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The role of atmospheric reactive nitrogen in environmental issues in China

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The role of atmospheric reactive nitrogen in environmental issues in China

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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

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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

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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 ($\mu g\ m^{-3}$) 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

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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

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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

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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

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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

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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.

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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 $\mu g \ m^{-3}$) to January 2014 (79 $\mu g \ m^{-3}$) [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.

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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]).

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Figure 1.

Figure 2.

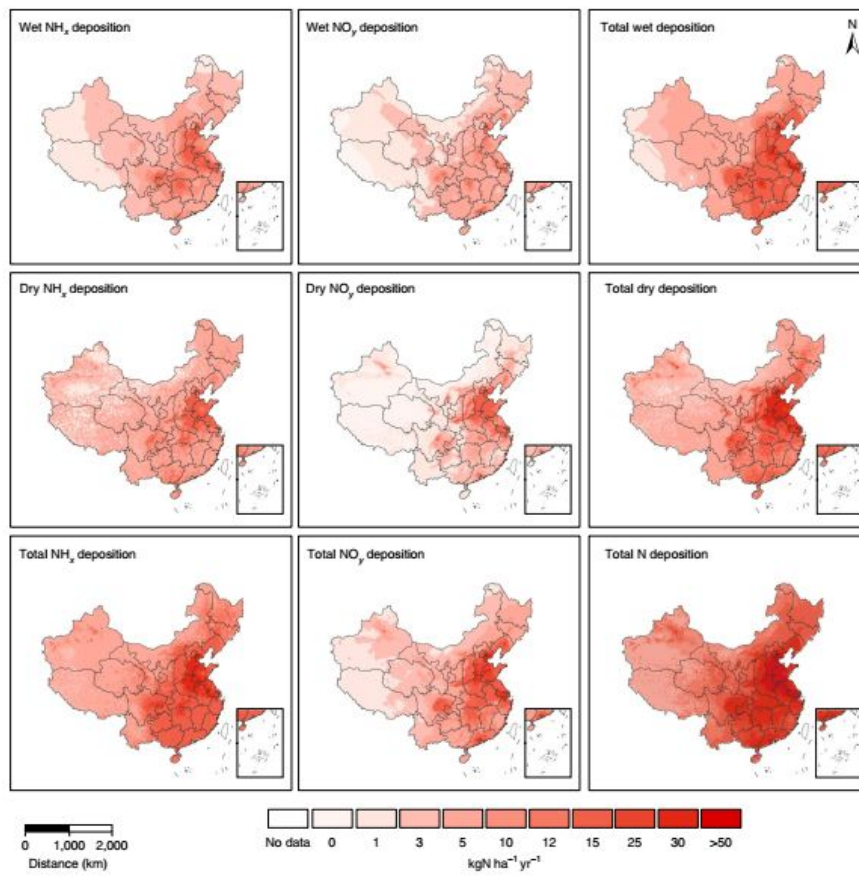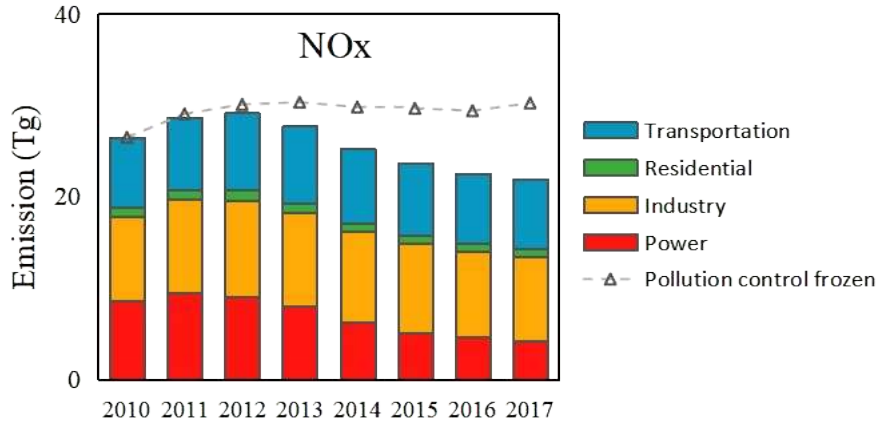Submitted to Phil. Trans. R. Soc. A - Issue

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Figure 3.

Figure 4.

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Figure 5.