Impacts of reactive nitrogen on climate change in China
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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 ± 163 Tg CO2e on a GTP20 and a GTP100 basis, respectively. Agriculture contributes to warming at 187 ± 108 and 186 ± 56 Tg CO2e on a 20-y and 100-y basis, respectively, dominated by long-lived nitrous oxide (N2O) from fertilized soils. On a 20-y basis, industry contributes to cooling at 287 ± 306 Tg CO2e, largely owing to emissions of nitrogen oxides (NOx) altering tropospheric ozone, methane and aerosol concentrations. However, these effects are short-lived. The effect of industry converts to warming at 136 ± 107 Tg CO2e on a 100-y basis, mainly as a result of the reduced carbon (C) sink from the NOx-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 N2O and industrial NOx 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 (NH4-N in water and NH3 in air), nitrogen oxides (NOx, nitrous oxide (N2O), and nitrate & nitrite) in circulation, and caused severe damage to the environment at local, regional and global scales. 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. The atmospheric N2O emission directly contributes to global warming and ozone depletion; NOx 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 (CH4)); NH3 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. 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. For an equivalent increase in the human population, the emission mass of CO2-C into the environment is about 40 times that of Nr. Although the absolute climate impact of Nr is dwarfed by the impact of CO2 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 Nr management.

N2O, as one of the GHGs, has gained worldwide attention, and has also been thoroughly assessed in China. It is well known that reducing N2O emissions offers the combined benefits of mitigating climate change and protecting the ozone layer. The increasing demands for food and energy in China have led to ever-increasing emissions of NOx and NH3. These short-lived Nr emissions have multiple and profound effects on climate, the environment and human health. 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, as well as many other ecosystem services associated with environmental quality. The terrestrial ecosystem C balance in China was...
investigated to under the influence of multiple global change factors\textsuperscript{16,17}. The effects of N deposition and N fertilizer use have been identified in a few studies\textsuperscript{18,19}.

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\textsubscript{2}O, but also the near-term climate fluctuations caused by emissions of the short-lived species NO\textsubscript{x} and NH\textsubscript{3}\textsuperscript{17,19}. Accordingly, we use GTP to convert the climate change impacts of each of the processes to common units of equivalent Tg CO\textsubscript{2} (CO\textsubscript{2}e).

In this analysis, we combine the set of Representative Concentration Pathway (RCP) scenarios, based on consistent radiative forcing levels\textsuperscript{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\textsuperscript{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\textsubscript{2}O, NO\textsubscript{x} and NH\textsubscript{3}, respectively (Fig. 1). The emission sectors of Nr also show large variations. N\textsubscript{2}O is the only form of Nr that has unambiguous global implications through the strong warming effect on climate (E1). Total anthropogenic N\textsubscript{2}O emissions in China for 2005 were estimated at 1.04–1.22 Tg N. The primary source of N\textsubscript{2}O is agriculture. N\textsubscript{2}O 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\textsubscript{2}O 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\textsuperscript{22,23}.

In addition, N\textsubscript{2}O 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\textsubscript{2}O exerts a strong warming effect at 345 ± 19 Tg CO\textsubscript{2}e on a GTP\textsubscript{20} basis, then transforms to 311 ± 17 Tg CO\textsubscript{2}e on a GTP\textsubscript{100} basis (Fig. 2, Table S7). The close climate impacts on both timescales imply lasting effects on the climate system resulting from N\textsubscript{2}O due to its long atmospheric lifetime of around 120 years. NO\textsubscript{x} and NH\textsubscript{3} contribute to climate change indirectly through altering the atmospheric constituents and corresponding climate forcers. NO\textsubscript{x} is a major byproduct of combustion. Fuel combustion in China resulted in a total of 4.04–6.08 Tg N of NO\textsubscript{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\textsubscript{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\textsubscript{x}. In comparison, the total emissions of NH\textsubscript{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\textsubscript{x} on ozone and CH\textsubscript{4} (i.e. ozone-CH\textsubscript{4} effect, E2) in China is −350 ± 166 and −13.2 ± 7.2 Tg CO\textsubscript{2}e on a 20-y and 100-y basis, respectively (Fig. 2, Table S7). Our results show that the net cooling effect of NO\textsubscript{x} on ozone and CH\textsubscript{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)\textsuperscript{41}. 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\textsubscript{2}e under controlled simulations for different scenarios\textsuperscript{42}. 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\textsubscript{x} and NH\textsubscript{3} can enhance the formation of light-scattering aerosols (E3). Compared with the NO\textsubscript{x} effect on the ozone-CH\textsubscript{4} effect (−350 ± 166 and −13.2 ± 7.2 Tg CO\textsubscript{2}e on a GTP\textsubscript{20} and a GTP\textsubscript{100} basis, respectively), the direct aerosol effect of NO\textsubscript{x} is much less, with only −97 ± 39 and −0.006 ± 0.004 Tg CO\textsubscript{2}e on a GTP\textsubscript{20} and a GTP\textsubscript{100} basis, respectively; while the corresponding NH\textsubscript{3} effects on aerosol are −59 ± 23 and −0.11 ± 0.13 Tg CO\textsubscript{2}e, respectively. As the time horizon is extended, the direct aerosol effects of NO\textsubscript{x} and NH\textsubscript{3} both become smaller, and can be effectively ignored on a GTP\textsubscript{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\textsuperscript{43}. 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\textsubscript{x} and NH\textsubscript{3} emissions used in our study. Without considering the response of N\textsubscript{2}O emissions mentioned above, N addition can stimulate terrestrial CO\textsubscript{2} assimilation in both
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$^{25}$. 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$^5$, 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$_2$e 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$_2$O 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, N 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$_2$e) and a warming effect (322 ± 163 Tg CO$_2$e) 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$_2$e on a GTP$_{20}$ basis but warming at 408 ± 112 Tg CO$_2$e 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$_2$e, 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$_2$e) on a GTP$_{20}$ basis but converts to warming (136 ± 107 Tg CO$_2$e) on a GTP$_{100}$ basis. The reason is that the cooling effects are largely from the short-lived agriculture, i.e., NO$_x$ emissions from fuel combustion. The effect of agriculture Nr is dominated by the long-lived effect of N$_2$O emissions. As a result, agriculture shows a net warming effect and a minor difference at two timescales, at 187 ± 108 Tg CO$_2$e on a GTP$_{20}$ basis and 186 ± 56 Tg CO$_2$e 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$_2$e on a GTP$_{20}$ basis and a warming effect at 528 and 939 Tg CO$_2$e 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$_2$e in 2005 to 57.3 Tg CO$_2$e 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$_2$e in 2005 to 585 Tg CO$_2$e in 2020 and 939 Tg CO$_2$e in 2050. The SRES II-improvement scenario is built on
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 GTP20 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 N2O emissions reduction. The SRES III-abatement scenario is built on the assumption of total quantity control for NOx in 2020 and an improved NOx removal rate in 2050, respectively. On a GTP20 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 NOx 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. N2O reduction) is the most timely and effective measure for the mitigation of the climate impact of Nr in China. On a GTP100 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 N2O and industrial NOx 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 GTP20 basis show a net warming effect, in contrary to our improvement scenario. However, the advantage of our improvement scenario diminishes with time. On a GTP100 basis, its warming effect in 2020 (379.4 Tg CO2-e) nearly approaches that of the lowest RCP scenario (RCP2.6, 356.4 Tg CO2-e), or even in 2050 (613.6 Tg CO2-e) surpasses that of the highest RCP scenario (RCP8.5, 499.6 Tg CO2-e). The warming effect of our abatement scenario on a GTP20 basis is lower than that of the RCP8.5 in 2020 and 2050. However, on a GTP100 basis, its warming effect in 2020 (452.3 Tg CO2-e) and in 2050 (646.1 Tg CO2-e) becomes higher than that of RCP8.5 (423.5 and 499.6 Tg CO2-e 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 NOx 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
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 N2O 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 N2O. The high N fertilizer input and low N use efficiency in China’s agricultural fields are responsible for the increasing N2O emissions. Our scenario analysis shows that a larger mitigation potential for climate change could be realized through improving N use efficiency to reduce agricultural N2O 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. Although the agricultural activities are the largest contributor, emission of N2O from chemical industries also shows a large increase potential; and the control options are the most-effective in this sector. Hence, the all-around measures to reduce N2O 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 NOx reflects a compilation of environmental policies and economic activities. NOx emissions in many industrialized countries are expected to decrease. For example, the NOx reductions in Europe in 2010 can be partly attributed to the global economic recession, and partly to the European NOx emission controls. Due to rapid economic development and the coal-dominated energy consumption structure, however, large increases in NOx from China have persisted over the past several decades. According to our results, the ozone-CH4 effect of NOx emissions in China is the largest cooling agency at a short time horizon, but is nearly negligible at a longer time horizon. The ozone-CH4 effect varies greatly across different regions. Investigations have shown that the ozone-CH4 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). Therefore, control strategies that reduce emissions of ozone precursors in China have a significant impact on the net climate forces from ozone and CH4. However, if only considering NOx alone, NOx emissions exert a negative force from decreased CH4, which far exceeds the smaller positive force from increased ozone, resulting in a net cooling effect. The scenario of NOx emissions reduction alone lead to a positive force and a net warming effect from overwhelming increased CH4, and as a result fails to produce mitigation potential for climate warming. Combined reductions in NOx 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 CH4. 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. The effect of NOx on plants is the secondary contributor to climate warming at both timescales. In China, the combined climate effects of current NOx

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 GTP20 and GTP100 basis.
emissions on ozone-CH₄, aerosol and the ozone-induced C sink were estimated at 336 ± 290 and 98 ± 97 Tg CO₂e on a GTP₂₀ and a GTP₁₀₀ basis, respectively. From the results of our scenario analysis, the NOₓ reduction has a secondary mitigation potential. Therefore, reduction of NOₓ in China can provide the co-benefits of climate-change mitigation and air pollution reduction in the long run. In especial, the NOₓ emission reduction still exist a large mitigation pressure when comparing with the RCP scenarios. China should confirm the implementation of total NOₓ quantity control required by taking policy measures. Meanwhile, continued

Figure 5 | Scenario analysis on climate forcers related to Nr in China, on a GTP₂₀ and GTP₁₀₀ basis for the year of 2020 and 2050. E1: N₂O → N₂O effect; E2: NOₓ → ozone-CH₄ effect; E3: NOₓ, NH₃ → Aerosol effect; E4: N deposition → CO₂, CH₄ effect; E5: N fertilization → CO₂, CH₄ effect; E6: NOₓ → ozone → CO₂ 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.

Figure 6 | Climate change impact of Nr in China between 2005–2050. (a), climate effect on a GTP₂₀ basis. (b), climate effect on a GTP₁₀₀ basis. The values in 2010, 2030 and 2040 are estimated on the basis of the logistic growth model.
push to improve the denitrification level is an essential measure for mitigating NOx pollution.

China is a large agricultural country with an enormous animal population and tremendous N fertilizer consumption. The characteristics of NH3 emissions reflect the intensity of synthetic fertilizer use and unique patterns of animal densities and types in China44. As the human population continues to demonstrate considerable growth, food production and associated NH4 emissions are likely to increase as well48. Increased NOx and NH3 produce radiative effects resulting from sulfate-nitrate aerosol and single-nitrate aerosol, respectively. The cooling effect of NO2 through the direct aerosol effect is larger than N2H on a 20- y basis, but its depletion rate with time is faster than the rate of NH3. Global model studies demonstrated that the increasing nitrate aerosol force would counteract the expected decline in the sulfate force by 201049. The future nitrate aerosol loads are projected to depend strongly on the emission strength of NH350. It can be deduced that a larger increase in nitrate aerosols in China will be accompanied by larger increases of NH3, as opposed to the smaller increases in Europe, the U.S. and Australia50. Although the cooling effect of nitrate aerosols is negligible in the long term, the future air pollution from nitrate in the atmosphere cannot be ignored.

Overall, the net cooling effects of NOx and NH3 are short-lived and NOx even plays a warming role on the long timescale. Nonetheless, the multiple environmental problems resulting from NOx and NH3, 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 areas49. 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 CO2 and CH4 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 there51; while almost all croplands can be regarded as N-saturated ecosystems due to continual application of chemical N fertilizers52. As a result, N-induced flux factor of CO2 is only 0.45–1.7 kg CO2/ 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 CO2 emissions53, and decrease CH4 uptake rate54. However, in N-limited pine forests, elevated N deposition has no significant effect on soil respiration or CH4 uptake55,56. Overall, the N-induced flux factor of CO2 in forest lands in China is 5–27 kg CO2/kg N, much lower than that of the United States, with 24–65 kg CO2/kg N. 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 limited57. 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 pathways58,59. In addition, globally N fertilizer application-induced CO2 uptake began to level off in the 1980s whereas N2O emission continued to grow49. As a result, the climate benefits of the enhanced C sequestration would likely be eliminated due to increased N use60. 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 air51,54.

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 cropland51,52,55,56. Rational Nr fertilizer application, no-tillage and preferential crop residues management are widely recognized measures for enhancing C sequestration52,56. 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 N2O and industrial NOx 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 fluxes42. 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 CO2 (CO2e) (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 N2O, NOx, and NH4) 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 from the extant studies (Table S4). Through the application of ARCGIS 10, an explicit spatial map of Nr 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 CO2 and CH4 fluxes, except for N2O, 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 stochastic 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 forces, 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 current situation and needs. (Long-term projections (to 2050), we applied a “storyline” approach similar to that used in the IPCC Special Report on Emission Scenarios (SRES) scenarios17. 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 N 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 use efficiency of cropland will increase with an aspirational target of 20% until 202020 and a further 20% until 2050, which approaches the level of developed countries17. (iii) SRES III-Abatement scenario, is designed to cut 10% of N emissions in accordance with the reduction target of the “Twelfth Five-Year Plan” in 2020, assumed that the nationwide N 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 emissions22.

1. Fowler, D. et al. The global nitrogen cycle in the twenty-first century. Philosophical Transactions of the Royal Society B: Biological Sciences 368, 2010164 (2013).
2. Galloway, J. N. et al. Transformation of the nitrogen cycle: Recent trends, questions, and potential solutions. Science 320, 889–892 (2008).
3. Erisman, J. W., Galloway, J., Seitzinger, S., Bleeker, A. & Butterbach-Bahl, K. Transformation of the nitrogen cycle: Recent trends, questions, and potential solutions. Science 320, 889–892 (2008).
4. Berntsen, T. K. et al. Response of climate to regional emissions of ozone precursors: sensitivities and warming potentials. Tellus B 57, 283–304 (2005).
5. Naik, V. et al. Net radiative forcing due to changes in regional emissions of tropospheric ozone precursors. J Geophys Res Atmos 110, D24306 (2005).
6. Collins, W. et al. Global and regional temperature-change potentials for near-term climate forcers. Atmos Chem Phys 13, 2471–2483 (2013).
7. Sitch, S., Cox, P. M., Collins, W. J. & Huntingford, C. Indirect radiative forcing of climate change through ozone effects on the land-carbon sink. Nature 448, 791–794 (2007).
8. Huang, X. et al. A high-resolution ammonia emission inventory in China. Global Biogeochem Cy 26, GB1030 (2012).
9. Xing, J. et al. Projections of air pollution emissions and its impacts on regional air quality in China in 2020. Atmos Chem Phys 11, 3119–3136 (2011).
10. Adams, P. J., Seinfeld, J. H., Koch, D., Mickley, L. & Jacob, D. General circulation model assessment of direct radiative forcing by the sulfate-nitrate-ammonium-water aerosol in the United States. Geophys Res Atmos 106, 1097–1111 (2001).
11. Baur, S. E. et al. Nitrate aerosols today and in 2030: a global simulation including aerosols and tropospheric ozone. Atmos Chem Phys 7, 5043–5059 (2007).
12. Liu, X. J. et al. Enhanced nitrogen deposition over China. Nature 494, 459–462 (2013).
13. Xia, Y. J., Niu, S. L. & Wang, S. Q. Response of ecosystem carbon exchange to warming and nitrogen addition during two hydrologically contrasting growing seasons in a temperate steppe. Global Change Biol 15, 1544–1556 (2009).
14. Vitousek, P. M. et al. Nutrient Imbalances in Agricultural Development. Science 324, 1519–1520 (2009).
15. Mo, J. M. et al. Nitrogen addition reduces soil respiration in a mature tropical forest in southern China. Global Change Biol 14, 403–412 (2008).
16. Zhang, W. et al. Methane uptake responses to nitrogen deposition in three tropical forests in southern China. J Geophys Res Atmos 113 (2008).
17. Mo, J. M. et al. Response of soil respiration to simulated N deposition in a disturbed and a rehabilitated tropical forest in southern China. Plant Soil 296, 125–135 (2007).
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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.

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