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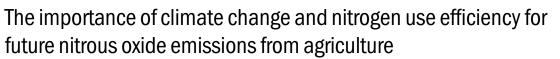
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## Abstract

Nitrous oxide (N<sub>2</sub>O) is an important greenhouse gas and ozone depleting substance. Previous projections of agricultural N<sub>2</sub>O (the dominant anthropogenic source) show emissions changing in tandem, or at a faster rate than changes in nitrogen (N) consumption. However, recent studies suggest that the carbon dioxide (CO<sub>2</sub>) fertilization effect may increase plant N uptake, which could decrease soil N losses and dampen increases in N<sub>2</sub>O. To evaluate this hypothesis at a global scale, we use a process-based land model with a coupled carbon-nitrogen cycle to examine how changes in climatic factors, land-use, and N application rates could affect agricultural N<sub>2</sub>O emissions by 2050. Assuming little improvement in N use efficiency (NUE), the model projects a 24%–31% increase in global agricultural N<sub>2</sub>O emissions by 2040–2050 depending on the climate scenario—a relatively moderate increase compared to the projected increases in N inputs (42%–44%) and previously published emissions projections (38%–75%). This occurs largely because the CO<sub>2</sub> fertilization effect enhances plant N uptake in several regions, which subsequently dampens N<sub>2</sub>O emissions. And yet, improvements in NUE could still deliver important environmental benefits by 2050: equivalent to 10 Pg CO<sub>2</sub> equivalent and 0.6 Tg ozone depletion potential.

## Introduction

A growing body of scientists and policy experts argue that  $N_2O$ , the third most important greenhouse gas (GHG) and the most abundantly emitted ozone depleting substance, requires more focused attention from the international policy community [1–3]. Humanity's ability to produce synthetic nitrogen (N) fertilizer via the Haber-Bosch process has dramatically increased N concentrations in agricultural soils, bolstering the major biogeochemical processes that produce  $N_2O$ : nitrification (the conversion of ammonium (NH<sub>4</sub><sup>+</sup>) to nitrate (NO<sub>3</sub><sup>-</sup>)), and denitrification (the conversion of NO<sub>3</sub><sup>-</sup> to

atmospheric dinitrogen ( $N_2$ )). Supplemented by nonagricultural emission sources such as industry, energy production and transport, atmospheric concentrations of  $N_2O$  have increased from mid-19th century levels of approximately 275 ppb to 328 ppb in 2015 [4]. Agriculture is the dominant source, responsible for two thirds of gross anthropogenic emissions, and the focus of this study [5].

However,  $N_2O$  is not only an important GHG that exacerbates climate change; its emissions are also affected by climate change. Rising temperatures, shifting precipitation patterns, and increasing atmospheric  $CO_2$  concentrations all impact  $N_2O$  emissions in a variety ways that are still being studied [6]. Consequently, this study uses a process-based land model (Princeton-GFDL LM3 N.1, which captures vegetation, hydrological, carbon (C) and N dynamics [7, 8]) to examine both the potential climate and ozone benefits of reducing agricultural N<sub>2</sub>O emissions at a global scale, and the direct and indirect effects of future climate change scenarios on N<sub>2</sub>O emission rates.

The LM3 N.1 model allows us to simulate both the processes that directly control agricultural N2O emissions (such as temperature and soil water content), and the indirect effects such as crop N uptake that change under different climate, N consumption, and atmospheric CO<sub>2</sub> concentration scenarios. Climate change is expected to impact all of these effects. Rising soil temperatures induced by climate change will likely increase the rate of soil organic matter decomposition, releasing more mineral N into the soil, and thereby make more N available for nitrification and denitrification-the major biogeochemical processes that produce N<sub>2</sub>O [9]. Meanwhile, nitrification and denitrification are a function of oxygen levels in the soil, which are inversely proportional to soil water content. Soil water content is a function of soil topography, texture, and the balance between precipitation and evapotranspiration [10]. Therefore, the effect of shifting precipitation patterns and rising temperatures could impact nitrification and denitrification rates across the globe.

In addition to these direct effects, there is evidence to suggest that increasing atmospheric CO<sub>2</sub> concentrations have an indirect effect on N2O emissions [6]. The  $CO_2$  fertilization effect has been shown to decrease N<sub>2</sub>O emissions by increasing plant N and C uptake, which can reduce N losses, including N2O [11]. The magnitude and persistence of the CO<sub>2</sub> fertilization effect depends on the availability of mineral N and the ability of plants to acquire it. Hence, previous modeling and empirical studies have focused on the effects of N limitation on CO2 fertilization in natural ecosystems and pasturelands. These land systems are often characterized by limited N inputs, and thus N limitation is believed to be the main limiting factor on their C storage capacity [9, 12-15]. N limitation is an important issue, particularly for natural ecosystems, where the main sources of N inputs (biological N fixation and N deposition) may not be able to sustain increased plant demand for N stimulated by CO2 fertilization [6]. However, in intensive agricultural systems (the dominant source of anthropogenic N<sub>2</sub>O emissions), N limitation is usually not a concern; there is, in fact, an excess supply of N, largely from synthetic fertilizers and manure [16]. Therefore, without N limitation cropping systems in certain regions could be more able to take advantage of the CO<sub>2</sub> fertilization effect and potentially increase their yields by up to 20% [17], improving N use efficiency (NUE) and water use efficiency [18] albeit with a possible deterioration in key micronutrient levels [19]. If certain



cropping systems can increase yields as a result of the  $CO_2$  fertilization effect, this could then reduce excess N in soils, thereby potentially reducing agricultural N losses and N<sub>2</sub>O emissions. This paper evaluates this hypothesis for the first time at a global scale under different climate scenarios. It builds on previous studies that have analyzed future N<sub>2</sub>O emissions using other process-based land models [20] by explicitly disaggregating the impacts of climate,  $CO_2$  concentrations and N inputs on agricultural biomass growth and agricultural N<sub>2</sub>O emissions.

The other focus of this study is to better understand the mitigation potential of agricultural N2O emissions via improvements in global NUE. Previously published scenarios of global NUE improvements and their impacts on agricultural N2O emissions in the 21st century have relied primarily on modified N2O emission factors and projections of global N fertilizer demand based on assumptions that are often opaque [3, 16, 20-22]. While these projections are useful in generating order of magnitude estimates of future N<sub>2</sub>O emissions, they do not simulate the effects of farmer N management practices that ultimately determine NUE. In the only other study to have evaluated future NUE scenarios using a process-based global land model [20], the assumptions underpinning the ambitious NUE improvement scenario are not clear. By contrast, this study builds on previous work by representing the effects of farmer N management practices that underpin NUE improvements using the LM3-N.1 model. While this is by no means the first process-based land model with a coupled C-N cycle to simulate the effects of farmer N management practices on N cycle dynamics [23], we believe it is the first to do so with the goal of projecting future agricultural N2O emissions under different climate and NUE scenarios. This could lead to a more complete assessment of how improvements in global NUE will affect N2O emissions, and thus the climate and stratospheric ozone benefits that could follow.

## Princeton GFDL-LM3 N.1 model

To conduct our analysis we introduce a new  $N_2O$  flux function based on Xu-Ri and Prentice into the LM3-N.1 model [24]. This model captures vegetation, C, N and water dynamics and was developed by Princeton University and the National Oceanic and Atmospheric Administration's (NOAA) GFDL [7]. The N submodel couples each C pool in vegetation and the soil with a N compartment [8]. The model allows for N limitation on plant growth and CO<sub>2</sub> assimilation, includes processes such as N fixation, leaching (both mineral and organic), N volatilization from fire, N plant uptake, mineralization, nitrification, denitrification and hydrological and gaseous N export. Global to site-scale evaluations of this model have been presented in previous work [7, 8]. The general



denitrification function is adapted from Heinen [25] and was first introduced into LM3-N.1 and evaluated in Lee et al [26]:

$$D = k_{\rm d} \times W_{\rm d} \times T_{\rm d} \times C_{\rm NO_3}^{-},$$

 $k_{\rm d}$  = denitrification rate coefficient (yr<sup>-1</sup>)

 $W_{\rm d} =$ soil water content function (dimensionless)

 $T_{\rm d} =$ soil temperature function (dimensionless)

 $C_{\text{NO}_3^-}$  = soil NO<sub>3</sub><sup>-</sup> concentration (kg  $NO_3^{-}-Nm^{-2}).$ 

The focus on soil temperature, water content and NO<sub>3</sub><sup>-</sup> concentrations emulates the approach of several other global denitrification models [27, 28]. The effect of labile C availability is reflected in the denitrification rate coefficient [29-31]. Meanwhile, the N<sub>2</sub>O flux function is adapted from Xu-Ri and Prentice [24]:

 $N_2O = (F_{denit} \times T_{N_2O} \times D) + (F_{nit} \times Nit),$ 

 $F_{\text{denit}} = Proportion$  of denitrification flux which is converted to N<sub>2</sub>O (0.02) independent of the temperature modifier  $(T_{N2O})$ 

 $D = \text{Denitrification rate } (\text{kg N m}^{-2} \text{ yr}^{-1})$   $T_{N_2O} = e^{\left[308.56 \times \left(\frac{1}{68.02} - \frac{1}{T + 46.02}\right)\right]} (T \text{ is in degrees})$ Celsius)

 $F_{\rm nit}$  = Proportion of nitrification flux which is converted to  $N_2O$  (fixed at 0.001)

Nit = Nitrification rate (kg N  $m^{-2} yr^{-1}$ ).

See supplementary information section 1 for more detail on each individual parameter.

To evaluate these functions, we simulate agricultural N2O emissions from 1990 to 2000 and find that our emission estimates are largely consistent with other model simulations and bottom-up inventories (see supplementary information section 1 for a model evaluation and sensitivity analysis). The denitrification function was also previously evaluated at the watershed scale, comparing model output with data from 16 monitoring stations in the Susquehanna River Basin. It was shown to capture fluxes well at multiple locations within the basin with different climate regimes and land-use types [26].

We define agricultural N2O emissions as direct and indirect N2O emissions (the latter from N leached into waterways and N deposition) from cropland and pasture, excluding manure management. Manure management is not currently simulated as emissions from this sector occur during the collection, handling, storage, and treatment of manure, practices that are not yet explicitly integrated into the model. The explicit simulation of indirect N2O emissions is currently in development for LM3-N.1 and partially implemented in this study. While indirect N<sub>2</sub>O emissions from N deposition are currently simulated (as the N deposited on land becomes part of the soil NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> pools which impact nitrification and denitrification fluxes), indirect emissions from N leaching are not. Consequently, for this study we follow a similar approach to other recent process-based land model simulations [20] and apply the IPCC emission factor for indirect

N<sub>2</sub>O emissions from N leaching (0.75% of leached mineral N is emitted as  $N_2O$  [32]). The dynamics of fertilizer and manure N application to cropland and pasture used in this study are described in supplementary information section 2.

# **Climate and NUE scenarios**

For model simulations from 1990-2000 to 2040-2050 we use a global fertilizer and manure dataset from Bouwman et al (described in detail in supplementary information section 2) under two climate and landuse change scenarios [33]. These scenarios are Representation Concentration Pathway (RCP) 2.6, representing a highly ambitious climate mitigation scenario that maintains the possibility of reaching the 2 °C target, and RCP 8.5, a high emission/business-asusual (BAU) scenario [34, 35]. The latter approximates the path the world is currently following.

In addition to simulating BAU agricultural N<sub>2</sub>O emissions, a central goal of this study is to better understand the potential environmental benefits of improving global NUE. Here we evaluate the impacts of a moderate (20%) and an ambitious (50%) increase in NUE by country by 2050 under both climate scenarios. These targets are based on results from previous studies of NUE improvements in cereal production, using a combination of plant breeding, proper technology and incentives [36-39]. NUE is measured in this study using partial factor productivity  $(\text{kg C kg N}^{-1})$ , one of the most widely used and easily measured NUE metrics, which measures the ratio of crop yield per unit of applied N [40]. All agricultural biomass in the model is represented as either a C3 or C4 grass type (determined as a function of climate, with C3 grasses more prevalent in temperate zones and C4 grasses more prevalent in tropical zones [7]), which precludes crop-specific analysis, a drawback shared by several similar models [6, 20] and an active area of model development. Net primary productivity on agricultural land is currently considered proportional to crop yields in the model, and is calculated as a function of gross primary productivity, growth and maintenance respiration, and N limitation [8]. All aboveground C and N in leaves and labile stores on cropland is annually harvested and released back to the atmosphere the following year. In pasture, 25% of leaf C and N is annually removed to represent livestock grazing and partitioned between a livestock respiration fraction (0.9) and a grazing residue fraction (0.1)that is returned to the soil pools. Given this relatively simplistic representation of agricultural activities currently in LM3 N.1, it is not possible to explicitly simulate specific agricultural management practices such as improving crop cultivars or using enhanced efficiency fertilizers. Instead, we evaluate three more basic approaches for achieving global NUE improvements of 20% and 50%, which represent the effects of a suite

of improved N management strategies that better synchronize soil N supply with crop N demand:

- Reduced N inputs: fertilizer and manure application rates are reduced to achieve a 20% and 50% increase in NUE on a country-by-country basis by 2050.
- (2) Increased N uptake: plant N uptake is increased by 20% and 50% by 2050.
- (3) Combined: a combination of the reduced N input and increased N uptake strategies.

See supplementary information section 4 for more detail on these approaches, including a description of the most commonly used NUE-improving best management practices and technologies. It should also be noted that we are focusing on NUE at the field-level (an approach taken by similar studies [20, 33]). However, in reality N can continue to cascade through the environment, from field to feedlot and ultimately to the consumer's plate, with N losses throughout. New metrics are emerging to estimate NUE across the entire supply chain in which N use occurs, notably 'full chain NUE' [16]. While these efforts are important, the focus of this study is on in-field NUE given the capacity of our model.

#### **Experimental set-up**

The model was spun up for 1000 years with preindustrial climate forcing (using atmospheric CO2 concentrations of 280 ppm) starting from bare ground without C-N coupling (i.e. a plant's capacity to photosynthesize was not limited by N concentrations in the soil) in order to accelerate vegetation growth. C-N coupling was then initiated and the spin-up continued for an additional 400 years to reach steady state. The model runs analyzed in this paper were begun in 1860 using historical CO<sub>2</sub> concentrations [41], high frequency forcings obtained from GFDL-ESM2 model output (temperature, wind, short- and long-wave downward radiation, specific humidity, and precipitation [42]) and the Bouwman et al fertilizer and manure data beginning in 1900. Depending on the run, forcings from RCP 2.6 or RCP 8.5 using the GFDL-ESM2 simulations were used from 2006 onwards, and modified Bouwman et al fertilizer and manure data were used to represent the various NUE scenarios (see supplementary materials section 1-3). Land use was prescribed from Hurtt *et al* [43].

# **Results and discussion**

# Agricultural N<sub>2</sub>O emissions in 2040–2050 assuming little improvement in NUE

Under the baseline fertilizer and manure consumption scenario [33], which assumes little improvement in

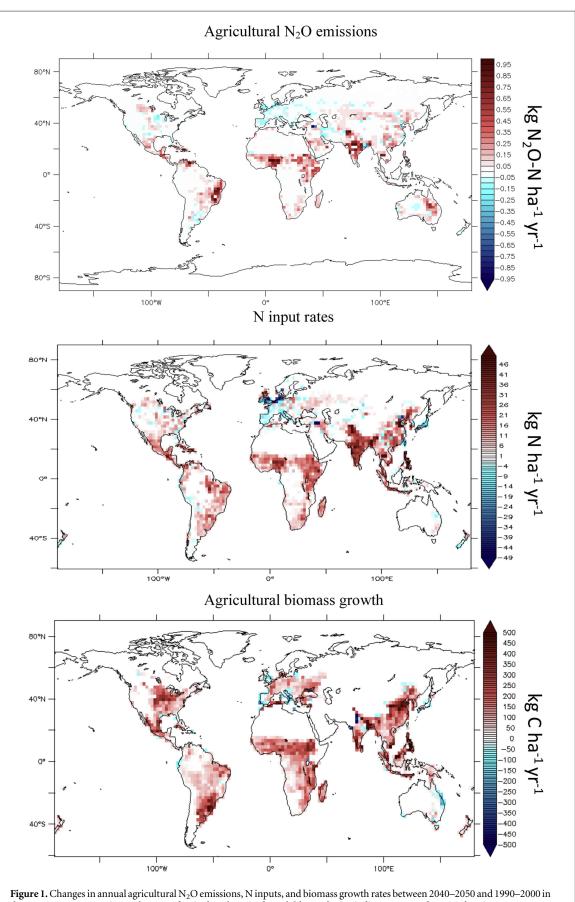


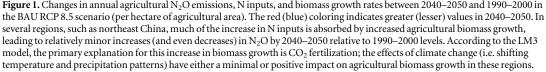
global NUE by 2050 (see supplementary information section 2), our model projects a 24%–31% increase in agricultural N<sub>2</sub>O emissions: from 2.9 Tg N<sub>2</sub>O-N yr<sup>-1</sup> in 1990–2000 to 3.6 and 3.8 Tg N<sub>2</sub>O-N yr<sup>-1</sup> by 2040–2050 for the RCP 2.6 and RCP 8.5 scenarios, respectively. Figure 1 globally maps the increases in emissions from 1990–2000 to 2040–2050 for the RCP 8.5 scenario (equivalent maps for RCP 2.6 are included in supplementary information section 5). The most significant increases in N<sub>2</sub>O emissions are concentrated in regions with rapidly increasing N input rates, driven by sustained population growth and food production (e.g. India, sub-Saharan Africa) over the first half of the 21st century.

Nevertheless, changes in N input rates only partly explain future emission trends. There are several regions in figure 1 where N inputs are projected to increase considerably, but N2O emissions are not. A likely explanation is that the CO<sub>2</sub> fertilization effect enhances agricultural biomass growth in regions where N is not limiting, which subsequently absorbs a significant portion of the increased N inputs, leaving less excess N to be lost as N2O. In northeast China, for example, N input rates are projected to increase by up to  $50 \text{ kg N} \text{ ha}^{-1} \text{ yr}^{-1}$  by 2040–2050 (a 30% increase relative to 1990–2000 levels) and yet N2O emissions are projected to rise less than 0.3 kg  $N_2$ O-N ha<sup>-1</sup> yr<sup>-1</sup> (a 10% increase). Meanwhile, agricultural biomass is projected to increase by over 0.2 kg C m<sup>-2</sup> yr<sup>-1</sup> (a 50% increase relative to 1990-2000 levels), indicating that much of the projected increase in N input is absorbed by an increase in crop yields. A similar dampening effect of agricultural biomass growth on agricultural N<sub>2</sub>O emissions can be seen in other important agricultural regions such as northeastern and southern India, and southeastern South America.

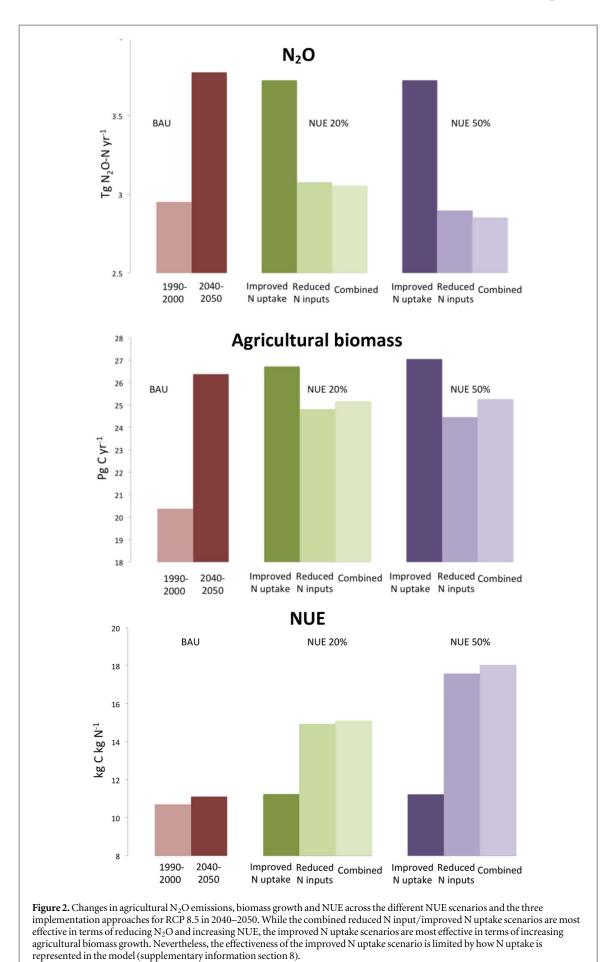
The LM3 model attributes approximately 80% of the globally projected agricultural biomass growth by 2040–2050 to the CO<sub>2</sub> fertilization effect and 24% to increases in N inputs, while changes in soil temperature and precipitation patterns alone lead to a 4% decrease in global agricultural biomass growth (see supplementary information section 8). The dominance of the CO<sub>2</sub> fertilization response over the warming response concurs with empirical experiments that have evaluated these effects [44]. The  $CO_2$ fertilization effect increases agricultural biomass levels by about 20% in RCP 8.5 and 10% in RCP 2.6 by 2040-2050 compared to model runs where atmospheric CO<sub>2</sub> concentrations are kept at 1990–2000 levels, which is within the range of previous estimates [17, 45, 46]. This increase in agricultural biomass growth takes up N that could otherwise be lost as N<sub>2</sub>O. Indeed, without the CO<sub>2</sub> fertilization effect, we project that global agricultural N2O emissions would be 0.5 Tg  $N_2$ O-N yr<sup>-1</sup> higher in 2040–2050, consistent with previously reported values [6]. A similar phenomenon is seen for RCP 2.6 (supplementary information section 5; see also section 6 for an analysis of the differences in





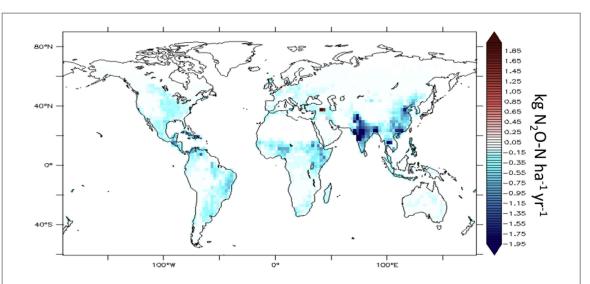


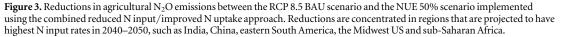












**Table 1.** The cumulative environmental benefits generated from achieving the 20% and 50% improvements in global NUE (integrated over the period 2015–2050) using the 'improved N uptake', 'reduced N input' and 'combined' implementation approaches. These benefits are reported in terms of reductions in N<sub>2</sub>O emissions (Tg N<sub>2</sub>O-N), climate forcing (Pg CO<sub>2</sub> eq.) and stratospheric ozone depletion (Tg ODP). The lower bound of the uncertainty range marks the benefits from achieving the NUE 20% scenario and the upper bound marks the benefits from achieving the NUE 50% scenario. The results represent the range of benefits achievable across both NUE scenarios and the RCP scenarios.

NUE approach	Nitrous oxide (Tg N <sub>2</sub> O-N)	Climate (Pg $CO_2$ eq.)	Ozone (Tg ODP)
Improved N uptake	0.4–2.6	0.2–1.2	0.01-0.07
Reduced N inputs	13.7–17.5	6.4-8.2	0.4-0.5
Combined	14.0–21.2	6.6–9.9	0.4–0.6

N<sub>2</sub>O emissions between RCP 2.6 and RCP 8.5). A common feature of the regions where CO<sub>2</sub> fertilization is most pronounced (e.g. northeast China)-and thus where N<sub>2</sub>O emissions are most dampened—is that the effects of climate change (i.e. shifting temperature and precipitation patterns) have either a minimal or positive impact on agricultural biomass growth (figure S10). Nevertheless, it should be noted that CO<sub>2</sub> fertilization only dampens N2O emissions during the growing season; a significant proportion of annual N2O emissions have been shown to occur during periods when agricultural land is bare (i.e. directly following fertilizer application or off-season during freeze-thaws events and post-harvest [47]), periods that are represented in the LM3 model. However, intra-annual variability is less important for the purposes of this study given our focus on decadal time-scales.

The dampening effect that agricultural biomass growth has on agricultural N<sub>2</sub>O emissions in certain regions suggests that simply applying N<sub>2</sub>O emission factors to projected N input rates might overestimate future emissions. For example, applying the Davidson emission factors (approximately 2% for manure and 2.5% for fertilizer) to the Bouwman *et al* N input projections suggests a 42%–44% increase in agricultural N<sub>2</sub>O emissions across the RCPs by 2040–2050 relative to 1990–2000, which is substantially larger than the 24%-31% increase projected in this study [33, 48]. Our results also contrast with the results published in Stocker et al using the LPX-Bern Earth System Model of intermediate complexity, which projects the relative increase in agricultural N<sub>2</sub>O emissions by 2040-2050 (38%-75%) to be greater than the relative increase in N inputs (20%-60%) which is counter to our findings [20]. While they do not offer an explanation for this dynamic, we believe the differences stem from the fact that the balance in their model between climatic factors that enhance N2O emissions (e.g. rising surface temperatures that can increase soil N mineralization and ultimately denitrification rates) and those that dampen emissions (i.e. the CO<sub>2</sub> fertilization effect) skew towards the former, while the balance in LM3-N.1 skews towards the latter. More research in this area is necessary to better understand which of the climatic drivers of N<sub>2</sub>O emissions can be expected to dominate in the future.

# Impact of global NUE increases on future agricultural N<sub>2</sub>O emissions

Despite the impact of the  $CO_2$  fertilization effect, improving NUE could still lead to significant reductions in agricultural N<sub>2</sub>O emissions, depending on the target set and the implementation strategy used. Figure 2 shows the impact of each NUE improvement and implementation strategy on global N2O emissions, agricultural biomass growth, and NUE for the RCP 8.5 scenario. RCP 2.6 has similar results (supplementary information sections 5 and 8). The largest N2O reductions are achieved with the scenario that combines N input reductions and N uptake increases (reducing emissions to 2.8 Tg  $N_2$ O-N yr<sup>-1</sup> in 2040-2050, a 28% reduction in emissions compared to the baseline scenario). In this scenario, agricultural biomass growth is slightly reduced relative to the baseline scenario (25.3 Pg C yr<sup>-1</sup> versus 26.4 Pg C yr<sup>-1</sup>). However this is still a 24% increase in agricultural biomass growth from 1990-2000 levels, with 24% less N inputs (see figure 3 for spatial distribution of N2O reductions). Reductions are concentrated in regions that are projected to have the highest N input rates in 2040–2050.

### Stratospheric ozone and climate benefits

The different NUE approaches produce a range of environmental benefits over 2015-2050 (table 1). The highest mitigation potential comes from a combination of the reduced N inputs/improved N uptake approach and switching from a RCP 8.5 to a RCP 2.6 forcing scenario. In this case, emissions in 2040-2050 are 2.7 Tg  $N_2$ O-N yr<sup>-1</sup> (instead of 3.9 Tg  $N_2$ O-N yr<sup>-1</sup>, a 31% reduction), with cumulative savings of 21.2 Tg  $N_2O-N$  over 2015–2050, equivalent to 9.9 Pg  $CO_2$  eq. and 0.6 Tg ODP. These estimates do not take into account the other indirect environmental and health benefits that could result from reducing N pollution, namely improved water quality from less N leaching and a reduction in air pollution due to decreases in NH<sub>3</sub> and NO<sub>x</sub> volatilization. A more holistic assessment of the entire N cascade is an area of active model development. In addition, a better representation of agricultural crops and practices is also being developed for the LM3-N.1 model. This will allow for a more nuanced and realistic evaluation of NUE scenarios, with an ability to analyze the effect of different fertilizer best management practices and technologies in different climates and growing cultures.

With growing calls for an effective international framework to manage N pollution, policy-makers require scientific information on a range of issues to better evaluate policy options. This study has attempted to contribute to this need for policy-relevant science by demonstrating the importance of climatic feedbacks in determining future agricultural N<sub>2</sub>O emissions, and that increasing NUE can deliver important global environmental benefits. This is an important step if humanity hopes to return within a safe planetary boundary for nitrogen.

#### Author contributions

DRK, XZ, DLM, SM and ES designed the study, analyzed the data, and wrote the paper. DRK, SM and ES designed the model experiments. DRK and XZ



designed and performed the model evaluation and sensitivity analysis. DRK wrote the new code and ran the model.

#### **Competing interests**

We have no competing interests.

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