A microbial functional group-based CH$_4$ model integrated into a terrestrial ecosystem model: model structure, site-level evaluation and sensitivity analysis

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Key Points:

- A new process-based model incorporating microbial mechanisms was developed to quantify CH4 emissions from natural wetlands.
- The model was applied to 24 different wetlands globally to compare the simulated CH4 emissions to observations.
- The sensitivity analysis showed the significant impacts of parameters regulating DOC and acetate production, and acetoclastic methanogenesis on simulated CH4 emissions.
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 multi-substance 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 (DOC) 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; (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 assess of process components of CH4 cycling demonstrated that 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 1803 ± 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 CH4 emissions source [Bridgham et al., 2013; Saunois et al., 2016]. The estimation of CH4 emissions from natural wetlands range from 153 to 227 Tg CH4 yr\(^{-1}\) for the 2003–2012 period, which accounts for 30% (top-down inversions) and 25% (bottom-up estimation) on the average of total CH4 emissions [Saunois et al., 2016], meaning natural wetlands play an important role in the global CH4 budget and climate change.

CH4 emission from natural wetlands is a net balance between microbial CH4 production and consumption. There exists two major CH4 production mechanisms: hydrogenotrophic methanogenesis and acetoclastic methanogenesis [Conrad, 1999; Krüger et al., 2001], two CH4 oxidation mechanisms: aerobic and anaerobic methanotrophy [Gerard and Chanton, 1993; Smemo and Yavitt, 2011] and three aggregated CH4 transport pathways: plant-mediated transport, diffusion and ebullition [Chanton, 2005; Mer and Roger, 2001; Whiting and Chanton, 1996], all of which are influenced by the availability and quality of substrate, soil temperature and pH, hydrological conditions, and vegetation composition, etc. [Walter and 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 and Stephen, 1998], ecosys [Grant, 1998, 1999], the Walter model [Walter and Heimann, 2000], Wetland-DNDC [Zhang et al., 2002], the Kettunen model [Kettunen, 2003], PEATLAND-VU [Van Huisseldon et al., 2006], TEM-CH$_4$ [Tang et al., 2010; Zhuang et al., 2004], DLEM [Tian et al., 2010], ORCHIDEE [Ringeval et al., 2010, 2011], LPJ-WHYMe [Wania et al., 2010], CLM4Me [Riley et al., 2011], VISIT [Ito and 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; (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 and Lettenmaier, 2010; Bohn et al., 2015]. A recent model inter-comparison project showed that the maximum estimate of global wetland CH$_4$ emissions is 264 Tg CH$_4$ yr$^{-1}$, which is 1.89 times the minimum estimate of 141 Tg CH$_4$ yr$^{-1}$ [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 and 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 which 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, etc. [e.g. Grant, 1998, 1999; Kettunen, 2003; Xu et al., 2015].

In this study, we developed a new process-based model to quantify CH$_4$ emissions from natural wetlands, which considered the interaction of different gases, including CH$_4$, O$_2$, CO$_2$ and H$_2$, and incorporated the main microbial mechanisms related to CH$_4$ production and
oxidation. Moreover, the new process-based model has been integrated in a terrestrial ecosystem model (Integrated Biosphere Simulator), 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, (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 Integrated Biosphere Simulator (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, carbon and nitrogen cycling in the terrestrial biosphere and have been integrated into a single, physically consistent, with different timescales, modeling framework [Foley et al., 1996; Kucharik et al., 2000; Yuan et al., 2014; Liu et al., 2014], here we only describe the development of novel wetland CH₄ dynamics. The new wetland CH₄ emission model (Fig. 1) is a multi-dimensional 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:

\[
\frac{\partial}{\partial t} C_{\text{CH}_4}(z, t) = \frac{\partial}{\partial z} F_{\text{diff}, \text{CH}_4} - Q_{\text{plant}, \text{CH}_4} - Q_{\text{ebull}, \text{CH}_4} + R_{\text{prod}, \text{CH}_4} - R_{\text{oxid}, \text{CH}_4} \tag{1}
\]

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

\[
\frac{\partial}{\partial t} C_{\text{CO}_2}(z, t) = \frac{\partial}{\partial z} F_{\text{diff}, \text{CO}_2} - Q_{\text{plant}, \text{CO}_2} - Q_{\text{ebull}, \text{CO}_2} + R_{\text{prod}, \text{CO}_2} - R_{\text{cons}, \text{CO}_2} \tag{3}
\]

\[
\frac{\partial}{\partial t} C_{\text{H}_2}(z, t) = \frac{\partial}{\partial z} F_{\text{diff}, \text{H}_2} - Q_{\text{plant}, \text{H}_2} - Q_{\text{ebull}, \text{H}_2} + R_{\text{prod}, \text{H}_2} - R_{\text{cons}, \text{H}_2} \tag{4}
\]

\[
\frac{\partial}{\partial t} C_{\text{Ace}}(z, t) = R_{\text{prod}, \text{Ace}} - R_{\text{cons}, \text{Ace}} \tag{5}
\]

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 \(X\), \(R_{\text{oxid}, \text{CH}_4}\) is the CH₄ 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 CH₄ is produced in the layers below the water table position, and mainly consumed in the layers above the water table position [Walter and Heimann, 2000].

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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 CH$_4$-related biogeochemical processes occurring in the nine soil layers. The detailed algorithms are described as follows.

2.1 CH$_4$ production

2.1.1 Available carbon decomposition

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

$$DOC = K_{cpool} \times c_{pool} \times f_T(DOCprodQ_{10}) \times f_{moist}$$

(6)

Here, DOC is the dissolved organic carbon concentration (mol m$^{-3}$), $K_{cpool}$ represents the ratio of dissolved organic carbon to soil organic carbon, $c_{pool}$ 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 $f_{moist}$ 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 $f_{root}$, which is calculated in the IBIS.

Under anaerobic conditions, the available carbon is fermented into acetate, CO$_2$ and H$_2$, which is governed by the following equation:

$$DOC \rightarrow 0.67Ace + 0.33CO_2 + 0.11H_2$$

(7)

Thus, the fermentation is presented as:

$$DOCprodAce = V_{DOCprodAce, max} \times \frac{DOC}{K_{DOCprodAce} + DOC} \times f_T(DOCprodQ_{10}) \times f_{pH}$$

(8)

$$DOCprodCO2 = 0.5 \times DOCprodAce$$

(9)

$$DOCprodH2 = \frac{1}{6} \times DOCprodAce$$

(10)

Under aerobic conditions, the available carbon is decomposed into acetate and CO$_2$, which can be calculated as:

$$DOCprodAce = V_{DOCprodAce, max} \times \frac{DOC}{K_{DOCprodAce} + DOC} \times \frac{[O_2]}{K_{AceprodO2} + [O_2]} \times$$

$$f_T(DOCprodQ_{10}) \times f_{pH}$$

(11)

$$DOCprodCO2 = 0.5 \times DOCprodAce$$

(12)

Here, DOCprodAce, DOCprodCO2 and DOCprodH2 are the production rate (mol m$^{-3}$ d$^{-1}$) of acetate, CO$_2$ and O$_2$, respectively. $V_{DOCprodAce, max}$ is the maximum acetate production rate.
(mol m$^{-3}$ d$^{-1}$), [O$_2$] is the O$_2$ concentration (mol m$^{-3}$), and $K_{DOCprodA}$ and $K_{AceprodO2}$ are the half saturation coefficients (mol m$^{-3}$) for DOC and O$_2$, respectively. $AceprodQ_{10}$ is the temperature sensitivity of acetate production, and $f_{pH}$ 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 and 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) [Liu and Conrad, 2011; Kotsyurbenko et al., 2001; Schulz and Conrad, 1996]. This process is governed by the following equation:

$$4H_2 + 2CO_2 \rightarrow CH_3COOH + 2H_2O \quad (13)$$

Thus, acetate production from homoacetogenesis is calculated as:

$$H2prodAce = V_{H2prodAce, max} \times Homoacetogens \times \frac{[H_2]}{K_{H2prodAce} + [H_2]} \times \frac{[CO_2]}{K_{CO2prodAce} + [CO_2]} \times f_{T1} \times f_{pH} \quad (14)$$

Here, $H2prodAce$ is the acetate production rate from homoacetogenesis (mol m$^{-3}$ d$^{-1}$), $V_{H2prodAce, max}$ 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$^{-3}$). $K_{H2prodAce}$ and $K_{CO2prodAce}$ are the half saturation coefficients (mol m$^{-3}$) for H$_2$ and CO$_2$, respectively, and $f_{T1}$ 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 [Hattori, 2008], which use CO$_2$ and H$_2$ to generate CH$_4$ (hydrogenotrophic methanogenesis). This process is governed by the following equation:

$$4H_2 + CO_2 \rightarrow CH_4 + 2H_2O \quad (15)$$

Thus, CH$_4$ production from hydrogenotrophic methanogenesis is calculated as:

$$H2prodCH4 = V_{H2prodCH4, max} \times H2methanogens \times \frac{[H_2]}{K_{H2prodCH4} + [H_2]} \times \frac{[CO_2]}{K_{CO2prodCH4} + [CO_2]} \times f_{T2} \times f_{pH} \quad (16)$$

Here, $H2prodCH4$ is the CH$_4$ production rate from hydrogenotrophic methanogenesis (mol m$^{-3}$ d$^{-1}$), $V_{H2prodCH4, max}$ is the maximum CH$_4$ production rate (mol m$^{-3}$ d$^{-1}$), H2methanogens is the microbial biomass of hydrogenotrophic methanogens (mol m$^{-3}$), $K_{H2prodCH4}$ and $K_{CO2prodCH4}$ are the half saturation coefficients (mol m$^{-3}$) for H$_2$ and CO$_2$, respectively, and $f_{T2}$ represents the soil temperature factor.
2.1.3 Acetoclastic methanogenesis

Product acetate is the substrate for acetoclastic methanogens [Grant, 1998; Grant and Roulet, 2002]. CH$_4$ production from acetoclastic methanogenesis is governed by the following equation:

\[
\text{CH}_3\text{COOH} \rightarrow \text{CH}_4 + \text{CO}_2
\] 
(17)

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:

\[
\text{Acecons} = V_{\text{Acecons,max}} \times \text{Acemethanogens} \times \frac{\text{Ace}}{k_{\text{AceprodCH}_4} + \text{Ace}} \times f_T(CH_4\text{prodQ}_{10}) \times f_{pH}
\] 
(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 and 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:

\[
R_{\text{oxid,CH}_4} = V_{\text{CH}_4\text{oxid,max}} \times \text{Methanotrophs} \times \frac{[\text{CH}_4]}{K_{\text{CH}_4\text{oxidCH}_4} + [\text{CH}_4]} \times \frac{[\text{O}_2]}{K_{\text{CH}_4\text{oxidO}_2} + [\text{O}_2]} \times f_T(CH_4\text{oxidQ}_{10}) \times f_{pH}
\] 
(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$^{-3}$), [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:
\[ R_{\text{aer}} = K_{\text{aer}} \times \frac{\text{DOC}}{K_{\text{aerDOC}} + \text{DOC}} \times \frac{[O_2]}{K_{\text{aerO2}} + [O_2]} \times f_T(\text{DOCprodQ}_{10}) \times f_{\text{pH}} \]  

(22)

Here, \( K_{\text{aer}} \) is the \( O_2 \) consumption rate by aerobic respiration (mol m\(^{-3}\) d\(^{-1}\)), and \( K_{\text{aerDOC}} \) and \( K_{\text{aerO2}} \) 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 and 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 \( f_{D, w} \) and \( f_{D, a} \) to calculate the effective diffusivities in water or air [Raivonen et al., 2017], respectively.

\[ D_{X, w} = f_{D, w} \times D_{X, \text{water}} \]  

(24)

\[ D_{X, a} = f_{D, a} \times D_{X, \text{air}} \]  

(25)

Here, \( D_{X, w} \) and \( D_{X, a} \) represent the effective diffusivities of compound \( X \) in water and air (m\(^2\) s\(^{-1}\)), and \( D_{X, \text{water}} \) and \( D_{X, \text{air}} \) 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.

\[ F_{\text{diff, X}} = -\varphi_X \times (C_{\text{surf, X}} - C_{\text{eq, X}}) \]  

(26)

Here, \( C_{\text{surf, X}} \) is the concentration of gas \( X \) in the top soil layer (mol m\(^{-3}\)), and \( C_{\text{eq, X}} \) is the equilibrium concentration of gas \( X \) in the atmosphere (mol m\(^{-3}\)). \( \varphi_X \) represents the transfer velocity of gas \( X \) (cm h\(^{-1}\)). We use a normalized transfer velocity \( \varphi_{600} \) [Cole and Caraco, 1998] to calculate the \( \varphi_X \):

\[ \varphi_X = \varphi_{600} \times \left( \frac{S_{X}}{600} \right)^n \]  

(27)

The \( \varphi_{600} \) (cm h\(^{-1}\)) is calculated as:

\[ \varphi_{600} = 2.07 + 0.215 \times U_{10}^{1.7} \]  

(28)

Here, \( S_{X} \) is the Schmidt number of gas \( X \), 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 cm h$^{-1}$, which can be further transformed into m d$^{-1}$.

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

$$C_{eq, X} = PP_X \times H_X$$

(29)

Here, $H_X$ is the Henry’s law constant for gas X (mol m$^{-3}$ Pa$^{-1}$). Based on the above equations, the diffusive flux for gas 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$^{-1}$).

$$Q_{plant, X} = \frac{D_{air, X}}{\tau} \times \epsilon(z) \times \frac{C_X(z, t) - C_{eq, X}}{z}$$

(30)

Here, $\epsilon(z)$ 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], the $\epsilon(z)$ is formulated as:

$$\epsilon(z) = a_{mA} \times \frac{f_{root}}{dz} \times \frac{LAI}{SLA}$$

(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 CH$_4$, O$_2$, CO$_2$, H$_2$ and N$_2$, when the criterion for bubble formation is reached; such that when

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

(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:

$$f_{ebull}(z) = \frac{\sum_X PP_X(z) \cdot (P_{atm} + P_{hyd})}{\sum_X PP_X(z)}$$

(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:

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

(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 CH$_4$ 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 CH$_4$ 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 includes daily climate data, daily water table depth and soil carbon data. We used the Modern-Era Retrospective Analysis for Research and Applications (MERRA) dataset [Gelaro et al., 2017] and the CPC Global Unified Precipitation data provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA (https://www.esrl.noaa.gov/psd/) to drive the model. The MERRA data provided the daily maximum, minimum and average air temperature, relative humidity and wind speed at 2m and the CPC Global Unified Precipitation data provided the daily precipitation. The soil carbon data for model initialization was obtained from the Global Gridded Surfaces of Selected Soil Characteristics (IGBP-DIS) dataset [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 (DSMW) by Land and Water Development Division, FAO (http://www.fao.org/geonetwork/srv/en/metadata.show?id=14116). A spin-up of 200 years (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 CH₄ emissions to input parameter (Table 1), the polynomial chaos expansions (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 aim to decompose the total variance of the model into the sum of the variances of each input variable or their interactions [Sudret, 2008; Wang et al., 2016; Ricciuto et al., 2018]. The polynomial chaos expansion is a powerful probabilistic technique that using the orthogonal stochastic polynomials in the random inputs to provide a functional approximation of the model output [Crestaux et al., 2009; Marellia and Sudret, 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 [Marellia and Sudret, 2019], and has been applied to some sensitivity studies of parameters in earth system model or land surface model [Shi et al., 2019; Ricciuto et al., 2018]. The PCE-based Sobol indices method is briefly introduced as follows:

For an input random vector \( X = \{X_1, \ldots, X_k\} \), the polynomial chaos expansion of model output \( Y = M(X) \) can be established as:

\[
Y = M(X) = \sum_{\alpha \in \mathbb{N}^k} \lambda_\alpha \Psi_\alpha(X)
\] (35)

Here, the \( \Psi_\alpha(X) \) are multivariate polynomials orthonormal with regard to the distribution of \( X \), \( \alpha \in \mathbb{N}^k \) is a indices vector that identifies the components of the multivariate polynomials \( \Psi_\alpha \), the \( \lambda_\alpha \in \mathbb{R} \) represent the corresponding coefficients.

The sum in Eq. (35) needs to be truncated to a finite sum, when applied in realistic situation, by the truncated polynomial chaos expansion:

\[
M(X) \approx M^{PC}(X) = \sum_{\alpha \in A} \lambda_\alpha \Psi_\alpha(X)
\] (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)

\[
V(M(X)) \approx \sum_{\alpha \neq 0} \lambda_\alpha^2,
\] (38)

\[
S_i = \frac{1}{V(M(X))} \sum_{\alpha \in A_{S_i}} \lambda_\alpha^2 \quad \text{with} \quad A_{S_i} = \{\alpha : \alpha_i > 0, \alpha_k = 0 \text{ for } k \neq i\},
\] (39)

\[
S_{Ti} = \frac{1}{V(M(X))} \sum_{\alpha \in A_{S_{Ti}}} \lambda_\alpha^2 \quad \text{with} \quad A_{S_{Ti}} = \{\alpha : \alpha_i > 0\},
\] (40)

\[
S_{ij} = \frac{1}{V(M(X))} \sum_{\alpha \in A_{S_{ij}}} \lambda_\alpha^2 \quad \text{with} \quad A_{S_{ij}} = \{\alpha : \alpha_i > 0, \alpha_j > 0, \alpha_k = 0 \text{ for } k \neq i, j\},
\] (41)
Here, $\mathbb{E}(M(X))$ and $\mathbb{V}(M(X))$ indicate the expectation and variance of $M(X)$. $S_i$, $S_{T_i}$ 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. $A_{S_i}$, $A_{S_{T_i}}$ and $A_{S_{ij}}$ is the set 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; 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:

$$\text{RMSE} = \sqrt{\frac{\sum_{i=1}^{n}(M_i - O_i)^2}{n}}$$

Here, $M_i$ is the modeled value and $O_i$ the observed value; $n$ is the number of days for which we had the observed values.

3) Relative predictive error (RPE), computed as:

$$\text{RPE} = \frac{\bar{M} - \bar{O}}{\bar{O}} \times 100\%$$

Here, $\bar{M}$ and $\bar{O}$ are the means of modeled and observed values, respectively.

4 Results

4.1 Model evaluation

Twenty-four sites were selected to evaluate the 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 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 (Fig. 2), with the relative predictive error (RPE) varying from −19.47% to 33.28% (Table 5). The magnitude and seasonality of simulated CH$_4$ emissions were consistent with the observations, especially at the Siikaneva and Lompolojänkkä sites (Fig. 2b and c). 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 (Fig. 2a). Meanwhile, the model had slight high predicted values in the growing season of 2004 at Lena Samoylov Delta (Fig. 2f). At these two sites, the 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 (Fig. 2g, i and k). 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 (Fig. 3). At the Minnesota and Mer Bleue sites (Fig. 3e1, e2 and h2), the model explained about 69%, 51% and 50% of the variation observed in CH$_4$ emissions, respectively (Table 5). The relative predictive error 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 (Fig. 3b1–b3). Although they had lower relative predictive errors, 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 (Fig. 3d1–d2 and i). The R$^2$ values were 0.118, 0.059 and 0.252 and the RMSE values were 0.0029 mol m$^{-2}$ d$^{-1}$, 0.0039 mol m$^{-2}$ d$^{-1}$ 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 (Fig. 3a2, e2, g and h4), 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 (Fig. 3a2 and h4). In addition, the model showed poor agreement between simulated and observed CH$_4$ emissions at the Luanhaizi, Kopytkowo and Kopuatai sites (Fig. 3c, k and l). The calculated R$^2$ and RPE values at these sites were 0.17 and −26.70%, 0.145 and −48.62%, 0.001 and −4.97%, respectively (Table 5). At the Kopytkowo site, the model significantly underestimated observations during the growing season in 2013 (Fig. 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 (Fig. 4). At the Sarawak1 site, the simulated CH$_4$ emissions did not reproduce the variation in observations, which had no clear seasonal patterns (Fig. 4a1). In addition, the model slightly overestimated the observed CH$_4$ emissions in November at the Sarawak2 site (Fig. 4a2). The R$^2$ and RPE values for these two sites were 0.016 and 47.04%, 0.003 and 25.08%, respectively (Table 5). The model captured the
magnitude and variation of CH₄ emissions during the measured period at the Sarawak3 site (Fig. 4a3). The R² and RPE values for this site were 0.426 and 6.25%, respectively (Table 5). The RMSE values for this peat swamp forest site ranged from 0.0003 mol m⁻² d⁻¹ to 0.001 mol m⁻² d⁻¹ (Table 5), indicating a small disparity between simulated and observed CH₄ emissions.

The average simulated and observed CH₄ emissions during measured periods for all evaluated, boreal and temperate sites are shown in Fig. 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 R² is 0.87 (Fig. 5a).

4.2 Sensitivity analysis

The PCE-based parameter sensitivity analysis of modeled CH₄ emissions with 500 model evaluations was performed at seven measured sites, covering the tropical, temperate and boreal biomes (Fig. 6 and 7). For the Siikaneva, Lompolojänkkä and Quebec sites, CH₄prodQ₁₀ (P29) is the most sensitive parameter for CH₄ emissions (Fig. 6a), which is also reflected in the evaluation of total Sobol indices (Fig. 6b). For the Sallies and Kopuatai sites, Kcpool (P1) has the largest impact on CH₄ emissions (Fig. 6a and b). In addition, The CH₄ emissions is also sensitive to VAcecons, max (P7), KAceprodCH₄ (P13) and DOCprodQ₁₀ (P27) at the Sallies site, and sensitive to KCH₄prod (P14) and CH₄prodQ₁₀ at the Kopuatai site (Fig. 6a and b). For the Schechenfilz site, the first-order Sobol indices are low for all parameters (Fig. 6a). However, the DOCprodQ₁₀ has a great total effect index of about 58% on the total variance of CH₄ emissions (Fig. 6b), which indicates the apparent impact of this parameter on the CH₄ emissions. For the Sarawak3 site, except for VAcecons, max and KCH₄prod, the first-order and total Sobol indices have low values for other parameters (Fig. 6a and b). CH₄ emissions at this site is obviously sensitive to these two parameters, with VAcecons, max becomes the most sensitive parameter (Fig. 6a and b). Moreover, compared to other sites, the Sobol indices of parameters related 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 KAceprodCH₄ and CH₄prodQ₁₀ have the largest interaction effects on CH₄ emissions at the Siikaneva, Lompolojänkkä, Quebec, Schechenfilz and Kopuatai sites (Fig. 7). The parameters VAcecons, max and KAceprodCH₄ have the largest interaction effects on CH₄ emissions at the Sallies site, and VAcecons, max and KCH₄prod have the largest interaction effects on CH₄ emissions at the Sarawak3 site (Fig. 7). In addition, VAcecons, max and CH₄prodQ₁₀ have the important interaction effects on CH₄ emissions at the Siikaneva, Lompolojänkkä and Quebec sites (Fig. 7). For the Lompolojänkkä site, the parameter subset of AceprodQ₁₀ and CH₄prodQ₁₀ also has the important interaction effects on CH₄ emissions (Fig. 7). The parameter set of Kcpool and VAcecons, max, and the parameter set of VAcecons, max and AceprodQ₁₀ also have the important interaction effects on CH₄ emissions at the Sallies site (Fig. 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
DOCprodQ_{10}, AceprodQ_{10}, CH4prodQ_{10}, K_{cpool}, V_{Acecons, max}, K_{AceprodCH4} and K_{CH4prod}, 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 (Fig. 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 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 (Fig. 8) and the proportion that each process relative to CH₄ production at all evaluated sites (Fig. 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 [Münchberger 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 mid-growing seasons (Fig. 8). This may reflect enhanced microbial activities caused by the high temperature in this period [Dunfield et al., 1993]. In addition, at all validated sites, CH₄ oxidation was observed to be about half or more of CH₄ production (Fig. 9).
Plant-mediated transport has been reported as the principal pathway for CH$_4$ emissions [Colmer, 2003; Green and Baird, 2012; King et al., 1998; Waddington et al., 1996; Whiting and 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 (Fig. 8). Moreover, plant-mediated transport also had the greatest proportion of total CH$_4$ emission at most sites (Fig. 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; Mikhailov 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 non-monotonic [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 4.3). At the Lompolojänkkä site, the water table position almost always above the soil surface (Fig. 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 Siikaneva site was controlled by soil temperature (Fig. 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 (Fig. 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].

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 site-level to evaluate these microbial dynamics, which needs to be improved in the future [Allison et al., 2010]. Ebullition 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].

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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 hydro-thermal 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$_4$ emissions 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 measurements 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 mol m$^{-2}$ d$^{-1}$ to 0.014 mol m$^{-2}$ d$^{-1}$ using global parameters, and the RMSE of our model are quite close with them (0.0029 mol m$^{-2}$ d$^{-1}$ 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 hydro-thermal 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 CH$_4$ production and oxidation.

Microbial dynamics related to CH$_4$ production and oxidation are represented by the following equations, which are derived from Kettunen [2003] and Grant [1998].
\[ \frac{\partial \text{Homoacetogens}}{\partial t} = \text{Homoacetogens}_{\text{growth}} - \text{Homoacetogens}_{\text{death}} \]  
(A1)

\[ \frac{\partial \text{H}_2\text{methanogens}}{\partial t} = \text{H}_2\text{methanogens}_{\text{growth}} - \text{H}_2\text{methanogens}_{\text{death}} \]  
(A2)

\[ \frac{\partial \text{Acemethanogens}}{\partial t} = \text{Acemethanogens}_{\text{growth}} - \text{Acemethanogens}_{\text{death}} \]  
(A3)

\[ \frac{\partial \text{Methanotrophs}}{\partial t} = \text{Methanotrophs}_{\text{growth}} - \text{Methanotrophs}_{\text{death}} \]  
(A4)

where

\[ \text{Homoacetogens}_{\text{growth}} = \text{Grow}_{\text{Homoacetogens}} \times 4 \times \text{H}_2\text{prodAce} \]  
(A5)

\[ \text{H}_2\text{methanogens}_{\text{growth}} = \text{Grow}_{\text{H}_2\text{methanogens}} \times 4 \times \text{H}_2\text{prodCH}_4 \]  
(A6)

\[ \text{Acemethanogens}_{\text{growth}} = \text{Grow}_{\text{Acemethanogens}} \times \text{Acecons} \]  
(A7)

\[ \text{Methanotrophs}_{\text{growth}} = \text{Grow}_{\text{Methanotrophs}} \times R_{\text{oxid}, \text{CH}_4} \]  
(A8)

\[ \text{Homoacetogens}_{\text{death}} = \text{Dead}_{\text{Homoacetogens}} \times \text{Homoacetogens} \times f_{T1} \]  
(A9)

\[ \text{H}_2\text{methanogens}_{\text{death}} = \text{Dead}_{\text{H}_2\text{methanogens}} \times \text{H}_2\text{methanogens} \times f_{T2} \]  
(A10)

\[ \text{Acemethanogens}_{\text{death}} = \text{Dead}_{\text{Acemethanogens}} \times \text{Acemethanogens} \times f_{T}(\text{CH}_4\text{prod}Q_{10}) \]  
(A11)

\[ \text{Methanotrophs}_{\text{death}} = \text{Dead}_{\text{Methanotrophs}} \times \text{Methanotrophs} \times f_{T}(\text{CH}_4\text{oxid}Q_{10}) \]  
(A12)

Here, \( \text{Homoacetogens}_{\text{growth}} \) and \( \text{Homoacetogens}_{\text{death}} \) are the growth and the death of homoacetogens, respectively; \( \text{H}_2\text{methanogens}_{\text{growth}} \) and \( \text{H}_2\text{methanogens}_{\text{death}} \) 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. \( \text{Grow}_{\text{Homoacetogens}}, \text{Grow}_{\text{H}_2\text{methanogens}}, \text{Grow}_{\text{Acemethanogens}}, \text{Grow}_{\text{Methanotrophs}} \) 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; \( \text{Dead}_{\text{Homoacetogens}}, \text{Dead}_{\text{H}_2\text{methanogens}}, \text{Dead}_{\text{Acemethanogens}}, \text{Dead}_{\text{Methanotrophs}} \) are the death rate (d\(^{-1}\)) for the corresponding microbial functional group, whose values were 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:

\[ D_{CH_4}^{\text{air}} = 1.9 \times 10^{-5} \times \left( \frac{T}{T_\theta} \right)^{1.82} \]  
(B1)

\[ D_{O_2}^{\text{air}} = 1.8 \times 10^{-5} \times \left( \frac{T}{T_\theta} \right)^{1.82} \]  
(B2)

\[ D_{CO_2}^{\text{air}} = 1.47 \times 10^{-5} \times \left( \frac{T}{T_\theta} \right)^{1.792} \]  
(B3)

\[ D_{H_2}^{\text{air}} = 6.68 \times 10^{-5} \times \left( \frac{T}{T_\theta} \right)^{1.82} \]  
(B4)

\[ D_{CH_4}^{\text{water}} = 1.5 \times 10^{-9} \times \left( \frac{T}{T_\theta} \right) \]  
(B5)

\[ D_{O_2}^{\text{water}} = 2.4 \times 10^{-9} \times \left( \frac{T}{T_\theta} \right) \]  
(B6)

\[ D_{CO_2}^{\text{water}} = 1.81 \times 10^{-6} \times \exp \left( \frac{-2032.6}{T} \right) \]  
(B7)

\[ D_{H_2}^{\text{water}} = 5.11 \times 10^{-9} \times \left( \frac{T}{T_\theta} \right) \]  
(B8)

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

\[ H_{CH_4} = 1.3 \times 10^{-3} \times \exp \left[ 1700 \times \left( \frac{1}{T} - \frac{1}{T_\theta} \right) \right] \]  
(B9)

\[ H_{O_2} = 1.3 \times 10^{-3} \times \exp \left[ 1500 \times \left( \frac{1}{T} - \frac{1}{T_\theta} \right) \right] \]  
(B10)

\[ H_{CO_2} = 3.4 \times 10^{-2} \times \exp \left[ 2400 \times \left( \frac{1}{T} - \frac{1}{T_\theta} \right) \right] \]  
(B11)

\[ H_{H_2} = 7.8 \times 10^{-4} \times \exp \left[ 530 \times \left( \frac{1}{T} - \frac{1}{T_\theta} \right) \right] \]  
(B12)

Here, \( T \) is the soil temperature (K), and \( T_\theta \) and \( T_\theta \) 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 multiplied the conversion factor \( \theta \), 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:
\[
\text{Sc}_{\text{CH}_4} = 1898 - 110.1 \times T_s + 2.834 \times T_s^2 - 0.02791 \times T_s^3 \\
(B13)
\]
\[
\text{Sc}_{\text{O}_2} = 1800.6 - 120.1 \times T_s + 3.7818 \times T_s^2 - 0.047608 \times T_s^3 \\
(B14)
\]
\[
\text{Sc}_{\text{CO}_2} = 1911 - 113.7 \times T_s + 2.967 \times T_s^2 - 0.02943 \times T_s^3 \\
(B15)
\]
\[
\text{Sc}_{\text{H}_2} = 629.95 - 34.691 \times T_s + 0.8681 \times T_s^2 - 0.0084 \times T_s^3 \\
(B16)
\]

Here, \( T_s \) is the soil temperature in °C.

**Appendix C** Environmental controls.

The soil temperature factors are calculated as:

\[
f_{T}(Q_{10}) = \begin{cases} 
0 & T < 0 \\
\frac{T_{-30}^{10}}{Q_{10}} & 0 \leq T \leq 30 \\
1 & T > 30 
\end{cases} \\
(C1)
\]

\[
f_{T1} = \frac{(T - T_{\text{min1}}) \times (T - T_{\text{max1}})}{(T - T_{\text{min1}}) \times (T - T_{\text{max1}}) \times (T - T_{\text{opt1}})^2} \\
(C2)
\]

\[
f_{T2} = \frac{(T - T_{\text{min2}}) \times (T - T_{\text{max2}})}{(T - T_{\text{min2}}) \times (T - T_{\text{max2}}) \times (T - T_{\text{opt2}})^2} \\
(C3)
\]

The soil pH factor is calculated as:

\[
f_{\text{pH}} = \frac{(pH - pH_{\text{min}}) \times (pH - pH_{\text{max}})^2}{(pH - pH_{\text{min}}) \times (pH - pH_{\text{max}}) \times (pH - pH_{\text{opt}})^2} \\
(C4)
\]

Here, \( T \) is the soil temperature (°C), \( Q_{10} \) represents the temperature sensitivity in different conditions, including \( \text{DOCprodQ}_{10} \), \( \text{AceprodQ}_{10} \), \( \text{CH4prodQ}_{10} \) and \( \text{CH4oxidQ}_{10} \), all of which have a value of 2.5, \( T_{\text{min1}} \) and \( T_{\text{min2}} \) are the minimum soil temperature (°C), whose values are 0 and 20, respectively, \( T_{\text{max1}} \) and \( T_{\text{max2}} \) are the maximum soil temperature (°C), whose values are 20 and 50, respectively, and \( T_{\text{opt1}} \) and \( T_{\text{opt2}} \) are the optimum soil temperature (°C), whose values are 10 and 35, respectively. \( pH \) is the pH value, and \( pH_{\text{min}} \), \( pH_{\text{max}} \) and \( pH_{\text{opt}} \) are the minimum, maximum and optimum pH, with values of 3, 9 and 6.2, respectively. Soil pH factor was calculated with reference to \( \text{Cao et al.} \ [1996] \) and \( \text{Meng et al.} \ [2012] \). The soil moisture factor \( f_{\text{moist}} \) was adopted from the IBIS.

**Acknowledgments**

The research was funded by the National Key Research and Development Program of China (No. 2016YFA0602701), National Youth Top-Notch Talent Support Program (2015-48), Changjiang Young Scholars Program of China (Q2016161), Fok Ying Tung Education Foundation and the Fundamental Research Funds for the Central Universities (No. 18lgpy09). The Modern-Era Retrospective Analysis for Research and Applications (MERRA) dataset is available at \( \text{https://gmao.gsfc.nasa.gov/} \). The CPC Global Unified Precipitation data is available at \( \text{https://www.esrl.noaa.gov/psd/} \). The Global Gridded...

References


Table 1. Major parameters in CH$_4$-related processes.

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<th>Parameter</th>
<th>Value</th>
<th>Range</th>
<th>Units</th>
<th>Descriptions</th>
<th>References</th>
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</thead>
<tbody>
<tr>
<td>P1</td>
<td>$K_{cpool}$</td>
<td>0.02</td>
<td>0.0001-0.02</td>
<td>mol m$^{-3}$ d$^{-1}$</td>
<td>ratio of dissolved organic carbon to soil organic carbon</td>
<td>Grant (1998)</td>
</tr>
<tr>
<td>P2</td>
<td>$V_{DOCprodAce, max}$</td>
<td>0.5</td>
<td>0.3-0.7</td>
<td>mol m$^{-3}$ d$^{-1}$</td>
<td>maximum acetate production rate from fermentation</td>
<td>Grant (1998)</td>
</tr>
<tr>
<td>P3</td>
<td>$K_{DOCprodAce}$</td>
<td>10.0</td>
<td>5-15</td>
<td>mol m$^{-3}$</td>
<td>half saturation coefficient</td>
<td>Kettunen (2003)</td>
</tr>
<tr>
<td>P4</td>
<td>$K_{AceprodO2}$</td>
<td>0.04</td>
<td>0.01-0.1</td>
<td>mol m$^{-3}$</td>
<td>half saturation coefficient</td>
<td>Kettunen (2003)</td>
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<tr>
<td>P5</td>
<td>$V_{H2prodAce, max}$</td>
<td>0.15</td>
<td>0.01-0.3</td>
<td>mol m$^{-3}$ d$^{-1}$</td>
<td>maximum acetate production rate from homoacetogenesis</td>
<td>Grant (1998)</td>
</tr>
<tr>
<td>P6</td>
<td>$V_{H2prodCH4, max}$</td>
<td>0.15</td>
<td>0.01-0.3</td>
<td>mol m$^{-3}$ d$^{-1}$</td>
<td>maximum CH$_4$ production rate from hydrogenotrophic methanogenesis</td>
<td>Grant (1998)</td>
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<tr>
<td>P7</td>
<td>$V_{Acecons, max}$</td>
<td>0.5</td>
<td>0.3-0.7</td>
<td>mol m$^{-3}$ d$^{-1}$</td>
<td>maximum acetate consumption rate by acetoclastic methanogenesis</td>
<td>Grant (1999)</td>
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<td>P8</td>
<td>$V_{CH4oxid, max}$</td>
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<td>0.3-0.7</td>
<td>mol m$^{-3}$ d$^{-1}$</td>
<td>maximum CH$_4$ oxidation rate</td>
<td>Grant (1999)</td>
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<td>P9</td>
<td>$K_{H2prodAce}$</td>
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<td>0.01-0.1</td>
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<td>half saturation coefficient</td>
<td>Kettunen (2003)</td>
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<td>P10</td>
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<td>0.01-0.1</td>
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<td>half saturation coefficient</td>
<td>Kettunen (2003)</td>
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<tr>
<td>P11</td>
<td>$K_{H2prodCH4}$</td>
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<td>0.01-0.1</td>
<td>mol m$^{-3}$</td>
<td>half saturation coefficient</td>
<td>Kettunen (2003)</td>
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<tr>
<td>P12</td>
<td>$K_{CO2prodCH4}$</td>
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<td>0.01-0.1</td>
<td>mol m$^{-3}$</td>
<td>half saturation coefficient</td>
<td>Kettunen (2003)</td>
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<tr>
<td>P13</td>
<td>$K_{AceprodCH4}$</td>
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<td>0.01-0.1</td>
<td>mol m$^{-3}$</td>
<td>half saturation coefficient</td>
<td>Kettunen (2003)</td>
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<tr>
<td>P14</td>
<td>$K_{CH4prod}$</td>
<td>0.5</td>
<td>0.3-0.7</td>
<td>mol mol$^{-1}$</td>
<td>CH$_4$ production ratio</td>
<td>Kettunen (2003)</td>
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<tr>
<td>P15</td>
<td>$K_{CH4oxidCH4}$</td>
<td>0.05</td>
<td>0.01-0.1</td>
<td>mol m$^{-3}$</td>
<td>half saturation coefficient</td>
<td>Kettunen (2003)</td>
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<tr>
<td>P16</td>
<td>$K_{CH4oxidO2}$</td>
<td>0.02</td>
<td>0.01-0.1</td>
<td>mol m$^{-3}$</td>
<td>half saturation coefficient</td>
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<tr>
<td>P17</td>
<td>$f_{D, w}$</td>
<td>0.8</td>
<td>0.7-0.9</td>
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<td>reduction factor for diffusion in water-filled peat</td>
<td>Raivonen et al. (2017)</td>
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<tr>
<td>P18</td>
<td>$f_{D, a}$</td>
<td>0.5</td>
<td>0.7-0.9</td>
<td></td>
<td>reduction factor for diffusion in air-filled peat</td>
<td>Raivonen et al. (2017)</td>
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<tr>
<td>P19</td>
<td>$\tau$</td>
<td>1.5</td>
<td>1-2</td>
<td></td>
<td>root tortuosity</td>
<td>Stephen et al. (1998)</td>
</tr>
<tr>
<td>P20</td>
<td>$a_{inA}$</td>
<td>0.085</td>
<td>0.01-0.1</td>
<td>m$^2$ kg$^{-1}$</td>
<td>root ending area per root dry biomass</td>
<td>Stephen et al. (1998)</td>
</tr>
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<td>P21</td>
<td>SLA</td>
<td>20</td>
<td>15-25</td>
<td>m$^2$ kg$^{-1}$</td>
<td>specific leaf area</td>
<td>Raivonen et al. (2017)</td>
</tr>
<tr>
<td>P22</td>
<td>$\sigma$</td>
<td>0.8</td>
<td>0.7-0.9</td>
<td></td>
<td>peat porosity</td>
<td>Raivonen et al. (2017)</td>
</tr>
<tr>
<td>P23</td>
<td>$k$</td>
<td>0.001</td>
<td>0.0001-0.0003</td>
<td>d$^{-1}$</td>
<td>time constant of ebullition</td>
<td>Raivonen et al. (2017)</td>
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<tr>
<td>P24</td>
<td>$K_{aer}$</td>
<td>0.1</td>
<td>0.1-0.2</td>
<td>mol m$^{-3}$ d$^{-1}$</td>
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<td>P25</td>
<td>$K_{aerDOC}$</td>
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<td>5-15</td>
<td>mol m$^{-3}$</td>
<td>half saturation coefficient</td>
<td>Grant (1998)</td>
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<td>P26</td>
<td>$K_{aerO2}$</td>
<td>0.22</td>
<td>0.01-0.3</td>
<td>mol m$^{-3}$</td>
<td>half saturation coefficient</td>
<td>Grant (1998)</td>
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<td>P27</td>
<td>DOCprod$Q_{10}$</td>
<td>2.5</td>
<td>1-5</td>
<td></td>
<td>temperature sensitivity of DOC production</td>
<td>Kettunen (2003)</td>
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<td>P28</td>
<td>Aceprod$Q_{10}$</td>
<td>2.5</td>
<td>1-5</td>
<td></td>
<td>temperature sensitivity of acetate production</td>
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<td>P29</td>
<td>CH4prod$Q_{10}$</td>
<td>2.5</td>
<td>1-5</td>
<td></td>
<td>temperature sensitivity of CH$_4$ production</td>
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<td>P30</td>
<td>CH4oxid$Q_{10}$</td>
<td>2.5</td>
<td>1-5</td>
<td></td>
<td>temperature sensitivity of CH$_4$ oxidation</td>
<td>Kettunen (2003)</td>
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Table 2. Description of sites in boreal regions.

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<th>Site Name</th>
<th>Location</th>
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<th>Dominant Vegetation</th>
<th>Time</th>
<th>Method</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Zackenberg, Greenland</td>
<td>74°30′ N, 21°00′ W</td>
<td>Arctic Fen</td>
<td>Eriophorum scheuchzeri, Carex stans and Dupontia psilosantha</td>
<td>2006-2010</td>
<td>Automatic chamber</td>
<td>Mastepanov et al. (2013)</td>
</tr>
<tr>
<td>3</td>
<td>Lompolojänkkä, Finland</td>
<td>67°59.8′ N, 24°12.5′ E</td>
<td>Open, nutrient-rich sedge fen</td>
<td>Betula nana, Menyanthes trifoliata, Salix lapponum and Carex ssp.</td>
<td>2006-2010</td>
<td>Eddy covariance</td>
<td>Raivonen et al. (2017)</td>
</tr>
<tr>
<td>4</td>
<td>Stordalen, Sweden</td>
<td>68°21′ N, 19°02′ E</td>
<td>Subarctic mire</td>
<td>Eriophorum angustifolium</td>
<td>2004-2006</td>
<td>Automatic chamber and Eddy covariance</td>
<td>Petrescu et al. (2008)</td>
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<tr>
<td>5</td>
<td>Degero Stormyr, Sweden</td>
<td>64°11′ N, 19°33′ E</td>
<td>Mixed acid mire</td>
<td>Eriophorum vaginatum, Vaccinium oxycoccos, Andromeda polifolia and Scheuchzeria palustris</td>
<td>1995-1997</td>
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<td>6</td>
<td>Lena Samoylov Delta, Russia</td>
<td>72°22′ N, 126°30′ E</td>
<td>Tundra</td>
<td>Sedges, Mosses and Shrubs</td>
<td>2003-2004</td>
<td>Eddy covariance</td>
<td>Wille et al. (2008)</td>
</tr>
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<td>7</td>
<td>Bakchar, Russia</td>
<td>56°51′ N, 82°50′ E</td>
<td>Open unforested mesotrophic fen</td>
<td>Eriophorum vaginatum, Carex rostrata, C. limosa and horse-tail Equisetum fluviatile</td>
<td>1999</td>
<td>Static chamber</td>
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<tr>
<td>8</td>
<td>Manitoba, Canada</td>
<td>58°40′ N, 93°50′ W</td>
<td>Eutrophic fen</td>
<td>Sedges, Grasses and Moss</td>
<td>2008-2011</td>
<td>Eddy covariance</td>
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<tr>
<td>9</td>
<td>Athabasca, Canada</td>
<td>54°57′ N, 112°28′ W</td>
<td>Moderately rich treed fen</td>
<td>Stunted trees, Shrub and Mosses</td>
<td>2007</td>
<td>Eddy covariance</td>
<td>Long et al. (2010)</td>
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<tr>
<td>10</td>
<td>Quebec, Canada</td>
<td>53°40′ N, 78°10′ W</td>
<td>Boreal bog</td>
<td>Lichen, Mosses and Shrubs</td>
<td>2012</td>
<td>Eddy covariance</td>
<td>Nadeau et al. (2013)</td>
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<td>11</td>
<td>Robinsons, Canada</td>
<td>48°16′ N, 58°40′ W</td>
<td>Boreal bog</td>
<td>Mosses, Lichens and Sedge</td>
<td>2014-2016</td>
<td>Eddy covariance</td>
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Table 3. Description of sites in temperate regions.

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<th>Wetland Type</th>
<th>Dominant Vegetation</th>
<th>Time</th>
<th>Method</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Duxinganling,</td>
<td>51°7.92′N, 125°8.22′E</td>
<td>Intermittently inundated marsh</td>
<td>Carex meyerian (#1)(^a)</td>
<td>2011-2012</td>
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<td>Liu et al. (2015)</td>
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<td>China</td>
<td>52°56′N, 122°51′E</td>
<td>Permafrost peatland</td>
<td>Shrubs, Sedges and Grass (#2)</td>
<td>2012-2013</td>
<td>Eddy covariance</td>
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<td>2</td>
<td>Sanjiang Plain,</td>
<td>47°53′N, 133°30′E</td>
<td>Permanently inundated marsh</td>
<td>Glyceria spiculosa (#1) and Deyeuxia angustifolia (#2)</td>
<td>2011</td>
<td>Static chamber</td>
<td>Sun et al. (2013)</td>
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<tr>
<td></td>
<td>China</td>
<td>47°35′N, 133°30′E</td>
<td>Permanently inundated marsh</td>
<td>Carex lasiocarpa and Deyeuxia angustifolia (#3)</td>
<td>2011</td>
<td>Eddy covariance</td>
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<td>Luanhaizi, China</td>
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<td>Static chamber</td>
<td>Ding et al. (2004)</td>
</tr>
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<td>Minnesota, USA</td>
<td>47°32′N, 93°28′W</td>
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<td>Sphagnum, Carex and Scheuchzeria palustris (#1)</td>
<td>1988-1990</td>
<td>Static chamber</td>
<td>Dise (1993)</td>
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<tr>
<td></td>
<td>47°30′N, 93°29′W</td>
<td>Peatland</td>
<td></td>
<td>Sphagnum papillosum, Carex spp., Eriophorum chamissonis and Saracenia purpurea (#2)</td>
<td>2009-2011</td>
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<td>6</td>
<td>Sallies Fen, USA</td>
<td>43°12.5′N, 71°3.5′W.</td>
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<td>Mosses, Shrubs and Sedges</td>
<td>2008-2011</td>
<td>Static chamber</td>
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<td>Michigan, USA</td>
<td>42°27′N, 84°01′W</td>
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<td>Sphagnum and Scheuchzeria palustris</td>
<td>1991-1993</td>
<td>Static chamber</td>
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<tr>
<td>8</td>
<td>Mer Bleue, Canada</td>
<td>45°41′N, 75°52′W</td>
<td>Temperate ombrotrophic bog</td>
<td>Chamaedaphne (#1), Eriophorum (#2) and Maianthemum (#3)</td>
<td>2009-2010</td>
<td>Triplicate auto chamber</td>
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<td></td>
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<td>Sphagnum spp., Ericaceous shrubs and Judd. Sedges (#4)</td>
<td>2011-2012</td>
<td>Eddy covariance</td>
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<tr>
<td>9</td>
<td>Federsee Moor,</td>
<td>48°06′N, 9°38′E</td>
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<td>Eddy covariance</td>
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<td>Germany</td>
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<td></td>
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<td></td>
</tr>
<tr>
<td>10</td>
<td>Schechenfilz,</td>
<td>47°48′N, 11°19′E</td>
<td>Temperate bog-pine forest</td>
<td>Bog-pines and Mosses</td>
<td>2012-2013</td>
<td>Eddy covariance</td>
<td>Hommeltenberg et al. (2014)</td>
</tr>
<tr>
<td>12</td>
<td>Kopuatai, New Zealand</td>
<td>37°56′S, 175°22′E</td>
<td>Ombrotrophic, raised bog</td>
<td>Wire rush and Empodisma robustum</td>
<td>2012-2014</td>
<td>Eddy covariance</td>
<td>Goodrich et al. (2015)</td>
</tr>
</tbody>
</table>

\(^a\)The observed site with corresponding dominant vegetation.
Table 4. Description of sites in tropical regions.

<table>
<thead>
<tr>
<th>No.</th>
<th>Site Name</th>
<th>Location</th>
<th>Wetland Type</th>
<th>Dominant Vegetation</th>
<th>Time</th>
<th>Method</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Sarawak, Malaysia</td>
<td>1°25′N, 111°07′E</td>
<td>Tropical peat swamp</td>
<td>Gonystlus bancanus, Dactylocladus stenostachys and Copaifera palustris (#1)</td>
<td>2012-2015</td>
<td>Closed chamber</td>
<td>Sangok et al. (2017)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>forest</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>1°27′N, 111°09′E</td>
<td>Tropical peat swamp</td>
<td>Shorea albida, Gonystlus bancanus, and Stemonurus spp. (#2)</td>
<td>2013</td>
<td>Eddy covariance</td>
<td>Tang et al. (2018)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>forest</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Shorea albida, Lithocarpus sp., Litsea sp. and Dillenia sp. (#3)</td>
<td>2014-2015</td>
<td>Eddy covariance</td>
<td>Wong et al. (2018)</td>
</tr>
</tbody>
</table>
Table 5. Statistic results for site-level evaluation.

<table>
<thead>
<tr>
<th>Site</th>
<th>R²</th>
<th>RMSE</th>
<th>RPE (%)</th>
<th>N</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Boreal sites</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Zackenberg</td>
<td>0.210</td>
<td>0.0019</td>
<td>-1.39</td>
<td>192</td>
</tr>
<tr>
<td>Siikaneva</td>
<td>0.517</td>
<td>0.0017</td>
<td>14.54</td>
<td>2285</td>
</tr>
<tr>
<td>Lompolojänkkä</td>
<td>0.815</td>
<td>0.0018</td>
<td>2.80</td>
<td>837</td>
</tr>
<tr>
<td>Stordalen</td>
<td>0.317</td>
<td>0.0057</td>
<td>17.38</td>
<td>126</td>
</tr>
<tr>
<td>Degero Stormyr</td>
<td>0.361</td>
<td>0.0028</td>
<td>-19.47</td>
<td>23</td>
</tr>
<tr>
<td>Lena Samoylov Delta</td>
<td>0.053</td>
<td>0.0007</td>
<td>15.95</td>
<td>79</td>
</tr>
<tr>
<td>Bakchar</td>
<td>0.586</td>
<td>0.0028</td>
<td>4.38</td>
<td>103</td>
</tr>
<tr>
<td>Manitoba</td>
<td>0.012</td>
<td>0.0030</td>
<td>30.08</td>
<td>127</td>
</tr>
<tr>
<td>Athabasca</td>
<td>0.563</td>
<td>0.0010</td>
<td>4.38</td>
<td>103</td>
</tr>
<tr>
<td>Quebec</td>
<td>0.455</td>
<td>0.0007</td>
<td>10.76</td>
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</tr>
<tr>
<td>Robinsons</td>
<td>0.313</td>
<td>0.0006</td>
<td>33.28</td>
<td>329</td>
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<tr>
<td><strong>Temperate sites</strong></td>
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<td></td>
</tr>
<tr>
<td>Daxinganling1</td>
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<td>0.0005</td>
<td>12.31</td>
<td>38</td>
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<tr>
<td>Daxinganling2</td>
<td>0.272</td>
<td>0.0002</td>
<td>15.38</td>
<td>242</td>
</tr>
<tr>
<td>Sanjiang1</td>
<td>0.258</td>
<td>0.0030</td>
<td>13.46</td>
<td>54</td>
</tr>
<tr>
<td>Sanjiang2</td>
<td>0.000</td>
<td>0.0025</td>
<td>-2.79</td>
<td>57</td>
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<tr>
<td>Sanjiang3</td>
<td>0.379</td>
<td>0.0032</td>
<td>-6.27</td>
<td>119</td>
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<tr>
<td>Sanjiang4</td>
<td>0.413</td>
<td>0.0027</td>
<td>7.04</td>
<td>142</td>
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<tr>
<td>Luanhaizì</td>
<td>0.170</td>
<td>0.0025</td>
<td>-26.70</td>
<td>133</td>
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<tr>
<td>Ruoergai1</td>
<td>0.118</td>
<td>0.0029</td>
<td>20.29</td>
<td>35</td>
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<tr>
<td>Ruoergai2</td>
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<td>0.0039</td>
<td>-19.26</td>
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<td>Minnesota1</td>
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<td>0.0057</td>
<td>9.13</td>
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<td>Minnesota2</td>
<td>0.506</td>
<td>0.0033</td>
<td>4.22</td>
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<tr>
<td>Sallies Fen</td>
<td>0.240</td>
<td>0.0052</td>
<td>-28.70</td>
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<tr>
<td>Michigan</td>
<td>0.142</td>
<td>0.0147</td>
<td>-27.30</td>
<td>62</td>
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<tr>
<td>Mer Bleue1</td>
<td>0.083</td>
<td>0.0018</td>
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<tr>
<td>Mer Bleue2</td>
<td>0.498</td>
<td>0.0034</td>
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<tr>
<td>Mer Bleue3</td>
<td>0.231</td>
<td>0.0028</td>
<td>-28.99</td>
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<tr>
<td>Mer Bleue4</td>
<td>0.000</td>
<td>0.0012</td>
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<tr>
<td>Federseemoor</td>
<td>0.252</td>
<td>0.0043</td>
<td>-3.55</td>
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<tr>
<td>Schechenfilz</td>
<td>0.231</td>
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<td>32.64</td>
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<tr>
<td>Kopytkowo</td>
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<td>0.0060</td>
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<tr>
<td>Kopuatai</td>
<td>0.001</td>
<td>0.0041</td>
<td>-4.97</td>
<td>124</td>
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<tr>
<td><strong>Tropical sites</strong></td>
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<tr>
<td>Sarawak1</td>
<td>0.016</td>
<td>0.0003</td>
<td>47.04</td>
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</tr>
<tr>
<td>Sarawak2</td>
<td>0.003</td>
<td>0.0010</td>
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<td>61</td>
</tr>
<tr>
<td>Sarawak3</td>
<td>0.426</td>
<td>0.0005</td>
<td>6.25</td>
<td>18</td>
</tr>
</tbody>
</table>

bThe unit of RMSE is mol m⁻² d⁻¹.
Figure 2. Daily variation in simulated and observed CH$_4$ emissions at boreal sites. The blue solid lines represent simulated values and the red cross dots represent observed values.
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.
**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. Comparison of the average observed and simulated CH$_4$ emissions (red dots). (a) all evaluated sites, (b) boreal sites, and (c) temperate sites.
Figure 6. Parameter sensitivity indices for modeled CH$_4$ emissions. (a) First-order Sobol indices, (b) Total Sobol indices. All parameters are listed in Table 1.
Figure 7. The second-order Sobol indices for modeled CH$_4$ emissions. All parameters are listed in Table 1.
Figure 8. Time series of simulated CH₄ production, oxidation and transportation.
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.