PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A
MATHEMATICAL, PHYSICAL AND ENGINEERING SCIENCES

The role of atmospheric reactive nitrogen in environmental issues in China

Journal:	*Philosophical Transactions A*
Manuscript ID	Draft
Article Type:	Review
Date Submitted by the Author:	n/a
Complete List of Authors:	Liu, Xuejun; China Agricultural University Xu, Wen; China Agricultural University Du, Enzai; 2; Beijing Normal University Tang, Aohan; China Agricultural University Zhang, Ying; China Agricultural University Zhang, Yangyang; China Agricultural University Wen, Zhang; China Agricultural University Pan, Yuepeng; Institute of Atmospheric Physics Chinese Academy of Sciences Zhang, Lin; Peking University Gu, Baojing; Zhejiang University Zhao, Yu; Nanjing University Shen, Jianlin; Institute of Subtropical Agriculture Chinese Academy of Sciences Gao, Zhiling; Hebei Agricultural University Feng, Zhaozhong; Nanjing University of Information Science and Technology Chang, Yunhua; Nanjing University of Information Science and Technology Goulding, K; Rothamsted Research Collett Jr, JL; Colorado State University, Department of Atmospheric Science Vitousek, Peter; Stanford University, Biological Sciences; Zhang, Fusuo; China Agricultural University, Plant Nutrition
Issue Code (this should have already been entered but please contact the Editorial Office if it is not present):	DM1119
Subject:	Atmospheric Chemistry (11) < CHEMISTRY (1002), Environmental Chemistry (67) < CHEMISTRY (1002)
Keywords:	Ammonia, nitrogen oxides, particulate pollution, eutrophication, integrated nitrogen management, China

Author-supplied statements

Relevant information will appear here if provided.

Ethics

Does your article include research that required ethical approval or permits?:
This article does not present research with ethical considerations

Statement (if applicable):
CUST_IF_YES_ETHICS :No data available.

Data

It is a condition of publication that data, code and materials supporting your paper are made publicly available. Does your paper present new data?:
My paper has no data

Statement (if applicable):
CUST_IF_YES_DATA :No data available.

Conflict of interest

I/We declare we have no competing interests

Statement (if applicable):
CUST_STATE_CONFLICT :No data available.

Authors' contributions

This paper has multiple authors and our individual contributions were as below

Statement (if applicable):
Liu, Xuejun: Conceptualization, Writing - original draft;
Xu, W: Writing - original draft;
Du, EZ: Writing - original draft;
Tang AH: Writing - original draft
Zhang Y: Writing - review & editing
Zhang YY: Writing - original draft
Wen Z: Writing - review & editing
Hao TX: Writing - review & editing
Pan YP: Writing - review & editing
Zhang L: Writing - review & editing
Gu BJ: Writing - review & editing
Zhao Y: Writing - review & editing
Shen JL: Writing - original draft
Zhou F: Writing - review & editing
Gao ZL: Writing - review & editing
Feng ZZ: Writing - original draft
Chang YH: Writing - review & editing
Goulding K: Writing - review & editing

Collett Jr JL: Writing - review & editing
Vitousek PM: Writing - review & editing
Zhang FS: Writing - review & editing

The role of atmospheric reactive nitrogen in environmental issues in China

Liu XJ[1], Xu W[1], Du EZ[2], Tang AH[1], Zhang Y[1], Zhang YY[1], Wen Z[1], Hao TX[1], Pan YP[3], Zhang L[4], Gu BJ[5], Zhao Y[6], Shen JL[7], Zhou F[8], Gao ZL[9], Feng ZZ[10], Chang YH[11], Goulding K[12], Collett Jr JL[13], Vitousek PM[14], Zhang FS[1]

1. College of Resources and Environmental Sciences, National Academy of Agriculture Green Development, School of Agriculture Green Development, China Agricultural University, Beijing 100193, China
2. State Key Laboratory of Earth Surface Processes and Resource Ecology, and School of Natural Resources, Faculty of Geographical Science, Beijing Normal University, Beijing 100875, China
3. State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC), Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China
4. Laboratory for Climate and Ocean-Atmosphere Studies, Department of Atmospheric and Oceanic Sciences, School of Physics, Peking University, Beijing 100871, China
5. College of Environmental and Resource Sciences, Zhejiang University, Hangzhou 310058, China
6. State Key Laboratory of Pollution Control & Resource Reuse and School of the Environment, Nanjing University, Nanjing 210023, China
7. Institute of Subtropical Agriculture, Chinese Academy of Sciences, Changsha 410125, China
8. Sino-France Institute of Earth Systems Science, Laboratory for Earth Surface Processes, College of Urban and Environmental Sciences, Peking University, Beijing 100871, China
9. College of Resources and Environmental Sciences, Hebei Agricultural University, Baoding 071000, China
10. Department of Applied Meteorology, Nanjing University of Information Science & Technology, Nanjing 210044, China
11. Yale-NUIST Center on Atmospheric Environment, Nanjing University of Information Science & Technology, Nanjing 210044, China
12. Sustainable Agricultural Sciences Department, Rothamsted Research, Harpenden AL5 2JQ, UK
13. Department of Atmospheric Science, Colorado State University, Fort Collins, CO 80523, USA
14. Department of Biology, Stanford University, Stanford, CA 94016, USA

Atmospheric reactive nitrogen (Nr) has been a cause of serious environmental pollution in China. Historically, China used too little Nr in its agriculture to feed its population. However, with the rapid increase in N fertilizer use for food production and fossil fuel consumption for energy supply over the last four decades, increasing gaseous Nr species (e.g. NH_3 and NO_x) have been emitted to the atmosphere and then deposited as wet and dry deposition, with adverse impacts on air, water and soil quality as well as plant biodiversity and human health. This paper reviews the issues associated with this in a holistic way. The emissions, deposition, impacts, actions and regulations for the mitigation of atmospheric Nr are discussed systematically. NH_3 emissions come mainly from the agricultural sector (including both crop and animal production systems), although other sources are important in urban areas, while NO_x emissions come mainly from vehicles, industry and power plants. Both NH_3 and NO_x make major contributions to environmental pollution but especially to the formation of secondary fine particulate matter ($PM_{2.5}$) pollution, which impacts human health and light scattering (haze). Regulations and practices introduced by China that meet the urgent need to reduce Nr emissions are explained, and recommendations for improving future N management for achieving "win-win" outcomes for Chinese agricultural production and food supply, and human and environmental health, described.

Key words: Ammonia, nitrogen oxides, particulate pollution, eutrophication, integrated nitrogen management, China

1. Introduction

The Haber-Bosch process, whereby N from atmospheric N_2 is made biologically available, has played an essential role in feeding an increasing global population [1]. However, the wide use of synthetic nitrogen (N) fertilizer (together with fossil fuel combustion) has also caused a number of environmental pollution problems worldwide, especially in China [2-4]. China has experienced a transition from a shortage of N for agriculture (1950s) to an initial N input-output balance (1980s) but then to large N surplus (2000s and 2010s) over the last six decades [5]. This has been accompanied by the rapid economic development since the 1980s [6]. China's environmental problems induced by excess reactive N (Nr) such as soil acidification [3,7], enhanced N deposition [8], biodiversity loss [9] and aquatic eutrophication [10] have been reported widely since the 2000s. The excess Nr inputs and subsequent losses from recipient systems have caused soil degradation, decline in water and air quality, and human health effects, as well as changing ecosystem functions and degrading ecosystem services in China [11]. The challenge in reducing Nr emissions to the environment and so limiting Nr pollution are growing not only in China [10] but around the world [12-13]. As a result, sustainable N management is essential to further increase crop production for a growing global population [14-16] while maintaining ecosystem functions and services [17]. Currently China is at a historic turning-point, needing to change its economic model from high resource inputs and high production to double high (high efficiency and high production) model [18]. To achieve this, more precise N management practices and stricter environmental regulations for both producers and consumers are required urgently.

This review is in five parts: (1) an overview of the environmental problems caused by excess Nr; (2) progress in reducing Nr emissions in relation to air pollution; (3) progress in reducing Nr deposition and its ecological impacts; (4) regulations and actions for mitigating atmospheric Nr emissions and improving air quality; (5) summary and recommendations. Figure 1 summarizes the three key parts of the review (Nr emissions and air pollution; Nr deposition and impact; national Nr emission regulations) as well as the relationships between the parts.

2. Atmospheric reactive N emissions and air pollution

(a) Atmospheric reactive N emissions

Reactive N compounds are emitted to the atmospheric mainly in the form of ammonia (NH_3) and nitrogen oxides (NO_x). NH_3 is the main alkaline gas species, and can neutralize sulfuric acid (H_2SO_4, a product of SO_2 oxidation) and nitric acid (HNO_3, a product of NO_x oxidation) to form ammoniated sulfate and ammonium nitrate aerosols in the air. These aerosols are secondary inorganic aerosols, reflecting their formation by atmospheric chemistry, and are a significant component of fine particulate matter (PM) pollution, accounting for 40–57 % of the $PM_{2.5}$ (particles smaller than 2.5 μm) mass in eastern China [19-20]. NO_x is also a precursor of tropospheric ozone, reacting with hydrocarbons to form ozone in the presence of sunlight. Although nitrous oxide is an important greenhouse gas, it is chemically inert in the troposphere and has a minor effect on air pollution, and so is not discussed further in this section.

Figure 2 shows the spatial distributions of annual total NH_3 and NO_x emissions in China averaged over 2008-2012. Sources of NH_3 and NO_x in China are mainly anthropogenic activities, but their magnitudes and variations are typically driven by different sectors of the economy. NH_3 is mainly released from the agricultural sector, i.e., fertilizer use and livestock production, although vehicles and other sources can be important in urban areas. NO_x is mostly generated by fuel combustion by industry, transportation, and power plants. China has the largest NH_3 emissions in the world due to its intensive agriculture [8]. Present-day Chinese NH_3 emissions are estimated to be in the range of 6.9-15 Tg N per year averaged over 2005-2012 [21-23]. The wide range and significant uncertainty in China's NH_3 emission estimates are largely attributed to missing agricultural statistics (activity data and emission factors), as well as a lack of NH_3 flux measurements to constrain the emission estimates. Recent developments in a nationwide N deposition monitoring network [24-25], the Chinese ammonia monitoring network [26] and multiple satellite retrievals [27-29] provide valuable information on Chinese NH_3 emissions and their spatial and temporal variation [23,30]. At the national scale, agricultural sources (fertilizer use and livestock manure management) are the dominant NH_3 sources, together contributing over 80% of total anthropogenic emissions [21,23]. However, urban sources such as transportation and waste disposal may be important in urban environments, as suggested by recent research using concentration ratios with NO_2 [31] and N isotope measurements [32-33].

NO_x emissions over China had been increasing rapidly over the past three decades, mainly driven by industrialization and urbanization, but have begun to decline in recent years. Anthropogenic NO_x emissions were estimated to be about

3.5 Tg N in 2000 [34], increasing to 8.9 Tg N in 2012, then gradually decreasing to 6.7 Tg N in 2017 due to the implementation of clean air regulations [35]. Satellite observations of NO_2 columns have been successfully applied in a growing number of studies to improve the estimates of NO_x emissions over China [36-38]. NO_x can also be emitted from natural and/or non-fossil fuel sources, such as lightning, microbial processes in soil and biomass burning. These may only account for about 10% of anthropogenic Chinese NO_x emissions, but have important contributions to surface ozone air quality [39].

Recent clean air actions implemented by the Chinese government have effectively decreased the emission of NO_x, as shown by satellite measurements [40], but emissions of NH_3 have not been regulated, resulting in increasing attention over the need to control NH_3 to further mitigate air pollution [35,41-42] and reduce N deposition exceedance in many regions of China [43].

(b) Reactive N and acid rain

Wet deposition is effective in scavenging many atmospheric pollutants. The chemical characteristics of rainwater provide insight into air pollution and help us to understand the sources and transportation of atmospheric pollutants [44]. In-cloud and below-cloud scavenging results in Nr becoming the dominant pollutant of precipitation. Gaseous acids (e.g., HNO_3 and H_2SO_4) and bases (NH_3) are critical in determining the acidity of rainwater. The average pH of precipitation in China was 5.58 in 2018 with 53.8 million ha, 5.5% of the Chinese land area, receiving acid rain (rain or precipitation that contains elevated levels of hydrogen ions with pH value under 5.6, an equilibrium pH of distilled water in contact with atmospheric CO_2). Most Chinese cities suffer from acid rain: approximately 77 out of 471 Chinese cities had a frequency of acid rain that exceeded 25%, and 39 had a frequency that exceeded 50% in 2018 [45]. Southwest China especially continues to suffer from severe acid precipitation, with the mean pH being 5.1 in Sichuan province from 2011- 2016 [46]. The situation in eastern China is becoming worse. For example, Shanghai experienced acid rain with a pH value less than 3.0 in 2005 [47] and the acidity was still very high, with an average pH of 4.96 from 2011 to 2016 [48]. In contrast to Southern and Eastern China, Northern China has experienced alkaline precipitation. In Gansu province, for example, the pH value of precipitation was from 6.63 to 8.10 during 2011-2014 [49]. In Beijing city, the precipitation has changed from sulfuric acid dominated to mixed, with both sulfuric and nitric acids [50-51].

The acidity of precipitation and its chemical components vary as a function of the local environment and anthropogenic activities. SO_4^{2-}, NO_3^-, NH_4^+, Ca^{2+} are the dominant water-soluble ions in precipitation. In China, SO_2 emissions from industrial, domestic and energy sectors decreased from 25.5 Tg in 2005 to 18.6 Tg in 2015 and even <10 Tg in 2018 because of the Chinese government's strict control strategies [52]. The remarkable decrease in SO_4^{2-} concentration (from 285 to 145 μeq L^{-1}) resulted in an increase in the pH of Sichuan precipitation from 5.24 in 2011 to 5.70 in 2016 [46] and the chemical composition of acid rain changed from sulfuric acid-dominated to mixed. NO_3^- was the second most abundant anion in precipitation, with vehicle emissions considered the dominant source [53]. From the point of view of reducing acidity, further control of NO_x emissions is essential. Anthropogenic NH_3 emissions increased 8% from 2005 to 2010 and were predicted to show further increase by 15% in 2050 (relative to 2010) [54-55]. The high concentration of NH_4^+ in precipitation is mostly due to NH_3 emissions from agriculture [56]. As the only alkaline gas in air, NH_3 neutralizes precipitation acidity and so reduces the occurrence of acid rain. However, the downside is that NH_3 promotes the formation of PM material and aggravates haze pollution [57]. Liu et al. [41] found that NH_3 emission abatement mitigates $PM_{2.5}$ and N deposition but worsens acid rain in China. Therefore, a comprehensive reduction of NH_3 emissions is urgently needed.

(c) Reactive N and haze pollution

Over the past decades, China's economy has been growing rapidly but this has resulted in severe air pollution, especially haze, which is largely due to increasing resource consumption and accompanied elevated emissions of NO_x, SO_2 and NH_3 [58]. Haze pollution threatens not only the human respiratory system and the heart [59-60] but also the regional climate [61], plant photosynthesis [62] and food security by affecting solar radiation [63].

The prime air pollutant of concern during haze is fine particulate matter ($PM_{2.5}$), ~40-57% of which is caused by secondary inorganic aerosols (SIA, i.e. sulfate (SO_4^{2-}), nitrate (NO_3^-) and ammonium (NH_4^+)) in China [19,20], as for other regions across the world [64-65]. Both acid (e.g., SO_2 and NO_x) and alkaline (e.g., NH_3) pollutants are crucial to the formation of SIA by acid-base neutralization reactions [66]. These reactions increase the size and solubility of particles, and aerosol growth becomes spontaneous once the bonded particles exceed the threshold size, i.e., the nucleation barrier [67]. Hence, emission control of these important gaseous precursors (SO_2, NO_x and NH_3) has been a critical factor in attempting to control haze pollution. However, since 2010 China has contributed more than one-fifth of global anthropogenic emissions of SO_2 (~30%), NO_x (~24%) and NH_3 (~20%) [68].

China has taken many strict and appropriate emission control measures to improve air quality, notably its 'Action Plan for the Prevention and Control of Air Pollution' in 2013. Consequently, national emissions of SO_2 and NO_x decreased by 62% and 17%, respectively, from 2010-2017 [69]. In contrast, because no official control measures have been implemented for NH_3 in China until 2017, NH_3 emissions have remained constant at ~10.0 Tg and showed relatively smaller decline over recent years. Although the absolute mass concentrations (μg m^{-3}) of $PM_{2.5}$ and SIA significantly decreased because of the decreasing emissions of SO_2 and NO_x, the unbalanced decline in emissions of SO_2 (large) and NO_x+NH_3 (small) increased the fraction of N-containing inorganic compounds (NO_3^- and NH_4^+) in $PM_{2.5}$ from 2013 [23,70].

The contribution of aerosol nitrate to $PM_{2.5}$ increased during 2006–2015 over eastern China, together with a decrease in SO_4^{2-} [71]. NO_3^-, rather than SO_4^{2-}, dominated haze formation in winter [72]. Zhang *et al.* [69] showed that regional haze may be effectively minimized by controlling NH_3 emissions, but there are still large uncertainties about the effectiveness of NH_3 reduction without simultaneously reducing emissions of SO_2 and NO_2. In future, the simultaneous reduction in emissions of NO_x and NH_3 together with SO_2 is key to limiting deadly haze pollution in China [73] and other countries [74].

(d) Reactive N and O_3 pollution

Ground-level ozone (O_3) is an important secondary air pollutant, which has a strong correlation with its precursors, such as NO_x and VOC_s [75]. The photolysis of NO_2 to give NO and an O atom, results in the O atom reacting with molecular oxygen (O_2) to form O_3; it can also react with NO to regenerate NO_2 [76]. In China, the large emissions of NO_x from rapid industrial and urban development have caused rapidly-increasing ground-level O_3 concentrations [77-79]. Ozone pollution is mainly concentrated in summer, which also coincides with the growing season of plants.

Li *et al.* [80] analyzed the spatial-temporal changes in ground-level O_3 concentrations in China, using data from 187 cities, from January 2014 to November 2016. They showed that the average O_3 concentration had a large spatial variation, from 50.61 ppb to 64.1 ppb, and increased from 46.1 ± 8.8 ppb in 2014 to 51.9 ± 7.8 ppb in 2016. Ozone pollution is more serious in economically developed areas, such as Jing-Jin-Ji, the Yangtze River Delta and the Pearl River Delta [80]. Zhu and Liao [81] used the high-resolution nested grid version of the GEOS-Chem model to simulate changes in ground-level O_3 concentrations from 2000-2050 under emission pathways for IPCC scenarios RCP2.6, RCP4.5, RCP6.0 and RCP8.5. They predicted the average O_3 concentration would increase by a maximum of 6-12 ppb each year if emissions are not effectively controlled, and so will become increasingly serious.

High ground-level O_3 concentrations have adverse effects on human health and vegetation [82]. Ozone causes damage to plants after entering through leaf stomata, including visible leaf injury [83], impairment of photosynthesis [84], and reductions in growth and yield [82,85]. A recent meta-analysis of Chinese woody plants showed that elevated O_3 concentrations (116 ppb) reduced total biomass by 14% compared with the control (21 ppb) [86]. Yue *et al.* [87] used a coupled chemical-carbon-climate model to suggest that the current O_3 concentration in China has reduced the annual net primary productivity by about 10.1–17.8%. Feng *et al.* [82] quantified the adverse impact of O_3 on human health and vegetation (forests and crops) based on data from > 1400 monitoring stations in China in 2015, and showed that the current O_3 level led to a 0.9% increase in premature mortality, and reduced annual forest tree biomass growth by 11–13% and the yield of rice and wheat by 8% and 6%, respectively. The total O_3-related cost to health, food production and the environment reached 761.7 billion US$, equivalent to 7% of the China's Gross Domestic Product in 2015.

3. Atmospheric N deposition and ecological impacts

(a) Atmospheric N deposition

As an important component of the N cycle, atmospheric N deposition has attracted increasing attention in China and worldwide [88-90]. With the continuous increase in Nr emissions, China has been a global hotspot for N deposition [2,8] and so has introduced several monitoring networks. Currently, the Nationwide Nitrogen Deposition Monitoring Network (NNMDN), led by China Agricultural University, the Center for Ecosystem Research Network (CERN) and the Ammonia Monitoring Network in China (AMoN-China) led by the Chinese Academy of Sciences, are the three main N deposition monitoring networks in China [26,91-92].

Nitrogen deposition exhibits clear spatial variability in China due to the large spatial heterogeneity of anthropogenic Nr emissions [93] and of factors such as land use type [94] and the amount of precipitation [93]. Based on field measurements at 43 sites of the NNDMN, the total wet/bulk and dry N deposition averaged 39.9 kg N ha^{-1} yr^{-1} in China and is ranked by land use as urban> rural> background sites or by region as North China> Southeast China> Southwest China> Northeast China> Northwest China> Tibetan Plateau [24]. Except for significantly higher values in northern rural sites, annual dry N deposition is comparable at urban and background sites in northern and southern regions [95]. Modeling and satellite observations have been used to elucidate the spatial characteristics of N deposition in China [43,88,96]. For example, total N deposition simulated by Zhao et al. [43] was generally less than 10 kg N ha^{-1} yr^{-1} in western China, and 15-50 kg N ha^{-1} yr^{-1} in eastern China. Using a remote sensing model, Yu et al. [88] showed that: 1) wet reduced (NH_x) and oxidized (NO_y) N deposition were both a maximum in the North, followed by East and Central

China; 2) the highest NH_x and NO_y dry deposition occurred in North China, and dry N deposition decreased from North China to other regions; 3) the spatial pattern of total N deposition (19.6 kg N ha^{-1} yr^{-1} on average) was similar to those of total NH_x and NO_y deposition (Figure 3). They also found that 62–99% of the spatiotemporal variation of total N deposition in China was mainly due to Nr emissions from energy consumption, N fertilizer use and livestock production [88], whereas for wet deposition, the amount of precipitation and N fertilizer use can explain 80–91% of its variability [92]. Although estimates of total N deposition were somewhat different in the aforementioned studies, they all agreed that dry N deposition is equally important as wet/bulk N deposition at the national scale [24,43,88].

As for temporal changes in N deposition, Zhang et al. [97] first reported a substantial increase in wet N deposition in the North China Plain from the 1980s to the 2000s. After summarizing historic data on bulk N deposition in the whole China, Liu et al. [8] found that China's N deposition showed a significant increase from 1980 to 2010, with an annual increase of 0.41 kg N ha^{-1}. Yu et al. [88] showed that total N deposition in China changed from an initial rapid increase to stability between 1980 and 2015, in which wet N deposition reached a maximum in 2001–2005 and declined thereafter, in contrast to a continuous increase in dry deposition. The stabilization of total N deposition was mainly caused by a gradual decline in wet NH_4^+ deposition. Total N deposition was dominated by wet N deposition between the 1980s and 2000s, with a shift to approximately equal wet and dry N deposition from 2011–2015 in all regions except Northwest, Central and South China. NH_x deposition dominated dry, wet, and total N deposition between 1980 and 2015, but its contribution gradually decreased due to increasing NO_3^- deposition. The recent stabilization and even decrease of atmospheric N deposition [98] reflected successful Nr emission control measures especially after 2013 [69].

Significant progress in determining the spatio-temporal trend in N deposition in China has been achieved, but some key points should be addressed in future studies to improve the accuracy of results [99]. Taking NH_3 as an example, all the reported deposition fluxes may be subjected to some uncertainties owing to the following major causes: 1) large uncertainties still exist in China's NH_3 emission inventory due to a lack of reliable data about the local agriculture and other activities and emission factors, which limits accurate modeling of temporal and geographical distribution of atmospheric NH_3 concentrations; 3) parameterization for bi-directional exchange of NH_3 is not included in chemistry and transport models and satellite-based estimates, which can result in an overestimation of NH_3 deposition, especially in agricultural areas.

(b) Impact on soil acidification

Soil acidification is a natural and gradual process in soil development, but which is dramatically enhanced by human activity. Elevated acid deposition, induced by growing NO_x and SO_2 emissions derived from industrial activities and increasing NH_3 emission due to agricultural N fertilization, has caused soil acidification in forests and/or grasslands especially in Europe [100], United States [101] and China [102-103]. Enhanced soil acidification has not only become a serious threat to ecosystem functioning and services in grasslands and forests [7,104] but also a threat on agricultural systems [105].

Increased N inputs to agriculture, especially of chemical N fertilizer, have enhanced soil acidification in croplands [3]. N input via deposition comprises ~8% of the N fertilizer application (farmers' practice) [106-107], making deposition a significant contributor to N-induced soil acidification of croplands, especially for those regions with high N deposition [96,108].

Chronic N deposition has significantly increased net aboveground primary productivity (NAPP) and plant uptake, but also and leaching losses (with excess nitrate) of base cations from the soil, especially in agricultural ecosystems [106-107]. Soil acidification reduces the availability of nutrients (e.g. base cations, phosphate, molybdenum and boron) and increases the concentrations of toxic elements (e.g. aluminum, iron and manganese), leading to restricted plant and soil biota growth due to nutrient deficiency and metal toxicity [109]. Liming is a common practice for alleviating soil acidification, but it is not a panacea [105].

Chinese croplands are facing an increasing risk of aluminum toxicity due to enhance soil acidification [110], with accompanying crop yield losses unless N deposition is mitigated [111]. Although '4R principles' (applying fertilizer as the Right product in the Right amount at the Right time and in the Right place), i.e. balanced N fertilization, combined with manure and straw recycling, can effectively reduce soil acidification [111-113], N deposition, as an important N input to croplands, must be included in calculating N fertilizer recommendations and management. Meanwhile, non-agricultural ecosystems (forests and grasslands) in China are still suffering from high N deposition and related soil acidification and reduced biodiversity, urgently requiring more vigorous controls on N pollution [8,11].

(c) Impact on plant biodiversity

The impact of N deposition on plant diversity has gained increasing attention because of potential consequences for ecosystem services. Elevated N deposition is considered to be one of the most important drivers of biodiversity loss [114]. A global assessment has suggested that N deposition can cause a shift in community species composition or/and a loss of plant species in terrestrial ecosystems [115]. Ecosystems in China, especially those in eastern and southern regions, have been subjected to long-term, high-level N deposition [8,88]. Comparing N deposition maps with critical load estimates, Zhao *et al.* [43] suggested that about 15% of the land cover in China experiences critical load exceedances, implying a high risk of negative ecological effects (e.g., biodiversity loss). However, there are few reports of observed losses of plant biodiversity in response to N deposition due to a lack of well-designed monitoring systems of long-term changes in plant biodiversity [116].

Current understanding of N deposition impacts on plant biodiversity come mainly from N addition experiments [91,116]. Generally, increasing N deposition decreases the dominance of N-sensitive species, while benefiting nitrophiles due to their ability to utilize available N. For example, nine-years of N additions (20, 50 and 100 kg N ha^{-1} yr^{-1}) significantly increased coverage of graminoids and decreased that of mosses and shrubs in a natural boreal forest in Northeast China [117-118]. In an old-growth subtropical forest in southern China, N additions of >100 kg N ha^{-1} yr^{-1} decreased the abundance of understory seedlings, ferns and mosses, but did not affect canopy trees and shrubs [119]. A recent meta-analysis of experimental results in China concluded that N addition negatively affected plant biodiversity in grasslands and forest understory communities; the effects varied by climatic zone, N addition level and duration [120]. However, there are relatively few reports from eastern and southern China [120] where high-level N deposition has occurred for decades. In these regions, N addition experiments likely underestimate the negative effects because high-level N deposition may have already caused a shift in plant species composition towards that better adapted to high N availability before the experiments began.

(d) Impact on aquatic eutrophication

Elevated atmospheric N deposition has increasingly impacted aquatic ecosystems particularly their N budget and phytoplankton structure [121-122]. It has resulted in eutrophication and enhanced phytoplankton biomass in unproductive lakes in Europe and North America [123]. With the increase in N deposition, the stoichiometric N/phosphorus (P) ratio increased and phytoplankton diversity was reduced by favoring those few species with the ability to use P more effectively [121]. In the recent years, much research has explored the impacts of N deposition on aquatic eutrophication across China. Nitrogen deposition to Lake Taihu, Lake Dongting and Lake Dianchi were reported to be as high as 50 to 80 kg N ha^{-1} yr^{-1} [124-126]. This has contributed a large proportion (15 to 48%) of the total N load into these lakes, increased the N concentration in the lakes and induced eutrophication. Research in Lake Dianchi also found that the when toxic blooms of the non-N_2-fixing cyanobacteria *Microcystis* Spp. were initiated and proliferated, the contribution of N deposition to the total N load was as high as 27-48%, which indicates that N deposition may be the main contributor to the blooms [125]. High N deposition was also reported to the coastal waters around China. A total N deposition of 20 to 50 kg N ha^{-1} was measured to the Yellow sea and South China Sea [127-129]. It is estimated that an additional primary biological productivity of 1.5 to 30 g C m^{-2} yr^{-1}, which accounted for 0.3 to 6.7% of the current productivity in the Yellow sea, was caused by N deposition [127]. Incubation experiments on the impacts of wet N deposition on phytoplankton community structure have shown that, when treated by 5 to 10% of the rainwater addition (filtered or not), the total chlorophyll *a* concentration increased 1.6 to 1.9-fold while microphytoplankton increased 1.7 to 3.2-fold [129]. The abundance of diatoms increased and they became the dominant species, accounting for 55% of total phytoplankton abundance. Atmospheric N deposition also causes N enrichment in rivers [95,130]. A study in a subtropical catchment showed that the N concentrations in the river water showed significant positive correlations with rainwater N concentrations, and N deposition contributed 21% of the total riverine N export in the whole catchment [130].

4 Regulations and actions mitigating atmospheric Nr emissions

(a) Regulations mitigating NH_3 emissions in agriculture

Mitigating NH_3 emission in agriculture was added to the updated version of the Clean Air Act (CAA) in 2018, before which the CAA mainly focused on pollutant reductions from industrial and transportation sources in China. Other national regulations, such as the Zero Increase in Chemical Fertilizer Use after 2020 [130], have been introduced by the Chinese government. Farmers are required to change of crop rotations, substitute manure for chemical fertilizers, etc., and these have resulted in some increases in NUE while stabilizing chemical fertilizer use. However, the overuse of chemical fertilizer in China is estimated to be still >30%, indicating that a substantial reduction in fertilizer use is still needed [131].

Compared to crop production, the reduction of NH_3 emission from livestock production is more important because the most serious air pollution occur mainly in winter and spring when agricultural NH_3 emissions are dominated by livestock production [42,132]. Noting the importance of NH_3 in the causes of $PM_{2.5}$ pollution and haze, explained above, effective reductions in haze episodes can be anticipated given that successful mitigation of NH_3 emission from livestock in winter and spring has already been achieved. However, due to the incomplete assessment of manure management practices, many current treatments such as air-drying, intended to reduce water pollution, may benefit manure recycling and reduce N losses to water but cause unintended increases in NH_3 emissions – 'Pollution Swapping' [133]. This creates imperative and critical requirements for establishing guidelines for environmental friendly practices for livestock production, including standards and protocols for monitoring NH_3 emissions from livestock production. In addition, the decoupling of livestock production from crop production increases the transportation costs and reduces manure recycling, increasing NH_3 emissions from livestock systems [134]. Policies that can rebuild the link between livestock and cropping are crucial.

Although not the major source of NH_3, optimizing fertilizer use in croplands and reducing NH_3 emissions is still important for environmental health. Many measures have been developed, including '4R stewardship', explained above, to manage N use in croplands [112]. However, the implementation of these measures all require appropriate farm size to make them practicable and reduce costs [135]. When Chinese farm sizes increase from the current average of 0.5 ha to 3-5 ha, agricultural fixed input costs for machinery and other facilities are much improved, benefitting the implementation of 4R stewardship and other measures, and China's N fertilizer use can be reduced by an estimated 30% [132]. A concomitant increase in NUE and a reduction in NH_3 emissions can be expected [136]. To increase farm size, urbanization and regulations for land transfer are critical: urbanization can move rural populations to urban areas and, at the same time, release arable land through the reclamation of land that was rural towns and villages, leading to larger farm sizes through an increase in croplands for a smaller population [137]. Land transfer can then reallocate cropland among rural households, facilitating the growth of alternative farming models such as family, cooperative/collective and industrial farms, which normally have larger farm sizes. However, this would be unacceptable in many countries.

Other non-technical measures, such as dietary change, can also help [137]. It is estimated that, in China, over half of crop production is consumed by livestock as feedstuff, due to the dramatic increase in consumption of animal products [138]. A change to a more plant-based diet, partially reversing the recent trend in China, could produce a significant decrease in agricultural NH_3 emissions. Managing consumption can have a major impact but will be difficult to achieve.

(b) Regulating NO_x emissions from traffic and industry

As one of the main atmospheric pollutants, NO_x plays a crucial role in the formation of acid rain and other regional environmental problems. Production of O_3 and SIA is very dependent on the abundance of NO_x [139]. Driven by rapid economic development, urbanization and intensive energy use, NO_x emissions in China were estimated to have increased significantly within the first decade of the 21st century [139-142]. Before 2010, the Chinese government took aggressive steps to improve energy efficiency and reduce emissions of primary aerosols and SO_2, but paid less attention to NO_x abatement. The annual growth rate of NO_x emissions from 1995 to 2004 was estimated at 6.3% [139] and the annual total emissions were calculated to reach 28.1 Mt in 2010 [142]. This rapid growth partially offset China's efforts on SO_2 emission reduction and exacerbated regional acid rain [143].

The national 12th Five-Year Plan issued in 2011 aimed to reduce NO_x emissions by 10% from 2010 to 2015 [144]. A series of emission control measures have been implemented in power plants, steel and cement industries, industrial boilers, and transportation. These measures were further refined in the Air Pollution Prevention and Control Action Plan issued in 2013 [145]. Thermal power has been the most important sector introducing NO_x control since 2010. An updated emission standard for the sector was issued in 2011, limiting NO_x concentrations in flue gas to <100 mg m^{-3} [146]. $DeNO_x$ technologies (e.g., selective catalytic reduction, SCR) were required and installed in 92% of China's thermal power sector in 2015 [147]. The government subsequently released the ultra-low emission policy for the sector, requiring NO_x concentrations in the flue gas of coal-fired units to be the same as those in gas-fired units, i.e. <50 mg m^{-3} [148]. Coal-fired units retrofitted with ultra-low emission controls totalled 810 million kilowatts, accounting for 80% of the total installed-capacity at the end of 2018 [149]. As a result, the fraction that the power sector contributed to total emissions was reduced from 32.5% in 2010 to 19.1% in

2017, as shown in Figure 4. Emission controls have gradually expanded to other non-electricity generating industries including cement and steel production. For example, the ultra-low emission policy was introduced in the steel industry in April 2019 [150], limiting NO_x concentrations in sintering flue gas to <50 mg m^{-3}. China is also paying increasing attention to emission controls in transportation, including the staged implementation of stringent emission standards on new vehicles, the retirement of old vehicles, and the expansion of renewable energy fueled vehicles. These policies have proved effective. For example, progress from the China III to China IV standard was estimated to reduce the NO_x emissions from on-road vehicles by 50% between 2011 and 2015 [151]: from 2013 to 2018, China's vehicle ownership increased by 33% while NO_x emissions decreased by 14% [149]. The most recent plans have included a series of policies released in early 2019 to control emissions from diesel trucks that account for over 70% of total vehicle emissions [152], and the nationwide implementation of China VI, the most stringent standard, from 2020. Besides on-road vehicles, official plans were also gradually announced to limit NO_x emissions from other mobile sources such as ships [153]. With the above-mentioned regulations, China's NO_x emissions from anthropogenic sources were estimated to have declined by 21% from 2013 until the end of the Action Plan in 2017 (Figure 4)[35]. If emission controls had been frozen at the 2010 level, as a comparison, emissions would have increased by 38%, as indicated by the triangles in Figure 4. To maintain the restraints on NO_x emissions in the future, a three-year plan [154] for 'Defending the Blue Sky' was announced in 2018, aiming for a further 15% reduction in NO_x emission from 2015 to 2020.

(c) National actions for mitigating Nr emissions and improving air quality

In China, a number of national/regional actions have been introduced for improving air quality through the mitigation of atmospheric Nr, as explained in previous sections [18,155]. In general, these can be divided into short-term and long-term measures. Figure 5 summarizes the major implementation steps in China's atmospheric environmental protection policies/regulations and emission mitigation measures related to atmospheric Nr emission controls and air quality improvement.

For short-term emission mitigation measures, the Chinese Government temporarily closed many factories and construction sites and controlled traffic flow during some important national celebrations such as the 2008 Beijing Summer Olympics, the 2010 Shanghai World Expo and the 2014 Beijing APEC meeting. These proved their effectiveness in improving air quality over short periods, but their weakness was very clear in the rebound observed in post-activity air pollution, which offset all previous efforts in air quality improvement [31,156-157]. Similarly, but not to be welcomed, the recent coronavirus epidemic (which was first reported in Wuhan, then extended to the whole of China and finally worldwide) abruptly stopped China's major economic activities and led to a reduction in NO_x and other air pollutant emissions of at least 36% due to the nationwide isolation needed to tackle the virus. However, $PM_{2.5}$ pollution events still occurred during the Chinese New Year holiday in many regions, such as Beijing and Shanghai, for reasons that are not yet known [158].

For long-term Nr mitigation measures, the main national actions included denitration from industry and coal-fired power plants, natural gas heating replacing coal heating in northern cities, and agricultural NH_3 mitigation [11]. These have produced continuously positive effects in reducing atmospheric Nr concentrations, N deposition and $PM_{2.5}$ pollution [98-99,155]. For example, the replacement of coal by natural gas for heating caused a >75% decline in $PM_{2.5}$ concentrations in Urumqi from January 2011 (322 μg m^{-3}) to January 2014 (79 μg m^{-3}) [155]. Another successful example was that of Quzhou in Hebei, where improved N management associated with the now well known 'Science and Technology Backyard Programme' (a technology innovation and extension platform linking universities, local government, private companies and farmers [159]) from 2009 increased grain yields and reduced fertilizer N inputs, reducing NH_3 losses [160]. As a consequence, the annual mean $PM_{2.5}$ concentration in Quzhou was 40% lower in 2015-2017 than in 2011-2014 [160-161].

5. Conclusion and recommendations

Concerns over Nr-induced environmental issues, in particular air pollution problems such as acid rain, haze and O_3 pollution, have increased dramatically in China. Ammonia and NO_x, mainly from agriculture and from the transport and industrial sectors, respectively, are two key Nr species inducing air, water and soil pollution and environmental degradation including loss of plant biodiversity. This overview clearly shows China's progress in reducing Nr emissions, deposition and related environmental issues through policies, regulations and practices introduced during a period of rapid economic growth over the last 4 decades.

To improve air quality (e.g. by reducing $PM_{2.5}$ pollution), the Chinese government has taken a series of actions covering policy, law/regulation and technology innovations to mitigate Nr and other pollutant emissions from the 2000s onwards and especially after 2013 [18]. As a result, Nr emissions and deposition and their ecological and environmental impacts have at least stabilized or even declined, especially after the recent strict emission control regulations and measures [88,98]. This represents considerable progress for China. However, the current atmospheric Nr emissions and deposition are still relatively high at approx. 20 Tg N yr^{-1} or 20 kg N ha^{-1} yr^{-1} [23,88].

Looking forward to 'Green Development' in the future, China needs to facilitate further national and international collaborations between various stakeholders from different regions, institutions and private companies that focus on increased NUE in agriculture through sustainable N management to decrease Nr emissions from agriculture. This will add to progress already being made in the transport and industrial sectors. NH_3 emission mitigation from both agricultural and non-agricultural sectors is particularly important for making further reductions in secondary $PM_{2.5}$ pollution now that increasingly strict SO_2 and NO_x emission mitigations are being introduced [162]. Finally, atmospheric Nr emission reduction should be placed in the context of global climate change, environmental sustainability, food security and human health. We should pursue "win-win" strategies over the long-term.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (41425007), the Chinese State Key Special Program on Severe Air Pollution Mitigation "Agricultural Emission Status and Enhanced Control Plan" (DQGG0208), the State Key R&D programme (2017YFC0210101, 2018YFC0213301-03), and the UK-China Virtual Joint Centre for Improved Nitrogen Agronomy (CINAg, BB/N013468/1).

Reference

1. Erisman JW, Sutton MA, Galloway J, Klimont Z, Winiwarter W. 2008 How a century of ammonia synthesis changed the world. *Nat. Geosci.* **1**, 636-639. (doi:10.1038/ngeo325)
2. Galloway JN, Townsend AR, Erisman JW, Bekunda M, Cai Z, Freney JR, Martinelli LA, Seitzinger SP, Sutton MA. 2008 Transformation of the nitrogen cycle: recent trends, questions, and potential solutions. *Science* **320**, 889-892. (doi:10.1126/science.1136674)
3. Guo JH, Liu XJ, Zhang Y, Shen JL, Han WX, Zhang WF, Christie P, Goulding KW, Vitousek PM, Zhang FS. 2010 Significant acidification in major Chinese croplands. *Science* **327**, 1008-1010. (doi:10.1126/science.1182570)
4. Kanter DR, Searchinger TD. 2018 A technology-forcing approach to reduce nitrogen pollution. *Nat. Sustain.* **1**, 544-552. (doi:10.1038/s41893-018-0143-8)
5. Liu XJ, De Vries W, Zhang Y, Ma L, Cui ZL, Zhu QC, Fan MS, Zhang WF, Goulding K, Misselbrook T, Chadwick D, Zhang FS. Balancing nitrogen inputs to reconcile China's food supply and environmental safety. *Nat. Commun.*, in preparation.
6. Liu JG, Diamond J. 2005 China's environment in a globalizing world. *Nature* **435**, 1179-1186. (doi:10.1038/4351179a)
7. Lu XK, Mao QG, Gilliam FS, Luo YQ, Mo JM. 2014 Nitrogen deposition contributes to soil acidification in tropical ecosystems. *Global Change Biol.* **20**, 3790-3801. (doi:10.1111/gcb.12665)
8. Liu XJ, Zhang Y, Han WX, Tang AH, Shen JL, Cui ZL, Vitousek P, Erisman JW, Goulding K, Christie P, Fangmeier A, Zhang FS. 2013 Enhanced nitrogen deposition over China. *Nature* **494**, 459-462. (doi:10.1038/nature11917)
9. Bai YF, Wu JG, Clark CM, Naeem S, Pan QM, Huang JH, Zhang LX, Han XG. 2010 Tradeoffs and thresholds in the effects of nitrogen addition on biodiversity and ecosystem functioning: evidence from inner Mongolia Grasslands. *Global Change Biol.* **16**, 358-372. (doi:10.1111/j.1365-2486.2009.01950.x)
10. Yu CQ, Huang X, Chen H, Godfray HCJ, Wright JS, Hall JW, Gong P, Ni SQ, Qiao SC, Huang GR, Xiao YC, Zhang J, Feng Z, Ju XT, Ciais P, Stenseth NC, Hessen DO, Sun ZL, Yu L, Cai WJ, Fu HH, Huang XM, Zhang C, Liu HB, Taylor J. 2019 Managing nitrogen to restore water quality in China. *Nature* **567**, 516-520. (doi:10.1038/s41586-019-1001-1)
11. Liu XJ, Du EZ. 2020 Chapter 1: An Overview of Atmospheric Reactive Nitrogen in China from a Global Perspective. In *Atmospheric Reactive Nitrogen in China* (eds. Liu XJ and Du EZ). Springer, Singapore pp. 1-10. (doi:10.1007/978-981-13-8514-8_1)

12. Davidson EA, Suddick EC, Rice CW, Prokopy LS. 2015 More food, low pollution (mo fo lo Po): a grand challenge for the 21st century. *J. Environ. Qual.* **44**, 305-311. (doi:10.2134/jeq2015.02.0078)
13. Stevens CJ. 2019 Nitrogen in the environment. *Science* **363**, 578-580. (doi:10.1126/science.aav8215)
14. Chen XP, Cui ZL, Fan MS, Vitousek P, Zhao M, Ma WQ, Wang ZL, Zhang WJ, Yan XY, Yang JC, Deng XP, Gao Q, Zhang Q, Guo SW, Ren J, Li SQ, Ye YL, Wang ZH, Huang JL, Tang QY, Sun YX, Peng XL, Zhang JW, He MR, Zhu YJ, Xue JQ, Wang G, Wu L, An N, Wu LQ, Ma L, Zhang WF, Zhang FS. 2014 Producing more grain with lower environmental costs. *Nature* **514**, 486-489. (doi:10.1038/nature13609)
15. Zhang X, Davidson EA, Mauzerall DL, Searchinger TD, Dumas P, Shen Y. 2015 Managing nitrogen for sustainable development. *Nature* **528**, 51-59. (doi:10.1038/nature15743)
16. Gu BJ, Lam SK, Reis S, van Grinsven H, Ju XT, Yan XY, Zhou F, Liu HB, Cai ZC, Galloway JN, Howard C, Sutton MA, Chen DL. 2019 Toward a Generic Analytical Framework for Sustainable Nitrogen Management: Application for China. *Environ. Sci. Technol.* **53**, 1109-1118. (doi:10.1021/acs.est.8b06370)
17. Howarth R, Ramakrishna K, Choi E, Elmgren R, Martinelli L, Mendoza A, Moomaw W, Palm C, Boy R, Scholes M. 2005 Chapter 9: Nutrient Management, Responses Assessment. Pages 295-311 in Ecosystems and Human Well-being, Volume 3, Policy Responses, the Millennium Ecosystem Assessment. Island Press, Washington, DC.
18. Liu XJ, Vitousek P., Chang Y.H., Zhang W.F., Matson P., Zhang F.S., 2016. Evidence for a historic change occurring in China. *Environ. Sci. Technol.*, **50**: 505-506. (doi: 10.1021/acs.est.5b05972)
19. Yang FM, *et al.* 2011 Characteristics of $PM_{2.5}$ speciation in representative megacities and across China. *Atmos. Chem. Phys.* **11**, 5207-5219. (doi:10.5194/acp-11- 5207-2011)
20. Huang RJ, *et al.* 2014 High secondary aerosol contribution to particulate pollution during haze events in China. *Nature* **514**, 218-222. (doi:10.1038/nature13774)
21. Huang X, *et al.* 2012 A high-resolution ammonia emission inventory in China. *Global Biogeochem. Cycles* **26**, GB1030. (doi:10.1029/2011GB004161)
22. Zhang XM, *et al.* 2017 Ammonia emissions may be substantially underestimated in China. *Environ. Sci. Technol.* **51**, 12089-12096. (doi:10.1021/acs.est.7b02171)
23. Zhang L, *et al.* 2018 Agricultural ammonia emissions in China: reconciling bottom-up and top-down estimates. *Atmos. Chem. Phys.* **18**, 339-355. (doi:10.5194/acp-18-339-2018)
24. Xu W, *et al.* 2015 Quantifying atmospheric nitrogen deposition through a nationwide monitoring network across China. *Atmos. Chem. Phys.* **15**, 12345-12360. (doi:10.5194/acp-15-12345-2015)
25. Xu W, Zhang L, Liu XJ. 2019a. A database of atmospheric nitrogen concentration and deposition from the nationwide monitoring network in China. *Sci. Data* **6**, 51. (doi:10.1038/s41597-019-0061-2)
26. Pan YP, *et al.* 2018 Identifying ammonia hotspots in China using a national observation network. *Environ. Sci. Technol.* **52**, 3926-3934. (doi:10.1021/acs.est.7b05235)
27. Shephard MW, *et al.* 2011 TES ammonia retrieval strategy and global observations of the spatial and seasonal variability of ammonia. *Atmos. Chem. Phys.* **11**, 10743-10763. (doi:10.5194/acp-11-10743-2011)
28. Warner JX, Dickerson RR, Wei Z, Strow LL, Wang Y, Liang Q. 2017 Increased atmospheric ammonia over the world's major agricultural areas detected from space. *Geophys. Res. Lett.* **44**, 2875-2884. (doi:10.1002/2016GL072305)
29. Van Damme M, Clarisse L, Whitburn S, Hadji-Lazaro J, Hurtmans D, Clerbaux C, Coheur P-F. 2018 Industrial and agricultural ammonia point sources exposed. *Nature* **564**, 99-103. (doi:10.1038/s41586-018-0747-1)
30. Kong L, *et al.* 2019 Improved Inversion of Monthly Ammonia Emissions in China Based on the Chinese

Ammonia Monitoring Network and Ensemble Kalman Filter. *Environ. Sci. Technol.* **53**, 12529-12538. (doi:10.1021/acs.est.9b02701)

31. Xu W, *et al.* 2017 Air quality improvement in a megacity: implications from 2015 Beijing Parade Blue pollution control actions. *Atmos. Chem. Phys.* **17**, 31-46. (doi:10.5194/acp-17-31-2017)
32. Pan YP, *et al.* 2016 Fossil fuel combustion-related emissions dominate atmospheric ammonia sources during severe haze episodes: Evidence from ^{15}N-stable isotope in size-resolved aerosol ammonium. *Environ. Sci. Technol.* **50**, 8049-8056. (doi:10.1021/acs.est.6b00634)
33. Chang Y, Zou Z, Zhang Y, Deng C, Hu J, Shi Z, Dore AJ, Collett Jr JL. 2019 Assessing contributions of agricultural and non-agricultural emissions to atmospheric ammonia in a Chinese megacity. *Environ. Sci. Technol.* **53**, 1822-1833. (doi: 10.1021/acs.est.8b05984)
34. Streets DG, *et al.* 2003 An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. *J. Geophys. Res. [Atmos.]* **108**, 8809. (doi:10.1029/2002JD003093)
35. Zheng B, *et al.* 2018 Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions. *Atmos. Chem. Phys.* **18**, 14095-14111. (doi:10.5194/acp-18-14095-2018)
36. Richter A, Burrows JP, Nüß H, Granier C, Niemeier U. 2005 Increase in tropospheric nitrogen dioxide over China observed from space. *Nature* **437**, 129-132. (doi:10.1038/nature04092)
37. Lin JT, McElroy MB, Boersma KF. 2010 Constraint of anthropogenic NO_x emissions in China from different sectors: a new methodology using multiple satellite retrievals. *Atmos. Chem. Phys.* **10**, 63-78. (doi:10.5194/acp-10-63-2010)
38. Geddes JA, Martin RV. 2017 Global deposition of total reactive nitrogen oxides from 1996 to 2014 constrained with satellite observations of NO_2 columns. *Atmos. Chem. Phys.* **17**, 10071-10091. (doi:10.5194/acp-17-10071-2017)
39. Lu X, *et al.* 2019 Exploring 2016-2017 surface ozone pollution over China: source contributions and meteorological influences. *Atmos. Chem. Phys.* **19**, 8339-8361. (doi:10.5194/acp-19-8339-2019)
40. Liu F, Zhang Q, Zheng B, Tong D, Yan L, Zheng YX, He KB. 2016 Recent reduction in NO_x emissions over China: synthesis of satellite observations and emission inventories. *Environ. Res. Lett.* **11**, 114002. (doi:10.1088/1748-9326/11/11/114002)
41. Liu MX, *et al.* 2019 Ammonia emission control in China would mitigate haze pollution and nitrogen deposition, but worsen acid rain. *Proc. Natl. Acad. Sci. U.S.A.* **116**, 7760-7765. (doi:10.1073/pnas.1814880116)
42. Xu ZY, *et al.* 2019. High efficiency of livestock ammonia emission controls in alleviating particulate nitrate during a severe winter haze episode in northern China. *Atmos. Chem. Phys.* **19**, 5605-5613. (doi:10.5194/acp-19-5605-2019)
43. Zhao YH, Zhang L, Chen YF, Liu XJ, Xu W, Pan YP, Duan L. 2017 Atmospheric nitrogen deposition to China: A model analysis on nitrogen budget and critical load exceedance. *Atmos. Environ.* **153**, 32-40. (doi:10.1016/j.atmosenv.2017.01.018)
44. Tang AH, Zhuang GS, Wang Y, Yuan H, Sun YL. 2005 The chemistry of precipitation and its relation to aerosol in Beijing. *Atmos. Environ.* **39**, 3397-3406. (doi:10.1016/j.atmosenv.2005.02.001)
45. China Ecological Environment Bulletin, 2018. The Ministry of Ecology and Environment of the People's Republic of China.
46. Li JL, Li R, Cui LL, Meng Y, Fu HB. 2019 Spatial and temporal variation of inorganic ions in rainwater in Sichuan province from 2011 to 2016. *Environ. Pollut.* **254**, 112941. (doi:10.1016/j.envpol.2019.07.109)

47. Huang K, Zhuang GS, Xu C, Wang Y, Tang AH. 2008 The chemistry of the severe acidic precipitation in Shanghai, China. *Atmos. Res.* **89**, 149-160. (doi:10.1016/j.atmosres.2008.01.006)

48. Meng Y, Zhao YL, Li R, Li JL, Cui LL, Kong LD, Fu HB. 2019 Characterization of inorganic ions in rainwater in the megacity of Shanghai: Spatiotemporal variations and source apportionment. *Atmos. Res.* **222**, 12-24. (doi:10.1016/j.atmosres.2019.01.023)

49. [49] Zhang FX. 2019 Spatial pattern and seasonal variation of alkaline precipitation observed in the Gansu Province, NW China. Environ. Earth Sci. **78**, 3-11. (doi: 10.1007/s12665-019-8442-6)

50. Yang F, Tan J, Shi ZB, Cai Y, He K, Ma Y, Duan F, Okuda T, Tanaka S, Chen G. 2012 Five-year record of atmospheric precipitation chemistry in urban Beijing, China. *Atmos. Chem. Phys.* **12**, 2025-2035. (doi:10.5194/acp-12-2025-2012)

51. Xu W, Wen Z, Shang B, Dore AJ, Tang AH, Xia XP, Zheng AH, Han MJ, Zhang L, Zhao YH. 2020 Precipitation chemistry and atmospheric nitrogen deposition at a rural site in Beijing, China. *Atmos. Environ.* **223**, 117253. (doi:10.1016/j.atmosenv.2019.117253)

52. Liu MX, Huang X, Song Y, Xu TT, Wang SX, Wu ZJ, Hu M, Zhang L, Zhang Q, Pan YP, Liu XJ, Zhu T. 2018 Rapid SO_2 emission reductions significantly increase tropospheric ammonia concentrations over the North China Plain. *Atmos. Chem. Phys.* **18**, 17933-17943. (doi:10.5194/acp-18-17933-2018)

53. Liu F, Beirle S, Zhang Q, van der AR, Zheng B, Tong D, He KB. 2017 NO_x emission trends over Chinese cities estimated from OMI observations during 2005 to 2015. *Atmos. Chem. Phys.* **17**, 9261-9275. (doi:10.5194/acp-17-9261-2017)

54. Riahi K, Grübler A, Nakicenovic N. 2007 Scenarios of long-term socio-economic and environmental development under climate stabilization. *Technol. Forecast. Soc. Change* **74**, 887-935. (doi:10.1016/j.techfore.2006.05.026)

55. Qiao X, Du J, Kota SH, Ying Q, Xiao WY, Tang Y. 2018 Wet deposition of sulfur and nitrogen in Jiuzhaigou National Nature Reserve, Sichuan, China during 2015–2016: possible effects from regional emission reduction and local tourist activities. *Environ. Pollut.* **233**, 267-277. (doi:10.1016/j.envpol.2017.08.041)

56. Calvo-Fernández J, Marcos E, Calvo L. 2017 Bulk deposition of atmospheric inorganic nitrogen in mountainous heathland ecosystems in North-Western Spain. *Atmos. Res.* **183**, 237-244. (doi:10.1016/j.atmosres.2016.09.006)

57. Jiang BF, Xia DH. 2017 Role identification of NH_3 in atmospheric secondary new particle formation in haze occurrence of China. *Atmos. Environ.* **163**, 107-117. (doi:10.1016/j.atmosenv.2017.05.035)

58. An ZS, et al. 2019 Severe haze in northern China: A synergy of anthropogenic emissions and atmospheric processes. *Proc. Natl. Acad. Sci. U.S.A.* **116**, 8657-8666. (doi:10.1073/pnas.1900125116)

59. Sun QH, Hong XR, Wold LE. 2010 Cardiovascular effects of ambient particulate air pollution exposure. *Circulation* **121**, 2755-2765. (doi:10.1161/CIRCULATIONAHA.109.893461)

60. Xing YF, Xu YH, Shi MH, Lian YX. 2016 The impact of $PM_{2.5}$ on the human respiratory system. *J. Thorac. Dis.* **8**, E69-74. (doi:10.3978/j.issn.2072-1439.2016.01.19)

61. Booth BB, Dunstone NJ, Halloran PR, Andrews T, Bellouin N. 2012 Aerosols implicated as a prime driver of twentieth-century North Atlantic climate variability. *Nature* **484**, 228-232. (doi:10.1038/nature10946)

62. Bergin M, Greenwald R, Xu J, Berta Y, Chameides W. 2001 Influence of aerosol dry deposition on photosynthetically active radiation available to plants: A case study in the Yangtze delta region of China. *Geophys. Res. Lett.* **28**, 3605-3608. (doi:10.1029/2001GL013461)

63. Zhou L, Chen X, Tian X. 2018 The impact of fine particulate matter ($PM_{2.5}$) on China's agricultural production

from 2001 to 2010. *J. Clean. Prod.* **178**, 133-141. (doi:10.1016/j.jclepro.2017.12.204)

64. Bell ML, Dominici F, Ebisu K, Zeger SL, Samet JM. 2007 Spatial and temporal variation in PM2. 5 chemical composition in the United States for health effects studies. *Environ. Health Perspect.* **115**, 989-995. (doi:10.1289/ehp.9621)
65. Jimenez JL, *et al.* 2009 Evolution of organic aerosols in the atmosphere. *Science* **326**, 1525-1529. (doi:10.1126/science.1180353)
66. Zhang RY, *et al.* 2015 Formation of urban fine particulate matter. *Chem. Rev.* **115**, 3803-3855. (doi:10.1021/acs.chemrev.5b00067)
67. Griffith SM, Huang XH, Louie P, Yu JZ. 2015 Characterizing the thermodynamic and chemical composition factors controlling $PM_{2.5}$ nitrate: Insights gained from two years of online measurements in Hong Kong. *Atmos. Environ.* **122**, 864-875. (doi:10.1016/j.atmosenv.2015.02.009)
68. Hoesly RM, et al. 2018 Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS). *Geosci. Model Dev.* **11**. (doi:10.5194/gmd-11-369-2018)
69. Zhang Q, Song Y, Li M, Zheng B. 2020 Chapter 2: Anthropogenic Emissions of SO_2, NO_x, and NH_3 in China. In *Atmospheric Reactive Nitrogen in China* (eds. Liu X.J. and Du E.Z.), Springer, Singapore pp. 13-40. (doi: 10.1007/978-981-13-8514-8_2)
70. Li HY, *et al.* 2018 Nitrate-driven urban haze pollution during summertime over the North China Plain. *Environ. Sci. Technol.* **18**, 5293-5306. (doi:10.5194/acp-18-5293-2018)
71. Wang Y, Zhang Q, He K, Zhang Q, Chai L. 2013 Sulfate-nitrate-ammonium aerosols over China: response to 2000-2015 emission changes of sulfur dioxide, nitrogen oxides, and ammonia. *Atmos. Chem. Phys.* **13**, 2635-2652. (doi:10.5194/acp-13-2635-2013)
72. Shao PY, *et al.* 2018 Characterizing remarkable changes of severe haze events and chemical compositions in multi-size airborne particles (PM_1, $PM_{2.5}$ and PM_{10}) from January 2013 to 2016–2017 winter in Beijing, China. *Atmos. Environ.* **189**, 133-144. (doi:10.1016/j.atmosenv.2018.06.038)
73. Wu YY, Gu BJ, Erisman JW, Reis S, Fang YY, Lu XH, Zhang XM. 2016 $PM_{2.5}$ pollution is substantially affected by ammonia emissions in China. *Environ. Pollut.* **218**, 86-94. (doi:10.1016/j.envpol.2016.08.027)
74. Plautz J. 2018 Piercing the haze. *Science* **361**, 1060-1063. (doi:10.1126/science.361.6407.1060)
75. Wang T, Xue L, Brimblecombe P, Lam YF, Li L, Zhang L. 2017 Ozone pollution in China: A review of concentrations, meteorological influences, chemical precursors, and effects. *Sci. Total Environ.* **575**, 1582-1596. (doi:10.1016/j.scitotenv.2016.10.081)
76. Cape J. 2008 Surface ozone concentrations and ecosystem health: past trends and a guide to future projections. *Sci. Total Environ.* **400**, 257-269. (doi:10.1016/j.scitotenv.2008.06.025)
77. Wang XP, Mauzerall DL. 2004 Characterizing distributions of surface ozone and its impact on grain production in China, Japan and South Korea: 1990 and 2020. *Atmos. Environ.* **38**, 4383-4402. (doi:10.1016/j.atmosenv.2004.03.067)
78. Feng ZZ, Hu EZ, Wang XK, Jiang LJ, Liu XJ. 2015 Ground-level O_3 pollution and its impacts on food crops in China: a review. *Environ. Pollut.* **199**, 42-48. (doi:10.1016/j.envpol.2015.01.016)
79. Lu X, Hong J, Zhang L, Cooper OR, Schultz MG, Xu XB, Wang T, Gao M, Zhao YH, Zhang YH. 2018 Severe surface ozone pollution in China: A global perspective. *Environ. Sci. Technol. Lett.* **5**, 487-494. (doi:10.1021/acs.estlett.8b00366)
80. Li R, Cui L, Li J, Zhao A, Fu HB, Wu Y, Zhang LW, Kong LD, Chen JM. 2017a Spatial and temporal variation of particulate matter and gaseous pollutants in China during 2014–2016. *Atmos. Environ.* **161**, 235-246.

(doi:10.1016/j.atmosenv.2017.05.008)

81. Zhu J, Liao H. 2016 Future ozone air quality and radiative forcing over China owing to future changes in emissions under the Representative Concentration Pathways (RCPs). *J. Geophys. Res. [Atmos.]* **121**, 1978-2001. (doi:10.1002/2015JD023926)
82. Feng ZZ, De Marco A, Anav A, Gualtieri M, Sicard P, Tian HQ, Fornasier F, Tao FL, Guo AH, Paoletti E. 2019 Economic losses due to ozone impacts on human health, forest productivity and crop yield across China. *Environ. Int.* **131**, 104966. (doi:10.1016/j.envint.2019.104966)
83. Feng ZZ, Sun JS, Wan WX, Hu EZ, Calatayud V. 2014 Evidence of widespread ozone-induced visible injury on plants in Beijing, China. *Environ. Pollut.* **193**, 296-301. (doi:10.1016/j.envpol.2014.06.004)
84. Shang B, Feng ZZ, Li P, Yuan XY, Xu YS, Calatayud V. 2017 Ozone exposure-and flux-based response relationships with photosynthesis, leaf morphology and biomass in two poplar clones. *Sci. Total Environ.* **603**, 185-195. (doi:10.1016/j.scitotenv.2017.06.083)
85. Hu TJ, Liu S, Xu YS, Feng ZZ, Calatayud V. 2020 Assessment of O_3-induced yield and economic losses for wheat in the North China Plain from 2014 to 2017, China. *Environ. Pollut.* **258**, 113828. (doi:10.1016/j.envpol.2019.113828)
86. Li P, Feng ZZ, Catalayud V, Yuan XY, Xu YS, Paoletti E. 2017b A meta-analysis on growth, physiological, and biochemical responses of woody species to ground-level ozone highlights the role of plant functional types. *Plant Cell Environ.* **40**, 2369-2380. (doi:10.1111/pce.13043)
87. Yue X, Unger N, Harper K, Xia XG, Liao H, Zhu T, Xiao JF, Feng ZZ, Li J. 2017 Ozone and haze pollution weakens net primary productivity in China. *Atmos. Chem. Phys.* **17**, 6073-6089. (doi:10.5194/acp-17-6073-2017)
88. Yu GR, *et al.* 2019 Stabilization of atmospheric nitrogen deposition in China over the past decade. *Nat. Geosci.* **12**, 424-429. (doi:10.1038/s41561-019-0352-4)
89. Li Y, *et al.* 2016 Increasing importance of deposition of reduced nitrogen in the United States. *Proc. Natl. Acad. Sci. USA* **113**, 5874-5879. (doi: 10.1073/pnas.1525736113)
90. Matson P, Lohse KA, Hall SJ. 2002 The globalization of nitrogen deposition: consequences for terrestrial ecosystems. *Ambio* **31**, 113-119. (doi:10.1579/0044-7447-31.2.113)
91. Liu XJ, et al. 2011 Nitrogen deposition and its ecological impact in China: An overview. *Environ. Pollut.* **159**, 2251-2264. (doi:10.1016/j.envpol.2010.08.002)
92. Zhu JX, He NP, Wang QF, Yan GF, Wen D, Yu GR, Jia YL. 2015 The composition, spatial patterns, and influencing factors of atmospheric wet nitrogen deposition in Chinese terrestrial ecosystems. *Sci. Total Environ.* **511**, 777-785. (doi:10.1016/j.scitotenv.2014.12.038)
93. Zhang J, *et al.* 2019 Impacts of climate change and emissions on atmospheric oxidized nitrogen deposition over East Asia. *Atmos. Chem. Phys.* **19**, 887-900. (doi:10.5194/acp-19-887-2019)
94. Deng OP, Zhang SR, Deng LJ, Lan T, Luo L, Gao XS. 2019 Atmospheric dry nitrogen deposition and its relationship with local land use in a high nitrogen deposition region. *Atmos. Environ.* **203**, 114-120. (doi:10.1016/j.atmosenv.2018.12.037)
95. Xu W, *et al.* 2018 Spatial–temporal patterns of inorganic nitrogen air concentrations and deposition in eastern China. *Atmos. Chem. Phys.* **18**, 10931-10954. (doi:10.5194/acp-18-10931-2018)
96. Han X, Zhang MG, Skorokhod A, Kou XX., 2017 Modeling dry deposition of reactive nitrogen in China with RAMS-CMAQ. *Atmos. Environ.* **166**, 47-61. (doi:10.1016/j.atmosenv.2017.07.015)
97. Zhang Y, Liu XJ, Fangmeier A, Goulding KTW, Zhang FS. 2008 Nitrogen inputs and isotopes in precipitation in

the North China Plain. *Atmos. Environ.* **42**, 1436-1448. (doi:10.1016/j.atmosenv.2007.11.002)

98. Wen Z, *et al.* 2020. Changes of nitrogen deposition in China during 1980 and 2018. *Environ. Int.*, in review.
99. Liu XJ, *et al.* 2017 Atmospheric nitrogen emission, deposition and air quality impacts in China: An overview. *Current Pollut. Reports* **3**, 65-77. (doi:10.1007/s40726-017-0053-9)
100. De Vries W, Hettelingh JP. Posch M. 2015 Critical loads and dynamic risk assessments: nitrogen, acidity and metals in terrestrial and aquatic ecosystems. Dordrecht, Springer. (doi:10.1007/978-94-017-9508-1)
101. Tian DS, Niu SL. 2015 A global analysis of soil acidification caused by nitrogen addition. *Environ. Res. Lett.* **10**, 024019. (doi:10.1088/1748-9326/10/2/024019)
102. Yang YH, Ji CJ, Ma WH, Wang SF, Wang SP, Han WX, Mohammat A, Robinson D, Smith P. 2012 Significant soil acidification across northern China's grasslands during 1980s-2000s. *Global Change Biol.* **18**, 2292-2300. (doi:10.1111/j.1365-2486.2012.02694.x)
103. Zhu QC, de Vries W, Liu XJ, Zeng MF, Hao TX, Du EZ, Zhang FS, Shen JB. 2016 The contribution of atmospheric deposition and forest harvesting to forest soil acidification in China since 1980. *Atmos. Environ.* **146**, 215-222. (doi:10.1016/j.atmosenv.2016.04.023)
104. Bowman WD, Cleveland CC, Halada Ĺ, Hreško J, Baron JS. 2008 Negative impact of nitrogen deposition on soil buffering capacity. *Nat. Geosci.* **1**, 767-770. (doi:10.1038/ngeo339)
105. Goulding K. 2016 Soil acidification and the importance of liming agricultural soils with particular reference to the United Kingdom. *Soil Use Manage.* **32**, 390-399. (doi:10.1111/sum.12270)
106. Hao TX, Zhu QC, Zeng MF, Shen JB, Shi XJ, Liu XJ, Zhang FS, de Vries W. 2019 Quantification of the contribution of nitrogen fertilization and crop harvesting to soil acidification in a wheat-maize double cropping system. *Plant Soil* **434**, 167-184. (doi:10.1007/s11104-018-3760-0)
107. Zhu QC, de Vries W, Liu XJ, Hao TX, Zeng MF, Shen JB, Zhang FS. 2018a Enhanced acidification in Chinese croplands as derived from element budgets in the period 1980–2010. *Sci. Total Environ.* **618**, 1497-1505. (doi:10.1016/j.scitotenv.2017.09.289)
108. Whitburn S, Van Damme M, Clarisse L, Bauduin S, Heald C, Hadji-Lazaro J, Hurtmans D, Zondlo MA, Clerbaux C, Coheur PF. 2016 A flexible and robust neural network IASI-NH_3 retrieval algorithm. *J. Geophys. Res. [Atmos.]* **121**, 6581-6599. (doi:10.1002/2016JD024828)
109. Kochian LV, Hoekenga OA, Piñeros MA. 2004 How do crop plants tolerate acid soils? Mechanisms of aluminum tolerance and phosphorous efficiency. *Annu. Rev. Plant Biol.* **55**, 459-493. (doi:10.1146/annurev.arplant.55.031903.141655)
110. Zhu QC, Liu XJ, Hao TX, Zeng MF, Shen JB, Zhang FS, De Vries W. 2018b Modeling soil acidification in typical Chinese cropping systems. *Sci. Total Environ.* **613**, 1339-1348. (doi:10.1016/j.scitotenv.2017.06.257)
111. Zhu QC, Liu XJ, Hao TX, Zeng MF, Shen JB, Zhang FS, de Vries W. 2020 Cropland acidification increases risk of yield losses and food insecurity in China. *Environ. Pollut.* **256**, 113145. (doi:10.1016/j.envpol.2019.113145)
112. Ju XT, Xing GX, Chen XP, Zhang SL, Zhang LJ, Liu XJ, Cui ZL, Yin B, Christie P, Zhu ZL, Zhang FS. 2009 Reducing environmental risk by improving N management in intensive Chinese agricultural systems. *P. Natl. Acad. Sci. U.S.A.* **106**, 3041-3046. (doi:10.1073/pnas.0813417106)
113. Zeng MF, de Vries W, Bonten LT, Zhu QC, Hao TX, Liu XJ, Xu MG, Shi XJ, Zhang FS, Shen JB. 2017 Model-based analysis of the long-term effects of fertilization management on cropland soil acidification. *Environ. Sci. Technol.* **51**, 3843-3851. (doi:10.1021/acs.est.6b05491)
114. Sala OE, Chapin FS, Armesto JJ, Berlow E, Bloomfield J, Dirzo R, Huber-Sanwald E, Huenneke LF, Jackson RB,

Kinzig A. 2000 Global biodiversity scenarios for the year 2100. *Science* **287**, 1770-1774. (doi:10.1126/science.287.5459.1770)

115. Bobbink R, Hicks K, Galloway J, Spranger T, Alkemade R, Ashmore M, Bustamante M, Cinderby S, Davidson E, Dentener F. 2010 Global assessment of nitrogen deposition effects on terrestrial plant diversity: a synthesis. *Ecol. Appl.* **20**, 30-59. (doi:10.1890/08-1140.1)
116. Du EZ, Lu XK, Tian D, Mao QG, Jing X, Wang C, Xia N. 2020 Chapter 9: Impacts of Nitrogen Deposition on Forest Ecosystems in China. In *Atmospheric Reactive Nitrogen in China* (eds. Liu X.J. and Du E.Z.). Springer, Singapore pp. 185-213. (doi:10.1007/978-981-13-8514-8_9)
117. Du EZ. 2017 Integrating species composition and leaf nitrogen content to indicate effects of nitrogen deposition. *Environ. Pollut.* **221**, 392-397. (doi:10.1016/j.envpol.2016.12.001)
118. Xing AJ, Xu LC, Shen HH, Du EZ, Liu XY, Fang JY. 2019 Long term effect of nitrogen addition on understory community in a Chinese boreal forest. *Sci. Total Environ.* **646**, 989-995. (doi:10.1016/j.scitotenv.2018.07.350)
119. Lu XK, Mo JM, Gilliam FS, Zhou GY, Fang YT. 2010 Effects of experimental nitrogen additions on plant diversity in an old-growth tropical forest. *Global Change Biol.* **16**, 2688-2700. (doi:10.1111/j.1365-2486.2010.02174.x)
120. Han WJ, Cao JY, Liu JL, Jiang J, Ni J. 2019 Impacts of nitrogen deposition on terrestrial plant diversity: a meta-analysis in China. *J. Plant Ecol.* **12**, 1025-1033. (doi:10.1093/jpe/rtz036)
121. Elser JJ, Andersen T, Baron JS, Bergström A-K, Jansson M, Kyle M, Nydick KR, Steger L, Hessen DO. 2009 Shifts in lake N: P stoichiometry and nutrient limitation driven by atmospheric nitrogen deposition. *Science* **326**, 835-837. (doi:10.1126/science.1176199)
122. Shou WW, Zong HB, Ding PX, Hou LJ. 2018 A modelling approach to assess the effects of atmospheric nitrogen deposition on the marine ecosystem in the Bohai Sea, China. *Estuarine Coastal Shelf Sci.* **208**, 36-48. (doi:10.1016/j.ecss.2018.04.025)
123. Bergstrom AK, Jansson M. 2006 Atmospheric nitrogen deposition has caused nitrogen enrichment and eutrophication of lakes in the northern hemisphere. *Global Change Biol.* **12**, 635-643. (doi:10.1111/j.1365-2486.2006.01129.x)
124. Ti CP, Gao B, Luo YX, Wang SW, Chang SX, Yan XY. 2018 Dry deposition of N has a major impact on surface water quality in the Taihu Lake region in southeast China. *Atmos. Environ.* **190**, 1-9. (doi:10.1016/j.atmosenv.2018.07.017)
125. Zhan XY, *et al.* 2017 Evidence for the importance of atmospheric nitrogen deposition to eutrophic lake Dianchi, China. *Environ. Sci. Technol.* **51**, 6699-6708. (doi:10.1021/acs.est.6b06135)
126. Zhang Y, Liu CM, Liu XJ, Xu W, Wen Z. 2019 Atmospheric nitrogen deposition around the Dongting Lake, China. *Atmos. Environ.* **207**, 197-204. (doi:10.1016/j.atmosenv.2019.03.034)
127. Qi JH, Shi JH, Gao HW, Sun Z. 2013 Atmospheric dry and wet deposition of nitrogen species and its implication for primary productivity in coastal region of the Yellow Sea, China. *Atmos. Environ.* **81**, 600-608. (doi:10.1016/j.atmosenv.2013.08.022)
128. Luo XS, *et al.* 2014 Chinese coastal seas are facing heavy atmospheric nitrogen deposition. *Environ. Res. Lett.* **9**, 095007. (doi:10.1088/1748-9326/9/9/095007)
129. Cui DY, Wang JT, Tan LJ, Dong ZY. 2016 Impact of atmospheric wet deposition on phytoplankton community structure in the South China Sea. *Estuarine Coastal Shelf Sci.* **173**, 1-8. (doi:10.1016/j.ecss.2016.02.011)
130. Shen JL, Liu JY, Li Y, Li YY, Wang Y, Liu XJ, Wu JS. 2014 Contribution of atmospheric nitrogen deposition to

diffuse pollution in a typical hilly red soil catchment in southern China. *J. Environ. Sci.* **26**, 1797-1805. (doi:10.1016/j.jes.2014.06.026)

131. Ju XT, Gu BJ, Wu YY, Galloway JN. 2016 Reducing China's fertilizer use by increasing farm size. *Global Environ. Chang.* **41**, 26-32. (doi:10.1016/j.gloenvcha.2016.08.005)

132. Wu YY, Xi XC, Tang X, Luo DM, Gu BJ, Lam SK, Vitousek PM, Chen DL. 2018 Policy distortions, farm size, and the overuse of agricultural chemicals in China. *P. Natl. Acad. Sci. U.S.A.* **115**, 7010-7015. (doi:10.1073/pnas.1806645115)

133. Bai ZH, *et al.* 2018 China's livestock transition: Driving forces, impacts, and consequences. *Sci. Adv.* **4**, eaar8534. (doi:10.1126/sciadv.aar8534)

134. Zhang CZ, Liu S, Wu SX, Jin SQ, Reis S, Liu HB, Gu BJ. 2019 Rebuilding the linkage between livestock and cropland to mitigate agricultural pollution in China. *Resour. Conserv. Recycl.* **144**, 65-73.(doi:10.1016/j.resconrec.2019.01.011)

135. Fan LC, Yuan YM, Ying ZC, Lam SK, Liu L, Zhang XC, Liu HB, Gu BJ. 2019 Decreasing farm number benefits the mitigation of agricultural non-point source pollution in China. *Environ. Sci. Pollut. Res.* **26**, 464-472. (doi:10.1007/s11356-018-3622-6)

136. Ren CC, Liu S, Van Grinsven H, Reis S, Jin SQ, Liu HB, Gu BJ. 2019 The impact of farm size on agricultural sustainability. *J. Clean. Prod.* **220**, 357-367. (doi:10.1016/j.jclepro.2019.02.151)

137. Gu BJ, Zhang XL, Bai XM, Fu BJ, Chen DL. 2019 Four steps to food security for swelling cities. *Nature* **566**, 31-33. (doi:10.1038/d41586-019-00407-3)

138. Gu BJ, Ju XT, Chang J, Ge Y, Vitousek PM. 2015 Integrated reactive nitrogen budgets and future trends in China. *Proc. Natl. Acad. Sci. U.S.A.* **112**, 8792-8797. (doi:10.1073/pnas.1510211112)

139. Zhang Q, *et al.* 2007 NO_x emission trends for China, 1995–2004: The view from the ground and the view from space. *J. Geophys. Res. [Atmos.]* **112**, D22306. (doi:10.1029/2007JD008684)

140. Ohara T, Akimoto H, Kurokawa J, Horii N, Yamaji K, Yan XY, Hayasaka T. 2007 An Asian emission inventory of anthropogenic emission sources for the period 1980-2020. *Atmos. Chem. Phys.* **7**. (doi:10.5194/acp-7-4419-2007)

141. Lei Y, Zhang Q, Nielsen C, He KB. 2011 An inventory of primary air pollutants and CO2 emissions from cement production in China, 1990-2020. *Atmos. Environ.* **45**, 147-154. (doi:10.1016/j.atmosenv.2010.09.034)

142. Zhao B, Wang SX, Liu H, Xu JY, Fu K, Klimont Z, Hao JM, He KB, Cofala J, Amann M. 2013 NOx emissions in China: historical trends and future perspectives. *Atmos. Chem. Phys.* **13**, 9869-9897. (doi:10.5194/acp-13-9869-2013)

143. Zhao Y, Duan L, Xing J, Larssen T, Nielsen C, Hao JM. 2009 Soil Acidification in China: Is Controlling SO_2 Emissions Enough? *Environ. Sci. Technol.* **43**, 8021-8026. (doi:10.1021/es901430n)

144. China State Council (CSC). 2011 The 12th Five-Year Plan on Energy Saving and Emission Reduction, China State Council, Beijing, China. (http://www.gov.cn/zwgk/2011-12/20/content_2024895.htm)

145. China State Council (CSC). 2013 Action Plan on Prevention and Control of Air Pollution, China State Council, Beijing, China, (http://www.gov.cn/zwgk/2013-09/12/content_2486773.htm)

146. Ministry of Environmental Protection in China (MEP). 2011 Emission Standard of Air Pollutants for Thermal Power Plants, GB 13223-2011, China Environmental Science Press, Beijing, China.

147. China Association of Environmental Protection Industry (CAEPI). 2017 Development of Desulfurization and Denitration Industries in 2015, Beijing, China (in Chinese).

148. Ministry of Environmental Protection in China (MEP). 2015 Work Plan for Fully Implementing Ultra-low Emissions and Energy Conservation in Coal-Fired Power Plants, Beijing, China. [148]

149. Clean Air Asia (CAA). 2019 Chinese Atmosphere 2019. (http://www.cleanairasia.org)

150. Ministry of Ecology and Environment (MEE). 2019a Opinions on Promoting the Implementation of Ultra-low Emissions in the Steel Industry, Beijing, China.

151. Wu Y, Zhang SJ, Hao JM, Liu H, Wu XM, Hu JN, Walsh MP, Wallington TJ, Zhang KM, Stevanovic S. 2017 On-road vehicle emissions and their control in China: A review and outlook. *Sci. Total Environ.* **574**, 332-349. (doi:10.1016/j.scitotenv.2016.09.040)

152. Ministry of Ecology and Environment (MEE). 2019b China Mobile Source Environmental Management Annual Report, Beijing, China.

153. Ministry of Transport (MOT) (2018) Notice on Printing and Distributing the Implementation Plan of Ship's Air Pollutant Emission Control Zone, (http://www.gov.cn/xinwen/2018-12/20/content_5350451.htm)

154. China State Council (CSC). 2018 Three-year Action Plan on Defending the Blue Sky, China State Council, Beijing, China, (http://www.gov.cn/zhengce/content/2018-07/03/content_5303158.htm)

155. Song W, et al. A multiyear assessment of air quality benefits from China's emerging shale gas revolution: Urumqi as a case study. *Environ. Sci. Technol.* **49**, 2066-2072. (doi:10.1021/es5050024)

156. Shen JL, Tang AH, Liu XJ, Kopsch J, Fangmeier A, Goulding K, Zhang FS. 2011 Impacts of pollution controls on air quality in Beijing during the 2008 Olympic Games. *J. Environ. Qual.* **40**, 37-45. (doi:10.2134/jeq2010.0360)

157. Chang Y, Liu XJ, Deng CR, Dore AJ, Zhuang GS. 2016 Source apportionment of atmospheric ammonia before, during, and after the 2014 APEC summit in Beijing using stable nitrogen isotope signatures. *Atmos. Chem. Phys.* **16**, 11635-11647. (doi: 10.5194/acp-2016-432)

158. Chang Y, *et al.* 2020 The puzzle of haze events during the Chinese New Year Holiday of the coronavirus (COVID-19) outbreak. *JGR-Atmosphere*, in review.

159. Zhang WF, *et al.* 2016 Closing yield gaps in China by empowering smallholder farmers. *Nature* **537**, 671-674. (doi:10.1038/nature19368)

160. Liu XJ, Xu W, Sha ZP, Zhang YY, Wen Z, Wang JX, Zhang FS, Goulding K. 2020 A green eco-environment for sustainable development: Framework and action. *Front. Agr. Sci. Eng.* **7**, 67-74. (doi:10.15302/J-FASE-2019297)

161. Xu W, Wu QH, Liu XJ, Tang AH, Dore AJ, Heal MR. 2016 Characteristics of ammonia, acid gases and $PM_{2.5}$ for three typical land use types in the North China Plain. *Environ. Sci. Pollut. Res.* **23**, 1158-1172. (doi:10.1007/s11356-015-5648-3)

162. Zhang YY, Benedict KB, Tang AH, Sun YL, Fang YT, Liu XJ. 2020 Persistent Non-agricultural and Periodic Agricultural Emissions Dominate Sources of Ammonia in Urban Beijing: Evidence from ^{15}N Stable Isotope in Vertical Profiles. *Environ. Sci. Technol.* **54**, 102-109. (doi:10.1021/acs.est.9b05741)

Figure Caption

Figure 1. Nitrogen emissions, deposition, impacts and regulation in China.

Figure 2. Annual NH_3 and NO_x emissions averaged for the years 2008-2012. The numbers are annual emission totals for China (Adapted from Zhao *et al.* [43]).

Figure 3. Spatial patterns of atmospheric wet and dry deposition of various N species over China, averaged for 2011–2015 (Adapted from Yu *et al.* [88]).

Figure 4. Anthropogenic emissions of NO_x in China by sector and year (Adapted from Zheng *et al.* [35]).

Figure 5. Implementation steps of China's atmospheric environmental protection policies and measures particularly related to atmospheric Nr emission control and air quality improvement (Modified from Wen *et al.* [98]).

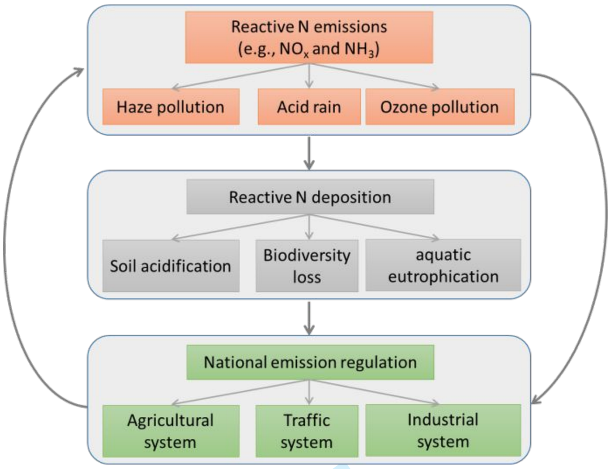
Reactive N emissions
(e.g., NO_x and NH_3)
Haze pollution
Acid rain
Ozone pollution
Reactive N deposition
Soil acidification
Biodiversity loss
aquatic eutrophication
National emission regulation
Agricultural system
Traffic system
Industrial system

Figure 1.

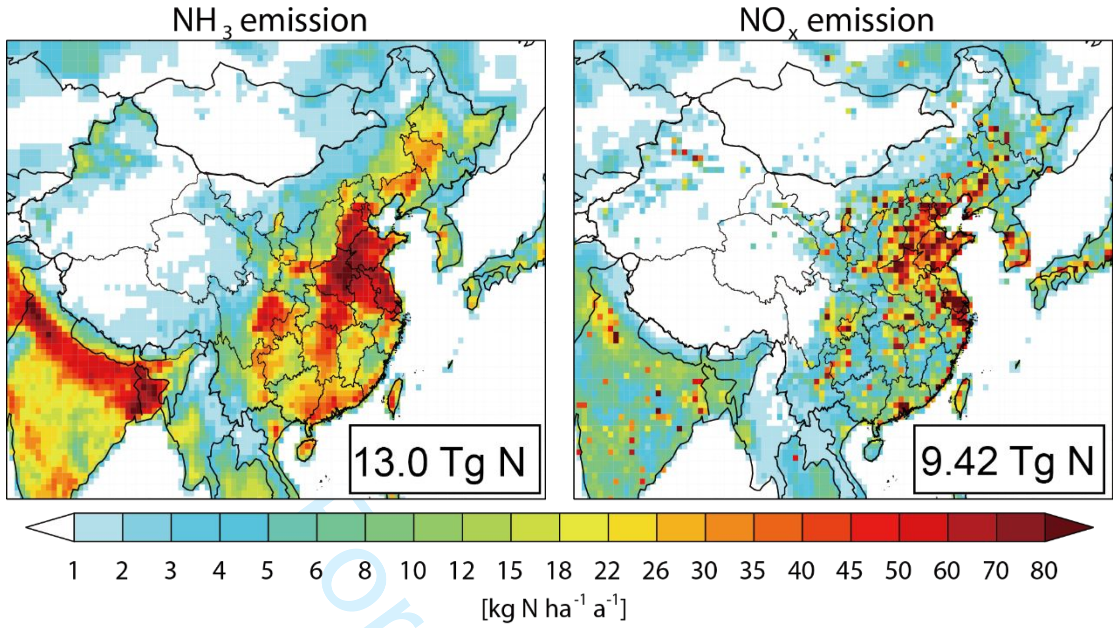
NH3 emission
NOx emission
13.0 Tg N
9.42 Tg N
1 2 3 4 5 6 8 10 12 15 18 22 26 30 35 40 45 50 60 70 80
[kg N ha-1 a-1]

Figure 2.

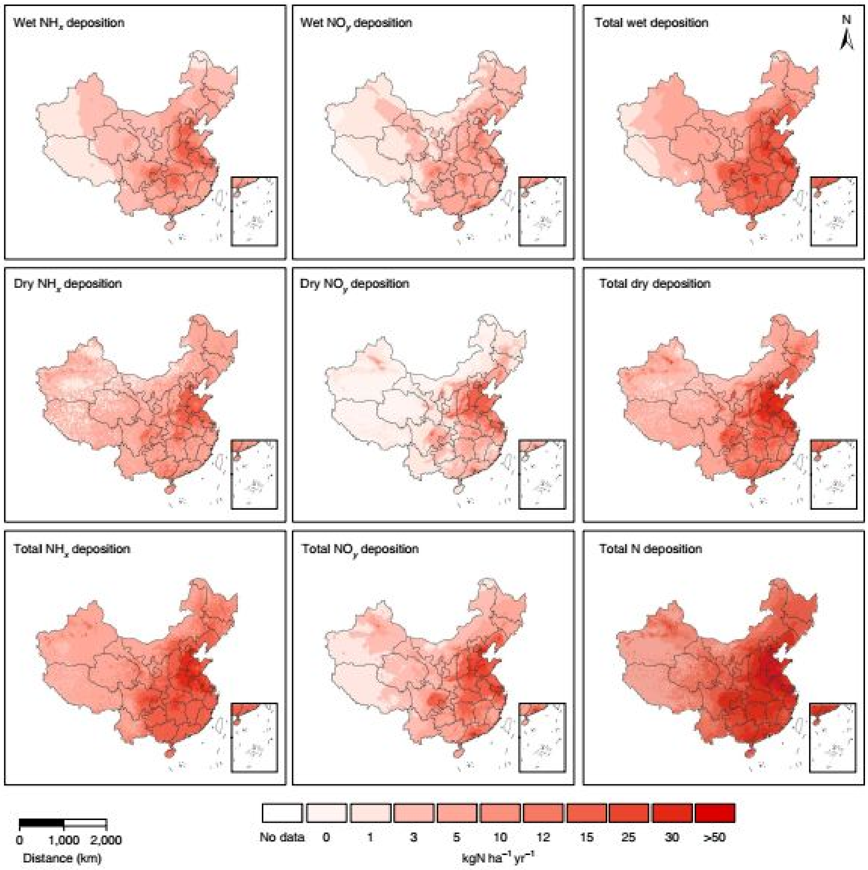
Wet NH_x deposition
Wet NO_y deposition
Total wet deposition
N
Dry NH_x deposition
Dry NO_y deposition
Total dry deposition
Total NH_x deposition
Total NO_y deposition
Total N deposition
0 1,000 2,000
Distance (km)
No data 0 1 3 5 10 12 15 25 30 >50
kgN ha^{-1} yr^{-1}

Figure 3.

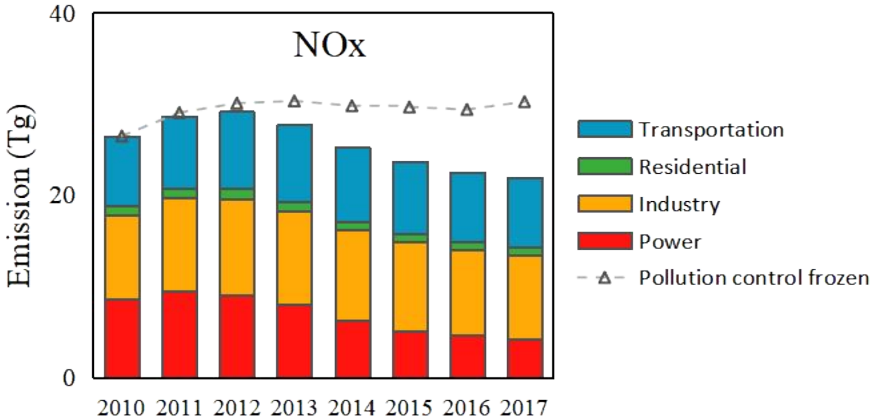
NOx
Emission (Tg)
40
20
0
2010 2011 2012 2013 2014 2015 2016 2017
Transportation
Residential
Industry
Power
Pollution control frozen

Figure 4.

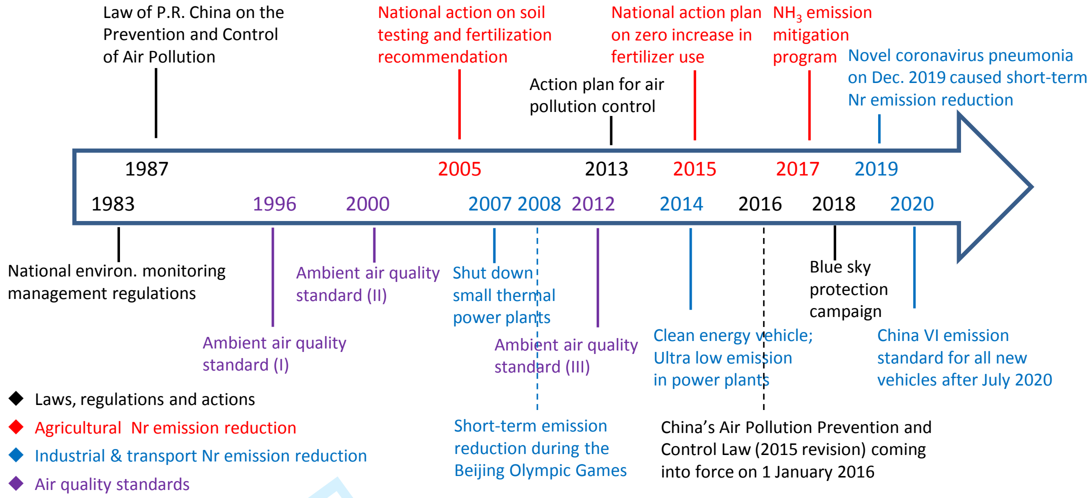
Law of P.R. China on the Prevention and Control of Air Pollution
National action on soil testing and fertilization recommendation
Action plan for air pollution control
National action plan on zero increase in fertilizer use
NH_3 emission mitigation program
Novel coronavirus pneumonia on Dec. 2019 caused short-term Nr emission reduction
1987
2005
2013
2015
2017
2019
1983
1996
2000
2007 2008
2012
2014
2016
2018
2020
National environ. monitoring management regulations
Ambient air quality standard (II)
Shut down small thermal power plants
Blue sky protection campaign
Ambient air quality standard (I)
Ambient air quality standard (III)
Clean energy vehicle; Ultra low emission in power plants
China VI emission standard for all new vehicles after July 2020
Laws, regulations and actions
Agricultural Nr emission reduction
Industrial & transport Nr emission reduction
Air quality standards
Short-term emission reduction during the Beijing Olympic Games
China's Air Pollution Prevention and Control Law (2015 revision) coming into force on 1 January 2016

Figure 5.