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## Land-atmosphere exchange of methane from soil thawing to soil freezing in a high-Arctic wet tundra ecosystem

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#### Abstract

The land-atmosphere exchange of methane (CH<sub>4</sub>) and carbon dioxide (CO<sub>2</sub>) in a high-Arctic wet tundra ecosystem (Rylekærene) in Zackenberg, north-eastern Greenland, was studied over the full growing season and until early winter in 2008 and from before snow melt until early winter in 2009. The eddy covariance technique was used to estimate CO<sub>2</sub> fluxes and a combination of the gradient and eddy covariance methods was used to estimate CH<sub>4</sub> fluxes. Small CH<sub>4</sub> bursts were observed during spring thawing 2009, but these existed during short periods and would not have any significant effect on the annual budget. Growing season CH<sub>4</sub> fluxes were well correlated with soil temperature, gross primary production, and active layer thickness. The CH<sub>4</sub> fluxes remained low during the entire autumn, and until early winter. No increase in CH<sub>4</sub> fluxes were seen as the soil started to freeze. However, in autumn 2008 there were two CH<sub>4</sub> burst events that were highly correlated with atmospheric turbulence. They were likely associated with the release of stored CH<sub>4</sub> from soil and vegetation cavities. Over the measurement period, 7.6 and 6.5 g C m<sup>-2</sup> was emitted as CH<sub>4</sub> in 2008 and in 2009, respectively. Rylekærene acted as a C source during the warmer and wetter measurement period 2008, whereas it was a C sink for the colder and drier period of 2009. Wet tundra ecosystems, such as Rylekærene may thus play a more significant role for the climate in the future, as temperature and precipitation are predicted to increase in the high-Arctic.

*Keywords:* Micrometeorology, eddy covariance, gradient method, methane, carbon balance, tundra, climate change, land–atmosphere interactions

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#### Introduction

Approximately 16% of the global soil area is underlain by permafrost (Tarnocai *et al.*, 2009). Ecosystems in these areas are vulnerable to the effects of predicted global climate change, and a range of possible feedback mechanisms to climate could arise from these ecosystems (Chapin *et al.*, 2000). Global warming is not evenly distributed over the world, and especially northern high-latitude areas are projected to see a larger increase in temperature, precipitation, and growing season length than the rest of the world (ACIA, 2005; IPCC, 2007).

Northern wet tundra ecosystems are characterized by waterlogged, anaerobic, and cold conditions, which effectively reduce decomposition rates and favor the formation of peat. The net carbon (C) emission from these ecosystems is the balance between the carbon dioxide  $(CO_2)$  absorbed by gross primary production

(GPP), and the C decomposed and released as  $CO_2$  or methane (CH<sub>4</sub>). During the Holocene, these areas acted as sinks for C, and approximately 50% of the global soil organic C pool is today stored in the northern permafrost regions (Smith *et al.*, 2004; McGuire *et al.*, 2009; Tarnocai *et al.*, 2009). As the permafrost areas are thawing (Serreze *et al.*, 2000), previously frozen C becomes available for decomposition, and could have a major impact on the global C cycle (Tarnocai, 2006).

The waterlogged and anaerobic conditions also make northern wetlands one of the largest natural sources of atmospheric  $CH_4$  (Whalen, 2005). A change in this major  $CH_4$  source would have significant effects on the greenhouse gas budgets of these areas. As concerns regarding the future climate change grow, there is an increasing need to quantify the greenhouse gas budget of global ecosystems. There are still large uncertainties regarding the  $CH_4$  source strength of wet tundra ecosystems, and its relationship to environmental variables. For example, Mastepanov *et al.* (2008) showed unexpectedly high  $CH_4$  fluxes during the refreezing of the active layer. Whalen & Reeburgh (1988), showed

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similar autumn  $CH_4$  fluxes for areas covered with mosses in Alaska where 40% of the annual fluxes were released during the freeze-in period.

Most CH<sub>4</sub> flux studies in high-latitude ecosystems have been based on the closed-chamber technique (e.g., Whalen & Reeburgh, 1988; Joabsson & Christensen, 2001; Corradi et al., 2005; Mastepanov et al., 2008). However, chamber-based measurements decouple the surface from the atmosphere, and the influence of , for example, atmospheric turbulence on the CH<sub>4</sub> fluxes can hereby not be studied. Net exchange of greenhouse gases between the ground surface and the atmosphere can be measured at a scale of hectares to km<sup>2</sup> with micrometeorological methods like the eddy covariance technique (EC) or in lack of fast response instruments, by using gradient methods. Only a few studies have reported CH<sub>4</sub> flux measurements with these methods from high-Arctic wet tundra ecosystems (Friborg et al., 2000; Sachs et al., 2008; Wille et al., 2008).

The high-Arctic region constitutes approximately 34% of the non-glaciated Arctic area (Walker et al., 2005) and is defined as areas where the average air temperature of the warmest month is below 7 °C (Bliss, 1997; Walker et al., 2005). The spatial coverage of wetlands north of 45° is  $3.72 \times 10^6$  km<sup>2</sup>, whereof 62% is underlain by permafrost (Avis et al., 2011). The primary aim of this study was to use micrometeorological methods to investigate ecosystem-scale CH<sub>4</sub> and CO<sub>2</sub> fluxes in a high-Arctic wet tundra ecosystem from spring thaw, throughout the growing season, and into the freeze-in and early winter period during 2008 and 2009. As study site, representative of the non-glaciated high-Arctic areas underlain by permafrost, we selected Rylekærene a wet area situated in the Zackenberg valley, Northeast Greenland. A long time series of CH4 fluxes in combination with long term data of other ecosystem properties, that is, CO2 fluxes and environmental conditions, will allow us to assess the seasonal behavior of CH<sub>4</sub> fluxes, and the influence of ecosystem properties. Subsequently, a second aim was to identify how well these ecosystem properties determine CH<sub>4</sub> fluxes at an ecosystem level. In addition, our measurements of both CH<sub>4</sub> and CO<sub>2</sub> fluxes allowed us to estimate C budgets for the land-atmosphere exchange of the measured area, and a final aim was to investigate inter-annual variation in these budgets.

#### Materials and methods

#### Site description

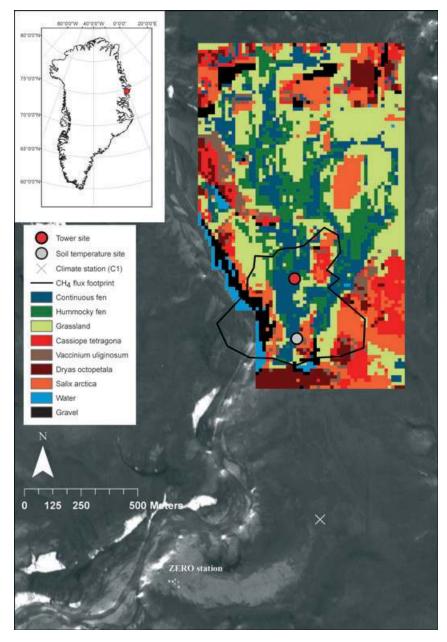
The field measurements were carried out in the fen area, Rylekærene, in the Zackenberg valley (74°28'N 20°34'W), situated in the Northeast Greenland National Park. The average air temperature of the warmest month in the valley is 5.8 °C, and the mean annual temperature is -9 °C (Hansen et al., 2008). The snow melt usually begins in late May and ends in mid-June. Rylekærene is a typical high-Arctic heterogeneous wetland area. It is a patterned fen characterized by alternating high, dry heath areas, and low, wet fen areas. During a field inventory in 2007, the dominant plant communities were registered for every 15 m<sup>2</sup> within a 1.4 km<sup>2</sup> area covering Rylekærene (Fig. 1) (Tagesson, 2011). The dominant plant communities were fen, grassland (dominated by Arctagrostis latifolia, Eriophorum triste, and Alopecurus alpinus), Cassiope tetragona heath, Dryas octopetala heath, Vaccinium uliginosum heath and Salix arctica snowbed (Fig. 1) (Tagesson, 2011). The fen areas were further divided into continuous fen (flat areas dominated by Eriophorum scheuchzeri, Carex stans and Dupontia psilosantha) and hummocky fen (hummocks dominated by E. triste, S. arctica and A. latifolia) (Bay, 1998). The moss species at the site was dominated by true mosses, for example, Tomenthypnum, Scorpidium, Aulacomnium and Drepanoclaudus. The maximum active layer thickness in the valley ranges between 0.40 m at the fen to about 0.80 m at Dryas sites (Elberling et al., 2008). The peat layer in the fen is 0.20-0.30 m thick (Christensen et al., 2000). There were mainly mineral soils below the peat and in the remaining plant communities. The pH of the fen pore water at 10 cm peat depth was  $6.9 \pm 0.2$  (n = 50; Ström, unpublished results). There have been extensive ecological, biogeographic, climatic, and hydrological research and monitoring carried out in the Zackenberg research area since 1995 (Meltofte et al., 2008).

#### Turbulence and CO<sub>2</sub> flux measurements

A 3 m tower was placed in the center of Rylekærene so that fen areas were located in the main upwind direction (Fig. 1). The study took place between 25 June and 28 October 2008, and between 16 May and 24 October 2009. An EC system consisting of a 3-axis sonic anemometer (Metek, Gmbh, Elmshorn, Germany), and an open-path  $CO_2/H_2O$  infrared gas analyzer (LI-7500: LI-COR Inc, Lincoln, NE, USA) was installed at 3.3 m above the surface. The gas analyzer was tilted 32° from vertical next to the sonic anemometer. This system provided both the direct  $CO_2$  fluxes, and the turbulence data (friction velocity and stability) necessary for the further  $CH_4$  flux calculations. The  $CO_2$  flux calculations followed standard EC methods (Kaimal & Finnigan, 1994).

The sonic anemometer and gas analyzer data were sampled at 20 Hz, but the statistics were calculated for 30 min periods. The raw data were screened for spikes four times with a running mean of 100 values (Hojstrup, 1993). All velocity and temperature values that deviated from the running mean more than 4.5, 5.0, 5.5 and 6.0 times the running standard deviation were removed, for each run respectively. These factors were determined empirically to give an optimum despiking. Higher factors (8.0, 8.5, 9.0 and 9.5) were needed for the  $CO_2$  concentration data. Linear detrending was applied to all data, and the 2D rotation to wind velocity data (Kaimal & Finnigan, 1994). Data were filtered for fully developed turbulent conditions according to Foken (2008) using the integral

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**Fig. 1** The Zackenberg valley and the investigation area surrounding the Zackenberg research station (ZERO). A field-inventory map of the dominant plant communities observed in Rylekærene in July 2007 (Tagesson, 2011) is put on top of an IKONOS satellite image from 23 July 2000. The red mark on the Greenland map is the location of Zackenberg.

turbulence characteristics.  $CO_2$  concentration data were filtered for steady-state conditions following Vickers & Mahrt (1997), with an empirically determined threshold value of 0.02. In addition, all half-hourly values with a spike removal of more than 1% were rejected (Foken, 2008). The  $CO_2$  data were additionally filtered according to a diagnostic value from the LI-7500, indicating problems caused by precipitation, icing, fog, and dirt on the sensor. In total, 43% of the combined  $CO_2$  flux and turbulence data were filtered away. Schotanus corrections were applied on the sensible heat fluxes (Schotanus *et al.*, 1983). Consequently, the CO<sub>2</sub> fluxes were Webb corrected (Webb *et al.*, 1980), which added between -5 and 10  $\mu$ mol CO<sub>2</sub> m<sup>-2</sup> s<sup>-1</sup> (on average 0.64  $\mu$ mol CO<sub>2</sub> m<sup>-2</sup> s<sup>-1</sup>). The CO<sub>2</sub> fluxes were also corrected for heating of the openpath sensor itself according to Burba *et al.* (2008) (on average 1.14  $\mu$ mol CO<sub>2</sub> m<sup>-2</sup> s<sup>-1</sup>). The correction for heating of the open-path sensor was originally designed for vertically mounted sensors. However, no correction is available for sensors mounted at an angle. The frequency losses were estimated by comparing the average of 450 half-hour co-spectra

of the vertical wind and the concentration of CO<sub>2</sub> against the average co-spectra of the vertical wind and the temperature. No transfer functions were applied for compensating for frequency losses (see below). Totally, all applied corrections added on average 1.77  $\mu$ mol CO<sub>2</sub> m<sup>-2</sup> s<sup>-1</sup>.

#### Methane flux measurements

We lacked a fast response instrument necessary for the EC technique to be applicable, and the CH<sub>4</sub> fluxes were thus estimated by combining gradient and EC methods. The CH<sub>4</sub> concentrations were measured at two levels (0.70 and 2.75 m) on the tower at 1 Hz rate. The system consisted of a laser off-axis integrated cavity output spectroscopy analyzer (LGR; DLT200, Fast Methane Analyzer, repeatability 1 ppb at 0.1 Hz, Los Gatos Research, Mountain View, CA, USA). Air was pumped at a rate of 4 L min<sup>-1</sup> with two membrane pumps (THOMAS EMS3025-63ri, Sheboygan, WI, USA) from the two heights through two 16 m long high-density polyethylene tubes (inner diameter 4 mm). The air from the two different heights was first passed through two 5 L mixing chambers to minimize fluctuations of CH<sub>4</sub> concentration. Subsamples were taken after the mixing chambers and entered the LGR at a rate of 0.4 L min<sup>-1</sup>. The measurements were switched between the two levels every 5 min. The first 1/2 minute was discarded and remaining 4.5 min of 1 Hz data were averaged. For the flux calculations, average and standard deviation for the concentrations over the 30 min periods were calculated. Data were filtered for steady-state conditions with an empirically determined standard deviation threshold value of 20 ppb. In total, 49% of the combined CH<sub>4</sub> concentrations and turbulence data were filtered away.

According to Monin–Obukhov similarity theory vertical turbulent fluxes ( $F_{CH4}$ ) can be calculated as

$$F_{\text{CH}_4} = -\rho \frac{\kappa u_* (C_{\text{CH}_{4, z_2}} - C_{\text{CH}_{4, z_1}})}{\ln(z_2/z_1) - \psi_H(\zeta)_{z_2} + \psi_H(\zeta)_{z_1}}$$
(1)

where  $\kappa$  (0.4) is the von Kármán constant,  $u_*$  is the friction velocity,  $C_{CH4,z1}$  is the CH<sub>4</sub> concentration at 0.7 m and  $C_{CH4,z2}$ 

is the CH<sub>4</sub> concentration at 2.75 m,  $z_1$  is the 0.7 m measurement height,  $z_2$  is the 2.75 m measurement height and  $\psi(\zeta)$  is the integrated form of the universal function (Foken, 2008). There are few studies with universal functions for trace gases, and the functions for sensible heat (*H*) is widely used (Högström, 1988). The CH<sub>4</sub> concentrations at  $z_1$  and  $z_2$  were measured and  $u_*$  and *H* were obtained by the sonic anemometer measurements.

The footprint of the CH<sub>4</sub> flux measurements was estimated by a model that combines a Lagrangian stochastic dispersion model and similarity theory (Hsieh *et al.*, 2000). The model relates footprint to atmospheric stability, measurement height, and surface roughness length. The measurement height was set to the arithmetic mean for stable conditions, and the geometric mean for unstable conditions (Horst, 1999). Surface roughness length was set to 0.01 m (Wieringa, 1993). The Obukhov length was given by the sonic anemometer measurements. The 80% cumulative flux distance was on average 260 m, and the average point of maximum contribution was 22 m from the tower. The average 80% cumulative flux distance is shown in Fig. 1. The fractions of different plant communities for different wind directions are included in Table 1.

#### Environmental variables

Soil temperature at 0.05, 0.10 and 0.15 m depth were measured (Tinytag Plus, Gemini Data Loggers, Chichester, UK) every 5 min at a site 265 m south of the tower (Fig. 1). In addition, soil temperature at a depth of 0.02 m ( $T_{0.02}$ ) was measured every 10 min at the tower site using Copper-Constantan thermocouples. Photosynthetic active radiation (PAR) was measured with a JYP-1000 sensor (SDEC, Tauxigny, France). Soil gas pressure profiles relative to the atmospheric pressure (24PCEFA6G, Honeywell S&C, Morristown, NJ, USA) were measured at four plots in the surroundings of the tower at 0.10, 0.20, 0.30 and 0.40 m depth from the 20 July until the soil temperature dropped down to 0 °C in 2008. No pressure sensors were used during 2009. Some of the sensors broke during the field season, and only data from two sensors at 0.10 m, one sensor at 0.20 m, four sensors at 0.30 m, and three sensors

**Table 1** Fractions of the plant communities in the CH<sub>4</sub> flux footprint area of the tower. Quadrant 1 is between  $315^{\circ}$  and  $45^{\circ}$ , quadrant 2 is between  $45^{\circ}$  and  $135^{\circ}$ , quadrant 3 is between  $135^{\circ}$  and  $225^{\circ}$  and quadrant 4 is between  $225^{\circ}$  and  $315^{\circ}$ . The fractions for the dominant wind direction  $\pm 20^{\circ}$  for the growing season and the autumn and early winter periods are also included

Plant community	Entire footprint	Quadrant 1	Quadrant 2	Quadrant 3	Quadrant 4	Dominant wind direction growing season	Dominant wind direction autumn/ early winter
Continuous fen	0.25	0.40	0.23	0.28	0.14	0.38	0.49
Hummocky fen	0.11	0.23	0.13	0.07	0.09	0.05	0.26
Grassland	0.25	0.23	0.18	0.21	0.37	0.27	0.22
Cassiope tetragona	0.07	0.03	0.15	0.07	0.03	0.06	0.02
Vaccinium uliginosum	0.02	0.02	0.01	0.03	0.00	0.02	0.01
Dryas octopetala	0.02	0.00	0.00	0.05	0.01	0.04	0.00
Salix arctica	0.16	0.07	0.30	0.14	0.13	0.11	0.00
Water	0.04	0.00	0.00	0.05	0.09	0.01	0.00
Gravel	0.08	0.01	0.00	0.10	0.14	0.06	0.00

at 0.40 m depth were used in the final analysis. The soil gas pressures were corrected for temperature effects on the sensors.

All sensors were connected to a CR-1000 data logger in combination with a multiplexer (Campbell scientific Inc., North Logan, UT, USA). Data were sampled every 30 s, and stored as 10 min averages.

Active layer thickness (AL) and water table depth (WtD) were measured manually during 15 occasions at 15 plots around the tower in 2008. Active layer thickness was measured at five plots and WtD at one plot during 33 occasions in 2009. Precipitation and snow depth were measured at the Climate station (C1) in Fig. 1 (ClimateBasis, 2010).

#### Data handling

Partitioning of CO<sub>2</sub> fluxes into GPP and ecosystem respiration. The measured net ecosystem exchange (NEE) of CO<sub>2</sub> from the growing season was partitioned into GPP, and ecosystem respiration (ER). The growing season was defined to start the day snow depth exceeded 0.10 m and end the first of two consecutive days with T<sub>0.02</sub> below 0 °C (Tamstorf et al., 2007). The 2008 growing season data were divided into 4 day data sets, except for the last period, which got 7 days. For 2009, the data were divided into 4 day data sets, except for the beginning (1-12 June 2009) and end of the growing season (after the 19 August), which were divided into 12 and 9.5 day intervals, respectively. The reason for the longer periods in the beginning and the end of the growing seasons was that GPP constitute a small fraction of the measured data and it was hereby harder to separate it from ER. A light response curve was fitted to each data set:

$$NEE = -(F_{csat} + R_d)(1 - e^{\frac{-\alpha(PAR)}{F_{csat} + R_d}}) + R_d$$
(2)

where  $F_{csat}$  is CO<sub>2</sub> flux at light saturation,  $R_d$  is the ecosystem dark respiration, and  $\alpha$  is initial slope of the light response curve (Falge *et al.*, 2001). By subtracting  $R_d$ , the curve was forced through zero, and GPP could hereby be estimated:

$$GPP = -(F_{csat} + R_d)(1 - e^{\frac{-\alpha(IAK)}{F_{csat} + R_d}})$$
(3)

Ecosystem respiration was calculated by subtracting the modeled GPP from the measured NEE values. The light response curves were well fitted for all periods, except the 1–12 June 2009. For partitioning of this period, an exponential regression was fitted with ER against air temperature, with data measured 7 days before, and after this period. Ecosystem respiration for the 1 June to 12 June 2009 was estimated using the regression on the air temperature data set, and GPP was estimated by subtracting modeled ER from measured NEE.

The budgets of the land-atmosphere exchange of carbon. For calculation of the  $CH_4$  and  $CO_2$  budgets, the time series needed to be gap-filled. Gaps were filled from 24 June to 31 October 2008, and 16 May to 31 October 2009. Gaps in data originated from, (1) the filtering described above, (2) a late start-up during the growing season 2008, (3) the measurements were stopped before the 31 October 2008 and 2009, (4)

power failures, (5) technical issues, and (6) a polar bear attack. The gaps in the  $CH_4$  fluxes were 57%, the gaps in GPP were 7%, and the gaps in ER were 49% of the full time series. Gaps that were shorter than or equal to 2 h were gap-filled by linear interpolation between the two neighboring values. Remaining gaps in the  $CH_4$  fluxes that were shorter than 24 h were set to daily averages. Gaps in daily average  $CH_4$  fluxes during the growing season were filled with a linear regression model against  $T_{0.02}$ , whereas outside the growing seasons, they were filled by linear interpolation between daily averages the days before, and after the gap. For the remaining gaps in the GPP and ER time series, a mean diurnal value of a 7 day gliding window was used (Falge *et al.*, 2001).

The errors in the CO<sub>2</sub> flux budgets were estimated following Aurela *et al.* (2002). These estimates do not cover all error sources, but gives an estimate of the key uncertainties in the flux measurements. Aurela *et al.* (2002) used closed-path EC measurements. The errors associated with frequency losses were thus estimated by comparing the average half-hour cospectra of the vertical wind and the concentration of CO<sub>2</sub> against the average co-spectra of the vertical wind and the temperature for five different groups with 90 half-hour values each.

The random errors in the CH<sub>4</sub> flux measurements were calculated by setting a measurement error of u<sub>\*</sub> ( $E_{u^*}$ ) to 0.05 m s<sup>-1</sup>, an error of 10 W for *H* ( $E_H$ ) (Foken, 2008) and an error of 1 ppb for the CH<sub>4</sub> concentration difference between 0.7 and 2.75 m ( $E_{\Delta CH4}$ ) (Los Gatos Research). The errors in CH<sub>4</sub> flux budgets associated with the gap filling ( $E_{gf}$ ) and the filtering of the measured data ( $E_f$ ) were estimated following Aurela *et al.* (2002). Finally all errors ( $E_t$ ) were added together using the error accumulation principle:

$$E_{\rm t} = \sqrt{E_{\rm u_*}^2 + E_{\rm H}^2 + E_{\rm ACH_4}^2 + E_{\rm gf}^2 + E_{\rm f}^2} \tag{4}$$

Seasonal dynamics of  $CH_4$  fluxes. We divided the measurement periods into three different seasons, based on the stages of physical and biological conditions; (i) spring, (ii) growing season, and (iii) autumn and early winter. As stated above the growing season was defined as ranging from the day the snow depth exceeded 0.10 m until the first of two consecutive days with soil temperature at 0.02 m depth below 0 °C (Tamstorf *et al.*, 2007). Autumn and early winter was defined to start the day after the end of the growing season until the 31 October. Spring 2009 was set to start on 16 May, that is, date of the first CH<sub>4</sub> flux measurements and end when the growing season started. Dates for the different seasons are given in Table 2.

**Table 2** The three different seasons in which the measurement periods were divided into for 2008 and 2009

Season	2008	2009
Spring	–	16 May–31 May
Growing	24 June–16	1 June–6
season	September	September
Autumn and	17 September–31	7 September–31
early winter	October	October

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The relationships between daily average CH<sub>4</sub> fluxes and daily average soil temperatures at the different depths, GPP, WtD, AL, and friction velocity were tested by fitting linear and exponential regressions. Data from the gaps in the daily average CH<sub>4</sub> fluxes were not used in the analysis against environmental variables, as they were not true measured data. An exponential regression between half-hourly CH<sub>4</sub> fluxes and friction velocity measured after soil was completely frozen 2008 was also fitted.

*Diurnal dynamics of*  $CH_4$  *fluxes.* We analyzed the different seasons for diurnal variation. The  $CH_4$  fluxes were normalized by dividing each half-hourly flux value with the mean daily flux. Only days with more than 70% data coverage were used in the analysis (in total 29 and 27 days for 2008 and 2009, respectively).

To avoid effects of the seasonal variation, 1 week data sets at the peak of the growing seasons were used to study variation in half-hourly  $CH_4$  fluxes. The data sets were from 26 July to 1 August 2008 and from 6 July to 12 July 2009. The variation in half-hourly  $CH_4$  fluxes during these periods was analyzed by fitting exponential and linear regressions against half-hour values of soil temperature at the different depths, GPP, fraction of emissions from the different plant communities in the footprint and friction velocity.

#### Results

#### Environmental conditions

Average air temperature during June-August was 6.9 °C in 2008 and 5.0 °C in 2009. Both years were above the long-term average of 4.6 °C since the start of the measurements in 1996 (ClimateBasis, 2010). The AL reached its maximum (~0.50 m) in the beginning of August both years (Figs 2c and 3c). Maximum snow depths were 1.30 and 0.17 m in the winters preceding the growing seasons 2008 and 2009, respectively. The area became snow free on 24 June 2008 and 0 n 1 June 2009. It rained 161 and 117 mm in 2008 and 2009, respectively. The difference in temperatures and precipitation lead to different hydrological conditions, and the water table was closer to the soil surface in 2008 (deepest WtD: -0.03 m) than in 2009 (deepest WtD: -0.10 m) (Figs 2d and 3d).

There were no changes in the soil gas pressures until the soil froze at any of the depths (Fig. 2f). After the soil started to freeze the 23 September 2008, there was a strong increase in soil gas pressure at all depths, gradually increasing as the frozen ground advanced downwards.

#### Diurnal variation in CH<sub>4</sub> fluxes

There were large variations in half-hourly CH<sub>4</sub> fluxes, but no systematic diurnal variation could be seen for

any of the seasons 2008 or 2009. Normalized halfhourly data from the growing seasons 2008 and 2009 are shown in Fig. 4. Shorter periods within the different seasons were also studied, but no diurnal variation was seen in these periods either.

None of the environmental factors gave any good degree of explanation to the variation in half-hourly CH<sub>4</sub> fluxes during the peak of the growing season. The fraction of emission that derived from continuous fen was the factor that best explained the variation in half-hourly CH<sub>4</sub> fluxes (linear correlations: 2008: *P*-value = 0.0002,  $R^2$ =0.17, n = 80; 2009: *P*-value < 0.0001,  $R^2$ =0.09, n = 181).

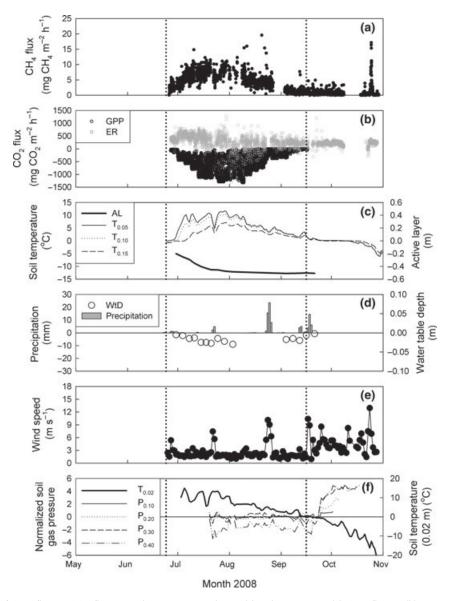
#### Seasonal dynamics in CH<sub>4</sub> fluxes

Spring 2009. The CH<sub>4</sub> flux was on average 0.18 mg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> during spring 2009. The highest CH<sub>4</sub> emissions with values up to 1.75 mg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> were observed on 21, 23 and 28 May 2009. These emissions do however not have any significant impacts on a seasonal or annual budget since they existed only for short periods of time (Fig. 3a). There were no correlations between variation in CH<sub>4</sub> fluxes and any of the measured variables. The reason for the lack of correlations was the low steady CH<sub>4</sub> fluxes without any large variations.

*Growing seasons 2008 and 2009.* The average growing season CH<sub>4</sub> fluxes were larger in 2008 than in 2009, that is, 4.60 mg and 2.94 mg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup>, respectively. Peak daily average CH<sub>4</sub> fluxes were 10.62 mg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> the 5 August in 2008, and 6.64 mg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> the 9 July in 2009 (Figs 2a and 3a).

All input variables were co-varying over the season. The temporal autocorrelation meant that the data could not be regarded as statistically independent, and the underlying assumptions for testing of significance in a regression analysis were hereby not fulfilled. It is consequently hard to tell which exact factors control the CH<sub>4</sub> fluxes. Despite this we still used regression analysis as an attempt to investigate how well the factors determine the CH<sub>4</sub> fluxes. We only present the coefficient of determination ( $R^2$  values), whereas the significance level (*P*-values) is not presented in the results. The lack of diurnal cycles allowed us to average fluxes daily, since missing data only increased uncertainty, but did not add any systematic errors.

Soil temperature at 0.02 m was the depth that best explained the variation in daily average CH<sub>4</sub> fluxes (linear correlation: 2008:  $R^2 = 0.79$ , n = 73; 2009:  $R^2 = 0.55$ , n = 98) (Fig. 5a). There was also a strong linear correlation between daily average CH<sub>4</sub> fluxes



**Fig. 2** Time series of  $CH_4$  fluxes,  $CO_2$  fluxes, and environmental variables during 2008. (a)  $CH_4$  fluxes, (b) gross primary production (GPP), and ecosystem respiration (ER), (c) soil temperature (T) at 0.05, 0.10 and 0.15 m depth, and active layer thickness (AL), (d) precipitation and water table depth (WtD), (e) average daily wind speed and (f) soil gas pressure (P) normalized by the annual mean together with soil temperature at 0.02 m depth. The dotted lines show the boundary between the different seasons.

and GPP (2008:  $R^2 = 0.72$ , n = 73; 2009:  $R^2 = 0.66$ , n = 98) (Fig. 5b).

No correlation between daily average CH<sub>4</sub> fluxes and WtD existed for the growing season in 2009 ( $R^2 = 0.03$ , n = 26) and a negative linear correlation ( $R^2 = 0.22$ , n = 14) was seen in 2008. There was a strong linear correlation between daily average CH<sub>4</sub> fluxes and AL, if only data up until the peak of the growing seasons were used (2008:  $R^2 = 0.58$ , n = 8; 2009:  $R^2 = 0.90$ , n = 10). No correlation was seen between CH<sub>4</sub> fluxes and friction velocity for either 2008 or 2009. Autumn and early winter 2008 and 2009. Average autumn and early winter CH<sub>4</sub> fluxes were 0.74 and 1.22 mg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> for 2008 and 2009, respectively. There were no increases in CH<sub>4</sub> fluxes after the soil started to freeze at 0.02 m depth on 23 September 2008, and on 3 October 2009. There were no correlations between daily average CH<sub>4</sub> fluxes and any of the environmental variables. Large CH<sub>4</sub> bursts were seen, at the 21 and 24–25 October 2008, and these occurred during very windy conditions. For the data measured after soil was completely frozen (18 October 2008) there was an exponential relationship ( $R^2 = 0.63$ ,

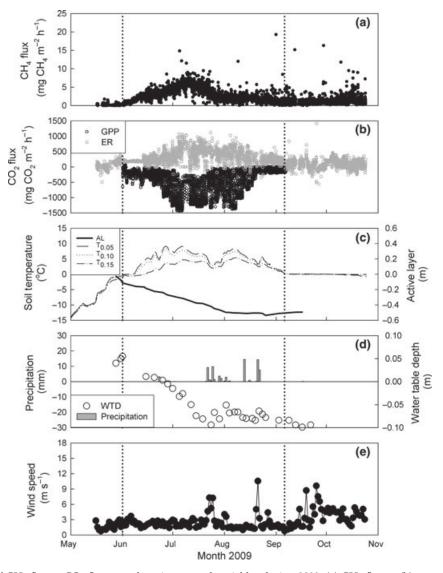


Fig. 3 Time series of CH<sub>4</sub> fluxes, CO<sub>2</sub> fluxes, and environmental variables during 2009. (a) CH<sub>4</sub> fluxes, (b) gross primary production (GPP), and ecosystem respiration (ER), (c) soil temperature (T) at 0.05, 0.10 and 0.15 m depth and active layer thickness (AL), (d) precipitation, and water table depth (WtD) and (e) average daily wind speed in ms<sup>-1</sup>. No soil gas pressure estimates were done during 2009. The dotted lines show the boundary between the different seasons.

n = 322) between half-hourly CH<sub>4</sub> fluxes and friction velocity (Fig. 6).

#### The budgets of the land-atmosphere exchange of carbon

If all C fluxes were added together (24 June to 31 October 2008 and 16 May to 31 October 2009), Rylekærene acted as a C source for the measurement period 2008 (+3.8 g C m<sup>-2</sup>), whereas it acted as a C sink for the measurement period 2009 (-47.6 g C m<sup>-2</sup>). Over the measurement period, 7.6 and 6.5 g C m<sup>-2</sup> were emitted as CH<sub>4</sub> in 2008 and in 2009, respectively. During the measurement period 2008, Rylekærene switched from being

a sink of  $-3.8 \text{ g C m}^{-2}$  to a source of  $+ 3.8 \text{ g C m}^{-2}$  because of the CH<sub>4</sub> emissions. During the measurement period 2009, 14% of the C balance was emitted as CH<sub>4</sub>. The C budgets for the different seasons are shown in Table 3.

The error analysis for the  $CO_2$  flux measurements and the gap filling procedure resulted in total accumulated errors of  $\pm$  5.68 and  $\pm$  4.45 g C for 2008 and 2009, respectively. The error analysis of the CH<sub>4</sub> flux measurements and gap filling of the CH<sub>4</sub> fluxes resulted in total accumulated errors of  $\pm$ 0.63 and  $\pm$ 0.77 g C for 2008 and 2009, respectively. According the co-spectrum analysis, 99% of the frequencies were measured by the

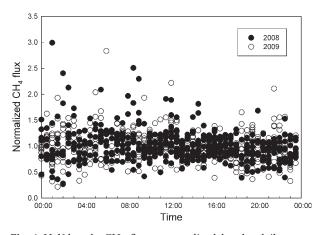


Fig. 4 Half-hourly  $CH_4$  fluxes normalized by the daily mean flux. Only data from the growing seasons 2008 and 2009 with daily data coverage larger than 70% were used.

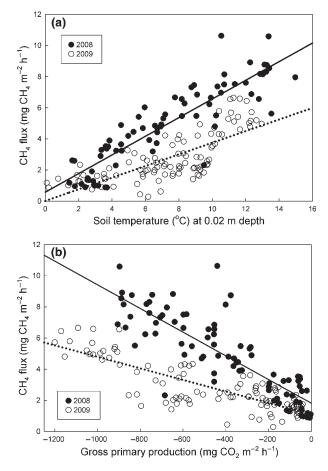
system. No transfer functions were thus applied for compensating for losses of high and low frequencies. The separate components of the error analysis are given in Table 4.

#### Discussion

#### Seasonal dynamics in CH<sub>4</sub> flux

*Spring* 2009. The increased spring-time emissions were observed simultaneously to the start of snow melt, possibly indicating a release of  $CH_4$  from the snow cover, as it settled during snow thawing. These were minor releases during short periods of time, and no extra  $CH_4$  bursts occurred as the ice covering the fen was melting. The increased  $CH_4$  emissions were very small in comparison to previous studies showing bursts as the soil thaws. Hargreaves *et al.* (2001) showed that 11% of total annual  $CH_4$  fluxes were released during spring thaw using the EC technique, whereas both Wille *et al.* (2008) (using the EC technique) and Tokida *et al.* (2007) (using closed chambers) showed spring thaw bursts equal in size to episodic ebullition releases during high summer.

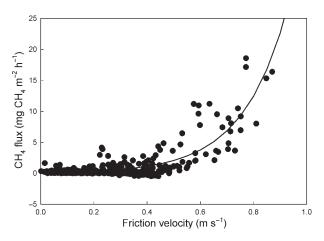
*Growing season*. The start of the growing season greenhouse gas exchange at high-Arctic areas is strongly controlled by timing of the snow melt (Groendahl *et al.*, 2007; Mastepanov, 2010). This was confirmed in the present study as both years had similar CH<sub>4</sub> fluxes 2 days after snow melt. The increase over the season was much faster in 2008 than in 2009. The main reason was most likely the more rapid increase in soil temperature in 2008 than in 2009. The relationship between CH<sub>4</sub> emissions and soil temperature has previously been shown to have both strong exponential relation-



**Fig. 5** (a) Daily average CH<sub>4</sub> fluxes against soil temperature at 0.02 m depth (T<sub>0.02</sub>) and (b) against gross primary production (GPP) for the growing seasons 2008 and 2009. The solid lines are regressions for the 2008 data series, and the dotted lines are regressions for the 2009 data series (2008: CH<sub>4</sub> flux =  $0.60 \times T_{0.02}$ +0.57,  $R^2 = 0.79$ , n = 73; 2009: CH<sub>4</sub> flux =  $0.37 \times T_{0.02}$ +0.01,  $R^2 = 0.55$ , n = 98), (2008: CH<sub>4</sub> flux =  $-0.007 \times \text{GPP} + 1.90$ ,  $R^2 = 0.72$ , n = 73; 2009: CH<sub>4</sub> flux =  $-0.004 \times \text{GPP} + 1.08$ ,  $R^2 = 0.66$ , n = 98).

ships (Hargreaves *et al.*, 2001; Rinne *et al.*, 2007; Wille *et al.*, 2008) and relationships that were weak or absent (Wickland *et al.*, 2006; Sachs *et al.*, 2008). When the relationship between the net  $CH_4$  fluxes and soil temperature deviates from an exponential one, this implies that other factors than  $CH_4$  production exert a strong control on the net emissions. Examples of such factors could be metabolic activity of the methanotrophs, sizes of the zones for  $CH_4$  production and oxidation, substrate availability, and storage or resistances along the transport pathways from the site of production to the atmosphere.

The WtD regulates the sizes of the zones for  $CH_4$  production and oxidation (Bubier & Moore, 1994). There was no consistent relationship between seasonal



**Fig. 6** Half-hourly CH<sub>4</sub> fluxes against friction velocity (u<sub>\*</sub>), after the soil was completely frozen the 18 October 2008. The solid line is an exponential regression with CH<sub>4</sub> flux against u<sub>\*</sub> (CH<sub>4</sub>flux =  $0.09 \times e^{6.39 \times u_*} - 0.18$ ,  $R^2 = 0.63$ , n = 322).

variation in CH<sub>4</sub> fluxes and WtD, that is, a lack of correlation 2009 and a negative correlation 2008. An explanation could be that the WtD is usually at its peak right after the snow melt, where after it decreases over the summer as the water runs off or is evapotranspired. Low CH<sub>4</sub> fluxes were consequently measured in the beginning of the growing seasons, when the WtD was high, and high fluxes at the peak of the growing season when the WtD was low. The water table was closer to the soil surface in 2008 than in 2009. It may be suggested that WtD explains the inter-annual variation in CH<sub>4</sub> flux whereas there are other factors with a stronger control on the annual CH4 fluxes. In agreement with this reasoning, many previous studies showing a positive relationship between CH4 flux and WtD have mainly showed spatial and long-term dependences (Hargreaves & Fowler, 1998; Huttunen et al., 2003; Bubier *et al.*, 2005). Another factor setting the zone for  $CH_4$  production is the AL. The main part of the  $CH_4$  fluxes originates from the upper part of the soil (Whalen, 2005), and  $CH_4$  fluxes were therefore not affected by AL when it approached its maximum.

The quantity and quality of the substrate to be decomposed are additional important environmental variables regulating  $CH_4$  fluxes (Whiting & Chanton, 1993; Joabsson & Christensen, 2001). The most important substrates for methanogens are fresh organic matter from root exudates, and recently produced litter (Chanton *et al.*, 1995; Ström & Christensen, 2007). The availability of fresh organic matter is closely linked to GPP (Joabsson & Christensen, 2001), hence the observed strong relationship between the  $CH_4$  fluxes and GPP. This has also been shown in previous studies (Whiting & Chanton, 1993; Friborg *et al.*, 2000; Joabsson & Christensen, 2001).

No correlation was seen between  $CH_4$  fluxes and friction velocity during the growing seasons. It has been shown that there are high correlations between atmospheric turbulence and diffusive transfers between water–air interfaces (MacIntyre *et al.*, 1995) and most studies reporting an effect of friction velocity have had a high surface coverage of water bodies (MacIntyre *et al.*, 1995; Hargreaves *et al.*, 2001; Sachs *et al.*, 2008; Wille *et al.*, 2008).

Autumn. Our study did not show any autumn  $CH_4$  emission pulse during the onset of soil freezing, as has been suggested by land-atmosphere  $CH_4$  flux measurements, and atmospheric concentration measurements at high northern latitudes (Khalil & Rasmussen, 1983; Dlugokencky *et al.*, 1994; Mastepanov *et al.*, 2008). There are few studies addressing the issue of wintertime  $CH_4$  fluxes in permafrost areas (Whalen &

				Autumn	and early	Entire measurement	
	Spring	Growing sea	ison	winter	and early	period	arement
Flux	2009	2008	2009	2008	2009	2008	2009
Daily CH <sub>4</sub> fluxes	0.003	0.084	0.051	0.011	0.027	0.059	0.039
Daily GPP	0.00	-2.57	-3.11	0.00	0.00	-1.68	-1.80
Daily ER	0.37	1.89	2.16	1.19	0.60	1.65	1.49
Daily net C exchange	0.37	-0.59	-0.90	1.20	0.63	0.03	-0.28
CH <sub>4</sub> flux budgets	0.1	7.1	5.0	0.5	1.5	7.6	6.5
GPP budgets	0.0	-218.1	-304.9	0.0	0.0	-218.1	-304.9
ER budgets	5.9	160.9	211.7	53.5	33.2	214.3	250.8
Net C exchange budgets	6.0	-50.1	-88.2	53.9	34.7	3.8	-47.6

**Table 3** The carbon budgets for the different seasons. The periods for the different seasons are given in Table 2. All daily average flux values are in g C  $m^{-2}$  per 24 h period. The Flux budgets are total accumulated flux in g C  $m^{-2}$ . No measurements were done during spring 2008

GPP, gross primary production; ER, ecosystem respiration.

**Table 4** The separate error components from the error analysis of  $CO_2$  and  $CH_4$  flux budgets. The errors are in g C for the entire measurement period. There were no errors associated with high and low frequency loss for the  $CH_4$  flux measurements as it was not based on fast gas concentration measurements

	2008	2009
Error $CO_2$ flux measurements (g C)		
Random	±0.26	±0.32
Gap filling	±5.36	±4.24
Frequency losses	±0.04	±0.54
Filtering for steady state conditions	±0.59	±0.60
Filtering for turbulent conditions	$\pm 1.77$	±1.05
Total	±5.68	±4.45
Error $CH_4$ flux measurements (g C)		
Friction velocity measurements	$\pm 0.41$	±0.55
Sensible heat flux measurements	±0.18	±0.23
CH <sub>4</sub> gradient measurements	±0.22	±0.29
Gap filling	±0.24	±0.08
Filtering for steady state conditions	±0.28	±0.39
Filtering for turbulent conditions	±0.10	±0.09
Total	±0.63	±0.77

Reeburgh, 1988; Mastepanov *et al.*, 2008; Wille *et al.*, 2008). Wille *et al.* (2008) did not see any increase in the wintertime CH<sub>4</sub> fluxes as the soil started to freeze in the Lena delta, north-eastern Siberia. Whalen & Reeburgh (1988) showed that 40% of the annual fluxes were winter fluxes from moss covered surfaces in Alaska, whereas no increases were seen for other vegetation covers. Mastepanov *et al.* (2008), showed that an equal amount of CH<sub>4</sub> was emitted during the freeze-in period as was emitted during the entire growing season in 2007 at the edge of Rylekærene.

Both Mastepanov et al. (2008) and Whalen & Reeburgh (1988) explained the increase in  $CH_4$  fluxes as the soil started to freeze with physical processes. The soil gas pressure increased as the soil started to freeze down toward the permafrost bottom. Accumulated CH<sub>4</sub> is prevented from diffusing downwards due to the impermeable permafrost, and it is hereby pressed up through the soil matrix. Pathways through the soil could both be voids in the frozen moss matrix (Whalen & Reeburgh, 1988), and through the aerenchyma of senescent vascular plants (Hargreaves et al., 2001; Mastepanov, 2010). In accordance with this hypothesis, there was indeed a strong increase in soil gas pressure as the soil started to freeze the 23 September 2008 (Fig. 2f). Despite the increase in soil pressure, there was no increase in the CH<sub>4</sub> fluxes.

A plausible explanation for the lack of autumn bursts in 2008 and 2009 is that other environmental conditions need to be fulfilled in order for the burst to occur. It was not enough with an increase in soil gas pressure. It could be that it was also necessary with an accumulation of CH<sub>4</sub> produced during the growing season. Whalen & Reeburgh (1988) only observed increased autumn emissions in moss covered areas, whereas measurements at plots with vascular plants did not show any increase in CH<sub>4</sub> fluxes during soil freezing. Shannon et al. (1996), have shown that CH<sub>4</sub> transporting plants deplete both the dissolved CH<sub>4</sub> pool in the pore water, and the concentration of CH<sub>4</sub> in trapped gas bubbles in the saturated rhizosphere. In case there are no vascular plants, a larger fraction of the produced CH<sub>4</sub> is stored in the soil. This CH<sub>4</sub> can then be released due to the increased soil pressure during soil freezing. There was no apparent difference in vascular plant coverage between the site where the Mastepanov et al. (2008) chambers were positioned and the main parts of Rylekærene (where the current study was conducted). The Mastepanov et al. (2008) site, however, has a slightly different vascular plant composition with a higher coverage of the grass A. latifolia, indicating lower plant mediated CH<sub>4</sub> transport. This could possibly result in a CH<sub>4</sub> accumulation at the Mastepanov et al. (2008) site.

An interesting feature was the bursts of CH<sub>4</sub> during the storm events in October 2008. It might be that accumulated CH<sub>4</sub> existed in cavities in the moss and vegetation layer, and in cracks in the frozen soil. The high wind speed increased the turbulence, which likely resulted in turbulence-induced large-scale ebullition, possibly triggered by changes in air pressure, and ventilation of these cavities. This has also been observed in other studies (Massman *et al.*, 1997; Black *et al.*, 2000; Aurela *et al.*, 2002). No similar phenomena were seen in 2009. However, there was a 0.30 m deep snow layer covering the area in 2009, and an increase in turbulence could not affect the soil due to this protective layer.

#### Diurnal variation in CH<sub>4</sub> flux

Diurnal variation in CH<sub>4</sub> fluxes is often site specific and has been observed at some sites (Shannon *et al.*, 1996; Hargreaves & Fowler, 1998; Kim *et al.*, 1998), whereas not at others (Kormann *et al.*, 2001; Rinne *et al.*, 2007). We did not find any indications of diurnal variations in the present study. It could be that the lack of diurnal variation was associated with the absence of true night conditions during the main part of the growing seasons. Average nighttime (22:00–6:30 hours) PAR was 103 and 120  $\mu$ mol m<sup>-2</sup> s<sup>-1</sup> for the growing seasons 2008 and 2009, respectively. The lack of diurnal variation indicates that the CH<sub>4</sub> fluxes were not directly affected by the stomatal closure or had other pathways than the stomata for CH<sub>4</sub> transport to the atmosphere (Morrissey *et al.*, 1993). It could also be that the vegetation had sufficient water and was not limited by transpiration.

Tagesson (2011) showed that there were CH<sub>4</sub> emissions from continuous and hummocky fen, whereas they were undetectable in the dry plant communities (grassland, S. arctica snowbed, and the heath communities). This could explain the relationship between halfhourly CH<sub>4</sub> fluxes and fraction of continuous fen in the footprint of the tower. The continuous fen had a water table close to the soil surface and a high coverage of vascular plants; consequently it had the largest CH<sub>4</sub> fluxes. Furthermore, species composition of vascular plants can affect CH<sub>4</sub> emissions and substrate availability for methanogens and pointed to the importance of Eriophorum species (E. scheuchzeri, E. angustifolium and E. vaginatum) in this respect (Ström et al., 2005, 2012; Ström & Christensen, 2007). These species are common in the continuous fen areas.

#### Representativeness of the site

Walker et al. (2005) made a hierarchical subdivision of the Arctic vegetation along an altitudinal temperature gradient from north to south, that is, (A) cushion-forb, (B) prostrate dwarf-shrub, (C) hemiprostrate dwarfshrub (D) erect dwarf-shrub, and (E) low shrub subzones. The main features distinguishing Subzone C from Subzone B are the presence of the hemiprostrate shrub C. tetragona and well-differerentiated plant communities in mires, snowbeds and creek sides. The mean July temperature at the southern boundary of Subzone B is approximately 5 °C and of Subzone C about 7 °C (Walker et al., 2005). The average air temperature of the warmest month in the Zackenberg valley is 5.8 °C. Based solely on temperature the Zackenberg valley would belong to subzone B. However, the presence of fen, grassland, C. tetragona heath, D. octopetala heath, V. uliginosum heath and S. arctica snowbed (Bay, 1998) would primarily put the valley in to subzone C. Combined subzone B and C covers about 32% of the tundra zone, including most of the islands in the Canadian Archipelago, most of northern Greenland, south-western Svalbard, Novaya Zemlya, most of the northern fringe of mainland Russia, and the New Siberian Islands (Walker et al., 2005). Thus, the plant communities in the Zackenberg valley and in Rylekærene comprise a good example of this part of the Arctic tundra.

#### The budgets of the land-atmosphere exchange of carbon

The carbon budgets were quite large, but in the same range as other Arctic studies (Corradi *et al.*, 2005; Wille *et al.*, 2008). Both total growing season and daily

averages of GPP and ER were larger in 2009 than in 2008 (Table 3), whereas the total growing season  $CH_4$ fluxes were larger in 2008. There was no difference in average incoming PAR, whereas both temperature and WtD were higher in 2008 than in 2009. The growing season constituted a larger fraction of the measurement period in 2008 than in 2009 and the sink function for 2008 should thus be larger in comparison to 2009. It could be that the vegetation was water stressed in 2008, as the WtD was almost at the soil surface throughout the entire growing season. Further, it could be that the drier conditions in 2009 increased the mineralization rates resulting in increased GPP; ER was also higher in 2009 than in 2008 in accordance with this hypothesis. An explanation to the low ER in 2008 could be the very shallow aerobic zone. This lowered the fraction of C respired as CO<sub>2</sub>, and resulted in a larger fraction respired as CH<sub>4</sub>. The% of the C balance that was emitted as CH<sub>4</sub> was similar to studies in the Siberian arctic tundra (Friborg *et al.,* 2003; Wille *et al.,* 2008). The year 2008 was warmer and wetter than 2009. Global circulation models predict increased temperatures and increased precipitation in the future for the northern latitude ecosystems. Arctic wetlands such as Rylekærene may thus play a more significant role for the climate in the future.

#### Acknowledgements

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