Uncertainties in the national inventory of methane emissions from rice cultivation: field measurements and modeling approaches

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Abstract. Uncertainties in national inventories originate from a variety of sources, including methodological failures, errors and insufficiency of supporting data. In this study, we analyzed these sources and their contribution to uncertainty in the national inventory of rice paddy methane emissions in China and compared the differences in the approaches used (e.g., direct measurements, simple regressions and more complicated models). For the 495 field measurements we collected from the scientific literature, the area-weighted 95% CI ranged from 13.7 to 1115.4 kg CH_4 ha^{-1}, and the histogram distribution of the measurements agreed well with parameterized gamma distributions. For the models, we compared the performance of methods of different complexity (i.e., the CH4MOD model, representing a complicated method, and two less complex statistical regression models taken from literatures) to evaluate the uncertainties associated with model performance as well as the quality and accessibility of the regional datasets. Comparisons revealed that the CH4MOD model may perform worse than the comparatively simple regression models when no sufficient input data for the model were available. As simulated by CH4MOD with data of irrigation, organic matter incorporation and soil properties of rice paddies, the modelling methane fluxes varied from 17.2 kg CH_4 ha^{-1} to 708.3 kg CH_4 ha^{-1}, covering 63% of the range of the field measurements. When applying the modeling approach to the 10 km × 10 km gridded dataset of the model input variables, the within-grid variations, made via the Monte Carlo method, were found to be 81.2%–95.5% to the grid means. Up-scaling the grid estimates to the national inventory, the total methane emission from the rice paddies was 6.43 (3.79–9.77) Tg. The fallacy of CH4MOD contributed 56.6% of the total uncertainty, with the remaining 43.4% being attributed to errors and the scarcity of the spatial datasets of the model inputs. Our analysis reveals the dilemma between model performance and data availability when using a modeling approach: a model with better performance may help in reducing uncertainty caused by model fallacy but increases the uncertainty caused by data scarcity, as greater levels of input are needed to improve performance. Reducing the total uncertainty in the national methane inventory depends on a better understanding of both the complexity of the mechanisms of methane emission and the spatial correlations of the factors that influence methane emissions from rice paddies.

Keywords: Uncertainty, source and contribution, spatial variation, national inventory, methane emission

1 Introduction

Rice cultivation is a major source of anthropogenic methane and a prime target of greenhouse gas mitigation efforts (Tian et al., 2016; Smith et al., 2008). Globally, the methane emission from rice cultivation was about 18.3Tg CH_4 yr^{-1} under intermittent irrigation and 38.8Tg CH_4 yr^{-1} under continuous flooding in the 2000s (Zhang et al., 2016). Methane fluxes in rice paddies varied extensively with environmental and agronomic factors. Certain factors, such as rice biomass (Bachelet and Neue, 1993), organic matter input (Kern et al., 1995), water management (Khosa et al., 2011;Mishra et al., 1997), paddy soil properties (Yao et al., 1999;Gaunt et al., 1997), climate Sass et al., 1991) and rice varieties (Su et al., 2015; Ding et al., 1999), have been recognized as having significant impacts on methane emissions. Other factors, such as atmospheric CO_2 and ozone contents (Dijkstra, et al., 2012; Bhatia et al., 2011;Inubushi et al., 2011), N fertilizer application (Banger et al., 2012;Xie et al., 2010a) and active soil organic C (Zhan et al., 2011), and even the field management of the previous crop that rotating with rice (Weller et al., 2016) are also receiving increasing attention. Because so many factors affect the production, oxidation and emission of methane from rice cultivation, the observed methane fluxes varied extensively both spatially and temporally.

Numerous methods have been applied for estimating national and global inventories of rice paddy methane emissions, including meta-analysis of direct measurements, process models and empirically based statistical models. However, the range of national/global source estimates remains large (Cao et al., 1996;Sass et al., 1999;Chen et al., 2013). The major factors that are known to regulate rice paddy methane emissions include agricultural management practices (Khosa et al., 2011;Sanchis et al., 2012;Sass et al., 1992;Bodelier and Laanbroek, 2006) and environmental conditions, such as climate and soil properties (Conrad et al., 2007;Inubushi et al., 2011;Sass et al., 1991). Currently, techniques for calculating methane emissions differ substantially and usually in the way of scenario simulations (Ito et al., 2012; Van Bodegom et al., 2002a, b; Verburg et al., 2006), without integrated consideration of methodological fallacy and data insufficiency.

By extrapolating field measurements obtained from experiments, methane emissions from the 30 million hectares or so of land under rice cultivation in China were estimated to range from 21.6 Tg CH_4 yr^{-1} to 30 Tg CH_4 yr^{-1} (Matthews et al., 1991;Taylor et al., 1991), much larger than the result of a recent study (Zhang et al., 2016). The extrapolation of methane emission rates from site measurements to larger regions is unlikely to yield reliable results because of the tremendous spatial heterogeneity in environmental conditions and agronomic activities (Ogle et al., 2010). Other studies have described the relationships between methane emissions and rice NPP (net primary productivity) (Bachelet and Neue, 1993) and organic matter inputs (Bachelet et al., 1995). Ambient temperature and the use of nitrogen (N) fertilizer have also been identified as determinants of methane emissions (Kern et al., 1995;Bachelet et al., 1995). Until the significant reduction in methane emissions caused by mid-season drainage was confirmed (Sass and Fisher, 1997;Yagi et al., 1997;Li et al., 2002;Yan et al., 2005), all previous regional and national estimates (obtained using extrapolation or regression equations) were derived from continuously flooded rice fields. More factors such as the rice cultivar involved (Watanabe et al., 1995;Butterbach-Bahl et al., 1997;Ding et al., 1999;Inubushi et al., 2011), soil properties (Sass et al., 1994;Yao et al., 1999) and atmospheric CO_2 (Dijkstra et al, 2012; Xie et al., 2010b) and ozone (Bhatia et al., 2011) concentrations have also been incorporated into models designed to estimate methane emissions from rice paddies. Complex interactions among these factors have spurred model development (Cao et al., 1995;Li, 2000;Matthews et al., 2001;Huang et al., 1998;Van Bodegom et al., 2001;Huang et al., 2004). To delineate variations in methane emissions and to reduce uncertainties, the impacts of these factors on the production, oxidation and emission of methane were mathematically incorporated into the models. Models with more factors involved are able to reduce uncertainties in estimating methane emissions, but the estimates generated by these models still differ significantly across multiple spatial and temporal scales (Butenhoff et al., 2009;Ren et al., 2011;Chen et al., 2013).

Reduction of the uncertainty in estimated methane emissions requires the development of an effective and reliable model that incorporates various paddy environments and agronomic activities. However, our understanding of the complex biogeochemical processes that occur in paddy soils is poor. When estimating methane emissions from rice agriculture, only factors that are thought to be key determinants of methane emissions have been incorporated into the models. Excluding other factors introduces errors into the model output (Equations C2 and C3 in the Supporting Information). Improving our knowledge of methane processes in the future will increase the number of factors that are integrated into models and potentially delineate details related to spatial/temporal variations.

Uncertainties in regional estimates of methane emissions from rice paddies stem not only from fallacy in the applied models but also from errors and inadequate data, which we discussed in a previous study (Zhang et al., 2014; Appendix D in the Supporting Information). A model with more factors generally performs better than a model with fewer factors but requires a larger amount of data to facilitate model application. A model with good performance (less fallacy) can still result in large uncertainties when the available input data (e.g., soil properties, rice irrigation, types and amount of organic matter) are insufficient (Zhang et al., 2014; Ito et al., 2012).

In the present study, we analyzed the uncertainties in experimental measurements of methane fluxes in different rice paddies. We also evaluated the performance of different methods involving a diversity of input variables and the influence of data availability on the performance of these methods. Finally, the uncertainty in the national emissions inventory as a consequence of variable model performance and according to the quality and availability of input data were discussed.

2 Materials and methods

2.1 Field measurements of methane emissions from rice paddies in China

The observational data used in this study (Table 1) consisted of field methane fluxes measured at 33 sites (Fig. 1). We obtained these measurements from the published literature concerning all crop rotations with rice cultivation in China (double rice, winter wheat and rice rotation, single rice crop cultivation, and so forth) (Wei, 2012). A total of 495 measurements were taken at the 33 sites, after excluding those had been use for the model calibration (Neue et al., 1990; Kern et al., 1997; Huang et al, 2004). The amount of organic matter added to the rice paddies ranged from 0 t C ha^{-1} to 15.3 t C ha^{-1} and included animal manure, green manure, crop straw, biogas residuals and their various components. The applied water regimes consisted of continuous flooding, single mid-season drainage and multi-drainage irrigations.

Model performance was assessed by comparing the model estimates with the measurements. To drive the models, data pertaining to rice yields, soil properties and crop phenologies were collected from the relevant literature (Appendix B in the Supporting Information).

2.2 Performance of the methods used to estimate methane emissions

The uncertainties produced by the models derive from model fallacy (Kennedy and O'Hagan, 2001, Appendix C in the Supporting Information) as well as from the quality and availability of data (Fig. 2). Model performance was assessed by comparing model outputs with the direct measurements (left part in Fig. 2). Errors in the input data of the model can be propagated in the obtained estimates (right side of Fig. 2, Appendix D in the Supporting Information).

Many techniques are available for calculating estimates of rice paddy methane emissions, such as extrapolation of measured emission rates (Khalil et al., 1991;Khalil et al., 1993), statistical regression equations (Bachelet et al., 1995;Kern et al., 1995;Kern et al., 1997) and the application of models of varying complexity (Cao et al., 1995;Matthews et al., 2001;Van Bodegom et al., 2001;Huang et al., 1998;Li, 2000). Here we chose two regression models (Neue et al., 1990; Kern et al., 1997) and CH4MOD (Huang et al., 2004) in that they differed explicitly in levels of structural complexity. We compared the performance of these methods under different levels of data availability (Table 1) using experimental field measurements as a point of reference (Fig. 1). In Table 1, R1 represents a simple regression equation in which the carbon (C) input is the sole predictor (Neue et al., 1990). Regression equation R2 is slightly more complicated in that it uses organic C and fertilizer N application as inputs (Kern et al., 1997). We assumed two data availability scenarios for R2. In R2－S0, both the C and N inputs are available; in R2－S1, only the C input is available (Table 1).

The third approach consists of a semi-empirical model, CH4MOD. This model was developed to simulate methane emissions from rice paddies under diverse environmental conditions and various agricultural practices (Huang et al., 1998;Huang et al., 2004). The input variables of the model include the climate, soil conditions, water management type, organic matter application and crop rotations. The model consists of two modules: the derivation of methanogenic substrates from added organic matter and rice root exudates and the production and emission of methane. Rice biomass is a key variable used to calculate the root exudates and the fraction of the methane emitted by rice plants and bubbles. The daily changes in the soil redox potential (Eh) were calculated according to various water manipulations conducted in the rice paddies (Xie et al., 2010b). The influences of other environmental factors, such as soil temperature and texture, on the decomposition of organic matter and the production of methane were expressed as specific coefficient functions (Huang et al., 1998). The input variables of the CH4MOD model (Appendix B in the Supporting Information) include the daily air temperature, soil sand percentage (*SAND*), organic matter amendment (*OM*), rice grain yield (*GY*), water management pattern (W_{ptn}) and rice cultivar index (*VI*).

Four model input scenarios (Table 1) were scheduled to evaluate the performance of CH4MOD under different levels of data availability. In M－S0, all of the model variables were assigned specific values. In M－S1, the application of organic matter was assigned the average value for all experiments, thus assuming a situation where no detailed information on organic matter application was available. In M－S2, detailed information on the water regime and soil properties was assumed to be unavailable. In M－S3, detailed information on all three major factors (organic matter application, soil properties and water regime) was assumed to be unavailable.

The estimation residuals (Δy, Equation 1), relative bias (r_b, Equation 2) and coefficient of variations (r_v, Equation 3) were thus evaluated as follows:

$$\Delta y_k = \hat{y}_k - y_k, \qquad i = 1,2,...,n \tag{1}$$

$$r_b = \frac{E(\Delta y)}{E(y)} \times 100\% \tag{2}$$

$$r_v = \frac{\sqrt{E((\Delta y)^2) - (E(\Delta y))^2}}{E(y)} \times 100\% \tag{3}$$

where y represents the measured methane fluxes; $\hat{y}$ is the estimate of y; and n is the total number of measurements. *E( )* indicates the statistical mean.. The mean of the squared errors (MSE) of the estimation is calculated as follows:

$$\begin{aligned} MSE &= E((\Delta y)^2) = (E((\Delta y)^2) - (E(\Delta y))^2) + (E(\Delta y))^2 \\ &= (F \times r_v)^2 + (F \times r_b)^2 \end{aligned} \tag{4}$$

where $F = E(y)$ represents the mean of the measured methane fluxes (y_k).

2.3 Uncertainties in estimating rice paddy methane emissions on national scales: data error and availability

In addition to model fallacy, the difficulties in estimating national rice paddy methane emissions also stem from errors in, and limited availability of, input data. To measure the uncertainties in model outputs due to insufficient data quality and availability, we applied Monte Carlo simulations (Penman, 2000) to the CH4MOD model. Statistical characteristics were derived from the available datasets to develop probability distribution functions (PDFs) for each model input variable (Table B1, B2 of Appendix B in Supporting Information). We performed Monte Carlo simulation in the way of randomly drawing values of the model input variables from their PDFs and then run the model. This process iterated 1000 times and at the last step, the mean and 95% CI of the calculated methane fluxes were derived from the iterations.

The factors involved in the uncertainty analysis included organic matter application, soil properties and water regimes; these variables (*OM*, *SAND*, and W_{ptn}) were parameterized as input variables in the CH4MOD model (Huang et al., 2006;Zhang et al., 2011). The other two model input variables were the rice grain yield and daily ambient air temperature. These two variables were not used in the uncertainty analysis because sufficient relevant data were available, which were characterized by less error compared with the other variables (Zhang et al., 2014).

The *SAND* data were obtained from a 10 km × 10 km grid dataset interpolated from soil survey data (Oberthür et al., 1999;Shi et al., 2004;Liu et al., 2006). It is possible that approximately half (Van Bodegom et al., 2002b) of the immense spatial variation in soil properties can be lost after spatial interpolation (Goovaerts, 2001); as a result, the missing spatial variation was attributed to the PDF of the gridded *SAND* data (Appendix B in Supporting Information).

The organic matter inputs in the rice fields consisted of various types of farm manure (green manure and animal feces), crop straw, and dead roots and stubble leftover from previous harvests. Root and straw biomass were calculated using the root/shoot ratio and harvest indices (Huang et al., 2007;Gao et al., 2002;Xie et al., 2010c). Stubble was assumed to represent one-tenth of the straw biomass (Huang et al., 2004). The proportions of incorporated straw and applied farm manure were derived from data obtained from two large-scale investigations, the First National Census of Pollution Sources conducted by China's Ministry of Environmental Protection (CFPC, 2011) and census conducted by the Institute of Atmospheric Physics, Chinese Academy of Sciences. The proportion of straw and the amount of manure incorporated into the crop fields were summarized by province. Table B1 shows the statistical parameters of the PDF of organic matter incorporation in each province.

The irrigation in rice cultivation were grouped into five general irrigation patterns: 1) flooding-drainage-flooding-intermittent irrigation, 2) flooding-drainage-intermittent irrigation, 3) flooding-intermittent irrigation, 4) continuous flooding and 5) continuously intermittent irrigation (Gao and Li, 1992; Huang et al., 2004). Data pertaining to W_{ptn} were only very rarely available on a regional scale. The limited information provided in a few studies (Mao, 1981;Liang, 1983;Xiong et al., 1992;Cai et al., 2003;Ma et al., 2005;Ministry of Water Resources and Utilization of China (MWRUC), 1996) could only yield rough estimates related to irrigation in regions of major rice cultivation. The PDF of field irrigation were defined by the occurrence percentage of each irrigation pattern (Table B2). (Table B2 in the supporting information).

The data pertaining to the rice grain yield and harvesting area as of 2005 were obtained from China's Statistical Yearbook (EBCAY, 2006) and the nation's agricultural database maintained by the Chinese Academy of Agricultural Sciences, respectively. The spatial distributions of all rice paddies in 2005 and the rice paddy area within each 1 km × 1 km grid were obtained from the Data Center for Resources and Environmental Sciences of the Chinese Academy of Sciences (RESDC, CAS). Daily mean air temperature data from 678 meteorological stations throughout China for 2005 were acquired from the National Meteorological Information Center (NMIC) of the China Meteorological Administration (CMA) (http://cdc.cma.gov.cn/). The temperatures were then spatially interpolated into 10 km × 10 km grids for each day according to the method described by Thornton et al. (1997). Details on the datasets used in this study can be found in Appendix B.

To preserve details related to spatial variations, all data input into the model were converted into 10 km × 10 km grids. The applied rasterization techniques and details of how the model was run on raster datasets were provided in previously published papers (Huang et al., 2006).

2.4 Combining uncertainty and spatial aggregation

In each 10 km × 10 km grid, the uncertainties in our estimates originated from both the model fallacy (Equation 4) and error in the input data. Equation 5 was used to merge the two uncertainty sources where MSE was again split into two parts as showed in Equation 4:

$$\begin{aligned}{\sigma_{T,i}}^2 &= \sigma_{b,i}^2 + \sigma_{v,i}^2 + \sigma_{d,i}^2 \\ &= (F_i \times r_b)^2 + (F_i \times r_v)^2 + {\sigma_{d,i}}^2\end{aligned} \tag{5}$$

where $\sigma_{T,i}$ represents the uncertainty of the methane flux in grid i, and F_i and $\sigma_{d,i}$ represent the mean and standard deviation of the Monte Carlo simulation results in grid i, respectively. r_b and r_v represent the same entities as in Equations 2 and 3. ${\sigma_{d,i}}^2$ signifies the uncertainty caused by the error and availability of data, $(F_i \times r_b)^2$ represents the modelling bias, and $(F_i \times r_v)^2$ represents the rest parts of the model fallacy apart from $(F_i \times r_b)^2$. To produce the uncertainty of the national inventory, the three components ($(F_i \times r_b)^2$, $(F_i \times r_v)^2$ and ${\sigma_{d,i}}^2$ in Equation 5) of the estimation uncertainties in all grids were separately aggregated (Equation D2, D3, D4 and D5 in Appendix D of the Supporting Information) and summed as follows:

$$ {\sigma_T}^2 = \sigma_b^2 + \sigma_v^2 + \sigma_d^2 \tag{6} $$

3 Results

3.1 Methane emissions and the uncertainties derived from field measurements

Among the 495 methane flux measurements (the accumulative methane emission from transplanting to harvesting), 184 (37% of all cases) came from paddies that were continuously flooded during the entire rice growing period; 50 (10% of all cases) came from paddies with single mid-season drainage; and 261 (53% of all cases) came from paddies under multi-drainage. The average methane fluxes associated with the three water regimes were 531.6 ± 512.6, 251.6 ± 231.1 and 224.1 ± 207.5 kg CH_4 ha^{-1}, respectively (Fig. 3a). The overall arithmetic average of the 495 measurements (represented hereafter by m_c) was 341.2 ± 383.2 kg CH_4 ha^{-1}. However, the simple arithmetic average might be a biased representation of the “true” mean methane flux of rice paddies in China, as far less than 37% of the rice paddies in China are continuously flooded. In the literature, 10%, 20% and 70% of the rice area was reported to be under continuous flooding, single drainage and multi-drainage water regimes, respectively (Xiong et al., 1992;Ministry of Water Resources and Utilization of China (MWRUC), 1996), and the harvested-area-weighted mean (Appendix A in the Supporting Information) of the measured fluxes (represented hereafter by m_w) was 260.4 ± 281.6 kg CH_4 ha^{-1} (Fig. 3a).

The 95% confidence intervals (CIs) of the methane flux measurements were 61.1–2,145.9 kg CH_4 ha^{-1}, 9.6–809.9 kg CH_4 ha^{-1} and 14.0–797.7 kg CH_4 ha^{-1}, respectively, for the three water regimes (continuous flooding, single drainage, and multi-drainage in Fig. 3a). The 95% CI of all combined area-weighted measurements (Appendix A in the Supporting Information) was 13.7–1,115.4 kg CH_4 ha^{-1}. The measurements were not normally or symmetrically distributed (Fig. 3b). The P-P plots (Fig. 4) showed that the parameterized gamma distributions matched the sample distributions. The 95% CIs calculated with the parameterized gamma functions were 16.8–1,900.8 kg CH_4 ha^{-1}, 10.4–863.4 kg CH_4 ha^{-1} and 8.9–774.2 kg CH_4 ha^{-1}, respectively, for the three water regimes; these values overlapped the CIs derived directly from the measurements by 88.2%, 99.9% and 97.0%, respectively.

The national methane emissions from rice agriculture calculated by multiplying the rice harvesting area (yearbook data in 2005) by the area-weighted mean flux (260.4 ± 281.6 kg CH_4 ha^{-1}) were 7.51 Tg CH_4 (Fig. 3a). When the measurements are statistically independent, the standard error (SE) of the summation is $n-1$ (n is the sample size of the measurements) times smaller than the standard deviation (± 281.6 kg CH_4 ha^{-1}), which consists of the representative and measurement errors of the measured fluxes (Van Bodegom et al., 2002a;Verburg et al., 2006). Assuming that the measurements were statistically independent, the 95% CI of the national inventory was 7.20–8.58 Tg CH_4 (Equation A3 in the Supporting Information). However, the independency assumption is questionable because of the spatial correlations between the spatially correlated background environmental conditions and agricultural activities (Legendre, 1993;Dormann et al., 2007). The equivalent sample size used to calculate SE may be smaller than 495, and the 95% CI of the national inventory is therefore larger than that with the independency assumption.

3.2 Model performance under different situations of data availability

The averaged bias of the estimate obtained with R1 was 212.0 kg CH_4 ha^{-1} (Table 2) or 62.1% of the measured mean (m_c = 341.2 kg CH_4 ha^{-1}). The average bias of R2, in contrast, was −1.3 kg CH_4 ha^{-1}. R1 was more likely to overestimate the amount of methane emitted than R2 (Table 2), especially when more organic matter was incorporated (Fig. 5a). For example, in one case the modeled CH_4 flux was more than 6,000 kg CH_4 ha^{-1}, whereas the measured flux was less than 3,000 kg CH_4 ha^{-1} (Fig. 5a). The estimates obtained using R2 did not show significant variations and appeared to decline when the measured methane fluxes increased (Fig. 5b). The CH4MOD model also produced a small averaged bias, representing 7.1% of the measured mean. The MSE was 253.0, 407.8 and 596.0 kg CH_4 ha^{-1} for the M－S0, R2－S0 and R1－

S0 scenarios, respectively (Table 2), which demonstrates that model performance improves when more factors are incorporated into the model.

Although the CH4MOD model produced better simulation results than the simple regression equations, its performance fundamentally depends on data availability. When no case-specific data were available (as in scenario M—S3), r_b was −32.2%, and MSE was 122.1% of the mean flux; the results obtained under this scenario were even worse than the results obtained under the R2—S0 scenario (Table 2). For the M—S1 scenario, where the data pertaining to the soil properties and water regime were case-specific, the magnitude of r_b decreased to 9.0% of the mean flux, and the MSE decreased to 101.2% of the mean flux. The M—S0 scenario produced much better results than the other scenarios, as more data were available for the key model input variables (Table 2). Even no case-specific input data used in M—S3 had smaller r_b, r_v and MSE than R1—S0. In Table 2, larger r_v of R1—S0 than M—S3 might come from the too simple explanation of the influence from organic matter inputs on methane emission that added extra error on the estimation.

3.3 Inventory of rice paddy methane emissions and the uncertainties with different approaches

Because of the spatial heterogeneity in the climate, soil properties, organic matter incorporation and field irrigation in rice cultivation, the methane fluxes simulated by CH4MOD varied spatially between 17.2 kg CH_4 ha^{-1} and 708.3 kg CH_4 ha^{-1} from grid to grid (Fig. 6). The national means for the simulated methane fluxes were 217.9 kg CH_4 ha^{-1}, 204.6 kg CH_4 ha^{-1} and 255.8 kg CH_4 ha^{-1} for single, early and late rice cultivation, respectively. The within-grid estimation error ($\sigma_{T,i}$, calculated with Equation 5) represented 81.2%–95.5% of the mean fluxes F_i in the grids. In the present study, model fallacy, represented by $U_{b,i}+U_{v,i}$, contributed 79.5%–88.9% to the uncertainty $\sigma_{T,i}^2$, with $\sigma_{d,i}^2$ accounting for the remaining 11.1%–20.5%. This implies that a model with better performance is needed to reduce the uncertainty of $\sigma_{T,i}$ in each grid.

As shown in Fig. 7, the highest levels of emitted methane occurred in southern China, with the northeast also representing a major source of methane, despite this region being climatically cool. The total amount of methane emitted, as calculated by the M–

S0 approach, was 6.43 (3.79–9.77) Tg CH_4 yr^{-1} (Table 3), which is close to the 7.51 Tg CH_4 yr^{-1} derived from the experimental field measurements.

In Table 3, the estimated national CH_4 emissions ranged from 6.43 (3.79–9.77) Tg CH_4 yr^{-1} to 13.59 (1.45–38.98) Tg CH_4 yr^{-1} for the M–S0 scenario and R1–S0 scenario, respectively. The 95% CIs of the national estimation differed more greatly among the approaches than those among the data availability scenarios of each approach. With M–S0, The fallacy of CH4MOD contributed 56.6% of the total uncertainty, with the remaining 43.4% being attributed to errors and the scarcity of the spatial datasets of the model inputs (Table 4). As an indicator of the trade-off between the complexity of the approach and data availability, the σ_d/σ_{b+v} ratio in Table 3 was 0.87 for M–S0, closer to 1 than those for the other approaches and scenarios, which also yielded the narrowest 95% CI in Table 3.

4 Discussion

4.1 Contributions of different error sources to the uncertainties in the inventory

In the experimental field measurements (Fig. 1), the variations in rice paddy methane fluxes ranged from 3.2 kg CH_4 ha^{-1} to 2,451.7 kg CH_4 ha^{-1}, averaging 341.2 ± 383.2 kg CH_4 ha^{-1}. The average simulated methane fluxes in the 10 x 10 km grids varied from 17.2 to 708.3 kg CH_4 ha^{-1} (Fig. 6). The extremely high methane fluxes obtained from experimental measurements were not reproduced by the model estimations. This was partly due to the discrepancy in the spatial representativeness of the methane fluxes in field observations and model estimations (Verburg et al., 2006). The experimental measurements represented methane fluxes from an area of less than one hectare, while the modeled fluxes were the averages from 10 x 10 km grids. This mismatch in spatial representativeness might also be due to errors in the model input data as well as to the impacts of other unknown factors (Singh and Dubey, 2012;Bhatia et al., 2011;Zheng et al., 2010;Gauci et al., 2008). Methane emissions could be estimated using a limited number of factors and simplified equations to express the complex relationships between methane emissions and influential factors, but such simplification resulted in poor performance of the methods (Table 2). In Equation 5, $\sigma_{d,i}$ is the uncertainty due to errors in the input data. With an increasing number of explanatory factors, r_b and r_v might decrease (which means better performance of the method), but $\sigma_{d,i}$ might increase because of the cumulative errors resulting from the increasing number of factors incorporated in the models. To reduce uncertainties in the estimates and improve the performance of the model, the input data need to be available and of good quality.

The aggregated uncertainty of the national inventory depended not only on the magnitude of $\sigma_{v,i}$ and $\sigma_{d,i}$ in each grid (i) but also on the spatial correlation between these variables (Equation C2 in the Supporting Information). The spatial correlation of $\sigma_{d,\,i}$ depends on the availability of input data for the model and on spatial aggregation (Table C1 in the Supporting Information). However, the spatial correlation of $\sigma_{v,i}$ could not be assessed analytically because it was a result of model fallacy and errors in measurements. In the case of a strong correlation of $\sigma_{v,i}$ values, the aggregated σ_v^2 will account for a large proportion of σ_T^2 (right side in Fig. 8). However, if the spatial correlation is confined to a short distance, such as less than four grids (Dormann et al., 2007;Dray et al., 2006), the contribution of σ_v^2 to σ_T^2 will be negligible (left side in Fig. 8). At the mid-point of D_C (Equation C2, 30 grids, equal to 300 kilometers), as shown in Fig. 8, the model uncertainty (σ_r^2 + σ_v^2) accounted for 56.6% of the uncertainty in σ_T^2 (Table 4).

4.2 Consistency of errors between model validation and model up-scaling

Up-scaling a site-scale model (e.g., CH4MOD in this study) to a national scale poses enormous challenges when data are scarce. Enhancing the spatial abundance of the input data minimizes the propagation of data error into the aggregated uncertainties. Many environmental and agricultural factors impact methane emissions from rice paddies. In the CH4MOD model, the key factors were parameterized as model inputs (Huang et al., 2004). However, when assessing the uncertainty of a model, the explanatory variables are arbitrarily included (Verburg et al., 2006). Li et al. (2004) found that soil properties were the “most sensitive factor” and therefore used this parameter in the uncertainty analysis. The inclusion of as many of the highly sensitive key factors as possible in the uncertainty analysis should generate more accurate and reliable results (right part in Fig. 2).

Experimental field studies have shown that the rice variety has substantial impacts on methane emissions (Aulakh et al., 2008;Inubushi et al., 2011;Jia et al., 2002). A study of field observations (Su et al., 2015) showed that transfer of the barley gene *SUSIBA2* to rice favors the allocation of photosynthates to the aboveground biomass over allocation to the roots and, moreover, that less biomass allocation to root exudates results in reduced methane emissions. The impact that the rice variety has on methane emissions was parameterized as the variety index (*VI*) in CH4MOD. According to Huang et al. (1998), *VI* ranges from 0.5 to 1.5 and averages 1.0 for most rice varieties. To validate the CH4MOD model (left portion of Fig. 2) using the 495 methane emission measurements included in the present study, *VI* was assigned a default value of 1.0 regardless of the rice variety because, until now, no dedicated attempts have been made to quantify the *VI* of different rice varieties. Therefore, the r_b and r_v values presented in Table 2 incorporate the uncertainty in model performance that can be attributed to different rice varieties ($M_f(x)$ in Equation C2 of the Supporting Information). To maintain consistency, *VI* was assigned the same default value (1.0) when the model was scaled-up to the national scale (right side of Fig. 2), and no PDF was built for the uncertainty calculation conducted with the Monte Carlo simulation. If a PDF had been incorporated into the uncertainty calculation when the model was scaled-up, the overall uncertainties (Table 4) would have been overestimated. However, if different *VI* values were assigned to rice varieties during model validation, the error caused by the inaccuracy of *VI* would also need to be considered during the scaling-up of the model to prevent underestimation of the overall uncertainty.

5 Conclusion

Due to the remarkable spatial variation in rice paddy methane emissions, the uncertainties in national estimates obtained either through field measurements or modeling remain considerably large. For field measurements, the reduction in uncertainty achieved by increasing the number of observations was shown to be inversely related to the spatial correlation between the measurements. To reduce the estimation bias, the number of measured emission fluxes should be proportional to the paddy area where the corresponding agronomic activities and environmental conditions occur homogenously.

Model performance depends not only on the effectiveness of the models themselves but also on the availability of the data needed to drive the model. We found that without a sufficient quantity of high-quality data, a well-developed model may perform even more poorly than simple regression approaches. When modeling methane emissions, uncertainties in the performance of the model remain the major obstacle to reliably estimating methane emissions. Estimate uncertainty could be reduced at the national scale by increasing the availability of input data and decreasing spatial correlations among the residues of the model output.

Modelling by CH4MOD with all the available data, the national methane emission from rice paddies was 6.43 (3.79–9.77) Tg CH_4 yr^{-1} in China. Comparing to other options, balancing between the uncertainties caused by the model fallacy and data scarcity produced national estimations of least total uncertainty.

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Tables

Table 1 Methods and their input scenarios

Methods	Input scenario	Reference
R1: $C_{CH4} = 0.3 \times C_{input}$	R1 - S0: Case-specific C input, adjusted with the water regime[†].	Neue et al., (1990)
R2: $CH_4 = -0.006 \times C_{input} + 0.078 \times N_{input} + 0.885 \times R_{C/N} + 21.15$	R2 - S0: Case-specific C and N input.	Kern et al., (1997)
	R2 - S1: Case-specific C input, averaged N input in all cases.	
M: CH4MOD model	M - S0: Case-specific inputs of all model variables: e.g., organic matter amendments, soil properties and water regimes[‡]	Huang et al., (1998, 2004); Xie et al., (2010)
	M - S1: Case-specific inputs of soil properties and water regimes; other model variables use averaged values for all 495 cases	
	M - S2: Case-specific inputs of organic matter amendments; all other model variables use averaged values for all 495 cases, the water regime was assumed to be multi-drainage irrigation.	
	M - S3: No case-specific inputs used for soil properties or organic matter amendments, the water regime was assumed to be multi-drainage irrigation.	

† Regression equation R1 was developed according to measurements conducted in continuously flooded fields, and the calculated flux was therefore adjusted by a scaling factor of 1.00, 0.65 or 0.56 for continuous flooding, single drainage or multi-drainage irrigation, respectively (IPCC, 2006).

‡ The water regimes in the CH4MOD model (Huang et al., 2004) are more specifically defined and differ from that of the IPCC (2006).

Table 2 Performance of the methods under different scenarios of data availability

Method	Bias of the estimation (r_b)	Std. of the estimation residues (r_v)	root of MSE (RMSE)
R1－S0	212.0 (62.1%)†	577.1 (163.3%)	596.0 (174.7%)
R2－S0	−1.3 (−0.4%)	407.8 (119.5%)	407.8 (119.5%)
R2－S1	−4.9 (−1.4%)	415.7 (121.8%)	415.7 (121.9%)
M－S0	−24.2 (−7.1%)	251.8 (73.8%)	253.0 (74.1%)
M－S1	−30.8 (−9.0%)	343.9 (100.8%)	345.2 (101.2%)
M－S2	−120.7 (−35.4%)	341.3 (100.0%)	362.9 (106.1%)
M－S3	−109.8 (−32.2%)	401.8 (117.8%)	416.6 (122.1%)

† Percentages in parentheses indicate the magnitude of the error relative to the overall average methane flux (341.2 kg CH_4 ha^{-1}) for all cases.

Table 3 Methane emissions inventory and the uncertainties caused by model imperfection and errors in model input data

Scenario	CH_4 emission (Tg)	σ_{b+v}[§]	σ_d	σ_d/σ_{b+v}	σ_T (Tg)	95% CI (Tg)
R1－S0	13.59	9.89	1.11	0.11	9.99	1.45－38.98
R2－S0	10.37	2.74	0.14	0.05	2.74	5.71－16.39
R2－S1	10.24	2.91	0.07	0.02	2.91	5.83－17.16
M－S0	6.43	1.15	1.00	0.87	1.53	3.79 － 9.77
M－S1	7.94	1.89	0.97	0.51	2.13	4.33 －12.62
M－S2	7.40	3.16	0.56	0.18	3.12	2.56 －14.75
M－S3	9.23	3.79	0.00	0.00	3.79	3.37 －18.01

§ Root of U_b+U_v, uncertainty owing to model fallacy in the national inventory.

Table 4 Components of the uncertainty in the national inventory

Rice	Due to model performance		Due to data quality and availability,	Total	
	U_b	U_v	U_d	U_T	σ_T
Early rice	0.01	0.06(0.00−0.81) ‡	0.08	0.15	0.39
Late rice	0.01	0.10(0.00−1.28)	0.05	0.16	0.40
Single rice	0.07	0.25(0.00−5.15)	0.24	0.56	0.75
All rice	0.21	1.12(0.00−22.56)	1.00	2.35	1.53

‡ Numbers in parentheses represent the range of U_v depending on the spatial correlation of the model simulation residuals. Long-distance correlation results in a large aggregated U_v, whereas short-distance correlation results in a small aggregated U_v.

Figure Legends

Figure 1 Locations of the experimental sites (red stars). The background map represents the spatial distribution of rice paddies in China. The size of the red stars is proportional to the number of measured methane fluxes at the site. The polygons show zones of different crop rotation systems involving rice: I—Double rice rotation; II—Mixed zone of rice/rice rotation and rice/upland crop rotation; III & IV—Rice/upland crop rotation or rice/fallow rotation; V & VI—Rice/fallow rotation; and VII—No rice.

Figure 2 Flowchart for estimating regional/national methane emissions and the uncertainties associated with field measurements and modeling

Figure 3 Statistical representations of the measured methane fluxes. (a) Statistical parameters and (b) histogram of the measurements. The solid circles represent the sample mean, and the vertical bars are the 95% confidence intervals of the samples, from the 2.5% percentile to the 97.5% percentile. The dashed line indicates the arithmetic average of all measured fluxes (m_c). The solid line is the area-weighted mean of the methane fluxes (m_w), in reference to the areal proportion of each water regime in the national total rice harvesting area: 10% continuous flooding (Flooded), 20% single drainage (Single-D) and 70% multi-drainage (Multi-D) (Xiong et al., 1992; MWRUC, 1996;Li et al., 2001;Zou et al., 2009).

Figure 4 P-P plots of the cumulative probability of the measured methane fluxes versus the gamma distribution. (a) Single drainage irrigation cases, (b) multi-drainage irrigation cases, (c) continuous flooding irrigation cases, and (d) all cases after being area weighted (Appendix A). *n*, *avg.* and *std.* represent the sample size, statistical mean and standard deviation of the sample methane fluxes, respectively. α and β represent the shape and scale parameters of the gamma distribution, which were calculated with the statistical mean and variance of the measured methane fluxes; $\beta = (std.)^2/(avg.)$, and $\alpha = (avg.)/\beta$. The diagonal line is the 1:1 straight line for a perfect gamma distribution match.

Figure 5 Methane fluxes in the experiments plotted against the respective simulation results through different methods. (a) R1—S0, (b) R2—S0 and (c) M—S0, which are described in Table 1.

Figure 6 Histograms and their fitting gamma probability lines for the calculated methane fluxes (via CH4MOD) of the 10 km × 10 km rice paddy grids in China. (a) Single rice rotations, including rice-fallow rotations, and rotations of rice with upland crops; (b) early and (c) late rice in double rice rotations. The vertical bars are the histograms of the calculated F_j (Equation 5), and the solid line is the theoretic gamma PDF line, the parameters for which were derived from the statistics for F_j via momentum methods.

Figure 7 Spatial distributions of rice paddy methane emissions ($\times 10^6$ g CH_4 per 10 km × 10 km grid).

Figure 8 Composition of the aggregated uncertainty of the national inventory along with the spatial autocorrelation of the variances of the model residues in grids. Distance criteria (D_c) are used to define the step functions of spatial autocorrelation: if two grids diverge by a distance beyond D_c, the autocorrelation is 0; otherwise, it is 1. The step function is a simplified version and represents the upper limit of the true spatial autocorrelation. With the step function, a larger D_c indicates stronger autocorrelation.

Figures

Figure 1

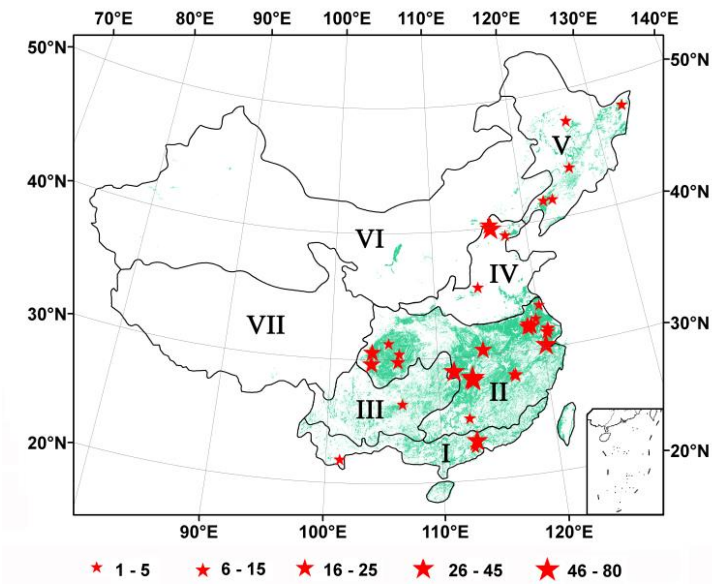
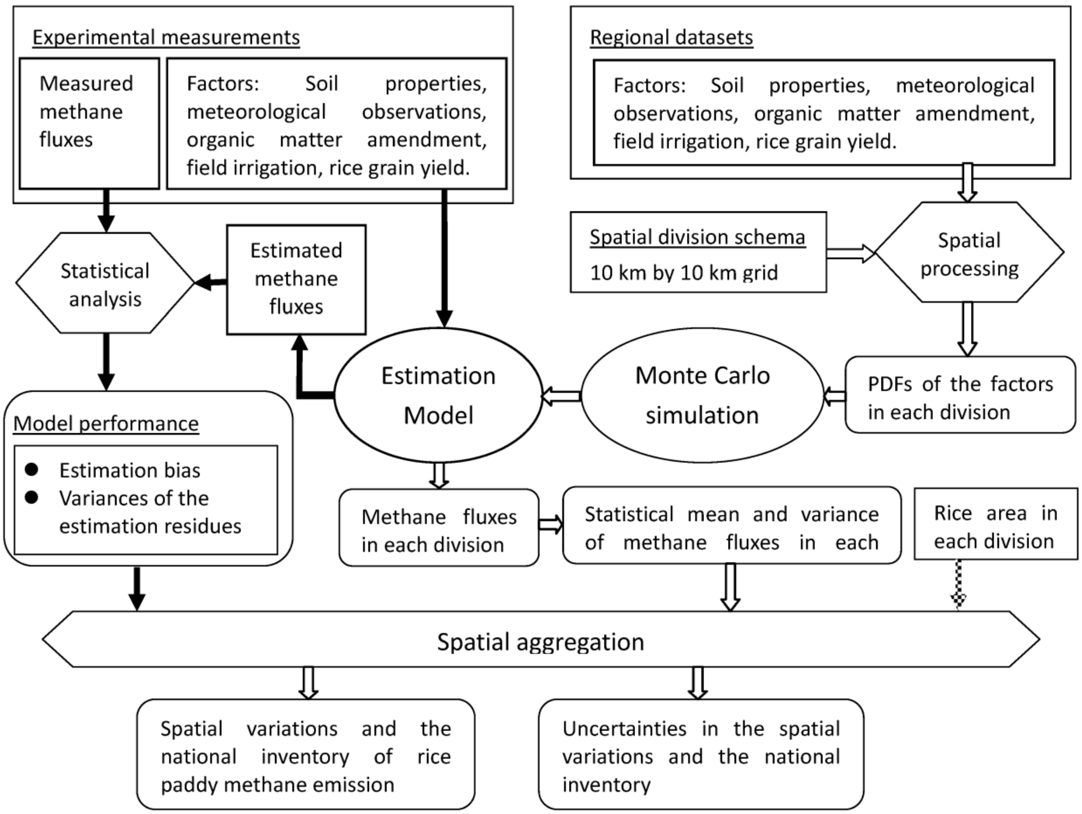


Figure 2



Experimental measurements
Measured methane fluxes
Factors: Soil properties, meteorological observations, organic matter amendment, field irrigation, rice grain yield.
Regional datasets
Factors: Soil properties, meteorological observations, organic matter amendment, field irrigation, rice grain yield.
Statistical analysis
Estimated methane fluxes
Spatial division schema
10 km by 10 km grid
Spatial processing
Estimation Model
Monte Carlo simulation
PDFs of the factors in each division
Model performance
Estimation bias
Variances of the estimation residues
Methane fluxes in each division
Statistical mean and variance of methane fluxes in each
Rice area in each division
Spatial aggregation
Spatial variations and the national inventory of rice paddy methane emission
Uncertainties in the spatial variations and the national inventory

Figure 3

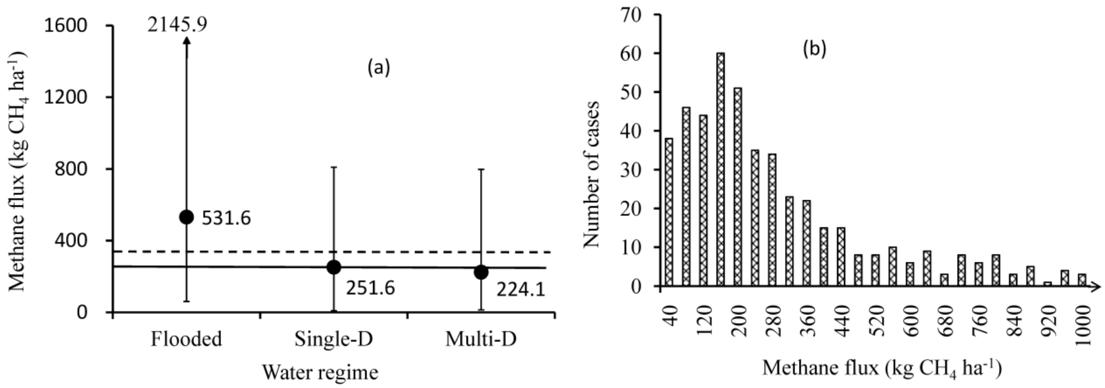

(a)
Methane flux (kg CH_4 ha^{-1})
1600
1200
800
400
0
2145.9
531.6
251.6
224.1
Flooded
Single-D
Multi-D
Water regime
(b)
Number of cases
70
60
50
40
30
20
10
0
40
120
200
280
360
440
520
600
680
760
840
920
1000
Methane flux (kg CH_4 ha^{-1})

Figure 4

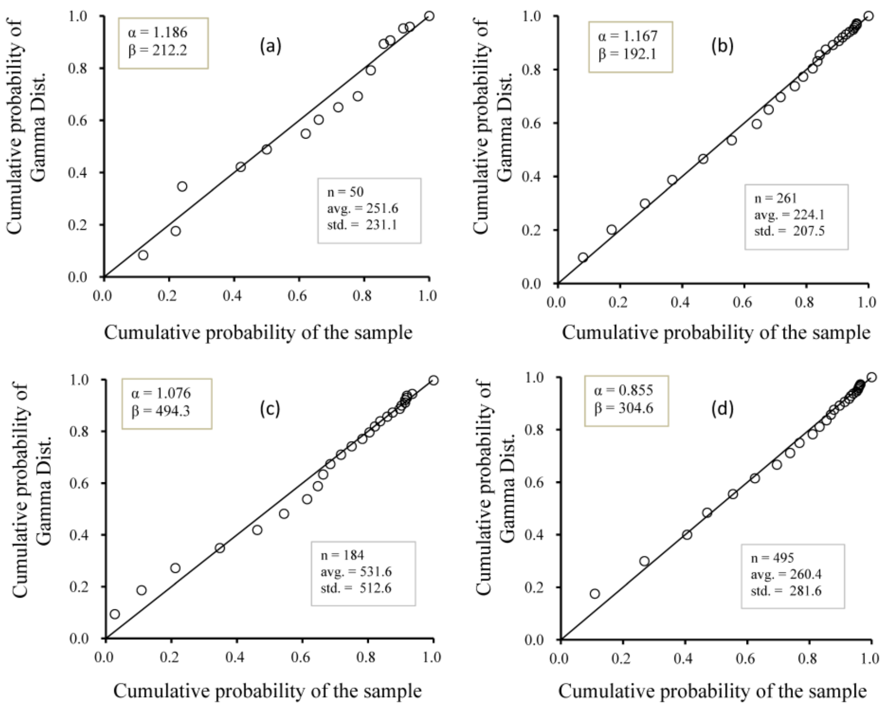

(a)
α = 1.186
β = 212.2
n = 50
avg. = 251.6
std. = 231.1
(b)
α = 1.167
β = 192.1
n = 261
avg. = 224.1
std. = 207.5
(c)
α = 1.076
β = 494.3
n = 184
avg. = 531.6
std. = 512.6
(d)
α = 0.855
β = 304.6
n = 495
avg. = 260.4
std. = 281.6
Cumulative probability of Gamma Dist.
Cumulative probability of the sample
0.0
0.2
0.4
0.6
0.8
1.0

Figure 5

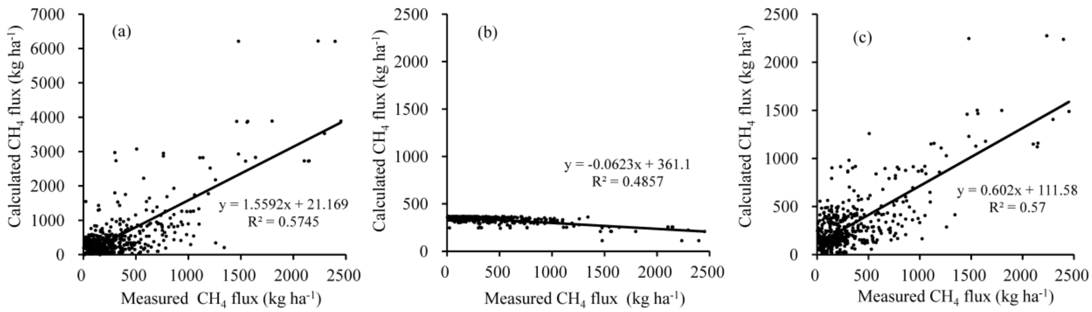

(a)
Calculated CH4 flux (kg ha-1)
Measured CH4 flux (kg ha-1)
y = 1.5592x + 21.169
R² = 0.5745
(b)
Calculated CH4 flux (kg ha-1)
Measured CH4 flux (kg ha-1)
y = -0.0623x + 361.1
R² = 0.4857
(c)
Calculated CH4 flux (kg ha-1)
Measured CH4 flux (kg ha-1)
y = 0.602x + 111.58
R² = 0.57

Figure 6

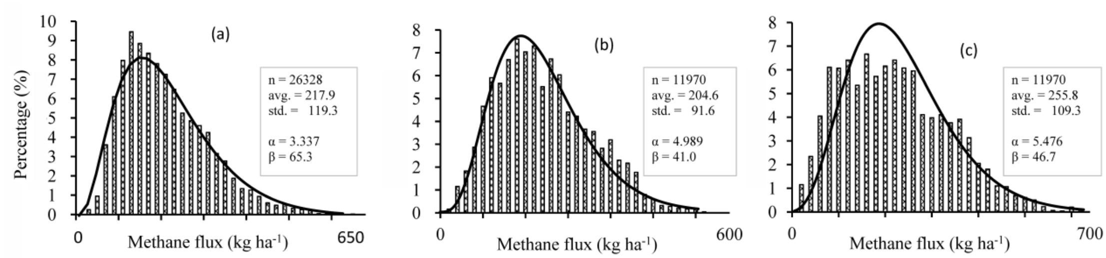

(a)
Percentage (%)
Methane flux (kg ha-1)
0
650
n = 26328
avg. = 217.9
std. = 119.3
α = 3.337
β = 65.3
(b)
Methane flux (kg ha-1)
0
600
n = 11970
avg. = 204.6
std. = 91.6
α = 4.989
β = 41.0
(c)
Methane flux (kg ha-1)
0
700
n = 11970
avg. = 255.8
std. = 109.3
α = 5.476
β = 46.7

Figure 7

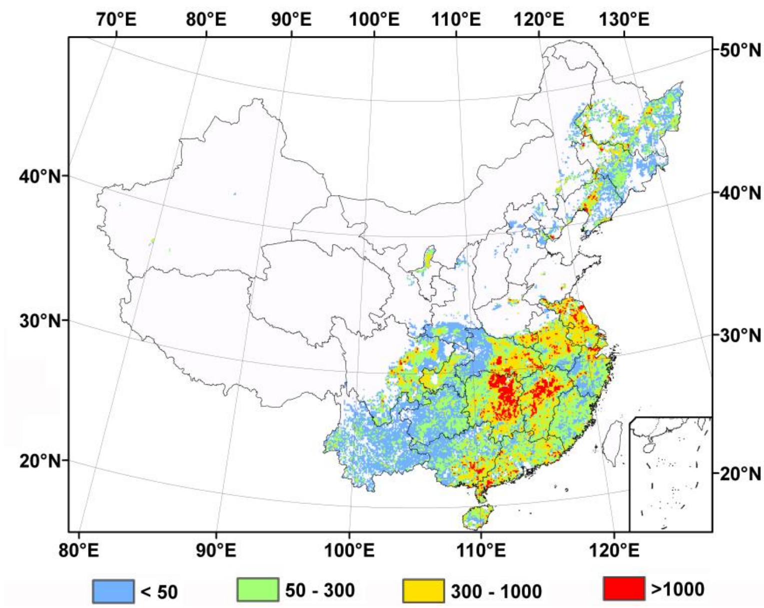
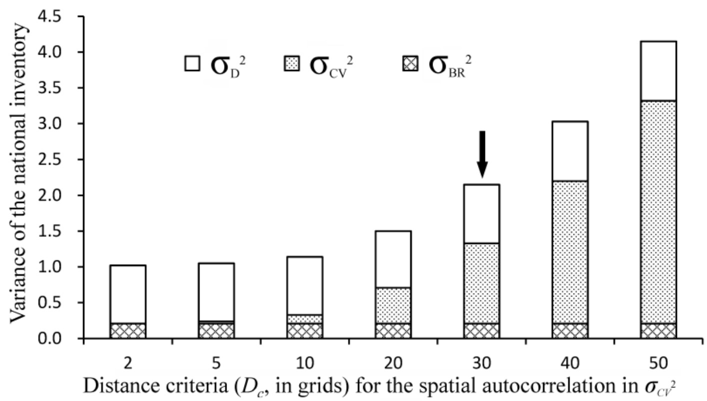

70°E
80°E
90°E
100°E
110°E
120°E
130°E
50°N
40°N
30°N
20°N
80°E
90°E
100°E
110°E
120°E
< 50
50 - 300
300 - 1000
>1000

Figure 8


σD2
σCV2
σBR2
Variance of the national inventory
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
2
5
10
20
30
40
50
Distance criteria (Dc, in grids) for the spatial autocorrelation in σCV2