

# Background nitrous oxide emissions from croplands in China in the year 2000

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**Abstract** It is important to accurately predict annual background nitrous oxide (N<sub>2</sub>O) emissions (BNE) at a national scale due to the considerable contribution of these emissions to the overall N<sub>2</sub>O emissions from croplands. We predicted the national background N<sub>2</sub>O emissions (BNE<sub>n</sub>) from croplands in China in the year 2000 within a geographical information system (GIS) framework. The spatial resolution was 10 km × 10 km. The BNE<sub>n</sub> was predicted as the sum of two parts. One part was from mineral soils, estimated using four monivariate models based on soil total nitrogen content (TN) and soil organic carbon content (SOC), while the other part was from organic soils, estimated by directly extrapolating the default IPCC (2006) emission factor of 8 kg N ha<sup>-1</sup> yr<sup>-1</sup> to the area of organic soils, which consisted of less than 0.2% of the total area in national croplands. Our estimates showed that the hectare-based annual background N<sub>2</sub>O emission rates (BNE<sub>h</sub>) <0.1, 0.1–2.0, and >2.0 kg N ha<sup>-1</sup> yr<sup>-1</sup> occurred in 11–24%, 71–84%, and 3–5% of national croplands, respectively. A

spatial distribution pattern of grid-based background emissions (BNE<sub>g</sub>), different from that of BNE<sub>h</sub>, was revealed. High BNE<sub>g</sub> (>6,000 kg N grid<sup>-1</sup> yr<sup>-1</sup>) occurred in the major agricultural regions of North-eastern, Northern, Eastern, and Middle China. The simulation based on soil properties yielded estimates of 99.0–116.9 Gg (1 Gg=10<sup>9</sup> g) N yr<sup>-1</sup> for BNE<sub>n</sub>, of which organic soils contributed 1.5–1.8%. Among the different models, the model based on TN yielded the smallest uncertainty (ranging from –22% to 30%). We recommend this model for BNE estimation at a national scale. Our estimates of BNE<sub>n</sub> accounted for 26–30% of the total N<sub>2</sub>O emissions from the croplands in China. Methods for improving BNE predictions were proposed for further study.

**Keywords** Empirical model · GIS · National estimation · Soil properties · Spatial distribution · Uncertainty assessment

## Introduction

Agricultural soil is recognized as a significant source of atmospheric nitrous oxide (N<sub>2</sub>O). As an essential part of the national greenhouse gases inventory to be submitted to the United Nations Framework Convention on Climate Change (UNFCCC), it is important to accurately estimate N<sub>2</sub>O emissions from croplands (Li et al. 2001, 2005; Sozanska et al. 2002; Zheng et al. 2004). N<sub>2</sub>O emissions from cultivated soils are most

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often estimated as the sum of two independent parts, i.e., background and fertilizer-induced emissions (Bouwman 1996; Bouwman et al. 2002; Yan et al. 2003; Lu et al. 2006; Stehfest and Bouwman 2006). Background emissions are defined as the emissions from cultivated soils that received no nitrogen fertilizer in the current year or season (Zheng et al. 2004; Gu et al. 2007). They originate from a) residual nitrogen that has remained in the soil from nitrogen addition in previous years or seasons, and b) other nitrogen sources, such as biological nitrogen fixation, that are present in the soil. Fertilizer-induced emissions are the direct emissions that are induced by nitrogen fertilizers that had been added within the current year or season (Bouwman et al. 2002; Yan et al. 2003; Zheng et al. 2004; Stehfest and Bouwman 2006). According to the methodology outlined by the IPCC (1997, 2000, 2006), direct emissions are the product of a direct N<sub>2</sub>O emission factor (EF<sub>d</sub>) and the total amount of input nitrogen, which can originate from the application of organic and/or inorganic fertilizers, biological nitrogen fixation, and crop residues. To date, most efforts have focused on how to accurately quantify EF<sub>d</sub>s, and subsequently the direct emissions; however, background emissions have been neglected or considered insignificant (Bouwman 1996; Bouwman et al. 2002; Yan et al. 2003; Zheng et al. 2004; Stehfest and Bouwman 2006). The national inventory requires that direct emissions are fully and independently accounted. Background emissions are considered as part of the total indirect emissions (IPCC 1997, 2000, 2006). However, it is of substantial importance to quantify background emissions accurately, at a national scale, due to their considerable contribution to the overall N<sub>2</sub>O emissions from croplands: 26–52% in China (Li et al. 2001; Yan et al. 2003; Lu et al. 2006; Gu et al. 2007), and 43–52% in the United Kingdom of Great Britain (Brown et al. 2002; Sozanska et al. 2002).

In general, two approaches may be applied for estimating background N<sub>2</sub>O emissions from croplands at a national scale. One is based on process-oriented models and the other is based on empirical models. DNDC (Li et al. 2001, 2005) is a popular process-oriented model, and includes features for simulating N<sub>2</sub>O emissions from agricultural soils. Validations of DNDC show that it is able of capturing the general patterns and magnitudes of N<sub>2</sub>O emissions observed in fertilized fields (Li et al. 2001, 2005). Thus, DNDC has been used to simulate

N<sub>2</sub>O emissions at a national scale (e.g., Li et al. 2001, 2005; Brown et al. 2002). However, DNDC requires complex definitions of parameters and inputs such as temperature, moisture, pH, redox potential, and other environmental factors that are not usually required for empirical models (Li et al. 2001, 2005). Some of these parameters are not available at regional or national scales. This increases the difficulties in applying the DNDC model. Presently, process-oriented models are not yet “universally valid” (Jungkunst et al. 2006; Stehfest and Bouwman 2006). Thus, the national estimates generated by these models need to be cross-verified with empirical models (Li et al. 2001; Brown et al. 2002; Stehfest and Bouwman 2006). The early empirical models assume that background emissions occur at a constant rate of 1.0 kg N ha<sup>-1</sup> yr<sup>-1</sup> (Bouwman 1996). This figure was obtained as the statistical mean of a few observations carried out in managed grasslands (Bouwman 1996, and references therein). It provides the simplest approach for estimating background emissions at regional/global scales. However, this figure ignores the spatial variation of background emissions, as well as the regulatory effects of soil and climate upon N<sub>2</sub>O production in soils. In fact, background emission rates, within a specific region, may greatly deviate from Bouwman’s value (Yan et al. 2003; Gu et al. 2007). Huge spatial and/or inter-annual variation is consistent in background emissions within a given region, which is due to the variations of soil and climate (Lu et al. 2006; Gu et al. 2007). Therefore, empirical models must be further developed in order to address spatial and temporal variations in background N<sub>2</sub>O emissions. In this regard, Gu et al. (2007) report that the annual background N<sub>2</sub>O emission (BNE) rates from cultivated mineral soils across various soil/climate regions of China and/or the world are significantly regulated by soil properties such as soil total nitrogen content (TN), soil organic carbon content (SOC), bulk density and/or clay fraction. These regulatory effects are described by univariate or multivariate regression functions, which suggest simple, but improved, empirical models to estimate BNE at various spatial scales. The determination coefficients of these functions indicate that multivariate functions might explain up to 95% of the spatial variation of BNE (Gu et al. 2007). However, the results from multivariate functions might

**Fig. 1** GIS (grid size: 10 km×10 km) databases of cropland soil properties and area. *a* cropland area per grid (%) in the year 2000. *b* soil total nitrogen content (TN, g N kg<sup>-1</sup> soil). *c* soil organic carbon content (SOC, g C kg<sup>-1</sup> soil). Data were provided by the Institute of Soil Sciences and Resources and Environmental Scientific Data Center, Chinese Academy of Sciences

be misleading because of the volume of the multidimensional spaces. Accordingly, one usually prefers monivariate regression functions when making regional estimations.

This study attempts to couple monivariate functions, of TN or SOC, with a geographical information system (GIS) to estimate the BNE from croplands in China, in the year 2000.

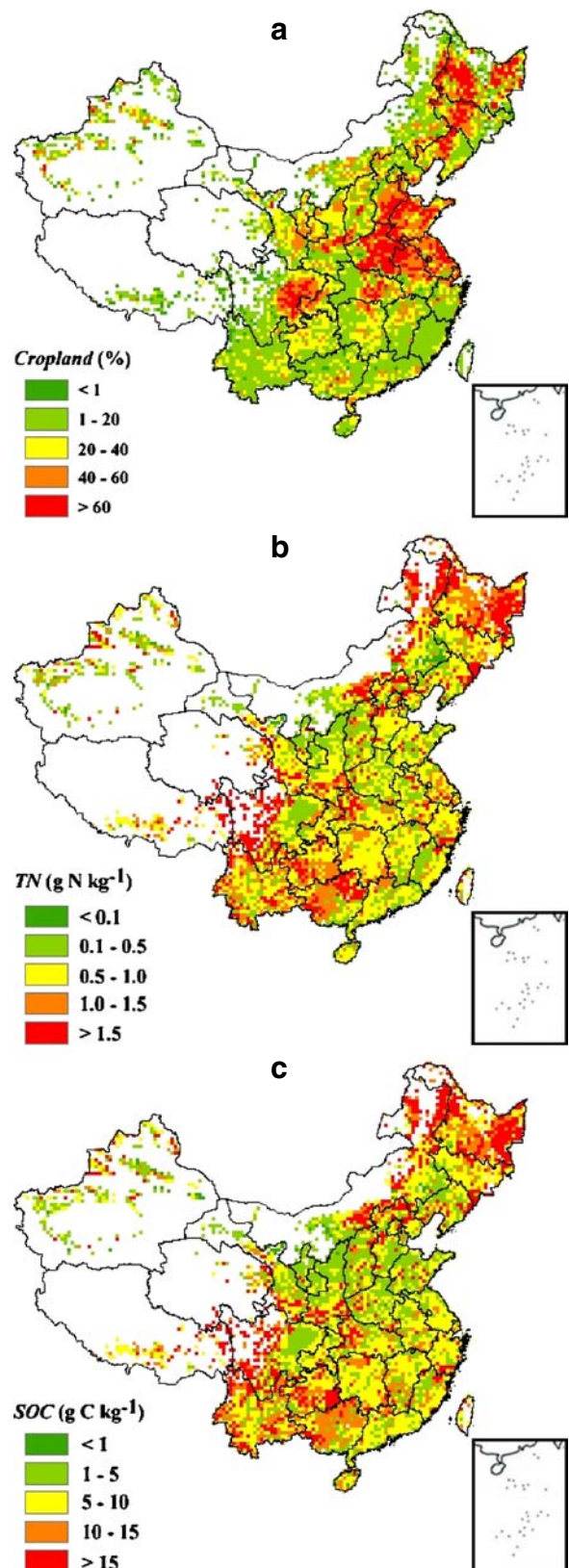
## Materials and methods

### GIS database

We applied a GIS software package, ArcGIS (ESRI, Redlands, California, USA) to map soil properties and cropland area, as well as estimated annual background N<sub>2</sub>O emissions on a per hectare basis (BNE<sub>h</sub>) and a grid basis (BNE<sub>g</sub>). The spatial resolution was 10 km×10 km.

The statistical data for cropland area at the county level were linked to a map of the administrative boundaries of China (1:100,000), and then re-assigned to the corresponding grids by overlaying the map with the thematic raster data of land-use. The total area of cultivated soils in China in the year 2000 amounted to 136 Mha (1 Mha=10<sup>6</sup> ha). The major agricultural regions were located in Northeastern, Northern, Eastern, Middle and Southwestern (Sichuan Basin) China (Fig. 1a).

The TN and SOC data for cultivated soils (Figs. 1b–c) were extracted by overlaying the maps of the latest soil GIS database of China with the land-use map and re-assigning the property data into individual grids. The TN ranged from 0.02 to 23.7 g N kg<sup>-1</sup> soil, and the SOC ranged from 0.17 to 361.4 g C kg<sup>-1</sup> soil, respectively. The soil GIS database integrates about 7,400 soil profiles surveyed all over China during the past three decades and was provided by the Institute of Soil Sciences, Chinese Academy of Sciences (CAS). The land-use map was provided by the Resources and Environmental Scientific Data Center, CAS.



## Empirical models for cultivated mineral soils

Network observations (six sites) of BNE rates were carried out across various soil/climate regions in China, during 2002–2006 (Gu et al. 2007). Based on these annual measurements, as well as previous measurements in China and abroad, four empirical equations were obtained. These equations describe the significant relationship between BNE<sub>h</sub> and individual soil properties of TN and SOC, respectively and apply to cultivated mineral soils. However, a number of the BNE measurements (Sanjiang, Shenyang, Yanting, Wuxi, and Jiangdu) involved in obtaining these empirical models were observed through opaque static chamber techniques coupled with using pure diatomic nitrogen (N<sub>2</sub>), as the carrier gas, in a gas chromatograph (GC) equipped with an electron capture detector (ECD) for N<sub>2</sub>O analysis (hereinafter referred to as the N<sub>2</sub> method). A recent study has shown that the N<sub>2</sub> method might overestimate the coherent N<sub>2</sub>O emissions from plants, forest floors, typical semi-arid steppes or other plant-soil systems. Accumulation of carbon dioxide (CO<sub>2</sub>) in the chamber enclosures has been identified as the major reason for overestimation in N<sub>2</sub>O emission quantification (Zheng et al. 2008). Thus, adding ascarite to the N<sub>2</sub> method (hereinafter referred to as the N<sub>2</sub>-Ascarite method), for removing CO<sub>2</sub> from the air samples, is one approach to avoid overestimation (e.g., Butterbach-Bahl et al. 1997; Holst et al. 2007, 2008). By conducting two contrasting sets of measurements, using the N<sub>2</sub> and N<sub>2</sub>-Ascarite methods, Zheng et al. (2008) have proposed correction terms for cropland N<sub>2</sub>O emission fluxes measured using the N<sub>2</sub> method. When the N<sub>2</sub>O fluxes measured by the N<sub>2</sub> method were <−30, −30–0, 0–30, 30–100, and 100–200 μg N m<sup>−2</sup> h<sup>−1</sup>, significant differences that amounted to −72, −22, 5, 38, and 64 μg N m<sup>−2</sup> h<sup>−1</sup>, respectively, appeared in comparison to the N<sub>2</sub>-Ascarite method. Using the correction terms, we corrected the raw N<sub>2</sub>O fluxes measured by the N<sub>2</sub> method at four sites (Sanjiang, Shenyang, Yanting, and Wuxi) and subsequently re-quantified the annual BNE rates. Table 1 lists the corrected annual BNE rates from these sites. At the Jiangdu site, we adopted the annual BNE rates that were directly measured simultaneously with the N<sub>2</sub>-Ascarite method, as provided by Zheng et al. (2008). As compared to the annual BNE rates originally quantified with the N<sub>2</sub>-measured fluxes, the corrected values, together with the one directly

quantified with the N<sub>2</sub>-Ascarite-measured fluxes, were significantly lower (20–48% with a mean of 33%). Using the data shown in Table 1, and substituting the annual BNE rates from the five sites with the corrected rates, we updated the parameters and parameter uncertainties of the models based on TN and SOC for predicting of annual BNE rates or BNE<sub>h</sub>. The updated models (Eqs. 1–4) are listed in Table 2. Equations 1 and 3 were obtained from the observations in China, and Eqs. 2 and 4 resulted from measurements acquired from around the world (including those in China). These could explain 47–73% of spatial variation of BNE<sub>h</sub> from cultivated mineral soils.

The cropping systems, from which BNE rates were measured, included both upland and paddy soils (Table 1). Since the 1980s, water management of Chinese rice paddies has changed substantially. Midseason drainage has gradually replaced continuous flooding, which would stimulate N<sub>2</sub>O emissions from the paddies (Zheng et al. 2000; Li et al. 2005). Results from the network observations (Gu et al. 2007) showed that the mean N<sub>2</sub>O fluxes from the rice-growing season are comparable with, or higher than, those from the non-rice season at Yanting (15.7±15.5 vs. 11.4±2.1 μg N m<sup>−2</sup> h<sup>−1</sup>, *n*=3, *p*>0.05 for rice-winter wheat rotation; 53.8±2.1 vs. 18.8±1.7 μg N m<sup>−2</sup> h<sup>−1</sup>, *n*=3, *p*<0.01 for rice-rape seed rotation), Jiangdu (12.7±1.7 vs. 15.6±1.9 μg N m<sup>−2</sup> h<sup>−1</sup>, *n*=6, *p*>0.05 for rice-winter wheat rotation), and Wuxi (11.8±2.5 vs. 17.5±3.0 μg N m<sup>−2</sup> h<sup>−1</sup>, *n*=6, *p*>0.05 for rice-winter wheat rotation). This evidence indicates that it is reasonable to estimate the BNE from upland and paddy soils together using Eqs. 1–4.

Using survey data of soil properties (TN and SOC) and cropland area, we applied Eqs. 1–4 in order to investigate the spatial distributions of BNE<sub>h</sub>, BNE<sub>g</sub>, and national total BNE (BNE<sub>n</sub>) from Chinese cultivated mineral soils in the year 2000.

## Estimate method for organic soils

Organic soil is characterized by a layer of organic matter (>30% or >174 g C kg<sup>−1</sup> soil) of more than 40 cm either extending down from the surface, or taken cumulatively within the upper 80 cm of the soil (Food and Agriculture Organization of the United Nations, <http://www.fao.org/AG/AGL/agll/prosoil/histo.htm>). By this definition, less than 0.2% of the area of cultivated soils in China is classified as organic soils,



**Table 1** Observed annual background N<sub>2</sub>O emission (BNE) rates (on a per hectare basis) in cultivated mineral soils and their corresponding cropping systems, measurement duration (year), thickness of the cultivated soil layer (cm), soil organic carbon content (SOC, g C kg<sup>-1</sup> soil), and soil total nitrogen content (TN, g N kg<sup>-1</sup> soil)

Nation	Site	Crop	Duration (year)	Thickness of soil (cm)	SOC (g C kg <sup>-1</sup> soil)	TN (g N kg <sup>-1</sup> soil)	BNE (kg N ha <sup>-1</sup> yr <sup>-1</sup> )	Reference
China	Sanjiang	Maize & spring wheat	2	0–25	30.8	3.1	2.18*	[1]
	Shenyang	Maize	1	0–20	9.4	0.8	0.43	[2]
	Fengqiu	Maize & spring wheat	2	0–20	9.6	0.9	1.12*	[1]
		Winter wheat-maize rotation	2	0–20	6.8	0.5	0.67	[3]
		Same as above	2	0–20	6.4	0.4	0.35	
	Yanting	Same as above	2	0–20	7.3	0.6	0.68	
		Winter wheat-rice rotation	1	0–15	7.1	0.8	1.28*	[1]
		Winter wheat-maize rotation	1	0–15	9.7	1.2	1.39*	
	Wuxi	Rape seed-rice rotation	1	0–15	8.6	1.2	0.90*	
		Winter wheat-rice rotation	2	0–15	15.0	1.6	1.70*	[1]
Same as above		1	0–15	18.3	1.4	1.0	[4]	
Winter wheat-maize rotation		1	0–20	7.3	0.8	0.49	[5], [6]	
Same as above		2	0–20	4.5	0.5	0.1	[7]	
Germany	Suzhou	Winter wheat-rice rotation	1	0–15	20.3	1.9	2.53	[8], [9]
	Yingtian	Double rice-dry fallow rotation	1	0–15	14.0	0.8	1.12	[10]
	Braunschweig	Sugar beet-winter wheat-winter barley-fallow rotation & winter wheat-winter rape rotation	3	0–20	9.4	1.1	2.1	[11]
	Rostock	-	2.6	-	8.5	0.8	0.6	[12]
	Potsdam	Cocksfoot, willow, poplar, rye, triticale, hemp & rape	6	0–30	9	-	0.9	[13]
France	Scheyern	Maize	1	0–20	15	1.7	1.5	[14]
	Freising	Same as above	1	0–20	14	1.5	1	
USA	Lower Saxony	Maize-winter wheat-winter barley rotation	1.5	0–20	11.4	1.7	2.5	[15]
	Burgundy	Winter wheat	1	0–15	12.5	1.2	1.8	[16]
Canada	Colorado	Rape seed	1	0–20	11	0.9	0.66	[17]
	Lethbridge	Wheat	1	0–15	10	-	0.44	[18]
Costa Rica	La Selva	Wheat & canola	1	0–15	20	-	0.43	[19]
		Double maize-double taro rotation	2	0–10	50.35	5.3	2.63	[20]
		Same as above	2	0–10	41.41	4.1	2.29	

\* Corrected from those annual BNE rates provided by Gu et al. (2007) following Zheng et al. (2008). [1] This study. [2] Huang et al. 1998. [3] Gu et al. 2007; [4] Zheng et al. 2008. [5] Wang et al. 1994. [6] Zeng et al. 1995. [7] Dong et al. 2001. [8] Xing and Zhu 1997. [9] Xing 1998. [10] Xiong et al. 2002. [11] Kaiser et al. 1998. [12] Jungkunst et al. 2006. [13] Hellebrand et al. 2003. [14] Sely et al. 2003. [15] Kilian et al. 1998. [16] Henault et al. 1998. [17] Röver et al. 1998. [18] Bronson and Mosier 1993. [19] Hao et al. 2001. [20] Weitz et al. 2001.

**Table 2** Dependences of annual background N<sub>2</sub>O emissions (BNE<sub>h</sub>, kg N ha<sup>-1</sup> yr<sup>-1</sup>) from cultivated mineral soils on soil total nitrogen content (TN, g N kg<sup>-1</sup> soil) and soil organic carbon content (SOC, g C kg<sup>-1</sup> soil)

No.	Equations
Eq. 1	$BNE_h = 1.044[\pm 0.354]\text{Ln}(\text{TN}) + 1.123[\pm 0.096]$ (n = 15, r <sup>2</sup> = 0.73, p < 0.01)
Eq. 2	$BNE_h = 0.994[\pm 0.280]\text{Ln}(\text{TN}) + 1.140[\pm 0.180]$ (n = 24, r <sup>2</sup> = 0.70, p < 0.01)
Eq. 3	$BNE_h = 1.085[\pm 0.413]\text{Ln}(\text{SOC}) + 1.457[\pm 0.980]$ (n = 15, r <sup>2</sup> = 0.68, p < 0.01)
Eq. 4	$BNE_h = 0.914[\pm 0.387]\text{Ln}(\text{SOC}) + 1.058[\pm 0.985]$ (n = 27, r <sup>2</sup> = 0.47, p < 0.01)

n, r<sup>2</sup> and p indicate sample size, determination coefficient and significance level, respectively. Data in each pair of square brackets is the uncertainty (at 95% confidence interval) for the corresponding parameter. Eqs. 1 and 3 were obtained with the observations in China while Eqs. 2 and 4 fitted with the observations around the world, including those data of China and other regions (see Table 1).

where these are distributed mainly in Northeastern China (Fig. 1c). Considering the huge N<sub>2</sub>O emission rate from organic soils (e.g., Terry et al. 1981), we independently estimated the background emissions from organic soils, and then added the estimates to those from mineral soils. However, the year-round measurements of N<sub>2</sub>O emissions from cultivated organic soils are rare in China. An emission factor for temperate organic crop and grassland soils provided by IPCC (2006), i.e., 8 kg N ha<sup>-1</sup> yr<sup>-1</sup>, was used in our estimations by directly extrapolating to the organic soil area in China.

#### Calculation assumptions for mineral soils

A cultivated soil that lacks the above characteristics for organic soils was regarded as a cultivated mineral soil in this study. As Gu et al. (2007) mentioned, the empirical models used in this study (Eqs. 1–4) could not adequately cover the entire extent of cultivated mineral soils at the national scale of China. This is because they were obtained with TN > 0.4 g N kg<sup>-1</sup> soil or SOC > 4.5 g C kg<sup>-1</sup> soil (Table 1). When TN < 0.3 g N kg<sup>-1</sup> soil for Eqs. 1–2, or when SOC < 3.0 g C kg<sup>-1</sup> soil for Eqs. 3–4, these models yield approximately zero, or negative estimates of annual BNE rates. Slight net uptake of atmospheric N<sub>2</sub>O might be caused by physical and/or biogeochemical processes in the soil, in addition to uncertainty of the measurements (Chapuis-Lardy et al. 2007). In fact, while net uptakes were occasionally detected in different ecosystems, including croplands (Chapuis-Lardy et al. 2007, and references therein), few negative annual BNE rates have been observed in cultivated mineral soils (Gu et al. 2007, and references therein). In total, 17% of the cropland area in China had TN

values < 0.3 g N kg<sup>-1</sup> soil, and 23% of cropland area had SOC values < 3.0 g C kg<sup>-1</sup> soil (Fig. 1). Applying Eqs. 1–4 to these areas would result in negative annual BNE rates. These values do not reflect the true situation, and might thus introduce considerable negative uncertainty into national estimates. In light of this, we set the lowest simulated BNE<sub>h</sub> at 0 kg N ha<sup>-1</sup> yr<sup>-1</sup> with the assumption that negative annual BNE rates would not occur in cultivated mineral soils.

#### Uncertainty assessment

The soil properties (TN and SOC), used as inputs into the empirical models (Eqs. 1–4), are inherently heterogeneous within a grid cell. However, data on the spatial variation within a grid cell was not available for either of the soil properties of interest. Thus, individual soil properties were assumed to be uniform within a grid cell. This assumption, however, introduced uncertainties into the estimates of BNE<sub>h</sub>, BNE<sub>g</sub> and subsequently BNE<sub>n</sub>. In addition, uncertainties were introduced by the model parameters. Fortunately, the parameter-related uncertainties could be easily quantified, as the uncertainty ranges of individual parameters were provided (Table 2). Considering that the data for the cropland area in a grid cell originated from the annual governmental statistics of each county, we assumed that this uncertainty could be ignored.

To estimate the uncertainties of TN or SOC within a grid cell, we picked the minimum and maximum property values within a 30 km × 30 km area. This area covers nine adjacent grid cells. This range was considered to be the most representative of the TN or SOC uncertainty (at the 95% confidence interval) of the central grid cell of the selected square. We then

calculated the standard deviation (SD) of the BNE<sub>h</sub> estimates, using Monte Carlo (MC) simulation. This SD was regarded to represent the uncertainty of the BNE<sub>h</sub> estimate for the central grid cell. In the MC simulation, random samples of the soil property, the model parameters, and the subsequent BNE<sub>h</sub> calculation, were acquired more than 10,000 times, until the mean of the BNE<sub>h</sub> estimates converged to a stable value, with a tolerance of <1%. The random sampling was performed under the assumption that TN and SOC, as well as any other parameters involved, were normally distributed. The uncertainty of the BNE<sub>g</sub> estimate for the central grid was determined by multiplying the SD for the BNE<sub>h</sub> estimates with the cropland area of the grid cell. The uncertainties for the BNE<sub>g</sub> estimates of organic soils were independently quantified. The uncertainty of the BNE<sub>g</sub> estimates for organic soils in a given grid cell was obtained directly by multiplying the fixed uncertainty range of 2–24 kg N ha<sup>-1</sup> yr<sup>-1</sup> (IPCC 2006) by the organic soil area. The uncertainties for the BNE<sub>g</sub> estimates of mineral soils and organic soils were then integrated. Ultimately, the uncertainty of the BNE<sub>n</sub> estimate was obtained by summing the BNE<sub>g</sub> uncertainty estimates for all cells.

## Results

### Spatial distributions of background N<sub>2</sub>O emission rates (BNE<sub>h</sub>)

Figure 2a shows the spatial distribution of BNE<sub>h</sub> from cultivated mineral soils, calculated with TN (Eq. 1). High BNE<sub>h</sub> (>2.0 kg N ha<sup>-1</sup> yr<sup>-1</sup>) occurred over 4% of the total national cropland area, which were located in Northeastern and Southwestern China. The area showing low BNE<sub>h</sub> (<0.1 kg N ha<sup>-1</sup> yr<sup>-1</sup>) covered 12% of the national croplands. These areas were distributed in Northern, Northwestern and Southern China. The BNE<sub>h</sub> of 0.1–2.0 kg N ha<sup>-1</sup> yr<sup>-1</sup> occurred over the remaining 84% of croplands. The exact same pattern of spatial distribution (Fig. 2b) was obtained when the other model, based on TN (Eq. 2), was applied. The percentages of national total cropland area were 11%, 84%, and 5% for BNE<sub>h</sub> <0.1, 0.1–2.0, and >2.0 kg N ha<sup>-1</sup> yr<sup>-1</sup>, respectively.

The spatial distribution of BNE<sub>h</sub> estimates based on SOC (Figs. 2c–d) was comparable with those

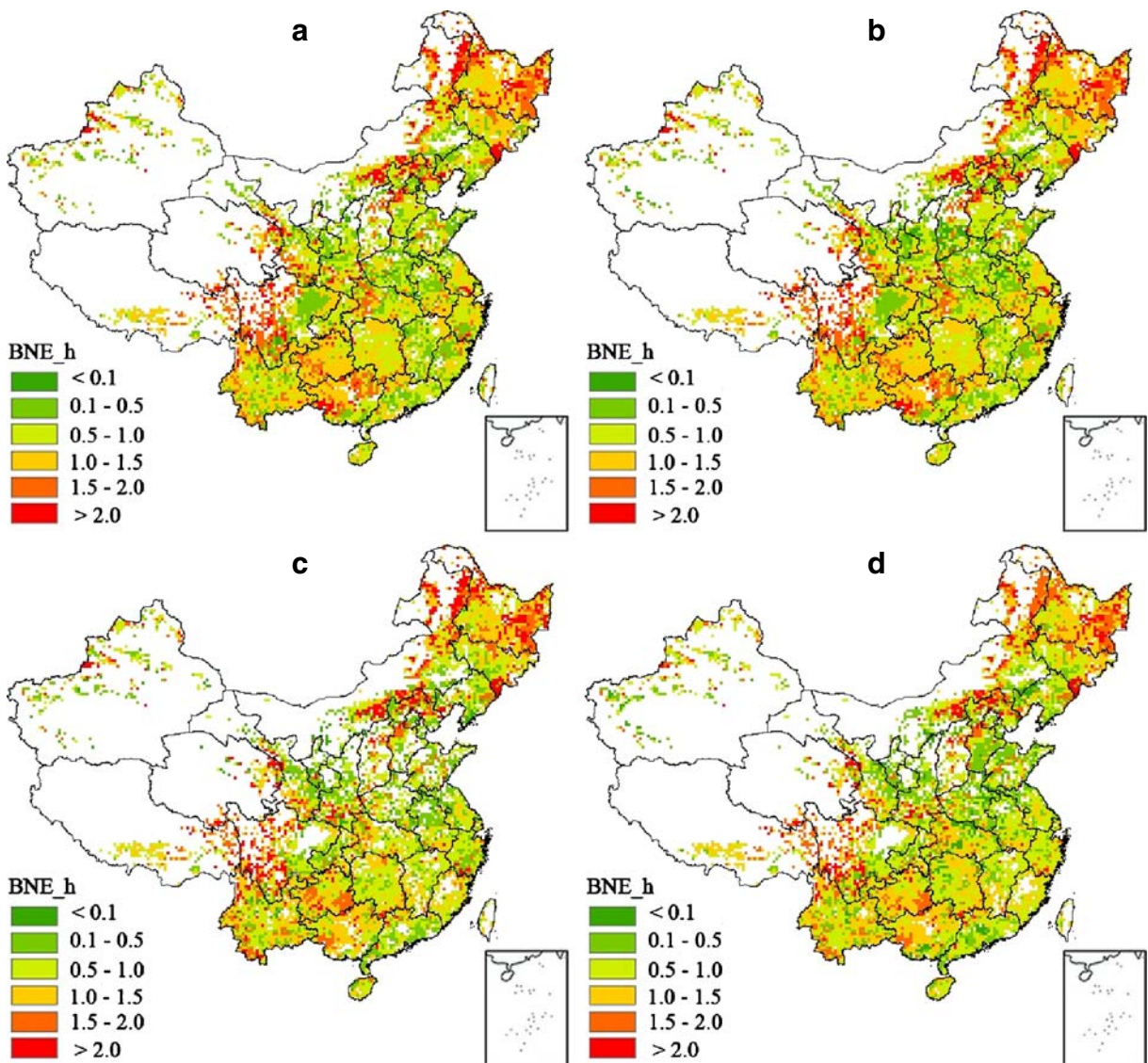
based on TN (Figs. 2a–b). High BNE<sub>h</sub> (>2.0 kg N ha<sup>-1</sup> yr<sup>-1</sup>) occurred over 3% (Fig. 2d) to 5% (Fig. 2c) of national croplands that were located in Northeastern and Southwestern China. The BNE<sub>h</sub> ranging from 0.1 to 2.0 kg N ha<sup>-1</sup> yr<sup>-1</sup> covered 71% (Fig. 2c) to 79% (Fig. 2d) of national cropland area. The main difference between the estimates, based on TN and SOC, appeared in Southwestern China (Sichuan Basin; Fig. 2). The estimates based on TN show low BNE<sub>h</sub> in this region. However, the estimates based on SOC yielded zero background emissions, following the calculation assumptions for mineral soils. This is attributed to the purplish soils that dominate this region, which are characterized by low SOC (Fig. 1c). Furthermore, Fig. 2c shows more zero BNE<sub>h</sub> area in Northern (Hebei and Shanxi province) and Eastern (Shandong province) China than what is shown in Figs. 2a, b and d.

### Spatial distributions of grid-based background N<sub>2</sub>O emissions (BNE<sub>g</sub>)

Figures 3a–d illustrate the spatial distributions of estimated BNE<sub>g</sub> from cultivated mineral soils. These distributions are products of the estimated BNE<sub>h</sub> (Fig. 2) and the cultivated land area (Fig. 1a). Figure 3a shows relatively high BNE<sub>g</sub> (>6,000 kg N grid<sup>-1</sup> yr<sup>-1</sup>) distributed over 19% of the national cultivated land area, and located over a majority of Northeastern, Northern, Eastern, and Middle China. These regions also correspond to the main agricultural areas in China (Fig. 1a). Figures 3a–d show a similar spatial distribution pattern of BNE<sub>g</sub>. However, there are more zero BNE<sub>g</sub> areas in Figs. 3c–d because of the assumed zero BNE<sub>h</sub>.

### Estimates of the national total background N<sub>2</sub>O emissions (BNE<sub>n</sub>) and uncertainties

We obtained the national total amounts of background N<sub>2</sub>O emissions from cultivated soils in China in the year 2000 (Table 3) by summing up the values of BNE<sub>g</sub> estimates for mineral soils, illustrated in Figs. 3a–d, and those for organic soils. The BNE<sub>n</sub> calculated with TN (Eqs. 1–2) and SOC (Eqs. 3–4) were between 99.0 and 116.9 Gg (1 Gg=10<sup>9</sup> g) N yr<sup>-1</sup>. The organic soils contributed 1.8 Gg N yr<sup>-1</sup>, and accounted for 1.5–1.8% of the BNE<sub>n</sub> estimates. The BNE<sub>n</sub> amount that relied on the estimates based on



**Fig. 2** Estimates of annual background N<sub>2</sub>O emission rates on a per hectare basis (BNE<sub>h</sub>, kg N ha<sup>-1</sup> yr<sup>-1</sup>) from cultivated mineral soils of China in the year 2000. *a* estimated with TN using Eq.1. *b* estimated with TN using Eq.2. *c* estimated with

SOC using Eq.3. *d* estimated with SOC using Eq.4. Definitions of TN and SOC are referred to in the captions of Fig. 1 or Table 1

SOC for mineral soils was slightly lower than that which relied on the estimates based on TN for mineral soils.

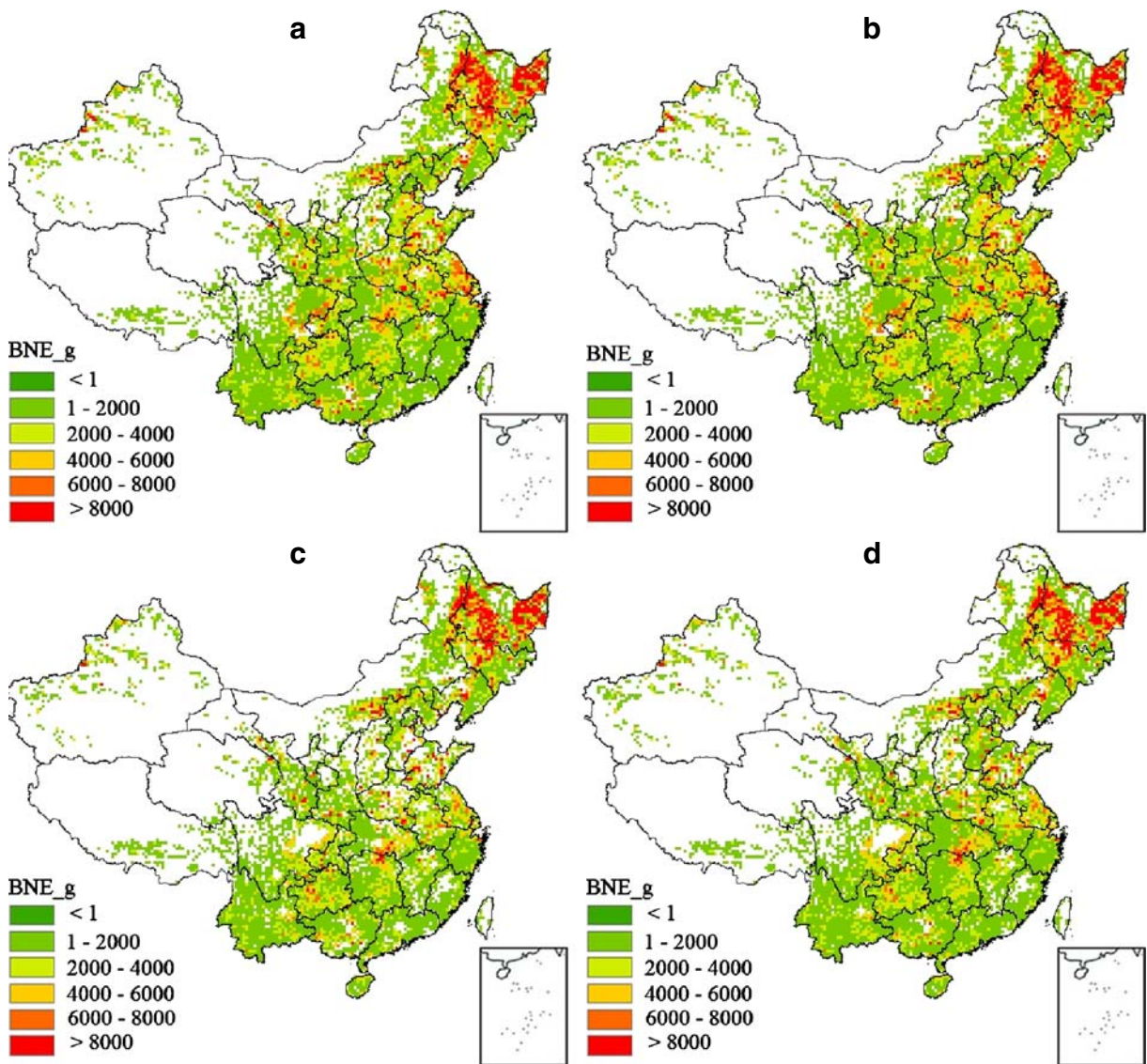
As shown in Table 3, the estimated BNE<sub>n</sub> uncertainties (at the 95% confidence interval), from models based on TN, ranged from -22% to 31% (by Eq. 1) and -22% to 30% (by Eq. 2), respectively. They were much narrower than those resulting from the two models based on SOC (i.e., from -61% to 98% by Eq. 3 and -62% to 91% by Eq. 4).

## Discussion

Estimates of BNE<sub>n</sub> from cultivated soils

In addition to the coupled model-GIS estimates of BNE<sub>n</sub> from cultivated soils, Table 3 lists some results from direct extrapolation of BNE<sub>h</sub> values, at a national scale (according to the total national cropland area). By directly extrapolating the constant BNE<sub>h</sub> of 1.0 kg N ha<sup>-1</sup> yr<sup>-1</sup> (Bouwman 1996) to a national





**Fig. 3** Estimates of the annual background N<sub>2</sub>O emissions on a grid basis (BNE<sub>g</sub>, kg N grid<sup>-1</sup> yr<sup>-1</sup>) from cultivated mineral soils of China in the year 2000. *a* estimated with TN

using Eq. 1. *b* estimated with TN using Eq. 2. *c* estimated with SOC using Eq. 3. *d* estimated with SOC using Eq. 4. Definitions of TN and SOC are referred to in the captions of Fig. 1 or Table 1

cropland area of 95 Mha in China, Li et al. (2001) have reported a BNE<sub>n</sub> estimate of 95 Gg N yr<sup>-1</sup> for the year 1990. By directly multiplying this constant annual BNE<sub>h</sub> with the national cropland area of 136 Mha, Gu et al. (2007) have presented an estimate of 136 Gg N yr<sup>-1</sup> for the year 2000. Yan et al. (2003) have reported a BNE<sub>n</sub> estimate of 127.1 Gg N yr<sup>-1</sup> for the year 1995. This was the product of the mean BNE<sub>h</sub> of 1.22 kg N ha<sup>-1</sup> yr<sup>-1</sup> from seven field observations in Asian croplands, and a national cropland area of 104 Mha. A national mean BNE<sub>h</sub>

of 1.06 kg N ha<sup>-1</sup> yr<sup>-1</sup> was the result of fifteen field observations in China (Table 1). Directly extrapolating this mean BNE rate to a national scale, we presented a BNE<sub>n</sub> estimate of 144.2 Gg N yr<sup>-1</sup> for the year 2000. In addition, we report a global mean BNE<sub>h</sub> of 1.21 kg N ha<sup>-1</sup> yr<sup>-1</sup> based on the 27 worldwide field observations (Table 1). Directly multiplying this BNE<sub>h</sub> value with the national cropland area yields a BNE<sub>n</sub> estimate of 164.6 Gg N yr<sup>-1</sup> for the year 2000. All of the above estimates fail to take into account the contribution of the high emission rates from cultivated

**Table 3** Estimated national total amounts of background N<sub>2</sub>O emission from croplands in China (BNE<sub>n</sub>) and their uncertainties

Calculation method	Year	Area (10 <sup>6</sup> ha)	BNE <sub>n</sub> (10 <sup>9</sup> g N yr <sup>-1</sup> )	Uncertainty <sup>f</sup> (%)	Remarks
Eq. 1 <sup>a</sup>	2000	136	113.6	-22–31	This study
Eq. 2 <sup>a</sup>	2000	136	116.9	-22–30	This study
Eq. 3 <sup>a</sup>	2000	136	99.0	-61–98	This study
Eq. 4 <sup>a</sup>	2000	136	102.6	-62–91	This study
Direct extrapolation <sup>b</sup>	2000	136	144.2 <sup>d</sup>	±128	This study
Direct extrapolation <sup>b</sup>	2000	136	164.6 <sup>e</sup>	±125	This study
Direct extrapolation <sup>b</sup>	2000	136	136	±274	Gu et al. 2007
Direct extrapolation <sup>b</sup>	1995	104	127.1	±106	Yan et al. 2003
Direct extrapolation <sup>b</sup>	1990	95	95	±274	Li et al. 2001
DNDC model	1990	95	160	-	Li et al. 2001
Empirical model <sup>c</sup>	1997	136	92.78	±55	Lu et al. 2006

<sup>a</sup>Eqs. 1–4 in Table 2 were applied for estimating the part of background emission from cultivated mineral soils. The part from organic soils was estimated by directly extrapolating the emission factor for temperate organic soils of 8 (ranging from 2 to 24) kg N ha<sup>-1</sup> yr<sup>-1</sup> (IPCC 2006) to the grid area of cultivated organic soils. The BNE<sub>n</sub> was estimated as the sum of both parts.

<sup>b</sup>BNE<sub>n</sub> estimated by multiplying a mean value of measured BNE<sub>h</sub> with the national total cropland area.

<sup>c</sup>BNE<sub>h</sub> = 1.49P (Lu et al. 2006), wherein BNE<sub>h</sub> and P denote annual background N<sub>2</sub>O emission on a per hectare basis (kg N ha<sup>-1</sup> yr<sup>-1</sup>) and annual precipitation (m), respectively. This model only applies for upland soils, and background emission during the paddy rice growing season was excluded for the national total estimates given here.

<sup>d</sup>Calculated with the mean BNE<sub>h</sub> value of the observations in China (Table 1).

<sup>e</sup>Calculated with the mean BNE<sub>h</sub> value of the observations around the world including those in China (Table 1).

<sup>f</sup>Uncertainty at the 95% confidence interval.

organic soils. For the national cropland area of 136 Mha in the year 2000, our BNE<sub>n</sub> estimates integrated both mineral and organic soils in this study, amounting to 99.0–116.9 Gg N yr<sup>-1</sup>. Compared to estimates based mainly on soil properties, those resulting from direct extrapolation of national or global mean BNE<sub>h</sub> to national total cropland area in the year 2000, were significantly higher ( $p < 0.05$ ) by ca. 38% (ranging from 16% to 66%). This is the case even though the contribution from cultivated organic soils was excluded. This indicates that ignoring the regulatory effects of the local soil properties on BNE<sub>h</sub> in direct extrapolation may overestimate the national total background N<sub>2</sub>O emissions. This overestimation is most likely due to the disregard of spatial variation of background emissions over a national scale. Therefore, including the regulatory effects of soil properties is necessary for accurately estimating the total national background N<sub>2</sub>O emissions.

Lu et al. (2006) proposed an empirical model to predict N<sub>2</sub>O emissions from upland agricultural soils, in which precipitation was the key factor for stimulating BNE. Using their model, Lu et al. (2006) presented a BNE<sub>n</sub> estimate of 92.78 Gg N yr<sup>-1</sup> for the

year 1997. This estimate was 6–21% lower than our results obtained from soil properties (Table 3). However, Gu et al. (2007) reported that annual rainfall might only be a key stimulating factor at relatively smaller scales (e.g., Eastern China), and might not be significant at the national scale of China. Thus, regarding rainfall as a key factor over the national area might introduce considerable uncertainty for the BNE<sub>n</sub> estimate. In addition, the estimate from Lu et al. (2006) excluded the BNE from paddy rice fields. This exclusion might have underestimated the BNE<sub>n</sub>. Nitrogen sources, such as biological nitrogen fixation (at a rate of 14–50 kg N ha<sup>-1</sup> yr<sup>-1</sup>; Kundu and Ladha 1995) and atmospheric nitrogen deposition, might have yielded considerable background N<sub>2</sub>O emissions from paddy rice fields.

Using DNDC, a process-oriented biogeochemical model, Li et al. (2001) simulated background N<sub>2</sub>O emissions at a county level and then calculated a BNE<sub>n</sub> estimate of 160 Gg N yr<sup>-1</sup> for a cropland area of 95 Mha for the year 1990. If their estimate was converted to a comparable cropland area of 136 Mha in the year 2000, the DNDC-based result would be 229 Gg N yr<sup>-1</sup>. This is 96–132% higher than our

estimates from soil property and 39–68% higher than the estimates from direct extrapolation of a mean BNE<sub>h</sub> (Table 3). This difference might be caused by high temporal resolution of the DNDC simulation, which is conducted on a daily basis. In comparison, all of the approaches for the non-DNDC estimates, listed in Table 3, were based on low-frequency field measurements. For instance, Eqs. 1–4 were obtained from field measurements with sampling frequencies of twice per week or less (Gu et al. 2007). A low frequency may sometimes lead to an underestimation of BNE<sub>h</sub> (e.g., Zheng et al. 2004) although its effects upon chamber-based measurements are not certain (Smith and Dobbie 2001; Stehfest and Bouwman 2006; Parkin 2008). Thus, there is a possibility that our estimates have underestimated the BNE<sub>n</sub>. However, another possibility is that DNDC has overestimated the BNE<sub>n</sub>. The DNDC model has never been adequately validated in China for simulating of background N<sub>2</sub>O emissions from croplands. In addition using the N<sub>2</sub> as a carrier gas in the GC-ECD system might induce overestimation of annual N<sub>2</sub>O emissions (Zheng et al. 2008). Although a few could be corrected in this study by using the correction terms in Zheng et al. (2008), many of the BNE observations extracted from references provide too little information to make a justified correction (Table 1; e.g., Kaiser et al. 1998; Weitz et al. 2001; Sehy et al. 2003). This suggests that the models based on TN or SOC presented in this study might be further modified after taking into account the overestimation of the measured BNE<sub>h</sub> data. The national estimates of BNE from mineral soils, calculated using Eqs. 1–4 in this study, were ca. 17% (ranging from 15% to 18%) less than those using the raw empirical models in Gu et al. (2007). Further studies of BNE with high-frequency field measurements that avoid the use of N<sub>2</sub> as a carrier gas in the GC-ECD system, as well as verification/validation of process-oriented models such as DNDC with respect to BNE simulation, will help elucidate these issues.

By adding the fertilizer-induced N<sub>2</sub>O emissions at 275 Gg N yr<sup>-1</sup> (the mean estimate for the 1990s; Zheng et al. 2004) to the amounts of BNE estimated from soil properties (i.e., 99.0–116.9 Gg N yr<sup>-1</sup>) the national total N<sub>2</sub>O emissions from agricultural soils in China would amount to 374.0–391.9 Gg N yr<sup>-1</sup>. The BNE component accounted for 26–30% of these estimates. The BNE contribution to the national total

amount of N<sub>2</sub>O from croplands in China was approximately 50% lower than the contribution at a global scale (Bouwman et al. 2002; Stehfest and Bouwman 2006). The higher contribution from the global BNE might be due to the large percentage of unfertilized global cropland area, which was as much as 32% (Bouwman et al. 2002; Stehfest and Bouwman 2006). In China, however, there was practically no unfertilized cropland in use. The lower contribution of the BNE in China might also be caused by the high application rates of nitrogen fertilizers (Zheng et al. 2002; Yan et al. 2003). The potentially large contribution from BNE indicates of the importance of accurately quantifying BNE.

#### Uncertainties in estimates of BNE<sub>n</sub> from cultivated soils

In geostatistics, the soil property uncertainties depend on the number of soil profiles within a given region. The TN and SOC uncertainties (at 95% confidence interval), in the top 20–30 cm soils in China, fell within 47–116% and 50–360%, respectively, from village to province scales (Cheng et al. 2004; Zhao et al. 2005; Wu and Cai 2006). In our estimation, the TN uncertainties (at 95% confidence interval) within a grid cell were within –100–6,671%, with a mean of –47–171%; while the SOC uncertainties (at 95% confidence interval) were within –100–19,305%, with a mean of –50–215%. The uncertainty ranges in our estimation of soil properties are somewhat wider than those from previous studies are, where these were based on soil profiles at different regional scales. However, the extent to which the magnitudes of uncertainty of soil properties estimated in this study represents the actual uncertainties for a grid cell remains unclear. This is because the 10 km × 10 km grid size, for the GIS database, is low in resolution relative to the high spatial variation of real soils. Furthermore, the uncertainty of the cropland area in each grid cell was not considered in the uncertainty assessment due to a lack of data. These shortcomings can only be overcome by populating the GIS database with additional data for soil properties and cropland area.

The BNE from organic soils was estimated to account for 1.5–1.8% of the BNE<sub>n</sub> estimates, although organic soils comprised less than 0.2% of the national croplands. This implies that N<sub>2</sub>O emissions from organic soils should not be ignored when

compiling the national BNE inventory of China. However, field measurements of BNE from this cultivated soil type are yet to be undertaken. Therefore, further studies, beginning with field measurements, are required for BNE from cultivated organic soils in China.

As Table 3 shows that the uncertainties of the BNE<sub>n</sub> estimates based on the soil properties were narrower than those estimated by direct extrapolation of a BNE<sub>h</sub>, as well as those from previous studies. The uncertainties of the BNE<sub>n</sub> estimates based on TN were narrower than those based on SOC. In principle, to judge whether or not an empirical model is good, there are at least two criteria: a) being as simple as possible; and, b) being capable of yielding an estimated/predicted uncertainty that is as small as possible. Following these criteria, we recommend the monivariate model based on TN, shown as Eq. 2, for estimating regional or national BNE from croplands with mineral soils.

## Conclusion

In this study, we predicted the national background N<sub>2</sub>O emission (BNE<sub>n</sub>) from croplands in China in the year 2000. The BNE from cultivated mineral soils was calculated by coupling four empirical monivariate models, based on soil total nitrogen content (TN) and soil organic carbon content (SOC), within a 10 km×10 km GIS framework. We then directly extrapolated a hectare-based annual background N<sub>2</sub>O emission (BNE<sub>h</sub>) rate of the IPCC (2006) default value to the organic soil area. Because of the small proportion of organic soil area relative to the national total croplands, the organic soils contributed only 1.5–1.8% of the BNE<sub>n</sub> in the year 2000. The approach adopted in this study could estimate the spatial distribution of BNE over the national region. With regard to BNE<sub>n</sub> from cultivated mineral soils, the models based on TN could yield comparable national estimates to those from the models based on SOC. Moreover, the models based on TN yield much smaller uncertainties for the national estimates, compared to the models based on SOC or other approaches, such as direct extrapolation of a mean BNE<sub>h</sub> rate to the national scale. Accordingly, we recommend the models based on TN, which could simulate BNE with the smallest uncertainty for regional or national cultivated mineral soils.

However, efforts are still required to further improve the BNE<sub>n</sub> predictions in this study. The efforts may focus on the following aspects. First, further parameterization of the models based on soil properties would require high-frequency field measurements that cover diurnal and day-to-day variations of BNE. It would be better to conduct the high-frequency field measurements with reliable GC-ECD methods. Second, field measurements are required to further parameterize the approach for simulating BNE from cultivated organic soils of China, or to further parameterize the models based on soil properties so as to improve their capability in simulating BNE<sub>h</sub> of cultivated mineral soils with extremely low TN or SOC contents. Finally, a GIS database, containing uncertainty information of soil properties and cropland area, is required to assess the uncertainty of national estimates from bottom-up models based on soil properties.

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