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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

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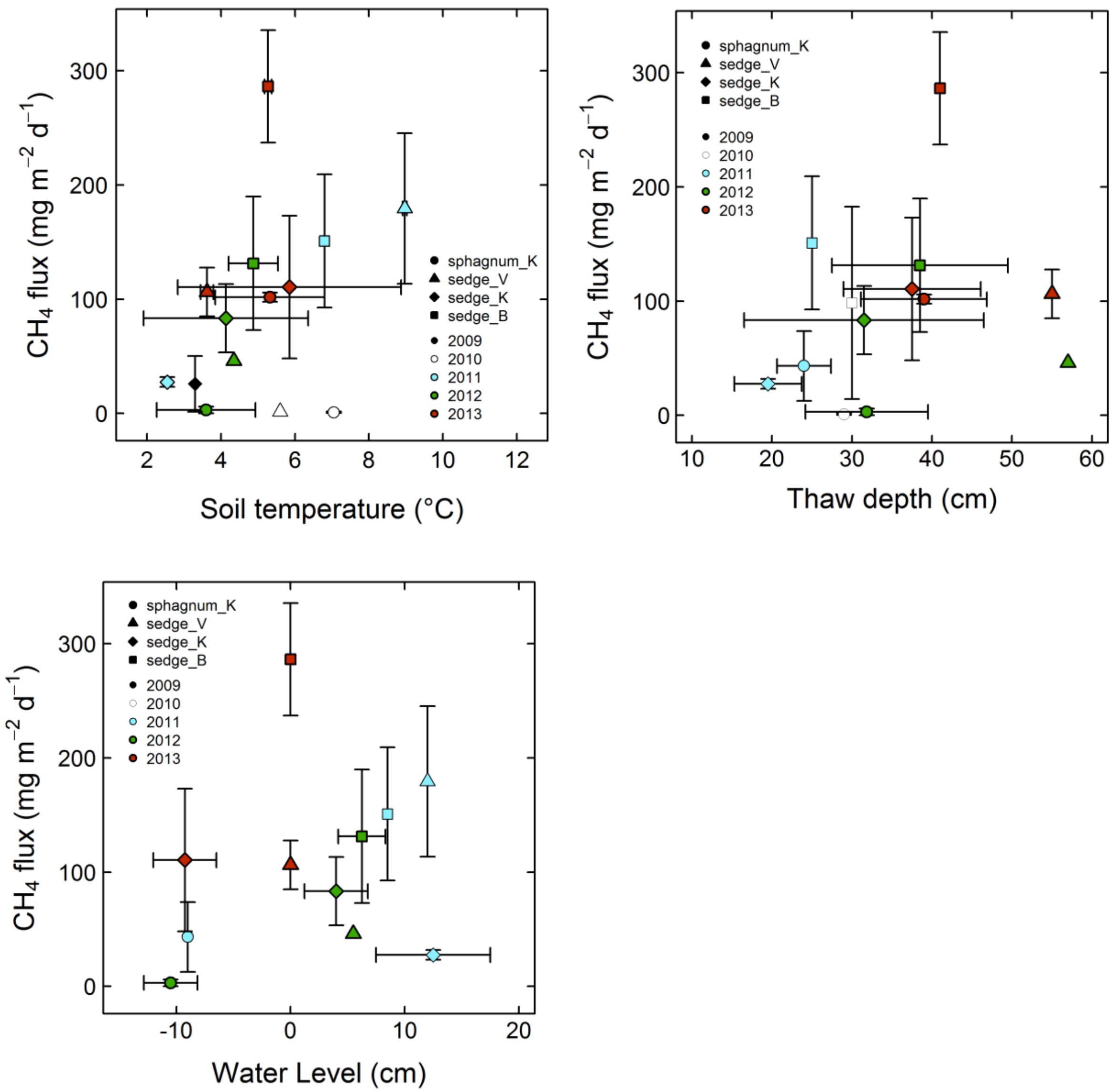

Figure S1. Averaged CH_4 flux over each wet area and each year plotted against (a) soil temperature (10 cm depth), (b) thaw depth, and (c) water level. Error bars represent standard deviations. Most observations for soil temperature and thaw depth were conducted at the same time of flux measurements, but not for all (Table S1).

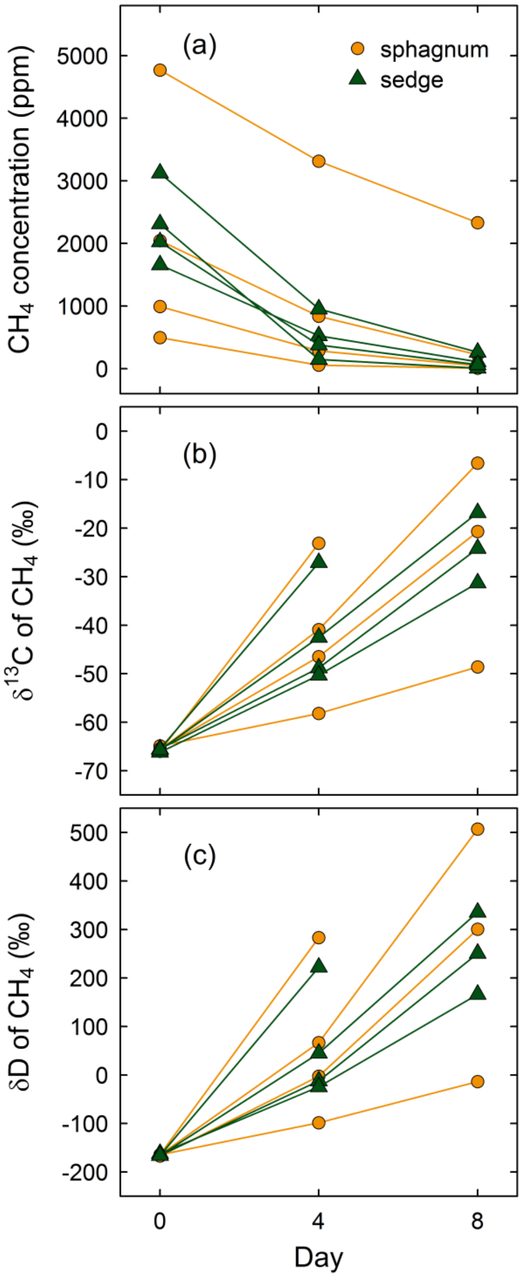

Figure S2. Temporal changes in (a) concentration, (b) $\delta^{13}C$, and (c) δD of the remaining headspace CH_4 in the soil incubation experiment for CH_4 production. Surface organic layers (0-13 cm) from wet areas (sphagnum_K and sedge_K) were incubated in quadruplicate at 8 °C.

Table S1. Thaw depths in tree mound and wet area observed along with CH_4 flux from 2010 to 2013. Averaged values and ranges are shown for each vegetation type and each year. Standard deviations are represented when $n \geq 3$. See Table S2 for each observation date.

Year	Thaw depth (cm)			
	Tree mound		Wet area	
	mean	range	mean	range
2010	23 (n = 2)	21–25 (Jul 20–21)	30 (n = 2)	29–30 (Jul 20–21)
2011	23 ± 6 (n = 6)	14–30 (Jul 9–30)	22 ± 4 (n = 9)	15–28 (Jul 9–21)
2012	21 ± 4 (n = 11)	16–27 (Jul 3 – Aug 9)	35 ± 14 (n = 24)[a]	9–57 (Jul 3 – Aug 9)[a]
2013	20 ± 3 (n = 3)	18–23 (Jul 15 – Aug 2)	40 ± 9 (n = 20)[b]	23–58 (Jul 11 – Aug 2)[b]

[a] During Jul 8–20, 2012 in wet area, the mean value was 25 ± 8 cm, and the range was 9–37 cm.

[b] During Jul 11–18, 2013 in wet area, the mean value was 35 ± 7 cm, and the range was 23–46 cm.

Table S2. Averaged CH_4 flux (in mg CH_4 m^{-2} day^{-1}) over each observation point and each year (2009–2013). Standard deviations are shown in case of $n \geq 3$. Dates of the flux observation are indicated in parenthesis. Superscripts represent observed environmental variables on each day: a) soil temperature (2009–2013), b) thaw depth (2010–2013), c) water level (2011–2013), and d) volumetric water content in surface soil (2011).

Observation points	Year				
	2009	2010	2011	2012	2013
tree mound_V	–	0 (Jul 16)	−1 ± 2 (Jul 23[b], 29[abd])	0 (Aug 7[ab])	0 (Aug 2[ab])
sedge_V (wet area)	3 ± 3 (Jul 23)	2 (Jul 16[a])	179 ± 66 (Jul 23[c], 29[acd])	46 (Aug 7[abc])	106 ± 21 (Aug 2[abc])
tree mound_K	0 (Jul 22)	−1 ± 3 (Jul 21[ab])	0 (Jul 15[bd], 18[bd])	0 (Jul 3[ab], 8[a], 12[ab], 24[ab]; Aug 2[ab], 6[ab])	1 (Jul 15[ab])
sphagnum_K (wet area)	–	1 ± 1 (Jul 21[ab])	43 ± 31 (Jul 11[bd], 17[b], 18[bcd], 21[bcd])	3 ± 3 (Jul 3[abc], 8[abc],12[abc], 24[abc]; Aug 2[abc], 6[abc])	102 ± 4 (Jul 11[ab], 18[ab], 25[ab], 31[ab])
sedge_K (wet area)	26 ± 24 (Jul 22[a])	–	28 ± 4 (Jul 11[bcd], 17[bc], 18[bcd], 21[abcd])	83 ± 30 (Jul 3[abc], 8[abc], 12[abc],20[abc], 21[a], 24[abc]; Aug 2[abc], 6[abc])	111 ± 63 (Jul 11[abc], 18[abc], 25[abc], 31[abc])
tree mound_B	–	0 (Jul 20[b])	0 (Jul 9[bd], 30[bd])	−1 ± 1 (Jul 13[ab], Aug 9[ab])	0 (Jul 16[ab])
sedge_B (wet area)	79 ± 80 (Jul 25)	98 ± 84 (Jul 20[b])	151 ± 58 (Jul 9[bcd], 30[acd])	131 ± 58 (Jul 13[ab], Aug 9[abc])	286 ± 49 (Jul 16[abc])

Table S3. Concentration, $\delta^{13}C$, and δD of dissolved CH_4 in surface water and soil pore water averaged over each wet area and each year. Standard deviations are shown in case of $n \geq 3$.

Wet area	Depth	Concentration (μmol CH_4 L^{-1})			$\delta^{13}C$ (‰)			δD (‰)		
		2011	2012	2013	2011	2012	2013	2011	2012	2013
sphagnum_K	surface water	—	—	—	—	—	—	—	—	—
	10 cm	6 ± 3 (*n* = 6)	318 (*n* = 1)	—	−56 ± 9 (*n* = 6)	−48 (*n* = 1)	—	−380 ± 23 (*n* = 6)	−408 (*n* = 1)	—
	20 cm	—	176 ± 125 (*n* = 3)	399 ± 68 (*n* = 3)	—	−55 ± 10 (*n* = 3)	−47 ± 2 (*n* = 3)	—	−412 ± 4 (*n* = 3)	−412 ± 3 (*n* = 3)
	30 cm	—	266 (*n* = 2)	351 (*n* = 1)	—	−49 (*n* = 2)	−50 (*n* = 1)	—	−407 (*n* = 2)	−406 (*n* = 1)
sedge_V	surface water	3 ± 2 (*n* = 3)	6 (*n* = 2)	105 (*n* = 2)	−58.1 ± 0.2 (*n* = 3)	−56 (*n* = 2)	−60 (*n* = 2)	−378 ± 7 (*n* = 3)	−383 (*n* = 1)	−414 (*n* = 2)
	10 cm	30 ± 17 (*n* = 3)	150 (*n* = 1)	249 (*n* = 1)	−59 (*n* = 2)	−51 (*n* = 1)	−46 (*n* = 1)	−405 (*n* = 2)	−409 (*n* = 1)	−415 (*n* = 1)
	20 cm	—	52 (*n* = 1)	168 (*n* = 1)	—	−53 (*n* = 1)	−51 (*n* = 1)	—	−405 (*n* = 1)	−416 (*n* = 1)
	30 cm	—	—	—	—	—	—	—	—	—
sedge_K	surface water	12 ± 14 (*n* = 4)	8 ± 8 (*n* = 11)	74 ± 27 (*n* = 6)	−46 ± 4 (*n* = 4)	−51 ± 9 (*n* = 10)	−45 ± 3 (*n* = 6)	−378 ± 27 (*n* = 4)	−349 ± 30 (*n* = 4)	−388 ± 11 (*n* = 6)
	10 cm	53 ± 31 (*n* = 6)	244 ± 143 (*n* = 7)	88 ± 27 (*n* = 5)	−50 ± 1 (*n* = 6)	−52 ± 3 (*n* = 7)	−49 ± 4 (*n* = 5)	−412 ± 4 (*n* = 6)	−405 ± 21 (*n* = 7)	−392 ± 11 (*n* = 5)
	20 cm	—	368 ± 59 (*n* = 3)	431 ± 123 (*n* = 5)	—	−46 ± 1 (*n* = 3)	−54 ± 10 (*n* = 5)	—	−412 ± 1 (*n* = 3)	−407 ± 4 (*n* = 5)
	30 cm	—	519 (*n* = 2)	450 ± 55 (*n* = 3)	—	−50 (*n* = 2)	−49 ± 2 (*n* = 3)	—	−412 (*n* = 2)	−400 ± 2 (*n* = 3)
sedge_B	surface water	14 (*n* =2)	8 ± 14 (*n* = 3)	4 (*n* = 2)	−40 (*n* = 2)	−56 (*n* = 1)	−48 (*n* = 2)	−320 (*n* = 2)	−392 (*n* = 1)	−350 (*n* = 1)
	10 cm	21 (*n* = 2)	380 (*n* = 1)	392 (*n* = 1)	−47 (*n* = 2)	−53 (*n* = 1)	−51 (*n* = 1)	−360 (*n* = 2)	−405 (*n* = 1)	−404 (*n* = 1)
	20 cm	—	351 (*n* = 2)	322 (*n* = 1)	—	−54 (*n* = 2)	−53 (*n* = 1)	—	−404 (*n* = 2)	−406 (*n* = 1)
	30 cm	—	—	194 (*n* = 1)	—	—	−53 (*n* = 1)	—	—	−405 (*n* = 1)

Table S4. Individual values of water level, dissolved CH_4 concentration (10 cm depth), and $\delta^{13}C$ and δD of dissolved CH_4 (10 cm depth) observed in each wet area on each date in 2011. Duplicated data are shown in some cases. Although water level increased during July 2011, clear temporal change was not found in the delta values.

Observation point	Date in 2011	Water level (cm)	Dissolved CH_4 concentration (μmol L^{-1})	$\delta^{13}C$ of dissolved CH_4 (‰)	δD of dissolved CH_4 (‰)
sphagnum_K	Jul 17	— (< 0)	6, 8	−43, −58	−386, −394
	Jul 18	−10	2, 2	−56, −51	−363, −341
	Jul 21	−8	8, 9	−66, −65	−398, −395
sedge_V	Jul 23	10	16	−58	−409
	Jul 29	14	49, 26	−60	−401
sedge_K	Jul 11	5	−	−	−
	Jul 17	15	27, 37	−50, −52	−407, −408
	Jul 18	15	32, 35	−51, −50	−413, −414
	Jul 21	15	88, 97	−49, −49	−415, −415
sedge_B	Jul 9	6	−	−	−
	Jul 30	11	17, 26	−45, −49	−350, −369

Table S5. Phylogenic composition of methanogenic archaea in wet areas. Soils (organic layers) were sampled in July 2016 from 10 cm depth in the same wet areas as the CH_4 production incubation experiment in triplicate. Then, microbial communities in the samples were analyzed by amplicon sequencing of 16S rRNA gene. See Fig. 8 for a plot by the level of orders.

Order	Family	Genus	Relative abundance in the total sequences (%)											
			sphagnum_K (S01)	sphagnum_K (S02)	sphagnum_K (S03)	sedge_V (S07)	sedge_V (S08)	sedge_V (S09)	sedge_K (S04)	sedge_K (S05)	sedge_K (S06)	sedge_B (S10)	sedge_B (S11)	sedge_B (S12)
Methanosarcinales	Methanosarcinaceae	*Methanosarcina*	0.01	0.00	0.00	0.21	0.09	0.47	0.06	0.08	0.08	0.25	0.20	0.16
	Methanosaetaceae	*Methanosaeta*	0.00	0.00	0.00	0.00	0.13	0.06	0.74	0.80	0.93	0.82	0.91	0.78
Methanomicrobiales	Methanoregulaceae	*Candidatus* Methanoregula	0.00	0.00	0.00	0.09	0.07	0.10	0.10	0.10	0.11	0.23	0.32	0.28
	Methanospirillaceae	*Methanospirillum*	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.02	0.00	0.01
	Methanoregulaceae	Other Methanoregulaceae	0.00	0.00	0.00	0.00	0.01	0.01	0.00	0.02	0.01	0.08	0.08	0.05
Methanocellales		Methanocellales	0.00	0.00	0.00	0.00	0.00	0.00	0.07	0.07	0.09	0.00	0.00	0.00
Methanobacteriales	Methanobacteriaceae	*Methanobacterium*	0.07	0.00	0.12	0.16	0.15	0.40	1.11	1.11	1.49	0.93	0.47	0.66
E2	[Methanomassiliicoccaceae]	[Methanomassiliicoccaceae]	0.01	0.00	0.00	0.04	0.02	0.03	0.07	0.07	0.08	0.08	0.06	0.05

Table S6. Redox potential observed in summers 2012 and 2013 by ORP meter (RM-20P or RM-30P, DKK-TOA Corporation, Japan) connected with an ORP electrode (PST-2739C). Measurement accuracy of the ORP meter is ±10 mV. Redox potential value was accepted when the potential stabilized after installing the ORP electrode into the soil.

Observation points	Depth (cm)	Redox potential (mV versus the normal hydrogen electrode)	
		2012	2013
tree mound_K	10	608 to 643	478 to 482
	20	627 to 631	—
sphagnum_K (wet area)	10	−183 to 814	547 to 617
	20	−129	—
sedge_K (wet area)	10	−177 to −121	−114 to −69
	20	−250 to −78	−223 to −194
	30	−152 to −118	—
sedge_B (wet area)	10	—	−113 to −102