Reply to reviewer 1

We thank the reviewer for providing these comments and provide responses below. Reviewer comments are in bold followed by our responses.

5

Major comment 1: My first big concern is about correspondence between your solution of eq. (2) (using eqs. (3) and (6-7)) and boundary conditions. […] So suggested general solution does not satisfy both of used boundary conditions. eqs. (3) and (6-7)) and boundary conditions. […] So suggested general solution does not satisfy both of used boundary conditions.

10 We apologize for an error in the sign of the denominator in Eq. (7) that resulted in this comment. The correct expression for the integration constant B is:

$$B = \frac{C_{CH4} * exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right)}{\left[exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right) - exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right)\right]} \qquad (7)$$

15 Inserting A and B into Eq. (3) and solving Eq. (3) for z = 0 and z = L now yields the correct boundary conditions CH_4 (0) = C_{CH4} and CH_4 (L)=0. Equation 7 has been corrected in the revised manuscript. We thank the reviewer for bringing this error to our attention.

The incorrect equation also was present in the code, which produced L values that were too large.
20 However, because the majority of CH_4 is consumed at shallow depth in soil, the overestimation of L resulted in a <1% error in estimates of regional uptake of atmospheric CH_4. We have corrected the MeMo code and rerun the simulations. The changes have not significantly altered the modelling outcomes or conclusions of our study and all numbers have been updated throughout the manuscript.

25 We note also that use of the term "99.9% consumption" has been changed to 'complete consumption' throughout the manuscript.

2. My second big concern is rationality of building this model in its current state. I suppose that each new model should provide substantial improvement of available models. But in your paper
30 **only one class of available models is described and improved (models of Curry, Ridgwell, Potter; further CRP models). To my knowledge there are much better models of methane consumption (as example, Saggar et al., 2007; Zhuang et al., 2013). The main their advantage is description of methane consumption and soil methane diffusion not as constant along the soil profile (like in your model and CRP models) but as dependent on soil depth. These models also take into account**
35 **all environmental controls considered in your paper. So it is not correct to ignore them. Ridgwell, Potter; further CRP models). To my knowledge there are much better models of methane consumption (as example, Saggar et al., 2007; Zhuang et al., 2013). The main their advantage is description of methane consumption and soil methane diffusion not as constant along the soil**

profile (like in your model and CRP models) but as dependent on soil depth. These models also take into account all environmental controls considered in your paper. So it is not correct to ignore them.

That's why it is necessary:

5   **-   to tell that these models exist and to give their brief description**
       -   to explain why it is important to build a new model and why your model is better than others. This explanation is necessary to give comparing your model with CRP models too. You consider the same factors as CRP models, so what are the reasons to improve these models? Are CRV models or models from (Saggar et al., 2007; Zhuang et al., 2013) predict measured
10   **methane fluxes worse than your model or not good enough?**

We developed MeMo to be a process-based global model for simulating past, present and future uptake of atmospheric CH_4 by soil. We chose to build on the Potter et al. (1996), Ridgwell et al. (1999) and Curry (2007) (PRC) models because mechanistic simulation of global CH_4 consumption in soil could be
15 forced using data from past archives, modern records and future simulations of climate. We acknowledge that higher resolution models and more complex approaches presently exist for modelling soil methanotrophy, in particular, at a local scale; however, comprehensive global datasets that contain driving data at an adequate spatial and temporal resolution are not available (specific examples are discussed below and in text that has been added to the manuscript).

20

We have not provided detailed descriptions of non-PRC class models in the manuscript because MeMo builds on the PRC models and demonstrating the advances offered by MeMo was the focus our manuscript. The reviewer notes the Zhang *et al.* (2013) global model (hereafter referred to as 'Z13') for simulating soil uptake of atmospheric CH_4, which can be regarded as separate from the PRC class of
25 models. The general analytical solution used in Z13 is the same as Curry (2007; C07), which has been improved in MeMo; however, Z13 incorporates differences in its parameterization of microbial activity that are based upon redox potential and maximum rates of CH_4 consumption instead of using a base rate for CH_4 oxidation. The Z13 model also differs in that it employs modelled ecosystem-specific inputs for Q10 and optimum soil moisture; however, that complexity requires that Z13 operate within the Terrestrial
30 Ecosystem Model (TEM) because global data sets for parameters such as optimum soil moisture and redox potential are not available. In short, driving data for the PRC and MeMo models are only a portion of the input needed for Z13 simulations and consequently, it was not possible to conduct the same level of comparison between MeMo and Z13 that was conducted for the PRC models. However, we note that the Z13 model was not ignored in our original manuscript and that a comparison of global soil uptake of
35 atmospheric CH_4 simulated by Z13 and MeMo (and a range of other models) was provided in Table 7. The similarity of the global uptake results is a notable outcome despite differences in the modelling approaches used to simulate CH_4 uptake by soil. It is important to note, however, that our study is the first time that a soil methanotrophy model has been validated against global observations, highlighting the importance of accurately quantifying regional variations.

40

As stated by the reviewer there are biochemical models available at present that are more complex than MeMo (and Z13). These models (e.g., NZ-DNDC and XHAM) have been used to simulate CH_4 dynamics

at specific sites based upon coupled reaction transport equations that require highly depth-resolved local input data (e.g., Saggar *et al.*, 2007; Oh *et al.*, 2006; Sabrekov *et al.*, 2016). All of these models can be driven by depth-variable parameters when high resolution local data are available; however, the models are impractical for global simulations of soil methanotrophy because of the limited availability of the high resolution global data required to drive the models (e.g. rhizosphere depth, specific soil management, specific metabolic data, enzyme concentrations).

In summary, attributes of MeMo that advance the state of global simulation of soil uptake of atmospheric CH_4 are (i) its use of an analytical (more complete) solution to quantify the depth and maximum consumption of atmospheric CH_4, (ii) its ability to quantify the influence of internal CH_4 sources (e.g., methane produced in anoxic microsites in soil) on soil methanotrophy and the impact of autochthonous CH_4 on regional uptake of atmospheric CH_4 by soil linked to seasonal or inter-annual changes in soil moisture or temperature, and (iii) its standalone nature, similar to the PRC models which it is built upon, that eliminates the need to operate within more complex models that provide driving data, and (iv) a detailed validation of simulations both globally and regionally against currently available CH_4 uptake rates for soil methanotrophy.

We did not originally describe all available models in the manuscript because MeMo builds explicitly on the PRC class of soil methanotrophy models. However, we concur with the reviewer that the manuscript would be improved by noting how MeMo differs from these other types of models, in particular Z13. We recognize that addition of the new text is contrary to the second reviewer's recommendation that the manuscript be shortened. The following text has been added on page 3 beginning at line 20 (replacing text formerly from line 20 page 3 to line 19 page 4) to address the concerns raised by reviewer 1:

"Several detailed biogeochemical models have been developed to quantify consumption of atmospheric CH_4 by soil. Saggar et al. (2007) produced a modified version (NZ-DNDC) of DNDC (Li et al., 2000) to evaluate local impacts of changes in climate, soil properties, fertiliser management and grazing regimes on soil methanotrophy. Sabrekov et al. (2016) developed a process-based model of soil CH_4 uptake that also incorporates rhizosphere methanotrophy. Oh et al. (2016) developed a model (XHAM) that explicitly simulates high affinity methanotrophy and active microbial biomass dynamics. These models are driven by high resolution local data sets, which presents challenges for conducting global simulations of soil methanotrophy because of limited availability of input data necessary to drive the models (e.g., global rhizosphere depth, specific soil management, specific metabolic data, enzyme concentrations).

Previous global models included Potter et al. (1996) (hereafter referred to as model 'P96'), which estimates terrestrial uptake of CH_4 by calculating diffusive flux of atmospheric CH_4 into soil using a modified version of Fick´s first law. Ridgwell et al. (1999) (hereafter referred to as model 'R99') improved the P96 model by explicitly accounting for microbial CH_4 oxidation in soil. The R99 model quantifies CH_4 oxidation rates as a function of soil temperature, moisture and N content. The latter parameter was estimated using agricultural land area as a proxy for fertilizer application. Solution of the resulting one-dimensional diffusion-reaction equation was approximated semi-numerically assuming steady state conditions. Curry (2007) (hereafter referred to as model 'C07') employed a steady state

analytical solution of the one-dimensional diffusion-reaction equation and introduced a scalar modifier to account for the regulation of CH_4 oxidation rates by soil moisture and the impact of temperature below 0°C. The C07 model continued to use the R99 agricultural land area approximation to evaluate the effect of N loading on CH_4 uptake. The C07 model is employed as a reference model for the Global Carbon Project (Saunois et al., 2016) to estimate global CH_4 uptake in dynamic global vegetation models, such as the Lund-Potsdam-Jena model (LPJ-WHy-Me; Wania et al., 2010; Spahni et al., 2011).

The model of Zhang et al. (2013) (hereafter referred to as model 'Z13') employs the same steady state analytical solution as model C07; however, parameterization of microbial activity in model Z13 is based upon redox potential, ecosystem-specific inputs for Q10 and optimum soil moisture, and maximum rates of CH_4 consumption instead of a base rate for CH_4 oxidation. Consequently, model Z13 operates within the Terrestrial Ecosystem Model (TEM) that provides the necessary driving data because global data sets for many of these parameters are not available. If external data were available, model Z13 presumably could be operated independently of the TEM in a manner similar to models P96, R99 and C07. However, such a stand-alone application (i.e. decoupled from TEM) would require a new implementation or presumably significant modifications to the code.

We have chosen to focus on refining the R99 and C07 models because availability of new observational and experimental data present an opportunity to re-evaluate global simulations of soil methanotrophy based upon an enhanced version of these models. For example, new global datasets quantifying N deposition and N input via fertilizers now enable better representation of this key inhibitory effect on soil uptake of atmospheric CH_4 (Lamarque et al., 2013). In addition, a new global inventory of CH_4 uptake rates in soil (Duataur and Verchot, 2007) provides a means to better compare and valid model simulations.

3. Using L, the depth of total methane consumption, is good idea, but total methane consumption (or consumption up to 0.1% of atmospheric methane level) does not occur in natural upland soils. There is a certain threshold of methane consumption by microorganisms. Methane consumption stops if this threshold is reached because microorganisms cannot get enough energy by methane oxidation for cell maintenance (Stackhouse et al, 2017). According to literature methane concentration is never close smaller than 0.1 ppm in deep soil horizons and consumption declines to zero in deep soil layers – about 50-70 cm (Bender and Conrad (1992), Whalen et al (1992), Czepiel et al (1995), Priemé and Christensen (1997), Jensen and Olsen (1998)). To my knowledge biological consumption of methane was not ever investigated on a depth more than 1 m in upland soils. Threshold of consumption varies depend on ecosystem type, climate and is defined by oxidation efficiency of methanotrophs (see references above and Stackhouse et al, 2017).

That's why I think that this approach of using total methane consumption depth is not correct.

We thank the reviewer for this suggestion. It is straightforward to incorporate a CH_4 threshold, CH_4 min, in Eqs. 6 and 7 for the case CH_4 (L) = CH_4 min. In the original manuscript CH_4 min = 0 but it is now a variable that can be set in the model:

$$A = -\frac{C_{CH4} * exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right) - CH4min}{\left[exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right) - exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right)\right]} \quad (6)$$

$$B = \frac{-CH4\ min + C_{CH4} * exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right)}{\left[exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right) - exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right)\right]} \quad (7)$$

Eq. (8) becomes:

$$0 = -D_{CH4}\sqrt{\frac{k_d}{D_{CH4}}}\frac{\left(2\ C_{CH4} - CH4min * exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right) - CH4min * exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right)\right)}{\left[exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right) - exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right)\right]} - F_{CH4} \quad (9)$$

To evaluate the effect of using a CH_4 threshold of 0 or 0.1 ppm, we compared the two scenarios. Figure 1 shows that the difference in L between CH_4 (L) = 0 and CH_4 (L) = 0.1 ppm is ~5 cm, which will have a minimal impact on CH_4 uptake flux because the majority of CH_4 is consumed in the top 10 to 30 cm of soil. Based on these changes and analysis we made the following modifications to the manuscript:

1) Updated Eqs. (6), (7) and (9) in the text.
2) Added Figure R1 to the Supplementary file (page 1; Figure S1).
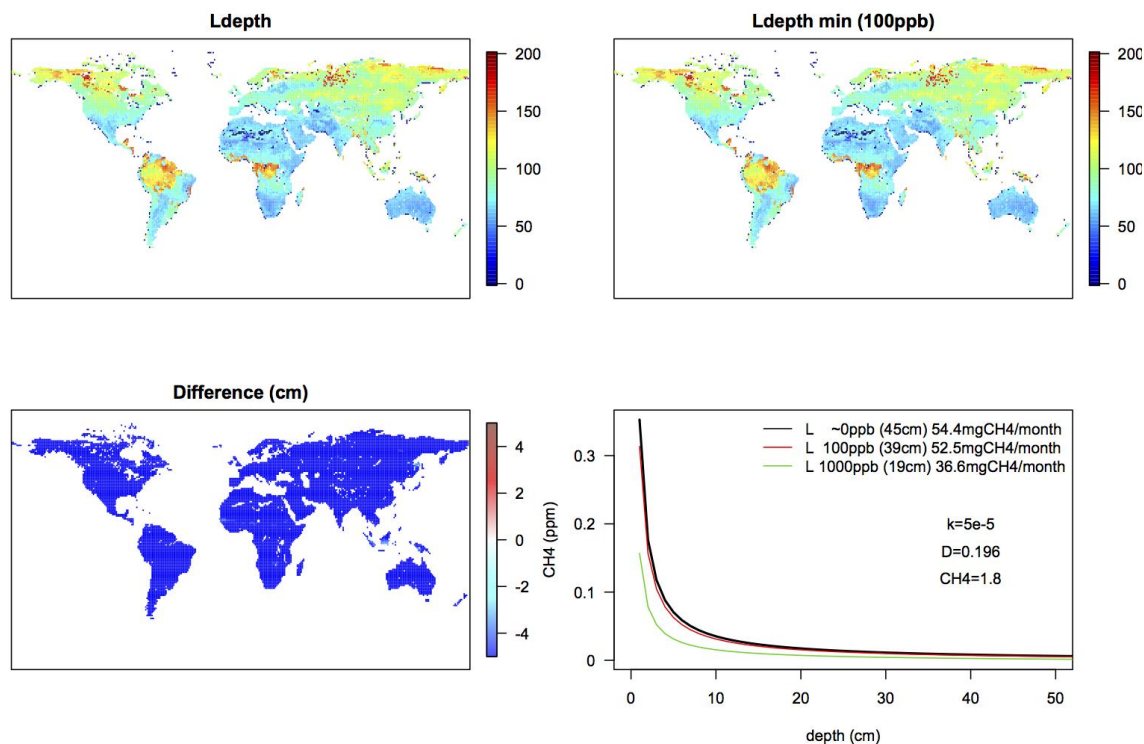3) Replaced Eq. (9) in the MeMo code for calculation of L.

Figure R1 (also Supplementary Fig S1): Comparison of model-derived depth L when $CH_4(L) = 0$ (top left), $CH_4 (L) = 0.1$ ppm (top right), the difference in depth L using the two approaches (bottom left), and CH_4 consumption profiles in soil and total uptake flux using fixed parameters of k, D and CH_4 (bottom right).

Under optimal conditions for methanotrophy, a CH_4 min = 0.1 ppm threshold results in a reduction in L of 6 cm (Supplementary Figure R1 bottom right panel); however, conditions for methanotrophy vary spatially and temporally, and hence use of the 0.1 ppm CH_4 threshold globally yields an average L reduction of 5 cm. The impact on CH_4 uptake rates is negligible because ~90% of atmospheric CH_4 entering soil is consumed within 10 cm of the ground surface. The effect on L size is important when CH_4 min is at least > 0.35 ppm, for example when CH_4 min =1.0 ppm the uptake flux decreases by ~57%.

Thus, the inclusion of a 0.1 ppm threshold for soil methanotrophy does not have an impact on the estimation of the global uptake of atmospheric CH_4, when compared with a scenario in which it is assumed that all CH_4 entering soil is consumed.

4. Page 5, rows 15-20. It would be better to give here any estimates, why only diffusive transport and biological consumption should be considered. What about convective transport? Is it always can be omitted? Why term on a right side of eq. (2) is not important? Are conditions really always steady state?

6

Several studies have shown that soil methanotrophy is limited by CH_4 diffusion and that advective fluxes (convective fluxes do not operate at this scale) play only a minor role in CH_4 under particular circumstances (Striegl, 1993; **Kruse et al., 1996**). Regardless, advective fluxes can be readily incorporated in the model (see below) although cannot be parameterized or constrained because of a lack
5   of driving data.

To incorporate advective flux in MeMo an additional advective term is added to the diffusion-reaction equation. Assuming the following boundary conditions: (i) $C(0) = C0$ and (ii) $\frac{dCH_4}{d_z}|_{z->\infty} = 0$ the solution of the advection-reaction-diffusion equation is given by:
10
$$J_{CH4} = -D_{CH4}(A * a + B * b) - wC$$

Where w is the advective velocity and C is defined as:

$$C(z) = A * \exp(a * z) + B * \exp(b * z)$$

15   Where:

$$a = \frac{\left(w - \sqrt{w^2 + 4 * D_{CH4} * k_d}\right)}{2 * D_{CH4}}$$

$$b = \frac{\left(w + \sqrt{w^2 + 4 * D_{CH4} * k_d}\right)}{2 * D_{CH4}}$$

20   Thus, if $w = 0$ the solution is Eq. 10 currently in the manuscript. Solution of the equation using different values of w yields the CH_4 depth profiles shown in Figure R2 below.
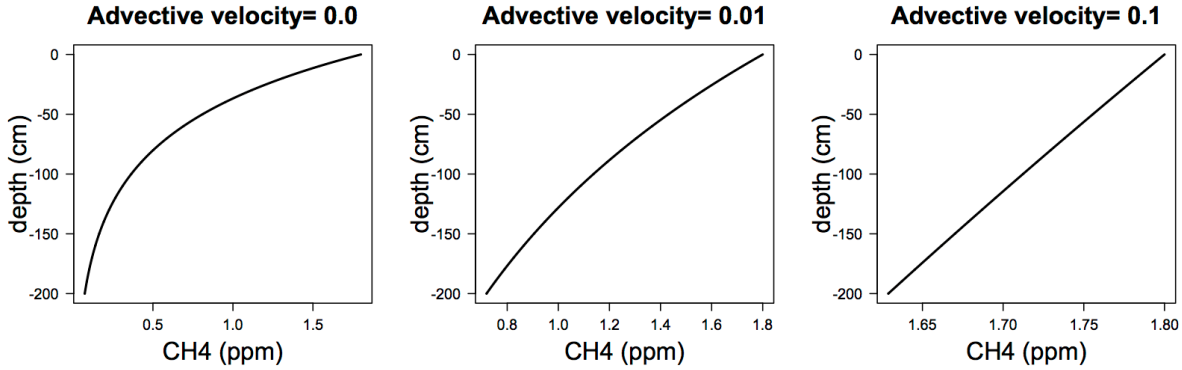

Figure R2: Calculation of CH_4 flux using different values of downward advective velocity (w). The depth (z) in soil at which CH_4 is 0.1 ppm occurs is shown in each panel.
25

The analysis shows that an advective velocity of 0.01 cm²/s can reduce the depth (L) of complete CH_4 consumption by up to 20% under optimal conditions. An advective velocity of 0.1 cm²/s (half the rate

of diffusion) can cause a decrease of up to 50%. However, as stated initially no data exist at present to parameterize or valid incorporation of advection into soil methanotrophy models.

4. Page 5, rows 15-20. Are conditions really always steady state?

It is reasonable to assume steady state conditions in global models such as MeMo because the timescale of boundary condition changes is long compared to the time required to attain steady state conditions in soil.

2. Page 10. What is the reason of using old Moldrup paper (same as in Curry paper) while there is much more recent and better soil gas diffusion model in (Moldrup et al., 2013)?

We have cited the Moldrup et al. (2013) paper in our work. While the authors evaluate several soil-diffusion models, it is important to note that their performance was only slightly better than the one employed in MeMo:

$$\frac{Dp}{Do} = \Phi^{4/3} \left(\frac{\Phi_{air}}{\Phi} \right)^{1.5+3/b}$$

The new version proposed by Moldrup et al. (2013) is:

$$\frac{Dp}{Do} = \Phi_{air}^2 \left(\frac{\Phi_{air}}{\Phi} \right)$$

Where Dp is the gas diffusion coefficient in soil (cm^3 air cm^{-1} soil s^{-1}), Do is the gas diffusion coefficient in free air (cm^2 air s^{-1}), Φ is total pore volume (cm^3 cm^{-3}), Φ_{air} is air-filled porosity (cm^3 cm^{-3}), b is a scalar that accounts for soil structure ($b = 15.9 f_{clay} + 2.91$).

The new version of the gas diffusion equation from Moldrup et al. (2013) provides only a marginal improvement in the RSME fit (0.017; Figure 2 in Moldrup et al., 2013) versus the model we used in MeMo (RSME=0.028). However, the main reason that we use this formulation of the equation is that the new model no longer includes the soil structure parameter (b) that accounts for the effects of clay content on gas diffusion in soil. Data for this parameter are available globally for different soil types which enables a more explicit assessment of the impact of soil texture on global uptake of atmospheric CH$_4$.

3. Page 30, row 11. Please fix, Sabrekov (like in list of references), not Savrekov.

This error has been corrected.

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Reply to reviewer 2

We thank the reviewer for providing these comments. Reviewer comments are shown in bold followed by our responses.

General comments

15 **1. In this manuscript, the authors present a new process-based model of upland soil oxidation by microbes, MeMo. They showed major results on global methane uptake, its latitudinal and spatial distribution, and seasonal change, in comparison with previous models by Potter et al. (1996), Ridgewell et al. (1999), and Curry (2007). I agree that global methane budget is gathering attention in terms of global climate change and so that the topic is timely.**

20

The manuscript provides a detailed description of basic concept and equations, mathematical solution, and environmental dependencies. I know that GMD accepts such a descriptive paper but still want to recommend shortening main text to some extent. The results presented in this manuscript are basic and lack scientific novelty; again, main text can be truncated by
25 **removing redundant statements of results in figures and tables.**

I'm not clearly sure what is the substantial advancement of the MeMO model, in comparison with previous models, because the new model used the similar framework for modeling soil methane oxidation. In fact, the estimated global total (34.3 Tg CH4/yr) is around the middle of
30 **the previous estimates (Table 7), and one apparent advantage is the better agreement with recent observations. In this regard, the low methane oxidation in humid tropics simulated by MeMO seems reasonable in comparison with previous ones.**

We thank the reviewer for highlighting the timely nature of our research. In order to convey to readers
35 how MeMo builds on earlier models it is necessary to provide details about the Potter et al, 1996, Ridgwell et al., 1999 and Curry, 2007 (PRC) class of models and the changes made in MeMo to enhance model capabilities. The equations that describe processes integrated into MeMo are not new

but nonetheless must be presented in order to explain how different facets of the model operate and interact. In short, the level of detail contained in the manuscript is necessary to convey the formulation and validation of MeMo.

Our study and the development of MeMo advances previous efforts to simulate global uptake of atmospheric CH_4 in the following ways:

    1. The performance of MeMo, and two previous PRC class models upon which MeMo is built, were evaluated against independent observational data (Figure 5 in the manuscript). This study is the first time that a global soil methanotrophy model has been evaluated in such a manner.

    2. MeMo simulates a total global soil sink for atmospheric CH_4 that is similar to previous models; however, refinements incorporated in MeMo improve the quality of regional simulations without calibration of the model to local data. For example, the new soil moisture and k_0 parameterizations in MeMo yield a significantly improved fit to observed CH_4 uptake rates in tropical and humid area soils, which other models presently overestimate (as noted by the reviewer).

    3. MeMo includes parameterization for the impacts of atmospheric N-deposition and application of N-fertilizer on soil methanotrophy (see response to comment 4). Models R99 and C07 both use agricultural land area as a proxy for application of N-fertilizer and attenuation of CH_4 uptake rates. Their approach was not validated rigorously against observational data, which resulted in attenuation rates that are significantly larger than field observations, and also did not account for atmospheric N-deposition.

    4. MeMo uses an analytical (exact) solution to quantify the depth and maximum consumption of atmospheric CH_4. This feature is particularly important for soil conditions (e.g., dry soil) where L is less than 50 cm (Supplementary 1, Figure S1). In such areas, previous models (C07) slightly overestimate CH_4 uptake fluxes because a larger fixed value of L has to be assumed.

    5. MeMo is capable of quantifying the influence of autochthonous CH_4 sources on rates of soil uptake of atmospheric CH_4. This feature provides the ability to examine the role of seasonal or inter-annual changes in soil moisture on soil methanotrophy as a result of anoxia and methanogenesis in finely textured soil

2. On the other hand, my serious concern is on the nitrogen limitation factor. The author seems to consider only atmospheric deposition, but in reality, fertilizer and manure input is much more important as nitrogen input into croplands. Previous models, Ridgwell et al. (1999) and Curry (2007), implicitly accounted for the effect by using land-cover data. If this is correct, the MeMo model underestimated the effect of nitrogen input on methane oxidation (as shown in Figure 9).

There are two different aspects of the R99, C07 and MeMo models that must be considered in responding to this comment: model parameterization and driving data.

Models R99 and C07 use land cover area as a proxy for N-fertilizer application and do not consider atmospheric deposition of nitrogen. The models are parameterized to inhibit soil uptake of atmospheric CH_4 by 75% of the agricultural land percentage per grid. Parameterization is based on data from a single location and those findings have not been replicated to date. Current data show only a 0.33% decrease in CH_4 uptake flux per g N m^{-2} y^{-1} (Figure 4 in manuscript). In contrast, parameterization in MeMo is based on multiple laboratory and field observations of the inhibitory effect of N addition on methanotrophy.

A more detailed assessment of the impact of N-fertilizer application on soil methanotrophy simulated using MeMo was planned for a future study; however, in response to the reviewer's comment we have incorporated the effects of fertilizer input and atmospheric deposition of nitrogen in the manuscript. The driving data for the former are global time-dependent inputs of N-fertilizer reported by Nischinia et al. (2017).

The new results (Figure R1) show that nitrogen input inhibits global soil methanotrophy simulated in MeMo by 4 to 5% compared to inhibition of 7.1% in models R99 and C07. Regionally, MeMo simulates 72% inhibition in Europe, China and India compared to a 75% reduction in soil methanotrophy in models R99 and C07. However, in regions where significant areas of agriculture land exist but rates of N-input are low (e.g. South America) the simulated attenuation of CH_4 uptake by soil becomes very different for models R99 and C07 compared to MeMo. In such areas models R99 and C07 continue to simulate an inhibition of 75% whereas MeMo responds to the lower rates of N-input, leading to an inhibition of only 5 to 10%.

The global impact of N inhibition on soil uptake of atmospheric CH_4 differs between models: 1.38 Tg y^{-1} in MeMo, 2.43 Tg y^{-1} in model R99, and 10.1 Tg y^{-1} in model C07. The much larger inhibition in model C07 results from the inhibition factor (r_N) residing outside of the parameterization for bacterial oxidation, which causes attenuation of soil methanotrophy to power of two relative to the formulation in model R99. As stated previously, a key benefit of the new formulation for N inhibition in MeMo relative to the earlier models is that the amount of inhibition simulated regionally and globally is consistent with field and experimental observations.
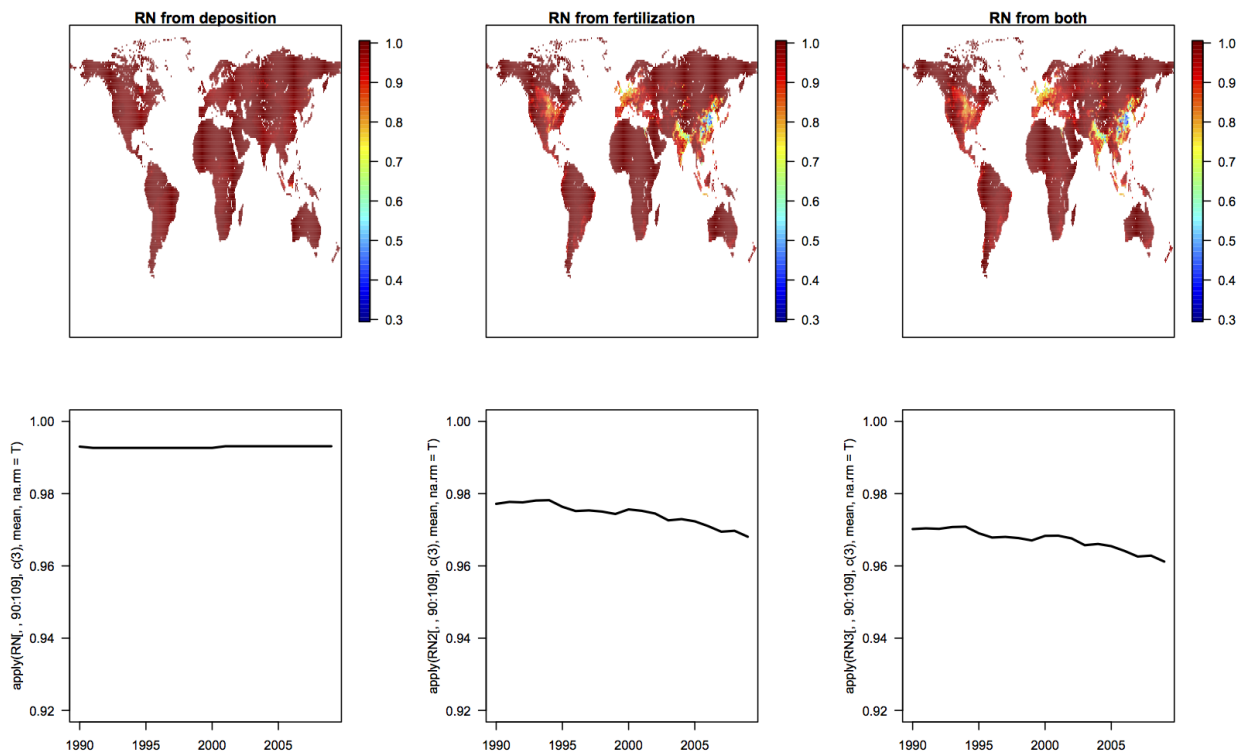
Figure R1: Nitrogen inhibitory effect (r_N) for atmospheric N-deposition (left), N-fertilizer application (middle), and both sources of N (right).

Overall, I conclude that the manuscript needs major revision and would be re- considered. I also recommend reinforcing discussion part with respect to implications to experimental and observational studies and potential impacts on climate projections and management.

In response to this comment we have incorporated new text in the manuscript on page 33, line 9:

"MeMo can be used to guide new field and laboratory experiments to address the lack of parameterization data, in particular, k0 and Q10 values for soil methanotrophy in different ecosystem and latitudes, and long-term *in situ* studies of N inhibition on CH_4 uptake by soil. It also can be used to compare results from short- and long-term investigations of CH_4 uptake in field and laboratory experiments."

On page 33, line 24 we added:

"Additionally, MeMo can be used to evaluate the impact of different proposed policies and mitigation strategies for managing the atmospheric burden and growth rate of CH_4 because of its capacity to

evaluate different future scenarios based upon parameterization of key drivers that impact rates of CH_4 uptake by soil globally."

Specific comments

1. Page 2 Line 20: Please cite more recent syntheses of global methane budget (e.g., Saunois et al., 2016, 2017)

We have included the references in the manuscript.

2. Page 8 Figure 1: Please show the atmospheric CH4 concentration for this example.

We have added the CH_4 concentration as requested.

3. Page 14 Line 1: "Grosso" should be "Del Grosso".

This reference has been corrected.

References

We have added these suggested references:

Saunois M, Bousquet P, Poulter B, Peregon A, Ciais P, Canadell JG, et al. The global methane budget: 2000–2012. Earth System Science Data 2016, 8: 697–751.

Saunois M, Bousquet P, Poulter B, Peregon A, Ciais P, Canadell JG, et al. Variability and quasi-decadal changes in the methane budget over the period 2000–2012. Atmo- spheric Chemistry and Physics Discussions 2017: doi:10.5194/acp-2017-5296.

Soil Methanotrophy Model (MeMo v1.0): a process-based model to quantify global uptake of atmospheric methane by soil

5

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Abstract.

Soil bacteria known as methanotrophs are the sole biological sink for atmospheric methane (CH_4), a powerful greenhouse gas that is responsible for ~20% of the human-driven increase in radiative forcing since pre-industrial times. Soil methanotrophy is controlled by a plethora of ~~different~~ factors, including temperature, soil texture and
20 **moisture or nitrogen content, resulting in spatially and temporally heterogeneous rates of soil methanotrophy. As a consequence, the exact magnitude of the global soil sink, as well as its temporal and spatial variability remains poorly constrained. We developed a process-based model (Methanotrophy Model; MeMo v1.0) to simulate and quantify the uptake of atmospheric CH_4 by soils on the global scale. MeMo builds on previous models by Ridgwell et al. (1999) and Curry (2007) by introducing several advances, including: (1) a general analytical solution of the one-dimensional**
25 **diffusion-reaction equation in porous media, (2) a refined representation of nitrogen inhibition on soil methanotrophy, and (3) updated factors governing the influence of soil moisture and temperature on CH_4 oxidation rates, and (4) the ability to evaluate the impact of autochthonous soil CH_4 sources on uptake of atmospheric CH_4.. We show that the improved representation of these key drivers of soil methanotrophy results ~~resulted~~ in a better fit to observational data. A global simulation of soil methanotrophy for the period 1990-2009 using MeMo yielded an average annual sink of**
30 **33.5~~34.3~~ ± 0.64~~.3~~ Tg CH_4 yr^{-1}. Warm and semiarid regions (tropical deciduous forest, ~~dense~~ and open shrubland) had**

the highest CH$_4$ uptake rates of 60**2**~~30~~ and 51**8**~~80~~ mg CH$_4$ m^{-2} y^{-1}, respectively. In these regions, favorable annual soil moisture content (~20% saturation) and low seasonal temperature variations (variations < ~6 ºC) provided optimal conditions for soil methanotrophy and soil-atmosphere gas exchange. In contrast to previous model analyses, but in agreement with recent observational data, MeMo predicted low fluxes in wet tropical regions because of refinements in ~~describing~~ formukation of the influence of excess soil moisture on methanotrophy. Tundra and **mixed**~~boreal~~ forest had the lowest simulated CH$_4$ uptake rates of 17**6**~~9~~ and 18**2**~~7~~ mg CH$_4$ m^{-2} y^{-1}, respectively, due to their marked seasonality driven by temperature. Global soil uptake of atmospheric CH$_4$ was **decreased** by **4**% **by the effect of nitrogen inputs to the system, however the direct addition of fertilizers attenuated the flux by 72% in regions with high agricultural intensity (*i.e*. China, India and Europe) and by 4-10% in agricultural areas receiving low rates of N-input (e.g. South America).** ~~,~~ Globally, nitrogen ~~deposition~~ inputs reduced soil uptake of atmospheric CH$_4$ by **1.38**~~0.34~~ Tg y^{-1}, which **is** two to **five**~~six~~ times smaller than reported previously. In addition to improved characterization of the contemporary soil sink for atmospheric CH$_4$, MeMo provides an opportunity to quantify more accurately the relative importance of soil methanotrophy in the global CH$_4$ cycle in the past and its capacity to contribute to reduction of atmospheric CH$_4$ levels under future global change scenarios.

1 Introduction

Methane (CH$_4$) is the most abundant organic trace gas in the atmosphere and responsible for approximately 20% of the human-driven increase in radiative forcing since preindustrial times (Myhre et al., 1998; Ciais et al., 2013). Anthropogenic activities during the last 200 years have increased the concentration of CH$_4$ in the atmosphere from pre-industrial levels of approximately 710 parts per billion (ppb) to the current mixing ratio of approximately 1800 ppb (Etheridge et al., 1998; Kirschke et al., 2013). The atmospheric lifetime of CH$_4$ is 9.1 ± 0.9 years (Prather et al., 2012) and most CH$_4$ is consumed in the troposphere via oxidation by OH radicals, which represents ~90% of the global CH$_4$ sink (Prather et al., 2012; Ciais et al., 2013). Soil bacteria known as methanotrophs consume ~9 to 10% of atmospheric CH$_4$ and a further ~1% is oxidized by reaction with chlorine radicals from sea salt in the marine boundary layer (Allan et al., 2007; Ciais et al., 2013).

Soil methanotrophy is the only biological sink for CH$_4$ and its rate is highly dependent on environmental conditions. The total global soil sink is similar in size to global emissions of CH$_4$ from rice paddies (Kirschke et al., 2013) and consequently year-to-year changes in factors that impact rates of soil CH$_4$ oxidation may contribute to variability in the interannual growth rate of atmospheric CH$_4$. Moreover, soil methanotrophy consumes up to 90% of CH$_4$ produced via methanogenesis in persistently or periodically wet soil and thus factors that impact soil uptake of atmospheric CH$_4$ may reduce the capacity of soil methanotrophs to attenuate emission of soil-produced CH$_4$ (Oremland and Culbertson, 1992; Singh et al., 2010).

The rate of methanotrophy in soil is controlled by several environmental factors including temperature, soil texture, moisture and nitrogen (N) content (Czepiel et al., 1995; Le Mer and Roger, 2001; Wang et al., 2005). The influence of these

factors on rates of CH$_4$ oxidation has been widely studied both at the ecosystem level and under laboratory conditions. Positive correlations have been consistently reported between temperature and rates of CH$_4$ oxidation in soil (Castro et al., 1995; Butterbach-Bahl and Papen, 2002; Rosenkranz et al., 2006; Luo et al., 2013). Atypically low and high soil moisture levels both have a negative impact on rates of atmospheric CH$_4$ consumption. A soil moisture content of ~20% appears to yield

5 optimum rates of CH$_4$ uptake in different ecosystems, including tropical forests, short grass steppe and tundra (Adamsen and King, 1993; Mosier, 2002; Burke et al., 1999; Castro et al., 1995; Epstein et al., 1998; Klemedtsson and Klemedtsson, 1997; McLain and Ahmann, 2007; West et al., 1999). Soil texture impacts the ability of soil to retain water and influences diffusion of atmospheric CH$_4$ and O$_2$ into soil because of its control on pore size and connectivity. Thus sandy soil generally exhibits higher rates of CH$_4$ uptake than silt-rich soil followed by clayey soil (Born et al., 1990; Dörr et al., 1993). The influence of N

10 input from atmospheric deposition and fertilizer application is more complex; however, the majority of studies report inhibition of soil methanotrophy with increased addition of N (Aronson and Helliker, 2010; Bodelier and Laanbroek, 2004; Fang et al., 2014).

There is a large year-to-year uncertainty in the accounting of the global CH$_4$ budget, particularly for processes that consume CH$_4$ (Kirschke et al., 2013). Our understanding of the main drivers of CH$_4$ uptake in soils and how those factors

15 respond to climate change is incomplete. Estimates of the soil CH$_4$ sink based upon field data (Dutaur and Verchot, 2007) show high variability globally and within different ecosystems. Numerical models provide an efficient means to deal with the spatial and temporal heterogeneity and to evaluate mechanistic understanding of physical and biological processes that influence soil methanotrophy. Ultimately, models enable derivation of regional and global estimates of soil uptake of atmospheric CH$_4$ and provide the ability to predict the response of soil methanotrophy to past and future global change. In

20 addition, they provide a platform of interdisciplinary knowledge synthesis, help identify the most important parameters and environmental controls and can thus inform future field and laboratory research.

Several detailed biogeochemical models have been developed to quantify consumption of atmospheric CH$_4$ by soil. Saggar et al. (2007) produced a modified version (NZ-DNDC) of DNDC (Li et al., 2000) to evaluate local impacts of changes in climate, soil properties, fertiliser management and grazing regimes on soil methanotrophy. Sabrekov et al. (2016) developed

25 a process-based model of soil CH$_4$ uptake that also incorporates rhizosphere methanotrophy. Oh et al. (2016) developed a model (XHAM) that explicitly simulates high affinity methanotrophy and active microbial biomass dynamics. These models are driven by high resolution local data sets, which presents challenges for conducting global simulations of soil methanotrophy because of limited availability of input data necessary to drive the models (e.g., global rhizosphere depth, specific soil management, specific metabolic data, enzyme concentrations).

30 Previous global models included Potter et al. (1996) (hereafter referred to as 'P96' model), which estimates estimated terrestrial uptake of CH$_4$ by calculating diffusive flux of atmospheric CH$_4$ into soil using a modified version of Fick´s first law. Ridgwell et al. (1999) (hereafter referred to as 'R99' model) improved the P96 model by explicitly accounting for microbial CH$_4$ oxidation in soil. The R99 model quantifies CH$_4$ oxidation rates as a function of soil temperature, moisture and N content. The latter parameter was estimated using agricultural land area as a proxy for fertilizer application. Solution of the

resulting one-dimensional diffusion-reaction equation was approximated semi-numerically assuming steady state conditions. Curry (2007) (hereafter referred to as 'C07'model) employed a steady state analytical solution of the one-dimensional diffusion-reaction equation and introduced a scalar modifier to account for the regulation of CH_4 oxidation rates by soil moisture and the impact of temperature below 0°C. The C07 model continued to use the R99 agricultural land area

5   approximation to evaluate the effect of N loading on CH_4 uptake. The C07 model has been employed as a reference model for the Global Carbon Project (Saunois et al., 2016) and has been used to estimate global CH_4 uptake in dynamic global vegetation models, such as the Lund-Potsdam-Jena model (LPJ-WHy-Me; Wania et al., 2010; Spahni et al., 2011).

        The model of Zhang et al. (2013) (hereafter referred to as model 'Z13') employs the same steady state analytical solution as model C07; however, parameterization of microbial activity in model Z13 is based upon redox potential, ecosystem-

10   specific inputs for Q10 and optimum soil moisture, and maximum rates of CH_4 consumption instead of a base rate for CH_4 oxidation. Consequently, model Z13 operates within the Terrestrial Ecosystem Model (TEM) that provides the necessary driving data because global data sets for many of these parameters are not available. If external data were available, model Z13 presumably could be operated independently of the TEM in a manner similar to models P96, R99 and C07. However, such a stand-alone application (i.e. decoupled from TEM) would require a new implementation or presumably significant

15   modifications to the code.

        We have chosen to focus on refining the R99 and C07 models because availability of new observational and experimental data present an opportunity to re-evaluate global simulations of soil methanotrophy based upon an enhanced version of these models. For example, new global datasets quantifying N deposition and N input via fertilizers now enable better representation of this ~~key~~ inhibitory effect on soil uptake of atmospheric CH_4 (Lamarque et al., 2013). In addition, a

20   new global inventory of CH_4 uptake rates in soil (Duataur and Verchot, 2007) provides a means to better compare and valid model simulations.

        Here we present an updated process-based model to quantity the global sink for atmospheric CH_4 by soil (hereafter referred to as 'MeMo': soil **Me**thanotrophy **Mo**del). MeMo ~~builds on the R99 and C07 models; however, it~~ is based on a general analytical solution of the one-dimensional diffusion-reaction equation, which makes obsolete the *a priori* assumption

25   of complete CH_4 consumption in the model domain applied in the C07 model. The refinement now also provides the opportunity to account for CH_4 flux from below (*i.e.*, due to CH_4 production in soil, if present) and to set a minimum methane concentration threshold~~s~~ at which metanotrophy can occur in the soil column. In addition, MeMo revisits and improves R99 and C07 model formulations to incorporate advances in the mechanistic understanding of soil methanotrophy that have resulted from availability of new data. Finally, MeMo utilizes for the first time data for atmospheric N deposition and N input from

30   fertilizers to explore more accurately the effect of land-use and land-use changes on the global CH_4 sink. We present a comprehensive description of the new model, a comparison of MeMo with the R99 and C07 models, and a critical discussion of model formulations and assumptions based on observational data. We then provide an assessment of global and regional soil uptake and variability across ecosystem types and seasons.

2.0 Model Description

The following sections provide a detailed description of MeMo in the context of existing global soil CH₄ uptake. Table 1 provides a summary of all terms, names and units used in the model description section, while Table 2 contains a short summary of the four global CH₄ uptake models ~~of~~based on the P96 family.

5

Table 1. Terms, names and units used in the model description section.

Terms	Name	Units
CH_4	CH₄ concentration	mg m⁻³
J_{CH4}	CH₄ flux uptake	mg CH₄ m⁻² mo⁻¹
C_{CH4}	Atmospheric CH₄ concentration	ppb
CH_4min	CH₄ threshold	ppb[AG1]
F_{CH4}	CH₄ flux through L	mg CH₄ m⁻² mo⁻¹
A and B	Integration constants	dimensionless
z	Depth in the soil profile	cm
L	Depth of 99.9% penetration of atmospheric CH₄ into the soil	cm
D_{CH4}	Diffusion coefficient of CH₄ into soil	cm² s⁻¹
k_d	CH₄ oxidation activity	s⁻¹
$D_{0CH4} = 0.196$	CH₄ diffusion in free air at standard temperature and pressure STP= 0°C and 1 atm pressure	cm² s⁻¹
G_T	Soil temperature response	°C
G_{soil}	Soil structure response	dimensionless
Φ	Total pore volume	cm³ cm⁻³
ρ	Bulk density	cm⁻³ g⁻¹
$ɗ = 2.65$	Soil particle density	g cm⁻³
Φ_{air}	Air-filled porosity	cm³ cm⁻³
θ	Soil water content	%
w	Saturation soil water potential	MPa
b	Clay soil content factor	dimensionless
f_{clay}	Clay soil content	%
k_0	Base oxidation rate constant for uncultivated moist soil at 0°C	s⁻¹
r_{SM}	Microbial CH₄ oxidation, soil moisture response	dimensionless
r_T	Microbial CH₄ oxidation, temperature response	dimensionless
r_N	Microbial CH₄ oxidation, nitrogen response	dimensionless
N_{soil}	Nitrogen deposition into soil	g N m⁻² mo⁻¹
$\alpha = 0.33$	Average coefficient of N deposition inhibition	% mol N⁻¹

2.1 Conservation Equation

10   The general, one-dimensional mass conservation equation for CH₄ in soil is given by:

$$\frac{\partial CH_4}{\partial t} = -\frac{\partial J_{CH4}}{\partial z} + \sum R \qquad (1)$$

Where J_{CH4} denotes the flux of CH$_4$ and ΣR is the sum of all production and consumption processes that affect CH$_4$ concentrations in soil. The flux J_{CH4} in the soil is generally controlled by diffusion. Consequently, the P96 model assumes that global uptake of atmospheric CH$_4$ by soil is diffusion limited and thus describes the soil CH$_4$ sink as a purely diffusive process (*i.e.*, $\sum R = 0$). However, CH$_4$ is consumed by microbial activity in the soil and the simplified diffusion model may thus underestimate total uptake of CH$_4$. Consequently, R99 extended the diffusion model by explicitly accounting for microbial oxidation of CH$_4$ through a first order rate expression. The resulting diffusion- reaction equation forms the basis of the R99 model, the C07 model and MeMo:

$$\frac{\partial CH4}{\partial t} = -D_{CH4} \frac{\partial^2 CH4}{\partial z^2} + k_d * CH_4 \tag{2}$$

Where D_{CH4} is the CH$_4$ diffusion coefficient and k_d the first-order rate constant for microbial CH$_4$ oxidation. Under steady-state conditions (*i.e.*, ∂CH$_4$/ ∂t=0), soil CH$_4$ uptake is controlled by the balance between diffusion of CH$_4$ into soil and the rate of microbial CH$_4$ oxidation. Hence, accurate characterization of D_{CH4} and k_d is essential for a robust quantification of CH$_4$ uptake by soil.

2.2 Solution of Reaction-Transport Equation

The R99 model solved Eq. (2) semi-numerically by (i) assuming steady-state, (ii) numerically approximating the diffusion term similar to the approach applied in the P96 model (Table 2, Eq. 11), and (iii) assigning CH$_4$ oxidation exclusively to a distinct soil layer of thickness ϵ at depth $z_d = 6$ cm (Table 2, Eq. 12). However, CH$_4$ consumption can occur throughout a soil profile and thus Eq. (12) (Table 2) may either overestimate or underestimate the CH$_4$ sink.

In the C07 model, Eq. (2) was solved analytically, providing a more accurate and mathematically robust estimate of CH$_4$ uptake Eq. (13) (Table 2). Assuming steady-state <u>conditions</u> and constant D_{CH4} and k_d throughout the soil profile, integration of Eq. (2) provides a general solution for determining CH$_4$ concentration at depth z in soil:

$$CH_4(z) = A * exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}z\right) + B\ exp\left(\sqrt{\frac{k_d}{D_{CH4}}}z\right) \tag{3}$$

Where A and B are integration constants that can be determined by setting upper and lower boundary conditions for the soil profile. The concentration of CH$_4$ at the soil-atmosphere interface is defined by the atmospheric concentration of CH$_4$ (C_{CH4}) and thus, a Dirichlet boundary (*i.e.*, fixed concentration) is applied at the upper boundary. Conditions at the lower boundary are more challenging to ascribe because the soil depth at which atmospheric CH$_4$ is completely consumed is not known *a priori*.

Negligible CH₄ flux through the lower boundary (C07 Solution)

The C07 model circumvents the problem by applying a homogenous Neumann (no-flux) condition at the lower model boundary: $\frac{dCH_4}{d_z}|_{z->\infty} = 0$

The application of this boundary condition allows derivation of the integration constants $A = C_{CH4}$ and $B = 0$, which simplifies Eq. (3) to:

$$CH_4(z) = C_{CH4} * exp\left(-\sqrt{\frac{k_d}{D_{CH4}}} * z\right) \tag{4}$$

The diffusive uptake of atmospheric CH₄ at the soil-atmosphere interface can then be calculated using the derivative of Eq. (4) at $z = 0$:

$$J_{CH4} = -D_{CH4} * \frac{dCH4}{d_z}|_{z=0} = D_{CH4} * C_{CH4} * \sqrt{\frac{k_d}{D_{CH4}}} = C_{CH4}\sqrt{D_{CH4}k_d} \tag{5}$$

This formulation of soil uptake of CH₄ is the simplest analytical solution to Eq. (2). It represents an improvement from the semi-numerical representation used in the R99 model and enables complete consumption of CH₄ to be accounted for within the soil layer; however, the homogeneous Neumann boundary condition applied here is only an approximation, which is not generally valid. The simulation will not be influenced if the Neumann boundary is infinitely far from the consumption depth of CH₄ and thus, the corresponding Neumann boundary conditions can be neglected. However, if this is not the case, it will result in simulation error.

Complete consumption of CH₄ at an a priori unknown depth L (MeMo solution)

Therefore, we adopted an approach similar to the model C07 model but one that is generally valid. We assume that methanotrophy consumes atmospheric CH₄ in the soil until CH₄ reaches a threshold ($CH_4(L) = CH_4min$) that can be imposed based on biological limits ($CH_4min=0.1ppm$) or when CH₄ is fully depleted ($CH_4min=0$). The integration constants in Eq. (3) thus become:

$$A = -\frac{C_{CH4} * exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right) - CH4min}{\left[exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right) - exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right)\right]} \frac{C_{CH4} * exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right)}{\left[-exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right) + exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right)\right]}$$

(6)

$$B = \frac{-CH4\,min + C_{CH4} * exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right)}{\left[exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right) - exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right)\right]} \quad - \quad \text{~~B=CCH4~~}$$

~~* exp-kdDCH4L-exp-kdDCH4L+expkdDCH4L~~

(7)

In addition to the concentration condition $CH_4(L) = CH_4 min$, a flux condition also is imposed on the lower boundary in order to determine depth L: $-D_{CH4} * \frac{dCH4}{d_z}|_{z=L} = F_{CH4}$

Where F_{CH4} denotes a potential CH_4 flux across the lower boundary that can be specified (i.e. $CH_4(L) = CH_4 min$) or set equal to zero (i.e. $CH_4(L) = 0$). The unknown depth L ~~for a general case at which $CH_4 = 0$~~ is then calculated by substituting the derivative of Eq. (3) into the expression for the lower boundary condition:

$$-D_{CH4} * \frac{dCH4}{dz}|L = -D_{CH4} * \left(A\left(-\sqrt{\frac{k_d}{D_{CH4}}}\right) * exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right) + B\sqrt{\frac{k_d}{D_{CH4}}} * exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right)\right) = F_{CH4} \qquad (8)$$

Rearranging Eq. (8) and finding its root allows for the determination of the initially unknown depth L when $CH_4(L) = CH_4 min$:

$$0 = -D_{CH4}\sqrt{\frac{k_d}{D_{CH4}}}\frac{\left(2\,C_{CH4} - CH4min*exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right) - CH4min*exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right)\right)}{\left[exp\left(-\sqrt{\frac{k_d}{D_{CH4}}}L\right) - exp\left(\sqrt{\frac{k_d}{D_{CH4}}}L\right)\right]} - F_{CH4} \qquad (9)$$

~~(9)~~

Once L is known total CH_4 uptake can be calculated from:

$$J_{CH4} = -D_{CH4} * \frac{dCH4}{d_z}|_{z->z=0} = -D_{CH4}\left(-A\sqrt{\frac{k_d}{D_{CH4}}} + B\sqrt{\frac{k_d}{D_{CH4}}}\right) \qquad (10)$$

Where A and B are defined by Eqs. (6) and (7). When L tends to infinity Eq. (10) is equivalent to the model C07 solution however Eq. (10) also allows for (i) complete consumption of CH_4 within the soil interval, (ii) influx of CH_4 from beneath the soil profile (*e.g.*, from thawing permafrost or production of CH_4 in oxygen-depleted microsites in soil ~~soils~~) and (iii) a minimum CH_4 concentration at which methanotrophy can occur in the soil column.

Figure 1 illustrates CH$_4$ soil profiles and the penetration depth of CH$_4$ into soil, L, for different k_d values, $F_{CH4} = 0$ and $D_{CH4} = D_{0CH4}$ (diffusivity in free air) (Table1). It is expected that L will vary spatially depending on local k_d, D_{CH4} and soil properties.
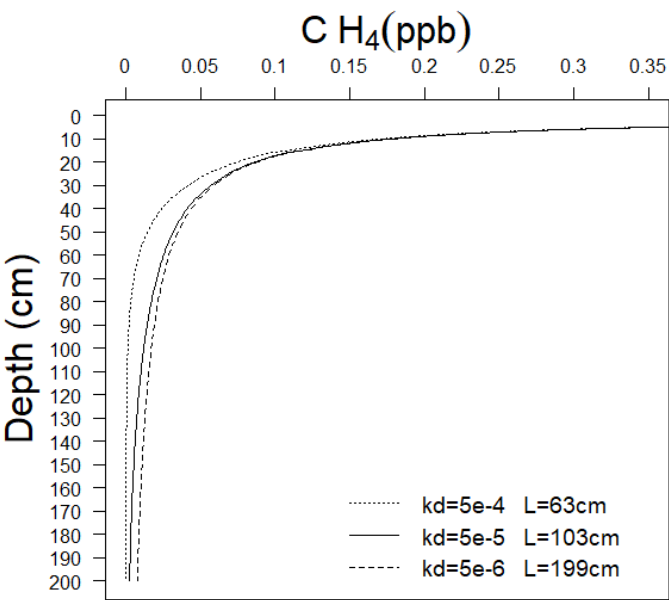

Figure 1: Computational solution of Eq. (9) for different values of k_d. Parameter L is defined as the depth where CH$_4$min = 0, assuming complete removal of CH$_4$ in soil pore spaces (assuming a total of atmospheric of CH$_4$ concentration of 1800 ppb).

Table 2. Descriptions of four soil methanotrophy models.

Model / Study	Description	CH$_4$ uptake calculation (J_{CH4})	Eq.
P96 Potter et al. (1996)	Model based on Fick's first law. The calculation of the uptake flux is approximated numerically and based on the diffusion of CH$_4$ into soil.	$J_{CH4} = D_{CH4} \dfrac{\Delta C_{CH4}}{\Delta_z}$	(11)
R99 Ridgwell et al. (1999)	R99 extends the P96 model by including an explicit term for microbial oxidation of CH$_4$ in soil. The uptake flux is approximated numerically, using	$J_{CH4} = \dfrac{C_{CH4} D_{CH4}}{z_d}\left(1 - \dfrac{D_{CH4}}{D_{CH4} + k_d z_d}\right)$	(12)

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	Fick's first law and adopting a first order rate law for microbial oxidation, assuming that oxidation occurs in a thin ϵ cm layer located at 6 cm depth.	
C07 Curry (2007)	C07 adopts the diffusion-reaction equation that underlies R99. However, C07 solves the equation analytically (as opposed to semi-numerically). The model also improves representation of soil moisture influence on the microbial oxidation rate. C07 refines methanotrophy response at subzero temperatures on the basis of observations.	$$J_{CH4} = C_{CH4} r_N r_w \sqrt{D_{CH4} k_d} \qquad (13)$$
MeMo This study	Incorporates a general mathematical description of CH$_4$ uptake flux, allowing for complete consumption of CH$_4$ at an initially unknown depth L and CH$_4$ flux through the lower boundary. Refines representation of the influence of soil moisture, temperature and nitrogen deposition on CH$_4$ oxidation.	$$J_{CH4} = -D_{CH4}\left(-A\sqrt{\frac{k_d}{D_{CH4}}} + B\sqrt{\frac{k_d}{D_{CH4}}}\right) \quad (10)$$

MeMo is based on the more general solution (Eq. (10)) and uses local methanotrophy rates (k_d) and diffusion coefficients (D_{CH4}) based upon soil conditions to determine CH$_4$ penetration depths (L). Additionally, Eq. (9) allows one to set a minimum CH$_4$ concentration if this parameter is known. In this case, we assume a minimum of 0 or complete consumption.

5 We assume no *in situ* production of CH$_4$ or upward CH$_4$ flux from below (*i.e.*, $F_{CH4} = 0$) because of a scarcity of field data for model validation. However, a flux from below can be employed in MeMo to enable a more comprehensive quantification of soil CH$_4$ uptake that also potentially accounts for consumption of upward migrating CH$_4$ and autochthonous CH$_4$ produced in oxygen-depleted microsites of finely textured soil.

2.3 Parameters

10 The rate of CH$_4$ uptake by soil is controlled by the balance between gaseous diffusion of atmospheric CH$_4$ into soil and the rate of CH$_4$ oxidation by methanotrophic bacteria as described by Eq. (14) and Eq. (20), respectively. Thus, D_{CH4} and k_d are key parameters and accurate characterization of their values is essential for robust quantification of the soil CH$_4$ sink.

2.3.1 Soil CH4 Diffusivity, D_{CH4}

Similar to the R99 and C07 model, D_{CH4} in MeMo is determined from the diffusivity of CH_4 in free air (D_{0CH4}; Table 1) adjusted for the influence of temperature (G_T) and soil structure (G_{soil}):

5 $$D_{CH4} = D_{0CH4} * G_T * G_{soil} \tag{14}$$

The gaseous diffusion coefficient of CH_4 in soil increases linearly with temperature T (°C) (Potter et al., 1996) according to the relationship:

10 $$G_T = 1.0 + 0.0055\, T \text{ (°C)} \tag{15}$$

The soil structure factor (G_{soil}) accounts for the effects of pore size, connectivity and tortuosity on gaseous diffusion and is determined according to the parameterization of Moldrup et al. (1996; 2013):

15 $$G_{soil} = \Phi^{4/3} \left(\frac{\Phi_{air}}{\Phi}\right)^{1.5+3/b} \tag{16}$$

Where Φ is total pore volume (cm^3 cm^{-3}), Φ_{air} is air-filled porosity (cm^3 cm^{-3}) and b is a scalar that accounts for soil structure. Total pore volume is defined as a function of bulk density ρ (g cm^{-3}) and average particle density d (Table 1) (Brady et al., 1999):

20

$$\Phi = 1 - \left(\frac{\rho}{d}\right) \tag{17}$$

The scalar b in Eq. (16) is calculated as a function of soil clay content (f_{clay}; %) as proposed by Saxton et al. (1986):

25 $$b = 15.9\, f_{clay} + 2.91 \tag{18}$$

Air-filled porosity (Φ_{air}) is determined from the difference between total pore volume and soil water content θ (%):

$$\Phi air = \Phi - \theta \tag{19}$$

25

2.3.2 Rate Constant for CH₄ Oxidation, k_d

The CH₄ oxidation rate (k_d) is defined as the base oxidation rate constant (k_0) for an uncultivated moist soil at 0° C scaled by three factors to account for the influence of soil moisture (r_{SM}), soil temperature (r_T), and nitrogen content (r_N):

5    $$k_d = k_0 * r_{SM} * r_T * r_N \tag{20}$$

The R99 and C07 models used a similar equation to estimate k_d but without the r_N parameter, opting instead to employ intensity of agricultural activity as a proxy to account for the inhibitory effects of N deposition on soil methanotrophy. Moreover, model C07 excluded r_N from the k_d formulation and used a N deposition term to modify total CH₄ uptake flux (Table 2, Eq. 13), 10   which results in a larger N inhibition effect. The approach employed in MeMo is to use N deposition data directly to modify k_d.

2.3.3 Base Oxidation Rate Constant, k_0

The base oxidation rate constant (k_0) is a key parameter that exerts significant control on k_d and thus the estimated CH₄ uptake 15   flux. For example, a 10-fold change in k_0 (and thus k_d) leads to a 3-fold decrease in the depth L at which CH₄ is fully depleted from soil pores (Fig. 1) and a ~3-fold increase in total uptake of CH₄ (Fig. 2).
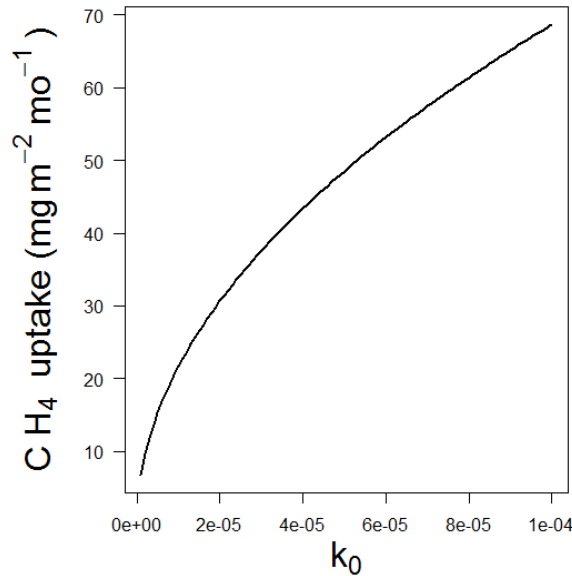

Figure 2: Total CH₄ uptake for different values of k_0 (s⁻¹), assuming a constant value of $D_{CH4} = D_{0CH4}$ and no modification by soil temperature, moisture or nitrogen deposition.

20

Rate constants can be defined either on the basis of theoretical considerations or through site-specific field and laboratory observations. Rates of soil microbial processes, such as CH_4 oxidation, are controlled by microbial biomass dynamics and community structure, and, thus a complex array of environmental factors, including temperature, substrate (CH_4) concentration, land use, moisture, pH and soil type (Ho et al., 2013). The influence of these environmental factors on microbial
5   CH_4 oxidation rates is not well characterized and thus all factors are not explicitly represented in models. Consequently, apparent rate constants implicitly account for some environmental factors via fitting field observations or laboratory experiments, resulting in parameter values that may be more environment- and model-specific. A possible limitation of such an approach is reduced transferability and predictive capacity in other environments or from a regional to the global scale. For example, Ridgwell et al. (1996) derived a single global estimate of $k_0 = 8.7 \times 10^{-4}$ s^{-1} by fitting Eq. (12) to 13 measured values
10   of J_{CH4}, D_{CH4} and soil temperature from four different studies. In contrast, Curry (2007) estimated a global k_0 of 5.0×10^{-5} s^{-1} based upon fitting Eq. (13) to a five-year time series of J_{CH4} and soil temperature, moisture and CH_4 flux measurements from a single site in Colorado (Mosier et al., 1996). The order of magnitude difference in k_0 between the R99 and C07 models illustrates the potential model-specific nature of parameter values derived from experimental and observational data, as well as the limits and challenges for transferability. Soil methanotrophy is not unique in this regard and parameterization of
15   microbially mediated processes remains a common problem more generally in modelling approaches (*e.g.*, Arndt et al., 2013; Bradley et al., 2016).

Parameterization of k_0 in MeMo has been refined using ~~recently~~ time-series data recently published by Luo et al., (2013), which ~~and~~ consist~~ing~~ of daily soil CH_4 uptake rate and~~,~~ temperature and soil moisture data from three contrasting environments: temperate forest (Hoglwald Germany), tropical rainforest (Bellenden Ker Australia) and steppe (Inner
20   Mongolia, China). The data sets were used to explore potential variations in apparent k_0 values in different environments, including comparison with k_0 values from models R99 and C07. Data from each site were interpolated according to Eq. (10) to derive an apparent k_0 value for each biome. The k_0 values for temperate forest and steppe are similar to the k_0 value employed in the C07 model; however, the apparent k_0 for tropical forest is approximately three times smaller than the model C07 k_0 value. The three newly derived k_0 values were employed in MeMo for their respective biomes and the k_0 value from the C07
25   model ($k_0 = 5.0 \times 10^{-5}$ s^{-1}) was used for all other regions for which no biome specific k_0 values exist (Table 3). Similar k_0 values of 5.0×10^{-5} s^{-1} for temperate forest, steppe and short grass steppe indicate that this magnitude of k_0 is appropriate for many ecosystems. Yet, apart from the tropical wet forest, the data clearly indicate additional controls and the use of $k_0 = 1.6 \times 10^{-5}$ s^{-1} will thus prevent an overestimation of simulated fluxes. Nevertheless, further research is required to better characterize this key parameter.

30

Table 3. k_0 values from models R99 and C07 and new k_0 values employed in MeMo that were determined based upon temperate forest, tropical forest and steppe data from Luo et al. (2013).

Model	Biome	k_0 (s^{-1})

R99	Global	8.7×10^{-4}
C07	Global	5.0×10^{-5}
MeMo	Temperate forest	4.0×10^{-5}
	Tropical forest	1.6×10^{-5}
	Steppe	3.6×10^{-5}
	Other ecosystems	5.0×10^{-5}

2.3.4. Soil Moisture Factor, r_{SM}

Both low and high soil moisture levels can negatively impact soil uptake of atmospheric CH_4 (Schnell and King, 1996; von Fischer et al., 2009). Scarcity of soil water generally inhibits soil microbial activity while excessive moisture attenuates gas
5 diffusion, limiting entry of atmospheric CH_4 and O_2 into soil (Burke et al., 1999; McLain et al., 2002; McLain and Ahmann, 2007; West et al., 1999).

TheModels R99 and C07 models incorporated parameters to address the limiting effects of low soil moisture levels on CH_4 uptake fluxes. The R99 model applied a soil moisture factor adopted from Potter et al. (1986) where r_{SM} was calculated as a proportional ratio of precipitation (P) plus soil moisture (SM) divided by potential evapotranspiration (ET; Table 4, Eq.
10 (21)). It was assumed that r_{SM} decreases linearly when (P+SM)/ET is less than one. The C07 model modified the response of soil methanotrophy to moisture using an empirical water stress parameterization and soil water potential based on findings from Clapp and Hornberger (1978) (Table 4, Eq. (22)). A consequence of that approach is that r_{SM} decreases logarithmically to zero at an absolute soil water potential of $w < 0.2$ MPa (Fig. 3).

In MeMo, soil moisture (%) is used to calculate r_{SM} and a formulation similar to the C07 model is used for low soil
15 moisture values. A threshold of <20% soil moisture is applied because that value corresponds to optimum conditions for CH_4 oxidation in soil (Castro et al., 1995; Whalen and Reeburgh, 1996) and because inclusion of a water stress parameter better captures CH_4 uptake flux in dry ecosystems (Fig. 3; Curry, 2007).

Establishing parameters to quantify the impact of excess moisture on soil methanotrophy has proven more challenging. The C07 model relied upon soil pore space characteristics in factor G_{soil} (Eq. (16)) to account for decreased gas
20 diffusion and limitation of k_d at high soil moisture content. However, attenuation of gas diffusion is only one impact of high soil water content and it is necessary also to account for the inhibitory effects of excessive moisture on k_d (Boeckx and Van Cleemput, 1996; Dasselaar et al., 1998; Visvanathan et al., 1999). Soil moisture content >20% reduces CH_4 uptake due to a restricted diffusion of CH_4 and supply of O_2. The R99 and C07 models assume that microbial CH_4 oxidation remains active at a soil moisture content of 80%, an assumption that contradicts field investigations, which show that CH_4 uptake decreases
25 rapidly at soil moisture levels >50% (Dasselaar et al., 1998). Thus, the soil moisture factor employed in MeMo also accounts for limitation of microbial CH_4 oxidation at a soil moisture content >20% after which rates of CH_4 uptake begin to decrease (Adamsen and King, 1993; Visvanathan et al., 1999). The r_{SM} factor used in MeMo was determined by fitting a Gaussian

function to laboratory experimental data (Table 4, Eq. (23); Fig. 3a), following the approach of Del Grosso et al. (2000). The mean r_{SM} and standard deviation determined using this approach were 0.2 ± 0.2.

Table 4. Model R99, C07 and MeMo formulations for r_{SM} response.

Model	Formulation	Eq.	Variable definitions
R99	$r_{SM} = 1$ for P+SM/ETp>1 $r_{SM} = P + SM/ETp$ for P+SM/ET p≤1	(21)	P=precipitation SM=soil moisture stored at 30 cm depth ETp=potential evapotranspiration
C07	$r_{SM} = 1$ for w<0.2MPa $r_{SM=}\left[1 - \dfrac{log_{10}w - log_{10}(0.2)}{log_{10}(100) - log_{10}(0.2)}\right]^{0.8}$ for w≥0.2≤100MPa	(22)	w=saturation soil water potential
MeMo	$r_{SM=}\left[1 - \dfrac{log_{10}\frac{1}{SM} - log_{10}(0.2)}{log_{10}(100) - log_{10}(0.2)}\right]^{0.8}$ for SM<0.2 $r_{SM=}\dfrac{1}{\sigma\sqrt{2\pi}}e^{-\frac{1}{2}\left(\frac{SM-0.2}{0.2}\right)^2}$ for SM>0.2	(23)	SM=soil moisture

A soil moisture factor (r_{SM}) was calculated for each set of observational data from independent field sites (Supplementary 1, Table S1) based upon an optimum rate of CH$_4$ uptake occurring at a soil moisture content of 20% ($r_{SM} = 1$). The remaining r_{SM} values were computed as a linear ratio of the CH$_4$ uptake rate at 20% water content. Figure 3b illustrates the pattern of response in methanotrophy rates to changes in soil moisture content in the R99, C07 models and MeMo and the net effect on CH$_4$ uptake fluxes across a range of absolute soil moisture levels used to force parameter r_{SM}. The CH$_4$ uptake fluxes were calculated by varying soil moisture content while holding constant all other environmental parameters (temperature, C_{CH4} and N_{dep}). The R99 and C07 models both predict greater CH$_4$ uptake fluxes than MeMo at soil moisture contents >20% with the R99 model yielding the highest flux rates; however, the C07 model and MeMo yield similar CH$_4$ uptake rates for much of the soil moisture range. Reduction of CH$_4$ uptake flux at high soil moisture levels due to attenuation of gas diffusion cannot be managed solely through the term G_{soil} (*i.e.*, reduction in free pore space). MeMo also accounts for inhibition of microbial CH$_4$ oxidation rates at elevated soil moisture content, predicting lower CH$_4$ uptake flux as a result of more realistic r_{SM} values determined from the Gaussian response observed in field data from three different global biomes (Luo et al., 2013).

2.3.5. Temperature Factor, r_T

Temperature exerts an important influence on rates of microbial processes and consequently all models parameterize for the effects of temperature on soil methanotrophy. The R99 model employs a Q10 function derived from experimental data with a Q10 factor of 2 change over the temperature interval 0 to 15°C. The model assumes that bacterial methanotrophy ceases at temperatures <0°C (Table 5, Eq. (24)). The C07 model adopts the same Q10 factor as R99 for temperatures >0°C but employs a different response below 0°C. Soil water generally does not freeze at a surface temperature of 0°C and observations from cold regions provide ample evidence for the presence of methanotrophic activity at temperatures <0°C (Vecherskaya et al. 2013). The C07 model allows for a parabolic decrease of methanotrophy rates from 0 to -10°C (Table 5, Eq. (25)) based upon observations of CH_4 uptake in soil at subzero temperatures (Del Grosso et al., 2000).

Parameterization of a temperature factor (r_T) is revisited in MeMo based upon availability of new experimental data for soil from different biomes (Supplementary 1, Table S2). A Q10 factor having a value of 1.95 was determined for the temperature interval 0 to 15 °C by curve fitting and minimizing linear errors ($r^2 = 0.75$, p=1.9 e^{-11}; Table 5, Eq. (26)). The factor r_T was determined by using the observed CH_4 uptake flux at 10°C at each site as the base of the Q10 function (Fig. 3c). An exponential decrease in CH_4 uptake flux was assigned to the temperature range 0 to -5°C as recommend by Castro et al. (1995) and Del Grosso et al. (2000). Moreover, the amount of frozen soil increases exponentially with decreasing temperatures (Low et al., 1968) and consequently, CH_4 uptake also should decline exponentially.

Table 5. Model R99, C07 and MeMo formulations for r_T response.

Model	T<0°C	T≥0°C	Eq.
R99	$r_T = 0$	$r_T = \exp(0.0693T - 8.56 \times 10^{-7} T^4)$	(24)
C07	$r_T = (0.1T + 1.0)^2$ if T>-10°C	$r_T = \exp(0.0693T - 8.56 \times 10^{-7} T^4)$	(25)
MeMo	$r_T = 1/\exp(-T)$	$r_T = \exp(0.1515 + 0.05238T - 5.946 \times 10^{-7} T^4)$	(26)

The pattern of change in the r_T factor and CH_4 uptake flux for the temperature range -10 to 60°C is shown in Fig. 3d. The CH_4 uptake fluxes shown were calculated by varying temperature while holding other environmental factors constant (*i.e.*, soil moisture, N deposition or agricultural land use, and C_{CH4}). All models exhibit an optimum in CH_4 uptake at 25°C characterized by a maximum r_T and CH_4 oxidation rate. The key differences between models are the behavior of r_T at temperatures below 0°C and the amplitude of response curves. The R99 model assumes that methanotrophic~~y~~ activity ceases at 0°C and consequently, CH_4 uptake rates decrease sharply at that temperature. In contrast, models C07 and MeMo both allow for methanotrophy at temperatures <0°C. In general, the exponential decrease of r_T employed in MeMo more closely resembles natural patterns of soil methanotrophy at subzero temperatures than the parabolic decline employed in the C07 model consistent with observations reported by Castro et al. (1999) and Del Grosso et al. (2000). Although our parameterization

yields a fit similar to C07 to the limited observations available at temperatures <0°C the r_T used in MeMo provides a simpler solution because it does not require multiple conditions to be met. In contrast, the C07 parameterization increases parabolically at temperatures <-10° C, which requires an additional condition to be incorporated into the model to prevent increased rates of CH4 uptake at very low temperatures. Soil CH4 uptake fluxes predicted by the C07 model are greater than those calculated using MeMo because of the different parameterization at temperatures <0°C. Finally, the amplitude of the temperature response curve is greater and similar in models C07 and MeMo compared to model R99, in particular, at temperatures >25°C as a result of differences in the formulation and solution for CH4 uptake flux (Fig. 3d).
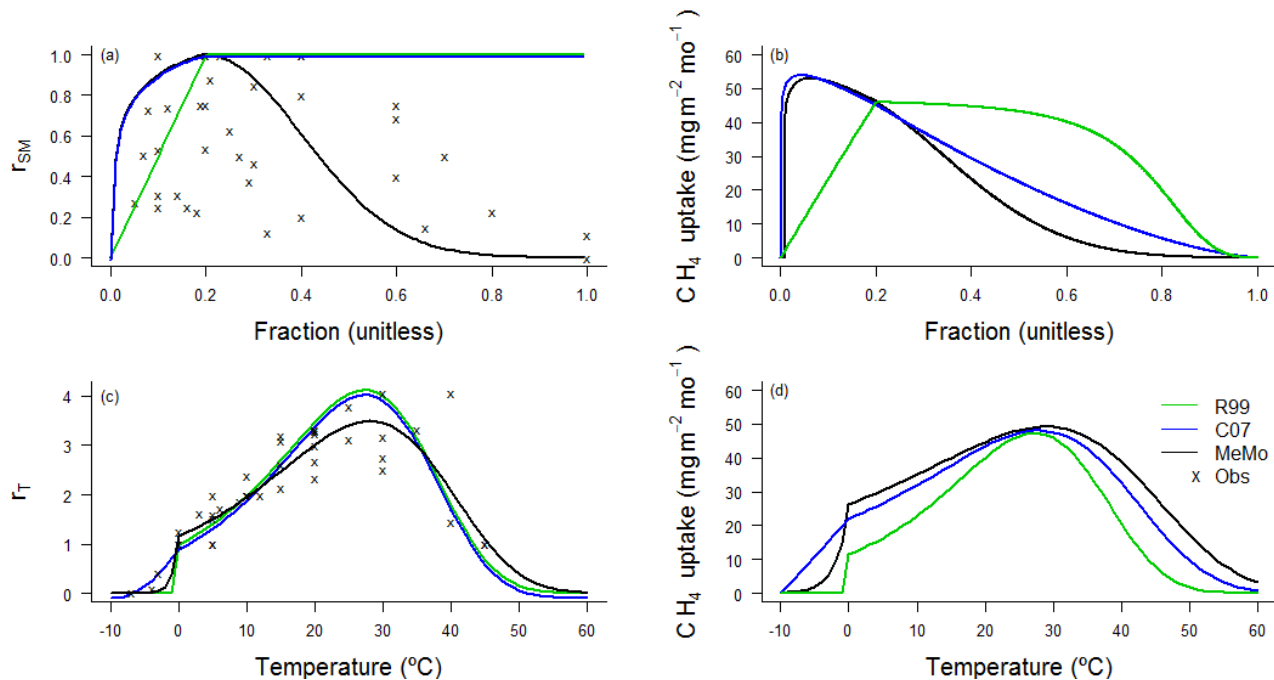

Figure 3: CH4 uptake response factors (a, c) and uptake fluxes (b, d) as a function of soil moisture (r_{SM}) and temperature (r_T). Observations (shown as crosses) (r_{SM}, Supplementary 1, Table S1; r_T, Supplementary 1, Table S2), MeMo (black line), C07 (blue line) and R99 (green line).

2.3.6 Nitrogen Deposition factor, r_N

The effect of nitrogen (N) deposition on CH4 uptake is not as well constrained as the effects of temperature and soil moisture. In general, field observations have shown that CH4 consumption rates and thus, uptake fluxes decrease with N additions (Aronson and Helliker, 2010; Butterbach-Bahl and Papen, 2002; Steinkamp et al., 2001). Different processes have been suggested to explain this negative effect. Firstly, methanotrophs and ammonia oxidizers are capable of switching substrates (although the latter microorganisms typically consume N compounds preferentially if available) and therefore the presence of N compound reduces CH4 consumption (Bradford et al., 2001; Gulledge and Schimel, 1998; Phillips et al., 2001;

Wang and Ineson, 2003; Whalen, 2000). In addition, intermediate and end products from methanotrophic ammonia oxidation (*i.e.*, hydroxilamida and nitrite) can be toxic to methanotrophic bacteria (Bronson and Mosier, 1994; MacDonald et al., 1996; Sitaula et al., 2000). Finally, large amounts of mineral fertilizers (*i.e.*, ammonium salts) can induce osmotic stress in methanotrophs inhibiting CH_4 consumption (Whalen, 2000). However, other studies suggest a positive effect of N fertilization

5   on CH_4 oxidation rates. One of the mechanism invoked to explain the positive effect is a stimulation of nitrifying bacteria to consume CH_4 by increased inputs of N due to an improvement in living conditions (Cai and Mosier, 2000; De Visscher and Cleemput, 2003; Rigler and Zechmeister-Boltenstern, 1999). The positive effect of N addition on CH_4 oxidation rates has been observed primarily under experimental conditions and also greatly depends on the local microbial community structure. Therefore, we assumed that N has an inhibitory effect on uptake of atmospheric CH_4 in all scenarios.

10   The C07 and R99 models both account for the negative effect of N inputs on CH_4 uptake fluxes via a the factor r_N. In model R99, r_N directly affects k_d while in model C07 r_N directly modifies the uptake flux. Both models parameterize the negative effect of N inputs on CH_4 oxidation rates as a function of agricultural intensity (as a fraction of area) as a proxy for fertilizer application (Table 6, Eq. (27)). However, the mathematical description of r_N used by models R99 and C07 does not account for the enhanced N deposition by anthropogenic activity or direct N input via fertilizers because its global distribution

15   was not well known at the time of model development. Here, we suggest a mathematical description of r_N that accounts for all anthropogenic N input sources: fossil fuel combustion, biomass burning and fertilizer application (Lamarque, 2013; Nischina et al., 2017).

The computation of r_N in MeMo is a function of: i) the inhibitory effect on CH_4 uptake, and ii) the distribution and amount of N input in soil (Zhuang et al., 2013). We estimated the percent reduction of CH_4 uptake per mol of N added based

20   on field and laboratory observations (Supplementary 1, Table S3). We determined an average inhibition α of 0.33 % mol N^{-1} based on the mean uptake reduction per mol of N added. The N response function r_N was governed by Eq. (29):

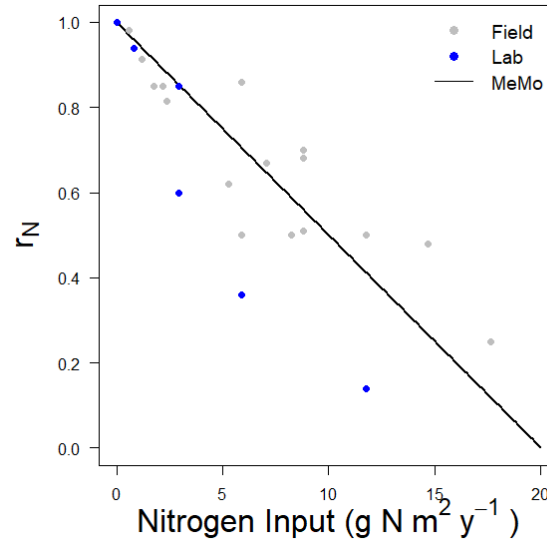$$r_N = 1 - (N_{soil} * \alpha) \tag{29}$$

Figure 4: CH₄ uptake response as a function of nitrogen deposition and fertilizer application factor r_N. The linear fit (black line) is based on observations from field and laboratory measurements (gray and blue dots; Supplementary 1, Table S3).

In cases where entry of N into soil is limited by bulk density ρ, ninety percent of N compounds tend to remain at depths $z <= 5$ cm before exponentially decreasing in concentration with depth (Schnell and King, 1994). Thus, N_{soil} was calculated as N input (kg N ha⁻¹ y⁻¹) divided by ρ at $z = 5$cm:

$$N_{soil} = \frac{Ndep+Nfert}{(\rho*z)} \tag{30}$$

Figure 4 shows the change in r_N in relation to N input rate and the form of Equation (29).

Table 6. Model R99, C07 and MeMo formulations for r_N response.

Model	Formulation	Eq.	Driving data
R99	$r_N = 1.0 - (0.75 \times I)$	(27)	I = fractional intensity of cultivation
C07	$r_N = 1.0 - (0.75 \times I)$	(28)	I = fractional intensity of cultivation (r_N outside of k_d parameterization)
MeMo	$r_N = 1-(N_{soil}) * \alpha$	(29)	$N_{soil} = \frac{Ndep+Nfert}{(\rho*z)}$ (30)

3.0 Model implementation

MeMo was implemented in R (version 3.0.1) and simulations were carried out with a spatial resolution of 1°x1° and a monthly temporal resolution for the period between 1990 and 2009. The model code, a simple model case study for year 2000 and output for 1990-2009 are available as a supplement to this manuscript. To enable model-model comparisons and assess the combined effect of all refinements introduced in MeMo on the global CH$_4$ uptake flux estimate, the R99 and C07 models also were also implemented in R at identical spatial and temporal resolutions and forced using the same driving data.

3.1 Forcing data

MeMo, and the C07 and the R99 models were forced using global, monthly observations of soil moisture, temperature, atmospheric CH$_4$ concentrations, N deposition, soil bulk density, and clay content for the period 1990-2009. Information about data sources and maps of the forcing data are provided in Supplementary 32.

Satellite observations of soil moisture at a spatial resolution of 1x1° and a monthly temporal resolution are available for the period 1990-2009 from Dorigo et al. (2011); however, the data set contains gaps in some regions (*e.g.*, in areas of high-density vegetation). The use of MeMo as a predictive tool to estimate the past and future global CH$_4$ soil sink relies strongly on the use of soil moisture from standard climate models, such as output from land surface models or Dynamic Global Vegetation Models (DGVMs). Therefore, gaps in the Dorigo et al. (2011) data set were filled using soil moisture data from an ensemble of 9 DGVMs (TRENDY; Sitch et al., 2015). The R99 model parameterizes the effect of soil moisture on CH$_4$ uptake fluxes as a function of precipitation and evaporation and therefore, R99 was forced using monthly data sets of precipitation (CRU3.1; Harris et al., 2014) and evapotranspiration (TRENDY; Sitch et al., 2015). Temperature forcing is constrained by global data sets for surface temperature as a proxy for soil temperature (CRU3.1; Harris et al., 2014). Monthly mean global atmospheric CH$_4$ concentrations multiplied by the latitudinal atmospheric CH$_4$ gradient were calculated from Rigby et al. (2008). The N deposition data were obtained from an atmospheric chemical transport model embedded in an Earth System Model (Lamarque et al., 2013) and the N input via fertilizers was obtained from Nischina et al. (2017). Because the R99 and C07 models express the influence of N deposition on CH$_4$ uptake fluxes as a function of fraction agricultural area fraction (see section 2.3.6), R99 and C07 were forced using annual global gridded land-use change data from Hurtt et al. (2011). Finally, global gridded observations for bulk density and clay content were taken from Shangguan et al. (2014).

Areas that have less than 0.5% average annual soil moisture content were masked (*e.g.*, Sahara Desert) because it was assumed CH$_4$ uptake is negligible under such conditions. If the areas were left unmasked, then MeMo would overestimate CH$_4$ uptake across the regions due to high temporal variability in the driving data (*e.g.*, a month with no moisture followed by a month with >20%). Irregular short-lived precipitation events in deserts lead to unreliable estimates of soil uptake of atmospheric CH$_4$ because such areas are unlikely to host well-established communities of methanotrophic bacteria capable of responding rapidly to short-term increases in soil moisture.

4.0 Results and Discussion

The following sections critically evaluate MeMo estimates of the global CH$_4$ sink (section 4.1), as well as the regional distribution of CH$_4$ uptake and its main drivers (section 4.2) in the context of available field data and published model predictions.

4.1 Global CH$_4$ Uptake by Soils

MeMo predicts an average annual global flux of 33.5~~34.3~~ ± 0.6 Tg CH$_4$ y^{-1} for the period 1990 to 2009. The estimated global uptake compares well with estimates from terrestrial ecosystem models, DGVMs and global atmospheric inversions (Table 7). Zhuang et al. (2013) determined a similar average global uptake flux of 34 ± 2 Tg CH$_4$ y^{-1} during the 21st century using a process-based model included in the Terrestrial Ecosystem Model (TEM) while Spahni et al. (2011) estimated an uptake flux of 38.9 Tg CH$_4$ y^{-1} using the LPJ-WHyMe DGVM. Hein et al. (1997) predicted a similar flux through atmospheric inversions but with a greater level of uncertainty (30 ± 15 Tg CH$_4$ y^{-1}). Upscaling of field measurements of soil methanotrophy rates from 120 different studies spanning a wide range of ecosystems yielded a value of 36 ± 23 Tg CH$_4$ y^{-1} (Dutaur and Verchot, 2007). In contrast, flux estimates based upon extrapolation of long-term records of CH$_4$ uptake in a smaller number of soil types resulted in an estimated flux of 28.7 Tg CH$_4$ y^{-1} (Dörr et al. 1993). Similarly, global extrapolation of measurements made solely on northern European soils yielded a sink strength of 29 Tg CH$_4$ y^{-1} (Smith et al. 2000).

Table 7. Global CH$_4$ uptake estimations

Methodology	Reference	Global uptake by soils (Tg CH$_4$ y^{-1})
Observation	Dörr et al. (1993)	28.7
Observation	Smith et al. (2000)	29
Observation	Dutaur and Verchot (2007)	36 ± 23
Atmospheric inversions	Hein et al. (1997)	30 ± 15
Model (P96)	Potter et al. (1996)	20 ± 3
Model (R99)	Ridgwell et al. (1999)	38.1 ± 1.1
Model	Spahni et al. (2011)	38.9
Model (C07)	Curry (2007)	29.3 ± 0.6
Model	Zhuang et al. (2013)	34 ± 2
Model (MeMo)	**(This study)**	**33.5 ~~34.3~~ ± 0.6**

The average annual soil sink for atmospheric CH$_4$ estimated by MeMo (33.5 ~~34.3~~ ± 0.6 Tg CH$_4$ y^{-1}) is greater than global uptake predicted using the P96 and C07 model (20 ± 3 Tg CH$_4$ y^{-1} and 29.3 ± 0.6 Tg CH$_4$ y^{-1}, respectively). The R99 model predicts a global sink of 38.1 ± 1.1 Tg CH$_4$ y^{-1}, which compares more favorably with the MeMo estimate. The observed differences in mean global soil uptake of atmospheric CH$_4$ estimated using models R99, C07 and MeMo forced with identical data are attributed primarily to three factors: (i) their respective mathematical solutions of reaction-transport equations (section 2.2), (ii) differences in parameterization of k_0 (section 2.3.3), and (iii) differences in formulation of r_N (section 2.3.6). The

R99 model predicts soil uptake that is 1~~2~~0% and 24% greater, respectively, than fluxes estimated using MeMo and the C07 model. These differences are due to the R99 model applying a k_0 that is one order of magnitude greater than k_0 values used in the C07 model and MeMo. The amplifying effect of the large k_0 is partially offset by the semi-numerical approximation (Eq. 12) employed in the R99 model, which results in the final global CH_4 uptake flux being of similar magnitude to the MeMo and the C07 model estimates. Finally, the low uptake predicted by the C07 model is a consequence of the parameterization of the nitrogen inhibition effect (r_N,) and its direct modification of the CH_4 flux rather than the CH_4 oxidation activity (k_d) (section 2.3.3). Nitrogen inhibition was responsible for a global reduction in CH_4 uptake of 1.4~~0.34~~ Tg y^{-1} in MeMo compared to 7.3 and 2.3 Tg y^{-1} in the C07 and R99 models, respectively.

4.2 Regional CH4 Uptake by Soils

The latitudinal distribution of soil uptake rates of atmospheric CH_4 predicted using the R99 and ~~model, the~~ C07 model__s__ and MeMo are shown in Fig. 5 accompanied by direct measurements of CH_4 oxidation rates from Dutaur and Verchot (2007) and a 10° running average. Figure 6 provides globally gridded maps of average simulated CH_4 uptake fluxes from the three models for the period 1990 to 2009 and a comparison between MeMo and the R99 and C07 models.
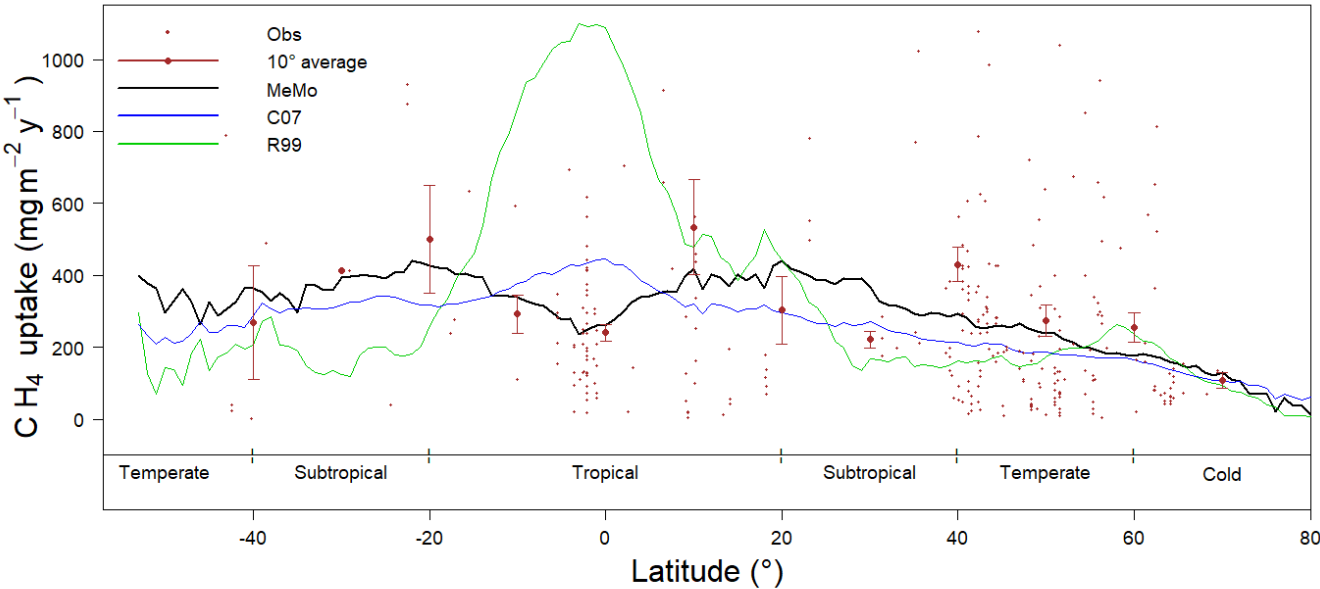

Figure 5. Latitudinal distribution of the soil uptake predicted by models R99 (green line), C07 (blue line) and MeMo (black line). Measurements of CH4 uptake (small brown dots; Dutaur and Verchot, 2007) and a 10º running mean of direct observations (large brown dots for average with bars representing one standard deviation error).

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The latitudinal distribution of observations reveals a scarcity of direct measurements of soil methanotrophy from sites in the southern hemisphere. Additionally, the frequency of measurements generally is low and rarely encompasses a full twelve-month period, which creates challenges for verifying model estimates of annual CH_4 uptake fluxes. Observations at specific latitudes typically exhibit a wide range of values, which are reflected in the large standard error bars calculated for the

5   10° running means (Fig. 5). Nevertheless, the averages of direct observations calculated for each $10°$ latitude interval show a distinct bimodal pattern with the lowest soil CH_4 uptake fluxes in the tropics and at high latitudes. Maximum rates of CH_4 uptake occur between 10 to $20°$ latitude in both hemispheres (Fig. 5). MeMo simulates a similar bimodal latitudinal distribution of CH_4 uptake fluxes with an RSME that is 16.86 mg CH_4 m^{-2} y^{-1} lower than other models when fitted to $10°$ latitudinal averages of observational data. In contrast, the C07 and R99 models both predict a latitudinal distribution of soil

10   methanotrophy that has CH_4 uptake maxima in equatorial regions and lower rates of CH_4 oxidation at mid-latitudes (~$40°$N and 20 to $40°$ S), resulting in higher RSMEs of 28.6 and 72.1 mg CH_4 m^{-2} y^{-1}, respectively, when fitted to the $10°$ latitude averaged data. The R99 model significantly overestimates CH_4 uptake fluxes in the tropics ($20°$N to $20°$S) and underestimates CH_4 oxidation in the subtropics (20 to $40°$ N and S), resulting in large differences for these regions relative to the MeMo simulations (Fig. 6e). The C07 model predicts a latitudinal pattern of simulated CH_4 fluxes that is similar to R99; however,

15   with much lower uptake fluxes in the tropics and no pronounced minima in the subtropics. Consequently, the RSME of the fit to observational data is much lower and regional differences relative to MeMo generally are smaller, ranging from 30% in the tropics to 20% in the subtropics (Fig. 6d).

The regional differences between MeMo and the R99 and C07 models result from differences in the parameterization of factors that govern CH_4 oxidation rates in the models: k_0, r_{SM}, r_T and r_N. The lower k_0 assigned to tropical wet forest (see

20   section 2.3.3) accounts for the reduction in CH_4 uptake by tropical soil in MeMo. The strong agreement between MeMo simulation results and CH_4 uptake measurements presented in Fig. 5 suggests that the empirically derived lower k_0 value more accurately reflects soil CH_4 oxidation rates in the tropics. However, we note the possibility that additional factors, or unexpected combinations of current factors, may influence rates of atmospheric CH_4 uptake in the tropics in ways that are not explicitly represented in the models.
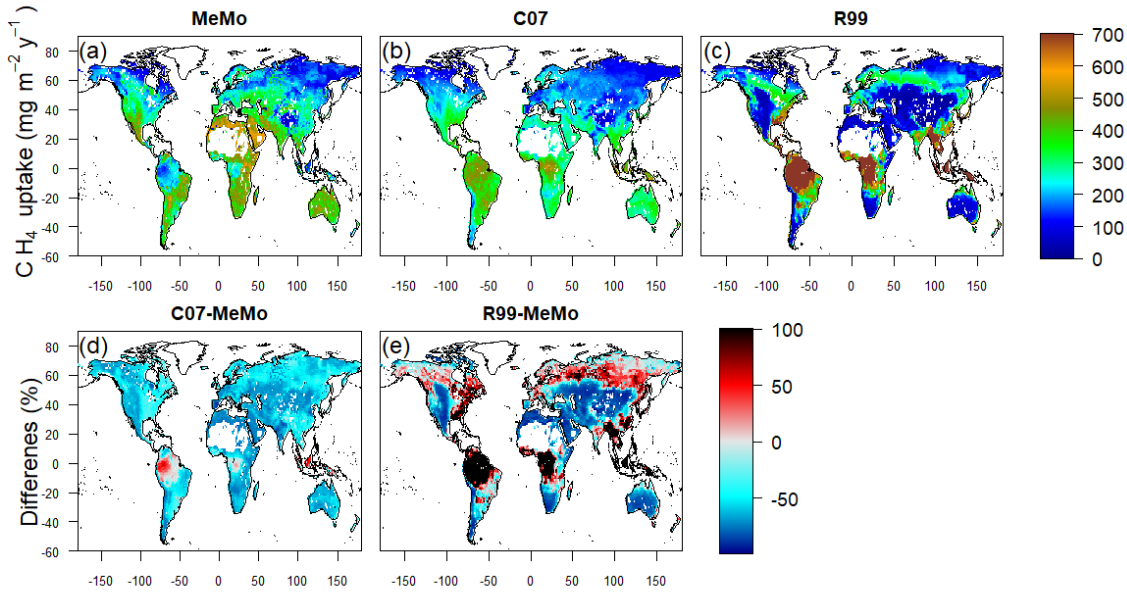
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Figure 6: Annual mean CH₄ uptake by soil predicted using models (a) MeMo, (b) C07 and (c) R99 for the period 1990-2009. Differences between models expressed in percent are shown in panels (d) C07 minus MeMo and (e) R99 minus MeMo.

The influence of different environmental factors on soil CH₄ uptake was assessed by calculating the global CH₄ uptake flux while varying each factor (temperature, soil moisture and nitrogen input) independently and keeping other factors constant (Figs. 7, 8 and 9). Comparison of r_{SM} values reveals large differences across models in tropical wet regions (Fig. 7), which explains the contrasting predictions of CH₄ uptake by MeMo (213 mg CH₄ m⁻² y⁻¹) versus models R99 (689 mg CH₄ m⁻² y⁻¹) and C07 (329 mg CH₄ m⁻² y⁻¹). Formulation of r_{SM} in MeMo (section 2.3.4) accounts for limitation of methanotrophic oxidation rates when soil moisture levels are >20% water content, a feature that is absent in the R99 and C07 models. In addition, the R99 model implements a linear decrease of r_{SM} for soil moisture conditions <20%, which results in a 60 to 80% reduction in CH₄ oxidation rates in the subtopics. The absence of this condition in models MeMo and C07 explains the significant differences in CH₄ uptake fluxes in subtropical regions (Figs. 5 and 6).
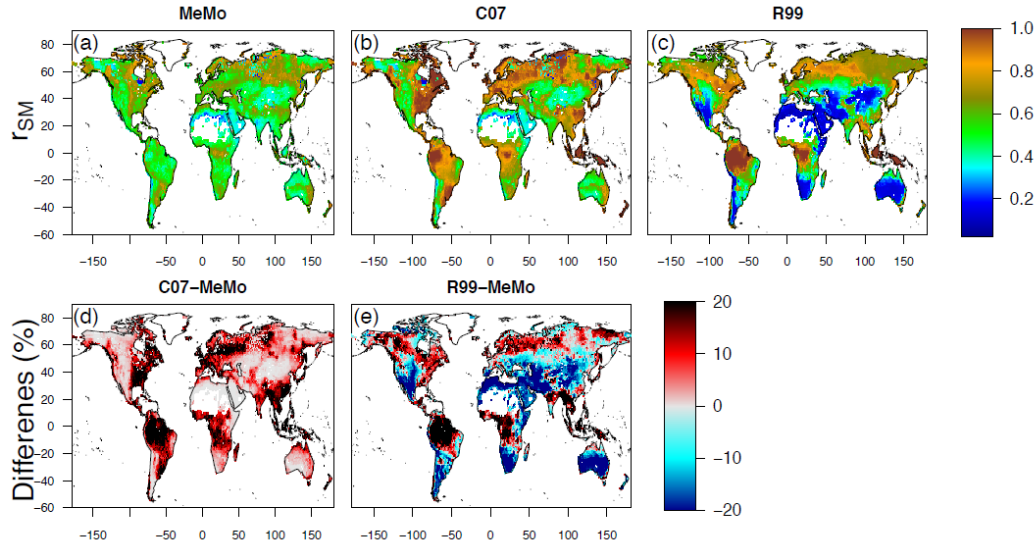
Figure 7. Soil moisture response (r_{SM}) of CH$_4$ oxidation simulated by models (a) MeMo, (b) C07 and (c) R99. Differences in model response expressed in percent are shown in panels (d) C07 minus MeMo, and (e) R99 minus MeMo.

Formulations of r_T are similar in the three models (section 2.3.5) and consequently, gridded maps of simulated r_T values exhibit broadly similar global patterns in which high r_T values are present at warm low latitudes and low r_T values are predicted at cold high latitudes. Notably, MeMo generally simulates r_T values that are approximately 20% lower than those predicted by the C07 and R99 models (Fig. 8) because of the revised formulation of the Q$_{10}$ value. MeMo and the C07 model simulate higher r_T values than R99 at high latitudes because of differences in parameterization of r_T at temperatures near 0°C.
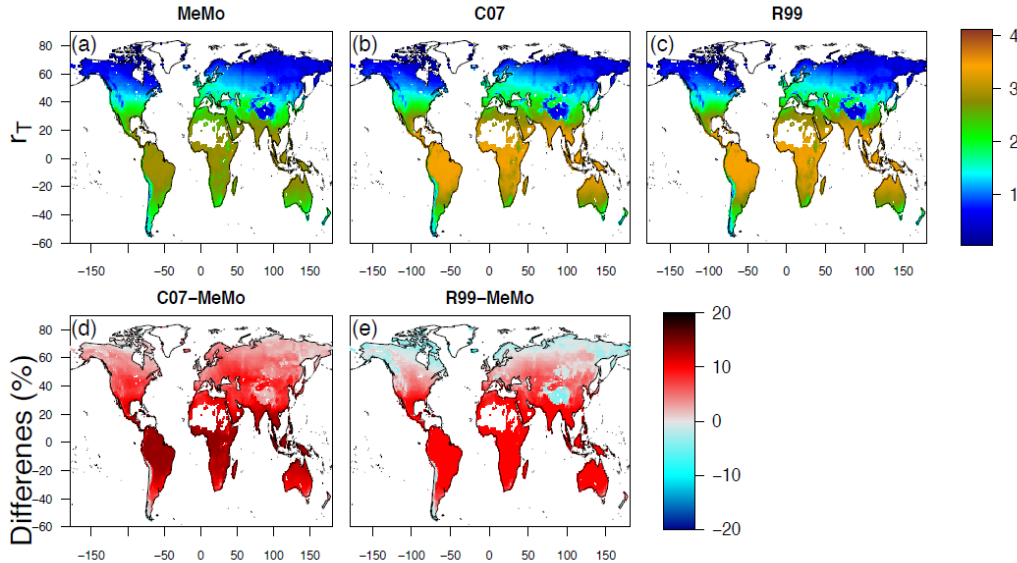
Figure 8: Temperature response (r_T) of soil methanotrophy simulated by models (a) MeMo, (b) C07, and (c) R99. Differences in model response expressed in percent are shown in panels (d) C07 minus MeMo, and (e) R99 minus MeMo.

Inhibition of soil methanotrophy due to N (r_N) differs significantly between the three models. Nitrogen inhibition of CH$_4$ oxidation rates is lower in MeMo compared to the R99 and C07 models, in particular, at mid-latitudes (Fig. 9). The R99 and C07 models formulate r_N as a function of agricultural intensity in contrast to MeMo which uses modelled N deposition and N input via fertilizers. The difference in approach results in an r_N factor that is up to ~20% higher ~~r_N factor~~ in MeMo across most regions with the exception of high latitude areas (Fig. 9).

In regions of intense agricultural activity and high N deposition (~150 kg N ha^{-1}), such as Europe, ~~or~~ the mid-western USA, China and India MeMo predicts a reduction in CH$_4$ uptake rates of up to 60% ~~o~~in average, which is consistent with R99 and C07 models. ~~.~~ ~~H~~however, inhibition of methanotrophy simulated by MeMo in areas experiencing low rates of N deposition is much smaller than R99 or C07 ~~is ~4%~~. The key limitation of the N effect approach adopted in the R99 and C07 models is the generalization of N inhibitory effects across different agricultural areas, crops and types of management, which results in a homogeneous and excessive attenuation of CH$_4$ oxidation rates. In contrast, the MeMo r_N parametrization employs a more conservative r_N factor and a realistic regional distribution, which is based upon observational data, that is consistent with recent studies reporting that high rates of N deposition (10 kg N ha^{-1} y^{-1}) can reduce soil uptake of atmospheric CH$_4$ by ~8.6% (Fang et al., 2014; Zhang et al., 2008). Direct application of fertilizers at more extreme rates >300 kg N ha^{-1} y^{-1} can entirely eliminate uptake of atmospheric CH$_4$ by agricultural soil (Veldkamp et al., 2001). Nevertheless, the importance of accurate characterization of the attenuating effects of N addition on soil methanotrophy highlights the need for additional efforts to verify and refine parameterization of this key factor.

~~, the R99 and C07 models predict a reduction of up to 60% in CH$_4$ oxidation rates compared to a decrease of only ~10% simulated in MeMo. Two recent studies reported that high rates of N deposition (10 kg N ha^{-1} y^{-1}) can reduce soil uptake of atmospheric CH$_4$ by ~8.6% (Fang et al., 2014; Zhang et al., 2008), which is consistent with the lower r_N values employed in MeMo. The key limitation of the N effect approach adopted in the R99 and C07 models is the generalization of N inhibitory effects across different agricultural areas, crops and types of management, which results in excessive attenuation of CH$_4$ oxidation rates. For example, the R99 and C07 models predict a 75% reduction in CH$_4$ uptake rates for areas characterized by an agricultural intensity of 100%; however, Veldkamp et al. (2001) reported that inhibition of soil methanotrophy at such levels requires N addition >300 kg N ha^{-1} y^{-1}, a loading rate which greatly exceeds N input levels typically associated with agriculture. Although a more conservative r_N factor is employed in MeMo, the importance of accurate characterization of the attenuating effects of N addition on soil methanotrophy highlights the need for additional efforts to verify and refine parameterization of this key factor.~~
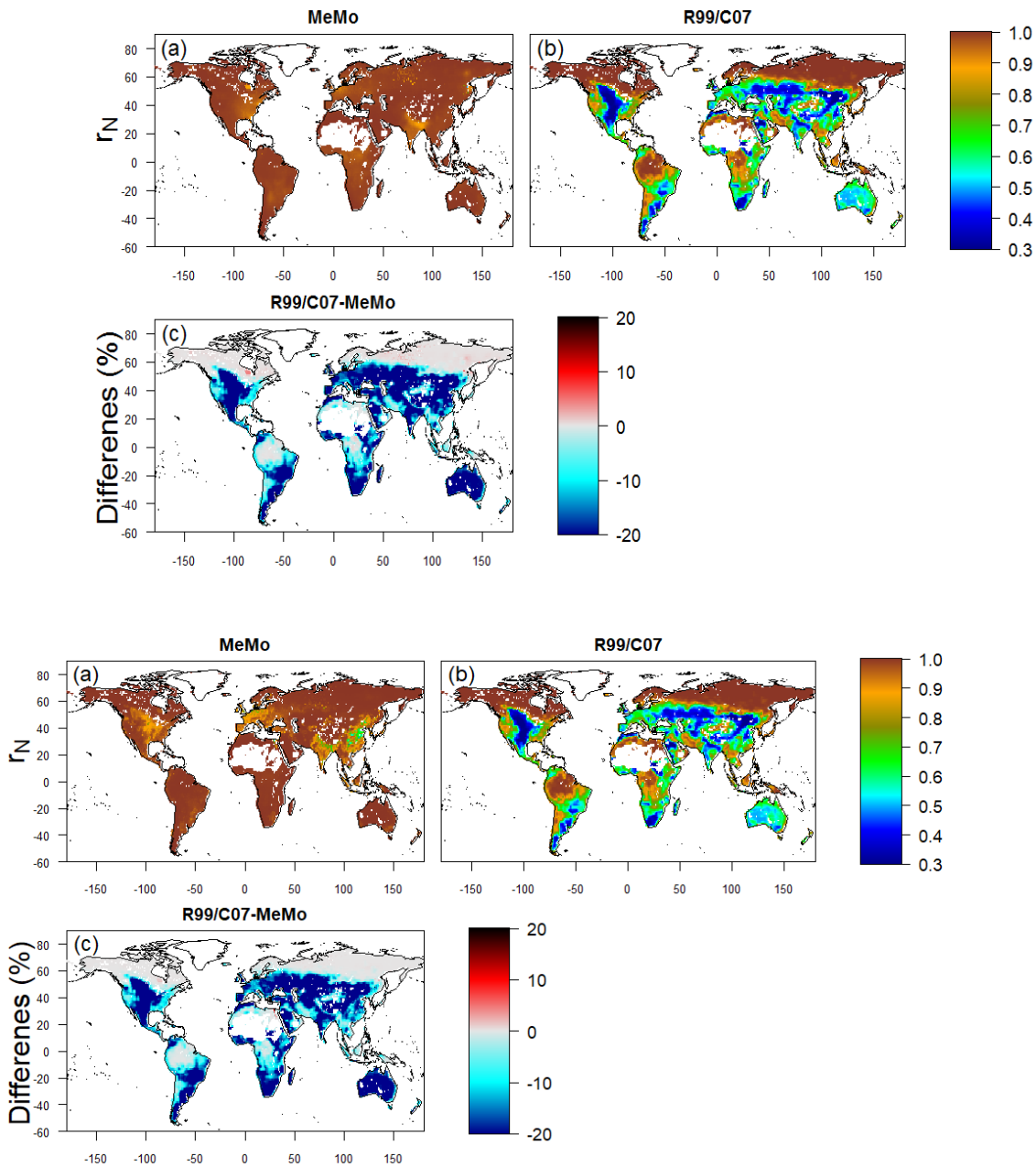
Figure 9. Response of soil methanotrophy to nitrogen effect (r_N) simulated by models (a) MeMo, (b) R99, and C07. The responses for models R99 and C07 are both shown in panel (b) because they have the same formulation. The difference in model response between models R99/C07 minus MeMo expressed in percent is shown in panel (c).

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4.3 Temporal and Spatial Variability of Soil CH$_4$ Uptake

Field observations of soil uptake of atmospheric CH$_4$ are generally sparse both spatially and temporally. Consequently, our quantitative understanding of CH$_4$ uptake fluxes across different ecosystems and seasons is limited. Models provide a means to quantitatively explore spatial and temporal patterns of soil methanotrophy on scales that cannot be readily captured by field-based observations. Therefore, once tested and validated (see section 4.2), MeMo was used to quantitatively assess the variability of soil CH$_4$ uptake in different climate zones and ecosystems on seasonal time scales.

4.3.1 Regional Variability

The relative contribution of soil in each climatic zone to global uptake of atmospheric CH$_4$ as predicted by MeMo is summarized in Table 8. Soil in the northern hemisphere is estimated to account for approximately two thirds (65~~7~~%) of the total global sink for atmospheric CH$_4$ because of the uneven distribution of landmasses between the northern and southern hemispheres. Notably, terrestrial areas in the northern subtropical and temperate zones collectively account for ~45~~6~~% of the global soil sink for atmospheric CH$_4$. The southern tropical zone contributes a further ~19% to soil uptake of CH$_4$. The southern subtropical and northern tropical zones are estimated to contribute almost equally (~14%) to total CH$_4$ uptake (Table 8). The smallest proportion of soil CH$_4$ oxidation occurs in the southern temperate (0.6~~5~~%) and northern polar (5~~6~~%) zones due to a combination of small land area and low rates of CH$_4$ uptake. Model predictions of CH$_4$ uptake by climatic zone provides insights into the relative importance of each region in the global CH$_4$ cycle but additionally begins to facilitate analysis of potential responses of the soil CH$_4$ sink within each zone to global change both due to climate and land management.

Table 8. MeMo CH$_4$ uptake estimates by region.

Regions	Regional gridded mean (mg CH$_4$ m^{-2} y^{-1})	Total land area (10^{12} m^{-2})	Total CH$_4$ uptake (Tg CH$_4$ y^{-1})	% of total
Cold zone (60°-90° N)	100.1~~2~~	18.7	1.87~~2.2~~	5.6~~4~~
Temperate zone (40°-60° N)	217.0	31.0	6.7~~9~~	20.0~~1~~
Subtropic zone (20°-40° N)	326.6~~44~~	26.4	8.6~~9.1~~	25.7~~26.5~~
Tropical zone (0°- 20° N)	309.2~~13~~	15.1	4.6~~7~~	13.9~~7~~
Northern Hemisphere Total:		**91.2**	**21.9~~2.9~~**	**65.3~~6.7~~**
Temperate zone (40°-60° S)	234~~12~~	1.1	0.2	0.6~~5~~
Subtropic zone (20°-40° S)	363.7~~85~~	13.3	4.8~~7~~	14.3~~13.7~~
Tropical zone (0°- 20° S)	313.9~~22~~	20.8	6.5	19.4~~18.9~~
Southern Hemisphere Total:		**35.2**	**11.6~~4~~**	**34.6~~33.3~~**

Further analysis of soil CH$_4$ uptake by ecosystem types (Table 9) shows that the highest gridded mean rates of CH$_4$ oxidation are associated with tropical deciduous forests (602~~30~~ mg CH$_4$ m^{-2} y^{-1}). The relatively low soil moisture content during the dry season (Supplementary 2, Figure S3) and the consistently high mean annual temperature (Supplementary 2,

Figure S7~~6~~) in such ecosystems promote high rates of soil methanotrophy. Furthermore, the soil typically possesses a low clay content (Supplementary 2, Figure S2), which results in higher porosity that enhances gas diffusion and promotes higher rates of CH_4 oxidation. In comparison, rates of CH_4 uptake by soil in open and dense shrubland, temperate evergreen forest and savanna ecosystems (Table 9) are ~100 mg CH_4 m^{-2} y^{-1} lower but still highly significant globally.

5   Dense and open shrubland are characterized by constant climatic conditions (temperate and relatively low soil moisture; Supplementary 2 Figures S7~~6~~ and S3, respectively) throughout the year, which in combination with a texture that typically is sandy results in high annual CH_4 uptake rates (Tate et al., 2007). In contrast, high annual rates of CH_4 uptake in temperate evergreen forests result from elevated rates of soil methanotrophy during summer months (section 2.3.4), indicating that temperature is a key driver of CH_4 oxidation in such ecosystems (Borken et al., 2006; Ueyama et al., 2015; Wang and

10 Ineson, 2003). Savannas share many climatic conditions with tropical deciduous forests but also commonly experience wildfire during the dry season. Both ecosystem types though are characterized by a marked seasonality driven by the presence or absence of precipitation in combination with a consistent high mean annual temperature (Supplementary 2, Figure S7~~6~~ and S3), which collectively support high rates of CH_4 uptake by soil.

     Tundra, taiga, polar desert and other ecosystem types that are common at high latitudes (Supplementary 2, Figure

15 S10~~9~~) are characterized by the lowest mean annual rates of soil methanotrophy (<1800~~8~~ mg CH_4 m^{-2} y^{-1}) because of low temperatures throughout most of the year. MeMo also predicts low rates of CH_4 uptake in tropical humid forest (332~~0~~ mg CH_4 m^{-2} y^{-1}) due to low rates of bacterial CH_4 oxidation and the negative impact of high soil moisture levels on gas diffusion (see section 2.3.5). The CH_4 uptake rates estimated by MeMo are consistent with field observations by Dasselar et al. (1998) and Luo et al. (2013) which indicate that excess soil moisture strongly attenuates CH_4 uptake rates across a range of ecosystem

20 types.

     Finally, the global significance of each ecosystem type as a CH_4 sink depends strongly on spatial extent as well as CH_4 oxidation rates. Open shrubland (19~~8~~.7%), grassland and steppe (15.0~~12.7~~%), and savanna (13.4~~11.3~~%) are the most important ecosystem types contributing to the global CH_4 soil sink (~48~~6~~% collectively; Table 9) in MeMo because of high mean rates of CH_4 uptake (392~~39~~ to 518~~71~~ mg CH_4 m^{-2} y^{-1}) in combination with a large areal extent globally (14 x 10^{12} to 23

25 x 10^{12} m^2). This finding is similar to the estima~~tion~~ted reported by Potter et al. (1996) that warm and relatively dry ecosystems, such as semi-arid steppe, tropical savanna, tropical seasonal forest, and chaparral, account for 40% of soil uptake of atmospheric CH_4 globally. Moreover, Luo et al. (2013) reported the highest annual CH_4 uptakes rates in dry savanna as part of a long-term field investigation of soil methanotrophy in several ecosystem types. Singh et al. (1997) also observed CH_4 uptake rates that were higher in savannah than temperate forest. Although both model simulations and available field

30 observations suggest these ecosystems are important global sinks for atmospheric CH_4 there is presently a dearth of field measurements for warm and dry environments relative to temperate ecosystems.

Table 9. MeMo CH_4 uptake estimates by ecosystem type from Ramankutty and Foley (1999) land cover classification.

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| Ecosystem type	Global gridded mean (mg CH_4 m^{-2} y^{-1})	Total land area ($x10^{12}$ m^{-2})	Total CH_4 uptake (Tg CH_4 y^{-1})	% of total
Tropical Deciduous Forest	60230 ± 6359	4.2	1.67	4.75.0
Dense Shrubland	580 ± 104	6.1	2.4	6.9
Open Shrubland	51871 ± 1342	23.3	6.67.3	19.721.2
Temperate Broadleaf Evergreen Forest	51226 ± 8270	2.0	0.67	1.72.0
Savanna	5004 ± 13228	14.1	4.5	13.41
Dense Shrubland	481 ± 90	6.1	2.4	7.1
Temperate Needleleaf Evergreen Forest	359 ± 90	3.9	1.2	3.4
Grassland/Steppe	39239 ± 11092	15.8	5.04.3	15.012.5
Temperate Needleleaf Evergreen Forest	347 ± 90	3.9	1.2	3.5
Temperate Deciduous Forest	32131 ± 7068	5.2	1.4	4.14.0
Tropical Evergreen Forest	3320 ± 454	12.5	2.5	7.42
Boreal Deciduous Forest	2828 ±1179	5.7	1.5	4.4 3
Boreal Evergreen Forest	26975 ± 946	9.1	2.4	7.10
Mixed Forest	1827 ± 826	13.4	2.7	8.07.8
Tundra	1769 ± 1434	6.2	1.1.7	3.24.9
Polar Desert/Rock/Ice	1058 ± 48	0.4	0.01	0.0
Total		**124.1**	**33.534.3**	**100**

4.3.2 Seasonal Variability

Global annual uptake of atmospheric CH_4 by soil exhibits a marked seasonality that reflects the dominance of the northern hemisphere in the soil sink methanotrophy. The highest simulated CH_4 uptake fluxes occur during June, July, August (JJA)

5 (10.34 Tg CH_4) followed by September, October and November (SON) (10.12 Tg CH_4), March, April and May (MAM) (6.87.2 Tg CH_4), and finally, December, January and February (DJF) (6.35 Tg CH_4) (Fig. 10).

Methane uptake in the cold and temperate regions of the northern hemisphere generally is characterized by the largest seasonality exhibiting an amplitude of 30 mg CH_4 m^{-2} mo^{-1}. In these regions, modeled uptake of CH_4 by soil is controlled strongly by temperature and consequently, ecosystems common at these latitudes (*e.g.,* boreal, needle leaf, temperate

10 deciduous, mixed forest, polar deserts/rock/ice and tundra) show pronounced seasonal trends (Fig. 11), which also are evident in field measurements (*e.g.*, Priemé and Christensen, 1997) and emphasized in local mechanistic models (*e.g.*, Oh et al., 2016). These finding suggest that the soil CH_4 sink in such ecosystems may be more sensitive to future change as a result of global warming.

In contrast, soil methanotrophy in temperate regions in the southern hemisphere are characterised by a weaker

15 seasonality having an amplitude of 17 mg CH_4 m^{-2} mo^{-1} due to the prevalence of grassland and steppe, which contrasts with a dominance of forest in the northern hemisphere. Seasonality of soil CH_4 uptake fluxes is even more muted in tropical and subtropical environments (<10 mg CH_4 m^{-2} mo^{-1}) because of favourable and stable environmental conditions. Tropical deciduous forest and tropical evergreen forest, which are common in these climate zones are characterized by relatively

constant CH$_4$ uptake fluxes throughout the year (Fig. 11); however, MeMo predicts greater seasonality (20 mg CH$_4$ m^{-2} mo^{-1}) of CH$_4$ uptake by soil in drier subtropical ecosystems, such as open shrubland, savanna and grasslands (Fig. 11) because of seasonality in soil moisture.

Notably, northern temperate forest in summer (JJA) was the ecosystem and time period possessing the highest average monthly CH$_4$ uptake fluxes (76.7~~137~~ mg CH$_4$ m^{-2} mo^{-1}) simulated by MeMo. During the rest of the year, the largest soil sink for atmospheric CH$_4$ occurred in the southern hemisphere in tropical deciduous forest of central Africa (DJF, 69.5~~132~~ mg CH$_4$ m^{-2} mo^{-1}; MAM, 73.5~~131~~ mgCH$_4$ m^{-2} mo^{-1}; SON, 75.5~~129~~ mg CH$_4$ m^{-2} mo^{-1}). This finding is significant because field observations of soil methanotrophy in northern temperate forest during summer are the measurements most commonly extrapolated to an annual basis, which may lead to a possible overestimation of global CH$_4$ uptake fluxes.

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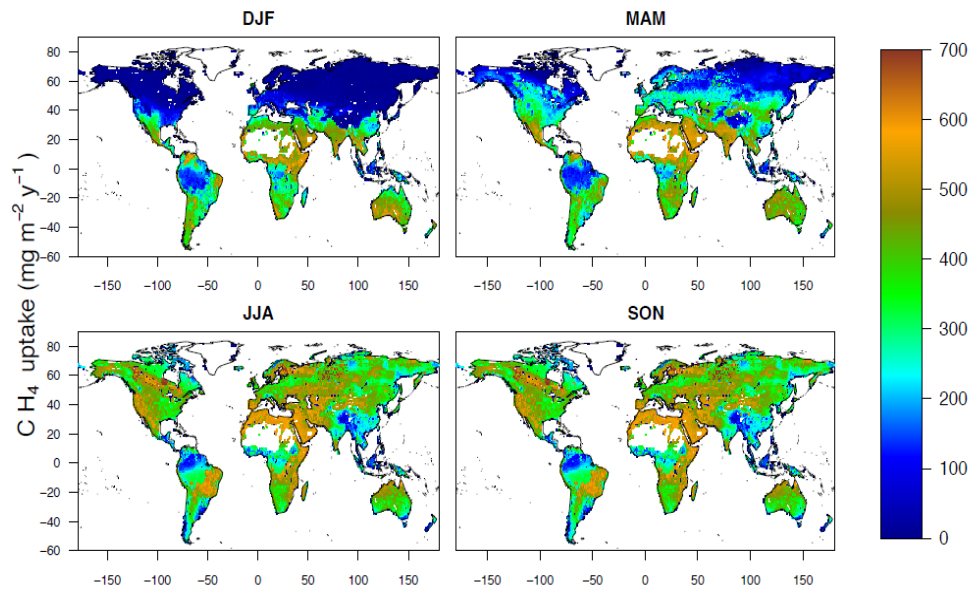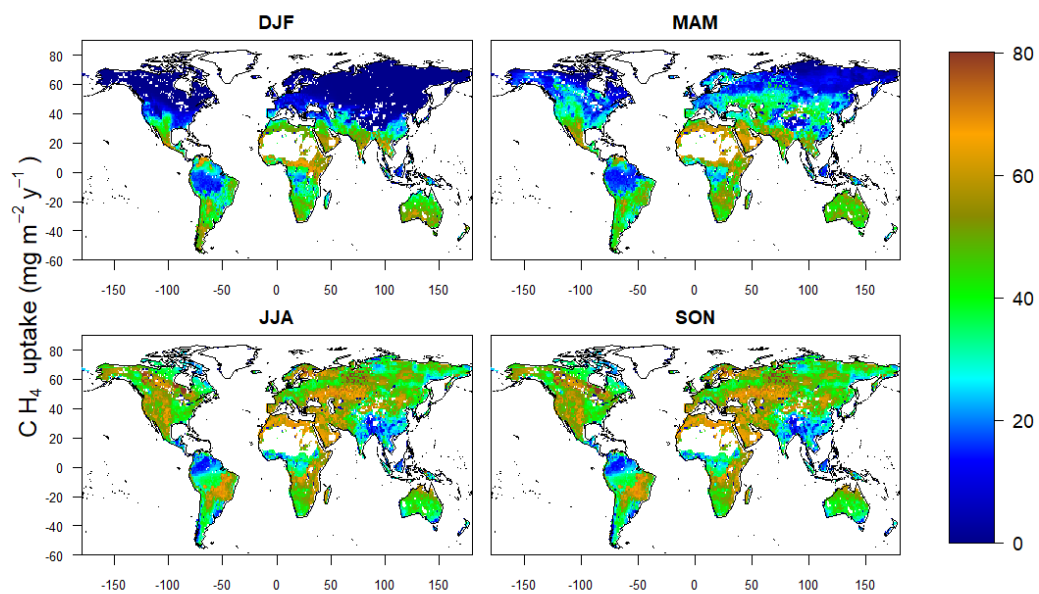
Figure 10: Seasonal uptake of atmospheric CH$_4$ by global soils predicted by MeMo for the period 1990 to 2009.
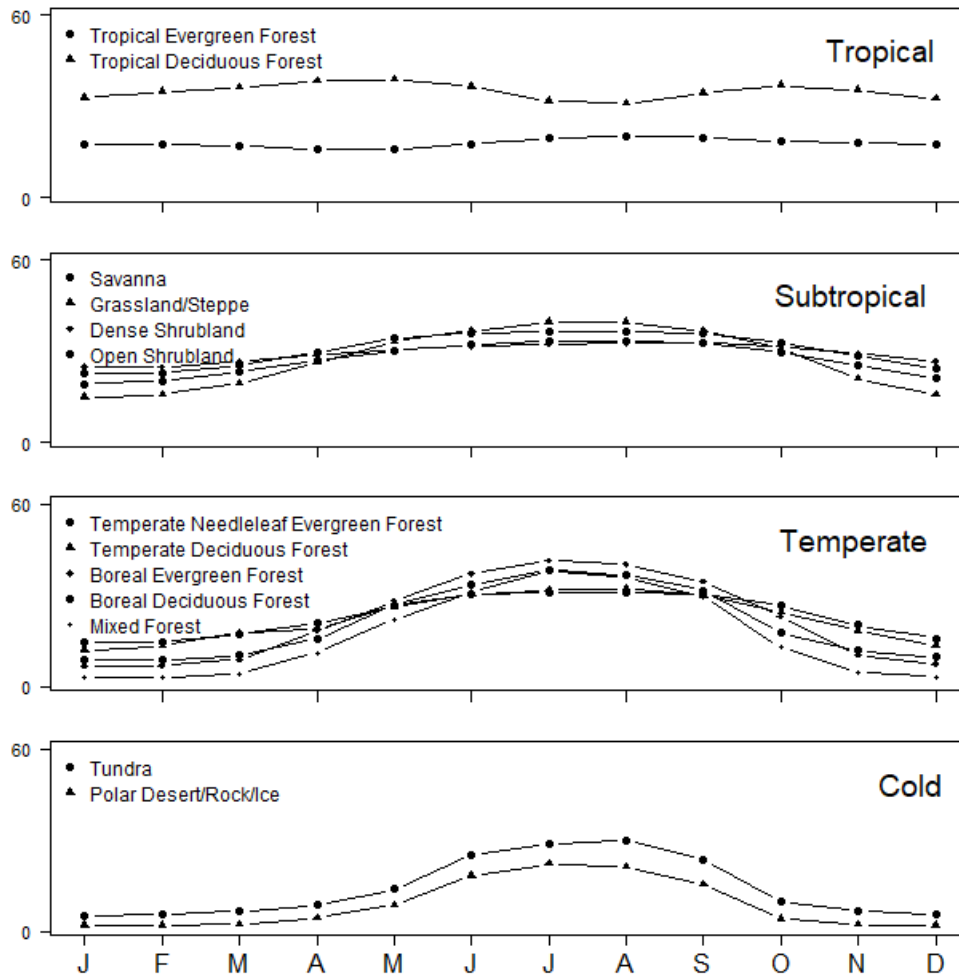
Figure 11: Seasonal patterns of soil uptake of atmospheric CH$_4$ by ecosystem for the four regions: cold, temperate, tropical and subtropical using MeMo model for the period 1990-2009.

4.4 Model Limitations and Scope of Applicability

Several aspects of MeMo can be developed further, pending availability of new additional field data, to improve estimation of global soil uptake of atmospheric CH$_4$. Firstly, the base oxidation rate of bacterial methanotrophy at 0°C (k_0) is a critical parameter necessary for accurate estimation of CH$_4$ uptake rates. There is presently a general dearth of published k_0 values for soil methanotrophy and moreover, ecosystem coverage is incomplete. Additionally, our parametrization for k_d accounts for methanotrophic activity in a one-dimensional soil matrix; however, other studies have separated CH$_4$ uptake in soil from methanotrophy in the rhizosphere to improve estimates of total CH$_4$ uptake (*e.g.*, Sabrekov et al., 2016). This refinement has been modeled for local conditions but insufficient data about rhizosphere CH$_4$ oxidation rates prevent inclusion in MeMo and

extension to a global scale. Secondly, the Q_{10} response of soil methanotrophy has been determined to date in only a small subset of ecosystems in which soils function as a sink for atmospheric CH_4. The majority of Q_{10} values have been determined for bacterial oxidation of CH_4 under laboratory conditions and there is considerable variability in values across different ecosystems. Thirdly, additional field observations of CH_4 uptake by soil are needed, in particular, long-term measurements at

5   individual sites that capture seasonality and inter-annual variability and from regions that presently have minimal or no representation (*i.e.*, the southern hemisphere, semi-arid ecosystems, etc.) in the current pool of observations. Fourthly, additional observations and characterization of the effects of N deposition on soil methanotrophy are needed. The measurements ideally should be conducted *in situ* using N input rates that are appropriate for different environments and land use practices. MeMo can be used to guide new field and laboratory experiments to address the lack of parameterization data,

10   in particular, k0 and Q10 values for soil methanotrophy in different ecosystem and latitudes, and long-term *in situ* studies of N inhibition on CH_4 uptake by soil. It also can be used to compare results from short- and long-term investigations of CH_4 uptake in field and laboratory experiments.

MeMo is also parameterized to accommodate input of CH_4 from below (*i.e.*, subsurface methanogenesis or upward migration of deeply sourced CH_4); however, rigorous validation of that aspect of the model will require additional field

15   observations, including better characterization of conditions under which CH_4 is produced in finely textured soils and deep sub-horizons. The presence, or periodic input, of high concentrations of CH_4 (*e.g.*, from permafrost melting) may impact competition for oxygen and niche space between low affinity CH_4-oxidizing bacteria and the high affinity methanotrophs responsible for uptake of atmospheric CH_4. Refinement and validation of the capacity for MeMo to account for upward migrating or autochthonous CH_4 will enable the model to be used to estimate CH_4 flux from intermittently wet environments,

20   which may currently fall outside the scope of process-based wetland models.

The process-based nature of MeMo and the breadth of conditions for which it has been validated provide scope for using the model to quantify CH_4 uptake in soil in a broad range of scenarios. For example, MeMo could be used to determine global uptake of CH_4 by soil in the past during glacial or former interglacial periods. It may also be used to assess potential uptake rates of atmospheric CH_4 in future climate scenarios and further elevated tropospheric CH_4 mixing ratios. Additionally,

25   MeMo can be used to evaluate the impact of different proposed policies and mitigation strategies for managing the atmospheric burden and growth rate of CH_4 because of its capacity to evaluate different future scenarios based upon parameterization of key drivers that impact rates of CH_4 uptake by soil globally.

5.0 Conclusions

We developed a processed-based model to simulate uptake of atmospheric CH_4 by soil, which was refined using newly reported

30   experimental data and the introduction of recent insights into physical and biological mechanisms that drive soil methanotrophy. We modified the general analytical solution proposed by Ridgwell et al. (1999) and Curry (2007) to account for a maximum depth of CH_4 uptake and to quantify upward migration and consumption of CH_4 produced *in situ*. Representation of the effects of N deposition and input via fertilizers, soil moisture and temperature on methanotrophy were improved based upon newly available data and recent advances in characterization of these processes. Finally, we proposed

utilization of a different base oxidation rate k_0 for methanotrophy in different regions because its value changes in relation to environmental conditions.

MeMo simulations produced a closer fit to observational data than two previous soil methanotrophy models (Ridgwell et al., 1999; Curry 2007). MeMo and observational data show a similar bi-modal latitudinal distribution of atmospheric CH_4 uptake by soil with the lowest fluxes at the equator and high-latitudes, and largest uptake fluxes at mid-latitudes. Previous models simulated a dissimilar pattern with large uptake fluxes in equatorial regions, a difference that results primarily from improved representation of the soil moisture effect in MeMo.

MeMo simulations supported by observational data indicate that warm and semiarid regions are the most efficient soil sink for atmospheric CH_4. In these regions, tropical deciduous forest and dense open shrubland are characterized by relatively low soil moisture and constant temperature during the year, which are key factors that promote high rates of CH_4 uptake by soil. In contrast, cold regions possessed the lowest CH_4 uptake rates, in particular, tundra and boreal forest, which have a marked seasonality driven by temperature, making soil methanotrophy in such areas potentially sensitive to future global climate change. The warm and wet tropical evergreen forest biome has CH_4 uptake rates that are ~50% less than warm and semiarid regions because excess soil moisture impacts soil-atmosphere gas exchange, resulting in a smaller k_0 (1.6×10^{-5} s^{-1}). The extensive area of shrubland, grassland, steppe and savanna globally yields a high total uptake of CH_4; however, there is presently a dearth of experimental data for these biomes and additional field observations are required to strengthen validation of MeMo simulations for these globally extensive areas.

MeMo simulations indicate that global soil uptake of atmospheric CH_4 is reduced 4% o̶n̶ in average and by as much as 60̶1̶0̶% in regions that receive high rates of atmospheric N̶nitrogen deposition and N input fertilizers.

Globally, N deposition and input via fertilizers attenuates the soil sink for atmospheric CH_4 by 1.38 Tg yr^{-1}, which is two to five times less than previously reported values because of the refined representation of the nitrogen inhibition on soil CH_4 oxidation in MeMo.

The accuracy of quantifying the modern soil sink for atmospheric CH_4 is improved using MeMo. In addition, the model can be used to explore changes in the relative importance of soil methanotrophy in the global CH_4 cycle in the past and the capacity of the soil sink to consume atmospheric CH_4 under future global change scenarios.

Code and Data Availability

MeMo was implemented in R (version 3.0.1). The model code and model output for 1990-2009 are available as a supplement to this manuscript. In addition, we also provide a post-processed driving dataset to run an a̶ ̶s̶i̶m̶p̶l̶e̶, example model case study for year 2000. All the forcing data used in this study are available from the following sources:

- temperature from CRU3.1, Harris et al. (2014): https://crudata.uea.ac.uk/cru/data/hrg/;

- vegetation mask from Ramankutty and Foley (1999): https://nelson.wisc.edu/sage/data-and-models/global-potential-vegetation/index.php;
- soil moisture from Dorigo et al. (2011) (Satellite): http://www.esa-soilmoisture-cci.org;
- soil moisture from TRENDY: (Sitch et al., 2015): http://www-lscedods.cea.fr/invsat/RECCAP/;
- nitrogen deposition from Lamarque et al. (2013):
  - 1) https://www.isimip.org/gettingstarted/downloading-input-data/
  - 2) https://www.isimip.org/gettingstarted/details/24/;
- N input via fertilizers from Nischina et al. (2017): https://www.earth-syst-sci-data.net/9/149/2017/.
- clay content and bulk density from Shangguan et al. (2014): http://globalchange.bnu.edu.cn

Author contributions. FMF and SA developed and modified the model. FMF and GMT created the code. FMF ran the simulations, analysed the data and created all figures. All authors contributed to the interpretation of the results and preparation of the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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