Intense Warming Will Significantly Increase Cropland Ammonia Volatilization Threatening Food Security and Ecosystem Health

Highlights
- By 2100 warming will increase US agricultural land ammonia emissions by up to 81%
- This increasing trend will be associated with significant reduction in crop yields
- It will increase ammonia concentrations in the air and subsequent deposition
- Feasible control strategies can completely offset this trend under global warming

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In Brief
Humans, through agricultural fertilizer application, inject more reactive nitrogen (N\textsubscript{r}) to terrestrial ecosystems than do natural sources. Ammonia volatilization is a major pathway of agricultural N\textsubscript{r} loss. Using a process-based dynamic model, Shen et al. show that ammonia volatilization from agricultural land in the US will increase by up to 81\% by the end of this century due to climate change alone, posing threats to food security, air quality, and ecosystem health, but mitigation strategies are available.
Intense Warming Will Significantly Increase Cropland Ammonia Volatilization Threatening Food Security and Ecosystem Health

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SUMMARY
Cropland ammonia volatilization ($V_{NH3,AG}$) is a major pathway of agricultural nitrogen loss. It remains unclear, however, how climate warming and human intervention (e.g., agricultural management) will affect $V_{NH3,AG}$. Here, we use a fully coupled agroecosystem/chemical transport model and multiple climate projections to quantify the changes in climate-induced $V_{NH3,AG}$ over the US. We show that climate change under an intensely warming scenario will increase $V_{NH3,AG}$ by 81% (95% confidence interval, 69%–92%) from 2010 to 2100. The increase in $V_{NH3,AG}$ will cause a 10% loss of nitrogen applied, decrease crop yields by 540 Gg-N year$^{-1}$, increase atmospheric burden of ammonia/ammonium by 18%, and increase ammonia/ammonium deposition to sensitive ecosystems by 14%. We have found that combining climate-adaptive agricultural management practices with attainable mitigation measures can completely offset the warming-induced increment in $NH3$ emissions and improve crop production, air quality, and ecosystem health.

INTRODUCTION
Ammonia ($NH3$) in the air is the key constituent controlling and stabilizing aerosol acidity$^{1,3}$ and is a major contributor to secondary aerosol that has adverse impacts on human health and climate.$^{3,4}$ Excessive $NH3$ deposition is harmful to sensitive ecosystems as it causes soil acidification, eutrophication, and biodiversity loss.$^{7-11}$ Globally, approximately 43% of the atmospheric $NH3$ emission is attributed to volatilization from soils and plants over fertilized agricultural land ($V_{NH3,AG}$)$^{12}$ due to the application of synthetic fertilizer and manure (agricultural land here refers to the cultivated land used for growing crops, hay, and pasture; $V_{NH3,AG}$ in this study denotes the $NH3$ volatilization from application of fertilizer and manure; $V_{NH3,AG}$ does not include other livestock management processes except land application of manure). Currently, 10%–20% of the agricultural nitrogen (N) applied worldwide is lost via $V_{NH3,AG}$ with important consequences for the global N cycle.$^{13-15}$

$V_{NH3,AG}$ may increase as climate warms$^{12,16-19}$ because warmer conditions are thermodynamically favorable for increased $NH3$ release from multiple surface layers (e.g., soil and stomatal and moisture layers on the leaf cuticle) regulated...
NH3 deposition (as a soil N source) enhances the loss of soil reactive N (N$_r$, which includes both oxidized and reduced N species, such as nitrate and N$_2$O), and (3) alters soil pH, which affects the NH$_3$/NH$_4^+$ equilibrium. Ambient NH$_3$ concentrations and deposition responding to $V_{NH3,AG}$ changes reveal negative and positive feedback, respectively. NH$_3$ deposition (as a soil N source) enhances $V_{NH3,AG}$, and ambient NH$_3$ concentrations suppress $V_{NH3,AG}$ by increasing cuticular resistances. Management practices, such as the type, amount, timing, and methods of N application will also evolve under climate change, which further complicates the climate dependency of $V_{NH3,AG}$. A quantitative assessment of $V_{NH3,AG}$’s dependency on future climate change requires integrating the dynamics and interactions of plants, soils, atmosphere, and human activities.

Past studies used empirical relationships between climatic drivers and NH$_3$ volatilization in natural circumstances (e.g., seabird colonies) to project future changes in $V_{NH3,AG}$ due to climate change. However, $V_{NH3,AG}$ differs from NH$_3$ volatilization over non-agricultural land that naturally occurs because human intervention plays a critical role in $V_{NH3,AG}$. So far, few studies have used process-based dynamic models to evaluate the changes in $V_{NH3,AG}$ in response to future climate change. Here, we use a process-based fully coupled agroecosystem-air quality model, FEST-C-EPIC-CMAQ_BIDI, to elucidate the climate change impacts on future $V_{NH3,AG}$ in the US, one of the nations with the largest coverage of agricultural land (Figure 1). Our model, FEST-C-EPIC-CMAQ_BIDI, consists of an agroecosystem model (Environmental Policy Integrated Climate [EPIC] model) and a chemical transport model (Community Multiscale Air Quality model [CMAQ]) using an interface (the Fertilizer Emission Scenario Tool for CMAQ [FEST-C]) and an NH$_3$ bidirectional exchange module (BIDI) to couple the two models (Experimental Procedures). For $V_{NH3,AG}$, we consider both synthetic fertilizer and manure application, which is estimated to contribute 30% of the total NH$_3$ emission in the US in 2011 (Table S1). FEST-C-EPIC simulates 42 rain-fed and irrigated crops (e.g., corn grain and soybean) and grasses (e.g., hay and alfalfa) (Table S2) and considers various fertilizer types (see Data and Code Availability) and application methods.34,35

Our model, FEST-C-EPIC-CMAQ_BIDI, simultaneously assesses the responses of multi-media environmental processes and agricultural management practices to climate change and is expected to be a suitable tool to investigate the climate dependency of $V_{NH3,AG}$ (Figure 1). We consider two climate scenarios comprised of the Representative Concentration Pathway (RCP) 4.5 (a climate scenario showing a moderate increasing trend in temperature) and RCP8.5 (a climate scenario showing an intensely increasing...
trend in temperature). Based on the integrated modeling framework, we evaluate the effectiveness of mitigation strategies for reducing \( V_{NH3,AG} \). These strategies include adaptation of management practices (i.e., amount and timing of fertilizer application) to climate change and deployment of mitigation measures (comprised of replacement of urea with non-urea fertilizers [RU], urea application with irrigation [UIR], application of urease inhibitor [UIN], and deep placement of fertilizers [DP]).

**RESULTS**

**Observed Climate Dependency of \( V_{NH3,AG} \)**

Satellite retrievals suggest an increasing trend (+40%) in \( NH3 \) concentration across the Midwest agricultural region (35°N–45°N, 87°W–101°W) during the last 13 years (Figure S1).\(^{16,36}\) The expansion of agricultural land (+0.5%) and an increasing use of fertilizer (+8%) can partly explain this increase in \( NH3 \) concentration.\(^{16,36}\) Reductions in \( NOx \) and \( SO2 \) emissions have played a minor role, given that Reductions in \( NOx \) and \( SO2 \) emissions have played a minor role, given that no significant trend in the observed \( NH3 \) levels is found in the eastern US despite large emission reductions in \( NOx \) and \( SO2 \).\(^{-1}\) On the other hand, the interannual \( NH3 \) variation from which the long-term trend is removed by detrending, shows a significantly positive correlation with annual mean 2-m temperature and a negative correlation with total precipitation \((p < 0.05)\) (Figure S1), which indicates a potential climate dependency. Satellite retrievals suggest that, in 2012, \( NH3 \) concentrations in the Midwest reached their highest levels, which coincides with the highest temperature on record and dryer-than-normal conditions.\(^{37}\) Consistent with the space-based observations, ground measurements operated by the Ammonia Monitoring Network peaked in 2012 at eight of the ten sites within this region (Figure S2).\(^{38}\)

The occurrence of high \( NH3 \) levels in 2012 could be attributed to emissions, gas-particle partitioning, and reduced removal processes that link to the abnormal climate conditions. To determine to what extent emission changes contributed to the \( NH3 \) level raise, we conduct simulations based on FEST-C_EPIC–CMAQ_BIDI. Compared with CMAQ simulations without coupling the agroecosystem feedbacks, simulations of FEST-C_EPIC–CMAQ_BIDI show better agreement with observations—the Normalized Mean Biases of annual mean \( NH3 \) concentrations and \( NH4^+ \) wet and dry deposition reduce from −52% to −30%, −47% to −32%, and −14% to −5%, respectively (Figures S3 and S4; Table S3) (Supplemental Information for more details on model evaluation). By comparing the modeled \( NH3 \) concentrations between 2011 (a year with generally normal temperature and precipitation levels over the Midwest agricultural region) and 2012, we estimate the sensitivities of \( NH3 \) concentration to 2-m temperature \((S_{C,T})\), which average +0.18 ppb K\(^{-1}\) in the Midwest and +0.33 ppb K\(^{-1}\) at the site locations, in line with the \( S_{C,T} \) values derived from space and ground-level observations (+0.15 and +0.34 ppb K\(^{-1}\), respectively). FEST-C_EPIC–CMAQ_BIDI estimates a 20% increase in \( V_{NH3,AG} \) (from 400 to 480 Gg-NH\(_3\) year\(^{-1}\)) between the two years that is solely caused by changes in meteorology. To isolate the emission-related impact, we turned off the agroecosystem feedbacks and applied the 2011 \( V_{NH3,AG} \) to the 2012 simulation. The simulation shows a 40% reduction in \( S_{C,T} \), meaning that the \( V_{NH3,AG} \) increase is responsible for about 40% of the large-scale increase in \( NH3 \) concentrations (Figure S5). The contribution can be up to 90% in certain areas of the Midwest. This analysis highlights the important role of \( V_{NH3,AG} \) leading to the observed \( NH3 \) pulse in 2012 and also supports the suitability of FEST-C_EPIC–CMAQ_BIDI in capturing the \( NH3 \) air-surface dynamics in response to climate change.

**Increased \( V_{NH3,AG} \) under Future Climate Warming**

We use FEST-C_EPIC–CMAQ_BIDI to address the \( V_{NH3,AG} \) response under future climate change. The meteorological fields used to drive FEST-C_EPIC–CMAQ_BIDI are generated by downsampling the bias-corrected climate projections generated by the National Center for Atmospheric Research’s Community Earth System Model version 1 (CESM1).\(^{39}\) We conduct FEST-C_EPIC–CMAQ_BIDI simulations under climate conditions of the 2010 (2008–2012), 2050 (2048–2052), and 2100 (2096–2100) periods (Experimental Procedures). To avoid bias caused by climate anomalies, each period is delineated by a 5-year simulation. To isolate the direct impact of climate change, land cover (LC) distributions and \( CO2 \) concentrations for this set of simulations remain unchanged (a brief discussion on the potential impacts of future changes in LC and \( CO2 \) concentration on \( V_{NH3,AG} \) can be found in the Supplemental Information). In the RCP4.5 climate scenario, the projected \( V_{NH3,AG} \) shows an average increase of 12% and 23% by 2050 and 2100 compared with 2010, respectively, over the CONUS (Table S4). Substantial increases are found in the Northern and Southern Plains (26% and 35% by 2100) and the Corn Belt (23%), while emissions in the eastern and western coastal regions show lower increases (Northeast −1%, Appalachia 7%, Southeast 12%, and Pacific 7%) (see Figure S6 for a map of the 10 farm production regions used in this study). In the RCP8.5 scenario, the projected \( V_{NH3,AG} \) shows a striking increase of 59% by 2100, with higher increases occurring in the Southern Plain (82%) and the Corn Belt (79%) and increases of >50% in all other regions except the Pacific (43%) and the Northeast (39%) (Table S4). Mean differences in \( V_{NH3,AG} \) between 2010 and 2100 RCP8.5 simulations (+658 Gg-NH\(_3\) year\(^{-1}\)) are significantly higher than the standard deviations within each period (90 and 70 Gg-NH\(_3\) year\(^{-1}\) for 2010 and 2100 periods, respectively) (Table S4), suggesting a persistent long-term increase despite well-marked interannual variations.

Based on the simulations, we develop three reduced-form models (RFMs) to decompose the spatiotemporal variations in \( V_{NH3,AG} \) into various factors, including LC types, climate factors, and agricultural management practices (Experimental Procedures). The first RFM (LC-based RFM) solely uses LC types to determine \( V_{NH3,AG} \); the second RFM (RFM\(_C\)) builds on the LC-based RFM to include interannual climate variation; the third RFM (RFM\(_{C,M}\)) further includes agricultural management practices (Experimental Procedures). The performances of these three RFMs are discussed below. The LC-based RFM explains 68% \((R^2)\) of the spatial variation. All types of vegetated land are subject to \( NH3 \) volatilization \((V_{NH3})\) denotes \( NH3 \) volatilization from any type of vegetated land.\(^{40}\) The regression coefficients \((\alpha)\) representing emission intensities vary by more than two orders of magnitude across different LC types (Figure S7). \( V_{NH3,AG} \) generally shows much higher emission intensities than \( NH3 \) volatilization from non-agricultural land. The highest emission
intensities are found for rice (18.7 kg-NH$_3$ ha$^{-1}$ year$^{-1}$) and cotton fields (12.0 kg-NH$_3$ ha$^{-1}$ year$^{-1}$) due to high N fertilizer input for crop growth.\textsuperscript{31,42} The lowest is found over forest (0.2 kg-NH$_3$ ha$^{-1}$ year$^{-1}$). Overall, $V_{\text{NH}_3}$ dominates (>80%) the $V_{\text{NH}_3}$ in the CONUS (Supplemental Information for more details).

We calculate the ratios of the FEST-C-EPIC–CMAQ_BIDI-modeled $V_{\text{NH}_3}$ to the RFM-reproduced $V_{\text{NH}_3}$ and find that the ratios are log-normally distributed with a spatial gradient increasing from the north to the south (Figure S8). After log-transformation, the ratios are positively correlated with 2-m temperature ($T$) ($r = 0.76$, $p < 0.001$), which largely explains the latitudinal gradient—higher ratios in the south due to warmer climates. In addition, the ratios show a positive correlation with near-surface wind speed ($W$, 10-m wind speed) ($r = 0.05$, $p < 0.001$) and a negative correlation with total precipitation ($P$) ($r = -0.21$, $p < 0.001$). These correlations warrant inclusion of meteorological variables in the RFM. We use an exponential term as the climate-adjustment factor to account for $T$, $W$, and $P$ (Experimental Procedures) following previous studies.\textsuperscript{12,43} The enhanced model (i.e., RFMC) explains 95% of the $V_{\text{NH}_3}$ variance, significantly improved from the previous version. The model predictions resemble the gridded $V_{\text{NH}_3}$ values simulated by FEST-C-EPIC–CMAQ_BIDI (Figures S9A and S9B). Decomposing the effects of LC and the three climate components (i.e., $T$, $W$, and $P$) shows that $T$ is the dominant factor leading to the $V_{\text{NH}_3}$ increases. Under RCP4.5, increased $T$ in 2100 leads to a net increase of 34% in $V_{\text{NH}_3}$, while the changes in $W$ and $P$ lead to decreases in $V_{\text{NH}_3}$ (−3.3% and −3.4%, respectively). Under RCP8.5, however, RFMC tends to overestimate the increase in $V_{\text{NH}_3}$ by 22% in 2100 (Figure S10).

In most studies as well as in RFMC, it is assumed that the long-term dependence of $V_{\text{NH}_3}$ over agricultural land follows a similar function as that of natural circumstances, e.g., seabird colonies, where $\ln(V_{\text{NH}_3})$ is a linear function of $1/T$ as $\Delta \ln(V_{\text{NH}_3}) = \alpha \cdot (1/T)$, where the values of $\alpha$ are always negative and differ by LC type (Experimental Procedures).\textsuperscript{12,43} The sensitivity of $V_{\text{NH}_3}$ to $T$ ($S_{\Delta T}$, denotes the percentage change in $V_{\text{NH}_3}$ to the absolute change in $T$) derived from the $V_{\text{NH}_3} - T$ function is inversely proportional to $T^2$ ($S_{\Delta T} = -\alpha/T^2$), indicating that $V_{\text{NH}_3}$ becomes less sensitive to $T$ at higher $T$ levels. Our RFMC shows an $\alpha$ value of −8.690 K for unirrigated crop, equivalent to an $S_{\Delta T}$ of 10.2% K$^{-1}$ at 18°C and a slightly lower $S_{\Delta T}$ (9.8% K$^{-1}$) at 25°C. The $S_{\Delta T}$ directly calculated from FEST-C-EPIC–CMAQ_BIDI via a year-by-year comparison shows a very similar $S_{\Delta T}$ (10.2% K$^{-1}$) at 18°C but a much lower $S_{\Delta T}$ (4.7% K$^{-1}$) at 25°C. The difference between FEST-C-EPIC–CMAQ_BIDI-modeled and RFMC-reproduced $S_{\Delta T}$ suggests that the $V_{\text{NH}_3}$ in RFMC is overly sensitive to $T$ changes at higher $T$ levels (Figure 2). For non-agricultural land, on the other hand, the $S_{\Delta T}$ obtained from RFMC and FEST-C-EPIC–CMAQ_BIDI match each other over a wide range of $T$ changes (for RFMC, 9.9%–14.8% and 9.5%–14.1% at 18°C and 25°C, respectively; for FEST-C-EPIC–CMAQ_BIDI, 12.0% and 11.4%, respectively) (Figure S11). The functional difference of $\Delta T$ at higher $T$ levels between agricultural and non-agricultural lands reflects LC-determined heterogeneous responses of $V_{\text{NH}_3}$ to warming and implies that the true response for agricultural land could be oversimplified if directly using the semiempirical relationship obtained from natural circumstances. Further investigation of the FEST-C-EPIC–CMAQ_BIDI simulations reveals that the climate change under RCP8.5 leads to a
adaptive to climate change (Figures 3 and 4; Tables S5–S7). All evaluated the benefits from adopting management practices. Vlyses consistently show the effect of management practices on climate models,46,47 isolated the impacts of increases in NH3,AG under climate warming, we introduce a nonlinear cubic polynomial function in RFM C to developed RFM C,M (i.e., RFM C = C0 + C1/T + C2/T^2 + C3/T^3). Based on RFM C and RFM C,M, we projected the future trends of NH3,AG. The analyses consistently show the effect of management practices on NH3,AG suppression in FEST-C_EPIC–CMAQ_BIDI and imply that the climate dependency of NH3,AG described by RFM C is close to a scenario where fixed management practices are adopted irrespective of changing climate.

To reflect the suppression effect of management practices on NH3,AG under climate warming, we introduce a nonlinear cubic polynomial function in RFM C to developed RFM C,M (i.e., RFM C = C0 + C1/T + C2/T^2 + C3/T^3). Using high-resolution meteorological fields downscaled from climate projections of 18 climate models,46,47 isolated the impacts of increases in T, and evaluated the benefits from adopting management practices adaptive to climate change (Figures 3 and 4; Tables S5–S7). All NH3,AG projections show increasing trends between 2010 and 2100 (Figure 3), with overall increases ranging from +1% (based on GFDL-ESM2G) to +51% (HadGEM2-CC36S) under RCP4.5 and from +47% (GFDL-ESM2M) to +128% (HadGEM2-CC36S) under RCP8.5 (Tables S6 and S7). Sixteen out of 18 projections show more than 50% increases over at least two-thirds of the CONUS land under RCP8.5, but the changes are not spatially consistent among models (Tables S6 and S7) due to the disparity in projected extents of warming among models. Excluding the impacts of T eliminates the increasing trends (Figure 3) and significantly narrows the variation across projections, indicating the dominant role of warming in future NH3,AG increases. The multi-projection means show an increase of 285 Gg-NH3 year^−1 (95% confidence interval [CI], 222–340 Gg-NH3 year^−1) or 26% (85% CI, 20%–31%) in NH3,AG between 2010 and 2100 under RCP4.5 and an increase of 900 Gg-NH3 year^−1 (95% CI, 767–1,020 Gg-NH3 year^−1) or 81% (95% CI, 69%–92%) under RCP8.5 (see Figure 4 for the spatial distributions of the increases under both climate scenarios). The NH3,AG increase under RCP8.5 is significantly higher than the single-projection estimate using CESM1. We have found that NH3,AG accounts for 10% of the total fertilizer N applied in 2010, and the fraction increases to 22% in 2100 under RCP8.5 (if not adopting adaptive management practices). We subsequently evaluate the impacts on crop yield (expressed as N yield in Gg-N year^−1), atmospheric burden of NH3/NH4^+, and N deposition. N deposition loss is defined as the reduction in the total yield of the 42 types of crops considered in this study (Table S2) caused by a decrease in the N input and is calculated using the hyperbolic relationship between yield and the effective total N input following previous studies.48,49 We have found that the NH3,AG increase is responsible for a 10% loss of total nitrogen applied and a 540 Gg-N year^−1 loss of crop yields (or 3.7% of the total crop yield), increases the atmospheric burden of NH3/NH4^+ by 18% across the CONUS, and enhances the NH3/NH4^+ deposition to sensitive ecosystems by 14% (or increases the total N deposition to sensitive ecosystems by 8%) (Experimental Procedures) (Figures S12 and S13, see Figure S14 for the definition of sensitive areas). RFM C,M shows that adopting adaptive management practices suppresses the NH3,AG increase in 2100 only mildly under RCP4.5 (40 Gg-NH3 year^−1 or 14% of the NH3,AG increase) but greatly under RCP8.5 (242 Gg-NH3 year^−1 or 27% of the NH3,AG increase).
increase) (Figure 3). Nevertheless, adaptive management practices cannot fully offset the \( V_{NH3,AG} \) increase induced by climate change.

**Mitigation Strategies**

The increasing \( V_{NH3,AG} \) under warming urges the need for adopting mitigation strategies that combine adaptive management practices with other feasible mitigation measures. We evaluate the effectiveness of four feasible mitigation measures (Experimental Procedures). Three of the measures (RU, UIR, and UIIN) target urea application, which amounts to one-third of \( V_{NH3,AG} \). The fourth, DP, is applicable to all types of fertilizers. We have found that full deployment of these individual measures by 2100 (RCP8.5) lowers the \( V_{NH3,AG} \) by 500 (RU), 180 (UIR), 280 (UIIN), and 700 (DP) Gg-N year\(^{-1}\), respectively. The most effective strategy, which combines RU and DP with adaptive management practices, reduces the \( V_{NH3,AG} \) by 1,186 Gg-NH\(_3\) year\(^{-1}\) (59%), where mitigation measures account for 83% of the reduction, and adaptive management practices account for the remaining 17%. The amount of \( V_{NH3,AG} \) reduced prevents loss of 13% of applied N, with subsequent crop yield gain of 735 Gg-N year\(^{-1}\) (5.0% of the total crop yield), and leads to an 18% decrease in the atmospheric burden of NH\(_3\)/NH\(_4^+\) in the CONUS and a 16% decrease in the NH\(_3\)/NH\(_4^+\) deposition to sensitive ecosystems (or a 10% decrease in the total N\(_r\) deposition to sensitive ecosystems) (Figures S12 and S13).

**DISCUSSION**

Our study uses a process-based model to project future \( V_{NH3,AG} \) trends associated with climate change. We have found that an intense warming (RCP8.5) between 2010 and 2100 will increase the \( V_{NH3,AG} \) by ~81% over the CONUS, and that a moderate warming (RCP4.5) can significantly prevent the increasing trend (only an increase of ~26%). All else unchanged, the increases in \( V_{NH3,AG} \) will lower agricultural nitrogen use efficiency (NUE). A first-order estimate shows that by the end of 21st century of RCP8.5, the amount of N loss via \( V_{NH3,AG} \) over the CONUS will be equal to the amount of crop N yield in California—the largest crop-producing state in the US. While our study focuses on the CONUS, similar \( V_{NH3,AG} \) increases are anticipated globally. Mid- and high-latitude regions may see higher \( V_{NH3,AG} \) increases due to the greater intensification of regional warming, which warrants further investigation. By the end of the 21st century, the world population will grow by approximately 35%–80%, with food demand increased by 30%–160%. The potential decline in NUE due to rising \( V_{NH3,AG} \) undermines the efforts to address the need to meet growing food demand, posing a global threat to food security.

Using the process-based model outputs, we develop RFMs to decompose the \( V_{NH3,AG} \) variations associated with various factors, including LC types, climate factors, and agricultural management practices. The RFM method represents an approach to effectively provide projections of \( V_{NH3,AG} \) under climate change and modulates the prescribed NH\(_3\) emission inventories that are currently used in most models in a way that takes climate impacts into consideration. However, it is not entirely appropriate to apply our RFMs directly to other world regions. Different parameterizations of the RFMs are expected for different regions given the differences in crop and fertilizer types, climate, soil property, and agricultural practice. Process-based models can be used to translate these regional differences into region-specific parameterizations for RFMs developed by others.

The \( V_{NH3,AG} \) increases have far-reaching implications for human and ecosystem health and the global climate because of the significance of the resulting enrichment of atmospheric burden and deposition of NH\(_3\)/NH\(_4^+\) species. Our simulations find that not only the boundary layer but also the free troposphere see considerable NH\(_3\)/NH\(_4^+\) enrichment owing to the \( V_{NH3,AG} \) increases (Figure S15). While in the boundary layer, NH\(_3\) contributes to the formation of secondary aerosols that are associated with adverse health effects, in the free troposphere, the increases in NH\(_3\) may have important implications for cloud formation.

Currently, anthropogenic activities inject more than 2-fold N\(_r\) to terrestrial ecosystems as natural sources do (190 versus 84 Tg-N year\(^{-1}\)). Synthetic fertilizer application dominates the anthropogenic input (~60%), followed by crop N fixation and fossil fuel combustion (~20% each). All else equal, the warming-induced increases in \( V_{NH3,AG} \) indicate a greater demand for fertilizer to achieve current crop yield, thereby releasing more N\(_r\) to the environment. As it cascades through the environment, the same atom of N transforms into a sequence of different forms and causes multiple effects in series. Adverse impacts of the N cascade are inevitable unless the process is restricted from where the N\(_r\) pollution starts. We show that adopting management practices adaptive to climate change and deploying feasible mitigation measures can decrease \( V_{NH3,AG} \), and that adaptive management practices will be increasingly more effective as climate warms. The reduction in \( V_{NH3,AG} \) caused by implementation of these adaptation actions will fully compensate for the warming-induced increase in \( V_{NH3,AG} \) and come along with improved NUEs, maintaining the crop yield while effectively reducing N\(_r\) input to terrestrial ecosystems.
We focus on the \(V_{\text{NH3,AG}}\) increases caused by climate change alone, but \(V_{\text{NH3,AG}}\) will also grow with global expansion of agriculture and associated fertilizer use driven by growing population and per-capita resource demand.\(^{62,63}\) In addition to \(V_{\text{NH3,AG}}\) there are other pathways of N loss that are likely disturbed by climate change, e.g., surface runoff, nitrate leaching, emissions of nitrogen oxide and nitrous oxide from nitrification and denitrification.\(^{62-64}\) In the context of increasing food demand on top of a changing climate, preventing the unexpected exacerbation of agricultural N loss will require better understanding of the dynamical coupling between human and natural systems, which is the key to integrative process-based modeling of agroecosystems, as demonstrated in part in this study. Extended ground- and space-based measurements are needed to better constrain the magnitudes of N loss from various pathways.\(^{62}\) The next generation of the process-based models should enable integration of the various pathways of N loss with consideration of multi-media consequences, and incorporation of strategies, technologies, and infrastructure into promising solutions for sustainable development of agriculture.

**EXPERIMENTAL PROCEDURES**

**Resource Availability**

**Lead Contact**

Further information and requests should be directed to and will be fulfilled by the Lead Contact, Huizhong Shen (hshen73@gatech.edu).

**Materials Availability**

This study did not generate new unique materials.

**Data and Code Availability**

The source code of FEST-C_EPIC-CMAQ_BIDI can be freely downloaded from the CMAS center at https://www.cmascenter.org/fest-c/ and from ZENODO at https://zenodo.org/record/3438888#.Xud8WWhkg2W. The bias-corrected CMIP5 outputs used to derive the future meteorological fields are available at https://rda.ucar.edu/datasets/da316.1/. A complete list of fertilizers modeled in FEST-C_EPIC_CMAQ-BIDI are available at https://osf.io/5efua/.

**Integrated Model**

We use FEST-C v.1.4, CMAQ v.5.3, and an adapted EPIC v.0509, which comprises the most updated version of FEST-C-EPIC-CMAQ_BIDI, to conduct our simulations. EPIC is used to determine the amount and timing of fertilizer application triggered to meet plant demand considering the impacts of both long-term and short-term climate/weather conditions on soil properties and plant growth.\(^{65}\) CMAQ simulates the transport, full chemistry, and wet and dry deposition of NH\(_3\) and relevant species in the atmosphere.\(^{66}\) BIDI simulates the air-surface bidirectional fluxes of NH\(_3\).\(^{67}\) Integrating EPIC and CMAQ, FEST-C enables the dynamic coupling between agriculture and atmosphere at large scales.\(^{68,69}\)

Downscaled meteorological fields for the periods 2008–2012, 2048–2052, and 2096–2100 under RCP4.5 and RCP8.5 are used to drive the integrated model. The meteorological fields are derived from the bias-corrected outputs of the National Center for Atmospheric Research’s CSM1.\(^{36}\) We use the Weather Research and Forecasting model version 3.8.1\(^{70}\) with spectral nudging to downscale the global climate projections into the grid containing 120 x 156 horizontal grid cells at the 36-km spatial resolution over the study domain. The downsampling procedure was detailed in a previous study.\(^{36}\) We use the US EPA’s 2011 National Emission Inventory\(^{69}\) as the baseline emission inventory. To isolate the impact of climate change on \(V_{\text{NH3,AG}}\), emission sources other than \(V_{\text{NH3,AG}}\) are kept constant as in the baseline emission inventory across the study periods and climate scenarios. We conduct six 5-year FEST-C_EPIC-CMAQ_BIDI simulations, corresponding to the three periods under the two climate scenarios. We also conduct two real-world simulations and several sensitivity tests to facilitate our analyses. Detailed information about the model setting and the simulation design can be found in Supplemental Experimental Procedures.

**Reduced-Form Models**

We developed RFMs to predict \(V_{\text{NH3}}\) using the outputs of the six 5-year FEST-C_EPIC-CMAQ_BIDI simulations as the training database. These RFMs were used to decompose the spatial variations in \(V_{\text{NH3}}\) into variations in LC and climate factors and to conduct multiple \(V_{\text{NH3}}\) projections using multi-model climate projections. We used the 4 km-resolution meteorological fields provided by the Multivariate Adaptive Constructed Analogs Datasets to drive the RFMs.\(^{46}\) Eighteen climate projections under two climate scenarios (RCP4.5 and RCP8.5) were considered in the multi-projection analysis. Detailed information on the model configuration and evaluation, RFM development and parameterization, uncertainty analysis, and the evaluation of the potential impacts of LC change and CO\(_2\) elevation on \(V_{\text{NH3}}\) can be found in Supplemental Experimental Procedures, Tables S8 and S9.

**Mitigation Measures**

We chose the four most commonly used mitigation measures and evaluated their individual and combined efficacy in reducing \(V_{\text{NH3,AG}}\) over the CONUS and resulted benefit impacts on crop yield, atmospheric burden of NH\(_3\)/NH\(_4\)\(_{\text{+}}\), and N deposition. The four mitigation measures are RU, UIR, UIN, and DP.\(^{14,48}\) The \(V_{\text{NH3,AG}}\) reduction rates of these mitigation measures were directly obtained from a literature review,\(^{14}\) which, based on a meta-analysis of 824 \(V_{\text{NH3,AG}}\) measurements, reported reduction rates of 74.5% for RU, 34.5% for UIR, 53.7% for UIN, and 54.7% for DP. We assumed that anhydrous ammonia and nitrogen solution were already deeply placed or injected,\(^{70}\) and therefore they were not considered when evaluating the mitigation potential of DP. We applied the reduction rates of the first three measures to cropland fertilized with urea only and the fourth to all cropland, and evaluated their efficacy in reducing \(V_{\text{NH3,AG}}\) separately by region. We chose one of the first three measures accompanying the fourth to evaluate the combined efficacy of two measures. We calculated the \(V_{\text{NH3,AG}}\) reduction associated with the most effective strategy, which adopts a combination of RU and DP with adaptive management practices.

**Impact Assessments**

Focusing on RCP8.5, we evaluated the impacts of climate change-induced \(V_{\text{NH3,AG}}\) increase and mitigation-induced \(V_{\text{NH3,AG}}\) reduction on crop yield, atmospheric burden of NH\(_3\)/NH\(_4\)\(_{\text{+}}\) over the CONUS domain, and N deposition to sensitive ecosystems. The crop yield loss due to the \(V_{\text{NH3,AG}}\) increase and the crop yield gain due to the \(V_{\text{NH3,AG}}\) mitigation were evaluated based on the hyperbolic relationship between yield and the effective N input following previous studies.\(^{74-76}\) The changes in the atmospheric burden of NH\(_3\)/NH\(_4\)\(_{\text{+}}\) and the N deposition were modeled using CMAQ. We used the protected biodiversity areas (GAP Status Code 1 and 2, areas permanently protected for the protection of biodiversity) defined by the Protected Areas Database of the United States as the sensitive areas of interest (Figure S14).\(^{71}\) Details can be found in Supplemental Experimental Procedures.

**SUPPLEMENTAL INFORMATION**

Supplemental Information can be found online at https://doi.org/10.1016/j.oneear.2020.06.015.

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**AUTHOR CONTRIBUTIONS**

H.S. conceived the idea. H.S. designed the study. A.G.R., Y.H., L.R., and F.Z. provided suggestions on the study design. H.S. and Y.C. conducted the model simulations under the guidance of Y.H., L.R., and J.E.P. H.S. and Y.C. analyzed the results. H.S. wrote the manuscript with input from all authors.
DECLARATION OF INTERESTS
The authors declare no competing interests.

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