Global gains in food production over the past decades have been associated with substantial agricultural nitrogen overuse and ammonia emissions, which have caused excessive nitrogen deposition and subsequent damage to the ecosystem health. However, it is unclear which crops or animals have high ammonia emission potential, how these emissions affect the temporal and spatial patterns of nitrogen deposition, and where to target future abatement. Here, we develop a long-term agricultural ammonia emission dataset in nearly recent four decades (1980–2018) and link it with a chemical transport model for an integrated assessment of global nitrogen deposition patterns. We found global agricultural ammonia emissions increased by 78% from 1980 and 2018, in which cropland ammonia emissions increased by 128%, and livestock ammonia emissions increased by 45%. Our analyses demonstrated that three crops (wheat, maize, and rice) and four animals (cattle, chicken, goats, and pigs) accounted for over 70% total ammonia emissions. Global reduced nitrogen deposition increased by 72% between 1980 and 2018 and would account for a larger part of total nitrogen deposition due to the lack of ammonia regulations. Three countries (China, India, and the United States) accounted for 47% of global ammonia emissions, and had substantial nitrogen fertilizer overuse. Nitrogen deposition caused by nitrogen overuse accounted for 10 to 20% of total nitrogen deposition in hotspot regions including China, India, and the United States. Future progress toward reducing nitrogen deposition will be increasingly difficult without reducing agricultural ammonia emissions.

In the past half-century, humans have increased the amount of reactive nitrogen (Nr) in the environment more than the amount of any other major element (1). Nr compounds in the atmosphere are mainly controlled by the emissions of nitrogen oxides (NOx) and ammonia (NH3) (2); NOx is mainly from the burning of fossil fuels for energy production and NH3 is mostly from agricultural sources including volatilized livestock waste and nitrogen (N)-based fertilizers (3, 4). Our analyses of Nr have shifted from how to increase food production to a realization that agricultural intensification adds excess Nr that damages environmental systems and degrades human health. Agriculture is responsible for approximately two-thirds of global Nr pollution (2). These emissions result in excessive N deposition in terrestrial and aquatic ecosystems, with adverse feedbacks on human and ecosystem health, water eutrophication, soil acidification, and biological diversity; they increase the risk of irreversible and sudden environmental change (5, 6).

Global crop production has doubled in the recent four decades, while synthetic N fertilizer use has tripled over the same period according to the Food and Agriculture Organization (FAO) of the United Nations. The global average nitrogen use efficiency (NUE) for crop production (NUE refers to the ratio of crop N uptake to applied N fertilizer) has decreased from 0.5 in 1961 to 0.4 in 2010 (7), indicating substantial N losses to the environment. Agricultural NH3 emissions are not regulated in most countries, with the exception of Western Europe (8). Due to substantial overuse of agricultural N and inappropriate management, large amounts of NH3 are volatilized from agricultural systems, and thereby affect atmospheric chemistry and cause high N2 deposition downwind (9, 10). Asian countries overuse N more than do others. The absolute worst case is China, whose average NUE has decreased from more than 60% in 1961 to 25% in 2010 (7). China has implemented a series of policies to encourage the production and use of synthetic fertilizer in the past three decades (11). In 2010, more than 30% of the global fertilizer consumption was used for China’s cultivated land (accounting for only 7% of the global cultivated land area). In India, where fertilizer application has doubled in 20 y, its NUE has decreased from 40% in 1961 to 30% in 2018. 

Significance

Agricultural systems are already major forces of ammonia pollution and environmental degradation. How agricultural ammonia emissions affect the spatio-temporal patterns of nitrogen deposition and where to target future mitigation efforts, remains poorly understood. We develop a substantially complete and coherent agricultural ammonia emissions dataset in nearly recent four decades, and evaluate the relative role of reduced nitrogen in total nitrogen deposition in a spatially explicit way. Global reduced nitrogen deposition has grown rapidly, and will occupy a greater dominant position in total nitrogen deposition without future ammonia regulations. Recognition of agricultural ammonia emissions on nitrogen deposition is critical to formulate effective policies to address ammonia related environmental challenges and protect ecosystems from excessive nitrogen inputs.
2010 (7). Decreasing NUE also occurs in many other countries in the world, making the need to increase agricultural productivity, while at the same time reducing environmental impacts related to the use of N fertilizer, a global challenge.

Sustainably meeting global food demands is one of humanity’s grand challenges, and it is crucial to produce sufficient food with less pollution in the near future. However, there have been few attempts to evaluate the effect of agricultural NH$_3$ emissions on N$_r$ deposition systematically while at the same time identifying where to target proposed solutions. In this study, we aimed to identify a small set of regions, crops, and actions that provide strategic global opportunities to reduce NH$_3$ emissions and N$_r$ deposition, while delivering more food more efficiently from existing farmlands. To determine which interventions may have a high degree of global impact in these categories, we use recently published geospatial data and models to analyze the potential reduction of agricultural NH$_3$ emission in terms of how effective they will be in mitigating N$_r$ deposition.

**Results**

Agricultural NH$_3$ Emissions. To start, we constructed a high-resolution global agricultural NH$_3$ emission dataset (0.083°) for 1980 to 2018 using recently published geospatial data (4, 12) and long-term statistics from the FAO of the United Nations and considering both N application and livestock waste. We use our estimated cropland and livestock NH$_3$ emission data in the year 2010 shown here as a case study (Fig. 1). We choose 2010 as a representative year in terms of very detailed spatial information of N fertilizer use and livestock data.

We analyzed 16 major crops (including wheat, maize, rice, soybean, etc.) from the FAO of the United Nations. Global agricultural N application was estimated at 144 Tg N in 2010, of which 68% and 32% came from N fertilizer and manure, respectively. The 16 major crops accounted for 73% of global N application (SI Appendix, Table S1). With current agricultural management, emission factors of NH$_3$ in croplands were highest over Asia (20%) and lowest in Europe (13%) and had a global average of 17% (SI Appendix, Table S3). Global N application produced 28 Tg N as NH$_3$ emitted to the atmosphere (Fig. 1A), in which wheat, maize, and rice accounted for 68% of cropland NH$_3$ emissions from the 16 major crops analyzed here (Fig. 1D). Three countries (China, India, and the United States) had cropland NH$_3$ emissions of 16 Tg N, accounting for 61% of cropland NH$_3$ emissions. These results highlight that cropland NH$_3$ emissions are not evenly distributed by crops or regions.

Besides crops, we also used the recently published gridded livestock database to calculate livestock NH$_3$ emissions (12) including 8 major animals (buffalo, cattle, chicken, duck, horse, goat, pig, and sheep) (Fig. 1D). We used a bottom-up approach to estimate livestock NH$_3$ emissions, combining animal numbers and emission factors of animals. Emission factors of NH$_3$ from animals ranged from 0.23 to 18.40 kg N head$^{-1}$ y$^{-1}$ (SI Appendix, Table S3), with high values in cattle (dairy cattle as 18.4 kg N head$^{-1}$ y$^{-1}$ and nondairy cattle as 8.5 kg N head$^{-1}$ y$^{-1}$), followed by horse, buffaloes, pigs, goats, sheep, chickens, and ducks (0.23–8.70 kg N head$^{-1}$ y$^{-1}$). Total livestock NH$_3$ emissions were estimated to be 30 Tg N in 2010 (Fig. 1B). Generally, NH$_3$ emissions differ substantially among animal types. Four animals (cattle, chicken, goats, and pigs) accounted for 90% of livestock NH$_3$ emissions. This suggests that mitigation measures should give priority to these animals with high NH$_3$ emission potential.

In summary, NH$_3$ emissions from livestock and cropland systems were 58 Tg N in 2010 (Fig. 1B). China, India, and the United States had total NH$_3$ emissions as 14.5, 8.9, and 4.1 Tg N, which summed to 27.5 Tg N and accounted for 47% of global NH$_3$ emissions from livestock and N fertilizer. Our estimates of NH$_3$ emissions had a consistency in the geospatial patterns of the Emission Database for Global Atmospheric Research (EDGAR) and Community Emissions Data System (CEDS) by country (SI Appendix, Fig. S3), and were also close to estimates of regional studies of China (14.5 Tg N in this study vs. 12.6 Tg N in Zhang et al. [9]), the United States (4.1 Tg N in this study vs. 3.9 Tg N in Cao et al. [13]), and Western Europe (3.0 Tg N in this study vs. 3.2 Tg N in European Environment Agency [14]) around 2010. Compared to other emission inventories, we not only estimated the total amounts of agricultural NH$_3$ emissions, but also estimated NH$_3$ emissions by each crop and animal, which was critical for understanding which crops or animals had high NH$_3$ emission potential, and thus helpful for formulating future N$_r$ abatement measures.

Regarding trends, global agricultural NH$_3$ emissions increased by 78% from 1980 to 2018, in which cropland NH$_3$ emissions increased by 128% (14–32 Tg N y$^{-1}$) and livestock NH$_3$ emissions increased by 45% (22–32 Tg N y$^{-1}$). For the spatial changes, both cropland and livestock NH$_3$ emissions increased substantially including China, India, the United States, South America, and Africa. The largest increase in NH$_3$ emissions occurred in China and India (for cropland above 10 kg N ha$^{-1}$ and for livestock 5–10 kg N ha$^{-1}$), while Western Europe had experienced a decline for both cropland and livestock (0–10 kg N ha$^{-1}$) (Fig. 2). The decline in agricultural NH$_3$ emissions in Western Europe were associated with NH$_3$ emission controls, such as the EU National Emissions Ceilings Directive (15, 16).

Reduced vs. Oxidized N$_r$ Deposition. We calculated the overall status of N$_r$ deposition in the base year 2010, using a state-of-the-art chemistry transport model (GEOS-Chem) to simulate global N$_r$ deposition with our estimates of global agricultural NH$_3$ emissions, while the nonagricultural NH$_3$ emissions and NO$_x$ emissions were obtained from the Community Emissions Data System (CEDS) (17). The modeled deposition of reduced N$_r$ (NH$_4$ = ammonia [NH$_4^+$] + ammonium [NH$_4^+$]) and oxidized N$_r$ (NO$_x$, the sum of all oxidized N$_r$ species) show a general agreement with measurements over regions with intensive monitoring sites (the overall bias was below 10% in SI Appendix, Fig. S4).

Global N$_r$ deposition was estimated at 119 Tg N in 2010 (land, 60%; seas, 40%) (Fig. 3). We found, for most countries, above 60% of N$_r$ emissions were deposited to their own lands (Fig. 3F). For instance, in China, N$_r$ deposition accounted for 75% of its N$_r$ emissions in 2010 (20.4 Tg N), while the remaining 25% was deposited outside China. The ratio of N$_r$ deposition to emissions (RDep/Emi) in China was close to that in the United States (68%) and Western Europe (71%), while RDep/Emi was close to or larger than 1 for remote regions (e.g., in Greenland), suggesting these regions can be substantially affected by other countries through atmospheric transports. N$_r$ deposition varied substantially with the highest values in the Eastern China (>40 kg N ha$^{-1}$) and India (20–40 kg N ha$^{-1}$), followed by the Eastern United States, Western Europe, Central South America, and Africa around the equator (5–20 kg N...
As the world’s most polluted region, China’s total Nr deposition budgets (15.6 Tg N) in 2010 were approximately three times those over the United States (5.0 Tg N) and Western Europe (4.6 Tg N) in 2010.

NH₃ and NOₓ deposition showed markedly different spatial patterns (Fig. 3 A and B). High NOₓ deposition mainly appeared in developed regions partly correlated with industrial sources. Instead, high NH₃ deposition mainly occurred in
agricultural regions, which can be evidenced clearly in Eastern China, India and Central Eastern United States. Both NOy and NHx deposition exhibited highest (>20 kg N ha⁻¹/year) over the Eastern China (including North China Plain and Yangtze River Delta Region), with highly developed economy and agriculture (18). High NHx deposition also occurred in Central South America and Africa around Equator, associated to the large-scale biomass burning (SI Appendix, Fig. S6). The ratio of NHx (NOy) deposition to NH3 (NOx) emissions was also calculated using the GEOS-Chem (SI Appendix, Fig. S5). A larger proportion of NHx was deposited close to the source region, while NOy could be deposited in more remote regions with atmospheric transports. For instance, the average ratio of deposition to emissions was 82% for NHx in China, which was higher than that (68%) for NOy; similar phenomena can be also found in the United States (71% for NHx and 65% for NOy), India (68% for NHx and 55% for NOy), and Brazil (81% for NHx and 67% for NOy).

Fig. 3D shows the ratio of NHx to NOy deposition (R_{NHx/NOy}) globally. R_{NHx/NOy} >1 represented that agriculture was the major source; otherwise fossil fuel combustion was the dominating source (19). As shown in Fig. 3D and E, NHx exceeded NOy deposition in agricultural regions including the Eastern United States, India, China, and biomass burning regions (Central South America and Africa around the equator), while the Middle East, North Africa, and Greenland were dominated by NOy deposition. The Greenland had near-zero NOx and NH3 emissions, but received more NOy deposition than NHx, reflecting that NOy can be deposited in nearly human untouched areas with a longer transport than NHx. India’s NHx deposition was approximately triple that of the NOy deposition (Fig. 3D), showing N deposition was mainly
dominated by NH₃. Over China, the RNHₓ/NOₓ was 1.7, indicating NH₃ deposition was 70% higher than NOₓ deposition. Over Western Europe and the United States, the RNHₓ/NOₓ was around 0.8, showing its NH₃ deposition was slightly lower than NOₓ deposition.

**Trends in Reduced and Oxidized N₂ Deposition.** We constructed the comprehensive long-term oxidized and reduced N deposition over the period 1980 to 2018 using the GEOS-Chem model with the high-resolution agricultural NH₃ emissions and CEDS NOₓ emissions. For the overlap time period (year 2010) between this study and Tan et al. (20), our estimates of NH₃ (63 Tg N) and NOₓ (56 Tg N) deposition were close to theirs (NH₃, 64 Tg N and NOₓ 59 Tg N). The major difference between the two studies was due to the different emission datasets used. We used our developed high-resolution NH₃ emission data and the CEDS NOₓ emissions overwritten by several comprehensive regional emission data, while they mainly adopted the EDGAR emissions. Our results show similar estimates with regional studies at corresponding years conducted, such as in China [16.4 Tg N by Zhao et al. (21) vs. 15.6 Tg N in this study during 2008–2010], the United States [6.7 Tg N by Zhang et al. (10) vs. 5.9 Tg N in this study during 2006–2008], and Western Europe [5.1 Tg N by Tan et al. (20) vs. 4.6 Tg N in this study in 2010]. This study provides a robust estimation of global N deposition in recent four decades with a consistent and reproducible methodology.

Regarding NOₓ deposition trends (Fig. 4 B and C), the United States and Western Europe have reduced by half (50–60%) due to effective regulations (such as the Clean Air Act) (22) designed to decrease NOₓ emissions between 1980 and 2018. NOₓ deposition in the United States and Western Europe reached the same level low in 2018 (2.4 vs. 2.4 Tg N). For some countries in Western Europe, the reduction of NOₓ deposition even exceeded 60% between 1980 and 2018, for instance in France (67%) and Netherlands (70%) (SI Appendix, Fig. S7). In contrast, China has experienced explosive growth of NOₓ deposition and increased by more than 5 times from 1980 (1.2 Tg N) to 2012 (6.2 Tg N). Fortunately, China’s NOₓ deposition has declined by 16% between 2012 and 2018 due to the adoption of emission control technologies for large combustion plants and boilers (23). The decrease in China’s NOₓ deposition since 2012 was consistent with an observational study (24) based on the Chinese National N Deposition Monitoring Network (NNDMN). Although great efforts have been made to reduce NOₓ emissions, China still faced a high level of NOₓ deposition (5.2 Tg N), which was more double that in the United States and Western Europe in 2018. Global NOₓ deposition on land has increased by 40% from 1980 (27 Tg N) to 2016 (38 Tg N), after then it has declined by 16% to 2018 (32 Tg N). Further reductions in NOₓ deposition are expected in coming years resulting from stringent policy actions including the vehicle emission control and the national ambient air quality standards (22).

Regarding NH₃ deposition trends (Fig. 4 A and C), Western Europe was identified as the only region with a continuous decline in recent decades (16%, 1980–2009) due to the agricultural emission reduction measures under the Union Common Agricultural Policy (25), while NH₃ deposition increased slightly after 2009 (from 2.0 Tg to 2.2 Tg in 2018). For instance, NH₃ abatement was required in policies of Denmark and Netherlands (15), leading to reduction of NH₃ deposition by 40% and 10% between 1990 and 2018 (SI Appendix, Fig. S7). In Western Europe, NH₃ emission reduction would be required for all sectors by 2030, with the guidance of European National Emission Ceilings directive (15). Notably, although NH₃ emissions in Western Europe have reduced since 1980, it does not mean that NH₃ emissions for all Western European countries have decreased. For instance, in Spain, NH₃ deposition has increased since 1980 (SI Appendix, Fig. S7). The 2020 European Environment Agency report also indicated that NH₃ emissions in Spain were 33% above ceiling levels with the highest exceedances for all 27 member states (26). Reducing NH₃ emissions will continue to be a major challenge in Western Europe. Although Western Europe had continuous decreasing NH₃ deposition in recent decades, other countries have experienced a rapid increasing trend. For the United States, despite its successful controls in NOₓ deposition, its NH₃ deposition has increased by 14% from 1980 (2.2 Tg N) to 2019 (2.5 Tg N) because NH₃ emissions in the United States are not strictly regulated. The estimated increasing NH₃ deposition in the United States was consistent with an observational study, which found NH₃ deposition increased by 10–20% from 1990 to 2010 (22). Compared to Western Europe and the United States, China’s NH₃ deposition increased greatly by 113% from 1980 (4.7 Tg N) to 2015 (10.0 Tg N). After 2015, China’s NH₃ deposition became stable with a slight decreasing trend. The stabilization in China’s NH₃ deposition after 2015 can be also supported by the NNDMN measurements (24), which may be partly explained by the implementation of Zero Increase Action Plan for N fertilizer after 2015. China’s NH₃ deposition was still much too high (9.4 Tg N in 2018), which was substantially larger (4 times) compared to that in the United States (2.5 Tg N) and Western Europe (2.2 Tg N). SI Appendix, Fig. S8 shows regional N deposition in China, the United States, and Western Europe expressed by per unit area (kg N ha⁻¹ y⁻¹), presenting similar regional trends and differences as indicated by Fig. 4. China’s N deposition in the 2010s (>15 Tg N) was much larger than those of the United States and Western Europe in 1980s (7–8 Tg N), when the Clean Air Act and emission control had not been fully implemented (11). Challenges faced by China are much higher than those in the United States and Europe. For other developing countries, a similar continuous increasing trend in NH₃ deposition since 1980 was also found in India, Pakistan, Mongolia, Brazil, and Sudan (SI Appendix, Fig. S9) because no limitation was implemented on NH₃ emissions. For the global totals on land, NH₃ deposition increased by 72% from 1980 (25 Tg N) to 2018 (44 Tg N). SI Appendix, Fig. S10 shows the time series of the relative contribution of NH₃ to total N deposition (RNH₃/N) on land globally in China, the United States, and Western Europe. Increasing importance of NH₃ to total N deposition for the globe can be clearly seen from 0.41 in 1980 to 0.54 in 2018. Similar phenomena with increased RNH₃/N can be also found for the United States (0.31–0.52) and Western Europe (0.33–0.47). However, for China, the RNH₃/N first decreased from 0.78 in 1980 to 0.59 in 2011, and then increased after 2011 (0.59–0.65). The decrease of RNH₃/N in China between 1980 and 2011 was mainly due to the speed of NOₓ deposition driven by fossil fuel combustion that exceeded that of NH₃ deposition driven by agriculture, while the increased RNH₃/N, after 2011 can be explained by reduction measures for NOₓ emissions by the Chinese government. Increasing agricultural NH₃ emissions and the success of control measures in reducing NOₓ emissions are changing the temporal patterns of global N deposition. NH₃ dominated total N deposition in most countries in the recent decade and would account for a larger part of total N deposition due to the lack of NH₃ regulations.
Discussion and Implications

Global total NH₃ deposition increased by 70% in recent four decades due to the lack of policies in reducing agricultural NH₃ pollution worldwide (except Western Europe). We foresee that, without NH₃ emission reduction in the future, NH₃ deposition will continue to increase, and occupy a greater dominant position in total Nr deposition. Previous studies pointed to the overuse of N fertilizer as one major contributor to excessive Nr deposition (1, 27). Our developed agricultural NH₃ emissions demonstrated that three crops (rice, wheat, and maize) accounted for 68% of cropland NH₃ emissions. Using a yield-response model (see Materials and Methods), 38% of N fertilizer can be reduced without impacting current yields (Fig. 5C and SI Appendix, Fig. S13 and Table S1), and three crops (rice, wheat and maize) are responsible for 72% of the excessive N fertilizer of the 16 crops globally. For the hotspot regions, China, India, and the United States accounted for 65% of global excessive N fertilizer.

Nitrogen fertilizer overuse accounted for 25% of cropland NH₃ emissions (0–55% by country), and 11% of total NH₃ emissions (cropland plus livestock) (Fig. 5F). Regionally, the contribution of N overuse to total NH₃ emissions in China, the United States, and India was 31%, 25%, and 22%, respectively, while it was close to 0 in Africa, Southern America, and Russia because these areas had low N fertilizer. Low-yield performing was found in Africa, South America, and Russia (Fig. 5D) with the ratio of the observed to attainable yields below 0.5. Taking wheat, rice, and maize as an example, the ratio of current yields to attainable yields was below 50% over many regions, especially for the Sub-Saharan Africa (Eritrea, 27%; Sudan, 40%; Rwanda, 35%; Congo, 19%), South America (Brazil, 47%) and Russia (38%) (Fig. 5D and SI Appendix, Fig. S12).

It is still unclear on how much N fertilizer overuse contributes to Nr deposition. We calculated the contribution of potential N fertilizer overuse to Nr deposition using the GEOS-Chem for the base year 2010 as a case study. NH₃ deposition induced by N overuse accounted for 17% globally, while the contribution of N overuse to total Nr deposition was 10%. Higher contribution of N overuse to total Nr deposition occurred in China (20%), followed by India (15%), the United States (12%), and Western Europe (10%) (Fig. 4F). This suggests that 10 to 20% of total Nr deposition is due to N fertilizer overuse.
deposition can be reduced if N overuse was avoided in these hotspot areas through appropriate nutrient management. Thus, reducing N overuse for a small number of crops and countries may have a significant impact on global NH3 emissions.

Although Western European countries (such as Denmark and Netherlands) have adopted the nutrient managements (such as optimizing N fertilizer) along with emission control policies, many other countries (such as China, India, and the United States) still have a long way to address NH3 pollution in terms of lacking policies regarding reducing N overuse. As the world’s highest NH3 emission region, China began to mitigate agricultural NH3 emission during the 13th Five Year Plan (2016–2020), and it was estimated that China’s NH3 emission could be reduced by half through nutrient managements and appropriate policies (such as subsidies for reductions of urea-based fertilizer, promotion of enhanced efficiency N fertilizer, and machine deep placement of fertilizer) (25). Such improved nutrient management, for example, will also help reduce the imbalances of N and phosphorus in the North China Plain, the Midwest United States, and the sub-Sahara region (e.g., Kenya) (28).

In addition, NH3 reduction by avoiding N fertilizer overuse would mitigate PM2.5 (with an aerodynamic diameter smaller than 2.5 μm) through dry deposition and wet deposition.
2.5 μm) pollution at the same time (15). High decrease in PM2.5 concentrations would occur in Eastern China with the reduction ranges of 5 to 10 μg m⁻³, followed by India, the Eastern United States, Western Europe (0–5 μg m⁻³) (SI Appendix, Fig. S14). The decrease in PM2.5 by reduction in agricultural NH₃ emissions support increased emphasis on nutrient management to address NH₃ environmental challenges. Although reducing the production of N₂O and its harmful effects will be a challenge, it is both possible and vital. Our comprehensive analysis shows appropriate agricultural strategies can reduce agricultural NH₃ loss and NH₃ deposition at large scales while improving food security at the same time. Our work provides guidance for decision-making that can promote a sustainable food system in the future. Besides cropping systems, more structural measures for livestock production systems could further reduce agricultural NH₃ emissions, such as livestock relocation to enhance manure recycling, manure acidification, aerobic composting, and improved animal feed (25). Agricultural nutrient management by improving the efficiency of N fertilizer application would have co-benefits on air quality, human health, food security, climate mitigation, and biodiversity conservation, helping to solve the global N pollution challenge.

Materials and Methods

Agricultural Data. We collected crop related datasets from the EarthStat geographic datasets and the long-term statistical datasets (1980-2018) from the FAO datasets (https://www.fao.org/faostat/en) for analysis. The EarthStat is collaboration between the Global Landscapes Initiative at the University of Minnesota’s Institute on the Environment and the Land Use and Global Environment laboratory at the University of British Columbia. The FAO collected relevant national statistical information related to food and agriculture including the crop and livestock data. We used the FAO “Fertilizers by Nutrient” data by country between 1980 and 2018 that over 16 major crops analyzed here. The FAO included N fertilizer input for each crop by country in unit of both kg N ha⁻¹ y⁻¹ and Tg N y⁻¹. We used FAO statistical data to scale the EarthStat geospatial data by country. We focused our analysis on 16 key crops, composing the 16 highest-calorie-producing crops consumed as food (SI Appendix, Table S1). These 16 crops accounted for 60% of the global cropland area harvested and 70% of all N application on croplands and produced 86% of the world's crop calories. N application rate and consumption data were compiled for nations and subnational units across the globe. The crop related subnational data were mainly from statistical bureaus and national-level fertilizer industry associations at the provincial or state levels. Crop-specific application rates were then distributed across detailed maps of crop and pasture areas, and rates were harmonized with subnational and national nutrient consumption data.

We used a new version of the Gridded Livestock of the World (GLW3) database, reflecting the most recently compiled and harmonized subnational livestock distribution data for the year 2010 (https://dataverse.harvard.edu/dataverse/glw_3). GLW3 provides global population densities of cattle, buffalos, horses, sheep, goats, pigs, chickens, and ducks in each land pixel at a spatial resolution of 0.083 decimal degrees. They are accompanied by detailed metadata on the year, spatial resolution and source of the input census data. The FAO included the 8 major animals analyzed in this study in the unit of head in each country since 1980. We have detailed spatial distribution data of each animal in 2010s from the GLW3, while it is still very challenging to obtain animal-specific information at the subnational scales especially in 1980s. For the long-term datasets (1980-2018), we used the FAO statistics of animal number by country to scale the 2010 spatial data and then estimated livestock NH₃ emissions. Our estimated livestock NH₃ emissions are particularly helpful for national comparisons and temporal analysis at the regional scales.

Agricultural NH₃ Emissions between 1980–2018. We developed an agricultural NH₃ emission dataset from N fertilizer and livestock during 1980 to 2018. The NH₃ emission model represented N fertilizer application for 16 crops and livestock for 8 animals. The NH₃ emission model we used was a bottom-up NH₃ emission estimation from both N application and livestock waste using recently published geospatial data (4, 12) and long-term statistics from the FAO of the United Nations: Agricultural NH₃ emissions, from livestock production and cropland planting, were calculated by the bottom-up methods based on activities data and corresponding condition-specific EFs, according to the following equation:

$$E(NH_3) = \sum_j \sum_i A_{ij} \times EF_{ij}$$

where $$E(NH_3)$$ was the total agricultural NH₃ emissions, $$t$$ represented the source type (including crop-specific N fertilizers and livestock production [head]), i and j represented the grid row and column of global map (0.083° grids), $$A_{ij}$$ was the activity data of a specific source, and $$EF_{ij}$$ was the corresponding EFs. Crop groups included rice, wheat, maize, cotton, soybean, sugar crops, roots, oil crops, and other cereals (SI Appendix, Table S1). Different livestock mainly focused on 8 significant animals (buffalos, cattle, chickens, ducks, goats, sheep, horses, and pigs). Further details on the estimate methods and gridded allocation of the various sources were presented in previous studies (6, 29). Emission factors of NH₃ from N fertilizer and livestock were obtained from observation-based measurements by regions from cropping and pasture systems. The results were categorized by country, and the average by continent was shown in SI Appendix, Table S3 including Asia, Europe, North America, and South America.

N. Deposition by GEOS-Chem. The GEOS-Chem, a widely used global chemical transport model (v1203, http://www.geos-chem.org), was applied to simulate long-term N deposition (1980–2018). It includes a detailed simulation of tropospheric gas-aerosol chemistry and has been applied in a number of studies to simulate atmospheric N deposition. GEOS-Chem parameterizes dry deposition of gases and aerosols by a standard big leaf resistance model and wet deposition separated by the convective updraft and large-scale precipitation scavenging (10, 21). The surface-atmosphere bidirectional NH₃ fluxes are not considered in current deposition model simulations. The long-term MERRA2 assimilated meteorological data from the NASA Global Modeling and Assimilation Office were used to drive GEOS-Chem. The “tropchem” scheme of GEOS-Chem model was run at a horizontal resolution of 2° latitude by 2.5° longitude and 47 levels in the vertical. Spin-up simulation was performed with 6 mo before the actual simulation time for our analysis typically exceeding atmospheric NH₃ lifetime usually within 24 h and NH₄⁺ lifetime usually with 1 wk (30). We used 30-min and 15-min time steps for chemistry and transmission, respectively, as set similar to a previous study (31).

For agricultural NH₃ emissions, we used our developed high-resolution data set, while the anthropogenic NOX emissions were obtained from the newly updated global CEDS during 1980 to 2014, overwritten by regional inventories including EMEP over Europe and REAS v3 over East Asia, NEI in the United States, and CAC in Canada. For the years 2015 to 2018, we adopted the methods of Geddes et al. (31) to scale the 2014 base emissions to 2015 to 2019 according to the satellite observations (i.e., for NOX by OMI and NH₃ by IASI). The natural NOX and NH₃ emissions include GFED biomass burning, lightning, soil, and the biosphere. All emissions were treated with the Harvard emission components (HEMCO) system designed for deriving emissions in atmospheric models.

N Fertilizer Overuse and Its Contribution to NH₃ Emissions. We explored the potential overuse of N fertilizer for 16 major crops. A crop- and climate-specific yield response model was used to calculate the amount of fertilizer application that could be reduced yet still get the same yields. We refer to this potential as avoidable N. The yields for each crop are divided into 100 climate categories that describe the percent of Ymax achieved at a given nutrient level. Climatic potential yields are defined as the 95th percentile range of yields for a given crop in a given bin.

We then used the model to estimate the required amount of N needed to obtain current yields:

$$Y = \text{Yield}_{\text{max}}(1 - b_0 e^{-c_0 N_{\text{fertilizer}}})$$

where $$Y_{\text{max}}$$ is the maximum yield possible within the climate bin; $$b_0$$ describes the y-intercepts for each nutrient-yield response curve; $$c_0$$ are response coefficients that describe the percent of $$Y_{\text{max}}$$ achieved at a given nutrient level. Climatic potential yields are defined as the 95th percentile range of yields for a given crop in a given bin.
The model was only used when R² is ≥0.30 for within-bin variability explained by Mitscherlich–Baule curves.

After estimating avoidable N fertilizer, we applied it to estimate new NH₃ emissions and calculated the contribution of N overuse to total N deposition globally using the GEO-S-Chem model. The NH₃ emissions caused by N overuse were calculated as:

\[
\Delta F_{\text{NH}_3} = \sum_j \Delta A_{\text{NH}_3} \times E F_{\text{NH}_3} \tag{4}
\]

where \(\Delta F_{\text{NH}_3}\) is the agricultural NH₃ emissions induced by N overuse, \(\Delta A_{\text{NH}_3}\) is the overuse N, and \(E F_{\text{NH}_3}\) is the original emission factor.

Data Availability. The crop and livestock related dataset and GEO-S-Chem model code in this study are publicly available for download. Crop data from the EarthStat can be downloaded from http://www.earthstat.org/nutrient-application-major-crops/. The FAO long-term statistic data can be freely accessed from https://www.fao.org/faostat/en/#data/EF. The livestock data are available from Harvard Dataverse at https://dataverse.harvard.edu/dataverse/glw_3. The GEO-S-Chem model code is open source (https://doi.org/10.5281/zenodo.3676008). All other study data are included in the article and/or the supporting information.

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