Geophysical Research Letters

RESEARCH LETTER
10.1029/2019GL086551

Key Points:
- We provide new high-resolution estimates of reactive gaseous nitrogen (Ngr) emissions from China’s croplands and their seasonal dynamics
- Across different prefectures, Ngr emission per capita follows an environmental Kuznets curve against urbanization rate
- The overall warming effect of N2O emissions dominate over local cooling effects ascribed to NH3 and NOx emissions each year

Supporting Information:
- Supporting Information S1

Correspondence to:
Y. Zheng, zhengy@sustech.edu.cn

Spatial Variation of Reactive Nitrogen Emissions From China’s Croplands Codetermined by Regional Urbanization and Its Feedback to Global Climate Change

Peng Xu1,2, Anping Chen3, Benjamin Z. Houlton4, Zhzhong Zeng1, Song Wei1,2, Chenxu Zhao5, Haiyan Lu1,2, Yajun Liao5, Zhonghua Zheng6, Shengji Luan5, and Yi Zheng1,7,8

1School of Environmental Science and Engineering, Southern University of Science and Technology, Shenzhen, China, 2School of Water Resources and Hydropower Engineering, Wuhan University, Wuhan, China, 3Department of Biology and Graduate Degree Program in Ecology, Colorado State University, Fort Collins, CO, USA, 4John Muir Institute of the Environment and Department of Land, Air and Water Resources, University of California, Davis, CA, USA, 5School of Environment and Energy, Peking University, Shenzhen, China, 6Department of Civil and Environmental Engineering, University of Illinois at Urbana-Champaign, Urbana, IL, USA, 7State Environmental Protection Key Laboratory of Integrated Surface Water-Groundwater Pollution Control, School of Environmental Science and Engineering, Southern University of Science and Technology, Shenzhen, China, 8Shenzhen Municipal Engineering Lab of Environmental IoT Technologies, Southern University of Science and Technology, Shenzhen, China

Abstract Reactive gaseous nitrogen (Ngr) emissions significantly affect Earth’s climate system. Disagreement exists, however, over Ngr contributions to short- versus long-term climate forcing, from local to global scales and among different gaseous forms, including NH3, NOx, and N2O. Here, we provide a comprehensive inventory of Ngr from China’s croplands based on a new bottom-up, mass flow-based approach integrated with fine-resolution agricultural activity data and nitrogen emission factors. We demonstrate that China’s croplands emit about 8.87 Tg N to the atmosphere in 2014. Across different prefectures, Ngr emission per capita conforms to a “Kuznets curve,” that is, first increases then decreases, along the gradient of increasing urbanization. Ngr emission per gross domestic productivity (GDP) decreases exponentially with increasing urbanization or per capita GDP. Furthermore, climate change impact analyses suggest that the global-scale warming effect of China’s cropland N2O emissions dominate over local cooling effects ascribed to its NH3 and NOx emissions.

Plain Language Summary Reactive gaseous nitrogen, sourced to the world’s highest fertilizer application rates in China, impacts local cooling effects on short time scales. The effects are ultimately counterbalanced by nitrous oxide emissions over the long term, leading to substantial warming effects on the global climate system. We base our conclusion on the first-ever high-resolution assessment of reactive nitrogen emissions to the atmosphere from China’s croplands. Owing to limited high-resolution reactive gaseous nitrogen emissions data, hitherto, large uncertainties have existed in the detailing the role of China’s cropland reactive gaseous nitrogen emissions on climate forcing. This paucity of information poses a challenge for informing China’s agricultural sustainability policies with regard to maintaining/increasing agricultural yields while reduces reactive gaseous nitrogen emissions and their warming potential. Our analyses provide essential information for further sustainable N management and the interactions between agriculture and climate change. We provide policy-relevant information for mitigation targets, including the 2°C limit goal set by the 2015 Paris Agreement of the United Nations Framework Convention on Climate Change, enabling progress related to multiple United Nations Sustainable Development Goals.

1. Introduction

Agriculture is critical to mitigating rising levels of greenhouse gas (GHG) pollution, given that ~25% of global GHG emissions are attributed to agriculture and land use change. A major facet of agriculture’s impact on climate change occurs via the use of nitrogen (N) fertilizers, which have more than doubled in use throughout the twentieth century (FAO, 2018; Frank et al., 2019). Given that on average ~50% of the N applied to croplands does not enter agricultural products (Houlton et al., 2019), worldwide emissions of agricultural...
nitrogen have grown substantially, altering both local and global radiative forcing (Pinder et al., 2012). For example, total reactive nitrogen emissions to the atmosphere (Ngr, including ammonia (NH₃), nitrogen oxides (NOₓ), and nitrous oxide (N₂O)) from global croplands rose from 14 to 35 Tg N·yr⁻¹ between 1970 and 2012 or ~31% of the global anthropogenic Ngr emissions today. This nitrogen flux could double by 2050 to keep pace with the ~50% increase in food demand needed to support ~9–10 billion people (Bodirsky et al., 2014).

Ngr emissions have long-lived warming effects from the production of N₂O and tropospheric O₃, as well as short-lived cooling effects via changing tropospheric aerosol, ozone (O₃) and methane (CH₄) (Galloway et al., 2008). Hence, accurate accounting of Ngr inventories is important for the calculation of global radiative forcing. Such information is widely used by policy makers for setting GHG mitigation targets including the 2°C limit goal set by the United Nations Framework Convention on Climate Change (UNFCCC) Paris Agreement of December 2015 (Bowles et al., 2018) and the climate goals established by the United Nations Sustainable Development Goals (SDGs) (Haines et al., 2017). However, large uncertainties exist in estimates of global cropland nitrogen emissions, perhaps as large as 47% across studies (Fowler et al., 2013; Galloway et al., 2008). Because of the different climate effects of N₂O (warming) and NH₃ and NOₓ (cooling), it is also critical to separate the components of Ngr, which are often ignored and remain uncertain (Fesenfeld et al., 2018; Liu et al., 2019).

China is the world’s largest producer of croplands Ngr emissions. Yet current estimates of China’s croplands Ngr emissions are highly uncertain, with NH₃ emissions ranging from 2.5 to 9.0 Tg N·yr⁻¹, N₂O emissions ranging from 0.40 to 0.64 Tg N·yr⁻¹, and NOₓ ranging from 0.07 to 0.40 Tg N·yr⁻¹ across studies (supporting information Table S1) (Chen et al., 2016; Cui et al., 2013; EDGAR v4.3.2, 2017; Fu et al., 2015; Gu et al., 2015; H. Wu, Wang, et al., 2018; Kang et al., 2016; Lin et al., 2019; Luo et al., 2018; M. R. Wang et al., 2017; NDRC, 2017; Ouyang et al., 2018; Paulot et al., 2014; Qu et al., 2017; Saikawa et al., 2014; X. Huang et al., 2012; Y. Huang & Li, 2014; Zhang et al., 2017, 2018; Zhou et al., 2014, 2016). This uncertainty is attributed to differences among methodologies, data on socioeconomic activities, and the emission factors (EFs) that relate agricultural activities (e.g., fertilizer usage) to soil Ngr emissions (H. Wu, Wang, et al., 2018; Zhan et al., 2019).

Top-down approach based on atmospheric Ngr concentrations and/or satellite observations provide an integrative benchmarking method (Pan et al., 2018; Van Damme et al., 2014); however, this approach is challenged from linking total Ngr emissions to local sources (Tian et al., 2016), which is important for practical N management (Gu et al., 2019) and for estimating their climate forcings (CFs; Liu et al., 2019). An alternative, bottom-up approach relies on collecting agriculture activity data (i.e., amounts of agricultural fertilizers application) and applying nitrogen EFs (the fraction of nitrogen inputs that is lost to various gases), which is in principle more capable of local-scale source attribution than top-down inventories. However, previous studies using bottom-up approaches have relied on agriculture activity data collected at the provincial (which exceed the size of many European countries) or national scale and applied Intergovernmental Panel on Climate Change (IPCC) default or uniform EFs for the whole country, ignoring the large spatiotemporal variation in agricultural EFs. As a result, significant discrepancies exist in China’s croplands Ngr emissions estimates.

In this study, we estimate the spatial and temporal distribution of the 2014 Ngr emissions from China’s croplands by combining bottom-up mass flow-based approaches with subprovincial level agricultural activity data and provincial condition-specific EFs (details refer to section 2). The subprovincial unit is called “prefecture” (Di Qu, Di Ji Shi in pinyin); it is an administrative unit in China and usually contains one center city and several mostly agricultural-dominated counties. Our estimates incorporate new crop-specific information, such as monthly rice irrigation rates at the provincial level. The data at subprovincial level offer a pathway toward more accurate estimates of Ngr emissions from China’s croplands. We compare our results to studies using bottom-up or top-down approaches (SI Table S1), revealing how our finer-resolution approach improves the estimate of China’s agricultural Ngr emissions, including magnitudes and the spatial-temporal pattern (SI Text S3).

Furthermore, we apply global temperature potential (GTP) to quantify the climate impacts of the Ngr emissions from China’s croplands. GTP represents the change in global average surface temperature at a chosen point in time via an equivalent pulse of CO₂, an approach which aligns with the UNFCCC goal of limiting future warming (Allen et al., 2016; Shine et al., 2005). With our new inventory data set, we estimate the

---

**Geophysical Research Letters**

10.1029/2019GL086551
relative contributions of China’s agricultural Ngr emissions to climate change using the GTP20 and GTP100 metrics, thereby addressing both short- and long-term climate impacts in parallel.

2. Methods

2.1. Data Sources

We consider five types of Ngr emissions sources from synthetic and organic fertilizers applied to croplands: chemical and compound fertilizers, livestock manure spreading, rural excrement, cake fertilizer, and straw returning. N₂O emissions from indirect sources (e.g., atmospheric N deposition and N leaching or runoff) are not considered due to their minor contribution fraction and the difficulty in separating them from other sources (Gu et al., 2015; Zhou et al., 2014). Ngr emissions are calculated by multiplying prefecture level activity data by provincial condition-specific EFs.

We collect emissions source data from China’s 387 prefectures based on the most recent administrative planning efforts. Notably, we also treat provincial level cities including Beijing, Shanghai, Tianjin, and Chongqing as prefectures in this study. Hong Kong and Macau have too little agricultural land and are thus excluded from the analysis. Prefecture-level data of the annual amounts of synthetic fertilizer (5 types), livestock (7 types), crop (17 types), and rural population for 2014 are directly derived from statistical yearbooks and bulletins of the 34 provinces in China (including Taiwan) (COA, 2015; EBCAHVY, 2015). The meteorological data are acquired from the TIANQIHOUBAO and the Central Weather Bureau. Soil pH data are obtained from the Harmonized World Soil Database v1.2. A land use map is acquired from the Data Center for Resources and Environmental Sciences (RESDC), Chinese Academy of Sciences. It is used as a proxy to rescale the prefecture-level Ngr emissions into a 1 km × 1 km spatial resolution, following the recommendation of previous studies (X. Huang et al., 2012).

2.2. Ngr Estimation Methodologies

For NH₃ EF calculations, we use the National Ammonia Reduction Strategy Evaluation System (NARSES) model to derive monthly and gridded condition-specific EFs from synthetic fertilizer application for each prefecture based on prefecture-level mean monthly temperature and precipitation, fertilization method, application rate and soil pH (1 km × 1 km) (Webb et al., 2006). Reference EFs, obtained from published field measurements in China, are adjusted by the above five impacting factors. Monthly proportions of synthetic fertilizer applied in each province are derived based on local information of cultivated crops (RESDC), corresponding fertilization timing (Zhang et al., 2018) and rates (Statistics of Cost and Income of Chinese Farm Produce, http://tongji.cnki.net/kns55/Navi/YearBook.aspx?id=N2015100281&floor=1). Note that the derived monthly proportions of a given province are assumed the same for all prefectures in this province. We consider 16 major crops that consume over 98% of China’s synthetic fertilizer applications (NBSC (National Bureau of Statistics of China), 2016). Then we obtain the monthly amounts of synthetic fertilizer applications of each prefecture based on the corresponding annual amounts of applications and monthly proportions. More details can be found in supporting information (SI) Text S1.1.

Provincial monthly condition-specific NH₃ EFs from livestock manure are based on the mass flow approach by considering the transformation of total ammoniacal nitrogen (TAN) embodied in livestock manure (TAN-flow model) (EEA, 2009). Manure management in China is grouped into four different stages: outdoor/grazing, housing, manure storage, and manure cropland application. TAN input to manure management is calculated by provincial-specific N excretion rate and free-range proportion, rearing period, percent of N in excreted feces transformed into TAN, and reference TAN emission coefficients. The obtained reference TAN emission coefficients of livestock manure housing stage are then adjusted by different temperature intervals based on provincial monthly average temperature. The detailed processes have been described in a previous study (Xu et al., 2017), which focus on manure cropland application and thus do not consider NH₃ emissions from outdoor/grazing, housing, and manure storage. Similar to previous studies (Kang et al., 2016; Xu et al., 2015, 2019), we assume that livestock population per month remained the same due to the small monthly fluctuation of meat, egg, and milk productions (http://www.caaa.cn/). The remaining NH₃ emissions sources are derived from a comprehensive EF database using the arithmetic mean of data collected from the literature and evenly distributed across the 12 months (X. Huang et al., 2012; Xu et al., 2016). Detailed model description, structure and parameters can be found in the SI Text S1.1.
We use the PKU-N$_2$O emissions model (Zhou et al., 2014), with further improvements, to quantify N$_2$O emissions. We identify rice irrigation months based on the types of rice cultivation at the provincial level considering China’s actual rice cropping systems (SI Table S2) (Clauss et al., 2016; NBSC, 2016; Yan et al., 2003). We then differentiate the EFs for irrigation and nonirrigation periods (Gao et al., 2011; Zhou et al., 2014; Zou et al., 2010) because flooded paddy fields have low N$_2$O emissions rates (Kritee et al., 2018). For monthly EFs during the irrigation period, we adopt the EFs of paddy fields in the corresponding month of each province, and for the nonirrigation period, we treat their EFs equal to that of drylands (SI Table S3). We do not account for monthly variation EFs of dryland fields in each province. Here, we adopt the improved PKU-N$_2$O emissions model at prefecture-level to calculate N in livestock manure, rural excrement, cake fertilizer, and straw returning applied to croplands, based on livestock and rural population sizes and crop yields. A detailed description of the model is provided in SI Text S2.

To calculate China’s cropland soil NO$_x$ (NO + NO$_2$) emissions caused by the application of agricultural fertilizers, we adopt a hybrid method combining the temperature dependence approach by Williams et al. (1992) and the YL95 scheme (Yienger & Levy, 1995). Due to the low emission rate from natural soils and limited data, we do not consider natural soil NO$_x$ emissions (Almaraz et al., 2018). We first use the arithmetic average of different experiments by collecting data from the literature to obtain the reference NO$_x$ N loss rates for drylands and paddy fields (Wang et al., 2005). We then use three soil temperature intervals and prefecture-level monthly average temperature to adjust the reference NO$_x$ N loss rates for the purpose of obtaining prefecture-level monthly emissions. We apply NO$_x$ N loss rate from cropland soil of 0 if the monthly average temperature is below 0°C (Yienger & Levy, 1995). Although frozen soil microbes may still be active, the effect of the freezing-thawing cycle on NO$_x$ N loss rate is negligible (Pilegaard, 2013). We use an empirical relationship between soil and air temperatures based on Feng and Cai (2004) to obtain soil temperature at a 50-cm depth. More details about the approach and the parameters are given in SI Text S3.

We estimate N$_2$O and NO$_x$ emissions also by separately considering agricultural fertilizers allocated to paddy and dryland fields in China. The amount of agricultural fertilizers applied to paddy fields in different provinces are calculated by multiplying the provincial mean nitrogen application rate (supporting information Table S3) (Wu et al., 2015) by the paddy fields area derived from RESDC. The remaining amount was assigned to dryland fields for each prefecture. The monthly proportions of livestock manure spreading are considered the same as those of synthetic fertilizer applications. The applications from the remaining sources of small contributions are evenly distributed across the 12 months.

2.3. Estimating GTP

To calculate the relative contribution of Ngr emissions from China’s agricultural fertilizer application to climate change, we use the GTP of CO$_2$e at 20 and 100 years (GTP$_{20}$ and GTP$_{100}$, respectively) for different Ngr gases. We adopt updates with more comprehensive agricultural emissions sources relevant to the China’s agriculture (Shi et al., 2015). Specifically, we applied the method of Pinder et al. (2012) to estimate four direct CFs from Ngr emissions, namely, the warming potential of N$_2$O itself (CF1), the aerosol effects of NH$_3$ (CF2), the aerosol effects of NO$_x$ (CF3), and the impacts of NO$_x$ on ozone and CH$_4$ (CF4). The direct cooling effect of nitrate and sulfate light-scattering aerosols attributed to NH$_3$ and NO$_x$ is included to estimating GTP$_{20}$ and GTP$_{100}$. We do not consider the indirect cooling effect of aerosols on clouds because it is highly localized and poorly quantified (Pinder et al., 2012). Although the aerosol effect of NH$_3$ and NO$_x$ shows considerable variations in space and time, currently there are no data of such variability available for China. A literature review and the latest IPCC assessment report provides the range of GTP values that reflect the current progress in calculating each of the climate change impacts from CF1 to CF4 (SI Table S4) (Boucher et al., 2009; Collins et al., 2013; IPCC, 2013; Pinder et al., 2012; Shi et al., 2015; Shindell et al., 2009). More details about the calculation and data sources are given in SI Text S2. In addition, to calculate China’s relative contribution to the GTP from agricultural fertilizer applications in 2014, we use the global croplands Ngr emissions in 2012 (latest available) obtained from EDGAR v4.3.2 (2017).

2.4. Uncertainty Analyses

We apply a Monte Carlo simulation with 10,000 runs to characterize the overall uncertainty of the Ngr emissions inventory and its climate impact based on the variations in socioeconomic activity data, EFs, and...
related calculation parameters. The results are characterized as medians and \( R_{50} \) (the difference between the 75th and 25th quartiles). The uncertainty analyses are introduced in detail in SI Text S5.

3. Results

3.1. Spatial and Seasonal Patterns of Ngr Emissions

Total Ngr emissions from China’s croplands (\( E_{\text{total}} \)) were approximately 8.87 Tg N yr\(^{-1} \) (\( R_{50} \): 6.69–11.06 Tg N yr\(^{-1} \)) in 2014. The majority of agricultural Ngr emissions occurred via NH\(_3\), which contributes approximately 8.06 (6.09–10.04) Tg N yr\(^{-1} \) or 91% of \( E_{\text{total}} \). N\(_2\)O and NO\(_x\) contribute 0.42 (0.34–0.50) and 0.38 (0.26–0.52) Tg N yr\(^{-1} \), respectively. Synthetic fertilizer was 71.5% of \( E_{\text{total}} \), with livestock manure spreading (19.1%), straw returning (4.3%), cake fertilizer (3.3%), and rural excrement (1.8%) making progressively smaller contributions.

The spatial distribution of the Ngr emissions densities is highly heterogeneous. Ngr emissions densities are highest (above 5.0 Mg N km\(^{-2} \) yr\(^{-1} \)) in the Northeast Plain, the North China Plain, the Huaihe River Basin, the Weihe Plain, the Sichuan Basin and the Lianghu Plain, which collectively compose China’s agricultural centers (Figure 1a). In particular, the largest emission densities of above 9.0 Mg N km\(^{-2} \) yr\(^{-1} \) are mostly present over the Henan-Shandong-Jianguo-Anhui border region, where the population and agriculture are highly concentrated. On the other hand, lower Ngr emissions densities below 1.0 Mg N km\(^{-2} \) yr\(^{-1} \) are concentrated in the northwest of the “Hu Huanyong line” (Figure 1a), an area accompanied by sparse population density (14.68 people·km\(^{-2} \)) and low agricultural production (Chen et al., 2016). The mean Ngr emissions per unit grain yield in China in 2014 was 16.8 kg N Mg\(^{-1} \) yr\(^{-1} \) (Figure 1b), which is higher than that of the European Union 28 (EU-28) countries (11.8 kg N Mg\(^{-1} \) yr\(^{-1} \)) and the United States (6.5 kg N Mg\(^{-1} \) yr\(^{-1} \)). Overall, the average N application rate in China was 250 kg N ha\(^{-1} \) yr\(^{-1} \), several times higher than rates in the EU-28 (47 kg N ha\(^{-1} \) yr\(^{-1} \)) and United States (76 kg N ha\(^{-1} \) yr\(^{-1} \)) over the period of 2010–2014 (FAO, 2018). Nitrogen use efficiency (NUE, the fraction of nitrogen input harvested as product) is only 0.25 in China compared to 0.52 in EU-28 and 0.68 in the United States during 2010 (Zhang et al., 2015). The mean Ngr emissions per unit cultivated area in 2014 was approximately 4.8 Mg N km\(^{-2} \) yr\(^{-1} \), and for most cultivated lands, the Ngr emission densities fell into the range of 1.2–6.0 Mg N km\(^{-2} \) yr\(^{-1} \) (Figure 1c).

Substantial seasonal variation is apparent nationwide (SI Figure S2), with highest and lowest peaks during the summer (4.4 Tg N yr\(^{-1} \)) and winter (0.8 Tg N yr\(^{-1} \)), respectively. Ngr emissions in the warmer months (April–September, 76.6%) are higher than in colder months (October–March, 23.4%), owing to a combination of higher temperature and intensive agricultural activities. The maximum amount of emissions occurs in July (1.6 Tg N yr\(^{-1} \)), which is 6.8 times that of the minimum amount of emissions (January). The differences between results of this study and those of many previous studies are mostly caused by the various sources of agricultural activity data and EF parameters such as soil temperature and precipitation (SI Table S1).

The seasonal variation in Ngr emissions is largely due to agricultural activities. In Hebei, Henan, Jiangsu, Shandong, and Anhui, Ngr emissions increase substantially in April due to top-dressing fertilizers for winter wheat production. Another emission peak occurs in June–August, attributable to basal and top dressings in preparation for summer maize plantings. In Hubei and Fujian, Ngr emissions in June are 52% and 55% greater than that in May, respectively, due to intensive fertilization for semiliterate rice. Additional comparisons and descriptions of results are shown in SI Texts S3 and S4.

3.2. Driving Factors of Ngr Emissions

Population density, economic development, and urbanization strongly impact the Ngr emissions from China’s croplands. Here, via application of same-year population and gross domestic productivity (GDP) data, we estimate that the mean Ngr emissions intensities per GDP and per capita (\( E_{\text{gdp}} \) and \( E_{\text{cap}} \)) in China are 1.2 g N·USD\(^{-1} \) yr\(^{-1} \) and 6.9 kg N·cap\(^{-1} \) yr\(^{-1} \), respectively (SI Figure S4). Correlation analysis at the prefecture-level (SI Table S7) demonstrates that Ngr emissions from Chinese croplands correlate positively with human population and GDP and negatively with urbanization rate (the ratio of urban population to total population). Note GDP per capita is positively correlated with urbanization rate and N fertilizer application per-unit cultivated area (SI Table S7). Prefecture-scale relationships between \( E_{\text{gdp}} \) and \( E_{\text{cap}} \) and socioeconomic development in China for 2014 are shown in Figure 2. \( E_{\text{gdp}} \) decreases exponentially as...
a function of urbanization rate and GDP per capita. However, the regression coefficient ($R^2$) between $E_{cap}$ and GDP per capita is much lower than that between $E_{cap}$ and urbanization rate, implying that urbanization rate is a better socioeconomic explanatory variable for the variation in $E_{cap}$ (Figure 2). The relationship between urbanization rate and $E_{cap}$ conforms to an inverted U-shape, where $E_{cap}$ first increases and then decreases as the urbanization rate grows. The turning point occurs at approximately 43% urbanization rate.

Figure 1. Spatial distribution of Ngr emissions from China's croplands in 2014 and its climate impacts. (a) Per unit cultivated area Ngr emissions. (b) Per unit grain yield (namely cereal yield) Ngr emissions. (c) Frequency histogram of Ngr emissions density. Note: HN-Henan, SD-Shandong, JS-Jiangsu, and AH-Anhui. The red line in (a) is the “Hu Huanyong line.” The climate impacts from China's cropland Ngr emissions measured by (d) GTP$_{20}$ and (e) GTP$_{100}$ at the prefecture level. The cumulative GTP contribution (by percentages) of China's prefectures (f) is summed from the highest to the lowest GTPs, and the 35 prefectures have negative GTP$_{20}$ values.
3.3. Spatial Characteristics of the GTP in China

With the new high-resolution inventory of China’s agricultural \( \text{N}_2\text{O} \) emissions and their EFs, we total climate impacts at approximately 79.0 Tg CO\(_2\)e (14.0 to 110.0 Tg CO\(_2\)e) for GTP\(_{20} \) and 203.4 Tg CO\(_2\)e (180.5 to 219.8 Tg CO\(_2\)e) for GTP\(_{100} \) (SI Table S8). We also estimate the relative contribution of China’s croplands \( \text{N}_2\text{O} \) emissions to each CFs of global croplands GTP. They account for approximately 24% and 32% of the current global croplands GTP\(_{20} \) and GTP\(_{100} \), given that the global GTP\(_{20} \) and GTP\(_{100} \) from agricultural fertilizer applications are 324.5 Tg CO\(_2\)e (88.6 to 469.0 Tg CO\(_2\)e) and 639.5 Tg CO\(_2\)e (595.3 to 661.8 Tg CO\(_2\)e) in this study. This contribution includes both the climate warming and cooling impacts of \( \text{N}_2\text{O} \) emissions (designated as CF\(_1 \)–CF\(_4 \)) below developed by Pinder et al. (2012) among which CF\(_1 \)–CF\(_4 \) (directly) represents the warming potential of \( \text{N}_2\text{O} \) itself (CF\(_1 \)), the aerosol effects of NH\(_3 \) (CF\(_2 \)), the aerosol effects of NO\(_x \) (CF\(_3 \)), and the impacts of NO\(_x \) on ozone and CH\(_4 \) (CF\(_4 \)). As a result, China’s croplands account for approximately 24% and 32% of the current global climate warming GTP\(_{20} \) and GTP\(_{100} \), respectively, by CF\(_1 \) and 42% and 58% of the current global climate cooling GTP\(_{20} \) and GTP\(_{100} \), respectively, by CF\(_2 \)–CF\(_4 \). The spatial distribution of \( \text{N}_2\text{O} \)’s contributions to GTP\(_{20} \) and GTP\(_{100} \) exhibit large spatial variation in China (Figures 1d, 1e, and SI Figure S7).

At the prefecture level, areas with high agricultural \( \text{N}_2\text{O} \) emissions-induced CFs (CF hot spots) are mostly located in the Northeast Plain and the middle and lower reaches of the Yangtze River for both GTP\(_{20} \) (Figure 1d) and GTP\(_{100} \) (Figure 1e). The top 10 CF hot spot prefectures measured by both GTP\(_{20} \) and GTP\(_{100} \) account for approximately 10% of China’s total cultivated area (SI Table S9). They are responsible for approximately 17% and 13% of the total GTP\(_{20} \) and GTP\(_{100} \), respectively. CF\(_2 \) and CF\(_4 \) (sum) of these
10 GTP20 cities together contribute to ~7.6 Tg CO2e and offset 34% of the warming effects of CF1. However, the climate cooling effects of CF2 and CF4 are largely short-lived, and CF2 and CF4 (sum) of these 10 GTP100 cities together offset 1% of the warming effects of CF1. Ngr in 35 (out of 387) prefectures impart a net cooling effect at ~1.9 Tg CO2e for GTP20 due to such offsetting effects (Figure 1f).

4. Discussion

We find that China accounts for approximately 26% of the current global croplands Ngr emissions, which is lower than its contribution to the global croplands GTP100 (32%). This difference occurs because short-lived compounds, NH3 and NOx, which account for a significant proportion of China's croplands Ngr emissions, and which impart a short-term cooling. When the spatial distributions of GTP20 (Figure 1d) and GTP100 (Figure 1e) are compared to that of NH3, N2O, and NOx emissions (SI Figure S3), higher net GTP20 and GTP100 are found in the middle and lower reaches of the Yangtze River and the Northeast Plain. In general, the spatial distribution of GTP100 is highly consistent with that of N2O's long-lived and powerful impact on global climate warming (~6% of global forcing) (Davidson, 2009). However, the spatial hot spots of Ngr emissions densities and high net GTP do not always overlap.

The inverted U-shape relationship between urbanization rate and Ecap revealed in this study resembles that of the environmental Kuznets curve (EKC). The EKC describes a phenomenon that environmental degradation first increases, then decreases, with economic development (Dinda, 2004). The EKC is sometimes considered as an optimist's view in curbing environmental deterioration (Zhang et al., 2015). Here we should also note that the EKC-like relationship between urbanization and Ecap is derived from the same-year data of different prefectures. While space-for-time substitution needs to be not overinterpreted, our observed spatial EKC-like phenomenon may still offer some insights on how urbanization may influence agricultural Ngr emissions. For prefectures at the low to middle urbanization levels, those with higher urbanization rates usually have higher incomes and more quests for food security, leading to increased N uses and decreases in NUE (Zhang et al., 2015). On the other hand, prefectures with even higher urbanization rates in China are usually richer and have the resources in technology and management practices (such as irrigation systems and machinery) that can increase NUE and thus reduce N fertilizer application per unit cultivated area (Y. Y. Wu, Xi, et al., 2018). However, highly urbanized prefectures usually also depend on other regions and countries for food supply; and the importation of food could also mean export of Ngr emissions (Zhang et al., 2015). Therefore, it is still an open question whether further urbanization may eventually lead to the reduction in Ngr emissions.

With continuous population growth, the world faces the substantial challenge of producing more food on less land area (M. R. Wang et al., 2018; Yu et al., 2019); hence, Ngr emissions are expected to rise in the absence of coordinated policies and innovations in N fertilizer technologies and management practices (Houlton et al., 2019; Lu et al., 2019; Zhang et al., 2015). Grain yields in China are substantially below potentially attainable levels (Zhang et al., 2016). In particular, the large gap between China and those developed countries in NUE implies a great potential for China in increasing its NUE (reducing Ngr emissions). China, United States, and EU-28 are all located in the northern temperate for most of their lands and share some similarity in their climate and soils, which offers a perspective for China's agricultural practice to learn from the United States and EU-28 in closing their gap in NUE. Genetic technologies for cereal breeding (Wu et al., 2020), for example, can be useful in enhancing grain yield yet without increasing Ngr emissions. Increasing grain yields and reducing Ngr emissions can be achieved by addressing N deficient regions with low N fertilizer use intensities. Our spatially refined yield-scaled Ngr emission data can inform policymakers to explore opportunities for increasing grain yields while maintaining or reducing Ngr emissions; regions with high yield/emission ratios should be given priorities for sustainable agriculture production.

Meanwhile, it is also crucial to understand what fertilizer management practices can be implemented to simultaneously increase food production, reduce emissions, and mitigate both long- and short-term climate changes. The derived provincial condition-specific Ngr EFs and the developed high spatiotemporal resolution emissions inventory in this study can help assess future mitigation policies and practices in China. For instance, China may consider increasing farm size to reduce fertilizer use due to the higher sensitivity of large farms to rising fertilizer prices (Ju et al., 2016). Y. Y. Wu, Xi, et al. (2018) suggested that a 1% increase in farm size would lead to a 0.3% decrease in fertilizer use per hectare. In addition, following the “4Rs” of N
fertilizer application (right type, right rate, right time, and right place) (IFA, 2009) and using improved agronomic management techniques (e.g., fertigation) are effective in reducing N use without compromising crop yields. For those provinces with high Ngr EFs, such as Henan and Shandong, local governments may consider substituting livestock manure for synthetic N fertilizer, because of the high NUE of manure and the potential benefit of the C sequestration in China’s croplands (Zhao et al., 2018). However, one also needs to bear in mind the differences between China and those developed countries. For instance, China has a higher population density and an even higher proportion of agricultural workers, resulting in a much smaller average farm size compared to that in EU-28 or United States.

Our study highlights additional opportunities to fill research gaps and needs. First, the activity data, EFs, and parameters used in NARSES, TAN-flow, PKU-N₂O and YL95 scheme models used for emission estimates impose various degrees of uncertainty. Additional uncertainties may be introduced by the lack of subprovincial data of fertilizer application rates. Hence, more Ngr field experiments and the development of process-based models and/or a reliable data-driven approach can help reduce such uncertainties. Second, the agricultural activity data we use span only 1 year in estimating Ngr emissions and are thereby limited from addressing interannual variation and dynamics in China’s cropland Ngr emissions. Earlier studies with coarse national/provincial-level data and IPCC default EFs or uniform EFs indicated an increasing trend in China’s cropland Ngr emissions from 1.9 Tg N yr⁻¹ in 1970 to 13.4 Tg N yr⁻¹ in 2012 (EDGAR v4.3.2, 2017). However, due to their coarse data resolution and potential systematic spatial biases, large uncertainties exist in both the annual Ngr emissions and their interannual variations (Zhou et al., 2014). In the future, our new bottom-up, mass flow-based approach can be extended to other years with improved agricultural activity data and therefore can inform interannual changes of China’s cropland Ngr emissions. Third, the climate impacts of the change in CO₂ and CH₄ flux from N deposition and fertilizer application are not considered in our study, and may cause incomplete an account of climate impacts. Their inclusion will further highlight the requirement for mitigation measures in preventing global warming, such as applying enhanced-efficiency fertilizers, better irrigation practices (Gao et al., 2018) and precision fertilization (Almaraz et al., 2018).

Data Availability Statement

The meteorological data are acquired from the website (http://www.tianqihoubao.com/lishi/and http://www.cwb.gov.tw). Soil pH data are obtained from the website (https://daac.ornl.gov/SOILS/guides/HWSD.html). A land use map is acquired from RESDC (http://www.resdc.cn). The output data of model simulations are available as described in Text S1 and Tables 2–6 in the supporting information.

Acknowledgments

This work was financially supported by the National Natural Science Foundation of China (41905079 and 51961125203), the China Postdoctoral Science Foundation (2019M662718), and the Strategic Priority Research Program of Chinese Academy of Sciences (XDA20100004).

References

Allen, M. R., Fuglestvedt, J. S., Shine, K. P., Reisinger, A., Pfeiferhertum, R. T., & Forster, P. M. (2016). New use of global warming potentials to compare cumulative and short-lived climate pollutants. *Nature Climate Change*, 6, 773.

Almaraz, M., Bai, E., Wang, C., Trousdell, J., Conley, S., Faloona, I., & Houlton, B. Z. (2018). Agriculture is a major source of NOx pollution in California. *Science Advances*, 4, eaao3477. https://doi.org/10.1126/sciadv.aao3477

Bodirsky, B. L., Popp, A., Lotze-Campen, H., Dietrich, J. P., Rolinski, S., Weindl, I., et al. (2014). Reactive nitrogen requirements to feed the world in 2050 and potential to mitigate nitrogen pollution. *Nature Communications*, 5, 3858. https://doi.org/10.1038/ncomms4858

Boucher, O., Friedlingstein, P., Collins, B., & Shine, K. P. (2009). The indirect global warming potential and global temperature change potential due to methane oxidation. *Environmental Research Letters*, 4, 044007. https://doi.org/10.1088/1748-9326/4/4/044007

Bowles, T. M., Atallah, S. S., Campbell, E. E., Gaudin, A. C., Wieder, W. R., & Grandy, A. S. (2018). Addressing agricultural nitrogen losses in a changing climate. *Nature Sustainability*, 1, 399–408. https://doi.org/10.1038/s41893-018-0106-6

Chen, M. P., Sun, F., & Shindo, J. (2016). China’s agricultural nitrogen flows in 2011: Environmental assessment and management scenario. *Resources Conservation and Recycling*, 111, 10–27. https://doi.org/10.1016/j.resconrec.2016.03.026

Chen, M. X., Gong, Y. H., Li, Y. L., Lu, D. D., & Zhang, H. (2016). Population distribution and urbanization on both sides of the Hu Huanyong line: Answering the Premier’s question. *Journal of Geographical Sciences*, 26(11), 1593–1610. https://doi.org/10.1007/s11442-016-1346-4

Clauss, K., Yan, H. M., & Kuenzer, C. (2016). Mapping Paddy Rice in China in 2002, 2005, 2010 and 2014 with MODIS time series. *Remote Sensing*, 8, 434. https://doi.org/10.3390/rs8050434

Collins, W. J., Fry, M. M., Yu, H., Fuglestvedt, J. S., Shindell, D. T., & West, J. J. (2013). Global and regional temperature-change potentials for near-term climate forcings. *Atmospheric Chemistry and Physics*, 13, 2471–2485. https://doi.org/10.5194/acp-13-2471-2013

Council of Agriculture (COA), 2015. Yearly report of Taiwan’s Agriculture Department of Agriculture and Forestry of Taiwan Provincial Government: Taipei, Taiwan, 2015.

Cui, S. H., Shi, Y. L., Groffman, P. M., Schlesinger, W. H., & Zhu, Y. G. (2013). Centennial-scale analysis of the creation and fate of reactive nitrogen in China (1910–2010). *Proceedings of the National Academy of Sciences of the United States of America*, 110, 2052–2057. https://doi.org/10.1073/pnas.1221638110

Davidson, E. A. (2009). The contribution of manure and fertilizer nitrogen to atmospheric nitrous oxide since 1860. *Nature Geoscience*, 2, 659–662. https://doi.org/10.1038/ngeo608
Dinda, S. (2004). Environmental Kuznets curve hypothesis: A survey. Ecological Economics, 49, 431–455. https://doi.org/10.1016/j.ecolecon.2004.02.011

EBCAHVY (Editorial Board of China Animal Husbandry and Veterinary Yearbook) (2015). China Animal Husbandry and Veterinary Yearbook 2015. Beijing: China Agriculture Press.

EDGAR v4.3.2 (2017). Emission Database for Global Atmospheric Research v4.3.2. http://edgar.jrc.ec.europa.eu/overview.php?v=432

EEA (European Environment Agency) (2009). EMEP/EEA air pollutant emission inventory guidebook 2009. Tech. Rep. 9/2009, Copenhagen.

FAO (Food and Agriculture Organization of the United Nations) (2018). Food and Agriculture Organization of the United Nations

Statistics Division: FAO Statistical Databases (Food Agric Organ UN, Rome, available at: http://faostat3.fao.org).

Feng, X. M., & Cai, D. L. (2004). Soil temperature in relation to air temperature altitude and latitude (in Chinese). Acta Pedologica Sinica, 41, 489–491. https://doi.org/10.11676/txb200303240327

Fesenfeld, L. P., Schmidt, T. S., & Schrode, A. (2018). Climate policy for short- and long-lived pollutants. Nature Climate Change, 8, 934–936.

Fowler, D., Coyle, M., Skiba, U., Sutton, M. A., Cape, J. N., Reis, S., et al. (2013). The global nitrogen cycle in the twenty-first century. Philosophical Transactions of the Royal Society B-Biological Sciences, 368.

Frank, S., Havlík, P., Stehfest, E., van Meijl, H., Witzke, P., Pöez-Dominguez, I., et al. (2019). Agricultural non-CO2 emission reduction potential in the context of the 1.5°C target. Nature Climate Change, 9, 66–72. https://doi.org/10.1038/s41558-018-0358-8

Fu, X., Wang, S. X., Ran, J. M., Pleim, J. E., Cooter, E., Bash, J. O., et al. (2015). Estimating NH3 emissions from agricultural fertilizer application in China using the bi-directional CMAQ model coupled to an agro-ecosystem model. Atmospheric Chemistry and Physics, 15, 6637–6649. https://doi.org/10.5194/acp-15-6637-2015

Galloway, J. N., Townsend, A. R., Erisman, J. W., Bekunda, M., Cai, Z. C., Freney, J. R., et al. (2008). Transformation of the nitrogen cycle: Recent trends, questions, and potential solutions. Science, 320, 889–892. https://doi.org/10.1126/science.1136674

Gao, B., Huang, T., Ju, X. T., Gu, B. J., Huang, W., Xu, L. L., et al. (2018). Chinese cropping systems are a net source of greenhouse gases despite soil carbon sequestration. Global Change Biochemistry, 24, 1–17. https://doi.org/10.1111/gcb.14425

Gao, B., Ju, X. T., Zhang, Q., Christie, F., & Zhang, F. S. (2011). New estimates of direct N2O emissions from Chinese croplands from 1980 to 2007 using localized emission factors. Biogeochemistry, 8, 3011–3024. https://doi.org/10.5194/bg-8-3011-2011

Gu, B. J., Ju, X. T., Chang, J. G., Ye, Y., & Vítousek, P. M. (2015). Integrated reactive nitrogen budgets and future trends in China. Proceedings of the National Academy of Sciences of the United States of America, 112, 8792–8797. https://doi.org/10.1073/pnas.1510211112

Gu, B. J., Lam, S. K., Reis, S., van Grinsven, H., Ju, X. T., Yan, X. Y., et al. (2019). Toward a general analytical framework for sustainable nitrogen management: Application for China. Environmental Science & Technology, 53, 1109–1118. https://doi.org/10.1021/acs.est.8b06370

Haines, A., Amann, M., Borgford-Pannell, N., Leonard, S., Kaylenstienza, J., & Shindell, D. (2017). Short-lived climate pollutant mitigation and the Sustainable Development Goals. Nature Climate Change, 7, 863–869. https://doi.org/10.1038/nclimate3588-017-0012-x

Houlton, B. Z., Almaraz, M., Aneja, V., Austin, A. T., Bai, E., Cassman, K. G., et al. (2019). A world of cobene. Nature Climate Change, 9, 714–720. https://doi.org/10.1038/s41558-019-0385-7

Huang, X., Song, Y., Li, M. M., Li, J. F., Hoo, Q., Cai, X. H., et al. (2012). A high-resolution ammonia emission inventory in China. Global Biogeochemical Cycles, 26, GB1030. https://doi.org/10.1029/2011GB004161

Huang, Y., & Li, D. J. (2014). Soil nitric oxide emissions from terrestrial ecosystems in China: A synthesis of modeling and measurements. Scientific Reports, 4, 7406.

IFA (International Fertilizer Industry Association) (2009). The Global “4R” Nutrient Stewardship Framework. Developing Fertilizer Best Management Practices for Delivering Economic, Social, and Environmental Benefits. AgCom/09/44, http://www.fertilizer.org/Library (IFA Task Force on Fertilizer Best Management Practices, IFA, 2009).

IPCC (2013). In T. F. Stocker, D. Qin, G.-K. Plattner, M. Tignor, S. K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex, & P. M. Midgley (Eds.), Climate change 2013: The physical science basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change (p. 1535). Cambridge, United Kingdom and New York, NY, USA: Cambridge University press.

Kang, Y. N., Liu, M. X., Song, Y., Huang, X., Yao, H., Cai, X. H., et al. (2016). High-resolution ammonia emissions inventories in China from 1980 to 2012. Atmospheric Chemistry and Physics, 16, 2043–2058. https://doi.org/10.5194/acp-16-2043-2016

Kriete, K., Nair, D., Zavala-Araiza, D., Provillie, J., Rudek, J., Adhya, T. K., et al. (2018). High nitrous oxide fluxes from rice indicate the need to manage water for both long- and short-term climate impacts. Proceedings of the National Academy of Sciences of the United States of America, 115, 9720–9725. https://doi.org/10.1073/pnas.1809276115

Lin, J., Khamnal, N., Liu, X., & Wang, X. (2019). China’s non-CO2 greenhouse gas emissions: Future trajectories and mitigation options and potential. Scientific Reports, 9.

Liu, M. X., Huang, X., Song, Y., Tang, J., Cao, J. J., Zhang, X. Y., et al. (2019). Ammonia emission control in China would mitigate haze pollution and nitrogen deposition, but worsen acid rain. Proceedings of the National Academy of Sciences of the United States of America, 116, 7780–7785. https://doi.org/10.1073/pnas.1804880116

Lu, C. Q., Zhang, J. E., Cao, P. Y., & Haftefield, J. L. (2019). Are we getting better in using nitrogen? Variations in nitrogen use efficiency of two cereal crops across the United States. Earth's Future, 7, 939–952. https://doi.org/10.1002/2019EF001155

Luo, Z. B., Hu, S. Y., Chen, D. J., & Zhu, B. (2018). From production to consumption: A coupled human-environmental nitrogen flow analysis in China. Environmental Science & Technology, 52, 2025–2035. https://doi.org/10.1021/acs.est.7b03471

NDRC (National Bureau of Statistics of China) (2016). China Agriculture Yearbook, 1980–2015. Beijing: China Statistics Press.

NDRC (Climate Change Department of National Development and Reform Commission) (2017). The People's Republic of China first biennial update report on climate change. Beijing: China: NDRC.

Ouyang, W., Tian, Z. M., Hao, X., Gu, X., Hao, P. H., Lin, C. Y., & Zhou, F. (2018). Increased ammonia emissions from synthetic fertilizers and land degradation associated with reduction in arable land area in China. Land Degradation & Development, 29, 3928–3939. https://doi.org/10.1002/ldr.3319

Pan, Y. F., Tian, S. L., Zhao, Y. H., Zhang, L., Zhu, X. Y., Gao, J., et al. (2018). Identifying ammonia hotspots in China using a National Observation Network. Environmental Science & Technology, 52, 3926–3934. https://doi.org/10.1021/acs.est.7b05235

Paulot, F., Jacob, D. J., Pinder, R. W., Bash, J. O., Travis, K., & Henze, D. K. (2014). Ammonia emissions in the United States, European Union, and China derived by high-resolution inversion of ammonium wet deposition data: Interpretation with a new agricultural emissions inventory (MASAGE_NH3). Journal of Geophysical Research: Atmospheres, 119, 4343–4364. https://doi.org/10.1002/2013JD021130
Zhou, F., Ciais, P., Hayashi, K., Galloway, J., Kim, D. G., Yang, C. L., et al. (2016). Re-estimating NH3 emissions from Chinese cropland by a new nonlinear model. *Environmental Science & Technology*, 50, 564–572. https://doi.org/10.1021/acs.est.5b03156

Zhou, F., Shang, Z. Y., Ciais, P., Tao, S., Piao, S. L., Raymond, P., et al. (2014). A new high-resolution N2O emission inventory for China in 2008. *Environmental Science & Technology*, 48, 8538–8547. https://doi.org/10.1021/es5018027

Zou, J. W., Lu, Y. Y., & Huang, Y. (2010). Estimates of synthetic fertilizer N-induced direct nitrous oxide emission from Chinese croplands during 1980–2000. *Environmental Pollution*, 158, 631–635. https://doi.org/10.1016/j.envpol.2009.08.026