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Sensitivity of wetland methane emissions to model assumptions: application and model testing against site observations

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Abstract

Methane emissions from natural wetlands and rice paddies constitute a large proportion of atmospheric methane, but the magnitude and year-to-year variation of these methane sources is still unpredictable. Here we describe and evaluate the integration of a methane biogeochemical model (CLM4Me; Riley et al., 2011) into the Community Land Model 4.0 (CLM4CN) in order to better explain spatial and temporal variations in methane emissions. We test new functions for soil pH and redox potential that impact microbial methane production in soils. We also constrain aerenchyma in plants in always-inundated areas in order to better represent wetland vegetation. Satellite inundated fraction is explicitly prescribed in the model because there are large differences between simulated fractional inundation and satellite observations. A rice paddy module is also incorporated into the model, where the fraction of land used for rice production is explicitly prescribed. The model is evaluated at the site level with vegetation cover and water table prescribed from measurements. Explicit site level evaluations of simulated methane emissions are quite different than evaluating the grid cell averaged emissions against available measurements. Using a baseline set of parameter values, our model-estimated average global wetland emissions for the period 1993–2004 were 256 Tg CH₄ yr⁻¹, and rice paddy emissions in the year 2000 were 42 Tg CH₄ yr⁻¹. Tropical wetlands contributed 201 Tg CH₄ yr⁻¹, or 78 % of the global wetland flux. Northern latitude (> 50 N) systems contributed 12 Tg CH₄ yr⁻¹. We expect this latter number may be an underestimate due to the low high-latitude inundated area captured by satellites and unrealistically low high-latitude productivity and soil carbon predicted by CLM4. Sensitivity analysis showed a large range (150–346 Tg CH₄ yr⁻¹) in predicted global methane emissions. The large range was sensitive to: (1) the amount of methane transported through aerenchyma, (2) soil pH (± 100 Tg CH₄ yr⁻¹), and (3) redox inhibition (± 45 Tg CH₄ yr⁻¹).

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1 Introduction

Methane (CH₄) is an important greenhouse gas and has made approximately a 12 ~ 15 % contribution to global warming (IPCC, 2007). Its atmospheric concentration has increased continuously since 1800 (Chappellaz et al., 1997; Etheridge et al., 1998; Rigby et al., 2008) with a relatively short period of decreases during 1999–2002 (Dlugokencky et al., 2003). Wetlands are the single largest source of atmospheric CH₄, although their estimated emissions vary from 80 to 260 Tg CH₄ annually (Matthews and Fung, 1987; Bartlett et al., 1990; Hein et al., 1997; Walter et al., 2001; Whalen, 2005). In addition, the spatial distribution of methane emissions from wetlands is still unclear. For instance, some studies suggest that tropical regions (20 N–30 S) release about 60 % of the total wetland emissions (Bartlett et al., 1990; Bartlett and Harriss, 1993), whereas other studies argue that northern wetlands contribute as much as 60 % of the total emissions (Matthews and Fung, 1987). For tropical regions, methane emissions are highly uncertain because (1) tropical wetlands have a large area (Matthews and Fung, 1987; Aselmann and Crutzen, 1989; Page et al., 2011) that fluctuates seasonally and (2) methane fluxes vary significantly across different wetland types (Nahlik and Mitsch, 2011). Rice paddies are human-made wetlands and are one of the largest anthropogenic sources of atmospheric methane. Methane emission rates from rice paddies have been estimated to be 20 to 120 Tg CH₄ yr⁻¹ (Yan et al., 2009) with an average of 60 Tg CH₄ yr⁻¹ (Wuebbles and Hayhoe, 2002; Denman et al., 2007). Together, rice paddies and wetlands can release 100–380 Tg CH₄ yr⁻¹ to the atmosphere. Further, recent studies identified a new source of tropical methane from non-wetland plants that could add as much as 10–60 Tg CH₄ yr⁻¹ to the global budget (Keppler et al., 2006; Kirschbaum et al., 2006), although this source has been disputed and is still poorly quantified (Dueck et al., 2007).

Process-based methane emission models have been previously used to estimate the global methane budget (Cao et al., 1996; Christensen et al., 1996; Potter, 1997; Walter et al., 2001; Zhuang et al., 2004; Wania et al., 2010). Due to the complexity

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of wetland systems and the paucity of field and laboratory measurements to constrain process representations, these models used different approaches to simulate the methane emissions. Zhuang et al. (2004) coupled a methane module to a process-based biogeochemistry model, the Terrestrial Ecosystem Model (TEM), with explicit calculation of methane production, oxidation, and transport in the soil and to the atmosphere. Walter et al. (2001) integrated a process-based methane model with a simple hydrologic model to estimate methane emissions from wetlands with external forcing of net primary production. Cao et al. (1996) developed a methane model based on substrate supply by plant primary production and organic matter degradation. The most recent methane model developed by Wania et al. (2010) is fully coupled into a global dynamic vegetation model designed specifically to simulate northern peatlands. This model avoids the use of some empirical relationships and parameters (such as the Q_{10} temperature-dependence) used previously. As discussed above, these models parameterize the biogeochemical processes and hydrological processes in different ways and use different inputs (e.g., inundated area and NPP). Thus, it is not surprising that they produce a large range of emissions for the global methane budget. For instance, Cao et al. (1996) estimated the global methane emissions from wetlands to be $92 \text{ Tg CH}_4 \text{ yr}^{-1}$ while Walter et al. (2001) calculated an emission of $260 \text{ Tg CH}_4 \text{ yr}^{-1}$ from global wetlands. This large range indicates a high degree of uncertainty in the global methane budget. Here we attempt to understand this uncertainty and the sources of this uncertainty by driving a complex process-based biogeochemical model with multiple observational constraints.

Here, and in a related article (Riley et al., 2011), we describe a process-based methane model that simulates the physical and biogeochemical processes regulating terrestrial methane fluxes. Specifically, we include physical and biogeochemical processes related to soil, hydrology, microbes and vegetation that account for microbial methane production, methane oxidation, methane and oxygen transport through aerenchyma of wetland plants, ebullition, and methane and oxygen diffusion through soil. The integration of processes into CLM4CN (called CLM4Me) has been described

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in detail by Riley et al. (2011). Although CLM4Me can be operated as part of a fully-coupled carbon-climate-chemistry model, here we force the global methane emission model with the best available information for the current climate, including satellite derived inundation fraction (Prigent et al., 2007; Papa et al., 2010), rice paddy fraction (Portmann et al., 2010), soil pH, and observed meteorological forcing (Qian et al., 2006). In contrast to the initial description of CLM4Me (Riley et al., 2011), we used satellite derived inundation, evaluated a new soil pH parameterization, and evaluated the predicted methane fluxes at wetland and rice paddy sites against site-level model simulations. We then extended our parameterization to the global scale and estimated the terrestrial methane flux and its sensitivities to model parameterization choices.

In this paper, Sect. 2 describes several new features of this model beyond those originally described in Riley et al. (2011). The data sets used to drive the model are described in Sect. 3. Model validation and comparisons with observations as well as sensitivity analysis are presented in Sect. 4. Discussion of the global methane flux is presented in Sect. 5 and conclusions are in Sect. 6.

2 Model descriptions and modifications

The methane biogeochemical component of CLM4 (CLM4Me) is composed of four processes: methane production, methane oxidation, methane ebullition, methane transport through wetland plant aerenchyma, and methane diffusion through soil. In CLM4Me, production of CH_4 below the water table ($P \text{ mol C m}^{-2} \text{ s}^{-1}$) is related to the gridcell estimate of heterotrophic respiration from soil and litter ($R_{\text{H}} \text{ (mol C m}^{-2} \text{ s}^{-1})$), soil temperature (Q'_{10}), pH (f_{pH}), redox potential (f_{pE}), and a factor accounting for the portion of the gridcell that is seasonally inundated (S):

$$P = R_{\text{H}} f_{\text{CH}_4} Q'_{10} S f_{\text{pH}} f_{\text{pE}} \quad (1)$$

Here, f_{CH_4} is the ratio between CO_2 and CH_4 production which is currently set to 0.2 for wetlands and rice paddies. We constrain the model simulations to observations by

using satellite inundation, pH, and temperature datasets. In CLM4Me, f_{pH} and are f_{pE} set to 1. The pH and redox potential functions and other modifications from CLM4Me are described in detail in the following subsections, and together are referred to as CLM4Me'.

5 2.1 Soil pH effects on methanogenesis

Soil pH has an important control on methane production with maximum rates at neutral pH conditions (Conrad and Schutz, 1988; Minami, 1989; Dunfield et al., 1993; Wang et al., 1993; Zhuang et al., 2004). We used the data from Dunfield et al. (1993) to develop a new soil pH function (f_{pH}):

$$10 f_{\text{pH}} = 10^{-0.2335 \text{pH}^2 + 2.7727 \text{pH} - 8.6} \quad (2)$$

The maximum methane production occurs at $\text{pH} \sim 6.2$ (Fig. 1). Compared with other functions used to specify the pH dependence of methane emissions (Cao et al., 1995; Zhuang et al., 2004), the advantage of this new pH function is that it allows for small but finite methane production at acidic pH. Several studies have shown that methane can be produced in acidic conditions, e.g., at pH of 4.0 in northern bogs (Williams and Crawford, 1985; Valentine et al., 1994). Another difference between our function and that in Cao et al. (1995) is the optimal pH for methanogenesis, which is 7.5 in Cao et al. (1995) and 6.2 here.

2.2 Redox potential effects on methanogenesis

20 Methane is produced in anoxic soils only when all oxidized species such as NO_3^- , Fe(III) , and SO_4^{2-} are consumed because these chemical species fuel microbial activities at the expense of methanogenesis (Lovley and Phillips, 1987). Theoretically, methane production occurs only when redox potentials (E_{h}) in soil are below -200 mV (Neue et al., 1990; Wang et al., 1993). E_{h} reflects the abundance of alternative electron

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acceptors (such as O_2 , NO_3^- , Fe^{+3} , Mn^{4+} , SO_4^{2-}) which can suppress methanogenesis through the reduction of H_2 (Conrad, 2002) and supply more energy than available through methanogenesis (Zehnder and Stumm, 1988). Once these alternative electron acceptors have been depleted, H_2 will increase to a level that methanogens can use to produce methane. The duration of suppression of the alternative electron acceptors on methanogenesis will depend on their concentrations in soils and availability of acetate and H_2 . The effect of redox potential has been incorporated in several previous methane models (e.g., Zhuang et al., 2004; Zhang et al., 2002; Li et al., 1999). For instance, Zhuang et al. (2004) calculated E_h based on the status of soil saturation assuming that O_2 is the dominant alternative electron acceptor that suppresses methanogenesis. Li et al. (1999) developed a simple dynamic model to estimate soil redox potential based on soil oxygen pressure which is calculated through soil oxygen diffusion and consumption. In submerged soil, reducible Fe (III) is one of the most abundant electron acceptors. Studies have suggested that methane production will not occur until a significant amount of Fe (III) has been reduced to Fe (II) (Conrad, 2002; Cheng et al., 2007). Based on laboratory experiments, Cheng et al. (2007) developed an empirical model to include soil chemical properties (such as available N and Fe (II)) in predicting methane emissions from Japanese rice paddy soils. They showed that methane production is significantly related to reducible Fe and decomposable C and found that methane production is delayed by 4–8 weeks for different types of soils due to the abundance of reducible Fe. Due to the lack of globally available datasets for reducible Fe and other species, we do not estimate the delay time on a spatially explicit basis.

Here we developed a simple parameterization of the effects of redox potential by assuming newly inundated wetlands will not produce methane initially because of the existing electron acceptors (such as O_2 , SO_4^{2-} , Fe^{3+} , etc) regenerated by O_2 prior to the flooding. As other electron acceptors are consumed following the flooding, the inundated fraction that can produce methane increases. We assumed a time constant of 30 days (the average time for other electron acceptors to be consumed) for the resumption

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of methane production. Such delayed impacts have been demonstrated in other studies (Lovley and Phillips, 1987; van Bodegom and Stams, 1999; Conrad, 2002; Cheng et al., 2007). We incorporated the redox potential into CLM4Me in inundated fraction and non-inundated fractions separately.

In the inundated fraction, we modified the inundation fraction that produces methane. In other words, we adjusted the fractional inundation in each grid cell to account for changing redox potential. Therefore, the redox potential factor f_{pE} in Eq. (1) is calculated as follows:

$$f_{i_lag}(t) = f_i(t) - f_{redox}(t) \quad (3)$$

$$f_{redox}(t) = f_i(t) - f_i(t-1) + f_{redox}(t-1) \cdot (1 - \Delta t / \tau) \quad (4)$$

$$f_{pE} = \frac{f_{i_lag}(t)}{f_i(t)} \quad (5)$$

where $f_i(t)$ is the fractional inundation, $f_{i_lag}(t)$ is the adjusted fractional inundation that is producing methane, $f_{redox}(t)$ is the fraction of grid cell where alternative electron acceptors (such as O_2 , NO_3^- , Fe^{+3}) are consumed (i.e., methane production is completely inhibited), Δt is the time step, and τ is the time constant currently set to 30 days. Thus $f_{redox}(t)$ is equal to the newly inundated fraction of land plus a relaxation of the previously inundated fraction to zero. These are new equations that we derived based on current understanding of the impact of redox potential on methane production. Figure 2 shows the adjusted fractional inundation (f_{i_lag}) against original fractional inundation.

In the non-inundated fraction, we estimated the delay in methane production as the water table depth increases by estimating an effective depth below which CH_4 production can occur (Z_{i_lag}):

$$Z_{i_lag}(t) = Z_i(t) - Z_{redox}(t) \quad (6)$$

$$Z_{redox}(t) = Z_i(t) - Z_i(t-1) + Z_{redox}(t-1) \cdot (1 - \Delta t / \tau) \quad (7)$$

where Z_{redox} is the depth of saturated water layer where alternative electron acceptors are consumed and Z_i is the actual water table depth. We then used Z_{i_lag} for methane

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production in the unsaturated portion in each grid cell. This approach is a simplification of the true dynamics of redox species concentrations and their impact on CH₄ production, which include vertical transport and multiple transformation processes. Future work in global-scale models should address this simplification.

2.3 Methane oxidation in the rhizosphere

In wetlands and rice paddies, plants develop aerenchyma to facilitate oxygen transport for root respiration and to support microbial activity in the soil-root rhizosphere. However, aerenchyma can also serve as conduits for methane to escape to the atmosphere (Colmer, 2003). Studies suggest that aerenchyma can be a dominant pathway for plant-mediated transfer of methane from soil to the atmosphere with up to 90% of the total methane emissions via transport in the aerenchyma from the rhizosphere (Cicerone and Shetter, 1981; Nouchi et al., 1990). While the methane is escaping through aerenchyma, some of it can be oxidized by the available oxygen. Therefore, rhizospheric methane oxidation can have a large control on global methane budgets. In CLM4Me, competition of root respiration and methanotrophy for the available oxygen determines the fraction of methane that is oxidized in the rhizosphere before being released into the atmosphere through aerenchyma. The balance between transport and oxidation depends on the availability of oxygen in the rhizosphere (Riley et al., 2011). The amount of O₂ that can be brought to the root depends on several factors including temperature, light intensity, water table change, and plant physiology (Whiting and Chanton, 1996; van der Nat and Middelburg, 1998). For instance, van der Nat and Middelburg (1998) investigated seasonal variation in rhizospheric methane oxidation of two common wetland plants (reed and bulrush) in a well-controlled environment and found that rhizospheric methane oxidation peaked during the early plant growth cycle and decreased after plants matured and root respiration decreased. We selected two sites where field-measured rhizospheric oxidation fractions were measured for comparison with model predictions. A sensitivity analysis was also conducted to characterize the impact of uncertainty in maximum oxidation fraction ($R_{o,max}$) on rhizospheric oxidation.

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2.4 Existence of aerenchyma in mostly inundated wetlands

In this study, we assumed that plant aerenchyma develop only in plants restricted to continuously-inundated land. Although, aerenchyma represent one adaptation to inundation, there are other differences between wetland plants and other plant types in their ability to deal with inundation. Studies suggest that some plants in dry land do not form aerenchyma (Voeselek et al., 1999), given the metabolic cost to construct and maintain tissue. Rather they adjust physiologically to seasonal flooding (Voeselek and Blom, 1989; Colmer, 2003). For instance, some cultivars of *Brassica napus* tend to develop new roots near the water surface in response to waterlogging (Daugherty et al., 1994; Voeselek et al., 1999). Because CLM4 does not have a wetland plant functional type (pft), the methodology adapted here is designed to improve our ability to simulate soil methane dynamics without adding a new wetland pft (which in the long term is a better solution). Here we define the fraction of continuously-inundated land (f_m) as the long-term (1993–2004) mean NPP flux weighted fractional inundation (f_i) at each grid cell:

$$f_m = \frac{\sum_i f_i \cdot \text{NPP}_i}{\sum_i \text{NPP}_i} \quad (8)$$

where NPP_i is the simulated average NPP at month i in the CLM4CN. To implement this feature into the model, at each gridcell we decrease plant aerenchyma area (T) evenly across all inundated area if current inundated fraction (f_i) is greater than f_m as follows:

$$T^* = T \cdot f_{\text{aere}} \quad (9)$$

$$f_{\text{aere}} = \min\left(1, \frac{f_m}{f_i}\right) \quad (10)$$

This new feature sets the limit of plants with aerenchyma to the mean inundated area and increases maximum aerenchyma area in plants when mean inundated fraction

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increases, which agrees with other studies that show the increase of aerenchyma in wetland plants in response to flooding (Fabbri et al., 2005; Kolb and Joly, 2009). However, this model feature may underestimate aerenchyma area in unflooded plants as formation of aerenchyma in some plants is not controlled by flooding conditions (Fabbri et al., 2005). This relationship only applies to natural wetlands since rice paddies are assumed to always be inundated in this study.

2.5 NPP-adjusted methane flux

Uncertainties in simulated methane fluxes could possibly come from errors associated with simulated NPP. By comparing with observation-based estimate NPP, we adjusted simulated methane fluxes and evaluated how improved NPP could increase the predictability of methane emissions. We applied the following equation to predict simulated methane flux (F_{CH_4}):

$$F'_{\text{CH}_4} = F_{\text{CH}_4} \frac{\text{NPP}_{\text{MODIS}}}{\text{NPP}_{\text{model}}} \quad (11)$$

where F'_{CH_4} is the NPP-adjusted daily methane flux ($\text{mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$), $\text{NPP}_{\text{MODIS}}$ is the annual mean NPP derived from MODIS, and $\text{NPP}_{\text{model}}$ is the annual mean NPP simulated in the CLM4CN. We applied this factor only to test the impact of substrate production uncertainty on methane emissions and not to modify our global emission estimates.

2.6 Modifications for rice paddies

In the model, the major differences between rice paddy and natural wetlands are that (1) rice paddies are treated as continuously inundated areas while natural wetlands are seasonally inundated and (2) we applied the crop PFT to represent rice, as the crop PFT is the closest to rice in CLM4. For wetland simulations, we used the spin-up described in Riley et al. (2011) to initialize model simulations. For rice paddy simulations,

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we used the year 2000 atmospheric forcing (Qian et al., 2006) with unlimited nitrogen to spin-up the CLM4 model offline simulation. We also assumed only one crop pft in each gridcell, so that the soil column would only contain rice; normally in CLM4, PFTs share a single soil column. This new spin-up is used to initialize the rice paddy simulation. In addition, only methane emissions from the inundated fraction in each gridcell are used to calculate the gridcell mean emissions in the rice paddy simulations. The methane emissions from non-inundated fraction were excluded when calculating gridcell mean emissions in rice paddy module.

2.7 Model setup for point and global simulations

We compared simulated methane emissions to site level observations by running the methane emission model in point simulations as well as at the global level. For point simulations, we used the atmospheric forcing data (Qian et al., 2006) from the overlapping grid cell. Then we spun-up the model for each site by running CLM4CN as a single-point model for more than 1000 yr until the soil carbon stabilized. For these single-point simulations, we did not consider the grid-cell averaged flux for the evaluation of our model. Instead, we calculated the methane emission fluxes from either the unsaturated or saturated portion of the grid cell depending on the local water table measurements at the site location. When the measured water table was above the surface we assumed the measured flux at the site was represented by the simulated flux in the saturated portion of the grid cell; when the measured water table is below the surface we assumed the measured flux is represented by the simulated flux in the unsaturated portion of the grid cell, where the simulated water table position is taken to be the monthly water table position at the measurement location. The imposed water table level is used for the methane-related calculation of anaerobicity, production, oxidation, etc., but does not include the expected impact of water table on soil temperature. For global wetland simulations, we used the spin-up described in Riley et al. (2011) to initialize an offline 1993–2004 run with observed meteorological forcing and evaluated the methane flux on a grid-cell averaged basis. In the global simulations

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the fraction of inundation was taken from the satellite measurements. For rice paddy simulations, we used the spin-up described in Sect. 2.6 to initialize an offline run for year 2000.

2.8 Calculation of rhizospheric methane oxidation fraction

In order to calculate the fraction of methane oxidized in the rhizosphere, we conducted two single-point simulations for each of the two sites with data on plant aerenchyma. One simulation assumed that all methane transported through aerenchyma from the rhizosphere was released into the atmosphere without loss (hereafter referred to as “NoLoss”), and the other considered methane oxidation loss in the rhizosphere before being emitted into the atmosphere (hereafter referred to as “WithLoss”). The rhizospheric methane oxidation fraction was computed as the ratio of calculated methane flux differences between NoLoss and WithLoss to methane flux that was transported through aerenchyma in NoLoss. This method for calculating rhizospheric oxidation is comparable to the way it was calculated in the field experiment. In our model, we assumed that vegetation communities at these two sites include significant amount of plants with aerenchyma.

2.9 Calculation of aerenchyma area

We also modified the Eq. (5) in Riley et al. (2011) to use fine root C instead of leaf area index in calculating aerenchyma area because fine root C calculated in CLM4-CN accounts for pft-specific and seasonal variations. This term better represents mass of tiller used in Wania et al. (2010) to calculate aerenchyma area. The equation is as follows:

$$T = \frac{F_{\text{rootc}}}{0.22} \pi R^2 \quad (12)$$

where F_{rootc} is pft-specific fine root Carbon (g C m^{-2}), R is the aerenchyma radius ($2.9 \times 10^{-3} \text{ m}$); and the 0.22 factor represents the amount of C per tiller. We will conduct

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a sensitivity analysis to test the impact of this change on global methane budget relative to that calculated using leaf area index.

3 Datasets

We used the datasets described below to force the methane emission model to the extent possible with observed data.

3.1 Global distributions of wetlands and rice cultivation fields

We used satellite inundation data (1993–2004) provided by Prigent et al. (2007) and Papa et al. (2010) to represent the extent of natural wetlands and to include seasonal and interannual variability in our global simulations. As discussed in Prigent et al. (2007), the satellite inundation does not discriminate among inundated wetlands and irrigated agriculture; therefore, we removed the irrigated agriculture from the satellite inundation by assuming rice cultivation areas were inundated agricultural land. Monthly mean distributions of rice cultivation areas compiled by Portmann et al. (2010) were used to define rice location and area. Irrigated, rain-fed, and deepwater rice (Kende et al., 1998) areas are included in the rice cultivation areas. Due to the lack of information on water management, draining, and re-flooding during the rice-growing season at the global scale, we assumed that rice fields were continuously flooded from the beginning of rice planting to the end of rice harvest. Overall, global coverage of rice paddies totals $1.67 \times 10^6 \text{ km}^2$, which is slightly larger than the areas estimated by Matthews and Fung (1991) and Asemann and Crutzen (1989), which are 1.47×10^6 and $1.3 \times 10^6 \text{ km}^2$, respectively. Rice growth areas peaked in July and August in this dataset (Fig. 3). Comparison of satellite-derived inundated areas with wetland extents compiled from other sources shows a large deficiency (Fig. 4). On average, satellite derived inundated areas in northern latitudes are $\sim 37\%$ and $\sim 45\%$ smaller than wetland extents compiled by Matthews and Fung (1987) (hereafter referred to as “MF”) and Aselmann and

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Crutzen (1989) (hereafter referred to as “AC”), respectively. The underestimation of the inundated area might be expected because satellites tend to underestimate small inland water bodies (inundated fraction less than 10 % of the pixels) that exist in high latitudes. Despite this weakness, the satellite-derived dataset provides a powerful tool to constrain methane emissions as it provides seasonal variations in inundated area that have large impacts on the seasonal variation in methane emissions (and will be discussed below). Satellite inundated areas are 36 % and 77 % larger than MF and AC wetland extents in temperate regions and are ~ 37 % and ~ 45 % smaller than MF and AC wetland extents in tropical regions, respectively. As demonstrated below, the assumption of wetland extent can result in large differences in simulated global methane fluxes.

3.2 Global soil pH datasets

Global soil pH datasets for this study are from the global soil data set of IGBP-DIS distributed by the International Soil Reference and Information Centre (Tempel et al., 1966) (<http://www.isric.org/>) (Fig. 5). The original sources of these datasets are from the combination of international soil reference and information center (ISRIC)'s soil information system (SIS) and CD-ROM of the Natural Resources Conservation Service (USDA-NRCS). The two datasets can be merged without issues of compatibility (Pleijzier, 1986). Note that this pH dataset does not necessarily represent wetland conditions, although soil pH is thought to be an important control on wetland pH (Magdoff and Bartlett, 1985). However, this is the only available global soil pH dataset. A site-level comparison between wetland pH at each measurement site and IGBP soil pH at the closest location is shown in Appendix A (Fig. A). The correlation between the two datasets is 0.69, with a root mean square error 1.07.

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3.3 Observed meteorological forcing

The observed meteorological forcing dataset that is provided with CLM4 extends from 1948 to 2004 at 3-hourly temporal and T62 (~ 1.875°) spatial resolution. The dataset is a combination of observed monthly precipitation and temperatures with model simulated intra-monthly variations from NCEP-NCAR 6-hourly reanalysis (Qian et al., 2006).

3.4 Rice paddies and wetland sites

A total of 11 rice paddy fields (Table 1) and 7 natural wetland sites (Table 2) were selected to test our model simulations. The rice paddy fields include sites in Italy, Chengdu (China), Nanjing (China), Japan, California (USA), Texas (USA), New Delhi (India), Cuttack (India), Beijing (China), Central Java (Indonesia), and Lampung (Indonesia). The common feature of the selected rice growing seasons at these sites was that there was no drainage until harvest. At each location, the flooding and drainage dates were provided in their corresponding references (Table 1). The pH values were set to 6.2 (optimal pH) when not available. The soil types on paddies are mainly loam and clay. These sites were chosen to cover major rice growing regions with a focus on Asia.

The wetland comparison includes sites in Panama, Indonesia, Florida, Minnesota, Michigan, Alberta (Canada), and Finland, covering the tropics, mid-latitudes, and high latitudes. Measured water table positions were integrated into the model to simulate methane emissions at these natural wetland sites (except the Panama site which used modeled water table positions). We assumed that soil was inundated below the water table. These wetland sites usually have peat soils with varying depths underlain by mineral soil. Methane is produced in the wetlands from litter and dead vegetation remnants in anoxic conditions. For these site-level comparisons, we used NCEP-NCAR reanalysis atmospheric forcing (including precipitation, temperature, wind speeds, and solar radiation) (Qian et al., 2006), pH from the site level measurement, and redox potential effects on production.

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4 Results: model testing and sensitivity analysis

Here we discuss the comparisons of the model against site-level observations. The selected wetland sites (Table 2) have varying water table positions obtained from measurements (except Panama where simulated water table was used). At the northern latitude sites, water table level will not control methane emissions during winter when the surface is frozen.

4.1 Net primary production (NPP)

The long-term annual mean NPP was derived from the MODerate Resolution Imaging Spectroradiometer (MODIS) and obtained from the Numerical Terradynamic Simulation Group (NTSG) (<http://www.ntsg.umd.edu>) (Zhao et al., 2005). At all sites, methane production in the model is dependent on the model simulation of the carbon cycle. One measure of carbon uptake is net primary productivity or NPP, which is calculated by CLM4CN. Measured and simulated NPP are highly correlated, although the simulated NPP tends to overestimate observations, particularly at higher levels of NPP (Fig. 6), consistent with previous comparisons (Randerson et al., 2009).

4.2 Methane oxidation fraction in the rhizosphere

Simulations suggest that the model tends to overestimate the magnitude of rhizospheric methane oxidation fraction at the two sites with measurements (Alberta, Canada and Florida, USA) (Fig. 7). With no change in aerenchyma transport there are three ways to decrease the rhizospheric methane oxidation in the model: 1) decrease the maximum oxidation fraction ($R_{o,max}$); 2) increase the CH_4 half-saturation oxidation coefficient (K_{CH_4}); and 3) increase the O_2 half-saturation oxidation coefficient (K_{O_2}). The values of these parameters are not well constrained and measurements generally vary over 2 orders of magnitude (Riley et al., 2011). We found that the simulated methane flux responded similarly to the three parameters and was most

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sensitive to $R_{o,max}$. Therefore, we focused on $R_{o,max}$ for our sensitivity analysis. We decreased $R_{o,max}$ from 1.25×10^{-5} to 1.25×10^{-6} , still within the estimated parameter uncertainty given in Riley et al. (2011), which led to a closer match of simulated rhizospheric methane oxidation fraction with observations (Fig. 7). We then tested the sensitivity of global methane budget to this parameter and applied this lower $R_{o,max}$ to the global simulation. The model estimated a 12% increase in global methane fluxes using the lower $R_{o,max}$ (Table 7). We also note that there is a spring peak in methane emission at Alberta (Canada) and Michigan (USA) sites in Fig. 7. A detailed description of this phenomenon is provided in Appendix B (Fig. B).

4.3 Impacts of pH on methane emission

There are three sites that have pH values more acidic than neutral conditions, allowing us to test our pH function against observed methane fluxes. In each case the site level pH is obtained from local measurements.

Soil pH plays an important role in constraining model simulations to the observations at several sites where soils are acidic (Fig. 8, Table 2). For example, at the Indonesian site, if we remove the pH impact, the model simulated methane emissions of $> 300 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ which is > 30 to 80 times larger than the measurements (approximately $10 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$). Soil pH is also an important control on methane emissions at Minnesota and Michigan sites. Removal of the pH factor at these sites increases the methane emissions by a factor of 4–5. Including the pH factor allows for better agreement with observations (Fig. 8). Table 3 shows that f_{pH} has reduced the RMSE at all sites, although, f_{pH} has negligible impacts on the ability to simulate the seasonal cycle (seen in the correlation coefficient) (Table 3). These results suggest that pH is an important control on regional methane budgets, and should be included in models to produce accurate spatial distribution and magnitudes of methane emission. A scatter plot of simulated annual mean fluxes with and without pH function against observations is provided in Appendix C (Fig. C).

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4.4 Impact of redox potential on methane emissions

Our simulations suggest that redox potential does not have substantial impacts on methane emissions at the sites where we have observations of water table levels (not shown). This low sensitivity is because of the relatively small changes in water table fluctuation at these sites (for detailed information see the description of each site given in the Table 2 references). At each individual site, the impact of redox potential on methane production is predominately through the change in water table levels. This dependence is different from the large-scale simulation where the impact of redox potential is largely seen through changes in the inundated fraction. In the large-scale simulation, the impact of redox potential in the unsaturated zone is through the change in water table levels and is negligible since very little methane is produced and released into the atmosphere. The redox potential factor does play an important role in large-scale methane emissions when the inundated fraction dramatically changes from season to season. Figure 9 shows the impact of redox potential on methane emission at a gridcell near Michigan extracted from a global CLM4 simulation. These simulations suggest that modeled methane emissions are reduced due to the fact that the inundated fraction that produces methane (f_{i_lag} , red dashed line) is much lower than the actual inundated fraction (f_i , blue dashed line). We want to emphasize that this proposed mechanism has not been tested against observations but matches theoretical expectations.

4.5 Site simulations: rice paddies

We simulated the rice paddies as single-gridcell cases and assumed that the fields were submerged during the simulation period between initial flooding and final drainage. In general, CLM4Me' as modified for rice paddies captures the magnitudes and temporal variations of methane emissions during the growing season (Fig. 10). In the model simulations, methane emissions have a large peak right after drainage in each simulation. This phenomena is consistent with the measurements at sites in

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California and Japan (Fig. 10d–f), but not at the other sites, possibly due to the duration and frequency of measurements (i.e., once a week). The sudden increase in simulated methane emissions immediately after drainage can be attributed to the release of methane previously trapped in the soil and water. This flush of methane has also been demonstrated in other studies (Wassmann et al., 1994; Jain et al., 2000). On a growing-season mean basis, the model performed relatively well for sites with observed mean fluxes less than $200 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$, and less well for sites with greater than a mean of $200 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ (Fig. 11a). Simulated maximum CH_4 emissions matched observations relatively well for sites with maximum daily fluxes less than $300 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$, but less well for sites with values greater than about $300 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ (Fig. 11b). For the latter sites, the model has a low bias.

4.6 NPP-adjusted methane fluxes

Scatter plots show that the NPP-based adjustment to simulated methane emissions only slightly increased the correlation with the measurements and did not improve the RMSE (Fig. 11). For instance, the correlation between modeled and observed mean fluxes increased from 0.5 to 0.61 using the NPP-based adjustment, primarily due to the adjustment at the Panama site (Fig. 11a). Overall, adjusting for NPP did not significantly improve model simulations at all other sites. This result suggests that the methane emission model biases are not just because of errors in the NPP.

4.7 Global simulations vs. observations

We note that our global simulations were forced with satellite inundation data and the same NCEP forcing data as used for the site simulations. To compare the global simulation against site level measurements, we extracted methane fluxes from the saturated portion of the closest grid cells to both the natural wetlands and rice paddies in the global simulation and compared with site level observations. This is the best comparison one can do usually for a global simulation (e.g., Riley et al., 2011), and

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thus a commonly used approach. We used methane fluxes from the saturated portion because they are very close to site-level conditions where the water table level is close to the surface, as is the case at most of the sites.

Comparison between mean methane fluxes in the global simulation and observations at sites shows a poor correlation ($r = 0.2$) (Fig. 12). Comparing with Fig. 11 suggests that the model's performance is worse in simulating the magnitude of methane fluxes when comparing grid-cell methane fluxes obtained from global simulations with point measurements. For instance, the correlation (r) decreased and the RMSE increased in Fig. 12. This result is not unexpected because of spatial heterogeneity and the large spatial resolution ($1.9^\circ \times 2.5^\circ$ resolution) used in the global simulation. We suggest that model should be validated at the site level if localized information is available, ideally forced by local vegetation characteristics, water table depth, and near-surface meteorology.

4.8 Sensitivity analysis at individual sites

Seven parameters were selected for sensitivity analysis (Table 4). The value for each parameter was varied from the lower end to the higher end of its range in the references listed in Table 4 to test its impacts on modeled methane emissions. The Panama site was selected for this analysis. The percentage change in annually averaged methane emission rate relative to the base simulation is listed in parenthesis in Table 5 for each parameter. The Q_{10} for production, f_{CH_4} , and the porosity of tillers have the most significant impacts on simulated methane emissions at this site. This result is consistent with the sensitivity analysis conducted by Wania et al. (2010) and Riley et al. (2011). The maximum oxidation rate ($R_{\text{o,max}}$) has a moderate impact on methane emissions. Other parameters, including K_{CH_4} , $Q_{\text{o,10}}$, K_{o2} , and $C_{\text{e,max}}$, have smallest influences on methane emissions. For instance, varying $C_{\text{e,max}}$ values within the range of current estimates negligibly affects methane emissions. Sensitivity analysis conducted at several other sites shows similar results (not shown).

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4.9 Sensitivity analysis on the global methane budget from natural wetlands

In this section, we focused our analysis on wetland emissions. For this sensitivity analysis, we conducted two year (1992–1993) simulations and used the second year for this analysis. We conducted the sensitivity analysis with the following parameters: soil pH (f_{pH}), redox potential (f_{pE}), and the limitation on aerenchyma area (f_{aere}). The processes these parameter impact have very different impacts on the global methane budget. We note that uncertainties in model structure and other model parameters listed in Table 4 could also have significant impacts on global emissions. These uncertainties have been discussed in Riley et al. (2011) and are not included in this paper.

In general, the inclusion of soil pH (f_{pH}) and redox potential (f_{pE}) decreased methane emissions. The limitation on aerenchyma area (f_{aere}) decreased methane oxidation, causing an increase in methane emissions. Model results suggest that the impacts of these factors on the global and regional methane budget vary (Fig. 13). Soil pH has the largest impacts on methane emissions. On the global scale, exclusion of soil pH in methane production ($f_{\text{pH}} = 1.0$) increased methane emissions by $100 \text{ Tg CH}_4 \text{ yr}^{-1}$, an approximate 41 % increase from the base simulation (Table 7). Removal of redox potential impacts ($f_{\text{pE}} = 1.0$) increases global methane emissions to $290 \text{ Tg CH}_4 \text{ yr}^{-1}$ (a 18 % increase from the base simulation). Unlimited aerenchyma ($f_{\text{aere}} = 1.0$) only decreased the global methane budget by 3%. At the regional scales, approximately 70 % of the global impacts of these factors occurred in the tropics (Fig. 13a), as tropical regions account for 80 % of the global methane wetland emissions and soil pH is generally low there (Fig. 5).

Our simulations suggest that the rhizospheric methane oxidation fraction is generally higher in temperate regions and lower in the tropics and high latitudes (Fig. 13b). The rhizospheric oxidation fraction is approximately 11.4 %, 25.4 %, and 23 % in the tropics, temperate, and high latitudes, respectively. On the global scale, ~ 15 % of methane was oxidized before being transported through aerenchyma and eventually being released to the atmosphere. Although aerenchyma is well known in grasses, some wetland trees

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also develop conduits (Grosse et al., 1992). The default value for aerenchyma in trees is set to be 17 % of that in grasses. Adjusting this proportion from 1 % to 35 % changed the methane flux by less than 25 Tg CH₄ yr⁻¹ (< 10 % of global methane budget).

4.10 Fine root carbon (FROOTC) vs. leaf area index (LAI)

5 Our modeling results suggest that simulated global methane budget is very sensitive to the way the aerenchyma area is calculated (Table 7). When the aerenchyma area was calculated based on FROOTC using Eq. (11) in this paper, the model's methane emissions are 245 Tg yr⁻¹. When LAI is used to calculate aerenchyma area, the methane emissions were 150 Tg yr⁻¹, an approximately 39 % decrease relative to FROOTC
10 method.

5 Estimation of global methane flux

5.1 Global simulations-wetlands

We estimated global wetland methane emissions of 256 Tg CH₄ yr⁻¹, which is close to the estimate of Walter et al. (2001), but higher than other estimates (Aselmann and Crutzen, 1989; Bartlett et al., 1990; Fung et al., 1991; Bartlett and Harriss, 1993) (Table 6). Figure 14 shows the spatial distribution of mean methane flux for the period
15 1993–2004 from natural wetlands. A comparison of the global methane emissions between CLM4Me' and other models is compared in Fig. 15 (Matthews and Fung, 1987; Aselmann and Crutzen, 1989; Bartlett et al., 1990; Bartlett and Harriss, 1993; Cao et al., 1996; Walter et al., 2001; Bousquet et al., 2006; Riley et al., 2011). The
20 CLM4Me' estimate is at the low end of current estimates for high latitude wetlands and at the high end for tropical and temperate wetlands.

Tropical wetlands released 201 Tg CH₄ yr⁻¹ to the atmosphere, comprising 78 % of the global wetland methane budget. This proportion is close to the estimate by

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Bartlett and Harriss (1993) who calculated global wetland CH_4 emissions using available methane flux measurements and the wetland areas compiled by MF and AC. This high proportion occurs even though mean satellite inundated areas in the tropics are 31 % and 39 % lower than the MF and AC wetland extents, indicating that the methane productivity in CLM4Me' is larger or oxidation is lower than other models. Higher tropical production may be partially attributed to the fact that CLMCN overestimates gross primary production over the tropical regions (Bonan et al., 2011). It is also demonstrated in the Panama site (one of the tropical sites) where accurate NPP could improve model estimation against observation (Fig. 11a). For middle-latitude regions, the CLM4Me' estimate is approximately 2 times larger than other process-based models (model 1–6 on Fig. 15) partially because satellite inundated areas are 48 % and 92 % larger than the MF and AC wetland extents used in other models.

CLM4Me' high latitude ($> 50^\circ \text{N}$) wetlands released $\sim 12 \text{ Tg CH}_4 \text{ yr}^{-1}$, which is much lower than other estimates. One of the primary reasons for the low high-latitude emissions could be that we assume a smaller inundation in the high latitudes, compared to other estimates (Fig. 4). Another reason is that CLM4CN under-predicts high latitude vegetation productivity and soil carbon storage (Lawrence et al., 2011). There is still considerable uncertainty in the wetland extent in the high latitudes (Finlayson et al., 1999; Papa et al., 2010), and the satellite inundated area we use may not capture all of the relevant wetland area (Prigent et al., 2007). Reducing the uncertainty associated with wetland extent might help further improve the estimation of the methane flux. For pan-arctic regions (North of 45°N), CLM4Me' estimated $15 \text{ Tg CH}_4 \text{ yr}^{-1}$ were released into the atmosphere. This value is lower than the estimates using various process-based models ($31\text{--}106 \text{ Tg CH}_4 \text{ yr}^{-1}$) (Cao et al., 1996; Walter et al., 2001; Zhuang et al., 2004; Wania et al., 2010) (Table 6), but is close to the estimate of Chen and Prinn (2006) in an inverse calculation. Wania et al. (2010) calculated an emission of $21.9\text{--}57.9 \text{ Tg CH}_4 \text{ yr}^{-1}$ from northern wetlands in Chen and Prinn's inverse model results assuming a methane uptake of $6.9 \text{ Tg CH}_4 \text{ yr}^{-1}$ (Chen and Prinn, 2006; Curry, 2009). Please note that our estimation includes a methane uptake in drylands.

Seasonal variations of methane emissions in the tropics, temperate climatic zones, and northern latitudes generally follow the pattern of the inundated areas measured by satellite (Fig. 14a,b). This relationship occurs because satellite inundated areas were used to derive methane emissions in each gridcell. As can be seen in Fig. 14a, seasonal variations between south and north of the equator have a different seasonality. Peak methane emissions occur in the rainy season, which is generally from June to October to the north of the equator and from October to March to the south of the equator. The seasonality of methane emissions in our model agrees well with that estimated in Cao et al. (1996) (their Fig. 3).

Even though our model simulated a low methane emission rate from northern latitudes ($> 50^\circ \text{N}$) in the summer, the seasonality of the satellite inundated areas is pronounced with maximum inundation in summer (Fig. 14b). The high inundated area in northern latitudes indicates that this region could potentially be a source of atmospheric methane that grows in importance because the duration and magnitude of methane production could increase as it experiences warming.

5.2 Global simulation of rice paddies

On average, our model estimates that global rice paddies emit approximately $42 \text{ Tg CH}_4 \text{ yr}^{-1}$ into the atmosphere, assuming no mid-season drainage. Our estimate is in the middle of current estimates of $26\text{--}120 \text{ Tg CH}_4 \text{ yr}^{-1}$ (Fig. 13c) (Seiler et al., 1984; Holzapfelschorn and Seiler, 1986; Bouwman, 1990; Sass, 1994; Cao et al., 1995, 1998; Scheehle et al., 2002; Wuebbles and Hayhoe, 2002; Olivier et al., 2005; Chen and Prinn, 2006; Yan et al., 2009). For instance, Cao et al. (1998) estimated global emissions from rice paddies to be $\sim 53 \text{ Tg CH}_4 \text{ yr}^{-1}$ using MF rice paddy areas. On the regional scale, CLM4Me' predicts $39 \text{ Tg CH}_4 \text{ yr}^{-1}$ is released from rice paddies in the Asian monsoon region ($10^\circ \text{S}\text{--}50^\circ \text{N}$, $65^\circ \text{E}\text{--}145^\circ \text{E}$) which contributes 92% of the global rice paddy methane emissions (Fig. 14d). This estimate agrees well with the most recent study using LPJ-WHyMe global dynamical vegetation model

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forced with fractional rice cover compiled by Leff et al. (2004) (Spahni et al., 2011). Chinese rice paddies release more CH_4 than any other country. In our model, Chinese rice paddies released $\sim 10 \text{ Tg CH}_4 \text{ yr}^{-1}$, which is in the middle of other estimates ($7 \sim 17 \text{ Tg CH}_4 \text{ yr}^{-1}$) derived using agricultural activity data and field measurements (Matthews et al., 2000; Li et al., 2004; Yan et al., 2009; Kai et al., 2010). For instance, Yan et al. (2009) estimated the methane emissions from Chinese rice paddies to be $7.41 \text{ Tg CH}_4 \text{ yr}^{-1}$ using the IPCC 2006 guidelines for national greenhouse gas inventories and methane emissions from rice paddies, and agricultural activity data for 2000. Yan et al. (2003) estimated emissions of $7.67 \text{ Tg CH}_4 \text{ yr}^{-1}$ in 1995 for China using measurements and region-specific CH_4 emission factors. Recently, Kai et al. (2010) revised the Huang model to include the effect of fertilizer use and water management and found that methane emissions from Chinese rice fields peaked in 1982 with an emission of $\sim 11 \text{ Tg CH}_4 \text{ yr}^{-1}$ in 2000. This estimate agrees with our estimates very well since we used the rice paddy fraction dataset developed by Portmann et al. (2010) for the year 2000.

Our model may overestimate methane emission from rice paddies for several reasons. First, we assumed continuous flooding during the growing season. Previous studies have suggested that mid-season drainages have been critical to reduce methane emissions in rice paddy fields. For instance, Yan et al. (2009) showed that one-time drainage in continuously flooded fields will reduce methane emissions by $4.1 \text{ Tg CH}_4 \text{ yr}^{-1}$ globally. Wassmann et al. (2000) suggested that mid-season drainage could reduce associated methane emissions by 7–80%. On average only about 30–40% of rice paddies experience continuous flooding (Yan et al., 2009). Simulation with one time drainage in August in our model (not shown) decreased methane emissions by $6 \text{ Tg CH}_4 \text{ yr}^{-1}$ globally, or by about 14% of the total methane emissions from rice paddies. This reduction is similar to the findings in other studies (Yan et al., 2009). Secondly, the increases in inorganic fertilizer use since 1982 also have contributed to a reduction in methane emissions (Kai et al., 2010), a feature not currently present in the model. Finally, our assumed rice cultivation area includes some rain-fed rice fields

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which might be only partially inundated. In order to accurately estimate rice paddy emissions, water management strategies (such as flooding and drainage), rice vegetation properties, and the use of fertilizers should be included in future methane models. Unfortunately, some of the required information is not readily available on the global scale.

6 Summary and conclusions

The purpose of this paper is to test several alternative parameterizations in CLM4Me, a global process-based biogeochemical methane model integrated with the Community Land Model version 4 (CLM4CN) (Riley et al., 2011) and to explicitly evaluate the model on the site level with measurements. To achieve this goal, we forced the model with explicit inundated fraction derived from satellites and other environmental variables (e.g., pH). We also tested several physically meaningful changes to the model, including the pH control on methane production, the limitation of aerenchyma to plants in mostly inundated areas, and a parameterization of redox potential. Site-level model comparisons to observations show that these changes improved the match between model simulations and observations at both wetland sites and rice paddies. We compiled data which allowed us to compare aerenchyma oxidation to simulations, and we used this data to adjust our maximum oxidation rate for sensitivity analysis. However, large sensitivities of the modeled emissions to other model parameters and sparse site-level observations make it difficult to be sure that these improvements are achieved via the right mechanisms. More data is needed to test alternative parameterization improvement options.

Our study suggests that models should be tested at the site level (not using global model simulations) when point measurements are used for evaluation. We only found limited data for tropical wetland methane fluxes, and unfortunately some datasets could not be used for the model evaluation exercise described here. For instance, many measurements on the Amazonian Basin cover a large area for a short period of time

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(Bartlett et al., 1990; Devol et al., 1990) and therefore temporal analysis could not be obtained between model simulations and measurements at site levels.

With our base parameterization choices, our model simulates an average annual methane flux over the period 1993–2004 of $256 \text{ Tg CH}_4 \text{ yr}^{-1}$ from natural wetlands.

This estimate is at the high end of current estimates, although alternative parameter choices can substantially reduce this estimate. Tropical wetlands accounted for 79 % of the global wetland methane budget. Northern latitudes only contributed $12 \text{ Tg CH}_4 \text{ yr}^{-1}$, partially due to the low inundated area derived from satellites and the low high-latitude productivity in CLM4CN. On average, 15 % of methane was oxidized in the rhizosphere before being released to the atmosphere through aerenchyma. This proportion varied from region to region and was highest in middle latitudes.

We applied the set of changes enumerated above to global model simulations and conducted sensitivity analysis to test their importance in constraining global methane fluxes. More sensitivity studies were conducted in a related paper (Riley et al., 2011). Sensitivity analysis suggests that global methane fluxes are most sensitive to the inclusion of a soil pH factor (f_{pH}). Sensitivity analysis suggests a large range ($150\text{--}346 \text{ Tg CH}_4 \text{ yr}^{-1}$) in the annual methane flux when some of the features described in this study were not taken into account (Table 7).

Rice paddies were simulated to release 42 Tg yr^{-1} to the atmosphere. This estimate is within the range of current estimates. In our model, Chinese rice paddies contribute $10 \text{ Tg CH}_4 \text{ yr}^{-1}$, which is similar to other estimates using localized data. Due to the lack of information on agricultural activities (such as water management strategies and use of fertilizers) on the global scale, CLM4Me' might overestimate emissions from rice paddies. In addition, our rain-fed rice paddies might include upland and deepwater rice areas which are probably not significant sources of atmospheric methane.

This study represents an effort to quantify global methane fluxes. However, there are still large uncertainties on the magnitude of global methane fluxes (Petrescu et al., 2010). In order to remove these uncertainties, further model improvement should focus on constraining the parameters that govern methane-related processes and possibly

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employ spatially variable parameters (such as Q_{10} for methane production). In addition, additional field measurements and studies of methane flux should focus on tropical wetlands; although tropical wetlands represent a significant source of atmospheric methane and seasonal and interannual variation of methane flux is primarily controlled by the tropical wetland extent, relatively few field studies have been done in this region. This study also suggests that rhizospheric methane oxidation is an important control on the global methane flux. Rhizospheric methane oxidation is affected not only by environmental factors but also by physiological factors. Detailed study of this process is necessary to further improve global methane flux estimates.

Appendix A

IGBP soil pH vs. wetland pH

We compared wetland pH and IGBP soil pH at each site in this study (Fig. A). It suggests a generally good agreement between the two different datasets ($r = 0.69$, RMSE = 1.07).

Appendix B

Spring peaks in methane emissions

Aerenchyma in plants serves as a conduit for (i) O_2 from the atmosphere to the soil and (ii) CH_4 from the soil to the atmosphere. Therefore, aerenchyma represents a faster pathway to the atmosphere than ebullition and diffusion in water. In this model, the specific aerenchyma area T ($m^2 m^{-2}$) depends on fine root C (FROOTC, $gC m^{-2}$). Thus, FROOTC will affect the partitioning of methane fluxes among diffusion, ebullition, and transport through aerenchyma. So, accurate estimation of FROOTC plays a critical role in methane emissions.

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Model simulations demonstrated an early spring spike in methane emissions at the Michigan site (Fig. B1a) lasting 2–3 days. This high methane flux was produced through the initial accumulation of methane in soils (Fig. B1c) followed by its rapid release through aerenchyma. Methane accumulates in the soil as methane production begins in the spring and upward transport is slow since aerenchyma, ebullition, and diffusion are slow (Fig. B1d). A sharp increase in aerenchyma releases methane in a burst because of abrupt increases in the simulated FROOTC, which increase from ~ 0 to highest values (~ 300) within ~ 15 days (Fig. B1b). Although it appears that this peak in CH₄ emissions does not match the observations, we caution that weekly observations could easily miss these emissions, and that flux chamber observations are notoriously unable to capture temporally and spatially heterogeneous fluxes. This quick increase in fine root C predicted by the CLM-CN might not be realistic and deserves further attention for model testing and improvement.

Appendix C

Simulated daily mean fluxes with (and without) pH function vs. observations

Here we only selected the sites with pH < 5 or pH > 8 for this analysis because other sites have optimal (or close to optimal) pH values and the inclusion of pH in methane production will not significantly affect their methane fluxes. As can be seen in Fig. C, pH function generally decreases RMSE (from 472 to 143 mg CH₄ m⁻² d⁻¹) and does not affect the correlations very much.

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Table 1. Site descriptions for rice paddy fields.

Site name	Year	Location	pH	Date of field flooded	Date of final drainage	Nitrogen added	Rice type (cultivar)	Measurement techniques	Soil type	References
Texas, USA	1994	29.95° N, 265.5° E	N/A	17 May	11 Aug	Yes	Lemont	Chamber	Bernard-Morey	Sigren et al. (1997)
Italy	1991	45.3° N, 8.42° E	6	7 May	30 Aug	Yes	Roma/Lido	Static (closed) chamber	Sandy loam	Butterbach-Baul et al. (1997)
Chengdu, China	2003	31.27° N, 105.45° E	8.1	9 May	7 Sep	Yes	hybrid II-You 162	Chamber	Purplish	Jiang et al. (2006)
Nanjing, China	1999	32.8° N, 118.75° E	N/A	18 Jun	13 Oct	Yes	# 9561	Chamber	Hydromorphic	Huang et al. (2001)
California, USA	1982 1983	40.2° N, 237.98° E	N/A	11 May 21 May	2 Oct 1 Oct	Yes Yes	M101	Static chamber	Capay silty clay	Cicerone et al. (1983, 1992)
Japan	1991 1993	36.02° N, 140.22° E	6.6–6.9 6.6–6.9	7 May 7 May	12 Aug 2 Sep	Yes Yes	Koshihikar Koshihikar	Automated chamber	Gley soil (sandy clay loam)	Yagi et al. (1996)
New Delhi, India	1995 1996	20.08° N, 77.12° E	8.2	1 Jul	1 Nov	Yes	IR72	Closed chamber, manual	Ustochrept (sandy loam)	Jain et al. (2000)
Cuttack, India	1996	20.42° N, 85.92° E	6.19	19 Jul	30 Oct	Yes	CR 749-20-2	Automatic chamber	Haplaquept (Alluvial)	Adhya et al. (2000)
Beijing, China	1995	40.55° N, 116.78° E	7.99	4 Jun	17 Oct	Yes	Zhongguo	Automatic chamber	Silty clay loam	Wang et al. (2000)
Central Java, Indonesia	2001–2002	6.63° S, 110° E	5.1	1 Nov	28 Feb	Yes	Memberamo, Cisadane, IR64, Way Apoburu	Automatic closed chamber	Aeric Tropaquept (silty loam)	Setyanto et al. (2004) (Indonesia Journal of Agricultural Science)
Lampung, Indonesia	1993	4.52° S, 105.3° E	5	21 Nov	4 Mar	Yes	Oryza Sativa var. IR-64	Chamber	Typic Paleudult (sandy clay)	Nugroho et al. (1994) (SSPN)

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Table 2. Descriptions of wetland sites used in this study.

Site name	Location	Wetland type	Dominant vegetation	Mean precipitation and temperature	Soil and climate characteristics	Measurement technique	Forcing data*	References
Central Kalimantan, Indonesia	2.33° S, 113.92° E	Ombrotrophic peatland	Evergreen broadleaved trees	Mean precipitation is 2331 mm and mean <i>T</i> is 26.3 between 2002 and 2005	Wet season from October to May and dry season from June to September, soil pH is 4.0	Dark static chamber	Measured water table positions	Jauhiainen et al. (2005)
Panama	9° N, 80° W	Swamp	Palms	Mean precipitation is 1600 mm in Panama city and mean temperature is 27 °C	Four-month dry season between February and May. Soil pH is 6.2	Static chamber	Modeled water table positions from Walter and Heimann (2000)	Keller (1990); Walter and Heimann (2000)
Florida, USA	30.07° N, 275.8° E	Swamp	Sagittaria lancifolia	Annual precipitation is about 1400 mm	Soil pH is 6.2	Open chamber	Fully saturated areas	Lombardi et al. (1997)
Salmisuo, Eastern Finland	62.75° N, 30.93° E	Minerotrophic, oligotrophic pine fen	Sphagnum papillosum	Mean temperature is about 10 °C	Wet conditions from July to September	Static chamber	Measured water table positions	Saarnio et al. (1997)
Michigan, USA	42.45° N, 84° W	Ombrotrophic peatland	Sphagnum, Scheuchzeria palustris, Vaccinium oxycoccos	Mean precipitation for 1948–80 is 761 mm	Soil pH 4.2	Static chamber	Measured water table positions	Shannon and White (1994)
Minnesota, USA	47.53° N, 266.53° E	Poorly-minerotrophic to ombrotrophic peatland	Sphagnum, Chamaedaphne calyculata, Scheuchzeria palustris	Average precipitation is 553 mm and mean temperature is about 13.6 °C for the May–October period	Soil pH is 4.6	Eddy correlation technique	Measured water table positions	Shurpali and Verma (1998)
Alberta, Canada	54.6° N, 246.6° E	Nutrient rich fen	Carex aquatilis and cares rostrata	N/A	The freeze-thaw cycle spans from May to October, pH = 7	Open chamber	Fully saturated areas	Popp et al. (2000)

* All sites use NCEP atmospheric forcing.



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Table 3. Model performance statistics for the Base and NopH simulations at selected wetland sites.

Site	Base		NopH	
	r	RMSE	r	RMSE
Indonesia	0.45	28.97	0.45	411.26
Minnesota, USA	0.57	27.92	0.57	162.43
Michigan, USA	0.09	76.29	-0.08	201.00

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Table 4. Parameters used for sensitivity test.

Parameter	Description	Value used	Units	Range	References
f_{CH_4}	CH ₄ /CO ₂	0.2		0.001–1.7	Segers (1998)
ρ	Porosity of tillers	0.3		0.08–0.43	Colmer (2003)
Q_{10}	Q_{10} for CH ₄ production	3		1.5–26	Segers (1998)
$R_{\text{o,max}}$	Maximum oxidation rate	45	μM h ⁻¹	5.0–50.0	Dunfield et al. (1993); Knoblauch (1994)
K_{CH_4}	CH ₄ half-saturation oxidation coefficient	5	μM	1.0–5.0	Walter and Heimann (2000); Knoblauch (1994)
$Q_{\text{o},10}$	Q_{10} oxidation constant	1.9		1.4–2.1	Knoblauch (1994)
K_{O_2}	O ₂ half-saturation oxidation coefficient	20	μM	17–25	Lidstrom and Somers (1984)
$C_{\text{e,max}}$	CH ₄ concentration to start ebullition	0.15		0.12–0.15	Kellner et al. (2006); Baird et al. (2004)

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Table 5. Results from sensitivity test for the Panama Site. Percentage values in parentheses are relative to the simulations using the base values.

Parameter	Description	Low	High	Base value
f_{CH_4}	CH ₄ /CO ₂ ratio	0.1 (−53.4 %)	0.3 (58.5 %)	0.2
ρ	Grass aerenchyma porosity	0.1 (+30 %)	0.43 (−49.6 %)	0.3
Q_{10}	Q_{10} for CH ₄ production	1.5 (−41.9 %)	5 (+11 %)	3
$R_{o,max}$	Maximum oxidation rate	5 (36.1 %)	50 (−1.7 %)	45
K_{CH_4}	CH ₄ half-saturation oxidation coefficient	1 (−5.57 %)	10 (+5.22 %)	5
$Q_{o,10}$	Q_{10} for CH ₄ oxidation	1.4 (7.1 %)	2.4 (−5.1 %)	1.9
K_{O_2}	O ₂ half-saturation oxidation coefficient	17 (−0.6 %)	25 (0.867 %)	20
$C_{e,max}$	CH ₄ concentration to start ebullition	0.13 (0 %)	0.17 (0 %)	0.15



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Table 6. Comparison of global wetland methane estimates between our model and other models.

Model	Climate zone			Total global budget
	Northern (> 50° N)	Temperate (20–50° N, 30° S–50° S)	Tropical (20° N–30° S)	
Matthews and Fung (1987)	65	14	32	111
Aselmann and Crutzen (1989)	25	12	43	80
Bartlett et al. (1990)	39	17	55	111
Bartlett and Harriss (1993)	34	5	66	105
Cao et al. (1996)	23.4	17.2	51.4	92
Walter et al. (2001)	48	26	186	260
Zhuang et al. (2004)	31–106*	N/A	N/A	
Wania et al. (2010)	40.8–73.7*	N/A	N/A	
Bousquet et al. 2006	31.55	25	103	159.55
Chen and Prinn (2006)	21.9–57.9*			143–148
This model	12 (15*)	43	201	256

* For high latitude > 45° N.

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Table 7. Global methane budget for different case simulations for year 1993.

Simulation	Global budget	Percentage change	Description
Base	245	0 %	All features are included
NoRedox	290	18 %	Same as base, except $f_{pE} = 1.0$
NopH	346	41 %	Same as base, except $f_{pH} = 1.0$
LowR _{o,max}	275	12 %	Same as base, except $R_{o,max} = 1/10$ default value
NoLimitAeren	237	−3 %	Same as base, except $f_{aere} = 1.0$
UseLAI	150	−39 %	Same as base, except that LAI is used in calculation of aerenchyma area

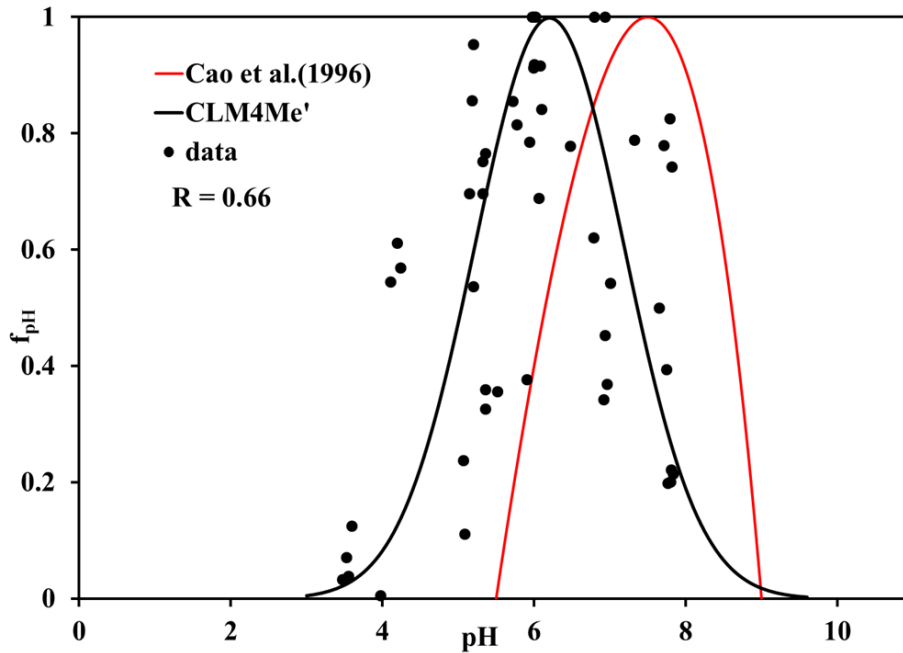


Fig. 1. pH function used in the model (black line). The optimal pH for methanogenesis is 6.2 in our pH function. The red line shows pH function used in Cao et al. (1996) with optimal pH 7.5.

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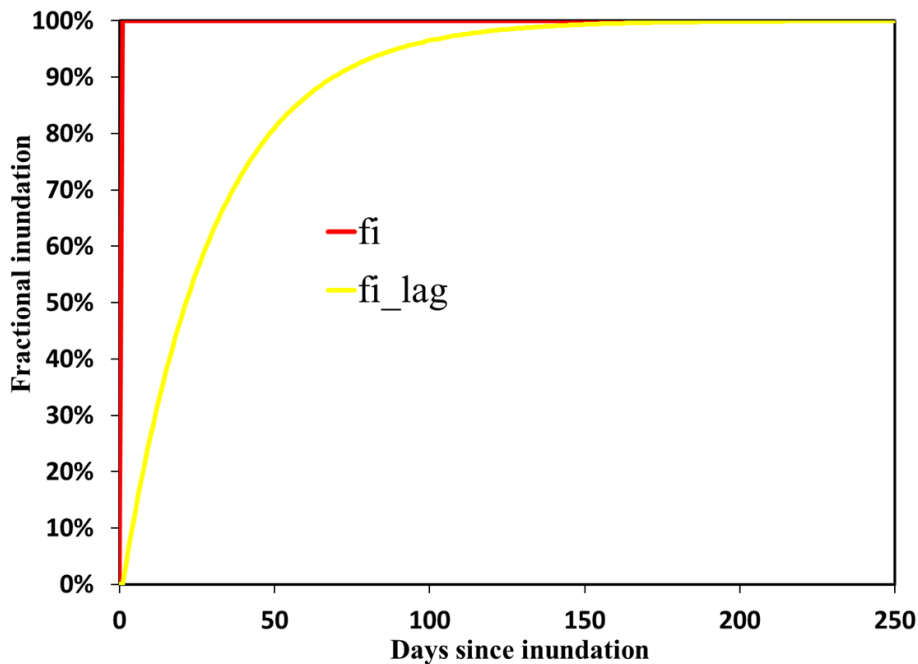


Fig. 2. An illustrative diagram of the impact of redox potential on inundated fraction. f_i is representative of the inundated fraction that is predicted by the model. f_{i_lag} is the inundated fraction that is actively producing methane.

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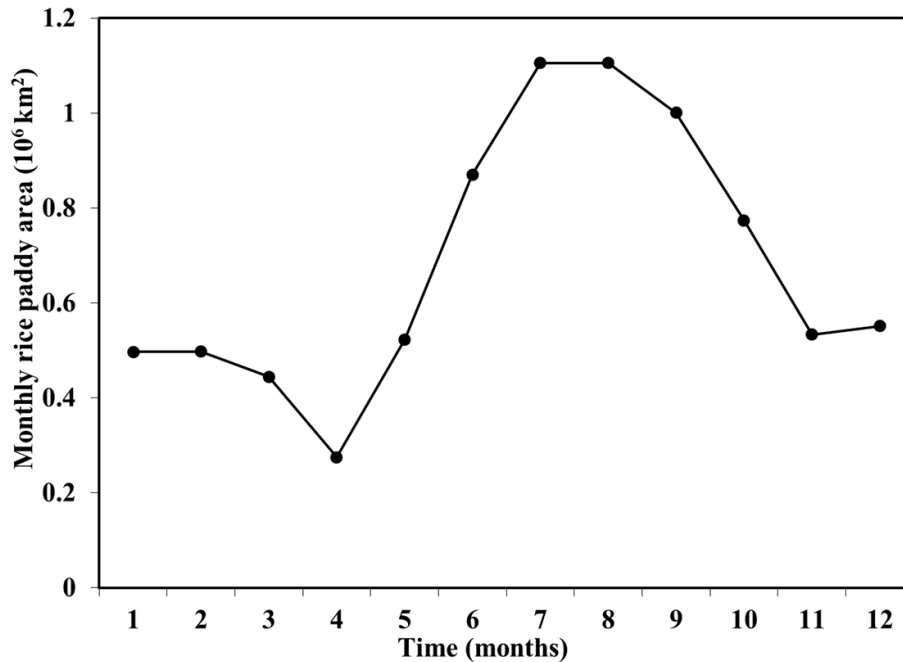


Fig. 3. Seasonal variation of global rice paddy areas in year 2000 (Portmann et al., 2010). The rice paddy area peaks in July and August.

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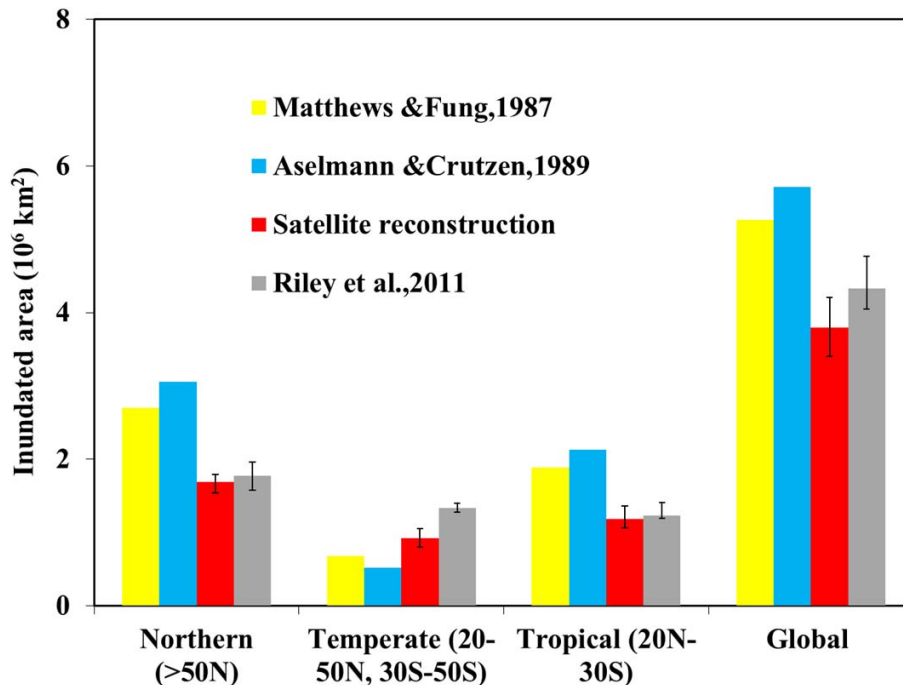


Fig. 4. Comparison of inundated areas used in different methane models with error bars indicating the range of annual mean inundated areas. Please note that for the northern regions the inundated area only in June, July, August, and September are used to calculate the annual mean inundated area in order to exclude frozen soils in winter.

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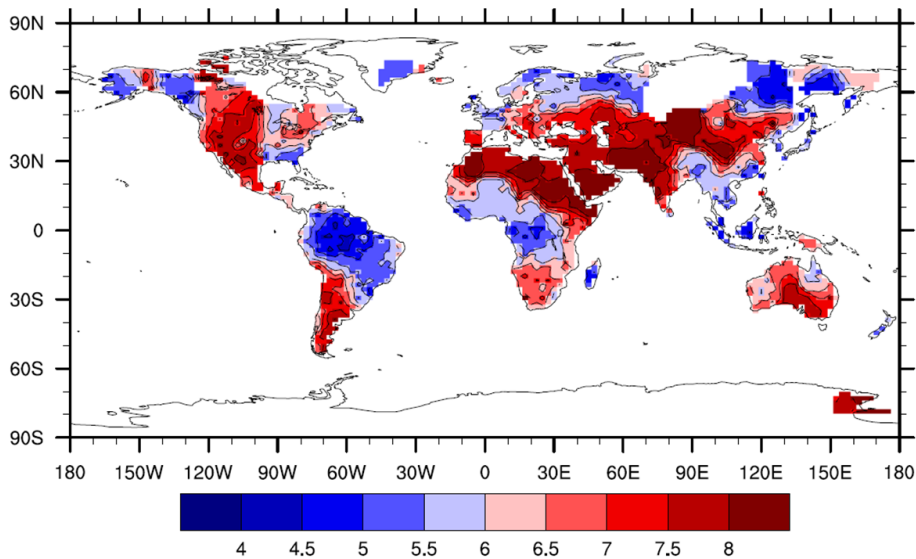


Fig. 5. The global distribution of soil pH. Data sources: IGBP-DIS (see Tempel et al., 1966; Pleijsier, 1986).

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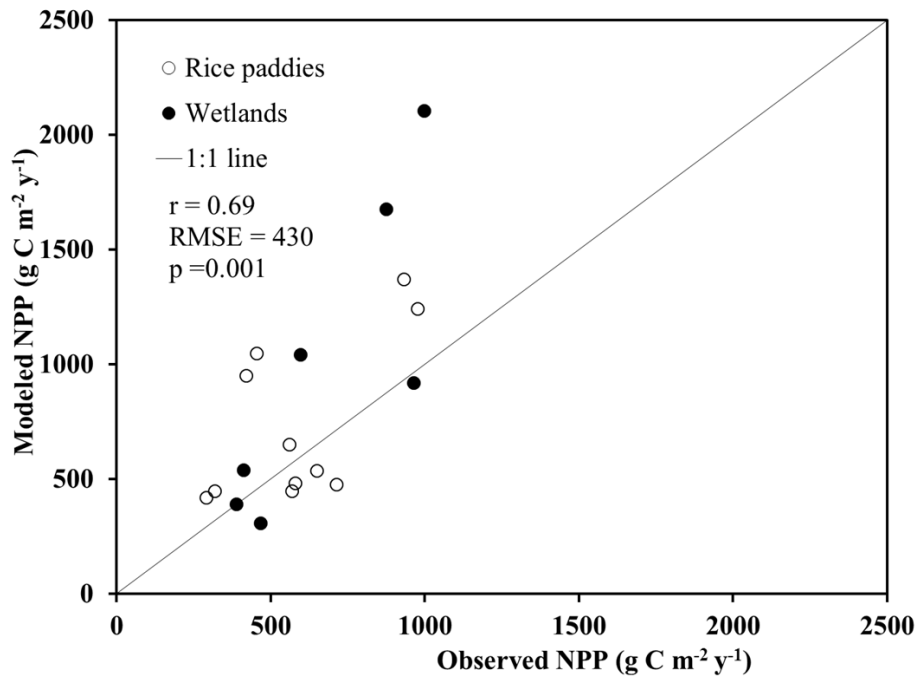


Fig. 6. Scatter plot of modeled annual NPP vs. observed annual mean NPP at the rice paddy and wetland sites. The observed annual mean NPP was obtained from MODIS (Zhao et al. 2005). r is correlation coefficient, rmse indicates root mean squared error, and p is probability level.

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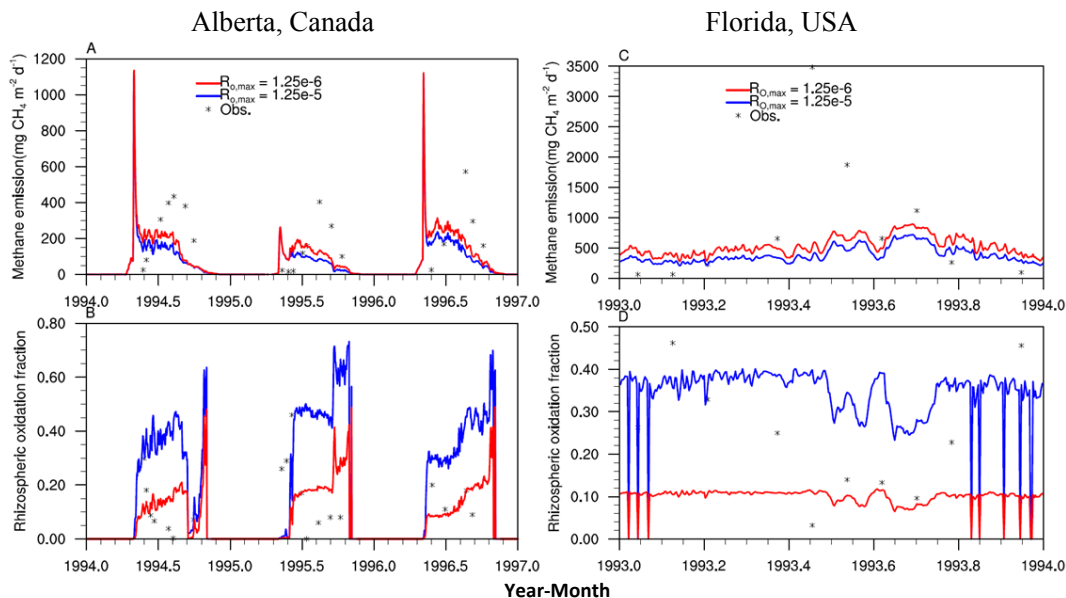


Fig. 7. Comparisons between model simulations and observation at Alberta, Canada and Florida, USA sites. For each site, the top figure shows comparison of methane emissions with different $R_{o,max}$ values; the bottom figure shows the comparison of estimated rhizospheric oxidation fraction with different $R_{o,max}$ with observations.

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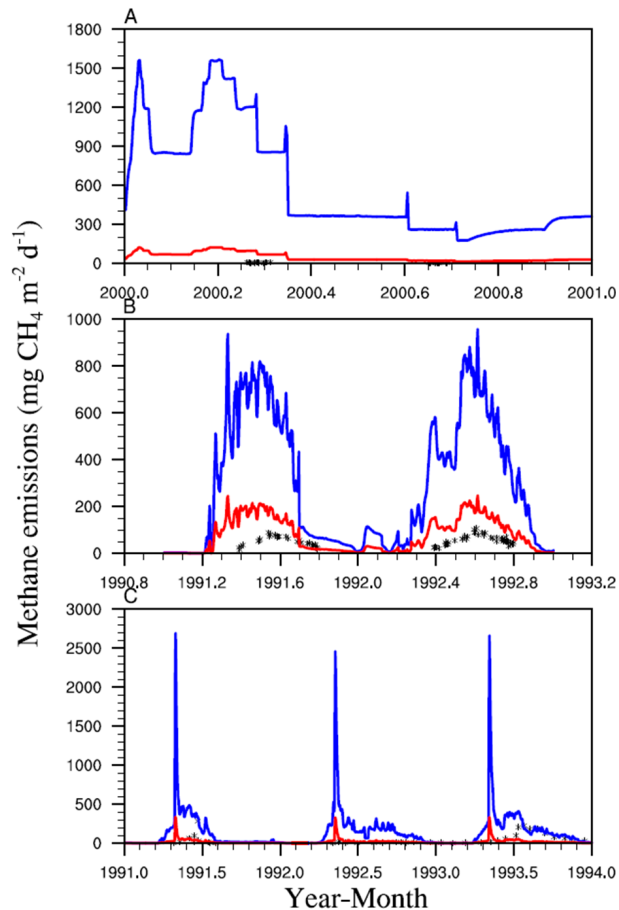


Fig. 8. Comparison between model simulations and observations at wetland sites. Red line indicates simulations with f_{pH} ; blue line shows simulations without f_{pH} . **(A)** Indonesia; **(B)** Minnesota; **(C)** Michigan. Observations are in dots. Please see Table 2 for site descriptions.

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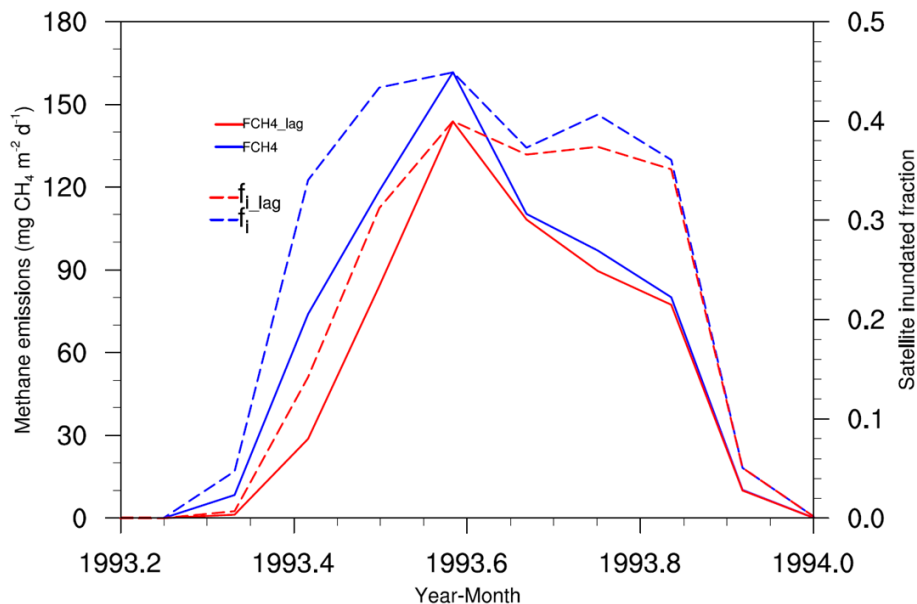


Fig. 9. Impact of redox potential on methane production and inundated fraction at a grid cell (lat: 48.31° N, lon: 267.5° E) extracted from global simulation. Dashed lines indicate satellite inundated fraction (f_i in blue) and delayed inundated fraction (f_{i_lag} in red); solid lines are methane emissions with (FCH_{4_lag} in red) and without (FCH₄ in blue) the inclusion of redox potential impact.

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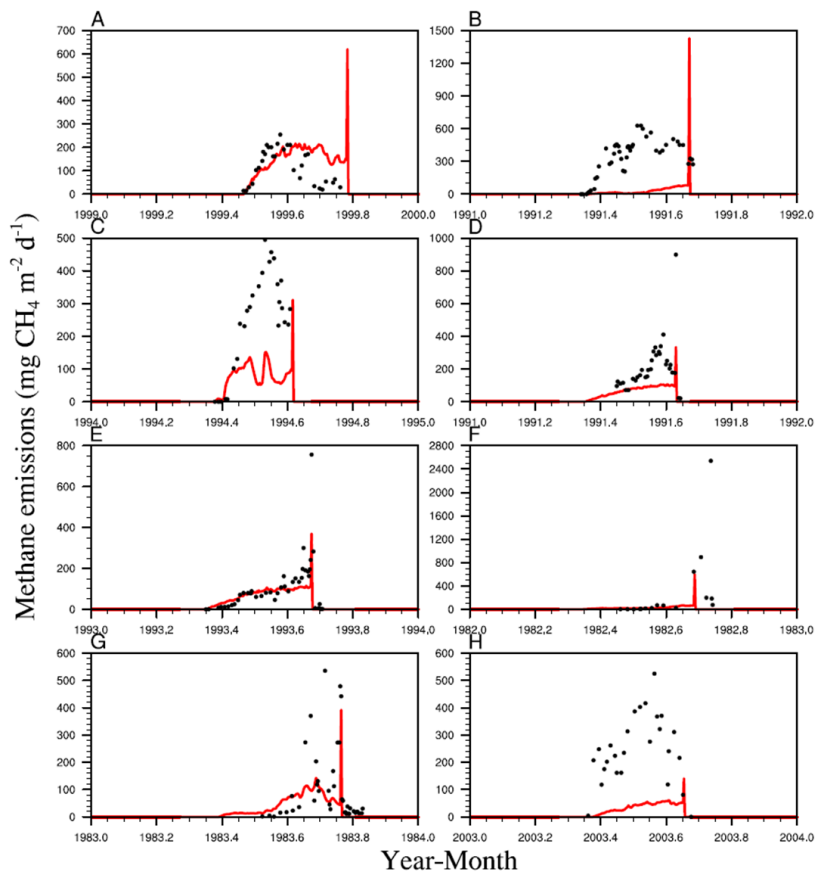


Fig. 10. Comparison between model ed methane fluxes (red lines) and observation (dots) at each rice paddy site. Note that the scale of y-axis varies between plots. **(A)** Nanjing, China; **(B)** Italy; **(C)** Texas, USA; **(D)** Japan, 1991; **(E)** Japan, 1993; **(F)** California, USA, 1982; **(G)** California, USA, 1983; **(H)** Chengdu, China. Please see Table 1 for site descriptions.

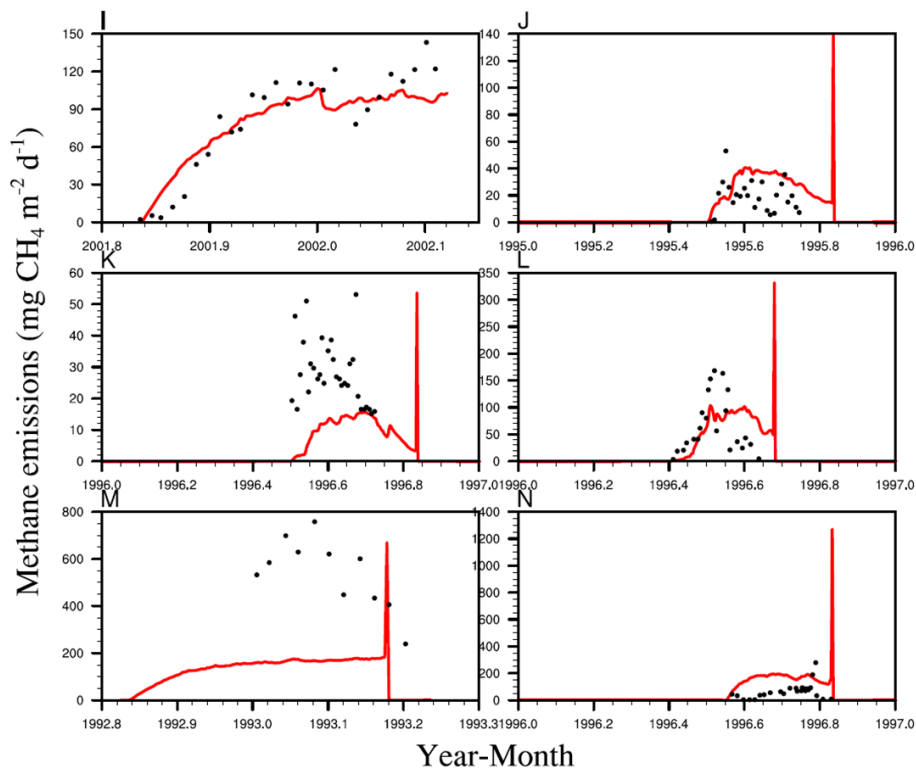


Fig. 10. (continued). **(I)** Central Java, Indonesia; **(J)** New Delhi, India, 1995; **(K)** New Delhi, India, 1996; **(L)** Beijing, China; **(M)** Lampung, Indonesia; **(N)** Cuttack, Indonesia. Please see Table 1 for site descriptions.

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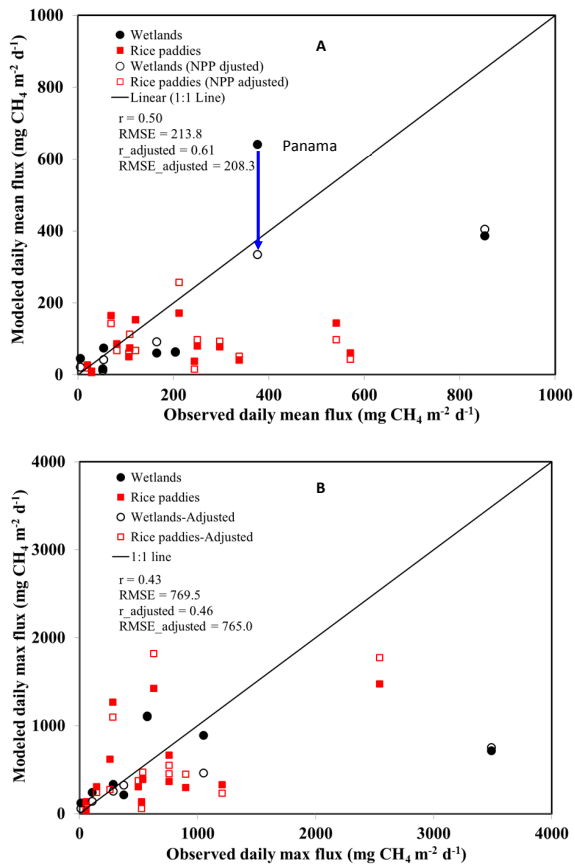


Fig. 11. Scatter plot of observed and model (and NPP adjusted) simulated mean (top) and annual daily maximum (bottom) methane emissions ($\text{mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$) at the rice paddies and wetlands.

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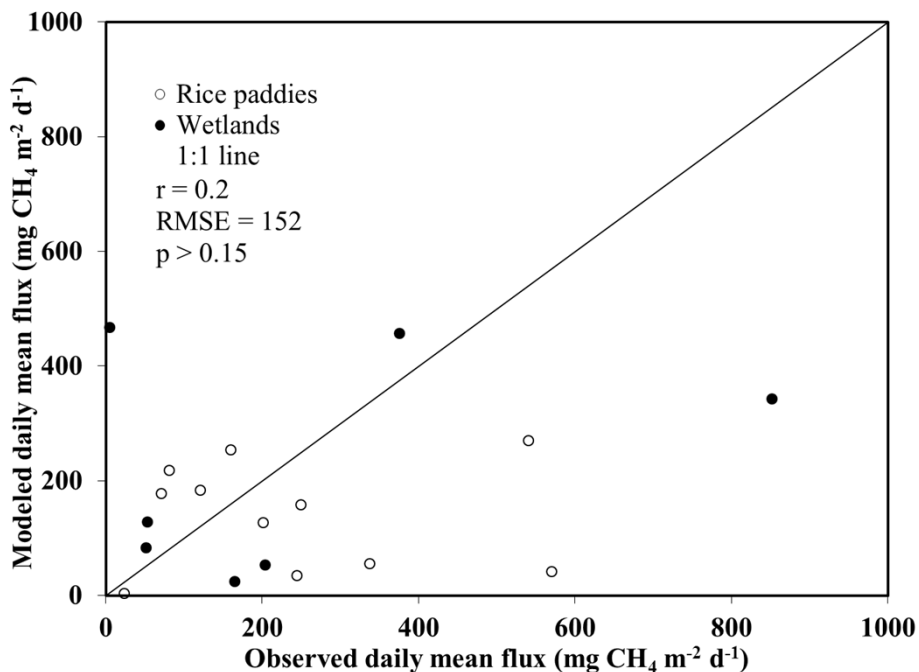


Fig. 12. Comparison of mean methane fluxes extracted from the closest gridcells in the global simulation with observations at sites. It demonstrates a poor correlation between them due to the spatial heterogeneity and large spatial resolution in the global simulation.

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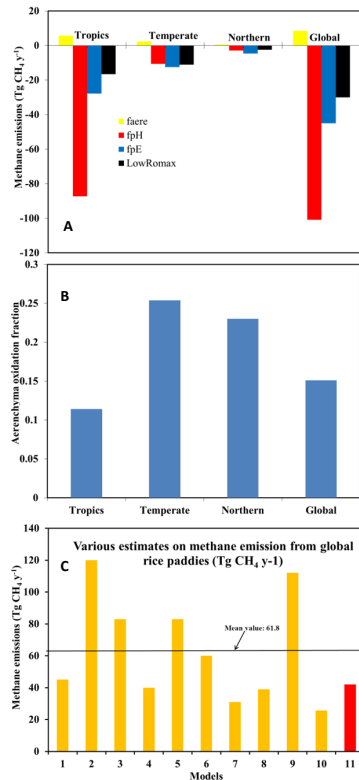


Fig. 13. (A) Sensitivity analysis of each variable. The number on the y-axis indicates the change in net annual mean methane emission associated with changes in each variable. (B) Prognostic aerenchyma oxidation fractions at different regions. (C) Comparison of global methane budget from rice paddies estimated in our model and other models. 1: Seiler et al. (1984); 2: Holzapfel-Pschorn and Seiler (1986); 3: Bouwman (1990); 4: Sass (1994); 5: Hein et al. (1997); 6: Wuebbles and Hayhoe (2000); 7: Scheehle et al. (2002); 8: Olivier et al. (2005); 9: Chen and Prinn (2006); 10: Yan et al. (2009); 11: this model (red).

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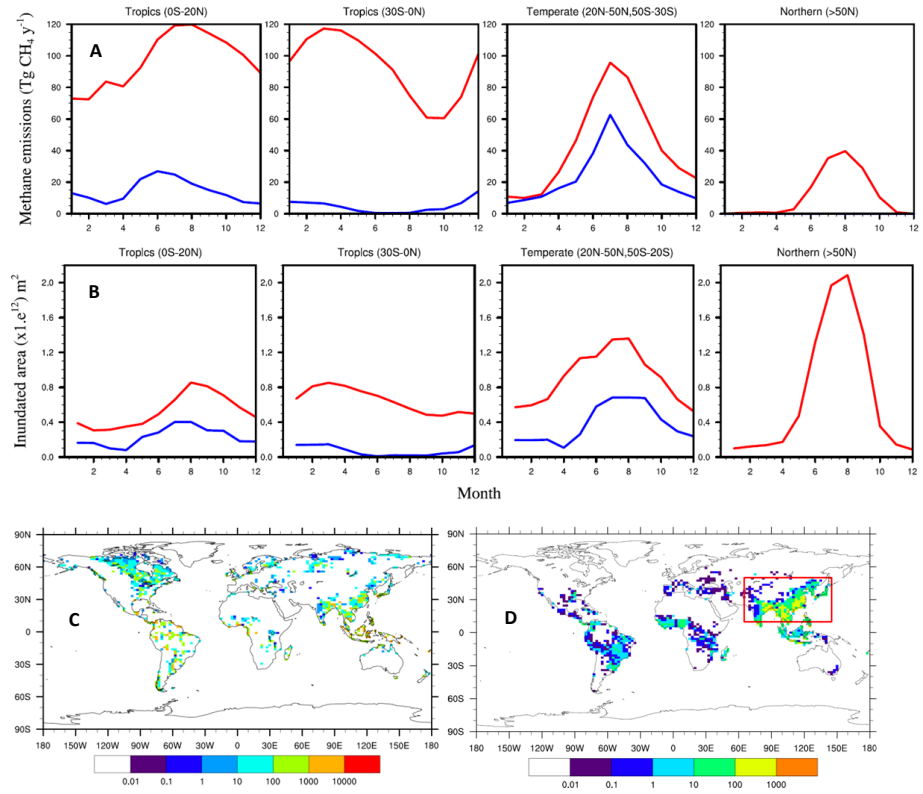


Fig. 14. Seasonal variation of methane emissions **(A)** and inundated areas **(B)** in the four defined regions for natural wetlands (red) and rice paddies (blue). **(C)** The global distribution of the mean methane emission rates (units: $\text{mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$) during the period 1993–2004 from natural wetlands. **(D)** The global distribution of annual averaged methane emissions (units: $\text{mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$) for the year 2000 from rice paddies. (Asian monsoon regions are in red box).

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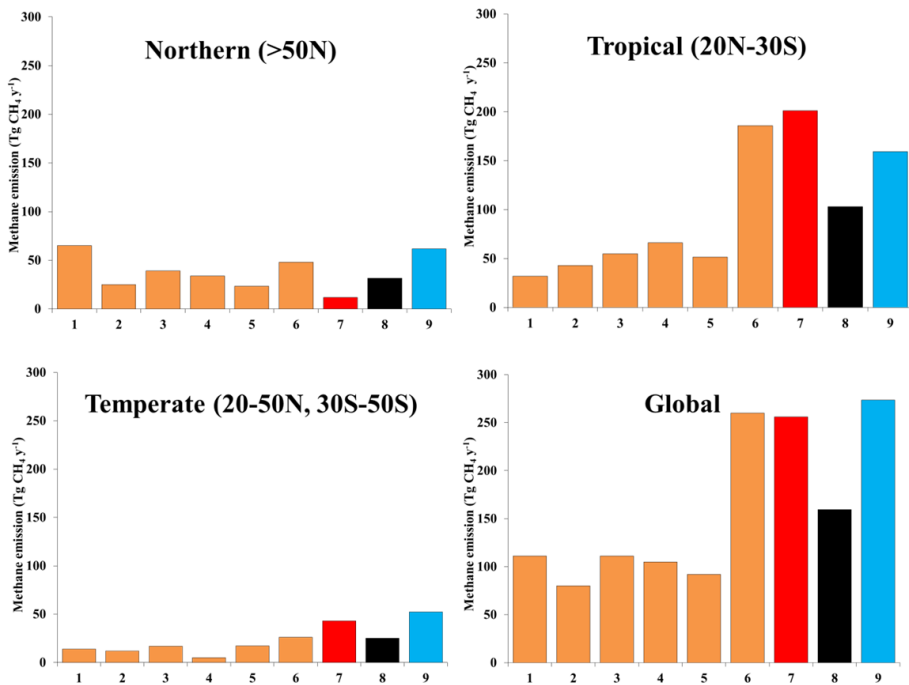


Fig. 15. Comparison of total CH₄ emissions (Tg CH₄ yr⁻¹) between our model and other models' estimations from natural wetlands. (1: Matthews and Fung (1987), 2: Aselmann and Crutzen (1989), 3: Bartlett et al. (1990), 4: Bartlett and Harriss (1993), 5: Cao et al. (1996), 6: Walter et al. (2001), 7: the CLM4Me' (this study), 8: Bousquet et al. (2006); 9: the CH₄Me model (Riley et al., 2011). Red indicates the CLM4Me' and Black is a top-down inversion.) Please note that estimates from the CH₄Me (9) may include rice paddy emissions since the rice paddy fraction was not removed from model simulated inundated fraction.

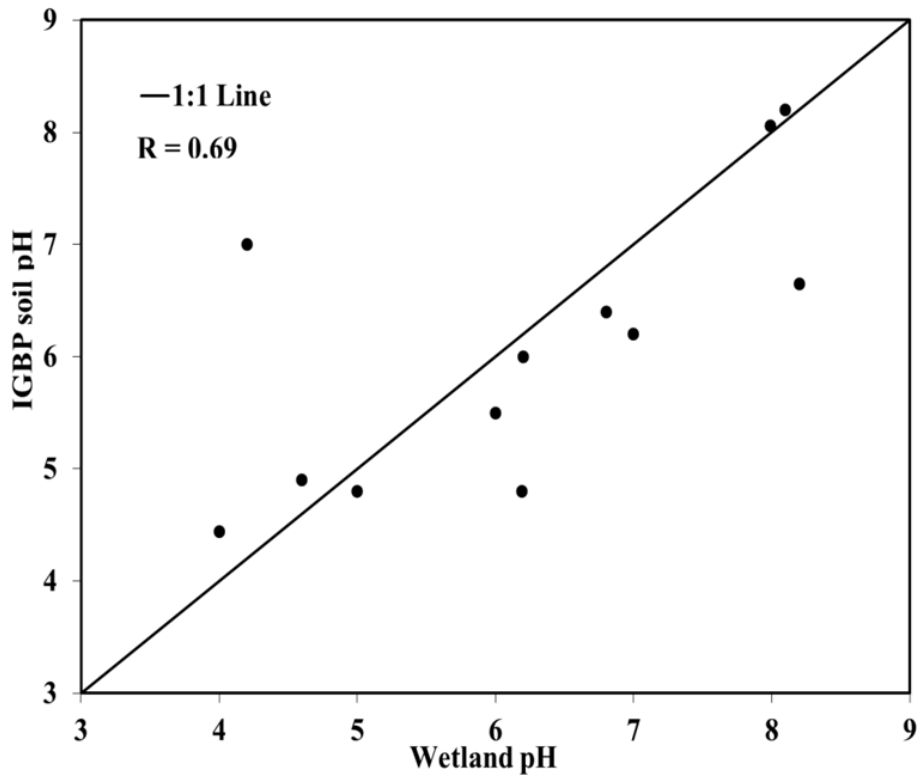


Fig. A. Comparison between wetland pH and IGBP soil pH.

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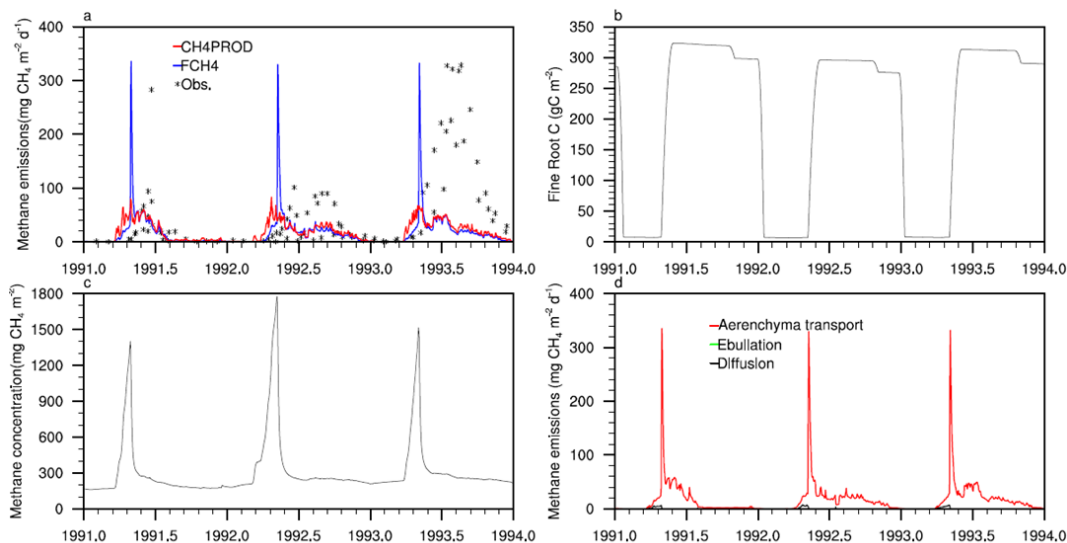


Fig. B. Model simulations at the Michigan site (Shannon and White, 1994). **(a)** Model simulated emission rates and production rates vs. observations; **(b)** model simulated fine root carbon; **(c)** vertically averaged methane concentration in the model; **(d)** partition of methane fluxes through aerenchyma transport (red), ebullition (green), and surface diffusion (black).

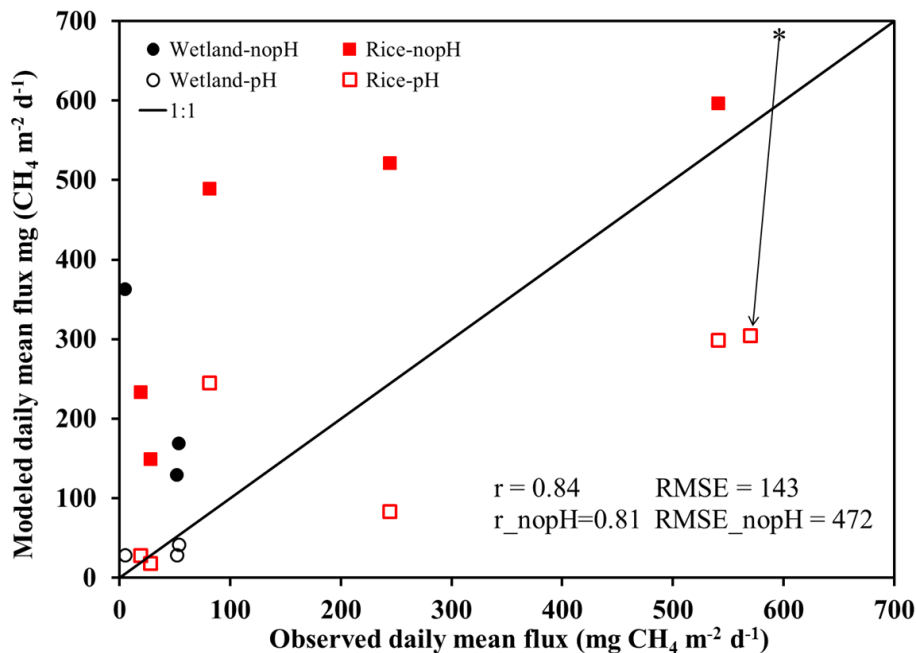


Fig. C. Scatter plot of modeled daily mean flux with (and without) pH function with observations. * indicates the corresponding modeled daily mean flux without pH function at this site is $1601 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ which is beyond the range and not shown in this figure. Because of the limited wetland sites (only 3), we used both wetland and rice paddy sites together to calculate the correlations and RMSE. A detailed analysis of these three wetland sites is provided in Sect. 4.3.

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