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Global Biogeochemical Cycles

RESEARCH ARTICLE

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Key Points:

- Global CH₄ uptake by soils doubled from 1900 to 2015
- Future global CH₄ uptake by soils ranges between 25.7 and 82.7 Tg yr⁻¹ in 2100
- Changes in CH₄ uptake by soils can be attributed to changes in atmospheric CH₄, temperature, soil moisture, and nitrogen inputs

Supporting Information:

Supporting Information may be found in the online version of this article.

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Global Uptake of Atmospheric Methane by Soil From 1900 to 2100

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Abstract Soil methanotrophy is the only biological process that removes methane (CH₄) from the atmosphere. There is good agreement about the size of the global sink but great uncertainty about its interannual variability and regional responses to changes in key environmental drivers. We used the process-based methanotrophy model Methanotrophy Model (MeMo) v1.0 and output from global climate models to simulate regional and global changes in soil uptake of atmospheric CH₄ from 1900 to 2100. The annual global uptake doubled from 17.1 ± 2.4 to 37.2 ± 3.3 Tg yr⁻¹ from 1900–2015 and could increase further to 82.7 ± 4.4 Tg yr⁻¹ by 2100 (RCP8.5), primarily due to enhanced diffusion of CH₄ into soil as a result of increases in atmospheric CH₄ mole fraction. We show that during the period 1980–2015 temperature became an important influence on the increasing rates of soil methanotrophy, particularly in the Northern Hemisphere. In RCP-forced simulations the relative influence of temperature on changes in the uptake continues to increase, enhancing the soil sink through higher rates of methanotrophic metabolic activity, increases in the global area of active soil methanotrophy and length of active season. During the late 21st century under RCP6.0, temperature is predicted to become the dominant driver of changes in global mean soil uptake rates for the first time. Regionally, in Europe and Asia, nitrogen inputs dominate changes in soil methanotrophy, while soil moisture is the most important influence in tropical South America. These findings highlight that the soil sink could change in response to drivers other than atmospheric CH₄ mole fraction.

1. Introduction

Methane (CH₄) consumption by soil methanotrophic bacteria is the second largest sink of atmospheric CH₄ after oxidation by the hydroxyl radical (Allan et al., 2007; Ciais et al., 2013). In addition, soil methanotrophy is the only known biological sink in the global CH₄ budget, accounting for 5%–7% of the global atmospheric CH₄ uptake per year (Saunio et al., 2020) and preventing the emission of as much as 90% of CH₄ produced in wet soil (Oremland & Culbertson, 1992; Singh et al., 2010).

Rates of soil methanotrophy are controlled by atmospheric CH₄ concentration, temperature, moisture, soil texture, nitrogen content, and land use practices (Castro et al., 1995). Global process-based models typically estimate soil methanotrophy rates based on a reaction-transport equation that describes diffusive transport of CH₄ into soil as well as microbial oxidation of CH₄. Both transport and reaction rates vary as a function of environmental conditions. Diffusive transport of CH₄ through the soil-atmosphere interface is controlled mainly by the concentration gradient of CH₄ across the interface and thus, is governed by atmospheric CH₄ concentration and soil porosity. The latter parameter varies according to soil texture and can be modified by land use practices such as tillage, which can further affect soil moisture, texture, and N content. Bacterial CH₄ oxidation is an enzymatic process that is influenced by temperature, oxygen availability, and nitrogen concentration (Curry, 2007; Murguia-Flores et al., 2018; Potter et al., 1996; Ridgwell et al., 1999; Zhuang et al., 2013).

Bacterial methanotrophy occurs in all soils under aerobic conditions (Hanson & Hanson, 1996). Process-based model simulations, atmospheric inversion modeling, and upscaling of field observations suggest that soil methanotrophy globally removes 11–49 Tg of CH₄ per year from the atmosphere (Curry, 2007; Dörr

et al., 1993; Dutaur & Verchot, 2007; Hein et al., 1997; Murguía-Flores et al., 2018; Potter et al., 1996; Ridgwell et al., 1999; Saunio et al., 2020; Smith et al., 2000; Spahni et al., 2011; Zhuang et al., 2013). This amount is in the range of annual global emissions of CH₄ from rice agriculture (Saunio et al., 2020). Despite its global significance, soil uptake of atmospheric CH₄ currently is not represented in most state-of-the-art Earth System Models (ESMs) (Nazaries et al., 2013), which compromises our ability to quantify and predict feedbacks between the soil CH₄ sink and changes in global climate and land use.

Environmental factors that control rates of soil methanotrophy have changed during the past century due to natural variation and anthropogenic activity. More specifically, the uptake of atmospheric CH₄ via soil methanotrophy has been altered by: (a) the increase of CH₄ mole fraction in the atmosphere, (b) the indirect influence of increases in atmospheric greenhouse gas concentrations on soil temperature and moisture, which alter rates of both gas diffusion in soil and methanotroph metabolism, (c) input of anthropogenic nitrogen to soil, and (d) land use practices that physically alter soil structure and texture. Relative to the preindustrial era, the atmospheric CH₄ mole fraction has nearly tripled (Etheridge et al., 1998; Saunio et al., 2020) and, since 1750, anthropogenic emissions of greenhouse gases have resulted in a global temperature increase of ~0.8°C (Hansen et al., 2010). Reactive nitrogen in soil also has doubled due to chemical fertilizer application (Nishina et al., 2017) and atmospheric deposition of nitrogen (Lamarque et al., 2013). In addition, more than 40% of the ice-free land has been modified by humans, in particular, for agriculture (Foley et al., 2011), which alters both the chemistry and structure of natural soil.

The net effect of these changes has been a two-fold increase in the rate of atmospheric CH₄ uptake by soil during the last century (Curry, 2009; Zhuang et al., 2013). Previous studies have quantified global soil methanotrophy and its drivers (Curry, 2009; Zhuang et al., 2013). Curry (2009) and Murguía-Flores et al. (2018) primarily quantified the present-day soil CH₄ sink. In addition, Murguía-Flores et al. (2018) also provided an analysis of how different factors such as temperature, soil moisture, and nitrogen deposition affect CH₄ oxidation rates globally. Zhuang et al. (2013) quantified the strength of the 20th and 21st century soil CH₄ sink. They forced their model with SRES A1FI and B1 and also examined spatial patterns in CH₄ uptake. However, significant knowledge gaps remain in the characterization of dominant drivers of the soil CH₄ sink, including how they vary globally and how they may change in the future. This information is critical for our ability to predict the impacts of global change on the strength of the global soil CH₄ sink and assess the capacity for soil to mitigate increases in atmospheric CH₄.

The objectives of this study are thus to use the process-based model MeMo v1.0 (hereafter referred to as MeMo) developed by Murguía-Flores et al. (2018) to (a) quantify global changes in CH₄ uptake by soil from 1900 to 2100, (b) characterize drivers (temperature, soil moisture, nitrogen input, and atmospheric CH₄ mole fraction) responsible for spatial changes in soil methanotrophy during that time period, and (c) estimate the potential capacity of global soil methanotrophy to offset CH₄ emissions during the next century under the Representative Concentration Pathways (RCPs) used in the Fifth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC, 2013).

2. Materials and Methods

2.1. Model Description

MeMo is a global, vertically resolved soil model used to simulate CH₄ diffusion and bacterial methanotrophy in soil. MeMo was built on the models of Potter et al. (1996), Ridgwell et al. (1999), and Curry (2007), which started with a general one-dimensional mass conservation equation that is a simplified diffusion model of CH₄ in soil given by:

$$\frac{\partial \text{CH}_4}{\partial t} = -\frac{\partial J_{\text{CH}_4}}{\partial z} + \sum R \quad (1)$$

where J_{CH_4} is the flux of CH₄ that is generally controlled by diffusion and is the sum of processes that produce and consume CH₄. Based on this first equation, a significant improvement in the mathematical representation for complete distribution of CH₄ into soil and a refined parameterization of the influence of soil moisture, temperature, and nitrogen inputs, are presented in Murguía-Flores et al. (2018), as well as a detailed description of MeMo, the model code and model output for 1990–2009 in a supplemental file.

Figure 1. Methanotrophy Model (MeMo) consists of two parts: (1) CH₄ diffusion into soil (D_{CH_4}) controlled by physical soil properties, water content and temperature, and (2) microbial CH₄ oxidation (k_d) determined from a based oxidation rate (for an uncultivated and moist soil at 0°C) modified by temperature, soil moisture and nitrogen inputs.

A schematic description of Memo is provided in Figure 1. MeMo calculates the soil uptake flux by analytically solving the vertically resolved reaction-diffusion equation, which accounts for (a) CH₄ diffusion into soil (D_{CH_4}), controlled by physical soil structure properties (G_{soil}), which are influenced by soil porosity, soil moisture (water filled pore space %), clay content, and by temperature (G_T); and (b) microbial CH₄ oxidation (k_d) calculated from a base oxidation rate (for an uncultivated and moist soil at 0°C), which is modified by temperature (r_T), with optimum conditions at 28°C moderated by a Q_{10} equation; soil moisture (r_{SM}), with an optimum moisture content at 20% with decreasing rates of CH₄ uptake above and below that level; and nitrogen inputs (r_N), via fertilizers and deposition.

2.2. Model Validation

A comprehensive, global model-data and a model-model comparison were reported in Murguía-Flores et al. (2018), which demonstrated good agreement between simulation results and global observations from a wide range of ecosystems. In model-model comparisons, MeMo simulated a global average annual CH₄ uptake of 33.5 ± 0.6 Tg CH₄ yr⁻¹ for the period 1990–2009, which agrees well with the range 20–38.1 Tg CH₄ yr⁻¹ from other modeled global estimates (Potter et al., 1996; Ridgwell et al., 1999; Spahni et al., 2011; Curry, 2007; Zhuang et al., 2013; Tian et al., 2015). In addition, MeMo estimations are similar to the atmospheric inversion estimate of 30 ± 15 Tg CH₄ yr⁻¹ from Hein et al. (1997), a value of 36 ± 23 Tg CH₄ yr⁻¹ based upon observational data from a recompile and meta-analysis by Dutaur and Verchot (2007), and global extrapolations of measurements yielding estimates of 28.7 and 29 Tg CH₄ yr⁻¹ reported by Dörr et al. (1993) and Smith et al. (2000), respectively.

In addition, Murguía-Flores et al. (2018) also provided a complete latitudinal model-data validation using observational data from Dutaur and Verchot (2007). The structural and parametric improvements in MeMo resulted in a better fit to observations and the lowest error in comparison to previous models. The improvement was especially evident when MeMo was used to simulate CH₄ uptake in wet tropical ecosystems where previous models markedly overestimated annual CH₄ uptake by soil.

In this work, we further validate MeMo temporally, by comparing annual output against the most up-to-date compilation of direct CH₄ soil uptake measurements for different forested ecosystems reported by Feng

Figure 2. The global rate of atmospheric CH₄ uptake by soil in Tg yr⁻¹ from 1900 to 2100. Soil uptake rate is simulated using the process model Methanotrophy Model (MeMo) based upon observational forcing data for the historical run (black) and CMIP5 output data for the four Representative Concentration Pathways (RCPs) for the period 2016 to 2100. Color shading represents the range of values calculated using a Monte Carlo method sampling different values of k_d and D_{CH_4} .

et al. (2020) (Figure S1). First, due to differences in spatial and temporal resolution between model output and observational data, we averaged observational points in the same gridcell and transformed the result to annual values. Second, we calculated the normalized mean bias (NMB) between every georeferenced point (with latitude and longitude) in the observational dataset and MeMo output, using the formula:

$$NMB = \frac{(\text{modelled} - \text{observed})}{\text{modelled}} \quad (2)$$

2.3. Model Scope

MeMo was run at a monthly temporal resolution and a $1^\circ \times 1^\circ$ spatial resolution for the historical period, which was divided into early (1900–1979) and late (1980–2015) intervals, and a future period, which was defined as 2016 to 2100. The boundary between the early and late historical intervals was set at 1980 because that was the year of maximum rate of change in global CH₄ uptake by soil (Figure 2). The future period begins at 2016 because historical data required to run the simulations are complete only up to 2015. Table 1 summarizes the forcing datasets for the different time periods.

2.4. Analyzed Model Output

2.4.1. Global Mean CH₄ Uptake Rate

We calculated the global annual mean CH₄ uptake rate for the two intervals in the historical period (1900–1979; 1980–2015) and the last 35 years of the future period (2065–2100 for RCPs 2.6, 4.5, 6.0, 8.5). We chose the

final 35 years of the future period to ensure consistency with the averaging period of the late historical period. We assessed the potential for soil methanotrophy to offset atmospheric CH₄ emissions by calculating the percentage of soil CH₄ uptake relative to total CH₄ emissions as defined by Ciais et al. (2013) for the historical periods and van Vuuren et al. (2011) for the future period.

2.4.2. Uncertainty Calculation

The rate of CH₄ uptake by soil is controlled by the balance between gaseous diffusion of atmospheric CH₄ into soil and the rate of CH₄ oxidation by methanotrophic bacteria. It is thus essentially controlled by two key parameters: the microbial CH₄ oxidation rate constant k_d , and the CH₄ molecular diffusion coefficient D_{CH_4} . The two key parameters are expressed as a function of a number of parameters that depend on climatic conditions and soil structure (Murguía-Flores et al., 2018). Parameters and their functional dependency on climatic conditions and soil structure have been constrained on the basis of field and laboratory studies (Figure S2). The resulting apparent parameters implicitly account for factors that are not explicitly

Table 1
Driving Data and Sources Used for the Historical and Future Simulations

Driver	Historical (1900–2015)	Future (2016–2100)
Temperature	CRU3.1 (Harris et al., 2014)	Mean of nine ESMs: CCSM4, GFDL-ESM2G, HadGEM2, IPSL-CM5A-LR, PSL-CM5A-MR, MIROC-ESM, MIROC-ESM-CHEM, NorESM1-M, NorESM1-ME (Taylor et al., 2012)
Soil moisture	Dorigo et al. (2011); Sitch et al. (2015)	
Atmospheric CH ₄ mole fraction	Rigby et al. (2008)	
Atmospheric N deposition	Lamarque et al. (2013)	Lamarque et al. (2013)
N fertilizer application	Nishina et al. (2017)	Nishina et al. (2017) extrapolated based on rate of change in nitrogen deposition
Bulk density and clay content	Shangguan et al. (2014)	Shangguan et al. (2014)
Tillage (agricultural area)	Chini et al. (2014)	Chini et al. (2014)

resolved in the model and thus, are associated with different degrees of uncertainty. We used a Monte Carlo approach to quantify uncertainty across all time periods. A temperature and soil moisture pair were randomly sampled from five CMIP5 models (CanESM2, IPSL-CM5A-LR, MIROC-ESM-CHEM, MPI-ESM-LR, and MPI-ESM-MR; Taylor et al., 2012) for each grid cell in each month for the historical, RCP2.6, RCP4.5, RCP6.0, and RCP8.5 simulations. The values were used to calculate gridded monthly key parameters, k_d and D_{CH_4} . Even though k_0 is a key parameter in calculating k_d , we used fixed values for specific ecosystems, and we could not calculate its uncertainty due to the scarcity of global data on this parameter. The effect of this parameter on CH_4 uptake were analyzed in Murguía-Flores et al. (2018). We then simulated CH_4 uptake for the particular condition using MeMo. The process was repeated 1000 times for each climate scenario. This analysis provides the potential range of variation in CH_4 uptake rates due to model and RCP variability.

2.4.3. Drivers of Global Changes in Soil CH_4 Uptake

We conducted a series of factorial experiments to determine the effect of each driver on the total change in global CH_4 uptake. For each experiment, one parameter was allowed to vary with time while the remainder were set to monthly values of the year that started the period (i.e., 1900 for the historical period and 2016 for the future period). We conducted 30 simulations in total: for each of the five scenarios (historical + four RCPs) we first ran simulations with all parameters varying simultaneously and then with each parameter (temperature, soil moisture, atmospheric CH_4 mole fraction, nitrogen input, and cropland tillage) varying individually.

For each simulation, the net change in the soil CH_4 sink during each time period was calculated as the year-to-year finite difference in the annual global rate of soil methanotrophy integrated over the period. This approach allowed us to quantify the contribution of each driver to the total change in uptake rate but did not account for interactions between drivers. However, simulation results showed that the sum of the individual driver contributions was similar to results obtained from simulations with the full suite of time varying parameters.

2.4.4. Spatial Distribution of CH_4 Uptake by Soils From 1900 to 2100

We calculated global gridded means of soil CH_4 uptake and differences were compared between future pathways and the late historical period to predict where substantive changes in soil methanotrophy are likely to occur as a result of global change.

2.4.5. Key Drivers of Change in the Spatial Distribution of Soil Methanotrophy

The dominant factor influencing the global distribution of methanotrophy in soil was assessed for each grid cell by dividing total uptake in the individual factorial runs (i.e., separately varying temperature, soil moisture, atmospheric CH_4 mole fraction, or nitrogen inputs) by the total uptake of the full factorial simulation (i.e., all factors varying). The variable that explained the greatest percentage of the trend was considered to be the dominant factor for each grid cell for the early and late historical and the end of the RCPs (2065–2100).

Mechanisms underlying each driver were explored using several approaches. The degree to which atmospheric CH_4 mole fraction influenced soil uptake rates was evaluated from the linear correlation between the two variables. Temperature effects were explored by (a) spatially evaluating the number of days that supported methanotrophic activity (i.e., when oxidation rates are not inhibited by low temperature and defined as $r_T > 0$, $r_T = \exp(0.1515 + 0.05238T - 5.946 \times 10^{-7}T^4)$ as explained in Murguía-Flores et al., 2018), (b) estimating the global area that previously did not support soil CH_4 oxidation due to frozen conditions, and (c) computing the difference between the simulated and the optimum temperature for methanotrophy. For soil moisture, Murguía-Flores et al. (2018) optimal CH_4 uptake by soil methanotrophy occurs at ~20% water content, and consequently, both an increase or decrease in water content will lead to a decrease in the rate of soil CH_4 uptake. We grouped spatial changes due to soil moisture into four categories: (a) positive changes in both soil moisture and CH_4 uptake, (b) negative changes in both soil moisture and CH_4 uptake, (c) positive change in soil moisture and negative change in CH_4 uptake, and (d) negative change in soil moisture and positive change in CH_4 uptake. The effect of nitrogen input on global uptake of CH_4 by soil was calculated based upon the change in cropland area between the late historical period and the RCPs.

3. Data or Model Forcing

For the historical simulations, MeMo was forced using global monthly observations of surface soil moisture (Dorigo et al., 2011; Sitch et al., 2015), surface temperature (CRU3.1, Harris et al., 2014), atmospheric CH₄ mole fraction (Rigby et al., 2008), gridded atmospheric nitrogen deposition (Lamarque et al., 2013), nitrogen input from fertilizers for croplands (Nishina et al., 2017) and soil bulk density and clay content (Shangguan et al., 2014). A tillage effect was parameterized as a 10% reduction in bulk density driven by aeration and a short-term increase in porosity (Bauer & Black, 1981; Dam et al., 2005; Grant & Lafond, 1993; Lampurlanés & Cantero-Martínez, 2003; Osunbitan et al., 2005; Unger & Jones, 1998). The tillage factor was imposed on agricultural area fraction in each grid cell based upon data from Chini et al. (2014). We used a vegetation mask (Ramankutty & Foley, 1999) to map the previously calculated ecosystem-specific values for k_0 in the steppe ($k_0 = 3.6 \times 10^{-5}$), temperate forest ($k_0 = 4.0 \times 10^{-5}$), tropical forest ($k_0 = 1.6 \times 10^{-5}$), and a general value for the remaining ecosystems ($k_0 = 5.0 \times 10^{-5}$) for all the periods (Murguía-Flores et al., 2018). Specific and general values for k_0 were constrained using available observational data. The calculations are described fully in Murguía-Flores et al. (2018).

For the future simulations, MeMo was forced by RCPs 2.6, 4.5, 6.0, and 8.5 (van Vuuren et al., 2011). Soil moisture, temperature, and atmospheric CH₄ mole fraction were extracted from the mean of nine Earth System Models (ESM), which simulate the effect of both anthropogenic and natural emissions into the future for each RCP (Taylor et al., 2012). Atmospheric nitrogen deposition rates were taken from Lamarque et al. (2013) and nitrogen input to soil via fertilizers and manure was assumed to change at the same rate as atmospheric nitrogen deposition in each RCP (Galloway et al., 2003). Soil bulk density and clay content values were kept the same as the historical period (Shangguan et al., 2014) except when altered by the tillage factor, which was applied to future predicted cropland areas (Chini et al., 2014), which varied with time using the same formulation as the historical period.

4. Results

Changes in global soil methanotrophy from 1900 to 2100 for all simulations are shown in Figure 2. During the early historical period, CH₄ uptake by soil increased globally from 17.1 ± 2.4 to 30.8 ± 2.7 Tg yr⁻¹, which corresponds to an annual rate of change of 0.16 ± 0.03 Tg yr⁻². During the late historical period, the annual uptake rate increased by 0.19 ± 0.05 Tg yr⁻² leading to a mean global uptake of 37.2 ± 3.3 Tg yr⁻¹ in 2015. The simulated changes in global soil methanotrophy for the period 1900–2015 are consistent with previously published estimates of an increase in global soil CH₄ uptake from 18 to 32 Tg yr⁻¹ between 1900 and 2000, based upon a process-based simulation forced with Terrestrial Ecosystem Model input and observational climate data (Mitchell & Jones, 2005; Zhuang et al., 2013). MeMo simulation results also agree well with the recent global median and range provided by the global methane budget for the period 2000–2017 (Saunio et al., 2020) of 30 (11–49) Tg CH₄ yr⁻¹ and with the modeled global mean uptake rate of 33.5 ± 5.0 Tg CH₄ yr⁻¹ for the period 2000–2009 estimated by Tian et al. (2015) using the Dynamic Land Ecosystem Model (DLEM), Spahni et al. (2011) using LPJ-WHyMe DGVM, and Curry (2007).

For the period 2016–2100, MeMo simulations predict significant differences in CH₄ uptake dynamics based upon the four future pathways. In the most extreme climate change pathway (RCP8.5), global uptake of atmospheric CH₄ by soil increases continuously from 35.9 ± 2.5 Tg yr⁻¹ in 2016 to 82.7 ± 4.4 Tg yr⁻¹ in 2100, which corresponds to a rate of change of 0.5 ± 0.01 Tg yr⁻². In RCP8.5, global soil CH₄ uptake increased by 230% by 2100 relative to uptake rates in 2015 or by 483% compared to the year 1900. In contrast, the simulation based upon the most conservative climate change pathway (RCP2.6) produced a continuous decrease in soil uptake of atmospheric CH₄. The global uptake rate decreased to 25.7 ± 2.5 Tg yr⁻¹, which is a rate of change of -0.11 ± 0.03 Tg yr⁻² and a 31% reduction from rates of soil CH₄ uptake in 2015. The intermediate climate change pathways RCP4.5 and 6.0 simulations predict increases in global CH₄ uptake by soil until the mid-21st century, reaching maximum uptake rates of 38.1 ± 1.3 and 41.6 ± 2.2 Tg yr⁻¹ in years 2051 and 2079, respectively, followed by decreases to rates that are similar to values in 2016 (34.9 ± 2.5 and 35.1 ± 2.4 Tg yr⁻¹, respectively). As a consequence, the overall net rate of change in CH₄ uptake by soil globally by year 2100 is small (-0.01 ± 0.01 and -0.006 ± 0.03 Tg yr⁻², respectively) for the intermediate pathways (Figure 2).

Table 2

Proportion of Total CH₄ Emissions Consumed by Soil Methanotrophy During the Early and Late Historical Periods, and the Late 21st Century for the Four Representative Concentration Pathways (RCPs)

Period	Total CH ₄ emissions (Tg yr ⁻¹) (Meinshausen et al., 2011)	Soil uptake (Tg yr ⁻¹)	% CH ₄ uptake of total emissions
1900–1979 Early historical	470 ± 104	22.2 ± 3.8	4.7 ± 0.8
1980–2015 Late historical	550 ± 68	34.9 ± 1.9	6.3 ± 0.4
2065–2100 RCP2.6	366 ± 33	26.3 ± 2.2	7.1 ± 0.7
2065–2100 RCP4.5	480 ± 47	35.4 ± 2.6	7.3 ± 0.6
2065–2100 RCP6.0	542 ± 62	39.9 ± 2.7	7.6 ± 0.2
2065–2100 RCP8.5	1273 ± 143	76.0 ± 3.8	5.9 ± 0.4

Note. Values are the mean and one standard deviation variability for each period.

The fraction of CH₄ emissions that has been consumed by soil methanotrophy has changed with time (Table 2). Global soil methanotrophy has consumed 4.7% of total CH₄ emissions from 1900 to 1979 and then increased to 6.3% during the period 1980 to 2015. In the RCPs, the fraction consumed is predicted to range between 5.9% and 7.6% of total CH₄ emissions.

4.1. Drivers of Change

Relative changes in CH₄ uptake rate and the contribution of individual drivers (atmospheric CH₄ mole fraction, temperature, soil moisture, and nitrogen input) are shown in Table S1 and Figure 3. During the historical periods, soil uptake of atmospheric CH₄ increased by 13.2 ± 2.1 Tg yr⁻¹ from 1900 to 1979 and by 6.9 ± 1.6 Tg yr⁻¹ from 1980 to 2015. Model results indicate that the increased strength of the soil sink was driven primarily by an increase in atmospheric CH₄ mole fraction; however, during the late historical period and under future climate change pathways other drivers appear to begin to exert a greater influence on changes in soil methanotrophy. For example, during the early historical period increased

temperature explains only 1.3 of the 13.2 Tg yr⁻¹ of total change in CH₄ uptake rates in contrast to 1.2 of the 6.9 Tg yr⁻¹ of simulated change for the late historical period and 2.4 of the 0.7 Tg yr⁻¹ of total change (there are offsetting effects that result in the 0.7 Tg yr⁻¹ net value, but almost half of the change in uptake is driving by the change in temperature) for RCP6.0 (Figure 3). Additionally, during the early historical period, increased nitrogen input to soil inhibited global uptake of atmospheric CH₄ by −0.5 Tg yr⁻¹. Under different RCPs, the absolute amount of inhibition due to nitrogen loading is predicted to remain similar, ranging from −0.1 to −0.6 Tg yr⁻¹, except in RCP4.5, in which a decrease in nitrogen input explains an increase of 1.1 Tg yr⁻¹ in the global uptake. The impact of tillage is sufficiently minor that relative changes to global CH₄ uptake rates are negligible in all historical and future pathways, accounting for less than 0.1% of change in soil CH₄ uptake rates.

The amount of future change in the global soil sink for atmospheric CH₄ will depend strongly on the climate change pathway. For example, for RCP2.6 and RCP4.5, simulations indicate a decreasing strength of the CH₄ sink that amounts to −11.4 ± 2.4 Tg yr⁻¹ and −1.0 ± 2.2 Tg yr⁻¹, respectively for the period 2016 to 2100. For RCP2.6, the decrease in soil uptake is driven mainly by the projected decrease in atmospheric CH₄ mole

Figure 3. Simulated change in global soil CH₄ uptake in Tg yr⁻¹ for the historical periods and four representative concentration pathways (RCPs). The net change during each period is indicated by the value in Tg yr⁻¹ on top of each bar. Each bar is divided into the changes attributed to the four main factors that drive soil CH₄ uptake: atmospheric CH₄ mole fraction, soil temperature, soil moisture, and nitrogen input via atmospheric deposition and fertilizer application.

fraction, which accounts for -12.1 Tg yr^{-1} , with the small positive difference in the net simulated change resulting from the opposing effects of temperature and nitrogen inputs. For RCP4.5, the predicted change is small because of a balance between the negative impact ($-4.5 \text{ Tg CH}_4 \text{ yr}^{-1}$) of the decreasing atmospheric CH_4 mole fraction and the positive influence of temperature and nitrogen inputs, which increase soil uptake of CH_4 by 2.1 and 1.1 Tg yr^{-1} , respectively (Table S1 and Figure 3).

In contrast to RCP2.6 and 4.5, RCP8.5 simulations predict an increase in global soil CH_4 uptake of $46.7 \pm 3.8 \text{ Tg yr}^{-1}$ for the period 2016 to 2100. Similar to the early and late historical periods and RCP2.6 and 4.5, the main driver of this increase in soil uptake of CH_4 in RCP8.5 is the increase in atmospheric CH_4 mole fraction, which explains 42.7 of 46.7 Tg yr^{-1} of the predicted change in RCP8.5. An increase in global temperature accounts for a further 3.9 of 46.7 Tg yr^{-1} of change.

For RCP6.0, the change in atmospheric CH_4 mole fraction is still important; however, it had a negative impact and explains -1.62 of the net 0.7 Tg yr^{-1} of change in CH_4 uptake. In contrast, the projected increase in temperature accounts for 2.4 of the 0.7 Tg yr^{-1} of the change in soil CH_4 uptake, thus becoming the dominant driver of change for the first time.

The spatial distribution of global soil CH_4 uptake is shown in Figure 4 for the late historical period (Figure 4a), each of the four RCPs (Figures 4b, 4d, 4f, and 4h) and the differences between late historical period and each RCP (Figures 4c, 4e, 4g, and 4i). For RCP 2.6, a decrease in soil CH_4 uptake is predicted for all regions driven by a decrease in atmospheric CH_4 mole fraction (Figure S3). In contrast, a widespread global increase in the soil CH_4 sink is simulated in RCP8.5 because of a significant increase in atmospheric CH_4 mole fraction. Regional patterns of soil methanotrophy are more complex for the intermediate climate change pathways (RCP4.5 and 6.0), which predict annual CH_4 uptake rates by soil at the end of the 21st century that are similar to the present day (Figure 2). The spatial pattern of predicted change in soil methanotrophy is similar for both RCP 4.5 and 6.0 with an increase in soil uptake of CH_4 at high latitudes and in wet regions of South America, and a decrease in soil CH_4 uptake rates in central and south Africa, central Australia and east Asia.

4.2. Spatial Evaluation of Drivers of Change in Soil CH_4 Uptake

Regional evaluation of drivers of change in soil uptake of atmospheric CH_4 reveals that the relative influence of the different factors has shifted during the different periods examined in this study (Figures 5 and S4). During the early historical period, the increase in atmospheric CH_4 mole fraction exerted the main control on change in soil uptake of CH_4 globally (Figure 5a). However, during the late historical period, temperature became an increasingly important control on soil CH_4 uptake in high latitudes, while increasing nitrogen inputs from intensifying agriculture began to exert a greater influence on soil methanotrophy in regions such as India, China, and eastern Europe (Figure 5b).

In all RCP simulations, changes in soil uptake in the Northern Hemisphere are mainly controlled by temperature. The spatial extent of this high latitude temperature control on soil methanotrophy is most prominent in RCP4.5 and 6.0. For RCP2.6 and 8.5, changes in soil CH_4 uptake in all other regions are mainly controlled by changes in the atmospheric mole fraction. However, the intermediate RCP scenarios reveal a more complex pattern. In these scenarios, soil moisture effects dominate changes in the soil CH_4 sink in South America while increased input of nitrogen to soil is the key driver of shifts in soil CH_4 uptake rates in India, East Asia, North America, and parts of Europe.

5. Discussion

Simulations conducted using the MeMo model confirm previous reports that temporal changes in global uptake of atmospheric CH_4 by soil during the 20th century have been controlled mainly by increases in atmospheric CH_4 mole fraction. However, our model results also indicate that temperature began to exert a greater influence regionally on soil methanotrophy during the late historical period and that it is likely to continue to be an important driver of future changes in soil CH_4 uptake under all future RCPs particularly in RCP6.0. This finding is consistent with outcomes reported by Zhuang et al. (2013).

Figure 4. Simulated gridded mean soil CH₄ uptake rates for (a) late historical and late 21st century, forced with (b) RCP2.6, (d) RCP4.5, (f) RCP6.0 and (h) RCP8.5. Simulated differences in soil CH₄ uptake between the late historical period and each representative concentration pathway (RCP) are shown in panels (c), (e), (g), and (i). Panels on the left represent the temporal means of the last 35 years in each period. Panels on the right represent the difference between the gridded means in the last 35 years in each period minus the late historical.

The atmospheric mole fraction of CH₄ remains important for establishing the CH₄ concentration gradient in soil and the diffusive supply of CH₄ substrate for soil methanotrophs. Model results confirm a linear relationship between atmospheric CH₄ mole fractions and rates of soil CH₄ consumption ($r = 0.84$) (Figures S3 and 2) consistent with previous studies (Lau et al., 2015; Nesbit & Breitenbeck, 1992). Notably, Whalen and Reeburgh (1990) observed that rates of CH₄ uptake in soil increased continuously even to a CH₄ mole fraction of 500 ppm, which suggests that the capacity of soil methanotrophs to consume atmospheric CH₄ is considerable.

Figure 5. Spatial attribution of the main drivers of change in soil uptake in the (a) early historical (1900–1979) and (b) late historical (1980–2015) periods, and from 2065 to 2100 based upon (c) RCP2.6, (d) RCP4.5, (e) RCP6.0, and (f) RCP8.5. Color denotes the driving factor that accounts for the largest proportion of change in the temporal trend of soil CH_4 uptake rate in each grid cell. The absolute change in CH_4 uptake by soil can be either positive or negative.

However, we show that changes in atmospheric CH_4 mole fraction do not fully explain observed regional changes in soil uptake of CH_4 . Soil CH_4 uptake exhibits an increased response to drivers other than atmospheric CH_4 mole fraction in regions that are predicted to be impacted to a greater extent by climate or land-use changes (e.g., the Arctic or wet forests in South America or East Asia). The response is particularly evident in RCPs 4.5 and 6.0 across the high latitudes and wet tropics. The increased importance of factors such as temperature and soil moisture can be explained by a change in the balance between the rates of microbial CH_4 consumption and CH_4 diffusion into soil (Figure S5). Diffusion of CH_4 across the atmosphere-soil interface is controlled primarily by concentration gradient and to a lesser extent by the apparent diffusion coefficient, which varies as a function of temperature, soil structure and water content. In contrast, the rate of CH_4 oxidation is determined by bacterial consumption of CH_4 , which is expressed via a rate constant that is controlled by temperature, soil moisture, nitrogen input, ecosystem type, and soil CH_4 concentration. Thus, the response of CH_4 uptake flux to changes in environmental conditions reflects the combined response of transport and microbial processes to environmental perturbations (Von Fischer et al., 2009). Increases in atmospheric CH_4 mole fraction will result in higher rates of soil CH_4 uptake only if methanotrophic bacteria can efficiently oxidize the additional substrate (Malghani et al., 2016), which MeMo simulations suggest will occur in soil at high northern latitudes and wet tropical South America (Figures 4 and S5). In contrast, regions of central Africa show a decrease in soil uptake of atmospheric CH_4 because excess moisture limits

the ability of soil methanotrophs to respond to an increased substrate supply provided by a higher atmospheric CH_4 mole fraction. These findings are supported by observational data, including a report by Ni and Groffman (2018) that CH_4 uptake by forest soils in the north-eastern United States decreased by ~60% as a result of increases in precipitation during the last 20 years. Although atmospheric CH_4 mole fraction has increased over a two-decade period, soil CH_4 uptake did not respond proportionately to the increased substrate supply due to high soil moisture conditions. Yu et al. (2017) reported that CH_4 uptake in cool temperate soil and polar and boreal grasslands increased during the period 1980–2010 due to increases in temperature, while soil CH_4 uptake rates decreased in tropical dry grasslands in response to an increase in precipitation during the same period. Thus, the biological component of the soil sink for atmospheric CH_4 can exert an important control in addition to atmospheric CH_4 mole fraction.

Temperature is the dominant controlling factor of global soil methanotrophy rates in RCP6.0. We propose three explanations to account for the increased role of temperature in global uptake of atmospheric CH_4 by soil: (a) increased rates of methanotroph activity, (b) a larger active area of CH_4 uptake by soil due to a reduction in permanently frozen ground, and (c) an increase in duration of the active season, in particular, at high latitudes.

Microbial enzymatic activity increases as a function of temperature and in the case of soil methanotrophs, several studies have shown that the optimum temperature for CH_4 oxidation is ~28°C (Castro et al., 1995; Czepiel et al., 1995; Whalen & Reeburgh, 1996). Thus, for regions where soil methanotrophy presently is operating at below optimal rates (e.g., the north temperate zone during winter) future increases in temperature will enhance rates of CH_4 uptake assuming that diffusive flux of atmospheric CH_4 into soil is sufficient to fulfill the consumption capacity of methanotrophs (Figure S6). Notably, there will be offset for this effect from regions that are currently functioning at or above the optimum temperature for methanotrophy. For example, in the tropics an increase in temperature could result in a decrease in uptake rate of atmospheric CH_4 if the 28°C optimum is surpassed.

Second, the global area over which soil methanotrophy can function is estimated to increase by 2.0%–5.4% in different RCPs as a result of loss of ground and surface ice (Figure S7). Extensive areas at high latitudes and higher elevations that are currently frozen year-round are predicted to become suitable habitat for high affinity methanotrophic bacteria, increasing the active area of the global soil CH_4 sink. However, such regions could also become sources of CH_4 and as this model does not simulate CH_4 emissions to estimate the net flux, further investigation is necessary on CH_4 dynamics in these regions.

Third, higher global temperatures in the different RCPs are predicted to increase the number of days in which soil temperature keeps soil methanotrophy active, in particular, in the Northern Hemisphere, extending the seasonal activity of soil methanotrophic bacteria (Figure S8). Depending on the RCP, seasonal lengthening is estimated to add between 14 and 62 days per year of soil CH_4 uptake at high northern high latitudes and globally between 6 and 36 additional days. These predictions are consistent with observations reported by L. Zhao et al. (2004) of a decrease of >20 days from 1967 to 1997 in the duration of seasonally frozen ground on the Tibetan Plateau and simulations conducted by Lau et al. (2015) that predict seasonal lengthening of 5–30 days in the Arctic during the next century.

Decreases in nitrogen input (primarily in RCP4.5) are predicted to dominate changes in soil uptake of atmospheric CH_4 in relatively small areas globally where forested land is expected to expand from its current state (Thomson et al., 2011) (Figure S9). When nitrogen input to soil is reduced in MeMo, uptake of atmospheric CH_4 increases immediately; however, several studies have shown that increased consumption of CH_4 by soil after cessation of agricultural activity takes months to years (Hütsch et al., 1994; King & Schnell, 1994; Mosier et al., 1991; Tate, 2015; Yue et al., 2016). These observations suggest that while a reduction in nitrogen input to soil will eventually increase rates of soil CH_4 uptake the recovery rate is likely to be slower than estimated by model simulations. The time required for soil to return to natural rates of CH_4 uptake after anthropogenic addition of nitrogen is discontinued remains a significant unknown for most ecosystems and agricultural soils.

An increase or decrease in soil moisture can have either a positive or negative effect on soil methanotrophy because CH_4 uptake is consistently optimal at 20 vol% H_2O content in different ecosystems worldwide and under laboratory conditions (Adamsen & King, 1993; Boeckx & Van Cleemput, 1996; Burke et al., 1999;

Castro et al., 1995; Epstein et al., 1998; Klemetsson & Klemetsson, 1997; McLain & Ahmann, 2008; Mosier et al., 2002; Van den Pol-van Dasselaar et al., 1998; West et al., 1999). A decrease in soil moisture in an arid region typically will diminish soil uptake of atmospheric CH₄ while a decrease in soil water content in a wet region will enhance rates of soil methanotrophy (Ni & Groffman, 2018; J. F. Zhao et al., 2019). Simulations conducted in MeMo under the different RCPs suggest that future changes in soil moisture will have a small net impact globally on soil uptake of atmospheric CH₄ (Figure 3). This finding is consistent with the report of Zhuang et al. (2013) that changes in soil moisture had minimal influence on global soil CH₄ uptake rates during the last century. In future scenarios, soil moisture is predicted to decrease in most areas (Figure S10), resulting in strong positive and negative regional patterns of CH₄ uptake by soil but overall yielding a relatively minor net global change in the soil sink for atmospheric CH₄ in all RCPs (Figure S11).

It is important to note that assessing the impact of changes in soil moisture on CH₄ uptake by high affinity methanotrophic bacteria is complicated by significant disagreement between ESMs about the predicted direction and intensity of change in soil moisture under the different RCPs (Collins et al., 2013). The issue is particularly evident for RCP2.6 in which a high level of confidence and agreement for soil moisture predictions are achieved for less than 10% of global area. The most favorable agreement occurs for RCP8.5 but is still only approximately one-third of global soil area. Consequently, while the change in the global mean in soil moisture is small, the impact due to variance remains uncertain. There is evidence that changes in soil moisture could exert a significant control on CH₄ uptake by soil across large regions (Blankinship et al., 2010; Bowden et al., 1998; Christiansen et al., 2016; J. F. Zhao et al., 2019), in particular, in the Northern Hemisphere (Ni & Groffman, 2018), where the mean soil moisture state is already optimal for methanotrophy. However, more work is needed to improve predictions of change in soil moisture globally before its impact on soil methanotrophy can be assessed with confidence in process-based models such as MeMo.

6. Conclusions

This study demonstrates that rates of soil CH₄ uptake during the 20th century increased due to enhanced transport of CH₄ across the atmosphere-soil interface. However, in future RCPs, other environmental factors, which differ across regions, will begin to have a greater impact on soil CH₄ uptake rates. Findings from this study are relevant for CH₄ emission mitigation because they provide an assessment of the potential future behavior of the sole biological sink for atmospheric CH₄ under a range of RCPs. Future work should refine Earth System Models to include soil CH₄ uptake to assess the significance of potential feedbacks between environmental conditions, soil CH₄ uptake, and atmospheric CH₄ concentrations to more accurately predict future emission trajectories.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

The model code and a postprocessed driving data set to run an example model case study for the year 2000 are available online as the Supplement in Murguia-Flores et al., 2018: <https://doi.org/10.5194/gmd-11-2009-2018-supplement>. Additionally, all the forcing data is available online from the following sources: Temperature from CRU3.1 (Harris et al., 2014): <https://doi.org/10.1002/joc.3711>, 2014. Soil moisture from Dorigo et al. (2011) and Sitch et al. (2015): <https://doi.org/10.5194/hess-15-1675-2011>, 2011 and <https://doi.org/10.5194/bg-12-653-2015>, 2015. Atmospheric CH₄ mole fraction from Rigby et al. (2008): <https://doi.org/10.1029/2008GL036037>, 2008. Atmospheric N deposition from Lamarque et al. (2013): <https://doi.org/10.5194/acp-13-7997-2013>, 2013. N fertilizer application from Nishina et al. (2017): <https://doi.org/10.5194/essd-9-149-2017>, 2017. Bulk density and clay content from Shangguan et al. (2014): <https://doi.org/10.1002/2013MS000293>, 2014. Agricultural area from Chini et al. (2014): <https://doi.org/10.3334/ORNLDAAAC/1248>. RCP's (Taylor et al., 2012): <https://doi.org/10.1175/BAMS-D-11-00094.1>. Vegetation land mask from Ramankutty and Foley (1999): <https://doi.org/10.1029/1999GB900046>.

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