Re-assessing Nitrous Oxide Emissions from Croplands Across Mainland China

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Abstract: Reliable quantification of nitrous oxide emission is a key to assessing efficiency of use and environmental impacts of N fertilizers in crop production. In this study, N_2O emission and yield were quantified with a database of 853 field measurements in 104 reported studies and a regression model was fitted to the associated environmental attributes and management practices from China's croplands. The fitted emission model explained 48% of the variance in N_2O emissions as a function of fertilizer rate, crop type, temperature, soil clay content, and the interaction between N rate and fertilizer type. With all other variables fixed, N_2O emissions were lower with rice than with legumes and then other upland crops, lower with organic fertilizers than with mineral fertilizers. We used the subset of the dataset for rice - covering a full range of different typical water regimes, and estimated emissions from China's rice cultivation to be 31.1 Gg N_2O-N per year. The fitted yield model explained 35% of the variance in crop yield as a function of fertilizer rate, temperature, crop type, and soil clay content. Finally, the empirical models for N_2O emission and crop yield were coupled to explore the optimum N rates (N rate with minimum N_2O emission per unit yield) for combinations of crop and fertilizer types. Consequently, the optimum N application rate ranged between 100 kg N ha^{-1} and 190 kg N ha^{-1} respectively with organic and mineral fertilizers, and different crop types. This study therefore improved on existing empirical methods to estimate N_2O emissions from China's croplands and suggests how N rate may be optimized for different crops, fertilizers and site conditions.

Keywords: nitrous oxide, croplands, nitrogen fertilizers, greenhouse gas emission, regression model, data synthesis, China's agriculture.

1 Introduction

Nitrogen (N) plays a key role in enhancing food production to support the world's growing population – being an essential nutrient supporting plant growth for food and feed (Zhang et al., 2012; Sutton et al., 2013). Apart from the natural conversion from nitrogen gas (N_2) by lightning fixation and bacterial fixation, reactive nitrogen (Nr) is increasingly produced through the Haber-Bosch process in industry of nitrogen fertilizers developed since early 20th century. Being a pivotal player in crop production, the ever-increased application of N fertilizers had dramatically increased food production albeit at significant environmental cost (Gruber & Galloway, 2008). Fertilized N in cropping systems could find its way to the atmosphere and aquatic systems via ammonia (NH_3) volatilization, leaching of nitrate/nitrite and emission of nitrous oxide (Wrage et al., 2001; Ju et al., 2009). These end-products of lost N are known to cause secondary inorganic aerosol formation and thus haze pollution (Liu et al., 2017), and destruction of the stratospheric ozone layer (Ravishankara et al., 2009), and again impact on human health (Galloway et al., 2008; Farnworth et al., 2017).

As a potent greenhouse gas, production and emission of nitrous oxide (N_2O) in global nitrogen (N) cycle is particularly important for climate change (Mosier et al., 1998). With a global warming potential (GWP) approximately 265 times as CO_2 over a 100-year time horizon (IPCC, 2013), N_2O emissions were around 8.4 Tg N_2O yr^{-1} globally, with, for example, 58% estimated to be contributed by agriculture in 2005 (Smith et al., 2007). Since global application of N fertilizer is projected to increase in world agriculture to meet the food demand of the increasing world population, N_2O emissions in global agriculture are also projected to increase in the coming decades (Reay et al., 2012). The key challenge this presents to the agricultural sector is to maximize crop productivity while minimizing N_2O emissions from fertilized field (Galloway et al., 2008).

In recent decades, a large number of field studies have been carried out to characterize N losses, including N_2O emissions, and exploring N use efficiency in various agricultural systems. Bouwman et al. (2002) created a global database of field N_2O emissions from a total of 388 studies, of which however only 3% of the data was from China. The existing field data has facilitated development of ecosystem N models to predict N_2O emissions from agricultural systems (Heinen 2006), including, for example, the dynamic process-based models of DNDC (Li et al., 1992), SUNDIAL (Smith et al., 1997) and DAYCENT (Ogle et al., 2010). The dataset had also been used directly by Bouwman et al. (2002) to develop an empirical model of N_2O emissions as a function of several field and management variables, which informed the choice of the emission factor of 1% (meaning 1% of fertilizer N is emitted as N_2O-N) adopted in the IPCC Tier I methodology (IPCC, 2006).

Accurate and precise prediction of N_2O emissions in croplands is difficult since the biotic and abiotic factors influencing N_2O emission in field are temporally dynamic and spatially heterogeneous, and influenced by a number of factors related to climate, soil quality, fertilizer application, cropping systems and management practices (Ladha et al., 2016; Tang et al., 2016; Lam et al., 2016; Yue et al., 2017). For instance, N_2O emission rates were lower for flooded or paddy rice than upland crops as the anaerobic conditions in wetland soils tend to encourage complete denitrification to N_2 (Gerber et al., 2016). Also, many existing models predicting N_2O emission from croplands were developed and parameterized in regions where agriculture was well-developed and fertilizer use efficiency was relatively high. However, much of the projected increased in food production, and thus N use, is expected to occur in the developing countries (Holland et al., 1999; Tilman, et al., 2001), particularly in the populous regions of the Indo-Gangetic Plain (IGP), southwest Asia and Yangtze and Yellow river plain of eastern Asia. Thus, quantifying N_2O emissions and developing more robust models suitable in these regions is critical to enable better prediction of global agricultural N_2O emission and identify improved management practices in these regions.

China is a country representing 19 % of the world's population and 7 % of net GHG emission from Agriculture, Forestry and Other Land Use (AFOLU) in 2014 (FAOSTAT, 2017). Total annual N_2O emissions from fertilized croplands in China had previously been estimated (Zou et al., 2007; Gerber et al., 2016), using the aforementioned existing models calibrated with global data in which China was under-represented. China's agriculture covered 166 M ha croplands and 23.6 Mt N was used for food production in 2015 (NBSC, 2017). Between 2002 and 2014, China had achieved a crop yield increase of 21% with an increase by 23.4% of N fertilizer application. The increase in N fertilizer application resulted in decreased N use efficiency (NUE) in China's croplands, resulting in negative environmental impacts such as soil acidification (Guo et al., 2010), water eutrophication (Le et al., 2010), air pollution (Sapkota et al. 2014;

Liu et al., 2017), and severe human health risks (Farnworth et al., 2017; Gu et al., 2012; Galloway et al., 2008). Better knowledge of the impacts of crop nitrogen use can be used to identify more efficient and lower emitting N management practices in China's agriculture which would in turn not only help the state to cut its GHG emissions as part of its commitments to the Paris Agreement (UNFCCC, 2015), but also to reduce other N losses while sustaining food production.

It is critical to identify ways to balance quantity of grain, NUE, and environmental impacts in China, given the increasing human population and limited resources (Galloway et al., 2008; Liu et al., 2016; Xia et al., 2017). Additional increase in N fertilization over the existing high rates might result in marginal yield benefits (Brentrup et al., 2004; Liu et al., 2016) but at the cost of proportionally higher N_2O emissions (Bellarby et al., 2014). As suggested by Van Groenigen et al. (2010), since yield response curves tended to flatten for higher N rates, above a certain point yield-scaled N_2O emissions increased progressively with N application rate. Yet, it is still unclear precisely how such yield based N_2O emissions change with N application for a given crop system, and soil and climate characteristics in the context of Chinese agriculture. Moreover, it was also questionable if the default global fertilizer-induced emission factor of 1% in Tier I approach by IPCC (2006) applies to croplands of China given that it the underpinning data contained few studies (only 3% of the total dataset) from China.

We hypothesized here that N_2O emissions from croplands varied with crop type, N fertilizer type and rate and climate, across various agricultural systems of China. We also hypothesized that such variation could be modeled to predict N_2O emissions and explore the main drivers for N_2O emissions from key Chinese croplands. In this study, field data of N_2O emissions in reported studies were reviewed to create a country-level database and a multi-variate empirical model fitted to predict N_2O emissions in China. Using the model, N_2O emission rates were compared between different fertilizer and crop types and the Emission Factors (EF) for China's croplands were derived for comparison to the IPCC default factors. Furthermore, a cross-system variability was elucidated with the model calculation of the cumulative N_2O emission for rice cultivation in 2014. With an additional multi-variate empirical model of crop yield derived from our database, yield-scaled N_2O emission were identified for different crops and fertilizer types to explore approaches to optimize N use efficiency in China's crop production.

2 Materials and methods

2.1 Database creation

A dataset with a total of 853 seasonal cumulative N_2O field emission measurements from 104 studies in China's agricultural fields was compiled for this study. A primary dataset was compiled from the scientific literature reporting field measurements of N_2O emissions from cropping systems of China published over a time span of 2001-2016. Firstly, papers were collected and archived via searching the databases of CNKI (China National Knowledge Infrastructure), ISI-Web of Knowledge and Google Scholar with keywords of "nitrous oxide" "emission" "chamber" "fertilizer" and "China". From the collected literature, data pairs of N_2O emissions under a fertilizer treatment and a non-fertilized control were retrieved and archived, retaining a total of 71 studies. In addition, 33 studies meeting our criteria in the dataset used by Albanito et al. (2017) were checked and added to the primary dataset. Finally, a dataset comprising 853 data pairs from a total of 104 studies were constructed and used in this study. The reported measurements were located across the mainland China, between the longitude of 85.0° to 139.6° and latitude of 21.9° to 47.4° (Fig. 1).

Information in the dataset included geographic location (latitude and longitude); climate data - annual average temperature (ranging between -0.4℃ and 21.3℃), annual average precipitation (values from 193 mm to 1795 mm); soil characteristics - including clay content, organic carbon and nitrogen content, bulk density, and pH; soil type - classified into 8 soil texture classes (Clay loam, Loam, Sand, Sandy clay loam, Sandy loam, Silt loam, Silty clay, Silty clay loam) following the United State Department of

Agriculture (USDA) classification; cropping system; crop types aggregated into 4 broad categories (Table 1); fertilizer types classified into 3 broad categories (Table 1); fertilizer application rate; management practices - including water management, tillage, straw return, and irrigation; length of experimental monitoring; seasonal cumulative N_2O emission; and grain harvest (more detailed information is shown in Table S1). This information was interpolated to every point in which data on N_2O emissions was considered.

In order to conduct the spatial analysis of individual variables, a 0.5×0.5 degree grid cell was created covering all the cultivated areas in China. Climate data were obtained from the China Meteorological Data Web (http://data.cma.cn). Soil data, including land use type, soil carbon content, pH and clay content were obtained from the Harmonized World Soil Database (FAO/IIASA/ISRIC/ISS-CAS/JRC, 2012). The area ratio of rice cultivation to non-rice cultivation in each grid cell was obtained from Monfreda et al. (2008). Data for N fertilizer application rates across years and regions were obtained from the China Agricultural Cost-benefit Data Assembly (DP-NDRC, 2015).

2.2 Modelling N_2O emissions

During the explanatory phase of the analysis, the data was tested for normality and it was observed that values of the cumulative seasonal N_2O emissions (*Cum* N_2O, in kg N ha^{-1} crop-cycle^{-1}) including the single or rotation cropping systems were highly skewed. Therefore, the original data of *Cum* N_2O was log-transformed to use natural logarithm ln (*Cum* N_2O), as the response variable throughout the analysis. This was required in order to meet the assumptions for performing a regression analysis (Zuur et al 2007). Eleven measurements were identified as obvious outliers and excluded from the analyses (Table S1). We suspected an accidental misreporting of these values and therefore excluded them. Interactions and co-dependence among variables were also examined to avoid co-linearity among explanatory variables in the model (Table S2).

The effect of different explanatory parameters on the log-transformed response variable, ln (*Cum N_2O*), was then investigated by fitting a set of linear models. To have a preliminary indication of the effect of the aforementioned variables on ln (*Cum N_2O*), univariate models including each covariate were initially fitted separately (Figure S3, Table S2). Subsequently, a stepwise approach to model selection was implemented as follows: a set of linear regression models were fitted, including systematically different combinations of the different potential explanatory parameters (Table S2). We firstly discarded the variables with the p-value >0.05, which was our significance threshold. Then, we chose the model based on R-square values. The functional forms of the fitted models were:

$$\ln(y) = \alpha + \sum_{i}^{N} \beta_i \ \ x + \varepsilon \qquad (1)$$

Where y is the target variable, *Cum N_2O*, in kg N ha^{-1}; x stands for the potential explanatory variables; α and β_i represent the model coefficients; ε indicates the model error.

For model diagnosis, the absence of pattern in the residuals, whether they were normal and centered was checked. To evaluate the model's accuracy, the bias and root-mean-squared error (RMSE) were calculated:

$$Bias_{(i)} = \sum(\widehat{V_i} - V_i)/n \qquad (2)$$

$$RMSE_{(i)} = \sqrt{\left(\sum(\widehat{V_i} - V_i)^2\right)/(n-p)} \tag{3}$$

Where, $\widehat{V_i}$ and V_i represent the estimated value of target variable from the fitted equation and the measured value by the original studies, n is the number of target values; and p is the number of parameters in the relevant model.

A variance analysis was carried out to calculate the variance explained by each of the significant factors and assess the importance of each covariate over the others. This was achieved by calculating the variance explained by each covariate divided by the total residual variance.

2.3 Evaluating the effect of fertilizer type on N_2O emissions

We used the model developed in section 2.2 to compare the rate of the N_2O emission as a function of fertilizer rate from different fertilizer types for particular crop types. Here we were interested in the effect of different fertilizer types on N_2O emissions with the changes in the amount of N fertilizer applied, thus in order to focus only on these particular variables (including N rate, crop type, and fertilizer type), we eliminated the variances due to soil type (clay content) and climate (temperature) by setting constant values for these variables in this particular case we aimed to evaluate. The fixed value we used was the average of these variables in the dataset. Considering cropland N_2O emission responses to fertilizers varying with crop types, the croplands were classified into groups “Legume”, “Rice”, “Rice with cover crop” and “Other”. Such classification was based partly on expected differences and partly on the need to achieve a balanced representation of data points in each class over the dataset.

2.4 Spatial distribution of N_2O emissions for rice in China

Rice paddy fields is of particular importance to China's agricultural development with a long history. Additionally, given its special water regimes in China, like continuous flooding, flooding-midseason drainage-reflooding, which make an effect on soil nitrification and denitrification (Zou et al., 2007), it's very necessary to test the accuracy of the model working on the N_2O emission calculating for rice paddy fields. Therefore, we used this crop as a case example to carry out a more detailed evaluation of N_2O emissions spatial distribution. For rice cultivation, the annual instead of the seasonal N_2O emissions from mineral fertilizer use were studied. The emission of rice cultivation was mapped for each 0.5° by 0.5° grid, using the model described in section 2.2 and the spatial covariates defined in section 2.1. Covariates of soil clay content and temperature were also used as described above. Firstly, the gridded N_2O emissions were calculated using R (version 3.4.0) using the package "Matrix" (Bates & Maechler, 2015) with gridded significant climate and the soil profile factors, and a map was generated using ArcGIS 10.2.

2.5 Optimum N use

2.5.1 Modelling yield

To identify the optimum N fertilizer rate, the factors affecting yield variation under different conditions were investigated. A similar modelling approach as for the N_2O emissions model (section 2.2) was carried out: a multivariate linear model was fitted, and its performance and accuracy (RMSE and bias) evaluated. Herein, the crop yield data were not log-transformed.

2.5.2 Identification of N rate for optimum yield and emission

The N_2O and yield models fitted in previous sections 2.2 and 2.5.1, were combined to identify the optimum N rates (*Opt N*). In other words, the N rate at which the lowest emissions intensity (N_2O-N/ton production) is obtained, which can be estimated as the minimum of the curve $N_2O/Yield$ in the unit of kg N_2O-N/ ton yield. We determined this optimum for each combination of crop type and fertilizer type from the covariate classes in the above models, with all other covariates set to the average of those in our dataset.

All the analyses in this study were conducted in R version 3.4.0 (R Core Team, 2017), using the R packages: “lattice” (Sarkar and Deepayan 2008), “car” (Fox and Weisberg, 2011), “mgcv” (Wood, 2003), “Matrix” (Bates & Maechler, 2015).

3 Results

3.1 N_2O emission model

Based on each variable with the p-value <0.05 (significance) and the best R-squared, the out-coming of the best model selection process was:

$$\ln(CumN_2O) = -2.7094 + 0.0045 \times N\ \ rate + 0.0742 \times Temp + 0.0134 \times Clay + \ \ C_1 crop\ \ type + C_2\ \ N\ \ rate \times fert\ \ type + \varepsilon \qquad (4)$$

Where *Cum N_2O* is the cumulative N_2O emissions in kg N ha^{-1}; *N rate* represents the application amount of nitrogen fertilizer in kg N ha^{-1}; *Temp* means the annual average temperature (℃); *Clay* indicates the fraction of clay (%). The significant variables were N rate, temperature, clay content, crop types, and the interaction between N rate and fertilizer type.

The coefficient values in equation (4) expressed as "Value ± Standard Error" (the same as below) were -2.7094 ± 0.1713, 0.0045 ± 0.0003, 0.0742 ± 0.0132, and 0.0134 ± 0.0030, respectively. Fitted values of C_1 for the different crop type classes were: background (0) for "Legume", 0.7002 ± 0.2150 for "Other", -0.1879 ± 0.2503 for "Rice", and -1.6339 ± 0.4893 for "Rice with cover crop". Values of C_2 for the different base fertilizer types were: background (0) for "Mineral" fertilizer type and -0.0018 ± 0.0003 for "Organic", Null for "Control" treatment (no fertilizer applied).

The R^2 of this model was 0.48, the RMSE was 5.5e-14 and the bias was -1.6e-15, with no pattern in the residuals (Figure S1). N rate was the main factor explaining the variation in emissions, accounting for 24 % (Fig. 2a). The variables Temperature, Crop type, Clay content and the interaction between N rate and fertilizer type explained 13%, 7%, 2% and 3% variance respectively.

3.2 Comparison of the emissions from different fertilizer and crop types

Regardless of fertilizer types, “Other” crops, meaning maize, wheat etc., always exhibited higher N_2O emissions than the other three crop types at the same N application rate (Fig. 3); followed by “Legume”; while the emission from “Rice” only was higher than rice in combination with cover crops (“Rice with cover crop”). In terms of fertilizer types, increase in N_2O emissions with N application rate was greater from “Mineral” (Fig. 3a) than from “Organic” (Fig. 3b), especially at rates over 100 kg N ha^{-1} (due to the interaction with N rate).

3.3 Spatial heterogeneity of emissions from rice cultivation

The calculated annual N_2O emissions were seen to be highly variable spatially (Fig. 4). Annual N_2O emissions per region were higher from the warm/humid climate regions of South, Southwest, and Yangtze River than from other regions in China owing to double rice cropping and a large rice cropping area; the high annual N_2O emission rate per hectare was identified in the agro-region of Inner Mongolia and along the Great Wall, Huang-Huai-Hai, and Gansu-Xingjiang as a result of high N application rates and/or temperatures.

3.4 Optimum values

3.4.1 Yield Model

The model for yield as a function of N rate and other significant variables was (details in Table S3):

$$Yield = -2.3626 + 3.1888 \times log(N\ \ rate) - 0.5271 \times Temp + 0.0426 \times Clay +$$

$$C_3 crop\ \ type + \varepsilon \qquad (5)$$

Where *Yield* is the grain yield of crops in t ha^{-1}; *N rate*, *Temp*, *Clay* (see above). The coefficient values in equation (5) expressed as “Value ± Standard Error” were -2.3626 ± 2.4254, 3.1888 ± 0.4000, -0.5271 ± 0.0793, and 0.0426 ± 0.0198, respectively. As for C_3, the base crop was “Legume” (no need to add the C_3 term), and C_3 was equal to 1.8263 ± 0.5174 for “Rice”, and 1.1850 ± 1.5880 for “Rice with cover crop”. The adjusted R^2 value was 0.35, the RMSE and bias were 0.35 and 8.3e-3, respectively. The main drivers explaining yield values were N rate and temperature (Fig. 2b).

3.4.2 Optimum N rates

Fig. 5 showed the relationship of yield-scaled N_2O emissions and N application rate for combinations of crop type and fertilizer type. In all cases a minimum in the yield-scaled N_2O emissions curve occurred between 98 and 190 kg N per hectare, and this value was achieved at a higher N rate for organic than mineral fertilizer. The slope was in general lower for organic fertilizer types than mineral fertilizers, especially for higher N rates, (Fig. 5) which might indicate a higher risk of oversupplying the highly mobile forms of N in mineral fertilizers compared with the relatively slow release forms in organic fertilizers (predominantly organically bound rather than in the form of NH_4^+ or NO_3^- ions.)

4 Discussion

4.1 Seasonal N_2O emissions in relation to crop and fertilizer types

For the model, the key drivers which had significant effect on N_2O emissions, in the order of their relative contributions, were: fertilizer application rate; temperature; clay content (positive in the three cases); crop type and the interaction between N rate and fertilizer type. Of course, these factors might have different loading depending on the crop or fertilizer type as described in Section 3.1. As already shown by the studies of Bouwman et al., (2002); Buckingham et al., (2014) and Zhou et al., (2017), N_2O emissions for agricultural land use were not only affected by N fertilizer rate, but also by climate, soil, crops and fertilizer types. Clay content was known to affect thus moisture status, water filled pore space and gas diffusion associated to soil texture (Dobbie & Smith, 2003). If clay soils were not completely water-saturated, fine soil texture with restricted drainage was prone to high N_2O emission for their high water holding capacity and capillary pores within aggregates (Bouwman et al., 2002). And this explained the positive correlation of N_2O emissions to clay content in the dataset. Meanwhile, clay content was also related to soil oxygen condition mediating the soil redox (Eh range) for N_2O production in nitrification and denitrification processes (Verstraete & Focht, 1977; Hou et al., 2000).

Emissions differed between fertilizer types in our model. With relatively high N input, N_2O emissions were significantly lower under organic (Fig. 3b) than under mineral fertilizers (Fig. 3a). This was in contrast to the finding of a meta-analysis of global data (Zhou et al. 2017) that reported manure N application significantly increased N_2O

emissions over mineral N application. There had been controversial debates on whether or not manure application led to increase in N_2O emissions compared to mineral fertilizers (Petersen et al., 1996; Meijide et al., 2007; Zhou et al., 2017). Rather, application of organic fertilizers could bring potential benefits to soil health through improved soil carbon storage and biodiversity (Tisdall & Oades, 1982; Karlen et al., 1997), which could help crops to exert a more steady response to increasing rate of N applied (Fig. 5). For this sake, China was encouraging the use of manure to save mineral fertilizers in terms of crop-specific and region-specific recommended rates (Hou et al., 2017).

However, the lower N_2O emission and higher optimum N rates with organic N sources may partly be due to the delayed N release particularly from organic amendments such as straw, compost or biochar. It is even possible that such materials release plant-available N not only within but also after the N_2O measurement periods. This possible long-term effect is not captured in the N_2O measurements and thus not considered in the calculation of optimum N rates, which in turn may even be overestimated.

4.2 Emission of N_2O in response to N application rate

The fertilizer-induced emission (FIE) reported in this study was derived from data covering one cropping season instead of one full year as in the determination of IPCC-FIEs. Calculating with the emission from fertilized plots minus the emission from unfertilized control plots on individual sites, the estimated FIE values was 0.52 ± 0.69 % on average. Of course, this estimation may result in discrepancy to those in the IPCC

EF database estimated using full crop year. Nevertheless, N_2O emissions in our study could be in exponential response to N application rate as the model derived this study was linear function of log-transformed dependent variables (Fig. 3). Therefore, cropland N_2O emissions from the studied Chinese crop systems could not be simply quantified or predicted using linear model of EFs. Non-linear response of N_2O emissions to N rates had been already challenged with the observations by Bouwman et al. (2002), Shcherbak et al. (2014) and Gerber et al. (2016). It had been well known that a greater portion of the applied N was subject to loss *via* leaching of nitrate and emissions of NH_3 and N_2O at higher N rates (Ju et al., 2009). In addition, the log-transformed N_2O emissions was observed in linear response to several factors other than N rate as described in Section 3.1. This may demand a more reliable Tier 3 model to estimate N_2O emissions in preference to emission factor based on approaches where robust data available.

The above mentioned non-response could be used to explore the optimum N application rate, which was indicated by a minimum yield-scaled emission among the existing N application rates for a given system. A minimum yield scaled N_2O emission was around 100 kg N ha^{-1} under mineral fertilizers (Fig. 5a-5d) but in a range of 160-190 kg N ha^{-1} under organic fertilizers, for all crop types (Fig. 5e-5h). The minimum yield scaled emissions were more or less variable but low with cover crops (Fig. 5d and 5h), probably due to the additional N input through cover crops. Moreover, cover crop may help to reduce soil nitrification (Cui et al., 2006; Xie, 2016), in line with increased soil organic carbon content (Dabney et al., 2001; Tripathi et al., 2014). In paddy rice, in particular, cover crop increased carbon substrate supply to promote the process of dissimilatory nitrate reduction to ammonium and thus to inhibit the denitrification process (Kelso et al., 1997).

4.3 N_2O emission from paddy rice

N_2O emissions from flooded rice were generally lower than for upland crops (Fig.3). Furthermore, emissions under rice with a cover crop were lower than under normal rice without cover crops at a given mineral N rate, likely due to biological nitrogen fixation by the cover crop, often as nitrogen fixing alfalfa (Supplement information).

Using the emission data from our database (Fig. 4), a seasonal direct emission of N_2O from paddy rice system in China was estimated to be 31.1Gg N_2O-N for 2014. Using the default linear emission factor of 0.3 % for flooded rice (IPCC, 2006), however, the direct emission would be estimated as 25.0 Gg N_2O-N for the same year. The estimation using the model in this study was close to the value of 29.0 Gg N_2O-N estimated by Zou et al. (2007) using an ordinary least square linear regression model. Using models of linear and nonlinear regressions, Gerber et al. (2016) proposed slightly higher emission factors for rice of 0.31% and 0.36% respectively. Clearly, our estimation using database in this study could match these proposed EF values.

Water management was often concerned as a key factor affecting N_2O emissions from paddy rice production. It should be noted that water management as a factor was not retained in our model. Overall, we found no significant differences in seasonal N_2O emission between continuous flooding and intermittent flooding, for the lack of reported data. However, significant differences between water management treatments were observed only in combination with regional factors. So, how rice water regime management impacted on N_2O emissions deserves further study.

4.5 Limitations of the study

Some limitations existed in our analysis and modeling primarily of data scope. Our data was from single crop cycle measurements and the analysis was largely based on crop season instead of a full year though our estimated EF was not intended to represent annual EFs. An issue of uncertainty may have arisen with annual average data of temperature and precipitation of the study locations as crop seasonal temperature and precipitation were not reported in most the studies. Moreover, for the absence of multi-year information in our dataset though estimation of annual emissions may vary with experiment length (Albanito et al., 2017). In addition, average data from large scale observations were used in cases where local data were missing, rising the uncertainties for our model.

It should be also noted that our optimum N fertilization rates were certainly functions of several other agronomic and environmental factors not contained in our model. For example, apart from N, supply of phosphorus and potassium also affect crop yield and thus potentially affect emission response to N fertilizer (Velde et al., 2014). The low R^2 value of 0.35 for our yield model implied that many other factors were not taken into account.

5 Conclusion

In this study we observed that total seasonal N_2O emission from China's cropping systems were controlled by both inherent attributes (soil and crop type, fertilizer rate) and external attributes (climate, management practices). Using the fitted regression model of N_2O emissions we derived an estimate of seasonal N_2O emission from rice cropping systems in 2014 of 31 Gg N_2O-N, compared to 25.0 Gg N_2O-N using the IPCC default emission factor. We also reported that optimal N rates may be in a range of 100-190 kg N ha^{-1} for the crop systems and fertilizer types explored in this study. However, the model only explained 48% of the variance in the current study. This lack of explanatory power might be improved by the addition of further studies which would add statistical power and allow significant effects to be identified for more refined classification of crop and fertilizer type.

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Supporting Information

Additional Supplementary Information may be found in the online version of this article.

Table captions

Table 1 Reclassified parameters used in the fitted models.

Figure captions

Fig. 1 Geographical distribution of the studies in China used in the analysis simulating *Cum* N_2O emissions.

Fig. 2 Mean proportions of factors showed in the relative *Cum* N_2O emission model (a) and grain yield model (b).

Fig. 3 Examples of calculations using the model for seasonal cumulative N2O emissions emission for selected combinations of factor classes: (a) “Mineral” and (b) “Organic” application, and varying N application rates for the categories of four crop types for (“Legume”; “Other”; “Rice”; “Rice with cover crop”).

Fig. 4 Annual *Cum* N_2O emission rates (a) and emission intensity per hectare (b) induced by mineral fertilizer application for rice growing in 2014.

Fig. 5 Optimum of N rates using the models for *Cum* N_2O emission and yield for crop types and fertilizer types with all other conditions equal (temperature 13.07, soil clay content 22.42%): (a) optimum N rate for “Mineral & Other”; (b) optimum N rate for “Mineral & Legume”; (c) optimum N rate for “Mineral & Rice”; (d) optimum N rate for “Mineral & Rice with cover crop”; (e) optimum N rate for “Organic & Other”; (f) optimum N rate for “Organic & Legume”; (g) optimum N rate for “Organic & Rice”; (h) optimum N rate for “Organic & Rice with cover crop”.