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New estimates of direct N_2O emissions from Chinese croplands from 1980 to 2007 using localized emission factors

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Abstract. Nitrous oxide (N_2O) is a long-lived greenhouse gas with a large radiation intensity and it is emitted mainly from agricultural land. Accurate estimates of total direct N_2O emissions from croplands on a country scale are important for global budgets of anthropogenic sources of N_2O emissions and for the development of effective mitigation strategies. The objectives of this study were to re-estimate direct N_2O emissions using localized emission factors and a database of measurements from Chinese croplands. We obtained N_2O emission factors for paddy fields (0.41 ± 0.04 %) and uplands (1.05 ± 0.02 %) from a normalization process through cube root transformation of the original data. After comparing the results of normalization from the original values, Logarithmic and cube root transformations were used because the frequency of the original data was not normally distributed. Direct N_2O emissions from Chinese croplands from 1980 to 2007 were estimated using IPCC (2006) guidelines combined with separate localized emission factors for paddy fields and upland areas. Direct N_2O emissions from paddy fields showed little change, increasing by 10.5 % with an annual rate of increase of 0.4 % from 32.3 Gg N_2O-N in 1980 to 35.7 Gg N_2O-N in 2007. In contrast, emissions from uplands changed dramatically, increasing by 308 % with an annual rate of 11 % from 68.0 Gg N_2O-N in 1980 to 278 Gg N_2O-N in 2007. Total direct N_2O emissions from Chinese croplands increased by 213 % with an annual rate of 7.6 % from 100 Gg N_2O-N in 1980 to 313 Gg N_2O-N in 2007, and were determined mainly by upland emissions (accounting for 67.8–88.6 % of total emissions from 1980 to 2007). Synthetic N fertilizers played a major role in N_2O emissions from agricultural land, and the magnitude of the contributions to total direct N_2O emissions made by different amendments was synthetic N fertilizer > manure > straw, representing about 78, 15, and 6 % of total direct N_2O emissions, respectively, between 2000 and 2007. The spatial pattern of total N_2O emissions in 2007 in China shows that high direct N_2O emissions occurred mainly in the north and in the Sichuan Basin in the southwest. The provinces with the highest emissions were Henan (35.4 Gg) and Shandong (31.6 Gg) and Tibet had the lowest (0.65 Gg). High direct N_2O emissions per unit of arable land occurred mainly on the North China Plain and the southeast coast. The mean value nationally was 2.52 kg N ha^{-1}, with 18 provinces above this value and with emissions of >4.0 kg N ha^{-1} in Beijing, Tianjin and in Jiangsu, Shandong, Fujian and Henan provinces.

1 Introduction

Nitrous oxide (N_2O) undoubtedly plays a key role in contributing to global warming and climate change (IPCC, 2007a). It had already reached a concentration of 319 ppb (10^{-9} mol mol^{-1}) in 2005 with a rate of increase of approximately 0.26 % per year (IPCC, 2007b). Agriculture plays a major role in the global emissions of N_2O (Robertson et al., 2000), explaining about 60 % of total anthropogenic N_2O emissions (IPCC, 2007c). Synthetic nitrogen (N) fertilizers are the largest source of N_2O emissions from croplands (EIA, 1997), with a linear or exponential effect of N fertilizer rate on N_2O emissions (Bouwman et al., 2002; Snyder et al., 2009). The total global consumption of N fertilizers reached 100 million tonnes in 2007 (Heffer and Prud, 2008) and will continue to increase to meet the food requirements of the increasing world population (Jiang et al., 2010). In some intensive agricultural areas excessive N fertilizer application

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is common because farmers always include an "insurance dressing" to avoid yield or economic losses. Lower N use efficiency will produce severe environmental problems including large emissions of N_2O (Dobermann, 2007; Ju et al., 2009). If N fertilizers and animal manures are not carefully managed in the future, N_2O emissions from agricultural soils will increase by 35–60 % up to 2030 according to the FAO (2003).

China became the largest producer and consumer of N fertilizers in the world during the 1990s (Heffer and Prud, 2008). It is important to estimate the direct N_2O emissions from N fertilizers and to devise measures of mitigation. Direct N_2O emissions have been defined as N_2O production from soils to which N has been applied during the current year or season and is the result of multiplying the direct N_2O emission factor (EFd) with the rate of N application (IPCC, 2006). Direct emissions contribute 75 % of the total N_2O emissions caused by agriculture in China according to Zheng et al. (2004). Several methods have been devised to estimate direct N_2O emissions, namely the emission fluxes aggregation extrapolation method (Mosier et al., 1991; Xing, 1998), the IPCC (1996) guideline method (Mosier et al., 1998; Xing and Yan, 1999; Yan et al., 2003; Zheng et al., 2004; Zou et al., 2009), the empirical formula method (Zou et al., 2010) and the mechanistic model computation method (Li et al., 2003; Zheng et al., 2004). The emission flux aggregation extrapolation method is a simple extrapolation using field measurements of emission flux in an area combined with the arable area of the cropping system and it is too approximate for a large country with diverse edaphic and climatic conditions and cropping systems. Empirical formula methods estimate through empirical formulae created by combining N_2O emissions with controlling factors and is also unsatisfactory for calculating emissions on a national scale. Mechanistic model computation calculates through a process model and links with a database of related parameters. Complex model structure and input parameters make it difficult to use this approach on a large scale. In addition, this type of model still awaits further validation using local observation data. IPCC methods mainly involve calculation by aggregating the inputs of N on cropland and combining these with emission factors. This is a universally accepted method for calculating the national inventory of N_2O emissions from different fertilizer N sources and the calculation parameters are easy to collect. It also easy to make comparisons among countries but the accuracy of estimation depends very much on the emission factor of each N input source. The estimation by localized EFs with the IPCC Tier 2 method is better than by default values of EFs with the IPCC Tier 1 method.

Most of the estimates of direct N_2O from Chinese croplands were calculated using the old version of the IPCC (1996) guidelines with the default value, and may not be suitable, especially since they do not differentiate between paddy fields and uplands. However, the mechanisms and quantities of N_2O emitted from these two systems were found to differ in recent studies (Ju et al., 2009, 2011). The objectives of the current study were therefore to summarize all the observations of direct N_2O emissions from N inputs in Chinese croplands and compute the emission factors of paddy fields and upland areas, to re-estimate direct N_2O emissions from both paddy fields and uplands using these localized emission factors together with IPCC (2006) guidelines from 1980–2007, and to compare the estimates with others reported in the literature.

2 Materials and methods

2.1 Basic data sources

The N_2O emission data from Chinese croplands were collected from previous studies published in the literature, books or research reports. The database mainly includes crop systems, distribution region, N application rates, correlation factors of soil and climatic conditions and emission factors range in different region and were shown in Table 1 which had been summarized from data collected in previous studies (e.g. Su et al., 1992; Cai et al., 1997, 2003; Chen et al., 1997, 2002; Song et al., 1997; Zheng et al., 2000; Dong et al., 2001; Xiong et al., 2002; Gao, 2004). The frequency of N_2O measurements were twice weekly, weekly or with longer time intervals.

N_2O emission factors were summarized with Meta Analysis (%) using the database described above. The direct emission factor (EFd) was calculated as total N_2O emission from nitrogen fertilizer plots minus the emission from unfertilized control plots expressed as a percentage of N applied (IPCC, 1996).

The numbers of livestock and poultry breeding in each province and over the whole country were obtained from the Ministry of Agriculture of the People's Republic of China (1980–2007b) and N fertilizer rate, crop yield, and population data from the Ministry of Agriculture of the People's Republic of China (1980–2007a). N application rates on paddy fields were sourced from "Information of the National Agricultural Costs and Returns" (Energy Bureau of National Development and Reform Commission, 1980–2007) and the doctoral dissertation of Wang (2007). Other parameters (Tables 2–4) such as quantities of human and animal excreta and N contents (fresh samples) were the mean value of different publications from 1994 to 2004 which were summarized by Ma et al. (2006) and Wang et al. (2006). Percentage of excreta returned to the field was the average of different publications during 1980–2007 (Zhu, 1997; Li et al., 1998; Liu et al., 2006; Ma et al., 2006). Ratio of oil crop seed conversion to cake fertilizer, N content in cake fertilizer and green manures were sourced from National Extension Center of Agriculture Techniques (1999a, b). Ratio of straw to grain, N content in straw and percentage return of main crop straw to the field were sourced from different literature

Table 1. Crop systems, distribution region, N application rates, correlation factors of soil and climatic conditions and emission factors range in different regions.

Type	Crop systems	Ratio to total measurements (%)	Distribution region	N application rate (kg N ha^{-1} $season^{-1}$)	Average temperature (°C)	Precipitation (mm)	Organic matter content (%)	Total N (%)	pH	Emission factors range (%)
Upland	Wheat, maize or wheat and maize rotation	42.4	Northeast China (NE) North China (N)							
	Cotton, soybean and peanut	10.5	East China (E) Northwest China (NW) and Southwest China (SW)	62–559	<10 (NE) 14.2 (N)	662 (NE) 534 (N)	2.4 (NE) 1.5 (N)	0.11 (NE) 0.09 (N)	6.5 (NE) 8.2 (N)	0.11–4.80 (NE) 0.13–1.61 (N)
Paddy fields	wheat (or green manure) and rice rotation or early rice and late rice rotation	4.3	East China (E) South China (S)	95–479	16.4 (E) 22.1 (S) 15.5 (NW) 14.2 (SW)	829 (E) 2107 (S) 428 (NW) 1200 (SW)	1.9 (E) 2.0 (S) 0.6 (NW) 1.2 (SW)	0.18 (E) 0.07 (S) 0.14 (NW) 0.07 (SW)	6.9 (E) 5.7 (S) 8.0 (NW) 7.5 (SW)	0.0036–4.71 (E) 0.11–2.16 (S) 0.04–1.25 (NW) 3.23–3.55 (SW)
	Rice (6.7 % seasonal continuous flooding and 93.3 % applying midseason drainage)	42.8	and Northeast China (NE)							

Table 2. Human or animal excreta parameters, their N contents (fresh samples) and percentage of return to the field in China*.

Category	Amount of excreta	N content (g kg^{-1})	Percentage return to field (%)
Pig	5.3 kg d^{-1}	2.38	65
Labor cow	10.1 t a^{-1}	3.51	30
Beef cow	7.7 t a^{-1}	3.51	30
Milk cow	19.4 t a^{-1}	3.51	30
Horse	5.9 t a^{-1}	3.78	44
Donkey and mule	5.0 t a^{-1}	3.78	44
Sheep	0.87 t a^{-1}	10.14	33
Meat chicken	0.10 kg d^{-1}	10.32	45
Layer chicken	53.3 kg a^{-1}	10.32	45
Duck and goose	39.0 kg a^{-1}	6.25	45
Rabbit	41.4 kg a^{-1}	8.74	45
Human	107 kg a^{-1}	6.43	33

* Data from Zhu (1997); Li et al. (1998); Wang et al. (2006); Ma et al. (2006); Liu et al. (2006).

Table 3. Ratio of oil crop seed conversion to cake fertilizer and N content in cake fertilizer and green manure*.

Organic material	Ratio of oil crop seed conversion to cake fertilizer (%)	N content (g kg^{-1})
Rape cake	0.55	53.5
Cotton seed cake	0.8	42.9
Soybean cake	0.85	66.8
Peanut cake	0.5	69.2
Sesame cake	0.5	50.8
Sunflower seed cake	0.7	47.6
Flax cake	0.7	56.0
Green manure	/	4.0

* Data from the National Extension Center of Agriculture Techniques (1999a, b).

or publications (Lu et al., 1996a, b; Li et al., 1998; National Extension Center of Agriculture Technique, 1999a, b; Liu et al., 2006).

2.2 Normalization of the emission factors

The frequency of emission factors of both paddy fields and upland areas (Fig. 2a and b) are not normally distributed and it is not valid simply to calculate the arithmetic or geometric mean values of this type of sample. A normalization process of the emission factor is necessary for obtaining the mean and standard deviation of the emission factor. The normalization process is by conversion of the skewed frequency distribution (positive bias or negative bias) on the original scale to a normal distribution on a probability scale (Group Section) (Xue, 1978; Tang, 1984.; Zhang et al., 1990). Figure 3c and f show that the data are normally distributed after cube root transformation of the original data on the probability scale. However, the method of cube root transformation is not valid simply to calculate the arithmetic or geometric mean values of the sample. The main step in normalization is to calculate the frequency of each group and the relative cumulative frequency after grouping the original data according to the order. The probability scale (Group Section) can be found in the area table under the curve of normalization and the group interval of two neighboring groups can be calculated based on relatively cumulative frequency. The normalized frequency (f) of each group can be calculated from the frequency divided by the group interval. The average group value ($\bar{t}$) and the standard deviation of the group value (δt) can then be calculated using formulas (1) and (2). The last obtained t-values ($\bar{t} \pm \delta t$) were used to deduce the original value by the method of linear interpolation. The range between maximum and

Table 4. Ratio of straw to grain, N content in straw, and percentage straw return to the field of main crops in China*.

Crop	Ratio of straw to grain	N content of straw ($g\,kg^{-1}$)	Percentage straw return to field (%)
Rice	0.9	9.1	30
Wheat	1.1	6.5	45
Maize	1.2	9.2	20
Millet	1.0	8.2	0
Sorghum	2.0	12.5	0
Other cereals	1.0	6.8	45
Bean	1.0	21.0	80
Potato	0.5	25.1	0
Cotton	3.0	12.4	0
Peanut	0.8	18.2	90
Rape seed	2.5	8.7	40
Sugarcane (leaves/stems)	0.3	11.0	90
Beet (leaves/roots)	0.5	2.5	90
Tobacco leaves	1.0	14.4	0

* Data from Lu et al. (1996a, b); Li et al. (1998); Liu et al. (2006); the National Extension Center of Agriculture Technique (1999a, b).

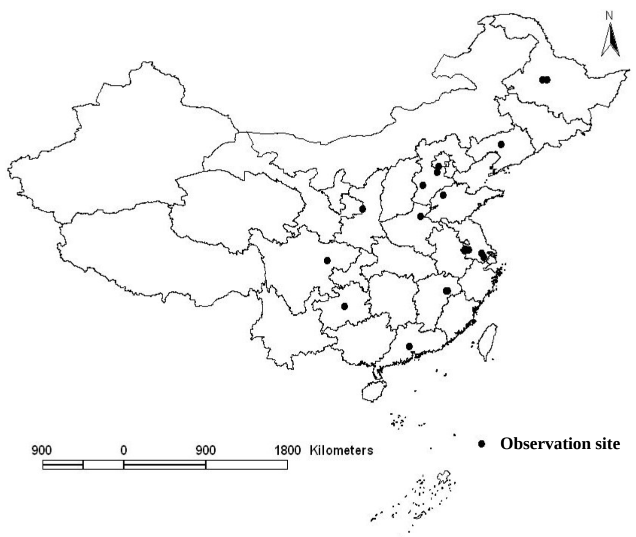

Fig. 1. Locations of the N_2O emission observation sites in Chinese croplands.

minimum, after cube transformation, is smaller than that of the original data and after original data is log transformed (see Supplement Tables S1 and S2). As a result, the data after cube transformation has less variation than the original data and the data after log transformation. So the mean values deduced from t-values ($\bar{t} \pm \delta t$) by normalization after cube root transformation also have lower standard error than data normalized through the original data and normalized after log transformation.

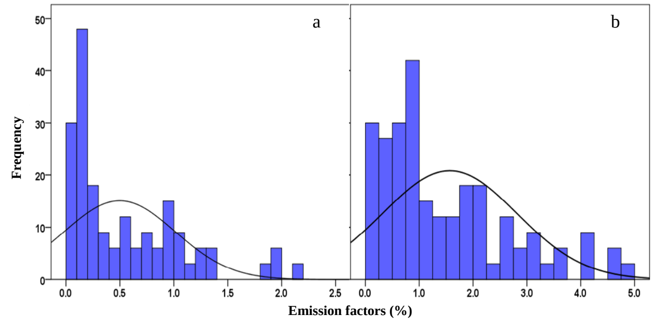

Fig. 2. Frequency distribution of the original data of N_2O emission factors of paddy fields **(a)** ($n = 195$) and uplands **(b)** ($n = 261$). The Gaussian curves superimposed on the two frequency distributions of the original data representing the original data are not normally distributed.

$$\bar{t} = \sum tf/n \tag{1}$$

$$\delta t = \sqrt{\frac{n\sum t^2 f - (\sum tf)^2}{n(n-1)}} \tag{2}$$

where n is the sum of each group frequency after normalization, and t is the group value, i.e. the mean of two neighboring group intervals. Finally, the emission factor can be calculated in reverse based on the t-value obtained through the mean group values $\pm$ one standard deviation (Xue, 1978; Tang, 1984). The steps above can be repeated after converting the original data using other transformation functions if they are still not normally distributed with graphics using group interval as the x-axis and f as the y-axis (Zhang et al., 1990).

2.3 Items included in the calculation

The method of calculating direct N_2O emissions from croplands in this study using IPCC (2006) guidelines with localized emission factors was as follows:

$$N_2O_{Direct}\text{-}N = N_2O\text{-}N_{N\,inputs} + N_2O\text{-}N_{OS} + N_2O\text{-}N_{PRP} \tag{3}$$

where N_2O_{Direct}-N, N_2O-$N_{N\,inputs}$, N_2O-N_{OS}, and N_2O-N_{PRP} represent direct N_2O emissions from fertilized soil on an annual basis, N_2O emitted from synthetic N amended soil, from Histosol areas and from grassland soil resulting from addition of manure and urine, respectively. The units are kilograms of N_2O-N per year.

This study did not consider the N_2O emissions from grassland by adding manure or urine (N_2O-N_{PRP}). In addition, Histosol areas comprise about 5 million ha and occur mainly in the northeast and in remote mountainous areas of southwest China which are not cultivated for arable crop production. The cultivated area of Histosols amounts to only 19 000 ha and the N_2O emissions amount to 0.95 Gg, a value basically unchanged since 1990 (Xing and Yan, 1999). The

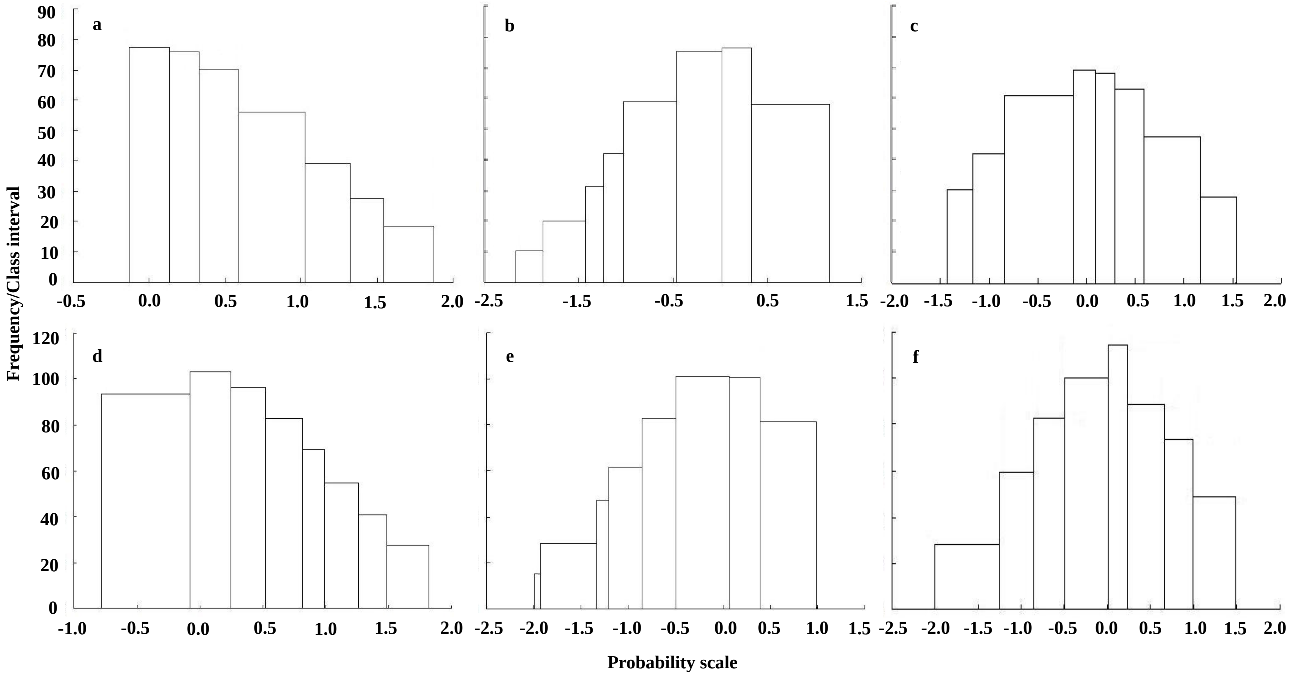

Fig. 3. Frequency distribution of N_2O emission factors of paddy fields ($n = 195$), **(a)**: normalized through original data; **(b)**: normalized after log transformation; **(c)**: normalized after cube transformation and uplands ($n = 261$), **(d)**: normalized through original data; **(e)**: normalized after log transformation; **(f)**: normalized after cube transformation.

N_2O emission data from cultivated Histosols are therefore taken as a default value since 1990.

2.4 Calculating direct N_2O emissions from different N input sources

The formula used to calculate the amount and sources of nitrogen from which N_2O emissions were estimated are included in Eq. (4).

$$N_2O\text{-}N_{\text{N inputs}} = [(F_{SN} + F_{AW} + F_{CR}) \cdot EF_1] + [(F_{SN} + F_{AW} + F_{CR}) \cdot EF_{1FR}] \quad (4)$$

Where the F_{SN}, F_{AW}, F_{CR} represent inputs of N from synthetic fertilizers, organic materials and crop residues. N content in compound fertilizer was calculated at $0.3\,\text{kg}\,\text{kg}^{-1}$. Nitrogen fixation by legumes is not considered in the calculation according to the IPCC (2006) guidelines. EF_1 and EF_{1FR} are the emission factors for paddy fields and uplands, respectively.

$$F_{AW} = \text{Manure/Urine N} + \text{Cake fertilizer N} + \text{Green manure N} \quad (5)$$

$$\text{Manure/Urine N} = \sum_i \text{Human or animal species } (i) \cdot \text{Discharge coefficient } (i) \cdot \text{Nitrogen excreted } (i) \cdot \text{Percentage return to field } (i) \quad (6)$$

$$\text{Cake fertilizer N} = \sum_i \text{Cake crop yield } (i) \cdot \text{Cake production rate } (i) \cdot \text{Nitrogen content in cake fertilizer } (i) \quad (7)$$

$$\text{Green manure N} = \text{Green manure production} \cdot \text{Nitrogen content}(\%) \quad (8)$$

We calculated the rural population only and the ratio of the total population to the adult population is 0.85 (Cao et al., 2006; Zhu, 1997).

$$F_{CR} = \sum_i \text{Crop yield } (i) \cdot \text{Ratio of straw to grain } (i) \cdot \text{Straw nitrogen content } (i) \cdot \text{Proportional return to field } (i) \quad (9)$$

3 Results

3.1 Emission factors of paddy fields and uplands

The mean emission factors were more normally distributed after normalization by cube root transformation of the original data compared to other transformations (see Supplement Tables S1 and S2 for detailed description) (Fig. 3a, b, c, d, e, f). We conducted further statistical testing for the effect of normal distribution after cube root transformation (Table 5) and the Z statistic values of one sample K-S test for paddy fields and uplands were 0.562 and 0.440, respectively. Both

Table 5. Normal test (one-sample K-S test) of the normalized frequency after cube root transformation.

Item		Value	
		Paddy fields	Uplands
Samples		10	10
Normal parameters	Mean	45.727	59.583
	Standard deviation	29.112	39.944
Most extreme differences	Absolute	0.178	0.139
	Positive	0.142	0.132
	Negative	−0.178	−0.139
Kolmogorov-Smirnov Z		0.562	0.440
Asymptotic significance (2-tailed)		0.911	0.990

Table 6. N_2O emission factors (%) of Chinese paddy fields and uplands calculated by different methods.

Land type	No. of samples	Method	Mean	Standard deviation	Range
Paddy fields	195	Original data	0.54	0.53	0.0036–2.13
		Normalized through origin data	2.18	0.25	1.93–2.43
		Normalized after logarithms conversion	0.23	1.07	0.21–0.25
		Normalized after cube root conversion	0.41	0.04	0.37–0.45
Uplands	261	Original data	1.49	1.23	0.035–4.8
		Normalized through origin data	1.90	0.98	0.92–2.88
		Normalized after logarithms conversion	0.79	0.36	0.28–2.19
		Normalized after cube root conversion	1.05	0.02	1.03–1.07

reject the null hypothesis and reach normal distribution because the corresponding concomitant probabilities are 0.911 and 0.990 larger than the significance level of 0.05. This was the most reasonable method of normalization after cube root conversion and gave smaller standard deviation and range than other methods (Table 6). We therefore selected 0.41 % and 1.05 % to replace the original average values of 0.54 % and 1.49 % as the emission factors of paddy fields and uplands, respectively (Table 6).

3.2 Direct N_2O emissions from paddy fields

Direct N_2O emissions from paddy fields have changed little from 1980 to 2007 (Fig. 4). They have increased from 32.3 Gg in 1980 to 35.7 Gg in 2007, with an annual rate of increase of only 0.4 %. Emissions were maintained at a stable level between 1980 and 1995 and changed markedly from 1995 to 2007. The dynamics of total direct N_2O emissions were dominated by N_2O emissions from synthetic N fertilizers. However, N_2O emissions caused by manures and straw showed only slight variation and the emissions of N_2O from Histosols remained horizontal because of the application of the default value of 1990. The order of contributions

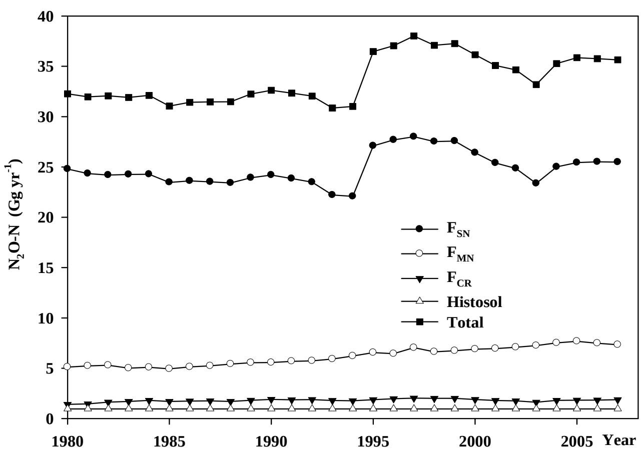

Fig. 4. Total direct N_2O emissions in paddy fields between 1980 and 2007; F_{SN}, F_{MN}, F_{CR} and Histosols represent direct N_2O emissions from synthetic fertilizers, manures, crop residues and Histosols, respectively.

Table 7. Contribution of different sources to direct N_2O emissions from Chinese croplands (%).

	Synthetic N fertilizers			Manures			Crop residues			Histosols		
Year	Paddy fields	Uplands	Total croplands	Paddy fields	Uplands	Total croplands	Paddy fields	Uplands	Total croplands	Paddy fields	Uplands	Total croplands
1980	76.84	52.16	60.11	15.85	38.54	31.24	4.37	9.30	7.71	2.94	/	0.95
1985	75.57	67.43	69.27	15.90	23.67	21.93	5.47	8.89	8.12	3.06	/	0.69
1990	74.18	75.30	75.11	17.06	17.77	17.65	5.85	6.93	6.75	2.91	/	0.49
1995	74.31	77.60	77.12	17.97	15.88	16.19	5.12	6.51	6.31	2.60	/	0.38
2000	73.06	79.05	78.26	19.08	14.84	15.40	5.24	6.12	6.00	2.63	/	0.35
2005	70.90	79.10	78.12	21.38	14.79	15.57	5.07	6.11	5.99	2.65	/	0.32
2007	71.50	80.43	79.42	20.58	13.54	14.34	5.26	6.03	5.94	2.66	/	0.30

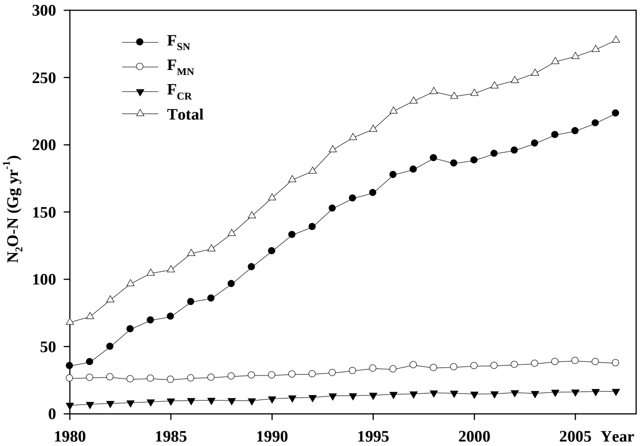

Fig. 5. Total direct N_2O emissions from uplands between 1980 and 2007.

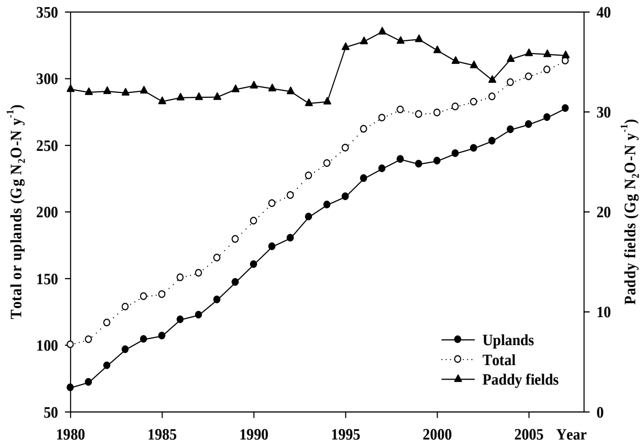

Fig. 6. Historical changes in direct N_2O emissions from Chinese croplands between 1980 and 2007.

to direct N_2O emissions in paddy fields was synthetic N fertilizers > organic matter > straw > Histosols.

The contributions of synthetic N fertilizers, manures, straw and Histosols to direct N_2O emissions showed slight variation after 2000 (Table 7) and were maintained at about 71–73 %, 19–21 %, 5.2 % and 2.6 %, respectively. From the perspective of historical change, the contribution from synthetic N fertilizers declined by 6.9 % from 76.8 % in 1980 to 71.5 % in 2007, from Histosols declined by 9.5 % from 2.9 % in 1980 to 2.7 % in 2007, and from manures and straw increased by 29.8 % and 20.4 %, respectively.

3.3 Direct N_2O emissions from uplands

Direct N_2O emissions from uplands showed rapid growth between 1980 and 2007 (Fig. 5). They increased by 308 % from 68.0 Gg in 1980 to 278 Gg in 2007, rising at an approximate rate of 11 % per year. Their change was also controlled by N_2O emissions from synthetic N fertilizers and the total N_2O emissions caused by manures and straw remained at a modest level. The order of contributions to direct N_2O emissions of uplands was synthetic N fertilizers > manures > straw.

The contributions of synthetic N fertilizers, manures and straw to direct N_2O emissions also showed slight variation after 2000 (Table 7) and were maintained at about 79–80, 13–15 and 6.0 %, respectively. From the perspective of historical change, the contribution of synthetic N fertilizers appeared to follow the opposite trend to paddy fields, increasing by 54.0 % from 52.2 % in 1980 to 80.4 % in 2007, rising at an average of 1.9 % per year, and the contribution from manure declined by 64.9 % from 38.5 % in 1980 to 13.5 % in 2007, and that from straw declined by 35.2 % from 9.30 % in 1980 to 6.03 % in 2007.

3.4 Overall direct N_2O emissions from Chinese croplands

The total direct N_2O emissions from Chinese croplands were estimated by summing the emissions from paddy fields and uplands. They increased rapidly from 100.2 Gg in 1980 to 313.2 Gg in 2007 (Fig. 6), rising at an approximate rate of

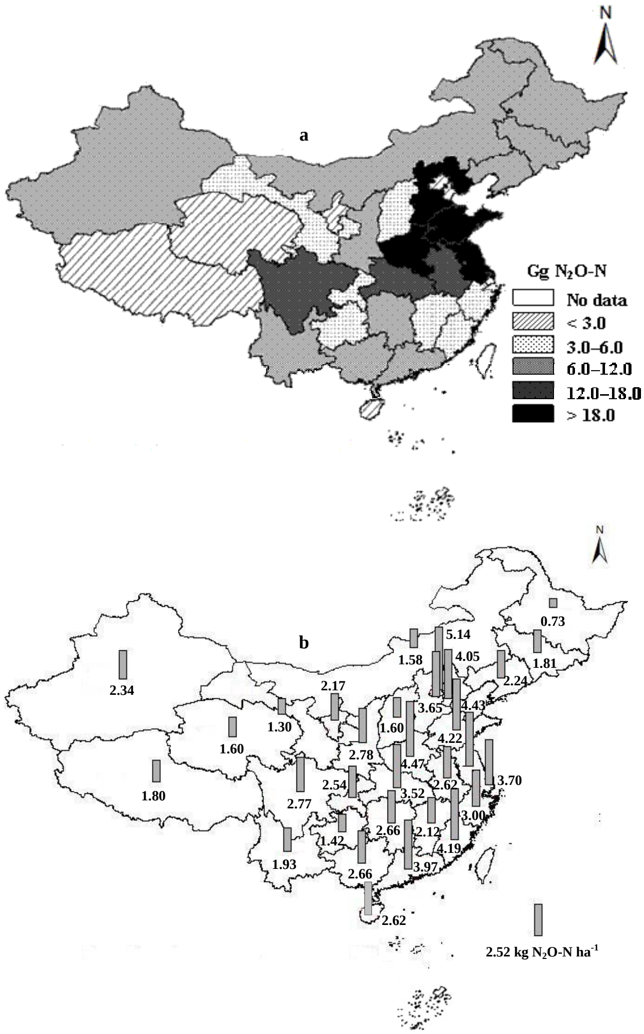

Fig. 7. The provincial pattern of total direct N_2O emissions from **(a)** Chinese croplands and **(b)** direct N_2O emissions per unit area of arable land in 2007; bar with 2.52 kg N_2O-N ha^{-1} represents the scale of average direct N_2O emissions per unit area of arable land of all provinces.

7.6 % per year. Direct N_2O emissions from uplands accounted for about 82.8 % of total direct N_2O emissions from croplands, with a range of 67.8–88.6 % from 1980 to 2007. The N_2O emissions from paddy fields were maintained at a stable level relative to uplands and had a slight effect on total direct N_2O emissions of croplands. Thus the historical changes in total direct N_2O emissions from croplands showed a similar trend to those from uplands. The order of contributions to total direct N_2O emissions of cropland was synthetic N fertilizers > manures > straw > Histosols.

The contributions of synthetic N fertilizers, manures, straw and Histosols to total direct N_2O emissions have changed year by year (Table 7) and have remained at about 78, 15, 6 and 0.32 % from 2000 to 2007, respectively. It is clear that the application of synthetic N fertilizers exerts an important influence on total direct N_2O emissions from cropland. The contribution of synthetic N fertilizers was about twice the world average of 36 %, and manures were much lower than the world average level of 30 % (Mosier et al., 1998).

3.5 Total direct N_2O emissions from croplands at provincial level in 2007

Total direct N_2O emissions from croplands on a provincial scale in 2007 were calculated by multiplying the rate of N applied from F_{SN}, F_{MN} and F_{CR} in each province with the corresponding emission factors of paddy fields or uplands and showed large spatial variability among provinces (Fig. 7a). This reflects the large variation in synthetic N inputs in different Chinese provinces. The larger emission provinces were mainly in north China and the Sichuan Basin, with the highest emission provinces, Henan (35.4 Gg) and Shandong (31.6 Gg), accounting for 11.6 % and 10.3 % of total direct N_2O emissions from cropland. Tibet had the lowest emissions (0.65 Gg), accounting for only 0.21 % of total direct N_2O emissions from cropland.

Direct N_2O emissions per unit of arable land had different distribution patterns from total emissions at provincial scale (Fig. 7b). Values were higher in the east than the west and the highest emission provinces were on the North China Plain and in the southeast coastal areas (Fig. 7b). The mean direct N_2O emissions per unit of arable land were 2.52 kg N ha^{-1} nationally and 18 provinces had higher values, including >4.0 kg N ha^{-1} in Beijing, Tianjin and in Jiangsu, Shandong, Fujian and Henan provinces. Beijing had the highest values of to 5.14 kg N ha^{-1}.

4 Discussion

4.1 Emission factors of paddy fields and uplands

The emission factors (0.41 % for paddy fields and 1.05 % for uplands) were obtained by the statistical approach in this study in order to estimate the total N_2O emissions on a national scale. It might be possible to make some comparison with IPCC methodology but there is no way of comparing site-specific results. Both emission factors are slightly higher than the default values of the IPCC guidelines (2006) (0.3 % and 1.0 % for paddy fields and uplands) and within their ranges of 0–0.6 % and 0.3–3.0 %, respectively. These increases may reflect the general and widespread overuse of N fertilizers in most Chinese croplands.

Recent studies have indicated that direct N_2O emission factors are very low in semiarid and low carbon soils. For example, they were only 0.08 % and 0.13 % in winter wheat and 0.45 % and 0.46 % in summer maize under conventional and optimum N fertilization when an automated N_2O flux measurement system was used in the North China Plain (Ju et al., 2011). The mean N_2O emission factor was only equivalent to 0.26 % of applied N fertilizer in the semiarid Northern Great Plains in the USA (Dusenbury et al., 2008). The value was as low as 0.02 % and 60 times lower than the IPCC default value in the semiarid climate of west Australia (Barton et al., 2008). All the above low direct N_2O emission factors may be attributed to drought and low carbon soil conditions, and not correspondingly decided by the N fertilizer rate, but rather to specific observations and cannot be compared with statistical emission factors on a national scale. N_2O emissions show little difference below an application rate of 100 kg N ha^{-1} and rise rapidly with higher rates (Bouwman et al., 2002) and there are larger N_2O emission factors when applications are in excess of uptake capacity (Grant et al., 2006). The higher emission factors in this study compared with IPCC guidelines (2006) may reflect the overuse of N fertilizer that is characteristic of most Chinese croplands on a national scale.

China is a very large country and the soil, climate and management practices (cropping systems, application rates of N fertilizer and manures etc.) have high spatial and temporal variation. It is preferable to use different emission factors for different regions to derive more reliable results. However, at present we do not have enough observation sites and databases to do this. For example, our N_2O emission data have originated mainly from the east of the country, with fewer observation sites in the vast central and western regions (Fig. 1). As the observations increase, we would expect the uncertainties associated with direct N_2O estimation to decrease. Another important aspect of our paper is the review of all the estimates of N_2O emissions using different approaches in Chinese croplands in comparison with our results to illustrate the differences and uncertainties among these estimates. We also give our readers a general overview of N_2O emissions from Chinese croplands.

The rice production area represents about 20 % of the world total and 23 % of the arable land area in China (Frolking et al., 2002). It is therefore necessary to separate paddy fields from uplands to estimate the direct N_2O emissions from Chinese croplands. The emission factor of paddy fields is 0.3 % by the IPCC guidelines (2006) which is lower than in this study (0.41 %) because the IPCC method does not consider the different water regimes and major contribution from aerobic paddy soils. Water-saving technology has increased rapidly in Chinese rice production because of scarce water resources and the development of cultivation techniques (Zou et al., 2009). Since the 1980s about 80 % of the paddy fields have included midseason drainage and the rate of N_2O emissions from this type of paddy field is very different from annually flooded paddy fields because the emissions have been stimulated by wetting and drying cycles in midseason drainage (Zou et al., 2009, 2010). Earlier estimates therefore incorporated larger errors because they did not consider this change in the management of paddy fields (Yan et al., 2003). The emission factors from continuously flooded paddy fields (0.02 ± 0.006 %), flooding-midseason drainage-reflooding (0.42 ± 0.12 %) and flooding-midseason drainage-reflooding-moist intermittent irrigation (0.73 ± 0.22 %) were calculated by Zou et al. (2007) by classifying the N_2O database into three categories taking into consideration differences in water regime and simulation of the N_2O emission factor using an ordinary least square (OLS) linear regression model. The flooding-midseason drainage-reflooding management accounted for about 80–90 % of the total rice growing area (Xing and Yan, 1999; Zou et al., 2009). We therefore used 0.41 % as the emission factor for paddy fields to minimize the uncertainties associated with estimation.

4.2 Direct N_2O emissions from Chinese croplands

The literature describes many methods from simple empirical formulae to complicated biogeochemical models for estimating N_2O emissions from Chinese croplands (Xing, 1998; Xing and Yan, 1999; Xing and Zhu, 2000; Yan et al., 2003; Li et al., 2003; Zheng et al., 2004; Zou et al., 2009, 2010). Comparing these estimates with the present study (Table 8), our direct N_2O-N emission results from paddy fields were higher than those of Zou et al. (2009) who only considered synthetic N fertilizer-induced direct N_2O-N emissions (16.5 and 20.0 Gg in 1980 and 2000, respectively) due to consideration of more N sources. The direct N_2O-N emissions from paddy fields in our study were 32.3 Gg and 36.2 Gg in 1980 and 2000, respectively, with a mean value of 34.5 Gg-N in the 1990s that was very close to the 32.3 Gg N_2O-N per year estimated by Zou et al. (2009). Yan et al. (2003) estimated 40.7 Gg N in 1995 and took into consideration the N fixed by legume crops in their study. Xing (1998) and Zheng et al. (2004) calculated very similar values of 88 Gg N in 1995 and 90.8 Gg N in the 1990s using different methods, and both of these were significantly higher than the present study. Xing (1998) calculated direct N_2O emissions from paddy fields by multiplying the average N_2O fluxes with their corresponding areas in different regions, and included background N_2O emissions and N_2O emissions induced by different N sources. Zheng et al. (2004) took into account the N fixed by legumes that was excluded in the new IPCC (2006) guidelines and deposition of atmospheric N derived from anthropogenic activities that belongs to the category of indirect emissions.

Direct N_2O-N emissions of uplands in the present study are similar to the results of Zou et al. (2010) in 1980 and 2000. Although considering different sources of direct N_2O emissions, the result of 184.2 Gg N in the 1990s from Zheng et al. (2004) is very similar to the average of 193.5 in 1990, 1994 and 1995 in this study because almost all of the direct N_2O emissions from synthetic N fertilizer, straw and manure, accounted for 99.1–99.7 % of total direct N_2O emissions in our study. The result of Xing (1998) in 1995 was very close to Yan et al. (2003) at about 310 Gg N and somewhat higher than the 211 Gg N in the present study.

Table 8. Comparison of national total N_2O emission estimates from Chinese croplands from different studies (Gg N_2O-N yr^{-1}).

Year	Method	N_2O-N sources considered	Emission factors	Paddy fields	Uplands	Total emissions	Reference
1980	Localized EFds	FSN+FAW+FCR+FOS[1]	Paddy fields 0.41 %, upland 1.05 %	32.3	68.0	100.2	This study
1985	Localized EFds	FSN+FAW+FCR+FOS	Paddy fields 0.41 %, upland 1.05 %	31.1	106.9	137.9	This study
1990	Localized EFds	FSN+FAW+FCR+FOS	Paddy fields 0.41 %, upland 1.05 %	32.6	164.0	193.1	This study
1994	Localized EFds	FSN+FAW+FCR+FOS	Paddy fields 0.41 %, upland 1.05 %	31.0	205.2	236.2	This study
1995	Localized EFds	FSN+FAW+FCR+FOS	Paddy fields 0.41 %, upland 1.05 %	36.5	211.4	247.8	This study
2000	Localized EFds	FSN+FAW+FCR+FOS	Paddy fields 0.41 %, upland 1.05 %	36.2	238.1	274.3	This study
2005	Localized EFds	FSN+FAW+FCR+FOS	Paddy fields 0.41 %, upland 1.05 %	35.9	265.5	301.4	This study
2007	Localized EFds	FSN+FAW+FCR+FOS	Paddy fields 0.41 %, upland 1.05 %	35.7	277.6	313.2	This study
1990	DNDC model	FSN+FAW+FCR+FDN+BNE	No	286.8	978.2	1265	Li et al. (2003)
1990	DNDC model	FSN+FAW+FCR+FDN+BNE	0.8 % or 3.3 kg N_2O-N ha^{-1} yr^{-1}			310	Li et al. (2001)
1990	Strictly empirical IPCC methodology[2]	FSN+FAW+FBN+ FOS+BNE	1.25 % and 5–10 kg N_2O-N ha^{-1} for histosols			360	Li et al. (2001)
1995	The average emission fluxes × their corresponding areas	All sources from cropland	Average N_2O fluxes in different area	88.0	310	398	Xing (1998)
1980	Revised IPCC (1996) guidelines	FSN+FAW+FBN+FCR+FOS	1.25 %			159	Xing and Yan (1999)
1985	Revised IPCC (1996) guidelines	FSN+FAW+FBN+FCR+FOS	1.25 %			253	Xing and Yan (1999)
1990	Revised IPCC (1996) guidelines	FSN+FAW+FBN+FCR+FOS	1.25 %			266	Xing and Yan (1999)
1995	Revised IPCC (1996) guidelines	FSN+FAW+FBN+FCR+FOS	1.25 %			336	Xing and Yan (1999)
1995	IPCC (1997) guidelines	FSN+FAW+FBN+FCR	upland 1.25 %, paddy fields 0.25 %	40.7	308.5	349.2	Yan et al. (2003)
1990s	The re-quantified site-scale EFds × Nr[3] input in individual cropland	FSN+FAW+FBN+FCR+FDN	The re-quantified site-scale EFds	90.8	184.2	275	Zheng et al. (2004)
1990s	The average of the re-quantified site-scale EFds × national Nr input	FSN+FAW+FBN+FCR+FDN	The mean of the re-quantified site-scale EFds			390	Zheng et al. (2004)
1990s	Empirical equations[4]	FSN+FAW+FBN+FCR+FDN	No			340	Zheng et al. (2004)
1990s	The IPCC (1997) default EFd × national Nr input	FSN+FAW+FBN+FCR+FDN	1.25 %			360	Zheng et al. (2004)
1994	Revised IPCC (1996) guidelines and IAP-N model[5]	FSN+FAW+FBN+FCR +FSSCON	Site-scale EFds			301.4	China National Coordination Committee on Climate Change (2004)
1980	Empirical models[6]	FSN	Upland 1.86 *P* %; paddy fields F 0.02 %, F-D-F 0.42 %, F-D-F-M 0.73 %	16.5	62.0	78.5	Zou et al. (2010)
1985	Empirical models	FSN	Upland 1.86 *P* %; paddy fields F 0.02 %, F-D-F 0.42 %, F-D-F-M 0.73 %			124.2	Zou et al. (2010)
1980s	Empirical models	FSN	Upland 1.86 *P* %; paddy fields F 0.02 %, F-D-F 0.42 %, F-D-F-M 0.73 %			115.7	Zou et al. (2010)
1990	Empirical models	FSN	Upland 1.86 *P* %; paddy fields F 0.02 %, F-D-F 0.42 %, F-D-F-M 0.73 %			177.2	Zou et al. (2010)

Table 8. Continued.

Year	Method	N_2O-N sources considered	Emission factors	Paddy fields	Uplands	Total emissions	Reference
1990s	OLS model[7]	FSN+FAW+FCR	paddy fields F 0.02 %, F-D-F 0.43 %	32.3			Zou et al. (2009)
1990s	Empirical models	FSN	Upland 1.86 *P* %; paddy fields F 0.02 %, F-D-F 0.42 %, F-D-F-M 0.73 %			210.5	Zou et al. (2010)
1995	Empirical models	FSN	Upland 1.86 *P* %; paddy fields F 0.02 %, F-D-F 0.42 %, F-D-F-M 0.73 %			217.7	Zou et al. (2010)
2000	Empirical models	FSN	Upland 1.86 *P* %; paddy fields F 0.02 %, F-D-F 0.42 %, F-D-F-M 0.73 %	20.0	230	250	Zou et al. (2010)

[1] FSN is the N from synthetic N fertilizer consumption. FAW is the animal waste or organic matter N used as fertilizer. FCR is the N in crop residue returned to soil. FOS is the N from organic soil. FDN is deposited atmospheric N derived from anthropogenic activities. BNE is N_2O emissions from the soils which no nitrogen fertilizer application. FBN is the N fixed by N-fixing crop. FSSCON is the N from sewage sludge and compounds of organic N. FTN is the N impacted by warmer temperatures raising +2° between maximum and minimum in each day's, the result of total N_2O emission would increased by 10 % if a DNDC set of simulations with a warming of +2° (Li et al., 2001).
[2] Applied IPCC (1997) methodology for calculating direct N_2O emissions from croplands + background N_2O emissions (croplands area × 1.0 kg N_2O-N ha^{-1} yr^{-1})
[3] Nr is the data of reactive nitrogen input from IAP-N model.
[4] Relationship between direct N_2O emissions and gross domestic product (GDP) on the basis of population or arable land area in 1990s in China (Zheng et al., 2004).
[5] IAP-N model for calculating anthropogenic Nr input into individual croplands, detailed information was introduced in Zheng et al. (2002).
[6] The synthetic fertilizer-induced N_2O emissions (FIE-N_2O) from fertilized upland soils in China = 0.0186(±0.0027) · P · N, P and N represent for precipitation (m) and nitrogen application rate (kg N ha^{-1} yr^{-1}). The FIE-N_2O emissions from continuous flooding (F) paddy fields = 0.0002(±0.00006) N, flooding-midseason drainage-reflooding (F-D-F) = 0.0042(±0.0012) N, and flooding-midseason-drainage-reflooding-moist intermittent irrigation but without water logging (F-D-F-M) = 0.0073(±0.0022)N, respectively (Zou et al., 2010).
[7] OLS model is an ordinary least square linear regression model (Zou et al., 2009).

Total N_2O emissions from Chinese croplands were about 340 Gg N_2O-N in 1995 as calculated by Xing and Yan (1999) and Yan et al. (2003), both of these intermediate between the 310 Gg N_2O-N of Li et al. (2001) and 398 Gg N_2O-N of Xing and Yan (1999). All of these are higher than the 247.8 Gg N_2O-N found in the present study, and this might be explained by the inclusion by Xing (1998) and Li et al. (2001) of background N_2O emissions. Furthermore, Xing and Yan (1999) and Yan et al. (2003) took into consideration the N fixed by legume crops. If we exclude the direct N_2O emission results (12.5–23.1 Gg N) from biological fixation during 1980–2000 as calculated by Xing and Yan (1999), this is also much higher than in our study due to the failure of Xing and Yan (1999) to distinguish between direct N_2O emissions from paddy fields and uplands by simply using the IPCC (1996) default value of 1.25 % as emissions factor, thus giving a substantial overestimate of direct N_2O emissions from paddy fields. Zou et al. (2010) used different emission factors in different croplands but they only estimated direct N_2O emissions induced by synthetic N fertilizers, giving an underestimate of direct N_2O emissions from Chinese croplands, but their results are close to those in the present study and can be explained by most of the direct N_2O-N emissions being dominated by synthetic N fertilizers. N_2O-N emissions reported by the Chinese National Coordination Committee on Climate Change (2004) were 301.6 Gg in 1994, about 28 % higher than our estimates due to the indirect N_2O emissions caused by atmospheric N deposition to croplands being incorporated and the N fixed by legumes also being included. The DNDC model gave an estimate of 1265 Gg N_2O-N in 1990 by calculating the sum of N_2O emissions from croplands in each province. This is much higher than any estimates from experimental work, and the province with the largest N_2O-N emissions was Heilongjiang (374 Gg N). The DNDC model is sensitive to soil organic matter and this result in overestimated N_2O emissions from farmland in northeast China (Li et al., 2003). When all the evidence is considered, the IPCC (2006) guidelines combined with localized emission factors as employed in the present study may give a more useful basis for estimates than complicated biogeochemical models for estimating direct N_2O emissions on a national scale.

4.3 The contribution of synthetic N fertilizers to direct N_2O emissions from paddy fields and uplands

The contribution of synthetic N fertilizers to direct N_2O emissions from paddy fields declined from 76.8 % in 1980 to 71.5 % in 2007. This is because the rate of synthetic N fertilizer application from statistical data in the "Compilation Materials of The National Agricultural Production Costs and Returns" changed slightly, increasing by only 3 % in 2007 relative to 1980. However, the N inputs from manures and crop straw increased by 43 and 33 % during the corresponding period. The increase in total direct N_2O emissions resulted mainly from N inputs from manures and straw. So the role of synthetic N fertilizers in direct N_2O emissions from paddy fields declined. In contrast, the rate of synthetic N fertilizer application in uplands increased by 530 % in 2007 relative to 1980. However, the increases in N inputs from manures and straw were 43 and 165 %. The increase in total direct N_2O emissions was dominated by synthetic N fertilizers. Thus the contribution of synthetic N fertilizers to direct N_2O emissions from uplands increased significantly from 1980 to 2007.

4.4 Further research

China is a very large country and the systems employed in upland cultivation systems are very diverse. The application rate of N fertilizers and manures has high spatial and temporal variation (Ti et al., 2011). In addition, Chinese vegetable production systems have expanded rapidly in recent years and the area has reached 2.5 million hectares (Li, 2005). Although the fraction of the total land area devoted to this production is small, the systems receive very high rates of N fertilizers together with poultry and other manures and frequent irrigation. N fertilization rates exceed 2800 kg N ha^{-1} yr^{-1} in Huimin, Shandong province (Ju et al., 2006), and the irrigation rates reach 1000 mm in each vegetable season in Shouguang, Shandong province (Li et al., 2001). Excessive N fertilization and uncontrolled water management may lead to high N_2O emissions from vegetable cropping systems. Unfortunately we found only limited published data for these systems. So our calculation does not include vegetable production systems because of insufficient available data and the likely increase in uncertainties of estimation in this study.

N_2O emission factors are not uniform across different regions because the factors controlling N_2O emissions are greatly affected by local soil conditions, climate, cropping systems and management practices (Bouwman, 1996; Ju et al., 2011). More credible results could be obtained by disaggregating of the data into the different production systems, followed by calculation of emission factors for each type of system, and then aggregating the calculated emissions to provincial or national level. We did not, however, use the different emission factors for the different regions because there are not enough experimental data from the vast central and western regions for the study period and the observation sites were mostly distributed in the eastern part of the country (Fig. 1). Therefore we only estimate total N_2O emissions from Chinese croplands at a national level and compare them with other similar studies. We may be able to use different emission factors for different regions in future estimates as measurements increase.

5 Conclusions

Between 1980 and 2007, direct N_2O emissions from Chinese croplands increased at the rate of 7.6 % per year. These emissions totaled an estimated 313 Gg N in 2007. The historical changes in direct N_2O emissions from Chinese croplands were determined by emissions from upland areas because these accounted for about 82.8 % of total direct N_2O emissions overall. The contributions of different N sources to direct N_2O emissions were synthetic N fertilizers > manures > straw and these were stable at 79.4, 14.3, and 5.9 % in 2007. The provinces with larger total emissions were mainly in north China and in the Sichuan Basin in 2007, and the highest and lowest direct N_2O emissions provinces were Henan (35.4 Gg N) and Tibet (0.65 Gg N), respectively. The highest direct N_2O emissions per unit area of arable land were mainly on the North China Plain and the southeast coast with a mean value of 2.52 kg N ha^{-1}, and 18 provinces gave higher values, with Beijing reaching the highest value of all at 5.14 kg N ha^{-1}.

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