A Microbial Functional Group-Based CH4 Model Integrated Into a Terrestrial Ecosystem Model: Model Structure, Site-Level Evaluation, and Sensitivity Analysis

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Abstract Wetlands are one of the most important terrestrial ecosystems for land-atmosphere CH4 exchange. A new process-based, biophysical model to quantify CH4 emissions from natural wetlands was developed and integrated into a terrestrial ecosystem model (Integrated Biosphere Simulator). The new model represents a multisubstance system (CH4, O2, CO2, and H2) and describes CH4 production, oxidation, and three transport processes (diffusion, plant-mediated transport, and ebullition). The new model uses several critical microbial mechanisms to represent the interaction of anaerobic fermenters and homoacetogens, hydrogenotrophic, and acetoclastic methanogens, and methanotrophs in CH4 production and oxidation. We applied the model to 24 different wetlands globally to compare the simulated CH4 emissions to observations and conducted a sensitivity analysis. The results indicated that (1) for most sites, the model was able to capture the magnitude and variation of observed CH4 emissions under varying environmental conditions; (2) the parameters that regulate dissolved organic carbon and acetate production, and acetoclastic methanogenesis had the significant impact on simulated CH4 emissions; (3) the representation of the process components of CH4 cycling showed that CH4 oxidation was about half or more of CH4 production, and plant-mediated transport was the dominant pathway at most sites; and (4) the seasonality of simulated CH4 emissions can be controlled by soil temperature, water table position, or combinations thereof.

Plain Language Summary CH4 emission from wetlands is an important part of global carbon cycle. A new process-based model was developed to quantify the CH4 emission from wetlands. The new model considered main microbial mechanisms and transport processes in wetland CH4 cycling, and the modeled results matched the observed CH4 emissions well at evaluation sites globally. A sensitivity analysis indicated the important role of parameters that controlled dissolved organic carbon and acetate production and acetoclastic methanogenesis. The assessment of process components of CH4 cycling demonstrated the importance of CH4 oxidation and plant-mediated transport in wetland CH4 emission.

1. Introduction

CH4 is well known to be an important greenhouse gas that has about 28 times the global warming potential of CO2 over a 100-year scale (Myhre et al., 2013). The concentration of atmospheric CH4 has increased from 722 ± 25 ppb in 1750 to 1,803 ± 2 ppb in 2011, about 2.5 times, since preindustrial periods (Hartmann et al., 2013). Natural wetlands, including wet soils, swamps, fens, bogs, and peatlands, are the largest...
individual natural CH$_4$ emissions source (Bridgham et al., 2013; Saunois et al., 2016). The estimation of CH$_4$ emissions from natural wetlands ranges from 153 to 227 Tg CH$_4$ per year for the 2003–2012 period, which accounts for 30% (top-down inversions) and 25% (bottom-up estimation) on the average of total CH$_4$ emissions (Saunois et al., 2016), meaning natural wetlands play an important role in the global CH$_4$ budget and climate change.

CH$_4$ emission from natural wetlands is a net balance between microbial CH$_4$ production and consumption. There exists two major CH$_4$ production mechanisms, hydrogenotrophic methanogenesis and acetoclastic methanogenesis (Conrad, 1999; Krüger et al., 2001); two CH$_4$ oxidation mechanisms, aerobic and anaerobic methanotrophy (Gerard & Chanton, 1993; Smemo & Yavitt, 2011); and three aggregated CH$_4$ transport pathways, plant-mediated transport, diffusion, and ebullition (Chanton, 2005; Mer & Roger, 2001; Whiting & Chanton, 1996), all of which are influenced by the availability and quality of substrate, soil temperature and pH, hydrological conditions, and vegetation composition, and so forth (Walter & Heimann, 2000). In anaerobic conditions, organic polymers are utilized by fermenters plus H$_2$ producing acetogens to generate acetate, CO$_2$, and H$_2$, which become the substrates for hydrogenotrophic methanogens and acetoclastic methanogens and are ultimately transformed into CH$_4$. After CH$_4$ is produced, it can be oxidized by methanotrophs to form CO$_2$ or be transported via vascular plants, molecular diffusion, and bubbles (Riley et al., 2011).

Numerous process-based models have been developed to quantify CH$_4$ emissions from natural wetlands during the past decades, including WMEM (Cao et al., 1996), the Arah model (Arah & Stephen, 1998), ecosys (Grant, 1998, 1999), the Walter model (Walter & Heimann, 2000), Wetland-DNDC (Zhang et al., 2002), the Kettunen model (Kettunen, 2003), PEATLAND-VU (Van Huisteden et al., 2006), TEM-CH$_4$ (Tang et al., 2010; Zhuang et al., 2004), DLEM (Tian et al., 2010), ORCHIDEE (Ringleval et al., 2010, 2011), LPJ-WHYMe (Wania et al., 2010), CLM4Me (Riley et al., 2011), VISIT (Ito & Inatomi, 2012), TRIPLEX-GHG (Zhu et al., 2014), CLM-Microbe (Xu et al., 2015), JSBACH-methane (Kaiser et al., 2017), TECO_SPRUCE_ME (Ma et al., 2017), HIMMELI (Raivonen et al., 2017), and the soil methane scheme of ISBA (Morel et al., 2019). These models explicitly consider CH$_4$-related biogeochemical processes and their responses to environmental factors, and many of them simultaneously account for CH$_4$ production, oxidation, and three transport pathways. On the whole, these models share the following features: (1) the majority of models use a simple function directly associated with soil heterotrophic respiration, dissolved organic carbon (DOC), or environmental factors to estimate CH$_4$ production, lacking the representation of key microbial mechanisms; (2) the diffusion and ebullition processes are usually simulated as a threshold phenomenon whereby gas is released once the gas concentration exceeds a prescribed threshold concentration; and (3) many models have only been evaluated with a few in situ observations, without being compared against continuous CH$_4$ measurements collected over large spatial scales (only Riley et al., 2011 and Zhu et al., 2014 compared simulated and observed CH$_4$ emissions across different wetlands globally).

Using existing observations and studies, these models predict wetland CH$_4$ fluxes at ecosystem, regional and global scales. Yet recent studies have indicated that most previous models reproduced regional- to global-scale observations poorly (Bohn et al., 2015; Bohn & Lettenmaier, 2010). A recent model intercomparison project showed that the maximum estimate of global wetland CH$_4$ emissions is 264 Tg CH$_4$ per year, which is 1.89 times the minimum estimate of 141 Tg CH$_4$ per year (Melton et al., 2013). In addition, it is important to accurately quantify the response of global wetland CH$_4$ emissions to climate change, but Melton et al. (2013) found that the sign and magnitude of CH$_4$ emissions response to changes in temperature and precipitation vary among the models. Furthermore, only a few of the above models consider the interaction of different gases during the vertical migration of CH$_4$; for example, the Arah model (Arah & Stephen, 1998), CLM4Me (Riley et al., 2011), and JSBACH-methane (Kaiser et al., 2017) account for CH$_4$ and O$_2$ simultaneously; LPJ-WHYMe (Wania et al., 2010), HIMMELI (Raivonen et al., 2017), and the soil methane scheme of ISBA (Morel et al., 2019) take CH$_4$, O$_2$, and CO$_2$ into consideration by keeping track of their dynamics throughout the process; and TEM-CH$_4$ (Tang et al., 2010) incorporates N$_2$-related processes to make it a four-substance model that can resolve CH$_4$ biogeochemical cycling. Meanwhile, only three models incorporate the critical microbial mechanisms for CH$_4$ production and oxidation, including hydrogenotrophic methanogenesis, acetoclastic methanogenesis, and methanotrophy (e.g., Grant, 1998, 1999; Kettunen, 2003; Xu et al., 2015).
In this study, we developed a new process-based model to quantify CH4 emissions from natural wetlands, which considered the interaction of different gases, including CH4, O2, CO2, and H2, and incorporated the main microbial mechanisms related to CH4 production and oxidation. Moreover, the new process-based model has been integrated in a terrestrial ecosystem model (Integrated Biosphere Simulator [IBIS]), which can reflect interactions between soil temperature, hydrology, vegetation, and CH4 biogeochemical processes.

The aims of this study are to (a) introduce a new wetland CH4 emission model that includes the explicit description of the microbial mechanisms related to CH4 generation, (b) show the capability of this model to simulate CH4 emissions from natural wetlands by comparing with 24 site-level observations globally, and (c) assess the process components of wetland CH4 cycling and the control on the seasonality of modeled CH4 emissions.

2. Model Description

A new representation of natural wetland CH4 emissions within a terrestrial ecosystem model, the IBIS, was developed for this study. The IBIS is a process-based, comprehensive model that describes land surface processes, terrestrial carbon balance, and vegetation dynamics. The main processes represented in the IBIS model include land surface physics, canopy physiology, phenology, vegetation structure and competition, and carbon and nitrogen cycling in the terrestrial biosphere and have been integrated into a single, physically consistent, with different time scales, modeling framework (Foley et al., 1996; Kucharik et al., 2000; Liu et al., 2014; Yuan et al., 2014); here, we only describe the development of novel wetland CH4 dynamics. The new wetland CH4 emission model (Figure 1) is a multidimensional substance system that simulates the biogeochemical processes related to CH4, O2, CO2, and H2 and accounts for the transient, vertically resolved dynamics of these gases. The model introduces specific microbial mechanisms like anaerobic fermentation and homoacetogenesis, hydrogenotrophic methanogenesis, acetoclastic methanogenesis, and methanotrophy to account for CH4 production and oxidation. To calculate net CH4 emissions, the model also explicitly considers three different transport pathways including molecular diffusion, plant-mediated transport, and ebullition.

The transient reaction-diffusion equations governing the concentrations of CH4, O2, CO2, H2, and Ace (acetate) are:

\[
\frac{\partial}{\partial t} C_{\text{CH}_4}(z, t) = \frac{\partial}{\partial z} F_{\text{diff,CH}_4} - Q_{\text{plant,CH}_4} - Q_{\text{ebull,CH}_4} + R_{\text{prod,CH}_4} - R_{\text{oxid,CH}_4},
\]

\[
\frac{\partial}{\partial t} C_{\text{O}_2}(z, t) = \frac{\partial}{\partial z} F_{\text{diff,O}_2} - Q_{\text{plant,O}_2} - Q_{\text{ebull,O}_2} - R_{\text{aero}} - 2R_{\text{oxid,CH}_4}.
\]
\[
\frac{\partial}{\partial t} C_{\text{CO}_2}(z,t) = \frac{\partial}{\partial z} F_{\text{diff,CO}_2} - Q_{\text{plant,CO}_2} - Q_{\text{ebull,CO}_2} + R_{\text{prod,CO}_2} - R_{\text{cons,CO}_2},
\]

\[
\frac{\partial}{\partial t} C_{\text{H}_2}(z,t) = \frac{\partial}{\partial z} F_{\text{diff,H}_2} - Q_{\text{plant,H}_2} - Q_{\text{ebull,H}_2} + R_{\text{prod,H}_2} - R_{\text{cons,H}_2},
\]

\[
\frac{\partial}{\partial t} C_{\text{Ace}}(z,t) = R_{\text{prod,Ace}} - R_{\text{cons,Ace}},
\]

where \( C_X(z,t) \) is the concentration of compound \( X \) at soil layer depth \( z \) and time \( t \); \( F_{\text{diff,X}} \) is the diffusive flux of compound \( X \); \( Q_{\text{plant,X}} \) and \( Q_{\text{ebull,X}} \) are the transport rates of compound \( X \) via plant aerenchyma and ebullition, respectively; \( R_{\text{prod,X}} \) is the production rate of compound \( X \); \( R_{\text{oxid,CH}_4} \) is the \( \text{CH}_4 \) oxidation rate; \( R_{\text{cons,X}} \) is the consumption rate of compound \( X \); and \( R_{\text{aero}} \) represents the aerobic respiration rate.

Water table depth is an important factor that separates the whole soil column into aerobic and anaerobic zones. In this study, site-level water table depth observations were used. We assume that \( \text{CH}_4 \) is produced in the layers below the water table position and mainly consumed in the layers above the water table position (Walter & Heimann, 2000). The original IBIS included six soil layers with thicknesses of 0.1, 0.15, 0.25, 0.5, 1.0, and 2.0 m, with a total soil depth of 4 m (Foley et al., 1996). In order to represent water table depth specifically, we divided the soil column into nine layers, with the first five layers having a thickness of 0.1 m and the other four layers having thicknesses of 0.2, 0.3, 0.5, and 0.5 m, respectively. We only considered the \( \text{CH}_4 \)-related biogeochemical processes occurring in the nine soil layers. The detailed algorithms are described as follows.

### 2.1. \( \text{CH}_4 \) Production

#### 2.1.1. Available Carbon Decomposition

Methanogens prefer fresh organic matter as their substrate (Joabsson & Christensen, 2001), so the initial carbon source available for methanogens mainly comprises easily decomposed plant litterfall and root exudates. Given that DOC is a key intermediate for anaerobic fermentation and is converted from hydrolysis products and eventually decomposed into acetate, \( \text{CO}_2 \), and \( \text{H}_2 \), we define it to be the original, available carbon for methanogens and use a simple formula for its calculation:

\[
\text{DOC} = K_{\text{cpool}} \times \frac{\text{cpool}}{dz} \times f_T (\text{DOCprodQ}_{10}) \times f_{\text{moist}}.
\]

Here, DOC is the dissolved organic carbon concentration (mol m\(^{-3}\)), \( K_{\text{cpool}} \) represents the ratio of DOC to soil organic carbon, cpool is the soil organic carbon content (mol m\(^{-2}\)), \( dz \) is the layer thickness (m), DOCprodQ\(_{10}\) is the temperature sensitivity of DOC production, \( f_T \) and \( f_{\text{moist}} \) are soil temperature and moisture factors, respectively. We use the approach adopted by Wania et al. (2010) and Raivonen et al. (2017) to distribute the available carbon for methanogens to all soil layers according to the root fraction \( f_{\text{root}} \), which is calculated in the IBIS.

Under anaerobic conditions, the available carbon is fermented into acetate, \( \text{CO}_2 \), and \( \text{H}_2 \), which is governed by the following equation:

\[
\text{DOC} \rightarrow 0.67\text{Ace} + 0.33\text{CO}_2 + 0.11\text{H}_2.
\]

Thus, the fermentation is presented as:

\[
\text{DOCprodAce} = V_{\text{DOCprodAce,max}} \times \frac{\text{DOC}}{K_{\text{DOCprodAce}} + \text{DOC}} \times f_T (\text{AceprodQ}_{10}) \times f_{\text{PH}},
\]

\[
\text{DOCprodCO}_2 = 0.5 \times \text{DOCprodAce},
\]

\[
\text{DOCprodH}_2 = \frac{1}{6} \times \text{DOCprodAce}.
\]

Under aerobic conditions, the available carbon is decomposed into acetate and \( \text{CO}_2 \), which can be calculated as
\[
\text{DOCprodAce} = V_{\text{DOCprodAce,\,max}} \times \frac{\text{DOC}}{K_{\text{DOCprod Ace}} + \text{DOC}} \times \frac{[O_2]}{K_{\text{AceprodO2}} + [O_2]} \times f_T(AceprodQ_{10}) \times f_{\text{pH}},
\]

\[
\text{DOCprodCO}_2 = 0.5 \times \text{DOCprodAce}.
\]

Here, DOCprodAce, DOCprodCO2, and DOCprodH2 are the production rate (mol m\(^{-3}\) d\(^{-1}\)) of acetate, CO2, and H2, respectively. \(V_{\text{DOCprodAce,\,max}}\) is the maximum acetate production rate (mol m\(^{-3}\) d\(^{-1}\)), \([O_2]\) is the O2 concentration (mol m\(^{-3}\)), and \(K_{\text{DOCprod Ace}}\) and \(K_{\text{AceprodO2}}\) are the half saturation coefficients (mol m\(^{-3}\)) for DOC and O2, respectively. AceprodQ\(_{10}\) is the temperature sensitivity of acetate production, and \(f_{\text{pH}}\) is the soil pH factor.

### 2.1.2. Homoacetogenesis and Hydrogenotrophic Methanogenesis

The products CO\(_2\) and H\(_2\) are substrates for homoacetogens and hydrogenotrophic methanogens (Grant, 1998; Grant & Roulet, 2002). At low temperature, CO\(_2\) and H\(_2\) appear to be the main substrates for homoacetogens, which use CO\(_2\) as the electron acceptor to convert H\(_2\) to acetate (chemolithotrophic acetogenesis/homoacetogenesis) (Kotsyurbenko et al., 2001; Liu & Conrad, 2011; Schulz & Conrad, 1996). This process is governed by the following equation:

\[
4H_2 + 2CO_2 \rightarrow CH_3COOH + 2H_2O.
\]

Thus, acetate production from homoacetogenesis is calculated as

\[
H_2\text{prodAce} = V_{H_2\text{prodAce,\,max}} \times \text{Homoacetogens} \times \frac{[H_2]}{K_{H_2\text{prodAce}} + [H_2]} \times \frac{[CO_2]}{K_{CO_2\text{prod Ace}} + [CO_2]} \times f_T \times f_{\text{pH}}.
\]

Here, \(H_2\text{prodAce}\) is the acetate production rate from homoacetogenesis (mol m\(^{-3}\) d\(^{-1}\)), \(V_{H_2\text{prodAce,\,max}}\) is the maximum acetate production rate (mol m\(^{-3}\) d\(^{-1}\)), Homoacetogens is the microbial biomass of homoacetogens (mol m\(^{-3}\)), \([H_2]\) and \([CO_2]\) are the concentrations of H\(_2\) and CO\(_2\) (mol m\(^{-3}\)), \(K_{H_2\text{prodAce}}\) and \(K_{CO_2\text{prod Ace}}\) are the half saturation coefficients (mol m\(^{-3}\)) for H\(_2\) and CO\(_2\), respectively, and \(f_T\) represents the soil temperature factor.

In contrast, at high temperature, produced CO\(_2\) and H\(_2\) have usually been found to be suitable substrates for hydrogenotrophic methanogens (Hattroi, 2008), which use CO\(_2\) and H\(_2\) to generate CH\(_4\) (hydrogenotrophic methanogenesis). This process is governed by the following equation:

\[
4H_2 + CO_2 \rightarrow CH_4 + 2H_2O.
\]

Thus, CH\(_4\) production from hydrogenotrophic methanogenesis is calculated as

\[
H_2\text{prodCH}_4 = V_{H_2\text{prodCH}_4,\,max} \times H_2\text{methanogens} \times \frac{[H_2]}{K_{H_2\text{prodCH}_4} + [H_2]} \times \frac{[CO_2]}{K_{CO_2\text{prodCH}_4} + [CO_2]} \times f_T \times f_{\text{pH}}.
\]

Here, \(H_2\text{prodCH}_4\) is the CH\(_4\) production rate from hydrogenotrophic methanogenesis (mol m\(^{-3}\) d\(^{-1}\)), \(V_{H_2\text{prodCH}_4,\,max}\) is the maximum CH\(_4\) production rate (mol m\(^{-3}\) d\(^{-1}\)), \(H_2\text{methanogens}\) is the microbial biomass of hydrogenotrophic methanogens (mol m\(^{-3}\)), \(K_{H_2\text{prodCH}_4}\) and \(K_{CO_2\text{prodCH}_4}\) are the half saturation coefficients (mol m\(^{-3}\)) for H\(_2\) and CO\(_2\), respectively, and \(f_T\) represents the soil temperature factor.

### 2.1.3. Acetoclastic Methanogenesis

Product acetate is the substrate for acetoclastic methanogens (Grant, 1998; Grant & Roulet, 2002). CH\(_4\) production from acetoclastic methanogenesis is governed by the following equation:

\[
CH_3COOH \rightarrow CH_4 + CO_2.
\]

Thus, CH\(_4\) production is calculated as
\[
\text{AceprodCH}_4 = K_{\text{CH}_4\text{prod}} \times (1 - \text{Grow}_{\text{Acemethanogens}}) \times \text{Acecons}.
\]  

(18)

The acetate consumption is calculated as

\[
\text{Acecons} = V_{\text{Acecons, max}} \times \text{Acemethanogens} \times \frac{\text{Ace}}{K_{\text{AceprodCH}_4} + \text{Ace}} \times f_T(\text{CH}_4\text{prod}Q_{10}) \times f_{\text{pH}}.
\]  

(19)

Here, AceprodCH\(_4\) is the CH\(_4\) production rate from acetate consumption (mol m\(^{-3}\) d\(^{-1}\)), \(K_{\text{CH}_4\text{prod}}\) is the CH\(_4\) production ratio, Grow\(_{\text{Acemethanogens}}\) is the growth efficiency of acetoclastic methanogens, Acecons is the acetate consumption rate for respiration by acetoclastic methanogens (mol m\(^{-3}\) d\(^{-1}\)), \(V_{\text{Acecons, max}}\) is the maximum acetate consumption rate (mol m\(^{-3}\) d\(^{-1}\)), Acemethanogens is the microbial biomass of acetoclastic methanogens (mol m\(^{-3}\)), Ace is the acetate concentration (mol m\(^{-3}\)), \(K_{\text{AceprodCH}_4}\) is the half saturation coefficient (mol m\(^{-3}\)) for acetate, and CH\(_4\)prodQ\(_{10}\) is the temperature sensitivity of CH\(_4\) production during acetate consumption.

2.2. CH\(_4\) Oxidation

The CH\(_4\) produced by hydrogenotrophic methanogens and acetoclastic methanogens is the substrate for methanotrophs (Grant, 1999; Grant & Roulet, 2002). In this process, heterotrophic methanotrophs are the main microbial functional group (Riley et al., 2011). We use double Michaelis-Menten kinetics to represent CH\(_4\) oxidation, which is governed by the following equation:

\[
\text{CH}_4 + 2\text{O}_2 \rightarrow \text{CO}_2 + 2\text{H}_2\text{O}.
\]  

(20)

Thus, the \(R_{\text{oxid,CH}_4}\) (mol m\(^{-3}\) d\(^{-1}\)) is calculated as

\[
R_{\text{oxid,CH}_4} = V_{\text{CH}_4\text{oxid, max}} \times \text{Methanotrophs} \times \frac{[\text{CH}_4]}{K_{\text{CH}_4\text{oxidCH}_4} + [\text{CH}_4]} \times \frac{[\text{O}_2]}{K_{\text{CH}_4\text{oxidO}_2} + [\text{O}_2]} \times f_T(\text{CH}_4\text{oxid}Q_{10}) \times f_{\text{pH}}.
\]  

(21)

Here, \(V_{\text{CH}_4\text{oxid, max}}\) is the maximum CH\(_4\) oxidation rate (mol m\(^{-3}\) d\(^{-1}\)), Methanotrophs is the microbial biomass of methanotrophs (mol m\(^{-3}\)), \([\text{CH}_4]\) is the CH\(_4\) concentration (mol m\(^{-3}\)), \(K_{\text{CH}_4\text{oxidCH}_4}\) and \(K_{\text{CH}_4\text{oxidO}_2}\) are the half saturation coefficients (mol m\(^{-3}\)) for CH\(_4\) and O\(_2\), respectively, and CH\(_4\)oxidQ\(_{10}\) is the temperature sensitivity of CH\(_4\) oxidation.

O\(_2\) is not only used to oxidize CH\(_4\) but also consumed by aerobic respiration. We assume that part of the available carbon will be used by aerobic microbes, and this process requires O\(_2\), so refer to Kettunen (2003); the \(R_{\text{aero}}\) (mol m\(^{-3}\) d\(^{-1}\)) is calculated as

\[
R_{\text{aero}} = K_{\text{aer}} \times \frac{\text{DOC}}{K_{\text{aer,DOC}} + \text{DOC}} \times \frac{[\text{O}_2]}{K_{\text{aerO}_2} + [\text{O}_2]} \times f_T(\text{DOCprod}Q_{10}) \times f_{\text{pH}}.
\]  

(22)

Here, \(K_{\text{aer}}\) is the O\(_2\) consumption rate by aerobic respiration (mol m\(^{-3}\) d\(^{-1}\)), and \(K_{\text{aer,DOC}}\) and \(K_{\text{aerO}_2}\) are the half saturation coefficients (mol m\(^{-3}\)) for DOC and O\(_2\), respectively.

In the above processes, \(R_{\text{prod, X}}\) (mol m\(^{-3}\) d\(^{-1}\)) is the sum of produced compound X, and \(R_{\text{cons, X}}\) (mol m\(^{-3}\) d\(^{-1}\)) is the sum of consumed compound X.

2.3. CH\(_4\) Transportation

2.3.1. Diffusion

Molecular diffusive flux \(F_{\text{diff, X}}\) (mol m\(^{-2}\) d\(^{-1}\)) within the soil profile depends on the vertical concentration gradient and the diffusion coefficients of compound X. Fick's first law is applied to calculate the diffusive flux (Walter & Heimann, 2000).

\[
F_{\text{diff, X}} = D_X \frac{\partial C_X}{\partial z}.
\]  

(23)

Here, \(D_X\) is the effective diffusivity of compound X (m\(^2\) s\(^{-1}\)). We also use the constant reduction factors \(f_{\text{D, w}}\) and \(f_{\text{D, a}}\) to calculate the effective diffusivities in water or air (Raivonen et al., 2017), respectively.
Here, $D_{X,w}$ and $D_{X,a}$ represent the effective diffusivities of compound X in water and air ($m^2 s^{-1}$), and $D_{X}^{water}$ and $D_{X}^{air}$ are the free-water and free-air diffusivities ($m^2 s^{-1}$), respectively.

At the air-water interface, diffusivities can vary by at least four orders of magnitude. The method adopted by Wania et al. (2010) is used to calculate gas fluxes from the top soil layer into the atmosphere.

$$F_{diff,X} = -\phi_X \times (C_{surf,X} - C_{eq,X}).$$  \hspace{1cm} (26)

Here, $C_{surf,X}$ is the concentration of compound X in the top soil layer (mol m$^{-3}$), and $C_{eq,X}$ is the equilibrium concentration of compound X in the atmosphere (mol m$^{-3}$). $\phi_X$ represents the transfer velocity of compound X (cm hr$^{-1}$). We use a normalized transfer velocity $\phi_{600}$ (Cole & Caraco, 1998) to calculate the $\phi_X$.

$$\phi_X = \phi_{600} \times \left( \frac{S_{C_X}}{600} \right)^n.$$  \hspace{1cm} (27)

The $\phi_{600}$ (cm hr$^{-1}$) is calculated as

$$\phi_{600} = 2.07 + 0.215 \times U_{10}^{1.7}.$$  \hspace{1cm} (28)

Here, $S_{C_X}$ is the Schmidt number of compound X, 600 is the Schmidt number for CO$_2$ at 20 °C, $n = -0.5$, and $U_{10}$ is the wind speed at 10-m height (m s$^{-1}$). In this study, we also assume that wind speed can be ignored within the wetland vegetation so use a constant value of 0 for $U_{10}$. The unit of $\phi_X$ is centimeter per hour, which can be further transformed into meter per day.

For the concentration $C_{eq,X}$, which is in equilibrium with the gas partial pressure $P_{P_X}$ (Pa) and can be computed as:

$$C_{eq,X} = PP_X \times H_X.$$  \hspace{1cm} (29)

$H_X$ is the Henry's law constant for compound X (mol m$^{-3}$ Pa$^{-1}$). Based on the above equations, the diffusive flux for compound X can be obtained. The solution for diffusion within the soil column is obtained using the Crank-Nicholson scheme (Press et al., 1996).

2.3.2. Plant Transport

Many wetland vascular plants develop aerenchyma in response to the inundation environment. These tissues can act as conduits for the transport of CH$_4$, O$_2$, CO$_2$, and H$_2$ between the soil and atmosphere. Thus, plant-mediated transport is a diffusion process through the aerenchyma and driven by the specific gas concentration gradient (Riley et al., 2011). We refer to the approach adopted by Stephen et al. (1998) to calculate the plant transport rate $Q_{plant,X}$ (mol m$^{-3}$ d$^{-1}$).

$$Q_{plant,X} = \frac{D_{air,X}}{\tau} \times (\varepsilon(z) - \varepsilon_{z}).$$  \hspace{1cm} (30)

Here, $\varepsilon(z)$ is the density of cross-sectional area of root endings at depth $z$ (m$^2$ m$^{-3}$), and $\tau$ is the root tortuosity. The effective diffusivities in air are used as the diffusion coefficients inside roots for each gas. Similar to Raivonen et al. (2017), $\varepsilon(z)$ is formulated as

$$\varepsilon(z) = a_{mA} \times \left( \frac{f_{root}}{d\varepsilon} \right) \times \frac{LAI}{SLA}.$$  \hspace{1cm} (31)

Here, $a_{mA}$ is the cross-sectional area of root endings per root biomass (m$^2$ kg$^{-1}$), SLA represents the specific leaf area (m$^2$ kg$^{-1}$), and LAI is the leaf area index (m$^2$ m$^{-2}$), which is modeled in IBIS.
2.3.3. Ebullition

Ebullition is a relatively rapid process and only occurs in water-filled soil when the total partial pressure of dissolved gases exceeds the sum of atmospheric and hydrostatic pressure. Our implementation of ebullition follows that of Tang et al. (2010) and Raivonen et al. (2017), which is a new algorithm based on hydrostatic equilibrium rather than concentration threshold. The ebullition algorithm considers the concentrations of CH4, O2, CO2, H2, and N2, when the criterion for bubble formation is reached; such that when

$$\sum_X PP_X(z) > P_{atm} + P_{hyd}, \quad (32)$$

ebullition occurs. Here, $P_{atm}$ and $P_{hyd}$ are the atmospheric and hydrostatic pressure (Pa), respectively.

The fraction of ebullition $f_{ebull}(z)$ is calculated as

$$f_{ebull}(z) = \frac{\sum_X PP_X(z) - (P_{atm} + P_{hyd})}{\sum_X PP_X(z)}. \quad (33)$$

We also use the ebullition rate constant $k$ (d$^{-1}$) in the equation, so the ebullition rate $Q_{ebull,X}$ (mol m$^{-3}$ d$^{-1}$) of compound $X$ is calculated as

$$Q_{ebull,X} = k \times \frac{\sigma \times f_{ebull}(z) \times PP_X(z)}{RT}. \quad (34)$$

Here, $\sigma$ is the porosity, $R$ is the universal gas constant (J mol$^{-1}$ K$^{-1}$) and $T$ is the soil temperature (K).

The total ebullition flux released into either the atmosphere or soil is determined by water table depth. If the position of the water table is below the soil surface, the bubbles of gas are transported into the overlying air-filled soil layer and are subsequently diffused into the soil or plant aerenchyma. Otherwise, they are directly released into the atmosphere.

The detailed descriptions and baseline values for all parameters of the CH4 model are listed in Table 1. The algorithms used to describe the dynamics of four microbial groups, the coefficients associated with gas transport processes (including diffusion coefficients, Henry law constants, and Schmidt numbers), and the specific expressions for environmental factors are presented in Appendices A, B, and C, respectively.

3. Data and Methods

3.1. Observations

Continuous observations of CH4 emissions were compiled from 24 natural wetland sites, covering tropical, temperate, and boreal regions. Detailed information regarding these sites has been reported in previous studies or field work, so site information is briefly introduced in Tables 2–4, including location, wetland types, dominant vegetation, years of observation, and the measurement methods.

3.2. Model Forcing Data

The forcing data for this model mainly include daily climate data, daily water table depth, and soil carbon data. We used the Modern-Era Retrospective Analysis for Research and Applications data set (Gelaro et al., 2017) and the Climate Prediction Center Global Unified Precipitation data provided by the National Oceanic and Atmospheric Administration/Oceanic and Atmospheric Research/Earth System Research Laboratory Physical Sciences Division, Boulder, Colorado, United States (https://www.esrl.noaa.gov/psd/) to drive the model. The Modern-Era Retrospective Analysis for Research and Applications data provided the daily maximum, minimum, and average air temperature, relative humidity, and wind speed at 2 m, and the Climate Prediction Center Global Unified Precipitation data provided the daily precipitation. The soil carbon data for model initialization were obtained from the Global Gridded Surfaces of Selected Soil Characteristics (International Geosphere-Biosphere Programme Data and Information System) data set (Global Soil Data Task Group, 2000), and the soil pH was obtained from the soil properties data set of the Digital Soil Map of the World by Land and Water Development Division, Food and Agriculture Organization (http://www.fao.org/geonetwork/srv/en/metadata.show?id=14116). A spin-up of 200 years
To characterize the sensitivity of the modeled CH$_4$ emissions to input parameters (Table 1), the polynomial chaos expansion (PCE)-based Sobol sensitivity indices method was employed to assess the response of model output to changes in parameters. The Sobol indices (Sobol, 1993) are convenient representations of the model sensitivity to its parameters and correspond to the variance output to changes in parameters. The Sobol indices are traditionally computed by Monte Carlo simulation, which makes them difficult to apply with computationally expensive models (Marelli et al., 2019) and has been applied to some sensitivity studies of parameters in earth system model or land surface model (Ricciuto et al., 2018; Shi et al., 2019). The PCE-based Sobol indices method is based on the orthogonal stochastic polynomials in the random inputs to provide a functional approximation of the model output (Crestaux et al., 2009; Marelli et al., 2019). The PCE-based Sobol sensitivity indices method was employed to assess the response of model output to changes in parameters. The Sobol indices (Sobol, 1993) are convenient representations of the model sensitivity to its parameters and correspond to the variance-based decomposition that aims to decompose the total variance of the model into the sum of the variances of each input variable or their interactions (Ricciuto et al., 2018; Sudret, 2008). Instead, the PCE-based Sobol indices method requires less computational effort and is more applicable for the computationally expensive models (Marelli et al., 2019) and has been applied to some sensitivity studies of parameters in earth system model or land surface model (Ricciuto et al., 2018; Shi et al., 2019). The PCE-based Sobol indices method is briefly introduced as follows: For an input random vector $X = [X_1, ..., X_d]$, the PCE of model output $Y = M(X)$ can be established as

### Table 1: Major Parameters in CH$_4$ Related Processes

| Index | Parameter                         | Value | Range       | Units          | Descriptions                                                                 | References                           |
|-------|-----------------------------------|-------|-------------|----------------|------------------------------------------------------------------------------|--------------------------------------|
| P1    | $K_{c_{pool}}$                     | 0.02  | 0.0001–0.02 | mol m$^{-3}$   | Ratio of dissolved organic carbon to soil organic carbon                     | Grant (1998)                         |
| P2    | $V_{DOCprodAce,max}$              | 0.5   | 0.3–0.7     | mol m$^{-3}$   | Maximum acetate production rate from fermentation                            | Grant (1998)                         |
| P3    | $K_{DOCprodAce}$                  | 10.0  | 5–15        | mol m$^{-3}$   | Half saturation coefficient                                                     | Kettunen (2003)                      |
| P4    | $K_{AceprodO2}$                   | 0.04  | 0.01–0.1    | mol m$^{-3}$   | Half saturation coefficient                                                     | Kettunen (2003)                      |
| P5    | $V_{H2prodAce,max}$               | 0.15  | 0.01–0.3    | mol m$^{-3}$   | Maximum acetate production rate from homoacetogenesis                         | Grant (1998)                         |
| P6    | $V_{H2prodCH4,max}$               | 0.15  | 0.01–0.3    | mol m$^{-3}$   | Maximum CH$_4$ production rate from hydrogenotrophic methanogenesis           | Grant (1998)                         |
| P7    | $V_{Acecons,max}$                 | 0.5   | 0.3–0.7     | mol m$^{-3}$   | Maximum acetate consumption rate by aceticlastic methanogenesis               | Grant (1998)                         |
| P8    | $V_{CH4oxid,max}$                 | 0.5   | 0.3–0.7     | mol m$^{-3}$   | Maximum CH$_4$ oxidation rate                                                  | Grant (1999)                         |
| P9    | $K_{H2prodAce}$                   | 0.01  | 0.01–0.1    | mol m$^{-3}$   | Half saturation coefficient                                                     | Grant (1998)                         |
| P10   | $K_{CO2prodAce}$                  | 0.05  | 0.01–0.1    | mol m$^{-3}$   | Half saturation coefficient                                                     | Grant (1998)                         |
| P11   | $K_{H2prodCH4}$                   | 0.01  | 0.01–0.1    | mol m$^{-3}$   | Half saturation coefficient                                                     | Grant (1998)                         |
| P12   | $K_{CO2prodCH4}$                  | 0.05  | 0.01–0.1    | mol m$^{-3}$   | Half saturation coefficient                                                     | Grant (1998)                         |
| P13   | $K_{AceprodCH4}$                  | 0.05  | 0.01–0.1    | mol m$^{-3}$   | Half saturation coefficient                                                     | Grant (1998)                         |
| P14   | $K_{CH4prod}$                     | 0.5   | 0.3–0.7     | mol mol$^{-1}$ | CH$_4$ production ratio                                                        | Kettunen (2003)                      |
| P15   | $K_{CH4oxidCH4}$                  | 0.05  | 0.01–0.1    | mol m$^{-3}$   | Half saturation coefficient                                                     | Kettunen (2003)                      |
| P16   | $K_{CH4oxidO2}$                   | 0.02  | 0.01–0.1    | mol m$^{-3}$   | Half saturation coefficient                                                     | Kettunen (2003)                      |
| P17   | $f_{D,W}$                         | 0.8   | 0.7–0.9     |                   | Reduction factor for diffusion in water-filled peat                          | Raivonen et al. (2017)               |
| P18   | $f_{D,A}$                         | 0.8   | 0.7–0.9     |                   | Reduction factor for diffusion in air-filled peat                            | Raivonen et al. (2017)               |
| P19   | $\tau$                           | 1.5   | 1–2         |                   | Root tortuosity                                                               | Stephen et al. (1998)               |
| P20   | $a_{mA}$                          | 0.085 | 0.01–0.1    | m$^2$ kg$^{-1}$  | Root ending area per root dry biomass                                         | Stephen et al. (1998)               |
| P21   | SLA                              | 20    | 15–25       | m$^2$ kg$^{-1}$  | Specific leaf area                                                            | Raivonen et al. (2017)               |
| P22   | $\sigma$                         | 0.8   | 0.7–0.9     |                   | Peat porosity                                                                | Raivonen et al. (2017)               |
| P23   | $k$                              | 0.0001| 0.0001–0.0003| d$^{-1}$        | Time constant of ebullition                                                    |                                    |
| P24   | $K_{aer}$                         | 0.1   | 0.1–0.2     | mol m$^{-3}$    | O$_2$ consume rate from aerobic respiration                                   |                                    |
| P25   | $K_{aerDOC}$                      | 10.0  | 5–15        | mol m$^{-3}$    | Half saturation coefficient                                                     |                                    |
| P26   | $K_{aerO2}$                       | 0.22  | 0.01–0.3    | mol m$^{-3}$    | Half saturation coefficient                                                     |                                    |
| P27   | DOCprodQ10                        | 2.5   | 1–5         |                   | Temperature sensitivity of DOC production                                      | Kettunen (2003)                      |
| P28   | acetoprodQ10                      | 2.5   | 1–5         |                   | Temperature sensitivity of acetate production                                  | Kettunen (2003)                      |
| P29   | CH4prodQ10                        | 2.5   | 1–5         |                   | Temperature sensitivity of CH$_4$ production                                   | Kettunen (2003)                      |
| P30   | CH4oxidQ10                        | 2.5   | 1–5         |                   | Temperature sensitivity of CH$_4$ oxidation                                    | Kettunen (2003)                      |

(repeat the climate data of observed years) was done to obtain the modeled soil temperature and moisture, and the last cycle of observed years was used to simulate the CH$_4$ emissions. The final output of the model was the daily fluxes of gases between soil and atmosphere.

### 3.3. Sensitivity Analysis

To characterize the sensitivity of the modeled CH$_4$ emissions to input parameters (Table 1), the polynomial chaos expansion (PCE)-based Sobol sensitivity indices method was employed to assess the response of model output to changes in parameters. The Sobol indices (Sobol, 1993) are convenient representations of the model sensitivity to its parameters and correspond to the variance-based decomposition that aims to decompose the total variance of the model into the sum of the variances of each input variable or their interactions (Ricciuto et al., 2018; Sudret, 2008; Wang et al., 2016). The PCE is a powerful probabilistic technique that uses the orthogonal stochastic polynomials in the random inputs to provide a functional approximation of the model output (Crestaux et al., 2009; Marelli et al., 2019). The Sobol indices are traditionally computed by Monte Carlo simulation, which makes them difficult to apply with computationally expensive models (Sudret, 2008). Instead, the PCE-based Sobol indices method requires less computational effort and is more applicable for the computationally expensive models (Marelli et al., 2019) and has been applied to some sensitivity studies of parameters in earth system model or land surface model (Ricciuto et al., 2018; Shi et al., 2019). The PCE-based Sobol indices method is briefly introduced as follows: For an input random vector $X = [X_1, ..., X_d]$, the PCE of model output $Y = M(X)$ can be established as
| No. | Site name | Location | Wetland type | Dominant vegetation | Time | Method | References |
|-----|-----------|----------|--------------|---------------------|------|--------|------------|
| 1   | Zackenberg, Greenland | 74°30′N, 21°00′W | Arctic Fen | Eriophorum scheuchzeri, Carex stans, Dupontia psilosantha | 2006–2010 | Automatic chamber | Mastepanov et al. (2013) |
| 2   | Siikaneva, Finland | 61°50′N, 24°12′E | Boreal minerotrophic fen | Sedges, Rannoch rush and Mosses | 2005–2015 | Eddy covariance | Rinne et al. (2018) |
| 3   | Lompolojänkkä, Finland | 67°59.8′N, 24°12.5′E | Open, nutrient-rich sedge fen | Betula nana, Menyanthes trifoliata, Salix lapponum, and Carex ssp. | 2006–2010 | Eddy covariance | Raivonen et al. (2008) |
| 4   | Stordalen, Sweden | 68°21′N, 19°02′E | Subarctic mire | Eriophorum angustifolium | 2004–2006 | Automatic chamber and eddy covariance | Petrescu et al. (2008) and Granberg et al. (2001) |
| 5   | Degero Stormyr, Sweden | 64°11′N, 19°33′E | Mixed acid mire | Eriophorum vaginatum, Vaccinium oxycoccos, Andromeda polifolia, and Scheuchzeria palustris | 1995–1997 | Static chamber | Grenberg et al. (2001) |
| 6   | Lena Samoylov Delta, Russia | 72°22′N, 120°30′E | Tundra | Sedges, mosses, and shrubs | 2003–2004 | Eddy covariance | Wille et al. (2008) |
| 7   | Bakchar, Russia | 56°51′N, 82°50′E | Open unfrozen minerotrophic fen | Eriophorum vaginatum, Carex rostrata, and Equisetum fluviatile | 1999 | Static chamber | Frilberg et al. (2003) |
| 8   | Manitoba, Canada | 58°40′N, 93°50′W | Eutrophic fen | Sedges, grasses, and mosses | 2008–2011 | Eddy covariance | Hans et al. (2013) |
| 9   | Athabasca, Canada | 55°17′N, 112°28′W | Moderately rich tundra mire | Sedges, shrubs, and mosses | 2007 | Eddy covariance | Long et al. (2010) |
| 10  | Quebec, Canada | 53°30′N, 78°10′W | Boreal bog | Betula spp. and mosses | 2012 | Eddy covariance | Nadeau et al. (2013) |
| 11  | Robinsons, Canada | 48°16′N, 80°40′W | Boreal bog | Mosses, lichens, and shrubs | 2014–2016 | Eddy covariance | Wang et al. (2018) |
| No. | Site name          | Location                          | Wetland type                      | Dominant vegetation                                      | Time       | Method           | References       |
|-----|--------------------|-----------------------------------|-----------------------------------|----------------------------------------------------------|------------|------------------|------------------|
| 1   | Daxinganling, China | 51°7.92’N, 125°8.22’E             | Intermittently inundated marsh     | Carex meyerian (#1)¹                                  | 2011–2012  | Static chamber   | Liu et al. (2015) |
|     |                    | 52°56’N, 122°51’E                  | Permafrost peatland               | Shrubs, sedges, and grass (#2)                          | 2012–2013  | Eddy covariance  | Sun et al. (2018) |
| 2   | Sanjiang Plain, China | 47°53’N, 133°30’E              | Permanently inundated marsh        | Glyceria spiculosa (#1) and Deyeuxia angustifolia (#2)  | 2011       | Static chamber   | Sun et al. (2013) |
|     |                    |                                    |                                   | Carex lasiocarpa, Glyceria spiculosa, and Deyeuxia angustifolia (#3) |           | Eddy covariance  |                  |
| 3   | Luanhaizi, China   | 37°35’N, 101°20’E                 | Permanently inundated marsh        | Carex lasiocarpa and Carex pseuduraica (#4)            | 2012–2013  | Eddy covariance  |                  |
| 4   | Ruoergai, China    | 32°47’N, 102°32’E                 | Peatland                          | Carex lasiocarpa and Carex pseuduraica (#4)            | 2011       | Eddy covariance  |                  |
| 5   | Minnesota, US       | 47°32’N, 93°28’W                  | Poor fen                          | Sphagnum, Carex, and Schuchzeria palustris (#1)        | 2001–2010  | Eddy covariance  | Olson et al. (2013) |
| 6   | Sallies Fen, US     | 43°12.5’N, 71°3.5’W               | Poor fen                          | Sphagnum papillosum, Carex spp., Eriophorum chamissonis, and Sarcenaria purpurea (#2) | 2009–2011  | Eddy covariance  |                  |
| 7   | Michigan, US        | 42°27’N, 84°01’W                  | Ombrotrophic peatland             | Schuchzeria palustris                                  | 1991–1993  | Static chamber   | Shannon and White (1994) |
| 8   | Mer Bleue, Canada  | 45°41’N, 75°52’W                  | Temperate ombrotrophic bog         | Chamaedaphne (#1), Eriophorum (#2), and Maianthemum (#3) | 2009–2010  | TriPLICATE auto chamber | Lai et al. (2014) |
| 9   | Federsee, Germany   | 48°06’N, 9°38’E                   | Minerotrophic peatland            | Phragmites                                              | 2013       | Eddy covariance  | Brown et al. (2014) |
| 10  | Schechenflitz, Germany | 47°48’N, 11°19’E              | Temperate bog-pine forest          | Bog-pines and mosses                                   | 2012–2013  | Eddy covariance  | van den Berg et al. (2016) |
| 11  | Kopytkowo, Poland  | 53°35’N, 22°53’E                  | Temperate mire                    | Reeds, sedges, and rushes                              | 2013–2014  | Eddy covariance  | Hommeltenberg et al. (2014) |
| 12  | Kopuatai, New Zealand | 37°56’S, 175°22’E                | Ombrotrophic, raised bog           | Wire rush and Empodisma robustum                       | 2012–2014  | Eddy covariance  | Fortunia et al. (2017) |

¹The observed site with corresponding dominant vegetation.
Here, the $\Psi_\alpha(X)$ are multivariate polynomials orthonormal with regard to the distribution of $X$, $\alpha \in \mathbb{N}^k$ is an indices vector that identifies the components of the multivariate polynomials $\Psi_\alpha$, and the $\lambda_\alpha \in \mathbb{R}$ represent the corresponding coefficients.

The sum in equation 35 needs to be truncated to a finite sum, when applied in realistic situation, by the truncated PCE.

$$M(X) \approx M_{PC}(X) = \sum_{\alpha \in A} \lambda_\alpha \Psi_\alpha(X)$$

(36)

Here, $A \in \mathbb{N}^k$ is the set of selected multi-indices of multivariate polynomials.

Therefore, when the model $M(X)$ is approximated by the PCE surrogate, the Sobol indices can be computed by its coefficients $\lambda_\alpha$ as follows:

$$E(M(X)) \approx \lambda_0,$$

(37)

$$V(M(X)) \approx \sum_{\alpha \in A} \lambda_\alpha^2,$$

(38)

$$S_i = \frac{1}{V(M(X))} \sum_{\alpha \in A_{S_i}} \lambda_\alpha^2 \text{ with } A_{S_i} = \{\alpha: \alpha_i > 0, \alpha_k = 0 \text{ for } k \neq i\},$$

(39)

$$S_{Ii} = \frac{1}{V(M(X))} \sum_{\alpha \in A_{S_{Ii}}} \lambda_\alpha^2 \text{ with } A_{S_{Ii}} = \{\alpha: \alpha_i > 0\},$$

(40)

$$S_{ij} = \frac{1}{V(M(X))} \sum_{\alpha \in A_{S_{ij}}} \lambda_\alpha^2 \text{ with } A_{S_{ij}} = \{\alpha: \alpha_i > 0, \alpha_j > 0, \alpha_k = 0 \text{ for } k \neq i, j\}.$$  

(41)

Here, $E(M(X))$ and $V(M(X))$ indicate the expectation and variance of $M(X)$. $S_i$, $S_{Ii}$, and $S_{ij}$ are the first-order Sobol indices, the total Sobol indices, and the second-order Sobol indices (also called the joint Sobol indices), respectively. $A_{S_i}$, $A_{S_{Ii}}$, and $A_{S_{ij}}$ are the sets of indices vectors that only include the interest term corresponding to the sensitivity index.

The first-order Sobol indices measure the fraction of the variance contributed by the $i$th parameter only; the total Sobol indices measure the total variance contribution due to the $i$th parameter and its interactions with other parameters; and the second-order Sobol indices measure the fractional variance contribution corresponding to the joint $i$th and $j$th parameter (Ricciuto et al., 2018).

### 3.4. Model Evaluation

Three metrics were used to evaluate model performance, including:

1. The coefficient of determination ($R^2$), which represents the variation in the observations interpreted by the model.
2. Root mean square error (RMSE), calculated as
Here, \( M_i \) is the modeled value and \( O_i \) the observed value; \( n \) is the number of days for which we have the observed values.

3. Relative predictive error (RPE), computed as

\[
\text{RPE} = \frac{\overline{M} - \overline{O}}{\overline{O}} \times 100\%
\]  

(43)

Here, \( \overline{M} \) and \( \overline{O} \) are the means of modeled and observed values, respectively.

4. Results

4.1. Model Evaluation

Twenty-four sites were selected to evaluate the \( \text{CH}_4 \) model (Tables 2–4). These evaluation sites spanned from boreal to tropical regions and covered several dominant wetland types including fen, bog, mire, marsh, peatland swamp, and tundra. We examined model performance by assessing its ability to accurately reproduce the magnitude and temporal variability of \( \text{CH}_4 \) emissions among these sites.

4.1.1. Boreal Sites

Observations from 11 sites in boreal regions, mainly located in northern Europe and Canada, were compiled for model evaluation, comprising six fen sites, two bog sites, two mire sites, and one tundra site. Overall, the model reliably predicted the observations (Figure 2), with the RPE varying from \(-19.47\%\) to \(33.28\%\) (Table 5).
Table 5  
Statistical Results for Site-Level Evaluation

| Site                  | $R^2$       | RMSE $^a$  | RPE (%) | $N$  |
|-----------------------|-------------|------------|---------|------|
| Boreal sites          |             |            |         |      |
| Zackenberg            | 0.210       | 0.0019     | -1.39   | 192  |
| Siikaneva             | 0.517       | 0.0017     | 14.54   | 2285 |
| Lompolojänkkä        | 0.815       | 0.0018     | 2.80    | 837  |
| Stordalen             | 0.317       | 0.0057     | 17.38   | 126  |
| Degero Stormyr        | 0.361       | 0.0028     | -19.47  | 23   |
| Lena Samoylov Delta   | 0.053       | 0.0007     | 15.95   | 79   |
| Bakchar               | 0.586       | 0.0028     | -16.55  | 35   |
| Manitoba              | 0.012       | 0.0030     | 30.08   | 127  |
| Athabasca             | 0.563       | 0.0010     | 4.38    | 103  |
| Quebec                | 0.455       | 0.0007     | 10.76   | 61   |
| Robinsons             | 0.313       | 0.0006     | 33.28   | 329  |
| Temperate sites       |             |            |         |      |
| Daxinganling1         | 0.297       | 0.0005     | 12.31   | 38   |
| Daxinganling2         | 0.272       | 0.0002     | 15.38   | 2425 |
| Sanjiang1             | 0.258       | 0.0030     | 13.46   | 54   |
| Sanjiang2             | 0.000       | 0.0025     | -2.79   | 57   |
| Sanjiang3             | 0.379       | 0.0032     | -6.27   | 119  |
| Sanjiang4             | 0.413       | 0.0027     | 7.04    | 142  |
| Luanhaizi             | 0.170       | 0.0025     | -26.70  | 133  |
| Ruoergai1             | 0.118       | 0.0029     | 20.29   | 35   |
| Ruoergai2             | 0.059       | 0.0039     | -19.26  | 35   |
| Minnesota1            | 0.689       | 0.0057     | -9.13   | 54   |
| Minnesota2            | 0.506       | 0.0033     | 4.22    | 644  |
| Sallies Fen           | 0.240       | 0.0052     | -28.70  | 80   |
| Michigan              | 0.142       | 0.0147     | -27.30  | 62   |
| Mer Bleue1            | 0.083       | 0.0018     | -0.19   | 194  |
| Mer Bleue2            | 0.498       | 0.0034     | -13.63  | 208  |
| Mer Bleue3            | 0.231       | 0.0028     | -28.99  | 185  |
| Mer Bleue4            | 0.000       | 0.0012     | -22.68  | 276  |
| Federeemoor           | 0.252       | 0.0043     | -3.55   | 253  |
| Schechenfliz          | 0.231       | 0.0012     | 32.64   | 145  |
| Kopytkowo             | 0.141       | 0.0060     | -48.62  | 239  |
| Kopuatai              | 0.001       | 0.0041     | -4.97   | 124  |
| Tropical sites        |             |            |         |      |
| Sarawak1              | 0.016       | 0.0003     | 47.04   | 33   |
| Sarawak2              | 0.003       | 0.0010     | 25.08   | 61   |
| Sarawak3              | 0.426       | 0.0005     | 6.25    | 18   |

$^a$The unit of RMSE is mol m$^{-2}$ d$^{-1}$.

The magnitude and seasonality of simulated CH$_4$ emissions were consistent with the observations, especially at the Siikaneva and Lompolojänkkä sites (Figures 2b and 2c). Long-term continuous CH$_4$ observations had been collected at these two sites, and the values of the coefficient of determination ($R^2$) were about 0.52 and 0.82, respectively (Table 5).

Although the model explained variability in CH$_4$ emissions well at most sites, large differences between simulated and observed CH$_4$ emissions were still observed to exist. The model underestimated CH$_4$ observations at the Zackenberg site; the discrepancies between modeled and observed values mainly occurred in the growing seasons of 2006 and 2007 (Figure 2a). Meanwhile, the model had slight high predicted values in the growing season of 2004 at Lena Samoylov Delta (Figure 2f). At these two sites, $R^2$ was very low with values of 0.21 and 0.053, respectively (Table 5). In addition, the model predicted slightly lower values for peak emissions at the Bakchar, Athabasca, and Robinsons sites (Figures 2g, 2i, and 2k). The lowest $R^2$ was produced at the Manitoba site, which simultaneously had a high RPE value ($R^2 = 0.012$, RPE = 30.08%) (Table 5), indicating that the model failed to capture the variation and magnitude of CH$_4$ observations at this site.

4.1.2. Temperate Sites

CH$_4$ flux observations from 12 sites in temperate regions were compiled for model evaluation. On the whole, the model predictions matched the observations well (Figure 3). At the Minnesota and Mer Bleue sites (Figures 3e1, 3e2, and 3h2), the model explained about 69%, 51%, and 50% of the variation in CH$_4$ emissions, respectively (Table 5). The RPE ranged from -28.99% to 32.64%, except at the Kopytkowo site, demonstrating that a lower deviation between simulated and observed CH$_4$ emissions existed at most sites (Table 5).

However, large differences also existed between modeled and observed CH$_4$ emissions at some sites. At the Sanjiang sites, the simulated peak emissions clearly lagged the observed peak values (Figures 3b1–3b3). Although they had lower RPEs, with values of 13.46%, -2.79%, and -6.27%, respectively (Table 5), the model predicted little of the variability in observed CH$_4$ emissions, with $R^2$ being 0.258, 0.000, and 0.379, respectively (Table 5). At the Ruoergai and Federeemoor sites, observed CH$_4$ fluxes had no general seasonal patterns (Figures 3d1, 3d2, and 3i). The $R^2$ values were 0.118, 0.059, and 0.252, and the RMSE values were 0.0029, 0.0039, and 0.0043 mol m$^{-2}$ d$^{-1}$, respectively (Table 5). Thus, the simulated results only matched the magnitude of observed CH$_4$ emissions at these two sites. The model underestimated observed CH$_4$ emissions at the Daxinganling, Minnesota, Michigan and Mer Bleue sites (Figures 3a2, 3e2, 3g, and 3h4), which mainly occurred in the 2013 growing season at Daxinganling, 2011 at Minnesota, 1991 at Michigan, and 2011 at Mer Bleue. Meanwhile, the model slightly overestimated CH$_4$ emissions at Daxinganling and Mer Bleue in 2012 (Figures 3a2 and 3h4). In addition, the model showed poor agreement between simulated and observed CH$_4$ emissions at the Luanhaizi, Kopytkowo, and Kopuatai sites (Figures 3c, 3k, and 3l). The calculated $R^2$ and RPE values at these sites were 0.17 and -26.70%, 0.145 and -48.62%, and 0.001 and -4.97%, respectively (Table 5). At the Kopytkowo site, the model significantly underestimated observations during the growing season in 2013 (Figure 3k).

4.1.3. Tropical Sites

Observations from a tropical peat swamp forest located in Sarawak, Malaysia were also collected for comparison. Overall, the model showed good agreement between simulated CH$_4$ emissions and observations at this site (Figure 4). At the Sarawak1 site, the simulated CH$_4$ emissions did not reproduce the variation in observations, which had no clear seasonal patterns (Figure 4a1). In addition, the model slightly overestimated the
observed CH$_4$ emissions in November at the Sarawak2 site (Figure 4a2). The $R^2$ and RPE values for these two sites were 0.016 and 47.04% and 0.003 and 25.08%, respectively (Table 5). The model captured the magnitude and variation of CH$_4$ emissions during the measured period at the Sarawak3 site (Figure 4a3). The $R^2$ and RPE values for this site were 0.426 and 6.25%, respectively (Table 5). The RMSE values for this peat

Figure 3. Daily variation in simulated and observed CH$_4$ emissions at temperate sites. The blue solid lines represent simulated values, and the red cross dots represent observed values.
swamp forest site ranged from 0.0003 to 0.001 mol m$^{-2}$ d$^{-1}$ (Table 5), indicating a small disparity between simulated and observed CH$_4$ emissions.

The average simulated and observed CH$_4$ emissions during measured periods for all evaluated, boreal and temperate sites are shown in Figure 5. On the whole, the average values of simulations and observations at all evaluated sites lie close to the 1:1 line over a range of 0 to 0.017 mol m$^{-2}$ d$^{-1}$, and $R^2$ is 0.87 (Figure 5a).

### 4.2. Sensitivity Analysis

The PCE-based parameter sensitivity analysis of modeled CH$_4$ emissions with 500 model evaluations was performed at seven measured sites, covering the tropical, temperate, and boreal biomes (Figures 6 and 7). For the Siikaneva, Lompolojänkkä, and Quebec sites, CH$_4$prod Q10 (P29) is the most sensitive parameter for CH$_4$ emissions (Figure 6a), which is also reflected in the evaluation of total Sobol indices (Figure 6b).

![Figure 4](image1)  
**Figure 4.** Daily variation in simulated and observed CH$_4$ emissions at a tropical site. The blue solid lines represent simulated values, and the red cross dots represent observed values.

![Figure 5](image2)  
**Figure 5.** Comparison of the average observed and simulated CH$_4$ emissions (red dots). (a) All evaluated sites, (b) boreal sites, and (c) temperate sites.
For the Sallies and Kopuatai sites, $K_{\text{pool}}$ (P1) has the largest impact on CH$_4$ emissions (Figures 6a and 6b). In addition, the CH$_4$ emissions are also sensitive to $V_{\text{Acecons, max}}$ (P7), $K_{\text{AceprodCH}_4}$ (P13), and DOC$_{\text{prodQ10}}$ (P27) at the Sallies site and sensitive to $K_{\text{CH}_4\text{prod}}$ (P14) and CH$_4$$_{\text{prodQ10}}$ at the Kopuatai site (Figures 6a and 6b). For the Schechenfilz site, the first-order Sobol indices are low for all parameters (Figure 6a). However, the DOC$_{\text{prodQ10}}$ has a great total effect index of about 58% on the total variance of CH$_4$ emissions (Figure 6b), which indicates the apparent impact of this parameter on the CH$_4$ emissions. For the Sarawak3 site, except for $V_{\text{Acecons, max}}$ and $K_{\text{CH}_4\text{prod}}$, the first-order and total Sobol indices have low values for other parameters (Figures 6a and 6b). CH$_4$ emissions at this site are obviously sensitive to these two parameters, with $V_{\text{Acecons, max}}$ becoming the most sensitive parameter (Figures 6a and 6b). Moreover, compared to other sites, the Sobol indices of parameters related to temperature sensitivity (P27–P30) are very low at the Sarawak3 site, which may be related to the high and relatively constant soil temperature in the tropics.

The second-order Sobol indices results indicate that $K_{\text{AceprodCH}_4}$ and CH$_4$$_{\text{prodQ10}}$ have the largest interaction effects on CH$_4$ emissions at the Siikaneva, Lompolojäntkkä, Quebec, Schechenfilz, and Kopuatai sites (Figure 7). The parameters $V_{\text{Acecons, max}}$ and $K_{\text{AceprodCH}_4}$ have the largest interaction effects on CH$_4$ emissions at the Sallies site, and $V_{\text{Acecons, max}}$ and $K_{\text{CH}_4\text{prod}}$ have the largest interaction effects on CH$_4$ emissions at the Sarawak3 site (Figure 7). In addition, $V_{\text{Acecons, max}}$ and CH$_4$$_{\text{prodQ10}}$ have the important interaction effects on CH$_4$ emissions at the Siikaneva, Lompolojäntkkä, and Quebec sites (Figure 7). For the Lompolojäntkkä site, the parameter subset of AceprodQ$_{10}$ and CH$_4$$_{\text{prodQ10}}$ also has the important interaction effects on CH$_4$ emissions (Figure 7). The parameter set of $K_{\text{pool}}$ and $V_{\text{Acecons, max}}$ and the parameter set of $V_{\text{Acecons, max}}$ and AceprodQ$_{10}$ also have the important interaction effects on CH$_4$ emissions at the Sallies site (Figure 7).
Although the results of first-order, total, and second-order Sobol indices among the parameters are somewhat different between these sites, the main sensitivity parameters are DOCprodQ10, AceprodQ10, CH4prodQ10, Kcpool, VAcecons,max, KAceprodCH4, and KCH4prod, which indicates that these parameters that control DOC and acetate production and acetoclastic methanogenesis have the significant impact on modeled CH4 emissions.

4.3. Environmental Controls on CH4 Emissions

At the above test sites, inconsistent relationships between simulated CH4 emissions, soil temperature, and measured water table position were observed (Figure S1). For example, the seasonality of simulated CH4 emissions at the Siikaneva and Lompolojänkkä sites depended on soil temperature dynamics and had no significant correlation with water table position. However, at the Sarawak3 site, the temporal variation of simulated CH4 emissions coincided well with water table position but showed no dependence on soil temperature. In addition, combined effects of soil temperature and water table position were observed on simulated CH4 emissions at other sites. The soil temperature and water table position exerted a positive effect on simulated CH4 emissions during the growing season at the Quebec site, where peak emissions

Figure 7. The second-order Sobol indices for modeled CH4 emissions. All parameters are listed in Table 1.
occurred when soil temperature and water table position reached their maximum. However, although soil temperature had a positive effect at the other sites, an increase of water table position had little influence on simulated CH₄ emissions, and a decrease substantially reduced CH₄ emissions, especially during the growing season.

5. Discussion

5.1. Modeled Process Components

The new CH₄ model explicitly represented CH₄-related microbial mechanisms, including anaerobic fermentation, homoacetogenesis, hydrogenotrophic methanogenesis, acetoclastic methanogenesis, and methanotrophy, and the interaction of different gases, including CH₄, O₂, CO₂, and H₂, in wetland CH₄ emission modeling. The new model has also been integrated into a terrestrial ecosystem model (IBIS) and evaluated at 24 different wetland sites globally. The simulated results captured the magnitude and variation of observed CH₄ emissions at most sites.

Net CH₄ emission is determined by the CH₄ production by methanogens, CH₄ oxidation by methanotrophs, and molecular diffusion, plant-mediated transport, and ebullition to the atmosphere (Chanton, 2005). To assess these processes represented by the model, we calculated the temporal variation in process components of wetland CH₄ emissions at the test sites (Figure 8) and the proportion that each process relative to CH₄ production at all evaluated sites (Figure 9).

Methanotrophy is an important process and can be up to 100% of CH₄ production (Fritz et al., 2011; Whalen, 2005). An almost completely CH₄ oxidation in a densely rooted bog has been reported recently from an analysis of isotopic δ¹³C-CH₄ values (e et al., 2019). The CH₄ oxidation of global wetland has also been estimated to be 40–70% of total CH₄ production (Megonigal et al., 2004). In our results, CH₄ production and oxidation showed apparent temporal variation at all test sites, with peak values of both processes occurring in the midgrowing seasons (Figure 8). This may reflect enhanced microbial activities caused by the high
temperature in this period (Dunfield et al., 1993). In addition, at all evaluated sites, CH$_4$ oxidation was observed to be about half or more of CH$_4$ production (Figure 9).

Plant-mediated transport has been reported as the principal pathway for CH$_4$ emissions (Colmer, 2003; Green & Baird, 2012; King et al., 1998; Waddington et al., 1996; Whiting & Chanton, 1992). In our analysis, plant-mediated transport also showed clear temporal variation and played a primary role in total observed period, followed by molecular diffusion, and ebullition had the smallest contribution and being characterized by pulse behavior (Figure 8). Moreover, plant-mediated transport also had the greatest proportion of total CH$_4$ emission at most sites (Figure 9).

5.2. Environmental Controls on CH$_4$ Emissions

Numerous studies have identified soil temperature as the fundamental control on wetland CH$_4$ emissions (Liu et al., 2015; Rinne et al., 2018; Sun et al., 2018), with an exponential dependence of CH$_4$ emissions on soil temperature being recorded (Marushchak et al., 2016; Mikhaylov et al., 2015). Water table position is also generally recognized as a major physical control on CH$_4$ emissions from wetlands (Moore et al., 2011; Wong et al., 2018). The relationship between water table position and CH$_4$ emissions is usually nonmonotonic (Brown et al., 2014; Christensen et al., 2003). However, several studies have reported no significant dependence of CH$_4$ emissions on water table position (Jackowicz-Korczyński et al., 2010; Liu et al., 2015; Rinne et al., 2018). In addition, combined effects of soil temperature and water table position are observed on CH$_4$ emissions from wetlands (Goodrich et al., 2015; Lai et al., 2014; Noyce et al., 2014).

This study analyzed the relationships between modeled CH$_4$ emissions and soil temperature and observed water table position at seven test sites. The different control patterns of these two variables on observed CH$_4$ emissions are reflected in our simulations (see section 3.3). At the Lompolojänkkä site, the water table position is almost always above the soil surface (Figure S1); this may maintain a relatively stable environment for wetland CH$_4$ dynamics. Thus, the water table position had little or no effect on CH$_4$ emissions, which was also observed in continuously inundated ecosystems (Strachanm et al., 2015; Sturtevant et al., 2016; Sun et al., 2013). The seasonal variation of simulated CH$_4$ emissions at the Silkaneva site was controlled by soil temperature (Figure S1), which was consistent with the analysis of observations (Rinne et al., 2018). For the opposite control pattern on simulated CH$_4$ emissions at the Sarawak3 site (Figure S1), which may be due to the high and narrow range of soil temperature in tropical regions (Hirano et al., 2014; Melling et al., 2005).

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**Figure 9.** Simulated contributions (%) of CH$_4$ oxidation, diffusion, plant transport, and ebullition processes to CH$_4$ production (which is set to 100%); the rest is left in the soil.
5.3. Model Limitations

Comparing to site-level observations inevitably leads to differences and occasionally substantial deviations. Our evaluation identified large differences between modeled and observed CH$_4$ emissions at some sites. An interpretation for the differences between predictions and observations is the inadequacy of model algorithms. Although we used several microbial mechanisms to represent CH$_4$ production and oxidation, no observations can be acquired at the site level to evaluate these microbial dynamics, which needs to be improved in the future (Allison et al., 2010). Ebulition is an episodic and complex process that depends on the total partial pressure of dissolved gases and atmospheric and hydrostatic pressure (Tokida et al., 2007). Although we adopted the hydrostatic equilibrium-based algorithm to describe this process, the modeled ebullition may also contribute to the differences to reproduce the observed CH$_4$ emissions, especially for peak values (Wania et al., 2010).

The gridded inputs play an important role in the prediction at regional and global scales, while local environmental conditions may differ significantly, especially for the meteorological conditions (Wania et al., 2010). Deviations in these conditions will be added into the simulation of vegetation production and soil hydrothermal dynamics and so will propagate into the CH$_4$ biogeochemical modeling (Riley et al., 2011). Moreover, the coarse time resolution of daily meteorological values may not capture the sharp changes accurately in the processes related to CH$_4$ emissions, like ebullition. Thus, the uncertainties from forcing data may also contribute to the differences between predictions and observations.

The performance of this new model is comparable with other models. Raivonen et al. (2017) developed a CH$_4$ emission model of peatland soils (Helsinki Model of MEthane buiLd-up and emission, HIMMELI) and evaluated at the Siikaneva site. Overall, HIMMELI model showed the coefficient of determination ($R^2$) between observations and of 0.63 at the Siikaneva site, which are comparable with our model of 0.52 (Table 5). Wania et al. (2010) evaluated the performance of LPJ-WHYMe (Lund-Potsdam-Jena Wetland Hydrology and Methane) at seven sites, and three of seven sites also are included in our model evaluation (i.e., Michigan, Minnesota, and Ruoergai). The RMSE of LPJ-WHYMe at these three sites range from 0.0011 to 0.014 mol m$^{-2}$ d$^{-1}$ using global parameters, and the RMSE of our model are quite close with them (0.0029 to 0.0147 mol m$^{-2}$ d$^{-1}$) (Table 5).

Continuous observation and modeling work should be conducted to improve our knowledge of wetland CH$_4$ dynamics. It is necessary to measure multiple gases at different temporal and spatial scales and separate their different transport pathways (Bridgham et al., 2013). For those areas with sparse observations, such as the tropics, more extensive measurements are particularly needed. Soil carbon and hydrothermal dynamics are important for CH$_4$ biogeochemical modeling (Kaiser et al., 2017). Although both have been integrated into terrestrial ecosystem models, the improvement of these processes, especially the modeling of water table dynamics, should be made at the fine scale (Zhu et al., 2014). In addition, CH$_4$ production and oxidation driven by microbes, the important predecessor processes of CH$_4$ emissions, require further breakthroughs in experiments and observations and should be better to serve to the evaluation of the microbial dynamics (Xu et al., 2016).

6. Conclusions

A new wetland CH$_4$ emission model was developed and integrated into a terrestrial ecosystem model (IBIS). The new model fully considered CH$_4$ production, oxidation, and three transport pathways and the interaction between CH$_4$ and other gases and used four main microbial mechanisms to represent CH$_4$ production and oxidation. We evaluated the model at 24 globally representative wetland sites. The simulated and observed results showed good agreement for most sites in terms of emission magnitude and variability, and the mean simulated and observed values were highly correlated with an $R^2$ of 0.87. Sensitivity analysis indicated that those controlling DOC and acetate production and acetoclastic methanogenesis are the main parameters that affect CH$_4$ emissions. The new process-based model is an attempt to incorporate the microbial mechanisms into the wetland CH$_4$ emission modeling. In the future, more complete observations and better integration with terrestrial ecosystem models will help to reduce the uncertainties in prediction.
Appendix A: Microbial Dynamics Related to CH₄ Production and Oxidation

Microbial dynamics related to CH₄ production and oxidation are represented by the following equations, which are derived from Kettunen (2003) and Grant (1998):

\[
\begin{align*}
\frac{\partial \text{Homoacetogens}}{\partial t} &= \text{Homoacetogens}_{\text{growth}} - \text{Homoacetogens}_{\text{death}}, \\
\frac{\partial \text{H₂methanogens}}{\partial t} &= \text{H₂methanogens}_{\text{growth}} - \text{H₂methanogens}_{\text{death}}, \\
\frac{\partial \text{Acemethanogens}}{\partial t} &= \text{Acemethanogens}_{\text{growth}} - \text{Acemethanogens}_{\text{death}}, \\
\frac{\partial \text{Methanotrophs}}{\partial t} &= \text{Methanotrophs}_{\text{growth}} - \text{Methanotrophs}_{\text{death}},
\end{align*}
\]

where

\[
\begin{align*}
\text{Homoacetogens}_{\text{growth}} &= \text{Grow}_{\text{Homoacetogens}} \times 4 \times \text{H₂prodAce}, \\
\text{H₂methanogens}_{\text{growth}} &= \text{Grow}_{\text{H₂methanogens}} \times 4 \times \text{H₂prodCH₄}, \\
\text{Acemethanogens}_{\text{growth}} &= \text{Grow}_{\text{Acemethanogens}} \times \text{Acecons}, \\
\text{Methanotrophs}_{\text{growth}} &= \text{Grow}_{\text{Methanotrophs}} \times R_{\text{oxidCH₄}}, \\
\text{Homoacetogens}_{\text{death}} &= \text{Dead}_{\text{Homoacetogens}} \times \text{Homoacetogens} \times f_{T1}, \\
\text{H₂methanogens}_{\text{death}} &= \text{Dead}_{\text{H₂methanogens}} \times \text{H₂methanogens} \times f_{T2}, \\
\text{Acemethanogens}_{\text{death}} &= \text{Dead}_{\text{Acemethanogens}} \times \text{Acemethanogens} \times f_{T}(\text{CH₄prodQ₁₀}), \\
\text{Methanotrophs}_{\text{death}} &= \text{Dead}_{\text{Methanotrophs}} \times \text{Methanotrophs} \times f_{T}(\text{CH₄oxidQ₁₀}).
\end{align*}
\]

Here, \(\text{Homoacetogens}_{\text{growth}}\) and \(\text{Homoacetogens}_{\text{death}}\) are the growth and the death of homoacetogens, respectively; \(\text{H₂methanogens}_{\text{growth}}\) and \(\text{H₂methanogens}_{\text{death}}\) are the growth and death of hydrogenotrophic methanogens, respectively; \(\text{Acemethanogens}_{\text{growth}}\) and \(\text{Acemethanogens}_{\text{death}}\) are the growth and the death of acetoclastic methanogens, respectively; and \(\text{Methanotrophs}_{\text{growth}}\) and \(\text{Methanotrophs}_{\text{death}}\) are the growth and the death of methanotrophs, respectively. \(\text{Grow}_{\text{Homoacetogens}}\), \(\text{Grow}_{\text{H₂methanogens}}\), \(\text{Grow}_{\text{Acemethanogens}}\), and \(\text{Grow}_{\text{Methanotrophs}}\) are the growth efficiency for the corresponding microbial functional group, the values of which are 0.2, 0.2, 0.3, and 0.4, respectively; \(\text{Dead}_{\text{Homoacetogens}}\), \(\text{Dead}_{\text{H₂methanogens}}\), \(\text{Dead}_{\text{Acemethanogens}}\), and \(\text{Dead}_{\text{Methanotrophs}}\) are the death rate (\(d^{-1}\)) for the corresponding microbial functional group, whose values are uniformly set to 0.06.

Appendix B: Coefficients Associated With Transportation of Gases.

The diffusivities of four gases in air and water (Tang et al., 2010) are calculated as

\[
\begin{align*}
D_{\text{CH₄}}^{\text{air}} &= 1.9 \times 10^{-5} \times \left( \frac{T}{T_\odot} \right)^{1.82}, \\
D_{\text{O₂}}^{\text{air}} &= 1.8 \times 10^{-5} \times \left( \frac{T}{T_\odot} \right)^{1.82}, \\
D_{\text{CO₂}}^{\text{air}} &= 1.47 \times 10^{-5} \times \left( \frac{T}{T_\odot} \right)^{1.792}, \\
D_{\text{H₂}}^{\text{air}} &= 6.68 \times 10^{-5} \times \left( \frac{T}{T_\odot} \right)^{1.82},
\end{align*}
\]
\[ D_{\text{water}}^{\text{CH}_4} = 1.5 \times 10^{-9} \times \left( \frac{T}{T_\theta} \right), \]  
(B5)

\[ D_{\text{water}}^{\text{O}_2} = 2.4 \times 10^{-9} \times \left( \frac{T}{T_\theta} \right), \]  
(B6)

\[ D_{\text{water}}^{\text{CO}_2} = 1.81 \times 10^{-6} \times \exp \left( -\frac{2.032.6}{T} \right), \]  
(B7)

\[ D_{\text{water}}^{\text{H}_2} = 5.11 \times 10^{-9} \times \left( \frac{T}{T_\theta} \right). \]  
(B8)

The Henry’s law constant for each gas is computed following Sander (2015), which can be expressed as

\[ H_{\text{CH}_4} = 1.3 \times 10^{-3} \times \exp \left[ 1.700 \times \left( \frac{1}{T} - \frac{1}{T_\theta} \right) \right], \]  
(B9)

\[ H_{\text{O}_2} = 1.3 \times 10^{-3} \times \exp \left[ 1.500 \times \left( \frac{1}{T} - \frac{1}{T_\theta} \right) \right], \]  
(B10)

\[ H_{\text{CO}_2} = 3.4 \times 10^{-2} \times \exp \left[ 2.400 \times \left( \frac{1}{T} - \frac{1}{T_\theta} \right) \right], \]  
(B11)

\[ H_{\text{H}_2} = 7.8 \times 10^{-4} \times \exp \left[ 530 \times \left( \frac{1}{T} - \frac{1}{T_\theta} \right) \right]. \]  
(B12)

Here, \( T \) is the soil temperature (K), and \( T_\theta \) and \( T_\phi \) are reference temperatures (K) with values of 273.15 and 298, respectively. In here, \( H_X \) is expressed in M atm\(^{-1}\) and can be converted to mol m\(^{-3}\) Pa by multiplying with the conversion factor \( \delta \), which value is 9.8623 \times 10^{-3} \) (Sander, 2015).

The Schmidt number for each gas is calculated following Wania et al. (2010), which can be expressed as

\[ \text{Sc}_{\text{CH}_4} = 1.898 - 110.1 \times T_s + 2.834 \times T_s^2 - 0.02791 \times T_s^3, \]  
(B13)

\[ \text{Sc}_{\text{O}_2} = 1.800.6 - 120.1 \times T_s + 3.7818 \times T_s^2 - 0.047608 \times T_s^3, \]  
(B14)

\[ \text{Sc}_{\text{CO}_2} = 1.911 - 113.7 \times T_s + 2.967 \times T_s^2 - 0.02943 \times T_s^3, \]  
(B15)

\[ \text{Sc}_{\text{H}_2} = 629.95 - 34.691 \times T_s + 0.8681 \times T_s^2 - 0.0084 \times T_s^3. \]  
(B16)

Here, \( T_s \) is the soil temperature in degree Celsius.

**Appendix C: Environmental Controls.**

The soil temperature factors are calculated as

\[ f_T(Q_{10}) = \begin{cases} 
0 & \text{if } T < 0 \\
\frac{T - 30}{10} & \text{if } 0 \leq T \leq 30 \\
1 & \text{if } T > 30
\end{cases}, \]  
(C1)

\[ f_T = \frac{(T - T_{\min 1}) \times (T - T_{\max 1})}{(T - T_{\min 2}) \times (T - T_{\max 2}) - (T - T_{\text{opt}})^2}, \]  
(C2)

\[ f_T = \frac{(T - T_{\min 1}) \times (T - T_{\max 2})}{(T - T_{\min 2}) \times (T - T_{\max 2}) - (T - T_{\text{opt}})^2}. \]  
(C3)

The soil pH factor is calculated as
Here, $T$ is the soil temperature ($°C$); $Q_{10}$ represents the temperature sensitivity in different conditions, including DOCprodQ10, AccepQ10, CH4prodQ10, and CH4oxidQ10, all of which have a value of 2.5; $T_{min1}$ and $T_{min2}$ are the minimum soil temperature ($°C$), whose values are 0 and 20, respectively; $T_{max1}$ and $T_{max2}$ are the maximum soil temperature ($°C$), whose values are 20 and 50, respectively; and $T_{opt1}$ and $T_{opt2}$ are the optimum soil temperature ($°C$), whose values are 10 and 35, respectively. pH is the pH value, and $pH_{min}$, $pH_{max}$ and $pH_{opt}$ are the minimum, maximum, and optimum pH, with values of 3, 9, and 6.2, respectively. Soil pH factor was calculated with reference to Cao et al. (1996) and Meng et al. (2012). The soil moisture factor $f_{moist}$ was adopted from the IBIS.

**References**

Allison, S. D., Wallenstein, M. D., & Bradford, M. A. (2010). Soil-carbon response to warming dependent on microbial physiology. *Nature Geoscience*, 3, 336–340. https://doi.org/10.1038/ngeo846

Araújo, R. J., & Stephen, K. D. (1998). A model of the processes leading to methane emission from peatland. *Atmospheric Environment*, 32(19), 3257–3264. https://doi.org/10.1016/S1352-2310(98)00052-1

Bohn, T. J., & Lettenmaier, D. P. (2010). Systematic biases in large-scale estimates of wetland methane emissions arising from water table formulations. *Geophysical Research Letters*, 37, L22401. https://doi.org/10.1029/2010GL045450

Bohn, T. J., Melton, J. R., Ito, A., Kleinen, T., Spahni, R., Stocker, B. D., et al. (2015). WETCHIMP-WSL: Intercomparison of wetland methane emissions models over West Siberia. *Biogeosciences*, 12(11), 3321–3349. https://doi.org/10.5194/bg-12-3321-2015

Bridgham, S. D., Cadillo-Quiroz, H., Keller, J. K., & Zhuang, Q. L. (2013). Methane emissions from wetlands: Biogeochemical, microbial, and modeling perspectives from local to global scales. *Global Change Biology*, 19(5), 1325–1346. https://doi.org/10.1111/gcb.12131

Brown, M. G., Humphreys, E. R., Moore, T. R., N. T., & Lafleur, P. M. (2014). Evidence for a nonmonotonic relationship between ecosystem-scale peatland methane emissions and water table depth. *Journal of Geophysical Research: Biogeosciences*, 119, 826–835. https://doi.org/10.1002/2013JG002576

Cao, M., Marshall, S., & Gregson, K. (1996). Global carbon exchange and methane emissions from natural wetlands: Application of a process-based model. *Journal of Geophysical Research*, 101(D9), 14,399–14,414. https://doi.org/10.1029/96JD00219

Chanton, J. P. (2005). The effect of gas transport on the isotope signature of methane in wetlands. *Organic Geochemistry*, 36(5), 753–768. https://doi.org/10.1016/j.orggeochem.2004.10.007

Christensen, T. R., Ekberg, A., et al. (2003). Factors controlling large-scale variations in methane emissions from wetlands. *Geophysical Research Letters*, 30(7), 1414. https://doi.org/10.1029/2002GL016848

Cole, J. J., & Caraco, N. F. (1998). Atmospheric exchange of carbon dioxide in a low-wind oligotrophic lake measured by the addition of SF6. *Limnology and Oceanography*, 43(4), 647–656. https://doi.org/10.4319/lo.1998.43.4.0647

Colmer, T. D. (2003). Long distance transport of gases in plants: A perspective on internal aeration and radial oxygen loss from roots. *Plant, Cell & Environment*, 26(1), 17–36. https://doi.org/10.1046/j.1365-3040.2003.00846.x

Conrad, R. (1999). Contribution of hydrogen to methane production and control of hydrogen concentration in methanogenic soils and sediments. *FEMS Microbiology Ecology*, 28(3), 193–202. https://doi.org/10.1016/S0168-6496(98)00086-5

Crestaux, T., Le Matre, O., & Martinez, J. (2009). Polynomial chaos expansion for sensitivity analysis. *Environmental Modelling & Software*, 24(3), 252–260. https://doi.org/10.1016/j.envsoft.2008.10.008

Ding, W. X., Cai, Z. C., & Wang, D. X. (2004). Preliminary budget of methane emissions from natural wetlands in China. *Atmospheric Environment*, 38(5), 751–759. https://doi.org/10.1016/j.atmosenv.2003.10.016

Dise, N. B. (1993). Methane emission from Minnesota peatlands: Spatial and seasonal variability. *Global Biogeochemical Cycles*, 7(1), 123–142. https://doi.org/10.1029/92GB02299

Dunfield, P., Dumont, R., & Moore, T. R. (1993). Methane production and consumption in temperate and subarctic peat soils, response to temperature and pH. *Soil Biology and Biochemistry*, 25(3), 321–326. https://doi.org/10.1016/0038-0717(93)90130-4

Foley, J. A., Prentice, I. C., Ramankutty, N., Levis, S., Pollard, D., Sitch, S., & Haxeltine, A. (1996). An integrated biogeochemical model of land surface processes, terrestrial carbon balance, and vegetation dynamics. *Global Biogeochemical Cycles*, 10(4), 603–678. https://doi.org/10.1029/96GB02692

Fortuniak, K., Pawlak, W., Bednorz, L., Grygoruk, M., Siedlecki, M., & Zieliński, M. (2017). Methane and carbon dioxide fluxes of a temperate mere in Central Europe. *Agricultural and Forest Meteorology*, 232, 306–318. https://doi.org/10.1016/j.agrformet.2016.08.023

Friborg, T., Soegaard, H., Christensen, T. R., Lloyd, C. R., & Panikov, N. S. (2003). Siberian wetlands: Where a sink is a source. *The New Phytologist*, 159(2), 398–408. https://doi.org/10.1111/j.1469-8137.2003.00604.x

Fritz, C., Pancotto, V. A., Elzena, J. T. M., Visser, E. J. W., Grootjans, A. P., Pol, A., et al. (2011). Zero methane emission bogs: Extreme rhizosphere oxygenation by cushion plants in Patagonia. *The New Phytologist*, 190(2), 398–408. https://doi.org/10.1111/j.1469-8137.2010.03604.x

Gelaro, R., McCarty, W., Suárez, M. J., Todling, R., Molod, A., Takacs, L., et al. (2017). The modern-era retrospective analysis for research and applications, version 2 (MERRA-2). *Journal of Climate*, 30(14), 5419–5454. https://doi.org/10.1175/JCLI-D-16-0785.1

Gerard, G., & Chanton, J. (1993). Quantification of methane oxidation in the rhizosphere of emergent aquatic macrophytes: Defining upper limits. *Biogeosciences*, 2(2), 79–97. https://doi.org/10.5194/bg-2-79-1995

Global Soil Data Task Group. (2000). Global gridded surfaces of selected soil characteristics (IGBP-DIS). ORNL DAAC, Oak Ridge, Tennessee, USA. https://doi.org/10.3334/ORNLDAAC/569.

Goodrich, J. P., Campbell, D. L., Roulet, N. T., Clearwater, M. J., & Schipper, L. A. (2015). Overriding control of methane flux temporal variability by water table dynamics in a Southern Hemisphere, raised bog. *Journal of Geophysical Research: Biogeosciences*, 120(5), 819–831. https://doi.org/10.1002/2014JG002844
Meng, L., Hess, P. G. M., Mahowald, N. M., Yavitt, J. B., Riley, W. J., Subin, Z. M., et al. (2012). Sensitivity of wetland methane emissions to model assumptions: Application and model testing against site observations. Biogeosciences, 9(7), 2793–2819. https://doi.org/10.5194/bg-9-2793-2012

Mer, J. L., & Roger, P. (2001). Production, oxidation, emission, and consumption of methane by soils: A review. European Journal of Soil Biology, 37(1), 25–50. https://doi.org/10.1016/S1164-5563(01)00167-6

Mikhailov, O. A., Miglovits, M. N., & Zagirova, S. V. (2015). Vertical methane fluxes in mesosoligotrophic boreal peatland in European northeast Russia. Contemporary Problems of Ecology, 8(3), 368–375. https://doi.org/10.13144/R102.5515.030099

Moore, T. R., De Young, A., Bubier, J. L., Humphreys, E. R., Laffle, P. M., & Roulet, N. T. (2011). A multi-year record of methane flux at the Mer Bleue bog, Southern Canada. Ecosystems, 14, 6465–657. https://doi.org/10.1007/s10021-011-9455-9

Morel, X., Decharme, B., Delire, C., Knirrer, G., Lund, M., Hansen, B. U., & Bastenope, M. (2019). A new process-based soil methane scheme for land surface modeling: Evaluation over Arctic field sites with the ISRA land surface model. Journal of Advances in Modeling Earth Systems, 11, 293–326. https://doi.org/10.1029/2018MS001329

Münchberger, W., Knorr, K.-H., Blodau, C., Pancotto, V. A., & Kleinebecker, T. (2019). Zero to moderate methane emissions in a densely rooted, pristine Patagonian bog—Biogeochemical controls as revealed from isotopic evidence. Biogeosciences, 16(2), 541–559. https://doi.org/10.5194/bg-16-541-2019

Myhre, G., Shindell, D., Bréon, F.-M., Collins, W., Fuglestvedt, J., Huang, J., et al. (2013). Anthropogenic and natural radiative forcing. In T. F. Stocker, D. Qin, G.-K. Plattner, M. Tignor, S. K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex, & P. M. Midgley (Eds.), Climate change 2013: The physical science basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, (pp. 659–740). Cambridge, UK and New York, NY, USA: Cambridge University Press.

Nadeau, D. F., Rousseau, A. N., Courtois, C., Margolis, H. A., & Parlangue, M. B. (2013). Summer methane fluxes from a boreal bog in northern Quebec, Canada, using eddy covariance measurements. Atmospheric Environment, 81, 464–474. https://doi.org/10.1016/j.atmosenv.2013.09.044

Noyce, G. L., Varner, R. K., Bubier, J. L., & Frolking, S. (2014). Effect of Carex rostrata on seasonal and interannual variability in peatland methane emissions. Journal of Geophysical Research: Biogeosciences, 119, 24–34. https://doi.org/10.1002/2013JG002474

Olson, D. M., Griffis, T. J., Noormets, A., Kolka, R., & Chen, J. (2013). Interannual, seasonal, and retrospective analysis of the methane and carbon dioxide budgets of a temperate peatland. Journal of Geophysical Research: Biogeosciences, 118, 226–238. https://doi.org/10.1002/2012JG002931

Petrescu, A. M. R., van Huissteden, J., Jackowicz-Korczynski, M., Yurova, A., Christensen, T. R., Crill, P. M., et al. (2008). Modelling CH₄ emissions from Arctic wetlands: Effects of hydrological parameterization. Biogeosciences, 5(1), 111–121. https://doi.org/10.5194/bg-5-111-2008

Press, W. H., Teukolsky, S. A., Vetterling, W. T., & Flannery, B. P. (1996). Numerical recipes in Fortran 90: The art of parallel scientific computing, (2nd ed.). New York: Cambridge University Press.

Raijonen, M., Smolander, X., Backman, L., Susiluoto, J., Aalto, T., Markkanen, T., et al. (2017). HIMMELI v1.0: Helsinki Model of MERthane buildUp and emission for peatlands. Geoscientific Model Development, 10(12), 4665–4691. https://doi.org/10.5194/gmd-10-4665-2017

Ricciuto, D., Sargoyan, K., & Thornton, P. (2018). The impact of parametric uncertainties on biogeochemistry in the ESM land model. Journal of Advances in Modeling Earth Systems, 10, 297–319. https://doi.org/10.1002/2017MS001062

Riley, W. J., Subin, Z. M., Lawrence, D. M., Swenson, S. C., Torn, M. S., Meng, L., et al. (2011). Barriers to predicting changes in global terrestrial methane fluxes: Analyses using CLM4Me, a methane biogeochemistry model integrated in CESM. Biogeosciences, 8, 1925–1955. https://doi.org/10.5194/bg-8-1925-2011

Ringeval, B., de Noblet-Ducoudré, N., Ciais, P., Bousquet, P., Prigent, C., Para, F., & Rosswos, W. B. (2010). An attempt to quantify the impact of changes in wetland extent on methane emissions on the seasonal and interannual time scales. Global Biogeochemical Cycles, 24, GB2003. https://doi.org/10.1029/2009GB003354

Ringeval, B., Friedlingstein, P., Koven, C., Ciais, P., de Noblet-Ducoudré, N., Decharme, B., & Cadule, P. (2011). Climate-CH₄ feedback from wetlands and its interaction with the climate-CO₂ feedback. Biogeosciences, 8(8), 2137–2157. https://doi.org/10.5194/bg-8-2137-2011

Rinne, J., Tuittila, E. S., Peltola, O., Li, X., Raijonen, M., Alekseyev, P., et al. (2018). Temporal variation of ecosystem scale methane emission from a boreal fen in relation to temperature, water table position, and carbon dioxide fluxes. Global Biogeochemical Cycle, 32, 1087–1106. https://doi.org/10.1002/2017GB005747

Sander, R. (2015). Compilation of Henry’s law constants (version 4.0) for water as solvent. Journal of Advances in Modeling Earth Systems, 1, 226. https://doi.org/10.1002/2015MS000139

Sato, I. M. (1993). Sensitivity estimates for nonlinear mathematical models. Mathematical Modeling and Computational Experiment, 1, 407–414.

Song, W., Wang, H., Wang, G., Chen, L., Jin, Z., Zhuang, Q., & He, J.-S. (2015). Methane emissions from an alpine wetland on the Tibetan Plateau: Neglected but vital contribution of the nongrowing season. Journal of Geophysical Research: Biogeosciences, 120, 1475–1490. https://doi.org/10.1002/2015JG003043

Stephen, K. D., Arah, J. R. M., Daulat, W., & Clymo, R. S. (1998). Root-mediated gas transport in peat determined by argon diffusion. Soil Biology and Biochemistry, 30(4), 501–506. https://doi.org/10.1016/S0038-0717(97)00042-9

Strachan, I. B., Nugent, K. A., Crombie, S., & Bonneville, M. (2015). Carbon dioxide and methane exchange at a cool-temperate freshwater marsh. Environmental Research Letters, 10(6). https://doi.org/10.1088/1748-9326/10/6/065006
