Ammonia emission control in China would mitigate haze pollution and nitrogen deposition, but worsen acid rain

Mingxu Liu[a,1], Xin Huang[b,1], Yu Song[a,2], Jie Tang[c], Junji Cao[d,e], Xiaoye Zhang[f], Qiang Zhang[g], Shuxiao Wang[h], Tingting Xu[a], Ling Kang[a], Xuhui Cai[a], Hongsheng Zhang[i], Fumo Yang[j], Huanbo Wang[k], Jian Zhen Yu[l], Alexis K. H. Lau[m], Lingyan He[n], Xiaofeng Huang[n], Lei Duan[h], Aijun Ding[b], Likun Xue[o], Jian Gao[p], Bin Liu[q], and Tong Zhu[a,r,2]

[a]State Key Joint Laboratory of Environmental Simulation and Pollution Control, Department of Environmental Science, Peking University, 100871 Beijing, China; [b]Joint International Research Laboratory of Atmospheric and Earth System Sciences, School of Atmospheric Sciences, Nanjing University, 210023 Nanjing, China; [c]Meteorological Observation Center, China Meteorological Administration, 100081 Beijing, China; [d]State Key Laboratory of Loess and Quaternary Geology, Institute of Earth Environment, Chinese Academy of Sciences, 710061 Xi'an, China; [e]Key Laboratory of Aerosol Chemistry and Physics, Institute of Earth Environment, Chinese Academy of Sciences, 710061 Xi'an, China; [f]State Key Laboratory of Severe Weather, Key Laboratory of Atmospheric Chemistry of China Meteorological Administration, Chinese Academy of Meteorological Sciences, 100081 Beijing, China; [g]Ministry of Education Key Laboratory for Earth System Modeling, Center for Earth System Science, Institute for Global Change Studies, Tsinghua University, 100084 Beijing, China; [h]State Key Joint Laboratory of Environmental Simulation and Pollution Control, School of Environment, Tsinghua University, 100084 Beijing, China; [i]Laboratory for Atmosphere-Ocean Studies, Department of Atmospheric and Oceanic Science, School of Physics, Peking University, 100871 Beijing, China; [j]National Engineering Research Center for Flue Gas Desulfurization, Department of Environmental Science and Engineering, Sichuan University, 610065 Chengdu, China; [k]School of Environment and Resource, Southwest University of Science and Technology, 621010 Mianyang, China; [l]Department of Chemistry, Hong Kong University of Science & Technology, Hong Kong, China; [m]Division of Environment & Sustainability, Hong Kong University of Science & Technology, Hong Kong, China; [n]Key Laboratory for Urban Habitat Environmental Science and Technology, School of Environment and Energy, Peking University Shenzhen Graduate School, 518055 Shenzhen, China; [o]Environment Research Institute, Shandong University, 250100 Jinan, China; [p]State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Science, 100012 Beijing, China; [q]Environmental Quality Forecast Center, China National Environmental Monitoring Center, 100012 Beijing, China; and [r]Beijing Innovation Center for Engineering Science and Advanced Technology, Peking University, 100871 Beijing, China

Edited by John H. Seinfeld, California Institute of Technology, Pasadena, CA, and approved March 5, 2019 (received for review August 29, 2018)

China has been experiencing fine particle (i.e., aerodynamic diameters ≤ 2.5 μm; $PM_{2.5}$) pollution and acid rain in recent decades, which exert adverse impacts on human health and the ecosystem. Recently, ammonia (i.e., NH_3) emission reduction has been proposed as a strategic option to mitigate haze pollution. However, atmospheric NH_3 is also closely bound to nitrogen deposition and acid rain, and comprehensive impacts of NH_3 emission control are still poorly understood in China. In this study, by integrating a chemical transport model with a high-resolution NH_3 emission inventory, we find that NH_3 emission abatement can mitigate $PM_{2.5}$ pollution and nitrogen deposition but would worsen acid rain in China. Quantitatively, a 50% reduction in NH_3 emissions achievable by improving agricultural management, along with a targeted emission reduction (15%) for sulfur dioxide and nitrogen oxides, can alleviate $PM_{2.5}$ pollution by 11–17% primarily by suppressing ammonium nitrate formation. Meanwhile, nitrogen deposition is estimated to decrease by 34%, with the area exceeding the critical load shrinking from 17% to 9% of China's terrestrial land. Nevertheless, this NH_3 reduction would significantly aggravate precipitation acidification, with a decrease of as much as 1.0 unit in rainfall pH and a corresponding substantial increase in areas with heavy acid rain. An economic evaluation demonstrates that the worsened acid rain would partly offset the total economic benefit from improved air quality and less nitrogen deposition. After considering the costs of abatement options, we propose a region-specific strategy for multipollutant controls that will benefit human and ecosystem health.

ammonia emission | China | PM2.5 | acid rain | nitrogen deposition

During recent decades, growing fossil fuel consumption resulting from rapid economic development and intense agricultural production has brought about huge emissions of multiple air pollutants and associated environmental issues in China: (*i*) acid rain, (*ii*) enhanced nitrogen deposition, and (*iii*) air quality deterioration caused by fine particulate matter (i.e., aerodynamic diameters ≤ 2.5 μm; $PM_{2.5}$). In the 1980s, acid rain was recognized as a top environmental issue in China; it was characterized by precipitation acidification with a pH less than 5.6 as a result of high levels of sulfur dioxide (i.e., SO_2) and nitrogen oxide (i.e., NO_x) emission from fossil fuel combustion (1–3). Moreover, large amounts of NH_3 emission from dense agricultural activities has made China a global hotspot for nitrogen deposition (4, 5). It is estimated that approximately 15% of the land area of China exceeds nitrogen critical load (6).

In recent years, China has been experiencing frequent haze pollution, which is characterized by extremely high $PM_{2.5}$ concentration and rapid deterioration of visibility (7, 8). In winter, severe haze pollution with $PM_{2.5}$ concentrations of hundreds of micrograms per cubic meter often engulfs tens of thousands

Significance

Atmospheric ammonia plays important roles in fine particle pollution, acid rain, and nitrogen deposition. China, known as the world's top emitter of gaseous ammonia, plans to control ammonia emissions to mitigate the haze pollution that has recently emerged. However, the complex side effects are still unclear. By integrating a chemical transport model, nationwide measurements, and a sophisticated ammonia emission model, we find that ammonia emission control would significantly aggravate acid rain pollution, thereby offsetting the benefit from reduced fine particle pollution and nitrogen deposition. Our work suggests that region-specific ammonia-control strategies provide a more rational and effective way to achieve the dual benefits of protecting human and ecosystem health in China.

Author contributions: Y.S. and T.Z. designed research; M.L., Xin Huang, Y.S., J.C., X.Z., T.X., L.K., L.D., A.D., L.X., and T.Z. performed research; M.L., Xin Huang, Y.S., J.T., X.Z., Q.Z., S.W., X.C., H.Z., F.Y., H.W., J.Z.Y., A.K.H.L., L.H., Xiaofeng Huang, J.G., and B.L. analyzed data; and M.L., Xin Huang, Y.S., J.T., and T.Z. wrote the paper.

The authors declare no conflict of interest.

This article is a PNAS Direct Submission.

[1]M.L. and Xin Huang contributed equally to this work.

[2]To whom correspondence may be addressed. Email: songyu@pku.edu.cn or tzhu@pku.edu.cn.

This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10.1073/pnas.1814880116/-/DCSupplemental.

Published online April 1, 2019.

of square kilometers and lasts for days or even weeks, drawing increasing attention from the public and the government. Many measurements are indicative of a remarkably high proportion (20–60%) of secondary inorganic aerosols in $PM_{2.5}$ mass concentration, including sulfate, nitrate, and ammonium, during haze pollution (9–11).

The Chinese government has devoted great effort to control SO_2 and NO_x emissions to mitigate acid rain and haze pollution (12, 13). In 1990s, China set out to control SO_2 emission for the purpose of mitigating acid rain pollution. Accordingly, SO_2 emissions significantly decreased during the 2000s as a result of widespread implementation of flue gas desulfurization (Fig. 1*A*). Measurements from ground and space also showed a notable decrease (>50%) of atmospheric SO_2 in the past decade (Fig. 1 *B–D*). As a result, acid rain pollution was gradually alleviated, even though it remains an important issue in the southern part of China (1, 14). In contrast, NO_x emission control started relatively late and has resulted in a moderate reduction of ∼20% from 2012 to 2015. Although air quality has been mitigated slightly in the past few years as a result of SO_2 and NO_x emission reduction, it is still a challenging problem facing China. According to a report issued by the government, 74.3% of 74 key cities in China exceeded the national air-quality standard of annual mean $PM_{2.5}$ concentrations (35 $\mu g \cdot m^{-3}$) in 2017 (15). To further improve the air quality, China will continue to implement stringent emission

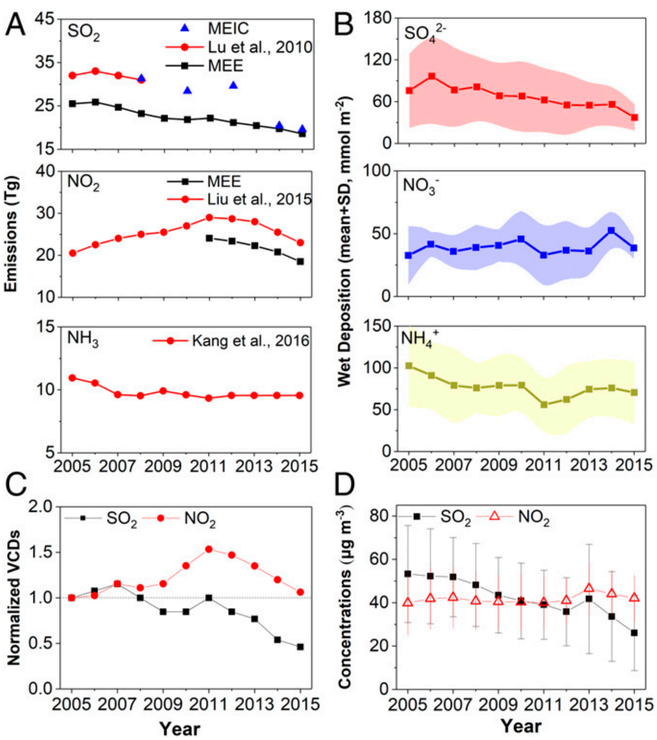

Fig. 1. (*A*) Interannual trends in SO_2, NO_x, and NH_3 emissions in China during 2005–2015. SO_2 emissions were provided by the Ministry of Ecology and Environment of China (MEE; www.mee.gov.cn/), Lu et al. (35), and Multi-resolution Emission Inventory for China; NOx emissions were derived from MEE and Liu et al. (12); and NH_3 emissions were derived from Kang et al. (25). (*B*) Interannual trends in wet deposition (mean ± SD) of sulfate, nitrate, and ammonium averaged over four Acid Deposition Monitoring Network in East Asia stations (Jinyunshan, Shizhan, Xiaoping, and Xiangzhou) in China. (*C*) Normalized vertical column densities (VCDs) of SO_2 and NO_2 retrieved from ozone monitoring instrument measurements over China relative to the 2005 levels. (*D*) Ground-based concentrations of SO_2 and NO_2 averaged over 31 cities in China. These annual averaged values for each city were provided by the China Environmental Statistics Yearbook (www.stats.gov.cn/).

controls on SO_2 and NO_x with the aim reduce their emissions by 15% during the period of 2016–2020 (16).

More recently in China, NH_3 control has been advocated as a potential measure by policy makers given that atmospheric NH_3 facilitates secondary $PM_{2.5}$ formation, i.e., ammonium sulfate/bisulfate and ammonium nitrate (17). The Chinese Premier recently announced that a new fund will be set up to discover the cause of frequent haze pollution, with special attention devoted to agricultural NH_3 emission control (18). In northern China, a highly polluted area, abatement options for NH_3 agricultural emissions have been integrated in the air pollution control plan for 2016–2020.

However, it should not be neglected that atmospheric NH_3 plays a vital role in neutralizing precipitation acidity and therefore helps to weaken acid rain (19, 20). Decrease in ambient NH_3 level is likely to induce more acidic precipitation and expose terrestrial systems to acid rain pollution, especially in southern and southwestern China, where soil systems are generally less buffered and particularly sensitive to acid input (21). Thus, a problem arises that, under the circumstance of tremendous emissions of acid gas (SO_2 + NO_x), China's NH_3 emission abatement may significantly worsen acid rain by increasing rainfall acidity. It is still unclear, and we are aware of no study that has quantified this. The impacts of NH_3 emissions are complex in China, with distinct effects on air quality and human and ecosystem health. Previous studies mainly focused on the role of NH_3 emission in particle pollution on global and regional scales, and there is still a lack of studies on these comprehensive impacts in China, particularly in terms of acid rain and nitrogen deposition.

In this study, we performed an integrated analysis of multiple impacts of NH_3 emission control strategy on $PM_{2.5}$ pollution, nitrogen deposition, and acid rain by combining the Weather Research and Forecasting model with Chemistry (WRF-Chem) simulation, ground-based and satellite-retrieved observations, as well as economic assessment. We have developed a high-resolution NH_3 emission model and used it to forecast the potential NH_3 emission reduction in the agricultural sector based on Chinese agricultural practice. We then intend to provide optimized and cost-effective strategies for China's NH_3 emission abatement policy in the near future.

NH_3 Emission and Its Potential Reduction in China

As a large agricultural country, China produces a huge amount of NH_3 emission (Fig. 1*A*), with agricultural activities accounting for more than 80% (22–24). In this study, we use a comprehensive high-resolution inventory developed by our research group (PKU-NH_3) that covers agricultural activities, biomass burning, industry, transportation, and other sectors (25). It has been demonstrated to accurately describe the magnitude and spatial and seasonal patterns of China's NH_3 emission and has been applied in chemical transport modeling to reproduce secondary inorganic aerosols (details are shown in the *SI Appendix*).

NH_3 emission in China (∼10.0 million tons annually) exceeds the sum of those in the European Union (3.7 million tons) and the United States (3.9 million tons) (24, 26). Moreover, the NH_3 emission rates per amount of fertilizer applied or animal excretion in farms are almost twice those in developed countries (*SI Appendix*, Table S2). Much evidence manifests that poor agricultural production management and low efficiency of nitrogen use are the main causes of the large NH_3 emissions over China (details are shown in the *SI Appendix*). The implementation of feasible abatement measures during agricultural activities could sharply reduce China's NH_3 emissions.

Here, a series of options for NH_3 emission reduction are compiled (*SI Appendix*, Table S1) based on existing literature and local agricultural practice. To abate NH_3 emission in croplands, rational fertilization (i.e., avoidance of overfertilization and implementation of deep application of fertilizers) can be implemented during growing seasons (mostly spring and summer months) when emission from nitrogen fertilizers peaks. For livestock NH_3 emissions, an optimization of animal manure management should be promoted. Based on the PKU-NH_3 emission model, it is estimated that a combination of the aforementioned measures can reduce China's NH_3 emissions by 30–50%. In this work, we choose a 50% NH_3 emission abatement strategy to examine its multiple environmental impacts, which can be taken as an upper bound of NH_3 emission reduction via feasible control options in the near future. In this case, the national annual emissions changed from 3.8 to 1.8 million tons during summer (June, July, and August; hereafter referred to as JJA) and from 1.4 to 0.7 million tons during winter (January, February, and December; referred to as DJF hereafter). The unit emission rates in fertilizer use and animal excretion are comparable with those in developed countries (*SI Appendix*, Table S2).

To quantitatively understand multiple impacts of NH_3 emission control, we performed WRF-Chem simulations in the summer months of 2015 with frequent rainfall and in the winter months with frequent haze pollution. Three parallel numerical experiments with different emission scenarios are designed: (*i*) standard emission levels in 2015 (Run_base), (*ii*) the same as (*i*) but reducing SO_2 and NO_x emissions by 15% on a national scale (Run_15SN; target for 2020), and (*iii*) the same as (*ii*) but with an additional 50% NH_3 emission reduction (Run_15SN50A). We validated the model by comparisons with nationwide measurements of gaseous NH_3 concentrations, $PM_{2.5}$ mass concentrations, particle chemical compositions, and rainfall acidity (*SI Appendix*, Figs. S3–S6 and Tables. S3–S5). After ensuring model performance, multiple environmental effects of NH_3 emission reduction could be disentangled by disparities among the simulations based on different emission scenarios.

Results and Discussion

Multiple Impacts of NH_3 Emission Control. China's huge anthropogenic emissions lead to severe particle pollution (Run_base), as shown in Fig. 2. In northern China, south-central China, and the Sichuan Basin (the geographical areas of interest in this study are shown in *SI Appendix*, Fig. S2), mean $PM_{2.5}$ concentrations exceed 75 $\mu g \cdot m^{-3}$, far greater than the Chinese Ambient Air Quality Standard of 35 $\mu g \cdot m^{-3}$ (interim target 1 set by World Health Organization) that would lead to potential public health risks. If China achieves its SO_2 and NO_x emission reduction targets as planned (15% reduction during 2015–2020; Run_15SN), $PM_{2.5}$ concentrations will decrease by 1–3 $\mu g \cdot m^{-3}$, or only ~3% of the baseline level (Fig. 2*B*). However, an additional 50% NH_3 emissions control (i.e., Run_15SN50A) can accomplish a decrease of 11–17% in $PM_{2.5}$ concentrations relative to the 2015 baseline in these three regions (Fig. 2*C*). Substantial decline in the average $PM_{2.5}$ concentrations (~10 $\mu g \cdot m^{-3}$) primarily occurs in southwestern, south-central, and parts of northern China, where $PM_{2.5}$ concentrations already exceed the air quality standard and NH_3 concentrations are relatively high. Clearly, synchronous NH_3 emission control is a more efficient way to mitigate $PM_{2.5}$ pollution, rather than merely implementing SO_2 and NO_x emission controls (i.e., Run_15SN).

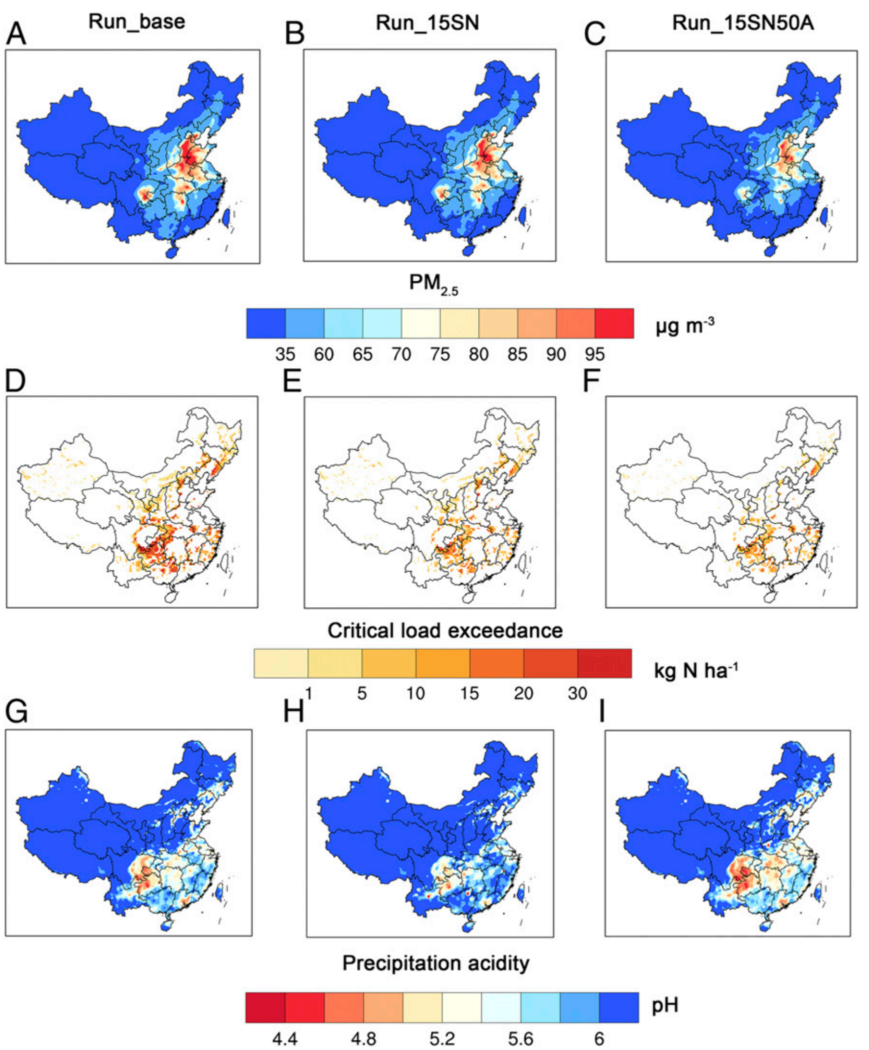

Fig. 2. Comparison of simulated $PM_{2.5}$ concentrations (*A*–*C*), critical load exceedance of nitrogen (*D*–*F*), and precipitation-weighted mean pH (*G*–*I*) over China in the Run_base, Run_15SN, and Run_15SN50A scenarios. The small islands in the South China Sea were excluded.

To shed more light on such a high efficiency of NH_3 emission control on $PM_{2.5}$ mitigation, we further investigate responses of different compositions of $PM_{2.5}$ in the Run_15SN50A case. Our results indicate that particulate nitrate dominates the reductions in $PM_{2.5}$ concentrations for the areas of interest, including northern China, southern China, and the Sichuan Basin. Particulate ammonium nitrate concentrations decrease by 30–60% relative to the baseline levels, which contributes approximately 70% of $PM_{2.5}$ mass reduction in these regions during JJA and DJF (other reductions are from particulate ammonium sulfate). According to the aerosol thermodynamic equilibrium, less available ambient NH_3 favors more HNO_3 staying in gas phase instead of partitioning into aerosols and then inhibits the formation of ammonium nitrate (17). The simulations demonstrate that particulate nitrate reductions resulting from NH_3 emission control are more pronounced in winter (mostly 3–9 $\mu g \cdot m^{-3}$) than in summer (<3 $\mu g \cdot m^{-3}$) in the regions on which we focused (*SI Appendix*, Fig. S7). This disparity can be ascribed to the seasonality of air temperature (i.e., highest in JJA and lowest in DJF) that determines the NH_4NO_3 phase equilibrium and NH_3 emission rate. First, higher temperature in summertime increases the NH_4NO_3 equilibrium constant and lowers the concentrations of ammonium nitrate. Moreover, NH_3 emissions are significantly enhanced in summer, leading to NH_3-rich regimes. These factors jointly make particle nitrate concentrations less sensitive to NH_3 reductions in summer. As a result, $PM_{2.5}$ mitigation, which is dominated by ammonium nitrate, is more substantial in DJF than in JJA (*SI Appendix*, Fig. S7).

Regionally, the mass reductions of nitrate in summer are greater in northern China than in southern China (*SI Appendix*, Fig. S7) because higher temperatures in the southern area of the country favor the thermodynamic phase partitioning of HNO_3 and NH_3 into gas phase. In contrast, the reductions of nitrate appear more pronounced in southern China in winter, which is likely attributable to relatively lower NH_3 emissions in this region that make the formation of NH_4NO_3 more sensitive to NH_3 reductions. In general, the NH_4NO_3 thermodynamic equilibriums and heterogeneous NH_3 emissions together determine the seasonal and regional responses of NH_4NO_3 to NH_3. It is also noteworthy that NH_3 emission control could be much more effective in reducing particulate nitrate during days with haze pollution than during days without it (*SI Appendix*, Fig. S5). It could be attributable to rapidly increased $PM_{2.5}$ nitrate concentrations during haze conditions, making the formation of ammonium nitrate NH_3-limited. The results highlight the importance of NH_3 abatement in $PM_{2.5}$ mitigation during winter haze events in China.

Given that NH_x-N (gaseous NH_3 and particulate NH_4^+) contributes to more than half of the inorganic nitrogen deposition in China (27), this NH_3 emission control will inevitably alter nitrogen deposition flux through dry and wet processes. By averaging simulation results in summer and winter, the annual nitrogen deposition over China is estimated to be 13.6 Tg $N \cdot y^{-1}$, in which 8.6 Tg $N \cdot y^{-1}$ (63%) is NH_x-N and 3.8 Tg $N \cdot y^{-1}$ (28%) is via dry deposition processes, consistent with previous studies (6, 27). To quantitatively investigate the effects of nitrogen deposition on terrestrial ecosystems, exceedance of critical load is introduced in this study. It is derived from the simulation of nitrogen deposition and a critical load map over China based on the steady-state mass balance method (28). Here, the critical loads of nitrogen deposition refer to quantitative estimates of the highest deposition amount that will not cause harmful effects on sensitive ecological systems, which range between 10–150 kg $N \cdot ha^{-1} \cdot y^{-1}$ over China (*SI Appendix*, Fig. S8).

As illustrated in Fig. 2 *D–F*, exceedance of critical load can reach more than 20 kg $N \cdot ha^{-1} \cdot y^{-1}$ in the northeastern, central, and southern regions of China in the baseline case of 2015. The area exceeding the critical load is estimated to account for approximately 17% of China's terrestrial land. If SO_2 and NO_x emission is cut by 15% (Run_15SN), the total nitrogen deposition decreases by only 1% and the area with critical load exceedance is almost the same. When NH_3 emission is halved (i.e., Run_15SN50A), nitrogen deposition could decrease by 34% (from 13.6 to 8.9 Tg $N \cdot y^{-}1$), thereby reducing the area with critical load exceedance to 9% of the country's land. Spatially, decrease in critical load exceedance is quite pronounced in central and southern China, which have high nitrogen deposition and vulnerable ecosystems. The reduction in NH_x-N deposition will help weaken soil and aquatic acidification and eutrophication over the terrestrial ecosystems with nitrogen saturation.

As mentioned earlier, another environmental concern that is closely linked to atmospheric NH_3 abundance is acid rain. We evaluated the model performance on reproducing precipitation acidity by comparing it with nationwide measurements (*SI Appendix*, Fig. S6). The measurements and simulations indicate that acid rain pollution (pH < 5.6) is mainly distributed in southern China, the Sichuan Basin, and parts of northern China in 2015 (Fig. 2*G*). On the contrary, almost no acid rain area is found in western parts of China because abundant windblown dust in the atmosphere is capable of neutralizing precipitation acidity in this region. Under the assumption of 15% SO_2 and NO_x emission reduction, we find that acid rain can be effectively mitigated. Specifically, precipitation pH values are elevated by 0.1–0.5 units in northern and southern China, and the area with pH values less than 5.6 simultaneously decreases from 1.7 (Run_base) to 1.0 million km^2 (Run_15SN).

However, unlike the impact on $PM_{2.5}$ pollution and nitrogen deposition, the NH_3 emission reduction aggravates precipitation acidification and expands the acid rain area over China (Fig. 2*I*). Our simulations show that 50% NH_3 emission reduction will significantly decrease the concentrations of ammonium ions in precipitation, with an average of 20–80 μeq/L over eastern China. This would enhance precipitation acidity via two pathways (*SI Appendix*, Fig. S9). One is the decreased wet removal of gas-phase NH_3 by rainwater. According to the gas–liquid equilibrium of the $NH_3(g)$-$NH_3(aq)$-$NH_4^+(aq)$ system, decreases in ambient gas-phase NH_3 concentrations make this equilibrium shift toward the gas phase, consequently releasing hydrogen ions (H_3O^+) in the aqueous phase. In addition, less ambient NH_3 favors the formation of ammonium bisulfate, which is more acidic than ammonium sulfate and thereby increases the aerosol acidity (29). When dissolved into cloud/rain water drops, ammonium bisulfate could produce more hydrogen ions than ammonium sulfate with the same wet scavenging amounts of sulfate ions, and the precipitation acidity therefore tends to be elevated. Our simulations demonstrate that the aforementioned two pathways account for almost all of the increase in H_3O^+ concentration in precipitation over China.

The simulations show that the volume-mean precipitation pH values decrease by as much as 1.0 unit in northern China and 0.1–0.5 units in southern areas. Given that precipitation acidity is much greater in southern China and the Sichuan Basin, even a small decrease in the pH would undoubtedly worsen the already grave situation. According to our simulations, acid rain area with pH < 5.6 over China increases from 1.7 (Run_base) to 2.0 million km^2 if synchronous control is applied on SO_2, NO_x, and NH_3 emission (Run_15SN50A). However, the heavy acid rain area (pH < 5.0) will expand by approximately 65% (from 0.23 to 0.38 million km^2), which is three times larger than that under the Run_15SN scenario (0.7 million km^2). In spatial terms, southern China and the Sichuan Basin undergo fast expansion of acid rain areas (Fig. 2*I*). Overall, NH_3 emission control inevitably results in a general increase in precipitation acidification over China. In addition, precipitation acidity under the Run_15SN50A scenario is almost the same or even higher than that under the Run_base scenario. In other words, the alleviation of acid rain over China caused by the control of SO_2 and NO_x emissions is very likely to be completely forfeited by NH_3 emission reductions.

Economic Valuation. As demonstrated, 50% NH_3 emission reduction can help to mitigate fine particle pollution and reduce nitrogen deposition to sensitive terrestrial land, but, in the meantime, worsen

ENVIRONMENTAL SCIENCES

the acid rain problem, a significant side effect that should not be neglected. To qualitatively compare these three impacts, we estimate the associated economic benefit or loss induced by the NH_3 emission reduction in terms of human health, crop production, and the ecosystem. Note that the differences between the scenario Run_15SN and Run_15SN50A represent the effects of the NH_3 emission reduction.

The assessment on the human health effects of decreased $PM_{2.5}$ concentrations is performed based on exposure–response functions (*SI Appendix, Materials and Methods*). The estimated total premature mortality in the baseline case is 1.15 million persons (0.54–1.75 million, within the 95% confidence level), which is similar to the findings of recent studies (30, 31). The NH_3 emission reduction in China would reduce the premature mortalities attributable to $PM_{2.5}$ exposure by approximately 80,000 people in 2015, and the resultant economic benefit is estimated to be 10.1 billion US$. Second, the reduction in nitrogen deposition decreases the area of critical load exceedance and yields an economic benefit of approximately 1.9 billion US$. However, the enhanced precipitation acidification caused by NH_3 emission control would cause an economic loss of approximately 5.0 billion US$ over China, with a dominant contribution from reduced crop yields (90%). This will partly offset the positive effects of the mitigation of $PM_{2.5}$ pollution and nitrogen critical load exceedance, and the overall benefit from these three effects is therefore 7.0 billion US$.

To figure out the overall cost effectiveness of the 50% NH_3 emission reduction in China, we further assess the cost of NH_3 abatement options in agricultural activities. The annual cost is estimated to be 6.6 billion US$, which is dominated by control measures for the improvement of livestock manure management (58%), followed by rational fertilizer application. Thus, the net benefit (subtracting the cost from the benefit) of NH_3 emission reduction is 0.4 US$, only approximately 6% of the expenditure. In other words, the loss from worsened acid rain and the abatement costs almost offset the benefits from mitigation of $PM_{2.5}$ pollution and nitrogen deposition. Therefore, under a target of a 15% reduction of SO_2 and NO_x emission in 2020, a reduction of 50% or more in NH_3 emissions may be not applicable because the benefits could not outweigh the costs.

To explore optimum control strategy for NH_3 emissions, we further vary the NH_3 emission reductions from 10% to 50% and compare the economic benefits with the corresponding abatement costs under each individual reduction scenario. Fig. 3 presents the comparisons in the three regions of concern. For northern China, NH_3 reductions always bring positive net benefits (i.e., revenue greater than cost), but the cost effectiveness (i.e., marginal revenue vs. marginal cost) varies with the NH_3 emission reductions. In response to the 0–20% options, the growth rate of economic benefit (i.e., marginal revenue) is greater than the abatement cost (i.e., marginal cost), which can be inferred from the slopes of the curves. A 20% reduction in NH_3 emission in northern China yields a relatively high benefit of 1.3 billion US$ relative to a cost of 0.3 billion US$, with a net benefit of 1.0 billion US$. Meanwhile, when NH_3 emissions are not as rigorously controlled (e.g., 20% reduction), the increase in acid rain area is relatively small ($<4 \times 10^4$ km^2), and famers would select cheaper and more feasible options to attain such a moderate NH_3 emission reduction, such as low-protein feeding and deep application of manure. However, further reductions ($\geq$ 30%) in NH_3 emissions result in an accelerated growth in acid rain pollution and rapidly increased expenditure for NH_3 emission abatement, making the unit reduction in NH_3 emissions less beneficial and more expensive. As a consequence, the marginal cost outweighs the marginal revenue when NH_3 emission reduction exceeds 30% (i.e., the slopes of the curves are <1 compared with the 1:1 line). Therefore, the most cost-effective strategy for NH_3 emission control is defined at a critical point at which marginal cost equals marginal revenue. In our study, that is estimated to be 20–30% for northern China.

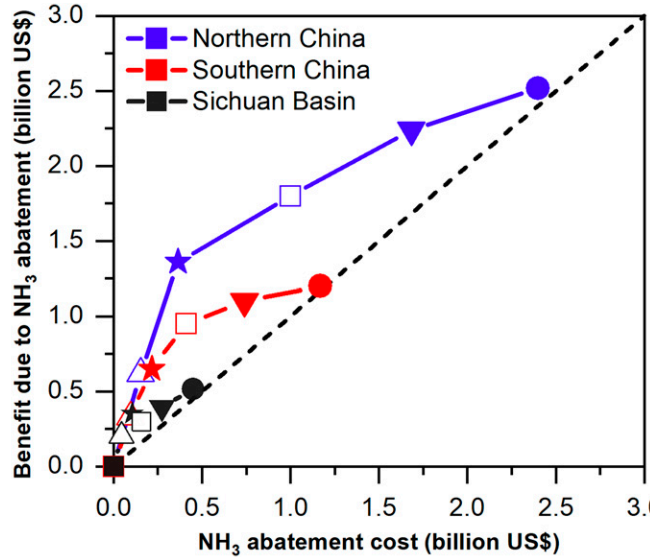

Fig. 3. Comparison of total economic benefit (in millions of US dollars; i.e., benefit from reduced $PM_{2.5}$ and nitrogen deposition minus loss from acid rain) vs. the costs of NH_3 abatement options under each NH_3 emission reduction scenario in northern China, southern China, and the Sichuan Basin. The cases shown in the figure are NH_3 emission reductions of 0 (■), 10% (△), 20% (★), 30% (□), 40% (▼), and 50% (●).

In southern China, the economic loss from worsened acid rain pollution is much more noteworthy. Under a 20% NH_3 reduction, the area exposed to acid rain in southern China increases by 2.1×10^5 km^2 (from 3.3 to 5.4×10^5 km^2), leading to a negative economic effect of as much as 0.6 billion US$. The resultant net benefit is only 0.4 billion US$ in the case that $PM_{2.5}$ pollution and nitrogen deposition is alleviated. Moreover, the economic loss from enhanced acid rain in southern China is 40–260% greater than that in northern China under the same emission reduction percentages. This further indicates that acid rain pollution remains a noteworthy problem in southern China and is quite sensitive to changes in NH_3 emissions.

Sichuan Basin is one of the most intensive emitters of NH_3 in China as a result of a large amount of livestock production. Heavy acid rain pollution also emerged in the region in the 1980s and has been a major environmental problem for decades (3). As shown in Fig. 3, the benefits of NH_3 emission reductions are almost the same as the corresponding costs (both $\leq$ 0.5 billion US$). Under a 20% NH_3 reduction, even though the marginal revenues are higher than the costs, acid rain pollution is obviously worsened. The simulation results show that the area exposed to heavy acid rain (pH < 5.0) in the Sichuan Basin increases by approximately 50% under the 20% case.

Overall, the results suggest a region-specific NH_3 emission abatement strategy. Over existing areas of acid rain like southern China and the Sichuan Basin (in southwestern China), it would be prudent to prioritize SO_2 and NO_x emission controls over NH_3 control to effectively alleviate the potential risks of precipitation acidification. In contrast, for areas with scarce acid rain but heavy fine particle pollution like northern China, implementation of 20–30% NH_3 emissions control would be cost-effective, particularly for reducing $PM_{2.5}$ pollution in winter haze conditions during 2016–2020.

Conclusion

NH_3 emission reduction has been proposed recently as a strategic option to mitigate severe haze pollution in China. However, atmospheric NH_3 is closely bound to other environmental concerns like nitrogen deposition and acid rain. In this study, we comprehensively investigate these multiple impacts of NH_3 emission reduction for further policy consideration in China. On the basis of a sophisticated NH_3 emission model, we indicate that improving agricultural production management can readily reduce the total NH_3 emission by as much as 50% across China. This reduction, together with a 15% reduction in SO_2 and NO_x emission, could accomplish a decrease of 11–17% in $PM_{2.5}$ concentrations, far more than when applying only SO_2 and NO_x control (3%). The dominant contributor is particulate nitrate, the concentration of which can be reduced by 30–60%, accounting for ~70% of $PM_{2.5}$ mitigation. As a result of NH_3 emission reduction, the nitrogen deposition is estimated to decrease by 33%, with the area of critical load exceedance decreasing from 17% to 9% of Chinese terrestrial land. However, as an important side effect, there arises an overall enhancement in precipitation acidity in China, with a decrease in pH values of as much as 1.0 unit and a 65% increase in heavy acid rain areas (pH < 5.0).

Our results emphasize the complexity embedded in NH_3 emission control in China: improved air quality, reduced nitrogen deposition, but worsened acid rain. Further quantitative assessments of the economic effects of NH_3 emission reduction demonstrate that the economic loss resulting from enhanced acid rain would partly offset the benefit from $PM_{2.5}$ mitigation and alleviated nitrogen deposition. After considering the costs of our abatement options, we propose regional-specific emission control strategies in the near future: implementing NH_3 emission reduction by 20–30% in areas with scarce acid rain but heavy fine particle pollution (e.g., northern China) while prioritizing SO_2 and NO_x emission controls in areas of existing acid rain like southern China. Such emission-control strategies will benefit air quality improvement and ecosystem health across China.

Materials and Methods

To investigate the multiple impacts of potential reductions in NH_3 emissions in China on $PM_{2.5}$ pollution, nitrogen deposition, and acid rain, we integrated numerical model simulations with a comprehensive NH_3 emission inventory (i.e., PKU-NH_3) as well as nationwide observations of precipitation acidity and aerosol chemical constituents. We performed simulations by using WRF-Chem (version 3.6.1) during summer (i.e., JJA) and winter (i.e., DJF) of 2015 to represent the pollution of acid rain, $PM_{2.5}$, and nitrogen critical load exceedance. The model domain and the geographic areas of interest are shown in *SI Appendix*, Fig. S2. Based on the PKU-NH_3 model and a combination of feasible emission abatement options, we designed a potential NH_3 emission reduction scenario and examined its influences on simulated $PM_{2.5}$ concentrations, nitrogen deposition, and precipitation acidity. We then estimated the economic effects of $PM_{2.5}$ pollution, nitrogen deposition, and acid rain caused by the NH_3 emission reduction. The premature mortality attributable to long-term exposure to $PM_{2.5}$ was calculated following the methods of Liu et al. (30). We quantitatively assessed the economic loss of reduced crop yields and forestry caused by enhanced precipitation acidification based on the experiments of Feng et al. (32). The potential benefit from reduced nitrogen deposition in those areas with critical load exceedance was estimated based on a unit damage cost for acidification and eutrophication of ecosystems (33). Furthermore, to obtain a cost-effective NH_3 control strategy, we estimated the cost of NH_3 abatement options applied in this study following the Greenhouse Gas—Air Pollution Interactions and Synergies model (34). Details of the observation datasets, evaluation of model performance, and economic assessment are provided in the *SI Appendix*.

ACKNOWLEDGMENTS. We thank Prof. A. R. Ravishankara (Departments of Chemistry and Atmospheric Science, Colorado State University) for valuable suggestions and improvement on the manuscript. Some observation data were provided by Prof. Shuiyuan Cheng (Beijing University of Technology), Dr. Shasha Yin (Zhengzhou University), and Nan Wang (Institute of Tropical and Marine Meteorology, China Meteorology Administration). This study was supported by National Natural Science Foundation of China Grants 91644212, 41421064, and 21625701 and National Key R&D Program of China Grant 2016YFC0201505.

1. Duan L, et al. (2016) Acid deposition in Asia: Emissions, deposition, and ecosystem effects. *Atmos Environ* 146:55–69.
2. Dianwu Z, Jiling X, Yu X, Chan WH (1988) Acid rain in southwestern China. *Atmos Environ (1967)* 22:349–358.
3. Larssen T, et al. (2006) Acid rain in China. *Environ Sci Technol* 40:418–425.
4. Vet R, et al. (2014) A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus. *Atmos Environ* 93:3–100.
5. Liu X, et al. (2013) Enhanced nitrogen deposition over China. *Nature* 494:459–462.
6. Zhao Y, et al. (2017) Atmospheric nitrogen deposition to China: A model analysis on nitrogen budget and critical load exceedance. *Atmos Environ* 153:32–40.
7. Huang R-J, et al. (2014) High secondary aerosol contribution to particulate pollution during haze events in China. *Nature* 514:218–222.
8. Guo S, et al. (2014) Elucidating severe urban haze formation in China. *Proc Natl Acad Sci USA* 111:17373–17378.
9. Zheng GJ, et al. (2015) Exploring the severe winter haze in Beijing: The impact of synoptic weather, regional transport and heterogeneous reactions. *Atmos Chem Phys* 15:2969–2983.
10. Elser M, et al. (2016) New insights into PM2.5 chemical composition and sources in two major cities in China during extreme haze events using aerosol mass spectrometry. *Atmos Chem Phys* 16:3207–3225.
11. Ding AJ, et al. (2016) Enhanced haze pollution by black carbon in megacities in China. *Geophys Res Lett* 43:2873–2879.
12. Liu F, et al. (2016) Recent reduction in NOx emissions over China: Synthesis of satellite observations and emission inventories. *Environ Res Lett* 11:114002.
13. Li C, et al. (2010) Recent large reduction in sulfur dioxide emissions from Chinese power plants observed by the ozone monitoring instrument. *Geophys Res Lett* 37: L08807.
14. Tang J, Xu X, Ba J, Wang S (2010) Trends of the precipitation acidity over China during 1992–2006. *Chin Sci Bull* 55:1800–1807.
15. Ministry of Ecology and Environment of the People's Republic of China (2018) Report of the state of the ecology and environment in China in 2017. Available at english.mee.gov.cn/Resources/Reports/soe/SOEE2017/201808/P020180801597738742758.pdf. Accessed October 5, 2018.
16. Central Committee of the Communist Party of China (2016) China's 13th Five-Year Plan (2016-2020). Available at en.ndrc.gov.cn/newsrelease/201612/P020161207645765233498.pdf. Accessed October 5, 2018.
17. Seinfeld JH, Pandis SN (2006) *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change* (John Wiley & Sons, New York), 2nd Ed, pp 434–485.
18. China Daily (2017) New fund will pay for research on smog. Available at www.chinadaily.com.cn/china/2017twosession/2017-03/16/content_28574400.htm. Accessed January 5, 2019.
19. Lajtha K, Jones J (2013) Trends in cation, nitrogen, sulfate and hydrogen ion concentrations in precipitation in the United States and Europe from 1978 to 2010: A new look at an old problem. *Biogeochemistry* 116:303–334.
20. Wang Y, Yu W, Pan Y, Wu D (2012) Acid neutralization of precipitation in northern China. *J Air Waste Manag Assoc* 62:204–211.
21. Larssen T, Carmichael GR (2000) Acid rain and acidification in China: The importance of base cation deposition. *Environ Pollut* 110:89–102.
22. Zhang L, et al. (2018) Agricultural ammonia emissions in China: Reconciling bottom-up and top-down estimates. *Atmos Chem Phys* 18:339–355.
23. Huang X, et al. (2012) A high-resolution ammonia emission inventory in China. *Global Biogeochem Cycles* 26:GB1030.
24. Paulot F, et al. (2014) Ammonia emissions in the United States, European Union, and China derived by high-resolution inversion of ammonium wet deposition data: Interpretation with a new agricultural emissions inventory (MASAGE_NH3). *J Geophys Res Atmos* 119:4343–4364.
25. Kang Y, et al. (2016) High-resolution ammonia emissions inventories in China from 1980 to 2012. *Atmos Chem Phys* 16:2043–2058.
26. Environmental Protection Agency (2014) Environmental Protection Agency National Emissions Inventory (NEI) Data. Available at https://www.epa.gov/air-emissions-inventories Accessed October 5, 2018.
27. Xu W, et al. (2015) Quantifying atmospheric nitrogen deposition through a nationwide monitoring network across China. *Atmos Chem Phys* 15:12345–12360.
28. Posch M, Duan L, Reinds GJ, Zhao Y (2015) Critical loads of nitrogen and sulphur to avert acidification and eutrophication in Europe and China. *Landsc Ecol* 30: 487–499.
29. Weber RJ, Guo H, Russell AG, Nenes A (2016) High aerosol acidity despite declining atmospheric sulfate concentrations over the past 15 years. *Nat Geosci* 9:282–285.
30. Liu J, Han Y, Tang X, Zhu J, Zhu T (2016) Estimating adult mortality attributable to PM2.5 exposure in China with assimilated PM2.5 concentrations based on a ground monitoring network. *Sci Total Environ* 568:1253–1262.
31. Hu J, et al. (2017) Premature mortality attributable to particulate matter in China: Source contributions and responses to reductions. *Environ Sci Technol* 51: 9950–9959.
32. Feng ZW, Miao H, Zhang FZ, Huang YZ (2002) Effects of acid deposition on terrestrial ecosystems and their rehabilitation strategies in China. *J Environ Sci (China)* 14: 227–233.
33. Sutton MA, et al. (2011) *The European Nitrogen Assessment: Costs and Benefits of Nitrogen in the Environment* (Cambridge Univ Press, Cambridge, UK), pp 513–540.
34. Klimont Z, Winiwarter W (2011) Integrated ammonia abatement–modelling of emission control potentials and costs in GAINS, IIASA interim report (International Institute for Applied Systems Analysis, Laxenburg, Austria).
35. Lu Z, et al. (2010) Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000. *Atmos Chem Phys* 10:6311–6331.