the significant higher concentration of dissolved CH_4 at the deep peat layer during the late-season, further demonstrated that the freezing ground not only affected the CH_4 transport mechanisms, but also may have a physical effect on the dissolved CH_4 changes.

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APPENDICES

Appendix 1. The procedure of calculating maximum CH₄ solubility

In accordance with previous study (Yamamoto *et al.*, 1976), we hypothesize that the concentration of CH_4 gas phase in peat water is 99.99% purity under both summer and autumn conditions. The estimation of maximum CH_4 solubility can be made by using the soil temperatures and salinity. The average soil temperature (mean of 0 - 30 cm depth) in summer (from 29 Jul. to16 Aug., 2011) and autumn (from 7 Oct. to 14 Oct., 2011) was 11°C and 2°C, respectively. The average salinity of peat water in summer and autumn is unknown, assumed 0 for them.

Bunsen solubility coefficient is one of the best predictors of the solubility of gases in liquids, β , defined as the volume of gas (corrected to standard temperature and pressure) is absorbed per unit volume of water at a specified temperature when the partial pressure of the gas is 1.013 bar (Yamamoto *et al.*, 1976; Dickerson *et al.*, 1979). The function represents Bunsen solubility coefficient respect to temperature and salinity as follows (Yamamoto *et al.*, 1976):

$$\ln \beta = A_1 + A_2 \left(\frac{100}{T}\right) + A_3 \ln \left(\frac{T}{100}\right) + S[B_1 + B_2 \left(\frac{T}{100}\right) + B_3 \left(\frac{T}{100}\right)^2]$$

Where A and B are constants,

 A_1 = -67.1962 A_2 = 99.1624 A_3 = 27.9015 B_1 = -0.072909 B_2 = 0.041674 B_3 = -0.0064603 T = soil temperature S = salinity

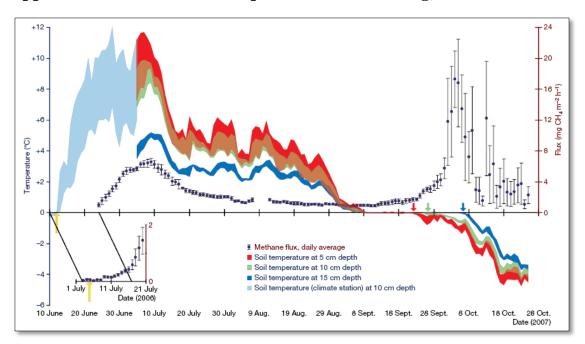
[K] [parts per thousand, ppt]

The results show β is 0.04252 and 0.05412 CH₄ ml/ml water for CH₄ solubility of pore water in summer and autumn, respectively. According to CH₄ gas density at the measurement temperature 11 °C and 2 °C, under the standard pressure of 1.013 bar, are 0.6895 and 0.7122 mg/ml. The results are rewritten as 0.0293 and 0.0385 (CH₄ mg/g water, 99.99% purity) in summer and autumn occasion, respectively. To convert into the traditional unit in µmol/L, they are expressed as 182.64 and 239.99 µmol/L, respectively.

			CH₄ Flux	AT	PWcon5	PWcon15	PWcon25	PWcon35	PWconAll	SoiIT_1	SoilT_5	SoiIT_10	SoilT_30	WtD
Correlation	CH₄ Flux	[mg CH ₄ m ⁻² h ⁻¹]	1.000	.931	.121	252	798	977	880	.870	.897	.993	.975	412
	AT	[deg C]	.931	1.000	247	518	912	987	961	.706	.705	.882	.827	071
	PWcon5	[umol/L]	.121	247	1.000	.682	.325	.090	.251	.349	.436	.235	.332	864
	PWcon15	[umol/L]	252	518	.682	1.000	.783	.427	.682	.234	.202	- .148	051	743
	PWcon25	[umol/L]	798	912	.325	.783	1.000	.888	.988	407	449	731	660	189
	PWcon35	[umol/L]	977	987	.090	.427	.888	1.000	.949	778	793	944	904	.212
	PWconAll	[umol/L]	880	961	.251	.682	.988	.949	1.000	544	578	823	761	057
	SoilT_1	[deg C]	.870	.706	.349	.234	407	778	544	1.000	.987	.905	.927	729
	SoilT_5	[deg C]	.897	.705	.436	.202	449	793	578	.987	1.000	.937	.963	757
	SoilT_10	[deg C]	.993	.882	.235	148	731	944	823	.905	.937	1.000	.994	515
	SoilT_30	[deg C]	.975	.827	.332	051	660	904	761	.927	.963	.994	1.000	602
	WtD	[cm]	412	071	864	743	189	.212	057	729	757	515	602	1.000
Sig. (1-tailed)	CH₄ Flux	[mg CH₄ m ⁻² h ⁻¹]		.034	.440	.374	.101	.012	.060	.065	.052	.004	.013	.294
	AT	[deg C]	.034		.376	.241	.044	.006	.019	.147	.148	.059	.086	.464
	PWcon5	[umol/L]	.440	.376		.159	.338	.455	.374	.326	.282	.383	.334	.068
	PWcon15	[umol/L]	.374	.241	.159		.108	.287	.159	.383	.399	.426	.474	.129
	PWcon25	[umol/L]	.101	.044	.338	.108		.056	.006	.296	.275	.135	.170	.405
	PWcon35	[umol/L]	.012	.006	.455	.287	.056		.025	.111	.104	.028	.048	.394
	PWconAll	[umol/L]	.060	.019	.374	.159	.006	.025		.228	.211	.089	.120	.472
	SoilT_1	[deg C]	.065	.147	.326	.383	.296	.111	.228		.006	.048	.037	.136
	SoilT_5	[deg C]	.052	.148	.282	.399	.275	.104	.211	.006		.031	.018	.122
	SoilT_10	[deg C]	.004	.059	.383	.426	.135	.028	.089	.048	.031		.003	.242
	SoilT_30	[deg C]	.013	.086	.334	.474	.170	.048	.120	.037	.018	.003		.199
		[cm]	201	181)))	100	105	200	011	136	200	242	100	

Table 2 The completed correlation matrix with significant level. The table shows correlations between the seasonal mean CH4 flux, air temperature (AT), pore water

Appendix 2. Completed version of correlation matrix



Appendix 3.The methane flux pattern in Zackenberg, NE Greenland

The diagram above presented full-season methane flux and soil temperature patterns at Zackenberg, 2007. Source: Mastepanov et al., 2008 (figure 1).

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