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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 %) and uplands (1.05 %) 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 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 11 % with an annual rate of increase of 0.4 % from 29.8 Gg $N_2O$-N in 1980 to 33.1 Gg $N_2O$-N in 2007. In contrast, emissions from uplands changed dramatically, increasing by 296 % with an annual rate of 10.9 % from 64.4 Gg $N_2O$-N in 1980 to 255.3 Gg $N_2O$-N in 2007. Total direct $N_2O$ emissions from Chinese croplands increased by 206 % with an annual rate of 7.6 % from 94.2 Gg $N_2O$-N in 1980 to 288.4 Gg $N_2O$-N in 2007, and were determined mainly by upland emissions (accounting for 68.4–88.5 % of total emissions from 1980 to 2007). Synthetic nitrogen 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 77, 16, and 6.5 % 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 north China and in the Sichuan Basin in the southwest. The provinces with the highest emissions were Henan (32.6 Gg) and Shandong (29.1 Gg) and Tibet had

the lowest (0.6 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 2.36 kg N $ha^{-1}$, with 17 provinces above this, and with emissions of >4.0 kg N $ha^{-1}$ in Beijing and in Jiangsu 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 tons in 2007 (Heffer and Homme, 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 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 (FAO, 2003).

China became the largest producer and consumer of N fertilizers in the world during the 1990s (Heffer and Homme, 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 et al., 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 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 are mainly calculated 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 be 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 includes emission rates and correlation factors of soil and climatic conditions, management practices and measurement conditions (Bouwman et al., 2002). The locations of the observation sites are shown in Fig. 1. For each measurement, data were saved on the latitude and longitude, the time of the experiment, soil type, soil texture, soil organic carbon and total nitrogen content, pH, crop type, average temperature and precipitation, fertilizer type, nitrogen fertilizer rate, duration of experiment, frequency of measurements, and rate of $N_2O$-N emissions. Microsoft Excel was used for data processing and for recording data from external sources. For example, climate data were obtained from the Chinese Weather Bureau (http://cdc.cma.gov.cn/). The database contained a total of 456 measurements, comprising 195 samples from paddy fields and 261 samples from upland sites.

$N_2O$ emission factors were summarized with Meta Analysis (%) using the database described above. The direct emission factor (EF) 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 Livestock Yearbook of China (Ministry of Agriculture of the People's Republic of China, 1980–2007a) and N fertilizer rate, crop yield, and





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population data from China Agriculture Statistical Report (Ministry of Agriculture of the People's Republic of China, 1980–2007b). 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 1–3) such as quantities of human and animal excreta and N contents (fresh samples), percentage of excreta returned to the field, ratio of oil crop seed conversion to cake fertilizer, N content in cake fertilizer and green manures, ratio of straw to grain, N content in straw and percentage return of main crop straw to the field were sourced from the literature or publications listed in Tables 1–3.

### 2.2 Normalization of the emission factor

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). The main step in normalization is to calculate the frequency of each group and the relatively 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 ($\delta t$) can then be calculated by the following formula:

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

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

where $n$ is the sum of each group frequency after normalization, $t$ is the group value, i.e. the mean of two neighboring group intervals. Finally, the emission factor can be calculated reversely based on the $t$ value obtained through the mean group values ± 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 x-axis and $f$ as 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:

$$N_2O_{Direct}{-}N = N_2O{-}N_{Ninputs} + N_2O{-}N_{OS} + N_2O{-}N_{PRP} \quad (3)$$

where $N_2O_{Direct} - N$, $N_2O - N_{Ninputs}$, $N_2O - N_{OS}$, 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 organic soil 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 ($N_2O$-$N_{PRP}$). The cultivated area of organic soils amounts to only 19 000 ha and the $N_2O$ emissions from this amount to 0.95 Gg, a value basically unchanged since 1990 (Xing and Yan, 1999). The $N_2O$ emission data from organic soil are therefore taken as a default value since 1990.



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### 2.4 Calculating direct $N_2O$ emissions from different N input sources

Nitrogen fixation by legumes is not considered in the calculation according to the IPCC (2006) guidelines. The formula used was therefore as follows:

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

Where the $F_{SN}$, $F_{AW}$, $F_{CR}$ represent inputs of N from synthetic fertilizers, organic materials and crop residues. $EF_1$ and $EF_{1FR}$ are the emission factors for paddy fields and uplands, respectively.

$$F_{SN} = N_{FERT} \cdot (1 - FRACGASF) \quad (5)$$

N content in compound fertilizer was calculated at 0.3 kg kg$^{-1}$. FRACGASF is the ratio of $NH_3$ and $NO_X$ emissions after synthetic N fertilizer application to the soil using the default value 0.1 kg kg$^{-1}$ and $N_{FERT}$ is the rate of synthetic fertilizer application.

$$F_{AW} = \text{Manure/Urine N} + \text{Cake fertilizer N} + \text{Green manure N} \quad (6)$$

$$\text{Manure/Urine N} = \sum_i \text{Human or animal species } (i) \cdot \text{Discharge coefficient } (i) \cdot \text{Nitrogen excreted } (i) \cdot \text{Percentage return to field } (i) \quad (7)$$

$$\text{Cake fertilizer N} = \sum_i \text{Cake crop yield } (i) \cdot \text{Cake production rate } (i) \cdot \text{Nitrogen content in cake fertilizer } (i) \quad (8)$$

$$\text{Green manure N} = \text{Green manure production} \cdot \text{Nitrogen content } (\%) \quad (9)$$

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

$$F_{CR} = \sum_i \text{Crop yield } (i) \cdot \text{Ratio of straw to grain } (i) \cdot \text{Straw nitrogen content } (i) \cdot \text{Proportional return to field } (i) \quad (10)$$



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## 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 and Tables S1 and S2 for detailed description) (Fig. 3a, b, c and d, e, f). We conducted further statistical testing for the effect of normal distribution after cube root transformation (Table 4) and the Z statistic values of one sample K-S test for paddy fields and uplands were 0.562 and 0.440, respectively. Both 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 5). 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 5).

### 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 29.8 Gg in 1980 to 33.1 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 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 6) and were maintained at





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about 68–71 %, 20–23 %, 5.6 % and 2.8 %, respectively. From the perspective of historical change, the contribution from synthetic N fertilizers declined by 7.5 % from 74.9 % in 1980 to 69.3 % in 2007, from histosols declined by 10 % from 3.2 % in 1980 to 2.9 % in 2007, and from manures and straw increased by 29.1 % and 19.7 %, 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 296 % from 64.4 Gg in 1980 to 255 Gg in 2007, rising at an approximate rate of 10.9 % 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 6) and were maintained at about 77–79 %, 14–16 % and 6.6 %, respectively. From the perspective of historical change, the contribution of synthetic N fertilizers appeared to follow the opposite trend to paddy fields, increasing by 58.9 % from 49.5 % in 1980 to 78.7 % in 2007, rising at an average of 2.2 % per year, and the contribution from manure declined by 63.8 % from 40.7 % in 1980 to 14.7 % in 2007, and the from straw declined by 33.1 % from 9.81 % in 1980 to 6.56 % 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 94.2 Gg in 1980 to 288.4 Gg in 2007 (Fig. 6), rising at an approximate rate of 7.6 % per year. Direct $N_2O$ emissions from uplands accounted for about 82.9 % of total direct $N_2O$ emissions from croplands, with a range of 68.4–88.5 % from 1980 to 2007. The $N_2O$ emissions from paddy fields were maintained at a stable level relative to uplands and had a slight





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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 6) and have remained at about 77 %, 16 %, 6.5 % and 0.35 % 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 – 32.6 Gg and Shandong – 29.1 Gg) accounting for 11.2 % and 10.0 % of total direct $N_2O$ emissions from cropland. Tibet had the lowest emissions (0.6 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 2.36 kg N $ha^{-1}$ nationally and 17 provinces had higher values, including >4.0 kg N $ha^{-1}$ in Beijing, Jiangsu and Henan provinces. Beijing had the highest values of to 4.73 kg N $ha^{-1}$.





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# 4 Discussion

## 4.1 Emission factors of paddy fields and uplands

The emission factor is a key parameter to estimate the direct $N_2O$ emissions from croplands. The IPCC (1996) guidelines use only one default value emission factor of 1.25 % with a range of 0.25–2.25 % for calculating the direct $N_2O$ emissions from fertilized fields (IPCC, 1997) and the IPCC (2006) guidelines revised the default value to 1 % with a range of 0.3–3 % for upland crops and 0.3 % with a range of 0–0.6 % for paddy fields (IPCC, 2006). These default values might increase the uncertainties involved in calculating direct $N_2O$ emissions from Chinese croplands because they have been derived from simple up-scaling and neglect some important effective factors (Zou et al., 2010). Zheng et al. (2004) re-quantified the site-scale emission factors with an average of 1.35 % changing from 0.57 % to 2.13 % and indicated that the IPCC default value was not suitable for calculating direct $N_2O$ emissions from Chinese croplands. Yan et al. (2003) distinguished the N inputs to paddy fields and uplands, and obtained 1.19 % with a large range of −1.17–12 % as emission factors for uplands and $0.25 \pm 0.24$ % for paddy fields during the rice growing season through establishing a database by assembling numerous reports. They also calculated an emission factor for Chinese croplands of 1.04 %. In our studies we distinguished between the direct $N_2O$ emissions from paddy fields and uplands and made our calculations using much lower emission factors of 0.41 % and 1.05 % than the IPCC default value, with a minor range of 0.37–0.45 % and 1.03–1.07 %, respectively. We believe that our calculations are likely to be more accurate because our emission factors have been calculated by summarizing experimental data collected in Chinese agricultural fields.

$N_2O$ emissions from paddy fields have been reported to be very small, under 0.1 % of the N fertilizer rate, and in some cases zero $N_2O$ emissions in early studies (Freney et al., 1990; Khalil et al., 1990). It is reasonable to estimate the $N_2O$ emissions from yearly flooded paddy fields which account for 10 % of paddy fields in China but in about 80 % of the paddy fields since the 1980s management practices have been implemented that include midseason drainage and the rate of $N_2O$ emissions from this type of paddy fields is very different from yearly flooded paddy fields because 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 management of paddy fields (Yan et al., 2003). $N_2O$ emissions from paddy fields cannot be ignored but they are much smaller than those from upland areas. The rice production area represents about 20 % of the world total and 23 % of 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 factors from continuously flooded paddy fields ($0.02 \pm 0.006$ %), flooding-midseason drainage-reflooding ($0.42 \pm 0.12$ %) and flooding-midseason drainage-reflooding-moist intermittent irrigation ($0.73 \pm 0.22$ %) were calculated by Zou et al. (2007) through classifying the database of $N_2O$ into three categories taking into considerations differences in water regime and simulating the $N_2O$ emission factor using an ordinary least square (OLS) linear regression model. The emission factor of paddy fields (0.41 % with a standard error of 0.04 %) in this study is similar to 0.42 % with a higher standard error of 0.13 % under flooding-midseason drainage-reflooding management and it accounted for about 80–90 % of total rice growing area (Xing, 1999; Zou et al., 2009). The average direct $N_2O$ emission factor, calculated by establishing an empirical model based on summarizing a worldwide database of $N_2O$ emissions from fertilized fields, was 1.02 % with a range of 0.81–1.08 % from 1981 to 1998 on a national scale (Zou et al., 2010) and this was very close to the default value of 1 % in the IPCC (2006) guidelines for uplands (IPCC, 2006). We used a similar value of 1.05 % with a significantly smaller range of 1.03–1.07 % and a low standard error of 0.02 %. The emission factors of paddy fields and uplands calculated from normalized values after cube root transformation in this study have notably smaller standard errors and ranges than other studies and make a good contribution to the accurate estimation of direct $N_2O$ emissions from Chinese croplands.

## 4.2 Direct $N_2O$ emissions from Chinese croplands

In the literature there are many methods from simple empirical formulae to complicated biogeochemical models used for estimating $N_2O$ emissions from Chinese croplands (Xing, 1998; Xing et al., 1999; 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 7), our results of direct $N_2O$-N emissions 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 29.8 Gg and 33.5 Gg in 1980 and 2000, respectively, with the mean value of 31.9 Gg-N in the 1990s was very close to 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 considered the N fixed by legume crops in the study. Xing (1998) and Zheng et al. (2004) calculated almost the same results of 88 Gg N in 1995 and 90.8 Gg N in 1990s used different methods, both of these 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 upland in this study were similar with the results of Zou et al. (2010) in 1980 and 2000, respectively. Although considering different sources of direct $N_2O$ emissions, the result of 184.2 Gg N in 1990s from Zheng et al. (2004) was much similar as 189.2 in 1994 in this study, that is because almost all of the direct $N_2O$ emissions from chemical N fertilizer, crops straw and manure, such three of them accounted for 96.7–97.8 % of total direct $N_2O$ emissions in this study. The result of Xing (1998) in 1995 was very close to Yan et al. (2003) about 310 Gg N quite higher than 195 Gg N in present study.





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Total $N_2O$ emissions from Chinese croplands were about 340 Gg $N_2O$-N in 1995 as calculated by Xing et al. (1999) and Yan et al. (2003), both of them between the results of 310 Gg $N_2O$-N by Li et al. (2001) and 398 Gg $N_2O$-N by Xing et al. (1999). All of them are higher than 228.7 Gg $N_2O$-N in this study, and this might be explained by Xing (1998) and Li et al. (2001) including background $N_2O$ emissions and Xing et al. (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 et al. (1999), this is also much higher than in our study due to Xing et al. (1999) not distinguishing between direct $N_2O$ emissions from paddy fields and uplands and simply using the IPCC (1996) default value of 1.25 % as emissions factor, 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 ours 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 38 % 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 through calculating the sum of $N_2O$ emissions from croplands in each province. This is much higher than any estimates from research, 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 resulted 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 model for estimating direct $N_2O$ emissions on a national scale.





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### 4.3 Further research

$N_2O$ 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). Our observation sites were mostly distributed in the east of the country and we still lack experimental data form central and western regions (Fig. 1). New observation sites should be developed to deal adequately with the high spatial variability of emission factors (Zheng et al., 2004). Measurements often involve large uncertainties due to low frequency of observations (usually 1 or 2 measurements per week and referred to as less intensive measurements), incomplete observations (most last for a short time or during the crop growing season, not on a yearly basis), no replication in field experiments, and no control treatment without N fertilizer application (Bouwman et al., 2002), all factors that increase the uncertainty of emission factors. In the future we may work on a much larger measurement database covering the spatial variability across the whole of China and develop a statistical model between $N_2O$ emissions and the main controlling factors (soil, climate, cropping system and management practices) to reduce the uncertainty of estimates of $N_2O$ emissions from Chinese croplands.

## 5 Conclusions

Direct $N_2O$ emissions from croplands have increased rapidly, with an annual rate of increase of 7.6 % per year, and total direct $N_2O$ emissions increased to 288.4 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 82.9 % of total direct $N_2O$ emissions from total croplands. The contributions of different N sources to direct $N_2O$ emissions were synthetic N fertilizers > manures > straw and these were stable at 77.6 %, 15.6 %, and 6.5 % in 2007. The provinces with larger total emissions were mainly in north China and in Sichuan Basin in 2007, and the





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highest and lowest direct $N_2O$ emissions provinces were Henan (32.6 Gg N) and Tibet (0.6 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 2.36 kg N $ha^{-1}$, 17 provinces higher than this value, and Beijing reaching the highest value of 4.73 kg N $ha^{-1}$.

**Supplementary material related to this article is available online at: http://www.biogeosciences-discuss.net/8/6971/2011/ bgd-8-6971-2011-supplement.pdf.**

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**Table 1.** Human or animal excreta parameter, its N content (fresh sample) and percentage of returning to 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 et al. (1997); Li et al. (1998); Wang et al. (2006); Ma et al. (2006); Liu et al. (2006).





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**Table 2.** The ratio of oil crop seed converting to cake fertilizer and N content in cake fertilizer and green manure*.

| Organic material | Ratio of oil crop seed converting to cake fertilizer (%) | N content ($g\,kg^{-1}$) |
|---|---|---|
| Rape cake | 0.55 | 53.5 |
| Cottonseed cake | 0.8 | 42.9 |
| Soybean cake | 0.85 | 66.8 |
| Peanut cake | 0.5 | 69.2 |
| Sesame cake | 0.5 | 50.8 |
| Sunflower seeds 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).





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**Table 3.** 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 (leaf/stem) | 0.3 | 11.0 | 90 |
| Beet (leaf/root) | 0.5 | 2.5 | 90 |
| Tobacco leaf | 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).





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**Table 4.** Normal test (one-sample K-S test) of the normalized frequency after cube root transformation.

| Item | | Value | |
|---|---|---|---|
| | | Paddy fields | Upland |
| 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 |

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**Table 5.** $N_2O$ emission factors (%) of Chinese paddy fields and uplands calculated by different methods.

| Land type | No. of samples | Method | Mean | Standard deviation | Range |
|---|---|---|---|---|---|
| | | Original data | 0.54 | 0.53 | 0.0036–2.13 |
| Paddy fields | 195 | 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 |
| | | Original data | 1.49 | 1.23 | 0.035–4.8 |
| Uplands | 261 | Normalized through origin data | 1.90 | 0.98 | 0.92–2.99 |
| | | 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 |

**Table 6.** Contribution of different sources to direct $N_2O$ emissions from Chinese croplands (%).

| Year | Synthetic N fertilizers | | | Manures | | | Crop residues | | | Histosols | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Paddy fields | Uplands | Total croplands | Paddy fields | Uplands | Total croplands | Paddy fields | Uplands | Total croplands | Paddy fields | Uplands | Total croplands |
| 1980 | 74.91 | 49.53 | 57.55 | 17.17 | 40.66 | 33.23 | 4.73 | 9.81 | 8.20 | 3.19 | / | 1.01 |
| 1985 | 73.57 | 65.08 | 66.98 | 17.20 | 25.39 | 23.56 | 5.91 | 9.53 | 8.73 | 3.31 | / | 0.74 |
| 1990 | 72.11 | 73.29 | 73.09 | 18.43 | 19.21 | 19.08 | 6.32 | 7.50 | 7.30 | 3.15 | / | 0.53 |
| 1995 | 72.25 | 75.52 | 75.21 | 19.41 | 17.22 | 17.54 | 5.53 | 7.06 | 6.83 | 2.81 | / | 0.42 |
| 2000 | 70.93 | 77.25 | 76.41 | 20.58 | 16.11 | 16.70 | 5.65 | 6.64 | 6.51 | 2.83 | / | 0.38 |
| 2005 | 68.68 | 77.30 | 76.27 | 23.01 | 16.06 | 16.89 | 5.46 | 6.64 | 6.50 | 2.85 | / | 0.34 |
| 2007 | 69.31 | 78.72 | 77.64 | 22.16 | 14.72 | 15.57 | 5.66 | 6.56 | 6.46 | 2.87 | / | 0.33 |





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**Table 7.** Comparative estimation of national total $N_2O$ emissions form Chinese croplands from different sources (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 % | 29.8 | 64.4 | 94.2 | This study |
| 1985 | Localized EFds | FSN+FAW+FCR+FOS | Paddy fields 0.41 %, upland 1.05 % | 28.7 | 99.7 | 128.4 | This study |
| 1990 | Localized EFds | FSN+FAW+FCR+FOS | Paddy fields 0.41 %, upland 1.05 % | 30.2 | 148.4 | 178.6 | This study |
| 1994 | Localized EFds | FSN+FAW+FCR+FOS | Paddy fields 0.41 %, upland 1.05 % | 28.8 | 189.2 | 218.0 | This study |
| 1995 | Localized EFds | FSN+FAW+FCR+FOS | Paddy fields 0.41 %, upland 1.05 % | 33.8 | 195.0 | 228.7 | This study |
| 2000 | Localized EFds | FSN+FAW+FCR+FOS | Paddy fields 0.41 %, upland 1.05 % | 33.5 | 219.3 | 252.8 | This study |
| 2005 | Localized EFds | FSN+FAW+FCR+FOS | Paddy fields 0.41 %, upland 1.05 % | 33.3 | 244.5 | 277.8 | This study |
| 2007 | Localized EFds | FSN+FAW+FCR+FOS | Paddy fields 0.41 %, upland 1.05 % | 33.1 | 255.3 | 288.4 | 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 et al. (1999) |
| 1985 | Revised IPCC (1996) guidelines | FSN+FAW+FBN+FCR+FOS | 1.25 % | | | 253 | Xing et al. (1999) |
| 1990 | Revised IPCC (1996) guidelines | FSN+FAW+FBN+FCR+FOS | 1.25 % | | | 266 | Xing et al. (1999) |
| 1995 | Revised IPCC (1996) guidelines | FSN+FAW+FBN+FCR+FOS | 1.25 % | | | 336 | Xing et al. (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) |



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**Table 7.** Continued.

| Year | Method | $N_2O$-N sources considered | Emission factors | Paddy fields | Uplands | Total emissions | Reference |
|---|---|---|---|---|---|---|---|
| 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) |
| 1990s | OLS model | 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).

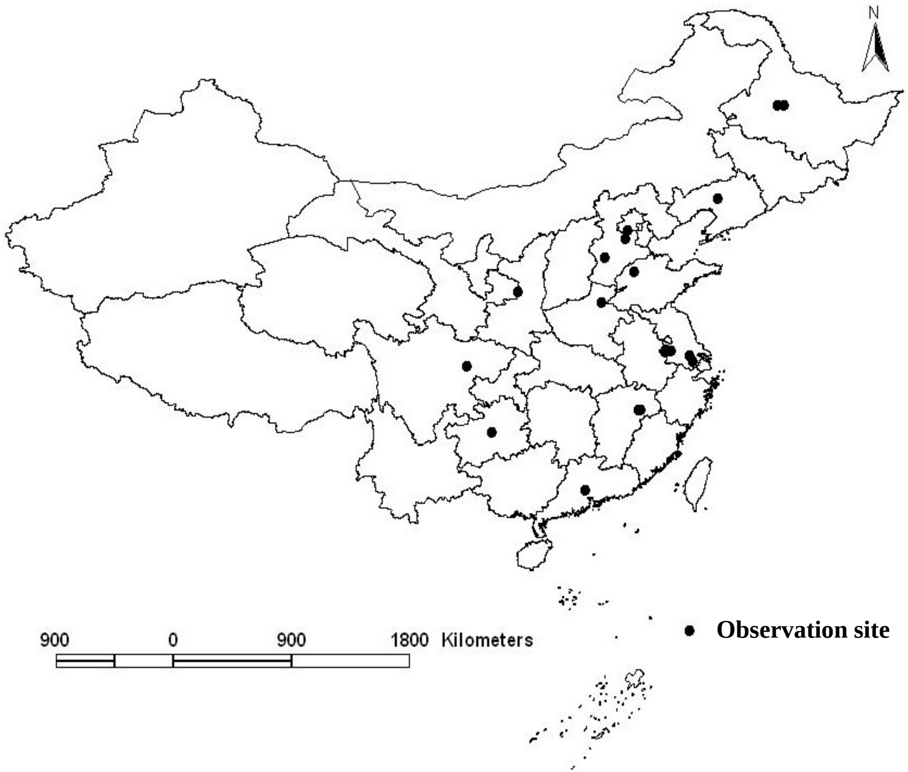


**Fig. 1.** Locations of the $N_2O$ emission observation sites from croplands in China.





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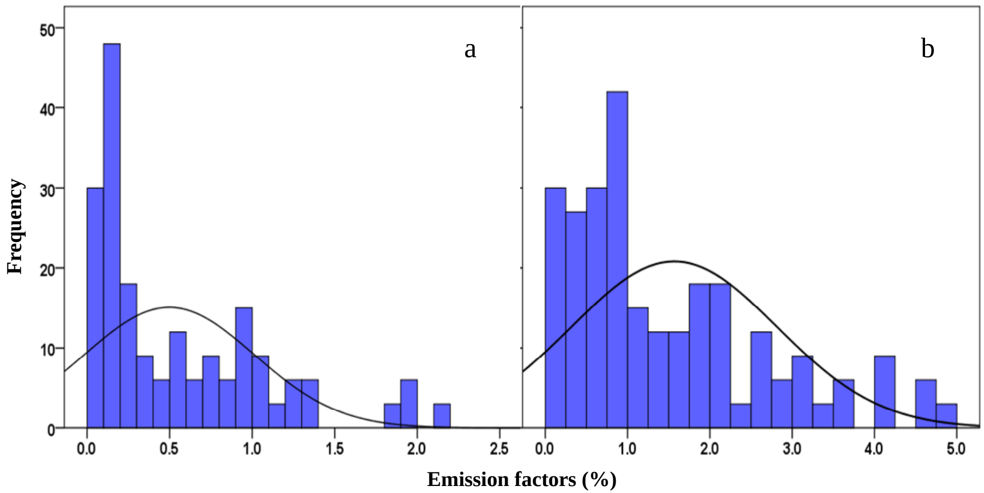


**Fig. 2.** Frequency distribution of the original data of $N_2O$ emission factor of paddy fields **(a)** ($n = 195$) and uplands **(b)** ($n = 261$).





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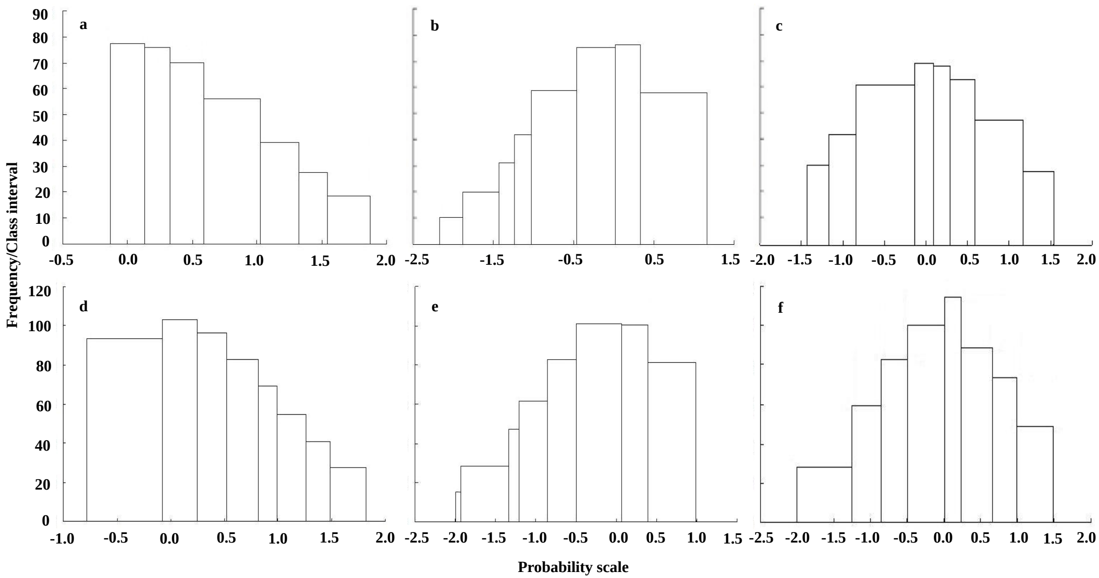


**Fig. 3.** Frequency distribution of $N_2O$ emission factors of paddy fields ($n = 195$, **(a)**: normalized through original data; **(b)**: normalized after log transformation; **(c)**: normalized after cube transformation) and uplands ($n = 261$, **(d)**: normalized through original data; **(e)**: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

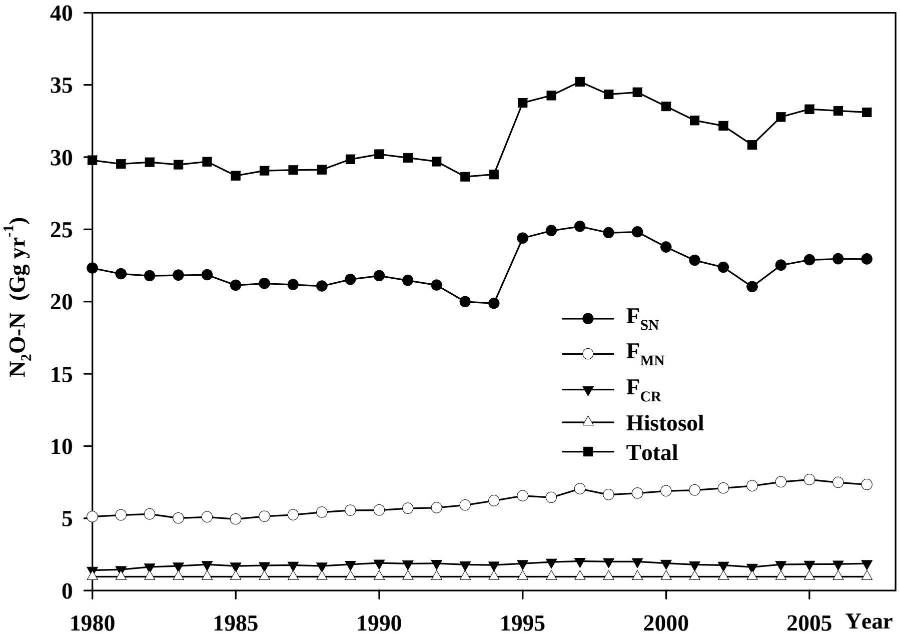


**Fig. 4.** Total direct $N_2O$ emissions from paddy fields between 1980 and 2007, $F_{SN}$, $F_{MN}$, $F_{CR}$ and Histosols represent direct $N_2O$ emissions from synthetic fertilizers, manures, crop residues and histosols, respectively.





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New estimates of direct $N_2O$ emissions from Chinese croplands

B. Gao et al.

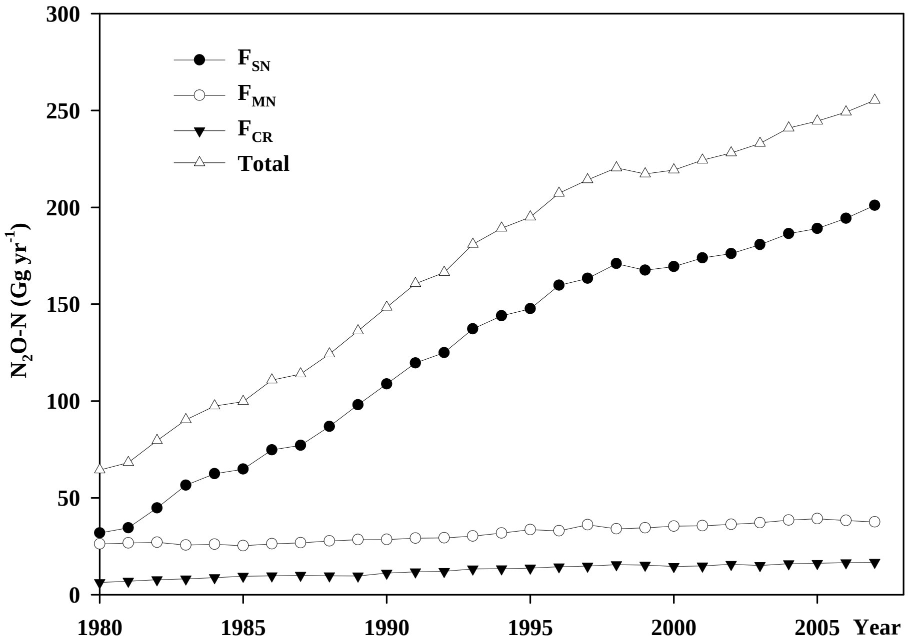


**Fig. 5.** Total direct $N_2O$ emissions from uplands between 1980 and 2007.





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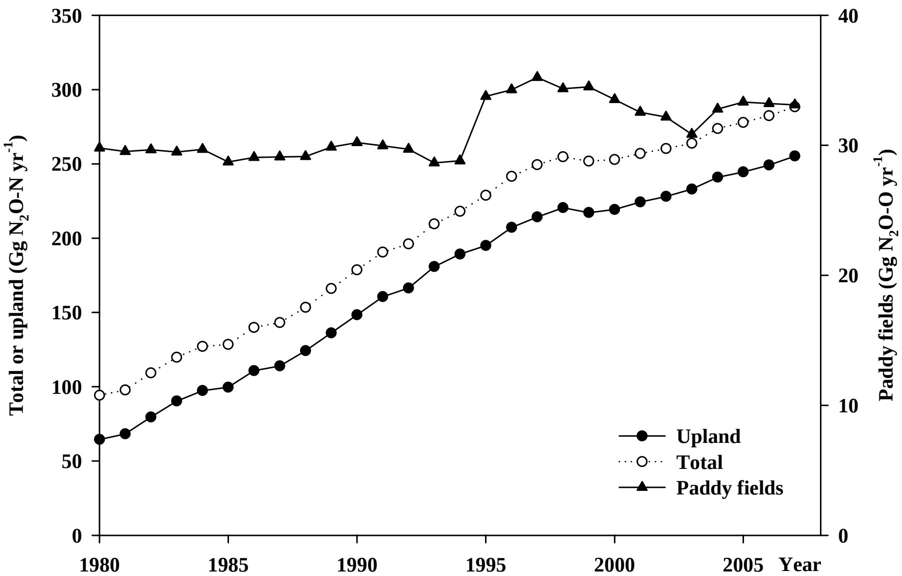


**Fig. 6.** Historical changes in direct $N_2O$ emissions from Chinese croplands between 1980 and 2007.

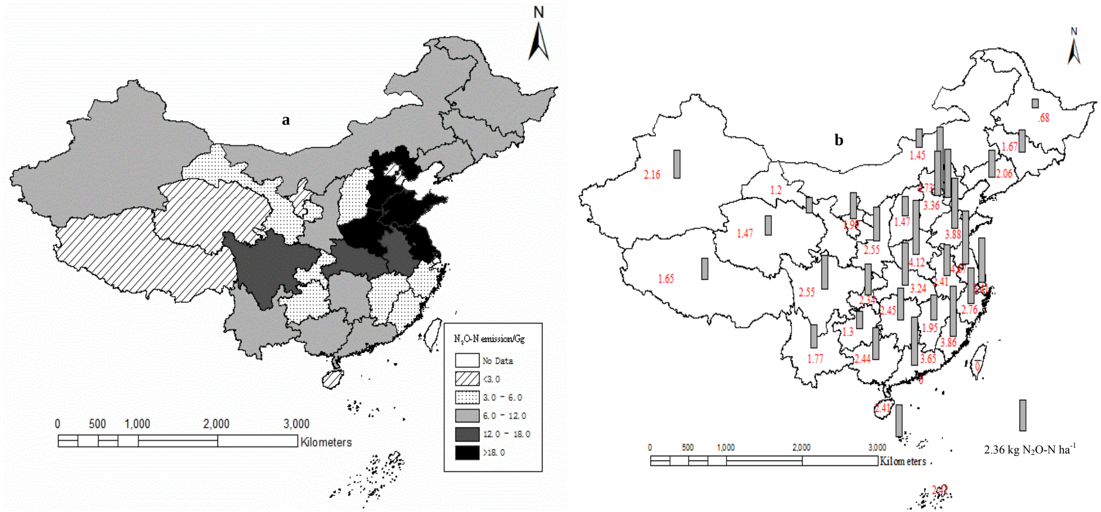


**Fig. 7.** The provincial pattern of total direct $N_2O$ emissions from Chinese croplands **(a)** and direct $N_2O$ emissions per unit area of arable land **(b)** in 2007, bar with 2.36 kg $N_2O$-N $ha^{-1}$ represents the scale of average direct $N_2O$ emissions per unit area of arable land of all provinces.





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