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# Supplement of

# Hysteretic temperature sensitivity of wetland CH<sub>4</sub> fluxes explained by substrate availability and microbial activity

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## **Model description**

The *ecosys* model has been rigorously examined in many high-latitude ecosystems, e.g.: modeled energy and carbon fluxes against data from eddy covariance flux towers at the Barrow Experimental Observatory (BEO) (Grant *et al* 2017b, 2017a) and a topographic gradient at Daring Lake, NWT (Grant 2015a, Grant *et al* 2015), and against long-term measurements from automated chambers in the Stordalen Mire(Chang *et al* 2019, n.d.). The model performance has also been tested and documented in sites from many biomes encompassing a range of latitudes in a long list of *ecosys* publications (5-92). The following qualitative model description is adapted from our earlier publication.

Ecosys represents multiple canopy and soil layers and fully coupled carbon, energy, water, and nutrient cycles solved at an hourly time step. Surface energy and water exchanges drive soil heat and water transfers to determine soil temperatures and water contents. These transfers drive soil freezing and thawing and, hence, active layer depth, through the general heat flux equation. Carbon uptake is controlled by plant water status calculated from convergence solutions that equilibrate total root water uptake with transpiration. Atmospheric warming increases surface heat advection, soil heat transfers, and hence active layer depth. Canopy temperatures affect CO<sub>2</sub> fixation rates from their effects on carboxylation and oxygenation modeled with Arrhenius functions for light and dark reactions. Soil temperatures affect heterotrophic respiration through the same Arrhenius function as for dark reactions.

Carbon uptake is also affected by plant nitrogen uptake. The model represents fully coupled transformations of soil carbon, nitrogen, and phosphorus through microbially driven processes. Soil warming enhances carbon uptake by hastening microbial mineralization and root nitrogen uptake. Carbon uptake is affected by phenology with leafout and leafoff (deciduous plants) or dehardening and hardening (evergreen plants) being determined by accumulated exposure to temperatures above set values while day length is increasing or below set values while day length is decreasing. Senescence is driven by excess maintenance respiration and by phenology in deciduous plant functional types.

Ecosystem-Atmosphere energy exchange:

Canopy energy and water exchanges in *ecosys* are calculated through a multi-layered soil-root-canopy system. The clumping effect for each leaf and stem surface is represented by a species-specific interception fraction to simulate non-uniformity in the horizontal distribution of leaves within each canopy layer. Coupled first-order closure schemes are solved between the atmosphere and each of leaf and stem surfaces in the multi-layered canopy to achieve energy balance at each model time step. Once the system converges to the required canopy temperature, latent and sensible heat fluxes of each canopy layer are calculated based on the simulated vapor pressure deficit, canopy-atmosphere temperature gradient, aerodynamic conductance, and stomatal conductance. Canopy heat storage is calculated from changes in canopy temperature and heat capacities of leaves, twigs, and stems.

# Canopy water relations:

A convergence solution is sought for the canopy water potential of each plant population at which the difference between its transpiration and total root water uptake equals the difference between its water contents at the previous and current water potentials. Canopy water potential controls transpiration and soil-root water uptake, which affects stomatal conductance and thereby all the processes (e.g., canopy temperature and vapor pressure) described in "Ecosystem-Atmosphere energy exchange". The water table depth in *ecosys* is calculated at the end of each time step as the depth to the top of the saturated zone below which air-filled porosity is zero. Changes in the simulated water table depth were driven by dynamical interactions among precipitation, evapotranspiration, vertical water transport, and lateral water transport.

# Canopy carbon and nutrient cycling:

Leaf carboxylation rates are adjusted from those calculated under non-limiting water potential to those under current water potential. The gross canopy CO<sub>2</sub> fixation is the sum of the leaf carboxylation rate of each leaf surface present on each branch of each plant species, which is then transported to a mobile pool of carbon storage. Storage carbon oxidized in excess of maintenance respiration requirements is used as growth respiration to drive the formation of new biomass. Net CO<sub>2</sub> fixation is calculated as the difference between gross fixation and the sum of maintenance, growth, and senescence respiration in the simulated canopy.

Nutrient (nitrogen and phosphorous) uptake is calculated for each plant species by solving for aqueous concentrations at root and mycorrhizal surfaces in each soil layer at which radial transport by mass flow and diffusion from the soil solution to the surfaces equals active uptake by the surfaces. This solution dynamically links rates of soil nutrient transformations with those of root and mycorrhizal nutrient uptake. The products of nitrogen and phosphorous uptake are transported to mobile pools of nitrogen and phosphorous stored in each root and mycorrhizal layer, which regulate vegetation growth.

### Plant functional type dynamics:

The model represents prognostic vegetation dynamics with internal resource allocation and remobilization. Shifts in plant functional types are modeled through processes of plant functional type competition for light, water, and nutrients within each canopy and rooted soil layer depending on leaf area and root length. Each plant functional type competes for nutrient and water uptake from common nutrient and water stocks held across multi-layer soil profiles, calculated from algorithms for transformations and transfers of soil carbon, nitrogen, and phosphorus, and for transfers of soil water. Modeled differences in plant functional type functional traits determine the strategy of resource acquisition and allocation that drive growth, resource remobilization, and litterfall, and therefore each plant functional type dynamic competitive capacity under different environmental conditions.

#### Soil microbial activity:

The modeling of microbial activity is based on six organic states: solid, soluble, sorbed, acetate, microbial biomass, and microbial residues. Carbon, nitrogen and phosphorous may move among these states within each of four organic matter-microbe complexes: plant litterfall, animal manure, particulate organic matter, and humus. Microbial biomass in *ecosys* is an active agent of organic matter transformation. The rate at which each component is hydrolyzed is a function of substrate concentration that approaches a first-order function at low concentrations, and a zero-order function at high concentrations. These rates are regulated by soil temperature through an Arrhenius function and by soil water content through its effect on substrate concentration. Similar to the growth and decline of vegetation biomass described above, the net change in microbial biomass is determined by the difference between heterotrophic respiration and maintenance

respiration. When heterotrophic respiration is greater than maintenance respiration, the excessive amount of respiration is used as growth respiration that drives microbial growth according to the energy requirements of biosynthesis.

*Methane production and transport:* 

The rate at which soil organic matter in *ecosys* is hydrolyzed during decomposition is a first-order function of the decomposer biomass of all heterotrophic microbial populations (functional types) generated from energy yields of the oxidation-reduction reactions conducted by each population. Hydrolysis rates are regulated by soil temperature through an Arrhenius function and by soil water content through its effect on aqueous microbial concentrations. Hydrolysis products are transferred to dissolved organic carbon (DOC) that is the substrate for respiration and growth according to energy yields from DOC oxidation-reduction by all heterotrophic microbial functional types (MFT) calculated from MFT biomass, a Michaelis-Menten function of DOC concentration, and from soil temperature and oxygen availability. DOC uptake by anaerobic MFTs (fermenters) drives fermentation products of which are partitioned among acetate (CH<sub>2</sub>COOH), CO<sub>2</sub>, and hydrogen (H<sub>2</sub>) according to Brock & Madigan (1991). These products are substrates for acetoclastic (AM) and hydrogenotrophic methanogenesis (HM), rates of which are driven by biomasses of AM and HM functional types, growths of which are generated from AM and HM energy yields. For all MFTs, respiration products beyond those used for microbial maintenance respiration drives microbial growth. Specifically, AM (HM) microbial growth is calculated by dividing the free energy change of AM (HM) by the energy required to construct new AM (HM) microbial carbon, which drives changes in AM (HM) microbial biomass after subtracting biomass loss from decomposition. CH<sub>4</sub> production rates are functions of microbial biomass. temperature, substrate concentrations, and moisture for AM and HM MFTs in each model soil layer during each hourly model time step. CH<sub>4</sub> produced by AM and HM can then be transported to the atmosphere through diffusion, plant aerenchyma transport, and ebullition, or oxidized by methanotrophs. Detailed equation sets for methanogenesis and methanotrophy used in *ecosys* were derived in Grant. (1998: 1999) and remain unchanged in other studies of methanogenesis (e.g. Grant et al., 2017, 2019) since then.

Supplementary Table 1. Evaluation of modeled apparent CH<sub>4</sub> emission temperature dependence calculated by fitting Boltzmann-Arrhenius functions with soil and air temperatures during the earlier, later, and full-season (entire thawed season) periods in the bog and fen at the Stordalen Mire.

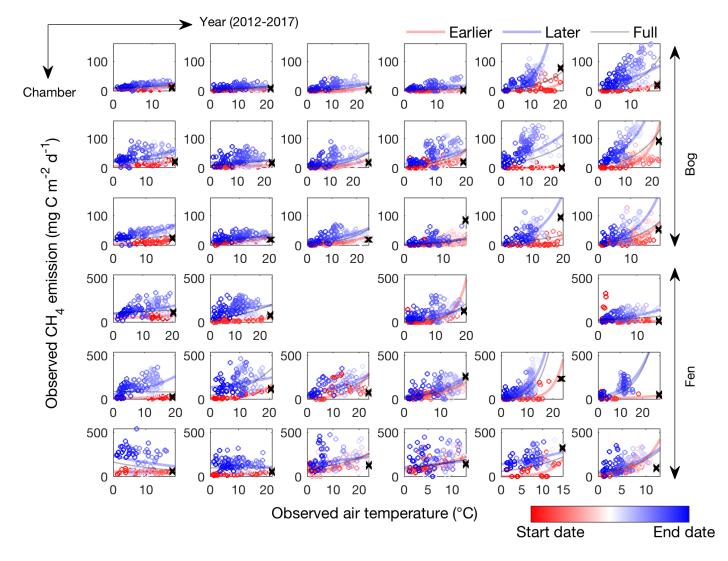
_	Soil temperature				Air temperature				
2003-2007									
	Earlier		Later		Earli	Earlier		Later	
	hysteretic	full- season	hysteretic	full- season	hysteretic	full- season	hysteretic	full- season	
· · · · · · · · · · · · · · · · · · ·		R	2			R <sup>2</sup>			
Bog	0.68	0.62	0.49	0.31	0.40	0.32	0.36	0.05	
Fen	0.84	-0.95	0.71	-0.93	0.11	-0.18	0.14	-0.24	
_	RMSE					RMSE			
Bog	10.75	11.68	14.93	17.34	13.39	14.22	17.07	20.92	
Fen	10.43	36.06	23.49	60.86	44.65	51.37	53.69	64.51	
	Percent error				Percent error				
Bog	-3.95	10.29	-3.02	-21.01	-14.20	20.48	-12.81	-51.17	
Fen	-2.45	81.22	-1.37	-40.35	-15.07	36.97	-8.02	-57.64	
_	E <sub>a</sub>				E <sub>a</sub>				
Bog	1.88	1.82	1.49	1.78	1.47	1.25	1.17	1.23	
Fen	1.34	0.98	0.42	0.97	1.57	1.25	1.17	1.27	
2011-2013									
-	R <sup>2</sup>				R <sup>2</sup>				
Bog	0.73	0.59	0.61	0.17	-1.34	-2.69	0.30	-0.17	
Fen	0.86	0.01	0.76	-1.12	-0.12	-1.19	0.20	-0.55	
-		RIV	ISE .		RMSE				
Bog	8.72	10.81	10.60	15.57	17.88	22.47	17.42	22.63	
Fen	29.21	76.88	39.50	119.42	90.78	126.80	112.14	156.44	
-	Percent error			Percent error					
Bog	-2.88	16.39	-1.92	-21.48	-0.93	56.28	-10.08	-38.17	
Fen	-2.61	71.76	-0.53	-34.25	-22.90	61.11	-11.11	-52.16	
<u>-</u>	E <sub>a</sub>				E <sub>a</sub>				
Bog	1.85	1.58	1.16	1.61	2.18	1.66	1.07	1.65	
Fen	1.21	1.04	0.45	1.05	1.91	1.53	1.10	1.61	

 $R^2$ , RMSE, and  $E_a$  are the correlation, root mean squared error (in mg C m<sup>-2</sup> d<sup>-1</sup>), and activation energy (in eV), respectively, from fitting Eq. 1 to the *ecosys*-modeled fluxes. Negative  $R^2$  implies the regression line fitted by Boltzmann-Arrhenius functions is worse than using the mean value.

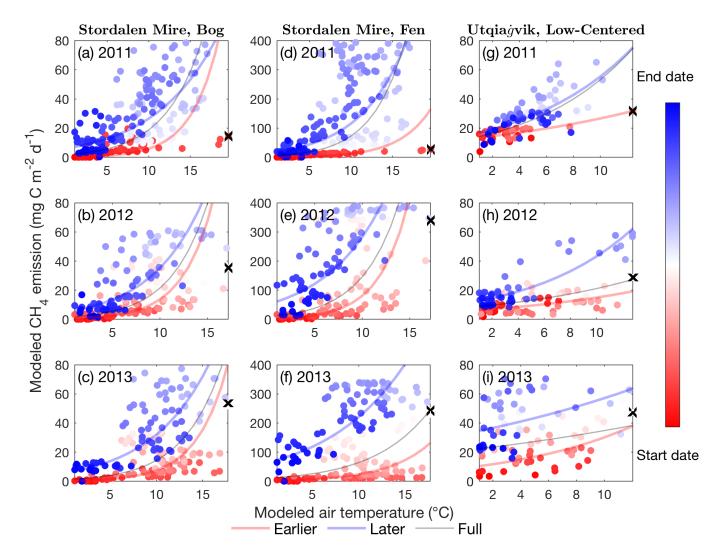
Supplementary Table 2. Evaluation of modeled apparent CH<sub>4</sub> emission temperature dependence calculated by fitting Boltzmann-Arrhenius functions with soil and air temperatures during the earlier, later, and full-season (entire thawed season) periods in the trough, rim, center, and polygon mean in a low-centered polygon at Utqiagvik (formerly Barrow) in 2013.

	Soil temperature				Air temperature				
	Earlier		Later		Earli	Earlier		Later	
	hysteretic	full- season	hysteretic	full- season	hysteretic	full- season	hysteretic	full- season	
		R	2			R <sup>2</sup>			
Trough	0.44	0.07	0.25	-0.67	0.18	0.15	-0.04	-1.03	
Rim	-0.11	-0.29	-0.69	-0.33	-0.20	-0.34	-0.47	-0.42	
Center	0.58	0.04	0.52	-1.41	0.25	-0.08	0.11	-1.75	
Polygon mean	0.60	0.06	0.42	-0.95	0.28	-0.13	0.08	-1.14	
	RMSE				RMSE				
Trough	8.14	9.40	20.79	27.91	10.13	10.28	21.74	30.36	
Rim	3.90	4.20	10.04	8.88	4.33	4.57	9.55	9.40	
Center	13.24	19.98	13.58	30.52	17.68	21.19	18.84	33.12	
Polygon mean	7.24	11.11	12.14	22.30	9.63	12.04	15.40	23.54	
	Percent error			Percent error					
Trough	-22.59	50.02	-5.98	-64.31	-31.66	32.49	-16.14	-61.37	
Rim	-27.87	-22.98	-43.55	-61.62	-30.95	-32.81	-57.49	-61.29	
Center	-5.46	40.77	-1.81	-38.38	-12.40	29.73	-3.43	-37.15	
Polygon mean	-5.77	45.40	-3.40	-44.45	-11.92	34.22	-6.15	-40.76	
	E <sub>a</sub>			E <sub>a</sub>					
Trough	2.55	1.56	1.91	1.56	1.18	0.62	0.86	0.62	
Rim	0.26	1.56	2.92	1.56	0.00	0.76	1.41	0.76	
Center	1.75	0.71	0.67	0.71	0.78	0.28	0.25	0.28	
Polygon mean	1.69	0.86	0.88	0.86	0.79	0.33	0.37	0.33	

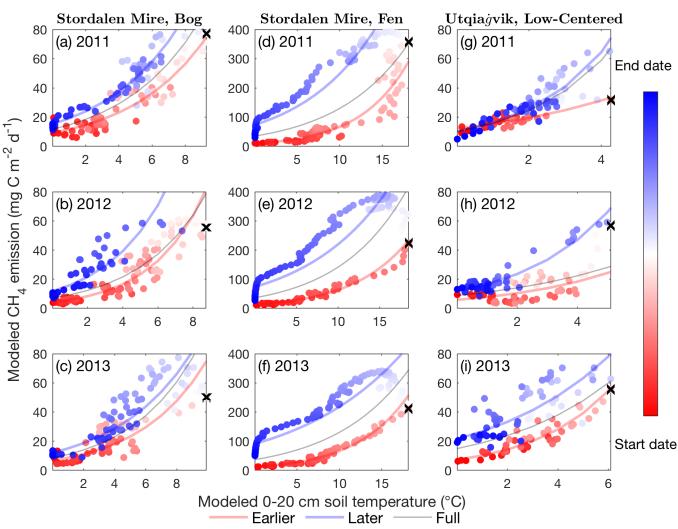
 $R^2$ , RMSE, and  $E_a$  are the correlation, root mean squared error (in mg C m<sup>-2</sup> d<sup>-1</sup>), and activation energy (in eV), respectively, from fitting Eq. 1 to the *ecosys*-modeled fluxes. Negative  $R^2$  implies the regression line fitted by Boltzmann-Arrhenius functions is worse than using the mean value.



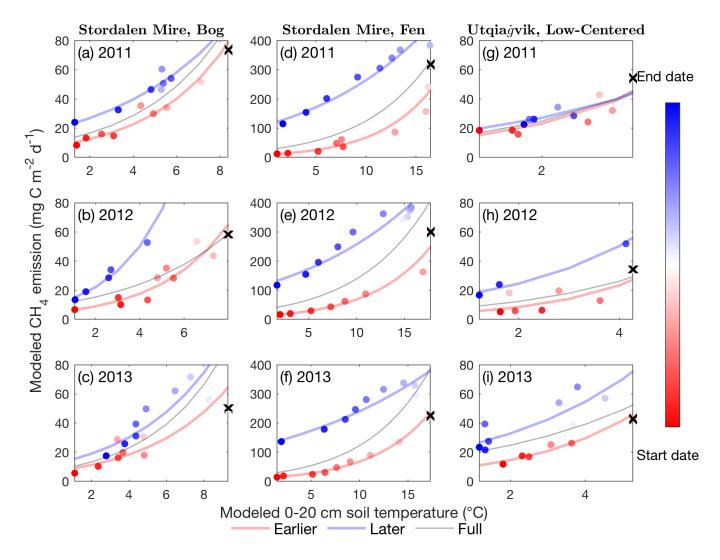
Supplementary Figure 1. CH<sub>4</sub> emissions are hysteretic to air temperature measured in individual automated chambers in the Stordalen Mire bog (top three panels) and fen (bottom three panels) sites from 2012 to 2017 thawed seasons (left to right). Open circles and lines represent the daily data points and the fitted apparent CH<sub>4</sub> emission temperature dependence, respectively. The earlier, later, and full-season periods are colored in red, blue, and black, respectively. Earlier and later periods are defined as the time before and after the seasonal maximum air temperature denoted by black cross signs. Start date and end dates represent the beginning and ending of a thawed season defined as the period when measured daily air temperature is above 1 °C, respectively.



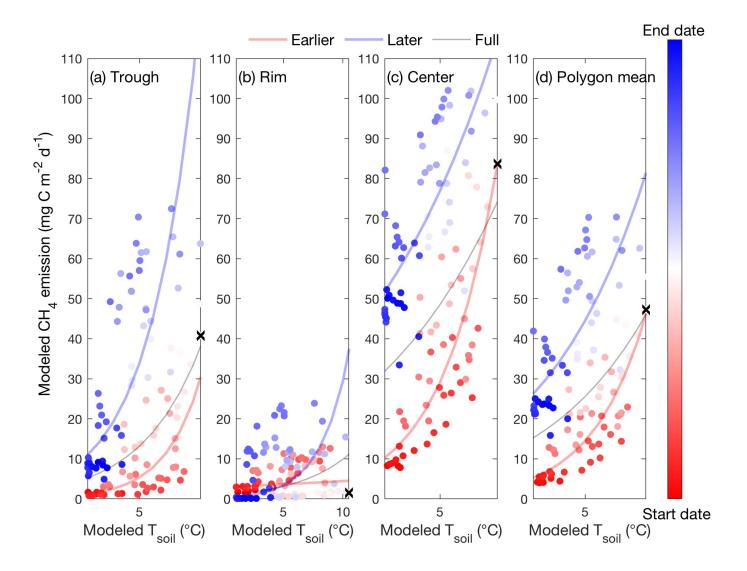
Supplementary Figure 2. CH<sub>4</sub> emissions are hysteretic to air temperature modeled at the Stordalen Mire bog (a to c) and fen (d to f) and the Utqiagvik low-centered polygon (g to i) from 2011 to 2013 thawed seasons. Dots and lines represent the daily data points and the fitted apparent temperature dependence, respectively. Earlier, later, and full-season periods are colored in red, blue, and black, respectively. Earlier and later periods are defined as the time before and after the seasonal maximum air temperature denoted by black cross signs. Start date and end dates represent the beginning and ending of a thawed season defined as the period when modeled daily air temperature is above 1 °C, respectively.



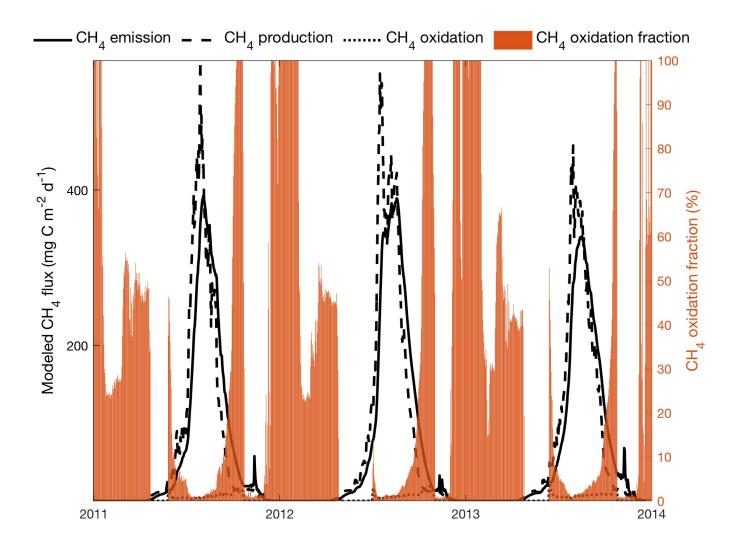
Supplementary Figure 3. CH<sub>4</sub> emissions are hysteretic to soil temperature modeled in the Stordalen Mire bog (a to c) and fen (d to f) and the Utqiagvik low-centered polygon (g to i) from 2011 to 2013 thawed seasons. Dots and lines represent the daily data points and the fitted apparent temperature dependence, respectively. Earlier, later, and full-season periods are colored in red, blue, and black, respectively. Earlier and later periods are defined as the time before and after the seasonal maximum 0-20 cm soil temperature denoted by black cross signs. Start date and end dates represent the beginning and ending of a thawed season defined as the period when modeled daily 0-20 cm soil temperature is above 0 °C, respectively.



Supplementary Figure 4. Weekly CH<sub>4</sub> emissions are hysteretic to weekly soil temperature modeled in the Stordalen Mire bog (a to c) and fen (d to f) and the Utqiagvik low-centered polygon (g to i) from 2011 to 2013 thawed seasons. Dots and lines represent the daily data points and the fitted apparent temperature dependence, respectively. Earlier, later, and full-season periods are colored in red, blue, and black, respectively. Earlier and later periods are defined as the time before and after the seasonal maximum 0-20 cm soil temperature denoted by black cross signs. Start date and end dates represent the beginning and ending of a thawed season defined as the period when modeled weekly 0-20 cm soil temperature is above 1 °C, respectively.



Supplementary Figure 5. Apparent soil temperature dependence of daily CH<sub>4</sub> emissions modeled in the trough (a), rim (b), center (c), and polygon mean (i.e., areal means of trough, rim, and center) (d) in a low-centered polygon at Utqiagʻvik, Alaska during the 2013 thawed season. Dots and lines represent the daily data points and the fitted apparent temperature dependence, respectively. The earlier, later, and full-season periods are colored in red, blue, and black, respectively. Earlier and later periods are defined as the time before and after the modeled maximum 0-20 cm soil temperature denoted by the black cross signs. Start date and end dates represent the beginning and ending of a thawed season defined as the period when modeled daily 0-20 cm soil temperature is above 1 °C, respectively. The hysteresis is less clear in the rim due to lower CH<sub>4</sub> emissions because of its drier conditions.



Supplementary Figure 6. Daily CH<sub>4</sub> emissions, CH<sub>4</sub> production, CH<sub>4</sub> oxidation, and CH<sub>4</sub> oxidation fraction modeled in the Stordalen Mire fen from 2011 to 2013. CH<sub>4</sub> oxidation fraction is defined as the ratio of daily CH<sub>4</sub> oxidation to daily CH<sub>4</sub> production.

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