ENVIRONMENTAL RESEARCH LETTERS

LETTER • OPEN ACCESS

Large methane emission upon spring thaw from natural wetlands in the northern permafrost region

To cite this article: Changchun Song et al 2012 Environ. Res. Lett. 7 034009

View the article online for updates and enhancements.

You may also like

- <u>Comparing process-based models with the</u> <u>inventory approach to predict CH₄</u> <u>emission of livestock enteric fermentation</u> Jianan Zhang, Lan Chen, Yizhao Chen et al.
- Pan-Arctic land—atmospheric fluxes of methane and carbon dioxide in response to climate change over the 21st century Xudong Zhu, Qianlai Zhuang, Xiang Gao et al.
- $\label{eq:states} \begin{array}{l} \mbox{ Isotopic composition and emission} \\ \mbox{ characteristics of CO_2 and CH_4 in glacial} \\ \mbox{ lakes of the Tibetan Plateau} \\ \mbox{ Fangping Yan, Zhiheng Du, Tao Pu et al.} \end{array}$



This content was downloaded from IP address 3.145.143.239 on 01/05/2024 at 20:46

Environ. Res. Lett. 7 (2012) 034009 (8pp)

Large methane emission upon spring thaw from natural wetlands in the northern permafrost region

Changchun Song^{1,4}, Xiaofeng Xu^{2,3,4}, Xiaoxin Sun¹, Hanqin Tian³, Li Sun¹, Yuqing Miao¹, Xianwei Wang¹ and Yuedong Guo¹

¹ Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences, Changchun, 130012, People's Republic of China

² Climate Change Science Institute and Environmental Science Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA

³ International Center for Climate and Global Change Research and School of Forestry and Wildlife Sciences, Auburn University, Auburn, AL 36849, USA

E-mail: songcc@mail.neigae.ac.cn and xux4@ornl.gov

Received 21 May 2012 Accepted for publication 25 June 2012 Published 19 July 2012 Online at stacks.iop.org/ERL/7/034009

Abstract

The permafrost carbon-climate feedback is one of the major mechanisms in controlling the climate-ecosystem interactions in northern high latitudes. Of this feedback, methane (CH₄) emission from natural wetlands is critically important due to its high warming potential. The freeze-thaw transition has been confirmed to play an important role in annual CH₄ budget, yet the magnitude of this effect is uncertain. An intensive field campaign was carried out in the Sanjiang Plain, Northeast China to estimate the CH₄ emission in the spring freeze-thaw transition period. The observation concluded that a large CH₄ source was caused by spring thaw; the maximum hourly emission rate was 48.6 g C m⁻² h⁻¹, more than three orders of the regularly observed CH₄ emission rate in the growing season. In some sporadically observed 'hot spots', the spring thawing effect contributed to a large CH₄ source of 31.3 ± 10.1 g C m⁻², which is approximately 80% of the previously calculated annual CH₄ emission in the same study area. If our results are typical for natural wetlands in the Northern Hemisphere permafrost region, we estimate a global CH₄ source strength of 0.5–1.0 Tg C (1 Tg $= 10^{12}$ g) caused by spring that in the Northern Hemisphere permafrost region in the year 2011. Combining with available satellite and flask data, a regional extrapolation reaches a temporal pattern of CH₄ emission during 2003–2009 which is consistent with recently observed changes in atmospheric CH₄ concentration in the high latitudes. This suggests that the CH₄ emission upon spring thaw in the high latitudes might be enhanced by the projected climate warming. These findings indicate that the spring thawing effect is an important mechanism in the permafrost carbon-climate feedback and needs to be incorporated in Earth system models.

Keywords: carbon-climate feedback, methane, natural wetland, permafrost S Online supplementary data available from stacks.iop.org/ERL/7/034009/mmedia

Content from this work may be used under the terms of the Creative Commons Attribution-NonCommercial-ShareAlike 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.

⁴ These authors have equal contributions to this research.

1. Introduction

As one of the potent greenhouse gases, methane (CH_4) plays an important role in terrestrial ecosystem–climate interaction (Bousquet *et al* 2006, Keppler *et al* 2006, Mastepanov *et al*



Figure 1. (A) The spatial distribution of the permafrost in the Northern Hemisphere (the red star indicates the location of the field campaign) and (B) the distribution of the sites with identified CH_4 emission pulses across the experimental field in the Sanjiang Plain Mire Station. (In total twenty sites with high CH_4 emission pulses were identified; S21, S22 and S23 are the observational sites for our growing season monitoring reported in our previous publication (Song *et al* 2009); it should be noted that the distance and location of each site might be not accurate since most of the distances were estimated without instruments.)

2008), especially under the changing global environment (Isaksen et al 2011, Walter et al 2006, Xu and Tian 2012, Xu et al 2010). The recently observed increase in atmospheric CH₄ concentration has drawn a large amount of attention to CH₄ emission from natural wetlands in northern high latitudes (Dlugokencky et al 2009). The northern permafrost region is essential in global CH₄ cycling, especially over the trajectory of global warming (Christensen et al 2004), because the dense carbon storage in permafrost soils is a potential source of atmospheric CH₄ (Christensen et al 2004, Koven et al 2011, Schuur et al 2009), and is predicted to release more and more CH₄ to the atmosphere by 2100 (Anisimov 2007, Koven et al 2011), even though the winter season had been previously thought to be a period with a weak CH₄ emission before a number of field studies confirmed that the biological processes in winter directly make relatively large contributions to the annual CH₄ budget (Campbell et al 2005, Hao et al 2006).

Given the isolation of the frozen surface in the permafrost region, a large portion of the produced CH₄ cannot be released to the atmosphere immediately (Tokida et al 2007). During the thawing season in the following year, the release of CH₄ is expected to occur in a very short period, forming an emission outburst (Christensen et al 2004). This effect is defined as the spring thawing effect, which might partially contribute to the recently observed increase in atmospheric CH₄ concentration (Dlugokencky et al 2009, Tokida et al 2007). Although the spring thawing effect on atmospheric CH₄ has been recognized for a long time (Friborg et al 1997), the magnitude of the spring thawing effect on CH_4 emission remains far from certain (Friborg et al 1997, Tokida et al 2007). This study was conducted to estimate the effects of the spring thaw on CH₄ release from natural wetlands in the permafrost region in the Northern Hemisphere by using a combination of field observations, satellite data and flask data.



Figure 2. Observed daily CH₄ flux in the Sanjiang station (47.6N, 133.5E), Northeast China (the red line shows the captured CH₄ pulse in the spring thawing period; the black line shows the normally observed CH₄ flux in the study area; four periods are divided based on local meteorological conditions; the fluxes are shown as daily average, and hourly results are listed in table 1).

2. Data and methods

A combination of field observations, NOAA (National Oceanic and Atmospheric Administration) flask data (GLOBALVIEW-CH₄), and a satellite-derived CH₄ concentration dataset (SCIAMACHY: Scanning Imaging Absorption Spectrometer for Atmospheric Chartography) were used in this study.

2.1. Field observations

We conducted an intensive field campaign in the Sanjiang Plain, Northeast China to monitor seasonal variations in CH₄ emission from natural wetlands and estimate the CH₄ emission rate caused by spring thaw at site level (figures 1, 2 and S3 available at stacks.iop.org/ERL/7/034009/mmedia). The Sanjiang Plain is in the largest freshwater wetland area

Table 1. Observed CH_4 fluxes in the natural freshwater marshland in the Sanjiang Plain from 6 April to 11 April 2011. (The bold indicates the largest CH_4 emission rate observed in this field campaign.) Note: in total, 23 high CH_4 emission pulses were measured. More than one measurement might be carried out for one single site. For example, H1-1 and H1-2 are two samplings and measurements for one single CH_4 emission pulse.

4/6/2011	Site Observed flux (mg CH ₄ -C $m^{-2} h^{-1}$) Duration (h) Site	H1-1 193.7 2 H9	H1-2 2.7 2 H10-1	H2 1179.9 2.5 H10-2	H3 4834.6 3 H10-3	H4 18092.9 2 H12	H5 37859.0 2 H13
	Observed flux (mg CH ₄ -C m ^{-2} h ^{-1})	345.0	9503.4	48560.0	11801.7	187.6	62.3
4/0/2011	Duration (h)	1.5	1.5	1.5	3.5	1.5	1.5
4/8/2011	Site Observed flux (mg CH, $C m^{-2} h^{-1}$)	H9-2	H16-1 1707-2	H16-2 4425.0	H16-3	HI/ 1282.6	H18 8560 1
	Duration (h)	42.2 24	24	24	40.4 3	1265.0	2
4/9/2011	Site	H19-1	H19-2				
	Observed flux (mg CH ₄ -C m ^{-2} h ^{-1})	96.8	96.9				
	Duration (h)	1	1				
4/10/2011	Site	H19-3	H19-4				
	Observed flux (mg CH ₄ -C m ^{-2} h ^{-1})	133.7	98.9				
	Duration (h)	1	1				
4/11/2011	Site	H19-5					
	Observed flux (mg CH ₄ -C m ^{-2} h ^{-1})	32.2					
	Duration (h)	1					

in China; there are approximately 10400 km² of natural wetlands in this region (Song et al 2009). The study sites are located at the Sanjiang Experimental Station of Wetland Ecology, Chinese Academy of Sciences (47°35'N, 133°31'E) at an altitude representative of the natural freshwater wetland in the Sanjiang Plain (56 m a.s.l.). This region is located at the southern boundary of Northern Hemisphere permafrost (Qiu et al 2002). Since the CH₄ emission pulses usually occur in a very short time period (Tokida et al 2007), our previous observations had not captured this event. In the year 2011, we increased the observational frequency and broadened the monitoring area. Fortunately, the CH₄ emission pulses were captured (figures 1 and 2, table 1). The observed CH₄ emission pulse can be viewed in our online supporting video clip (video S1 available at stacks.iop.org/ ERL/7/034009/mmedia).

Across our sampling area, twenty sites with high CH₄ emission pulses were identified (figure 1), of which thirteen were measured at least once by using a static chamber method (Song et al 2009). We used 5-7 persons walking through the area to identify the potential bubbling spots; after each bubbling spot was identified, we measured the flux rate if labor resources allowed. Measurements for several spots were missed due to shortage of labor; we recorded the locations of these spots. Twenty-three measurements were conducted for CH₄ emission pulses (table 1). There are fourteen high emission plots in a 30 m \times 30 m study area; the measurements define CH₄ emission from a 0.5 m \times 0.5 m plot (figure 1 and table 1). So we estimated that approximately 0.39% of the area had high CH₄ pulses. This fractional coverage of high CH₄ emission was then used to estimate the spring thawing effects on CH₄ emission at regional scale.

The gas flux measurements were conducted by using static dark chamber and gas chromatography techniques (Song *et al* 2009). The 0.5 m \times 0.5 m \times 0.5 m chamber

was put on the ground which was covered by 5-10 cm surface water as sealer. A fan was installed in the sampling chamber to keep the air mixed during the gas sampling. Since the emission rate was quite large, the gas sampling process was carried out four times with a 5 min interval, rather than with a 10 min interval as in previous studies (Song et al 2009, Xu et al 2005). The gas samples were stored in syringes for less than 12 h before being measured. Gas chromatography was used to measure the gas concentrations. The gradient of gas concentration during sampling was used to calculate the gas flux between the ecosystem and the atmosphere. Positive values mean flux from the ecosystem to the atmosphere, and negative values mean flux from the atmosphere to the ecosystem (Song et al 2009). The mixing ratios of CH₄ were analyzed with a modified gas chromatograph (Agilent 4890D) equipped with a flame ionization detector (FID). N2 was used as the carrier gas with a flow rate of 30 ml min⁻¹. The CH₄ was separated with a 2 m stainless-steel 13 XMS column (60/80 mesh) with an inner diameter of 2 mm. CH₄ was directly measured by FID with an operation temperature of 200 °C. The temperature for gas separation was 55 °C. Fluxes were determined from the slope of the mixing ratio changes in four samples taken at 0, 5, 10 and 15 min after chamber closure. Sample sets were rejected unless they yielded a linear regression value of r^2 greater than 0.9.

The gas flux was calculated according to the following equation from Song *et al* (2009):

$$F = \frac{\mathrm{d}c}{\mathrm{d}t} \frac{M}{V_0} \frac{P}{P_0} \frac{T_0}{T} H_{\mathrm{c}}.$$
 (1)

Here, *F* is the CH₄ flux (mg C m⁻² h⁻¹). dc/dt is the slope of the linear regression for the gas concentration gradient with time (mol mol⁻¹ h⁻¹). *M* is the molecular mass of each gas (mg mol⁻¹). *P* is the atmospheric pressure (Pa). *T* is the absolute temperature during sampling (K). V_0 , T_0 , and P_0 are

the gas mole volume (m³ mol⁻¹), absolute air temperature (K), and atmospheric pressure under standard conditions (Pa), respectively. H_c is the height of the chamber during sampling (m).

2.2. Satellite and flask data

The satellite data (available at http://www.sciamachy.org/) were used to estimate the potential area with high CH₄ emission caused by spring thawing effects. Combining the estimate of high CH₄ emission area with the CH₄ emission rate derived from field observational data, we provided a large-scale estimation of the spring thawing effects on CH₄ emission from natural wetlands in the northern permafrost region. Flask data provided by NOAA were used to verify the increase in atmospheric CH₄ concentration over the study area and to compare with the regional estimate of the CH₄ emission from natural wetlands in the northern permafrost region. The GLOBALVIEW-CH₄: Cooperative Atmospheric Data Integration Project methane, available at ftp://ftp.cmdl.noaa. gov/ccg/ch4/GLOBALVIEW/, was used in this study with the last access on 16 June 2011. GLOBALVIEW-CH4 is a product of the Cooperative Atmospheric Data Integration Project. While the project is coordinated and maintained by the Carbon Cycle Greenhouse Gases Group of the National Oceanic and Atmospheric Administration, Earth System Research Laboratory (NOAA ESRL), it is a cooperative effort among the many organizations and institutions making high-quality atmospheric CH₄ measurements. In this study, three sites located in the northern permafrost region were selected (table S1 available at stacks.iop.org/ERL/7/034009/mmedia); the annual CH₄ concentrations measured at the three sites are shown in figure S4 (available at stacks.iop.org/ERL/7/034009/ mmedia).

2.3. Auxiliary data

The regional permafrost data were from the National Snow and Ice Data Center at http://nsidc.org/data/ggd318.html. The circumpolar permafrost and ground ice data depict the distribution and properties of permafrost and ground ice in the Northern Hemisphere (20°N-90°N). The data set classifies permafrost into four categories: continuous, discontinuous, sporadic, and isolated. The fractions of permafrost in each category are 90-100%, 50-90%, 10-50%, <10%. The relative abundance of ground ice in the upper 20 m is estimated in per cent volume (>20%, 10-20%, <10% and 0%) (Brown et al 1998). The data set also contains the location of subsea and relict permafrost. The fractional distribution of natural wetlands was retrieved from and improved on the basis of the data from Aselmann and Crutzen (1989). Finally, we generated the natural wetlands distribution in the northern permafrost region by overlaying them in the same spatial domain (figures S1 and S2 available at stacks.iop.org/ERL/ 7/034009/mmedia).

2.4. Spatial extrapolation of spring thaw-induced CH₄ emission

We processed the SCIAMACHY (satellite-derived methane concentration in air volume) data to define the grids which have potential high CH₄ emission (Bergamaschi et al 2009, Frankenberg et al 2011, Schneising et al 2011) (figure S6 available at stacks.iop.org/ERL/7/034009/mmedia). Due to technical problems, there are a lot of spatial gaps in the CH₄ concentration derived from the SCIAMACHY data (figure S6 available at stacks.iop.org/ERL/7/034009/ mmedia), especially in the period of 2005-9 (Bergamaschi et al 2007, 2009, Frankenberg et al 2011, Schneising et al 2011). The percentage of the area with high CH₄ emission was calculated (figure S8 available at stacks.iop.org/ ERL/7/034009/mmedia). Then we combined the estimated percentage area of the 'hot spots' of CH₄ emission with the spatial distribution of the natural wetlands across the northern permafrost region to estimate the spring thawing effect on atmospheric CH₄ concentration. The criteria we used to identify potential high CH₄ pulses based on satellite were: (1) the CH_4 concentration in April is 100 ppb higher than that in May; (2) the estimated CH_4 concentration in April is higher than those in any other months (non-April) at the significance level of P = 0.05. The value of 100 ppb was used because the uncertainties of the CH₄ concentration retrieved from the SCIAMACHY data were approximately 50 ppb (Frankenberg et al 2011, Schneising et al 2011), so the threshold of sufficient difference should be approximately 100 ppb on the basis of normal distribution. Based on the delineation of the spatial coverage of the area with potential high CH₄ emission, we further calculated the contribution of the spring thawing effect to the variations of atmospheric CH₄ concentration (table S2 available at stacks.iop.org/ERL/ 7/034009/mmedia).

2.5. Satellite data in capturing spring thaw-induced CH₄ emission

To evaluate the satellite data in capturing the spring thaw effects on CH₄ emission, we analyzed the basin-level CH₄ concentration derived from satellite data (figure S5 available at stacks.iop.org/ERL/7/034009/mmedia). The higher April CH₄ concentration was observed for a few years in Ob River basin, Lena River basin, Amur River basin and Mackenzie River basin (figure S7 available at stacks.iop.org/ERL/7/ 034009/mmedia). We used the Δ CH₄ (CH₄ concentration in April minus that in May) to quantify the magnitude of spring thaw effects on CH₄ emission. After 2006, a strong impact is shown as the Δ CH₄ became larger and larger in Ob River basin, Amur River basin and Mackenzie River basin (figure S7 available at stacks.iop.org/ERL/7/034009/mmedia).

3. Results and discussion

3.1. Observed high CH₄ emission pulses

The field observations showed that the CH₄ emission pulses occurred sporadically in some 'hot spots'; the hourly emission



Figure 3. Air temperature and soil temperature from January 2010 to April 2011. ((A) daily air and soil surface temperature over the time period; (B) temperature along the soil profile over the time period; the red rectangle indicates the time period of 6–8 April.)

rate could be as high as 48.6 g C m⁻² h⁻¹ via bubbling emission (figure 2, table 1), which is almost three orders of the previously observed flux rate in the growing season (Song *et al* 2009). The CH₄ emission pulse lasts for a very short time period; it could be a few hours for each outburst event and three days for the study region. The observed CH₄ bubbling in the Sanjiang Plain lasted from 6 to 8 April and after that the CH₄ emission rate decreased to the normal emission rate for the spring season (figure 2); meanwhile, the observed CH₄ outburst during 9–11 April was from one site only, thus we assumed that the time period of observed CH₄ emission pulses was 6–8 April. The total CH₄ emission via bubbling from 6 April to 8 April was summed to 31.3 ± 10.1 g C m⁻².

In order to verify the effects of spring thaw on the observed CH₄ emission pulse, we analyzed the temporal variations of air and soil temperatures in the study area (figure 3). The temperature change along the soil profile (up to 1 m depth) showed that the surface 20 cm soil profile was completely frozen from mid-November 2010 to early April of 2011 (figure 3). The soil profile completely thawed during 6-8 April, which exactly matches the time period of observed CH₄ emission pulse (figures 2 and 3). Therefore it is concluded that the observed CH₄ emission pulses were associated with the spring thaw along the soil profile. This mechanism for CH₄ emission is very similar to that of nitrous oxide during spring thaw as reported by a field study (Christensen and Tiedje 1990). The CH_4 produced beneath ice in winter is stored in the soil porosity/water; only a small portion of it is oxidized (Mer and Roger 2001, Roslev and King 1996); when the frozen surface is completely thawed in the following spring, the stored CH₄ is released to the atmosphere in a very short time period, normally in hours as observed in this study. Given the large spatial heterogeneity of soil and vegetation, the soil thawing and associated CH₄ emission pulse over the study area usually occur episodically and in some 'hot spots' as observed (figure 1 and table 1).

The high CH_4 emission rate in the spring thawing period might be caused by three reasons: winter CH_4 production (Dise 1992, Melloh and Crill 1996), lack of winter CH_4

oxidation beneath the ice due to ice's isolation of atmospheric oxygen (Melloh and Crill 1995), and the outburst of CH₄ emission (Tokida et al 2007). A number of studies have found that the CH₄ emissions in the winter season are very important (Hao et al 2006, Song et al 2009). A few studies have reported that the winter season could contribute 2.32%-4.62% to the annual budget of CH₄ (Hao et al 2006). One study even concluded a 21% contribution of winter flux to annual CH₄ budget (Dise 1992). Based on the calculated temperature sensitivity of CH₄ emission in the same study area in our previous study (Song *et al* 2009), we estimated that the CH_4 emission in the winter season contributes 18% of the annual budget. After translating to unit emission, the average CH₄ emission rate in winter is approximately 23% of that in the growing season in the Sanjiang Plain. Relatively low CH₄ oxidation in winter also contributes to the observed CH₄ outbursts. The frozen surface isolates atmospheric oxygen and impedes oxidation of the produced soil CH₄. Therefore, a large amount of CH₄ is stored before release. Finally, the short time window allows the CH₄ emission to form the observed pulses.

3.2. Quantification of the spring thawing effect on CH₄ emission

For the reasons mentioned in section 3.1, high CH_4 emission pulses are expected. To estimate the spring thawing effect on CH_4 emission, we used the following calculation to quantify the spring thawing effect on CH_4 emission:

$$F_{\rm g} = (1 - \text{Oxid}_{\rm g})q_0 H L_{\rm g} Q_{10}^{\frac{12}{10}}$$
(2)

$$F_{\rm w} = (1 - \text{Oxid}_{\rm w})q_0 H L_{\rm w} Q_{10}^{\frac{1}{10}}$$
(3)

$$F_{\rm og} = \frac{F_{\rm g}}{D_{\rm g}} \tag{4}$$

$$F_{\rm ow} = \frac{F_{\rm w}}{D_{\rm w}} \tag{5}$$

$$F_{\rm STE} = \frac{F_{\rm ow}}{F_{\rm og}} \tag{6}$$

where F_{g} is the total CH₄ emission in the growing season, and Oxid_g is the fraction of produced CH₄ being oxidized in the growing season, q_0 is the potential CH₄ production rate at $0 \,^{\circ}$ C, H is the depth of the soil profile for CH₄ production, $L_{\rm g}$ is the duration of CH₄ production in the growing season, Q_{10} is the temperature sensitivity of CH₄ production, $T_{\rm g}$ is the average temperature in the soil profile during the growing season, F_w is the total CH₄ emission in the winter season, and Oxidw is the fraction of produced CH4 being oxidized in the winter season, L_w is the duration of CH₄ production in the winter season, T_w is the average temperature in the soil profile during the winter season, F_{og} and D_{g} are the observed CH₄ emission rate and duration of the CH₄ release in the growing season, F_{ow} and D_w are the observed CH₄ emission rate and duration of the CH₄ release in the winter season, and F_{STE} , dimensionless, is the magnitude of spring effects on CH₄ emission. The values for all parameter are provided in table 2.

Parameter	Value (unit)	Ecological meaning	Reference
$T_{ m w}$	0.27 (°C)	Mean temperature of the soil profile (0–50 cm) in the winter season	This study
Tg	16.16 (°C)	Mean temperature of the soil profile (0–50 cm) in the growing season	This study
Q_{10}	2.5	Temperature sensitivity of methane production in the study site	Song <i>et al</i> (2009)
$Oxid_w$	0.1	Fraction of produced CH_4 in the soil porosity oxidized in the winter season	Mer and Roger (2001), Roslev and King (1996)
Oxid _g	0.7	Fraction of produced CH ₄ in the soil porosity oxidized in the growing season	Conrad (1996), Mer and Roger (2001)
$D_{ m w}$	2–12 (h)	Duration of the CH_4 outburst during the spring thawing period	This study
$L_{\rm w}$	2904 (h)	Length of the winter season	This study
$L_{\rm g} = D_{\rm g}$	3672 (h)	Length and duration of CH_4 production and emission over the growing season	This study

Table 2. Parameters used in the calculation of the potential CH₄ emission in the spring thaw and their ecological meanings. (All parameters are for the Sanjiang Plain, Northeast China.)

It is reported that oxidation could consume up to 90% of produced CH₄ in the soil before releasing it to the atmosphere (Oremland and Culbertson 1992). A field study in a continuously flooded rice paddy system concluded on a 70% oxidation of produced CH₄ (Mer and Roger 2001), and a study on wetland found a 72% oxidation of produced CH₄ (Freeman et al 2002). Thus, we used 70% as the fraction of CH₄ oxidation in soils in the growing season. It is reported that the oxidation of produced CH₄ in soil is highly dependent on oxygen (King 1990). Meanwhile, a field study reported winter oxidation as approximately 5%-15% of that which occurs in summer (Chanton and Liptay 2000), therefore we used 10% as the fraction of CH₄ oxidation in soil/water in the winter season (table 2), and two hours as the normal duration of CH_4 emission during the thawing period (table 1). The temperature sensitivity dependence of CH₄ production was set as $Q_{10} = 2.5$ as reported in our previous study (Song *et al* 2009). The growing season at the Sanjiang Plain lasts from 1 May to 30 September, and the winter season, the frozen season referred in this study, lasts from 1 December to 31 March the following year. Thus the length of the winter season is 121 days. F_{STE} represents the relative magnitude of hourly CH₄ emission rate during the spring thaw compared to that in the growing season. The calculation suggests the spring thawing effect could be as high as 167-1002 times. The CH₄ emission rate during spring thawing could be more than three orders of the growing season CH₄ emission at its peak. This is consistent with our field observations (table 1).

3.3. Spatial extrapolation of spring thaw-induced CH₄ emission

Combining these site observations with other data, we further estimated the spring thawing effects on CH₄ emission from the natural wetlands across the entire northern permafrost region by using two methods (figures S1 and S2 available at stacks.iop.org/ERL/7/034009/mmedia). Firstly, based on the areal representativeness of each observed CH₄ pulse in the field campaign, we estimate that approximately 0.39% of the natural wetland served as outlets for the stored CH₄ in





Figure 4. Temporal variation of year-to-year change in observed CH₄ concentration and calculated CH₄ emission from the spring thawing effect (CH₄ change is defined as changes in atmospheric CH₄ concentration at yearly time steps derived from NOAA flask data; STE: spring thawing effect).

spring across the monitoring area (figure 1, section 2.1). If the measurements and the areal percentages are representative, we estimate that the spring thawing effect was 0.5-.97 Tg C of CH₄ from natural wetlands across the northern permafrost region in the year 2011 (table S2 available at stacks.iop.org/ ERL/7/034009/mmedia).

Meanwhile, we combined the time-series column CH₄ concentration derived from SCIAMACHY data with field observations to estimate the temporal variation of spring thaw-induced CH₄ emission in the northern permafrost region (figure 4). The SCIAMACHY data were partially verified in capturing CH₄ concentration variations induced by spring thaw (section 2.5). The results showed that spring thaw-induced CH₄ emission increased from 2003 through 2008 and slightly decreased in 2009, which is consistent with temporal changes in CH₄ concentration derived from NOAA flask data except for the year 2003 (figure 4). The increase in CH₄ concentration in the year 2003 could be attributed to biomass burning in Boreal regions of Asia and North America (Dlugokencky et al 2009, van der Werf et al 2006). We acknowledge that the spring thaw-induced CH₄ emission has existed for quite a long time since it was observed more than a decade ago (Christensen and Tiedje 1990, Christensen *et al* 2004). This study confirmed that the spring thawing effect has been enhanced recently, which is probably associated with climate warming as pointed out by several recent studies (Dlugokencky *et al* 2009, Frankenberg *et al* 2011).

The contribution from the enhanced spring thawing effect to the recent increase in atmospheric CH_4 concentration might be an important positive feedback of terrestrial ecosystem–climate system caused by recent warming in the Arctic region (Phelps *et al* 1998, Tokida *et al* 2007). It might also serve as the first trigger of permafrost carbon–climate feedback; if so the positive feedback might take place earlier than previously expected.

3.4. Uncertainty and research needs

We acknowledge that there are some uncertainties that need to be addressed in our further research effort. The extrapolation might cause large uncertainties in the regional estimate since we assume constant CH₄ emission over entire north permafrost region. To improve spatial extrapolation of CH₄ emission, it is clearly necessary to develop accurate spatial data on the distribution of the hot spots of CH₄ emission from natural wetlands in the northern permafrost region. Furthermore, all the field observations were conducted during the daytime and nighttime CH₄ emission pulses were not covered. Thus the current estimate might underestimate the CH₄ emission resulting from spring thawing effects. Yet we argue that this study captures the primary CH₄ outburst because nighttime usually has lower temperature and thaw events should not occur as often as in daytime. Meanwhile, high-frequency observations are needed to accurately estimate the CH₄ emission pulses. The comparison between satellite data and flask data shows partial verification of SCIAMACHY's capacity in capturing variations in atmospheric CH₄ concentration driven by spring thaw effect-induced increase in CH₄ concentration (figure 4). In addition, integration with an atmospheric transport chemistry model would be an improvement for evaluating the spring thawing effect on atmospheric CH₄ concentration.

4. Concluding remarks

This study shows the dramatic contribution of the spring thawing effect to atmospheric CH_4 variations caused by recent Arctic warming. The extremely high CH_4 emission observed in this study confirmed that natural wetland might also emit a large amount of CH_4 via bubbling in the spring season, in addition to the observed CH_4 emission from thaw lakes (Walter *et al* 2006). This high CH_4 emission enhances the positive feedback through greenhouse gases in the Artic region (Chapin *et al* 2005). Thus Earth system models should take this spring thawing effect into consideration to make more accurate examinations of permafrost carbon–climate feedback. We anticipate that the estimated contribution of the spring thawing effect to atmospheric CH_4 will stimulate

further research into the large-scale feedback from terrestrial ecosystems to the climate system. Given the projected change in the climate system in the 21st century, the spring thawing effect might get stronger and stronger, serving as one of the most important mechanisms for permafrost carbon–climate feedback. To better understand this feedback, therefore, more efforts are needed to investigate the processes or factors responsible for the spring thawing effect on CH_4 emission.

Acknowledgments

This work has been supported by the National Natural Science Foundation of China (41125001, 40930527), National Key Basic Research and Development projects (2009CB421103), NASA IDS program (NNG04GM39C), and NASA LCLUC program (NNX08AL73G_S01). This research was sponsored in part by the US Department of Energy, Office of Science, Biological and Environmental Research (BER) program and performed at Oak Ridge National Laboratory (ORNL). ORNL is managed by UT-Battelle, LLC, for the US Department of Energy under contract DE-AC05-00OR22725. We thank Dr Oliver Schneising at the University of Bremen FB1, Germany for providing the satellite data and some constructive comments. The authors are grateful to Drs Wilfred M Post and Peter E Thornton at Oak Ridge National Laboratory for reviewing this manuscript.

Notice

The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

Author contributions

CS, XX and HT initiated the effort; XS, LS, YM, XW and YG conducted the field observations; XX performed the overall analysis of field, flask and satellite data. All authors contributed to interpreting the results and writing the paper.

© US Government

References

- Anisimov O A 2007 Potential feedback of thawing permafrost to the global climate system through methane emission *Environ. Res. Lett.* **2** 045016
- Aselmann I and Crutzen P J 1989 Global distribution of natural freshwater wetlands and rice paddies, their net primary productiity, seasonality and possible methane emissions *J. Atmos. Chem.* 8 307–58
- Bergamaschi P *et al* 2007 Satellite chartography of atmospheric methane from SCIAMACHY on board ENVISAT: 2. Evaluation based on inverse model simulations *J. Geophys. Res.* **112** D02304
- Bergamaschi P *et al* 2009 Inverse modeling of global and regional CH₄ emissions using SCIAMACHY satellite retrievals J. Geophys. Res. 114 D22301

Bousquet P, Ciais P, Miller J, Dlugokencky E, Hauglustaine D, Prigent C, Van der Werf G, Peylin P, Brunke E and Carouge C 2006 Contribution of anthropogenic and natural sources to atmospheric methane variability *Nature* 443 439–43

Brown J, Ferrians O J, Heginbottom J A and Melnikov E S 1998 Circum-arctic map of permafrost and ground ice conditions *Report* (Boulder, CO: National Snow and Ice Data Center/World Data Center for Glaciology)

Campbell J, Mitchell M, Groffman P, Christenson L and Hardy J 2005 Winter in northeastern North America: a critical period for ecological processes *Front. Ecol. Environ.* **3** 314–22

Chanton J and Liptay K 2000 Seasonal variation in methane oxidation in a landfill cover soil as determined by an *in situ* stable isotope technique *Glob. Biogeochem. Cycles* **14** 51–60 Chapin F S *et al* 2005 Role of land-surface changes in Arctic

summer warming *Science* **310** 657–760

Christensen S and Tiedje J M 1990 Brief and vigorous N₂O production by soil at spring thaw *Eur. J. Soil Sci.* **41** 1–4

Christensen T R, Johansson T, Akerman H J and Mastepanov M 2004 Thawing sub-arctic permafrost: effects on vegetation and methane emissions *Geophys. Res. Lett.* 31 L04501

Conrad R 1996 Soil microorganisms as controllers of atmospheric trace gases (H₂, CO, CH₄, OCS, N₂O, and NO) *Microbiol. Rev.* **60** 609–40

Dise N B 1992 Winter fluxes of methane from Minnesota peatlands Biogeochemistry 17 71–83

Dlugokencky E J *et al* 2009 Observational constraints on recent increases in the atmospheric CH₄ burden *Geophys. Res. Lett.* **36** L18803

Frankenberg C, Aben I, Bergamaschi P, Dlugokencky E J, van Hees R, Houweling S, van der Meer P, Snel R and Tol P 2011 Global column-averaged methane mixing ratio from 2003 to 2009 as derived from SCIAMACHY: trends and variability J. Geophys. Res. 116 D04302

Freeman C, Nevison G B, Kang H, Hughes S, Reynolds B and Hudson J A 2002 Contrasted effects of simulated drought on the production and oxidation of methane in a mid-Wales wetland *Soil Biol. Biochem.* **34** 61–7

Friborg T, Christensen T R and Segaard H 1997 Rapid response of greenhouse gas emission to early spring thaw in a subarctic mire as shown by micrometeorologial techniques *Geophys. Res. Lett.* **24** 3061–4

 Hao Q J, Wang Y S, Song C C and Huang Y 2006 Contribution of winter fluxes to the annual CH₄, CO₂ and N₂O emissions from freshwater marshes in the Sanjiang Plain *J. Environ. Sci.-China* 18 270–5

Isaksen I S A, Gauss M, Myhre G, Anthony K M W and Ruppel C 2011 Strong atmospheric chemistry feedback to climate warming from Arctic methane emissions *Glob. Biogeochem. Cycles* **25** GB2002

Keppler F, Hamilton J T G, Brass M and Rockmann T 2006 Methane emissions from terrestrial plants under aerobic conditions *Nature* 439 187–91

King G M 1990 Dynamics and controls of methane oxidation in a Danish wetland sediment *FEMS Microbiol. Lett.* **74** 309–23

Koven C D, Ringeval B, Friedlingstein P, Ciais P, Cadule P, Khvorostyanov D, Krinner G and Tarnocai C 2011 Permafrost carbon-climate feedbacks accelerate global warming *Proc. Natl Acad. Sci.* **108** 14769–74

- Mastepanov M, Sigsgaard C, Dlugokencky E J, Houweling S, Strom L, Tamstorf M P and Christensen T R 2008 Large tundra methane burst during onset of freezing *Nature* 456 628–30
- Melloh R A and Crill P M 1995 Winter methane dynamics beneath ice and in snow in a temperate poor fen *Hydrol. Process.* 9 947–56

Melloh R A and Crill P M 1996 Winter methane dynamics in a temperate peatland *Glob. Biogeochem. Cycles* **10** 247–54

Mer J L and Roger P 2001 Production, oxidation, emission and consumption of methane by soils: a review *Eur. J. Soil Biol.* 37 25–50

Oremland R S and Culbertson C W 1992 Importance of methane-oxidizing bacteria in the methane budget as revealed by the use of a specific inhibitor *Nature* **356** 421–3

Phelps A R, Peterson K M and Jeffries M O 1998 Methane efflux from high-latitude lakes during spring ice melt J. Geophys. Res. 103 29029–36

Qiu G, Zhou Y, Guo D and Wang Y 2002 Maps of Geocryological Regions and Classifications ed T Zhang (Boulder, CO: National Snow and Ice Data Center/World Data Center for Glaciology)

Roslev P and King G M 1996 Regulation of methane oxidation in a freshwater wetland by water table changes and anoxia *FEMS Microbiol. Ecol.* 19 105–15

Schneising O, Buchwitz M, Reuter M, Heymann J, Bovensmann H and Burrows J P 2011 Long-term analysis of carbon dioxide and methane column-averaged mole fractions retrieved from SCIAMANHY Atmos. Chem. Phys. 11 2863–80

Schuur E A, Vogel J G, Crummer K G, Lee H, Sickman J O and Osterkamp T E 2009 The effect of permafrost thaw on old carbon release and net carbon exchange from tundra *Nature* 459 556–9

Song C, Xu X, Tian H and Wang Y 2009 Ecosystem-atmosphere exchange of CH_4 and N_2O and ecosystem respiration in wetlands in the Sanjiang Plain, Northeastern China *Glob. Change Biol.* **15** 692–705

Tokida T, Mizoguchi M, Miyazaki T, Kagemoto A, Nagata O and Hatano R 2007 Episodic release of methane bubbles from peatland during spring thaw *Chemosphere* **70** 165–71

van der Werf G R, Randerson J T, Giglio L, Collatz G J, Kasibhatla P S and Arellano A F 2006 Interannual variability in global biomass burning emissions from 1997 to 2004 Atmos. Chem. Phys. 6 3423–41

Walter K M, Zimov S A, Chanton J P, Verbyla D and Chapin F S 2006 Methane bubbling from Siberian thaw lakes as a positive feedback to climate warming *Nature* 443 71–5

Xu X and Tian H 2012 Methane exchange between marshland and the atmosphere over China during 1949–2008 *Glob. Biogeochem. Cycles* **26** GB2006

Xu X, Song C and Song X 2005 Linking of microorganisms to CO₂, CH₄ and N₂O dynamics in Calamagrostis angustifolia rhizosphere soil *Acta Botan. Sin.* 25 182–7

Xu X, Tian H, Zhang C, Liu M, Ren W, Chen G, Lu C and Bruhwiler L 2010 Attribution of spatial and temporal variations in terrestrial ecosystem methane flux over North America *Biogeosciences* 7 3637–55