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Changes in fertilizer-induced direct N_2O emissions from paddy fields during rice-growing season in China between 1950s and 1990s

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Abstract

Nitrogen fertilizer-induced direct nitrous oxide (N_2O) emissions depend on water regimes in paddy fields, such as seasonal continuous flooding (F), flooding–midseason drainage–reflooding (F-D-F), and flooding–midseason drainage–reflooding–moist intermittent irrigation but without water logging (F-D-F-M). In order to estimate the changes in direct N_2O emission from paddy fields during the rice-growing season in Mainland of China between the 1950s and the 1990s, the country-specific emission factors of N_2O-N under different water regimes combined with rice production data were adopted in the present study. Census statistics on rice production showed that water management and nitrogen input regimes have changed in rice paddies since the 1950s. During the 1950s–1970s, about 20–25% of the rice paddy was continuously waterlogged, and 75–80% under the water regime of F-D-F. Since the 1980s, about 12–16%, 77%, and 7–12% of paddy fields were under the water regimes of F, F-D-F, and F-D-F-M, respectively. Total nitrogen input during the rice-growing season has increased from 87.5 kg N ha^{-1} in the 1950s to 224.6 kg N ha^{-1} in the 1990s. The emission factors of N_2O-N were estimated to be 0.02%, 0.42%, and 0.73% for rice paddies under the F, F-D-F, and F-D-F-M water regimes, respectively. Seasonal N_2O emissions have increased from 9.6 Gg N_2O-N each year in the 1950s to 32.3 Gg N_2O-N in the 1990s, which is accompanied by the increase in rice yield over the period 1950s–1990s. The uncertainties in N_2O estimate were estimated to be 59.8% in the 1950s and 37.5% in the 1990s. In the 1990s, N_2O emissions during the rice-growing season accounted for 8–11% of the reported annual total of N_2O emissions from croplands in China, suggesting that paddy rice development could have contributed to mitigating agricultural N_2O emissions in the past decades. However, seasonal N_2O emissions would be increased, given that saving-water irrigation and nitrogen inputs are increasingly adopted in rice paddies in China.

Keywords: agriculture, decadal and spatial variations, emission factor, greenhouse gas, modeling, nitrogen input, nitrous oxide, paddy rice, uncertainty, water regime

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Introduction

Nitrous oxide (N_2O) is one of the key greenhouse gases that cause global warming. It continues to rise at a rate of approximate 0.26% per year and has reached a concentration of 319 ppb (10^{-9} mol mol^{-1}) in 2005 (IPCC, 2007a). Agriculture accounts for about 60% of the global anthropogenic N_2O emissions. Globally, agricultural N_2O emissions have increased by nearly 17% from 1990 to 2005 (IPCC, 2007b), and are projected to increase by 35–60% up to 2030 due to increased nitrogen fertilizer use and increased animal manure production (FAO, 2003). The emission of N_2O that results from anthropogenic N inputs to agricultural soils occurs through a direct pathway (i.e. directly from soils to

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which the N is added), and through two indirect pathways: volatilization of compounds such as NH_3 and NO_x and subsequent redeposition, and through leaching and runoff. Relative to the indirect pathways, the direct emission contributes ~ 75% to the agricultural N_2O sources (e.g. Zheng *et al.*, 2004). Thus, a good estimate of direct N_2O emission from agricultural fields will help assess its global source strength.

Rice is the staple food for nearly 50% of the world's people, mainly in Asia. China is the most important rice-producing country in the world. Rough rice production in China contributes ~ 30% to the world total (IRRI, 2004). Its planting area accounts for about 20% of the world total and 23% of all cultivated land in China (Frolking *et al.*, 2002). Of which, ~ 93% is irrigated rice paddies; ~ 5% is distributed in rainfed lowlands, and ~ 2% in uplands (IRRI, 2004). Various water management regimes are currently practiced in China's rice paddies, such as seasonal continuous flooding (F), flooding–midseason drainage–frequent water logging with intermittent irrigation (F-D-F), and flooding–midseason drainage–reflooding–moist intermittent irrigation but without water logging (F-D-F-M) (Gao & Li, 1992; Huang *et al.*, 2004). Since the 1950s, an episode of midseason drainage for 7–10 days rather than continuous flooding has been commonly employed in China to inhibit ineffective tillers, remove toxic substances, and improve root activities (Peng *et al.*, 1998). Because of water resource scarcity in the North China Plain, water-saving irrigation and aerobic rice paddies instead of anaerobic paddies have been suggested as potential options for rice production in the past few years (Geng *et al.*, 2001).

Water regime often incurs a sensitive change in N_2O emission in rice paddies (Akiyama *et al.*, 2005). It is well documented that midseason drainage in rice paddies triggers substantial N_2O emission in contrast with continuous flooding (e.g. Cai *et al.*, 1997; Zheng *et al.*, 2000). In addition, N_2O fluxes during intermittent irrigation periods depend strongly on whether or not water logging is present in paddy fields, which often begets a significant difference in seasonal total of N_2O emissions between the water regimes of F-D-F and F-D-F-M (Zou *et al.*, 2005a).

Some studies have gone into quantifying N_2O emissions from rice paddies. A general approach is based on process-oriented models such as the Denitrification–Decomposition (DNDC) model (Li *et al.*, 2001, 2006; Li, 2007). The DNDC model has been frequently used to quantify N_2O emissions from rice paddies (Li *et al.*, 2002, 2004, 2005, 2006), and is expected to be capable of minimizing the uncertainty in estimates. However, it was originally developed for simulating carbon sequestration and trace gas emissions from upland fields. Relative to other methodology estimates, N_2O emissions from rice paddies in China predicted by the DNDC model were high, and showed high uncertainty range (e.g. Xing, 1998; Li *et al.*, 2004, 2005; Zheng *et al.*, 2004).

Besides the process-oriented model methodology, empirical models [i.e. emission factor (EF) methodology] are also recommended by the IPCC to estimate fertilizer-induced direct N_2O emissions (IPCC, 2006). The EF is defined as N_2O emission from nitrogen fertilizer plots minus the emission from unfertilized control plots (all other conditions being equal to those of the fertilized plots) expressed as a percentage of N applied. In the IPCC methodology, cropping-specific and country-specific emission factors are encouraged to be used where possible, in order to reflect the specific conditions of the country and the agricultural practices involved (IPCC, 2006).

Based on the IPCC methodology and summary of available data, some studies have provided an insight into fertilizer-induced N_2O emission factors and background emissions from rice paddies at the regional and global scales (Yan *et al.*, 2003; Zheng *et al.*, 2004; Akiyama *et al.*, 2005). Yan *et al.* (2003) estimated N_2O emission factors and background emissions in irrigated paddy fields during the rice-growing season, but they did not differentiate N_2O emissions under different water regimes. In contrast, Akiyama *et al.* (2005) recently reported that the EFs averaged 0.22% for the continuous flooding paddies and 0.37% for the fertilized paddies with midseason drainage. These EFs have been adopted by IPCC (2006) as recommended default values for which countries could calculate the national inventory of N_2O emissions from rice paddies. In the dataset employed to estimate N_2O emission factors by Akiyama *et al.* (2005), however, only five field studies were carried out in China (i.e. Cai *et al.*, 1997; Chen *et al.*, 1997; Hou *et al.*, 2000; Zheng *et al.*, 2000; Xiong *et al.*, 2002). In addition, some N_2O measurements from rice paddies commonly found under the F-D-F-M water regime were treated as statistical high outliers (e.g. Xing & Zhu, 1997; Zheng *et al.*, 2000), and thus they were excluded by Akiyama *et al.* (2005) as well. Consequently, estimates of emission factors and background N_2O emissions in these studies may not sufficiently reflect N_2O emissions from rice paddies in China where various water regimes are practiced.

In order to quantify the cropping-specific direct emission factor of N_2O dependent on water regime during the rice-growing season in China, we have compiled and statistically analyzed field data on N_2O emission from paddy fields. Thereby, some empirical models have been established to quantify direct emission factors and background emissions for N_2O under different water regimes (Zou *et al*., 2007). Here, we used these statistical models to quantify seasonal fertilizer-induced direct N_2O emissions from the rice paddies during the 1950s–1990s. We collected paddy rice production data (area, yield, water regime type, organic and chemical nitrogen inputs) from the database of National Greenhouse Gases Inventories of Agriculture. The objective of this study was to estimate changes in seasonal direct N_2O emission from paddy rice production in Mainland China between the 1950s and the 1990s.

Materials and methods

Statistical models on direct N_2O emission

Based on 71 measurements from 17 field studies on N_2O emission from paddy fields during the rice-growing season, the direct emission factor and background emission of N_2O dependent on water regime were specified by different empirical models [i.e. Eqns (1)–(3), Zou *et al*., 2007]. Specifically, seasonal total N_2O was, on average, equivalent to 0.02% of the nitrogen applied in the continuous flooding rice paddies [Eqn (1)]. Applying an ordinary least square (OLS) linear regression model resulted in an emission factor of 0.42% with a standard error of 0.06% for N_2O, and in negligible background N_2O emission under the water regime of F-D-F [Eqn (2)]. Under the F-D-F-M water regime, N_2O emission factor and background emission were, on average, estimated to be 0.73% and 0.79 kg N_2O-N ha^{-1} during the paddy rice-growing season, respectively [Eqn (3)].

$$\text{Model F}: \ N_2O\text{-N} = 0.0002(\pm 0.00003)\text{N}(\text{kg ha}^{-1}), \quad (1)$$

$$\text{Model F-D-F}: \ N_2O\text{-N} = 0.0042(\pm 0.0006)\text{N}(\text{kg ha}^{-1}), \quad (2)$$

$$\begin{aligned}\text{Model F-D-F-M}: \ N_2O\text{-N} &= 0.79(\pm 0.28) \\ &\quad + 0.0073(\pm 0.0011)\text{N}(\text{kg ha}^{-1}). \quad (3)\end{aligned}$$

Here, the EFs 0.0002, 0.0042, and 0.0073 kg N_2O-N kg N ha^{-1} were adopted to estimate the fertilizer-induced direct N_2O emissions from rice paddies under the water regimes of F, F-D-F, and F-D-F-M, respectively.

Input data of rice production during 1950s–1990s

County-level ground-based agricultural survey data for China during the 1950s–1990s were acquired from the database of National Greenhouse Gases Inventories of Agriculture, which was established by the Institute of Atmospheric Physics of the Chinese Academy of Sciences and the Nanjing Agricultural University. They were collected from official annals of agriculture and agricultural census data for each county. The database contains county-level statistics on paddy rice planting area, yield, water regime, percentage of crop residue retained, and manure and synthetic nitrogen fertilizer use during the rice-growing season in Mainland China from the 1950s to the 1990s. Rice paddies were assigned to five crop-zone regions in Mainland China (Frolking *et al*., 2002): I, Northeast/North; II, North China Plain and central/western; III, Southwest; IV, Middle and lower Yangtze River; and V, Southern China. No data were available for Taiwan, Hong Kong, and Macao. For each county, the census data were not used if the difference in any statistical items (e.g. area) between the census dataset and official annals was larger than 10% (37 survey data in total). Eventually, more than 300 survey samples were averaged on a provincial scale for each year. The datum of 1950s referred to an average of data from 1950 to 1959, similarly so for other decades.

Seasonal total nitrogen input was composed of crop residue nitrogen returned to soils, organic manure, and chemical nitrogen fertilizer inputs. Major types of chemical N fertilizer, such as urea, compound fertilizer, ammonium sulfate, and ammonium bicarbonate have been applied in rice paddies in China. Besides chemical nitrogen fertilizer application, organic manure and crop residue amendments are commonly used as basal fertilizer before the rice transplanting. Crop residues amended to paddy fields are wheat, barley, soybeans, vegetable, and rapeseed (Frolking *et al*., 2002). The nitrogen content of crop residue was derived from the IPCC guidelines, with an average of 0.45% (IPCC, 2006). Total nitrogen input of crop residue was estimated by multiplying the amount of crop biomass retained in the fields by their respective nitrogen contents. The nitrogen content of organic manure averaged 0.61% (Huang & Cai, 1997). Because the models [Eqns (1)–(3)] did not distinguish between organic- and inorganic nitrogen fertilizer-induced N_2O emissions, seasonal total nitrogen input was used to estimate direct N_2O emissions in this study.

To validate the model F-D-F-M, we collected seasonal N_2O emissions reported in 2005–2007 that were independent of those used to establish the model (Table 1). To check the quality of the input data, the yield of paddy rice was compared with that obtained from the IRRI (2004) dataset. Water regime and nitrogen application were validated against those gathered from Chinese monographs, conference reports, and journals (Appendix S1–S5).

Uncertainties in N_2O estimate

Total uncertainty in N_2O inventory is generally combined by uncertainties in emission factors and activity

Table 1 Observed and modeled seasonal N_2O emissions from paddy fields under the water regimes of F-D-F and F-D-F-M during rice growing season

Location	Year	N input (kg N ha^{-1})	Observed N_2O emission (kg N ha^{-1})	Modeled N_2O emission (kg N ha^{-1})	References
F-D-F					
Nanjing, Jiangsu province 32°00′N, 118°48E	1994	100	0.17	0.42	Xu *et al.* (1997)
		300	0.62	1.26	
		100	0.17	0.42	
		300	0.98	1.26	
	1994	0	0.14	0.01	Cai *et al.* (1997)
		100	0.17	0.43	
		300	0.80	1.27	
	2000	277	1.55	1.17	Zou *et al.* (2005a, b)
		295	1.43	1.25	
	2007	200	1.33	0.85	Zou *et al.* (unpublished results)
Ningxiang, Hunan province 28°12N, 112°27E	2006	160	0.65	0.67	Xiao *et al.* (2007)
		160	0.45	0.67	
		160	0.37	0.67	
F-D-F-M					
Heshan, Guangdong province 22.7°N, 112.9°E	2003	331	3.32	3.20	Liu *et al.* (2006a, b)
Yanting, Sichuan province, 31.3°N, 105.5°E	2002	133	1.66	1.76	Jiang *et al.* (2006)
	2003	133	3.21	1.76	
		133	1.62	1.76	
		133	1.75	1.76	
		133	2.06	1.76	
Jiaxing, Zhejiang province, 30.7°N, 120.8°E	2003	90	2.74	1.45	Huang *et al.* (2005)
		180	2.78	2.10	
		360	3.54	3.42	
Nanjing, Jiangsu province 32.0°N, 118.8°E	2003	300	3.80	3.20	Chen *et al.* (2005)
	2004	300	3.19	3.20	Chen *et al.* (2007a)
Shengyang, Liaoning province, 41.5°N, 123.4°E	2004	150	1.80	1.89	Zheng *et al.* (2006)
	2004	0	1.21	0.79	Wang *et al.* (2006)
		150	1.80	1.89	
Wangcheng, Hunan province 112.6°N, 28.5°E	2004	0	1.03	0.79	Qin *et al.* (2006a, b)
		0	0.20	0.79	
		75	0.83	1.34	
		90	1.60	1.45	
Sanjiang Plain, Helongjiang province 113.5°N, 47.8°E	2004–2005	150	1.84	1.89	Chen *et al.* (2007b)

F-D-F, flooding–midseason drainage–reflooding; F-D-F-M, flooding–midseason drainage–reflooding–moist intermittent irrigation but without water logging.

data. Similar to uncertainty estimate in the IPCC methodology (IPCC, 2006), we used the error propagation equation to calculate the uncertainties in seasonal N_2O emissions from rice paddies under each water regime as follows:

$$U_C = \sqrt{U_A^2 + U_E^2}, \tag{4}$$

where U_C is the combined uncertainty expressed as a percentage for each water regime; U_A and U_E are the percentage uncertainties for the activity data and emission factor, respectively. In this study, the activity data is the input data of the estimate method and U_A is principally determined by the reliability of nitrogen input data. The confidence interval of parameter estimates in the simulated model was used to calculate U_E. A confidence interval of 95% is suggested by the IPCC guidelines (2006), and thus U_E was expressed as half the 95% confidence interval divided by the mean. Eventually, the total uncertainty in seasonal N_2O estimates during the rice-growing season for each decade was calculated by

$$U_{\mathrm{total}} = \frac{\sqrt{(U_{\mathrm{F}} \times x_{\mathrm{F}})^2 + (U_{\mathrm{F\text{-}D\text{-}F}} \times x_{\mathrm{F\text{-}D\text{-}F}})^2 + (U_{\mathrm{F\text{-}D\text{-}F\text{-}M}} \times x_{\mathrm{F\text{-}D\text{-}F\text{-}M}})^2}}{x_{\mathrm{F}} + x_{\mathrm{F\text{-}D\text{-}F}} + x_{\mathrm{F\text{-}D\text{-}F\text{-}M}}}, \tag{5}$$

where U_{total} is the total uncertainty expressed as a percentage for each decade; x_i and U_i (i represents the water regimes of F, F-D-F, and F-D-F-M) are the uncertain quantities (i.e. N_2O estimates) and the percentage uncertainties [i.e. U_C in Eqn (4)] associated with them under different water regimes, respectively.

Results

Model validation

The model F-D-F-M was validated against independent N_2O flux measurements in China's paddy fields under the water regime of F-D-F-M that were reported in 2005–2007. These measurements were made in seven provinces (Guangdong, Helongjiang, Hunan, Jiangsu, Liaoning, Sichuan, Zhejiang provinces; Table 1), covering main rice cultivation regions except for the II region of North China Plain and central/western. The validation of the model F-D-F-M against observed N_2O flux suggests that the model F-D-F-M did well in estimating N_2O emissions from paddy fields during the rice-growing season, although the computed N_2O emissions were generally 13% lower than the observed fluxes (Table 1; F-D-F-M, $y = 0.87x$, $r^2 = 0.68$, $P < 0.001$). Higher N_2O emissions observed relative to the model is probably because the soil was only kept moist instead of water logging during the late growing stage of rice for a longer period in paddy fields than before.

On country scale, no independent N_2O emission measurements in the continuous flooding rice paddies or under the water regime of F-D-F were available to validate the F and F-D-F models. Instead, we used N_2O flux measurements in rice paddies under the water regime of F-D-F at Nanjing, Jiangsu province, and Ningxiang, Hunan province, to validate the model F-D-F. The validation results showed that N_2O field measurements can be well predicted by the model F-D-F (Table 1; F-D-F, $y = 0.99x$, $r^2 = 0.86$, $P < 0.001$). Using the global N_2O emission measurements shown in Table 1 by Akiyama *et al.* (2005), we estimated the EF to be 0.43% by the OLS model, which is close to the emission factor of N_2O produced by the model F-D-F (Model: MSD-Akiyama; Zou *et al.*, 2007). For the continuous flooding rice paddies, the model predicted seasonal N_2O emission accounting for 0.02% of the total N applied, which is similar to the results of earlier studies at other countries (0.01–0.05%; e.g. Smith *et al.*, 1982; Granli & Bøckman, 1994). Together these results suggest that the statistical models [Eqns (1)–(3)] could be introduced to estimate N_2O emissions from paddy fields during the rice-growing season in Mainland China.

Rice production data input

Changes in paddy rice area. Rice-planted area in Mainland China totaled about 29.2–29.4 million hectares in the 1950s–1960s. It was as high as 34.9 million hectares in the 1970s, 7% and 10% greater than in the 1980s and 1990s, respectively (IRRI, 2004). Relative to the 1970s, the decrease in area in the 1990s mainly occurred in the IV (Middle and lower Yangtze River) and the V (Southern China) crop-zone regions. In general, rice-planted area in the I and II regions only contributed 5–10% to the national total, while the IV region accounted for 45–55% of the total rice-planted area in Mainland China.

Changes in water regime of rice paddies. Various water regime patterns have been practiced in rice paddies in China since the 1950s (Fig. 1). During the 1950s–1970s, about 20–25% of the total rice paddy was continuous water logging, and rice paddies with the midseason drainage (F-D-F) accounted for 75–80% of the total area. Since the 1980s, moisture instead of water logging during the late growing stage of rice has been adopted in paddy fields. In the 1980s, 16% of rice paddies were continuous flooding, 77% under the water regime of F-D-F, and 7% under the F-D-F-M, while their respective percentages have changed to 12%, 76%, and 12% in the 1990s (Fig. 1).

Changes in nitrogen input. Seasonal total N input in rice paddies has significantly increased since the 1950s (Table 2). Total N input during the rice-growing season in the 1950s ranged from 67.6 kg N ha^{-1} in the V region to 110.0 kg N ha^{-1} in the I region, with an average of 87.5 kg N ha^{-1} for the five crop-zone regions in Mainland China, which is half of the seasonal total N input in the 1970s. During the 1980s–1990s, the seasonal total N input was, on average, estimated to be 215–225 kg N ha^{-1}. Variation in total N among crop-zone regions was higher in the 1950s–1970s than in the 1980s–1990s. The highest N input generally occurred in the I (North and Northeast) and II (North China Plain and central/western) regions during the 1950s–1970s, while it was the highest in the II and III (Southwest) regions during the 1980s–1990s (Table 2).

Nitrogen management regime has also greatly changed in rice paddies over the period 1950s–1990s (Table 2). Chemical fertilizer application rate has

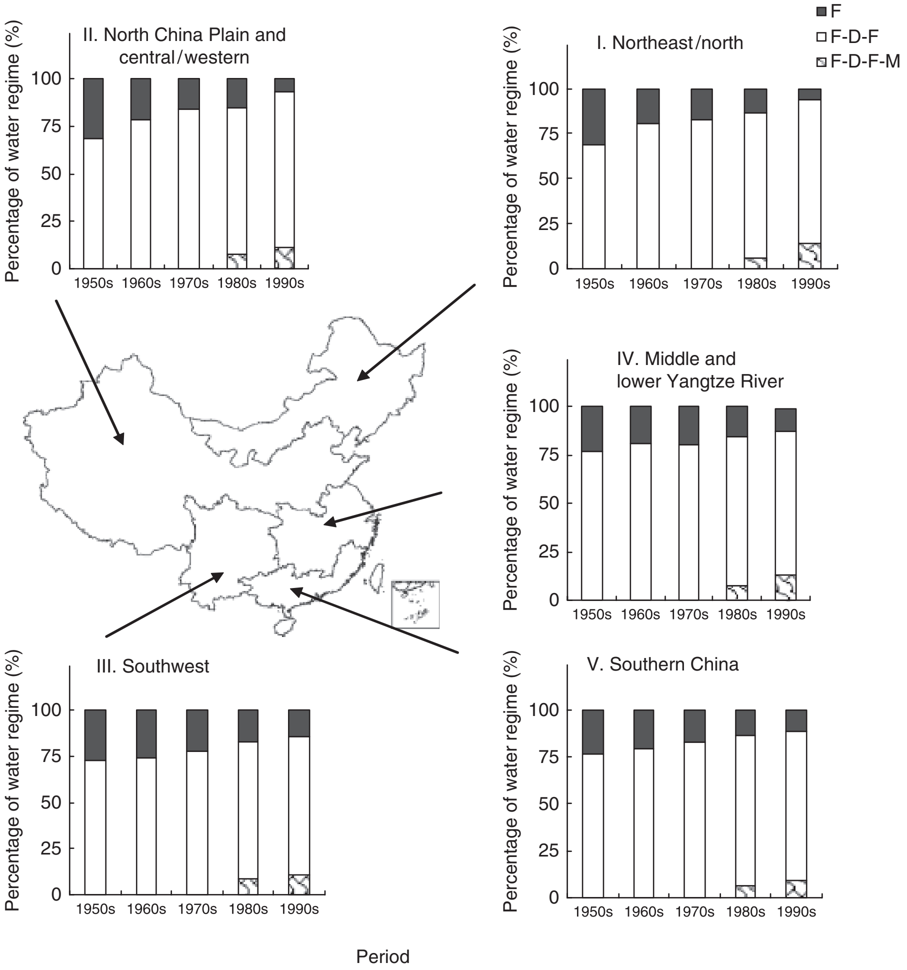

Fig. 1 Shifts in water regime of rice paddies for the five crop-zone regions in Mainland China from the 1950s to 1990s. The rice paddies assigned to the five crop-zone regions: I, North and Northeast, covering Heilongjiang and Inner Mongolia, Liaoning, and Jilin provinces; II, North China Plain and central and west, covering Henan, Hebei, Tianjin, Beijing, Ningxia, Shaanxi, Shanxi, Shandong, Xinjiang, and Gansu provinces; III, Southwest, covering Guizhou, Sichuan, Chongqing, and Yunnan provinces; IV, Middle and lower Yangtze River, covering Anhui, Hubei, Hunan, Jiangsu, Jiangxi, Shanghai, and Zhejiang provinces; V, Southern China, covering Fujian, Guangdong, Guangxi, and Hainan provinces.

increased from 37.4 kg N ha^{-1} in the 1950s to 198.8 kg N ha^{-1} in the 1990s, accounting for 43% and 88% of the total N input during the rice-growing season, respectively. Seasonal chemical N input rate was highest in the II and III crop-zone regions in the 1990s, and in the I crop-zone region in the 1950s–1960s (Table 2). The contribution of manure nitrogen to total N inputs decreased from 52% in the 1950s to 9% in the 1990s (Table 2). During the 1950s–1970s, manure nitrogen input rate was generally stable, ranging from 45.2 to 48.2 kg N ha^{-1}, but thereafter it decreased over time. Seasonal organic manure nitrogen input rate in the 1980s was almost as twice as that in the 1990s.

The harvest biomass of crop residue was estimated to be 2.2–4.5 t ha^{-1} during the 1950s–1990s. Over the period 1950s–1980s, percentage of crop residue retained in paddy fields did not change much, with an average of 33%, while it decreased to 18% of the total aboveground crop biomass in the 1990s. Crop residue N retained during the rice-growing season increased from 4.9 kg N ha^{-1} in the 1950s to 6.3 kg N ha^{-1} in the 1980s due to increased crop biomass from the 1950s to 1980s. In contrast, decreased percentage of crop biomass retained in the rice paddies resulted in lower crop residue N input in the 1990s, although the crop biomass was higher than before. It was, on average, estimated to be 4.9 kg N ha^{-1} in the 1990s, which is almost equivalent to that in the 1950s for the five crop-zone regions in Mainland China (Table 2).

Table 2 Organic and chemical nitrogen inputs and direct N_2O emissions during paddy rice-growing season from 1950s to 1990s

Period	Region*	Area ($\times 10^6$ ha)	Crop residue N	Manure N	Chemical N	Total N	N_2O flux ($kg\,N\,ha^{-1}$)	Percent of the N applied (%)
			(Mean ± SE, $kg\,N\,ha^{-1}$)					
1950s	I	0.6	1.1 ± 0.1	34.9 ± 9.8	74.0 ± 10.3	110.0 ± 17.4	0.28	0.25
	II	0.9	3.7 ± 0.5	68.2 ± 15.7	34.7 ± 6.8	106.6 ± 12.5	0.27	0.26
	III	5.4	3.7 ± 0.8	41.8 ± 6.7	28.7 ± 7.2	74.2 ± 12.0	0.34	0.46
	IV	14.2	5.3 ± 1.2	54.6 ± 4.4	37.2 ± 9.5	97.1 ± 11.2	0.38	0.39
	V	8.3	6.4 ± 0.6	25.6 ± 6.1	35.6 ± 11.7	67.6 ± 12.2	0.24	0.36
	Total	29.4	4.9 ± 0.5	45.2 ± 4.0	37.4 ± 4.9	87.5 ± 6.7	0.32	0.37
1960s	I	0.7	2.3 ± 0.6	43.2 ± 4.4	101.6 ± 21.0	147.0 ± 18.0	0.66	0.45
	II	1.1	4.1 ± 1.7	58.4 ± 19.8	95.9 ± 26.3	158.4 ± 36.2	0.35	0.22
	III	4.8	3.5 ± 0.6	44.4 ± 5.9	42.6 ± 7.2	90.5 ± 11.1	0.32	0.35
	IV	14.8	6.7 ± 1.0	57.4 ± 3.9	64.6 ± 7.8	128.8 ± 9.1	0.43	0.33
	V	7.7	7.3 ± 0.6	27.3 ± 6.5	71.5 ± 10.1	106.0 ± 11.3	0.42	0.39
	Total	29.2	5.6 ± 0.5	46.7 ± 3.9	69.6 ± 6.6	121.9 ± 8.0	0.40	0.33
1970s	I	0.9	2.6 ± 0.7	39.7 ± 8.4	132.8 ± 35.2	175.1 ± 35.7	0.78	0.45
	II	0.9	3.0 ± 0.7	70.3 ± 11.3	155.7 ± 24.4	229.0 ± 29.4	0.79	0.35
	III	5.2	3.5 ± 0.5	46.2 ± 4.3	105.7 ± 6.8	155.4 ± 9.2	0.52	0.34
	IV	19.0	7.1 ± 0.9	56.6 ± 4.8	116.0 ± 11.1	179.7 ± 14.4	0.61	0.34
	V	8.9	8.6 ± 0.6	27.2 ± 6.5	107.9 ± 11.2	143.8 ± 11.2	0.56	0.39
	Total	34.9	6.0 ± 0.5	48.2 ± 3.5	118.5 ± 6.9	172.7 ± 8.4	0.59	0.34
1980s	I	1.2	3.4 ± 1.3	20.8 ± 9.5	157.9 ± 29.6	182.1 ± 37.5	0.81	0.45
	II	0.9	2.8 ± 0.6	49.1 ± 11.8	180.0 ± 17.4	231.8 ± 21.0	1.04	0.45
	III	4.9	4.2 ± 0.8	59.4 ± 14.7	178.4 ± 16.4	242.0 ± 23.1	1.03	0.43
	IV	17.5	7.4 ± 0.6	44.0 ± 4.4	167.7 ± 6.0	219.1 ± 7.0	0.83	0.38
	V	8.3	8.6 ± 1.1	27.8 ± 4.4	160.5 ± 11.1	196.9 ± 13.5	0.86	0.44
	Total	32.8	6.3 ± 0.5	40.6 ± 3.6	168.2 ± 5.3	215.2 ± 7.0	0.88	0.41
1990s	I	1.8	3.9 ± 1.5	14.8 ± 4.3	194.3 ± 33.1	212.9 ± 32.8	0.92	0.47
	II	1.1	2.2 ± 0.8	18.2 ± 7.5	227.0 ± 20.4	247.4 ± 24.9	1.08	0.4
	III	5.2	3.3 ± 0.9	21.1 ± 6.8	206.1 ± 21.1	230.5 ± 20.0	1.03	0.43
	IV	16.4	5.3 ± 0.7	26.3 ± 2.3	197.8 ± 10.6	229.4 ± 10.9	1.05	0.44
	V	7.0	7.4 ± 1.0	16.1 ± 3.5	176.3 ± 12.0	199.7 ± 14.5	0.92	0.5
	Total	31.5	4.8 ± 0.5	21.0 ± 2.0	198.8 ± 7.2	224.6 ± 7.7	1.00	0.46

*The rice paddies assigned to the five crop-zone regions: I, North and Northeast, covering Heilongjiang and Inner Mongolia, Liaoning, and Jilin provinces; II, North China Plain and central and west, covering Henan, Hebei, Tianjin, Beijing, Ningxia, Shaanxi, Shanxi, Shandong, Xingjiang, and Gansu provinces; III, Southwest, covering Guizhou, Sichuan, Chongqing, and Yunnan provinces; IV, Middle and lower Yangtze River, covering Anhui, Hubei, Hunan, Jiangsu, Jiangxi, Shanghai, and Zhejiang provinces; V, Southern China, covering Fujian, Guangdong, Guangxi, and Hainan provinces.

Quantifying direct N_2O emissions during paddy rice-growing season

The emission factors and background emissions estimated under different water regimes (models: F, F-D-F, and F-D-F-M) are assumed to be applicable for N_2O emissions from rice paddies in Mainland China during the 1950s–1990s. Thus, N_2O emission during the rice-growing season was estimated to be 9.6 Gg N_2O-N for each year in the 1950s, which was less than one-third of seasonal N_2O emission (32.3 Gg N_2O-N) in the 1990s (Table 2). The increasing rate of N_2O emission was, on average, estimated to be 6.7 Gg N_2O-N per decade over the period 1950s–1990s ($y = 6.7x$, $r^2 = 0.96$; Table 2). Correspondingly, seasonal N_2O flux was estimated to be 0.32 kg N_2O-N ha^{-1} and 1.00 kg N_2O-N ha^{-1}, which was equivalent to 0.37% and 0.46% of the seasonal total N input in the 1950s and 1990s, respectively (Table 2). Substantial N_2O emission occurred in the IV region of Middle and lower Yangtze River, contributing 51–56% to the national total N_2O emission during the rice-growing season (Fig. 2).

Uncertainties in seasonal N_2O estimate

According to the IPCC methodology [Eqns (4)–(5)], the uncertainties in the emission factor were estimated to be 28.6%, 28.0%, and 29.5% for the F, F-D-F, and F-D-F-M

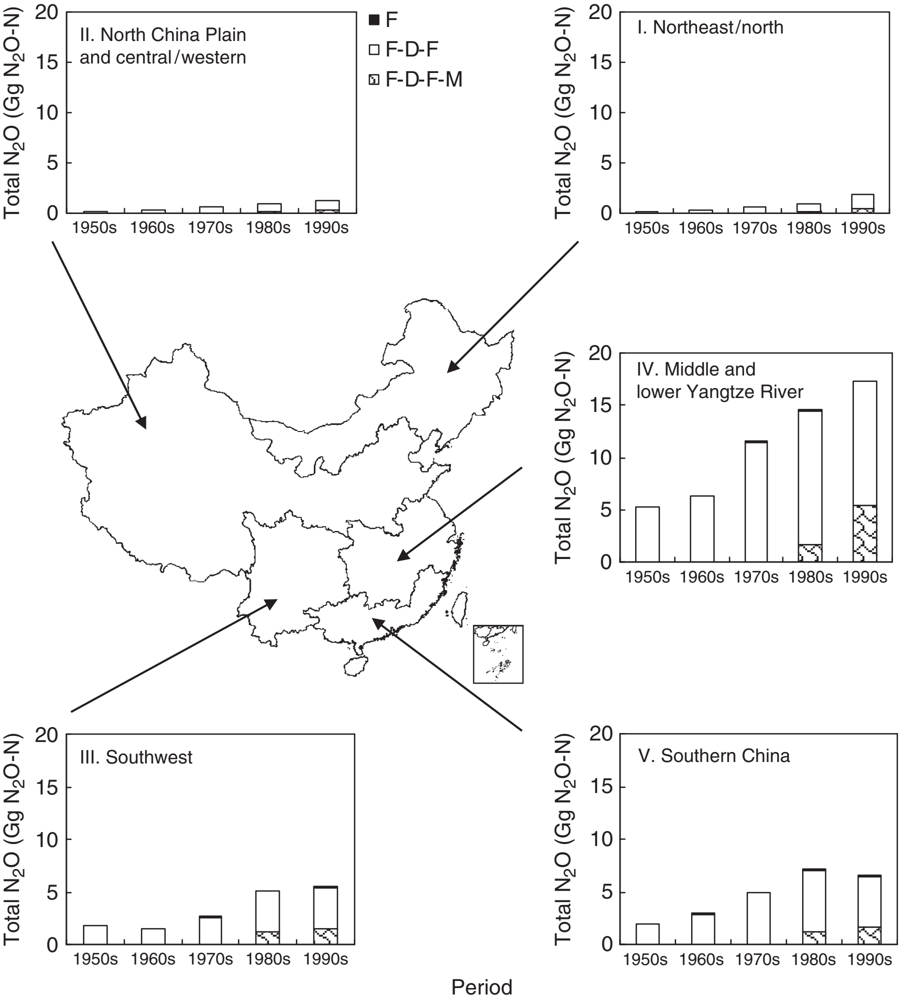

Fig. 2 Decadal and spatial distributions of seasonal N_2O total from paddy fields in Mainland China in the 1950s–1990s. Provincial boundaries of the five crop-zone regions are the same as in Fig. 1.

models, respectively. The uncertainty in seasonal total nitrogen input decreased from 52.5% in the 1950s to 34.4% in the 1970s, while it was estimated to be 22.2–24.3% in the 1980s–1990s. Thus, their combined total uncertainties in seasonal N_2O estimate were estimated to be 59.8% in the 1950s and 44.7% in the 1970s. Relatively, the total uncertainties were lower in the 1980s–1990s, ranging from 36.2% to 37.5% (Fig. 3).

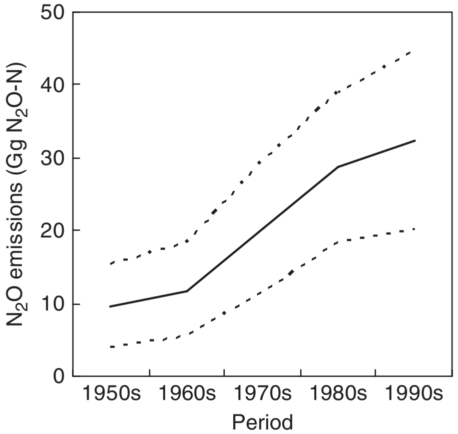

Fig. 3 Uncertainty range of N_2O estimate during the rice-growing season in paddy fields over the period 1950s–1990s. Solid line represents the mean estimate of seasonal N_2O total. Block lines refer to the upper and lower ranges of N_2O estimate.

Discussion

Input data quality

The percentage of various water regime derived from surveyed dataset of this study is fairly consistent with previous estimates (Table 3). During the 1950s–1970s, about 20–25% of the total rice paddy was continuously water logged, and rice paddies with the midseason

Table 3 Consistency of rice production data in this study with those reported in the literature

Time	Water regime (%) F	F-D-F	F-D-F-M	Reference	Proportion of organic N to total N input (%) Proportion	Reference	Chemical N ($kg\,N\,ha^{-1}$)	Total N ($kg\,N\,ha^{-1}$)	Reference
1950s	24.2	75.8		This study	57.3	This study	37.4	87.5	This study
1960s	20.6	79.4		This study	43.3	This study	69.6	121.9	This study
1970s	19.4	80.6		This study	31.4	This study	118.5	172.7	This study
1980s	15.6	77.3	7.1	This study	21.8	This study	168.2	215.2	This study
1990s	11.8	76.9	12.0	This study	11.5	This study	198.8	224.6	This study
1950s	20.0–25.0	80.0–85.0		Mao (1981); Li (1982)	63.0	Huang (1985)	36.1 ($n = 8$)	85.3 ($n = 7$)	Appendix S1
1960s	18.8–22.8	77.2–81.2		Zheng (1990)	45.0	Huang (1985)	57.7 ($n = 9$)	125.9 ($n = 7$)	Appendix S2
1970s	17.5	82.5		Wu & Zou (1981)	18.9–32.4	Xiong *et al.* (1980); Wen (1983)	117.6 ($n = 16$)	179.8 ($n = 22$)	Appendix S3
1980s	15.0	75.0	10.0	Gao & Li (1992)	16.6–21.0	Wang & Huang (1989)	163.0 ($n = 34$)	215.7 ($n = 16$)	Appendix S4
1990s	10.0	90.0		Xing (1998)	12.0–15.4	Xu *et al.* (1998)	193.6 ($n = 41$)	229.8 ($n = 23$)	Appendix S5
	20.0	80.0		Li *et al.* (2002)	12.0	Zheng *et al.* (2004)			
	25.0	75.0		Yan *et al.* (2003)					

F, flooding; F-D-F, flooding–midseason drainage–reflooding; F-D-F-M, flooding–midseason drainage–reflooding–moist intermittent irrigation but without water logging.

drainage accounted for 75–80% of the total area, as documented by Mao (1981), Wu & Zou (1981), Li (1982), and Zheng (1990). In the 1980s–1990s, the water regimes of F, F-D-F, and F-D-F-M were practiced in 12–16%, 77%, and 7–12% of the total rice paddy, respectively, close to the estimates in other studies (Table 3). Xing (1998) estimated that continuous flooding rice paddies account for 10% of the total in 1995. Li *et al.* (2002) reported that rice paddies with midseason drainage contribute $\sim 80\%$ to the total in China in 2000. Yan *et al.* (2003) estimated that two-thirds of rice paddy is under intermittent irrigation or middle season drainage in China. Above all, the dataset of this study could have reflected water regime history information of rice paddies in Mainland China.

Provincial rice yield derived from this surveyed (Y_S) county-scale dataset is comparable with that officially reported (Y_I) by the IRRI ($Y_S = 1.05 Y_I$, $r^2 = 0.635$, $P < 0.0001$, data not shown). The proportion of organic N to seasonal total N input decreased from 57.3% in the 1950s to 11.5% in the 1990s in this study, which is generally consistent with other estimates, ranging from 63.0% in the 1950s to 12.0% in the 1990s (Table 3). Based on summary of data, chemical and total N inputs averaged 36.1 and 85.3 $kg\,N\,ha^{-1}$ in the 1950s, and they have increased to 193.6 and 229.8 $kg\,N\,ha^{-1}$ in the 1990s, respectively (Appendix S1–S5), also comparable with those estimated in this study (Table 3). Overall, N inputs derived from the dataset of this study are close to those reported in the literature (Table 3). The spatial variation of nitrogen inputs is primarily due to the differences in agricultural practice, population growth, and economic development among different regions in the past decades (Zheng *et al.*, 2004).

Factors contributing to uncertainties in N_2O estimate

Uncertainty estimates are an essential element of a complete emissions inventory. Indeed, uncertainty information provides some insights into prioritizing efforts to improve the accuracy of inventories in the future and guiding decisions on methodological choice. High uncertainty in nitrogen input data contributed largely to the total uncertainty in seasonal N_2O estimates. Seasonal nitrogen input among the five crop-zone regions varied greatly in the 1950s, while its variation in the 1990s was relatively low (Table 2), which resulted in higher uncertainties in N_2O estimate in the 1950s relative to the 1990s (Fig. 3).

Some uncertainties in N_2O estimate in the present study were also derived from the emission factor model. The variation in N_2O emissions due to differences in soil properties, temperature, and other agricultural practices could have been included in the uncertainty of the emission factor, because the emission factors are based on measurement among fields with a large spectrum of these driving variables. However, we do not know which driving variable contributes how much to the uncertainty of the emission factor. In addition, the models did not differentiate fertilizer types, and the total nitrogen input was used to estimate N_2O emissions, despite that fertilizer type has been recognized as another factor influencing N_2O emissions in agricultural fields (e.g. Bouwman *et al*., 2002). Although seasonal N_2O emissions generally increased with fertilizer input, they varied with the type of fertilizer as well in rice paddy fields. Compared with urea, application of ammonium sulfate or ammonium bicarbonate induced higher N_2O emission under an identical water regime of F-D-F or F-D-F-M (e.g. Cai *et al*., 1997; Zheng *et al*., 2000). In contrast to pure chemical fertilizer application, on the other hand, organic manure and crop residue amendments increased seasonal N_2O emissions in some studies (e.g. Zheng *et al*., 2000; Zou *et al*., 2004, 2005b), while they decreased N_2O emissions in other studies (e.g. Xiong *et al*., 2003).

Difference in frequency of N_2O measurements may also contribute to their estimate uncertainty (Akiyama *et al*., 2005). Ideally, N_2O emissions should be measured frequently enough to capture their peak fluxes. Besides that N_2O fluxes can be measured continuously on an hourly basis by combining tunable diode laser technology with micrometeorological techniques (Edwards *et al*., 2003; Pattey *et al*., 2006), sharp peaks of N_2O fluxes in paddy fields were observed in a study using an automated monitor system (Zheng *et al*., 2000). Relative to measurements once a week, measurements twice weekly showed more peak fluxes of N_2O, particularly after nitrogen fertilizer was applied in rice paddies (Zou *et al*., 2005a). However, most studies in the dataset measured flux only once a week. As a consequence, some N_2O flux peaks might have been missed and seasonal N_2O emissions could have been underestimated in these studies.

IPCC EF statistical model vs. process-oriented DNDC model

Recently, the DNDC model has been frequently used to estimate N_2O emissions from rice paddies in China (Li *et al*., 2001, 2004, 2005, 2006). Besides the complex input requirements, however, validation of the DNDC model against N_2O flux measurements has produced inconsistent results in Chinese rice paddies. In Li *et al*.'s studies (2001, 2004, 2005, 2006), the DNDC model fitted a case field measurement of N_2O flux well under the water regime of F-D-F-M in Wujiang County. In contrast, the DNDC model has been validated to be not suitable for simulating N_2O emissions from rice paddies under the water regime of F-D-F in Fengqiu and Nanjing, China (Cai *et al*., 2003). Indeed, N_2O emissions from rice paddies in China projected by the DNDC model are extremely higher than those estimated by other methodologies. For example, the DNDC model estimated N_2O emissions to be 290–410 Gg N_2O-N yr^{-1} in the continuous flooding rice paddies and 420–610 Gg N_2O-N yr^{-1} in rice paddies with midseason drainage in China (Li *et al*., 2004, 2005), while direct N_2O emissions from rice paddies in China were estimated to be 88 Gg N_2O-N yr^{-1} by Xing (1998), or 91 Gg N_2O-N yr^{-1} by Zheng *et al*. (2004). Nevertheless, the DNDC model has been changing rapidly over the last several months, and some current estimates of N_2O emissions from rice paddies using a revised version 92 of the model no longer show such large emissions (personal communication, unpublished results).

In contrast to the DNDC model, the IPCC EF methodology primarily concentrates on fertilizer-induced direct N_2O emissions. Although the EF models adopted in present study seem to be too simplistic, the results of this study are comparable with existing country estimates. Using the 'top-down' methodology, Zheng *et al*. (2004) estimated EFs to be 0.60% in rice paddies, and direct N_2O emissions to be 49.5 Gg N_2O-N during the rice-growing season in the 1990s. Based on field N_2O flux measurements extrapolation, Xing (1998) reported that direct N_2O emissions from paddy fields totaled 88 Gg N_2O-N in 1995, consisting of 35 Gg N_2O-N emitted during the rice-growing season and 53 Gg N_2O-N during the upland crop seasons. However, the author did not distinguish N_2O emissions among different water regimes and thus failed to address the spatial and temporal variations of N_2O emissions from rice paddies in China.

Although direct N_2O emissions are generally influenced by soil parameters, agricultural practice, and environmental conditions on field site scales, no significant relationships between N_2O emissions and these factors were found for Chinese rice paddies on national and regional scales. Stehfest & Bouwman (2006) summarized and statistically analyzed 1008 N_2O emission measurements for agricultural fields, and concluded that agricultural N_2O emissions were significantly influenced by N application rate, crop type, fertilizer type, soil organic C content, soil pH, and texture. To our knowledge, however, few studies have presented statistical models incorporating the dependence of N_2O emissions on soil parameters and agricultural practices for rice paddies on country scale, although consideration of these factors could minimize the uncertainty in N_2O estimates (Stehfest & Bouwman, 2006; Kimura *et al*., 2007).

Decadal and spatial variations of seasonal N_2O fluxes

In the present study, the models addressing the dependence of fertilizer-induced N_2O emissions on water regime provided some insights into decadal and spatial variations of direct N_2O emission. On average, the flux of N_2O during the rice-growing season has tripled over the period 1950s–1990s, which largely occurred before 1990. Particularly in the V crop zone region (Southeast), seasonal N_2O flux at the highest increasing rate has fourfolded in the 1990s relative to the 1950s (Table 2). In addition, various water regime types incurred an obvious spatial variation in seasonal N_2O fluxes before the 1990s. In the 1950s, for example, seasonal N_2O flux was estimated to be 0.24 kg N_2O-N ha^{-1} in the V region, while it was 0.34 kg N_2O-N ha^{-1} in the IV region. Seasonal total N_2O emission was equivalent to 0.25% of the applied N in the I region and 0.46% in the III region. In contrast, no significant spatial variation in seasonal N_2O flux was found among different crop-zone regions in the 1990s.

Changes in seasonal N_2O flux in rice paddies were accompanied by rice yields over the period 1950s–1990s (Fig. 4). The relationship between rice yields and seasonal N_2O fluxes for the five crop-zone regions suggests that both increased N_2O emissions and rice yields were closely associated with shifts in water regime and increased nitrogen input. Obviously, rice yields were largely dependent on nitrogen application levels in rice paddies. On the other hand, adoption of midseason drainage and moisture instead of water logging as a saving-water irrigation incurring substantial N_2O emission has improved rice production as well.

Contribution of rice production to agricultural total of N_2O emissions in China

The present study estimated direct N_2O emissions to be 32.3 Gg N_2O-N, with an uncertainty of 37.5%, during the rice-growing season in paddy fields for each year in the 1990s, which is equivalent to 0.46% of the seasonal total N input. Direct N_2O emission from croplands in China was estimated to be 340 Gg N_2O-N yr^{-1} in 1995 by Li *et al.* (2001), 398 Gg N_2O-N in 1995 by Xing (1998), or 275 Gg N_2O-N yr^{-1} in the 1990s by Zheng *et al.* (2004). These estimates suggest that rice production occurring on 23% of the cultivated land accounts for 8–11% of the total N_2O emission from croplands in China. Because of rice planting area increase in the past decades and lower emission factor, therefore, paddy rice relative to upland crop production could have greatly contributed to mitigating N_2O emissions from agriculture in China.

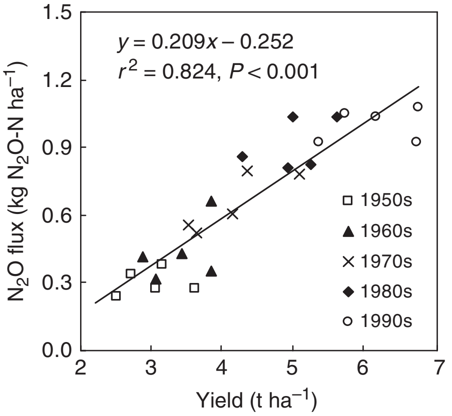

Fig. 4 Decadal changes in seasonal N_2O flux consistent with paddy rice yield over the period 1950s–1990s.

Note that this study only estimated direct N_2O emissions during the rice-growing season, and did not count those during the following nonrice seasons (e.g. rice–winter wheat, rice–rapeseed rotations) in paddy fields. Although water regime has distinguished N_2O emissions in rice paddies from upland crops and natural wetlands, some agricultural practices such as water management and organic incorporation during the rice-growing season may have a substantial effect on the following seasonal N_2O emissions (Zou *et al.*, 2005b). Results of our previous study in a paddy rice–winter wheat rotation system indicated that compared with the water regime of F-D-F, continuous flooding in the rice season significantly increased N_2O emissions from the winter wheat-growing season. As well, wheat residue incorporation before rice transplanting had a far-lasting effect on N_2O emissions during the winter wheat-growing season (Zou *et al.*, 2003). Therefore, annual total of N_2O emissions in rice paddies would be underestimated by extrapolating N_2O data during the rice-growing season.

Effects of water regime on direct N_2O emissions

Primarily, N_2O is produced in soils via the biogeochemical processes of nitrification and denitrification that are greatly influenced by the soil water status (Dobbie & Smith, 2003). In contrast to paddies with the water regime of F-D-F, or the seasonal continuous flooding paddies, the dry–wet alteration after midterm drainage created a favorable soil environment to both nitrification and denitrification processes, which contributed greatly to higher N_2O emissions under the water regime of F-D-F-M. Under continuous flooding, in contrast, a large proportion of N_2O produced from denitrification would be further reduced to N_2 before leaving the soil (Firestone & Davidson, 1989). On the other hand, water regime might influence the availability of nitrogen, labile C compounds, and O_2 in paddy soils that are key factors to N_2O production in general denitrification models (Firestone & Davidson, 1989). The midseason drainage and dry–wet alteration may create soil moisture suitable for translating fertilizer N into N_2O, and they are also able to improve root activities and accelerate soil organic C decomposition, which might produce more available C and N for soil microbes, and thereby favor N_2O emissions.

Because of water resources scarcity and cultivation technique development, the water regime of F-D-F-M as a water-saving irrigation technology has been increasingly practiced in China's rice production. For example, water is especially scarce in the North China Plain that contains 26% of the China's cultivated land, 30% of its irrigated land, and 24% of its total grain production (Geng *et al.*, 2001). The water regime of F-D-F-M and aerobic rice paddies instead of anaerobic paddies have been suggested as potential options for rice production in this area. However, shifting water management from continuous flooding to midseason drainage could increase N_2O emissions from Chinese rice paddies by 0.13–0.20 Tg N_2O-N yr^{-1} (Li *et al.*, 2004) or 0.15 Tg N_2O-N yr^{-1} (Li *et al.*, 2005). The EFs in this study predicted that N_2O emissions during the rice-growing season would be increased by 44 Gg N_2O-N if the water regime is shifted to the F-D-F-M for rice paddies in China. In addition, N_2O emissions have been shown to be significantly higher from aerobic rice paddies compared with anaerobic paddies (Xu *et al.*, 2004). A recent study suggests that water-saving rice production systems relative to conventional paddy rice systems would typically lead to a net increase in greenhouse gas emissions (Kreye *et al.*, 2007). Therefore, these options would increase N_2O emissions from rice production in China. Indeed, how to reconcile increasing N_2O emissions and scarcity of water resources with the development of rice production has become a real challenge in Mainland China.

Conclusions

The statistic models on N_2O measurements in rice paddies under different water regimes projected that due to increased paddy rice-planted area, increased nitrogen input and changes in water regime, seasonal N_2O emissions have increased from 9.6 Gg N_2O-N in the 1950s to 32.3 Gg N_2O-N in the 1990s, which is accompanied by the increase in rice yield over the period 1950s–1990s. The uncertainties in N_2O estimate were estimated to be 59.8% in the 1950s and 37.5% in the 1990s. N_2O emissions during the rice-growing season in the 1990s accounted for 8–11% of the reported estimates of annual total emission from croplands in Mainland China. Relative to upland crop production, paddy rice development in the past decades could have greatly contributed to mitigating agricultural N_2O emissions in China. However, seasonal N_2O emissions would be increased, given that saving-water irrigation and nitrogen inputs are increasingly practiced in the rice paddies in China.

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Supporting Information

Additional Supporting Information may be found in the online version of this article:

Appendix S1. 1950s.
Appendix S2. 1960s.
Appendix S3. 1970s.
Appendix S4. 1980s.
Appendix S5. 1990s.