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Estimating NH_3 emissions from agricultural fertilizer application in China using the bi-directional CMAQ model coupled to an agro-ecosystem model

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Abstract

Atmospheric ammonia (NH_3) plays an important role in atmospheric chemistry. China is one of the largest NH_3 emitting countries with the majority of NH_3 emissions coming from the agricultural practices, such as fertilizer application and livestock. The current NH_3 emission estimates in China are mainly based on pre-defined emission factors that lack the temporal or spatial details, which are needed to accurately predict NH_3 emissions. In this study, we estimate, for the first time, the NH_3 emission from the agricultural fertilizer application in China online using an agricultural fertilizer modeling system coupling a regional air quality model (the Community Multi-Scale Air Quality model, CMAQ) and an agro-ecosystem model (the Environmental Policy Integrated Climate model, EPIC), which improves the spatial and temporal resolution of NH_3 emission from this sector. Cropland area data of 14 crops from 2710 counties and the Moderate Resolution Imaging Spectroradiometer (MODIS) land use data are combined to determine the crop distribution. The fertilizer application rate and method for different crop are collected at provincial or agriculture-regional level. The EPIC outputs of daily fertilizer application and soil characteristics are inputed into the CMAQ model and the hourly NH_3 emission are calculated online with CMAQ running. The estimated agricultural fertilizer NH_3 emission in this study is about 3 Tg in 2011. The regions with the highest modeled emission rates are located in the North China Plain. Seasonally, the peak ammonia emissions occur from April to July.Compared with previous researches, this method considers more influencing factors, such as meteorological fields, soil and the fertilizer application, and provides improved NH_3 emission with higher spatial and temporal resolution.

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1 Introduction

Ammonia (NH_3) is the most important and abundant alkaline constituent in the atmosphere, which has a wide range of impacts. First, it plays a key role in atmospheric chemistry and ambient particle formation. NH_3 partitions to the sulfate (SO_4^{2-}) aerosol and nitric acid (HNO_3) to generate secondary non-organic aerosol (SNA), including sulfate, nitrate and ammonium. Field measurements indicate that SNA is a major contributing factor during haze days (He et al., 2014; Wang et al., 2012; K. Huang et al., 2012). Ye et al. (2011) observed a strong correlation between peak levels of fine particles and large increases in NH_3 concentrations. High aerosol concentrations have a significant effect on visibility range, climate forcing, and human health (Cheng et al., 2013; Ding et al., 2013; Pope et al., 2011). In addition, the deposition of ammonium particles (NH_4^+) and gaseous ammonia (NH_3) can cause soil acidification, water eutrophication, loss of biodiversity, and the perturbation of ecosystems (Lepori et al., 2012; Stevens et al., 2004; Zhu et al., 2013). China is the largest or among the largest producers of crops and meat agricultural products in the world (FAO 2013), which leads to a large amount of NH_3 emission. Previous studies indicated that the China's ammonia emissions contributed 23 % of the global NH_3 budget (EDGARv4.1, http://edgar.jrc.ec.europa.eu/datasets_list.php?v=41) and presents a continuously increasing trend (Dong et al., 2010).

Nitrogen fertilizer use is one of the largest sources of NH_3 emissions in China, accounting for 35–55 % of the national total (X. Huang et al., 2012; Zhao et al., 2013). There are many studies focusing on NH_3 emission from agricultural fertilizer in China, but they are mostly based on traditional "emission factor" (EF) methods. Some of them (Klimont, 2001; Streets et al., 2003; Dong et al., 2010; Zhao et al., 2013) use uniform simple United States or European-based emission factors (EF) for the whole of China. However, ammonia volatilization from nitrogen fertilizer application depends strongly on environmental parameters, such as ambient temperature and soil acidity (Roelle et al., 2002; Corstanje et al., 2008). In addition, fertilizer application dates and applica-

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tion amounts vary by geographical regions and crop types. Therefore, these estimates are subject to high uncertainty, especially in their temporal and spatial distributions. Zhang et al. (2011) and X. Huang et al. (2012) use some relative correction factors to introduce the impacts of temperature, soil properties and fertilization method, which somewhat reduce the temporal and spatial uncertainty. One shortcoming of this approach is that those correction factors are empirical and too simple. In recent years, some scientists from outside of China have begun to focus on estimating NH_3 emissions based on a bidirectional surface flux model (Cooter et al., 2010; Wichink Kruit et al., 2012). For example, a group at the U.S. Environmental Protection Agency (U.S. EPA) (Cooter et al., 2012; Bash et al., 2013; Pleim et al., 2013) modified the Community Multi-Scale Air Quality (CMAQ) model to include a bidirectional NH_3 exchange module, which was coupled to the Fertilizer Emission Scenario Tool for CMAQ (FEST-C) system (Ran et al., 2011; CMAS, 2014) containing the Environmental Policy Integrated Climate (EPIC) model (William et al., 2008). It was used to simulate the bidirectional exchange of NH_3 in the United States, which includes the influences of meteorology, air–surface exchange, and human agricultural activity. Compared with a traditional emission inventory, the model performances for NO_3^- concentration and NH_3 deposition were improved in the United States (Bash et al., 2013). However, until now this method has not yet been used to estimate the agricultural fertilizer NH_3 emission in China.

For the first time in this study, we estimate China's NH_3 emission from agricultural fertilizer use in 2011 based on the CMAQ model with the bidirectional NH_3 exchange module coupled to the FEST-C system with an agro-ecosystem model, EPIC. The structure of this modeling system and input data processing are described in detail in the next section. The results of the simulated fertilizer use and NH_3 emission, and comparison with other studies are discussed in Sect. 3. The results of CMAQ modeling are also discussed and compared with field measurements.

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2 Methodology and inputs

2.1 General description of the modeling system

Figure 1 shows the structure of the modeling system, which contains three main components (1) the FEST-C system containing the EPIC model, (2) the meso-scale meteorology Weather Research and Forecasting (WRF) model and (3) the CMAQ air quality model with bi-directional ammonia fluxes. A detailed description of CMAQ version 5.1 with bi-directional ammonia flux estimates of NH_3 emission from agriculture fertilizer use and bi-directional module can be found in Bash et al. (2013). The soil NH_4^+ content and agriculture activity data, such as fertilizer application rate, timing and depth of application are simulated by the EPIC model in the FEST-C system. In order to run the EPIC model, local Chinese information such as crop distribution, soil characteristics, climate patterns, fertilizer use characteristics is collected and processed. The details regarding these data sources and the processing methods are described in Sect. 2.2. In addition to the agriculture activity and soil information, this system also considers the influence of WRF simulated weather on NH_3 emission. The tools in the FEST-C system can be used to process the EPIC input data and also extract the EPIC daily output that is required for CMAQ (CMAS, 2014).

The CMAQ simulation domain, shown in Fig. 2, is based on a Lambert projection with the two true latitudes of 25° N and 40° N and covers most of East Asia with a grid resolution of 36 km × 36 km. The EPIC data and micrometeorological parameters are estimated for each modeled 36 km CMAQ grid cell.

2.2 The EPIC modeling in the FEST-C system

The EPIC model is a semi-empirical agro-ecosystem model which is designed to simulate agriculture fields that are characterized by soil, landscape, weather, crop management (William et al., 2008). A wide range of vegetative systems, tillage systems, and other crop management practices can be simulated in this model (Gassman et al.,

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2005). Additionally, soil nitrogen (N), carbon (C) and phosphorus (P) biogeochemical process models were incorporated into EPIC. Therefore, it's well-suited for simulation of fertilizer management and soil nitrogen content in agricultural systems. The input information required by EPIC includes crop site information, soil characteristics, weather and crop management, which will be described in detail in the next. All the data were processed to a 36 km × 36 km grid for integration with the air quality model, CMAQ.

2.2.1 Crops

Fourteen crop types were modeled in this study, including early rice, middle rice, late rice, winter wheat, spring wheat, corn, sorghum, barley, soybean, potato, peanuts, canola, cotton and "other crops" which is assumed to represent all remaining crops. There are 2710 counties in China. Data of Cropland area for each crop in each county was collected and processed based on each county-level statistical yearbook. The Moderate Resolution Imaging Spectroradiometer (MODIS; https://lpdaac.usgs.gov/products/modis_products_table/mcd12q1) was used to provide finer level land use information. The MODIS land use product provides annual 500 m pixel-scale information for 20 land use categories. MODIS classes 12 (Cropland) and 14 (Cropland/Natural Vegetation Mosaic) are of particular interest for this study. In addition, irrigation is an important factor for crop growth and soil characteristics. Here, we use the global irrigated area map (GIAM) at 1 km resolution (Thenkabail et al., 2008) to divide each crop into irrigated and non-irrigated classes. The BELD4 tool in the FEST-C system was used to process these data into 36 km grid cell (CMAS, 2014).

2.2.2 Soil information

The dominant soil type in each grid was taken from the Harmonized World Soil Database (HWSD, http://webarchive.iiasa.ac.at/Research/LUC/External-World-soil-database/HTML/), which gives the soil distribution with 30 arc-second resolution in China. Soil variables such as layer depth, soil texture, soil carbon

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content, carbonate content, bulk density, cation exchange capacity and pH for each soil type were obtained from China Soil Scientific Database (http://www.soil.csdb.cn/), the soil dataset in HWSD and the US soil profile data (Cooter et al., 2012; http://www.nrcs.usda.gov/wps/portal/nrcs/main/national/technical/nra/nri/). EPIC was initialized using this soil data and then was run for a 25 yr spin-up period to allow soil characteristics and nutrient pools to adjust to the long-term management environment.

2.2.3 Weather

The weather parameters required by EPIC include maximum and minimum temperature, radiation, precipitation, relative humidity and 10 m wind speed. For the spin-up run, these variables were extracted from NASA Modern Era Reanalysis for Research and Applications (MERRA; http://disc.sci.gsfc.nasa.gov/mdisc/overview/index.shtml) data, which provided the weather information from 1979 to the present with $0.5^\circ \times 0.667^\circ$ grid resolution. The climatological characteristics of the closest grid-cell in MERRA to each EPIC model grid-cell were selected as the weather input for the EPIC spin-up simulation run in each grid. For the year-specific EPIC run, the output of the Weather Research Forecast Model (WRF) was processed to generate the gridded weather conditions on the CMAQ 36 km × 36 km grid using the *WRF/CMAQ to EPIC* tool in the FEST-C system (CMAS, 2014).

2.2.4 Crop management

In the EPIC model, the timing of crop management, e.g., tilling, planting, irrigating, fertilization and harvesting, can be prescribed or scheduled based on a heat-unit (HU) method, as described in Cooter et al., (2012). In this study, a combination of prescribed and heat-unit scheduled timing was used. The HU scheduled timing allowed for adaptation to inter-annual and interregional temperature variability and more realistically represents a farmer's dynamic decision-making. At the same time, the planting and harvesting timing were also limited to a fixed time range based on the information from

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the Chinese planting information network (http://www.zzys.moa.gov.cn/) and the unpublished research about crop management from the Chinese Academy of Agriculture Sciences, which allowed the timing to be more suitable for Chinese agriculture.

Nitrogen fertilizer application information is necessary to accurate estimate NH_3 emission in this study. The application rate for specific fertilizer type, crop and province was extracted from Chinese statistical materials (National Bureau of Statistics of China (NBSC), 2012c). The fertilizer types include urea, ammonium bicarbonate(ABC), diammonium phosphate (DAP), N-P-K compound fertilizer (NPK) and others (e.g. ammonium nitrate, ammonium sulfate). Table 1 shows the national-average application rate for some major crops. We can see that the nitrogen fertilizer application rates for different crops are varied. The largest nitrogen amount is required by cotton and wheat, which were 228.11 and 196.22 $kg\,N\,ha^{-1}$, respectively. However, the nitrogen-fixing crops (e.g. soybean and peanuts) require much less nitrogen input. Among all the fertilizer types, the use of urea and ammonium bicarbonate were dominant.

Besides application rate, the ratio of basal and topdressing fertilizer is also important for ammonia volatilization. Figure 3 presents the Chinese agriculture regions used to characterize these management practices. Each region is a geographic area where crop management practices are assumed to be similar. Based on the results of some previous field investigations (Wang et al., 2008; Zhang, 2008), the ratios of basal and topdressing for different crops in each agriculture region were identified. Table 2 shows the results for three major crops in China and the divergence can be seen. For example, the ratio for wheat in the middle and lower Yangtze river region is 1.39, but that for wheat in the southwest region is only 0.33. In general, the ratio for corn is the highest among these three major crops. That is, much more fertilizer is applied to corn just prior to or at planting than is applied later in the growing season. The information in Tables 1 and 2 is combined to determine the amount of fertilizer applied to each crop in each grid cell during basal and topdressing activities.

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2.3 The bi-directional CMAQ model system

Bash et al. (2013) implemented the bi-directional ammonia flux module in CMAQv5.1 based on the two-layer (soil and vegetation canopy) resistance model described by Pleim et al. (2013) which is similar to the model presented by Nemitzet al. (2001). The soil inorganic nitrogen budget in CMAQ was parameterized following the method in EPIC (Williams et al., 2008). The EPIC results regarding soil condition, fertilizer application and fertilizer amount are input to the bi-directional CMAQ model. Other input data was the same as in the traditional CMAQ model. The WRF (Weather Research & Forecasting Model) version 3.5.1 (http://www.wrf-model.org) was used to generate the meteorological input. The configuration options used in WRF and CMAQ are same as described by Fu et al. (2014).

In order to evaluate the performance of this NH_3 emission, fate and transport model, two simulations were conducted in this study, including Base-case and Bidi-case. For Base-case, the emission inventory from Zhao et al. (2013) was used. For Bidi-case, the NH_3 emission from fertilizer use was calculated online by CMAQ while other emissions were from Zhao et al. (2013).

3 Results and discussion

3.1 Nitrogen fertilizer application

The nitrogen fertilizer application is a key aspect of EPIC for use in this system. This is explored by comparison of the EPIC simulation results to statistical data. The N use in each grid cell per day was calculated by the following formula:

$$\mathrm{USE}_i = \sum_{j=1}^{\mathrm{crop}} \left(N_{ij} \times f_{ij}\right) \times 129\,600 \tag{1}$$

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where USE_i (kg) is the N application in grid cell i; N_{ij} (kg ha^{-1}) is the N application rate in the grid cell i for crop j; f_{ij} is the fraction of the crop j in grid cell i; and 129 600 ha grid^{-1} is a conversion factor accounting for the area of the grid cell.

Figure 4a and b show the patterns of annual fertilizer use at province level between the statistical data of NBSC (2012a) and the EPIC output. We can see that the EPIC results well capture the general pattern, especially for the largest fertilizer use provinces (> 1750 million kg), such as Henan, Shandong, Jiangsu and Hebei provinces, where the biases are −9.7, −5.1, −1 and −0.6 %, respectively. At the same time, relatively large biases also exist for some provinces, such as Hunan (−20.6 %) and Heilongjiang (19.2 %). This may be due to the uncertainty of the statistical data or that the 36 km grid is relatively coarse and uncertainty exists for gridded crop area calculation based on the county-level crop statistical data and MODIS crop data. Because the provinces with larger bias apply relatively small amount of fertilizer, these modeled biases are not expected to lead to large biases in the simulations.

Figure 5 shows the comparison of the fraction of N fertilizer use by each month between statistics and EPIC output. The statistical data was derived from the field investigation of Zhang et al. (2008) for 2004. It can be seen that the model results well capture the temporal characteristics. The fertilizer amounts used from March to July, and October are dominant, which are closely related to the fertilizer timing of crops in China. For example, the North China Plain is the most important agriculture production region, where the winter wheat-summer corn rotation is the major crop planting system. Winter wheat is usually planted in October with the application of basal fertilizer, and the topdressing fertilizer is used in March and April of the next year. For summer corn, the timing for basal fertilizer is usually in June and that for topdressing fertilizer is in July. In another major agriculture production region, the Northeast Plain, rice is the dominant crop. Due to the temperature limitation, rice here is usually seeded in April and May and the topdressing fertilizer is applied in June and July.

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3.2 NH_3 emission

3.2.1 Spatial and temporal distribution

The NH_3 emission from N fertilizer application in 2011 estimated in this study is approximately 3.0 Tg. The spatial distribution of annual NH_3 emission in 36 km grid is presented in Fig. 6. It can be seen that the NH_3 volatilization was concentrated in Henan, Shandong, Hebei, Jiangsu and Anhui provinces, accounting for 11.1, 9.9, 8.8, 6.7 and 7.1 %, respectively. The highest NH_3 emission intensity in this region is above 5000 kg grid^{-1} cell. The crop production here is the most intense in the whole of China and the total crop area in these five provinces accounts for about 31.4 % China's total. These five provinces consumed approximately 37.3 % of the nitrogen fertilizer for the whole country in 2011 (NBSC, 2012c). Besides the large crop production, the high emissions are also due to the high fertilizer application rate. For example, the rate of N fertilizer use for rice in Jiangsu province is above 300 kg ha^{-1}, which is much higher (2 times) than the national average. The smaller contributors of NH_3 emission are located mostly in western China, like Tibet, Qinghai and Gansu, where the amount of arable land and N fertilizer use is small.

Figure 7 shows the monthly distribution of ammonia emissions. It can be seen that the emissions are dominant from March to July, and October, accounting for 88.7 % of the annual total, which agrees with the pattern of N fertilizer consumption described in Sect. 3.1. Besides N fertilizer use, the weather parameters, like temperature and precipitation, also affect the temporal and spatial distribution of emissions. For example, the emission in March is much smaller than April and May due to lower temperature, even though the amount of consumed fertilizer is nearly equivalent. Similarly, the emission in June is a little smaller than April and May. A possible reason is that precipitation in June is much larger than that in the earlier two months. Based on the statistical data of the major cities in China (NBSC, 2012b), the total precipitation in June of 2011 was 170.9 mm, while in April and May, it was 28.6 and 71.9 mm, respectively. Figure 8

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presents the spatial distribution for each month. Some differences for the months with large emissions still can be seen. For example, in North China Plain, like Hebei, Henan and Shandong, NH_3 emissions are relatively small in May for little fertilizer application in this month. In Northeast China, including Liaoning, Jilin and Heilongjiang, the NH_3 emissions in May, June and July are dominant. In November, major NH_3 emissions occur in Jiangsu, Hubei and Anhui, where the basal fertilizer for winter canola is applied in this month.

3.2.2 Comparison with other studies

The ammonia emissions from N fertilizer use in China have been estimated for different base years by different methods. The results of comparisons between this study and some previous studies relisted in Table 3. As presented, the results of this study are generally equivalent to the research of X. Huang et al. (2012), which is lower by 30–70 % compared to the other studies. The discrepancies are mostly caused by the various estimating methods and emission factors employed. Streets et al. (2003), Dong et al. (2010) and Zhao et al. (2013) used uniform, simple emission factors for all agriculture in China. These values were extracted from European or US experiments rather than Chinese local measurements. At the same time, they also did not consider the impacts of some environmental parameters, e.g. temperature, soil pH, etc. Zhang et al. (2011) used the NARSES (National Ammonia Reduction Strategy Evaluation System) model developed for the UK to estimate the emission. Similarly, the parameters in the model were set based on conditions in UK, which may be very different from conditions in China. X. Huang et al. (2012) characterized the emission factors at 1 km grid level by introducing the impacts of fertilizer types, fertilizing methods, soil properties and temperature based on the Chinese measurements and investigations, which introduces more local variability. However, it uses empirical correction factors coefficients that do not capture the processes modeled, including impacts of precipitation, and may introduce uncertainty in applications other than where they were developed.

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Figure 9 shows the comparisons of province ammonia emissions from N fertilizer among different studies. Similar with these previous studies, the emissions from Henan, Shandong, Jiangsu, Hebei and Anhui dominate the country annual total emissions. At the same time, some discrepancy also exists for the specific province between different studies, which may be caused by distinct fertilizer consumptions and emission rate employed. For example, for Henan, the estimation of X. Huang et al. (2012) is the highest among these studies. The possible reason is that alkaline soil is dominant in Henan and X. Huang et al. (2012) set a uniform high emission factor for alkaline soil, which is twice as high as that in Dong et al. (2010). In addition, comparing with the results in Dong et al. (2010) and Zhao et al. (2013), the emission fractions in Guangdong and Guangxi are lower in this study. This difference could be attributed to the acidic soil and large amount of precipitation in this region which were not considered in the previous studies.

3.3 Evaluation of the CMAQ results by ground observations

As the dominant alkaline gas in the atmosphere, NH_3 preferentially partitions to SO_4^{2-} and then partitions to NO_3^-. In NH_3-rich regions, the NO_3^- concentration is sensitive to NH_3 changes, but NH_3 changes don't lead to large differences in SO_4^{2-} concentration (Wang et al., 2011). In order to evaluate the reliability of this NH_3 emission estimation, we compared the CMAQ modeled NO_3^- concentration using different NH_3 emission with observations. In China, the observation data for chemical components of fine particulates was very spare and not publicly available. Here, we collected the observation data at three monitoring sites, including Shanghai station (121.5° E, 31.2° N), Suzhou station (120.6° E, 31.3° N) and Nanjing station (118.7E, 32.1N). Ion chromatography (Dionex-3000, Dionex Corp, CA, US) was used to measure the daily NO_3^- concentration in $PM_{2.5}$ particles (Cheng et al., 2014). Some statistical indices including mean observation (Mean Obs.), mean prediction (Mean Pred.), bias, normalized mean bias (NMB), normalized mean error (NME) and correlation coefficient (R) were calculated

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for Base-case and Bidi-case in June, August and November, as shown in Table 4. For Base-case, the emission inventory from Zhao et al. (2013) was used. For Bidi-case, the NH_3 emission from fertilizer use was calculated online by CMAQ, while other emissions were also from Zhao et al. (2013). It can be seen that the model performance of the Bidi-case was comparable or better in general compared with the Base-case. For August and November, the NMB and the NME were improved by 3.29–66.85 and 0.22–46.32 %, respectively. The correlation coefficients for the Bidi-case were also comparable or better than the base case. For June, even though the bias of the Bidi-case was a little larger, some other statistical indices were acceptable. For example, the NME decreased from 57.3 to 45.1 % and the correlation coefficient increased from 0.83 to 0.91 at Shanghai station. The correlation coefficient at Suzhou station and the NME at Nanjing station were comparable for these two cases.

3.4 Uncertainty analysis

Uncertainties still exist for this method at some aspects. On one hand, calculating results, including regional and temporal distribution of NH_3 emissions, are strongly related to the quality of the input data. For example, it's hard to count fertilizer application for some scattered farmers, so uncertainties may exist for the statistical data of fertilizer application rate for different crops. Besides, it is difficult to capture entirely the exact date of fertilizing, which may bring uncertainties to the temporal distribution of NH_3 emissions. In addition, the bi-directional ammonia flux module in the CMAQ is parameterized based on the field measurements in the United States, which might be not fully applicable to China. In order to lower the uncertainties, more local field measurements of agriculture NH_3 emissions should be conducted in the future.

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4 Conclusions

In this study, for the first time, the NH_3 emissions of 2011 from the N fertilizer use in China were estimated using the bi-directional CMAQ model rather than the traditional "emission factor" method. The hourly NH_3 emission can be calculated online with CMAQ running. Compared with previous researches, this method considers more influencing factors, such as meteorological fields, soil and the fertilizer application, and provides improved spatial and temporal resolution.

The China's NH_3 emission from N fertilizer application was about 3.0 Tg in 2011. The major contributors were Henan, Shandong, Hebei, Jiangsu and Anhui, accounting for 11.1, 9.9, 8.8, 6.7 and 7.1 % of total emissions, respectively. The monthly distribution of ammonia emissions was in line with the pattern of N fertilizer consumption, which were dominant from March to July, and October, accounting for 88.7 % of the whole year. Compared with the traditional "emission factor" method, this coupled modeling system improved the simulation of ambient nitrate aerosol concentrations.

Although uncertainties still exist in the NH_3 emission estimation due to the uncertainties of model parameterization and various input data, the CMAQ-EPIC modeling system allows for some interesting future research. This system is a combination of air quality and agro-ecosystem models and couples the processes and impacts that human activity has on food production with air-quality. The model could be applied at finer grid resolutions for China in order to more accurately capture spatial gradients in NH_3 emissions and resulting impacts on air quality. Secondly, this system reflects the impacts of weather and climate on the NH_3 emission. Therefore, it can be coupled with climate model to explore the interaction of climate change and NH_3 emission. If linking it to a water quality and transport model, the impacts of atmospheric nitrogen deposition from CMAQ and nutrient run off from EPIC on the water eutrophication can be estimated.

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Table 1. The national-average fertilizer application rate for major crops in China, 2011 ($kg\,N\,ha^{-1}$).

	Total	Urea	ABC[a]	DAP[b]	NPK[c]	Others
Early rice	183.48	125.03	20.03	4.00	21.87	12.55
Middle rice	185.62	117.38	33.15	4.04	18.69	12.36
Late rice	181.14	124.20	19.13	4.02	21.63	12.17
Wheat	196.22	123.90	19.05	16.14	29.98	7.16
Corn	186.75	123.45	19.05	12.63	18.85	12.77
Soybean	45.92	19.50	1.65	10.48	11.51	2.77
Peanuts	95.14	36.30	11.70	3.43	29.03	14.68
Canola	128.14	75.90	30.90	2.35	11.02	7.97
Cotton	228.11	152.40	9.45	24.34	27.45	14.46

[a] Ammonium bicarbonate (ABC); [b] diammonium phosphate (DAP); [c] N-P-K compound fertilizer (NPK).

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Table 2. Ratio of basal and topdressing fertilizer for major crops in each agriculture regions.

Region	Wheat		Corn		Rice	
	basal	topdressing	basal	topdressing	basal	topdressing
The Northeast Region	1.00	0.80	1.00	1.23	1.00	0.88
The Gan-Xin Region	1.00	0.44	1.00	3.50	1.00	1.00
The Southern China Region	1.00	1.00	1.00	2.98	1.00	2.91
The Huang-Huai-Hai Region	1.00	0.80	1.00	2.07	1.00	1.29
The Loess Plateau Region	1.00	0.44	1.00	3.50	1.00	1.00
The Inner Mongolia and along the Great Wall Region	1.00	0.44	1.00	3.50	1.00	1.00
The Tibetan Plateau Region	1.00	0.44	1.00	3.50	1.00	1.00
The Southwest Region	1.00	0.33	1.00	2.33	1.00	1.88
The middle and lower Yangtze River Region	1.00	1.39	1.00	1.66	1.00	1.29

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Table 3. Comparison of the NH_3 Emissions from fertilizer use in our study with other published results.

Reference	Year	NH_3 Emission ($Tg\,yr^{-1}$)
Streets et al. (2003)	2000	6.7
Zhang et al. (2011)	2005	4.3
Huang et al. (2012b)	2006	3.2
Dong et al. (2010)	2006	8.7
Zhao et al. (2013)	2010	9.8
This study	2011	3.0

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Table 4. The performance statistics of CMAQ modeled daily NO_3^- concentration for Base-case and Bidi-case compared to the observations at three monitoring stations.

			Shanghai station	Suzhou station	Nanjing station
June (1–30 Jun 2011)		Mean Obs. ($\mu g m^{-3}$)	7.27	13.43	12.81
	Base-case	Mean Pred. ($\mu g m^{-3}$)	8.41	9.32	13.44
		Bias ($\mu g m^{-3}$)	1.14	−4.10	0.63
		NMB (%)	15.65	−30.56	4.90
		NME (%)	57.34	40.71	59.93
		R	0.83	0.81	0.24
	Bidi-case	Mean Pred. ($\mu g m^{-3}$)	8.60	7.16	7.59
		Bias ($\mu g m^{-3}$)	1.32	−6.26	−5.23
		NMB (%)	18.21	−46.63	−40.81
		NME (%)	45.07	50.63	60.40
		R	0.91	0.83	0.14
August (20 Jul–20 Aug 2011)		Mean Obs. ($\mu g m^{-3}$)	2.99	7.04	6.24
	Base-case	Mean Pred. ($\mu g m^{-3}$)	6.42	14.51	12.02
		Bias ($\mu g m^{-3}$)	3.43	7.46	5.78
		NMB (%)	114.84	105.95	92.68
		NME (%)	142.48	115.89	97.18
		R	0.62	0.28	0.87
	Bidi-case	Mean Pred. ($\mu g m^{-3}$)	4.42	10.36	8.85
		Bias ($\mu g m^{-3}$)	1.43	3.31	2.62
		NMB (%)	47.99	47.01	41.92
		NME (%)	96.16	79.43	62.64
		R	0.64	0.24	0.90
November (1–30 Nov 2011)		Mean Obs. ($\mu g m^{-3}$)	9.42	11.59	14.57
	Base-case	Mean Pred. ($\mu g m^{-3}$)	12.59	16.72	22.62
		Bias ($\mu g m^{-3}$)	3.17	5.14	8.05
		NMB (%)	33.68	44.32	55.24
		NME (%)	83.85	53.68	74.81
		R	0.71	0.72	0.68
	Bidi-case	Mean Pred. ($\mu g m^{-3}$)	12.28	12.41	12.88
		Bias ($\mu g m^{-3}$)	2.86	0.82	−1.68
		NMB (%)	30.39	7.05	−11.56
		NME (%)	65.33	53.46	43.35
		R	0.78	0.72	0.79

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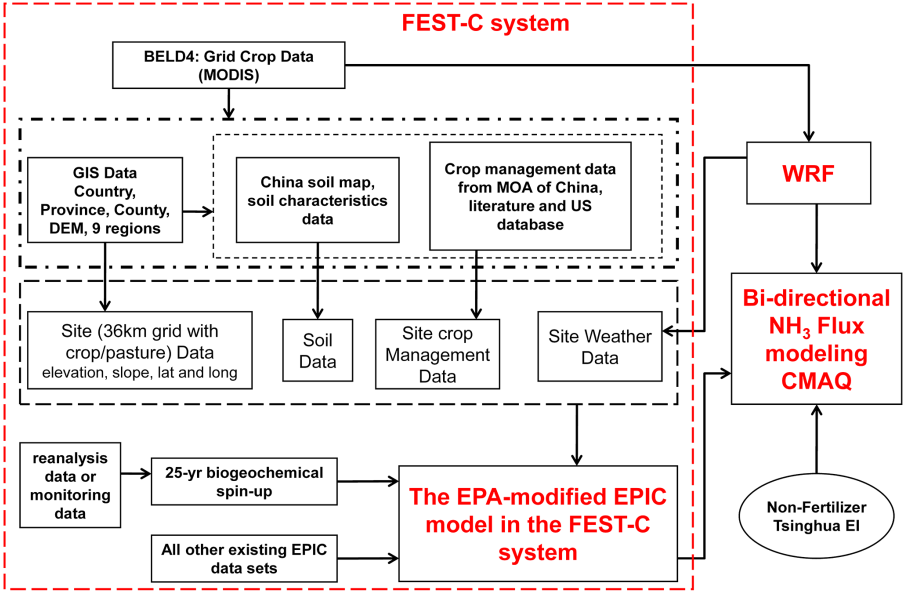

Figure 1. The modeling system of the agricultural fertilizer NH_3 emission for China.

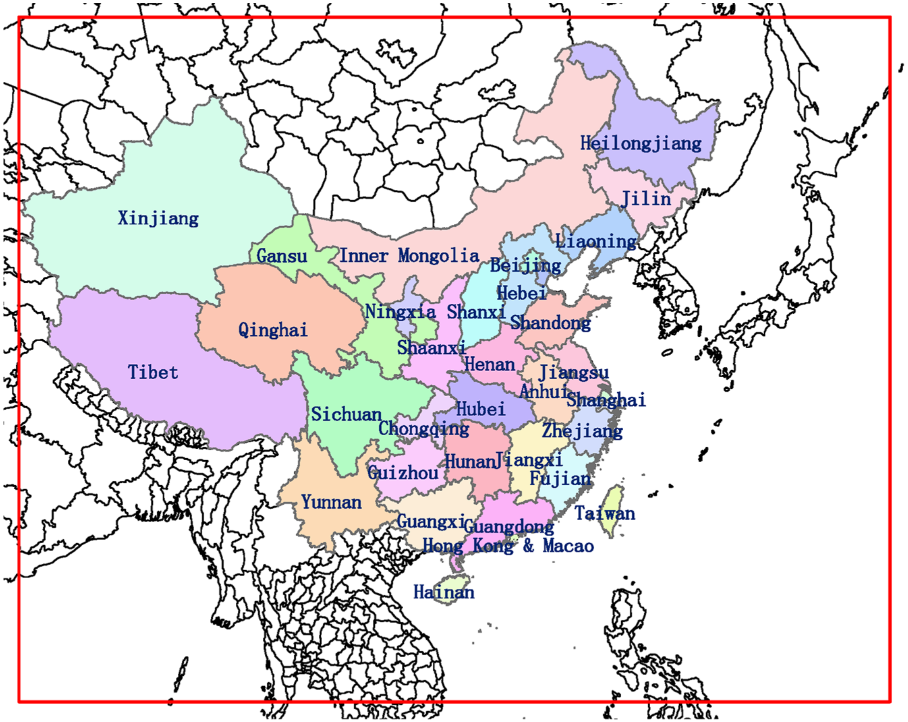

Figure 2. The modeling domain.

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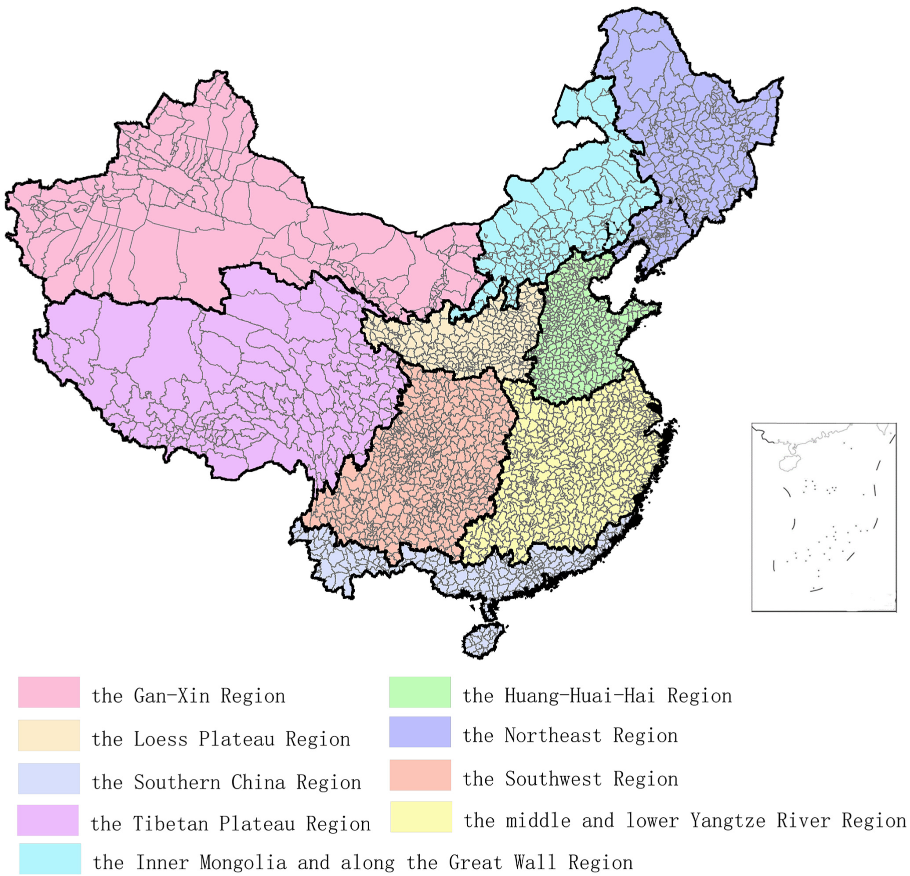

Figure 3. The nine agriculture regions in China.

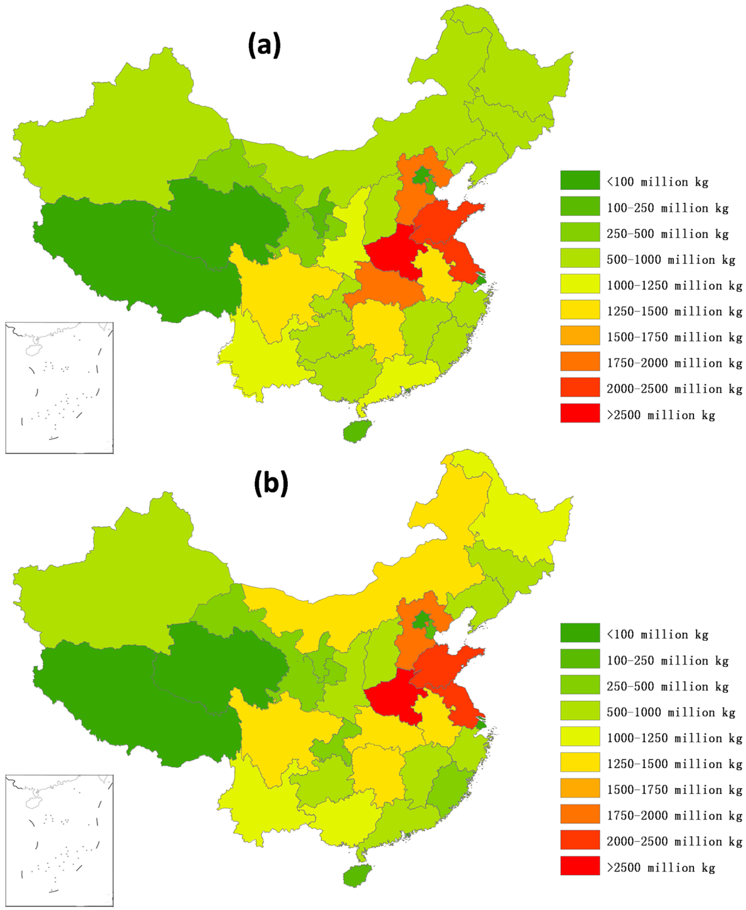

Figure 4. Comparison of annual N fertilizer use at province level from the statistic data **(a)** and the EPIC output **(b)**.

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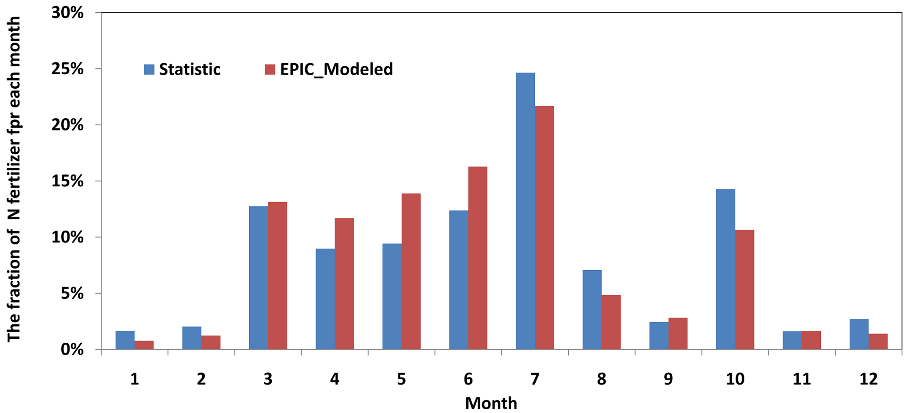

Figure 5. Comparison for the fraction of N fertilizer use by month from statistic and EPIC output.

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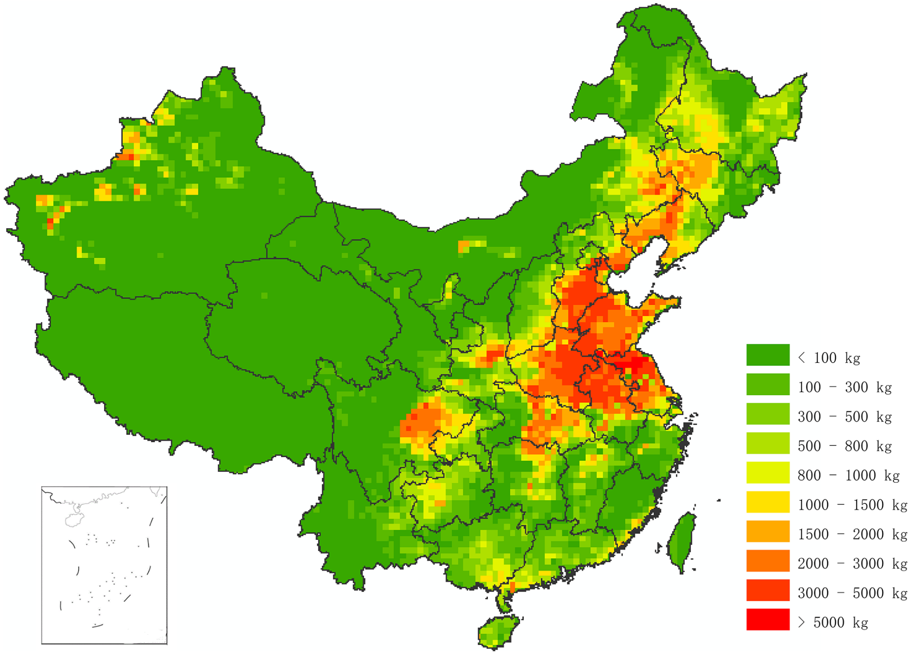

Figure 6. Spatial distribution of NH_3 emissions from N fertilizer use in 36 km grid cell ($kg\,yr^{-1}$).

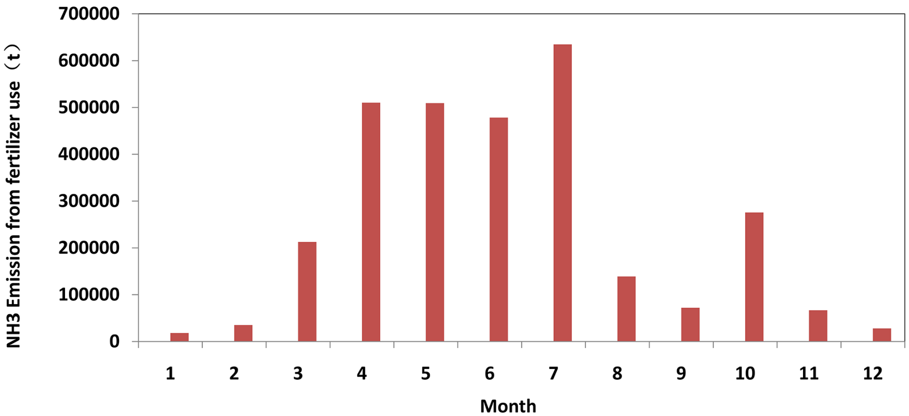

Figure 7. Monthly NH_3 emissions from N fertilizer use.

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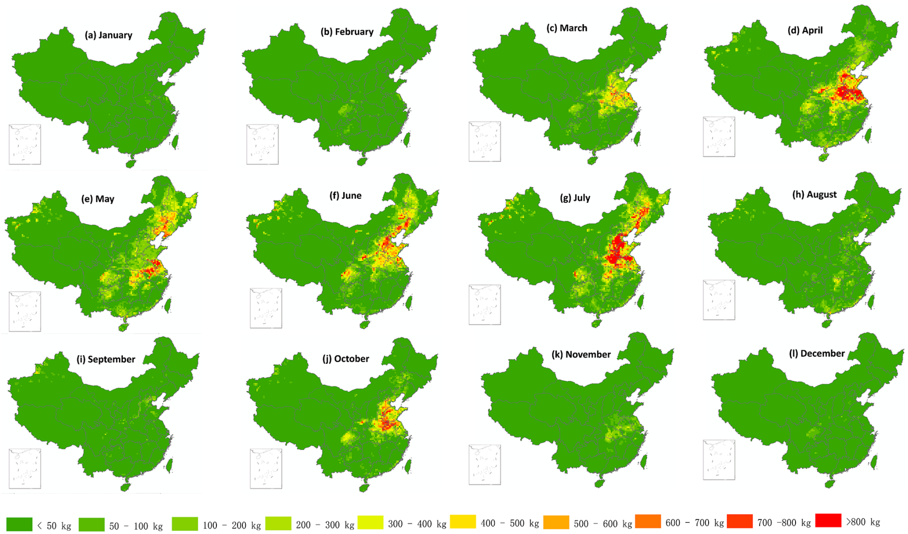

Figure 8. Spatial distribution of NH_3 emissions from N fertilizer use for each month ($kg\,yr^{-1}$).

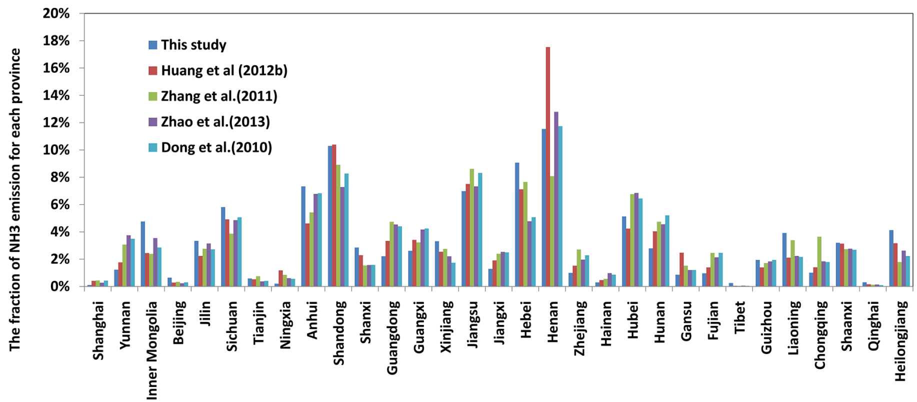

Figure 9. Comparison of provincial-level results on the NH_3 emissions from N fertilizer use.

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