Supplement of Biogeosciences, 14, 163–176, 2017
http://www.biogeosciences.net/14/163/2017/
doi:10.5194/bg-14-163-2017-supplement

Biogeosciences Open Access

Supplement of

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

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Section S1. Area-weighted methane flux measurements

For the area-weighted analysis, the mean (m_w) and variances (V_w) of the methane flux measurements (O_i) were calculated as:

$$m_w = \sum_{i=1}^{N} w_i \times O_i \tag{S1}$$

$$V_w = \sum_{i=1}^{N} w_i \times (O_i - m_w)^2 \tag{S2}$$

constrained by $\sum_{i=1}^{N} w_i = 1$, where N is the total number of all methane flux measurements. Ordinarily, $w_i = \dfrac{1}{N}$. However, here, we scaled w_i by the area proportional to that of each water regime applied in rice cultivation in China:

$$w_{wr} = \frac{R_{wr}}{N_{wr}}, wr \in \{continuously\ flooding,\ single\ drainage,\ multi-drainage\} \tag{S3}$$

where N_{wr} is the number of measurements belonging to water regime wr, and R_{wr} is the proportion of the area of rice paddies irrigated with each of the three water regimes. R_{wr} assumes values of 0.1, 0.2 and 0.7, respectively, for the three water regimes according to previous research (Mao, 1981; Xiong et al., 1992;Li, 2002;Zou et al., 2009). If a methane flux measurement (i) in Equations S1 and S2 belongs to water regime wr, then $w_i = w_{wr}$.

The standard error (SE) of the area-weighted mean (m_w) is calculated as follows:

$$SE = \sqrt{\frac{V_w}{N-1}} \tag{S4}$$

It should be noted that Equation S4 only holds when the measurements are statistically independent; if this is not the case, mostly due to spatial correlations of the environmental conditions that support the measurements, then the value for N should be smaller, depending on the strength of the correlation (Bence, 1995).

Section S2. CH4MOD model and the datasets used for simulating national rice paddy methane emissions

CH4MOD is an empirical model that simulates methane production and emissions from rice paddies under various environmental conditions and agricultural practices (Huang et al., 1998, 2004; Xie et al., 2010). It calculates methanogenic substrate production from rice plant root exudates and added organic matter (OM) decomposition. Both OM decomposition and rice plant-induced substrate production are significantly influenced by environmental factors, including the soil texture and temperature, with the soil moisture content controlling the fraction of transformation of the substrates into methane. The amount of the substrate derived from rice root exudate was simulated by a power function of the rice biomass, scaled by the parametric influence of the soil context and the rice cultivar. The substrate derived from the added organic matter was calculated by a first-order kinetic decomposition equation of the organic matter in soil, also scaled by the parametric influence of the soil context and the temperature. Details can be found in Huang et al (2004). There are two major routes by which methane produced in rice paddy soils escapes into the atmosphere: via the arenchyma system of the rice plants and via methane bubbles. Both of these pathways are incorporated into the model.

CH4MOD runs in a daily step, driven by the daily air temperature. Its input parameters include the soil sand percentage (*SAND*), organic matter amendment (*OM*), rice grain yield (*GY*), water management pattern (W_{ptn}) and rice cultivar index (*VI*).

Rice harvest area and grain production

Data on rice production and the harvest area for each province in 2005 were obtained from China's Statistical Yearbook (EBCAY, 2006) for early, late and middle rice. County-level rice production data were obtained from censusing conducted by the Chinese Academy of Agricultural Sciences. Although the fractions of early, late and single rice cultivation are not included in the county-level data, the rotation type for each county was formulated using the approach of Frolking et al. (2002) by referring to the climatic zones of each cropping system in China (Han et al., 1987).

Several studies have shown that methane emissions differ significantly among rice varieties (Singh et al., 1997; Wang et al., 1999). In CH4MOD, the impact of the methane variety on methane emissions was parameterized as the variety index (*VI*) (Huang et al., 1998, 2004). *VI* ranges from 0.5 to 1.5 but is typically approximately 1.0 for most rice varieties (Huang et al., 1997, 2004).

Climate data and rice phenologies

The daily mean air temperature is the only meteorological data required to drive the CH4MOD model. Air temperature data were obtained for 2005 from 678 meteorological stations included in the National Meteorological Information Center (NMIC), China Meteorological Administration (CMA) (http://data.cma.cn/) database. For counties that lack a meteorological station, air temperature data from the nearest neighboring station were substituted.

Rice phenologies (specifically transplanting and harvesting dates) control the start and end of the CH4MOD run for simulating methane emissions. Data regarding rice phenologies were originally derived from iso-line maps edited by Zhang et al. (1987) in the Agricultural Climate Atlas of China. The transplanting and harvesting dates within each grid were spatially interpolated from the iso-lines via the TIN (triangular irregular network) technique (Aumann et al., 1991) and assigned to each county.

Soil properties

Soil properties have extremely high spatial variation and may vary largely from one place not far from another. We obtained the data from Institute of Soil Sciences, Chinese Academy of Sciences. They collected more than 7000 soil profile measurements sampled during during the Program of the Second Soil Survey of China and subsequent surveys from 1980s to the present, and linked them to the a soil database of 1:1,000,000 scale (Shi et al., 2004) to produce the gridded data of soil properties with geostatistical methods. The database comprises of 10 km × 10 km raster datasets of soil properties at 10-cm intervals from the surface down through the profile, making the spatial resolution of soil the finest of the CH4MOD input parameters. We compared the spatial variation explained in the gridded datasets of soil properties against the variations in the profile measurements to analyze the ‘missing spatial variation’ (Bodegom et al., 2002b). The ‘missing spatial variation’ is the proportion of spatial variation of the soil properties (the sand content of the surface soil layer in the present study) that were not accounted for by the gridded datasets. We used the missing variation to build the PDF of SAND in Monte Carlo simulation by assuming normal distributions of the missing variation.

Organic matter amendment and water regimes in rice paddies

The organic matter inputs into rice fields include various types of farm manure (e.g., green manure, animal feces) and crop straw as well as dead roots and stubble from previous crops. Root biomass remaining in the soil can be calculated using the root/shoot ratio (Huang et al., 2007). Stubble biomass was assumed to be one-tenth the aboveground straw biomass. However, the fractions of straw incorporation and farm manure application are not well known, and the data are therefore limited. In the First National Census of Pollution Sources conducted by the Ministry of Environmental Protection of China (CFPC, 2011), straw application in croplands was summarized at the provincial level in the census data (Table S1); thus, the value for straw application given in Table S1 is not rice specific but accounts for all crops in each province. This bias may not be significant in provinces where crop cultivation is dominated by rice. In addition to crop straw, the incorporated crop residues include dead crop roots and stubble; according to Zhao and Li (2001), stubble accounts for approximately 13% of the total dry weight of straw.

No regular statistical data or comprehensive census data were available for manure application in rice cultivation. In this study, we estimated OM application in rice cultivation by examining more than 1000 research papers; estimates of farmyard manure application in each province are shown in Table S1.

Since the mid-1960s, a diverse array of irrigation regimes have been adopted that diverge from the traditional approach of continuous flooding, representing an important development for rice cultivation in China (Xiong et al., 1992; Li, 2002; Peng et al., 2007). As such, different compositions of flooding, drainage and moisture irrigation have been applied according to the climate, soil and topographic conditions of the rice fields and factors such as the rice variety being grown, its developmental stage and hydrological construction. To simplify CH4MOD, the forms of irrigation used for 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). Despite being the agronomic factor that is most sensitive to methane emissions (Table S2), the available data on irrigation are the scarcest among all of the inputs needed for CH4MOD up-scaling. Except for a few brief mentions in the literature (Mao, 1981; Xiong et al., 1992; MWRUC, 1996; Cai, 2000; Ma et al., 2005), almost no detailed data addressing spatial variations in rice irrigation are available. Given this limitation, we made rough assumptions about irrigation for each grand region of rice cultivation (Fig. 1, Table S2).

Table S1 Fraction of straw incorporation and farm manure application in rice cultivation£

Province	Fraction of straw incorporation†	Farm manure (kg OM ha^{-1}) ‡		Province	Fraction of straw incorporation	Farm manure (kg OM ha^{-1})	
		Mean	Range			Mean	Range
Beijing	0.41	821.6	321.6—1321.6	Henan	0.56	1539.2	776.2—2302.1
Tianjin	0.29	927.4	123.1—1731.6	Hubei	0.20	2101.3	981.1—3221.6
Hebei	0.62	1519.3	959.5—2079.1	Hunan	0.34	1836.9	846.7—2827.2
Shanxi	0.44	1824.8	1195.5—2454.2	Guangdong	0.23	1243.2	634.5—1851.8
Inner Mon.	0.12	1837.5	1042.4—2632.7	Guangxi	0.27	1384.7	645.4—2124.1
Liaoning	0.03	1108.5	657.8—1559.3	Hainan	0.22	1408.5	964.8—1852.1
Jilin	0.03	1308.4	421.5—2195.4	Chongqing	0.17	1608.7	801.5—2415.8
Heilongjiang	0.23	1800.8	836.0—2765.6	Sichuan	0.18	1922.7	940.7—2904.7
Jiangsu	0.23	1263.5	605.6—1921.4	Guizhou	0.09	1793.2	740.2—2546.1
Zhejiang	0.35	1276.2	734.1—1818.3	Yunnan	0.10	1802.3	853.1—2751.5
Anhui	0.19	1507.5	424.3—2590.7	Shaanxi	0.34	1769.6	555.3—2983.9
Fujian	0.32	1123.1	852.6—1393.6	Gansu	0.03	1923.0	375.9—3470.1
Jiangxi	0.38	1612.2	842.3—2382.1	Ningxia	0.15	1448.6	515.5—2381.7
Shandong	0.55	1032.8	530.8—1534.7	Xinjiang	0.45	1612.0	407.7—2816.3

£ No data on farm manure application were available for Shanghai and Tibet; as such, data for Jiangsu and Guizhou, respectively, were used as substitutes.

† Statistics derived from the First National Pollution Source Census conducted by the Ministry of Environmental Protection of China (CFPC, 2011); however, the range of variation was not provided in the publication.

‡ Statistics derived from an investigation of organic application in crop cultivation performed by the Institute of Atmospheric Physics, Chinese Academy of Sciences. Green manure was not included because it accounts for a minor proportion of the total organic matter application in rice cultivation.

Table S2 Proportions of different water irrigation patterns† in each grand region

Grand region‡	Baseline fraction	Uncertainty fraction
I	3: 0.92; 4: 0.08£	1: 0.31; 2: 0.31; 3: 0.30; 4: 0.08
II	2: 0.95; 4: 0.05	1: 0.32; 2: 0.32; 3: 0.31; 4: 0.05
III	2: 0.82; 4: 0.18	1: 0.27; 2: 0.28; 3: 0.27; 4: 0.18
IV	1: 1.0	1: 0.34; 2: 0.33; 3: 0.33
V	1: 1.0	1: 0.34; 2: 0.33; 3: 0.33

† Refer to Huang et al. (2004) for the definition of water irrigation patterns

‡ Grand region I: Guangdong, Guangxi, Hainan, Hunan and Jiangxi; Grand region II: Fujian, Hubei, Zhejiang, Jiangsu, Shanghai and Anhui; Grand region III: Chongqing, Sichuan, Yunnan and Guizhou; Grand region IV: Heilongjiang, Liaoning and Jilin; Grand region V: other provinces.

£ Indicates that water irrigation pattern 3 was applied in 92% of the rice cultivation area in Grand region I (Fig. 2a), and the remaining 8% of the rice area was under continuous flooding (water irrigation pattern 4).

Section S**3** Uncertainties in regional estimates obtained via the modeling approach

$F(x)$ is a spatial process that has a determinative component, $A(x)$, and a random component, $e(x)$:

$$F(x) = A(x) + e(x) \tag{S5}$$

where x is the location of two dimensions in the spatial domain, D.

When $A(x)$ is implemented in a model $M(x)$ that simulates the spatial variation of $A(x)$, there is unavoidably an error component, $M_f(x)$ (the model fallacy), due to the imperfection of the model, and therefore

$$A(x) = M(x) + M_f(x) \tag{S6}$$

Combining $M_f(x)$ and $e(x)$ into one component, $e_m(x)$, $F(x)$ this expression can be rewritten as

$$F(x) = M(x) + e_m(x) \tag{S7}$$

where $e_m(x)$ is typically used to evaluate model performance. To explicitly address the model input variables, e.g., environmental factors and anthropogenic activities of the model mechanism, $M(x)$ can be expressed as

$$F(x) = M(v_1,v_2,v_3,\ldots) + e_m(x) \tag{S8}$$

where v_1, v_2, v_3, … are the model input variables. Averaging over the spatial domain D, Equation S8 yields:

$$\overline{F(x)} = \overline{M(v_1,v_2,v_3,\ldots)} + \overline{e_m(x)} \tag{S9}$$

To implement the averaging of the model simulation over the spatial domain, the theoretical approach is

$$\overline{M(v_1,v_2,v_3,...)} = \frac{\int_D M(v_1,v_2,v_3,...)dx}{D} \quad \text{(S10)}$$

However, because it is impossible to obtain data for the model input variables at every location x of the domain D, $\overline{M(v_1, v_2, v_3,...)}$ has to be represented by the model simulation at a specific location p, $M(v_1^{(p)}, v_2^{(p)}, v_3^{(p)},...)$, and there emerges the representative error, e_s, which conforms to

$$\overline{M(v_1,v_2,v_3,...)} = M(v_1^{(p)},v_2^{(p)},v_3^{(p)},...) + e_s \quad \text{(S11)}$$

The representative error e_s comes from both the error and poor spatial availability of the model inputs over the spatial domain. The magnitude of e_s therefore depends on the model input errors and how sensitive the model simulation is in response to the variation of the model inputs (Zhang et al. 2014). Combining Equations S9 and S11 gives us

$$\overline{F(x)} - M(v_1^{(p)},v_2^{(p)},v_3^{(p)},...) = e_s + \overline{e_m(x)} \quad \text{(S12)}$$

The right side of Equation S12 is therefore the error of the model simulation over the spatial domain. From its definition in Equation S11, the statistical expectation of e_s is 0. By referring to the structural analysis of the model residue error given in Allen and Raktoe (1981), $\overline{e_m(x)}$ can be summarized into two parts, the bias and the variance of the modelling residuals. We, thereafter, itemized the total uncertainty (U_T), the right side of Equation S12, as:

$$U_T = U_d + U_b + U_v \quad \text{or} \quad \sigma_T^2 = \sigma_d^2 + \sigma_b^2 + \sigma_v^2 \quad \text{(S13)}$$

where U_d (or σ_d^2) signifies the spatially representative error corresponding to e_s in Equation S12, and U_b+U_v is the uncertainty attributed to the model fallacy, $e_m(x)$. U_b (or σ_b^2) represents the model performance bias at the site scale, whereas U_v (or σ_v^2) represents the model fallacy apart from U_B, which is the combination of the regression error and the random error, as described in Allen & Raktoe (1981). The assumed independence between e_s and $e_m(x)$ originates from the fact that they are due to separate causes. For a specific model, the model fallacy is independent of the accuracy and spatial availability of the model inputs that facilitate modeling in a spatial domain. However, changes in the model mechanism may regulate the relationship between U_d and U_b+U_v; for example, improving model performance by incorporating more factors as input variables may reduce the model's fallacy but increase the representative error due to the additional input data requirements necessary to run the model .

Due to substantial heterogeneities in spatial processes, such as fluxes in methane emissions from rice cultivation, the large area under study is usually split into several smaller regions. These regions may consist of grids of the same size or irregular patches of different sizes. Each division is a spatial domain with less heterogeneity to which modeling can be applied. To summarize the modeling results for each division, the spatial aggregation of U_d was discussed by Zhang et al. (2014) and is briefly addressed in Section S4. Section S4 also provides the rationale for the spatial aggregation of U_b and U_v.

Section S4 Spatial aggregation of the estimation uncertainties in grids

S4.1 Correlation coefficients of the model estimates between two grids due to data sharing of the model inputs

In each grid, i, the model estimates obtained via Monte Carlo iteration produce a numeric depiction of a random variable $V_i(m_i, \sigma_i)$, where m_i and σ_i are the statistical mean and standard deviation, respectively, of the random variable V_i. Thereafter, model up-scaling involves summation of the random variables $V_0=V_1+V_2+...+V_N$. The aggregation of uncertainty, represented by the statistical variance or standard deviation, is generalized as $Var(\sum_{i=1}^{N} V_i) = \sum_{i=1}^{N}\sum_{j=1}^{N} Cov(V_i, V_j)$ (Ross, 2006), and it can be transformed into a quadratic summation of the elementary variances via the standardized variance–covariance coefficient matrix:

$$\sigma_d^2 = \sum_{i,j} \sigma_i \times C_{i,j} \times \sigma_j \text{ , } (i=1...\text{N}, j=1...\text{N}) \qquad \text{(S14)}$$

where σ_d^2 is the aggregated variance of the regional estimation, and σ_i and σ_j are the standard deviations of the within-cell variations in cells i and j, respectively. Matrix **C** is composed of $C_{i,j}$ coefficients, which represent "correlations" between the spatially representative errors (e_s in Equation S12) of the individual cells. "Correlation" here is a measure of how the model outputs in two cells vary concurrently when they share common data for the model inputs. If the estimate in cell i is over-/underestimated, then the estimate in cell j will most likely be over-/underestimated as well, and vice versa, because they share common data. It is noteworthy that the correlation represented by $C_{i,j}$ is different from that between the "real" processes represented by $F(x)$ in Equation S12. The aggregation of the model outputs can be quite simple if the model estimate is generated with independent data in each cell. In this case, matrix **C** will be an identity matrix in which the diagonal elements will be 1, and all of the off-diagonal elements will be 0. The aggregation in Equation S14 will thereafter indicate the arithmetic sum of the within-cell variances, as addressed by the *Law of Large Numbers*. However, when there are not sufficient data to support independent calculation among cells, the off-diagonal elements, $C_{i,j}$, of matrix **C** will no longer be zero.

In the present study, $C_{i,j}$ was empirically calculated through numerical experiments. For a different level of data sharing between two cells (Table S3), the model estimates in the two cells were iteratively calculated with CH4MOD. The model inputs were randomly selected from the range of values for the variables. When data sharing occurred between the two cells for a variable in Table S3, the value of the variable was selected once for the two cells; for the variables for which there was no data sharing, the value of the variable was selected separately for the two cells. The correlation coefficients ($C_{i,j}$) of the model estimates in the two cells were statistically calculated with 1000 iterations of the paired model estimates in the two cells in the present study.

Table S3 Look-up table of correlation coefficients of the model outputs in two cells due to data sharing

Data sharing between cell *i* and *j*					$C_{i,j}$	Data sharing between cell *i* and *j*					$C_{i,j}$
Yield	*OM*	*Sand*	W_{Ptn}	*VI*		*Yield*	*OM*	*Sand*	W_{Ptn}	*VI*	
0†	0	0	0	1	0.069	1	0	0	0	1	0.136
0	0	0	1	0	0.347	1	0	0	1	0	0.430
0	0	0	1	1	0.413	1	0	0	1	1	0.520
0	0	1	0	0	0.295	1	0	1	0	0	0.343
0	0	1	0	1	0.375	1	0	1	0	1	0.478
0	0	1	1	0	0.674	1	0	1	1	0	0.776
0	0	1	1	1	0.796	1	0	1	1	1	0.900
0	1	0	0	0	0.082	1	1	0	0	0	0.170
0	1	0	0	1	0.167	1	1	0	0	1	0.225
0	1	0	1	0	0.436	1	1	0	1	0	0.481
0	1	0	1	1	0.519	1	1	0	1	1	0.616
0	1	1	0	0	0.396	1	1	1	0	0	0.458
0	1	1	0	1	0.499	1	1	1	0	1	0.575
0	1	1	1	0	0.760	1	1	1	1	0	0.849
0	1	1	1	1	0.878	1	1	1	1	1	1.000
1	0	0	0	0	0.066						

†1 means that the two cells share data for the variable, and 0 means that they do not share data for the variable

S4.2 Spatial aggregation of estimation uncertainties

Analogous to Equation S14, Equation S15, S16 and S17 was used to aggregate the uncertainty of U_b, U_v and U_d, of the estimated methane emission, respectively.

$$U_b = {\sigma_b}^2 = \sum_{i=1}^{N}\sum_{j=1}^{N}(A_i \times F_i \times r_b) \times E_{i,j} \times (A_j \times F_j \times r_b) \tag{S15}$$

$$U_v = {\sigma_v}^2 = \sum_{i=1}^{N}\sum_{j=1}^{N}(A_i \times F_i \times r_v) \times Q_{i,j} \times (A_j \times F_j \times r_v) \tag{S16}$$

$$U_d = {\sigma_d}^2 = \sum_{i=1}^{N}\sum_{j=1}^{N}(A_i \times \sigma_{d,i}) \times C_{i,j} \times (A_j \times \sigma_{d,j}) \tag{S17}$$

where A_i and A_j represent the rice harvesting area in grids i and j, respectively. No errors in the rice harvesting area were considered in Equation S15. F_i and F_j represent the average methane fluxes from the Monte Carlo simulation in grid i and j, and $\sigma_{d,i}$ and $\sigma_{d,j}$ are the standard deviations associated with F_i and F_j, respectively. No cross-correlation between the three components was considered here. Because of limited data availability, the neighboring grids were assigned probabilities of sharing data for the model input variables. The aggregation of $\sigma_{d,i}$ in the grids was therefore kernelled using data-sharing matrix **C** ($C_{i,j}$ represents its element, Table S3). $E_{i,j} = 1$ is the element of a constant matrix, **E**, which refers to the bias of the model estimates in all grids and is statistically under/overestimated concurrently in all grids. $Q_{i,j}$ is the element of matrix **Q**. $Q_{i,j}$ is not specifically known. The two extremes of matrix **Q** correspond to matrix **E** and the identity matrix, **I**. The estimation error, $F_i \times r_v$, is related to the factors that are not explicitly accounted for in the model, for instance, mineral fertilizer application (Xie et al., 2010) and soil organic carbon content (Zhan et al., 2011). Because the between-grid relationships of these "unknown" factors could not be explicitly accounted for, we assigned **U** the values for the mid-point of the two extremes **E** and **I**.

Combining Equation S15, S16 and S17, Equation S18 is then used to aggregate the uncertainties in all grids to calculate the uncertainty in the national inventory:

$$\begin{aligned} {\sigma_T}^2 &= \sigma_b^2 + \sigma_v^2 + \sigma_d^2 \\ &= \sum_{i=1}^{N}\sum_{j=1}^{N}(A_i \times F_i \times r_b) \times E_{i,j} \times (A_j \times F_j \times r_b) \\ &+ \sum_{i=1}^{N}\sum_{j=1}^{N}(A_i \times F_i \times r_v) \times Q_{i,j} \times (A_j \times F_j \times r_v) \\ &+ \sum_{i=1}^{N}\sum_{j=1}^{N}(A_i \times \sigma_{d,i}) \times C_{i,j} \times (A_j \times \sigma_{d,j}) \end{aligned} \tag{S18}$$

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