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# Methane oxidation in contrasting soil types: responses to experimental warming with implication for landscape-integrated CH<sub>4</sub> budget

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## Abstract

Arctic ecosystems are characterized by a wide range of soil moisture conditions and thermal regimes and contribute differently to the net methane (CH<sub>4</sub>) budget. Yet, it is unclear how climate change will affect the capacity of those systems to act as a net source or sink of CH<sub>4</sub>. Here, we present results of *in situ* CH<sub>4</sub> flux measurements made during the growing season 2014 on Disko Island (west Greenland) and quantify the contribution of contrasting soil and landscape types to the net CH<sub>4</sub> budget and responses to summer warming. We compared gas flux measurements from a bare soil and a dry heath, at ambient conditions and increased air temperature, using open-top chambers (OTCs). Throughout the growing season, bare soil consumed  $0.22 \pm 0.03$  g CH<sub>4</sub>-C m<sup>-2</sup> ( $8.1 \pm 1.2$  g CO<sub>2</sub>-eq m<sup>-2</sup>) at ambient conditions, while the dry heath consumed  $0.10 \pm 0.02$  g CH<sub>4</sub>-C m<sup>-2</sup> ( $3.9 \pm 0.6$  g CO<sub>2</sub>-eq m<sup>-2</sup>). These uptake rates were subsequently scaled to the entire study area of 0.15 km<sup>2</sup>, a landscape also consisting of wetlands with a seasonally integrated methane release of  $0.10 \pm 0.01$  g CH<sub>4</sub>-C m<sup>-2</sup> ( $3.7 \pm 1.2$  g CO<sub>2</sub>-eq m<sup>-2</sup>). The result was a net landscape sink of 12.71 kg CH<sub>4</sub>-C (0.48 tonne CO<sub>2</sub>-eq) during the growing season. A nonsignificant trend was noticed in seasonal CH<sub>4</sub> uptake rates with experimental warming, corresponding to a 2% reduction at the bare soil, and 33% increase at the dry heath. This was due to the indirect effect of OTCs on soil moisture, which exerted the main control on CH<sub>4</sub> fluxes. Overall, the net landscape sink of CH<sub>4</sub> tended to increase by 20% with OTCs. Bare and dry tundra ecosystems should be considered in the net CH<sub>4</sub> budget of the Arctic due to their potential role in counterbalancing CH<sub>4</sub> emissions from wetlands – not the least when taking the future climatic scenarios of the Arctic into account.

**Keywords:** Arctic, bare soil, dry heath, Greenland, methane budget, methane uptake, soil moisture, warming

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## Introduction

Methane (CH<sub>4</sub>) is a powerful greenhouse gas (GHG), which has a global warming potential (GWP) 28 times higher than carbon dioxide (CO<sub>2</sub>) estimated on a mass basis over a time scale of 100 years (Myhre *et al.*, 2013). The concentration of CH<sub>4</sub> in the atmosphere depends upon the balance between its sources and sinks (Ciais *et al.*, 2013). CH<sub>4</sub> can be emitted by natural (from lands and oceans) and anthropogenic sources, for example, fossil fuel combustion, waste management, livestock and land-use change (Dlugokencky *et al.*, 2011; Tate, 2015). Methane sinks are mainly natural and depend upon abiotic (tropospheric oxidation and loss to the stratosphere) and biotic reactions (microbial oxidation in soils) (Mosier *et al.*, 1991; Chen *et al.*, 2011). Compared to pre-industrial values, the current atmospheric

concentration of CH<sub>4</sub> has increased by a factor of 2.5 (Dlugokencky *et al.*, 2011), despite significant year-to-year variations not yet fully understood (Kirschke *et al.*, 2013).

Ice core CH<sub>4</sub> records from the Arctic and Antarctic regions show evidence on the close relationship between fluctuations in atmospheric CH<sub>4</sub> concentrations and climate changes, such as air warming (Brook *et al.*, 2000; Nisbet & Chappellaz, 2009). In the Arctic, the projected trend in air temperature for the end of the 21st century indicates a postindustrial warming of about 4–6 °C (Collins *et al.*, 2013). On land, the increased air temperature may cause thawing of permafrost and consequently vast amounts of organic carbon (C) stored in the permafrost will be available for biological decomposition, potentially followed by a release of CH<sub>4</sub> into the atmosphere (Schoor *et al.*, 2008, 2015). The sensitivity of arctic ecosystems to changes in temperature regimes has

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therefore raised the need to understand how the natural sources and sinks of CH<sub>4</sub> will respond to the future climatic scenarios. The ice-free terrestrial areas of the Arctic (covering about  $5 \times 10^6$  km<sup>2</sup>) are characterized by various soil and vegetation types with different soil moisture contents (Walker *et al.*, 2005). The relative contribution of those areas to the net CH<sub>4</sub> budget of the Arctic is therefore expected to be dependent upon their specific soil water content and temperature regimes which are considered among the main abiotic factor influencing CH<sub>4</sub> dynamics in arctic soils (Smith *et al.*, 2000; Tate, 2015). Soil moisture regulates the diffusion of gases throughout the soil profile (Hiltbrunner *et al.*, 2012) and may consequently regulate the subsurface availability of both CH<sub>4</sub> and oxygen (Jørgensen *et al.*, 2015). Thus, moist to wet lands function as net sources of CH<sub>4</sub> while dry heath tundra and nonvegetated areas act as net sinks (Jørgensen *et al.*, 2015). Water table fluctuations related to changes in soil temperature have been the subject of previous CH<sub>4</sub> budget studies which focused mainly on arctic water saturated soils (Mastepanov *et al.*, 2013; Tagesson *et al.*, 2013; Natali *et al.*, 2015; Ström *et al.*, 2015). Only recently the contribution of dry arctic landscapes has been investigated to calculate and model the net exchange of CH<sub>4</sub> (Brummell *et al.*, 2014; Emmerton *et al.*, 2014; Jørgensen *et al.*, 2015; Lau *et al.*, 2015; Stackhouse *et al.*, 2015). However, those studies assessed the relationship between CH<sub>4</sub> uptake rates and changes in soil temperature and moisture mostly under controlled conditions of laboratory experiments, with the exception of Lau *et al.* (2015) who estimated the *in situ* temperature sensitivity of soil CH<sub>4</sub> oxidation of arctic mineral cryosols.

Hence, there is a need for more *in situ* measurements of CH<sub>4</sub> fluxes to clarify the sensitivity of bare grounds and dry tundra soils to *in situ* changes of soil temperature and water content and their influence as CH<sub>4</sub> sinks in a changing climate (Graham *et al.*, 2012; Brummell *et al.*, 2014).

The aims of this study were accordingly (1) to quantify the capacity of bare and dry heath soils to consume atmospheric CH<sub>4</sub>, (2) to assess the sensitivity of CH<sub>4</sub> consumption with respect to summer warming and (3) to estimate a landscape-integrated CH<sub>4</sub> budget of the area at both ambient and increased summer temperatures. The latter is meant to provide insight into the present and possible future contribution of the different soil and vegetation cover types of Greenland to the exchange of CH<sub>4</sub> between soil and atmosphere. The underlying hypothesis is that dry arctic ecosystems have an underestimated capacity to oxidize CH<sub>4</sub>, which is important for the CH<sub>4</sub> budget of Greenland.

## Material and methods

### The Blæsedalen area

The study was carried out in the Blæsedalen valley which is located on Disko Island in West Greenland (69°16'N, 53°27'W). The mean annual soil temperature at 5 cm depth is  $-0.9$  °C (1991–2004) (Hansen *et al.*, 2006; Hollesen *et al.*, 2015). The warmest monthly mean air temperature is  $7.9 \pm 1.6$  °C (SD) during July, whereas the coldest monthly mean is  $-14.0 \pm 5.0$  °C during February–March (Hollesen *et al.*, 2015). Frozen soil conditions prevail from October to late May. The area is characterized by the presence of discontinuous permafrost, and the soil type is <10 000 years old. Soil development is generally weak. The area of sampling is characterized by four main soil cover types: bare ground, dry heath, fen and water-logged fen (Table 1). The vegetation cover at the dry heath is dominated by deciduous dwarf shrubs (*Vaccinium uliginosum*, *Betula nana*, *Salix glauca*) and evergreen low shrubs (*Empetrum nigrum*, *Cassiope tetragona*). In the fen and water-logged fen, the main vegetation components are sedges (*Carex aquatilis* ssp. *stans*, *Carex rariflora*, *Eriophorum angustifolium*), mosses (*Paludella squarrosa* and *Tomentypnum nitens*) and deciduous shrubs (*Salix arctophila*), while the bare ground presents few scattered patches of *Dryas octopetala* and other dwarf shrubs.

### Soil parameters

At the dry heath site, intact soil cores (5 cm diameter) were collected during two sampling campaigns in July and August 2013 from the top 10 cm at each plot ( $n = 5$  samples per sampling time) and split into 0–5 and 5–10 cm subsamples. Soil pH was measured on the soil samples collected during both campaigns by adding Milli-Q H<sub>2</sub>O to dry soil in a 1 : 10 ratio. Soil pH values at the fen and water-logged fen sites were based on *in situ* measurements made during August 2015 by inserting the pH probe directly into the ground at 2.5 cm and 7.5 cm as pH measurements in organic soils previously have been shown to be highly sensitive to drying (Elberling & Matthiesen, 2007). At the dry heath site, soil dry bulk density and total soil carbon (C) and nitrogen (N) were measured on the soil samples collected in August 2013, using a Eurovector EA coupled to an Isoprime isotope ratio mass spectrometer. At the bare soil site, intact soil cores (4.5 cm diameter) were collected in September 2014 outside the measurement plots ( $n = 3$ ) and split into 0–10 and 10–20 cm subsamples. The soil analyses were identical to the sampling from the dry heath.

Volume-specific soil samples at 5 cm depth were collected in July 2013 at the water-logged fen and in August 2015 in the fen. The total carbon and nitrogen contents in the samples from the water-logged fen (2013) and those from fen sites (2014) were measured in solid samples by Dumas combustion (1020 °C) on an elemental analyzer (EA Flash 2000, Thermo Scientific, Bremen, Germany). The soil samples (10 mg), previously homogenized and dried, were weighed out into tin combustion cups for elemental analysis. Acetanilide (Merck, Darmstadt, Germany), peach leaf material NIST 1547 (National Institute of Standards and Technology, Gaithersburg, MD,

**Table 1** Overview of the main soil characteristics and vegetation cover type in the area of sampling in Blæsedalen. Soil temperature and moisture are averages of manual measurements ( $\pm$  SE) during the gas measurement campaigns ( $n = 5$  bare ground, dry heath and water-logged fen;  $n = 6$  fen)

	Bare ground	Dry heath	Fen	Water-logged fen
Soil temperature 5 cm ( $^{\circ}$ C)	11.55 ( $\pm$ 0.88)	9.20 ( $\pm$ 0.66)	6.41 ( $\pm$ 0.36)	8.91 ( $\pm$ 0.45)
OTCs soil temperature 5 cm ( $^{\circ}$ C)	12.66 ( $\pm$ 0.96)	10.48 ( $\pm$ 0.75)	6.68 ( $\pm$ 0.39)	–
Soil moisture 0–7 cm (%Vol)	6.95 ( $\pm$ 0.57)	26.82 ( $\pm$ 1.79)	77.31 ( $\pm$ 3.61)	100
OTCs soil moisture 0–7 cm (%Vol)	12.45 ( $\pm$ 1.10)	15.24 ( $\pm$ 1.75)	75.85 ( $\pm$ 3.99)	–
Main vegetation cover	Barren with sparse dwarf shrubs	Dwarf shrub heath	Fen with sedges, mosses and willow	Fen with sedges
Soil depth (cm)	0–10	0–5		
C (%)	0.91 ( $\pm$ 0.35)	17.37 ( $\pm$ 2.10)	21.08 ( $\pm$ 1.50)	28.07 ( $\pm$ 1.38)
N (%)	0.09 ( $\pm$ 0.02)	0.78 ( $\pm$ 0.09)	1.30 ( $\pm$ 0.15)	1.44 ( $\pm$ 0.23)
C : N	9.81 ( $\pm$ 1.34)	22.50 ( $\pm$ 1.15)	17.20 ( $\pm$ 2.90)	20.73 ( $\pm$ 4.05)
pH	7.21 ( $\pm$ 0.07)	5.08 ( $\pm$ 0.09)	7.15 ( $\pm$ 0.07)	6.29 ( $\pm$ 0.19)
Bulk density ( $\text{g cm}^{-3}$ )	0.36 ( $\pm$ 0.04)	0.26 ( $\pm$ 0.03)	0.04 ( $\pm$ 0.01)	0.08 ( $\pm$ 0.04)
Soil depth (cm)	10–20	5–10		
C (%)	1.05 ( $\pm$ 0.32)	2.61 ( $\pm$ 0.29)	18.65 ( $\pm$ 0.64)	28.27 ( $\pm$ 7.36)
N (%)	0.10 ( $\pm$ 0.02)	0.17 ( $\pm$ 0.02)	1.49 ( $\pm$ 0.09)	1.54 ( $\pm$ 0.16)
C : N	9.96 ( $\pm$ 1.39)	15.15 ( $\pm$ 0.40)	12.73 ( $\pm$ 1.10)	17.68 ( $\pm$ 3.17)
pH	7.03 ( $\pm$ 0.12)	5.81 ( $\pm$ 0.03)	6.88 ( $\pm$ 0.09)	6.36 ( $\pm$ 0.13)
Bulk density ( $\text{g cm}^{-3}$ )	0.34 ( $\pm$ 0.02)	0.38 ( $\pm$ 0.09)	0.12 ( $\pm$ 0.04)	0.10 ( $\pm$ 0.02)

USA) and soil standards (Elemental Microanalysis, Okehampton, UK) were used for elemental analyzer mass calibration.

Manual records of soil temperature and moisture were carried out together with the gas flux measurements with a portable thermometer (Spectrum Technologies Inc., Plainfield, IL, USA) and a Theta Probe soil moisture sensor (ML2x Delta-T Devices Ltd, Cambridge, UK).

### Experimental design

The typical range of soils and vegetation types of ice-free west Greenland is also found in Blæsedalen at Disko. In this study, we focus on the  $\text{CH}_4$  fluxes in a bare soil and dry heath tundra, but also seasonal-integrated fluxes from wet fen areas are included to upscale the *in situ* plot measurements and estimate the landscape-integrated  $\text{CH}_4$  budget for the whole area of Blæsedalen (0.15  $\text{km}^2$ ). The four vegetation types (or landscape units) are indicated throughout the manuscript as: bare ground (BG), dry heath (DH), fen and water-logged fen.

At the four vegetation types, the exchange of  $\text{CH}_4$  between soil and atmosphere was measured at ambient conditions (control plots) and at BG, DH and fen also at increased summer air temperature by open-top chambers (OTCs). The plots at the dry heath were established in June 2012, and the plots of the fens and the bare site in July 2013. Each control plot is coupled to a plot with experimental warming by OTC forming a block unit, in total five replicate blocks for three of the vegetation types, and six for the fen site.

### Methane gas measurements

Fluxes of  $\text{CH}_4$  at the BG and at the DH sites were measured in six measurement campaigns during July and September 2014

(5th, 12th, 18th and 25th of July, 6th and 18th of September). At the fen and water-logged fen sites, the measurements were carried out during six campaigns between July, August and September 2014 (fen: 9th and 21th of July, 5th and 19th of August, 4th and 14th of September. Water-logged fen: 1st, 14th and 27th of July, 13th and 24th of August and 8th of September).

Measurements were carried out in random order using polycarbonate closed-static chambers placed onto water-sealed frames permanently installed into the soil. Depending on the height of the frames in the soil, and the water table level in the wet plots, the chamber volume varied from 8.6 to 11 L at the BG and DH sites and from 6 to 13 L in the wet areas. Larger chambers were used at the fen and water-logged fen to avoid damages on the vegetation. Measurements at the BG and DH sites were made in darkness using white blackout cloth to cover the chambers with little influence on the temperature inside the chambers. At the fen and water-logged fen, instead, the chambers were not covered due to the potential linkage between light and plant transport of  $\text{O}_2$  into and  $\text{CH}_4$  out of the wet soil. Light and dark measurements at the BG and DH sites have previously shown that including or excluding the light is not influencing the soil  $\text{CH}_4$  exchange (Fig. S1). This is due to the fact that dry tundra ecosystems are mainly sinks for  $\text{CH}_4$ , and in contrast to wetlands, soil  $\text{CH}_4$  oxidation rates in dry tundra are controlled by soil properties rather than vegetation (Emmerton *et al.*, 2014; Jørgensen *et al.*, 2015). Thus, the indirect effects of photosynthesis on the fluxes of  $\text{CH}_4$  at the DH site can be ruled out, making the  $\text{CH}_4$  flux measurements between the dry and wet sites comparable despite the fact that they were measured under dark and light conditions.

During measurements, air volume in the chamber headspace was circulated in a closed loop at a flow rate of ca. 0.8

standard L min<sup>-1</sup> into an Ultra-portable Greenhouse Gas Analyzer, model 915-0011 (©2013; Los Gatos Research, Inc., San Jose, CA, USA). CH<sub>4</sub> concentrations were continuously measured and logged over 10-min periods with 10-s sampling frequency. Air temperature outside the chambers was measured using a temperature sensor placed next to the lid of the chambers (107 temperature probe; Campbell Scientific Ltd., Shepshed, UK). A temperature sensitivity coefficient (Q<sub>10</sub>) was estimated based on the Arrhenius equation by plotting the natural logarithm of the *in situ* CH<sub>4</sub> uptake rates against 1000/soil temperature estimated in Kelvin degrees (Davidson & Janssens, 2006).

### Blæsedalen mapping

The study area of 0.15 km<sup>2</sup> was mapped with an unmanned aerial vehicle (UAV) on July 24, 2014, to allow for detailed areal integration of the point-based CH<sub>4</sub> flux measurements. Onboard the UAV, there were sensors to record visible red, green and blue light (RGB) and near-infrared (NIR) (modified Canon Inc., Tokyo, Japan sx260hs cameras). The UAV mapping resulted in a four-band orthophoto (RGB and NIR) at 3 cm ground resolution. The high-resolution orthophoto formed the basis for an object-based classification conducted in eCognition (Trimble, Sunnyvale, CA, USA). Classifications based on objects can be advantageous when the classification is based on a limited number of bands (Blaschke, 2010). Moreover, it allows for segmentation of areas with similar characteristics to mitigate noise in classifications based on orthophotos with high ground resolution. Auxiliary data consisted of soil moisture data covering a gradient from <10 Vol % to 100 Vol %. The classification was trained with 30 ground control points (GCPs) and tested on 30 independent GCPs equally distributed across the following surfaces: water-logged, fen, dry heath, shrubs (tall willow thicket) and barren ground (Fig. S2). Final accuracy for the five surface classes was 92.8% with a kappa of 0.89 (Table S1).

The soil/vegetation cover classification of the study area was furthermore compared to the general distribution of surface classes at the southern part of Disko, which was evaluated by the classification of a WorldView-2 satellite scene taken on July 12, 2012 (upper left corner: 69.31°N, 53.57°W; lower right corner: 69.23°N, 53.41°W) covering almost 40 km<sup>2</sup> (Fig. S3). The accuracy and results of this classification are reported in Tables S2 and S3.

### Data analysis

The fluxes of CH<sub>4</sub> were calculated by fitting a second-order polynomial regression model to the changes in gas concentration in the chamber headspace over 10 min. The slope after 10 min was subsequently used for calculating the rates if significantly different from zero by  $P \leq 0.05$  ( $R^2 \geq 0.85$  based on 61 records). A total of 120 fluxes were used for the analyses at the BG and DH sites and 100 fluxes for the fen and water-logged sites. The criteria for acceptance of fluxes from the fen and water-logged fen were  $P \leq 0.05$  and  $R^2 > 0.6$ . To perform the statistical analyses, the soil CH<sub>4</sub> fluxes at the BG and DH

sites were box cox-transformed ( $\lambda = 0.101$ ), while soil temperature and soil moisture were log<sub>10</sub>-transformed to meet the model assumptions of normal distribution and homogeneity of variance.

The CH<sub>4</sub> fluxes measured during each campaign were integrated over the number of days between the midtime points of two consecutive campaigns (g CH<sub>4</sub>-C m<sup>-2</sup> days<sup>-1</sup>). The final seasonal-integrated flux (g CH<sub>4</sub>-C m<sup>-2</sup>) was estimated as the sum of the CH<sub>4</sub> fluxes integrated over six time intervals, which at the BG, DH and water-logged fen sites spanned over 92 days, while at the fen site over 90 days.

An ANOVA mixed model with random and repeated factors was used to analyze the treatment and site effect on the CH<sub>4</sub> fluxes at the DH and BG sites. The model was run including 'site' (DH and BG), 'treatment' (C and W), day of sampling (indicated as day of the year, DOY) and their combination as fixed factors, 'block' as random factor, DOY as repeated factor and 'plot' as subject identifier. A similar repeated mixed model, without 'site' as factor, was used to assess the treatment effect within each site. The treatment effects during each measuring campaign, instead, were tested with an ANOVA mixed model with only a random factor ('block'). Pairwise comparisons of least squares means were performed on single factors and interactions to interpret the results of the ANOVA mixed model. Multiple linear regression analysis was used to understand the relationship at ambient conditions and under warming by OTCs between CH<sub>4</sub> uptake rates, soil temperature and moisture at the BG and DH sites. Statistical significance was accepted by values of  $P \leq 0.05$ , and tendencies are reported by values of  $P \leq 1$ . The error values reported in the text as well as in the figures are standard error of the mean ( $\pm$  SE).

## Results

### Site description

The Blæsedalen area is in the transition zone between the bioclimatic subzone C and subzone D of the Arctic, according to the circumpolar arctic vegetation map (CAVM) (Walker *et al.*, 2005). It is mainly characterized by dwarf and hemi-prostrate shrubs (DH site) with scattered areas of bare soil (BG site) and depressions dominated by fen and water-logged fen. The soil properties of the area are therefore heterogeneous and representative of the specific characteristics of each soil and vegetation type (Table 1). Generally, the pH was moderately acidic to slightly alkaline (Table 1). The seasonal averaged values of soil water content varied between low (<10%) at the BG and high values (>80%) at the fen sites. Soil temperature at 5 cm depth also showed a consistent difference between the sites; as expected, the BG site was the warmest while the fen had the lowest soil/water temperature. The area contribution of the

**Table 2** Averaged seasonal and total CH<sub>4</sub> fluxes at each site ( $n = 6 \pm \text{SE}$ ), and final net CH<sub>4</sub> budget estimated for the 0.15 km<sup>2</sup> area of Blæsedalen, Disko Island, West Greenland

	Area (m <sup>2</sup> )	Ambient		Warming with OTCs	
		Seasonal flux (g CH <sub>4</sub> -C m <sup>-2</sup> )	Total CH <sub>4</sub> flux (kg CH <sub>4</sub> -C)	Seasonal flux (g CH <sub>4</sub> -C m <sup>-2</sup> )	Total CH <sub>4</sub> flux (kg CH <sub>4</sub> -C)
Dry heath	103 253	-0.10 ± 0.02	-10.78 ± 1.79	-0.14 ± 0.04	-14.36 ± 3.88
Bare soil	19 683	-0.22 ± 0.03	-4.24 ± 0.61	-0.21 ± 0.07	-4.16 ± 1.42
Fen	23 467	0.10 ± 0.01*	2.32 ± 0.58	0.15 ± 0.13†	3.50 ± 2.96
Net CH <sub>4</sub> budget			-12.71		-15.02

\*Average for fen and water-logged fen.

†Value calculated only for the fen site.

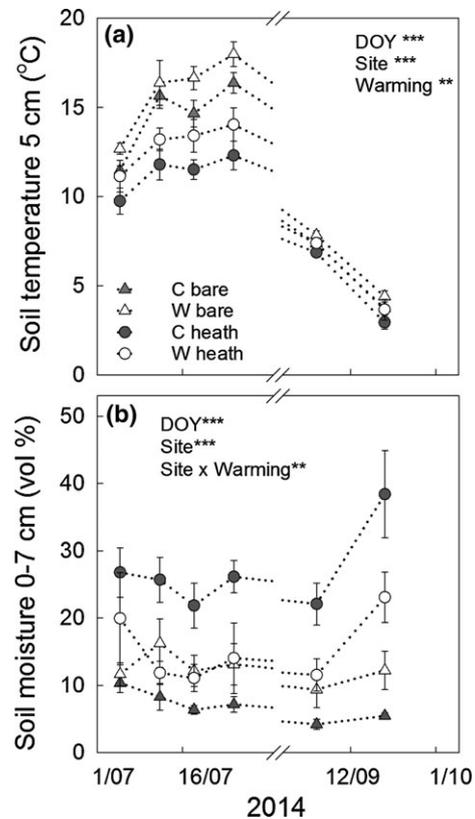
different soil/vegetation cover types is presented in Table 2.

#### Soil temperature and moisture

During the measurement campaigns, the manual records at ambient conditions of soil temperature at 5 cm depth of the DH site (Fig. 1a) reached a maximum by the end of July of  $12.3 \pm 0.8$  °C and dropped to  $3.0 \pm 0.4$  °C by the end of September. During the same period, soil moisture (Fig. 1b) ranged between a minimum of  $22 \pm 3.1\%$  on the 18th of July and a maximum of  $38.4 \pm 6.5\%$  in September. At the BG plots, soil temperature and moisture followed the same trends observed at the DH plots. Soil temperature reached a maximum by the end of July,  $16.4 \pm 0.6$  °C, and decreased to  $4.0 \pm 0.2$  °C in September while soil moisture ranged between a maximum of  $10.3 \pm 1.4\%$  at the beginning of July and a minimum of  $4.0 \pm 0.8\%$  in September. The repeated-measures ANOVAS indicated a significant effect of the day of sampling (indicated as DOY) on the seasonal trend of soil temperature ( $P < 0.001$ ;  $F = 264.14$ ) and moisture ( $P < 0.001$ ;  $F = 6.99$ ). The two soil parameters varied significantly according to the site; the BG site had the highest soil temperature ( $P < 0.001$ ;  $t = -5.52$ ) and the lowest soil moisture ( $P < 0.001$ ;  $t = 4.64$ ). However, plots with OTCs at the DH site showed a significant reduction in soil moisture ( $P = 0.02$ ;  $t = 2.64$ ), while plots with OTCs at the BG site showed a significant increase in soil moisture ( $P = 0.05$ ;  $t = 2.11$ ).

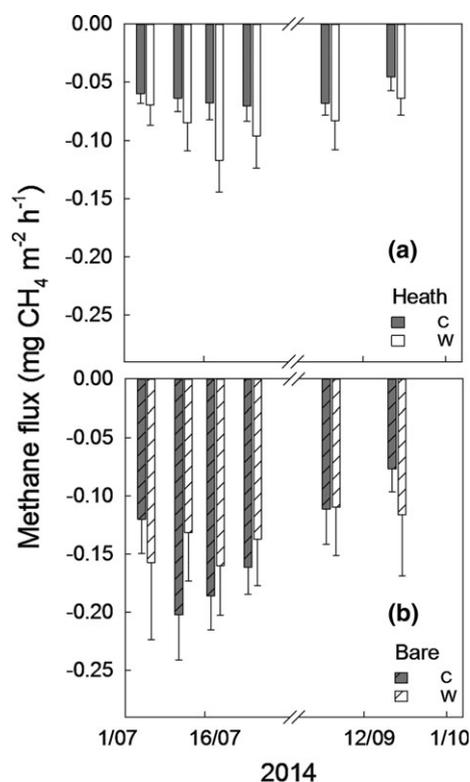
#### Methane uptake rates

The rates of soil CH<sub>4</sub> uptake at the control plots of the DH site ranged between a maximum of  $-0.07 \pm 0.01$  mg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> at the end of July and a minimum of  $-0.05 \pm 0.01$  mg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> during the last campaign in September (Fig. 2a). At the control plots of the BG site, the rates of soil CH<sub>4</sub> uptake ranged



**Fig. 1** Manual measurements ( $n = 5 \pm \text{SE}$ ) of (a) soil temperature and (b) soil moisture recorded during gas measurements at the two sites, 'bare' and 'heath' in Blæsedalen, west Greenland. In legend, control (C) and warming with OTCs (W). Significant repeated fixed treatment effects are indicated by \*\* $P \leq 0.01$ , \*\*\* $P \leq 0.001$ .

between  $-0.20 \pm 0.04$  mg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> in mid-July and  $-0.08 \pm 0.02$  mg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> in the last measurement campaign in September (Fig. 2b). The effects of warming by OTCs are contrasting for the two sites, however not statistically significant. The results of the ANOVA mixed model run by campaign indicated an increase in soil CH<sub>4</sub> uptake rates at the plots with



**Fig. 2** Methane uptake rates measured in the control plots (C) and plots with warming by OTCs (W) at (a) the dry heath (DH) site and (b) the bare ground (BG) site ( $n = 5 \pm \text{SE}$ ) in Blåsedalen, west Greenland.

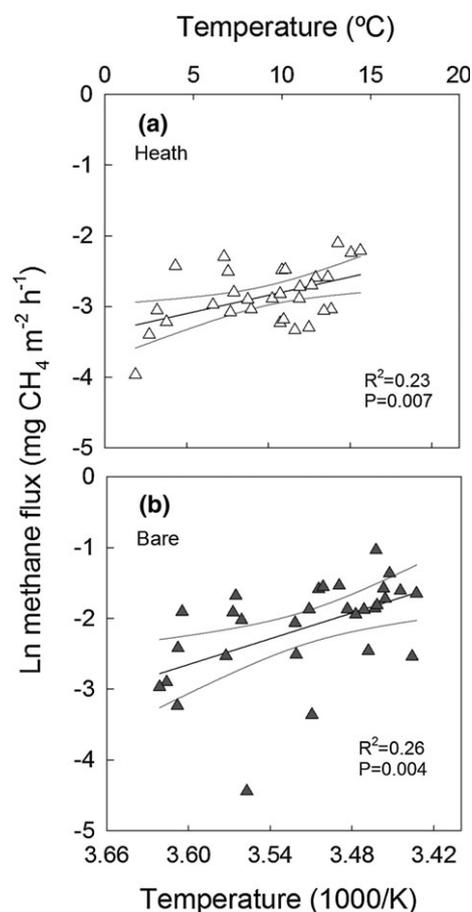
warming by OTCs of the DH site where activity peaked in mid-July (18th of July:  $P = 0.09$ ,  $F = 4.87$ ), and then gradually decreased toward the end of September (Fig. 2a). By contrast, the rates of soil CH<sub>4</sub> uptake at the plots with warming by OTCs of the BG site were generally lower than those recorded at ambient conditions, although not significantly. Based on the results of the repeated-measures ANOVA run across sites, there was a tendency ( $P = 0.06$ ,  $F = 3.90$ ) of higher rates of soil CH<sub>4</sub> uptake at the BG plots as compared to those recorded at the DH plots. At both sites, a seasonal trend was observed with significant variations in CH<sub>4</sub> uptake rates according to the day of sampling ( $P < 0.001$ ,  $F = 7.26$ ).

#### Linkages between CH<sub>4</sub> fluxes, soil temperature and soil moisture

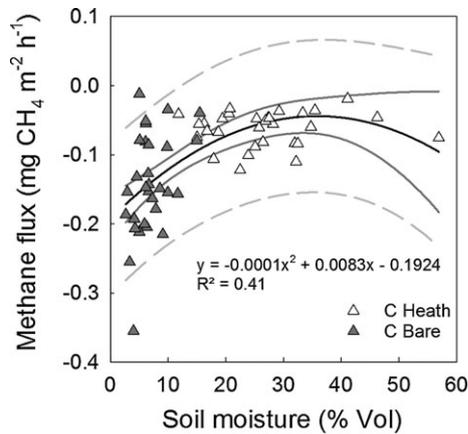
Multiple regression analysis was used to test whether soil temperature at 5 cm depth and soil moisture significantly affected soil CH<sub>4</sub> uptake rates at both ambient and increased air temperature at the BG and DH sites. At the BG site, at ambient conditions, the soil parameters explained 29% of the variance in CH<sub>4</sub> uptake rates ( $P = 0.008$ ) and soil temperature was the

main control on the process ( $P = 0.003$ ). At the DH site, soil temperature and moisture at ambient conditions explained 41% of the variance ( $P < 0.001$ ) and the changes in CH<sub>4</sub> uptake rates were mainly controlled by soil moisture ( $P = 0.004$ ). The CH<sub>4</sub> fluxes under increased air temperature by OTCs were not influenced at each site in the same manner as at ambient conditions. At the BG site, the soil parameters explained 33% of the variance ( $P = 0.004$ ) and soil moisture significantly influenced CH<sub>4</sub> uptake rates ( $P = 0.002$ ); at the DH site, the soil parameters explained 24% of the variance and soil moisture was still the main driver of soil CH<sub>4</sub> uptake, although only marginally ( $P = 0.07$ ).

Temperature sensitivity of *in situ* CH<sub>4</sub> fluxes at ambient conditions was assessed by calculating the rate of change of the reaction as a consequence of an increase in soil temperature by 10 °C ( $Q_{10}$ ). Based on the Arrhenius plot, both the DH and the BG sites showed a significant relationship between CH<sub>4</sub> fluxes and soil



**Fig. 3** Arrhenius relationship of ln-CH<sub>4</sub> oxidation rates and soil temperature at 5 cm depth measured during the growing season at the control plots of (a) dry heath (DH) and (b) bare ground (BG) sites in Blåsedalen, west Greenland. Expected ln-CH<sub>4</sub> fluxes (black line) and 95% confidence intervals (gray line) are reported in the figure.



**Fig. 4** Relationship between *in situ* soil CH<sub>4</sub> oxidation rates and soil moisture content (0–7 cm depth). The measures were recorded at control plots (C) of both dry heath (DH) and bare ground (BG) during the growing season 2014. Expected CH<sub>4</sub> oxidation rates (black line), 95% confidence intervals (gray lines) and prediction lines (dashed lines) are reported in the figure.

temperature at 5 cm depth (Fig. 3). At the DH site, soil CH<sub>4</sub> uptake had a  $Q_{10}$  of 1.7 and an activation energy ( $E_a$ ) of 36 kJ mol<sup>-1</sup> (Fig. 3a), while at the BG site, soil CH<sub>4</sub> uptake had a  $Q_{10}$  of 2.1 and an  $E_a$  of 50.5 kJ mol<sup>-1</sup> (Fig. 3b). At the fen and water-logged fen, no significant relationship was found. The sensitivity at ambient conditions of *in situ* soil CH<sub>4</sub> oxidation rates to soil moisture was further investigated by analyzing the direct relationship of the fluxes at the DH and BG site (Fig. 4). The significant relationship ( $P < 0.001$ ) fitted a quadratic regression.

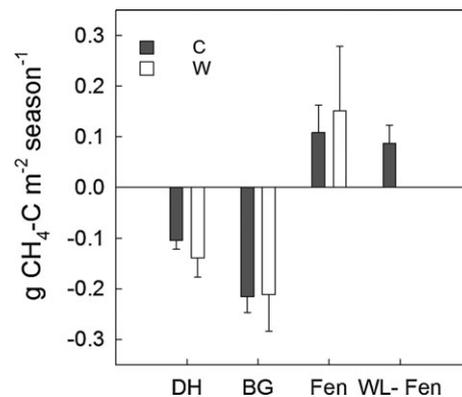
#### Net CH<sub>4</sub> budget

The net CH<sub>4</sub> budget is based on seasonal-integrated CH<sub>4</sub> fluxes calculated for all four soil types over 90- to 92-day period between the beginning of July and the end of September 2014 (Fig. 5). Over the season at ambient conditions, the DH site consumed  $0.10 \pm 0.02$  g CH<sub>4</sub>-C m<sup>-2</sup> and the BG site consumed  $0.22 \pm 0.03$  g CH<sub>4</sub>-C m<sup>-2</sup> while the wet area sites emitted  $0.10 \pm 0.03$  g CH<sub>4</sub>-C m<sup>-2</sup> to the atmosphere (fen:  $0.11 \pm 0.05$  g CH<sub>4</sub>-C m<sup>-2</sup>; water-logged fen  $0.09 \pm 0.04$  g CH<sub>4</sub>-C m<sup>-2</sup>) (Fig. 5). The seasonal-integrated CH<sub>4</sub> fluxes estimated at ambient conditions at the fen and water-logged sites differed significantly from the DH and BG sites ( $P < 0.001$ ;  $F = 15.66$ ), further the BG site tended to be a larger sink for atmospheric CH<sub>4</sub> than the DH site ( $P = 0.07$ ;  $t = 1.95$ ). Similarly, a significant effect of site was noted for the seasonal-integrated fluxes estimated at the plots with warming by OTCs ( $P = 0.04$ ;  $F = 4.32$ ), but not between the DH and the BG sites.

These rates were scaled to the landscape using the surface classification of the study site showing that the soil/vegetation cover types representative of potential sinks of atmospheric CH<sub>4</sub> cover 81% of the total area (0.15 km<sup>2</sup>), while the potential sources of CH<sub>4</sub> cover 15% (Table 2; Fig. S2). The water-logged area covered 0.19% of the total area and was considered as 'fen' in the estimation of the landscape-integrated CH<sub>4</sub> budget (Table 2). The 'shrubs' covered 4% of the total area and no data on CH<sub>4</sub> fluxes was available for this landscape unit, which therefore was excluded from the final landscape-integrated CH<sub>4</sub> budget. The resulting landscape-integrated seasonal CH<sub>4</sub> flux measured at ambient temperatures indicates that the area of Blæsedalen is overall a sink for CH<sub>4</sub> (Table 2), based on growing season measurements (July–September 2014).

The fluxes of CH<sub>4</sub> at the plots with warming by OTCs as compared to ambient conditions showed nonsignificant patterns such as an increase in CH<sub>4</sub> consumption by 33% at the DH and an increase in CH<sub>4</sub> emission by 39% at the fen sites, while at the BG site, a nonsignificant decrease in CH<sub>4</sub> consumption by 2% was observed (Table 2). Based on the landscape-integrated seasonal fluxes, the soil at plots with experimental warming by OTCs oxidized 20% more CH<sub>4</sub> than the soil at ambient conditions (Table 2), but the effect was statistically not significant ( $P = 0.94$ ;  $F = 0.01$ ). The net result of applying the same OTC with contrasting warming effects across soil types reveals that a potential increase in summer temperature could enhance the capacity of the soil to oxidize CH<sub>4</sub>.

The soil/vegetation cover classification of the study area in Blæsedalen and the related landscape-integrated CH<sub>4</sub> budget were considered representative of the entire southern part of Disko Island (Table S3), as



**Fig. 5** Seasonal-integrated CH<sub>4</sub> flux ( $n = 6 \pm$  SE) estimated at the control (C) and warming with OTCs (W) plots of: dry heath (DH), bare ground (BG), fen and water-logged fen (WL-Fen). In Blæsedalen valley, west Greenland (July–September 2014).

indicated by the classification based on the satellite scene (Table S3 and Fig. S3). In fact, the soil/vegetation cover classes which may act as potential sinks of atmospheric CH<sub>4</sub> (dry heath and barren) cover about 71% of terrestrial area of the southern part of Disko Island (24 km<sup>2</sup>), while the potential sources (fen) cover 2.5% (Table S3) as compared to 15% in Blæsedalen valley. The landscape-integrated net CH<sub>4</sub>-C budget estimated for these soil/vegetation cover classes of southern Disko (17.8 km<sup>2</sup>) confirmed that this area is a sink for CH<sub>4</sub>, which oxidized about -2.31 tonne CH<sub>4</sub>-C (-1.30 kg CH<sub>4</sub>-C ha<sup>-1</sup>) between July and September 2014.

## Discussion

### *Treatment and site effects on soil temperature and moisture*

The soil temperatures at 5 cm depth during July and September (Fig. 1a) show a typical temperature range of the active layer during the growing season in arctic ecosystems, which is characterized by positive soil temperatures only between June and September (Kane *et al.*, 1992). The BG sites were significantly warmer than DH sites, both at ambient air temperature and at plots with air warming by OTCs ( $P < 0.001$ ). As expected, the vegetation cover at the DH shaded the soil surface and kept it cooler and moister than the exposed bare soil at the BG site. Air warming by OTCs increased soil temperature by 1.1 and 1.3 °C at the BG and DH sites, but resulted in contrasting soil moisture levels. At the DH site, increased air temperature by OTCs led to lower soil moisture content than its controls as a consequence of increased evapotranspiration. A similar effect of the OTCs on soil moisture has been discussed by Dabros *et al.* (2010) who found that the soil inside the OTCs dried out due to the increased plant water uptake and therefore increased evapotranspiration. This indirect effect of the OTCs on soil moisture is not considered as a bias in our experiment as projected increases in air temperature, everything else equal, will also influence soil moisture. Hence, we conclude that the use of OTCs at the DH site was effective to mimic direct and indirect effect of summer warming. However, at the BG site, the presence of the OTCs on the bare ground caused a significant increase in soil water content compared to the control plots (Table 1). This is due to the OTCs shielding the soil from the wind and therefore reducing loss of soil water in bare plots. At the fen site, OTCs did not have a significant effect on subsurface soil temperature as compared to the DH and BG sites (Table 1). In fact, this site is characterized by a horizontal water flow during the

growing season mainly coming from melting snowdrifts to the east of the site. This flow transfers the heat laterally away from the plot surface impeding the underlying soil from heating up. In such wet ecosystems, the reduced effect of summer warming by OTCs on subsurface temperature suggests that both enzymatic reactions of CH<sub>4</sub> production and consumption are less dependent on temperature as compared to the same processes taking place in a dry tundra or bare soil.

### *Methane uptake rates*

The range of CH<sub>4</sub> uptake rates recorded at ambient conditions at the BG and at the DH sites were comparable to those measured in previous *in situ* studies carried out in similar arctic environments (Christiansen *et al.*, 2014; Emmerton *et al.*, 2014; Jørgensen *et al.*, 2015; Lau *et al.*, 2015) as well as in temperate forest soils (Smith *et al.*, 2000; Hiltbrunner *et al.*, 2012).

At both sites (BG and DH), the comparison between CH<sub>4</sub> uptake rates at the plots with warming by OTCs with those measured at ambient conditions highlighted the influence that soil water content and temperature have on CH<sub>4</sub> uptake rates especially in regard of gas diffusivity over the soil profile (Crill, 1991; Del Grosso *et al.*, 2000; Smith *et al.*, 2000). The nonsignificant trend toward lower rates at the plots with warming by OTCs of the BG site than at ambient conditions (Fig. 2b) followed the trend of soil water content, which in those plots was higher than at the control plots (Fig. 1b). That was supported by the results of the multiple linear regressions, showing that the soil water content was the limiting factor for diffusion and associated oxidation of CH<sub>4</sub> through the soil profile at both the control plots of the DH site, and at the plots with OTCs at the BG site. During the growing season, the plots characterized by low soil moisture (7–10.5% as averaged values at the control plots of BG and warming plots of DH) and a soil temperature above 10 °C (Table 1) showed the highest rates of CH<sub>4</sub> oxidation. A similar observation is reported by Chen *et al.* (2011) who investigated the temperature and moisture dependency of CH<sub>4</sub> uptake rates in temperate steppe vegetation. In that study at soil temperatures above 5 °C, the *in situ* CH<sub>4</sub> uptake rates were reduced by soil moisture <5% and >15% whereas within this soil moisture range CH<sub>4</sub> was oxidized at stable high rates.

Using OTCs on a bare soil, in an environment characterized by strong winds and low precipitation regimes, the evapotranspiration is reduced and the resulting increase in soil water content may have a negative effect on CH<sub>4</sub> oxidation. This suggests that subsurface methane availability is important and soil water content

can become a dominant factor limiting CH<sub>4</sub> uptake rates in exposed dry soils.

At Blæsedalen, the *in situ* CH<sub>4</sub> fluxes measured at the control plots of the dry heath and the bare soil sites had a temperature dependency of  $Q_{10} (2-15\text{ }^{\circ}\text{C}) = 1.7$  and  $Q_{10} (3-18\text{ }^{\circ}\text{C}) = 2.1$ , respectively. Both estimates were within the range of most biological reactions (Davidson & Janssens, 2006) and in the same range of *in situ* temperature sensitivity estimates reported by previous studies. Lau *et al.* (2015) reported  $Q_{10} (5.6-16\text{ }^{\circ}\text{C}) = 1.8$  for soil CH<sub>4</sub> uptake measured in a Canadian acidic tundra. Earlier studies investigating *in situ* CH<sub>4</sub> oxidation in forest soils also report temperature dependency values fluctuating between a  $Q_{10} (5-15\text{ }^{\circ}\text{C}) = 1.35$  in a Danish spruce forest (Priemé & Christensen, 1997), a  $Q_{10} (5-15\text{ }^{\circ}\text{C}) = 1.5$  in a mixed spruce/beechn forest in Germany (Born *et al.*, 1990) and  $Q_{10} (5-15\text{ }^{\circ}\text{C}) = 2.0$  in a temperate woodland in New Hampshire between April and July (Crill, 1991). The lower thermal sensitivity of atmospheric CH<sub>4</sub> oxidation at those sites, as compared to the estimated  $Q_{10}$  at the BG site at Blæsedalen, can be explained by limitations in gas diffusion due to seasonal changes or site-specific concentration of soil water (King & Adamsen, 1992; Priemé & Christensen, 1997). The low soil water content at the BG site did not hamper the gas diffusion as well as the substrate availability for the microbial communities involved in the oxidation of CH<sub>4</sub>. On the other hand, the effect of soil moisture on CH<sub>4</sub> uptake rates was clear at DH site which showed a lower temperature dependency as compared to the BG site (Fig. 3a, b), possibly due to the reduced gas diffusion (Fig. 4).

#### Net CH<sub>4</sub> budget of Blæsedalen Valley

The area of Blæsedalen is dominated by large areas of dry heath tundra with dispersed patches of barren ground while 15% of the total area is characterized by wet zones moderately emitting CH<sub>4</sub> to the atmosphere (Table 2; Fig. S2). The soil/vegetation cover classification of Blæsedalen, despite its relatively small area (0.15 km<sup>2</sup>), is representative of the entire southern part of Disko Island, as shown by the classification based on the satellite scene (Fig. S3 and Table S3). Accordingly, the seasonal net CH<sub>4</sub>-C budget estimated for this landscape (terrestrial part 17.8 km<sup>2</sup>) confirmed that southern Disko during summer 2014 was a sink for CH<sub>4</sub>-C (−1.30 kg CH<sub>4</sub>-C ha<sup>−1</sup>).

Thus, at the actual climatic conditions the area of Blæsedalen was a net sink for atmospheric CH<sub>4</sub> oxidizing 0.87 kg CH<sub>4</sub>-C ha<sup>−1</sup> between the 1st of July and the 31st of September (−0.0003 ± 0.001 g CH<sub>4</sub>-C m<sup>−2</sup> day<sup>−1</sup>). Our estimation of the seasonal-integrated fluxes of CH<sub>4</sub> at the BG and DH sites (Table 2)

matches well with those reported by Jørgensen *et al.* (2015) for Northeast Greenland (−0.216 ± 0.099 g CH<sub>4</sub>-C m<sup>−2</sup> for soils with water content between 5 and 25%, and −0.081 ± 0.045 g CH<sub>4</sub>-C m<sup>−2</sup> for soils with water content between 25% and 50%) showing the region to be a net CH<sub>4</sub> sink due to the dominance of dry tundra areas over wetlands and seasonally flooded grasslands. In the present study, the wet areas in the Blæsedalen landscape were weak emitters of CH<sub>4</sub> (Fig. S4) with a low seasonal-integrated flux in comparison with previous studies investigating CH<sub>4</sub> fluxes from wetlands in the high Arctic (6.05 ± 1.05 g CH<sub>4</sub>-C m<sup>−2</sup>, Tagesson *et al.*, 2012; 4.5 g CH<sub>4</sub>-C m<sup>−2</sup>, Mastepanov *et al.*, 2008). However, the low seasonal and landscape-integrated CH<sub>4</sub> emissions at Blæsedalen are in accordance with the *in situ* measurements reported by Christiansen *et al.* (2014) for a fen dominated by sedges which emitted 0.2 ± 0.02 mg CH<sub>4</sub> m<sup>−2</sup> h<sup>−1</sup>. As argued by Christiansen *et al.* (2014), the relatively young age of those wetlands with moderate amounts of soil organic carbon (SOC) accumulated in the active layer explains the low CH<sub>4</sub> emissions recorded during the growing season.

Despite the increased soil water content at the plots with warming by OTCs of the BG site, due to the presence of the OTCs, we expect that the projected increases in summer air temperature in the Arctic (Collins *et al.*, 2013) will lead to increased soil evaporation, in soils without vegetation cover and exposed to the wind. Thus, at the BG site, which at present conditions is characterized by an average soil water content <10% (Table 1), it is difficult to predict the response to a possible increase in summer temperature. In fact, as reported by King (1997), in soils that at present have water statuses at or near optimum water content CH<sub>4</sub> oxidation may decrease as a consequence of a significant drying. More *in situ* soil water manipulation experiments are therefore necessary to constrain the range of soil water levels supporting a maximum enzymatic reaction of CH<sub>4</sub> oxidation in bare soils.

Yet, given the large distribution over the Arctic of bare to dry soils with low SOC, it is important to take their strong capacity to take up CH<sub>4</sub> into account when estimating the net CH<sub>4</sub> budget of those ecosystems at present and future climatic conditions. One perspective is increasing summer air temperature and an earlier onset of the growing season which is therefore expected to last longer than present days (Schwartz *et al.*, 2006). Another perspective is increased winter air temperature, hence potentially more snow leading to a delay of the growing season and higher initial water content in the soil (Semenchuk *et al.*, 2015). Our results for the Blæsedalen valley, which is an ecosystem representative of the ice-free areas of Western Greenland, indicate that future changes in summer air temperature and soil

hydrology may increase the capacity of the soil to act as a sink for CH<sub>4</sub>-C. However, under this climatic scenario, not only the increased air temperature but also the earlier onset of the growing season should be taken into account in the seasonal landscape-integrated CH<sub>4</sub> budget. Thus, the net CH<sub>4</sub> budget reported in this study at experimentally increased summer temperature (−1.03 kg CH<sub>4</sub>-C ha<sup>−1</sup>) may be representative of the low range of the future estimates for this area, also due to the confounding effect of the OTCs on the soil moisture of the BG site. Whereas in the perspective of increased winter precipitation, the shorter and initially wetter growing season may offset or reduce the CH<sub>4</sub>-C soil sink capacity at the site. Overall, this study shows that site-specific soil water content can be used as a proxy to estimate the sensitivity of CH<sub>4</sub> dynamics to variations in soil temperature across different landscape units. However, further investigations are needed to clarify the net response of contrasting soil types to combined effects of future climate change.

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## Supporting Information

Additional Supporting Information may be found in the online version of this article:

**Figure S1.** Comparison between ambient CH<sub>4</sub> fluxes measured in light and dark chamber conditions at the dry heath.

**Figure S2.** Soil/vegetation cover classification at the Blæsedal site, Disko Island, Greenland.

**Figure S3.** Soil/vegetation cover classification of the southern part of Disko Island, Greenland.

**Figure S4.** Methane fluxes ( $n = 6 \pm \text{SE}$ ) measured at (a) the fen site and (b) the water-logged site during the growing season in 2014.

**Table S1.** Soil/vegetation classification accuracy of Blæsedal, Greenland.

**Table S2.** Soil/vegetation classification accuracy of the southern part of Disko Island, Greenland.

**Table S3.** Results of the soil/vegetation classification of the southern part of Disko Island, Greenland.