Assessing methane emissions for northern peatlands in ORCHIDEE-PEAT revision 7020.

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Abstract. In the global methane budget, the largest natural source is attributed to wetlands that encompass all ecosystems composed of waterlogged or inundated ground, capable of methane production. Among them, northern peatlands that store large amounts of soil organic carbon have been functioning, since the end of the last glaciation period, as long-term sources of methane (CH₄) and are one of the most significant methane sources among wetlands. To reduce uncertainty of quantifying methane flux in global methane budget, it is of significance to understand the underlying processes for methane production and fluxes in northern peatlands. A methane model that features methane production and transport by plants, ebullition process and diffusion in soil, oxidation to CO₂ and CH₄ fluxes to the atmosphere has been embedded in the ORCHIDEE-PEAT land surface model that includes an explicit representation of northern peatlands. The ORCHIDEE-PCH4 was calibrated and evaluated on 14 peatland sites distributed on both Eurasian and American continents in the northern boreal and temperate regions. Data assimilation approaches were employed to optimized parameters at each site and at all sites simultaneously. Results show that methanogenesis is sensitive to temperature and substrate availability over the top 75 cm of soil depth. Methane emissions estimated using single site optimization (SSO) of model parameters are underestimated by 9 g CH₄ m⁻² year⁻¹ on average (i.e. 50% higher than the site average of yearly methane emissions). While using the multi-sites optimization (MSO), methane emissions are overestimated by 5 g CH₄ m⁻² year⁻¹ on average across all investigated sites (i.e. 37% lower than the site average of yearly methane emissions).

1 Introduction

Atmospheric methane level estimated from ice cores analysis (Etheridge et al., 1998) and in situ measurements (Blake et al., 1982; Dlugokencky, 2019; Prinn et al., 2018) has nearly tripled since the pre-industrial equilibrium value i.e., from 680 ppb to reach a value of 1892 ppb in December 2020 (Dlugokencky, 2021; Saunois et al., 2020). This increase is consistent with the world population increase and the industrialization, such as the increase of fossil fuel extraction and use, of organic waste generation, and of livestock numbers (Raynaud et al., 2003).

Methane is the second most important anthropogenic greenhouse gas (GHG) after CO₂, and accounts for about 23% of the cumulative total radiative forcing (Etminan et al., 2016). In the troposphere methane is an ozone precursor and, in the stratosphere, methane interacts with hydroxyl radicals and carbon monoxide to produce water vapor. About 90% of CH₄ is oxidized by hydroxyl radical in the troposphere (Smith et al., 2003) and reactions with chlorine in the stratosphere or in the marine boundary layer (Allan et al., 2007; Thornton et al., 2010) leading to a residence time of about 9 years (Prather et al., 2012). At the continental surface, 5% to 10% of all methane sources are removed from the atmosphere by diffusion in soils and oxidation by soil microorganisms (Krüger et al., 2002; Prather et al., 1995; Smith et al., 2003, 1991; Tokida et al., 2007a, 2007b). Among natural sources, natural wetlands are the largest contributor and the most uncertain one in the global budget (Kirschke et al., 2013; Saunois et al., 2016). They contribute 25 – 30% of total methane emissions estimated by Saunois et al., (2020) and encompass anaerobic ecosystems composed of waterlogged or inundated ground that are capable
of methane production which include peatlands, mineral soil wetlands and floodplains. Peatlands are of particular interests because peat is composed of organic detritus and has an average carbon content of 52% dry mass (Gorham, 1991). Consequently, peatlands are large soil organic carbon reservoirs that could be functioning as a source of CH\textsubscript{4} and source or a sink of CO\textsubscript{2} to the atmosphere. They cover around 3% of surface continental lands but store approximately one third of the global soil carbon (Gorham, 1991). They are located in boreal and sub-arctic regions (80%, Strack et al., 2008) albeit some smaller areas are found in temperate and tropical regions (10-12%). Since the end of the last glaciation period (around 16500 years ago), northern peatlands have been functioning as long-term carbon sinks. This storage results from a delicate balance between carbon inputs (CO\textsubscript{2} absorbed by photosynthesis) and carbon outputs (CO\textsubscript{2} and CH\textsubscript{4} production, dissolved and particulate carbon). Clearly, in these ecosystems, processes controlling methane production, fluxes between the land surface and the atmosphere and feedback on climate are intimately connected.

The major pathway for methane production is via microbial processes which is limited by the availability of substrates (polymeric and monomeric compounds derived from carbohydrates, fatty acids, amino-acids, acetate and hydrogen; Blodau, 2002; Le Mer and Roger, 2001), the low oxygen content that is directly correlated to soil water content and by soil temperature. After its production, CH\textsubscript{4} migrates to the soil surface and is emitted to the atmosphere through three main processes (Bridgham et al., 2013): (1) diffusion through porous soil media; (2) ebullition, bubbles form in pores filled with water then quickly migrate to the surface; (3) plant-mediated fluxes via some vascular plant adapted to live in flooded environments. These plants developed aerenchyma to channel gas fluxes, oxygen is transported to roots and cells and CH\textsubscript{4} is transported from roots to the atmosphere (Bridgham et al., 2013; Smith et al., 2003).

Since the late 1980s, many CH\textsubscript{4} cycling processes has been mathematically described and included in terrestrial ecosystem models (Xu et al., 2016). These terrestrial ecosystems models have been outlined into two broad categories by Xu et al. (2016) review: (1) empirical models employed to evaluate observed processes of the CH\textsubscript{4} cycling and (2) process-based models used for budget quantification and to study sensitivity of CH\textsubscript{4} processes to environmental drivers. Unfortunately, so far only few global scale models feature peatland ecosystems, permafrost dynamics and CH\textsubscript{4} fluxes which are essential features to evaluate future climate changes and interactions between land surface and the atmospheres (Anav et al., 2013). Recent developments of ORCHIDEE land surface model lead to simulate soil hydrology, permafrost thermodynamic and carbon cycle in the northern latitudes (Guimberteau et al., 2018) and in the northern peatland specifically (Qiu et al., 2018), including peat carbon decomposition controlled by soil water content and temperature as well as CO\textsubscript{2} production and consumption processes (Largeron et al., 2018; Qiu et al., 2018). In the present study we adapt the Khvorostyanov et al. (2008a, 2008b) methane model to ORCHIDEE-PEAT (section 2.1) and calibrate and evaluate simulated emissions at northern peatland sites. To achieve model calibration, parameters were optimized with a data assimilation approach described in section 2.3. Parameters were optimized against methane fluxes at each site and from multiple sites simultaneously (section 3) in order to highlight parameter uncertainties while scaling up simulations from site to larger scale. The model evaluation is performed by discussing both optimization methods.
2 Model description

A general presentation of ORCHIDEE-PCH4 and associated processes are provided in section 2.1. Implementation of methane production and oxidation and transport are specified respectively in sections 2.1.1 and 2.1.2, whereas parameters values established for the site simulations conditions before observation periods are given in section 2.2. Section 2.3 describes the parameter optimization approaches.

2.1 ORCHIDEE-PCH4

The ORCHIDEE land surface model is a dynamic global process-oriented model that simulates carbon, water and energy fluxes between the biosphere, the land surface geosphere and the atmosphere. The carbon scheme describes photosynthesis, respiration, soil carbon cycle and CO$_2$ production and emissions. One of the branches of the ORCHIDEE land model aimed to improve the implementation of high latitude physical, hydrological and biogeochemical processes such as soil thermal processes, hydraulic processes, snowpack properties, and plant and soil carbon fluxes (ORCHIDEE-MICT, Guimberteau et al., 2018).

A northern peatlands scheme has been recently integrated to the model (ORCHIDEE-PEAT, Largeron et al., 2018; Qiu et al., 2018) which includes a peatland PFT (plant functional type) with adapted biological parameters was created allowing a separate calculation of the water balance. This PFT is defined as a flood tolerant C$_3$ grass with reduced productivity due to the lack of nutrients, and with a reduced rooting depth. For the present study, the ORCHIDEE-PEAT v2.0 (Qiu et al., 2019) has been further enriched with a module simulating methane production, oxidation and transport in northern peatlands and named ORCHIDEE-PCH4. To achieve this, the methane scheme described by Khvorostyanov et al., (2008a, 2008b) was revised according to high latitude processes and peatland ecosystem features. This early version was an idealized 1D soil model that accounted for heat and gas transport and soil organic carbon decomposition and production of CO$_2$ and CH$_4$ driven by soil water content and temperature in the soil column. In that early version, only a moss layer that serves as a thermal insulator was considered for the vegetation above ground (Khvorostyanov et al., 2008a). Soil moisture and carbon were treated as a single layer bucket scheme of 1 m depth containing a fixed amount of soil carbon content. In contrast, the ORCHIDEE-PCH4 is integrated into the peatland soil hydrological diffusion model (Largeron et al., 2018; Qiu et al., 2018) that incorporates water supply by precipitations and by runoff collected from other soils surrounding the peatland in the same grid-cell. The deep drainage is blocked to maintain soil water content at saturation in the bottom part of the peat soil. At the top of the water column, a dynamic water reservoir was added to represent standing water above the soil surface when water inputs exceed outputs and that soil is fully saturated. ORCHIDEE-PEAT simulates peat accumulation and decomposition to CO$_2$, of the three soil carbon pools, active, slow and passive that are vertically discretized in 32 layers accounting for a total maximum depth of 38 m (Qiu et al., 2019).
The methane scheme in Figure 1, delineates (1) methanogenesis of the three carbon pools, (2) methane and oxygen transport in the soil and snow layers, (3) transport of methane to the atmosphere by ebullition, (4) plant mediated transport and (5) methanotrophy by soil oxic conditions and roots exudates.

Each of these processes is constrained by soil temperature, soil water content ($\theta_{\text{soil}}$), soil O$_2$ concentration, atmospheric CH$_4$ concentration, leaf area and snow cover. The temporal variation of CH$_4$ in the soil layer ($z_2$) is assessed by:

$$\frac{\partial [\text{CH}_4(z,t)]}{\partial t} = f_{\text{MG}}, f_{\text{MG}_2} + f_{\text{MG}_3} + f_{\text{MG}_4} - f_{\text{MT}} - f_{\text{FMT}} - f_{\text{Ebu}} - f_{\text{Diff}} - f_{\text{Ebu}}$$

(1)

where each term varies in time (t) and with depth (z), expresses methane production ($f_{\text{MG}}$, MG: methanogenesis, a: active pool, s: slow pool, p: passive pool), transport by diffusion, ebullition, and plants ($f_{\text{Diff}}, f_{\text{Ebu}}, f_{\text{MT}}$) and oxidation ($f_{\text{MT}}, MT$: methanotrophy) processes. Net methane fluxes to the atmosphere is the sum of methane transport processes ($f_{\text{Ebu}}$ (Ebullition) and $f_{\text{MT}}$ (PMT: plant mediated transport)) and the amount of CH$_4$ that diffuses from the top soil layer at $z = 0$ to the atmosphere. Prognostic variables are defined per air volume i.e. the volume of gas in the air-filled pores ($v$) and gas dissolved in the water-filled pores (Khvorostyanov et al., 2008; Tans, 1998; Tang et al., 2013; Tang and Riley, 2014) assuming a constant equilibrium between gas concentrations in the air-filled and the water-filled part of pores. This gas volume is linked to the soil volume by the total CH$_4$ and O$_2$ in pores ($f_{\text{MG}}, f_{\text{MG}_2}, f_{\text{MG}_3}, f_{\text{MG}_4}$) defined as:

$$\epsilon_{\text{gas}} = v + \theta_{\text{soil}}\epsilon_{\text{gas}}$$

(2)

where $\theta_{\text{soil}}$ is the volumetric water content of the soil, $\epsilon_{\text{soil}}$ is the soil porosity and $B_{\text{gas}}$ is the Bunsen gas solubility coefficient defined for CH$_4$ and O$_2$, respectively, with $B_{\text{CH}_4}=0.043$ and $B_{\text{O}_2}=0.038$ (Hodgman, 1960; Wiesenburg and Guinasso, 1979).
2.1.1 Methane production and oxidation

Methanogenesis in soil occurs when oxygen concentration is limited for microorganisms and is considered for each type of soil carbon pools (i.e., i = a, s, p, in gC m⁻³ of soil), active, slow and passive:

\[ f_{MGi} = [C_i] \frac{k_{MG}}{q_{MG}} e^{-[(O_2)p]/[O_2]_{anoxia}} f_{clay} \]  

(3)

where the rate of methanogenesis (k, in s⁻¹) depends on soil temperature and moisture according to the same function as for the heterotrophic respiration (Qiu et al., 2019). This rate has been defined by (Klironostyanov et al., 2008a) to be 10 times lower than the rate of heterotrophic respiration. Here, q_{MG} determine the ratio between both rate of soil oxic and anoxic decomposition. \( [O_2] \) is the oxygen concentration in the soil \( [O_2]_{soil} \) (in gO₂ m⁻³ of soil) per unit porous volume \( \frac{e_{soil}}{m_{soil}} \) r_{oil} is the soil porosity). \( [O_2]_{anoxia} \) is the soil oxygen concentration at which anoxic conditions are reached and enable methane production. This oxygen concentration threshold is assumed to be 2 g m⁻³ (Duval and Goodwin, 2000). Soil clay content affects the decomposition of the active soil carbon pool (Parton et al., 1988):

\[ f_{clay} = 1 - (0.75 \times clay) \]  

(4)

where clay is the clay fraction and has a value of 0.2, neither the slow nor the passive pools are modified by \( f_{clay} \). Methane is oxidized to CO₂ in aerated soil layers. The amount of methane consumed by methanotrophy is limited by the soil oxygen concentration \( [O_2]_{soil} \) following a 1:2 CH₄:O₂ molar ratio:

\[ f_{MT} = k_{MT} \frac{1}{2} \frac{[O_2]_{soil}}{M_{O_2}} e_{CH₄} \]  

(5)

where \( k_{MT} \) is the rate of methanotrophy which value range from 0.06 to 5 d⁻¹ (Morel et al., 2019), the conversion of oxygen to methane content is provided by methane and oxygen molecular weight \( M_{CH₄} \) and \( M_{O_2} \) and their respective total gas porosities \( e_{CH₄} \) and \( e_{O₂} \).

2.1.2 Methane transport

The formation of methane bubbles in water-filled pores is determined by:

\[ f_{Ebu} = k_{Ebu} [CH₄]_{soil} - [CH₄]_{ET} P_{Ebu} \]  

(6)

where \( k_{Ebu} \) is a rate constant of 1 h⁻¹, methane ebullition occurs when methane concentration exceeds a concentration threshold that depends on soil temperature \( (T_{soil}) \) and pressure \( (P_{soil} \ in \ Pa) \) above 0.75 m depth as depth:

\[ [CH₄]_{ET} = \frac{\max_{CH₄} P_{soil} M_{CH₄}}{K_{T_{soil}} K_{CH₄}} \]  

(7)
Methane concentration gradient is also modified by the vertical distribution of roots in the soil around them in which methane will be oxidized. The proportion of methane oxidized to the atmosphere, \( f_{\text{FMT}} \), is expressed as:

\[
f_{\text{FMT}} = k_{\text{FMT}} \times T_{\text{weg}} \times f_{\text{root}} \times LAI \times \left( [CH_4]_{\text{soil}} - [CH_4]_{\text{atmos}} \right) \times (1 - M_{\text{rom}}),
\]

where \( k_{\text{FMT}} \) is a rate constant of the unit \( 0.01 \text{ h}^{-1} \) and \( T_{\text{weg}} \) has been defined by Walter and Heimann, (2000) as a factor that describes the efficiency of plants in methane transport depending on the type and the density of these plants. Its value ranges between 0-15 with shrubs and trees being poorly efficient and grasses and sedges being very efficient in gas transport.

Methane concentration gradient is also modified by the vertical distribution of roots in the soil as:

\[
f_{\text{root}} = 2 \times \left( \frac{z_{\text{root}} - z_{\text{soil}}}{z_{\text{root}}} \right),
\]

where \( z_{\text{root}} \) is the rooting depth and \( z_{\text{soil}} \) the soil depth. The leaf area index (LAI) influences the methane flux which varies by growing stage of the plants.
The gas diffusion scheme features the diffusion of CH₄ and O₂ in the 3 top layers of snow when snow cover is formed and in the 32 soil layers that correspond to 38 m depth. This scheme considered (1) the diffusion of oxygen from the top soil to the snow layer, (2) the diffusion of methane produced and remaining in the soil and (3) methane exchange between the soil and the atmosphere at z=0:

\[ f_{\text{diff}} = D_{\text{gas}}(x) \frac{\partial \rho_{\text{gas,soil}}(x,t)}{\partial x} \]  

(11)

Diffusion coefficients, \( D_{\text{gas}} \), are based on the diffusivity of each gas in air (\( D_{\text{gas,air}} \)) and in water (\( D_{\text{gas,water}} \)):

\[ D_{\text{gas}} = \left( D_{\text{gas,air}} \nu + D_{\text{gas,water}} \eta \right) \]  

(12)

where \( \nu \) is the volume of gas in the air-filled pores, \( \eta_{\text{soil}} \) is the volumetric water content of soil, \( \pi_{\text{soil}} \) is the soil porosity and \( B_{\text{gas}} \) is the Bunsen coefficient of the gas, the tortuosity \( \eta \) is defined to be 2/3 (Hillel, 1982). Diffusivity of O₂ in air and in water are defined respectively to 1.6 \( \times \) 10⁻⁵ and 1.6 \( \times \) 10⁻⁹ m²/s and for methane 1.7 \( \times \) 10⁻⁵ and 2.0 \( \times \) 10⁻⁹ m²/s (Khvorostyanov et al., 2008a). The diffusion is discretized using a forward time centered space method (Press et al., 1997) and converted in a tridiagonal system of equations before being solved using forward then backward substitution method. A time-splitting option is also implemented for the diffusion of large concentrations of gas per time step.

The only source of oxygen considered is from the atmosphere and is determined using atmospheric surface pressure, temperature and an atmospheric O₂ mixing ratio of 20.9%. Atmospheric methane content is also defined in the same way employing a methane mixing ratio of 1.7 ppm and is used as a boundary condition when the topsoil layer is in contact with the atmosphere. In winter, when snow accumulates above the topsoil, these atmospheric boundary conditions are applied to the top snow layer then gases diffuse from and to the atmosphere through the snow layers then soil layers. Methane and oxygen diffusivity in the snow are defined by:

\[ D_{\text{gas}} = D_{\text{gas,air}} \left( 1 - \frac{\rho_{\text{snow}}}{\rho_{\text{ice}}} \right) \eta_{\text{snow}} \]  

(13)

where \( D_{\text{gas,air}} \) the diffusion coefficient of each gas in free air, the snow porosity that is defined by the ratio of density of snow \( \rho_{\text{snow}} \) and ice \( \rho_{\text{ice}} \) and the tortuosity \( \eta_{\text{snow}} \) is equal to \( \sqrt{\left( 1 - \frac{\rho_{\text{snow}}}{\rho_{\text{ice}}} \right)} \). Snow density is determined by the snowpack scheme (Wang et al., 2013) with the density of the ice is of 920.0 kg/m³.

2.2 Sites description and simulation setup

The model was evaluated on 14 peatland sites distributed on the Eurasian and American continents in boreal and temperate northern regions (from 41°N to 69°N). These sites are a subset of the 30 peatlands sites collected for the calibration of ORCHIDEE-PEAT (Qiu et al., 2018) for which, in addition of eddy-covariance data and physical variables (water table, snow depth, soil temperature), methane emissions were measured by eddy-covariance at daily time scale at US-Los, hourly timescale at DK-Nuf and otherwise at half-hourly timescale or chamber measurements at monthly timescale for FR-Lag and RU-Chq. All methane emissions data were monthly average. At DE-Sfn, DE-Himm, FI-Lom, PL-Kpt, PL-Wet,
and US-Wpt, year-round data were available and zero values were filled for the first and the last month of years at the beginning and the end of the observation period. Otherwise, winter months were filled with zero and during spring, summer and fall months missing data were filled gapped using a linear regression. Descriptions of the sites were provided in Qiu et al., (2018). In Table 1, sites are assembled by increasing extreme values of mean monthly measurements of methane emission then by locations and ecological characteristics. The extreme values of mean monthly measurements are the most reliable quantity of methane fluxes since periods of observation and monitoring frequency differ. Among the 14 peatlands, 9 sites are located in temperate regions, 3 in boreal regions and 2 in arctic permafrost regions. The majority of the sites are fen (9 sites) and the others are 3 bogs (DE-Sfn, US-Bog, DE-Hmm), a marsh (US-Wpt) and a tundra (RU-Che). It is worth noticing that there is no obvious correlation between the magnitude of the monthly mean fluxes and types of ecosystems. Indeed, US-Los and DE-Spw are temperate fens that release less than 10 mg CH₄ m⁻² d⁻¹. Sites emitting 10 - 150 mg m⁻² d⁻¹ are located in Germany, Northwestern America and France among which half are fens and the other half are bogs. Half of them, DE-Sfn, US-Bog and CA-Wp1 are forested peatlands and release less than 55 mg CH₄ m⁻² d⁻¹. While the others, DE-Zrk, DE-Hmm and FR-Lag experienced a temporary drainage event because of anthropogenic activities during years earlier than the observed period. Sites located in Finland, Denmark and Poland are fens emitting between 150 and 400 mg m⁻² d⁻¹. The largest methane emitters are the arctic tundra RU-Che and the marsh US-Wpt which released more than 500 mg m⁻² d⁻¹. All sites are covered with some snow during winter and US-Bog and RU-Che are underlaid with permafrost located below 0.5 meter.

Table 1. Sites characteristics. Sites identification includes the country initials and the short three letters name of each site, locations of the sites are provided by the country, latitude (Lat) and longitude (Lon) values. Hydrological characteristics are distinguished by the type of ecosystem, fen, bog, tundra and marsh. Y and N indicate presence and absence of snow cover in winter, permafrost soil, forest above the peat. Temporary drawdown of the water table level is specified by presence and absence indicators Y or N. Grey color highlight peatlands with methane emissions of 0-10, 10-150, 150-400, 400-600 mg m⁻² d⁻¹.

| Sites  | Site name     | Country  | Lat  | Lon  | Climatic zone | Types            | Observed period (year range) | Monthly mean methane emissions (mg m⁻² d⁻¹, min, max) | Forest (Y/N) | Drained (Y/N) | Snow (Y/N) | Permafrost (active layer depth in m, Y/N) |
|--------|---------------|----------|------|------|---------------|------------------|-------------------------------|--------------------------------|--------------|--------------|------------|----------------------------------------|
| US-Los | Lost Creek    | United States | 46.08 | -89.98 | temperate | fen              | 2006                          | -1.1, 3.6                     | N            | Y            | Y          | N                                      |
| DE-Spw | Spreewald     | Germany  | 51.89 | 14.03 | temperate | fen              | 2011                          | -1.4, 6.5                     | Y            | N            | N          | N                                      |
| DE-Sfn | Schechenfliz Nord | Germany  | 47.81 | 11.33 | temperate | bog              | 2012-2014                    | 0.7, 38.0                     | Y            | N            | Y          | N                                      |
| DE-Zrk | Zarnekow      | Germany  | 53.88 | 12.89 | temperate | fen              | 2013                          | 0, 37.9                       | N            | Y            | Y          | N                                      |
| CA-Wp1 | AB-Western Peatland | Canada  | 54.95 | -112.47 | boreal | fen              | 2007                          | 0, 49.3                       | Y            | N            | Y          | N                                      |
| US-Bog | Bog at Bonanza Creek | United States | 64.7 | -148.32 | boreal | bog              | 2013                          | 0, 54.4                       | Y            | N            | Y          | (0.5-0.9)                               |
| FR-Lag | LaGuette      | France   | 47.3 | 2.3   | temperate | fen              | 2014-2016                    | 0.99, 2                       | N            | Y            | Y          | N                                      |
Each peatland site is a sub-grid area embedded in the 0.5° x 0.5° grid cells whose extent is determined by a fraction of grid area as defined in Table 2. These sub-grid areas enable the representation of ecosystems variability in which a specific scheme simulates soil hydrology, vegetation characteristics and soil carbon cycling for northern peatlands. The fraction of peatlands per grid cell was defined by modifying the prescribed values employed by Qiu et al., (2018) in order to collect enough water to fill the peatland by runoff from the other soil fractions and elevate the water table level for northern peatlands. We employed vegetation phenotype properties and peatland fractions described in (Qiu et al., 2019) and peatlands hydrology and carbon model as described in Qiu et al., (2019). Site simulations were then constrained at the grid cell scale with a half hourly time series of meteorological conditions e.g. air temperature, wind speed, wind direction, longwave incoming radiation, shortwave incoming radiation, specific humidity, atmospheric pressure, and precipitation. These time series are flux tower measurements that were gap filled by 6-hourly CRU-NCEP 0.5° global climate forcing dataset (Qiu et al., 2018). Other variables measured on a half-hourly time step at sites e.g. CO₂ and energy (latent heat: LE; sensible heat: H) fluxes, water table position, soil temperature, and snow depth served for the calibration of peatland soil and vegetation phenotype characteristics such as the maximum rate of carboxylation (Vcmax). Optimized Vcmax values (Qiu et al., 2018) are utilized to capture spatial carbon fluxes gradients (gross primary production, ecosystem respiration and net ecosystem exchange) at each peatland site. The peat model (Qiu et al., 2019) enables a vertical buildup of peat by simulating a downward movement of C when the discretized organic layers reach a threshold defined from a regression relationship between the carbon fraction and measured bulk density. This scheme in ORCHIDEE-PCH4 serves to constrain the vertical distribution of the soil carbon stock to the observed maximum peat depth. Simulations with ORCHIDEE-PCH4 driven by repeated site-specific meteorological conditions were performed for various periods of time to reach the observed soil carbon content and maximum peat depth (Table 2).
Table 2. Simulations conditions and framework to constrain peatlands soil carbon stock. Grey color reports sites with equivalent levels of methane emissions (Table 1).

| Sites identification | Peat fraction | Vcmax | Carbon accumulation period | Maximum peat depth | Soil carbon stock | References |
|----------------------|--------------|-------|---------------------------|-------------------|------------------|------------|
|                      | fraction     | µmol m⁻² s⁻¹ | numbers of years | Observed | Simulated | Observed | Simulated |
| US-Los               | 0.16         | 65    | 214                       | 0.5                   | 0.75             | 27.5      | 28.0      | Sulman et al., 2009; Chason and Siegel, 1986 |
| DE-Spw               | 0.14         | 89    | 272                       | 1.2                   | 1.5               | 84.0      | 84.2      | Dettmann et al., 2014 |
| DE-Sfn               | 0.18         | 45    | 4 544                     | 5                     | 5                 | 372.8     | 372.5     | Hommeltenberg et al., 2014 |
| DE-Zrk               | 0.9          | 33    | 10 060                    | 10                    | 7                 | 696.7     | 696.6     | Zak et al., 2008 |
| CA-Wp1               | 0.16         | 38    | 620                       | 2                     | 2                 | 51.0      | 51.0      | Benscoter et al., 2011; Long et al., 2010 |
| US-Bog               | 0.27         | 42    | 4 305                     | 2                     | 3                 | 207.4     | 207.7     | Manies et al., 2017 |
| FR-Lag               | 0.22         | 42    | 937                       | 1.6                   | 2                 | 121.0     | 121.4     | Gogo et al., 2011; Leroy et al., 2019 |
| DE-Hmm               | 0.9          | 35    | 8 963                     | 3                     | 3                 | 265.0     | 266.4     | VybornoVa, 2017 |
| FI-Lom               | 0.27         | 28    | 6 396                     | 3                     | 3                 | 200.3     | 200.5     | Lohila et al., 2010 |
| DK-NuF               | 0.5          | 31    | 8 959                     | 0.75                  | 1.5               | 54.6      | 54.6      | Bradley-Cook and Virginia, 2016 |
| PL-Kpt               | 0.14         | 52    | 3 819                     | 2.5                   | 3                 | 250.0     | 250.3     | Jaszczyński, 2015 |
| PL-Wet               | 0.11         | 52    | 261                       | 0.5                   | 0.75              | 37.6      | 37.8      | Milecka et al., 2016; Zak et al., 2008 |
| US-Wpt               | 0.27         | 80    | 32                        | 0.3                   | 0.75              | 5.3       | 5.4       | Chu et al., 2014 |
| RU-Che               | 0.05         | 35    | 2 968                     | 0.56                  | 0.75              | 45.8      | 45.8      | Dutta et al., 2006 |

During the first part of those simulations, atmospheric CO₂ concentration was set to preindustrial value at 285 ppm, and then between 1860 until the beginning of the respective observation period of methane emissions listed in Table 1, CO₂ concentration had risen. During soil carbon accumulation simulations, methane model parameters were defined to the default values defined in Table 3. Then during the site-specific measurement periods (Table 1), methane variables are calibrated.
against observed monthly average methane fluxes times series. Then site-specific simulation over the observed period is run again using the optimized parameters.

Table 3. List of parameters driving the methane production, oxidation and transport scheme in ORCHIDEE-PCH4.

| Parameters | Description | Unit | default values | ranges | References |
|------------|-------------|------|----------------|--------|------------|
| $q_\text{MG}$ | Ratio of soil oxic and anoxic decomposition | proportion | 10.0 | 9.0, 11.0 | Khvorostyanov et al., 2008a, Wania et al. 2010 |
| $k_\text{MT}$ | Methanotrophy rate | 1/d | 5.0 | 1.0, 5.0 | Khvorostyanov et al., 2008a; Morel et al., 2019 |
| $M_{\text{root}}$ | Root methane oxidation | fraction | 0.5 | 0.0, 1.0 | Walter and Heimann, 2000 |
| $Z_{\text{root}}$ | Root depth | m | 0.3 | 0.01, 0.5 | Walter and Heimann, 2000 |
| $T_{\text{veg}}$ | The efficiency of methane plant mediated transport | proportion | 7.0 | 0.0, 15.0 | Walter and Heimann, 2000 |
| $w_{\text{size}}$ | Connectivity of soil moisture | m | 0.01 | 0.001, 0.1 | Khvorostyanov et al., 2008a |
| $m_{\text{XCH4}}$ | Methane mixing ratio in bubbles | fraction | 0.27 | 0.05, 0.53 | Walter and Heimann, 2000; Riley et al., 2011; Morel et al., 2019 |

2.3 Optimization of methane parameters

The methane scheme revisited in ORCHIDEE-PCH4 (described in section 2.1) is driven by 7 parameters (Table 3) that constrain methane production ($q_\text{MG}$), oxidation ($k_\text{MT}$, $M_{\text{root}}$) and transport ($m_{\text{XCH4}}, w_{\text{size}}, T_{\text{veg}}, Z_{\text{root}}$). In order to optimize these parameters, we employed the ORCHIDEE data assimilation system (Bastrikov et al., 2018) that relies on the minimization of a cost function employing a Bayesian statistical formalism that expresses the discrepancy between observations and simulated methane emissions and the difference between the optimized parameter values and the prior information on them, weighted by the uncertainties assigned to both observations and parameters. A random search algorithm based on the genetic algorithm (GA) serves to randomly iterate the set of seven parameters following the principles of genetics and natural selection similar to chromosome genetic sequencing (Goldberg, 1989; Haupt and Haupt, 2004). At each iteration, 8 sets of parameters are defined from the previous iteration following crossover and mutation rules (Bastrikov et al., 2018). The frequency at which these rules are used, is governed by the crossover/mutation ratio fixed to 4 : 1, the number of parameter blocks exchanged during crossover that is 2 and the number of parameters perturbed during mutation that is equal to 1. In addition, a ranking in ascending order of the corresponding cost function values of all sets of parameters serves to selectively preserve the set of parameters that reduce the gap between observation and simulation data.
Two types of simulations are performed over the site-specific observation period defined in Table 1: single site (SS) experiment for which parameters are optimized for each site and a multi-site (MS) experiment that aims at refining one set of parameters considering all sites together. The single site experiments are performed for 100 iterations and aim at finding the lowest cost function employing the model-data root mean square difference (RMSD). Prior conditions for the single site experiment are described and listed in Table 3. Initial parameters values and range were derived from literature and of expert knowledge and parameter uncertainties are defined as 40% of the prescribed ranges. Across sites, mean values of each parameter serve as prior conditions for the multi-site experiment. This later was performed for 50 iterations and aims to evaluate methane emissions uncertainties at hemispheric-scale when only one set of parameters is employed.

3 Results

3.1 Single site optimization (SSO)

For each site, to minimize the discrepancy between observed and simulated methane emissions, iterative single site simulations were performed. Successive runs serve to ensure that the minimum reached is not a local minimum. Results from the last minimization experience are reported in Table 4 (uncertainties in parameters at sites are in Table S1). As expected, most optimized parameters fit within the initial range defined in Table 3 except for four of the sites. One of these four sites, DE-Spw, is among the sites that emit the fewest amount of methane (up to 7 mg m\(^{-2}\) d\(^{-1}\)) and features a larger stock of carbon of 84 kg C / m\(^2\) than at US-Los that features 27 kgC / m\(^2\) and emits up to 4 mg m\(^{-2}\) d\(^{-1}\). This explains, at the DE-Spw site, that the optimized value of wsize was reduced to 0.5 mm to maintain low methane emissions. The other three sites for which some of the optimized parameters are out of the initial range, DK-Nuf, PL-Wet and US-Wpt, are among the sites that emit more than 150 mg CH\(_4\) m\(^{-2}\) d\(^{-1}\). The carbon stock at DK-Nuf and PL-Wet are respectively 55 and 38 kg C / m\(^2\) which is lower than at FI-Lom and PL-Kpt that accumulated more than 200 kg C / m\(^2\). Three parameter ranges were modified for DK-Nuf, the minimum value of \(q_{\text{MG}}\) was lowered to 7.0, \(z_{\text{max}}\) maximum is increased to the maximum peat depth at 0.75 m in order to consider plant mediated transport in all the peat layers. the maximum value of \(T_{\text{veg}}\) was increased to 40.0 and the maximum rate of methanotrophy \(k_{\text{MT}}\) was enlarged up to 8 d\(^{-1}\) to decrease the methane oxidation and to obtain in the simulation methane emissions higher than 150 mg CH\(_4\) m\(^{-2}\) d\(^{-1}\). PL-Wet required also to modify range values of \(q_{\text{MG}}\) to 1.0-11.0 leading to the lowest optimized \(q_{\text{MG}}\) value of 4.0 which significantly reduced the RMSD from 227.4 to 80.5 (Fig. S1 and Table S2). For the US-Wpt site, \(q_{\text{MG}}\), \(k_{\text{MT}}\) and \(T_{\text{veg}}\) were adjusted to increase methane production and fluxes in order to balance the carbon stock of 5 kg C / m\(^2\) that is lower than the one at RU-Che.
Table 4. Single site optimized values of methane scheme parameters for each peatland site. In parenthesis are indicated prior parameter ranges which differ from the values in Table 3. Uncertainties for these ranges are specified in parentheses.

| Sites     | \( q_{\text{MG}} \) | \( k_{\text{MT}} \) | \( M_{\text{ex}} \) | \( Z_{\text{root}} \) | \( T_{\text{veg}} \) | wsize | \( \text{mxr}_{\text{CH}_4} \) |
|-----------|----------------------|----------------------|----------------------|----------------------|----------------------|-------|----------------------|
|           | proportion 1/d       | fraction m           | proportion m         | fraction m           |                      |       |                      |
| US-Los    | 9.9                  | 1.92                 | 0.994                | 0.057                | 3.8                  | 0.0319| 0.306                |
| DE-spw    | 9.9                  | 1.00                 | 0.595                | 0.188                | 0.003                | 0.0005| 0.530                |
| DE-Sfn    | 10.5                 | 1.98                 | 0.493                | 0.399                | 0.01                 | 0.0010| 0.377                |
| DE-Zrk    | 10.0                 | 1.98                 | 0.756                | 0.418                | 9.8                  | 0.0015| 0.259                |
| CA-Wp1    | 10.2                 | 2.99                 | 0.471                | 0.122                | 0.45                 | 0.0059| 0.193                |
| US-Bog    | 9.2                  | 2.45                 | 0.500                | 0.173                | 4.4                  | 0.0098| 0.117                |
| FR-Lag    | 10.7                 | 1.74                 | 0.857                | 0.291                | 0.5                  | 0.0085| 0.463                |
| DE-Hmm    | 9.4                  | 3.94                 | 0.147                | 0.118                | 3.7                  | 0.0011| 0.164                |
| FI-Lom    | 9.5                  | 3.97                 | 0.491                | 0.174                | 5.7                  | 0.0040| 0.140                |
| DK-NuF    | 8.5 (7.0, 11.0)      | 4.38                 | 0.068                | 0.677 (0.01,0.75)    | 23.6 (0.0, 40.0)     | 0.0255| 0.203                |
| PL-Kpt    | 10.3                 | 1.32                 | 0.541                | 0.071                | 9.1                  | 0.0030| 0.061                |
| PL-Wet    | 4.0 (1.0, 11.0)      | 1.95                 | 0.165                | 0.328                | 6.0                  | 0.0110| 0.136                |
| US-Wpt    | 7.9 (7.0, 11.0)      | 5.25 (1.0, 8.1)      | 0.035                | 0.304                | 22.3 (0.0, 40.0)     | 0.0023| 0.120                |
| RU-Che    | 9.8                  | 1.36                 | 0.004                | 0.404                | 8.4                  | 0.0171| 0.294                |
| Uncertainty | 0.8 (1.6)         | 1.6 (2.8)           | 0.4                  | 0.196 (0.296)        | 6.0 (16.0)           | 0.0398| 0.192                |

Across sites, \( q_{\text{MG}} \) values extend between 9.0 and 10.7, optimized \( k_{\text{MT}} \) values vary between 1 and 5.25 d\(^{-1}\). The fraction of methane that is oxidized at the root (\( M_{\text{ex}} \)) level fluctuate between 0.004 and 0.99 with the lowest values obtained at US-Wpt and RU-Che sites that emitted up to 500 mg CH\(_4\) m\(^{-2}\) d\(^{-1}\) and the largest values at US-Los that released the fewest amount of methane. The optimization of the maximum root depth (\( Z_{\text{root}} \)) results in values ranging between 0.057 and 0.68 with a maximum value at the DK-Nuf site which is an arctic fen in Greenland. Optimized values for plant mediated transport efficiency (\( T_{\text{veg}} \)) fell between 0.003 and 23.6. The largest \( T_{\text{veg}} \) values of 23.6 and 22.3, were obtained for DK-Nuf and US-Wpt, respectively and the lowest value of 0.003 at DE-Spw. The dimension of water droplets dispersed in the soil depict the probability of methane rich bubbles to be released to the atmosphere (wsize). The optimized wsize values vary within the range 0.005 and 0.032. And the optimized mixed ratios of methane involved in the ebullition process (\( \text{mxr}_{\text{CH}_4} \)) are ranging between 0.06 and 0.53.
Table 5. Discrepancies between observed and simulated methane emissions are quantified by the root mean square difference (RMSD) approach. Minimization efficiency of each test is indicated by the relationship between the prior using default values and posterior RMSD as (1 - RMSDpost / RMSDprior)×100. Normalized root mean square difference (NRMSD) is defined by the RMSD posterior normalized by the annual mean of observed methane emissions.

| Sites       | RMSD prior | RMSD posterior | 1-(RMSDpost/RMSDprior) | Observed emissions annual mean (mgCH₄ m⁻² d⁻¹) | NRMSD |
|-------------|------------|----------------|------------------------|-----------------------------------------------|-------|
| US-Los      | 69.6       | 1.1            | 0.98                   | 0.1                                           | 9.85  |
| DE-spw      | 687.9      | 9.5            | 0.99                   | 0.5                                           | 19.00 |
| DE-Sfn      | 263.3      | 9.2            | 0.97                   | 3.9                                           | 2.36  |
| DE-Zrk      | 16.2       | 4.6            | 0.71                   | 6.2                                           | 0.74  |
| CA-Wp1      | 73.6       | 11.8           | 0.84                   | 8.9                                           | 1.32  |
| US-Bog      | 33.0       | 6.7            | 0.80                   | 28.6                                          | 0.23  |
| FR-Lag      | 91.4       | 23.0           | 0.75                   | 26.9                                          | 0.85  |
| DE-Hmm      | 34.4       | 25.3           | 0.26                   | 21.2                                          | 1.19  |
| FI-Lom      | 44.0       | 38.3           | 0.12                   | 25.2                                          | 1.52  |
| DK-NuF      | 44.6       | 40.1           | 0.10                   | 52.7                                          | 0.76  |
| PL-Kpt      | 146.5      | 54.6           | 0.63                   | 56.1                                          | 0.97  |
| PL-Wet      | 181.3      | 80.5           | 0.56                   | 93.2                                          | 0.86  |
| US-Wpt      | 265.5      | 249.0          | 0.06                   | 196.0                                         | 1.27  |
| RU-che      | 157.4      | 139.7          | 0.11                   | 80.4                                          | 1.74  |

Differences between observed and simulated methane fluxes employing initial and optimized parameters are quantified by the RMSD prior and posterior respectively. At sites where methane fluxes were small such as US-Los and DE-Spw, RMSD posterior values are respectively of 1.1 and 9.5 whereas at US-Wpt and RU-Che where monthly mean methane emissions had reached up to 550 mgCH₄ m⁻² d⁻¹ RMSD posterior are larger i.e. respectively 249 and 140. At sites that emitted between 10 and 150 mgCH₄ m⁻² d⁻¹, RMSD values fluctuate between 4 and 26 and when methane fluxes were between 150 and 400 mgCH₄ m⁻² d⁻¹, RMSD is of 38 - 80. Performances of the optimization at each site are also evaluated utilizing the relationship (1-RMSDpost/RMSDprior)x100 which compares the RMSD prior defined by using the prior values and ranges and the RMSD posterior obtained after parameters optimization. It might seem that optimizations are more efficient at sites with low methane emissions than at sites that emitted the most whereas NRMSD values which are the RMSD posterior normalized by the annual mean of the observed emissions are close to 1 at each site except for US-Los and DE-Spw for which NRMSD are 10 and 19 respectively. This suggests that the optimizations are less efficient for sites that emitted the least amount of methane. Direct comparison during the period of observation between observed and simulated methane emissions are displayed for each site in Figure 2b, 3b, 4b and 5b. The temporal and the average magnitude are
equivalent than in measurements except for the US-Wpt and RU-Che for which simulated emissions are much lower than observed emissions.

In addition of the mismatch between observed and simulated methane emissions during the observed period, figures 2, 3, 4 and 5 show the simulated water table position, the amount of methane that is emitted by diffusion, plant transport and ebullition, temporal methane concentration in the soil and in the snow and the depth at which the largest amount of methane is produced together with the rate of production at that depth. These variables show the consistency of the model regarding peatlands functioning. US-Los and DE-Spw were emitting less than 10 mg CH₄ m⁻² d⁻¹ and their simulated water table positions fluctuate below the surface between 10 and 60 cm while showing a clear seasonal pattern and are lower in summer than in winter. In winter, simulated emissions are the result of methane diffusion between the soil and the atmosphere while in spring and summer methane mainly diffuses through aerenchyma of vascular plants. At DE-Spw, simulated methane concentration in the soil that ranges between 40 and 140 g/m², is more than 10 times higher than at US-Los which observed concentration barely reaching 5 gCH₄/m² in the fall. Model simulates a methane accumulation in the soil at DE-Spw that stimulates a small release of methane to the atmosphere by ebullition. In the model, the largest production of methane occurs consistently around 20 cm for US-Los and 40 cm for DE-Spw which is above the simulated water table position. It is commonly expected for methanogenesis to take place below the observed water table position. However, here the simulated water table position is a prognostic variable defined by the cumulative amount of soil water content over the soil column (Fig. S2 and Fig. S3). Indeed, in these simulations above the water table position soil moisture is still higher than 80% (Fig. S4 and Fig. S5). At those depths the simulated methane productions reach up to respectively 0.2 and 1.0 g CH₄/m² in the summer. In the winter, simulated methane productions are nearly very small and some methane is diffused in the simulated snow pack covering the peatlands, up to 0.025 g CH₄/m² at US-Los and 0.17 g CH₄/m² at DE-Spw. This explains the negative methane flux (Figure 2c) produced in winter by the model via simulated diffusion of atmospheric methane in the snow cover (Figure 2d). Then the positive flux that appears in the spring that occurs simultaneously to snow melting.
Figure 2: Temporal distribution of methane at sites emitting less than 10 mg CH$_4$ m$^{-2}$ d$^{-1}$. (a) Simulated water table position estimated from the soil water content; (b) Simulated (dark line) and observed (gray line) methane emissions released to the atmosphere; (c) Cumulative amount of simulated methane emitted by diffusion, plant mediated transport and ebullition; (d) Methane concentration in the soil layers (dark line) and in the snow layers of the model (gray line); (e) On the left, depth at which simulated methane production is the highest in the soil, scaled to the maximum peat depth. On the right, the amount of simulated methane produced at these depths.

Other sites that emitted less than 150 mg CH$_4$ m$^{-2}$ d$^{-1}$ are shown in Figure 3. Except for CA-Wp1 and US-Bog, during winter these peatlands are nearly inundated in the simulations with a simulated water table position near 10 cm above ground level. CA-Wp1 and US-Bog are respectively fen and bog boreal peatlands and their simulated water table position is lower than at the other sites. US-Bog is affected by permafrost which might explain the unexpectedly low position of the
simulated water table. At DE-Sfn, methane is mainly transported in the model via vascular plants and by ebullition whereas at the other sites, simulated methane is predominantly carried via vascular plants only. As for US-Los and DE-Spw, at CA-Wp1, during the winter simulations show that in the top soil layers some methane is transferred by diffusion (Figure 3c) to the snow cover (Figure 3d). Then a small part of the simulated methane is temporarily stored in the snow (Figure 3d) and the other part is released to the atmosphere via diffusion (Figure 3c). More simulated snow accumulated at DE-Sfn, DE-Zrk, CA-Wp1 and US-Bog where up to 0.8 - 0.04 gCH₄/m² are temporarily stored in the snow (Figure 3d). At FR-Lag and DE-Hmm, fewer methane, less than 0.005 gCH₄/m², are contained in the simulated snow cover (Figure 3d). As for DE-Spw, at DE-Sfn, simulation results show that up to 140 gCH₄/m² accumulate in the soil layers of the model during winter and provide sufficient methane to be expelled to the surface by ebullition. In contrast, methane accumulated up to 80 gCH₄/m² in the soil layers of the model at CA-Wp1 is not sufficient to trigger the methane ebullition process. In all the other sites, methane concentrations in the soil layers of the model are smaller between 5 and 35 gCH₄/m². The maximum of simulated methanogenesis takes place steadily at around 20 cm depth at DE-Sfn, FR-Lag and DE-Hmm which is in winter about 30 cm under the simulated water table position. At this depth simulated methane production fluctuated at 0.01-0.12 gCH₄/m². At DE-Sfn, CA-Wp1 and US-Bog, simulations show that in the winter most of the methane is produced at around 75 cm depth then in spring and summer the depth of maximum simulated production becomes shallower to reach 20 cm. Early spring at US-Bog, temporarily the maximum simulated production is near the surface at 1 cm depth which correlates with an increase of methane that accumulates in the simulated snow. At DE-Sfn, the depth at which the maximum simulated production occurred fluctuates more than at both other sites, CA-Wp1 and US-Bog. Unlike CA-Wp1 and US-Bog, during the first two years the maximum simulated production deepens at 75 cm when the maximum value of simulated production is reached.
Figure 3: Temporal distribution of methane for sites emitting between 10 and 150 mg CH₄ m⁻² d⁻¹. (a) Simulated water table position estimated from the soil water content; (b) Simulated (dark line) and observed (gray line) methane emissions released to the atmosphere; (c) Cumulative amount of simulated methane emitted by diffusion, plant mediated transport and ebullition; (d) Methane concentration in the soil layers (dark line) and in the snow layers (gray line) of the model; (d) On the left, depth at which simulated methane production is the highest in the soil, scaled to the maximum peat depth. On the right, the amount of simulated methane produced at these depths.

Sites that emitted between 150 and 400 mg CH₄ m⁻² d⁻¹ are temperate, sub-arctic and arctic fens (Figure 4). Simulated water table positions at FI-Lom, DK-Nuf and PL-Wet are lower in winter than in summer. During the observed period of three years, the simulated water table position at PL-Kpt is lower in summer the first and the last year of observations and higher in summer during the second year. In the winter the methane fluxes are stored in the simulated snow cover at FI-Lom (Figure 4d), therefore the simulated surface fluxes above the snow are driven by diffusion (Figure 4c). However, during summer simulated methane fluxes essentially originate from plant mediated transport. At DK-Nuf, PL-Kpt and PL-Wet, simulation results show that fewer methane, less than 0.4 g CH₄ m⁻² d⁻¹, accumulates in the simulated snow during winter (Figure 4d). Methane is transported by vascular plants in summer at DK-Nuf and PL-Wet whereas at PL-Kpt simulated methane fluxes are provided by both vascular plants and ebullition. This is consistent with high soil methane concentrations at PL-Kpt during summer that are near 70 g CH₄ m⁻² the first year and near 90 g CH₄ m⁻² the last two years of observation. In contrast, at FI-Lom simulated soil methane concentrations are near 50 g CH₄ m⁻² during summer whereas the...
winter concentrations are near 80 gCH₄ m⁻² (Figure 4d) which is not sufficient to cause methane ebullition (Figure 4c). Indeed, the ebullition (equation 7 and 8) results from the balance of soil temperature, pressure, gas content and porosity, which explain the large diversity of methane fluxes response by ebullition at each site. At DK-Nuf and PL-Wet simulated soil methane concentrations are less than 10 g CH₄ m⁻² therefore ebullition is not engendered. At FI-Lom, PL-Kpt and PL-Wet, the highest simulated methane production rates are maximum at 0.3 g CH₄ m⁻² d⁻¹ and are steadily near 20 cm at PL-Wet and about at 20 cm depth in summer and deepen down to 75 cm depth in winter for the two other sites. While at DK-Nuf the highest simulated methane production rates are lower with values up to 0.08 g CH₄ m⁻² d⁻¹ and take place around 20 cm in the summer and 40 cm in winter.
Figure 4: Temporal distribution of methane for sites emitting between 150 and 400 mg CH$_4$ m$^{-2}$ d$^{-1}$. (a) Simulated water table position estimated from the soil water content; (b) Simulated (dark line) and observed (gray line) methane emissions released to the atmosphere; (c) Cumulative amount of simulated methane emitted by diffusion, plant mediated transport and ebullition; (d) Methane concentration in the soil layers (dark line) and in the snow layers (gray line) of the model; (e) On the left, depth at which simulated methane production is the highest in the soil, scaled to the maximum peat depth. On the right, the amount of simulated methane produced at these depths.

The highest simulated methane fluxes of 600 mg CH$_4$ m$^{-2}$ d$^{-1}$ were observed at US-Wpt and RU-Che that are respectively a temperate marsh and an arctic tundra site. The simulated water table positions at both sites are lower in the summer than in the winter and vary for US-Wpt between 10 cm above ground and 40 cm below ground level. At RU-Che the prognostic water table depth is very low i.e. 60 to 90cm below the soil surface as for US-Bog. Indeed, both sites are underlaid with permafrost which limits water infiltration to the deepest soil layers and can explain these deeper simulated water table positions. At US-Wpt and RU-Che, site simulations could only provide methane fluxes up to 100 mg CH$_4$ m$^{-2}$ d$^{-1}$ despite the expansion of ranges for the optimization of the parameters. These simulated fluxes are entirely transported via vascular plant tissues. During the year of highest fluxes at both sites, simulated methane concentrations are around 0.2 g CH$_4$ m$^{-2}$ of soil however simulated methane concentrations in snow are 10 times lower at the marsh site, 0.3mg CH$_4$ m$^{-2}$ than at the tundra site, 3.0-4.0 mg CH$_4$ m$^{-2}$. At US-Wpt, simulations show that methane is primarily produced around 20 cm depth at a rate of 40-60 mg CH$_4$ m$^{-2}$ d$^{-1}$. Though, at RU-Che, simulated methane production rate is higher around 100 mg CH$_4$ m$^{-2}$ d$^{-1}$ and occurs at 20 cm depth during summer and few centimeters below the surface during winter. a suppressé: when snow covers the surface
Figure 5: Temporal distribution of methane for sites emitting more than 400 mg CH$_4$ m$^{-2}$ d$^{-1}$. (a) Simulated water table position estimated from the soil water content; (b) Simulated (dark line) and observed (gray line) methane emissions released to the atmosphere; (c) Cumulative amount of simulated methane emitted by diffusion, plant mediated transport and ebullition; (d) Methane concentration in the soil layers (dark line) and in the snow layers (gray line) of the model; (e) On the left, depth at which simulated methane production is the highest in the soil, scaled to the maximum peat depth. On the right, the amount of simulated methane produced at these depths.
3.2 Multi-site optimization (MSO)

For large scale simulations only one set of parameters is needed for the simulation of methane emissions to achieve that the average of each parameter value optimized on site are commonly employed. Here, a multi-site optimization has been performed for which prior values correspond to the average values of each parameter obtained from the single site optimizations described in section 3.1. This multi-site optimization serves to assess with which extent a multi-site optimization is more efficient than using average values of parameters optimized on site independently. Multi-site optimized parameters values acquired by using average values of parameters defined at each site and the initial ranges (Table 3) are shown in Table 6. Compared to the prior values, qMEG stayed about the same, optimized kMET shifted to values that promote lower oxidation of methane and near to the root area the proportion of methane oxidation MROX is increased. Plant mediated transport rate is stimulated by the increase of T_est to a value of 9 and the rooting depth is about the same, 0.27 for the prior and 0.26 for the posterior. Then the capability of methane ebullition in the model is decreased by the increase of the ebullition threshold deriving from mxFCH4 and the decrease of the probability of bubbles to reach the surface (wsize).

In Table 7, RMSDMS_prior constitutes the difference between observed and simulated emissions resulting from average single site optimized parameters values. RMSDMS_post is generated from the multi-site optimization of the parameters. For 8 sites, posterior values of the RMSDMS are smaller than prior values (RMSDMS_prior) thereby reducing the deviation of simulated emissions from the observation. The RMSDMS_post of the six other sites are larger than the RMSDMS_prior. Among those RMSDMS, posterior and prior values are very similar by less than one unit for FI-Lom and DK-Nuf. At DE-Hmm, PL-Wet and US-Bog the differences are lower than 16 units whereas at RU-Che RMSDMS_post is larger by more than 100 units than the RMSDMS_prior. NRMSDMS values are larger at US-Los, DE-Spw and DE-Sfn where methane emissions are lower. At the other sites, the difference of NRMSDMS and NRMSDMS are lower than 1.7 units. These results suggest that for global scale simulations parameters defined by the multi-site optimization should provide methane emissions estimation with lower uncertainties than when parameters are defined from the average of single site optimization values. Indeed, differences using single site and multi-site optimized parameters, displayed in Figure 6, are of the same order of magnitude for most of sites expect for the three sites that emitted the largest amount of methane, PL-Wet, RU-Che and US-Wpt and the lowest amount of methane, US-Los, DE-Spw and DE-Sfn. However, for those six sites methane emissions differences between observations and simulations is lower when using multi-site optimized parameters.

A multi-site optimization has also been performed employing extended ranges of parameter values that are enlarge to the maximum and minimum values obtained for the single site optimizations (Table S4 to S6 and Fig S9). Despite that a different set of parameters were defined (Table S3), discrepancies between observed and simulated emissions (Table S5 and S6 and Fig. S10) are similar than the ones obtained using default parameter ranges.
Table 6. Multi-site prior and optimized values of methane scheme parameters. Parameter prior values are the average value of the parameters optimized at each site. Parameter’s descriptions and references are in Table 3.

| Parameters | Unit | Prior values | Ranges | Posterior values | Uncertainty |
|------------|------|--------------|--------|-----------------|-------------|
| \(q_{MG}\)  | [-]  | 9.28         | 9.0, 11.0 | 9.64           | 0.8         |
| \(k_{MT}\)  | 1/d  | 2.59         | 1.0, 5.0  | 3.29           | 1.6         |
| \(M_{los}\) | fraction | 0.44 | 0.0, 1.0 | 0.70 | 0.4 |
| \(Z_{root}\) | m    | 0.27         | 0.01, 0.5 | 0.26           | 0.196       |
| \(T_{eq}\)  | [-]  | 6.99         | 0.0, 15.0 | 8.62           | 6.0         |
| \(wsize\)   | m    | 0.0088       | 0.001, 0.1 | 0.0018  | 0.396 |
| \(m_{CH4}\) | fraction | 0.24 | 0.05, 0.53 | 0.57 | 0.28 |

Table 7. Discrepancies between observed and simulated methane emissions are quantified by the root mean square difference (RMSD) approach. Minimization efficiency of the multisite optimization is indicated by the relationship between the prior using average values of parameters optimized by the single site optimization and posterior RMSD as \(1 - \frac{\text{RMSD}_{\text{post}}}{\text{RMSD}_{\text{prior}}}\)\(\times100\). Normalized root mean square difference (NRMSD) is defined by the RMSD posterior normalized by the annual mean of observed methane emissions of Table 5.

| Sites      | RMSD\(_{\text{prior}}\) | RMSD\(_{\text{posterior}}\) | \(1-\frac{\text{RMSD}_{\text{post}}}{\text{RMSD}_{\text{prior}}}\) | NRMSD |
|------------|------------------------|-----------------------------|-------------------------------------------------|-------|
| US-Los     | 56.1                   | 24.6                        | 0.56                                            | 224.00|
| DE-spw     | 855.9                  | 400.1                       | 0.53                                            | 800.20|
| DE-Sfn     | 325.8                  | 144.6                       | 0.56                                            | 37.08 |
| DE-Zrk     | 26.5                   | 6.6                         | 0.75                                            | 1.07  |
| CA-Wp1     | 91.7                   | 9.0                         | 0.90                                            | 1.01  |
| US-Bog     | 32.2                   | 43.9                        | -0.36                                           | 1.53  |
| FR-Lag     | 138.7                  | 67.6                        | 0.51                                            | 2.51  |
| DE-Hmm     | 31.8                   | 36.3                        | -0.14                                           | 1.71  |
| FI-Lom     | 52.2                   | 53.0                        | -0.01                                           | 2.10  |
| DK-NuF     | 43.9                   | 44.3                        | -0.01                                           | 0.84  |
| PL-Kpt     | 188.4                  | 78.0                        | 0.59                                            | 1.39  |
| PL-Wet     | 181.1                  | 197.4                       | -0.09                                           | 2.12  |
| US-Wpt     | 272.2                  | 159.4                       | 0.41                                            | 0.81  |
| RU-Che     | 159.0                  | 273.3                       | -0.72                                           | 3.40  |
Figure 6: Simulated and observed (gray line) methane emissions using single site (dashed dark line) and multi-site (solid dark line) optimized parameters.

4. Discussion

4.1 Parametrization’s sensitivity

Sensitivity analyses were previously performed to assess methane emission models responsiveness to parameters values (Meng et al., 2012; Riley et al., 2011; Spahni et al., 2011a; Wania et al., 2009; Zhu et al., 2014). These studies (Van Huissteden et al., 2009; Riley et al., 2011; Khvorostyanov et al., 2008b; Wania et al., 2009, 2010; Zhu et al., 2014; Morel et al., 2019) suggested that temperature dependency of methanogenesis is the most influential parameter affecting methane production whereas methane emissions are mostly sensitive to oxidation and plant transport. Indeed, in large scale models such as CLM4Me, LPJ-GUESS, LPX-Bern, CNRM and ORCHIDEE (Potter, 1997; Riley et al., 2011; Khvorostyanov et al., 2008b; Wania et al., 2009, 2010; Zhu et al., 2014; Morel et al., 2019) methane production result from anoxic decomposition of soil organic matter which rate is constrained by the soil oxic and anoxic decomposition ratio ($q_{ao}$). Therefore, methanogenesis rate is driven by the same variables as the oxic decomposition that depends on soil temperature and primary production. This ratio was first established from experimental studies that determine the microbial production ratio CO$_2$ to CH$_4$ (Potter et al., 1996; Segers, 1998) for various water table positions. These ratio values were
found to be between 0.58 and 10000. Because of this wide range of values, process-based models employed this CO\textsubscript{2} to CH\textsubscript{4} ratio as an adjustable parameter that is weighted by environmental factors such as soil moisture and temperature. Wania et al., (2009) performed a sensitivity analysis study of the LPJ-WHyMe model using 7 sites in which the multi-site optimization value of the CO\textsubscript{2}/CH\textsubscript{4} ratio was defined at 10 while other models as CLM4Me use a value of 5. Khvorostyanov et al., (2008a) and Morel et al., (2019) used respectively \( q_{MO} \) value of 9 and 10 to simulate methane emissions from arctic peatlands therefore in the present study at first \( q_{MO} \) were optimized in the range of 9-11 then this range was enlarged only for sites that underestimate methane emissions. Results show that for 13 sites out of 14, \( q_{MO} \) values ranging between 8.0 - 10.7 for the single site optimization approach and using multi-site approach a value of 9.6 were found. As in the previous sensitivity analysis studies (Riley et al., 2011) lower \( q_{MO} \) values were obtained at sites located in the highest latitudes.

After methanogenesis, methane is mobilized in pores and ultimately emitted to the atmosphere or being oxidized by methanotrophs depending on whether methane travels along the anoxic or the oxic parts of the soil. In large scale models, methanotrophy is formulated employing a Michaelis-Mentens or a first order kinetic framework based on soil methane and oxygen content (Morel et al., 2019). These formulations are then driven by the oxidation rate which values vary from few hours to days. In the present work, we employed the first order kinetic formulation of Khvorostyanov et al., (2008a) that is driven by methane and oxygen content. Optimization of the oxidation rate leads to values that are spread over the full range of 1 to 5 days. This is consistent with the review paper of Smith et al., (2003) that highlight that methanotrophy is more sensitive to soil moisture than soil temperature and that there is a direct link between methane oxidation rate and gas diffusivity. Thus, the optimization of the oxidation rate results from the balance between model inputs and outputs that are respectively available methane and oxygen substrates and methane fluxes which explain this large variability in oxidation rate. In addition, in our model, snow is considered in the diffusion scheme which is in part controlling diffusivity of oxygen from the atmosphere to the ground in winter (e.g. Figure 2c).

Methane emissions mediated by vascular plants result from series of processes that include (1) the diffusion and advective transport of methane and oxygen in aerenchym tissues (2) autotrophic respiration of a fraction of oxygen transiting in aerenchylma of vascular plants (Colmer, 2003; Nielsen et al., 2017) (3) methane production by microbial decomposition of plant exudates and (4) methane oxidation by exudates and by remaining oxygen at the root level brought through aerenchyma that increase methanotrophs activities. Modelling these processes requires (1) to understand and quantify them (Kaiser et al., 2017; Raivonen et al., 2017; Riley et al., 2011; Wania, 2007) and (2) to evaluate average density of vascular plants that are capable of significant gas transport across ecosystems. While a significant number of studies provide insight on gas exchanges through vascular plants, densities of vascular plants with aerenchyma in peatlands is poorly characterized.

In the most recent models, formulations of various complexity were used to simulate vegetation mediated gas transport considering mainly CH\textsubscript{4} and O\textsubscript{2} (Kaiser et al., 2017; Morel et al., 2019; Raivonen et al., 2017; Riley et al., 2011; Wania, 2007). These schemes considered plant transport at the scale of the plant and are based on gas concentration gradients between the atmosphere and the soil and some plant traits and properties such as plant height, root diameters, aerenchyma porosity and permeability. Because of the biodiversity of peatlands, calibration of parameters accounting for plant traits and
properties of each plant species or family is a cumbersome achievement and the lack of quantification of aerenchymatous plants at the scale of the ecosystem reduces the benefit in considering these characteristics. In the present scheme, vegetation transport of methane is simulated employing the rather simple scheme of Walter and Heimann (2001) that is driven by the rooting depth ($Z_{root}$) of vascular plants with aerenchyma and by the proportion of methane that is oxidized by the rhizosphere ($M_{ox}$). Optimized $Z_{root}$ values at sites ranges between 6 and 68 cm depth with the average depth defined at 26 cm which is also the value obtained using the multi-sites approach. These values are consistent with values utilized by Walter and Heimann (2001) that ranged between 0 and 74 cm. It could be expected for $Z_{root}$ to be set near the depth of maximum methanogenesis as it is the case at DE-Sfh where $Z_{root}$ is defined at 40 cm. Half of the sites have a $Z_{root}$ defined between 10 and 60 cm above the depth of maximum methanogenesis and the other remaining values are established between 10 and 50 cm below the depth of maximum methanogenesis. In the rhizosphere methane can also be oxidized at a rate ($M_{ox}$) that is independent of the rate of methanotrophy. Results of the optimization at site level provided $M_{ox}$ values that are scattered over the range of 0 to 1 with the highest values, 0.99, at site, US-Los that emitted the fewest methane and the lowest value, 0.003 at RU-Che that is the site emitted the largest amount of methane. Two trends can be distinguished, for sites that emitted less than 150 mg CH$_4$ m$^{-2}$ d$^{-1}$ an average of 60% of methane is oxidized by the rhizosphere against 22% at sites emitting more. Across all sites the average proportion of methane oxidized is 44% whereas the optimized value obtained with the multi-site approach is 70%. In previous models, Zhuang et al., (2004) and Wania et al., (2010) employed at the global scale a fixed value of 40 and 50% respectively. With a more realistic and complex formulation in CLM4Me, Riley et al. (2011) estimated that 60% of methane that would have been transferred to the atmosphere by aerenchyma tissues, is instead oxidized by the rhizosphere. $T_{reg}$ has been introduced by Walter et al., (1996) to describe the density of plants and their efficiency in methane transport for site estimation. It is an adjustable parameter that was scaled to be between 0 and 15 with lower values for ecosystems dominated by trees and shrubs and the highest values for ecosystems dominated by grasses and sedges. For our 14 sites, optimization at site established $T_{reg}$ values between 0.003 and 24 with an average value of 7 and an optimized value at 8.6 for the multi-site approach. Only two values have been defined above 10 at US-Wpt and DK-Nuf which are two sites that are limited in methane substrates in the model which explains these high values of $T_{reg}$.

When methane is significantly produced in the soil, the accumulation of methane in the water saturated pores involves the formation of methane rich bubbles that will migrate in the soil layers and eventually deliver methane to the atmosphere. This flux of methane is commonly prompted in land surface models by the amount of methane that is no longer soluble in saturated water-filled pores. This excess amount is defined here from the mixing ratio ($m_{CH}_4$) of methane in bubbles. Then this volumetric content of methane is converted to methane concentration per soil volume in each layer depending on soil temperature and pressure. The optimization of $m_{CH}_4$ at each site leads to values ranging between 3 and 53% with a mean value at 24% whereas the multi-site optimization evaluates $m_{CH}_4$ at 57%. It has been suggested in the literature that the methane partial pressure is sensitive to fluctuations of the hydrostatic and the atmospheric pressure (Tokida et al., 2007b) and of the water table position (Fechner-Levy and Hemond, 1996). Vegetation also impacts the ebullition flux by increasing substrate availability and by stabilizing indefinitely bubbles around roots (Klapstein et al., 2014). Migration of
methane rich-bubbles to the soil surface can be modelled as an instantaneous transport to the atmosphere or to upper layers depending on the connectivity between water filled pores (wsize). Khvorostyanov et al., (2008a) defined wsize at 1cm which establishes a probability of 1 at the surface that decreases to zero at 1.5m depth when soil is saturated. Probability is increasing when wsize increases and quickly decreases when soil moisture decreases. In the present study, at each site wsize is optimized to values of 0.05 - 3cm. At most sites, optimized wsize values are near or below 1cm except for US-Los, DK-Nuf and RU-Che. This might be explained by the low methane concentration in the model soil layers at these sites which annihilate possible emissions by ebullition in the model. The average value across sites corresponds to the same value determined by Khvorostyanov et al., (2008a) at 0.9 cm. A lower value is obtained for the multi-sites optimization of 0.2 cm which reduces occurrence of methane flux by ebullition in our model.

4.2 Methane sources

Soil and litter organic carbon and plant exudates are recognized to be the main substrates for methanogenesis (Chang et al., 2019; Riley et al., 2011; Whalen, 2005). Recent work of Hopple et al., (2019) demonstrates that dissolved organic carbon (DOC) also contributes significantly to anoxic decomposition in peatlands. Some field studies suggested that high latitude methanogenesis can be substrate limited (Chang et al., 2019; Riley et al., 2011; Whalen, 2005). In large scale models, soil organic carbon (SOC) is considered as primary source of methane however in order to increase the rate of methanogenesis labile organic matter such as litter carbon and plant exudates are directly combined to soil carbon bypassing oxic decomposition processes to account them as substrates for the methane production scheme (Morel et al. 2019; Khvonostianov et al; 2008). In the present study, SOC is the only substrate for methanogenesis for which total soil carbon stock and maximum peat depth has been adjusted to observation data at each site (Table 2). Simulation results show that at sites that emitted more than 400mg CH₄ m⁻² d⁻¹, US-WPT and RU-Che, methane emissions are lower than expected reflecting the lack of substrate for methanogenesis. Indeed, in land surface models, soil carbon is distributed in three types, the active, the slow and the passive pool. The active pool features labile SOC whereas the slow and the passive pools exert more stable SOC with slower decomposition rates. In Figures 2e to 5e that display the depth of maximum methane production, reveal that the deepest methane production depth is 0.75 meters in all the simulation results. Integrated SOC accumulated up to 0.75 m by our model for each site is reported in Table 8. These carbon stocks correspond to available substrate for methanogenesis occurring at lower depth than 0.75m depth. The lowest carbon stocks were obtained at US-Los, CA-Wp1, PL-Wet, US-Wpt and RU-Che with a total SOC lower than 50 kg/m². Unlike the other sites, the active SOC contents at US-Wpt and RU-Che are very small respectively 4 and 3.5 kg/m² which limit methane production in the model. At both sites, simulated vertical carbon contents were constrained using observed soil bulk density and the carbon accumulation model described in (Qiu et al., 2019). Khvorostyanov et al., (2008b) previously performed site simulation at RU-Che in which they prescribed an amount of 15gC m⁻² y⁻¹ of root exudates that was added to the active SOC leading to emissions up to 300 mg m² d⁻¹. As US-Wpt is a marsh it is expected to have a lower total SOC than the other peatland sites.
It is also expected that root exudates and DOC in pore water and in above ground reservoirs contribute significantly to methanogenesis which is not explicitly considered in the present version of the model.

### Table 8. Integrated simulated soil organic carbon content of peatlands sites up to 0.75 m depth.

| Sites identification | Soil organic carbon content |
|----------------------|----------------------------|
|                      | active | slow | passive | total |
|                      | kg/m² | kg/m² | kg/m² | kg/m² |
| US-Los               | 13.94 | 13.85 | 0.05  | 27.84 |
| DE-Spw               | 33.54 | 41.09 | 0.17  | 74.80 |
| DE-Sfn               | 28.15 | 49.40 | 0.28  | 77.83 |
| DE-Zrk               | 44.81 | 75.92 | 0.44  | 121.18|
| CA-Wp1               | 12.30 | 21.75 | 0.12  | 34.17 |
| US-Bog               | 14.16 | 66.55 | 0.69  | 81.40 |
| FR-Lag               | 33.67 | 52.02 | 0.25  | 85.94 |
| DE-Hmm               | 27.49 | 84.08 | 0.76  | 112.34|
| FI-Lom               | 13.95 | 63.89 | 0.85  | 78.69 |
| DK-NuF               | 4.18  | 49.20 | 1.18  | 54.56 |
| PL-Kpt               | 14.19 | 98.61 | 1.63  | 114.44|
| PL-Wet               | 15.36 | 22.08 | 0.11  | 37.55 |
| US-Wpt               | 3.94  | 0.84  | 0.001 | 4.78  |
| RU-Che               | 3.51  | 40.04 | 2.14  | 45.69 |

### 4.3 Methane fluxes

Sensitivity of methane fluxes to model parameters was evaluated by comparing annual methane emissions obtained by employing single site (SS) and multi-site (MS) optimized parameters. Table 9 reports annuals observed and simulated methane fluxes and the contributions among the three types of methane transport, i.e., diffusion, ebullition and plant mediated. Considering all 14 sites, average annual methane emissions for the observed values is $18 \pm 18$ g m$^{-2}$ y$^{-1}$, and $9 \pm 6$ and $25 \pm 38$ g m$^{-2}$ y$^{-1}$ for simulations using respectively SS and MS optimized parameters. Diffusion of methane in the topsoil layers of the model was minor compared to the other emissions and appeared to act as a sink of methane rather than a source.

Plant mediated transport (PMT) were the largest simulated fluxes, during the plant’s growth period. For SSO simulations these PMT fluxes represent between 52 and 74% of the total fluxes at US-Los, DE-Spw, DE-Sfn and PL-Kpt and more than 97% at all the other sites whereas for MSO simulations PMT fluxes are all higher than 98%. Given that diffusion released little amounts of methane to the atmosphere, remaining fluxes are emitted by ebullition. The largest ebullition fluxes were obtained in SSO simulations whereas fewer methane was released by ebullition in MSO simulations. For about half of sites, 3 - 11% of fluxes were furnished via ebullition and less than 1% at the other sites using SSO parameter values. In simulation employing MSO parameters values, ebullition contributed to less than 2% of the total fluxes at each site.
Discrepancies between the observation data and SSO and MSO simulations are displayed in Figure 6. At sites that emitted the largest amount of methane e.i. PL-Wet, RU-Che and US-Wpt, SSO and MSO simulations were underestimated up to 46 and 53 g CH$_4$ m$^{-2}$ y$^{-1}$, respectively (Fig. S6 to S8). At the other sites when using SSO parameters methane emissions were still underestimated even though this was only about 7 g CH$_4$ m$^{-2}$ y$^{-1}$. While in MSO simulations only three sites, DE-Hmm, FI-Lom and DK-Nuf underestimated methane emissions of 11 g CH$_4$ m$^{-2}$ y$^{-1}$ compared to observation data.

Simulations that display, in Figure 7, an overestimation of methane emissions were all performed using MSO parameters. At DE-Spw and DE-Sfn methane emissions were overestimated by 118 and 95 g CH$_4$ m$^{-2}$ y$^{-1}$. This large excess of methane emissions results from a significant increase of the parameters Tveg between the SSO and MSO. Indeed, optimized Tveg values at these sites are 0.003 and 0.1 when optimized at site level whereas it was defined at 8.6 with the multi-sites approach. In the model, Tveg established the magnitude of plant mediated fluxes which are constrained by soil methane content, plant growth and root expansion in the soil. This shows that for peatlands where methanogenesis is not subtract limited, Tveg is a key parameter to evaluate methane fluxes. Other sites that display an overestimation of methane emissions using MSO parameters are US-Los, CA-Wp1 and PL-Kpt. For these sites the excess of emissions compared to the observations only extend up to 12 g CH$_4$ m$^{-2}$ y$^{-1}$. Across sites, differences between observed emissions and simulated emissions employing SSO parameters averages around 9 g CH$_4$ m$^{-2}$ y$^{-1}$ of methane deficiency. On the contrary, emissions obtained with MSO parameters are in excess of about 5 g CH$_4$ m$^{-2}$ y$^{-1}$ on average compared to observations. Average differences between observation and simulation results significantly decrease to -1.2 and 0.5 g CH$_4$ m$^{-2}$ y$^{-1}$ for SSO and MSO simulations when excluding sites that emitted more than 300 and less than 20 mg CH$_4$ m$^{-2}$ d$^{-1}$, i.e. PL-Wet, RU-Che and US-Wpt for the SSO simulations and DE-Spw, DE-Sfn, PL-Wet, RU-Che and US-Wpt for the MSO simulations. This shows that the model is better constrained at sites emitting between 20 and 300 mg CH$_4$ m$^{-2}$ d$^{-1}$.

Average methane emissions estimated from these 14 sites can be utilized to roughly calculate emissions from peatlands located northern of 30°N. In Qiu et al. (2019), northern peatlands extent has been estimated using ORCHIDEEPEAT v2.0 and compare with three other peatlands inventories and soil data (Batjes, 2016; Joosten, 2009; Xu et al., 2018). All four estimates of northern peatlands areas range between 2823 and 3896 x 10$^6$ km$^2$. Assessment of methane emissions for these northern peatland areas estimated using the average fluxes from measurements yield annual methane fluxes of 51-71 Tg CH$_4$ y$^{-1}$ (Table 9). These annual fluxes are in good agreement with annual methane emissions determined from upscaling of flux measurements of 44-54 Tg CH$_4$ y$^{-1}$ by Zhu et al. (2013). Estimates of annual methane fluxes obtained from the SSO and MSO simulations lead to values of 25 - 35 and 70 - 96 Tg CH$_4$ y$^{-1}$, respectively. Estimates from SSO simulations are consistent with annual methane emissions reckoned from inversion models (Bruhwiler et al., 2014; Sphnhi et al., 2011b) and other process-based models (Chen et al., 2015; Peltola et al., 2019; Treat et al., n.d.; Zhang et al., 2016). While annual methane emissions assessed from MSO simulations are above the upper range of annual methane fluxes provided by the global methane budget for natural wetlands located northern of 30°N of 12-61 Tg CH$_4$ y$^{-1}$ for bottom-up approach and 31-64 Tg CH$_4$ y$^{-1}$ for top-down approach (Saunois et al. 2020).
Table 9. Yearly methane emissions defined from the observed data (Obs), from simulations employing optimized parameters obtained by the single site optimization (SSO) and by multi-site optimization (MSO). The methane fluxes combine methane emitted by diffusion, plant mediated transport and ebullition.

| Site       | Data | CH₄ fluxes | Diffusion | Plant mediated transport | Ebullition |
|------------|------|------------|-----------|--------------------------|------------|
|            | g m⁻² y⁻¹ | g m⁻² y⁻¹ | g m⁻² y⁻¹ | g m⁻² y⁻¹                 |            |
| US-Los     | Obs 0.03 | 0.01 | -0.01 | 0.01 | 0.0                      |
|            | SSO 6.70 | 0.031 | 6.71 | 0.0                        |
|            | MSO 0.46 | 0.07 | 0.34 | 0.02                       |
| DE-spw     | Obs 118.23 | -0.48 | 117.54 | 1.17                      |
|            | SSO 14.01 | 0.022 | 5.03 | 4.82                       |
|            | MSO 108.65 | -0.20 | 106.47 | 2.38                      |
| DE-Sfn     | Obs 5.60 | 0.0013 | 5.53 | 0.15                       |
|            | SSO 6.27 | -0.0013 | 6.27 | 0.01                       |
| US-Bog     | Obs 5.74 | 0.047 | 5.44 | 0.0                        |
|            | SSO 5.48 | 0.050 | 5.80 | 0.0                        |
| CA-Wp1     | Obs 3.29 | 0.012 | 3.12 | 0.19                       |
|            | SSO 15.63 | -0.10 | 15.72 | 0.0                        |
| FR-Lag     | Obs 9.91 | 0.0066 | 9.58 | 0.0                        |
|            | SSO 9.57 | 0.268 | 9.00 | 29.68                      |
| DE-Hmm     | Obs 12.19 | 0.0022 | 10.68 | 0.09                       |
|            | SSO 5.03 | 0.0022 | 4.97 | 0.06                       |
| FI-Lom     | Obs 21.15 | 0.0022 | 14.60 | 0.11                       |
|            | SSO 14.48 | 0.0040 | 9.54 | 0.0                        |
| DK-NuF     | Obs 8.69 | 0.05 | 9.21 | 0.26                       |
|            | SSO 0.54 | 0.01 | 0.54 | 0.0                        |
| PL-Kpt     | Obs 21.22 | 0.03 | 13.78 | 6.61                       |
|            | SSO 20.35 | 0.03 | 33.16 | 0.08                       |
| PL-Wet     | Obs 58.96 | 0.04 | 21.25 | 0.10                       |
|            | SSO 21.31 | 0.005 | 5.53 | 0.0                        |
| RU-che     | Obs 38.92 | 0.0001 | 8.46 | 0.0                        |
|            | SSO 8.46 | 0.0007 | 0.16 | 0.0                        |
| US-Wpt     | Obs 53.40 | 0.0 | 7.61 | 0.0                        |
|            | SSO 7.61 | 0.0 | 7.61 | 0.0                        |
|            | MSO 1.55 | 0.0 | 1.55 | 0.0                        |
5. Conclusion

The methane model developed by Khvorostyanov et al., (2008a) has been modified to encompass northern peatlands and permafrost features embedded in the most recent version of ORCHIDEE-PEAT v2.0. This modified version, ORCHIDEE-PCH4 which was used in this study, integrates a vertical discretization of oxic and anoxic decomposition of soil organic carbon of northern peatlands and subsequent methane production, oxidation and transport by vascular plants, ebullition and diffusion in soil and snow layers. A sensitivity analysis of methane emissions was performed on changes of 7 model parameters optimized with site-level measurements of 14 sites located northern than 41°N on the Eurasian and American continents. ORCHIDEE data assimilation system (Bastrikov et al., 2018) with a genetic algorithm for random search approach have been successfully employed to optimize these 7 parameters at each site and consider methane emissions from all sites simultaneously. Our results show that as in previous methane emissions models (Meng et al., 2012; Riley et al., 2011; Spahni et al., 2011a; Wania et al., 2009; Zhu et al., 2014), simulated methanogenesis is strongly correlated to simulated soil temperature and moisture content whereas methane emissions are more strongly correlated to plant mediated fluxes and to soils methane oxidation proportion. Surprisingly, a weak correlation has been established between the observed water table positions and the prognostic water table positions established from simulated soil moisture content. A correlation between soil moisture content and water table position in the field is needed to improve representation of the water table position in models.
Single site optimization results highlighted that the depth of the highest methane production fluctuates between 20 cm during warmer season and 75 cm during the cold season. This demonstrates the sensitivity of methanogenesis to soil temperature and provides insight on the extent to which methanogenesis takes place in the soil layers. This also serves in identifying sites that are substrate limited and to emphasize the need in global scale models to consider dissolved organic matter as a source of methane substrate. Indeed, in some site simulation studies prescribed methane substrate originating from litter decomposition or plant exudates were added to soil organic content in order to balance out the lack of labile substrate. In the scheme of ORCHIDEE-PCH4, the addition of methane diffusion in the snow layers during winter exposes the potential of snow to delay methane emissions coming from the soil.

Optimization of parameters employing simultaneously methane emissions from all 14 sites engender a reduction in the rate of methanotrophy and in methane transport in the soil by ebullition promoting methane oxidation at the root level and transport of methane by vascular plants. These involve a large overestimation of sites emitting small amounts of methane. Nonetheless, on average methane emissions simulated employing the multi-site optimization approach are only overestimated by about 5 g CH₄ m⁻² y⁻¹ because the overestimation of low emitting sites is counterbalanced by the high emitting sites that are limited in methane substrates. In contrast, average methane emissions obtained from the simulations using parameters from the single site optimization underestimate the average observed fluxes by 9 g CH₄ m⁻² y⁻¹. Nevertheless, extrapolation of these average methane emissions to northern peatland areas reveal that emissions estimated from the multi-site simulations are much larger than emissions estimated from other peatlands process-based models and inventories whereas emissions calculated from the single site optimizations are in good agreement with other estimates. This demonstrates the complexity of the interactions of the methane cycle with environmental conditions considered at various scales and the need for more detailed on-site studies.

**Code availability**

The source code (doi: 10.14768/d385219a-787a-439c-b128-2e2d30a21f87 ) is available online via https://forge.ipsl.jussieu.fr/orchidee/wiki/GroupActivities/CodeAvalaibilityPublication/ORCHIDEE_mict_peat_ch4. Readers interested in running the model should follow the guideline at http://orchidee.ipsl.fr/index.php/you-orchidee

The optimization tool is available through a dedicated web site for data assimilation with ORCHIDEE (https://orchidas.lsce.ipsl.fr).

**Author contributions**

ES revised and modified the implementation of the methane module in ORCHIDEE-PEAT, performed model optimization simulations in ORCHIDEE-PCH4 employing ORCHIDEE data assimilation system, investigated simulations results and prepared the manuscript with contributions from all co-authors. FJ, BG, CG, PC, SG, FL-D conceptualized, secured funding and supervised the project. CQ, DZ, BG provided ORCHIDEE-PEAT and assisted with model set up and development. LJ, FJ, BG assisted in implementing ORCHIDEE-PCH4 and investigating simulations results. VB, PP provided the model and
the expertise on the ORCHIDEE data assimilation system and contribute to the interpretation of simulation results. CG, SG, FL-D, MA, MSB-H, JC, BHC, HC, CWE, ESE, LBF, KF, DH, JK, OK, NK, AL, LM, WP, TS, KZ produced and provided quality field dataset employed to constrain and validate ORCHIDEE-PEAT and ORCHIDEE-PCH4.

890 **Acknowledgments**

The modelling work was supported by the European Union’s Horizon 2020, Project CRESmENDO under contract 641816 and Labex VOLTAIRE ANR-10-LABX-100-01. The authors acknowledge supports of staffs at each site. Research at US-Los was supported by the Ameriflux Network Management Project contract #7544821 to the ChEAS core site cluster. Funding for the measurements in Biebrza National Park was provided by the Polish National Science Centre under projects: UMO-2015/17/B/ST10/02187 and UMO-2020/37/B/ST10/01219. US-Bog was support by the United State National Science Foundation: NSF OPP 1107892, 1503912, 1936712. L.B. Flanagan acknowledges funding from the Natural Sciences and Engineering Research Council of Canada (NSERC), the FLUXNET-Canada Network (NSERC, the Canadian Foundation for Climate and Atmospheric Sciences (CFCAS), and BIOCAP Canada), and the Canadian Carbon Program (CFCAS). Field work at FR-Lag was funded as part of the Labex VOLTAIRE and the PIVOTS project of the Région Centre – Val de Loire (ARD 2020 program and CPER 2015–2020) in the framework of the French Peatland Observatory, SNO Tourbières, endorsed by CNRS-INSU. Research work at DE-Hmm was funded by the Deutsche Forschungsgemeinschaft under Germany’s Excellence Strategy – EXC 177 ’CliSAP - Integrated Climate System Analysis and Prediction’ – contributing to the Center for Earth System Research and Sustainability (CEN) of Universität Hamburg. The work at PL-Wet is based on use of Large Research Infrastructure CzeCOS supported by the Ministry of Education, Youth and Sports of CR within the CzeCOS program, grant number LM2018123. N. K. acknowledges the support by SustES – Adaptation strategies for sustainable ecosystem services and food security under adverse environmental conditions (CZ.02.1.01/0.0/0.0/16_019/0000797).

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