Ammonia emission abatement does not fully control reduced forms of nitrogen deposition

Jiani Tan[a], **Joshua S. Fu**[a,b,1], **and John H. Seinfeld**[c,1]

[a]Department of Civil and Environmental Engineering, The University of Tennessee, Knoxville, TN 37996; [b]Computational Earth Sciences Group, Computational Sciences and Engineering Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831; and [c]Division of Chemistry and Chemical Engineering, California Institute of Technology, Pasadena, CA 91125

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Human activities and population growth have increased the natural burden of reactive nitrogen (N) in the environment. Excessive N deposition on Earth's surface leads to adverse feedbacks on ecosystems and humans. Similar to that of air pollution, emission control is recognized as an efficient means to control acid deposition. Control of nitrogen oxides ($NO_x = NO + NO_2$) emissions has led to reduction in deposition of oxidized nitrogen (NO_y, the sum of all oxidized nitrogen species, except nitrous oxide [N_2O]). Reduced forms of nitrogen (NH_x = ammonia [NH_3] + ammonium [NH_4^+]) deposition have, otherwise, increased, offsetting the benefit of reduction in NO_y deposition. Stringent control of NH_3 emissions is being considered. In this study, we assess the response of N deposition to N emission control on continental regions. We show that significant reduction of NH_x deposition is unlikely to be achieved at the early stages of implementing NH_3 emission abatement. Per-unit NH_3 emission abatement is shown to result in only 60–80% reduction in NH_x deposition, which is significantly lower than the demonstrated 80–120% benefit of controlling NO_x emissions on NO_y deposition. This 60–80% effectiveness of NH_x deposition reduction per unit NH_3 emission abatement reflects, in part, the effects of simultaneous reductions in NO_x and SO_2 emissions.

nitrogen deposition | reduced forms of nitrogen | ammonia emission | emission control strategy

Atmospheric deposition is the principal pathway for the exchange of nitrogen (N) between Earth's compartments. Elevated N deposition on Earth's surface has occurred (1–5), due to increasing human consumption of energy, with estimates of continuing growth in the near future (1, 3, 4, 6, 7). However, future increases of N deposition depend largely on the projection of future N emissions, which in some cases show slowdown or even stabilization trends (1). Response to excessive N deposition is exhibited mainly by eutrophication and acidification of terrestrial and aquatic ecosystems (8–11). The well-studied influences are damage and toxicity to vegetation and soil systems (12), impact on natural dominant species (13, 14), and even loss of ecosystem biodiversity (15). Therefore, prevention of excessive atmospheric N deposition is of vital importance in the protection of a sustainable ecosystem and in avoiding irreversible future damages.

Implementation of emission controls is an effective avenue to reduce acid deposition (3, 16–18). In the United States, oxidized nitrogen (NO_y, sum of all oxidized nitrogen species except nitrous oxide [N_2O]) deposition, a result of nitrogen oxides ($NO_x = NO + NO_2$) emissions from combustion, has been reduced since implementation of the Clean Air Act (CAA) in the 1970s. Nonetheless, rapid growth of the deposition of reduced forms of nitrogen (NH_x = ammonia [NH_3] + ammonium [NH_4^+]) has taken place (19) and offsets the benefits of NO_y deposition reduction. NH_x deposition results from wet deposition (precipitation) and dry deposition (driven by turbulent and molecular diffusion processes that bring NH_3 into contact with surfaces) of gaseous NH_3 and particulate NH_4^+. Relatively few studies have examined the effect of reducing NH_3 emissions on NH_x deposition. However, the effectiveness of NH_3 emission abatement on reducing airborne fine-particle levels has proven lower than expected (20–23), since conversion between NH_3 and NH_4^+ particulate matter is highly dependent on acidic aerosol components such as sulfate (SO_4^{2-}) and nitrate (NO_3^-). A similar situation is likely to arise associated with the control of NH_x deposition.

Here, we address the impact of adding NH_3 emissions control to NO_x and SO_2 emission control as a means to reduce NH_x deposition over continental regions, as well as a comparison with the benefit of NO_x emission abatement on reducing NO_y deposition. We first review the consistency between the spatiotemporal trends of NH_3 emissions and NH_x deposition over the conterminous United States (CONUS) based on both national data and site measurement data. Then, we examine the response of NH_x deposition to controlling NH_3 emissions via modeling analysis of emission control scenarios.

Results and Discussion

Temporal Changes of N Emissions and N Deposition. We analyzed the temporal trends of N emissions, N wet deposition, and NO_2 ambient concentrations with measured data in nine regions of CONUS from 2000 to 2017 (Fig. 1). Site measurements of NH_3 ambient concentrations are available after 2007, but the numbers of sites in regions (*SI Appendix*, Table S1) are considerably lower than those of NO_2 ambient concentration and N deposition (*SI Appendix*, Table S2), especially before 2011. Thus, temporal trends of NH_3 ambient concentration are not included in Fig. 1,

Significance

Control of nitrogen oxides emissions has successfully reduced the deposition of oxidized nitrogen over the United States. Recent studies have called attention to the rapid growth of deposition of reduced forms of nitrogen, which would offset the benefits of reduction in oxidized nitrogen deposition. Our analysis, based on both monitoring and modeling approaches, illustrates that implementing ammonia emission abatement may not lead to full response in nitrogen deposition over continental regions. Here we provide important information to the community and policy makers concerning nitrogen deposition, given the urgent needs for developing control strategies on ammonia emissions.

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[1] To whom correspondence may be addressed. Email: jsfu@utk.edu or seinfeld@caltech.edu.

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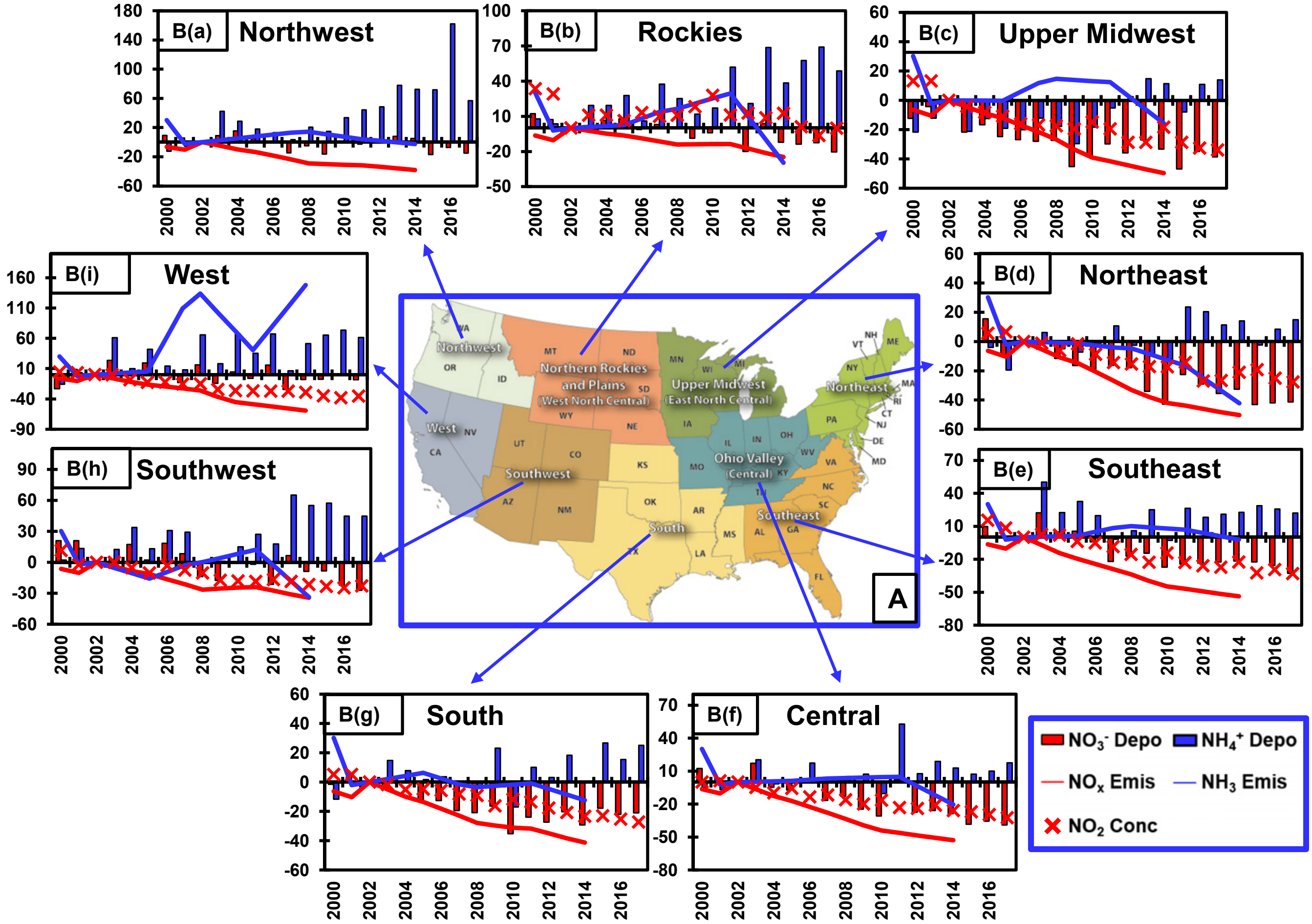

Fig. 1. Trends of annual N emissions and N deposition over nine CONUS regions from 2000 to 2017. (*A*) Definition of the nine CONUS regions in this study. (*B*, *a–i*) Percentage changes of N emission, NO_2 concentration, and N deposition in regions (unit: %). Airborne concentration and deposition values were averages over all sites in the regions. The percentage changes were calculated based on the 2002 level, calculated as 100 × (Emission of target year − Emission of 2002)/(Emission of 2002).

but are shown with the trends of N emission and deposition in *SI Appendix*, Fig. S1. NO_3^- and NH_4^+ wet deposition and ambient NO_2 concentration are annual average values over all sites in the regions. NO_x and NH_3 emissions are state-level values derived from the National Emissions Inventory (NEI) (see *Materials and Methods* for details).

NO_x emissions (Fig. 1*B*, red line) peaked in 2002 in all CONUS regions, with amounts dropping by ∼10% each year following for most regions, except the Northern Rockies and Plains (hereafter denoted Rockies) and the Southwest. This reduction of NO_x emissions, associated with implementation of regulations and policies subsequent to the 1970 CAA, is reflected in the reduction of NO_2 levels (Fig. 1*B*, red X's). NO_3^- wet deposition (Fig. 1*B*, red bars) also started to decrease after 2002 in all regions, although levels in all regions did not decrease at the same rate. For instance, 7 y were required for the Rockies to show a significant decrease of NO_3^- wet deposition, while clear and rapid (1–2 y after 2002) responses were found in the Upper Midwest, Northeast, South, and Central United States. Response times between decreases of NO_x emissions and NO_y wet deposition were associated with the reduction rates of NO_x emissions, and the atmospheric acidity and humidity (23–25), which determine the gas-aerosol partitioning of NO_3^-.

Overall, clear consistency exists between changes of NO_x emissions and NO_y wet deposition in the temporal trends in all regions. The relationship between NH_3 emissions and NH_x wet deposition, on the other hand, is considerably more complex than that between NO_x emissions and NO_y deposition. For example, a decrease in NH_4^+ wet deposition occurred during an increase of NH_3 emissions during 2004–2012 in the Upper Midwest (Fig. 1 *B*, *c*). As well, with continuously decreasing NH_3 emissions after 2006 (Fig. 1 *B*, *d*), the Northeast continued to experience increases in NH_4^+ wet deposition after 2011. Increases occurred in NH_3 airborne concentration in the Central and Northeast regions during 2011–2014 when NO_x and NH_3 emissions and NO_3^- and NH_4^+ wet deposition were decreased (*SI Appendix*, Fig. S1). The increased NH_3 air concentration is likely caused by release of NH_4^+ from the aerosol phase as SO_4^{2-} and NO_3^- concentrations and associated aerosol water decreased. Generally, it is difficult to establish a clear relationship between the trends of NH_3 emissions and NH_x wet deposition.

Temporal changes of NO_3^- wet deposition exhibited strongly positive linear correlations with those of NO_x emissions for several regions, such as Northeast, Central, and South United States (Fig. 2*A*), with correlations (R values) between these trends of ~0.8 (Fig. 2*C*, green bars). On the other hand, correlations between NH_3 emissions and NH_4^+ wet deposition were either unclear (Central, Rockies, South, Southeast, and West) or weakly negative (Northeast, Northwest, Southwest, and Upper Midwest) (Fig. 2*B*). Moreover, R values between the trends were low (Fig. 2*C*, blue bars).

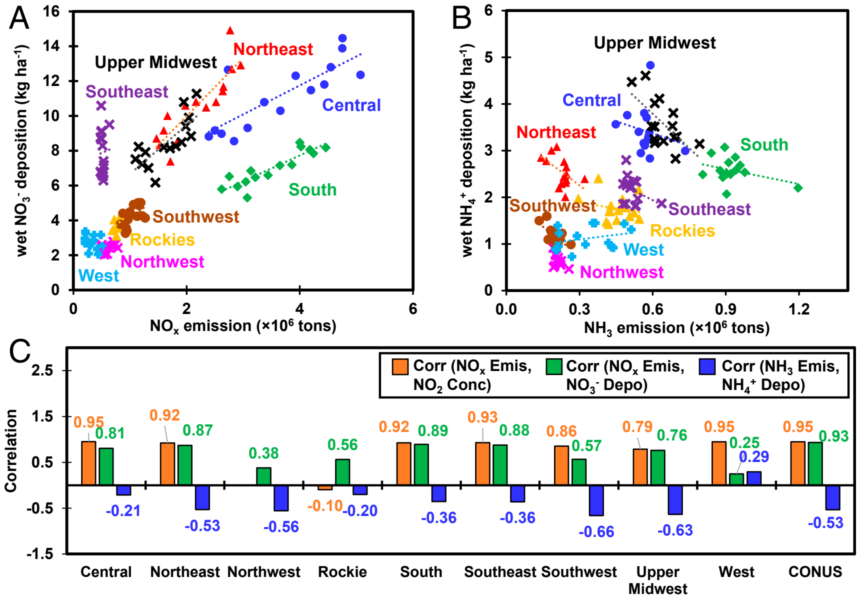

Fig. 2. Relationships between N emissions and N deposition. Scatter plots of regional emissions with deposition for (*A*) NO_x emission and NO_3^- wet deposition, and (*B*) NH_3 emission and NH_4^+ wet deposition from 2000 to 2014 for nine CONUS regions. (*C*) Correlations between NO_x emissions and NO_2 concentration (orange bars), between NO_x emissions and NO_3^- wet deposition (green bars), and between NH_3 emissions and NH_4^+ wet deposition (blue bars). Northwest lacked sufficient NO_2 observations.

Spatial Changes of N Emissions and N Deposition. Spatial distributions of percentage changes in N emissions and N deposition (wet + dry) from 2001 to 2010 are shown in Fig. 3. Annual maps of N deposition were developed by combining the site observation data with deposition fields simulated by a chemical transport model provided by the Total Deposition Science Committee (TDEP) of the National Atmospheric Deposition Program (NADP) (*Materials and Methods*). Over this period, NO_x emissions (Fig. 3*A*) decreased by 15–30% over CONUS, with slight increases in some regions, such as Montana (MT) and Nevada (NV). NO_y deposition responses (Fig. 3*B*) were generally consistent with the changes in NO_x emissions in both amount and distribution. NH_3 emissions (Fig. 3*C*) increased by 10–30% over CONUS, whereas NH_x deposition (Fig. 3*D*) exhibited mixed responses in different states. We found that the locations of areas with increased NH_x deposition agreed well with areas of increased NO_y deposition (Fig. 3*B*, contours with yellow and orange) and increased S deposition (*SI Appendix*, Fig. S2) in large extended regions around the states of MT, California (CA), Nebraska (NE), and Texas (TX). Changes in NH_4^+ wet

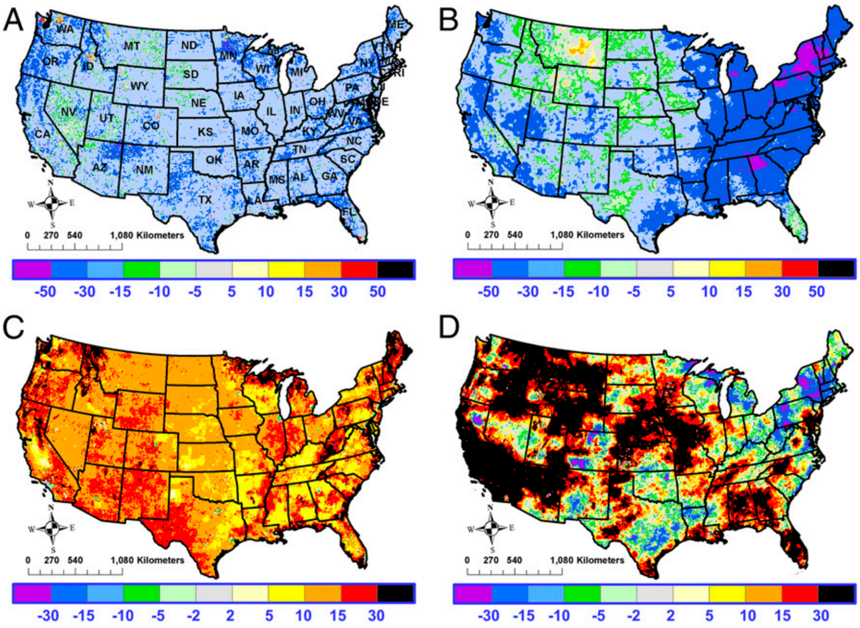

Fig. 3. Spatial distributions of the percentage changes of N emissions (derived from NEI) and deposition (derived from TDEP) from 2001 to 2010. Percentage changes of (*A*) NO_x emissions, (*B*) NO_y deposition, (*C*) NH_3 emissions, and (*D*) NH_x deposition. The percentage changes were calculated as 100 × (Emission of 2010 − Emission of 2001)/(Emission of 2001).

deposition were strongly influenced by the acidic components of wet deposition (mainly SO_4^{2-} and NO_3^-) since NH_4^+ is the main alkali neutralizer. Thus, in regions with decreased NO_y deposition, only a portion of the increased NH_3 emissions partitioned to the aerosol phase (NH_4^+). Once NH_3 emissions are controlled, the remaining NH_3 in the atmosphere might partition to particulate NH_4^+ as the aerosol becomes more acidic (26).

Response of N Deposition to N Emission Abatement. We calculated the response (Res) of deposition of NO_y or NH_x to emission control over continental regions with Eq. **1**, using results from 11 global climate models under base case (5) and emission perturbation scenarios (27) (*Materials and Methods*).

$$\text{Res} = \frac{\text{Predicted \% change of } NO_y \text{ or } NH_x \text{ deposition under emission changes over continental regions}}{\text{Predicted \% change of } NO_x \text{ or } NH_3 \text{ emissions under emission changes over continental regions}}. \quad \textbf{[1]}$$

The response of NO_y deposition to controlling NO_x emissions (Fig. 4*A*) generally ranged from 80 to 120% over most CONUS regions. The ±20% variation around 100% could be a result of meteorological factors such as precipitation and long-range transport, in agreement with findings from Hemispheric Transport of Air Pollution, second phase (HTAP II) simulations (25), and could be viewed as impacts of long-term climate change. Some areas in MT, South Dakota (SD), and NE exhibited extraordinary values (~200%) (Fig. 4*A*, rust color), owing to very low (<5%) reductions in NO_x emissions (*SI Appendix*, Fig. S3*A*). Overall, the average Res value of NO_y deposition approached the ideal value of 100%.

The response of NH_x deposition to controlling NH_3 emissions ranged from 60 to 80% (Fig. 4*B*) in most CONUS regions. Spatial distributions of the Res values were more consistent among nearby regions for NH_x deposition (Fig. 4*B*) than NO_y deposition (Fig. 4*A*). A possible explanation is that NO_x can be transported over relatively long distances via reservoir species (27). Gaseous NH_3 is removed mainly by wet and dry deposition, with only minor consumption in slow oxidation of NH_3 by OH (24). Importantly, the 20–40% shortfall to "full" effects indicates that additional reduction of NH_3 emissions is needed to reach the target for NH_x deposition.

Implications and Summary. Despite a tight connection between NO_y deposition and NO_x emissions, such a strong relationship does not exist between NH_3 emissions and NH_x deposition. In this study we controlled the emissions of SO_2, NO_x, and NH_3 simultaneously. It mimicked the situation that we are likely to face in the future, in which NO_x and SO_2 emission control strategies remain in effect. As NH_x deposition increased, NH_3 emission control was added to the existing control policies. We find that NH_x deposition would resist decrease after NH_3 emissions are reduced. Reduction in SO_2 and NO_x emissions leads to reduction of aerosol SO_4^{2-} and NO_3^- but also to reduction in the associated aerosol water; thus, the level of acidity does not change appreciably (24). The overall result is promotion of the release of NH_4^+ to the gas phase, which increases the dry deposition of NH_x.

The sensitivity of NH_x dry deposition to NH_3 emission reduction is not included in the present study. Vegetation can serve as either a sink or source of atmospheric NH_3 depending on the balance between the NH_3 concentrations in the atmosphere and those in the stomata of leaves, which is referred to as the compensation point (28). This bidirectional NH_3 flux can increase the model prediction of NH_3 emission flux by 0–70% (29). It also increased the model prediction of NH_3 air concentration by 0–75%, with high influences during June–October and low impacts during December–February (30). A modeling study employing the Long Term Ozone Simulation–European Operational Smog (LOTOS-EUROS) model (31) with compensation point found that NH_3 dry deposition flux would only decrease about 10–15% (seasonal variations) when NH_3 emissions were decreased by 30%, and would increase about 15–25% when NH_3 emissions were increased by 30%. The compensation can be viewed as a buffering effect that weakens the sensitivity of NH_x dry deposition to NH_3 emission change.

It is concluded that significant reduction of NH_x deposition is not likely to emerge in the early stage of NH_3 emission control. Our modeling results estimate that a 60–80% reduction of NH_x deposition can be achieved per unit of NH_3 emission control, considerably lower than the full benefits of controlling NO_x emissions to reduce NO_y deposition. The management of NH_x deposition is likely to be a focus for many regions in the near future. This "weakened" sensitivity needs to be taken into consideration

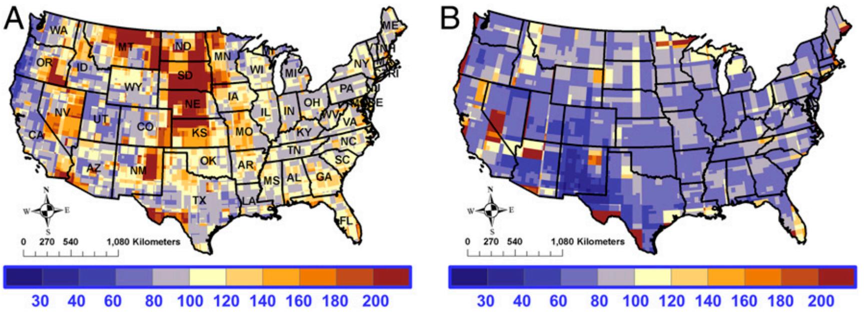

Fig. 4. Predicted responses of N deposition (wet plus dry) to N emissions abatement in 2010. (*A*) Response of NO_y deposition to NO_x emission control. (*B*) Response of NH_x deposition to NH_3 emissions control.

when developing control strategies for NH_3 emissions. The results of the present study for the United States are likely applicable to regions with intensive NH_x deposition rates, such as India, and regions in which the dominant deposition components are in transition from NO_y to NH_x, such as Europe.

Materials and Methods

Site observations of wet NO_3^- deposition and wet NH_4^+ deposition were provided by the National Trends Network (NTN) of the NADP (http://nadp.slh.wisc.edu/NTN/). NTN measured the wet deposition at 373 sites located throughout the entire United States (*SI Appendix*, Fig. S4) from 1980s until present day. This study used the annual accumulated wet deposition data from 244 sites with available data in the study period (*SI Appendix*, Table S2). Site observations of airborne NO_2 concentrations were provided by Air Quality System Monitoring Network of US Environmental Protection Agency (US EPA) (https://www.epa.gov/air-trends/nitrogen-dioxide-trends) (*SI Appendix*, Fig. S5 and Table S2). Spatial distributions of NO_y and NH_x deposition were developed by the TDEP by spatial interpolation of quality-controlled observation sites with model simulations (http://nadp.slh.wisc.edu/NTN/maps.aspx). Annual amounts of NO_x and NH_3 emissions were provided by US EPA's NEI (https://www.epa.gov/air-emissions-inventories/national-emissions-inventory-nei). Spatial distributions of emissions were obtained from the Emissions Database for Global Atmospheric Research (EDGARv4.3.2) (https://edgar.jrc.ec.europa.eu/overview.php?v=432) at 0.1 × 0.1° resolution (∼11.1 km × 11.1 km). The EDGAR emissions over CONUS were developed based on information from US EPA's NEI (32).

The responses of deposition to emission changes were determined from simulated NO_y and NH_x deposition from multimodel mean (MMM) results of 11 global climate models from HTAP II, directed by the United Nations Economic Commission for Europe (http://www.htap.org/) (33). The models used were CAM-Chem, CHASER_rel, CHASER_t106, EMEP_rv48, GEMMACH, GEOS5, GEOSSCHEMAJOINT, OsloCTM3v.2, GOCARTv5, SPRINTARS, and C-IFS_v2. Simulations were conducted for 2010, with an additional 6 mo as spin-up. The modeled NO_y deposition comprises deposition of NO_2, HNO_3, aerosol NO_3^-, peroxyacyl nitrate (PAN), and organic nitrates other than PAN. MMM performance on wet deposition of NO_y and NH_x was evaluated with the NADP observation network (ref. 5 and *SI Appendix*, Fig. S6). Simulation of dry deposition is subject to uncertainty, owing to lack of comprehensive observation data (5, 34). The base case utilized the HTAP v2.2 emissions inventory (35). In the emission control scenario, a modified emissions inventory was used with 20% reduction of all anthropogenic emissions, including SO_2, NO_x, and NH_3 over North America. Since the meteorological fields and model parameters remained unchanged in the base case and control scenarios, emissions change is the main factor contributing to the variation of deposition.

Data Availability. All data are publicly available. Details about measurement data, emission, and modeling datasets are given in *SI Appendix*.

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