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REGULAR ARTICLE

Estimating N_2O emissions from soils under natural vegetation in China

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Abstract

Background Natural and managed soils have been identified as the largest sources of atmospheric nitrous oxide (N_2O). However, the quantification of N_2O emissions from soils under natural vegetation in China and their possible responses to changing climate and atmospheric nitrogen deposition remains uncertain. In particular, information regarding N_2O emissions from Chinese shrublands is lacking.

Method This study used 28 sets of N_2O field measurements in China to validate a process-based dynamic nitrogen cycle model (DyN-LPJ), which was then used to investigate the N_2O fluxes from soils under natural vegetation in China from 1970 to 2009.

Results N_2O emissions from Chinese forests, grasslands, and shrublands in the 2000s were estimated to be 0.10 ± 0.06 Tg N $yr.^{-1}$, 0.09 ± 0.09, Tg N $yr.^{-1}$ and 0.14 ± 0.07 Tg N $yr.^{-1}$, respectively. Monthly N_2O fluxes were linearly correlated with precipitation, and exponentially ($Q_{10} = 3$) with air temperature. The total N_2O fluxes from natural terrestrial ecosystems in China increased from 0.28 ± 0.03 Tg N $yr.^{-1}$ in the 1970s to 0.46 ± 0.03 Tg N $yr.^{-1}$ in the 2000s. Warming and atmospheric nitrogen deposition accounted for 37% (or 0.07 ± 0.03 Tg N) and 63% (0.11 ± 0.01 Tg N) of this increase respectively.

Conclusions Our results indicate that when compared to grassland ecosystems, N_2O emissions from forest and shrubland ecosystems contain larger uncertainties due to either their uncertain areal extent or their emission rates. Long-term and continuous field measurements should be conducted to obtain more representative data in order to better constrain shrubland N_2O emissions.

Keywords N_2O · Natural terrestrial ecosystems · China · Shrubland · Forest · Grassland · Atmospheric nitrogen deposition · Climate change

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Introduction

Nitrous oxide (N_2O) is a potent long-lived greenhouse gas with a global warming potential that is nearly 300 times greater than that of carbon dioxide (CO_2), and is a major contributor to stratospheric ozone destruction (Wuebbles 2009). Atmospheric concentrations of N_2O have been increasing rapidly for a number of decades. Recent analyses indicate that the increase is caused by disturbance of the natural N cycle and associated N_2O production through nitrification and denitrification (Ciais et al. 2014; Kroeze et al. 1999). Estimates of the global total emission from soils under natural vegetation vary from 3.3 to 9.9 Tg N $yr.^{-1}$. This amount is similar to the sum of all anthropogenic sources, including agriculture (Ciais et al. 2014; Denman et al. 2007).

The land area in China is approximately 9.6×10^{12} m^2, accounting for 7% of the global terrestrial land area (Hou et al. 1982). The cropland area is about 1.68×10^{12} m^2 according to Vegetation Map of China (VMC) (1:4000000) (Hou et al. 1982) (http://westdc.westgis.ac.cn) and accounts for 17.5% of the Chinese land area. N_2O emissions from cultivated soils are most often estimated as the sum of two independent parts, i.e., background and fertilizer-induced emissions (Bouwman 1996; Bouwman et al. 2002; Stehfest and Bouwman 2006; Yan et al. 2003). The N_2O emissions from Chinese cropland including background emissions have been estimated to be 0.15 Tg N $yr.^{-1}$ by Chen et al. (2000), 0.37 Tg N $yr.^{-1}$ (range: 0.29–0.47 Tg N $yr.^{-1}$) by Cai (2012) and 0.34 Tg N $yr.^{-1}$ (range: 0.17–0.51 Tg N $yr.^{-1}$) by using the DeNitrification-DeComposition (DNDC) model (Li et al. 2001); The emissions at 0.20 Tg N $yr.^{-1}$ (Yan et al. 2003) and 0.28 Tg N $yr.^{-1}$ (range: 0.06–0.65 Tg N $yr.^{-1}$) (Zheng et al. 2004) were also estimated, when excluding the background emissions. Large uncertainties still exist in the estimation of background emissions for China. For example, using the revised 1996 IPCC Guidelines for National Greenhouse Gas Inventories, Yan et al. (2003) estimated emissions at 0.13 Tg N $yr.^{-1}$, while Li et al. (2001) estimated emissions at 0.18 Tg N $yr.^{-1}$ using the DNDC model, and the emissions were 0.10–0.12Tg N $yr.^{-1}$ based on observational data (Gu et al. 2007; Gu et al. 2009). Background N_2O emissions across different soil types and climate regions are significantly regulated by the climate, but the current IPCC approach does not consider these effects (Gu et al. 2009; Lu et al. 2006).To date, a number of studies have estimated natural ecosystem soil N_2O emissions. For example, a direct extrapolation from field studies estimated the emissions at 0.42 Tg N $yr.^{-1}$ (Cai 2012), which were very similar to process-based models estimates of 0.44 Tg N $yr.^{-1}$ (Lu and Tian 2013). Emissions from Chinese unmanaged grassland soils were estimated ranging from 0.04 to 0.13 Tg N $yr.^{-1}$ through direct extrapolations (Cai 2012; Chen et al. 2000; Huang et al. 2003) and using process-based models (Tian et al. 2011; Zhang et al. 2010). In contrast, the uncertainty range for Chinese forest soil emissions is much larger, ranging from 0.09 Tg N $yr.^{-1}$ (Chen et al. 2000) to 0.29 ± 0.17 Tg N $yr.^{-1}$(Cai 2012). In addition, the soil N_2O emissions from Chinese shrubland has never been estimated as a separate entity (Cai 2012). These major uncertainties are from either uncertain natural vegetation coverage data or uncertain emissions rates for specific vegetation types. Especially for Chinese forest ecosystems, there are only a few soil N_2O emission observations and even less for tropical and subtropical forest ecosystems before the 2000s.

Atmospheric N deposition rates have dramatically increased with the increase of anthropogenic nitrogen use (Liu et al. 2013; Lü and Tian 2007). This increase in atmospheric N deposition may have altered the terrestrial N cycle by increasing the rates of the microbial processes nitrification and denitrification and thereby the leaching of soil mineral N, such as NO_3^- concentration and N_2O emissions (Lu et al. 2011). Relatively little is known about how the increase in N deposition have altered N_2O emissions from natural ecosystems in China.

In this study we used a process-based dynamic nitrogen cycle model (DyN-LPJ) that has previously been comprehensively evaluated using 66 sets of measurements of total annual N_2O emissions from global natural ecosystems (Xu-Ri et al. 2012). We used an additional 28 sets of measurements covering 18 sites of grassland and forest ecosystems in China to further calibrate the model. The calibrated model was then used to quantify the N_2O emissions from the soils under natural vegetation and the background emissions from croplands in China. The contributions from grasslands, forests, and shrublands to total natural soil N_2O emissions in China was analyzed and their emission responses to changing climate and atmospheric N deposition were quantified.

Materials and methods

Model description

DyN-LPJ (Xu-Ri and Prentice 2008) is based on the Lund–Potsdam–Jena (LPJ) dynamic global vegetation modelling framework (Sitch et al. 2003) (Fig. 1). In addition to the coupled carbon and water cycling and vegetation dynamics simulated in LPJ, DyN includes following sub-models: (1) plant N uptake, (2) plant N allocation, turnover, reproduction, and mortality, (3) plant and soil N mineralization, (4) biological N_2-fixation, (5) nitrification, (6) NH_3 volatilization, (7) nitrate leaching, (8) denitrification, and (9) N_2, N_2O and NO production and emission. Sub-model (2) uses an annual time step; the others use a daily time step. The model runs at a spatial resolution of 0.5° × 0.5° (Xu-Ri and Prentice 2008). N_2O fluxes were modelled as byproducts of nitrification and denitrification. Nitrification is assumed to occur in aerobic microsites within the top 50 cm of soil, while denitrification is assumed to occur in anaerobic microsites within the same soil layer. Soil moisture (water filled pore space, WFPS) was used as an indicator to allocate substrates into two soil fractions with different aeration status. Nitrification and denitrification rates were then calculated directly from substrate availability (NH_4^+ for nitrification; labile C, NO_3^- and NO_2^- for denitrification) and soil environmental factors (soil temperature and WFPS). The rate of each transformation was regulated by the substrate concentrations (NH_4^+, labile C, NO_3^- or NO_2^-) following Michaelis–Menten kinetics and soil temperature. The detailed functions and parameters for nitrification and denitrification are documented in Xu-Ri and Prentice (2008). We applied the maximum fraction of N_2O losses from nitrification and denitrification as 0.05% (R_{N2ON}) and 1.8%(R_{N2ODN}), respectively (Xu-Ri et al. 2012).

Model evaluation

Twenty-eight sets of measurements, covering 18 sites were used in this study for model evaluation (Fig. 2). As shown in Table 1, the first 5 sites have been used in a previous global-scale evaluation (Xu-Ri et al. 2012), the additional 23 sets of measurements of annual N_2O emissions (covering 13 sites), conducted during the period 2003–2006, were used in this study for model calibration for grassland, forest, desert and alpine steppe

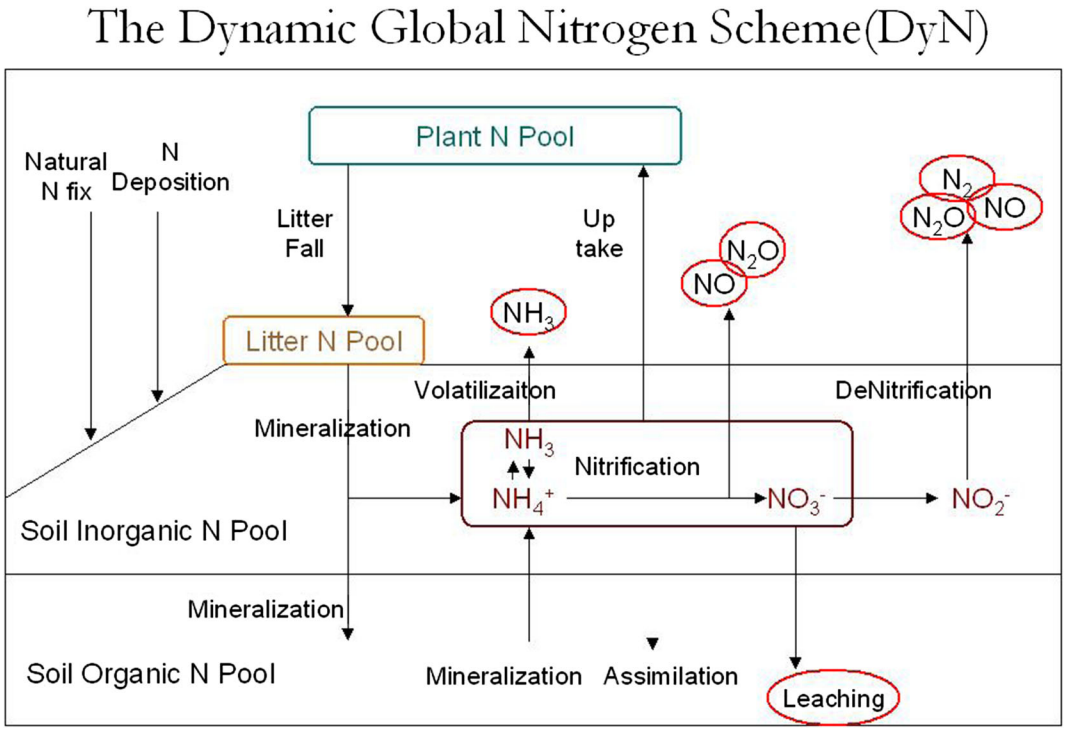

Fig. 1 Model structure for the process-based model DyN-LPJ

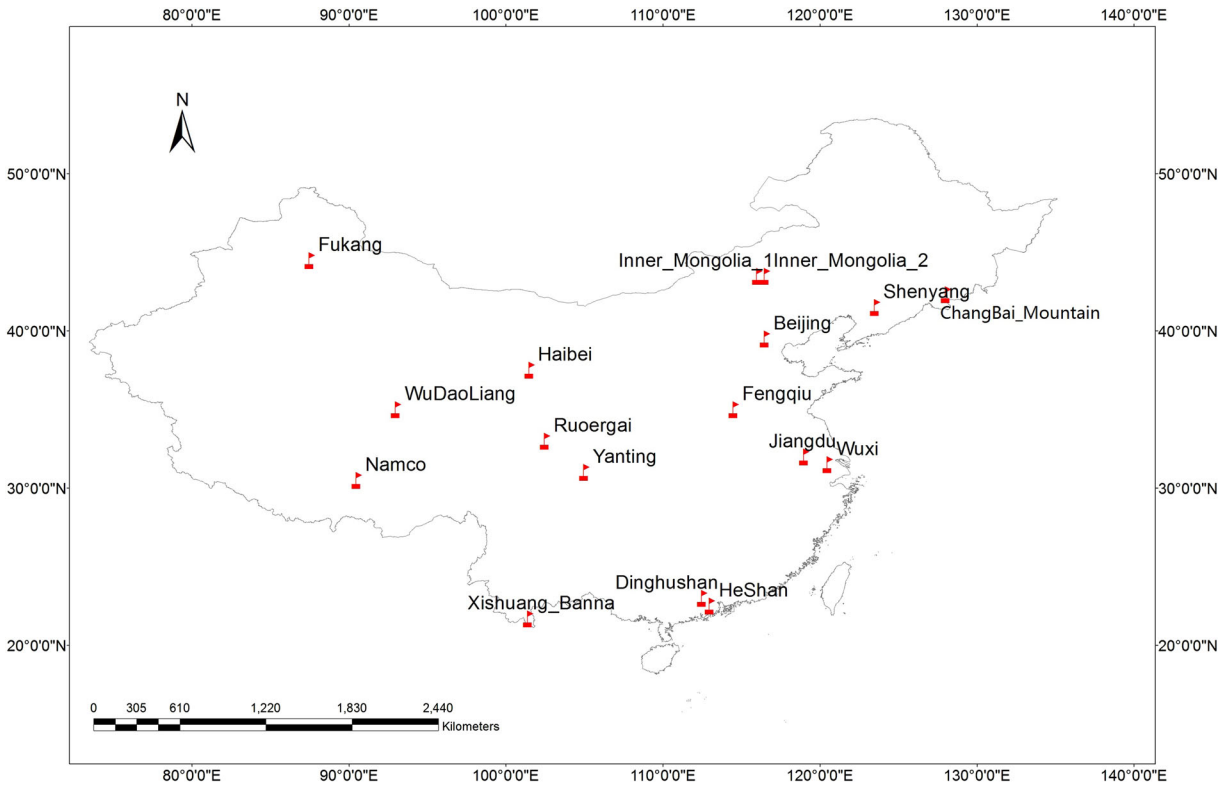

Fig. 2 Locations of the soil N_2O measurements from different terrestrial ecosystems in China, used for model evaluation; details provided in Table 1

ecosystems in China (Table 1, Fig. 2). These measurements were all conducted in the field, using closed chamber techniques and gas chromatography. Three gas flux measurement locations within each of the above sites were established randomly and N_2O fluxes were typically measured at the same time period (usually mid-morning, 10:00–12:00 h) of each sampling day and 1–2 times per week during the growing season and 1–2 times per month during winter.

The N_2O fluxes were determined using opaque static chambers with subsequent analysis on a gas chromatograph (GC) fitted with an electron capture detector (ECD). In studies 6–22 and 24–28 the carrier gas used was nitrogen gas (N_2). However, Zheng et al. (2008) showed that only using N_2 as carrier gas can overestimate the N_2O concentration. The reason is an interference of the presence of large concentrations of carbon dioxide (CO_2) in the gas samples collected. To combat this problem ascarite was added to the carrier gas stream to remove the CO_2 from the air samples. By comparing the N_2 only (the DN method) with the N_2-Ascarite method, Zheng et al. (2008) proposed correction terms for N_2O flux measurements. Using the correction terms, we adjusted the N_2O fluxes for studies No. 6–22 (Table 1). Emissions in study 24–28 had already been corrected by Gu et al. (2009), using the same method, and studies 1–5 and 23 did not require any correction.

For comparison, annual N_2O emissions were simulated at the geographic location of each site following the transient simulation protocol described below. The simulated results were compared with the specific grid cell location and specific year of each measurement. The simulated cropland background N_2O emissions represent the potential natural soil N_2O emissions in cropland areas.

Historic simulations for China

The transient model runs were set up using identical parameter values, model protocols and forcing to the simulations described by Xu-Ri et al. (2012) except that the transient runs were extended to 2009. TS 3.10.1 climate data (http://www.cru.uea.ac.uk/cru/data/hrg/) from the Climatic Research Unit and updated

Table 1 Observed annual N_2O emissions from China for comparison with the model results

No.	Long.	Lat.	Vegetation type	Year observed	N_2O fluxes (uncorrected data) kg N ha^{-1} $yr.^{-1}$ ± SD	N_2O fluxes (corrected data[1]) kg N ha^{-1} $yr.^{-1}$ ± SD	Location	Sources
Soils under natural vegetation, used for model calibration in Xu-Ri et al. (2012)								
1*	93	35	Alpine steppe	2000		0.07	Wudaoliang, Tibet	(Pei 2003)
2*	116.5	39.5	Temperate forest	1997–1998		0.28	Beijing	(Sun and Xu 2001)
3*	127	41.5	Alpine tundra	1994–1995		0.28	ChangBai Mountain	(Chen et al. 2000)
4*	116.0	43.5	Temperate steppe	1995		0.27	Inner Mongolia	(Chen et al. 2000)
5*	116.5	43.5	Temperate steppe	1998		0.37	Inner Mongolia	(Xu-Ri et al. 2003)
Soils under natural vegetation								
6	101.5	37.5	Alpine meadow	2003	2.55 ± 1.91	0.23 ± 0.31	Qinghai, Haibei	Data provided in this study
7	101.5	37.5		2004	1.84 ± 1.71	0.16 ± 0.29		
8	101.5	37.5		2005	2.67 ± 2.76	0.92 ± 1.03		
9	101.5	38	Alpine shrub	2003	2.37 ± 1.25	0.70 ± 0.47		
10	101.5	38		2004	1.90 ± 1.80	0.51 ± 0.46		
11	102.5	33	Alpine swamp	2004	0.67 ± 1.15	0.30 ± 0.53	Ruoergai, Sichuan	
12	102.5	33		2005	0.73 ± 0.77	0.31 ± 0.45		
13	87.5	44.5	Temperate desert	2004	0.83 ± 1.68	0.31 ± 1.24	Fukang, Xinjiang	
14	87.5	44.5		2005	0.25 ± 1.66	0.05 ± 0.96		
15	113	22.5	Subtropical forest	2004	3.14 ± 3.83	1.60 ± 3.26	HeShan, Guangdong	
16	113	22.5		2005	3.78 ± 4.20	2.26 ± 3.66		
17	102	22	Tropical forest	2003	3.01 ± 2.66	1.22 ± 0.90	Xishuang banna, Yunnan	
18	102	22		2004	1.82 ± 1.47	0.84 ± 0.92		
19	102	22		2005	1.61 ± 1.88	1.05 ± 0.92		
20	112.5	23	Tropical forest	2003	3.46 ± 2.67	1.51 ± 1.20	Dinghushan, Guangdong	
21	112.5	23		2004	3.80 ± 2.86	1.39 ± 0.89		
22	112.5	23		2005	4.72 ± 2.70	2.06 ± 1.06		
23	90.5	30.5	Alpine steppe	2009		0.01 ± 0.02	Namco, Tibet	(Wei et al. 2012)
Background emissions from croplands								
24*	123.5	41.5	Maize–spring wheat	2004–2005		1.12	Shenyang Liaoning	(Gu et al. 2007) and subsequently corrected by (Gu et al. 2009)
25*	114.5	35	Winter wheat–maize	2004–2006		0.67	Fengqiu Henan	
26*	119	32	Winter wheat–rice	2004–2006		1.00	Jiangdu Jiangsu	
27*	120.5	31.5	Winter wheat–rice	2002–2004		1.70	Wuxi, Jiangsu	
28*	105	31	Winter wheat–rice	2005–2006		1.28	Yanting Sichuan	

*the value provided for site 1–5 and site 24–28 are directly cited from the original literature, SD values were not available

atmospheric CO_2 concentration data from Keeling et al. (2009) were used. For a model spin-up period of 2000 years, we used climate input from 1901 to 1930. Thereafter, the model was run with transient monthly climate data from 1901 to 2009. These transient simulations were performed at both site level and national-scale.

Six historic simulations were performed: (1) all factors including CO_2, climate and N deposition varied throughout the twentieth century, (2) CO_2 and climate varied without N deposition, (3) climate varied but kept CO_2 constant, (4) only temperature varied, (5) only precipitation varied, (6) only cloud cover varied.

In simulation (4), (5) and (6), CO_2 was kept at 296 ppm, while the "fixed" climate variables were held at their 1901–1920 averages. Simulations were conducted for the period of 1901 to 2009. The simulations for the period 1970 to 2009 were analyzed.

The atmospheric N deposition data for China for the years 1860 to 1993 were taken from Dentener (2006) and were linearly interpolated to daily time steps and extrapolated to the period after 1993. The datasets include annual N deposition rates for ammonium and nitrate and were added to the soil ammonium and nitrate compartment. This simple approach estimates the magnitude, trend, and distribution of N deposition during the twentieth century. We recognize these estimates will not realistically represent true N deposition dynamics but will allow us to approximately assess the effects of atmospheric N deposition on N_2O emission rates. The land areas of natural terrestrial ecosystems in China were extracted from the Vegetation Map of China (1:4000000) (Hou et al. 1982) (http://westdc.westgis.ac.cn), as shown in Fig. S1 and Table 2. In this vegetation map, crop type includes the staple crops (rice, wheat, maize), economic crops, fruit orchards, and economic forests.

Results

Comparison between simulated and observed N_2O fluxes

Our 28 sets of measurements cover 18 sites (Fig. 2), comprising alpine steppe, alpine tundra, alpine meadow,

Table 2 N_2O emissions from terrestrial ecosystems in China

		Vegetation type	Area ($\times 10^{12}$ m^2)	Average fluxes (2000–2009) (kg N ha^{-2} $yr.^{-1}$ ± SD*)	Total fluxes (Tg N $yr.^{-1}$)
Natural vegetation	Forests	Temperate, coniferous forest (TeConf)	0.168	0.183 ± 0.069	0.003 ± 0.001
		Subtropical coniferous forest (SubTrConf)	0.541	0.595 ± 0.456	0.032 ± 0.025
		Temperate deciduous forest (TeDecf)	0.309	0.281 ± 0.205	0.009 ± 0.006
		Tropical and subtropical deciduous forest (TrDecf)	0.440	1.181 ± 0.669	0.052 ± 0.029
	Total		1.458		0.096 ± 0.061
	Shrublands	Temperate shrublands (TeShrub)**	1.225	0.405 ± 0.384	0.050 ± 0.047
		Tropical and subtropical shrublands (TrShrub)***	0.846	1.073 ± 0.206	0.091 ± 0.022
	Total		2.070		0.141 ± 0.069
	Grasslands	Temperate desert (TeDes)	1.467	0.159 ± 0.220	0.023 ± 0.032
		Temperate grasslands and meadow (TeGras)	1.034	0.467 ± 0.344	0.041 ± 0.029
		Alpine grassland and meadow (AlGras)	1.090	0.096 ± 0.101	0.011 ± 0.011
		Subtropical grassland (SubTrGras)	0.051	1.266 ± 1.078	0.006 ± 0.006
		Temperate and alpine swamp (Te-AlSwa)	0.054	0.206 ± 0.178	0.001 ± 0.001
	Total		3.695		0.090 ± 0.086
Total cropland, background emissions		Crop (Crop)	1.682	0.784 ± 0.471	0.132 ± 0.079
	Total		1.682		0.132 ± 0.079
No vegetation area		No vegetation (NoVege)	0.518	0.126 ± 0.141	0.007 ± 0.007
Lakes		Lakes (Lakes)	0.065	0.552 ± 0.447	0.004 ± 0.003
Total			9.489		0.462 ± 0.298

*Standard deviations are for the spatial variation of simulated N_2O emissions per grid cell

**The vegetation types within the TeShrub includes temperate and subtropical deciduous shrub, brushwood and low forest

***The vegetation types within the TrShrub include the tropical and subtropical evergreen shrub, brush wood and low forest

alpine shrubland, alpine swamp, temperate desert, temperate steppe and temperate forest, tropical forest and subtropical forest (Table 1, No. 1–23). The remaining five sites were background emissions from cultivated soils (Table 1, No. 24–28).

Simulated and observed annual N_2O fluxes were highly correlated either with ($R^2 = 0.75$, $n = 28$, $p < 0.001$) or without ($R^2 = 0.70$, $n = 28$, $p < 0.001$) N deposition. However, the slope was closer to the 1:1 line when atmospheric N deposition was considered (slope = 0.94, as shown in Fig. 3), especially for the tropical and subtropical forest areas, and the background emissions from cultivated soils, where the N_2O emissions rate was higher than 1 kg N ha^{-1} $yr.^{-1}$(Table 1, Fig. 3).

We compared the observed and simulated monthly soil N_2O emissions (kg N ha^{-1} $month^{-1}$) for two temperate grasslands in Inner Mongolia in 1998 and Haibei in 2004; and two tropical forests sites in Heshan in 2004 and DingHushan in year of 2005. The correlation coefficients of these measurement and model results are 0.93 (Inner Mongolia), 0.86 (Haibei), 0.65 (Heshan), and 0.68 (Dinghushan), respectively (Fig. 4). The model generally captured the seasonal variation of soil N_2O emissions in monsoon climate regions of China, which is highly consistent with the precipitation seasonality (Fig. S2).

N_2O emissions correlated linearly and exponentially with precipitation and temperature on either monthly or annual time scale, respectively (Fig. 5). The linear and exponential correlation at monthly time step was significant ($R^2 = 0.64$, $R^2 = 0.45$, $n = 96$, $p < 0.01$). The

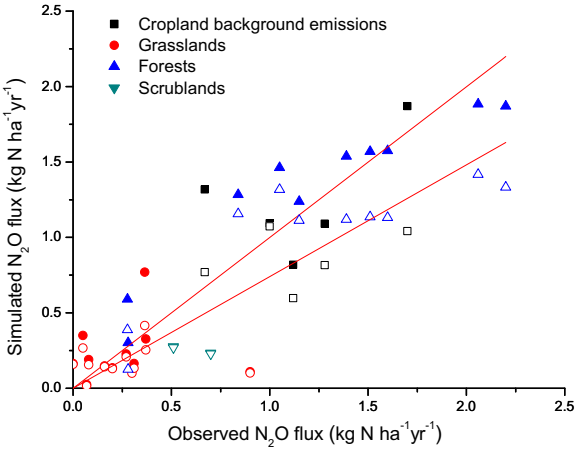

Fig. 3 Observed versus simulated annual N_2O fluxes from soils of terrestrial ecosystems in China, before (open symbols) and after (closed symbols) atmospheric N deposition was included

exponential fit showed a Q_{10} (the factor by which the rate increases with a 10 °C rise in temperature) value of 2.99, with a temperature range between 10 °C and 20 °C. Our model simulations of N_2O fluxes correlated well with temperature and precipitation at monthly and annually time scales. Large soil N_2O emissions occurred when the soil moisture and temperature were high. Some of the discrepancies between modeled and simulated N_2O fluxes on the monthly time scale were expected as the observation were conducted at the site scale while our climate data for driving the model are at 0.5° × 0.5° spatial resolution. Additionally, our model might have failed to capture the enhanced soil N_2O emissions during spring thawing events and the soils N_2O uptake during the non-growing season (Fig.4).

N_2O emissions from natural terrestrial ecosystems in China in the 2000s

N_2O emissions were highest in the humid tropics but limited by low temperatures in temperate and boreal ecosystems, and by low soil moisture in the desert areas of China (Figs. 6a, b). Tropical and subtropical deciduous forests, shrubland, grassland and coniferous forests were predicted to emit 1.18 ± 0.67, 1.07 ± 0.21, 1.27 ± 1.08, and 0.60 ± 0.46 kg N_2O-N ha^{-1} $yr.^{-1}$, respectively (Table 2). The N_2O emissions of temperate deciduous forests, shrubland, grassland, deserts, swamps and coniferous forests were predicted to be 0.28 ± 0.21, 0.41 ± 0.38, 0.47 ± 0.34, 0.16 ± 0.22, 0.21 ± 0.18 and 0.18 ± 0.07 kg N_2O-N ha^{-1} $yr.^{-1}$, respectively. Alpine grassland and meadow, with a very low emissions rate, were predicted to be 0.01 ± 0.10 kg N_2O-N ha^{-1} $yr.^{-1}$. The potential natural soil N_2O emissions from the cropland area were predicted as 0.78 ± 0.47 kg N_2O-N ha^{-1} $yr.^{-1}$ (Fig. S3).

When we aggregated the fluxes for different ecosystems according to their specific distribution maps (Fig. S1), we found that N_2O emissions from shrubland played an important role either in temperate or in subtropical regions, because of their relatively larger distribution area when compared to forest and grassland (Fig. S4, Table 2). The total emissions from shrublands were 0.14 ± 0.07 Tg N, and N_2O emissions from forest and grassland amounted to 0.10 ± 0.06 and 0.09 ± 0.09 Tg N, respectively (Fig. 7, Table 2). The potential background N_2O emissions from cropland areas were 0.13 ± 0.08 Tg N (as shown in Table 2 and Fig. 7). The total

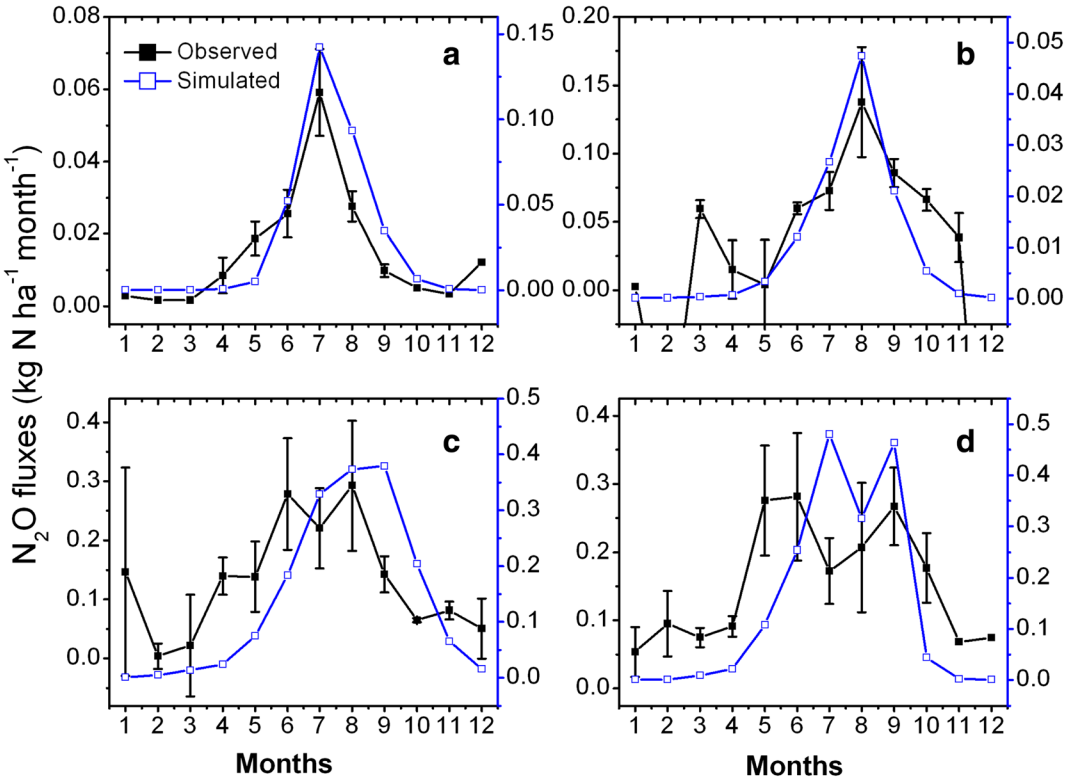

Fig. 4 Comparison of observed (closed symbols) and simulated (open symbols) monthly soil N_2O emissions (kg N ha^{-1} $month^{-1}$) from temperate grasslands: (**a**) Inner Mongolia (1994) and (**b**) Haibei (2004); and tropical forests: (**c**) Heshan 2004 and (**d**) DingHushan 2005)

amount of N_2O emissions from the soil of natural terrestrial ecosystems in China in the 2000s was estimated to be 0.46 ± 0.30 Tg N with, and 0.35 ± 0.34 Tg N without N deposition (Table 2).

Responses of N_2O emissions in China to a changing climate and atmospheric N deposition

The total amount of N_2O emissions from natural terrestrial ecosystems in China increased from 0.28 ± 0.03 Tg N in the 1970s (average for 1970–1979) to 0.46 ± 0.03 and 0.35 ± 0.02 Tg N with and without considering N deposition, respectively, in the 2000s (average for 2000–2009). Warming and atmospheric N deposition accounted for 37% (or 0.07 Tg N) and 63% (or 0.11 Tg N) of the increase in N_2O, respectively, while the effects of precipitation, cloud and atmospheric CO_2 concentrations were negligible (Fig. 8).

Atmopsheric N deposition rates during the period 2000–2010 (2000s) were estimated to be 8.36 ± 8.56 Tg N. The N deposition rates used were an average of 10.19 kg N ha^{-1} $yr.^{-1}$ and a maximum of 40 kg N ha^{-1} $yr.^{-1}$ (Fig. S5). Their distribution patterns and magnitudes were closer to the observation based estimates of 12.89 kg N ha^{-1} $yr.^{-1}$ by Lü and Tian (2007). The N_2O increase resulting from atmospheric N deposition was 0.11 ± 0.01 TgN (Figs. 6a, b), with an average emission factor of 1.19% (Fig. S6). Similarly, the increase of 0.17 Tg N from Lü and Tian (2007) might be due to their higher estimated atmospheric N deposition for China.

N_2O emissions versus net primary productivity

Satellite derived net primary productivity (NPP) has been used as an important variable to simulate global soil N_2O emissions (Potter and Klooster 1998; Potter et al. 1996). In this study, we examined the relationships between the modeled N_2O emissions and NPP on both spatial and temporal scales (Fig. 9). For China, spatially, the modeled soil N_2O emissions (g N m^{-2} $yr.^{-1}$) were highly correlated with NPP (kg C m^{-2} $yr.^{-1}$) with a slope

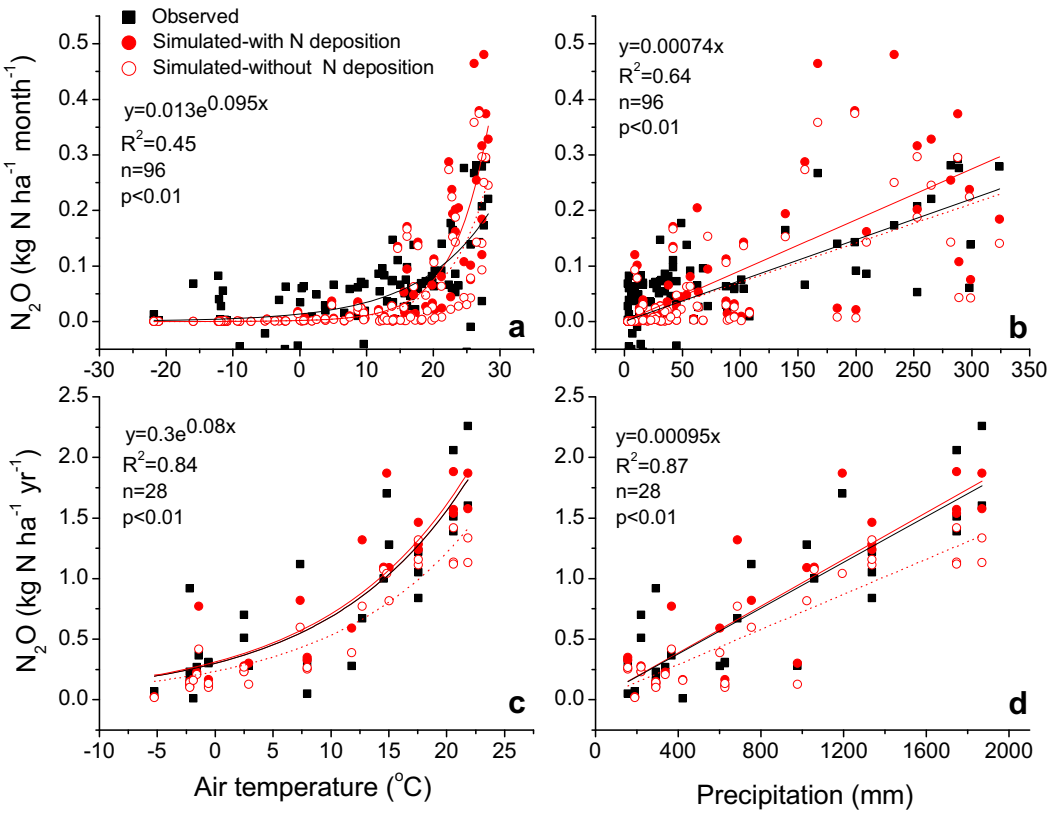

Fig. 5 Comparison between observed and simulated monthly (**a**, **b**) and annual (**c**, **d**) soil N_2O emissions (kg N ha^{-1} $month^{-1}$) with monthly and annual air temperature (**a**, **c**) and precipitation (**b**, **d**). Open circles do not include atmospheric N deposition rates, and closed circles represent data were N deposition was included. Monthly comparison includes sites 5, 7, 10, 11, 13, 15, 18, 21 and the annual comparison includes all 28 sites listed in Table 1

of 0.13 ($R^2 = 0.83$, $p < 0.001$). Temporally, the correlations were also significant with a slope of 0.17 ($R^2 = 0.99$, $p < 0.001$).

Discussions

N_2O emissions from grassland ecosystems

N_2O emissions from grassland ecosystems have been estimated in many studies, either by direct extrapolation, ranging from 0.04 to 0.13 ± 0.16 Tg N (Cai 2012; Chen et al. 2000; Huang et al. 2003), or via process-based model estimations, using either the DNDC model (0.08 ± 0.01 Tg N; Zhang et al. 2010), or the Dynamic Land Ecosystem Model (DLEM) model (0.10 ± 0.01 Tg N; Tian et al. 2011). Our estimation of 0.09 ± 0.09 Tg N falls within the above ranges; but from the perspective of the annual average emissions rate, the estimated value of 0.24 ± 0.23 kgN ha^{-1} $yr.^{-1}$ (Table 3) is closer to the value of 0.22 ± 0.04 kgN ha^{-1} $yr.^{-1}$ estimated by the DNDC model (Zhang et al. 2010).

N_2O emissions from forest ecosystems

Our model estimates of N_2O emissions from forest ecosystems were 0.10 ± 0.06 Tg N and were similar to the emissions of 0.09 ± 0.09 Tg N from grassland soils (Table 3). But, forest ecosystems cover a smaller area of China and per unit area therefore have a higher emission rate of 0.66 ± 0.42 kg N ha^{-1} $yr.^{-1}$ compared to grasslands (0.24 ± 0.23 kg N ha^{-1} $yr.^{-1}$). Our estimate is consistent with those of Chen et al. (2000), 0.10 Tg N, but much lower than that of Cai (2012), 0.29 ± 0.19 Tg N. The higher estimate of Cai (2012) may be partly because most of the N_2O emissions in China during the 2000s were measured using the DN-method, as detailed in section "Model evaluation".

This study includes three observation sites in tropical and subtropical forests: Guangdong Heshan, Guangdong

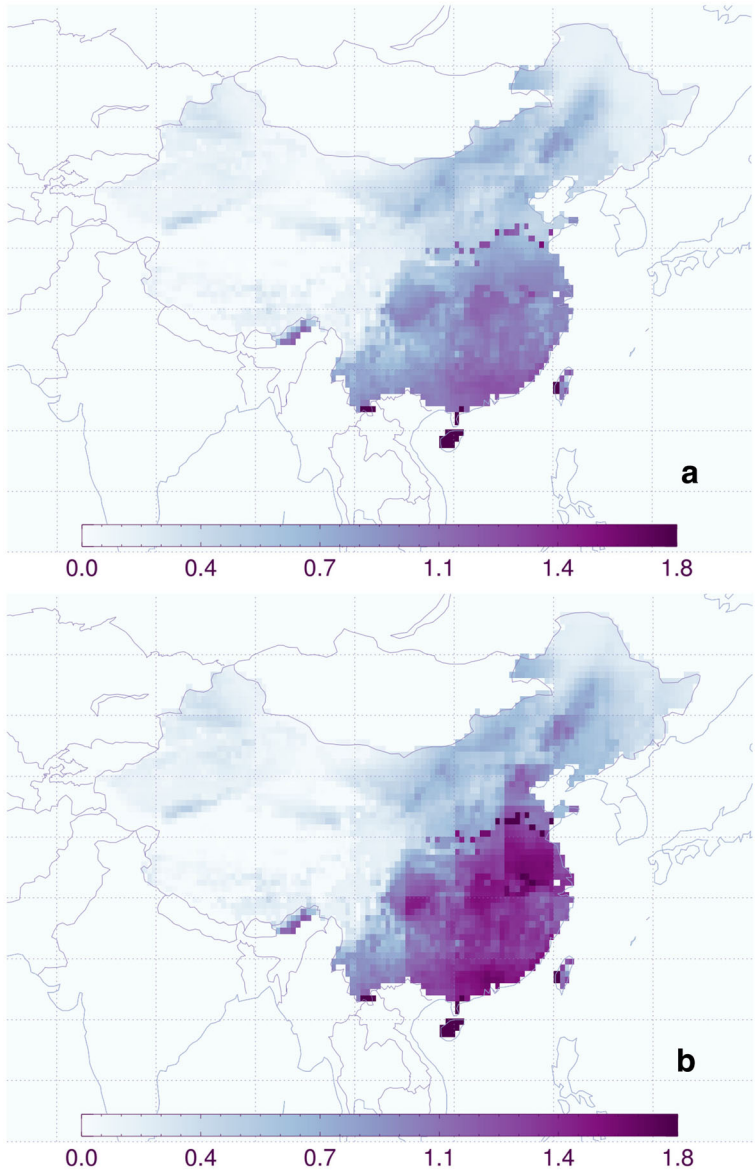

Fig. 6 Spatial distribution of N_2O fluxes (kg N ha^{-1} $yr.^{-1}$) from China (**a**) without and (**b**) with considering atmospheric N deposition rates, disaggregated to 0.5° × 0.5°

DingHuShan, and Yunnan Xishuangbanna (Table 1). The annual precipitation at these sites ranged between 1300 mm and 1900 mm, the annual air temperature between 17.5 °C and 22.0 °C, and annual N_2O fluxes between 0.84 to 2.26 kg N ha^{-1} $yr.^{-1}$(Table 1). These values fall within the range of N_2O fluxes 0.88 to 4.36 kg N ha^{-1} $yr.^{-1}$ for tropical forests worldwide (Stehfest and Bouwman 2006; Werner et al. 2007; Xu-Ri et al. 2012). Annual N_2O emissions were generally less than 2 kg N ha^{-1} $yr.^{-1}$ when annual precipitation was lower than 2000 mm and annual temperature was lower than 22 °C (Xu-Ri et al. 2012) (Fig. S7). However, there are still large uncertainties in N_2O emission estimates for tropical forests in China. For example, emissions ranging from 2.7 to 3.4 kg N ha^{-1} $yr.^{-1}$ have been reported based on short-term observations (Zhu et al. 2013). Future long-terms and year-round continuous measurements are needed to reduce these large uncertainties.

N_2O emissions from shrubland

N_2O emissions from the shrubland ecosystems are not well researched globally and have not previously been estimated for China. In this study, after model

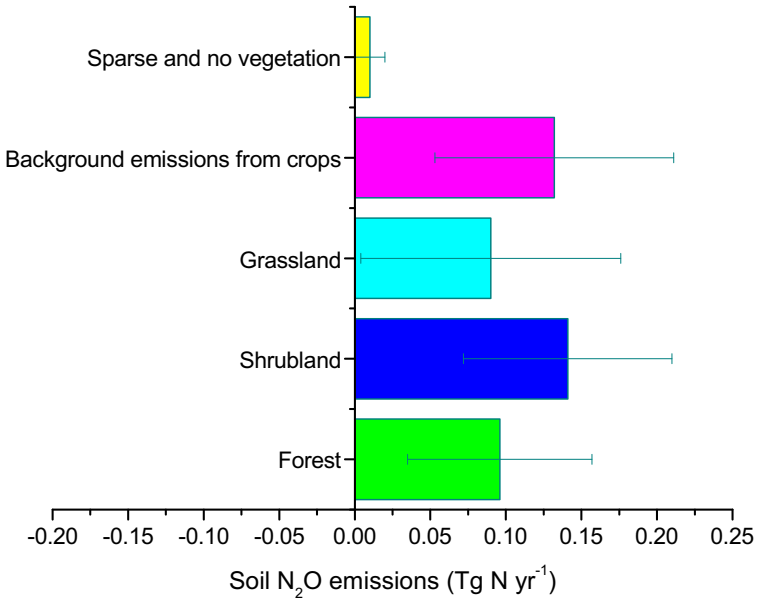

Fig. 7 Comparison of total annual N_2O fluxes, averaged over the period 2000–2009 from the natural ecosystems in China and background emissions from crops

calibration against forest and grassland ecosystems in China, N_2O emissions from shrubland ecosystems were calculated at 0.14 ± 0.07 Tg N, in which temperate shrublands contributed 0.05 ± 0.05 Tg N (with an average annual N_2O emissions of 0.41 ± 0.38 kg N ha^{-1} $yr.^{-1}$) and tropical/subtropical shrubland contributed approximately 0.09 ± 0.02 Tg N (with annual emissions of 1.07 ± 0.21 kg N ha^{-1} $yr.^{-1}$) (Table 3). Recent N_2O flux measurements from an alpine shrub meadow in China ranged from 0.18 to 0.27 kg N ha^{-1} $yr.^{-1}$ (Fu et al. 2018) and falls within our uncertainty range for temperate shrubland. The values for the temperate shrublands are also roughly consistent with those from European temperate shrublands (0.14 to 0.42 kg N ha^{-1} $yr.^{-1}$; Carter et al. 2012).

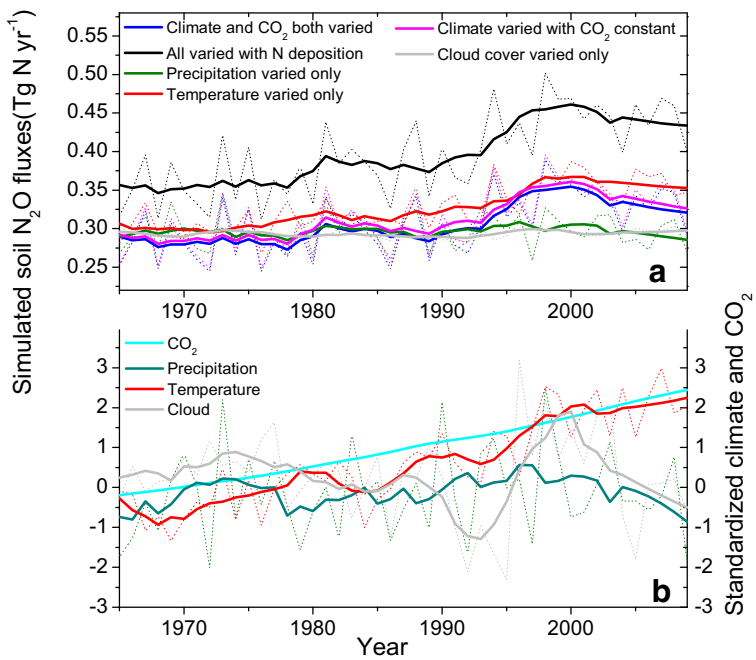

Fig. 8 **a** N_2O emissions from soils under natural vegetation in China for the period 1970–2009. Dashed lines represent year-to-year variations and full lines smoothed with 10-year splines (**b**) Observed atmospheric CO_2 concentration and climate variables in China: annual means over all land grid points in China, as 10-year splines of standardized values

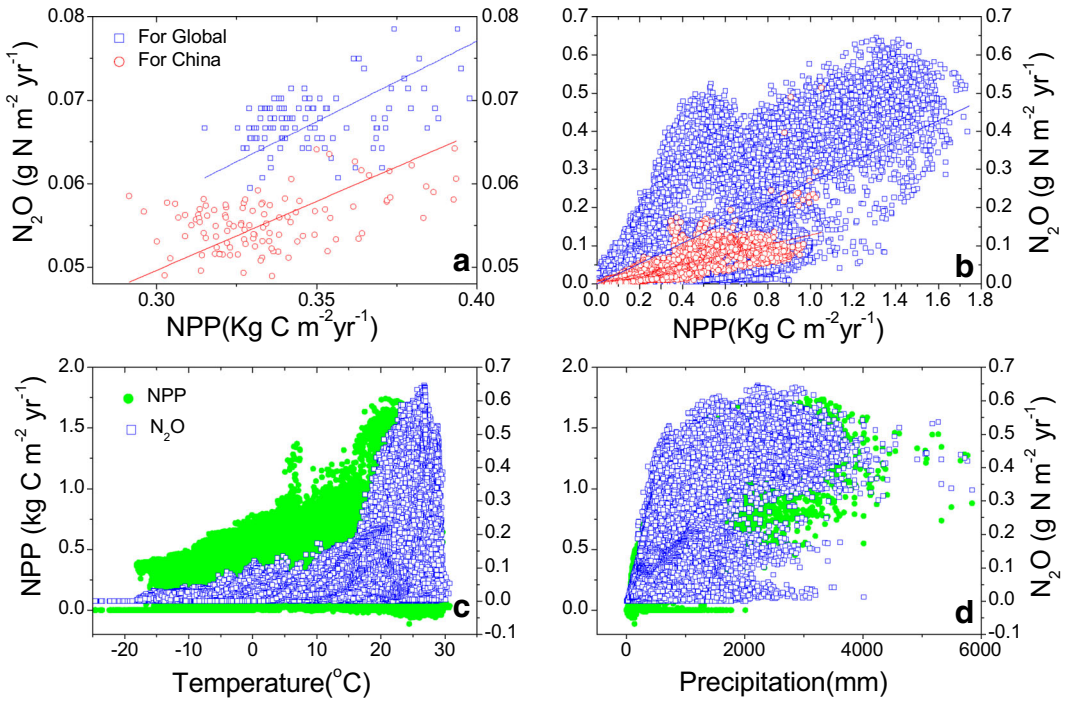

Fig. 9 Comparison between simulated N_2O emissions and NPP for: (**a**) average annual values for 1901–2009, and (**b**) the spatial variation at the global scale and for China in 1990s. Spatial comparison between model simulations of N_2O emissions and NPP with spatial patterns of temperature (**c**) and precipitation (**d**)

Background N_2O emissions from cropland

Background N_2O emissions are defined as emission from cultivated soil that has not received nitrogen fertilizer during the current season or year (Gu et al. 2007; Zheng et al. 2004). The background N_2O emissions from croplands may originate from the following nitrogen sources: natural microbial nitrification and denitrification processes from soils driven by natural N fixation; atmospheric N deposition, mineralization of soil organic matter or crop residues (Gu et al. 2007). Bouwman (1996) reported a background emissions rate of 1.0 kg N ha^{-1} $yr.^{-1}$, which was based on five sets of field observations conducted in European and American unfertilized grasslands. The spatial variation of background N_2O emissions proved to be highly climate dependent (Gu et al. 2009; Lu et al. 2006).

In the present study, background N_2O emissions from cropland are referring to the potential natural soil N_2O emissions on cropland areas. After calibrating against the site scale observations (Gu et al. 2009), we estimated an average background N_2O emission from cropland of 0.13 ± 0.08 Tg N (Fig. 7, Table 3), with an average flux rate of 0.79 ± 0.47 kg N ha^{-1} $yr.^{-1}$ for the period 2000 to 2009. This result falls within the uncertainty range of 0.72–1.66 kg N ha^{-1} $yr.^{-1}$ reported by previous studies (Li et al. 2001; Gu et al. 2009) and is close to the IPCC default value of 1 kg N ha^{-1} $yr.^{-1}$ (Lu et al. 2006).

N_2O emissions from natural ecosystems in China

Our estimate of average N_2O emissions from natural terrestrial ecosystems using the model DyN-LPJ was 0.46 ± 0.23 Tg N for the period 2000 to 2009. This estimate is comparable with a previous estimate of 0.44 Tg N, calculated from the difference between total terrestrial ecosystem emissions (0.81 Tg N) and those from N fertilized soils (0.37 Tg N) in China (Lu and Tian 2013). Cai (2012), estimated a similar value of 0.42 Tg N for total emissions from terrestrial ecosystems, but only forest and grassland emissions and not background emissions from croplands were included (Table 3). Since the1970s, the average N_2O emissions from natural ecosystems have increased from 0.28 to 0.46 Tg N $yr.^{-1}$ in the 2000s. These values are similar to even exceed the changes of agricultural fertilizer induced emissions, which were about, 0.10 in the 1980s (Gao et al. 2011),

Table 3 Summary of estimated soil N_2O emissions from terrestrial ecosystems in China

Ecosystem	Estimates (Tg N $yr.^{-1}$)	Area ($\times 10^{12}$ m^2)	Rate (kg N ha^{-1} $yr.^{-1}$ ± SD)	Description	Reference
Forest	0.10	1.34	0.70	Direct extrapolation	(Chen et al. 2000)
	0.29 ± 0.19	1.20	2.41 ± 1.55	Direct extrapolation	(Cai 2012)
	0.10 ± 0.06	1.46	0.66 ± 0.42	DyN-LPJ model	This study
Grassland	0.11	2.87	0.39	Direct extrapolation[a]	(Chen et al. 2000)
	0.04	2.87	0.14	Direct extrapolation[a]	(Huang et al. 2003)
	0.08 ± 0.01	3.37	0.22 ± 0.04	DNDC model	(Zhang et al. 2010)
	0.10 ± 0.01	1.73–1.74	0.58 ± 0.06	DLEM model[a]	(Tian et al. 2011)
	0.13 ± 0.156	4.82	0.27 ± 0.33	Direct extrapolation[b]	(Cai 2012)
	0.07 ± 0.01	3.37	0.20 ± 0.03	Direct extrapolation	(Fu et al. 2018)
	0.09 ± 0.09	3.70	0.24 ± 0.23	DyN-LPJ model[c]	This study
Shrubland	0.14 ± 0.069	2.07	0.68 ± 0.33	DyN-LPJ model	This study
Background emissions from cropland[d]	0.18	0.95	1.66	DNDC model	(Li et al. 2001)
	0.13	1.04	1.22	Direct extrapolation	(Yan et al. 2003)
	0.09	1.36	0.68	Empirical model	(Lu et al. 2006)
	0.11–0.18	1.36	0.84–1.35	Empirical model	(Gu et al. 2007)
	0.10–0.16	1.36	0.72–1.21	Direct extrapolation	(Gu et al. 2009)
	0.13 ± 0.08	1.68	0.79 ± 0.47	DyN-LPJ model	This study[e]
Natural terrestrial ecosystems in China	0.44	n.a.	n.a.	DLEM model[e]	(Lu and Tian 2013)
	0.42	6.01	0.70	Direct extrapolation[f]	(Cai 2012)
	0.46 ± 0.30	9.49	0.49 ± 0.30	DyN-LPJ model	This study

[a] Temperate grassland only

[b] part of the shrublands were included

[c] deserts included

[d] the background N_2O emissions obtained in this study including the potential natural soil N_2O emissions in cropland areas

[e] derived from the estimation for total terrestrial ecosystem emissions of 0.81 Tg N, minus the N_2O emissions from N fertilizations of 0.37 Tg N for China

[f] Background emissions from cropland not included

0.28 Tg N $yr.^{-1}$ in the 1990s (Lu et al. 2006; Zheng et al. 2004) and 0.31 Tg N $yr.^{-1}$ in the 2000s, primarily due to global warming and the increasing rates of atmospheric N deposition (Fig. 8).

Uncertainties in estimating N_2O emissions considering land use change

Our model simulations assume a constant land area for terrestrial ecosystems in China. The Vegetation Map of China (1:4000000) (http://westdc.westgis.ac.cn) represents the potential distribution of Chinese terrestrial ecosystems during the 1970s. However, the land areas of grassland, forest and cropland are not constant. Remote sensing products indicate that, during the 2000s, the Chinese cropland area had decreased, and forest and grassland areas had expanded. The limited data of changes in Chinese shrubland areas with time, have shown that Chinese cropland areas decreased from 1.68×10^{12} to $1.36–1.54 \times 10^{12}$ m^2 (Liu and Tian 2010; Xiu et al. 2014), while grasslands areas increased from 3.70×10^{12} to $4.03–4.20 \times 10^{12}$ m^2 (He and Shi 2015; Xiu et al. 2014), and forest areas from 1.46×10^{12} to $1.63 \pm 0.22 \times 10^{12}$ m^2 (Liu and Tian 2010; Wang et al. 2015) (Table 4). These changes in land areas are related to the Chinese national policy of "returning cropland to forest and grassland" and ecological environment restoration (He and Shi 2015).

Table 4 Soil N_2O emissions from grasslands, forests and background emissions from croplands in China before and after considering land use change

No.	Average emission rate (kg N ha^{-2} $yr.^{-1}$ ± SD)	Area ($\times 10^{12}$ m^2) according to vegetation map of China*	Area ($\times 10^{12}$ m^2) during the 2000s	Fluxes assuming fixed land area (Tg N $yr.^{-1}$)	Fluxes under land area change (Tg N $yr.^{-1}$)	Change	Land area sources
Grassland	0.24 ± 0.23	3.70	4.10 ± 0.07	0.09 ± 0.09	0.10 ± 0.00	+ 0.01	(Xiu et al. 2014), (He and Shi 2015)
Forest	0.66 ± 0.42	1.46	1.63 ± 0.22	0.10 ± 0.06	0.11 ± 0.01	+ 0.01	(Liu and Tian 2010), (Wang et al. 2015)
Background emissions from cropland	0.79 ± 0.47	1.68	1.46 ± 0.07	0.13 ± 0.08	0.12 ± 0.01	- 0.01	(Xiu et al. 2014), (He and Shi 2015), (Liu and Tian 2010),
Total change						+ 0.01	

*The Vegetation Map of China (1:4000 000) (http://westdc.westgis.ac.cn), this map representing the vegetation conditions of 1970s

The N_2O emission estimates calculated from the new land area extent during the period of 2000s for grassland, forest and cropland suggest that the increasing N_2O emissions due to increasing grassland and forest areas, might be offset by decreasing background emissions due to decreasing cropland area (Table 4). However, the overall estimates still fall within the uncertainty range of our estimates based on constant land area for various types of ecosystems.

Here we also examined vegetation area differences between various data sources (Table S1). The vegetation area of shrubland used in this study are 2.07×10^{12} m^2 based on the VMC (1:4000000), which is closer to the shrubland area of 2.15×10^{12} m^2 used in Piao et al. (2009) and 2.09×10^{12} m^2 according to the VMC (1:1000000) in Wang et al. (2017). We observed a large discrepancy in natural vegetation area extent between VMC and GLC 2000. Especially for the shrublands area, a large decrease in shrublands is obviously due to a systematic mismatch between two sources, VMC is mainly based on the field survey and the GLC2000 is mainly derived from the remote sensing production. Consequently, the large uncertainties in shrubland N_2O emissions come from land area uncertainties for various ecosystems, while the limited amount of observational data of N_2O emissions also contributed to the uncertainty of model extrapolation.

N_2O emissions and their controls

The observed soil N_2O fluxes were found to be significantly correlated with precipitation and temperature at seasonal and annual time scales (Fig. 5), which is consistent with other field studies (Smith et al. 2018). Nitrous oxide is produced naturally through nitrification and denitrification by soil microorganisms. Nitrification is an aerobic process in which ammonium (NH_4^+) is oxidized to nitrate (NO_3^-) and with nitric oxide (NO) and N_2O as byproducts. Denitrification is an anaerobic process in which NO_3^- is reduced to the denitrification products N_2O and N_2. In the DyN-LPJ model, soil moisture (represented as water filled pore space, WFPS) is used as an indicator of N_2O production rates by either nitrification or denitrification pathways (Xu-Ri and Prentice 2008). The close correlation between WFPS and N_2O emission rates is supported by many field observations (Bai et al. 2014; Fu et al. 2018; Gütlein et al. 2018). The fraction of N_2O loss from gross

denitrification can be 10 times larger than the maximum rate of N_2O production from nitrification processes (Xu-Ri and Prentice 2008). Both nitrification and denitrification process are highly temperature-dependent, with a Q_{10} value of ~2.0 to 4.0 over the range 15–25 °C. The exponential relationship between temperature and N_2O production has been well documented in field studies (Griffis et al. 2017; Song et al. 2018; Zhou et al. 2018). The above discussed empirical relations were implemented in the DyN-LPJ model (Xu-Ri and Prentice 2008).

Nitrous oxide fluxes were nonlinearly controlled by both precipitation and temperature (Fig. 9, Fig. S7) at the global scale and for China. However, the N_2O fluxes cannot be simply predicted by either of these single factors (Fig. 9, Fig. S7). The net primary productivity of CO_2 (NPP) was distributed similarly under the control of precipitation and temperature on the spatial scale (Fig. 9). These results verify the findings of a previous study in which satellite-derived NPP was used to predict global N_2O emissions (Potter et al. 1996). More recently, a close correlation between ANPP (Aboveground Net Primary Productivity) and N_2O fluxes was also observed in a field study (Zhang et al. 2018); The underlying mechanism is that ecosystem N loss (e.g., N_2O emissions) is affected by the rate of N fixation, while the NPP is one of the good indicators for the ecosystem scale N fixation (Xu-Ri and Prentice 2017). However, the correlation coefficients need to be calibrated locally according to local measurements when the upscaling of N_2O emissions is based on NPP data. On the temporal scale, however, NPP is a less reliable predictor, both globally and for China, because NPP more strongly responds to the increasing CO_2 concentration (Cramer et al. 1999; Xu-Ri et al. 2012).

On a decadal timescale, temperature appeared to be the dominant factor for natural soil N_2O emissions (Fig. 8). This could be due to the fact that in the twentieth century, there was no significant variation in annual precipitation, whereas the air temperature has significantly increased in China (Fig. 8).

Conclusions

This study compiled 28 sets of measurements of N_2O emissions from natural ecosystems in China, including temperate grassland, alpine grassland, temperate forest and tropical forest. The process-based dynamic nitrogen cycle model, DyN-LPJ, was calibrated using these data. The verified model captured observed seasonal and annual variability of N_2O fluxes and their significant correlations with precipitation and temperature very well. For the whole of China and globally a close correlation between NPP and soil N_2O fluxes from natural ecosystems became evident.

The average N_2O emissions from natural terrestrial ecosystems in China during 2000–2009 were estimated to be 0.46 ± 0.30 and 0.35 ± 0.34 Tg N with and without atmospheric N deposition effects, respectively. N_2O emissions resulting from atmospheric N deposition was estimated to be approximately 0.11 ± 0.01 Tg N. The total Chinese natural terrestrial ecosystem soil emissions account for 5.0% of global N_2O emissions from natural soil sources. Shrublands may contribute to one-third of the total natural terrestrial ecosystem soil N_2O emissions in China, however are also the most uncertain in terms of areal extend and N_2O emission rates. Our uncertainty analysis suggests that long-term and continuous field measurements should be conducted to obtain more representative flux data to better constrain model estimates.

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