OPEN

Impacts of reactive nitrogen on climate change in China

Yalan Shi[1], Shenghui Cui[1], Xiaotang Ju[2], Zucong Cai[3] & Yong-Guan Zhu[1,4]

[1]Institute of Urban Environment, Chinese Academy of Sciences, 1799 Jimei Road, Xiamen 361021, PR China, [2]College of Resources and Environmental Sciences, China Agricultural University, Beijing 100193, PR China, [3]College of Geography Science, Nanjing Normal University, Nanjing 210023, PR China, [4]Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, 18 Shuangqing Road, Beijing 10085, PR China.

China is mobilizing the largest anthropogenic reactive nitrogen (Nr) in the world due to agricultural, industrial and urban development. However, the climate effects related to Nr in China remain largely unclear. Here we comprehensively estimate that the net climate effects of Nr are -100 ± 414 and 322 ± 163 Tg CO_2e on a GTP_{20} and a GTP_{100} basis, respectively. Agriculture contributes to warming at 187 ± 108 and 186 ± 56 Tg CO_2e on a 20-y and 100-y basis, respectively, dominated by long-lived nitrous oxide (N_2O) from fertilized soils. On a 20-y basis, industry contributes to cooling at -287 ± 306 Tg CO_2e, largely owing to emissions of nitrogen oxides (NO_x) altering tropospheric ozone, methane and aerosol concentrations. However, these effects are short-lived. The effect of industry converts to warming at 136 ± 107 Tg CO_2e on a 100-y basis, mainly as a result of the reduced carbon (C) sink from the NO_x-induced ozone effect on plant damage. On balance, the warming effects of gaseous Nr are partly offset by the cooling effects of N-induced carbon sequestration in terrestrial ecosystems. The large mitigation potentials through reductions in agricultural N_2O and industrial NO_x will accompany by a certain mitigation pressure from limited N-induced C sequestration in the future.

Anthropogenic interventions of the global biogeochemical nitrogen (N) cycle have substantially raised the levels of reactive N [Nr, in general, including organic N, ammonium (NH_4-N in water and NH_3 in air), nitrogen oxides (NO_x), nitrous oxide (N_2O), and nitrite & nitrate] in circulation, and caused severe damage to the environment at local, regional and global scales[1,2]. Such dramatic increases can aggravate the impacts on the balance of climate forces in many ways, either through changes to the atmospheric constituents, or through feedbacks within the terrestrial ecosystem[3]. The atmospheric N_2O emission directly contributes to global warming and ozone depletion; NO_x compounds indirectly produce the short-lived climate-forcing agents (e.g. ozone, aerosol particles) and remove the long-lived climate-forcing agents (e.g. methane (CH_4)); NH_3 also increases the formation of aerosols. In addition, the deposited and applied N into the terrestrial ecosystem alters the carbon (C) exchange and greenhouse gas (GHG) fluxes between land and air[3]. There are many cooling and warming interactions between N and the climate; the dominant effect of N on climate change is likely to vary across regions[4]. For an equivalent increase in the human population, the emission mass of CO_2-C into the environment is about 40 times that of Nr[3]. Although the absolute climate impact of Nr is dwarfed by the impact of CO_2 emissions, the variety of Nr species and sources, and the complexity of climate forcers, indicates a need to better assess the integrated effects of Nr and to identify the key forcers at different scales, so as to take targeted measures for N management. A few studies have attempted to assess the combined impacts of Nr at global and regional scales[5,6]. The lack of systemic study of Nr on climate at the national level has also been noted by Pinder et al.[7], taking the United States as a case study. China is becoming the largest producer of Nr from both agricultural and industrial activities[8]. The climate effects of Nr emissions in China are global rather than regional. The magnitude and acceleration of Nr creation in China warrants an integrated assessment of the significance of anthropogenic Nr on climate change.

N_2O, as one of the GHGs, has gained worldwide attention, and has also been thoroughly assessed in China[9,10]. It is well known that reducing N_2O emissions offers the combined benefits of mitigating climate change and protecting the ozone layer[10]. The increasing demands for food and energy in China have led to ever-increasing emissions of NO_x and NH_3[11]. These short-lived Nr emissions have multiple and profound effects on climate, the environment and human health[12]. However, their relevance to climate in China is still unclear in previous studies. In addition, ecosystem N availability strongly affects the terrestrial net C balance[13], as well as many other ecosystem services associated with environmental quality[14]. The terrestrial ecosystem C balance in China was

investigated to under the influence of multiple global change factors[15,16]. The effects of N deposition and N fertilizer use have been identified in a few studies[17,18].

In this context, a comprehensive assessment of Nr emissions and use on climate impacts in China is needed to fully understand the consequences of Chinese agricultural, industrial and urban development. To address this need, we seek to answer three questions: (i) What are the climate change impacts of Nr emissions in China, (ii) what are the relative contributions from agriculture and industry sources, and (iii) what are the future climate impacts of Chinese Nr based on different development scenarios? A key challenge is to establish a calculation database for the different climate forcers related to anthropogenic Nr in China. We review and reconcile the relative data and parameters from a variety of literatures related to the Nr emissions in China and their radiative forcing intensities and atmospheric lifetimes. Global temperature potential (GTP) is the common metric to quantify and intercomparing the relative effects of climate forcers over varying timescales. GTP evaluates not only the long-term climate effect caused by emissions of long-lived species like N_2O, but also the near-term climate fluctuations caused by emissions of the short-lived species NO_x and NH_3[7,19]. Accordingly, we use GTP to convert the climate change impacts of each of the processes to common units of equivalent Tg CO_2 (CO_2e).

In this analysis, we combine the set of Representative Concentration Pathway (RCP) scenarios, based on consistent radiative forcing levels[20], and the set of Special Report on Emission Scenarios (SRES) scenarios, based on the best estimates of the current emissions and further mitigation potential[21]. The comparison between RCPs and SRES is useful to determine the mitigation potentials or pressures of China's development and make the reasonable N management measures.

Results

Gaseous Nr effects on climate forcers in the atmosphere. The different gaseous Nr species show the different increase rates during 2000–2005 in China, with 11.4%, 44.0% and 8.3% for N_2O, NO_x and NH_3, respectively (Fig. 1). The emission sectors of Nr also show large variations. N_2O is the only form of Nr that has unambiguous global implications through the strong warming effect on climate (E1). Total anthropogenic N_2O emissions in China for 2005 were estimated at 1.04–1.22 Tg N. The primary source of N_2O is agriculture. N_2O emissions from agricultural soils (including emissions from applied fertilizer, manure and crop residues) amounted to a total of 0.59–0.87 Tg N, it can be further categorized as either direct (0.40–0.59 Tg) or indirect (0.19–0.28 Tg) emissions. Direct emissions are
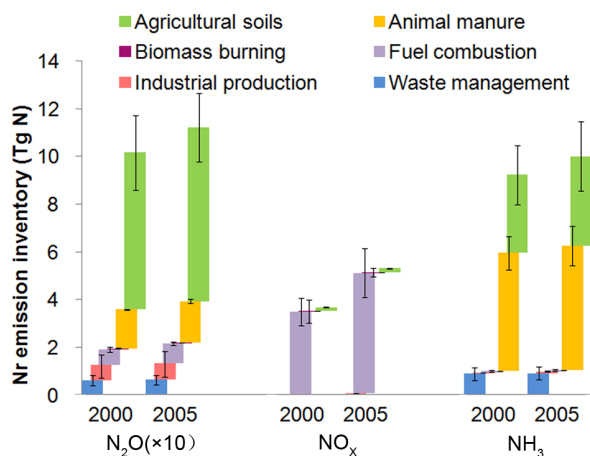
caused by N applied to the soils, while indirect emissions are related to subsequent processes after Nr emissions and re-deposition, or after leaching and runoff. Animal manure, including emissions from manure management and grazing animals, contributed 0.17–0.18 Tg N to the atmosphere. The emission factors for direct and indirect N_2O emissions from agriculture in China were 1.87% and 0.66%, a little higher than the global mean values of 1.48% and 0.42%, respectively[22]. In addition, N_2O from fuel combustion, industrial production, waste disposal and biomass burning delivered only 0.14–0.30 Tg N to the atmosphere. As a result, N_2O exerts a strong warming effect at 345 ± 19 Tg CO_2e on a GTP_{20} basis, then transforms to 311 ± 17 Tg CO_2e on a GTP_{100} basis (Fig. 2, Table S7). The close climate impacts on both timescales imply lasting effects on the climate system resulting from N_2O due to its long atmospheric lifetime of around 120 years.

NO_x and NH_3 contribute to climate change indirectly through altering the atmospheric constituents and corresponding climate forcers. NO_x is a major byproduct of combustion. Fuel combustion in China resulted in a total of 4.04–6.08 Tg N of NO_x emissions in 2005, mainly from the combustion sectors of power plants (1.15–2.08 Tg N), transportation (1.26–1.66 Tg N), industry (1.35–1.79 Tg N) and household use (0.27–0.54 Tg N). Agricultural soils contribute a minor part with only 0.17 Tg NO_x-N, mainly through microbial activity from agricultural soils. In sum, 4.34–6.24 Tg N is released to the atmosphere in the form of NO_x. In comparison, the total emissions of NH_3 were 7.69–12.3 Tg N in 2005, becoming the largest source of atmospheric Nr in China. Agricultural soils (including volatilization and applied fertilizer and manure) and animal and human manure management are the main contributors, at 2.30–5.20 and 4.37–6.02 Tg N, respectively; other sources are negligible for the country as a whole. As a result, the combined climate impact of NO_x on ozone and CH_4 (i.e. ozone-CH_4 effect, E2) in China is −350 ± 166 and −13.2 ± 7.2 Tg CO_2e on a 20-y and 100-y basis, respectively (Fig. 2, Table S7). Our results show that the net cooling effect of NO_x on ozone and CH_4 is notable only at a shorter time horizon, and gradually wears off with increasing time horizon. In addition, exposure to tropospheric ozone is known to inhibit photosynthesis, damage plants, and reduce plant primary productivity and land C sequestration capacity (E6)[23]. The reduction in C sequestration due to the ozone effect for China has been simulated by the terrestrial ecosystem model and estimated at 111 ± 100 Tg CO_2e under controlled simulations for different scenarios[24]. Therefore, the negative effects on the climate system and crop yields due to the indirect radiative forcing of ozone in China cannot be ignored. Both NO_x and NH_3 can enhance the formation of light-scattering aerosols (E3). Compared with the NO_x effect on the ozone-CH_4 effect (−350 ± 166 and −13.2 ± 7.2 Tg CO_2e on a GTP_{20} and a GTP_{100} basis, respectively), the direct aerosol effect of NO_x is much less, with only −97 ± 39 and −0.006 ± 0.004 Tg CO_2e on a GTP_{20} and a GTP_{100} basis, respectively; while the corresponding NH_3 effects on aerosol are −59 ± 23 and −0.11 ± 0.13 Tg CO_2e, respectively. As the time horizon is extended, the direct aerosol effects of NO_x and NH_3 both become smaller, and can be effectively ignored on a GTP_{100} basis (Fig. 2, Table S7).

Nr enrichment effects on net C exchange in the terrestrial ecosystem. The inputs of Nr into the terrestrial ecosystem are in the forms of N deposition and N fertilization. N addition into ecosystems can alter the physiology of soil microbes and vegetation, and concomitantly alter the balance of biogenic fluxes of GHGs[25]. Our literature analysis results provide an explicit spatial map of N deposition over China (Fig. 3a). N deposition sums to 15.1 Tg N/yr across China, with 3.56, 5.28, 5.68 and 0.57 Tg N deposited into cropland, forest, grassland and wetland, respectively (Fig. 3b–e). The estimated N deposition is closed to the published inventories of NO_x and NH_3 emissions used in our study. Without considering the response of N_2O emissions mentioned above, N addition can stimulate terrestrial CO_2 assimilation in both

Figure 1 | Emission inventory of gaseous Nr. Uncertainty bars represent the min and max ranges of Nr compounds. The data are collected from a variety of inventory models, including IIASA GAINS, EDGAR v4.2, RCPs, and MACCity.
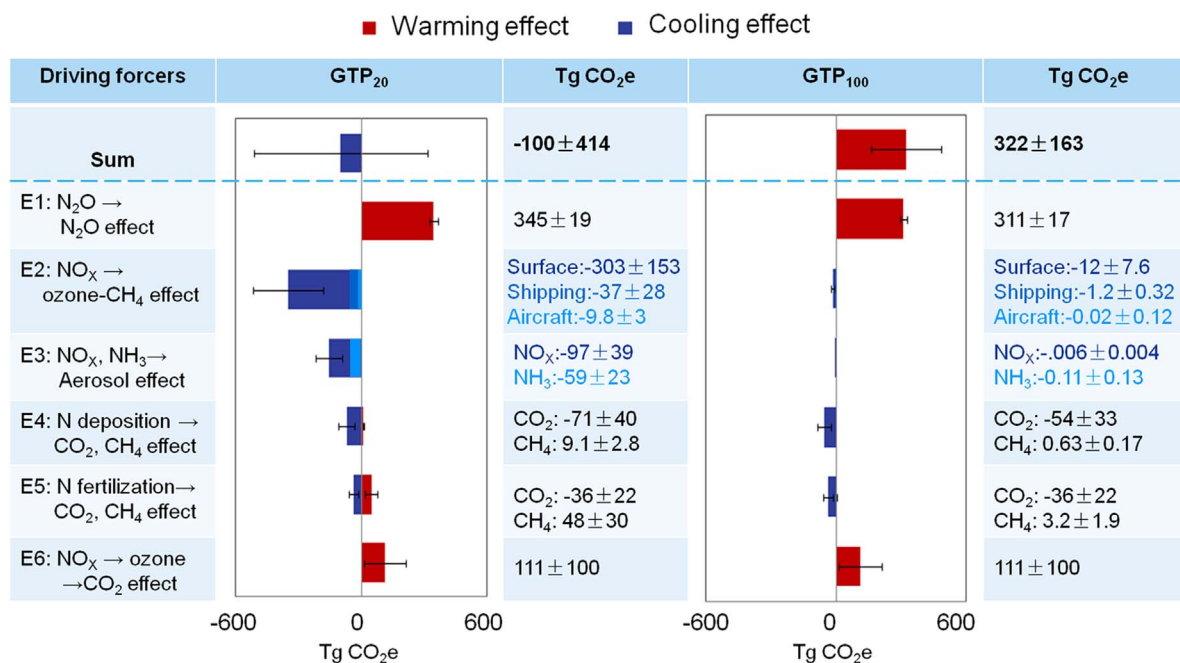
Figure 2 | Climate change impact of Nr in China. The warming effect is represented in red color, while the cooling effect is represented in blue color. Uncertainty bars represent the variation ranges of climate effects on a GTP_{20} and GTP_{100} basis.

natural vegetation and agricultural land; meanwhile, CH_4 emissions caused by N simulation to some extent offset the benefit of N-induced CO_2 uptake in terms of climate change[25]. These opposed impacts of Nr enrichment differ markedly depending on the land cover type[25,26]. The assessed N-induced CO_2 and CH_4 emission/uptake factors for different ecosystem types of China are summarized in Table S6. Our results show that in China forest land has the largest potential for CO_2 uptake (-27 to -5 kg CO_2-C/kg N), while cropland has the largest potential for CH_4 emissions (0.01 to 0.03 kg CH_4-C/kg N). Applying the flux factors of CO_2 uptake and CH_4 emission by ecosystem types in China from Table S6 to estimate the net C exchange (NCE) due to N deposition, the overall result indicates that the uptake of CO_2-C (27 to 176 Tg C) overwhelmingly surpasses the CH_4-C emissions (0.11 to 0.21 Tg C), resulting in an NCE of 27 to 176 Tg in terrestrial ecosystems during 2000–2005 (Table S6). Most regions in China's terrestrial system function as a net C sink; only a minor area in western China is an N-induced C source (Fig. 3f). Our results further show that, during 2000–2005, forest land has the largest C sequestration potential, with 31.9 to 136 Tg C as a result of N deposition, followed by grassland (4.5 to 45 Tg N) and cropland (1.8 to 5.6 Tg N). For fertilized cropland, in addition to N deposition, applied N, including chemical and organic fertilizers, was evaluated at 35.7 Tg in 2005[8], resulting in a net CO_2 uptake of 14 to 58 Tg C and CH_4 emissions of 0.40 to 1.06 Tg C. The NCE of croplands due to N fertilization amounts to 13.6 to 57.0 Tg C.

On the whole, N addition has a positive effect on terrestrial C sequestration over the past several years. The C sequestration simulated by both N deposition and fertilization exerts a cooling effect, -50 ± 86 and -86 ± 51 Tg CO_2e on a GTP_{20} and a GTP_{100} basis, respectively (Table S7). The climate cooling effects of N-induced C sequestration, however, offset only 18% and 35% of the warming effects of N_2O emissions from agriculture in terms of GTP_{20} and GTP_{100}, respectively. Basically, taking the effects of these three GHG fluxes between air and land into account, Nr enrichment of the terrestrial ecosystem can be recognized as a strong warming agency.

Integrated climate change impact of Nr in China. On the national scale, anthropogenic Nr in China's ecosystem has a cooling effect on a GTP_{20} basis in theory (-100 ± 414 Tg CO_2e) and a warming effect (322 ± 163 Tg CO_2e) on a GTP_{100} basis (Fig. 2, Table S7). However, the uncertainty of GTP_{20} is so large that it is hard to conclude whether Nr causes cooling or warming in practice. The uncertainty is largely caused by the climate effects of atmospheric Nr. The collective climate-change impact of gaseous Nr emissions is cooling (in theory) at -50 ± 328 Tg CO_2e on a GTP_{20} basis but warming at 408 ± 112 Tg CO_2e on a GTP_{100} basis, respectively, while the climate-change impact of Nr addition into the terrestrial ecosystem is cooling at -50 ± 86 and -86 ± 51 Tg CO_2e, respectively. The climate forcers of these two components offset each other through the Nr flows and exchanges between the atmosphere and the land.

The relative contributions of agriculture and industry sectors are illustrated in Fig. 4. The overall impact of industrial Nr is cooling (-287 ± 306 Tg CO_2e) on a GTP_{20} basis but converts to warming (136 ± 107 Tg CO_2e) on a GTP_{100} basis. The reason is that the cooling effects are largely from the short-lived agency, i.e., NO_x emissions from fuel combustion. The effect of agriculture Nr is dominated by the long-lived effect of N_2O emissions. As a result, agriculture shows a net warming effect and a minor difference at two timescales, at 187 ± 108 Tg CO_2e on a GTP_{20} basis and 186 ± 56 Tg CO_2e on a GTP_{100} basis, respectively. The climate effects of Nr from agricultural and industrial sources cancel each other out on a GTP_{20} basis, but together contribute to warming effect on a GTP_{100} basis.

Projection of Nr impact on climate. Under the SRES I-Business as Usual (BAU) scenario, we assume that Nr emissions and Nr addition will keep the current annual rates of increase. In 2020 and 2050, Nr in China will produce a cooling effect at -314 and -725 Tg CO_2e on a GTP_{20} basis and a warming effect at 528 and 939 Tg CO_2e on a GTP_{100} basis (Fig. 5 and Fig. 6, Table S8). The results indicate that the increase of Nr compounds without any measures will mitigate the climate change on a 20-y basis however aggravate the warming effect on a 100-y basis. On a 100-y basis, the cooling effect of N-induced C sequestration declines significantly, from 86.0 Tg CO_2e in 2005 to 57.3 Tg CO_2e in 2020, then approaches zero in 2050. By contrast, the impact of Nr emissions on climate change becomes more important, increasing from 408 Tg CO_2e in 2005 to 585 Tg CO_2e in 2020 and 939 Tg CO_2e in 2050. The SRES II-improvement scenario is built on
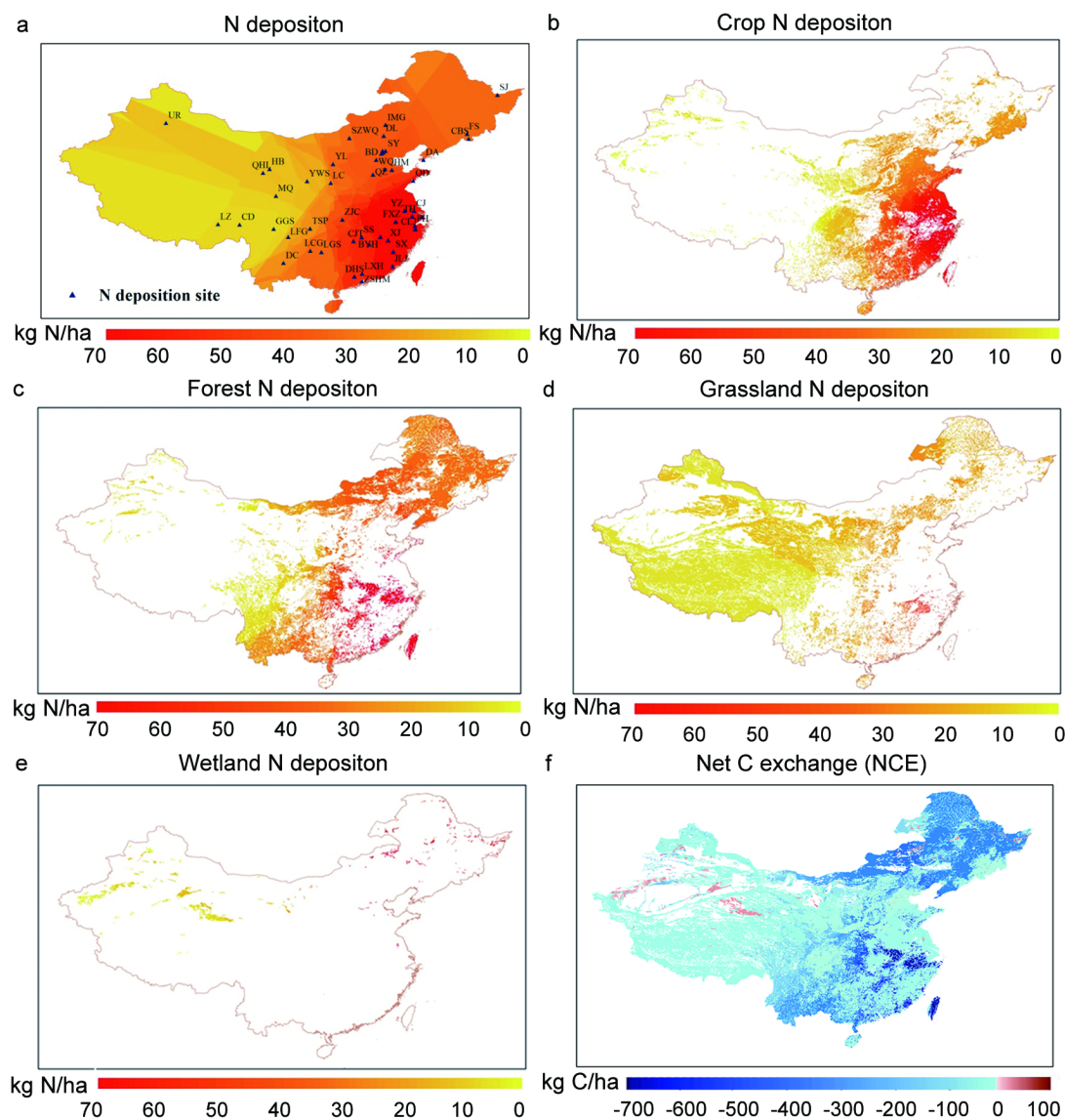
Figure 3 | N deposition into terrestrial ecosystems and net C exchange due to N deposition. (a), Spatial distribution of N deposition rate in China. (b), N deposition into cropland ecosystem. (c), N deposition into forest ecosystem. (d), N deposition into grass land ecosystem. (e), N deposition into wetland ecosystem. (f), Net C exchange (NCE) due to N deposition. The blue color represents C uptake; the red color represents C emission. The spatial maps of N deposition rate around China were developed from the spatial interpolation technique through the application of ARCGIS 10.

the assumption that the N use efficiency of cropland will increase by 20% in 2020 and another 20% in 2050. Compared to the BAU scenario, on a GTP_{20} basis, the improvement scenario enhances the net cooling effect with 41% and 38% for the years of 2020 and 2050, largely due to the agricultural N_2O emissions reduction. The SRES III-abatement scenario is built on the assumption of total quantity control for NO_x in 2020 and an improved NO_x removal rate in 2050, respectively. On a GTP_{20} basis, the abatement scenario reduces the net cooling effect by 95% and 139% compared to the BAU scenario, ultimately appearing as a net warming effect in 2050. The reduction in NO_x emissions has a strong negative feedback on climate change in the short term, whereas the increase in N use efficiency represents a positive effect on the mitigation of climate warming. Improving agricultural N use efficiency (i.e. N_2O reduction) is the most timely and effective measure for the mitigation of the climate impact of Nr in China. On a GTP_{100} basis, the improvement and abatement scenarios both reduce the warming effect by 28% and 14% in 2020, respectively; the reduction effects transfer to 35% and 31% in 2050. Hence, reductions in agricultural N_2O and industrial NO_x emissions have a comparatively large mitigation potential in the long term.

The detailed comparisons between SRES and RCP scenarios are showed in Fig. 5 and Fig 6. All RCP scenarios on a GTP_{20} basis show a net warming effect, in contrary to our improvement scenario. However, the advantage of our improvement scenario diminishes with time. On a GTP_{100} basis, its warming effect in 2020 (379.4 Tg CO_2e) nearly approaches that of the lowest RCP scenario (RCP2.6, 356.4 Tg CO_2e), or even in 2050 (613.6 Tg CO_2e) surpasses that of the highest RCP scenario (RCP8.5, 499.6 Tg CO_2e). The warming effect of our abatement scenario on a GTP_{20} basis is lower than that of the RCP8.5 in 2020 and 2050. However, on a GTP_{100} basis, its warming effect in 2020 (452.3 Tg CO_2e) and in 2050 (646.1 Tg CO_2e) become higher than that of RCP8.5 (423.5 and 499.6 Tg CO_2e in 2020 and 2050, respectively). The comparison results between SRES and RCP indicate that, (i) our reported SRES scenarios reveal the obvious mitigation potential on climate change but only on a short timescale not on a long timescale. (ii) the objectives of our scenarios in 2020 (i.e. realizing the total quantity control for NO_x and improving the N use efficiency with 20%) is well-targeted to the concentration levels of RCP. However, the objectives in 2050 are too cautious to meet the emission under
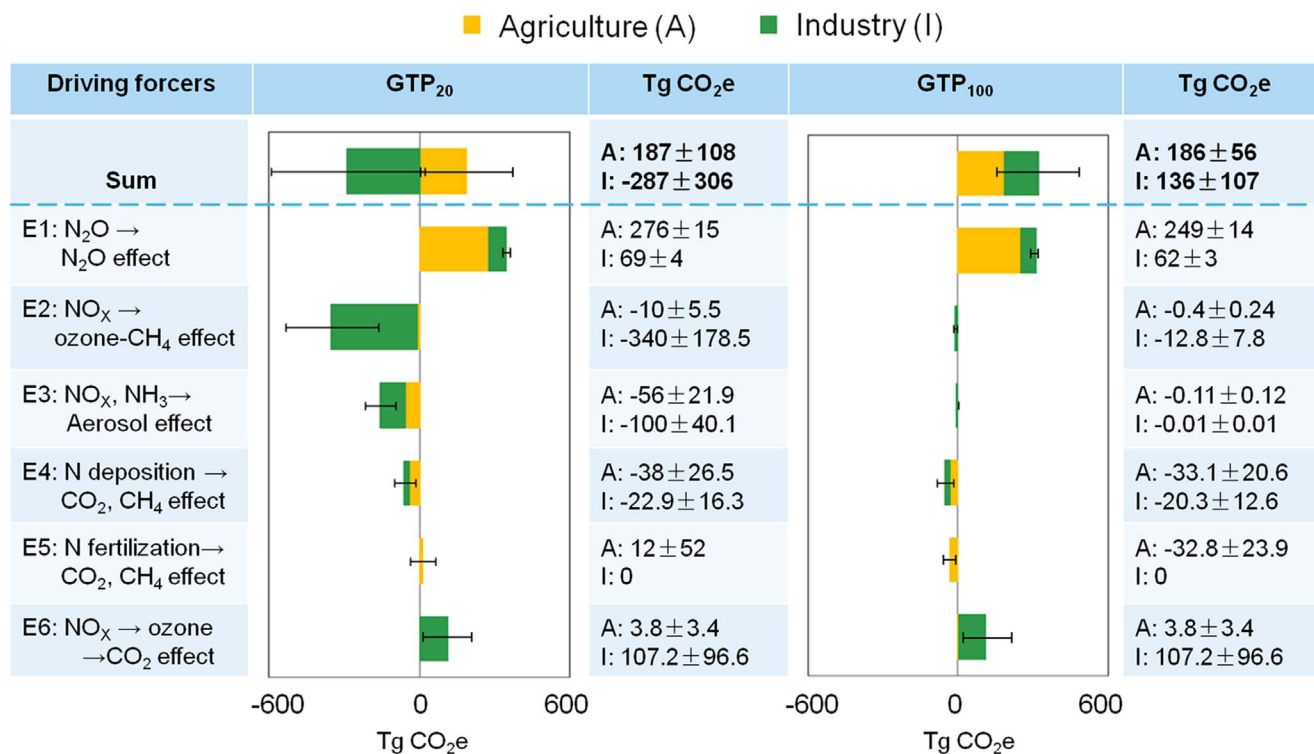
Figure 4 | Relative contribution to climate change of Nr from agriculture and industry. The effect of agriculture is represented in yellow color, while the effect of industry is represented in green color. Uncertainty bars represent the variation ranges of climate effects on a GTP_{20} and GTP_{100} basis.

the RCP scenarios. More aggressive target should be made for the year of 2050.

Discussion

China is mobilizing the largest anthropogenic Nr in the world due to agricultural, industrial and urban development. The climate effects of different Nr compounds are experiencing either the aggravation or the transformation processes. Based on the understanding of the dynamic of Nr on climate system, the investigation of possible changes and future trends is more meaningful than current status. Therefore, in this section, we analyze the dynamics and development trajectory of each climate forcer related to Chinese Nr, figuring out the key different points of China from other regions. Here, the differences between the SRES and BAU scenarios are defined as mitigation potential; while the differences between the SRES and RCP scenarios (here using RCP6.0) are defined as mitigation pressure. We further discuss the mitigation potential and mitigation pressure of individual N-related climate forcers (Fig. 5).

The strong GTP and long lifetime of N_2O makes it the largest and most lasting contributor to global warming in China. Agriculture is responsible for about 80% of the climate warming effect of N_2O. The high N fertilizer input and low N use efficiency in China's agricultural fields are responsible for the increasing N_2O emissions[27]. Our scenario analysis shows that a larger mitigation potential for climate change could be realized through improving N use efficiency to reduce agricultural N_2O emissions. Many approaches can be taken to enhance N use efficiency, for example, systematic crop rotation, wise use of fertilizer, and expanded use of nitrification inhibitors[28]. Although the agricultural activities are the largest contributor, emission of N_2O from chemical industries also shows a large increase potential; and the control options are the most-effective in this sector[10]. Hence, the all-around measures to reduce N_2O emission from agriculture and industry could lead to the co-benefits of climate mitigation and positive environmental effect through protecting ozone layer.

The realistic trajectory of NO_x reflects a compilation of environmental policies and economic activities[29]. NO_x emissions in many industrialized countries are expected to decrease[30]. For example, the NO_x reductions in Europe in 2010 can be partly attributed to the global economic recession, and partly to the European NO_x emission controls[29]. Due to rapid economic development and the coal-dominated energy consumption structure, however, large increases in NO_x from China have persisted over the past several decades[31]. According to our results, the ozone-CH_4 effect of NO_x emissions in China is the largest cooling agency at a short time horizon, but is nearly negligible at a longer time horizon. The ozone-CH_4 effect varies greatly across different regions[32-34]. Investigations have shown that the ozone-CH_4 effect is more sensitive to ozone precursor changes from tropical regions (e.g. East Asia) than from mid-latitude and high-latitude regions (e.g. Europe and North America)[35]. Therefore, control strategies that reduce emissions of ozone precursors in China have a significant impact on the net climate forces from ozone and CH_4. However, if only considering NO_x alone, NO_x emissions exert a negative force from decreased CH_4, which far exceeds the smaller positive force from increased ozone, resulting in a net cooling effect. The scenario of NO_x emissions reduction alone lead to a positive force and a net warming effect from overwhelming increased CH_4, and as a result fails to produce mitigation potential for climate warming. Combined reductions in NO_x and other ozone precursor emissions (such as CO) are beneficial, because they can produce a net cooling effect through the stronger negative force from decreased ozone[34]. Hence, in order to obtain co-benefits from reducing radiative forces and mitigating air pollution, simultaneous emission reductions of various ozone precursors at the regional scale are feasible. In addition, the indirect radiative force on climate change by ozone effects on plant production has proved to contribute more to global warming than the direct radiative force of elevated ozone levels[36]. The effect of NO_x on plants is the secondary contributor to climate warming at both timescales. In China, the combined climate effects of current NO_x
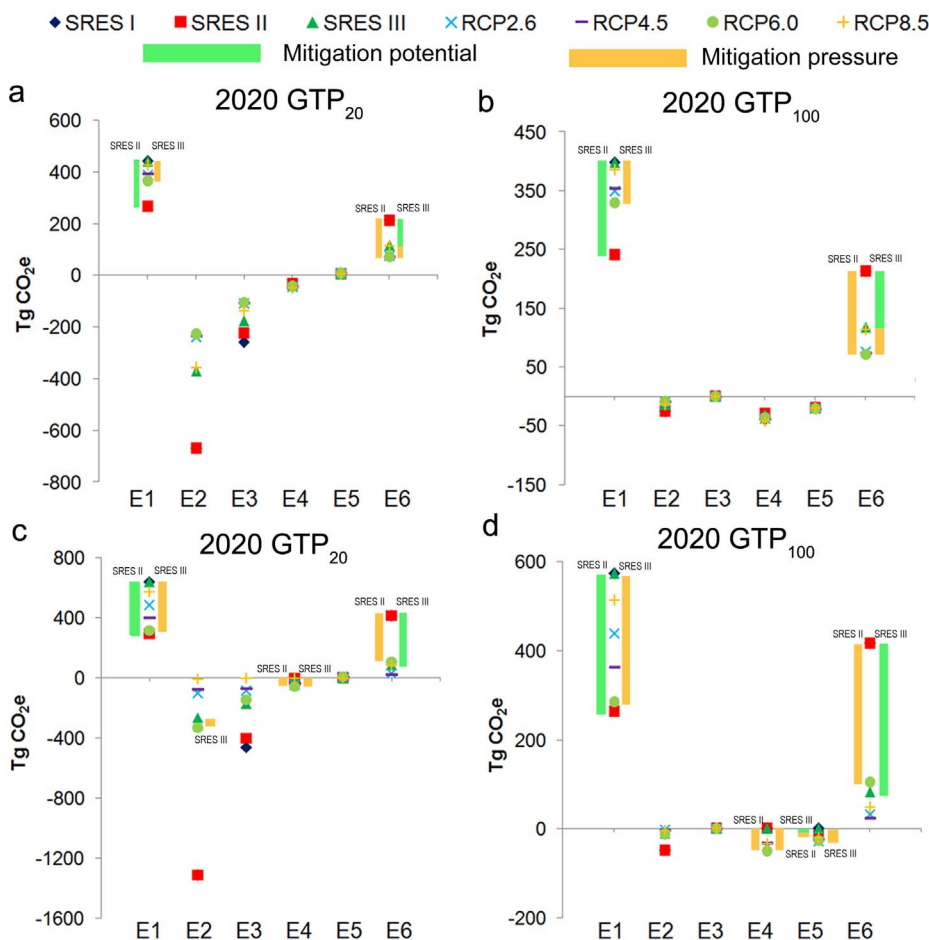
Figure 5 | Scenario analysis on climate forcers related to Nr in China, on a GTP_{20} and GTP_{100} basis for the year of 2020 and 2050. E1: $N_2O \rightarrow N_2O$ effect; E2: $NO_x \rightarrow$ ozone-CH_4 effect; E3: NO_x, $NH_3 \rightarrow$ Aerosol effect; E4: N deposition $\rightarrow CO_2$, CH_4 effect; E5: N fertilization $\rightarrow CO_2$, CH_4 effect; E6: $NO_x \rightarrow$ ozone $\rightarrow CO_2$ effect. The differences between SRES II, SRES III and BAU scenario are defined as mitigation potentials, with green column; while the difference between SRES II, SRES III and RCP scenario are defined as mitigation pressures, with orange column. The detailed mitigation potentials and pressures of SRES II and SRES III are represented on the left and right of the points, respectively.

emissions on ozone-CH_4, aerosol and the ozone-induced C sink were estimated at -336 ± 290 and 98 ± 97 Tg CO_2e on a GTP_{20} and a GTP_{100} basis, respectively. From the results of our scenario analysis, the NO_x reduction has a secondary mitigation potential. Therefore, reduction of NO_x in China can provide the co-benefits

of climate-change mitigation and air pollution reduction in the long run. In especial, the NO_x emission reduction still exist a large mitigation pressure when comparing with the RCP scenarios. China should confirm the implementation of total NO_x quantity control required by taking policy measures. Meanwhile, continued
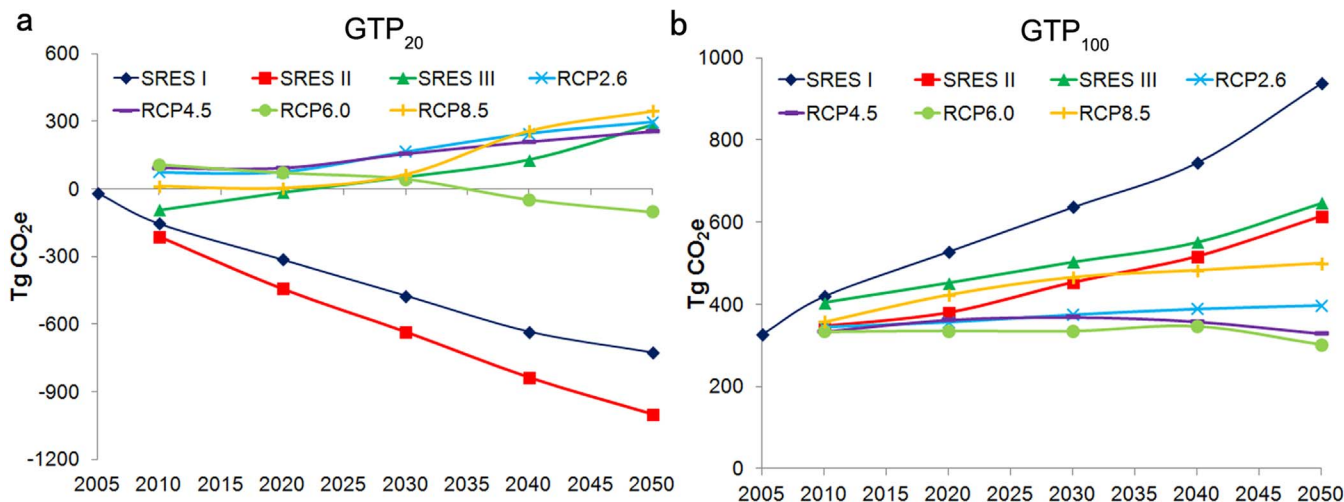

Figure 6 | Climate change impact of Nr in China between 2005–2050. (a), climate effect on a GTP_{20} basis. (b), climate effect on a GTP_{100} basis. The values in 2010, 2030 and 2040 are estimated on the basis of the logistic growth model.

push to improve the denitration level is an essential measure for mitigating NO_x pollution.

China is a large agricultural country with an enormous animal population and tremendous N fertilizer consumption. The characteristics of NH_3 emissions reflect the intensity of synthetic fertilizer use and unique patterns of animal densities and types in China[37]. As the human population continues to demonstrate considerable growth, food production and associated NH_3 emissions are likely to increase as well[38]. Increased NO_x and NH_3 produce radiative effects resulting from sulfate-nitrate aerosol and single-nitrate aerosol, respectively. The cooling effect of NO_x through the direct aerosol effect is larger than NH_3 on a 20-y basis, but its depletion rate with time is faster than the rate of NH_3. Global model studies demonstrated that the increasing nitrate aerosol force would counteract the expected decline in the sulfate force by 2010[39]. The future nitrate aerosol loads are projected to depend strongly on the emission strength of NH_3[40]. It can be deduced that a larger increase in nitrate aerosols in China will be accompanied by larger increases of NH_3, as opposed to the smaller increases in Europe, the U.S. and Australia[40]. Although the cooling effect of nitrate aerosols is negligible in the long term, the future air pollution from nitrate in the atmosphere can not be ignored.

Overall, the net cooling effects of NO_x and NH_3 are short-lived and NO_x even plays a warming role on the long timescale. Nonetheless, the multiple environmental problems resulting from NO_x and NH_3, such as air pollution, eutrophication, etc., are more remarkable than the climate impact in China. In this regard, it is important to assess the trade-offs between the environmental and climate effects of the Nr compounds in the future studies.

China—especially the east region—is one of the global heavy N-deposition areas[41]. Under the action of N deposition, our results show forest land in China is the single largest C sink, followed by grassland and cropland. The N status of different ecosystems in China can be reflected and further analyzed from the flux factors of CO_2 and CH_4 in Table S6. For example, N is usually found to be a limiting factor in plant growth in grassland ecosystems in China, and it is mostly unintended fertilization that has stimulated plant growth there[42]; while almost all croplands can be regarded as N-saturated ecosystems due to continual application of chemical N fertilizers[43], as a results, the N-induced flux factor of CO_2 is only 0.45–1.7 kg CO_2/kg N. Additionally, there have been very different response patterns of forest ecosystems in China depending on the forest type, the N status of the soil, and the level of N deposition. In N-saturated mature tropical forests, high N addition can reduce soil respiration and CO_2 emissions[44], and decrease CH_4 uptake rate[45]. However, in N-limited pine forests, elevated N deposition has no significant effect on soil respiration or CH_4 uptake[45,46]. Overall, the N-induced flux factor of CO_2 in forest lands in China is 5–27 kg CO_2/kg N, much lower than that of the United States, with 24–65 kg CO_2/kg N[7]. Throughout the whole of China, the terrestrial system acts as a net C sink due to N deposition. However, the potential for sustaining more terrestrial C storage as a result of atmospheric N deposition is limited[47]. As the N deposition increases, the net primary productivity induced by per-unit N deposition has leveled off or declined since the 1980s, indicating that part of the deposited N may not be invested to stimulate plant growth, but instead leave the ecosystem by various pathways[18,48]. In addition, globally N fertilizer application-induced CO_2 uptake began to level off in the 1980s whereas N_2O emission continued to grow[49]. As a result, the climate benefits of the enhanced C sequestration would likely be eliminated due to increased N use[50]. Finally, fertilized N will show an increasing warming effect in the future. This trend is more obvious in China. Excessive N-fertilization limits C sequestration. The declining rate of manure recycling to fields causes more N discharge into water or emitted into the air[51–54].

N saturation in forest land and excessive N fertilization in cropland in China limit the C sequestration capacity. The effect of N enrichment on C sequestration, as a result, has appeared to diminish in recent years. Hence, through the results of our scenario analysis, there is a certain GHG mitigation pressure in C sequestration through improving N management or reduction gaseous Nr emission in China. Efficient forest development and improved field management have shown the large potential of C sequestration in China's forest and cropland[23,55,56]. Rational N fertilizer application, no-tillage and preferential crop residues management are widely recognized measures for enhancing C sequestration[52,56]. Climate and environmental quality trade-offs and co-benefits, especially if they are applied nationwide in the near term, provide strong additional motivation for reducing N fertilizer use without yield loss in China.

Overall, anthropogenic Nr in China's environmental system has an obvious warming effect on a long timescale. The reductions in agricultural N_2O and industrial NO_x emissions have a comparatively great mitigation potential for climate change in the long term; while a certain mitigation pressure appears in N-induced C sequestration only if realizing the efficient forest development and improved field management. The mitigation pressure of climate warming of anthropogenic Nr is still under great pressure with time. In 2050, more aggressive targets and more powerful measures should be taken to maintain in a specific radiative forcing level.

Methods

Climate forcers related to Nr. Nr can exert a series of effects on GHG fluxes and on climate systems during its cascade flow. We needed to distinguish these "direct" biogeochemical consequences (i.e., radiative forcing) of atmospheric Nr emission from the "indirect" bio-geophysical climate feedbacks of terrestrial N dynamics through terrestrial GHG fluxes[47]. We considered six climate-driving forcers of Nr (designated as E1–E6 below) and show detailed interpretations in the Supplementary Information (Fig. S1). The common metric GTP is used to reconcile the climate change impacts of diverse climate forcers into the common unit of equivalent CO_2 (CO_2e) (Table S1) for the inter-comparison of the relative contributions over 20-y and 100-y timescales, respectively.

The climate forcers E1–E3 refer to climate change impacts through the effects of the atmospheric Nr species (including N_2O, NO_x, and NH_3) on trapping heat or scattering solar energy. So far, there has been no official national inventory of gaseous Nr species for China. The results from many published studies show a certain inconsistency due to the different approaches used. Rather than calculating the Nr emissions inventory, we compiled Nr emissions from the existing regional and global emissions inventories associated with China, including IIASA GAINS, EDGAR v4, RCPs, and MACCity. The data covered total volumes and sectoral structures of Nr emissions within the period 2000–2005 (Table S2).We defined the uncertainty (i.e., standard deviation) using the variation ranges of all these inventories. The estimated results are proved to be comparable with the published local studies (Table S3).

The climate forcers E4–E6 refer to climate-change impacts through the feedback of the atmospheric NO_x and terrestrial Nr enrichment (N deposition and N fertilization) on GHG's biogenic fluxes. We collected the monitoring data of N deposition rates from different sites around China mainly covering the period 2000–2005 from the extant studies (Table S4). Through the application of ARCGIS 10, an explicit spatial map of N deposition rate around China was developed using the spatial interpolation technique (kriging). The deposition to each land cover type is obtained by overlapping with the nationwide land-use distribution map. In addition, the fertilization of cropland was adopted from the results of Cui et al.[8]. Because of the greater heterogeneity of global terrestrial ecosystems, we reviewed and aggregated the data for N-induced GHG emission/uptake of China (Table S5). N-induced GHG emission/uptake factors were estimated for different ecosystem types (Table S6). In this section, we only accounted for the change in CO_2 and CH_4 fluxes, except for N_2O, which has been included in the previous atmospheric Nr emission inventory.

Uncertainty analysis. The quantification of the significances of climate forcers E1–E6 were calculated normally through multiplying the effective Nr volumes by the corresponding metric GTP, so as to reconcile and inter-compare the diverse climate forcers. We used the Monte Carlo Sampling (MCS) and Latin Hypercube Sampling (LHS) methods to simulate and propagate the uncertainties through the calculations, respectively (Table S7). These methods are based on the principle that the empirical process of actually drawing many random samples and observing the outcomes can assess the behavior of a statistic in random samples. The uncertainties of parameters, activity data, and formulas are propagated through the sequence of calculations. In the MCS approach, we assumed each value in the uncertainty range had equal likelihood and no correlation between the variables. A population of 10,000 estimates was built and ranked in order. The median and 90% confidence intervals were reported. The LHS approach was further used to calculate the uncertainties in our research. All variables were divided into 300 equal intervals. An ensemble of 300 sets of all parameters was selected randomly and ranked in order. The mean and standard

deviation were reported. The results of the uncertainty analysis using MCS and LHS were similar. Hence, we only discuss the results of the LHS method here.

Scenario analysis. Based on the possible or expected changes of different driving forcers, we set up a new set of three scenarios to depict the future trend of climate change (Table S8). Short-term projections (to 2020) were developed using a "baseline" path of development, which considered abatement options consistent with China's situation and policy. For long-term projections (to 2050), we applied a "storyline" approach similar to that used in the IPCC Special Report on Emission Scenarios (SRES) scenarios[21] . The situation in 2005 was used as the reference and those in 2020 and 2050 as the target years. (i) SRES I–Business as Usual (BAU) scenario is based on the assumption that Nr emissions will maintain the annual increase rate of 2000–2005 with no change. (ii) SRES II–Improvement scenario, builds on the BAU scenario and assumes that the N use efficiency of cropland will increase with an aspirational target of 20% until 2020[57] and a further 20% until 2050, which approaches the level of developed countries[27]. (iii) SRES III-Abatement scenario, is designed to cut 10% of NO_x emissions in accordance with the reduction target of the "Twelfth Five-Year Plan" in 2020, and assumed that the nationwide NO_x removal rate reaches 80% in 2050. Because of the limited C storage of terrestrial ecosystems, the N-induced C sequestration potential is projected to decline 50% and be negligible with the increased N use in 2020 and 2050. The values in 2010, 2030 and 2040 are calculated on the basis of the logistic growth model. In addition, we compared our SRES scenarios with the published Representative Concentration Pathway (RCP), with four scenarios based on different concentration levels (RCP2.6, RCP4.5, RCP6.0 and RCP8.5, with the numbers referring to different radiative forcing levels) rather than emissions[20].

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Author contributions

Y.S., S.C. and X.J. contributed equally to this work. S.C., Y.-G.Z. and Y.S. conceived of the study. Y.S. and S.C. prepared meteorological inputs. Y.S., S.C. and X.J. performed research and analyzed data; S.C., Y.S., X.J., Z.C. and Y.-G.Z. wrote the paper. All authors gave comments on the manuscript.

Additional information

Supplementary information accompanies this paper at http://www.nature.com/scientificreports

Competing financial interests: The authors declare no competing financial interests.

How to cite this article: Shi, Y., Cui, S., Ju, X., Cai, Z. & Zhu, Y.-G. Impacts of reactive nitrogen on climate change in China. *Sci. Rep.* **5**, 8118; DOI:10.1038/srep08118 (2015).