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For more information, contact the Program office:

MIT Joint Program on the Science and Policy of Global Change
Postal Address:
Massachusetts Institute of Technology
77 Massachusetts Avenue, E19-411
Cambridge, MA 02139 (USA)
Location:
Building E19, Room 411
400 Main Street, Cambridge
Access:
Tel: (617) 253-7492
Fax: (617) 253-9845
Email: ***globalchange@mit.edu***
Website: ***http://globalchange.mit.edu/***

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Hanqin Tian[1,7]**, Chaoqun Lu**[1]**, Jerry Melillo**[2]**, Wei Ren**[1]**, Yao Huang**[3]**, Xiaofeng Xu**[1]**, Mingliang Liu**[1]**, Chi Zhang**[1,4]**, Guangsheng Chen**[1]**, Shufen Pan**[1]**, Jiyuan Liu**[5] **and John Reilly**[6]

[1] International Center for Climate and Global Change Research, and School of Forestry and Wildlife Sciences, Auburn University, Auburn, AL 36849, USA
[2] The Ecosystem Center, Marine Biological Laboratory, Woods Hole, MA 02543, USA
[3] Institute of Botany, Chinese Academy of Sciences, Beijing 100093, People's Republic of China
[4] Xinjiang Institute of Ecology and Geography, Chinese Academy of Sciences, Urumqi 830011, People's Republic of China
[5] Institute of Geographical Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing 100101, People's Republic of China
[6] Joint Program on Science and Policy of Global Change, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, MA 02139, USA

E-mail: tianhan@auburn.edu

Abstract
Chemical nitrogen (N) fertilizer has long been used to help meet the increasing food demands in China, the top N fertilizer consumer in the world. Growing concerns have been raised on the impacts of N fertilizer uses on food security and climate change, which is lack of quantification. Here we use a carbon–nitrogen (C–N) coupled ecosystem model, to quantify the food benefit and climate consequence of agronomic N addition in China over the six decades from 1949 to 2008. Results show that N fertilizer-induced crop yield and soil C sequestration had reached their peaks, while nitrous oxide (N_2O) emission continued rising as N was added. Since the early 2000s, stimulation of excessive N fertilizer uses to global climate warming through N_2O emission was estimated to outweigh their climate benefit in increasing CO_2 uptake. The net warming effect of N fertilizer uses, mainly centered in the North China Plain and the middle and lower reaches of Yangtze River Basin, with N_2O emission completely counteracting or even exceeding, by more than a factor of 2, the CO_2 sink. If we reduced the current N fertilizer level by 60% in 'over-fertilized' areas, N_2O emission would substantially decrease without significantly influencing crop yield and soil C sequestration.

Keywords: China, crop yield, CO_2 uptake, N_2O emission, nitrogen fertilizer

S Online supplementary data available from stacks.iop.org/ERL/7/044020/mmedia

[7] Address for correspondence: School of Forestry and Wildlife Sciences, Auburn University, 602 Duncan Drive, Auburn, AL 36849, USA.

1. Introduction

To meet increasing food demands for feeding 22% of the world's population, China has used chemical nitrogen (N) fertilizer for more than 50 years as one of the most important

agronomic practices to stimulate crop production (Huang *et al* 2007, Galloway *et al* 2008, Vitousek *et al* 2009). During 2007–8, China ranked as the top consumer of N fertilizer, accounting for 32% of global N consumption (Heffer 2009). Per-hectare anthropogenic N additions in many fields of China, especially for typical double-, triple-cropping systems, surpassed those in the United States and Northern Europe (Ju *et al* 2009, Vitousek *et al* 2009).

Numerous field-based studies have proved that excessive N fertilizer application would reduce air and water quality, rather than further stimulate crop yield (Tilman *et al* 2002, Yan *et al* 2010, Sutton *et al* 2011). Besides, N fertilization has the potential to affect the climate system in two important ways. On the one hand, it can stimulate carbon storage in soils associated with increased plant production, and thereby reduce the atmosphere's carbon dioxide (CO_2) content (Melillo *et al* 2010, Tian *et al* 2011a). On the other hand, it can lead to the increased production of nitrous oxide (N_2O), a more potent greenhouse gas (GHG) than CO_2 (Liu and Greaver 2009, Zaehle *et al* 2011). A recent study (Lu *et al* 2009) showed that N fertilizer uses were estimated to sequester 5.96 Tg C yr^{-1} (1 Tg = 10^{12} g) in China's cropland and that the potential C sequestration rate may reach up to 12.1 Tg C yr^{-1}, a significant 'carbon benefit'. But some scientists cast doubt on this 'carbon benefit', arguing a complete analysis of the benefit must consider N_2O emission induced by N fertilizer application (Huang and Tang 2010, Schlesinger 2010). The concurrent effects of N fertilizer application on crop yield, soil C sequestration and biogenic N_2O emissions across the entire nation are not well studied. The inherent complexity of this issue requires a systems approach that is capable of addressing non-linear and interactive ecosystem processes in agricultural land.

In this study, we simultaneously estimated changes in the yields of major crops (maize, rice, spring wheat, winter wheat, soybean, barley and others) and the net fluxes of CO_2 and N_2O induced by N fertilizer uses across China's cropland during 1949–2008, by using a C–N coupled, processes-based ecosystem model, the Dynamic Land Ecosystem Model (DLEM, Tian *et al* 2011a, 2011b). The model simulations were driven by spatially-explicit data on N fertilizer uses and other environmental factors such as climate, atmospheric CO_2, tropospheric O_3, N deposition, land use and land cover change. Besides the historical model simulation, we set up several additional experiments to examine how crop yield, soil C sequestration, and net balance of CO_2 and N_2O would respond to four different levels of N fertilizer reduction.

2. Methods

2.1. Data and model

We developed historical gridded data sets to characterize changes in N fertilizer application and other environmental factors, including climate, atmospheric CO_2 concentration, O_3 pollution, N deposition, land use and cover patterns, in China's terrestrial ecosystems with a spatial resolution of

10 km × 10 km during 1900–2008. Historical N fertilizer application data were retrieved from the county-level census data during 1981–2008 and the provincial tabular data during 1950–2008 shown in China Statistical Yearbook (National Bureau of Statistics, NBS, www.stats.gov.cn/). We counted 30% of compound fertilizer as chemical N input. N fertilizer applied on agricultural land averaged 21.11 g N m^{-2} yr^{-1} in 2008, increasing by 4 g N m^{-2} yr^{-1} per decade since 1950. The highest N fertilizer uses occurred in some areas of the North China Plain for double-cropping systems and lower reaches of the Yangtze River for triple-cropping systems (figure 1). The detailed information on other input drivers can be found in our published papers (Tian *et al* 2011a, 2011b, Ren *et al* 2011, Lu *et al* 2012). These input drivers were used to force the global C–N coupled ecosystem model, the DLEM, to examine how N fertilizer uses in China's cropland have affected crop yield, soil C sequestration, and net fluxes of two major greenhouse gases (CO_2 and N_2O) since 1949. The DLEM model has been well calibrated and intensively validated against various field observations from the Chinese Ecological Research Network (CERN), and other long-term N fertilization experiments (Tian *et al* 2011a, 2011b, Ren *et al* 2011, Lu *et al* 2012). For this study, we adopted global warming potential (GWP) to calculate the time-integrated radiative forcing of 1 kg greenhouse gas emission relative to that of 1 kg CO_2 (Forster *et al* 2007). According to the Fourth IPCC Report, N_2O has a 298 times higher radiative forcing than CO_2 at a 100 yr time horizon. Our study focused on the period 1949–2008, since chemical fertilizer uses were assumed to be 0 in China before 1949 due to the lack of long-term records.

2.2. Simulation experiments

We set up a series of model simulation experiments to distinguish the impacts of N fertilizer application in agricultural land: (1) all combined experiment, in which all the driving factors change over the period 1900–2008. (2) Combined without N fertilization change, with no application of chemical N fertilizer while other factors change throughout the study period. The difference between these two experiments indicated the effects of N fertilizer application and its interaction with other environmental changes.

In order to examine the modeled responses of crop yield, soil organic carbon (SOC) storage change and GHG balance to better N management practices, we conducted sensitivity analysis by setting up five additional simulation experiments driven by four N fertilizer reduction scenarios and one control scenario (no N fertilizer application change since 1900) spanning another 20 yr after 2008 to capture the legacy effect of N fertilizer use. These sensitivity simulation experiments did not include future climate scenarios. From climate change perspectives, we firstly identified areas with average N-induced GWP of CO_2 and N_2O larger than 0 during 2000–8 as 'over-fertilized' regions where the proportion of CO_2 sink offset by N_2O emission exceeds 100%. Then we assigned four levels of N fertilizer reduction in those 'over-fertilized' areas: 0% (baseline, N fertilizer use keeping
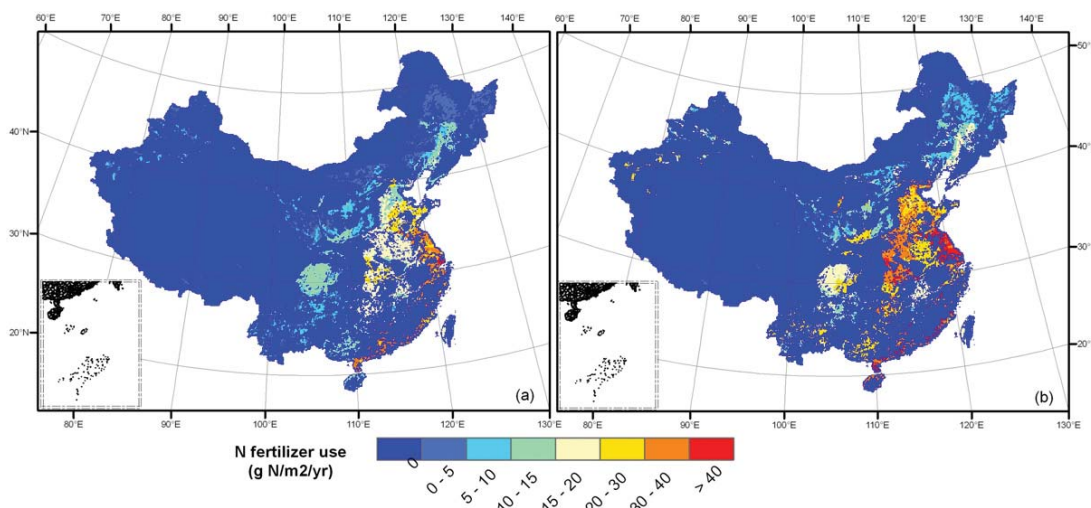
Figure 1. Spatial distribution of N fertilizer uses in China's agricultural land in 1990 (a) and 2008 (b).

constant at the current level, i.e., 2008), 20% (total amount of N fertilizer applied is equivalent to the level of late 1990s), 40% (equivalent to the level of early 1990s) and 60% (equivalent to the level of the late 1980s), while keeping N input in the remaining areas constant at the current level. Except N fertilizer application, other environmental forces were kept unchanged since 2008. The differences between the four N reduction experiments and control experiment were calculated to show the responses of China's cropland to different gradients of N fertilizer reduction. We used the N-induced changes relative to baseline simulation (0% reduction) to analyze the sensitivity of key variables to different gradients of N fertilizer reduction.

2.3. Evaluation of model performance

In this study, we validated model performance in simulating responses of crop yield, soil C sequestration and N_2O emission against the estimates from census and multiple N fertilization experiments (SBC 2000, Lu *et al* 2009, Gao *et al* 2011). The detailed data source and validation results can be found in the supplementary information (available at stacks. iop.org/ERL/7/044020/mmedia). Overall, the DLEM model is proved to perform well in simulating the dynamics in crop yield, soil C storage and GHG balance in response to N enrichment in China's cropland.

2.4. Statistical analysis

By using SPSS 20, we performed a Durbin–Watson (DW) test to examine whether serial correlation exists in N-induced CO_2 uptake and N_2O emission during 2000–8. We found negative serial correlation in N_2O emission, but the DW test is inconclusive for CO_2 uptake. After eliminating serial correlation through a Cochrane–Orcutt iterative procedure, we conducted a T test to check if the absolute means of modified CO_2 uptake and N_2O emission are statistically different from each other regardless of their opposite direction. In order to

assess the uncertainty of model estimates, we compared the modeled soil C sequestration and N_2O emission induced by N enrichment with field experiments. RMSD (root-mean-square deviation) was adopted to indicate the difference between the model estimate and observed values in groups of upland crop and paddy fields. We then used CV (RMSD), the RMSD normalized to the mean of multi-site observed values, to calculate the weighted-average uncertainty of regional estimates for N-induced CO_2 uptake and N_2O emission.

3. Results and discussion

3.1. N fertilizer-induced crop yield and soil organic C sequestration

As simulated by DLEM, crop yield and SOC were enhanced by N fertilizer application at the annual rates of 1.8 Tg C yr^{-1} and 1.3 Tg C yr^{-1}, respectively, during the period 1949–90. Crop yield and SOC storage reached their peaks in the early 2000s and 1990s, respectively. The N-induced soil C sequestration rate even began to decline thereafter (figure 2(a)). Although the N-induced crop yield slightly increased from 93 Tg C yr^{-1} in the 1990s to 98 Tg C yr^{-1} during the recent 9 yr (2000–8), we found that the relative contribution of chemical N fertilizer uses to crop yield was estimated to decrease from 53% to 49% over the past two decades. It implied that the importance of chemical N fertilizer in determining crop yield declined in recent years due to overuses of N fertilizer and increased contributions from other factors such as genetic improvement. Our findings are supported by emerging lines of evidence (Zhu and Chen 2002, Guo *et al* 2010) that partial factor productivity from fertilized N (PFP_N, calculated as the crop yield per unit nitrogen applied) declined in many fields of China in recent years, although the amount of N added as chemical fertilizer kept rising (supplementary figure S1 available at stacks.iop. org/ERL/7/044020/mmedia), implying that N fertilizer has been increasingly overused and N use efficiency in sustaining
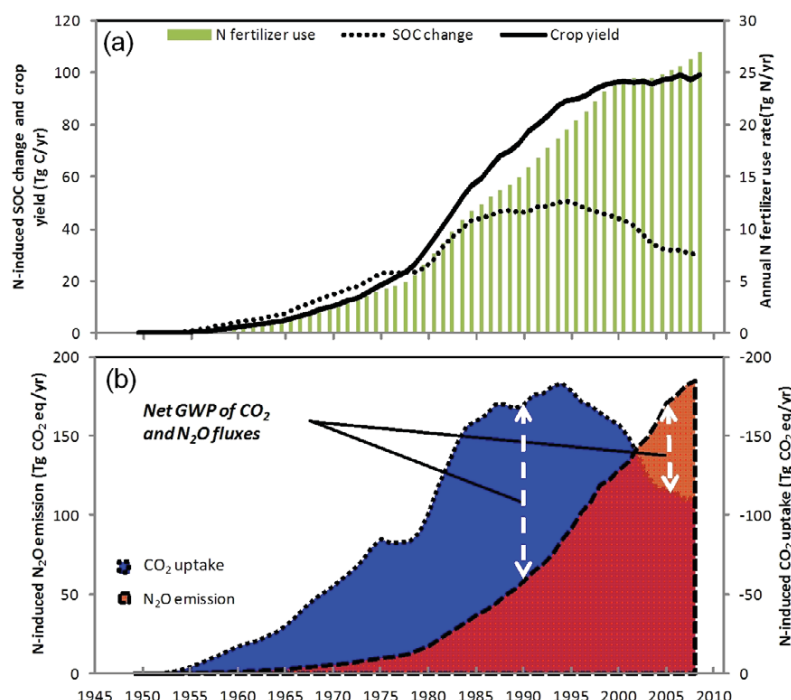
Figure 2. (a) Soil carbon sequestration, and crop yield induced by N fertilizer application in China's cropland during 1949–2008. (b) Global warming potential (GWP, unit: Tg CO_2 eq yr^{-1}) of CO_2 uptake and N_2O emission induced by N fertilizer application in China's cropland during 1949–2008. Note: GWP of CO_2 uptake is negative and surpassed N_2O emission before 2002, whereas climate change mitigation potential estimated from an increased CO_2 sink under N fertilizer application was totally offset by N_2O in recent years.

China's food security shrank. Experimental evidence also shows that N added in intensive double-cropping systems of China can be cut by 30–60% without significantly diminishing crop yield, yet reducing N loss to the environment by more than 50% (Ju *et al* 2009).

The accumulation rate of SOC due to anthropogenic N input also slowed, averaging 32 Tg C yr^{-1} during 2000–8, compared to 48 Tg C yr^{-1} estimated across China's cropland in the 1990s. The continuous rise in N fertilization rate might not further stimulate C accumulation in agricultural soils because crop productivity in response to N addition began to decline in large areas of China, which resulted in less C entering into the soil. In addition, long-term N fertilization experiments revealed that the more labile soil carbon fraction, including microbial and plant residues, is prone to being decomposed rather than accumulating with more available N (Neff *et al* 2002).

3.2. N fertilizer-induced GHG fluxes

Over the entire study period, N_2O emissions continued to rise as N fertilization rate increased. Direct N_2O emissions from N fertilizer uses in China was estimated to increase from 222 Gg N yr^{-1} (1 Gg = 10^9 g) in the 1990s to 369 Gg N yr^{-1} throughout the recent 9 years. On a national scale, the average N_2O emission factor grew from 1.0% of fertilizer applied in the 1990s to 1.4% during 2000–8, indicating that the surplus N was increasingly released, instead of being retained in the agricultural ecosystem.

In terms of GWP, our simulations indicated that N fertilizer application acted as a small mitigator of climate warming in the early part of the study period through an N-stimulated CO_2 sink. This sink began to be offset by N_2O emissions from the 1970s onwards. Since 2002, N fertilizer-induced climate warming through N_2O emissions has exceeded the cooling effect due to CO_2 storage in China's croplands (figure 2(b)). Over the past decade (2000–8), we found that agronomic N-induced CO_2 uptake (-117.3 ± 42.5 Tg CO_2 eq yr^{-1}, mean $\pm$ RMSD) was statistically different ($p < 0.05$) from N_2O emission (172.8 ± 43.0 Tg CO_2 eq yr^{-1}, mean $\pm$ RMSD) even in their absolute magnitude. Up to 2008, the net GWP totaled 73 Tg CO_2 eq yr^{-1}, with N_2O emissions offsetting CO_2 uptake by 166%. CO_2 uptake and N_2O emission showed an even larger statistical difference ($p < 0.001$) after removing serial correlations.

Climate consequences of N fertilizer application in terms of GHG balance showed large spatial variability (figure 3). The largest CO_2 sink and N_2O source both occurred in the North China Plain and the middle and lower reaches of Yangtze River Basin, where typical double- and triple-cropping systems have been used and high-level N fertilizer has been applied (figures 3(a)–(d)). Compared to 1990s, the largest CO_2 sink shown in the mid-mainland of China began to diminish, while the maximum N_2O source continued to expand over the recent 9 years. Our study shows that the net warming effect associated with N fertilizer uses mainly centered in intensively managed crop systems, with N_2O emission completely counteracting or even exceeding,
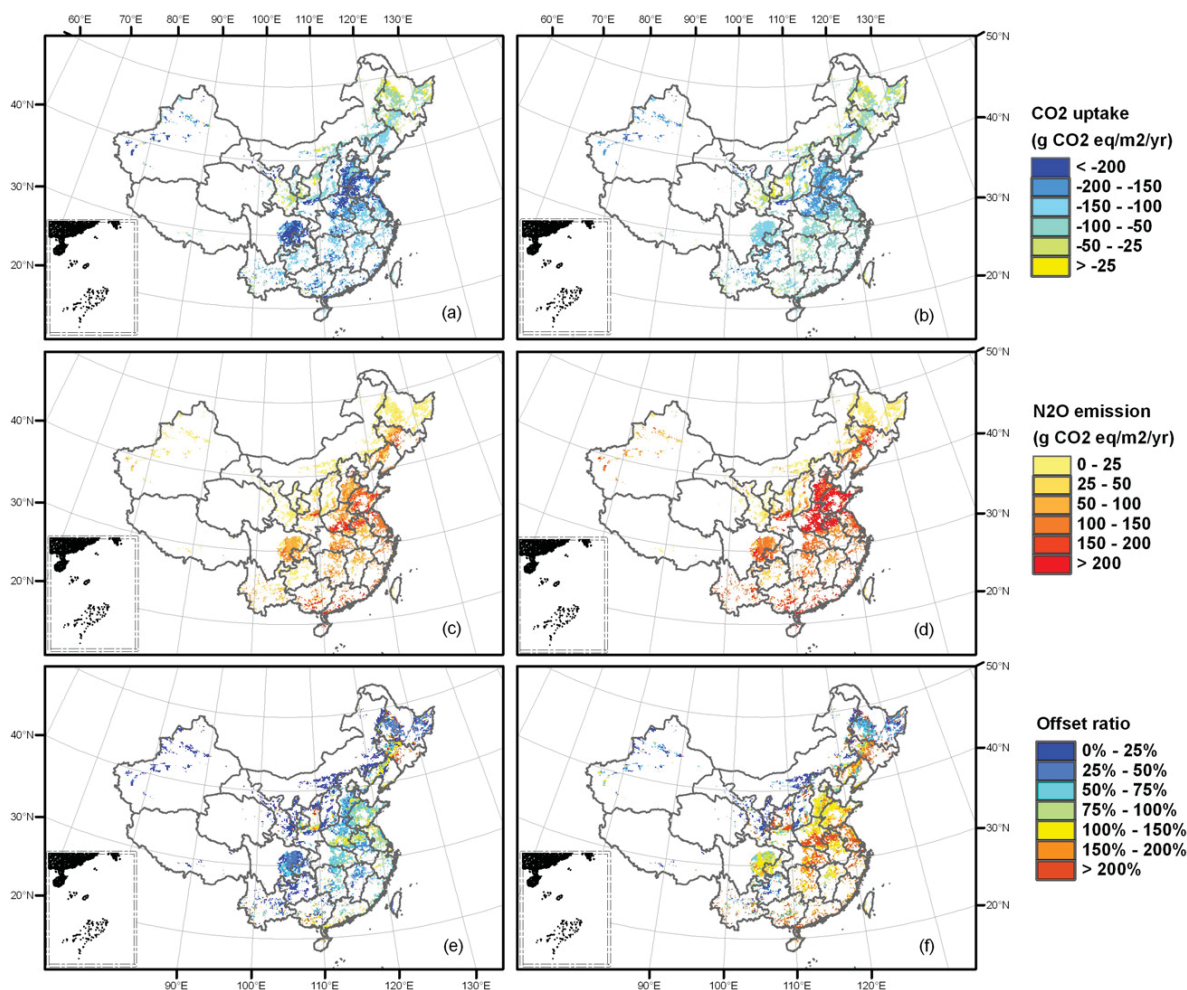
Figure 3. Global warming potential of (a), (b) CO_2, and (c), (d) N_2O, and (e), (f) offset ratio of N_2O emission to CO_2 sink induced by N fertilizer application in China's cropland during the 1990s (left panel) and the period 2000–8 (right panel).

by more than a factor of 2, the CO_2 sink during the recent 9 years (color yellow to orange in figure 3(f)). Only a small portion of China's cropland still played a mitigating role for the local and global climate system, with CO_2 uptake largely offset by the concurrently released N_2O from soils (color blue to green in figure 3(f)).

3.3. Sensitivity analysis of N fertilizer reductions

We assumed that N fertilizer uses in 'over-fertilized' areas of China (defined as offset ratio larger than 100% in figure 3(f)) would be reduced by 0% (baseline, continue applying N fertilizer in current rate), 20%, 40% and 60%, and used these four N fertilizer reduction levels to extend the model simulations for another 20 years. The 'what-if' experiments are able to reveal the likely patterns of crop yield, soil C storage and net GWP of CO_2 and N_2O fluxes in response to different N management strategies. Amazingly, our simulation showed that both crop yield and soil C sequestration induced by N fertilizer uses would decrease, but not change much with N fertilizer reductions compared to those driven by

current N fertilizer use rate. For example, 60% of N reduction would lead to an N-induced crop yield decrease of 1.88 Tg C yr^{-1} or 2%, compared to the experiment continuing fertilizer application at the current rate. However, direct agricultural N_2O emission would significantly decrease along with less N fertilizer inputs (table 1). Compared to the baseline, N-induced N_2O emission would decrease by 99 Tg CO_2 eq yr^{-1}, or 49.8%, with N fertilizer reduced by 60% of the current level in 'over-fertilized' areas. We also found that the amount of N leached out of the ecosystem largely decreased with N fertilizer reductions while plant N uptake remained with no significant changes. This implied that in 'over-fertilized' areas N fertilizer reduction as high as 60% would substantially decrease the loss of excessive N, alleviate air and water pollution, while not apparently suppressing crop yield.

With N fertilizer reduced by 40%–60%, the modeled CO_2 uptake in China's cropland showed a small decrease of 8.5%–15.8% compared to that driven by current fertilizer use rate (table 1). However, N_2O emission would largely decrease as N fertilizer use reduces. For example, as simulated by
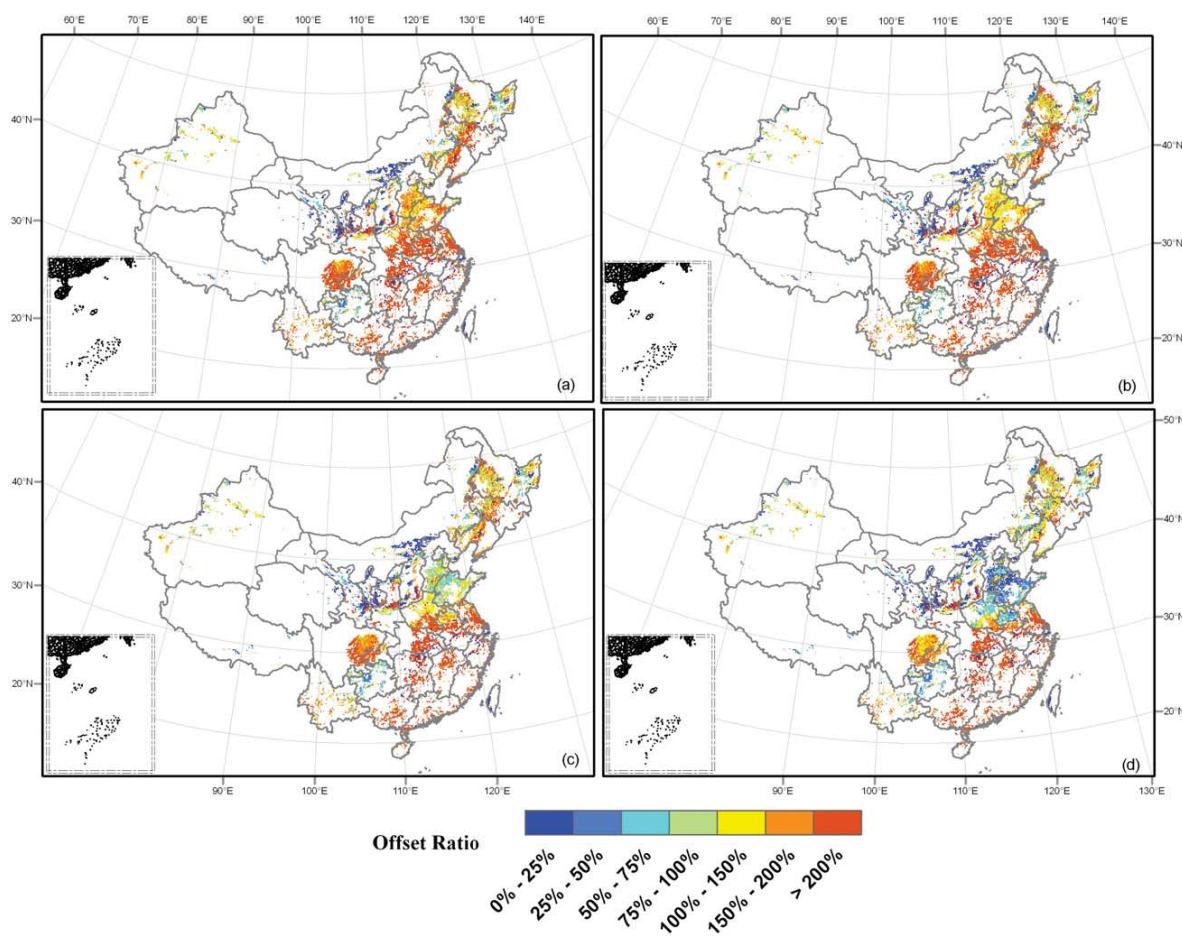
Figure 4. Offset ratio of N_2O emission to CO_2 sink induced by 4 levels of N fertilizer reduction in China's cropland: (a) N fertilizer kept constant at current level; (b) N fertilizer reduced by 20% of current level; (c) N fertilizer reduced by 40% of current level; (d) N fertilizer reduced by 60 per cent of current level in the 'over-fertilizer' areas (offset ratio $\geq$ 1 during 2000–8).

Table 1. Sensitivity analysis on impacts of different N reduction levels on crop yield, soil organic C (SOC) sequestration, GHG balance and leaching of N (including both dissolved inorganic N and dissolved organic N) in China's cropland. (Note: (a) the percentages shown in the table are the simulated changes in crop yield, soil C sequestration and GHG balance induced by three levels of N reduction relative to those corresponding values driven by the current (2008) N fertilizer application rate. (b) Nfer_20%, Nfer_40% and Nfer_60% stand for simulation experiments with N fertilizer use reduced by 20%, 40% and 60% of the current level in 'over-fertilized' areas, respectively.)

N reductions	N-induced changes	Yield (Tg C/yr)	SOC change (Tg C/yr)	CO_2 uptake (Tg CO_2 eq/yr)	N_2O emission (Tg CO_2 eq/yr)	Net GWP (Tg CO_2 eq/yr)	N leaching (Tg N/yr)
Nfer_20%	**change**	−0.88	−0.89	−3.29	−22.38	−19.08	−3.58
	change perc	−0.9%	−3.3%	−3.3%	−11.2%	−19.2%	−21.60%
Nfer_40%	**change**	−1.88	−2.31	−8.56	−56.25	−47.70	−6.82
	change perc	−2.0%	−8.5%	−8.5%	−28.2%	−48.0%	−41.10%
Nfer_60%	**change**	−1.88	−4.26	−15.82	−99.38	−83.55	−9.70
	change perc	−2.0%	−15.6%	−15.8%	−49.8%	−84.1%	−58.40%

DLEM, the net GWP values would decrease by 84%, and the CO_2 sink offset ratio would be lowered to 119% with 60% N fertilizer reduction. Therefore, if we reduce N fertilizer uses by ∼60% in 'over-fertilized' areas, China's cropland would act as a small to neutral climate heater while keeping crop yield relatively stable.

With N fertilizer use reduction, the N-induced climate warming area in China's cropland would significantly shrink.

The decreased offset ratio primarily occurred in the North China Plain, across which large areas of cropland would turn to a mitigator of climate warming with N fertilizer use reduced by 40%–60% (figure 4). However, such a reduction would not completely change the climatic role of cropland in the southeastern part. This might be related to the following facts: (1) typical double- and triple-cropping systems distributed in this area were characterized by intensive or excessive

agronomic N inputs; (2) other N sources such as higher N deposition and mineralized N due to faster turnover in this region provide a large amount of soil available N, which is the major substrate of N_2O production.

3.4. Uncertainty and research needs

While using a systems approach to simultaneously examine N fertilizer impact on crop yield, soil C storage and GHG balance, our estimated responses may have some uncertainties for several reasons. First, we ignored growth limitations from other nutrients such as potassium (K) and phosphorus (P), and made the assumption of an optimized irrigation strategy without water logging (Ren *et al* 2011) which might, to a small degree, have led us to overestimate the responses of crop yield to anthropogenic N additions. Second, we included maize, rice, spring wheat, winter wheat, soybean, barley and other crops in China to represent both upland and paddy fields, and then assumed N fertilizer use per unit harvest area of grain crops in each county was the same as that of cash crops such as vegetable and fruits. The harvest area of cash crops accounted for more than one fifth of China's arable and permanent crop area in 2005, with N fertilizer consumption over 30% of the national total (Heffer 2009). This simplification might, on the one hand, cause our underestimation on the decline of N-induced crop yield and the increase of N_2O emission, since cash crops tend to be more fertilizer intensive, and less effective in using N than grain crops. On the other hand, our simulation might overestimate N_2O emission, since implementation of a soil testing and fertilizer recommendation (STFR) program in some areas of China over 2001–8 was shown to reduce chemical N fertilizer uses and consequently decrease N_2O emission from wheat, rice and maize crops (Sun and Huang 2012). Third, our optimized water management (mid-season drainage) assumption would partly overestimate N_2O emission, as dry–wet alteration in paddy fields was proved to stimulate nitrification and denitrification processes, and therefore favor N_2O emission (Zou *et al* 2010). However, the resulting bias would not radically change our major conclusion.

This study does not consider CO_2 emission from N fertilizer production, transport and application (Huang and Tang 2010, Schlesinger 2010). Indirect N_2O emissions derived from re-deposition of volatilized N and N leached to downstream aquatic ecosystems (Zaehle *et al* 2011) are excluded here. If considering these two components, the stimulating effect of N fertilizer uses on climate warming would be higher than that estimated in this study. In addition to stimulating climate warming, excessive N fertilizer uses contributed to increasing the potential N pollution and eutrophication in downstream aquatic ecosystems (table 1). Taking both atmospheric and aquatic environment costs into account is urgently needed for assessing the impacts of agronomic N input in future studies.

4. Conclusion

Past research efforts have led to a growing recognition that N fertilizer is commonly overused in China's agricultural

production and has widely caused environmental problems. However, few studies have considered the climate consequences of excessive N fertilizer application in China. From our study we draw the following conclusions: (1) crop yield in China was substantially boosted by chemical N fertilizer uses over the past decades, but it was shown to barely further increase with surplus N input in recent years; (2) SOC accumulation induced by N added in cropland reached its peak in the 1990s and has since declined ; (3) overuse of chemical N fertilizer in China has led to an expanding warming effect, with direct N_2O emission offsetting the soil carbon sink by 166% in 2008; (4) the largest net sources of greenhouse gases associated with agronomic N addition have occurred in the North China Plain and in the middle and lower reaches of the Yangtze River Basin, where intensively managed crop systems are located; (5) if we reduce N fertilizer use by 60% in the 'over-fertilized' region, N-induced crop yield and SOC sequestration would slightly decrease, but direct N_2O emission would substantially decrease, and China's cropland would act as a small to neutral climate heater while keeping crop yield relatively stable; (6) The most apparent mitigation of climate warming due to N fertilizer reduction practices would occur in the North China Plain. Our research suggests that enhancing N use efficiency instead of increasing N inputs in the future would be more effective to sustain China's food security and cropland soil C sequestration, and diminish the climate warming and water pollution aggravated by anthropogenic N enrichment.

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