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Multi-year effect of wetting on CH_4 flux at taiga-tundra boundary in northeastern Siberia deduced from stable isotope ratios of CH_4

Ryo Shingubara[1,a], Atsuko Sugimoto[2,3,4], Jun Murase[5], Go Iwahana[2,6], Shunsuke Tei[2,3], Maochang Liang[1,b], Shinya Takano[1], Tomoki Morozumi[1], Trofim C. Maximov[7,8]

[1]Graduate School of Environmental Science, Hokkaido University, Sapporo, 060-0810, Japan
[2]Arctic Research Center, Hokkaido University, Sapporo, 001-0021, Japan.
[3]Faculty of Environmental Earth Science, Hokkaido University, Sapporo, 060-0810, Japan.
[4]Global Station for Arctic Research, Global Institution for Collaborative Research and Education, Hokkaido University, Sapporo, 060-0808, Japan.
[5]Graduate School of Bioagricultural Sciences, Nagoya University, Nagoya, 464-8601, Japan.
[6]International Arctic Research Center, University of Alaska Fairbanks, Fairbanks, 99775-7340, USA.
[7]Institute for Biological Problems of Cryolithozone, Siberian Branch, Russian Academy of Sciences, Yakutsk, 677890, Russia.
[8]Institute of Natural Sciences, North-Eastern Federal University, Yakutsk, 677000, Russia.
[a]Currently at: Graduate School of Environmental Studies, Nagoya University, Nagoya, 464-8601, Japan.
[b]Currently at: College of Horticulture and Gardening, Yangtze University, Jingzhou, 434023, China.

Correspondence to: Atsuko Sugimoto (atsukos@ees.hokudai.ac.jp)

Abstract. The response of CH_4 emission from natural wetlands to meteorological conditions is important because of its strong greenhouse effect. To understand relationship between CH_4 flux and wetting, we observed interannual variations in chamber CH_4 flux, and concentration, $\delta^{13}C$, and δD of dissolved CH_4 in summers from 2009 to 2013 at the taiga-tundra boundary in the vicinity of Chokurdakh (70° 37′ N, 147° 55′ E) on the lowland of the Indigirka River in northeastern Siberia. We also conducted incubation experiments to interpret $\delta^{13}C$ and δD of CH_4 to investigate variations in CH_4 production and oxidation processes. Methane flux showed large interannual variation in wet areas of sphagnum mosses and sedges (36–140 mg CH_4 m^{-2} day^{-1} as emission). Increased CH_4 flux was recorded in summer 2011 when a wetting event with extreme precipitation occurred. Although water level decreased from 2011 to 2013, CH_4 flux remained relatively large in 2012, and increased further in 2013. Concurrently, dissolved CH_4 concentration rose by one order of magnitude from 2011 to 2012, and increased further from 2012 to 2013. Large variations in $\delta^{13}C$ and δD of dissolved CH_4 were observed in 2011, and less variations were seen in 2012 and 2013, suggesting both enhancement of CH_4 production and depression of CH_4 oxidation. These multi-year effects of wetting on CH_4 dynamics may have been caused by continued soil reduction across multiple years after wetting, which suggests that duration of water saturation in the active layer can be important for predicting CH_4 emission following a wetting event in permafrost ecosystem.

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1 Introduction

Atmospheric CH_4 has an important greenhouse effect (Myhre et al., 2013). The greatest source of atmospheric CH_4 is emission from natural wetlands, which is considered to be the main driver of interannual variation in global CH_4 emission, depending on meteorological conditions such as air temperature and precipitation (Ciais et al., 2013). For instance, Dlugokencky et al. (2009) attributed observed high growth rates in atmospheric CH_4 concentration in 2007–2008 to high CH_4 emissions from natural wetlands that resulted from anomalous high temperatures in the Arctic and greater than average precipitation in the tropics. Atmospheric CH_4 has been increasing since 2007 to the present (Nisbet et al., 2014).

Methane flux from wetland soil to the atmosphere (we define positive flux values as CH_4 emissions) is determined by three processes: CH_4 production, oxidation, and transport (Lai, 2009). Methane is produced as an end product of organic matter decomposition by strictly anaerobic *Archaea* (methanogens) mainly via hydrogenotrophic methanogenesis ($4H_2 + CO_2 \rightarrow CH_4 + 2H_2O$) or acetoclastic methanogenesis ($CH_3COOH \rightarrow CH_4 + CO_2$). In the aerobic zone of soil, CH_4 is oxidized to CO_2 with O_2 by methanotrophic bacteria, which reduces CH_4 emission to the atmosphere. Underground CH_4 is transported to the atmosphere via bubble ebullition, diffusion through soil layers and surface water, and via aerenchyma of vascular plants.

High water levels can lead to development of reductive conditions in soil, which can promote CH_4 production or depress CH_4 oxidation, both leading to increases in CH_4 flux (Lai, 2009). This is reflected in a widely observed positive relationship between water level and CH_4 flux, found in a meta-analysis across the circum-Arctic permafrost zone (Olefeldt et al., 2013). Meanwhile, Desyatkin et al. (2014) observed increases in CH_4 flux in the second consecutive year of flooding at a thermokarst depression in boreal eastern Siberia. Treat et al. (2007) reported observations at a temperate fen in northeastern USA showing that high water level coincided with high CH_4 flux in interannual variations, whereas water level correlated negatively with CH_4 flux over shorter timescales, namely as monthly means or individual measurements. These observational results imply that wetting is not directly related to CH_4 flux in wetlands. To understand the relationship between wetting and CH_4 flux, it is necessary to assess the underlying processes.

Stable isotopes of CH_4 have been used to estimate production pathways of CH_4 (Sugimoto and Wada, 1993; Sugimoto and Wada, 1995; McCalley et al., 2014; Itoh et al., 2015), the fraction of oxidized CH_4 versus produced CH_4 (Marik et al., 2002; Preuss et al., 2013) and to study mechanisms of CH_4 transport by plants (Chanton, 2005). When CH_4 in soil is lost by oxidation or diffusion, both $\delta^{13}C$ and δD of the remaining CH_4 increase. While the hydrogen isotope ratio increases more than that of carbon in oxidation, both ratios are considered to change to the same extent in diffusion. Thus it is useful to analyze both carbon and hydrogen isotopes of CH_4 to distinguish the effects of these processes (Chanton, 2005).

The taiga-tundra boundary ecosystem (or transition zone) has vegetation types of both taiga and tundra. Liang et al. (2014) reported that the distribution of vegetation types at the taiga-tundra boundary on the lowland of the Indigirka River in northeastern Siberia is controlled by soil moisture, which corresponds to microtopography. Larch, the dominant tree species of taiga forest in eastern Siberia, grows on high and dry microsites, while wetland vegetation of sphagnum mosses and

sedges, typically seen in wet tundra (van Huissteden et al., 2005; van der Molen et al., 2007), dominates low and wet microsites. Thus, it is reasonable to assume that the taiga-tundra boundary ecosystem has various microsites in terms of interannual variation in soil wetness conditions: constantly wet microsites, constantly dry microsites, and microsites with large interannual variation. Hence, this ecosystem is a suitable area to evaluate the processes controlling CH_4 flux in relation to soil wetting and/or drying on an interannual timescale.

In this study, we observed interannual variations in chamber CH_4 flux, and concentration, $\delta^{13}C$, and δD of dissolved CH_4 in summer from 2009 to 2013 at the taiga-tundra boundary on Indigirka River lowland in northeastern Siberia to understand relationships between CH_4 flux and environmental factors. We also conducted incubation experiments to investigate how δ values of CH_4 reflect CH_4 production and oxidation processes in this ecosystem. In 2011, a wetting event with a prominent amount of precipitation occurred. We focused in particular on the responses of CH_4 flux and those underlying processes to the unusual wetting event.

2 Methods

2.1 Study sites

The taiga-tundra boundary on the lowland of the Indigirka River was selected as our study area and observations and samplings were conducted at three sites (V: Verkhny Khatistakha, K: Kodac, and B: Boydom) in the vicinity of Chokurdakh (70° 37′ N, 147° 55′ E), Republic of Sakha (Yakutia), Russia (Fig. 1 and Table 1). The study sites are located in the Russian Arctic with an annual mean air temperature of −13.9 °C and annual mean precipitation of 208 mm for the period 1950–2008, according to the Baseline Meteorological Data in Siberia Database (Yabuki et al., 2011). Sites V, K, and B are alongside the Indigirka River or its tributary, and tree density decreases from V to B.

These study sites are underlain by continuous permafrost (Iwahana et al., 2014). Normally, snowmelt and the start of active layer thawing occur from the latter half of May to the first half of June and growing season is from the end of June to the beginning of August. Air temperature and surface soil temperature (10 cm depth) peak in July, whereas the maximum thaw depth occurs from the latter half of August to the first half of September. The freezing of active layer starts from the latter half of September to October and whole active layer freezes up from November to December.

Observations of CH_4 flux were conducted at seven points with three typical vegetation types, as summarized in Table 1. These vegetation types are patchily distributed (Liang et al., 2014). Larch trees are found with shrubs (willows or dwarf birches) in dry areas with relatively high levels of micro-relief, where green mosses cover the surface (hereafter termed tree mounds). On the other hand, ground with relatively low levels of micro-relief form wetlands covered by sphagnum mosses and sedges. In this study, observation points covered predominantly by sphagnums are termed ‘sphagnum’ with site code K, and those with sedges are termed ‘sedge’ with one of the site codes V, K, or B (Table 1). Snapshot measurements of volumetric water content in the surface soil layer (0–20 cm) by TDR (time domain reflectometry; TDR-

341F, Fujiwara Scientific Company, Japan) showed that tree mounds were drier than wet areas as will be described in Sect. 3.1 (Table 1).

2.2 Field observations and samplings

Methane flux was observed by the chamber method in each of the typical vegetation types described in Sect. 2.1 in the summers from 2009 to 2013. A transparent cylindrical flux chamber (acrylic resin, base area 4.7×10^2 cm^2, height 25 cm) was installed on the ground. The headspace gas of the chamber (ca. 12 L) was circulated with a pump (ca. 1 L min^{-1}). The chamber was principally closed for 30 min and headspace gas was sampled at 0 min, 15 min, and 30 min after chamber closure. Samples were kept in pre-evacuated glass vials with butyl rubber septa. To minimize soil disturbance, we stepped on wooden boards at observation points. In 2009 and 2010, CH_4 flux measurements were conducted in the latter half of July, and from 2011 to 2013, observations were conducted continuously from early July to the end of July or early August. For all of these years, the observation period included the warmest season when CH_4 emission was expected to be the most active (Table S1).

For measurements of dissolved CH_4, surface water and soil pore water were sampled at wet areas from 2011 to 2013. Surface water was directly taken up by a 50 mL plastic syringe with a three-way cock attached to its tip, whereas soil pore water was sampled to a 50 mL syringe with a three-way cock through a plastic tube inserted in the soil. Soon after collecting water samples, dissolved CH_4 was extracted inside the syringes by the headspace method, after adding 15–35 mL of the atmosphere prepared in a 10 L aluminum bag. This atmosphere was analyzed for CH_4 concentration and isotopic compositions later as a background sample. Syringes were vigorously shaken for one minute and left standing for five minutes to ensure equilibration. Finally, headspace gas and background air samples were preserved in 10–20 mL pre-evacuated glass vial with rubber septa and 100–300 mL glass bottle with O-ring stopcocks, respectively.

Concurrently with each flux measurement, soil temperature around the flux chamber was measured by a temperature sensor in an ORP electrode (PST-2739C, DKK-TOA Corporation, Japan) with an ORP meter (RM-30P or RM-20P). After flux measurement, thaw depth was observed on the same day around each chamber by inserting a steel rod to the ground. Water level was also measured around each chamber in wet areas since 2011 with a scale, and expressed in height relative to the ground surface.

2.3 Incubation experiments and microbial community

Incubation experiments were conducted to estimate $\delta^{13}C$ and δD of produced CH_4 and fractionation factors of CH_4 oxidation for carbon and hydrogen isotopes. For CH_4 production experiments, surface soil was sampled in all the wet areas in Table 1 (sedge_V, sphagnum_K, sedge_K, and sedge_B) in summer 2013. Samples were taken from 10 cm depth at each sampling point. To observe vertical variation in δ values of produced CH_4 within the thaw layer, we also collected samples from two

additional depths (20 cm and 30 cm) at sedge_K, which is a location typical of the taiga-tundra boundary region. These samples were organic layers, except for the samples from 30 cm, which were the top of the mineral layer.

About 10 mL of soil was directly transferred into plastic syringes (60 mL maximum capacity) along with in situ water (about 50 mL) to prevent the sample from being oxidized by the atmosphere. Syringes were preserved in water to ensure no leakage and were immediately pre-incubated for 4–8 days, then incubated in triplicate for 8 days. Pre-incubation and incubation temperatures were set at 5 °C. We also incubated syringes at 10 °C for samples from 10 cm depth at sedge_K to investigate temperature dependence of δ values of produced CH_4. Water in the syringes was sampled at the start and end of incubation, and dissolved CH_4 was extracted by the headspace method described in Sect. 2.2.

To interpret CH_4 production in the incubation experiment (Sect. 2.3), phylogenic composition of methanogen in surface soil was additionally analyzed in 2016. In July, soil samples at 10 cm depth were collected to 10 mL plastic tubes in triplicate in the same four wet areas as the anaerobic incubation experiment, and kept frozen until analysis. DNA was extracted as described in the caption of Figure S3.

For CH_4 oxidation, surface organic layers (0–13 cm depth) were sampled at sphagnum_K and sedge_K in July 2012, and then kept in a refrigerator until the experiment (6 days). Samples were cut into small pieces and mixed well with air. Ten grams of each sample (about 40 mL) were transferred into plastic syringes (maximum 120 mL) in quadruplicate. Approximately 80 mL of air and 0.2–2 mL of 25% CH_4 gas were added to each syringe so that the total volume in the syringe was 120 mL and the headspace CH_4 concentration was 5.0×10^2–4.8×10^3 ppm. Syringes were preserved in water and incubated at 8 °C for 8 days. Headspace gas was sampled on day 0, day 4 and day 8 into 20 mL pre-evacuated glass vials with rubber septa.

2.4 Sample analysis and data processing

Methane concentrations in air samples were analyzed on a gas chromatograph (HP6890 series G1530A, Hewlett Packard, USA) equipped with a flame ionization detector and a CP-carboplot capillary column (Varian, USA). Methane flux was calculated from CH_4 concentration in chamber headspace by linear regression of two to three concentration values against the time elapsed since chamber closure. The detection limit of CH_4 flux in each observation was calculated as 0.8–2.4 mg CH_4 m^{-2} day^{-1}, based on whether the increment of chamber CH_4 concentration during the observation was significant relative to the precision of CH_4 concentration analysis. Regression r^2 was calculated (formally) as ≥ 0.87, when the flux value was larger than 2 mg CH_4 m^{-2} day^{-1}. Dissolved CH_4 concentrations were obtained from calculation of the headspace method where equilibrations of CH_4 between gas and water phases are described by the Bunsen absorption coefficient of CH_4 (Yamamoto et al., 1976).

Carbon and hydrogen isotope ratios of dissolved CH_4 in situ and CH_4 samples in both incubation experiments were analyzed on a GC/GC/C/IRMS (modified after Sugimoto, 1996) —which is a continuous flow system consisting of two gas chromatographs, a combustion reactor, and an isotope ratio mass spectrometer (MAT253, Thermo Fisher Scientific, USA)

—and on a GC/GC/P/IRMS (P: pyrolysis in a HTC reactor of GC IsoLink, Thermo Fisher Scientific), respectively. When calculating $\delta^{13}C$ and δD of dissolved CH_4, the effect of CH_4 in background air was removed based on the mass balance. In the aerobic incubation experiments, the fractionation factors of CH_4 oxidation for carbon and hydrogen were calculated from the following Rayleigh distillation equation:

$$\ln\frac{R_t}{R_0} = \left(\frac{1}{\alpha_{ox}} - 1\right)\ln\frac{[CH_4]_t}{[CH_4]_0}, \quad (1)$$

where R_0 and R_t represent isotope ratios under initial conditions and at time t, respectively; α_{ox} is the fractionation factor for CH_4 oxidation (defined so that $\alpha_{ox} > 1$); and $[CH_4]_0$ and $[CH_4]_t$ are CH_4 concentrations under initial conditions and at time t, respectively.

All statistical tests for detecting differences in CH_4 fluxes or dissolved CH_4 concentrations were conducted using R software (version 3.3.3). Mann-Whitney's U test was applied to compare magnitudes between two years and Steel-Dwass's multiple comparison test was used to compare magnitudes among three years or more.

2.5 Meteorological data

Air temperature and precipitation observed at a weather station in Chokurdakh (WMO station 21946) were used to investigate interannual variation in meteorological conditions during our observations of CH_4 flux. These data were available from GHCN-Daily, a NOAA database (Menne et al., 2012a, 2012b).

3 Results

3.1 Environmental factors

Soil wetness conditions and thaw depth differed among vegetation types (Table 1). Tree mounds had lower surface water content (2.1–17%) than wet areas (42–48%). Among the two types of wet areas, the water level was lower in wet area of sphagnum mosses than those of sedges (Fig. 2). Wet areas of sedges experienced water levels higher than the ground surface (defined as 0 cm), reaching more than 10 cm above the ground surface. Corresponding with soil water content, the thaw depth was shallower at dry tree mounds (20–23 cm), and deeper in wet areas (31–56 cm). The overall average thaw depth observed on days when flux measurements were taken from early July to early August was 31 cm (n = 78).

During the observation period (2009–2013), monthly mean air temperature in July showed large interannual variation compared to other months, ranging from 8.5 to 15.5 °C (Fig. 2). Monthly mean air temperature was highest in July 2010, while the monthly precipitation was low (8 mm), showing dry weather conditions in this period. In contrast, precipitation in July 2011 was extremely high (94 mm) with a milder temperature (13.0 °C), causing unusual wetting. High precipitation continued in August (74 mm) and September (67 mm) of this year. Corresponding to this heavy rainfall, water

levels were also high in 2011, showing a clear decrease from 2011 to 2013 in wet areas of sedges ($p < 0.005$). Water levels also declined in wet area of sphagnum mosses, with values of −9 cm, −10 cm, and less than −12 cm in 2011, 2012, and 2013, respectively.

3.2 CH_4 flux and dissolved CH_4 concentration

Obtained CH_4 flux showed clear spatial and interannual variations (Fig. 3). Individual flux measurements (n = 143 in total) are summarized as mean values for the main summer seasons. From 2011 to 2013, continuous flux observation (Table S1) was conducted in combination with dissolved CH_4 analysis, and the interannual variation during this period will be discussed in detail.

As regards the flux spatial variation, tree mounds had consistently small values around the detection limit including all the measurements (−4.9 to 1.9 mg CH_4 m^{-2} day^{-1}), while wet areas showed active CH_4 emissions. From 2009 to 2013, the CH_4 flux in wet areas showed large interannual variation ranging from 36 to 140 mg CH_4 m^{-2} day^{-1}. The flux increased in 2011 when the wetting event occurred, then remained relatively large in 2012 (compared to 2009 and 2010). Interestingly, the flux became even higher from 2011 and 2012 to 2013 ($p < 0.05$). No statistically significant correlation was found when CH_4 flux was plotted against soil temperature (10 cm depth), thaw depth, and water level using all the data from wet areas (Fig. S1).

In addition to CH_4 flux, dissolved CH_4 concentration increased after the wetting event in 2011 (Fig. 4). From 2011 to 2012, CH_4 concentration in soil pore water at 10 cm depth (Fig. 4b) exhibited a sharp rise by one order of magnitude ($p < 0.005$). It remained high from 2012 to 2013, while the concentrations in surface water and that at 20 cm depth (Fig. 4a and c) increased significantly over the same period ($p < 0.05$). No significant difference was observed in the concentration at 30 cm depth between 2012 and 2013. In terms of vertical variation, the concentration in surface water was lower than that in soil pore water (10, 20, and 30 cm depth).

3.3 $\delta^{13}C$ and δD of dissolved CH_4 in situ

Variability of both $\delta^{13}C$ and δD of dissolved CH_4 was smaller in deeper layers, showing different patterns between $\delta^{13}C$ and δD, and across years (Fig. 5). The $\delta^{13}C$ of dissolved CH_4 had similarly large ranges (−68‰ to −40‰) in surface water, and at 10 cm and 20 cm depths, compared to a small range (−53‰ to −46‰) at 30 cm depth. The δD of dissolved CH_4 was variable only in surface water (−415‰ to −308‰) and at 10 cm depth (−417‰ to −341‰), whereas it had a constant value of around −408‰ at 20 cm and 30 cm depths. Additionally, $\delta^{13}C$ values approached to a relatively high value (about −50‰) along depth, while δD values converged to almost its lowest value. In terms of interannual variations in $\delta^{13}C$ and δD of dissolved CH_4 from 2011 to 2013, both $\delta^{13}C$ and δD values in surface soil pore water (10 cm depth) were scattered more widely in

2011, showing standard deviations (SD) of 6.6‰ and 24‰, respectively, whereas their ranges were smaller in 2012 and 2013 (SD: 3.3‰ and 17‰ at maxima, respectively).

As shown in Fig. 6, convergence of $\delta^{13}C$ and δD of dissolved CH_4 was associated with dissolved CH_4 concentration. The $\delta^{13}C$ and δD values of dissolved CH_4, including surface water and 10 cm depth, converged to those at deeper layers at high CH_4 concentrations ($\delta^{13}C = -50 \pm 5‰$ and $\delta D = -408 \pm 5‰$ at > 200 μmol CH_4 L^{-1}).

3.4 Soil incubation experiments

In the anaerobic incubation experiment, the CH_4 production rate was different among sampling points (Fig. 7); the rate was higher for sedge_K and sedge_B than sphagnum_K and sedge_V. In sedge_K, the sampling point tested in detail, the production was more rapid for shallower soil layers among 10 cm, 20 cm and 30 cm deep, while no difference in the rate was found between two incubation temperatures (5 °C and 10 °C). When the CH_4 production rate was high, the $\delta^{13}C$ and δD values of produced CH_4 were less variable irrespective of sampling point, sampling depth, or incubation temperature. The $\delta^{13}C$ value of produced CH_4 at a high production rate (> 0.26 μmol day^{-1} gdw^{-1}) was −55 ± 4‰ (n = 12). Similarly, δD under rapid CH_4 production was −410 ± 9‰ (n = 12). These $\delta^{13}C$ and δD values of CH_4 obtained under rapid production were mostly comparable with the δ values of dissolved CH_4 that converged in deep soil layers in situ ($\delta^{13}C = -50 \pm 2‰$ at 30 cm depth and $\delta D = -408 \pm 5‰$ at 20–30 cm depth; Fig. 5c and d), although $\delta^{13}C$ values in the incubation experiment were slightly lower than those in situ.

In the CH_4 oxidation experiment, CH_4 concentration in headspace declined continuously in every sample (Fig. S2). As CH_4 oxidation proceeded, both δD and $\delta^{13}C$ of the remaining CH_4 increased with a linear relationship between them (Fig. 8). Observed slope $\Delta(\delta D)/\Delta(\delta^{13}C)$ was 11, indicating much larger fractionation for hydrogen than carbon, regardless of vegetation types in wet areas (sphagnum or sedge). The hydrogen isotope fractionation factors of CH_4 oxidation calculated from the data shown in Fig. 8 were 1.25 and 1.16 for sphagnum and sedge wet areas, respectively, while carbon isotope fractionations were 1.021 and 1.015, respectively.

4 Discussion

4.1 CH_4 flux at tree mounds and wet areas at the taiga-tundra boundary on Indigirka River lowland

Methane flux observed in our study was clearly larger at wet areas than dry tree mounds (Table 1, Fig. 3). Such a difference in CH_4 flux between wetland vegetation and dry areas with trees or shrubs is generally observed (van Huissteden et al., 2005; van der Molen et al., 2007; Flessa et al., 2008) and is consistent with the fact that CH_4 production requires reductive conditions in soil (Conrad, 2007). Our CH_4 flux in wet areas was comparable to that reported from northern wetlands (Olefeldt et al., 2013; Turetsky et al., 2014). In forests, many studies have observed CH_4 absorption instead of emission

(King et al., 1997; Dutaur and Verchot, 2007; Flessa et al., 2008; Morishita et al., 2014). However, our observations at tree mounds rarely found neither CH_4 absorption nor emission. In addition, CH_4 was not consumed even under O_2- and CH_4-rich conditions in incubation experiments of tree mound soil from site K (Murase et al., 2014), indicating that a lack of methanotrophic bacterial activity limited CH_4 absorption. Our results show that the CH_4 emission from wet areas is expected to make a greater contribution to ecosystem-scale CH_4 exchange at the taiga-tundra boundary on Indigirka River lowland.

4.2 Responses of CH_4 flux, production, and oxidation to the wetting event

In 2009 and 2010 the CH_4 emission in wet areas was low (Fig. 3), even at relatively high soil temperature in 2010 (Fig. S1), under dry conditions which was not directly observed in this study. The wetting event in 2011 initiated the high CH_4 emission that continued up to 2013 despite of decreasing water levels (Fig. 2). Moreover, further flux increase was observed in 2013, accompanying build-up of dissolved CH_4 (2011– 2013) as shown in Fig. 4.

These interannual variations from 2011 to 2013 could be caused by the development of soil reductive conditions over multiple years after the wetting event. Soil reductive conditions may have been formed to some extent due to the extreme precipitation in the summer of 2011 (Fig. 2). The surface soil layer, particularly under high water levels, could eliminate O_2 from the soil pores due to water saturation. Remarkably, these soil reductive conditions may be preserved by freezing of soil throughout the following winter. Moreover, a surface soil layer saturated with ice could have prevented snowmelt water (rich in O_2) from infiltrating the soil during the spring thaw season of 2012. These processes would have led the continuation of soil reductive conditions created, which was created in summer 2011, to 2012. Through further decomposition of soil organic matter with consumption of O_2, soil reductive conditions may have been exacerbated in the water-saturated soil layer to a greater extent in summer 2012. Water in the saturated soil layer may be retained without exchange for a relatively long time in our study sites, because lateral runoff is assumed to be small in the flat floodplain that comprises Indigirka River lowland. In addition, deep percolation loss is prevented by the impermeable permafrost layer. From summer 2012 to summer 2013, soil reductive conditions may have been similarly prolonged, especially in the deep soil layer, despite the decrease in water level from summer 2011 to summer 2013. This development of soil reductive conditions from 2011 to 2013 could have promoted CH_4 production and/or depressed CH_4 oxidation, which may explain the increase in dissolved CH_4 concentration and CH_4 flux in wet areas after the wetting event until 2013 (Fig. 3 and 4).

In 2011, $\delta^{13}C$ and δD of dissolved CH_4 (10 cm depth) were scattered broadly across a wide range, whereas in 2012 and 2013 the ranges were reduced and they were clustered around a high $\delta^{13}C$ value (−50‰) and low δD value (−408‰; Fig. 5b). Considering that δD increased much more rapidly than $\delta^{13}C$ in our oxidation experiment (Fig. 8), δD can be considered as a sensitive indicator of CH_4 oxidation. In contrast, $\delta^{13}C$ is not a good indicator because its fractionation factor of CH_4 oxidation (1.015–1.021) was similar to that of CH_4 diffusion (1.019; Chanton, 2005), and the effects of CH_4 oxidation and diffusion cannot be discerned by $\delta^{13}C$. Additionally, δD of dissolved CH_4 (Fig. 5) was clearly lower in deeper layers (20 cm and 30 cm depths) than in shallow layers (surface water and 10 cm depth), which indicates δD showed CH_4 oxidation in situ

as well, because shallow layers are close to atmospheric O_2. Thus, δD values at 10 cm in 2011 were scattered broadly compared with those in 2012 and 2013 which were gathered around a low value, suggesting that CH_4 oxidation was still active in the surface soil layer during the year of the wetting event (2011) and was subsequently depressed in 2012 and 2013. In the CH_4 production incubation experiment, $\delta^{13}C$ and δD of produced CH_4 were less variable at higher production rates ($\delta^{13}C = -55 \pm 4‰$ and $\delta D = -410 \pm 9‰$ as in Fig. 7). Analogously, those of dissolved CH_4 in situ converged at high CH_4 concentration towards similar values ($\delta^{13}C = -50 \pm 5‰$ and $\delta D = -408 \pm 5‰$ in Fig. 6). This suggests that δ values of produced CH_4 became almost constant under rapid CH_4 production in situ and that the convergence of δ values of dissolved CH_4 observed in situ reflect rapid CH_4 production. Hence, the narrow ranges of $\delta^{13}C$ and δD values of dissolved CH_4 at 10 cm depth observed in 2012 and 2013 ($\delta^{13}C$: around −50‰ and δD: around −408‰, Fig. 5b) suggest enhanced CH_4 production compared to the wetting year (2011).

Multi-year effects of wetting on CH_4 flux through soil reduction process have been previously proposed by Kumagai and Konno (1998) and Desyatkin et al. (2014) as one possible factor for explaining the increase in CH_4 flux after wetting. Kumagai and Konno (1998) reported a CH_4 flux increase at a temperate rice field in Japan one year after the rice field was irrigated and restored from a farmland that had been drained for eight years. Desyatkin et al. (2014) observed flux increases at a thermokarst depression in boreal eastern Siberia in the second consecutive year of flooding after large volumes of precipitation. On the other hand, studies at natural wetlands in northeastern USA (Smemo and Yavitt, 2006; Treat et al., 2007; Olson et al., 2013) and southern Canada (Moore et al., 2011) reported that interannual variations of CH_4 flux correspond with those of water level and/or precipitation in the current year. In our study area, multi-year soil reduction may be important because soil temperature is generally lower than 11 °C (10 cm to 30 cm depth; Fig. S1a and Iwahana et al., 2014) due to a shallow active layer underlain by permafrost. Therefore, decomposition of organic matter can be slow (Treat et al., 2015), which would slowly decrease soil redox potential, allowing it to remain relatively high in the first year of wetting.

4.3 Process behind CH_4 production response

When CH_4 production is initiated after the onset of anoxia in rice paddy soil, it first occurs via hydrogenotrophic methanogenesis, and then by both hydrogenotrophic and acetoclastic methanogeneses, which increases CH_4 production rate (Conrad, 2007). Afterwards, the ratio of acetoclastic to hydrogenotrophic methanogenesis can stabilize (Roy et al., 1997). Considering that this ratio is an important control on isotopic compositions of produced CH_4, stabilization of production pathways might explain the convergence in δ values of dissolved CH_4 at our study sites under high CH_4 concentration (Fig. 6), and the reduced variability of δ values of produced CH_4 in our experiment under rapid production (Fig. 7). As acetoclastic methanogenesis leads to higher $\delta^{13}C$ in produced CH_4 than hydrogenotrophic methanogenesis (Sugimoto and Wada, 1993), acetoclastic methanogenesis may have been activated when dissolved CH_4 concentration or CH_4 production rate were high in our study. Therefore, the high and less-variable $\delta^{13}C$ values observed at 10 cm depth in 2012 and 2013 (Fig.

5b) suggest a greater contribution from acetoclastic methanogenesis compared to the wetting year (2011). Similarly to findings from rice paddy soil (Conrad, 2007), acetoclastic methanogenesis may have experienced delayed activation after anoxia in 2011, which could also have promoted CH_4 production in 2012 and 2013.

Microbial community analysis by amplicon sequence of 16S rRNA gene was applied to soil samples at 10 cm depth at the same locations as the CH_4 production incubation experiment (Fig. S3, Table S3). Soils with high rates of CH_4 production and high $\delta^{13}C$ of CH_4 produced in incubation (sedge_K and sedge_B as in Fig. 7) had higher proportion of acetoclastic methanogens in the order Methanosarcinales than those with low CH_4 production rates and low $\delta^{13}C$ of produced CH_4 (sphagnum_K and sedge_V). This supports the interpretation that the ratio of acetoclastic to hydrogenotrophic methanogenesis controlled the $\delta^{13}C$ of produced CH_4 in incubation.

5 Concluding remarks

At the taiga-tundra boundary on Indigirka River lowland, we observed an increase in CH_4 flux in wet areas following the wetting event in 2011, and a further increase in flux in 2013. Our results show interannual variations in $\delta^{13}C$ and δD of dissolved CH_4, and when compared with our incubation experiments, suggest both promotion of CH_4 production and depression of CH_4 oxidation in 2012 and 2013 in comparison to 2011. This promotion of production could be partly caused by activation of acetoclastic methanogenesis following soil reduction after the wetting event. Analyses of isotopic compositions of CH_4 in situ and in incubation experiments can be combined to investigate the effects of CH_4 production and oxidation on these isotopic compositions, and to clarify the relationship between CH_4 flux and wetting. In future, measuring the $\delta^{13}C$ of dissolved CO_2 would be useful to further validate activation of acetoclastic methanogenesis. Additionally, analyzing δ values of emitted CH_4 would allow discussion of the response of CH_4 transport to wettings.

In recent years, strong storm activity and wetting events in terrestrial ecosystems have been observed in northern regions (Iijima et al., 2016). A wetting event at the taiga-tundra boundary can switch microsites with large interannual variations in soil wetness conditions to significant CH_4 sources; we observed clear increases in CH_4 flux at wet areas (sphagnum_K and sedge_V) after the wetting event. In order to predict CH_4 flux following a wetting event in permafrost ecosystem, our results show the multi-year process of soil reduction affected by the duration of water saturation in the active layer.

Author contribution

Ryo Shingubara and Atsuko Sugimoto designed the experiments and Ryo Shingubara carried them out. Go Iwahana, Shunsuke Tei, Liang Maochang, Shinya Takano, Tomoki Morozumi, and Trofim C. Maximov helped sampling, data

collection in situ, and preparing resources for the fieldwork. Jun Murase contributed to laboratory analysis. Ryo Shingubara prepared the manuscript with contributions from all co-authors.

Competing interests

The authors declare that they have no conflict of interest.

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Table 1. Observation points of chamber CH_4 flux. Concentration and isotopic compositions of dissolved CH_4 were also observed in the following wet areas.

Site	Landscape	Observation points and surface conditions	Dominant vegetation	Volumetric water content (%)[b]	Thaw depth (cm)[c]
V (Verkhny Khatistakha) 70° 15' N 147° 28' E	Larch forest and wetland	tree mound_V	Green moss, *Larix gmelinii*	17 ± 5 (*n* = 3)	23 ± 3 (*n* = 5)
		sedge_V (wet area)	*Carex sp.*	48 ± 4 (*n* = 3)	56 ± 3 (*n* = 4)
K (Kodac)[a] 70° 34' N 148° 16' E	Typical taiga-tundra boundary	tree mound_K	Green moss, *Larix gmelinii*	2.1 ± 0.6 (*n* = 4)	23 ± 4 (*n* = 9)
		sphagnum_K (wet area)	*Sphagnum spp.*	42 ± 5 (*n* = 6)	31 ± 8 (*n* = 15)
		sedge_K (wet area)	*Eriophorum angustifolium*	44 ± 4 (*n* = 6)	32 ± 13 (*n* = 28)
B (Boydom) 70° 38' N 148° 09' E	Low-centered polygon	tree mound_B	Green moss, *Larix gmelinii*	6 ± 2 (*n* = 5)	20 ± 4 (*n* = 8)
		sedge_B (wet area)	*Eriophorum angustifolium*	46 ± 2 (*n* = 5)	36 ± 9 (*n* = 8)

[a] Site K was previously named as Kryvaya (Iwahana et al., 2014) or Kodak (Liang et al., 2014).

[b] Observed for surface soil layer down to 20 cm in July 2011 (see Table S1 for detailed observation dates). Standard deviations are shown.

[c] Observed from early July to early August during 2010–2013 (see Table S1 for observation dates). Standard deviations are shown.

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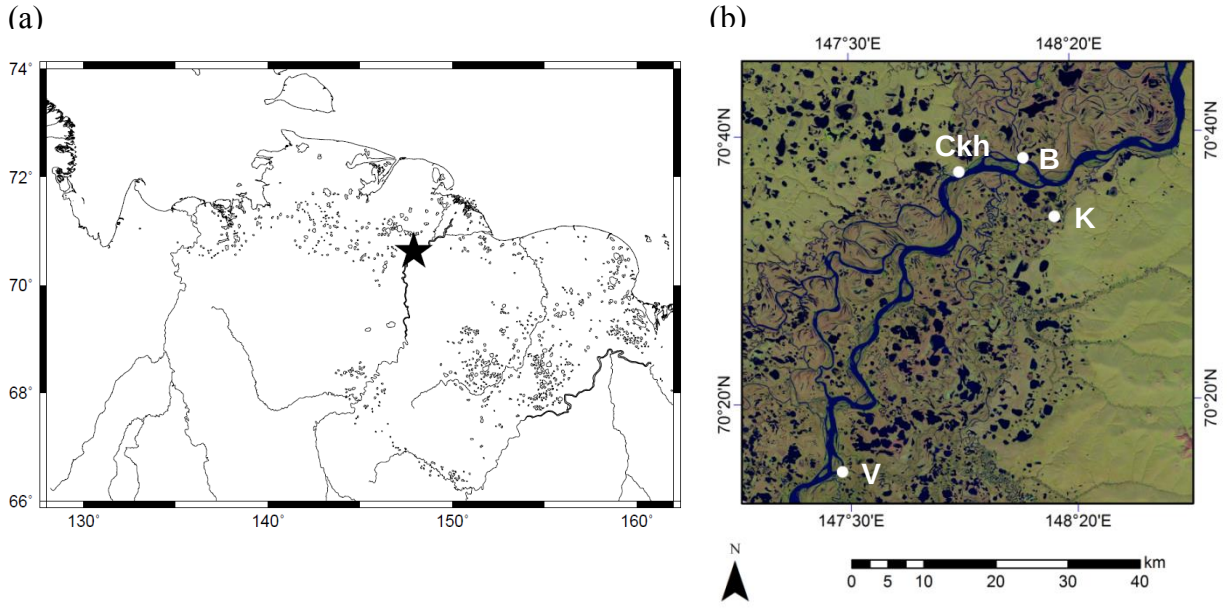

Figure 1: Locations of study sites. (a) Location of study region in Northeastern Siberia (the Generic Mapping Tools 5.0.0). (b) Satellite image of Indigirka River lowland around Chokurdakh village (Ckh: 70° 37' N, 147° 55' E) from Landsat 8. Observation sites (V, K, B) were selected in this region alongside the main stem and a tributary of Indigirka River.

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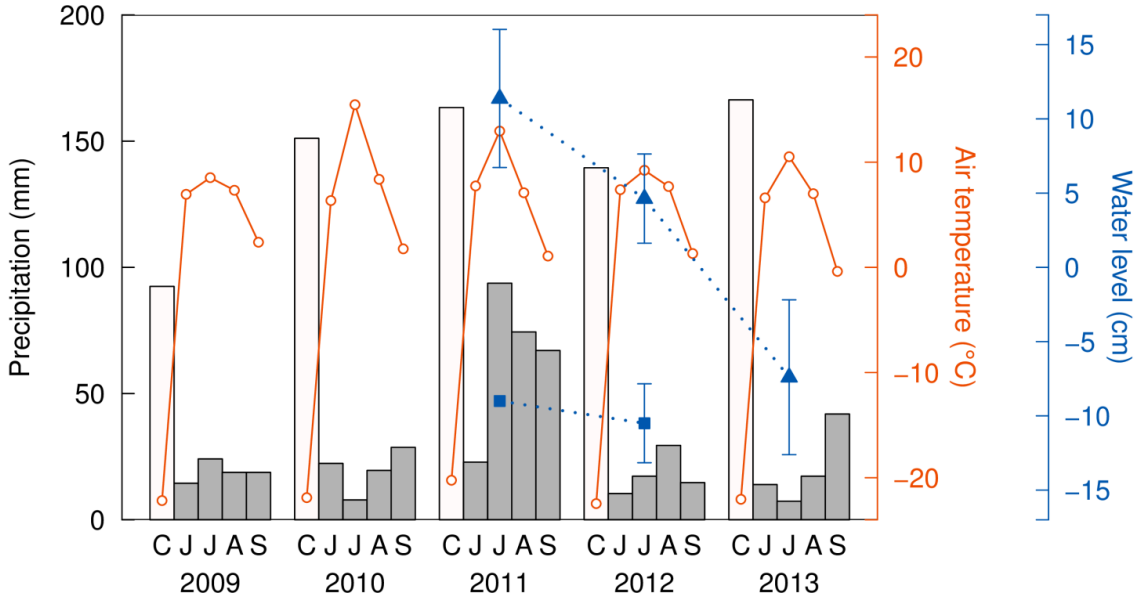

Figure 2: Interannual variations in precipitation (bars) and air temperature (solid lines) observed at a weather station at Chokurdakh for the cold season with snow cover (C: total from October in the previous year to May in the current year) and the warm season (JJAS), and those in water level (dotted lines) measured in wet areas of sedges (triangle) and sphagnum mosses (square). Methane flux was observed from early July to early August in these years. Error bars represent standard deviations.

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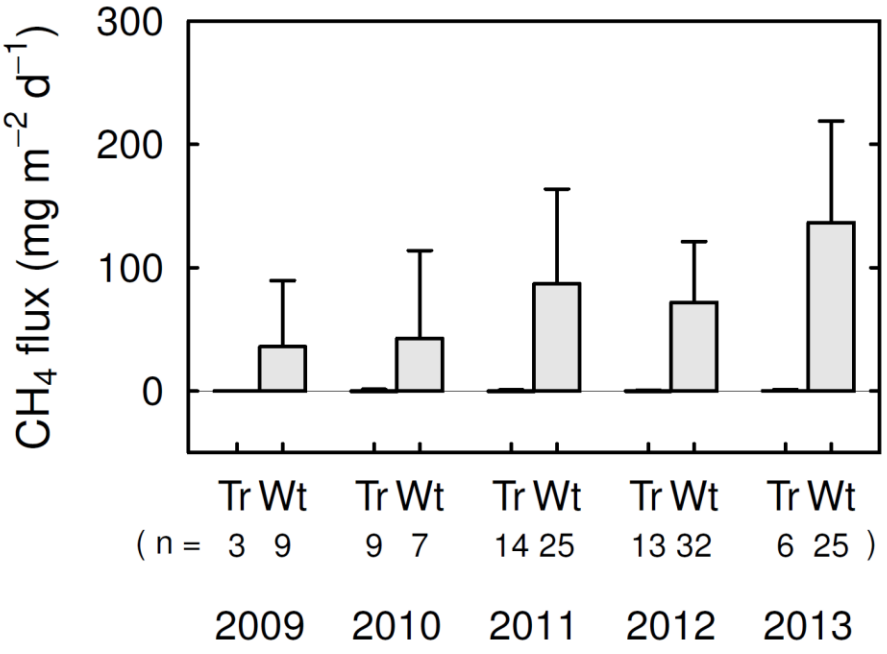

Figure 3: Interannual variations in averaged CH_4 flux in tree mounds (denoted as "Tr") and wet areas ("Wt") for main summer seasons from 2009 to 2013. Replication numbers ("*n*") are shown for every averaged flux values and standard deviations are represented by error bars. See Table S1 for flux values at respective observation points.

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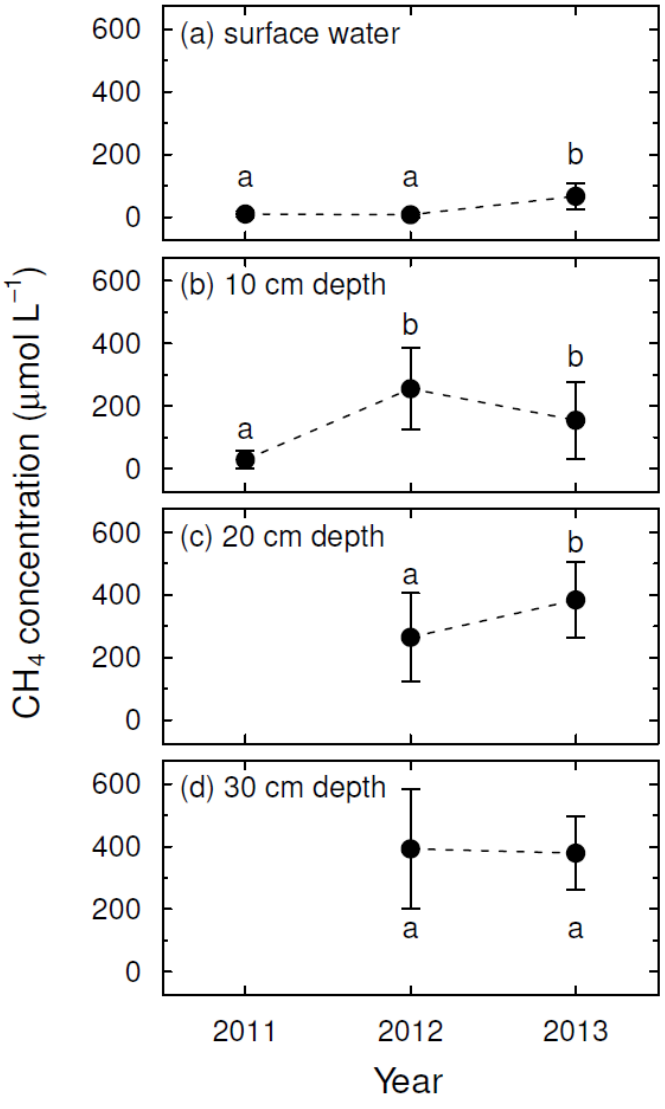

Figure 4: Interannual variation in averaged dissolved CH_4 concentration in (a) surface water and soil pore water at (b) 10 cm, (c) 20 cm, and (d) 30 cm depths in wet areas from the wet event in 2011 to 2013. Different letters in each panel denote statistical difference among years ($p < 0.05$). Error bars represent standard deviations. See Table S2 for concentration values at respective observation points.

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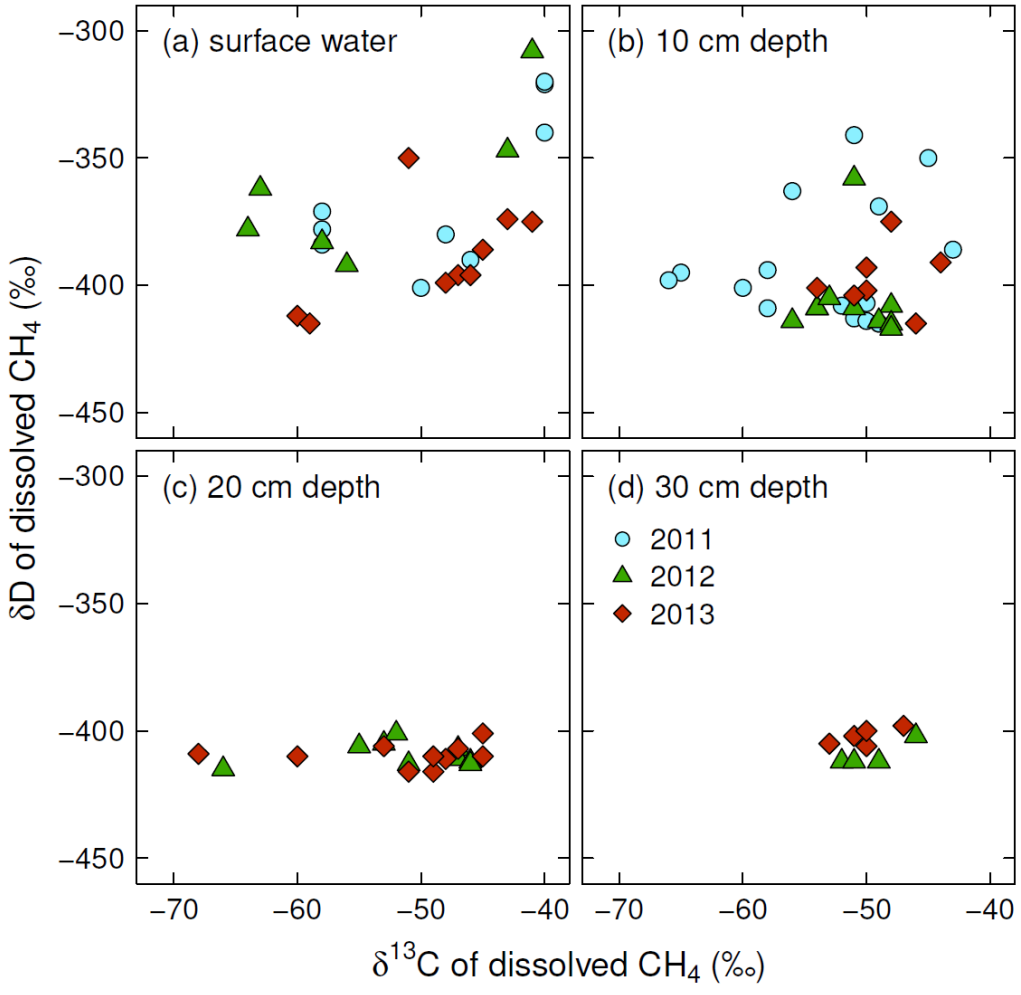

Figure 5: In-situ $\delta^{13}C$ versus δD of dissolved CH_4 in (a) surface water and soil pore water at (b) 10 cm depth, (c) 20 cm depth, and (d) 30 cm depth from the wet event in 2011 to 2013.

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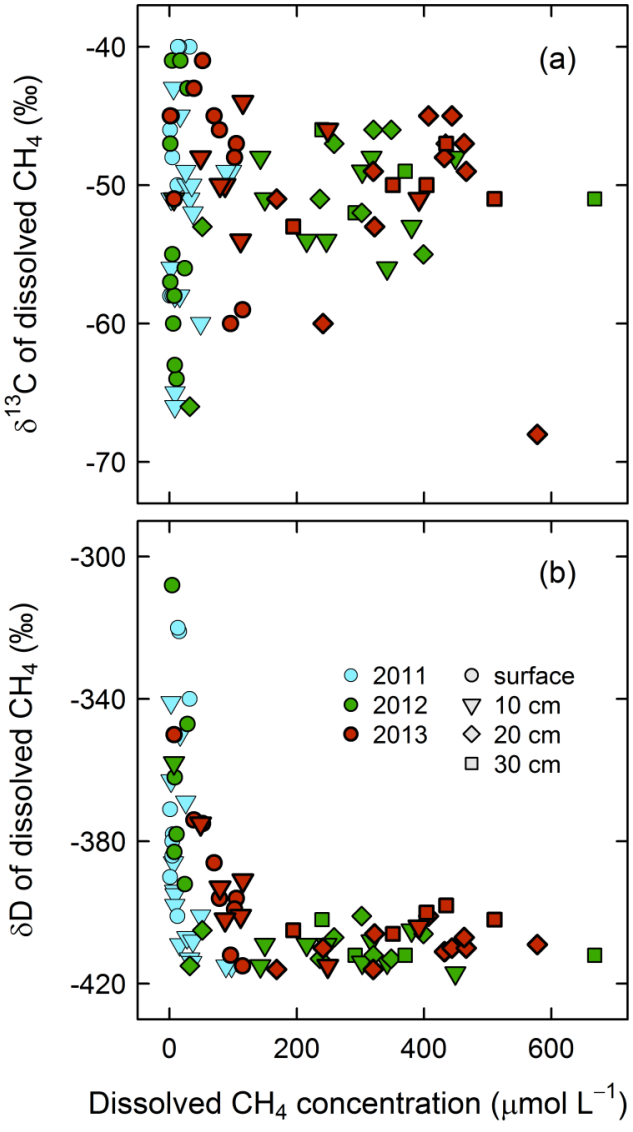

Figure 6: In-situ (a) $\delta^{13}C$ and (b) δD versus concentration for dissolved CH_4 at four depths (surface water, 10 cm, 20 cm, and 30 cm) in wet areas from 2011 to 2013.

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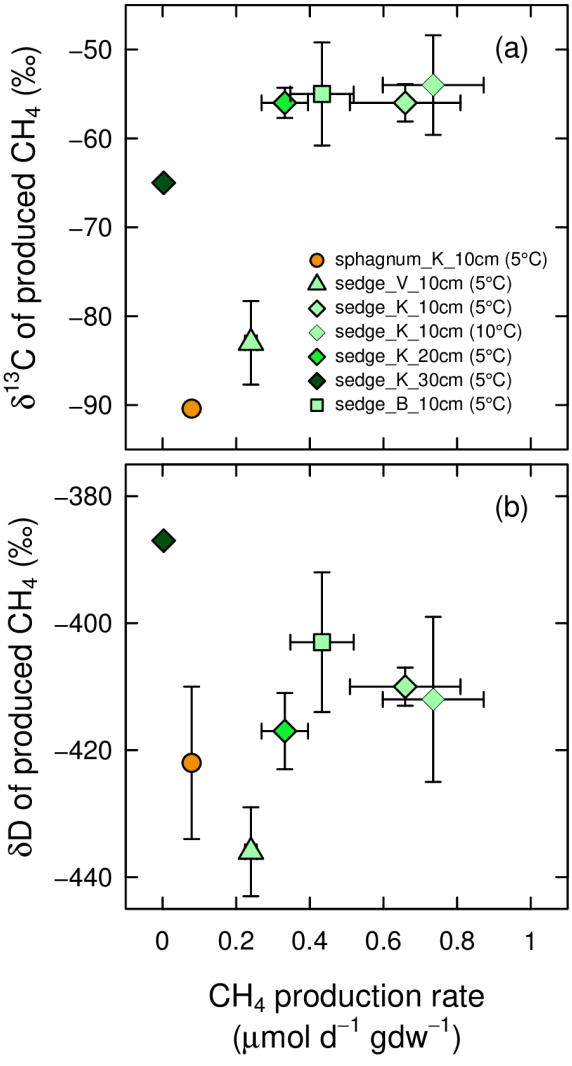

Figure 7: (a) $\delta^{13}C$ and (b) δD of produced CH_4 versus CH_4 production rate in the anaerobic soil incubation experiment. Production rates are shown in moles of produced CH_4 per day and per weight of dry soil in gram. Soil samples were collected at four observation points (sphagnum_K, sedge_V, sedge_K, and sedge_B) at three depths (10 cm, 20 cm, and 30 cm) and incubated at two temperatures (5 °C and 10 °C). These samples are organic layers except for that of 30 cm. Error bars represent standard deviations.

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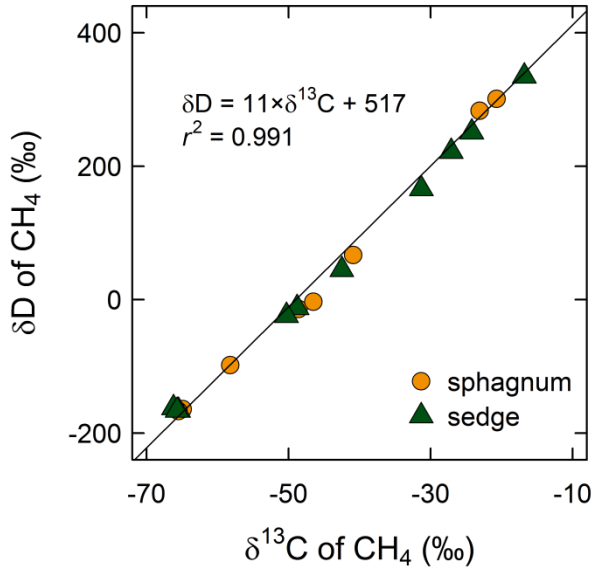

Figure 8: Enrichment of D/H (CH_4) and $^{13}C/^{12}C$ (CH_4) through CH_4 oxidation during the aerobic incubation experiment of surface organic layers in wet areas of sphagnum mosses and sedges in site K. Initial isotopic compositions of headspace CH_4 in incubated syringes are all situated in the bottom left corner ($\delta^{13}C = -66‰$, $\delta D = -165‰$).