

## Responses to Anonymous Referee #1

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We thank the referees for their helpful comments and improvement suggestions for our paper. Below, we enumerate the referees' comments and give our responses following each comment. The corresponding changes we have made in the revised manuscript are indicated with red font and the line numbers are reported below.

### 5 **Anonymous Referee #1, C8, January 31, 2015**

1. *The number of function evaluations will be high for high-resolution models. How your method faces this challenge?*

10       Reply: There is always the question of how many function evaluations one needs to perform  
to get a fixed level of accuracy for optimization. Our results on many difficult problems (e.g.  
problems with multiple local minima) indicate that surrogate global optimization methods can  
usually obtain more accurate results compared to non-surrogate methods with the same limited  
number of evaluations (references including, for example, Mugunthan et. al (2005): *Compar-*  
15       *ison of function approximation, heuristic, and derivative-based methods for automatic cali-*  
*bration of computationally expensive groundwater bioremediation models*, Water Resources  
Research, vol 41, W11427). It is a very difficult problem to find the best values of the param-  
eters for climate models, and the more evaluations one does, in general the better the answer.  
The number of evaluations needed depends upon the problem, for example, in Figure 4, a  
very good answer is obtained by all three trials within 40 evaluations whereas other problems  
20       require more evaluations. Given this is a problem with multiple local minima, we would not  
expect any other method (for example, evolutionary algorithm or multi-start local optimization

with numerical derivatives) to do better with this same number of model evaluations on a specific problem (Simple numerical inversion by solving a matrix equation is not possible since the problem has multiple local minima and hence is not linear or convex.). It is not necessary  
25 to do all the evaluations presented in the graph. For example, Figure 2 shows that quite a good result (reducing the error by almost an order of magnitude) is obtained for the five parameter case.

Given the importance of climate modeling projections, using the computer time for hundreds  
30 of evaluations to get better parameters (based on data) that provide better forecasts is certainly worthwhile. The parameter estimation is usually only done once based on historical data and does not need to be repeated for different CH<sub>4</sub> scenarios unless the model equations change. We addressed this issue on lines 397-406, red text, in the main manuscript.

2. *The expensive evaluation at step 5 in Algorithm 1 should provide a different value even for the same vector  $x_{new}$ , this is because realistic models are able to reproduce different results for the same input vector in different runs (the results are similar but they are not exactly the same one) Are there any problems in the use of realistic models in this step?*  
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Reply: The algorithm we apply here is for deterministic models, i.e., the simulated CH<sub>4</sub> emissions given a variable vector  $x^*$  will always be the same whenever we run the model for  $x^*$ .  
40 CLM4.5bgc and related codes are deterministic models. This is because we are using the land model forced by observationally-based datasets, not in the climate model mode. It is possible to use surrogate optimization codes for noisy functions, but that is not what we have done here since the NCAR codes like CLM4.5 are deterministic. We included a comment about this topic in the revised manuscript on page 2, lines 43-45, red text, and also in lines 162-164, red  
45 text.

3. *According to this: “the matrix in equation 8 is invertible if and only if  $\text{rank}(P) = d+1$ ” Whenever this is true, how do you guaranty that this matrix is well-conditioned? Is it based on the set of basis  $\Phi$ , what about having a degenerated basis?*

Reply: The matrix would become ill-conditioned if the selected sample points were too close  
50 to each other. We prevent this from happening during the sampling procedure in which we require that the sample points must have a minimum distance to each other. The details of the algorithm are given in the cited paper (Regis and Shoemaker, 2013, *Combining radial basis function surrogates and dynamic coordinate search in high-dimensional expensive black-box optimization*, Engineering Optimization, 45, pages 529-555). See also lines 368-371, red text,  
55 in the main manuscript.

60 *Another question from this, if I have a problem with 80 parameters in a model whose resolution is  $10^7$  (an atmospheric model can easily reach this dimension) should I run the model 81 times per iteration? Seems to be a quiet expensive. Should be nice to have a subsection discussing these points.*

65 Reply: We do not run the CLM4.5bgc model  $d+1$  times per iteration, where  $d$  is the dimension (and  $d = 80$  in the referee's example). In this paper, we use a serial code, and we only run the climate model once for each observation site in each iteration. So the answer to the question is "No, we do not run the model 81 times per iteration". Before the optimization, we do once a number of CLM evaluations based on an initial experimental design, which can be of arbitrary size (often with  $d + 1$  points, which would be 81 points evaluated once for the referee's example). We made this point clearer in the text on line 372 (we added "one", red text).

**Anonymous Referee #2, C933, June 02, 2015**

75 1. *The applied screening mechanism evaluates the change in output behaviour by changing inputs values locally to the default values and will therefor select the parameters responsible for significant output variability in that part of the parameter space. The excluded parameters potentially influence output variability further away from the default values. Furthermore, the screening does not take into account possible interaction effects of parameters although the authors mention in section 5.1.2 the CLM effectively makes use of interactions. Therefor I'm not convinced the applied method produces the best feature ranking. Perhaps methods such as Elementary Effect [1] or Locating Arrays [2] may identify a different subset of features.*

80 Reply: Sensitivity analysis for computationally expensive functions is very hard because to thoroughly examine all the interactions among the parameters over all sizes of perturbations and over multiple parameter combinations can easily take thousands of simulations. The computation time of the CLM4.5 is very expensive for each combination of parameters considered, and so we only devoted a limited number of evaluations to sensitivity analysis, but we did so in an "intelligent" way. We use information not only from the sensitivity studies, but also by considering the terms the parameters come from and what they will introduce sensitivity to in order assess which terms to include. We added text to describe how we do this to the main manuscript in the methods section (line 292-310, red text). We selected the perturbation range that we use in the sensitivity analysis as a small but potentially significant change in the parameter values.

90 2. *The authors correctly re-weight the error contribution of each site to avoid biased results as an absolute error function is used. Also they mention the different sites are geographically*

varying but does this guarantee all sites differ significantly? If some sites are related at the level of the data (in other words, representing the same part of feature space) it means that group starts to dominate the error function. My opinion is some additional information on the diversity amongst the sites at the level of the data is needed (for instance by clustering the union of all observation data), to provide proof that the relative contribution of each site to the total error should be equal.

Reply: We did the weighting according to the amount of methane flux we observed for each location. We furthermore investigated the possibility of giving equal weights to each site and the possibility of zonal weighting. There are various other possibilities on how to assign weights to observation sites, and the referee's suggested clustering method is one of these other possibilities. For the suggested clustering, different clustering methods ( $k$ -means clustering, hierarchical clustering, etc.) and different cluster numbers, which must be specified by the user, will give different cluster memberships for each site. For each cluster assignment, weights could be derived and a separate optimization would have to be done, which in turn requires many computationally expensive evaluations. We addressed this topic in the manuscript on lines 247-253.

3. *From the results and the discussion on the real data it seems there is a difference between methane production in northern and southern regions. Have the authors considered applying multi-objective surrogate based optimization [3] to identify a set of pareto-optimal configurations? Having 16 independent objectives is perhaps too much, but splitting the sites in two groups (northern, southern) and computing a weighted RMSE as in Equation 1 for both could result in several distinct solutions of which some are more suited for different regions.*

Reply: It would have been possible to do additional experiments, but each objective function evaluation is very expensive so the number of evaluations that can be done is limited. Our focus was on demonstrating that single objective global optimization analysis is useful in identifying reasonable parameter values. The reviewer suggested that we could have done a weighted RMSE, but the results would have been dependent on the weights chosen. If the weights are 1 on all quantities, then we would just get the answer already shown. We could have done multiple combinations of weightings, but then it would have taken a large number of very expensive simulations, and it is not clear what the answers would tell us unless the weights had some physical meaning. The reviewer also suggests using a surrogate multi-objective method. However, this would take vastly more evaluations to get a reasonable tradeoff curve (Pareto Front) so it is beyond the scope of this project. There are only few surrogate multi-objective methods known to the authors and the more recent one of them is an accepted paper by Akhtar and Shoemaker, *Multi Objective Optimization of Computationally Expensive Multi-Modal Functions with RBF Surrogates and Multi-Rule Selection*, Jn of Global Optimization

130 (accepted and online) with the second author also being an author on this paper. So the decision  
not to do global multi-objective optimization was an informed decision about the number of  
simulations required to do multi-objective optimization even with a very efficient surrogate  
method. We considered 16 separate objectives and groupings of them, but we decided analysis  
of this type would have to be considered in future work and future funding. See also the main  
manuscript lines 253-259, red text.

135 4. *Technical remarks: Although it may be implied, is the applied simulator deterministic (pro-  
ducing the same value for a combination of input parameters every time)?*

Reply: We included a comment about this topic on page 2, lines 43-45, red text.

5. *Equation 2 should be part of Equation 1*

Reply: We updated the numbering (see equations (1a) and (1b)).

# CH<sub>4</sub> Parameter Estimation in CLM4.5bgc Using Surrogate Global Optimization

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**Abstract.** Over the anthropocene methane has increased dramatically. Wetlands are one of the major sources of methane to the atmosphere, but the role of changes in wetland emissions is not well understood. The Community Land Model (CLM) of the Community Earth System Models contains a module to estimate methane emissions from natural wetlands and rice paddies. Our comparison  
5 of CH<sub>4</sub> emission observations at 16 sites around the planet reveals, however, that there are large discrepancies between the CLM predictions and the observations. The goal of our study is to adjust the model parameters in order to minimize the root mean squared error (RMSE) between model predictions and observations. These parameters have been selected based on a sensitivity analysis. Because of the cost associated with running the CLM simulation (15 to 30 minutes on the Yellow-  
10 stone Supercomputing Facility), only relatively few simulations can be allowed in order to find a near-optimal solution within an acceptable time. Our results indicate that the parameter estimation problem has multiple local minima. Hence, we use a computationally efficient global optimization algorithm that uses a radial basis function (RBF) surrogate model to approximate the objective function. We use the information from the RBF to select parameter values that are most promising with  
15 respect to improving the objective function value. We show with pseudo data that our optimization algorithm is able to make excellent progress with respect to decreasing the RMSE. Using the true CH<sub>4</sub> emission observations for optimizing the parameters, we are able to significantly reduce the overall RMSE between observations and model predictions by about 50%. The CLM predictions with the optimized parameters agree for northern and tropical latitudes more with the observed data  
20 than when using the default parameters and the emission predictions are higher than with default settings in northern latitudes and lower than default settings in the tropics.

## 1 Introduction and Motivation

Methane is the second most important greenhouse gas in terms of radiative forcing (Myhre et al., 2013) and thus a major concern regarding climate change. Natural wetlands as well as human activities such as agriculture (for example, rice cultivation) contribute to the methane emissions (Ciais et al., 2013). The role of wetlands in the total budget of methane, as well as in driving inter-annual variability and changes in the methane growth rate is not well understood (e.g. Bloom et al. (2010); Dlugokencky et al. (2011)). The Community Land Model (CLM), which is the land component of the Community Earth System Model (CESM), is equipped with a methane module that models methane emissions (Meng et al., 2012; Riley et al., 2011). There are several parameters in CLM related to the methane emission computations. The methane emissions estimated by the model are sensitive to the exact parameter values although these parameters are not well known (e.g. Meng et al. (2012); Riley et al. (2011); Wania et al. (2010)). Riley et al. (2011) and Meng et al. (2012) reported significant differences in model simulations and observations in both site-level methane emissions and the global budget. One important source of uncertainty is associated with the parametrization since the methane module has numerous parameters and they are yet to be identified empirically due to the lack of field data (Riley et al., 2011). In this study our goal is to use surrogate model optimization techniques in order to adjust the methane-related parameters of the CLM such that the differences between the simulated and observed methane emissions at 16 sites around the globe are minimized.

For computing an objective function value, we have to do a computationally expensive simulation with CLM4.5bgc in order to obtain the methane emission predictions at each observation site. **CLM4.5bgc and related codes are deterministic models, i.e., the simulated CH<sub>4</sub> emissions for a given parameter set will always be the same whenever we run the model for the same parameter set.** In an optimization framework where the goal is to find the best set of parameters to minimize the objective function, one obstacle is the computation time that is needed to obtain a single objective function value. Only a few hundred function evaluations can be allowed in order to obtain a solution within reasonable time. Moreover, the objective function value must be computed by running a simulation model, and thus an analytic description of the objective function is not available (black-box). Therefore, gradient information, which is important for many optimization algorithms, is not available. Due to the black-box nature of the objective function, it is also not known whether or not the objective function is convex and has only one local minimum (which corresponds to the global minimum) or if there are several local and global minima in the objective function landscape.

These characteristics of the objective function (computationally expensive, black-box, possibly multi-modal) do not allow the application of a gradient-based optimization algorithm because, on the one hand, the derivatives would have to be computed numerically (which may be inaccurate and requires many expensive function evaluations), and, on the other hand, gradient-based algorithms

generally stop at a local minimum if the initial guess is not close to the global minimum.

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For calibrating the parameters of other CLM modules, Markov Chain Monte Carlo (MCMC) methods and Kalman filters have been used in the literature (Lo et al., 2010; Prihodko et al., 2008; Schuh et al., 2010; Solonen et al., 2012; Sun et al., 2013; Tian et al., 2008; Turner et al., 2009; Zeng et al., 2013). MCMC, however, requires generally thousands of function evaluations (Ray and Swiler, 2014) and is thus not applicable for obtaining solutions in an acceptable time for computationally expensive problems. When using Ensemble Kalman Filters, assumptions about the underlying parameter distributions must be made and generally a large number of observations is necessary for the method to be effective. Furthermore, evolutionary strategies such as simulated annealing, particle swarm, and differential evolution methods have been used for parameter tuning in the climate area (Yang et al., 2012, 2013). However, these methods generally require many function evaluations in order to obtain good solutions.

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Other methods that have recently gained interest for parameter tuning are based on data assimilation (see, for example, Han et al. (2014); Moore et al. (2008)). In order to produce good parameter estimates, these methods require in general many observations. In our optimization problem, however, the number of observations at each site is very low (between 10 and 79 observations distributed over one to three years), and thus data assimilation techniques are not suitable because of the low number of observations. Ray and Swiler (2014) use a computationally cheap surrogate for CLM on which MCMC is used to reduce the number of costly simulations required during the optimization. In contrast to Ray and Swiler (2014), we apply an adaptive surrogate model during the optimization. Instead of relying on a surrogate that is based only on a limited number of initial sample points, we iteratively improve our surrogate by incorporating new data (new objective function values) that become available during the optimization.

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We use surrogate model based global optimization algorithms because they have been shown to find near-optimal solutions within few hundred function evaluations for computationally expensive multimodal black-box problems (Aleman et al., 2009; Giunta et al., 1997; Regis, 2011; Simpson et al., 2001). Surrogate models are used as computationally cheap approximations of the objective function. During the optimization, information from the surrogate model is used to carefully select a new promising point in the variable domain at which the computationally expensive objective function will be evaluated. The surrogate model is updated throughout the optimization whenever new data are obtained.

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Several surrogate model algorithms have been developed in the literature that use different surrogate model types. The efficient global optimization algorithm by Jones et al. (1998), for example,

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uses a kriging surrogate model and selects a new sample point by maximizing an expected improvement function. Gutmann (2001) uses radial basis function (RBF) surrogate models to approximate the expensive objective function and a new sample point is selected by minimizing a so-called bumpiness measure. Regis and Shoemaker (2007, 2013) also use RBF models and new function evaluation points are selected by a stochastic method. Müller and Piché (2011) developed a framework for automatically computing ensembles of various surrogate model types and Müller and Shoemaker (2014) extended the study to investigate the influence of different sampling strategies on the solution quality. Here for the first time, we apply a state-of-the-art RBF surrogate optimization algorithm to the problem of land surface emissions of methane and describe the results. As far as we know, no other groups have applied optimization techniques to find better parameters for methane emission models, and thus our work represents an innovative approach to an important land-atmosphere interaction.

The remainder of this paper is organized as follows. In Section 2 we briefly describe the CLM and the configuration we used for predicting the methane emissions and we give information about the individual observation sites. We also provide the mathematical description of the optimization problem. In Section 3 we summarize the methane-related parameters in CLM4.5bgc and show the results of a sensitivity analysis with which we determined the parameters that are most important for the optimization. We describe the surrogate optimization approach for solving the problem in Section 4. Section 5 contains information about the setup of our numerical experiments and we discuss the results of the optimization. We draw conclusions in Section 6. The appendix contains additional information about the methane equations and the observation sites.

## 2 Model Description, Configuration, and Mathematical Problem Description

### 2.1 Model Description

We used the Community Land Model Version 4.5 (CLM4.5), a land component of the Community Earth System Model (CESM) (Hurrell et al., 2013) which contains a detailed biophysics, hydrology, and biogeochemistry representation (Koven et al., 2013; Oleson et al., 2013). CLM4.5 is fully prognostic with respect to the carbon and nitrogen state variables in the vegetation, litter, and soil organic matter, as well as methane emissions (Koven et al., 2013; Thornton et al., 2007, 2009) and it is the most updated version of the model available.

We selected the latest version of CLM with improved biogeochemistry (CLM4.5bgc) over CLM4.0-CN. The major improvements in CLM4.5bgc include the incorporation of vertically-resolved soil carbon dynamics, an alternate decomposition cascade from the Century soil model, and a more detailed representation of nitrification and denitrification based on the Century nitrogen model (Koven et al., 2013). The hydrology of CLM4.5 has been improved to better represent the hydraulic proper-

ties of frozen soils, perched water tables, snow cover fraction, and lakes (Subin et al., 2012; Swenson and Lawrence, 2012; Swenson et al., 2012).

In previous versions, simulation of ecosystem productivity was too low in high latitudes and perhaps too high in low latitudes (Thornton et al., 2007, 2009). However, CLM4.5bgc has substantially increased the productivity in high latitudes, which may be overpredicted (Koven et al., 2013).

We used a mechanistic methane emission model, which is a module integrated in CLM4.5bgc (Meng et al., 2012; Riley et al., 2011). The model simulates the physical and biogeochemical processes regulating terrestrial methane fluxes such as methane production, methane oxidation, methane and oxygen transport through aerenchyma of wetland plants, ebullition, and methane and oxygen diffusion through soil (Riley et al., 2011). Meng et al. (2012) added constraints on methane emissions such as the effects of redox potential and soil pH to improve the predictions of methane emissions as well as the ability to simulate satellite derived inundation fraction (Prigent et al., 2007; Ringeval et al., 2010).

The model has been compared to the limited site-level observations of methane emissions (many of the sites have very sparse spatial and temporal data coverage, and directly measured climate forcing was unavailable at any of the sites) (Meng et al., 2012; Riley et al., 2011). Additionally, the model was compared with the results from three recent global atmospheric inversion estimates of methane emissions (Riley et al., 2011). In these comparisons, simulated emissions agreed relatively well with the observed emissions at some of the sites. However, there are considerable differences in seasonality and magnitude at other sites. The simulated patterns and magnitudes of annual-average methane emissions are consistent with the results from atmospheric inversion across most latitude bands. The limitations are discussed in Riley et al. (2011).

## 2.2 Model Configuration and Data

Although the land model can be used interactively within CESM, we use it at specific points driven by appropriate meteorology (Oleson et al., 2013). At each site, we forced the model with NCEP/NCAR's reanalysis atmospheric forcing data sets (Qian et al., 2006). These data sets include precipitation, temperature, wind speeds, and solar radiation. We also forced the model with transient atmospheric carbon dioxide concentrations, aerosol deposition data, and nitrogen deposition data that is available in CLM4.5. **Please note that this model is a deterministic model, and thus will give the same answer every time it is simulated when driven by observationally-based datasets as done here.**

In this study we used a total of six natural wetland sites and ten rice paddy sites (see Tables B1 and B2 in Appendix B). We chose the wetland sites from varying geographical regions such as the tropics, mid-latitudes, and high-latitudes to account for the zonal variability. We selected the rice paddy sites such as to cover the major rice-growing regions with a focus on Asia.

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The water table depth is one of the critical factors for methane emissions from natural wetlands because it determines the extent of anoxic and oxic soil zones where methane is produced and oxidized, respectively (Bloom et al., 2010; Grunfeld and Brix, 1999). Methane is produced in the wetlands from litter and dead vegetation remnants in anoxic conditions. The changes in the water table position also influence the moisture conditions of the soil and therefore affect the methane emissions. Here, we prescribed the measured water table position at each wetland site (except Panama) based on previous studies. Since the measured water table depths at Panama were not available, we used modeled water table positions (similar to Walter and Heimann (2000)). For the point simulations, the methane emissions were calculated only from the saturated portion of the soil (i.e. below the water table) when the water table is below the surface. The prescribed water table depth is used in the methane model for calculating anaerobic conditions, production, and oxidation.

Most of these wetland sites usually have peat soils with varying depths underlain by mineral soil. We also forced each wetland site with measured pH and a specific plant functional type (PFT). The PFT reflects the phenological and physiological characteristics of the vegetation (Oleson et al., 2013). Since the wetland PFT was not available in CLM4.5, we choose PFTs that are available in CLM4.5 and that closely match the specific vegetation types of the individual sites. We use  $C_3$  arctic grass for Salmisuo,  $C_3$  non-arctic grass for Alberta, Michigan, and Minnesota, and  $C_4$  grass for Florida and Panama. Other surface data required to perform the point simulation include soil color and soil texture which we extracted from the global grid data sets available in CLM4.5.

For the point simulations at the rice paddy sites we only considered the rice growing season. The flooding and drainage dates are shown in Table C1 in Appendix C. We assumed that the fields were submerged during the simulation period between initial flooding and final drainage. A common feature of these sites during the growing season is that the water was not drained until harvest. We prescribed the  $C_3$  crop PFT for all rice paddy sites, and assumed an optimal pH for the methane production whenever the pH value was not available. The dominant soil types at these sites are loam and clay. Other soil-related information such as soil color and texture are derived from the global grid datasets.

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To bring the terrestrial carbon and nitrogen cycles close to steady-state conditions, we spun up both wetland and rice paddy sites for 1850 conditions (atmospheric CO<sub>2</sub> concentrations, nitrogen

deposition, aerosol deposition, and land use) driven by a repeating 25-year subset (1948-1972) of the meteorological forcing data for more than 2000 years. Then, we performed transient simulations from 1850 to the simulation starting year of each site to generate the initial conditions file.

205 Additionally, we conducted global simulations of methane emissions from natural wetlands for 1993-2004. For these simulations, the grid cell averaged methane emissions were considered which accounts for methane emissions from both the inundated and non-inundated portion of the grid cell. Since the CLM4.5 simulated saturated fraction (an index of inundation) was substantially greater than the estimates from satellite observations and did not match the spatio-temporal pattern of variability (Riley et al., 2011), we prescribed the model with inundation fraction derived from multi-satellite observations for 1993-2004 (Prigent et al., 2007). Similar to point simulations, the global simulations were forced with NCEP/NCAR reanalysis atmospheric forcing data from 1948 to 2004 (Qian et al., 2006). The simulations were also spun up to steady-state conditions driven by atmospheric CO<sub>2</sub>, nitrogen deposition, aerosol deposition, and land use in the year 1850 and a repeated 25-year (1948-1972) subset of the meteorological forcing.

### 2.3 Mathematical Problem Formulation

The goal of our study is to improve the methane emission predictions of CLM4.5bgc by tuning the methane-related parameters such that the model better fits the observations. We use the CH<sub>4</sub> emission observation data for the locations and observation periods shown in Tables B1 and B2. Given the observation data at the  $M = 16$  locations, the goal is to minimize the root mean squared errors (RMSEs) between the CLM4.5bgc methane emission predictions and the observations at each site simultaneously. In order to tackle the problem, we formulate it such that we minimize the weighted sum of the RMSEs as follows:

$$\min f(\mathbf{x}) = \sum_{i=1}^M w_i r_i(\mathbf{x}) \quad (1a)$$

$$\text{s.t. } -\infty < x_k^l \leq x_k \leq x_k^u < \infty, \quad k = 1, \dots, d, \quad (1b)$$

230 where  $d$  denotes the problem dimension (the number of optimization parameters), and  $x_k^l$  and  $x_k^u$  are the lower and upper bounds of variable  $x_k$ , respectively. The RMSE

$$r_i(\mathbf{x}) = \sqrt{\frac{1}{N_i} \sum_{j=1}^{N_i} [O_{i,j} - S_{i,j}(\mathbf{x})]^2}, \quad i = 1, \dots, M, \quad (2)$$

is computed for each location  $i$ .  $N_i$  is the number of observations available at location  $i$ ,  $O_{i,j}$  denotes the  $j$ th methane emission observation at location  $i$ , and  $S_{i,j}$  denotes the corresponding methane emission predicted by CLM4.5bgc. The weights  $w_i$  are computed based on the means of the CH<sub>4</sub>

235 emissions at the observation locations as follows. Denote

$$a_i = \frac{1}{N_i} \sum_{j=1}^{N_i} O_{i,j} \quad (3)$$

the mean CH<sub>4</sub> emission at location  $i$ ,  $i = 1, \dots, M$ . The weight  $w_i$  for the  $i$ th location is then defined by

$$w_i = \frac{g_i}{\sum_{i=1}^M g_i}, \quad (4)$$

240 where

$$g_i = \frac{\max_{i=1, \dots, M} a_i}{a_i}, \quad (5)$$

where it is assumed that  $a_i > 0$  for all  $i$ . The goal is to give each location approximately equal influence in the weighted sum of RMSEs, i.e., we assign locations with large mean CH<sub>4</sub> values small weights such that these locations have approximately the same influence on the weighted sum as locations with low emissions. Otherwise, locations with large emissions would dominate the sum (1a) because their RMSEs would accordingly be larger. In that case the optimization would be driven by minimizing the RMSE of the site(s) with the largest emissions. **There are also other methods of how  $w_i$  could be determined. In the numerical experiments, we will investigate also the possibilities of assigning equal weights to each observation site and assigning weights derived from grouping the observation sites into zones. Another possibility would be to apply clustering algorithms in order to determine groups of observation sites with similar characteristics. For this possibility, however, different clustering methods and different numbers of desired clusters will lead to different groups and different weight adjustments. Lastly, the problem could be formulated as multi-objective optimization problem, for example, with 16 objectives and the goal of minimizing each observation site's RMSE individually, or as bi-objective optimization problem by minimizing the sum of the weighted RMSE values of northern and southern locations at the same time. However, each objective function evaluation is very expensive, and thus the number of evaluations that can be done to obtain the Pareto front in a multi-objective setting is limited. Our focus is on demonstrating that single objective global optimization analysis is useful in identifying reasonable parameter values.**

### 260 3 Methane-Related Parameters in CLM4.5bgc and Sensitivity Analysis

CLM4.5bgc has 21 parameters related to the methane emission predictions. The parameter names, their upper and lower bounds, and default values are shown in Table 1. The upper and lower bounds have been derived based on reported values in the literature (see Table C1 in Appendix C). How these parameters are used in the model is detailed in Riley et al. (2011) and Meng et al. (2012) and we repeat the important equations in Appendix A. The default parameter values  $v_k$  are available in

the CLM4.5bgc (see Table 1).

Optimization problems become increasingly more complex and difficult to solve as the number of parameters increases (curse of dimensionality). Thus, we determine first which of these 21 parameters are the most sensitive and thus the most important for the optimization. By sensitive we refer to parameters that when changed slightly lead to a significant change in emission predictions. Insensitive parameters, on the other hand, can be changed and do not (or comparatively only very mildly) change the emission predictions and can thus be excluded from the optimization, which decreases the problem dimension.

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We conducted analyses for each observation site in which we investigated to which of these 21 parameters the methane emission predictions of CLM4.5bgc are the most sensitive. We altered the value of each parameter  $k = 1, \dots, d$  by, respectively, adding and subtracting 20% of the variable range and we recorded the absolute change in emission predictions, i.e. we ran CLM4.5bgc with perturbed parameter values

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$$(a) \ x_k = \min\{v_k + 0.2(x_k^u - x_k^l), x_k^u\}, \forall k = 1, \dots, d \text{ when increasing } v_k \text{ for 20\%, and}$$

$$(b) \ x_k = \max\{v_k - 0.2(x_k^u - x_k^l), x_k^l\}, \forall k = 1, \dots, d \text{ when decreasing } v_k \text{ for 20\%}$$

for each parameter separately.

There are several parameters that are relatively important to the sensitivity test for all 16 observation sites, but there are also parameters that are important for some locations and less important for others. Tables 2 and 3 show the sensitive and insensitive parameters together with the number of locations (out of 16) for which these parameters are important and unimportant, respectively. Thus, in the optimization we consider only the parameters in Table 2 since these parameters are the most important at most locations. Please note that, due to (nonlinear) relationships between the parameters, for many parameters the effects of individual parameters will be opposite but act in a similar manner, indicating that some parameters may be difficult to optimize for. **In order to limit the number of parameters we consider, while allowing for the largest range in behavior, we combine information from the sensitivity study with information about the methane flux equations themselves (described in more detail in Appendix A). The most important parameters from the sensitivity study come from the dominant three terms in the methane flux equation, which are production (parameters 1, 2, and 21), oxidation (parameters 7, 8, 9, and 10), and aerenchyma transport (parameters 13, 15, 16, and 17). The first four parameters chosen are also the most important parameters at all 16 sites (see Table 2). Because production is the most important term, there are two parameters from production that the sensitivity studies indicate are the most important, namely one that controls globally the methane production flux ( $F_{CH4}$ , parameter 2), and one term that controls the temperature**

300

dependency of the methane production (Q10CH4, parameter 1). Another parameter that influences methane at all the sites comes from the oxidation equation (VMAX\_CH4\_OXID, parameter 7), and the final parameter that is important at all 16 sites is the parameter controlling the aerenchyma transport (SCALE\_FACTOR\_AERE, parameter 13). The above four parameters are the most sensitive parameters, and thus are easy to choose, as well as cover most of the important processes we want to investigate. For the last parameter, we include one parameter that controls how inundation affects methane production (MINO2LIM, parameter 21). Inundation is an important process for controlling methane flux, since there is an order of magnitude more methane coming from wet areas than dry, and thus having one parameter which changes the model’s sensitivity to inundation is appropriate.

## 4 Surrogate Models and Surrogate Model Algorithms

### 4.1 Surrogate Models

Surrogate models are used in optimization algorithms that aim to solve computationally expensive black-box problems. Surrogate models serve as computationally cheap approximations of the expensive simulation model (Booker et al., 1999), i.e.,  $f(\mathbf{x}) = s(\mathbf{x}) + e(\mathbf{x})$ , where  $f(\cdot)$  denotes the true expensive objective function,  $s(\cdot)$  denotes the computationally inexpensive surrogate model, and  $e(\cdot)$  denotes the difference between both. Surrogate models are used throughout the optimization to guide the search for promising solutions. The computationally expensive objective function is evaluated only at few selected points, and thus it is possible to find near-optimal solutions with only very few expensive function evaluations.

There are different surrogate model types such as radial basis functions (RBFs) (Gutmann, 2001; Müller et al., 2013; Powell, 1992; Regis and Shoemaker, 2007, 2009; Wild and Shoemaker, 2013), kriging (Davis and Ierapetritou, 2009; Forrester et al., 2008; Jones et al., 1998; Simpson et al., 2001), polynomial regression models (Myers and Montgomery, 1995), and multivariate adaptive regression splines (Friedman, 1991). There are also mixture models (also known as ensemble models) that exploit information from several different surrogate model types (Goel et al., 2007; Müller and Piché, 2011; Müller and Shoemaker, 2014; Viana et al., 2009). In general any type of surrogate model may be used in a surrogate model optimization algorithm. In this study, we use RBFs because they have been shown to perform better in comparison to other surrogate model types (Müller and Shoemaker, 2014).

An RBF interpolant is defined as follows:

$$s(\mathbf{x}) = \sum_{\iota=1}^n \lambda_{\iota} \phi(\|\mathbf{x} - \mathbf{x}_{\iota}\|) + p(\mathbf{x}), \quad (6)$$

335 where  $\phi(\tau) = \tau^3$  denotes the cubic radial basis function whose corresponding polynomial tail is linear ( $p(\mathbf{x}) = b_0 + \mathbf{b}^T \mathbf{x}$ ), and  $\mathbf{x}_\ell, \ell = 1, \dots, n$ , denotes the points at which the objective function has already been evaluated. The parameters  $\lambda_\ell \in \mathbb{R}, \ell = 1, \dots, n$ , and the parameters  $b_0 \in \mathbb{R}$  and  $\mathbf{b} = [b_1, \dots, b_d] \in \mathbb{R}^d$  are determined by solving the following linear system of equations

$$\begin{bmatrix} \Phi & \mathbf{P} \\ \mathbf{P}^T & \mathbf{0} \end{bmatrix} \begin{bmatrix} \boldsymbol{\lambda} \\ \mathbf{c} \end{bmatrix} = \begin{bmatrix} \mathbf{F} \\ \mathbf{0} \end{bmatrix}, \quad (7)$$

340 where  $\Phi_{\ell\nu} = \phi(\|\mathbf{x}_\ell - \mathbf{x}_\nu\|), \ell, \nu = 1, \dots, n$ ,  $\mathbf{0}$  is a matrix with all entries 0 of appropriate dimension, and

$$\mathbf{P} = \begin{bmatrix} \mathbf{x}_1^T & 1 \\ \mathbf{x}_2^T & 1 \\ \vdots & \vdots \\ \mathbf{x}_n^T & 1 \end{bmatrix}, \quad \boldsymbol{\lambda} = \begin{bmatrix} \lambda_1 \\ \lambda_2 \\ \vdots \\ \lambda_n \end{bmatrix}, \quad \mathbf{c} = \begin{bmatrix} b_1 \\ b_2 \\ \vdots \\ b_d \\ b_0 \end{bmatrix}, \quad \mathbf{F} = \begin{bmatrix} f(\mathbf{x}_1) \\ f(\mathbf{x}_2) \\ \vdots \\ f(\mathbf{x}_n) \end{bmatrix}. \quad (8)$$

The matrix in (7) is invertible if and only if  $\text{rank}(\mathbf{P}) = d + 1$  (Powell, 1992).

## 4.2 Surrogate Global Optimization Algorithm

345 Surrogate global optimization algorithms follow in general the steps shown in Algorithm 1.

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### Algorithm 1 General Surrogate Global Optimization Algorithm

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- 1: Select points from the variable domain to create an initial experimental design.
  - 2: Do the expensive objective function evaluations (here the CLM4.5bgc simulations) at the points selected in Step 1.
  - 3: Fit the surrogate model (here the RBF model) to the data from Steps 1 and 2.
  - 4: Use the information from the surrogate model to select the new evaluation point  $\mathbf{x}_{\text{new}}$ .
  - 5: Do the expensive evaluation at  $\mathbf{x}_{\text{new}}: f_{\text{new}} = f(\mathbf{x}_{\text{new}})$  (here, run CLM 4.5bgc for the parameter input vector  $\mathbf{x}_{\text{new}}$ ).
  - 6: **if** Stopping criterion is not met (the maximum number of allowed function evaluations has not been reached)  
**then**
  - 7:   Update the surrogate model and go to Step 4.
  - 8: **else**
  - 9:   Return the best solution found during the optimization.
  - 10: **end if**
- 

We use the DYCORS algorithm by Regis and Shoemaker (2013) for the optimization of the methane-related parameters of CLM4.5bgc. The reader is referred to this publication for the details of the algorithm. Since the parameters have significantly differing ranges (see Table 1), we scale all parameters to the interval  $[0, 1]$  when selecting new sample sites. When doing the computationally



350 expensive CLM4.5bgc simulations, we scale the parameters back to their original ranges. Thus, the  
perturbation radius used in DYCORS is the same for each variable.

We create a symmetric Latin hypercube initial experimental design with  $2(d + 1)$  points and run  
CLM4.5bgc at the selected parameter vectors in order to compute the objective function values. We  
355 then fit the cubic RBF model to the data and generate two sets of candidate points for the next expen-  
sive function evaluation (the next CLM4.5bgc run at the 16 sites). The first set of candidate points is  
generated as described by Regis and Shoemaker (2013) by randomly perturbing the best point found  
so far. The second set of candidate points is generated by uniformly selecting random points from  
the whole variable domain. Thus, we create twice as many candidate points as DYCORS. The goal  
360 of using uniformly random points from the whole variable domain is to obtain candidates that are  
far away from the best point found so far, and hence if selected as new evaluation point, the search  
is more global (exploration by function evaluation at points that are far away from already sampled  
points).

365 We use the same criteria as in DYCORS for determining the best candidate point (using the RBF  
approximation to predict the objective function values at the candidate points, compute the distance  
of the candidate points to the set of already sampled points, and compute a weighted score of these  
two measures where the weights cycle through a predefined pattern). **In order to guarantee that the  
matrix in equation (7) is well-conditioned, we ensure (as done in Regis and Shoemaker (2013)) that  
370 the sample points are sufficiently far away from previously evaluated points by discarding candi-  
date points that are closer than a given threshold distance to previously evaluated points.** We run  
CLM4.5bgc at each of the 16 observation sites using the **one** newly selected sample point as input  
parameter vector to obtain the corresponding objective function value. We update the RBF model  
with the new data and iterate until we have reached the maximum number of allowed function eval-  
375 uations.

## 5 Numerical Experiments

In this section we discuss the setup and results of the numerical experiments. In a first set of ex-  
periments (pseudo data case), we generate synthetic (pseudo) data and treat it as if it were the real  
measurement data in order to assess how well our optimization approach performs. For these ex-  
380 periments we know the optimal solution. In the second set of experiments (real data case), we use  
the measured methane emission data and apply the optimization algorithm. The goal in the second  
set of experiments is to find a parameter set that reduces the objective function value (the weighted  
RMSE in equation (1a)) from its default value (the RMSE when using CLM4.5bgc’s default param-  
eter settings, see also Table 1, column  $v_k$ ). Finally, we run CLM4.5bgc globally with the best set of

385 parameters found during the optimization of the real data case and investigate how much the default  
model predictions and the model predictions with the optimized parameter values differ from each  
other.

We did experiments with  $d = 5$  and  $d = 11$  parameters respectively. For the  $d = 5$  experiments,  
390 we used parameters 1, 2, 7, 13, and 21 (Table 2). Thus, we have parameters related to three types  
of  $\text{CH}_4$  emission, namely oxidation (parameter 7), aerenchyma (parameter 13), and production (pa-  
rameters 1, 2, 21). For the 11-parameter optimization, we used all variables shown in Table 2.

For each set of experiments we ran the optimization algorithm three times in order to examine the  
395 influence of the random component in the algorithm (random initial experimental design and ran-  
dom generation of candidate points). We allowed 800 function evaluations for the five-dimensional  
problem and 1000 evaluations for the 11-dimensional problem. **The question of how many function  
evaluations need to be performed in order to obtain a fixed level of solution accuracy is problem  
dependent. For computationally-expensive optimization problems, such as the problem we consider  
400 here, the time for evaluating the objective function and the totally available time for obtaining a  
solution usually defines how many evaluations can be done with any algorithm. Results for many  
difficult computationally-expensive optimization problems (for example, problems with multiple lo-  
cal minima) indicate that surrogate global optimization methods can usually obtain more accurate  
results compared to non-surrogate methods with the same limited number of evaluations (see, for  
405 example, Mugunthan et al. (2005)). It is a very difficult problem to find the best values of the param-  
eters for climate models, and the more evaluations one does, in general the better the answer.**

The weights  $w_i$  in equation (1a) were for the pseudo data case computed based on the pseudo  
observations (see Section 5.1) at each of the 16 sites at the same dates for which we also have real  
410 measurements. For the real data case, the weights were computed based on the actual measurements.  
The weights are given in Table D1 in Appendix D.

Solving problem (1a) requires running CLM4.5bgc for each input vector  $\mathbf{x}$  of parameter values  
and for each of the 16 observation sites. We run CLM4.5bgc on the Yellowstone Supercomputing  
415 Facility (Computational and Information Systems Laboratory, 2012). Each simulation at a single  
location takes between 15 and 30 minutes. We do the simulations for the 16 sites in parallel in order  
to speed up the objective function evaluation time.

## 5.1 Pseudo Data Case

We assessed the performance of the optimization algorithm by investigating how well the algorithm  
420 could find the model parameters that were used for creating the pseudo data. For this purpose, we

ran CLM4.5bgc with default parameter values  $v_k, k = 1, \dots, d$ , at all 16 sites for the same time span for which we also have observation data (see Tables B1 and B2 in Appendix B) and we record the model's predictions for the same dates at which the methane emissions were measured. We use this as our pseudo observation data that we want to match in the optimization, i.e., the goal of the optimization is to start from a set of parameter vectors that is different from the default parameter values and to recover the default parameter values by optimization. For the default parameter values, the objective function value will be zero, which is the global minimum of the pseudo data case.

### 5.1.1 Results for $d = 5$

Figure 1 shows the progress plots of the three optimization trials T1, T2, and T3. Illustrated is the development of the best objective function value found within the given number of function evaluations (horizontal axis). The fewer evaluations needed for reducing the objective function value, the better. The plot shows that the objective function value is reduced significantly in each of the three trials from a value of over 30 to about 5 within less than 150 function evaluations and close to zero towards the end of the optimization. Table 4 shows the best parameter values found during each of the three optimization trials together with the default parameter values. The table shows that the RMSE after 800 function evaluations is not exactly zero (which can be expected from an approximation method), but the default parameter values are matched closely.

### 5.1.2 Results for $d = 11$

Figure 2 shows the objective function value development as the number of function evaluations increases for the 11-dimensional case for the three trials T1, T2, and T3. The figure shows a rapid decrease of the objective function value from over 50 to less than 10 within 100 evaluations, which shows that the surrogate model algorithm is very efficient at finding improved solutions. Although the objective function value improvement over the following function evaluations is lower, we can see that the algorithm still makes progress and if we allowed more than 1000 evaluations, the objective function value would be further improved (which also follows from the global convergence property of the DYCORS algorithm).

Table 5 shows the parameter values of the best of the three trials (T3) together with the default parameter values and the variable vector CP that was evaluated during the optimization and that has a *worse* objective function value than the best solution, but that is *closer* to the default parameter values. This point has the same parameter values as T3 for all but two parameters, namely, parameters 10 (Q10\_CH4OXID) and 21 (MINO2LIM), which we indicate by bold numbers. For these two parameters, the point CP is closer to the global optimum, but it has a worse objective function value. This indicates a multimodality of the objective function (getting closer to the true global minimum

requires an increase in the objective function value, i.e., the algorithm has to escape from a local basin of attraction). This multimodality makes the search for the global optimum significantly more difficult.

460 In order to examine the impact of the differences between default and optimized parameter values on the model prediction, we use the best parameter vector of each trial and plot the corresponding CH<sub>4</sub> emission predictions against the predictions when using the default parameter values in Figure 3. We can see that although we do not exactly match the default parameter values, the model's predictions when using the optimized parameters are very close to the predictions when using the  
465 default parameter values (all points in the scatter plot lie close to or on the dashed line which represents agreement of default and optimized predictions). As also reflected in the best RMSE value reported in the legend, T3 matches the default data best and T2 has the largest differences.

This result indicates that the calibration problem is not "identifiable" for all parameter sets, indicating that more than one parameter set can give a very similar result in terms of the objective  
470 function value. For example, for the model  $y = \frac{\alpha}{\beta}x + \gamma$ , there are many combinations of values for  $\alpha$  and  $\beta$  that lead to the same value of  $y$  as long as  $\alpha = \kappa\beta$  for some constant  $\kappa$ . With only five parameters as described in the previous section, the parameter values obtained from the optimization did match very closely those of the default case used to create the pseudo data, and thus with this  
475 small set of parameters the problem was identifiable. However, for 11 parameters, we did encounter the identifiability problem. In some disciplines such parameters are called "hidden". For example, estimating  $\alpha$  and  $\gamma$  in the previous example with  $y = \frac{\alpha}{\beta}x + \gamma$  when  $\beta$  is given would be identifiable. However, estimating  $\alpha$ ,  $\beta$ , and  $\gamma$  is no longer identifiable.

480 It would be desirable to have an identifiable model, but the CLM (and probably other climate modules) have a number of interacting parameters and multiplicative nonlinearities, and thus there is no guarantee that all parameters are identifiable. This is reinforced by the data in Table 5, which indicates that the surface over which the optimization algorithm searches in the 11 parameter case is multi-modal, i.e., there are multiple local minima and it is possible for two (or more) parameter  
485 sets to yield the same objective function value (here RMSE). Hence the inability of the optimization to find the exact set of parameters that was used for generating the pseudo data is a problem caused by the complexity and multiplicative nonlinearities of the CLM model, not by the choice of the optimization method. However, the optimization analysis for both pseudo data cases (with 5 and 11 parameters, respectively) shows that the chosen optimization method is able to find a set of  
490 parameter values that has a low prediction error. The multi-modality in Table 5 does indicate the need for a global (not a local) optimization method.

## 5.2 Real Data Case

In the real data case, we use the actual methane emission measurements at each of the 16 observation sites for computing the objective function value. Since we only have very few observations for each site and no information about measurement errors, we did not exclude any of the measurements from the optimization although there might be outliers. Also for the real data case we examine the case for  $d = 5$  and  $d = 11$  variables.

### 5.2.1 Results for $d = 5$

The progress of the development of the objective function value for the three trials T1, T2, and T3, respectively, is illustrated in Figure 4 which also shows in the legend the lowest RMSE value found in each of the three trials. The RMSE was efficiently reduced from over 155 to below 115 within the first 150 function evaluations. Thereafter the objective function value improvement was at a significantly lower rate. All three trials return a solution with approximately the same objective function value.

The parameter values of the best solutions found in the three trials are shown in Table 6 where also the default parameter values are given for comparison. We can see that the three optimized solutions are approximately the same and significantly different from the default case. We can also see that three of the five optimized parameter values are on or very close to the boundary of the variable domain (shown in bold), indicating that improvements of the objective function value may be possible by increasing the parameter range. However, it is not possible due to physical constraints and at this point, we do not have information about possible wider parameter ranges than the ones we used in this study.

Figures 5 and 6 show the CH<sub>4</sub> emission predictions of CLM4.5bgc when using the default and the optimized parameter values for two selected observation sites (one wetland and one rice paddy site) together with the actual observation data. The legends show the associated RMSE value before applying the weights for computing (1a). We can see that the optimized solution actually worsens the predictions for Alberta (the RMSE value with default parameters is about 209 and with optimized parameters, the value is about 221, which is about 6% worse). For Central Java, on the other hand, the RMSE values of the optimized solutions are significantly better than for the default values (the default RMSE is about 221 and the optimized RMSE values are about 48, which is an improvement of over 350%). In both figures we can also see that despite the large differences between optimized and default parameter values, the trend in the predictions of CLM4.5bgc is the same, i.e., when the predicted CH<sub>4</sub> emissions with default parameters increase so do the predicted emissions when using the optimized parameters and vice versa.

## 5.2.2 Results for $d = 11$

Figure 7 shows the progress plots for each of the three trials together with the best objective function values found (legend) for the 11-dimensional case. The best objective function value found is about  
530 equal for each of the three trials. The figure shows that in each trial the algorithm is able to efficiently reduce the objective function value within the first 200 function evaluations. The improvement after 200 function evaluations is significantly slower.

Table 7 shows the parameter values of the best solution found in each of the three trials and the  
535 default parameter values. The table shows that for some parameters, for example, parameters 1, 7, and 8, all trials lead to approximately the same values (which are different from the default parameter values). For the remaining parameters, the values corresponding to the best solution found differ significantly for each trial and differ also from the default parameter values. Also for the 11-dimensional problem, some parameter values corresponding to the best solution found are on the  
540 upper or lower boundary of the parameter range (for example, parameters 1, 8, 13, 15, indicated in bold).

Since all three solutions have approximately the same objective function values, but the points differ greatly, it is an indicator that we either have a multi-modal surface in which some minima  
545 assume approximately the same objective function values, or we have a very flat valley in which many points assume similar objective function values. Both possibilities make it very difficult for gradient-based optimization algorithms to find the global optimum. In the first case, the optimization algorithm will get trapped in a local optimum if it is not started close to the global minimum. In the second case, the gradient-based algorithm would require many function evaluations because many  
550 steps and gradient computations are necessary due to a very small step size. The surrogate optimization algorithm overcomes this problem.

Table 8 shows the unweighted RMSE values (before applying the weights in (1a) for computing the objective function value) between observations and simulations using the default parameters  
555 (column 5), the best parameters of optimization trial T1 of the 11-dimensional case (column 4), and the best parameters of trial T2 of the 5-dimensional case, respectively. The table shows that with our optimization we were able to decrease the default RMSE for four sites in the 5-dimensional case and for six sites in the 11-dimensional case. The RMSE is lower at seven sites for the 11-dimensional case than for the 5-dimensional case. Since we minimized a weighted sum of all RMSE values, it  
560 can be expected that the RMSE at some locations may be worse for the optimized case than for the default case. We can see that for two of the improved sites (Java and Cuttack), the improvement is very large, and thus the overall RMSE of the optimized solution is lower than for the default param-

eters.

565 Figures 8 and 9 show the observed CH<sub>4</sub> emissions, the predictions with the default parameter values, and the predictions using the optimized parameter values for Alberta (Canada) and Central Java (Indonesia). For both sites we can see that the predictions with the optimized parameters have lower RMSEs than when using the default parameter values (note that the reported RMSEs in the legend are not weighted as done in equation (1a)). For Central Java, for example, the optimized parameters  
570 greatly improved the model's predictions, but we can also see that the temporal variability in the predictions stays the same although not as pronounced. We noticed this "temporal variability preserving" behavior for several sites such as Beijing, California, Cuttack, New Delhi, Florida, Japan, Michigan, Minnesota, Salmisuo, Texas, and Vercelli. Compared to the case where we optimized only five parameters, the solution for Alberta has improved and the RMSE values for all three trials are  
575 for the  $d = 11$  case better than the default RMSE value. On the other hand, the solution for Central Java is worse for T1 in the  $d = 11$  case than in the  $d = 5$  case.

The temporal variability in the model's predictions does not necessarily follow the temporal variability in the observation data (see, for example, Figure 10). Note that in Figure 10 the temporal  
580 variability is the same for each of the three trials although the best solutions found in the three trials were very different (see Table 7). Thus, it seems that the improvement of the model's predictions is restricted by an underlying model component that enforces the temporal variability. This is likely to be associated with structural errors either in the methane or in the carbon model. Notice that the methane emission is dependent on the temporal variability predicted in the carbon and land model,  
585 especially on the heterotrophic respiration rate, which could have the wrong magnitude or temporal evolution.

Figure 11 shows a scatter plot of the mean values of the CH<sub>4</sub> predictions using default and optimized parameter values versus the mean values of the observed CH<sub>4</sub> emissions. Ideally, if the  
590 simulated emissions agreed with the observations, all points would lie on the dashed line. Thus, the closer a point to the dashed line, the more simulation and observation are in agreement. The figure shows that with the optimized parameters, we obtain better or similar results for Beijing, Cuttack, Minnesota, Central Java, Nanjing, Japan, Salmisuo, Alberta, and Michigan. Although not all sites have been strictly improved by the optimization, the overall RMSE has been improved (indicated in  
595 the legend).

Figure 11 also shows that with default parameters, CLM4.5bgc predicts less CH<sub>4</sub> emissions than observed for both observation sites in the northern latitudes (Alberta (ID=1) and Salmisuo (ID=16)), which is corrected by the optimization such that the mean emissions at these sites are closer to

600 the dashed line. Thus, based on the observation data, CLM4.5bgc with default parameters does not predict enough emissions in the northern latitudes. On the other hand, CLM4.5bgc over-predicts the emissions for four locations, namely Cuttack (ID=14), Central Java (ID=12), Nanjing (ID=5), and Japan (ID=8), which are located in the tropical/subtropical zone. For those four locations, the predictions with the optimized parameters are closer in agreement with the observations. Hence, the  
605 observation data force the model predictions to increase in the northern latitudes and to decrease in the tropics. This can also be seen in Figures 12 and 13 in the following section where we simulated the model globally and compared default and optimized model predictions for the individual zones (discussed below).

### 5.2.3 Global CH<sub>4</sub> Emission Simulations

610 We simulated CLM4.5bgc to obtain predictions for the CH<sub>4</sub> emissions on a global scale and compared the predictions when using the default parameter values and the optimized parameter values from the 11-dimensional cases. Figure 12 shows spatial plots of the average methane emissions (mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) and the zonal means (right hand side of the plots) when using the default parameters (panel a), and the difference between the predictions when using the default and the optimized  
615 parameters for trial T1 (panel b). The figure shows that with the optimized parameters, the CH<sub>4</sub> emission predictions in the northern regions are larger than for the default parameters. For the tropics, the predictions with the optimized parameters are lower than when using the default values.

Figure 13 shows a comparison of the CH<sub>4</sub> emission predictions from several different models  
620 (models 1-10). We can see that globally the predictions with the optimized parameters (model 12) were only slightly higher than with the default parameters (model 11). However, the predictions of CH<sub>4</sub> emissions in the tropics are significantly lower than for the default model and the predictions are also lower in comparison to all other models (1-10). On the other hand, for the northern latitudes, CLM4.5bgc with optimized parameters predicts significantly more CH<sub>4</sub> emissions than the default  
625 model and models 1-10 in the comparison. Hence, even though the global average of predicted emissions did not change much, the distribution of the predicted emissions between the tropical and the northern latitudes changed significantly.

As indicated in the previous section, the observation data drives the model to predict more CH<sub>4</sub>  
630 emissions in northern latitudes and fewer emissions in the tropics. We investigated whether our weighting scheme in equation (1a) may give too much influence to individual observation sites or zones. Thus, we did an additional optimization trial of the parameters in Table 2 where we give each observation site the same weight  $w_i = 1$ ,  $i = 1, \dots, 16$  (“unweighted”). We also did a second additional optimization trial of the parameters in Table 2 where we give each zone the same influence  
635 on the total RMSE in order to account for the location of the various observation sites (“zonally



weighted”). Thus, each location in the temperate zone (12 sites totally) has  $w_i = 1/36$ , and each location in the northern (2 sites) and tropical (2 sites) zone, respectively, has the weight  $w_i = 1/6$ .

The spatial plots of the differences between the average methane emissions when using default  
640 and optimized parameters for the unweighted trial are shown in panel (c) of Figure 12, and the spa-  
tial plots of the differences when using the zonally weighted objective function is shown in panel  
(d) of Figure 12. The figures show that for both additional trials, the CH<sub>4</sub> emissions in the northern  
latitudes is even further increased. Moreover, the bars for models 13 and 14 in Figure 13 show the  
total methane emissions of the unweighted and the zonally weighted trials, respectively. The zonally  
645 weighted trial increases the global emissions, which is caused by larger emission predictions in the  
temperate zone and the northern latitudes. In comparison to the default CLM4.5bgc predictions, the  
unweighted trial shows a decrease in the predicted emissions in the tropics and an increase in the  
predicted emissions in the northern latitudes. Thus, even though it is suggested that CLM4.5bgc with  
default parameter settings over-predicts the CH<sub>4</sub> emissions in high latitudes (Koven et al., 2013), the  
650 observation data argues that the predictions should even be increased.

## 6 Conclusions

In this paper we used a surrogate optimization approach for calibrating the parameters of the methane  
module of the Community Land Model (CLM4.5bgc). Given only relatively few measurements at  
655 16 observation sites (wetlands and rice paddies) our goal was to explore the use of a surrogate op-  
timization method to improve the model prediction capability in a computationally efficient way  
by minimizing the root mean squared error between the measurements and the model’s predictions.  
We identified important methane-related parameters in CLM4.5bgc by doing a sensitivity analysis  
and we were thus able to reduce the problem dimension from 21 to 11. We then used a surrogate  
660 optimization approach for tuning the most important parameters in order to solve the problem. We  
investigated two cases, namely a problem with 5 of the most important parameters and a problem  
with all 11 parameters, respectively.

We first used pseudo data in order to asses how well the surrogate optimization performs and  
665 showed that we are able to closely match the pseudo observations. We were able to reduce the RMSE  
to less than a fifth within the first 150 function evaluations for both pseudo data cases. The objective  
function was shown to have multiple local minima, which indicates that the problem is probably  
not identifiable when 11 parameters were optimized. Although the RMSE was greatly reduced by  
the optimization for the 11 parameter pseudo data case, the optimization results did not generate the  
670 same values of the parameters in some cases as were used to generate the pseudo data. This is a

problem with the model, not with the optimization method used. The multiple local minima detected in Table 5 indicate that a global optimization method was needed. We used a surrogate global optimization method because the objective function was expensive to evaluate and has multiple local minima. The surrogate has been shown to reduce the number of objective function evaluations (e.g. 675 climate model simulations) required to obtain accurate approximations of the global minimum and so it is designed for computationally expensive models like climate modules.

By conducting the simulations globally and comparing the average predicted emissions with default and optimized parameters, we could show that the total global CH<sub>4</sub> emissions did not change significantly. However, the distribution of the predicted emissions between latitudes changed significantly. 680 The observation data force the optimized model's CH<sub>4</sub> emission predictions in the northern latitudes to increase and the predicted emissions in the tropics to decrease. In comparison to other models, CLM4.5bgc with both default and optimized parameters predicts significantly more emissions in the northern latitudes and less emissions in the tropics.

## Appendix A: Model Equations

685 The methane biogeochemical model used in this study is integrated in the Community Land Model  
 version 4.5 (CLM4.5), which is the land component of the Community Earth System Model (CESM,  
 Hurrell et al. (2013)). As discussed in more detail in Riley et al. (2011) and Meng et al. (2012), the  
 model represents five primary processes relevant to methane emission predictions. These processes  
 690 include methane production ( $P$ ), oxidation ( $R_{\text{oxic}}$ ), ebullition ( $E$ ), transport through wetland plant  
 aerenchyma ( $A$ ), and diffusion through soil ( $FD_e$ ) (described below). The methane gas and aqueous  
 phase concentrations ( $R_C$ ) in each soil layer of each grid box is calculated at every time point using  
 the following equation:

$$\frac{\partial R_C}{\partial t} = \frac{\partial FD_e}{\partial z} + P - E + A - R_{\text{oxic}} \quad (\text{A1})$$

In the following sections we consider each of these terms in more detail.

### 695 A1 Methane Production

Methane production ( $P$ ) in the anaerobic portion of the soil column is related to the grid cell estimate  
 of heterotrophic respiration from soil and litter corrected for various factors:

$$P = R_H \cdot F_{\text{CH}_4} \cdot Q_{10\text{CH}_4} \cdot f_{\text{pH}} f_{\text{pE}} S, \quad (\text{A2})$$

700 where  $R_H$  is the heterotrophic respiration from soil and litter ( $\text{mol C m}^{-2} \text{ s}^{-1}$ ), and  $F_{\text{CH}_4}$  is  
 the baseline fraction of anaerobically mineralized C atoms becoming  $\text{CH}_4$  (i.e.,  $\text{CO}_2/\text{CH}_4$ ).  $R_H$  is  
 corrected for its soil temperature dependence through a Q10 factor ( $Q_{10\text{CH}_4}$ ), pH ( $f_{\text{pH}}$ ), redox po-  
 tential ( $f_{\text{pE}}$ ), and a factor accounting for the seasonal inundation fraction ( $S$ ).

We adjusted the fractional inundation in each grid cell to account for a changing redox potential.

$$705 f_{\text{pE}} = \frac{f_{i_{\text{lag}}}(t)}{f_i(t)}, \quad (\text{A3})$$

where the redox potential factor  $f_{\text{pE}}$  is computed based on the fractional inundation  $f_i(t)$  and the  
 adjusted fractional inundation  $f_{i_{\text{lag}}}(t)$  that is producing methane.

The adjusted fractional inundation  $f_{i_{\text{lag}}}(t)$  is computed as

$$710 f_{i_{\text{lag}}}(t) = f_i(t) - f_{\text{redox}}(t), \quad (\text{A4})$$

where

$$f_{\text{redox}}(t) = f_i(t) - f_i(t-1) + f_{\text{redox}}(t-1) \left( 1 - \frac{\Delta t}{\text{REDOXLAG}} \right) \quad (\text{A5})$$

is the fraction of the grid cell where alternative electron acceptors (such as  $O_2$ ,  $NO_3^-$ ,  $Fe^{+3}$ ,  $SO_4^{2-}$  etc.) are consumed (methane production is completely inhibited),  $\Delta t$  is the time step, and REDOXLAG is the time constant parameter.

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

$$Z_{i_{lag}}(t) = Z_i(t) - Z_{redox}(t), \quad (A6)$$

where

$$Z_{redox}(t) = Z_i(t) - Z_i(t-1) + Z_{redox}(t-1) \left( 1 - \frac{\Delta t}{REDOXLAG} \right) \quad (A7)$$

is the depth of the saturated water layer where alternative electron acceptors are consumed at time  $t$  and  $Z_i(t)$  is the actual water depth at time  $t$ .

725

Additionally, we constrained the methane production using the soil pH function  $f_{pH}$  which is represented as

$$f_{pH} = 10^{-0.2335pH^2 + 2.7727pH - 8.6}, \quad (A8)$$

where pH represents the soil pH.  $f_{pH}$  is bounded by two parameters, namely PHMIN and PHMAX (i.e., PHMIN < pH < PHMAX). The maximum methane production occurs at pH $\approx$ 6.2.

We used a scaling factor ( $S$ ) to mimic the impacts of seasonal inundation on methane production which is represented as

$$S = \frac{MINO2LIM(f - \bar{f}) + \bar{f}}{f}, \quad S \leq 1, \quad (A9)$$

735 where  $f$  and  $\bar{f}$  are the instantaneous inundation fraction and annual average inundation fraction weighted by heterotrophic respiration, MINO2LIM is the anoxia factor that relates the fully anoxic decomposition rate to the fully oxygen-unlimited decomposition rate.

## A2 Methane Oxidation

Methane oxidation ( $R_{oxic}$ ) is represented with double Michaelis-Menten kinetics:

$$R_{oxic} = VMAX\_CH4\_OXID \left[ \frac{C_{CH_4}}{K\_M + C_{CH_4}} \right] \left[ \frac{C_{O_2}}{K\_M\_O2 + C_{O_2}} \right] Q10\_CH4OXID \cdot F_{\theta}, \quad (A10)$$

where VMAX\\_CH4\\_OXID is the maximum oxidation rate ( $mol\ m^{-3}\ s^{-1}$ ), Q10\\_CH4OXID is the temperature dependence of the reaction, K\\_M and K\\_M\\_O2 are the half saturation coefficients with

respect to  $\text{CH}_4$  and  $\text{O}_2$  concentrations ( $\text{mol m}^{-3}$ ),  $C_{\text{CH}_4}$  and  $C_{\text{O}_2}$  are the methane and oxygen concentrations in the soil ( $\text{mol m}^{-3}$ ), and  $F_{\vartheta}$  is the soil moisture limitation factor for oxidation applied  
745 above the water table to represent water stress for methanotrophs.

$F_{\vartheta}$  is represented as:

$$F_{\vartheta} = \exp \left\{ \frac{-P}{P_C} \right\}, \quad (\text{A11})$$

where  $P$  and  $P_C$  are the soil moisture potential and optimum water potential ( $-2.4 \times 10^5$  mm).  
750 If the soil layer is above the water table, the soil moisture limitation factor  $F_{\vartheta}$  is applied. To account for high- $\text{CH}_4$ -affinity methanotrophs in upland soils, we used a lower oxidation rate constant (VMAX\_OXID\_UNSAT) and half saturation coefficient with respect to  $\text{CH}_4$  concentrations (K\_M\_UNSAT).

### A3 Methane Transport Through Plant Aerenchyma

755 The diffusive transport through aerenchyma  $A$  ( $\text{mol m}^{-2} \text{s}^{-1}$ ) from each soil layer is represented in the model as:

$$A = \frac{C(z) - C_a}{r_a + \frac{\text{ROB} \cdot z}{DpT\rho_f}}, \quad (\text{A12})$$

where  $D$  is the free-air gas diffusion coefficient ( $\text{m}^2 \text{s}^{-1}$ ),  $C(z)$  and  $C_a$  are the gaseous concentrations at depth  $z$  and at the atmosphere ( $\text{mol m}^{-3}$ ),  $r_a$  is the aerodynamic resistance between the  
760 surface and the atmospheric reference height ( $\text{s m}^{-1}$ ), ROB is the ratio of root length to vertical depth (obliquity),  $p$  is the porosity,  $T$  is the specific aerenchyma area ( $\text{m}^2 \text{m}^{-2}$ ), and  $\rho_f$  is the root density as a function of depth. Oxygen concentrations can also diffuse into the soil layer from the atmosphere via the reverse of the  $\text{CH}_4$  pathway.

765 Here, aerenchyma porosity is parameterized based on the plant functional types (PFTs). A ratio is used to multiply upland vegetation aerenchyma porosity by comparing to inundated systems:

$$p = p \cdot \text{UNSAT\_AERE\_RATIO} \quad (\text{A13})$$

If the PFT is c3\_arctic\_grass, c3\_nonarctic\_grass, or c4\_grass, then  $p = 0.3$ . For the remaining PFTs, the porosity is multiplied by NONGRASSPOROSRATIO (ratio of root porosity in non-grass to  
770 grass):

$$p = p \cdot \text{NONGRASSPOROSRATIO}. \quad (\text{A14})$$

A minimum aerenchyma porosity is set to 0.05. Therefore,  $p$  is modified as:

$$p = \max\{p, \text{POROSMIN}\}. \quad (\text{A15})$$

The aerenchyma area varies over the course of the growing season. Therefore, it is parameterized  
 775 using the simulated leaf area index as

$$T = \frac{f_N N_a L}{0.22} \pi R^2, \quad (\text{A16})$$

where  $L$  is the leaf area index ( $\text{m}^2 \text{m}^{-2}$ ) (used from CLM4.5 model simulation),  $N_a$  is the maximum  
 annual net primary production (NPP,  $\text{mol m}^{-2} \text{s}^{-1}$ ),  $R$  is the aerenchyma radius ( $2.9 \times 10^{-3} \text{ m}$ ), and  
 $f_N$  is the below-ground fraction of the current NPP.

780

The aerenchyma area  $T$  is multiplied by a scale factor to adjust it:

$$T = T \cdot \text{SCALE\_FACTOR\_AERE}. \quad (\text{A17})$$

The default value is 1.

#### A4 Methane Ebullition

785 The representation of the ebullition fluxes in the methane model is based on Wania et al. (2010). The  
 simulated aqueous  $\text{CH}_4$  concentration in each soil level is used to estimate the expected equilibrium  
 gaseous partial pressure as a function of temperature and pressure. When this partial pressure exceeds  
 $\text{VGC\_MAX}$ , bubbling occurs to remove  $\text{CH}_4$  to below this value, modified by the fraction of  $\text{CH}_4$  in  
 the bubbles (taken as 57%). The  $\text{VGC\_MAX}$  parameter is the ratio of saturation pressure triggering  
 790 ebullition.

#### A5 Aqueous and Gaseous Diffusion

Gaseous diffusivity in the soil depends on several factors such as molecular diffusivity, soil structure,  
 porosity, and organic matter content. The relationship between effective diffusivity ( $D_e$ ,  $\text{m}^2 \text{s}^{-1}$ ) and  
 soil properties is represented as

$$795 \quad D_e = D_0 \theta_a^2 \left( \frac{\theta_a}{\theta_s} \right)^{\frac{3}{b}} \cdot \text{SCALE\_FACTOR\_GASSDIFF}, \quad (\text{A18})$$

where  $\theta_a$  and  $\theta_s$  are the air-filled and saturated water-filled porosity,  $b$  is the slope of the water  
 retention curve, and  $\text{SCALE\_FACTOR\_GASSDIFF}$  is the scale factor for the gas diffusion (the default  
 value is 1).

### Appendix B: Observation Sites

800 Tables B1 and B2 show the information about the wetland and rice paddy observation sites, respec-  
 tively, where methane emissions have been measured.

## **Appendix C: Parameters and References for Bounds**

Table C1 shows the CH<sub>4</sub> related parameters in CLM4.5bgc and their literature reference information.

## **Appendix D: Weights Used for RMSE Computation in Equation (1) of the Manuscript**

805 Table D1 contains information about the weights used for each observation site when computing the  
objective function value.

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**Table 1.** CH<sub>4</sub> related parameters in CLM4.5bgc and their upper and lower bounds  $x_k^u$  and  $x_k^l$ , respectively, and the default parameter values  $v_k$ .

Parameter ID	Parameter name	$x_k^l$	$x_k^u$	$v_k$
1	Q10CH4	1	4	1.33
2	F_CH4	0.1	0.4	0.26
3	REDOXLAG	15	45	30
4	OXINHIB	200	600	400
5	PHMAX	8	10	9
6	PHMIN	2	4	2.2
7	VMAX_CH4_OXID	1.25e-6	1.25e-4	1.25e-5
8	K_M	0.0005	0.05	0.005
9	K_M_O2	0.002	0.2	0.002
10	Q10_CH4OXID	1	4	1.9
11	K_M_UNSAT	0.00005	0.005	0.0005
12	VMAX_OXID_UNSAT	1.25e-7	1.25e-5	1.25e-6
13	SCALE_FACTOR_AERE	0.2	2	1
14	NONGRASSPOROSRATIO	0.2	0.5	0.33
15	POROSMIN	0.01	0.2	0.05
16	ROB	2	4	3
17	UNSAT_AERE_RATIO	0.1	0.25	0.1667
18	VGC_MAX	0.05	0.3	0.15
19	SCALE_FACTOR_GASDIFF	1	5	1
20	ATMCH4	1.7e-7	1.7e-5	1.7e-6
21	MINO2LIM	0.1	0.3	0.2

**Table 2.** Parameters that are sensitive for most observation sites (out of 16).

Parameter ID	Parameter name	# sensitive sites
1	Q10CH4	16
2	F_CH4	16
7	VMAX_CH4_OXID	16
13	SCALE_FACTOR_AERE	16
9	K_M_O2	15
15	POROSMIN	14
16	ROB	11
8	K_M	10
17	UNSAT_AERE_RATIO	10
10	Q10_CH4OXID	9
21	MINO2LIM	9



**Table 3.** Parameters that are least sensitive for observation sites (out of 16).

Parameter ID	Parameter name	# insensitive sites
3	REDOXLAG	16
4	OXINHIB	16
5	PHMAX	16
6	PHMIN	16
14	NONGRASSPOROSRATIO	16
18	VGC_MAX	16
20	ATMCH4	15
11	K_M_UNSAT	13
19	SCALE_FACTOR_GASDIFF	13
12	VMAX_OXID_UNSAT	10

**Table 4.** Default and optimized parameter values of optimization trials T1, T2, and T3 for the 5-dimensional pseudo data case. We report four decimal places because the model output is sensitive to very small changes for some variables. Note that we scaled the numbers to the interval [0,1].

Param.	Default	T1	T2	T3
1	0.1100	0.1088	0.1099	0.1091
2	0.5333	0.5366	0.5385	0.5458
7	0.0909	0.0912	0.0943	0.0967
13	0.4444	0.4461	0.4454	0.4443
21	0.5000	0.4936	0.4934	0.4856
RMSE	0	0.28	0.46	0.40

**Table 5.** Default and optimized parameter values of optimization trial T3 and parameter values for the point CP that was sampled during the same optimization trial and that is closer to the default point, but that has a worse objective function value (11-dimensional pseudo data case).

Param.	Default	T3	CP
1	0.1100	0.1148	0.1148
2	0.5333	0.5806	0.5806
7	0.0909	0.1336	0.1336
8	0.0909	0.1785	0.1785
9	0.0909	0.1248	0.1248
10	0.3000	0.4375	<b>0.4302</b>
13	0.4444	0.7107	0.7107
15	0.2105	0.1778	0.1778
16	0.5000	0.9583	0.9583
17	0.4444	0.2740	0.2740
21	0.5000	0.4436	<b>0.4583</b>
RMSE	0	2.28	2.35

**Table 6.** Default and optimized parameter values of optimization trials T1, T2, and T3 for the 5-dimensional real data case. Bold indicates optimized parameters that are on (or close to) the variable boundary (all variables are scaled to [0,1]).

Param.	Default	T1	T2	T3
1	0.1100	<b>0</b>	<b>0</b>	<b>0</b>
2	0.5333	0.1705	0.1747	0.1699
7	0.0909	0.7878	0.7518	0.7865
13	0.4444	<b>0</b>	<b>0</b>	<b>0.0267</b>
21	0.5000	<b>1</b>	<b>1</b>	<b>1</b>
RMSE	156.40	114.24	114.11	114.24

**Table 7.** Default and optimized parameter values of optimization trials T1, T2, and T3 for the 11-dimensional real data case. Bold indicates optimized parameters that are on the variable bound (all variables are scaled to [0,1]).

Param.	Default	T1	T2	T3
1	0.1100	<b>0</b>	<b>0</b>	<b>0</b>
2	0.5 333	0.4220	0.3298	0.3813
7	0.0909	0.7093	0.6889	0.7260
8	0.0909	<b>1</b>	<b>1</b>	0.9754
9	0.0909	<b>0</b>	0.2335	0.6971
10	0.3000	0.7702	0.6195	0.6195
13	0.4444	<b>1</b>	<b>1</b>	0.8063
15	0.2105	0.6987	<b>1</b>	<b>1</b>
16	0.5000	0.0865	0.4274	0.2473
17	0.4444	0.8543	0.3113	0.5359
21	0.5000	0.5064	0.7449	0.5586
RMSE	164.46	107.24	107.58	107.41

**Table 8.** Unweighted RMSE values for each site using the best parameters found during optimization trial T1 of the  $d = 11$  real data case and trial T2 of the  $d = 5$  real data case and with default parameter values.

Site	Name	Unweighted RMSE $d = 5$	Unweighted RMSE $d = 11$	Unweighted RMSE default
1	Alberta	220.34	203.82	209.25
2	Florida	1247.70	1280.29	1180.99
3	Michigan	334.01	337.51	328.10
4	Minnesota	41.05	35.16	34.31
5	Nanjing	97.88	96.14	212.18
6	Vercelli	325.34	326.04	293.36
7	Texas	179.21	139.09	116.85
8	Japan	132.31	161.22	184.88
9	California	372.71	374.59	360.37
10	New Delhi	18.67	19.96	14.21
11	Beijing	66.79	60.89	56.99
12	Java	49.09	54.61	221.52
13	Chengdu	231.93	241.91	198.42
14	Cuttack	72.01	63.75	364.75
15	Panama	446.83	464.59	422.86
16	Salmisuo	156.79	132.16	146.52
Total RMSE		3792.66	3991.73	4345.56

**Table B1.** Wetland site data. P = precipitation, T = temperature

Site Name	Location	Time	Wetland type	Dominant vegetation	Mean P & T	Soil and climate characteristics	Meas. techniques	Forcing datasets*	References
Michigan, USA	42.45 N, 84.00 E	1991 - 1993	Ombrotrophic peatland	Sphagnum, Scheuchzeria palustris, Vaccinium oxycoccos	P: 761mm (1948-1980)	Soil pH: 4.2	Static chamber	Measured positions	Shannon and White (1994)
Minnesota, USA	47.53 N, 266.33 E	1991 - 1992	Poorly minerotrophic to ombrotrophic peatland	Sphagnum, Chamaedaphne calyculata, Scheuchzeria palustris	P: 553mm, T: $\approx 13.6^{\circ}\text{C}$ for May-October period	Soil pH: 4.6	Eddy correlation technique	Measured positions	Shurpali and Verma (1998)
Alberta, Canada	54.60 N, 246.60 E	1994 - 1996	Nutrient rich fen	Carex aquatilis and Carex rostrata	-	Soil pH: 7; the freeze-thaw cycle spans from May to Oct	Open chamber	Measured positions	Popp et al. (2000)
Salmisuo, Eastern Finland	62.75 N, 30.93 E	1993	Minerotogenic, oligotrophic pine fen	Sphagnum papillosum	T: $\approx 10^{\circ}\text{C}$	Wet condition from Jul to Sept	Static chamber	Measured positions	Saario et al. (1997)
Florida, USA	30.07 N, 275.80 E	1993	Swamp	Sagittaria lancifolia	Annual $\approx 1400\text{mm}$	Soil pH: 6.2	Open chamber	Measured positions	Lombardi et al. (1997)
Panama	9.00 N, 80.00 E	1987	Swamp	Palms	-	Soil pH: 6.2; Feb to May is the dry season	Static chamber	Modeled WT positions	Keller (1990); Walter and Heimann (2000)

**Table B2.** Rice paddy site data

Site Name	Location	Year	Date of Field flooded	Date of final drainage	pH	Measurement techniques	Soil type	References
Texas, USA	29.95 N, 265.50 E	1994	17-May	11-Aug	N/A	Chamber	Bernard-Morey	Sigren et al. (1997)
Vercelli, Italy	45.30 N, 8.42 E	1991	7-May	30-Aug	6	Static chamber	Sandy loam	Butterbach-Bahl et al. (1997)
Chengdu, China	31.27 N, 105.45 E	2003	9-May	7-Sep	8.1	Chamber	Purplish	Jiang et al. (2006)
Nanjing, China	32.80 N, 118.75 E	1999	18-Jun	13-Oct	N/A	Chamber	Hydromorphic	Huang et al. (2001)
Beijing, China	40.55 N, 116.78 E	1995	4-Jun	17-Oct	7.99	Automatic chamber	Silty clay loam	Wang et al. (2000)
California, USA	40.20 N, 237.98 E	1982&1983	11-May (1982); 21-May (1983)	2-Oct (1982); 1-Oct (1983)	N/A	Static chamber	Capay silty clay	Cicerone et al. (1992, 1983)
Japan	36.02 N, 140.22 E	1991&1993	7-May	12-Aug (1991); 2-Sept (1993)	6.6-6.9	Automatic chamber	Gley soil (Sandy clay loam)	Yagi et al. (1996)
New Delhi, India	28.63 N, 77.12 E	1995	1-Jul	1-Nov	8.2	Closed chamber, manual	Ustochrept (sandy loam)	Jain et al. (2000)
Cuttack, India	20.42 N, 85.92 E	1996	19-Jul	30-Oct	6.19	Automatic chamber	Haplaquept (Al-luvial)	Adhya et al. (2000)
Central Java, Indonesia	6.78 S, 110.15 E	2001-2002	1-Nov	28-Feb	5.1	Automatic closed chamber	Aeric Tropaquept (Silty loam)	Setyanto et al. (2004)

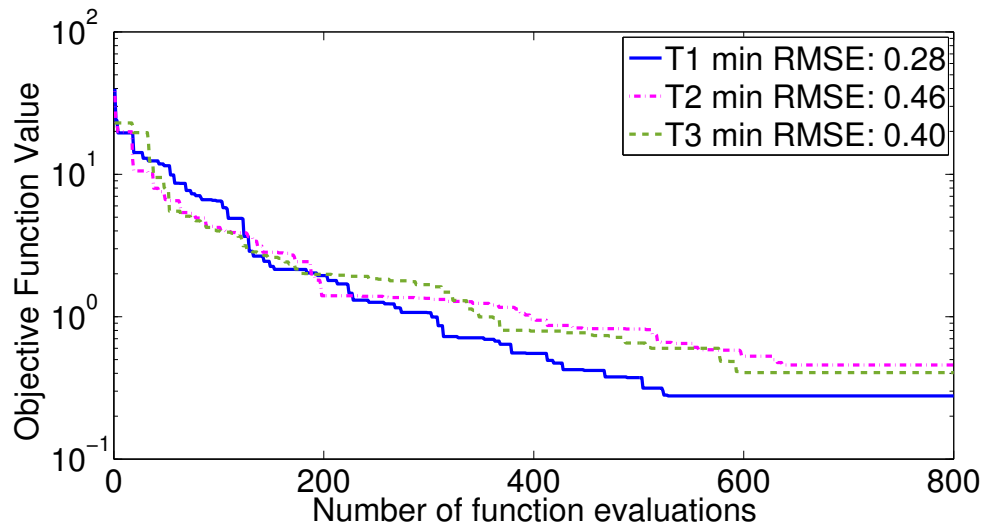


**Table C1.** Parameter names, descriptions, ranges, and literature references

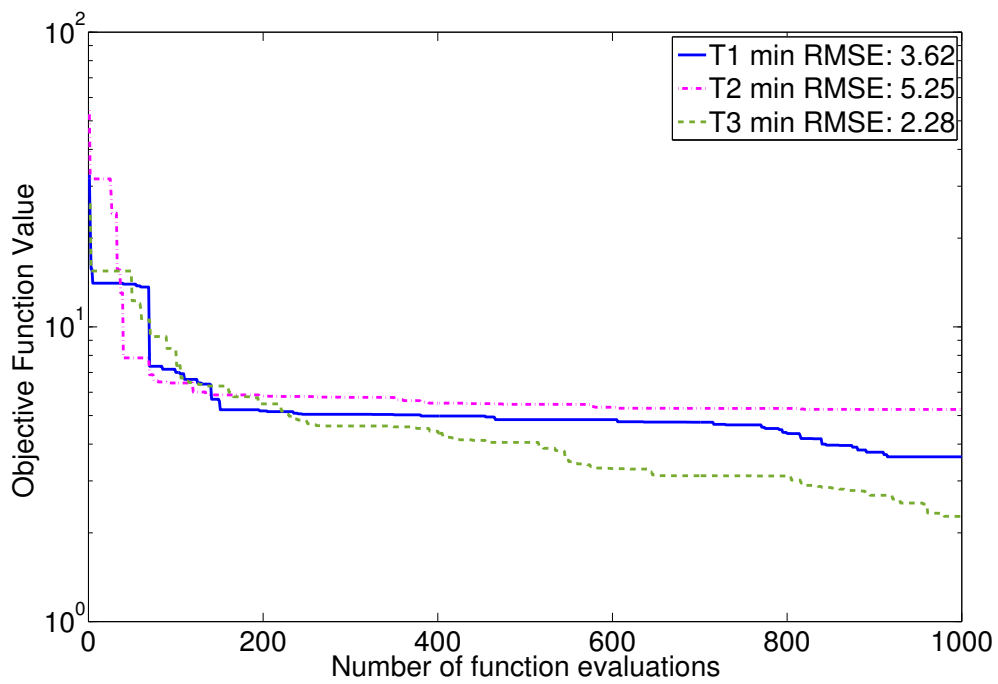
Number	Parameter	Description	Units	Range	References
1	q10ch4	Q10 for methane production	unitless	1 - 10	Dunfield et al. (1993); Walter and Heimann (2000); Riley et al. (2011)
2	f_ch4	Ratio of CH <sub>4</sub> production to total C mineralization	unitless	0.05 - 0.5	Wania et al. (2010); Zhang et al. (2002); Zhu et al. (2013), Effective value will depend on temperature, redox and pH but cannot exceed 50% based on stoichiometry (Bill Riley, personal communication)
3	redoxlag	Number of days to lag for production	days	15 - 45	Meng et al. (2012); Conrad (2002); Cheng et al. (2007)
4	oxinhib	Inhibition of methane production by oxygen	m <sup>3</sup> /mol	200 - 600	Arah and Stephen (1998); Riley et al. (2011)
5	pHmax	Maximum pH for methane production	unitless	8 - 10	Cao et al. (1996); Zhang et al. (2002); Zhuang et al. (2004); Meng et al. (2012)
6	pHmin	Minimum pH for methane production	unitless	2 - 4	Cao et al. (1996); Zhang et al. (2002); Zhuang et al. (2004); Meng et al. (2012)
7	vmax_ch4_oxid	Oxidation rate constant	mol/m <sup>3</sup> -w/s	1.25e-6 - 1.25e-4	Riley et al. (2011); Walter and Heimann (2000); Dunfield et al. (1993); Knoblauch (1994)
8	k_m	Michaelis-Menten oxidation rate constant for CH <sub>4</sub> conc.	mol/m <sup>3</sup> -w	5e-4 - 5e-2	Segers and Kengen (1998); Walter and Heimann (2000); Riley et al. (2011)
9	k_m_o2	Michaelis-Menten oxidation rate constant for O <sub>2</sub> conc.	mol/m <sup>3</sup> -w	0.002 - 0.2	Segers (1998); Walter and Heimann (2000); Riley et al. (2011)
10	q10_ch4oxid	Q10 oxidation constant	unitless	1 - 4	Meng et al. (2012); ?; Walter and Heimann (2000); Zhu et al. (2013); Zhang et al. (2002)
11	k_m_unsat	Michaelis-Menten oxidation rate constant for CH <sub>4</sub> conc. in upland areas	mol/m <sup>3</sup> -w	5e-5 - 5e-3	Whalen and Reeburgh (1996); Bender and Conrad (1992); Riley et al. (2011)
12	vmax_oxid_unsat	Oxidation rate constant in upland areas	mol/m <sup>3</sup> -w/s	1.25e-7 - 1.25e-5	Whalen and Reeburgh (1996); Bender and Conrad (1992); Riley et al. (2011)
13	scale_factor_aere	Scale factor on the aerenchyma area	unitless	0.2 - 5	Riley et al. (2011)
14	nongrassporosratio	Ratio of root porosity in non-grass to grass	unitless	0.2 - 0.5	Colmer (2003)
15	porosmin	Minimum aerenchyma porosity	unitless	0.01 - 0.2	Colmer (2003); Cronk and Fennessy (2001)
16	rob	Ratio of root length to vertical depth ("root obliquity")	unitless	2 - 4	Arah and Stephen (1998); Riley et al. (2011). This parameter is poorly constrained.
17	unsat_aere_ratio	Ratio to multiply upland vegetation aerenchyma porosity by compared to inundated systems	unitless	0.1 - 0.25	Not available in literature. The reasonable range could be between 0.1 and 0.25. Meng et al. (2012) used this range for sensitivity.
18	vgc_max	Ratio of saturation pressure triggering ebullition	unitless	0.05 - 0.3	Kellner et al. (2006); Baird et al. (2004)
19	scale_factor_gasdiff	Scale factor for gas diffusion	unitless	1 - 5	Range not available. Reasonable range is 1-5 for sensitivity analyses.
20	atmch4	Atm. CH <sub>4</sub> mixing ratio to prescribe	mol/mol	1.7e-7 - 1.7e-5	Range not available. Variable range; global average is $\approx 1.7e-6$
21	mino2lim	Min. anaerobic decomposition rate as a fraction of potential aerobic rate	unitless	0.05 - 0.45	Range not available in the literature. The default value (0.2) is from Riley et al. (2011). The reasonable range could be between 0.05 and 0.45 to adjust effect of anoxia on decomposition rate (used to calculate seasonal inundation factor). The range is considered based on knowledge.

**Table D1.** ID, name of observation sites, and associated weights for real data and pseudo data case (equation (1) of the main document)

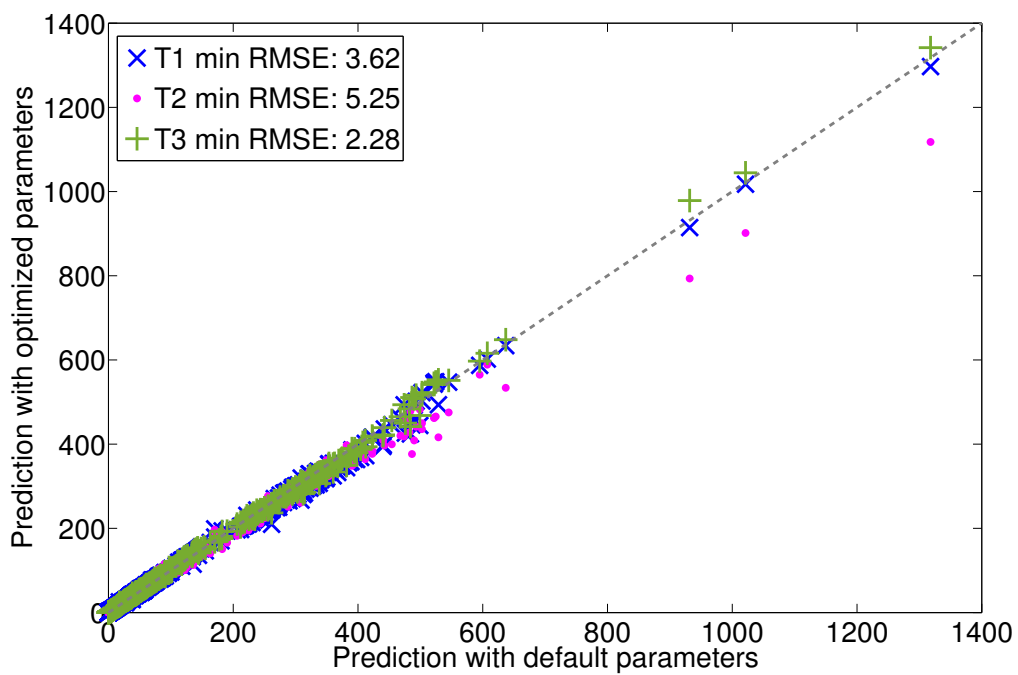
ID	Location	$w_i$ real data	$w_i$ pseudo data
1	Alberta	0.0327	0.0656
2	Florida	0.0078	0.0067
3	Michigan	0.0280	0.1599
4	Minnesota	0.0938	0.0783
5	Nanjing	0.0566	0.0149
6	Vercelli	0.0198	0.0382
7	Texas	0.0267	0.0189
8	Japan	0.0441	0.0153
9	California	0.0421	0.0684
10	New Delhi	0.2787	0.1707
11	Beijing	0.1053	0.1189
12	Central Java	0.0810	0.0143
13	Chengdu	0.0283	0.0571
14	Cuttack	0.0968	0.0104
15	Panama Swamp	0.0177	0.0795
16	Salmisuo	0.0405	0.0827



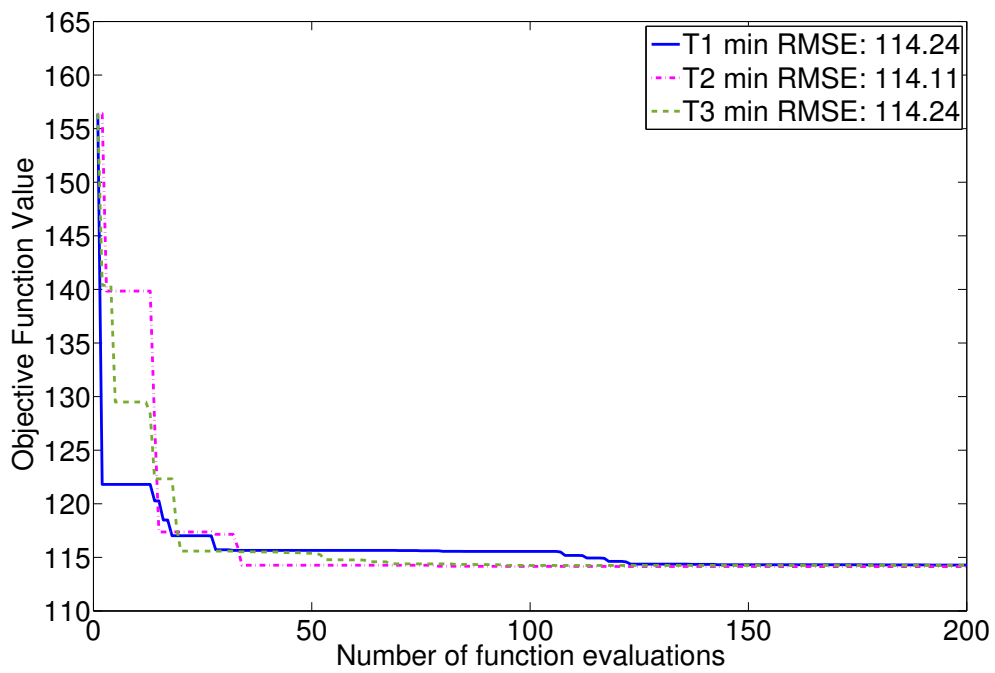
**Figure 1.** Progress plot that shows the development of the best objective function value found versus the number of function evaluations for the pseudo data case with  $d = 5$  parameters for optimization trials T1, T2, and T3. The legend shows the lowest RMSE value found in each trial.



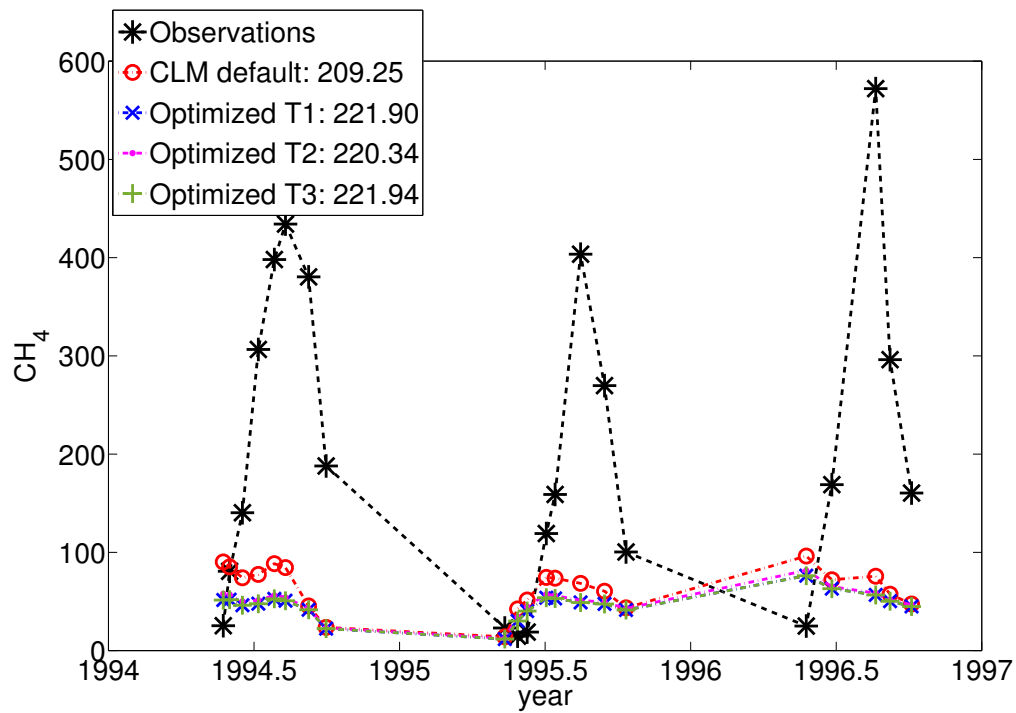
**Figure 2.** Progress plot that shows the development of the best objective function value found versus the number of function evaluations for the pseudo data case with  $d = 11$  parameters for optimization trials T1, T2, and T3. The legend shows the lowest RMSE value found in each trial.



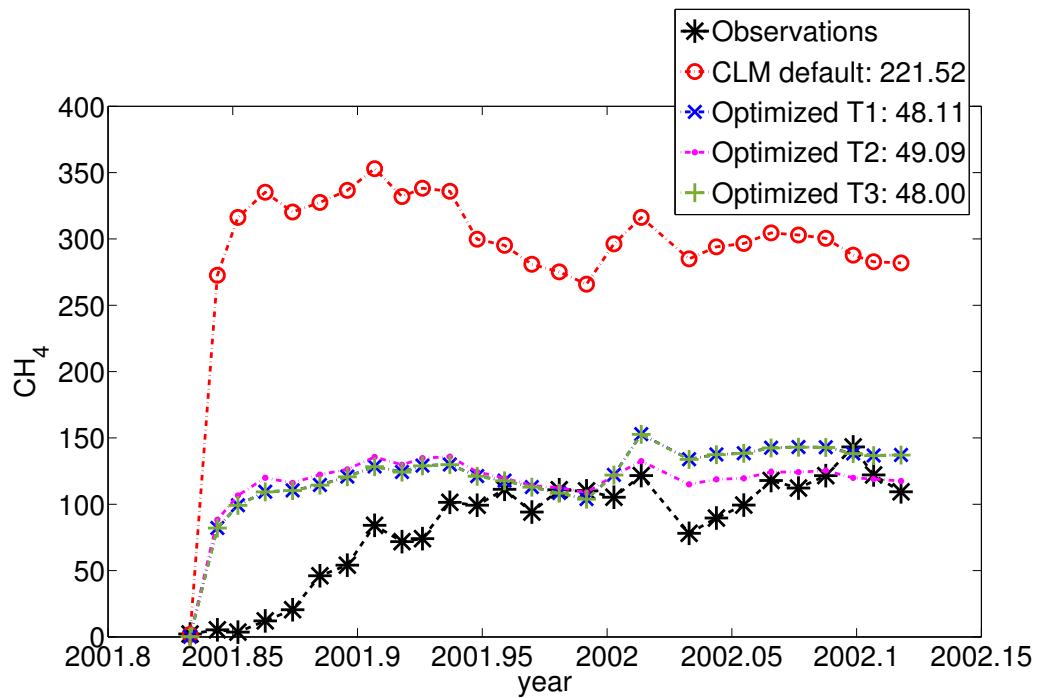
**Figure 3.** CLM4.5bgc CH<sub>4</sub> predictions when using the default parameter values versus the predictions when using the best solution found in each of the three optimization trials T1, T2, and T3, respectively, for the pseudo data case with  $d = 11$  parameters. The legend shows the lowest RMSE value found in each trial.



**Figure 4.** Progress plot that shows the development of the best objective function value found versus the number of function evaluations for the real data case with  $d = 5$  parameters for optimization trials T1, T2, and T3. The legend shows the lowest RMSE value found in each trial. The first function evaluation (left side of the graphs) corresponds to the RMSE when using the default parameters.

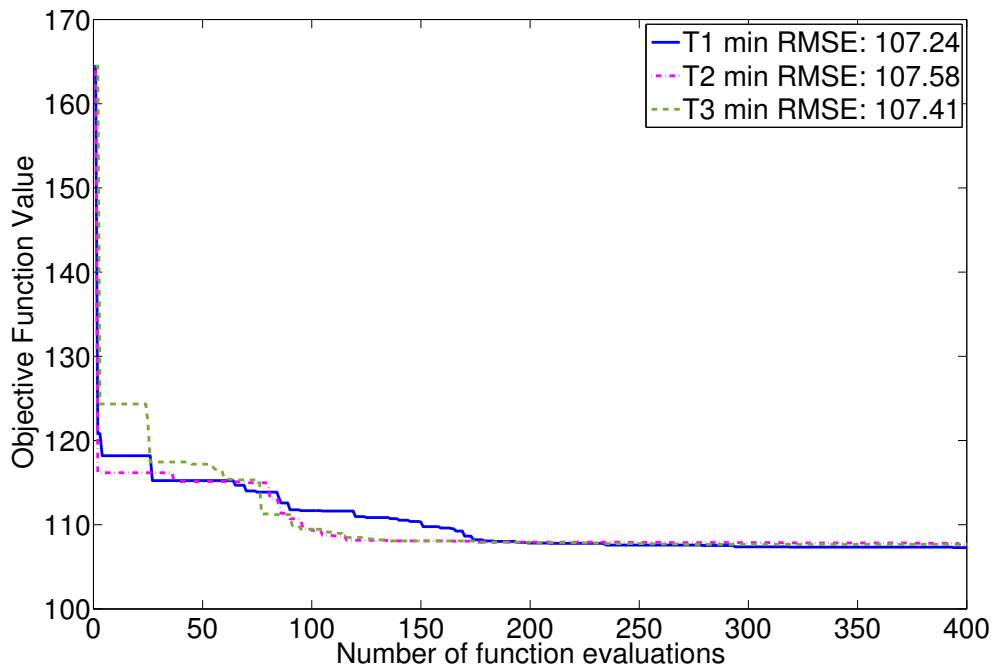


**Figure 5.** CH<sub>4</sub> emission observations and predictions when using the optimized parameters of optimization trials T1, T2, and T3, respectively, and when using the default parameters for the wetland site Alberta, Canada, for the real data case with  $d = 5$  parameters. The legend shows the lowest RMSE value found in each trial.

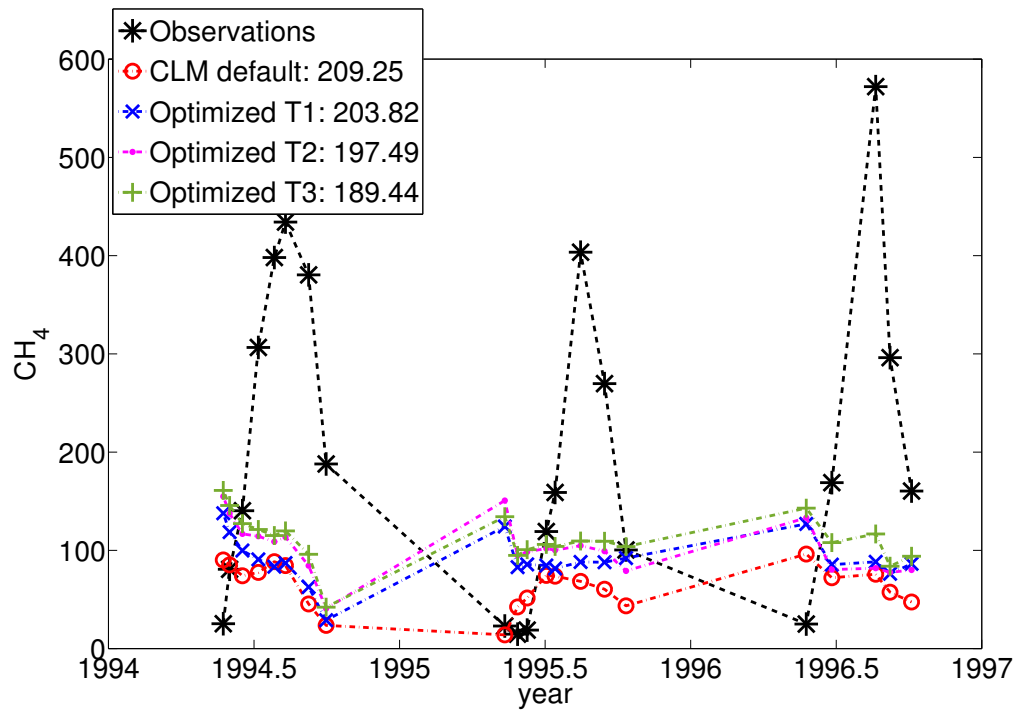


**Figure 6.** CH<sub>4</sub> emission observations and predictions when using the optimized parameters of optimization trials T1, T2, and T3, respectively, and when using the default parameters for the rice paddy site Central Java, Indonesia, for the real data case with  $d = 5$  parameters. The legend shows the lowest RMSE value found in each trial.

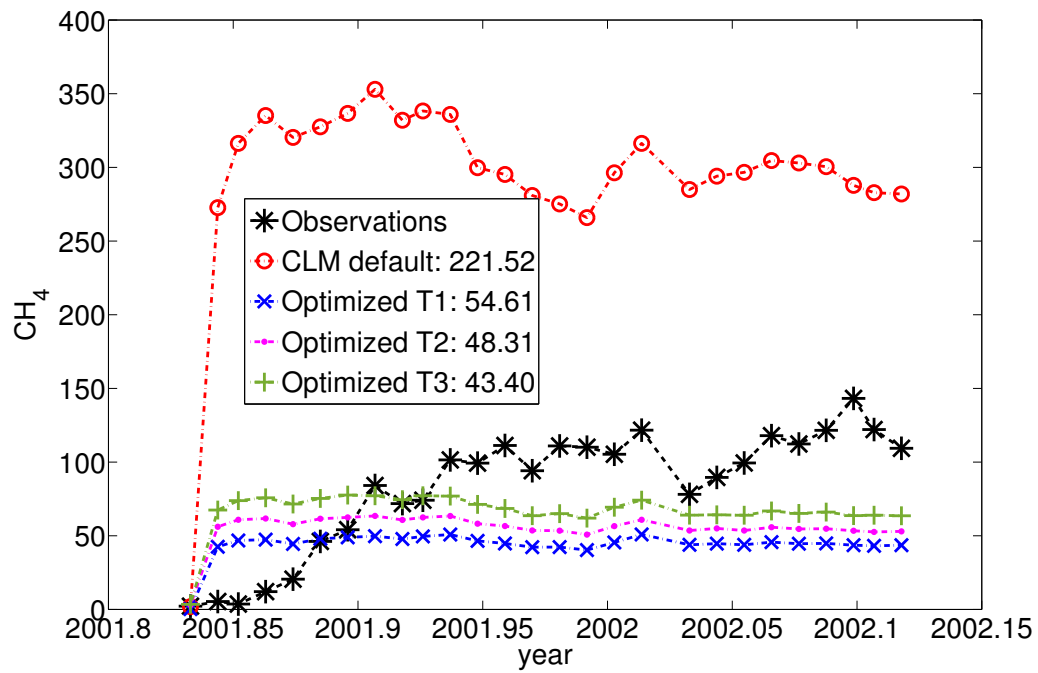




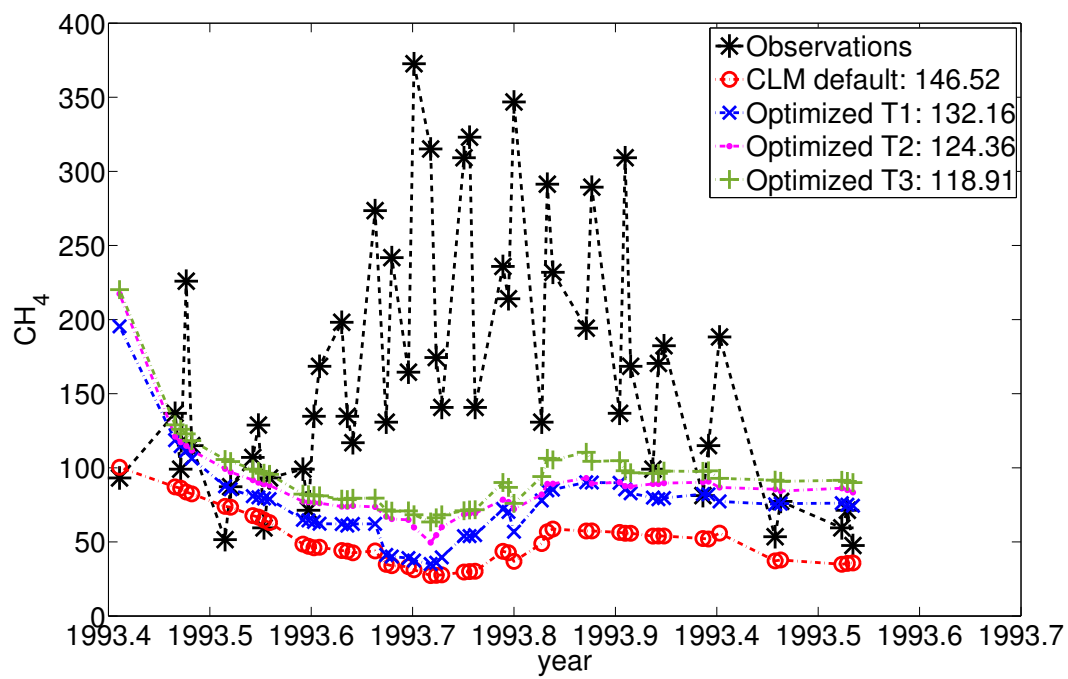
**Figure 7.** Progress plot that shows the development of the best objective function value found versus the number of function evaluations for the real data case with  $d = 11$  parameters for optimization trials T1, T2, and T3. The legend shows the lowest RMSE value found in each trial. The first function evaluation (left side of the graphs) corresponds to the RMSE when using the default parameters.



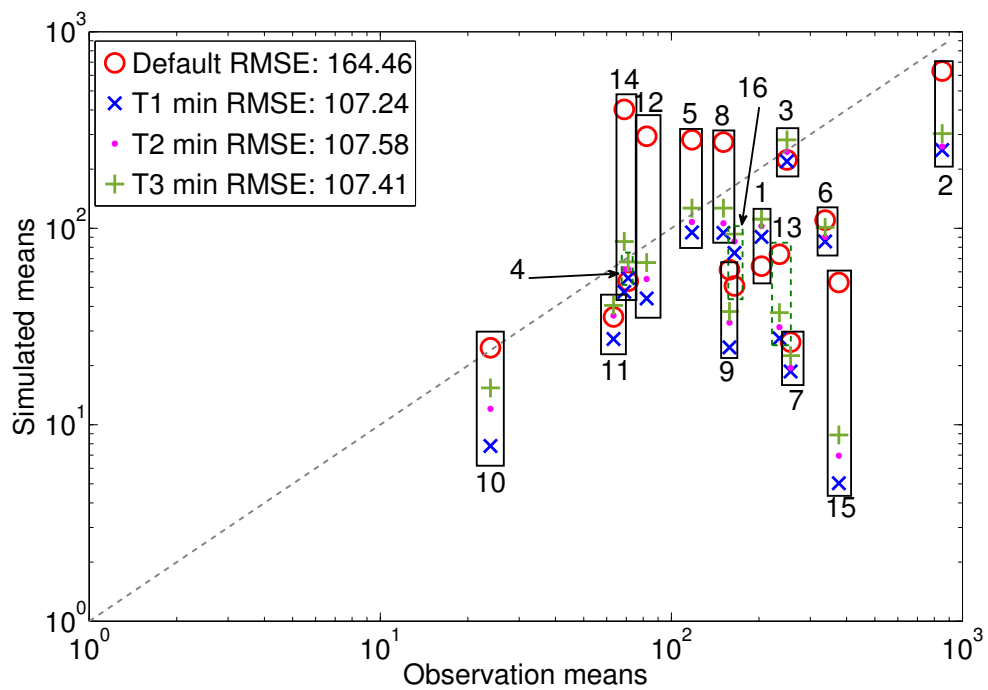
**Figure 8.** CH<sub>4</sub> emission observations and predictions when using the optimized parameters of optimization trials T1, T2, and T3, respectively, and when using the default parameters for the wetland site Alberta, Canada, for the real data case with  $d = 11$  parameters. The legend shows the lowest RMSE value found in each trial.



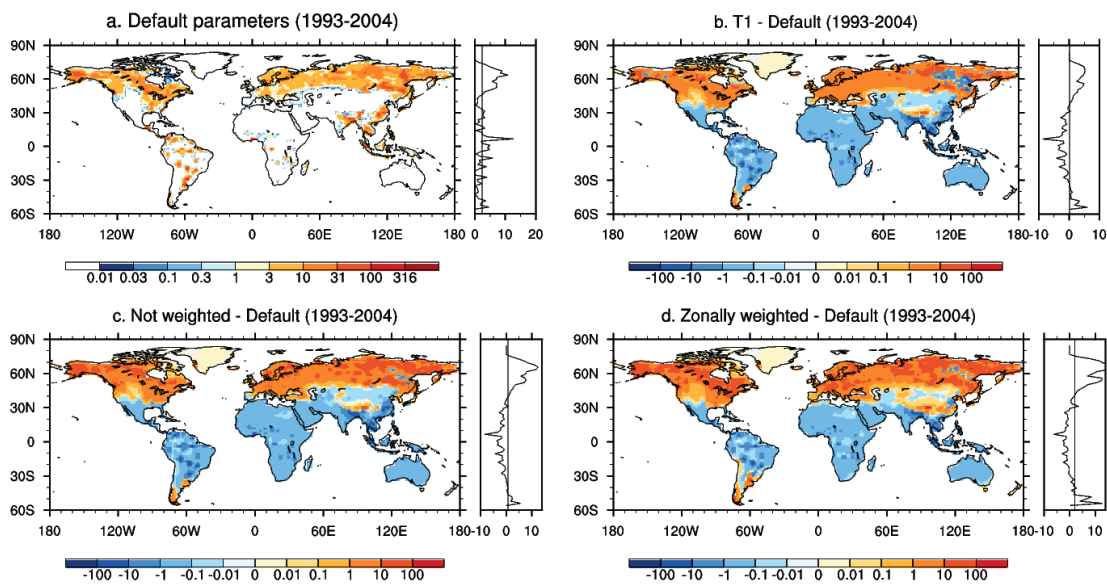
**Figure 9.** CH<sub>4</sub> emission observations and predictions when using the optimized parameters of optimization trials T1, T2, and T3, respectively, and when using the default parameters for the rice paddy site Central Java, Indonesia, for the real data case with  $d = 11$  parameters. The legend shows the lowest RMSE value found in each trial.



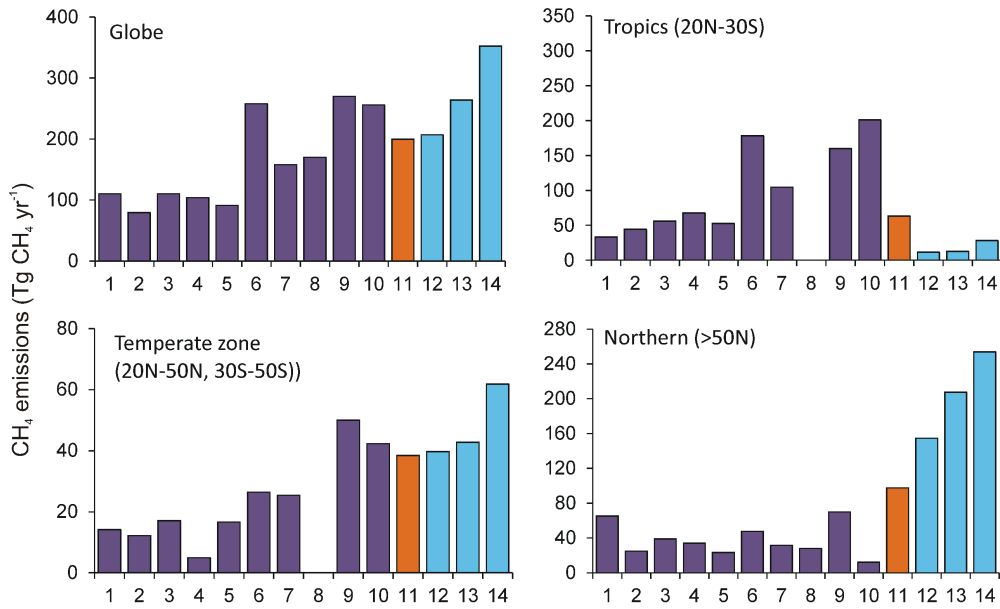
**Figure 10.** CH<sub>4</sub> emission observations and predictions when using the optimized parameters of optimization trials T1, T2, and T3, respectively, and when using the default parameters for the wetland site Salmisuo, Finland, for the real data case with  $d = 11$  parameters. The legend shows the lowest RMSE value found in each trial.



**Figure 11.** Scatterplot showing the mean values of the CH<sub>4</sub> predictions using the default and optimized parameter values of trials T1, T2, and T3, respectively, versus the mean values of the observations. The numbers in the legend show the best RMSE value corresponding to each trial. The numbers above/below the boxes indicate the observation site ID (1-Alberta, 2-Florida, 3-Michigan, 4-Minnesota, 5-Nanjing, 6-Vercelli, 7-Texas, 8-Japan, 9-California, 10-New Delhi, 11-Beijing, 12-Central Java, 13-Chengdu, 14-Cuttack, 15-Panama, 16-Salmisuo).



**Figure 12.** Average methane emissions ( $\text{mg CH}_4\text{m}^{-2} \text{d}^{-1}$ ) simulated by CLM4.5bgc for (a) default parameters, (b) differences between default parameters and 11-dimensional optimization trial T1, (c) differences between default parameters and optimization trial with unweighted sum of RMSE, and (d) differences between default parameters and optimization trial with zonally weighted sum of RMSE. Zonal means are shown on the right side of each spatial plot.



**Figure 13.** Comparison of total methane emissions ( $\text{Tg CH}_4 \text{ yr}^{-1}$ ) between CLM4.5bgc and other models from natural wetlands. 1: Matthews and Fung (1987), 2: Aselmann and Crutzen (1989), 3: Bartlett et al. (1990), 4: Bartlett and Harriss (1993), 5: Cao et al. (1996), 6: Walter et al. (2001), 7: Bousquet et al. (2006), 8: Bloom et al. (2010), 9: CLM4Me, Riley et al. (2011), 10: CLM4Me', Meng et al. (2012), 11: This study, CLM4.5bgc with default parameters, 12: This study, CLM4.5bgc with  $d = 11$  optimized parameters of T1, 13: This study, CLM4.5bgc with  $d = 11$  optimized parameters of unweighted sum of RMSE, and 14: This study, CLM4.5bgc with  $d = 11$  optimized parameters of zonally weighted RMSE. Note that number 7 is a top-down approach and number 9 may include the rice paddy emissions. For number 8, no data was available for the tropics and the temperate zone.