

# Climate change impacts of US reactive nitrogen

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**Fossil fuel combustion and fertilizer application in the United States have substantially altered the nitrogen cycle, with serious effects on climate change. The climate effects can be short-lived, by impacting the chemistry of the atmosphere, or long-lived, by altering ecosystem greenhouse gas fluxes. Here we develop a coherent framework for assessing the climate change impacts of US reactive nitrogen emissions, including oxides of nitrogen, ammonia, and nitrous oxide (N<sub>2</sub>O). We use the global temperature potential (GTP), calculated at 20 and 100 y, in units of CO<sub>2</sub> equivalents (CO<sub>2</sub>e), as a common metric. The largest cooling effects are due to combustion sources of oxides of nitrogen altering tropospheric ozone and methane concentrations and enhancing carbon sequestration in forests. The combined cooling effects are estimated at –290 to –510 Tg CO<sub>2</sub>e on a GTP<sub>20</sub> basis. However, these effects are largely short-lived. On a GTP<sub>100</sub> basis, combustion contributes just –16 to –95 Tg CO<sub>2</sub>e. Agriculture contributes to warming on both the 20-y and 100-y timescales, primarily through N<sub>2</sub>O emissions from soils. Under current conditions, these warming and cooling effects partially offset each other. However, recent trends show decreasing emissions from combustion sources. To prevent warming from US reactive nitrogen, reductions in agricultural N<sub>2</sub>O emissions are needed. Substantial progress toward this goal is possible using current technology. Without such actions, even greater CO<sub>2</sub> emission reductions will be required to avoid dangerous climate change.**

Combustion, fertilizer use, and biological nitrogen fixation transform inert N<sub>2</sub> into reactive nitrogen—forms of N that are chemically, biologically, or radiatively active (1). Reactive nitrogen includes oxides of nitrogen (NO<sub>x</sub>), ammonia (NH<sub>3</sub>), and nitrous oxide (N<sub>2</sub>O). NO<sub>x</sub> is largely from combustion, whereas NH<sub>3</sub> and N<sub>2</sub>O are largely from agriculture. These compounds can impact the climate in a myriad of interconnected ways. NH<sub>3</sub> and NO<sub>x</sub> contribute to climate change indirectly. They alter the production and loss of climate forcers, atmospheric constituents that perturb the Earth's energy balance by trapping heat (greenhouse gases) or scattering incoming solar energy (aerosols). NO<sub>x</sub> impacts greenhouse gases by (i) increasing the formation of ozone, contributing to warming, and (ii) increasing the removal of methane (CH<sub>4</sub>), contributing to cooling. Both NO<sub>x</sub> and NH<sub>3</sub> can enhance light-scattering aerosols. When deposited out of the atmosphere into ecosystems, reactive N can stimulate plant growth and alter the uptake of greenhouse gases. N<sub>2</sub>O has a direct effect on climate change; it is a powerful greenhouse gas. These climate change impacts are summarized in Table 1.

Previous studies have examined a subset of these impacts on atmospheric climate forcers (2–4) and greenhouse gas fluxes (5–9). A few studies have assessed the combined impacts on a global (10) and European (11) scale. However, prior efforts have not specifically assessed the impacts of US reactive nitrogen emissions on climate change. Because the United States has a unique mix of emission sources, photochemical conditions, and impacted ecosystems, a US assessment is needed to fully understand the consequences of agroindustrial policy decisions. To address this need, we seek to answer two questions: (i) What are the climate change impacts of US reactive nitrogen emissions, and (ii) what are the relative contributions from agriculture and combustion sources?

Credibly answering these questions, however, requires a framework that can intercompare both atmospheric climate forcers and greenhouse gas uptake. The first part of this framework is to

develop a common metric. A key challenge is that the relevant climate forcers have different radiative forcing intensities and atmospheric lifetimes. For example, aerosols can be powerful cooling forcers but only remain in the atmosphere for a few weeks, whereas N<sub>2</sub>O can persist in the atmosphere for more than 100 y (12). We propose using the “global temperature potential” (GTP<sub>t</sub>) for intercomparing the relative impact of climate forcers over varying timescales. The GTP<sub>t</sub> is the global average temperature change, at time *t*, due to a pulse of emissions (13, 14), relative to a pulse of CO<sub>2</sub> emissions. For example, the GTP<sub>20</sub> for CH<sub>4</sub> is 57 (15), which means a pulse of CH<sub>4</sub> will yield a 57 times greater global temperature increase after 20 y compared with the same mass of CO<sub>2</sub>. In contrast, the GTP<sub>100</sub> for CH<sub>4</sub> is 3.9. Because CO<sub>2</sub> remains in the atmosphere much longer than CH<sub>4</sub>, the GTP<sub>100</sub> for CH<sub>4</sub> is much lower (15). The GTP<sub>t</sub> can be defined on a 20-y timescale to identify emission reductions that reduce the rate of warming in the coming decades, or on a 100-y timescale, to identify emissions reductions that reduce the overall magnitude of climate change. Accordingly, we use both the GTP<sub>20</sub> and GTP<sub>100</sub> metrics to convert the climate change impacts of each of the processes in Table 1 to common units of equivalent Tg CO<sub>2</sub> (CO<sub>2</sub>e).

In addition to a common metric, a consistent treatment of uncertainty is necessary. For each climate change impact of reactive N, the science is evolving and there is a diversity of empirical and theoretical results reported in the literature. We acknowledge that there are real uncertainties. The second part of our framework is to bound these uncertainties in a consistent way. For each variable in our calculation, we survey the literature to assess the range of credible values. These uncertainty ranges are propagated through the calculation using Monte Carlo simulation. This analysis reports the overall uncertainty as the 90th percentile of the resulting probability distribution.

Consequently, our approach is both to reconcile the varied climate forcers using the GTP<sub>t</sub> metric and to propagate the uncertainty through the calculations. The first step is to calculate the US reactive nitrogen emissions from combustion and agricultural sources. For each process summarized in Table 1, we quantify the production or loss of climate forcers due to US reactive nitrogen. For climate change impacts R1–R4, we survey the literature and report the range of values that reflect the state of the science. For R5 and R6, we use nitrogen deposition simulations coupled with US-specific empirical studies of the change in greenhouse gas flux per unit N deposition. Next, we use the GTP<sub>t</sub> to convert to CO<sub>2</sub>e. For each step in this process, we propagate the uncertainty through the calculation. Finally, we report the median and 90th percentile for the contribution of combustion and agricultural emissions in CO<sub>2</sub>e on a 20-y and

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**Table 1. Impacts of reactive nitrogen on climate change included in this study**

	Process altered by reactive N	Climate forcer	Description
R1	N <sub>2</sub> O	N <sub>2</sub> O	N <sub>2</sub> O is a potent greenhouse gas emitted directly from combustion and agricultural soils. Anthropogenic N from deposition, infiltration, and runoff can cause indirect emissions from natural soils and surface waters.
R2	NO <sub>x</sub> → ozone and CH <sub>4</sub>	Ozone, CH <sub>4</sub>	NO <sub>x</sub> perturbs the chemical production and destruction of the greenhouse gases ozone and CH <sub>4</sub> .
R3	NO <sub>x</sub> → aerosol	Nitrate, ammonium, and sulfate aerosol	NO <sub>x</sub> can enhance the formation of light-scattering aerosols.
R4	NH <sub>3</sub> → aerosol	Nitrate, ammonium, and sulfate aerosol	NH <sub>3</sub> can enhance the formation of light-scattering aerosols.
R5	N deposition → CO <sub>2</sub> , CH <sub>4</sub> flux	CO <sub>2</sub> , CH <sub>4</sub>	As a plant nutrient, N deposition in natural systems enhances the storage of CO <sub>2</sub> and can perturb the uptake and emission of CH <sub>4</sub> .
R6	N fertilizer → CO <sub>2</sub> , CH <sub>4</sub> flux	CO <sub>2</sub> , CH <sub>4</sub>	On croplands, nitrogen from fertilizer and manure may enhance the storage of CO <sub>2</sub> and can perturb the uptake and emission of CH <sub>4</sub> .
R7	NO <sub>x</sub> → ozone → CO <sub>2</sub> uptake	CO <sub>2</sub>	NO <sub>x</sub> enhances surface ozone, which can in turn damage plant foliage and decrease carbon storage.

100-y basis. These calculations are described in detail in *SI Text*; the results are summarized below.

## Results and Discussion

**N<sub>2</sub>O: A Potent Greenhouse Gas.** N<sub>2</sub>O is itself a greenhouse gas, with a GTP<sub>100</sub> of 290 (13). Most N<sub>2</sub>O is directly emitted during microbial processes of nitrification and denitrification in agricultural soils and manure (12, 16), whereas combustion contributes only a minor amount (17). Indirect emissions of N<sub>2</sub>O arise from N that is first introduced onto agricultural fields, but N<sub>2</sub>O emissions occur elsewhere. An example is enhanced runoff of soluble forms of N from agricultural fields into rivers and groundwater that contribute to microbial N<sub>2</sub>O production in downstream aquatic and terrestrial ecosystems (18). Reactive nitrogen in the atmosphere can deposit in soils and also cause indirect emissions of N<sub>2</sub>O, but our calculations suggest this contribution is minor. We calculate N<sub>2</sub>O emissions and uncertainty using the Greenhouse Gas Emission Inventory (19), which reports 0.38, 0.090, and 0.099 Tg N for direct agricultural, combustion, and indirect N<sub>2</sub>O emissions, respectively. Converting to CO<sub>2</sub>e on a 20-y basis, agriculture contributes 180–380 Tg CO<sub>2</sub>e and combustion sources contribute 22–48 Tg CO<sub>2</sub>e. These values are lower on a 100-y basis: 160–350 and 20–45 Tg CO<sub>2</sub>e for agriculture and combustion, respectively. Whereas N<sub>2</sub>O directly affects the Earth's energy balance, NO<sub>x</sub> and NH<sub>3</sub> have a myriad of indirect effects that are detailed in the next sections.

**US Anthropogenic Emissions of NH<sub>3</sub> and NO<sub>x</sub>.** US anthropogenic NO<sub>x</sub> emissions are largely from combustion sources, whereas NH<sub>3</sub> is predominantly from agriculture (20). *SI Text* lists the emissions of each species by emission source. Combustion NO<sub>x</sub> emissions are split nearly equally between stationary sources (such as power plants), on-road vehicles, and off-road mobile sources (such as construction equipment). Agriculture contributes a minor amount to NO<sub>x</sub> emissions, largely from NO volatilization from fertilized fields. In contrast to NO<sub>x</sub>, NH<sub>3</sub> is largely emitted from fertilized fields and livestock manure. Combustion of fossil fuels contributes a minor amount. Wildfire, natural soils, and lightning are also sources of reactive nitrogen, but because these emissions are not man-made, they are not included in this analysis.

**Atmospheric Radiative Effects from Oxidants, Ozone, and Aerosols.** After NO<sub>x</sub> and NH<sub>3</sub> are emitted, they are transformed by chemical and physical processes, leading to numerous climate effects. NO<sub>x</sub> rapidly initiates photochemistry and leads to the formation of oxidants, namely the hydroxyl radical and ozone. This has important

implications for the Earth's energy balance. NO<sub>x</sub> increases production of ozone, the third most powerful greenhouse gas (21). On the other hand, NO<sub>x</sub> can also contribute to cooling by increasing hydroxyl radical concentrations, which remove CH<sub>4</sub> (2) from the atmosphere. Furthermore, the ozone concentration in the upper troposphere is enhanced by CH<sub>4</sub>, so NO<sub>x</sub> can also reduce ozone production over longer timescales (22).

These oxidants also play an important role in the formation of sulfate and organic aerosols, which scatter incoming solar radiation and contribute to cooling. NO<sub>x</sub> can be oxidized to form nitric acid, which in the presence of NH<sub>3</sub> forms ammonium nitrate aerosol (23). NH<sub>3</sub> can further enhance the scattering of incoming solar radiation, by altering the physical properties of aerosols (24) and clouds (25).

Prior studies have used models to estimate the radiative effects of NO<sub>x</sub> or NH<sub>3</sub> on aerosols, ozone, or CH<sub>4</sub> (e.g., 3, 4, 10, 25–28). To quantify these radiative effects per unit of emissions in a common framework, we have summarized from the literature or derived the GTP<sub>t</sub> for NO<sub>x</sub> and NH<sub>3</sub>. Because there are differences across models and few observational constraints (29), we quantify the uncertainty based on the range of reported values. The method and ranges are described in *SI Text*. The climate change impacts are calculated as the product of the emissions and the GTP<sub>t</sub>, accounting for differences between shipping, aircraft, and surface emissions (28).

The choice of metric has important consequences for the interpretation of the result. For example, a recent assessment of European reactive nitrogen and climate change (11) used instantaneous radiative forcing as a metric and found larger impacts due to aerosols compared with ozone and CH<sub>4</sub>. We find, on a 20-y basis, that the impact of NO<sub>x</sub> on ozone and CH<sub>4</sub> is –270 (–180 to –380) Tg CO<sub>2</sub>e and the aerosol effects are much less, –29 Tg CO<sub>2</sub>e and –7.3 Tg CO<sub>2</sub>e for NO<sub>x</sub> and NH<sub>3</sub>, respectively. On a 100-y basis, aerosols, ozone, and CH<sub>4</sub> are negligible, indicating that as the time horizon of the analysis becomes longer, short-lived compounds have less effect.

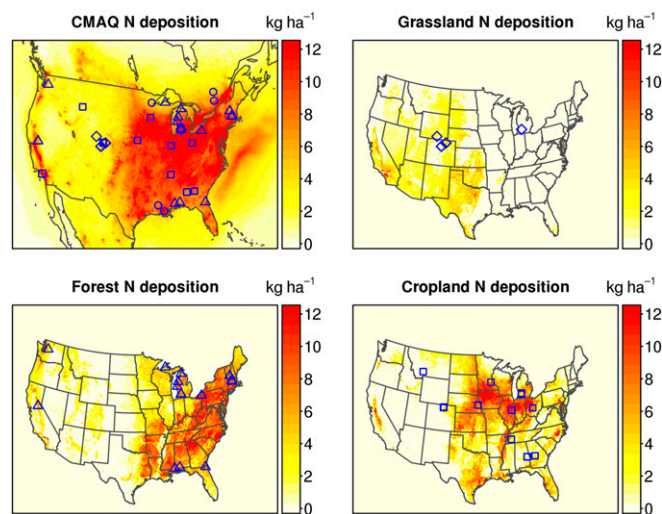
**Impact of Nitrogen Deposition on CO<sub>2</sub> and CH<sub>4</sub> Fluxes.** Ultimately, atmospheric NO<sub>x</sub>, NH<sub>3</sub>, and their chemical products are washed out by precipitation or settle onto surfaces. When deposited to nitrogen-limited ecosystems, reactive nitrogen can serve as a nutrient source for plants. This can cause an increase in productivity in many tree species, removing CO<sub>2</sub> from the atmosphere at a higher rate. However, not all tree species respond positively, and some show increased mortality with increased N deposition (8). Forest decline can occur in older forests and with chronic exposure to high rates of N deposition. Nonetheless, the

aggregate response of growth for US forests in response to current rates of deposition appears to be broadly one of growth enhancement. In addition to affecting plant growth, N additions can affect soil C balance, partly through suppression of rates of decomposition and reduced CO<sub>2</sub> emissions to the atmosphere (30). The combination of enhanced growth and reduced decomposition serves to act as a negative feedback on climate by reducing atmospheric CO<sub>2</sub> concentrations. However, the long-term extent of this effect will depend on the fate and persistence of this additional carbon in soils, forest biomass, or wood products in response to various changes in forest management, disturbance, and environmental conditions. In addition to the CO<sub>2</sub> effects, N additions also alter microbial processes that cause greater emissions and less uptake of CH<sub>4</sub>. The impacts of N deposition differ depending on the land cover type. In the United States, forests have the largest potential for nitrogen-enhanced CO<sub>2</sub> uptake, whereas croplands have the largest potential for enhanced CH<sub>4</sub> emissions (7).

According to our calculations, there are substantial levels of N deposition to US ecosystems that likely have considerable effects on greenhouse fluxes (Fig. 1 and Table 2). The net result is that the uptake of CO<sub>2</sub> is substantially more important than CH<sub>4</sub> emissions, and the net impact is cooling:  $-190$  and  $-110$  Tg CO<sub>2</sub>e on a 20-y and 100-y basis, respectively. The uncertainty is  $-50\%$  to  $+90\%$ , largely due to the range of the flux factor and the fraction of carbon that is returned to the atmosphere.

This estimate of N deposition effects on climate does not include potential biogeophysical responses of forests (31). We have not considered effects of reactive N on latent heat flux, atmospheric water vapor, or the formation of clouds. Nitrogen deposition could also affect forest albedo by favoring hardwood over conifer abundance (8, 31) or by affecting canopy processes (32), although the exact mechanisms of these relationships merit further study (33).

**Fertilized Cropland and CO<sub>2</sub> and CH<sub>4</sub> Fluxes.** For fertilized cropland, it is also important to capture the direct impact of nitrogen from fertilizer and manure application on CO<sub>2</sub> and CH<sub>4</sub> fluxes. We calculate this as the product of fertilizer N application for the United States [ $108.1$  kg N·ha<sup>-1</sup> (34)], the total US cropland acreage, and the cropland flux factors for CH<sub>4</sub> and CO<sub>2</sub> from Liu and Greaver (7). The resulting increase in CO<sub>2</sub> uptake is  $12 \pm 2$  Tg CO<sub>2</sub>-C, and the increase in CH<sub>4</sub> flux is  $470 \pm 240$  Gg CH<sub>4</sub>-C. These values are much larger than the cropland N deposition



**Fig. 1.** Total N deposition: total (Upper Left) and to each ecosystem type. N addition experiments reported in a recent review (7) are marked for crops (squares), forest (triangles), grassland (diamonds), and wetland (circles).

**Table 2.** Impacts of N deposition on CO<sub>2</sub> and CH<sub>4</sub> fluxes

	Area (10 <sup>8</sup> ha)	N deposition (Tg N·y <sup>-1</sup> )	CO <sub>2</sub> (Tg C·y <sup>-1</sup> )	CH <sub>4</sub> (Gg C·y <sup>-1</sup> )
Forest	3.1	2.3	$-100 \pm 40$	$39 \pm 12$
Grassland	2.2	0.81	0	0
Wetland	0.54	0.0044	0	$0.035 \pm 0.018$
Cropland	2.2	1.9	$-1.0 \pm 0.2$	$38 \pm 19$
Sum	8.0	5.0	$-101 \pm 40$	$77 \pm 31$

impacts listed in Table 2, because the direct application of fertilizer N is much larger than N deposition to croplands. The enhancement in CO<sub>2</sub> uptake is largely canceled by CH<sub>4</sub> emissions, and the net impact of fertilizer applied to croplands is  $-8.2$  to  $-25$  Tg CO<sub>2</sub>e on a 20-y basis and  $-37$  to  $-50$  Tg CO<sub>2</sub>e on a 100-y basis.

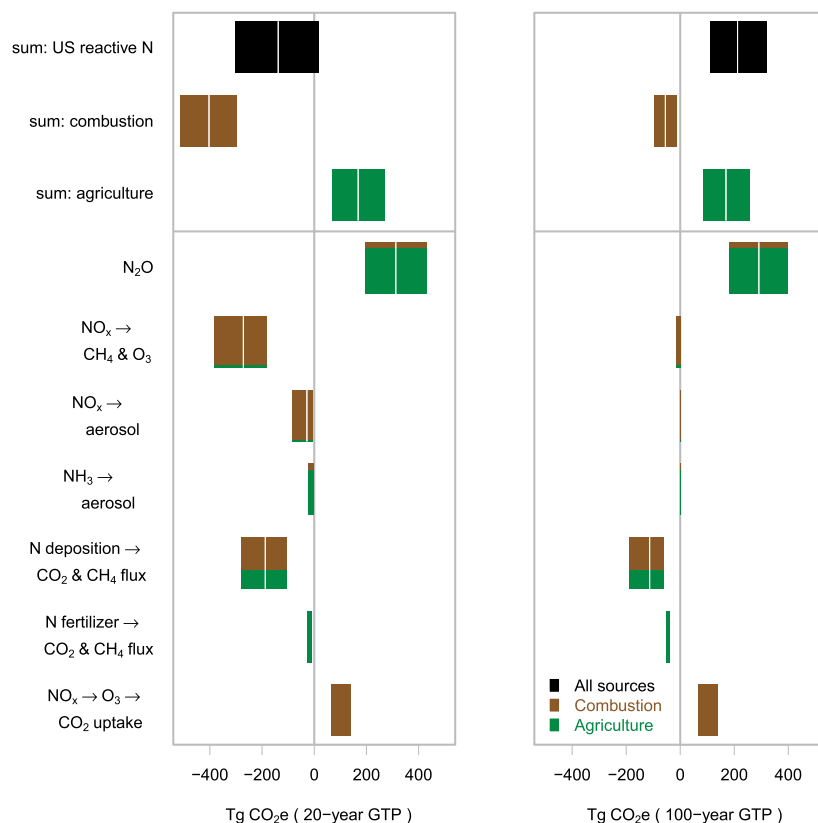
**Surface Ozone and CO<sub>2</sub> Fluxes.** An additional issue is that emissions of NO<sub>x</sub> increase ozone, which can damage plants and decrease carbon uptake (6, 35). Felzer et al. estimate that ozone decreases CO<sub>2</sub> sequestration by  $66$ – $140$  Tg CO<sub>2</sub>·y<sup>-1</sup> (36). This is the only estimate that we are aware of for the entire continental United States. This estimate was calculated for the years 1950–1995 by running the terrestrial ecosystem model, which incorporates spatially explicit ozone exposures, vegetation distribution, and empirical ozone response equations for trees and crops. The magnitude of this estimated loss compared favorably with other modeling estimates at different scales (35, 37). There are many uncertainties, including comparing past ozone exposures to future ozone exposures, unknown variability of response between various species, and future management of crops and forests. However, we believe that this is the most reasonable range of estimates for a change in carbon sequestration due to ozone exposure in the United States.

**Net Climate Change Impact of US Reactive Nitrogen.** To quantify the total reactive nitrogen climate change impacts, Monte Carlo simulation is used to propagate the uncertainties through the calculations described above (details available in *SI Text*). The climate change effects of US reactive nitrogen, in Tg CO<sub>2</sub>e, for each of the seven effects listed in Table 1 are summarized in Fig. 2. The length of the bar denotes the 5th and 95th percentile uncertainty bounds, and the white line denotes the median. The relative contribution from agriculture (green) and combustion (brown) is indicated by the color shading. The largest effects are from NO<sub>x</sub> impacts on ozone and CH<sub>4</sub> radiative forcing, N<sub>2</sub>O emissions, and enhanced CO<sub>2</sub> uptake. Likewise, the uncertainties that contribute the most are (i) the CO<sub>2</sub> uptake factor for US forest systems and the persistence of this uptake over time, (ii) the combined effects of NO<sub>x</sub> on ozone and CH<sub>4</sub> radiative forcing, and (iii) the total direct and indirect emissions of N<sub>2</sub>O.

Overall, the impacts of combustion sources are offset by agricultural sources. The largest effects are from combustion emissions, which are between  $-290$  and  $-510$  Tg CO<sub>2</sub>e on a GTP<sub>20</sub> basis. Much of these effects are short-lived, and the values on a GTP<sub>100</sub> basis are lower ( $-16$  to  $-95$  Tg CO<sub>2</sub>e). Agricultural sources are net warming, as they contribute more to N<sub>2</sub>O and less to N deposition on forests. We calculate a range of  $+74$  to  $+270$  Tg CO<sub>2</sub>e on a GTP<sub>100</sub> basis. For agriculture, the difference between GTP<sub>20</sub> and GTP<sub>100</sub> values is minor. Because of these offsetting effects, the overall climate change effects are slight cooling on a 20-y basis ( $-130$  Tg CO<sub>2</sub>e) and warming on a 100-y basis ( $+210$  Tg CO<sub>2</sub>e).

**Implications for Climate Change Mitigation.** On balance, we find that cooling effects of combustion emissions offset the warming effects of agricultural emissions. This has important implications





**Fig. 2.** Climate change impact of US reactive nitrogen emissions, in Tg CO<sub>2</sub> equivalents, on a 20-y (*Left*) and 100-y (*Right*) global temperature potential basis. The length of the bar denotes the range of uncertainty, and the white line denotes the best estimate. The relative contribution of combustion (brown) and agriculture (green) is denoted by the color shading.

for climate change mitigation. First, the cooling effects are largely from combustion NO<sub>x</sub> emissions, which have decreased considerably over the past decade (20). By decreasing the ratio of NO<sub>x</sub> to CO<sub>2</sub> in combustion emissions, each ton of combusted fuel contributes more to warming. Even more CO<sub>2</sub> reductions will be required to avoid dangerous climate change. Second, N<sub>2</sub>O from agriculture, which contributes the most to warming, is globally increasing (16). The warming components of reactive nitrogen could be mitigated by increasing the nitrogen-use efficiency in agriculture and by managing denitrification in agricultural runoff. Examples include improved matching of fertilizer applications to crop needs, expanded use of winter cover crops and nitrification inhibitors, improved drainage management, improved manure management, constructed wetlands and denitrification bioreactors, and riparian zone vegetation strips (38, 39). Using current technology, it is possible to improve agricultural N efficiency by up to 20–25% (40) and reduce N losses, including N<sub>2</sub>O emissions, by 30–50% (41, 42). Third, the long-term cooling impacts from both combustion and agricultural emissions are largely due to nitrogen enhancement of carbon storage in forests. Since 1990, the carbon storage in US forests has increased and the total forested acreage has grown (43). If forestry management changes such that carbon storage is not increasing, then the 100-y cooling impacts would be reduced by a factor of three.

In summary, to prevent warming from US reactive nitrogen emissions, it is important to reduce emissions of N<sub>2</sub>O from agriculture and to support continued carbon sequestration in forests. Achieving this goal will require continued advances in agricultural efficiency and forestry management. Not achieving

this goal means that even greater CO<sub>2</sub> emission reductions will be required to avoid dangerous climate change.

## Materials and Methods

This section describes our method to estimate the impact of N deposition on CO<sub>2</sub> and CH<sub>4</sub> fluxes. The first step is to calculate the total N deposition to each N-sensitive ecosystem. Because atmospheric NH<sub>3</sub> can rapidly settle onto plant surfaces within 1 km of the source (44) or can form aerosol and be transported hundreds of kilometers (45), simulations with resolutions on the order of 10 km are needed to capture N deposition gradients (46). In addition, ecosystems can be homogeneous over large areas, such as the prairie of the midwestern United States, or can vary substantially across mountainous regions. Capturing the collocation of nitrogen deposition and sensitive ecosystems requires a spatially explicit representation of deposition and land cover.

To capture the spatial variability in N deposition, we use the Community Multiscale Air Quality (CMAQ) model at 12-km horizontal resolution. CMAQ simulates atmospheric transport, chemistry, aerosol physics, and deposition across the continental United States with results that are consistent with observations (47) and prior N deposition assessments (48). We calculate the N deposition to four ecosystem types (forest, cropland, grassland, and wetland) by mapping the CMAQ N deposition to a 1-km characterization of land cover (Fig. 1). The variability in N deposition across ecosystem types is due to differences in location relative to emission sources. Substantial ammonia emissions cause high deposition on midwestern croplands, whereas industrial and urban emissions contribute to the high deposition on eastern forests.

To calculate the change in CO<sub>2</sub> and CH<sub>4</sub> flux, we multiply the N deposition by a flux factor (kg C·kg N<sup>-1</sup>). There are two approaches to estimate these flux factors that are broadly representative of US conditions: (i) fertilizer experiments and (ii) gradient studies. The first approach is to apply a known quantity of N to an ecosystem and measure the change in CO<sub>2</sub> uptake or CH<sub>4</sub> flux. Liu and Greaver (7) have synthesized 68 publications and reported flux factors and uncertainty ranges for forests, grasslands, croplands, and wetlands. Fig. 1 shows the spatial distribution of these studies; note that many locations with high N deposition have not yet been studied using fertilized plot experiments.

With 24 kg CO<sub>2</sub>-C·kg N<sup>-1</sup> reported by Liu and Greaver, forest CO<sub>2</sub> is the most significant flux and therefore merits special attention. Accordingly, we also examine gradient studies, which use observed gradients in N deposition and forest growth across the northeastern United States, as used by Thomas et al. (8). This approach is representative of the actual N deposition, species composition, and observed uptake for a large eastern US region, rather than of a collection of individual sites. Because the Thomas et al. study examined aboveground biomass only, we modify their result to account for belowground biomass, soil carbon, and differences in measured and CMAQ total N deposition. After this adjustment, the Thomas et al. estimate is 65 kg CO<sub>2</sub>-C·kg N<sup>-1</sup> (SI Text). However, ~15% of the N deposition to US forests occurs west of 100° W longitude. These western forests may be less responsive to N deposition compared with eastern forests (49). Accordingly, we treat the Thomas et al. estimate as an upper bound and select an uncertainty range of 24–65 kg CO<sub>2</sub>-C·kg N<sup>-1</sup>. A recent European synthesis

found a best estimate of 41 kg CO<sub>2</sub>-C·kg N<sup>-1</sup> (11), which falls near the middle of our range.

The additional carbon sequestered in forests that is stimulated by N addition is not permanent. This carbon can return to the atmosphere when harvested, transformed into products, or combusted in wildfire (43). We estimate that 40–60% of the annual growth in sequestered carbon is returned to the atmosphere within 20 y, and 60–80% within 100 y (SI Text).

For all other land cover types, we use the N deposition from CMAQ and the flux factor uncertainty ranges for CO<sub>2</sub> and CH<sub>4</sub> from Liu and Greaver (7). The results are summarized in Table 2.

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