Response to comments of Referee #1

Manuscript number: acp-2021-492
Authors: Xueying Liu, Amos P. K. Tai, Ka Ming Fung
Title: Responses of surface ozone to future agricultural ammonia emissions and subsequent nitrogen deposition through terrestrial ecosystem changes

This study provides a very interesting modelling study of the potential global impacts of changing agricultural demand, and thus ammonia emissions, on future surface ozone concentrations. The study provides a comprehensive set of scenarios looking at different vegetation responses to increased fertiliser use on ozone concentrations between 2000 and 2050, using both prescribed and dynamic meteorology. Overall it was shown that increased nitrogen fertiliser use by 2050 leads to increased LAI and thus enhanced surface ozone concentrations, with the biggest impact seen when dynamic meteorological affects were allowed. This study is suitable for publication in ACP after the following comments are addressed.

We would like to thank you for the thoughtful and insightful comments. The manuscript has been revised accordingly, and our point-by-point responses are provided below. The reviewer' comments are *italicized*, our new/modified text cited below is highlighted in **bold**. The revised manuscript with tracked changes is also included at the end for easy reference.

Specific Comments
Does this version of the CLM include the impacts and feedbacks of ozone damage on stomatal resistances? If so were they included in the simulations preformed in this work? This could potentially have further impacts on the nitrogen deposition effects on vegetation, particularly through stomatal uptake.

The reviewer aptly pointed out that ozone damage on stomatal conductance could affect nitrogen deposition through stomatal uptake. We have now addressed it in P15 L7 that "**One limitation of this study is that we did not consider ozone damage on stomatal conductance and photosynthesis as in the study by Sadiq et al. (2017). If ozone damage on stomatal conductance is considered, higher ozone concentrations could have positive feedbacks on ozone itself via reduced dry deposition and enhanced isoprene emission. Meanwhile, ozone damage on plant productivity may also diminish the fertilization effect of nitrogen and foliar nitrogen content, which is itself vital for photosynthetic capacity (Franz and Zaehle, 2021). Therefore, if ozone damage is considered, lower LAI and canopy height are expected, compensating some of the enhanced LAI and canopy height induced by higher nitrogen deposition found in this study. These changes in LAI and canopy height could further affect ozone via various biogeochemical and biogeophysical pathways, but such a secondary feedback effect is expected to be relatively minor (Zhou et al., 2018). More work is warranted to investigate the individual and combined effects of nitrogen deposition and ozone damage on plant growth and terrestrial carbon uptake, especially in light of the**

possible nonlinear interactions between ozone and nitrogen in plants (e.g., Shang et al., 2021).”

In Section 5 the authors present a very good summary of the potential feedbacks caused by changes in nitrogen deposition in response to future changes in agricultural practices. In particular they focus on the feedbacks through changes in LAI and canopy height. However, they do not cover the potential feedbacks involved where changes in ozone concentrations could lead to plant damage and thus impacts on not only ozone concentrations themselves but also uptake of nitrogen species. It is appreciated that given the current setup of the modelling system a further simulation is not possible but would the authors be able to give a more detailed comparison with the potential effects of ozone damage on the results observed or comment on how this could affect the results simulated by the model.

Please see above response.

Technical Comments
Page 1, Line 24: Please change to emissions

Revised as suggested.

Page 2, Line 4: The start of this sentence seems a little repetitive, please correct to something like ‘Crops typically take up only about 40-60% of the nitrogen fertiliser applied…….’

Revised as suggested.

Page 5, Line 13: Do you mean Fig 2 here?

Revised as suggested.

Response to comments of Referee #2

Manuscript number: acp-2021-492
Authors: Xueying Liu, Amos P. K. Tai, Ka Ming Fung
Title: Responses of surface ozone to future agricultural ammonia emissions and subsequent nitrogen deposition through terrestrial ecosystem changes

Comments on:
Responses of surface ozone to future agricultural ammonia emissions and subsequent nitrogen deposition through terrestrial ecosystem changes
Liu et al., submitted to Atmospheric Chemistry and Physics, August 2021
Decision: accept with minor revision and clarification

General comments:
In this manuscript, authors present a novel linkage between agricultural activities and ozone air quality, by examining the responses of surface ozone air quality to terrestrial changes caused by 2000-to-2050 increased ammonia emission and resulted increased nitrogen deposition. Authors make use of CESM model to investigate each individual and combined effects of LAI, canopy height and soil NOx, and try to isolate biogeochemical effects by using prescribed meteorology. In general, the manuscript is very well written! I think this manuscript meets the criteria for publication on Atmospheric Chemistry and Physics:
- It is an advancement in understanding the linkage between ozone air quality and agricultural activities.
- Evidence provided by the authors are strong for the conclusion drawn
- This work is of importance to researchers studying atmospheric chemistry, physics and atmosphere-biosphere interactions
However, there are some questions and details needed to be further addressed from my perspective:

> We would like to thank you for the thoughtful and insightful comments. The manuscript has been revised accordingly, and our point-by-point responses are provided below. The reviewer' comments are *italicized*, our new/modified text cited below is highlighted in **bold**. The revised manuscript with tracked changes is also included at the end for easy reference.

General revision suggestions:
Figure 1. This illustration is very helpful to readers who are not very familiar with the complex interactions between atmospheric chemistry and terrestrial ecosystem. Since one of the major conclusions is that ozone changes are typically larger when meteorology is dynamically simulated, I am wondering whether some biogeophysical effects/pathways could be added to this diagram. I understand it could get overcomplicated very fast, but maybe one or two pathways explained in Figure 7 should be added.

> We have now added biogeophysical pathways in Figure 1 and its caption: "Figure 1. "Biogeochemical" **and "biogeophysical"** pathways of nitrogen deposition affecting surface ozone concentration. Biogeochemical pathways via canopy height (yellow-

colored), leaf area index (LAI; green-colored), and soil NO_x (blue-colored), **as well as some of the biogeophysical pathways relevant for this study (red-colored)** are shown. The sign associated with each arrow indicates the correlation between the two variables; the sign of the overall effect (positive or negative) of a given pathway is the product of all the signs along the pathway. "Biogeochemical" pathways affect gas exchange (i.e. biogenic VOC emission and ozone deposition) though plant stomata or microbe-mediated soil processes. **"Biogeophysical" or "meteorological" pathways are mediated through a modification of the local and nonlocal overlying meteorological environment above the surface layer**."

Spin-up period for the model. I see that CLM45BGC mode has been spun-up for 150 years, and then 50 years for steady state. Perturbation experiment is then done for another 60-70 years. This seems an impressively long period of time for spin-up and perturbation. Is this a common practice for this mode of CLM model? Or how did you determine that the model has reached a steady state? Did the model start from zero vegetation (LAI=0)? I am interested to look at maybe just one figure showing the evolution of mean LAI over certain region during these hundreds of years of simulation. You don't have to include it in the appendix.

The 200-year simulation was to provide a steady-state initial condition for the perturbation experiments later. It started from the default initial condition files with certain LAI values (see right panel of Figure R1). We wanted to make sure that the LAI was stabilized at year-2000 level, so looping over year-2000 for 200 simulation years was adopted. The same practice is also used in Sadiq et al. (2017), Zhou et al. (2018), and Wang et al. (2020). After this, the actual perturbation experiments were simulated for 70 years. Figure R2 shows the LAI differences between year-2000 and year-2050 for the first 10–20 years. For all four regions, we observed the LAI differences are stabilized within the first 10–20 years, and then averaged the remaining 50 years as year-2050 steady state.

We have now explained further in P5 L34 that "**We used the year-2000 steady state as initial conditions for the following perturbation experiments.** We then perturbed the present-day steady state with future nitrogen deposition fluxes following the year-2050 agricultural emission scenario, allowing the vegetation and soil variables to come into a "new" steady state, which took 10–20 simulations years. After that, the simulation was conducted for another 50 years, which were **considered to be year-2050 steady state and then** averaged to determine the differences in LAI, canopy height and soil NO_x emission from the 50-year present-day averages."

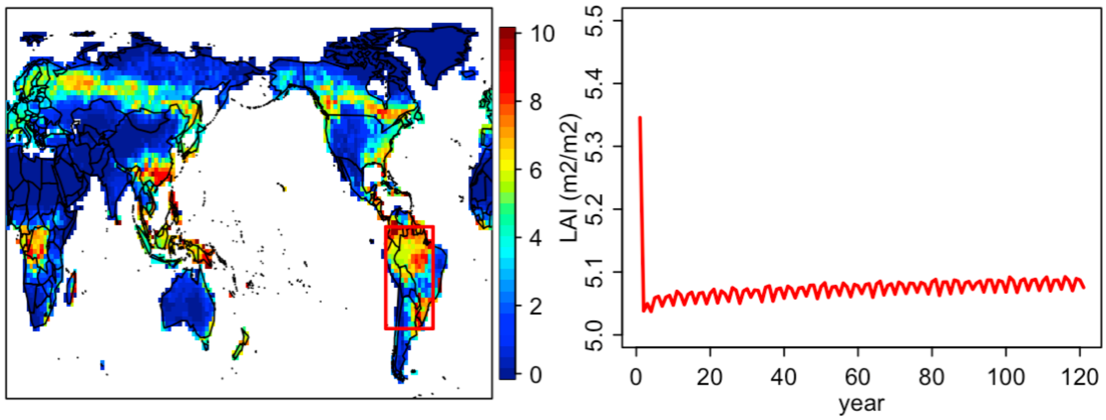

Figure R1. Left panel shows mean LAI of the 200-year simulation, and right panel shows LAI evolution of South America (red box in left panel).

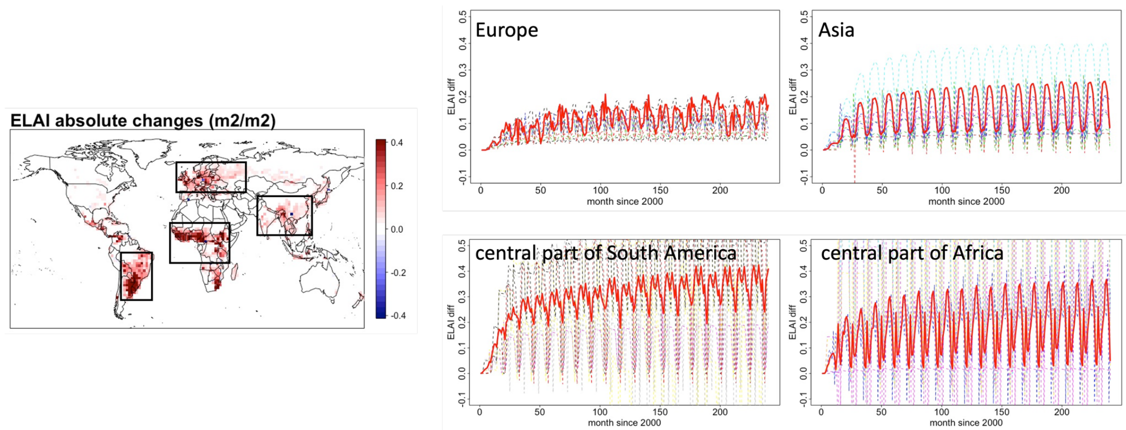

Figure R2. The LAI differences between year-2000 and year-2050 for the first 10–20 simulation years.

Reference:

Sadiq, M., Tai, A. P. K., Lombardozzi, D., and Val Martin, M.: Effects of ozone-vegetation coupling on surface ozone air quality via biogeochemical and meteorological feedbacks, Atmos. Chem. Phys., 17, 3055–3066, https://doi.org/10.5194/acp-17- 3055-2017, 2017.

Zhou, S. S., Tai, A. P. K., Sun, S., Sadiq, M., Heald, C. L., and Geddes, J. A.: Coupling between surface ozone and leaf area index in a chemical transport model: strength of feedback and implications for ozone air quality and vegetation health, Atmos. Chem. Phys., 18, 14133–14148, https://doi.org/10.5194/acp-18-14133-2018, 2018.

Wang, L., Tai, A.P., Tam, C.Y., Sadiq, M., Wang, P. and Cheung, K.K.,: Impacts of future land use and land cover change on mid-21st-century surface ozone air quality: distinguishing between the biogeophysical and biogeochemical effects, Atmos. Chem. Phys., 20, 11349–11369, https://doi.org/10.5194/acp-20-11349-2020, 2020.

P7L12, '..., we estimated that year-2050 NH3 budget to be 71 Tg N yr-1, ...'. I noticed and you discussed later as well that this number is the same as RCP8.5 projection. It is probably worth mentioning the fact and that FAO makes similar assumption as RCP 8.5 scenario here.

We have now mentioned this in P7 L14 that "**This estimate is comparable to the RCP8.5 estimate of 71 Tg N yr^{-1} as both studies assumed a business-as-usual scenario where future NUE in agroecosystems is not expected to be improved much.**"

Figure 3b. I think it would be more beneficial to have this figure in percentage changes rather than absolute changes.

We are happy to show the percentage changes of nitrogen deposition over 2000–2050. The current setting of Figure 3 is year-2000 nitrogen deposition and percentage GPP reduction on the left, and the absolute differences by year-2050 minus year-2000 on the right. If we change absolute difference in panel (b) to percentage difference, we would also need to change panel (d) to percentage difference to be consistent. Yet in this case, panel (d) becomes percentage difference of panel (c) percentage GPP reduction, which is less straightforward and complicates the explanation we show in Sect. 4.

As an alternative, we have now put the percentage changes in supplementary Figure S3, and state in P8 L6 that "**Relative changes over 2000–2050 can be found in supplementary Figure S2.**".

In Figure 3c, you have shown GPP reduction due to nitrogen limitation. I noticed some discussion about it is given in Section 4. However, I am wondering how you obtained this variable. I might have missed the part where you introduce this, but did you compute it by comparing two simulations (one with and the other without nitrogen limitation), or is it from some nitrogen limitation parameter in the model? Some introductions could be added in Section 3 or 4.

Nitrogen limitation is from a model output variable called "downregulation", which stands for downregulation of potential carbon allocation based on soil nitrogen availability.

We have now further explained this in P8 L14 that "…**In CLM, the plant nitrogen demand for new growth is calculated by the carbon available for allocation to new growth allocation, given the C:N stoichiometry of a given plant type and plant part. From the soil side, soil mineral nitrogen supply is calculated by adding various nitrogen sources (e.g., atmospheric nitrogen deposition, fertilizer, biological nitrogen fixation) and subtracting nitrogen sinks (e.g., leaching, assimilation by heterotrophs). When the plant nitrogen demand is greater than the soil nitrogen supply, the plants are not able to take up enough nitrogen to support the carbon allocation for new growth, which would then be reduced ("downregulated") by a percentage in the model, which we refer as soil "nitrogen limitation" on plant growth here.** When the soil is "nitrogen-limited", the plants are not able to take up enough nitrogen for maximum photosynthesis and unmet plant nitrogen demand is translated back to a carbon supply surplus which is eliminated through reduction of GPP in the CLM model. **Figure 3c shows**

the year-2000 GPP percentage reductions due to nitrogen limitation. Most of the nitrogen-limited soils are found over the boreal forests because of slow soil decomposition and turnover with litter of high C:N content and cold climate. Savannas and grasslands in the tropics are also mildly nitrogen-limited because of low foliar nitrogen concentrations and plant density. **Figure 3d shows the differences of GPP reductions, i.e., year-2050 GPP reductions minus year-2000 GPP reductions. We found smaller GPP reductions induced by nitrogen limitation in 2050 than 2000, reflecting higher plant productivity and growth over 2000–2050. However, this nitrogen fertilization effect is found only over nitrogen-limited regions, but not over** nitrogen-abundant regions such as India and northern China where the critical nitrogen loads are almost always exceeded (Zhao et al., 2017) despite of substantial increases of nitrogen deposition over 2000–2050.

Also, some technical corrections need to be made before the publication:
P3L4, 'facilities' to 'facilitates'.

Revised as suggested.

Figure 7 and 8, labels are inconsistent between caption and subpanels. Also, there are two subpanels labelled f.

Revised as suggested.

Response to comments of Referee #3

Manuscript number: acp-2021-492
Authors: Xueying Liu, Amos P. K. Tai, Ka Ming Fung
Title: Responses of surface ozone to future agricultural ammonia emissions and subsequent nitrogen deposition through terrestrial ecosystem changes

This manuscript presented a modelling study that aimed to quantify how future changes in atmospheric nitrogen deposition as driven by rising agricultural food production affect surface ozone levels via air-biosphere interactions. Asynchronously coupled air-biosphere modelling simulations were conducted using the atmosphere and land components of the Community Earth System Model (CESM), so that the individual biogeoschemical and biogeosphysical pathways of the nitrogen deposition-surface ozone air quality linkage. The results emphasize the importance of biogeophysical pathways or the meteorological variations induced by vegetation changes in modulating surface ozone.

The manuscript is overall well conducted and presented. The simulations are well designed, and the analyses identify a new linkage of agricultural nitrogen and air pollution. I suggest publish on ACP after the following comments been addressed.

We would like to thank you for the thoughtful and insightful comments. The manuscript has been revised accordingly, and our point-by-point responses are provided below. The reviewer' comments are *italicized*, our new/modified text cited below is highlighted in **bold**. The revised manuscript with tracked changes is also included at the end for easy reference.

Specific comments:
1) Page 5, Eq. 1: A few more sentences describing the growth factor are suggested. How it treats different crops? Could it consider ammonia emission factors may be different for different crops? Please clarify.

We generated growth factors for major crops and obtained an average growth factor from these crop-specific production growths. We have now clarified this in P5 L16 that "…**We generated the growth factors for major crops (Fig. S1) and obtained an average growth factor from these crop-specific production growths.**"

Agricultural NH_3 emission rates are different for different crops in the MASAGE_NH3 inventory (Paulot et al., 2014), which stands for year-2000 conditions. We assumed emission rates of each specific crop to remain the same in the future, which can be regarded as a representation of the "worst-case" scenario where fertilizer nitrogen use remains as inefficient as it is today.

2) Page 5, Line 20: Each atmospheric chemistry simulation was conducted for 20 years. What meteorology fields were used to represent the 2000 and 2050 conditions? Please clarify.

We have now clarified it further in P5 L21 that "…For each scenario of the sensitivity experiments, CAM-Chem simulations were conducted for 20 simulation years.

Throughout the CAM-Chem component was coupled online with CLM45SP with prescribed vegetation structures, which computed land-atmosphere fluxes for CAM-Chem to simulate atmospheric dynamics and chemistry. Both simulations were performed with prescribed sea surface temperature and sea-ice cover following the HadISST dataset (Rayner et al., 2003) at the year-2000 level. Long-lived greenhouse gases and their radiative forcing were kept at year-2000 level to exclude the effects of increasing temperature on NH_3 emissions. The first five years…"

There are also more details on meteorological fields in P4 L16: "…CAM-Chem provides the flexibility of performing climate simulations online (i.e., "dynamic meteorology") and simulations with specified meteorological fields (i.e., "prescribed meteorology"). For simulations with dynamic meteorology, it was driven by the Climatic Research Unit – National Centers for Environmental Prediction (CRU-NCEP) climate forcing dataset. For simulations with prescribed meteorology, year-2000 and 2001 horizontal wind components, air temperature, surface temperature, surface pressure, sensible and latent heat flux and wind stress of the Goddard Earth Observing System Model version 5 (GEOS-5) forcing data at six hour interval were used (see Table 1). This version of CAM-Chem…"

3) Page 9, Line 29: "Ozone dry deposition velocity decreases by 0.002-0.004 …". Should it be increases in ozone dry deposition velocity as shown by figure 5?

Revised as suggested.

4) Page 11, Figure 6: It appears that the individual effects do not add up when with dynamic meteorology. As shown in this figure, ozone changes due to LAI (figure 6d) and due to HTOP (figure 6g) show large positive values in the central US, while the combined effects (due to ALL, figure 6m) become much weaker. The same issue can be seen for deposition velocity changes over the US (figure 6f/i/6o). Can you explain why?

We have now attempted to address the issue more in P13 L9: "**It is noteworthy that unlike with prescribed meteorology, individual effects may not add up linearly with dynamic meteorology for a given location due to the complex and far-reaching changes in atmospheric circulation and the associated cascade of local and nonlocal changes in climate that are dynamically simulated following terrestrial changes.**"

5) Page 11, Line 15: "increase local albedo, which results in enhancement in absorbed solar radiation". It is not clear why higher albedo could lead to higher absorbed solar radiation, as higher albedo tends to reflect more solar radiation back to the atmosphere. Please clarify.

We have now revised Figure 7 and also revamped the explanation of the biogeophysical mechanisms behind in P11 L21, which does not involve the questionable changes in albedo anymore: "Therefore, here we choose the US which shows obvious ozone enhancement following vegetation changes, as an example to illustrate the biogeophysical effects further. **Figure 7 shows that in the forest regions in the eastern US where LAI and canopy height changes are relatively large following higher nitrogen deposition, albedo decreases, absorbed radiation increases, latent heat flux increases, and such changes**

appear to have shifted the surface energy balance and circulation patterns in a way that enhances moisture convergence, precipitation and soil moisture in the originally wetter places (i.e., the forested eastern US), but reduces the moisture convergence in the originally drier places (i.e., the grassland regions in the central US). This constitutes a feedback loop in these grassland regions that reduces transpiration, increases temperature, increases aridity and thus the plant stomata close more, all leading to the relatively large enhancements in surface ozone there. Our mild vegetation changes **only** have modest local impacts in places with dense vegetation to begin with (e.g., the eastern US). …"

6) Page 11, Line 22: Wang et al. (2020) is not listed in the References;

Revised as suggested.

Page 12, Figure 7: There are two (f) panels in the figure;

Revised as suggested.

Page 16: Line 26-31, missing journal and page information for the two citations.

Revised as suggested.

Responses of surface ozone to future agricultural ammonia emissions and subsequent nitrogen deposition through terrestrial ecosystem changes

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Abstract. With the rising food demands from the future world population, more intense agricultural activities are expected to cause substantial perturbations to the global nitrogen cycle, aggravating surface air pollution and imposing stress on terrestrial ecosystems. Much less studied, however, is how the terrestrial ecosystem changes induced by agricultural nitrogen deposition may modify biosphere-atmosphere exchange and further exert secondary feedback effects on global air quality. Here we examined the responses of surface ozone air quality to terrestrial ecosystem changes caused by 2000-to-2050 changes in agricultural ammonia emission and the subsequent nitrogen deposition by asynchronously coupling between the land and atmosphere components within the Community Earth System Model framework. We found that global gross primary production is enhanced by 2.1 Pg C yr^{-1} following a 20% (20 Tg N yr^{-1}) increase in global nitrogen deposition by the end of year 2050 in response to rising agricultural ammonia emissions. Leaf area index was simulated to be higher by up to 0.3–0.4 m^2 m^{-2} over most tropical grasslands and croplands, and 0.1–0.2 m^2 m^{-2} across boreal and temperate forests at midlatitudes. Around 0.1-0.4 m increases in canopy height were found in boreal and temperate forests, and ~0.1 m increases in tropical grasslands and croplands. We found that these vegetation changes could lead to surface ozone changes by ~0.5 ppbv when prescribed meteorology was used (i.e., large-scale meteorological responses to terrestrial changes were not allowed), while surface ozone could typically be modified by 2–3 ppbv when meteorology was dynamically simulated in response to vegetation changes. Rising soil NO_x emission from 7.9 to 8.7 Tg N yr^{-1} could enhance surface ozone by 2–3 ppbv with both prescribed and dynamic meteorology. We thus conclude that following enhanced nitrogen deposition, the modification of the meteorological environment induced by vegetation changes and soil biogeochemical changes are the more important pathways that can modulate future ozone pollution, representing a novel linkage between agricultural activities and ozone air quality.

1 Introduction

Increased food production for the ever-growing world population has been enabled by the widespread agricultural expansion and intensification with heavy fertilizer applications, which have correspondingly led to an enhancement in ammonia (NH_3) emission from the land by a factor of two to five since preindustrial times (Behera et al., 2013; Gu et al., 2015; Zhu et al., 2015). For instance, Asia (excluding Siberia), home to more than 60% of the world population (FAOSTAT, 2016), has experienced rapid expansion of agricultural activities (Liu & Tian, 2010; Tian et al., 2014), accounting for ~50% of the global total consumption of synthetic fertilizer and 30–40% of global manure production (FAOSTAT, 2016). Agriculture-related activities are known to be the most significant sources of atmospheric NH_3, of which the vast majority (~60%) originates from the excessive use of nitrogenous fertilizer and concentrated operations of livestock feeding on a global scale (Huang et al., 2012; Paulot et al., 2014; Zhang et al., 2018); for Asia the percentage is even higher (80–90%) (Streets et al., 2003; Reis et al., 2009; Gu et al., 2012; Kang et al., 2016; Zhang et al., 2017; Zhang et al., 2018). Crops typically take up only about 40–60% of the nitrogen fertilizer applied to croplands (Tilman et al., 2002; Zhang et al., 2015; Liu et al., 2016; Muller et al., 2017), and only 25–35% of the nitrogen fed to dairy cows is converted into milk (Bittman et al., 2009), while most of the remainder is chemically transformed into a variety of simple and complex forms and leaked to the environment. The release of gaseous NH_3 into the atmosphere is one of the major nitrogen leakages from agricultural soils. Under a business-as-usual scenario where future nitrogen use efficiency (NUE; i.e., the fraction of nitrogen input finally harvested as output) in agricultural systems is not expected to be substantially improved, increasing food production will undoubtedly continue to intensify agricultural NH_3 emission into the overlying air (Erisman et al., 2008; Lamarque et al., 2011; Zhang et al., 2017).

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Reactive nitrogen, from emissions of nitrogen oxides (NO_x; $NO+NO_2$) and NH_3, is deposited over land and ocean through a variety of processes collectively known as wet and dry deposition. As combustion-driven NO_x emission is projected to slow down due to regulatory efforts (van Vuuren et al., 2011) while agricultural NH_3 emission will continue to increase (Lamarque et al., 2011), future nitrogen deposition is expected to increase overall in the global budget (Galloway et al., 2004; Paulot et al., 2013; Lamarque et al., 2013; Kanakidou et al., 2016) and shift from a nitrate-dominated to ammonium-dominated condition (Ellis et al., 2013; Paulot et al., 2013; Li et al., 2016). Atmospheric nitrogen deposition onto the land surface is an important source of soil mineral nitrogen and thus enhances plants growth; this is known as the "nitrogen fertilization effect" (Reay et al., 2008; Templer et al., 2012). The fertilization effect depends on the soil "nitrogen limitation" defined as the nitrogen constraint on the productivity of many terrestrial ecosystems (Vitousek et al., 2002; Gruber and Galloway, 2008; LeBauer et al., 2008; Heimann et al., 2008; Reay et al., 2008; Zaehle et al., 2010). Nitrogen limitation is often found in natural soils where severe nitrogen competition among plants and microbes exists, and the unmet plant nitrogen demand can be translated to a reduction in the potential gross primary production (GPP) of the terrestrial ecosystems, representing a direct downregulation of photosynthetic carbon gain.

Nitrogen deposition affects the terrestrial carbon and nitrogen cycle, but much less is known about how nitrogen deposition affects atmospheric chemistry via terrestrial changes and feedbacks. As nitrogen limitation is relaxed, enhanced carbon assimilation can be translated to changes in the carbon mass allocated to different plant parts, ultimately manifested as an enhancement in vegetation structural variables such as leaf area index (LAI) and canopy height. Meanwhile, nitrogen deposition can also alter soil inorganic nitrogen composition and a variety of abiotic and biotic processes including uptake by plants, nitrification, denitrification, immobilization by microbes, and fixation in clay minerals. Soil NO_x is produced as a by-product of nitrification and denitrification, two microbial processes that first convert NH_3 aerobically to nitrate (NO_3^-) and then NO_3^- to nitrous oxide (N_2O) or nitrogen gas (N_2) under anoxic conditions. As LAI, canopy height and soil NO_x are known to affect surface air quality, nitrogen deposition can potentially affect atmospheric chemistry through affecting vegetation structure and ecophysiology, as well as soil biogeochemistry.

Nitrogen-mediated changes in vegetation and soil can affect surface ozone air quality via various pathways (Fig. 1). Among them, "biogeochemical" effects are processes mediated via direct exchange (i.e., emissions or deposition) of relevant chemical species between the terrestrial biosphere (vegetation and soil microbes) and the atmosphere, while "biogeophysical" or "meteorological" effects are mediated through a modification of the overlying meteorological environment (i.e., temperature, humidity, turbulence structure, etc.), as defined in Sadiq et al. (2017), Zhou et al. (2018) and Wang et al. (2020). One possible biogeochemical pathway is that LAI enhancement could elevate surface ozone by increasing biogenic volatile organic compound (VOC) emissions in high-NO_x environments, but could also reduce ozone by increasing dry-depositional uptake via leaf stomata (Zhao et al., 2017). Another possible biogeochemical effect is via the increase in canopy height, which further enhances surface roughness length, turbulent mixing and thus higher aerodynamic conductance for land-atmosphere exchange including ozone dry deposition (Bonan, 2016; Oleson et al. 2013). Another possible biogeochemical effect is that increased inorganic nitrogen availability facilitates soil NO_x emission through nitrification and denitrification processes, which further causes rapid NO and NO_2 cycling for ozone formation. Biogeophysical effects or meteorological effects are through vegetation-induced changes in the surface energy balance (e.g., absorbed solar radiation, sensible and latent heat fluxes) and subsequent changes in surface temperature, precipitation, humidity, circulation patterns, moisture convergence (Wang et al., 2020). Higher temperature enhances ozone mainly through increased biogenic emissions and higher abundance of NO_x, while lower humidity reduces the chemical loss rate of ozone (Jacob and Winner, 2009; Fiore et al., 2012). Surface ozone changes via each individual process are heterogeneous over the globe, and the overall ozone response through various biogeochemical and biogeophysical pathways is highly complex (Zhao et al., 2017).

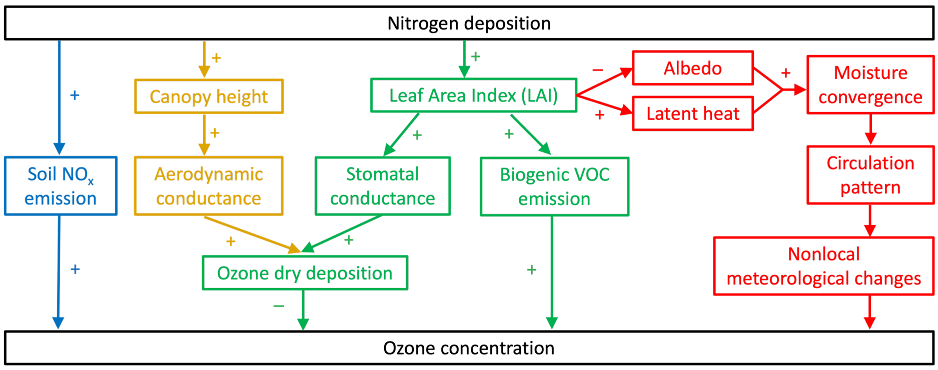

Figure 1. "Biogeochemical" and "biogeophysical" pathways of nitrogen deposition affecting surface ozone concentration. Biogeochemical pathways via canopy height (yellow-colored), leaf area index (LAI; green-colored), and soil NO_x (blue-colored), as well as some of the biogeophysical pathways relevant for this study (red-colored) are shown. The sign associated with each arrow indicates the correlation between the two variables; the sign of the overall effect (positive or negative) of a given pathway is the product of all the signs along the pathway. "Biogeochemical" pathways affect gas exchange (i.e. biogenic VOC emission and ozone deposition) though plant stomata or microbe-mediated soil processes. "Biogeophysical" or "meteorological" pathways are mediated through a modification of the local and nonlocal overlying meteorological environment above the surface layer.

Here we present a study that investigates how agriculture-induced increases in NH_3 emission and subsequent nitrogen deposition could affect surface ozone air quality via terrestrial ecosystem changes in terms of LAI, canopy height and soil NO_x emission. We used an asynchronously coupled modeling framework based on the atmosphere (CAM-Chem) and land (CLM) components of the Community Earth System Model (CESM) to quantify the corresponding responses of surface ozone air quality to terrestrial changes. We first examined the responses of vegetation and soil variables to the present-day vs. future scenarios of nitrogen deposition and then use those terrestrial changes to drive factorial simulations for surface ozone. To evaluate the relative importance of LAI, canopy height and soil NO_x emission, we evaluated ozone responses to the three individual effects and the overall combined effects using prescribed meteorology (i.e., large-scale meteorological responses to terrestrial changes are not allowed). Furthermore, we evaluated the effects of changing meteorology to surface ozone by conducting simulations using dynamic meteorology (i.e., where overlying boundary-layer meteorology and large-scale circulation also responds to terrestrial changes). Model configuration with dynamic meteorology represents the overall effects

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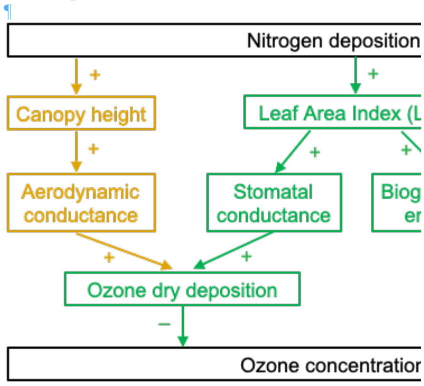

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from regional terrestrial changes and associated meteorological changes (an integration over both biogeochemical and biogeophysical effects to surface ozone), whereas the setting with prescribed meteorology provides limited above-surface layer meteorological changes directly caused by terrestrial changes and represents the biogeochemical effects only. Our study emphasizes the complexity of biosphere-atmosphere interactions and their indirect modulating effects on air quality and atmospheric chemistry, which are important for evaluating the impacts from future food production trends on air quality and health beyond the direct effects of agricultural emissions alone.

2 Model and Method

2.1 Model description

We used the Community Earth System Model (CESM), which includes atmospheric, land, ocean and sea ice model components. We employed CESM version 1.2.2 with fully interactive atmosphere and land components, but with prescribed ocean and sea ice consistent. For the atmosphere component, we used the Community Atmosphere Model version 4 (CAM4) (Neale et al., 2010) fully coupled with an atmospheric chemistry scheme (i.e., CAM-Chem) that contains full tropospheric O_3-NO_x-CO-VOC-aerosol chemistry based on the MOZART-4 mechanism (Emmons et al., 2010; Lamarque et al., 2012). Emissions are from the combined emission inventories of the Emissions Database for Global Atmospheric Research (EDGAR), Regional Emission inventory in ASia (REAS) and Global Fire Emissions Database (GFED2) and others. CAM-Chem provides the flexibility of performing climate simulations online (i.e., "dynamic meteorology") and simulations with specified meteorological fields (i.e., "prescribed meteorology"). For simulations with dynamic meteorology, it was driven by the Climatic Research Unit – National Centers for Environmental Prediction (CRU-NCEP) climate forcing dataset. For simulations with prescribed meteorology, year-2000 and 2001 horizontal wind components, air temperature, surface temperature, surface pressure, sensible and latent heat flux and wind stress of the Goddard Earth Observing System Model version 5 (GEOS-5) forcing data at six hour interval were used (see Table 1). This version of CAM-Chem simulates the concentrations of 56 atmospheric chemical species at a horizontal latitude-by-longitude resolution of 1.9°×2.5° and a vertical resolution of 26 layers for dynamic meteorology and 52 layers for prescribed meteorology.

For the land component, we used the Community Land Model version 4.5 (CLM4.5) (Oleson et al., 2013) with Satellite Phenology (CLM45SP) mode where vegetation structures are prescribed (e.g., using satellite-derived LAI data), or with active carbon–nitrogen biogeochemistry (CLM45BGC) that contains prognostic treatment of terrestrial carbon and nitrogen cycles (Lawrence et al., 2011), depending on the cases of concern. In CLM4.5, the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 was used to compute biogenic emissions online as functions of LAI, vegetation temperature, solar radiation, soil moisture and other environmental conditions (Guenther et al., 2012). For dry deposition of gases and aerosols we used the resistance-in-series scheme in CLM4.5 as described in Lamarque et al. (2012) with updated, optimized coupling of stomatal resistance to LAI (Val Martin et al., 2014). Soil NO_x emission was implemented by Fung et al. (2021) as a function of N_2O emission, soil air-filled pore space and volumetric soil water content during nitrification and denitrification (See Supplementary for details). We also applied a temperature factor to correct the soil NO_x overestimation at high latitudes as previous studies (Zhao et al., 2017). Evapotranspiration rate was calculated based on the Monin-Obukhov similarity theory for turbulent exchange and the diffusive flux-resistance model with dependence on vegetation, ground and surface temperature, specific humidity, and an ensemble of resistances that are functions of meteorological and land surface conditions (Oleson et al., 2013; Lawrence et al., 2011; Bonan et al., 2011).

2.2 Asynchronously coupled atmosphere chemistry-biosphere modeling framework

An asynchronously coupled system with CAM-Chem and CLM was adopted to investigate the vegetation structural changes induced by nitrogen deposition and their potential to modulate surface ozone under both dynamic and prescribed meteorology. Asynchronous instead of synchronous coupling was used because currently CESM does not have the capacity to allow "online" bidirectional exchange of reactive nitrogen fluxes between the atmosphere and land components; it also conveniently facilitates sensitivity experiments to be conducted to isolate individual drivers of changes and processes. First, present-day and future scenarios of nitrogen deposition are obtained by CAM-Chem simulations with the corresponding NH_3 emission of year 2000 and 2050. Year-2000 NH_3 emission was from the prescribed emission inventory inherent in CAM-Chem (see Sect. 2.1), which includes anthropogenic, ocean, soil and biomass burning sources. We split the year-2000 anthropogenic NH_3 emission into agricultural and non-agricultural parts by using the corresponding ratios based on the Magnitude And Seasonality of Agricultural Emissions model for NH_3 (MASAGE_NH3) (Paulot et al., 2014). We kept natural and non-agricultural emissions the same in both the year-2000 and year-2050 scenarios, and only scaled the year-2000 agricultural NH_3 by a growth factor g (Fig. 2c)

$$g = \frac{\text{crop production in 2050}}{\text{crop production in 2000}} \qquad \text{Eq.1}$$

based on crop production estimates from Alexandratos and Bruinsma (2012) accounting for technology-driven yield improvements and cropland area changes, as in Tai et al. (2014; 2017). We generated the growth factors for major crops (Fig. S1) and obtained an average growth factor from these crop-specific production growths. Such a linear scaling assumes nitrogen-use efficiency (NUE) of fertilization applications to remain the same in the future. In practice NUE is expected to rise with technological advancements, the extent of which is however highly uncertain and region-specific; we therefore regarded our linear scaling as a representation of the "worst-case" scenario where fertilizer nitrogen use remains as inefficient as it is today. For each scenario of the sensitivity experiments, CAM-Chem simulations were conducted for 20 simulation years. Throughout the CAM-Chem component was still coupled online with CLM45SP with prescribed vegetation structures, which computed land-atmosphere fluxes for CAM-Chem to simulate atmospheric dynamics and chemistry. Both simulations were performed with prescribed sea surface temperature and sea-ice cover following the HadISST dataset (Rayner et al., 2003) at the year-2000 level. Long-lived greenhouse gases and their radiative forcing were kept at year-2000 level to exclude the effects of increasing temperature on NH_3 emissions. The first five years of outputs were treated as spin-up and thus discarded in the analysis, and we calculated the annual averages of the last 15 years to obtain the corresponding nitrogen deposition fluxes for the year-2000 and year-2050 scenarios.

The CLM45BGC mode was used to investigate vegetation and soil changes in response to perturbations in the nitrogen input to the land. We first obtained steady-state vegetation and soil variables including LAI, canopy height and soil NO_x emission following present-day nitrogen deposition (obtained above CAM-Chem) for 200 years in CLM. The first 150 years of outputs were treated as spin-up, while the last 50-year average was used to represent the vegetation and soil conditions in a steady state. We used the year-2000 steady state as initial conditions for the following perturbation experiments. We then perturbed the present-day steady state with future nitrogen deposition fluxes following the year-2050 agricultural emission scenario, allowing the vegetation and soil variables to come into a "new" steady state, which took 10–20 simulations years. After that, the simulation was conducted for another 50 years, which were considered to be year-2050 steady state and then averaged to determine the differences in LAI, canopy height and soil NO_x emission from the 50-year present-day averages.

Last, we investigated the individual and combined impacts of the above changes in the three terrestrial pathways (i.e., via LAI, canopy height and soil NO_x emission) on surface ozone air quality, with both prescribed meteorology (i.e., large-scale meteorological responses to terrestrial changes are not allowed) and dynamic meteorology (i.e., overlying boundary-layer

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meteorology and large-scale circulation also responds to terrestrial changes). Terrestrial changes with prescribed meteorology included only biogeochemical pathways, while terrestrial changes with dynamic meteorology included the combined effects of biogeochemical and biogeophysical processes as well as larger meteorological and circulation pattern changes. Therefore, we were able to examine the effects from land-atmosphere feedbacks with dynamic meteorology, while prescribed meteorology provided limited atmospheric changes directly caused by terrestrial changes without much land-atmosphere feedbacks. To evaluate the relative importance of individual pathways to the overall effects, we conducted four sets of fully coupled land-atmosphere simulations: (1) a control case without any nitrogen-mediated changes in LAI, canopy height and soil NO_x emission ([CTR]); (2) a simulation with LAI change only ([LAI]); (3) a simulation with canopy height change only ([HTOP]); (4) a simulation with soil NO_x emission change only ([NOX]); (5) a simulation with all changes in LAI, canopy height and soil NO_x emission ([ALL]). Simulation [LAI], [HTOP] and [NOX] in relation to [CTR] allowed us to quantify the relative contribution from LAI, canopy height and soil NO_x emission respectively, while simulation [ALL] reflected the overall ozone changes due to three combined effects. The experiments were summarized in Table 1. We conducted the same set of simulations with both dynamic and prescribed meteorology to examine how meteorological responses to these terrestrial changes would modify the importance of these pathways (Table 2). We focused on average changes in the last 15-year northern summer (June, July and August: JJA) for most of the variables in the rest of this paper, since summer was both the high-ozone season and the growing season of the majority of global vegetation, when ozone-vegetation coupling appeared to be the strongest and significant.

Table 1. Meteorological inputs for simulations with dynamic and prescribed meteorology.

	Dynamic	Prescribed
Meteorology	Simulated within CAM	GEOS-5 reanalysis data
Terrestrial changes	[CTR], [LAI], [HTOP], [NOX], [ALL]	[CTR], [LAI], [HTOP], [NOX], [ALL]

Table 2. Experimental design to quantify surface ozone responses to terrestrial changes including leaf area index (LAI), canopy height, and soil NO_x emission.

	[CTR]	[LAI]	[HTOP]	[NOX]	[ALL]
LAI	Year 2000	Year 2050	Year 2000	Year 2000	Year 2050
Canopy height	Year 2000	Year 2000	Year 2050	Year 2000	Year 2050
Soil NO_x	Year 2000	Year 2000	Year 2000	Year 2050	Year 2050

3 Year-2000 vs. year-2050 NH_3 emissions and nitrogen deposition

We first show the year-2000 emissions of reactive nitrogen as NO_x (48 Tg N yr^{-1}, Fig. 2a) and NH_3 (53 Tg N yr^{-1}, Fig. 2b), with a global budget of 101 Tg N yr^{-1}, in good agreement with Ciais et al. (2013). NO_x is densely emitted from industrial and populated regions, while hotspots for NH_3 emission are India and eastern China with intensive agricultural activities and inefficient fertilizer use. Global year-2050 NH_3 emission is projected to reach 67, 57, 65 and 71 Tg N yr^{-1} in Representative Concentration Pathway (RCP) RCP2.6, RCP4.5, RCP6.0 and RCP8.5 respectively, mainly due to rising agricultural production (RCP database version 2.0.5). Yet RCP projections did not include a sufficient representation of the spatial patterns of agricultural NH_3 emissions worldwide and especially in Asia, the world's most productive croplands (RCP database version 2.0.5). To capture 2000-to-2050 agricultural intensification, we therefore estimated future NH_3 emission based on FAO 2000-to-2050 crop production changes.

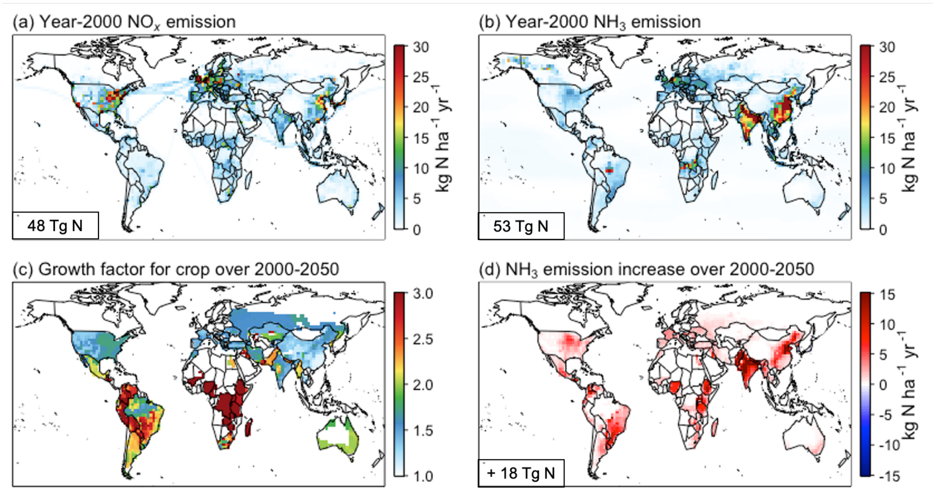

Figure 2. Global year-2000 emissions of **(a)** NO_x and **(b)** NH_3, **(c)** growth factor *g* of crop production increase over 2000–2050 from the Food and Agriculture Organization of the United Nations (FAO), and **(d)** projected increases in NH_3 emission over 2000–2050.

FAO projects global year-2050 crop production to be higher than year-2000 level due to changes in yield, crop intensity (i.e., multiple cropping, shortening of fallow periods), and arable land (Alexandratos and Bruinsma, 2012). The major increases occur in South America and Central Africa due to yield increases and arable land expansion. Production growth factor *g* in Fig. 2c can go up to 2–3 for South America and 3–5 for Central Africa, while it is 1.5–2 for some of the world's most productive croplands at northern midlatitudes, suggesting that the Southern Hemisphere will be playing an increasingly important role in producing food for the future global population. By scaling up year-2000 NH_3 emission by growth factor in FAO crop production, we estimated that year-2050 NH_3 budget to be 71 Tg N yr^{-1}, a 34% increase (18 Tg N yr^{-1}) compared to year-2000 emission, with major increases over East China, India, Midwestern United States, Brazil, Argentina and East Africa (Fig. 2d). This estimate is comparable to the RCP8.5 estimate of 71 Tg N yr^{-1} as both studies assumed a business-as-usual scenario where future NUE in agroecosystems is not expected to be improved much. We fed both year-2000 and year-2050 NH_3 emissions into the CESM model to simulate the corresponding nitrogen deposition. Global budget of both reduced (NH_x) and oxidized (NO_y) nitrogen deposition is 101 Tg N yr^{-1} in year 2000 (Fig. 3a), which almost balances out the emission totals of both NH_3 and NO_x. Nitrogen deposition in 2050 is 121 Tg N yr^{-1}, a 20% (20 Tg N yr^{-1}) increase from the year-2000 total (Fig. 3b). Increases in 2000-to-2050 nitrogen deposition mostly result from increased NH_x deposition, since we fixed the NO_x emission at year-2000 level to isolate the deposition changes due to agricultural intensification alone. These increased nitrogen deposition serves as an important input of mineral nitrogen from the atmosphere to the biosphere.

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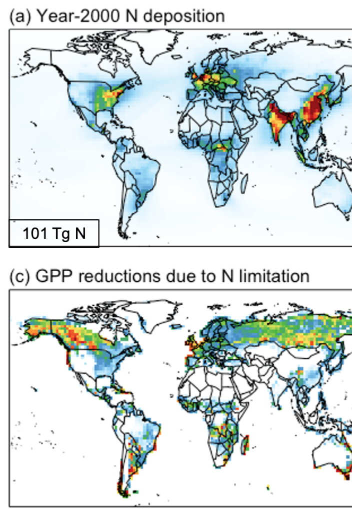

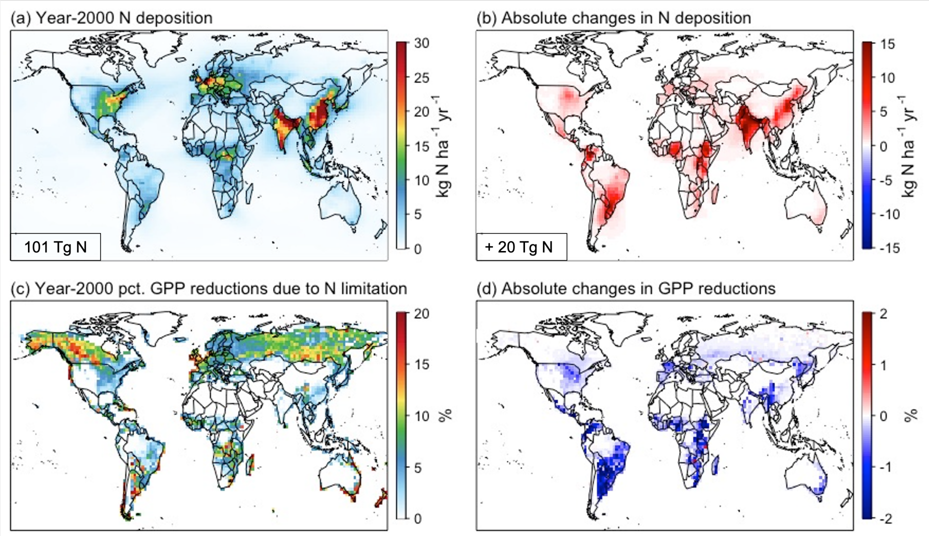

Figure 3. (a) Year-2000 atmospheric nitrogen deposition and **(b)** absolute changes in nitrogen deposition over 2000–2050. **(c)** Year-2000 gross primary production (GPP) percentage reduction due to nitrogen limitation as presented in the CLM model. In nitrogen-limited soils (i.e., colored areas), plant growth is limited by insufficient soil nitrogen supply due to plant-microbe competition. **(d)** Absolute changes in nitrogen limitation-induced GPP reductions because of enhanced nitrogen availability from atmospheric nitrogen deposition over 2000–2050. Relative changes over 2000–2050 can be found in supplementary Figure S2.

4 Responses of terrestrial ecosystems to nitrogen deposition

We present in this section the fertilization effect of year-2050 nitrogen deposition and associated enhancements in vegetation structure (i.e., LAI and canopy height) and soil NO_x emission compared with those of year-2000 nitrogen deposition. Nitrogen uptake from the soil is an important determinant of plant growth, as nitrogen is a major component of chlorophyll (i.e., pigments absorbing light energy for photosynthesis) and Rubisco (i.e., enzyme necessary for carbon fixation). Meanwhile, mineral nitrogen availability is also vital for nitrification and denitrification microbial processes where NO_x is produced as a by-product. In CLM, the plant nitrogen demand for new growth is calculated by the carbon available for allocation to new growth allocation, given the C:N stoichiometry of a given plant type and plant part. From the soil side, soil mineral nitrogen supply is calculated by adding various nitrogen sources (e.g., atmospheric nitrogen deposition, fertilizer, biological nitrogen fixation) and subtracting nitrogen sinks (e.g., leaching, assimilation by heterotrophs). When the plant nitrogen demand is greater than the soil nitrogen supply, the plants are not able to take up enough nitrogen to support the carbon allocation for new growth, which would then be reduced ("downregulated") by a percentage in the model, which we refer as soil "nitrogen limitation" on plant growth here. When the soil is "nitrogen-limited", the plants are not able to take up enough nitrogen for maximum photosynthesis and unmet plant nitrogen demand is translated back to a carbon supply surplus which is eliminated through reduction of GPP in the CLM model. Figure 3c shows the year-2000 GPP percentage reductions due to nitrogen limitation. Most of the nitrogen-limited soils are found over the boreal forests because of slow soil decomposition and turnover with litter of high C:N content and cold climate. Savannas and grasslands in the tropics are also mildly nitrogen-limited because of low foliar nitrogen concentrations and plant density. Figure 3d shows the differences of GPP reductions, i.e., year-2050

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GPP reductions minus year-2000 GPP reductions. We found smaller GPP reductions induced by nitrogen limitation in 2050 than 2000, reflecting higher plant productivity and growth over 2000–2050. However, this nitrogen fertilization effect is found only over nitrogen-limited regions, but not over nitrogen-abundant regions such as India and northern China where the critical nitrogen loads are almost always exceeded (Zhao et al., 2017) despite of substantial increases of nitrogen deposition over 2000–2050.

Due to nitrogen fertilization, GPP, LAI, canopy height and soil NO_x emission over nitrogen-limited regions are generally higher with year-2050 nitrogen deposition (Fig. 4). Specifically, we found that year-2050 nitrogen deposition to the land enhances global GPP by 2.1 Pg C yr^{-1} (Fig. 4e), and the enhanced carbon assimilation can be translated into changes in the carbon mass allocated to different plant parts such as leaves, stems and roots. The two vegetation structural proxies in the CLM model, LAI and canopy height, which characterize the carbon allocation to plant tissues leaf and stem, respectively. LAI was simulated to be higher by up to 0.3–0.4 m^2 m^{-2} over tropical grasslands and croplands in Brazil, savannas in Sub-Saharan Africa, and 0.1–0.2 m^2 m^{-2} across boreal and temperate forests at midlatitudes (Fig. 4f). Canopy heights from broadleaf deciduous trees and needleleaf evergreen trees were simulated to be higher by up to 0.1–0.3 m over the eastern US, southern Europe, southern Russia and southeastern China, and increases of 0.3–0.4 m were found over broadleaf deciduous trees in South America, and ~0.1 m increases were found for grasses and crops over Sub-Saharan Africa (Fig. 4g). Meanwhile, global soil NO_x emission budget rises from 7.9 Tg N yr^{-1} to 8.7 Tg N yr^{-1} (Fig. 4h) due to faster and greater nitrification and denitrification processes under year-2050 atmospheric nitrogen deposition.

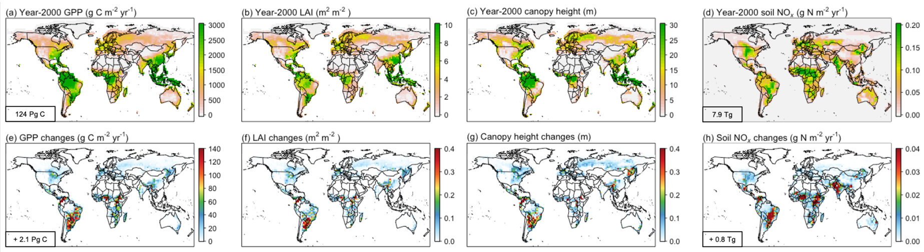

Figure 4. Annual mean of year-2000 **(a)** gross primary production (GPP), **(b)** leaf area index (LAI), **(c)** canopy height, **(d)** soil NO_x emission, and corresponding increases **(e-h)** due to increased nitrogen deposition over 2000–2050.

5 Impacts of terrestrial changes on surface ozone air quality

5.1 Surface ozone changes with prescribed meteorology

We first examined the responses of surface ozone air quality to changes in LAI, canopy height and soil NO_x separately, as well as the combined effects of all, with prescribed meteorology (i.e., large-scale meteorological responses to these terrestrial changes are not accounted for in the ozone changes). With prescribed meteorology, the responses of ozone are seen mostly where the changes in vegetation cover or soil emission take place. Figure 5d shows that LAI modulates surface ozone biogeochemically (i.e., without perturbing the overlying meteorology) by ±0.5 ppbv depending on the counteracting effects from enhanced biogenic VOC emission (Fig. 5e) and surface conductance for ozone deposition (Fig. 5d). We estimated a 3.0 Tg yr^{-1} increase in global biogenic isoprene emission (Fig. 5e), a key source of reduced atmospheric hydrocarbons that are the chief precursors of tropospheric ozone. Yet, rises in dry deposition velocity (Fig. 5f) reduce ozone concentration. The sensitivity of isoprene emission to LAI is higher than that of dry deposition, rendering the effects of isoprene emission

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dominant in northern midlatitude regions with low LAI to begin with (Wong et al., 2018). As shown in Fig. 5g, increased canopy height decreases ozone by 0.2 ppbv through stronger aerodynamic conductance and thus stronger turbulent exchange and dry deposition within the surface layer (without the corresponding changes in the overlying boundary-layer meteorology, however, due to prescribed meteorology). Ozone dry deposition velocity increases by 0.002–0.004 cm s^{-1}, with increased canopy height in central Africa and the northern US. Figure 5j shows that surface ozone is elevated biogeochemically by 1–3 ppbv in certain low-NO_x equatorial regions due to increased soil NO_x emission. Overall ozone changes with prescribed meteorology (Fig. 5m) are mostly local and can be explained predominately (80–90%) by biogeochemical effects from soil NO_x emission.

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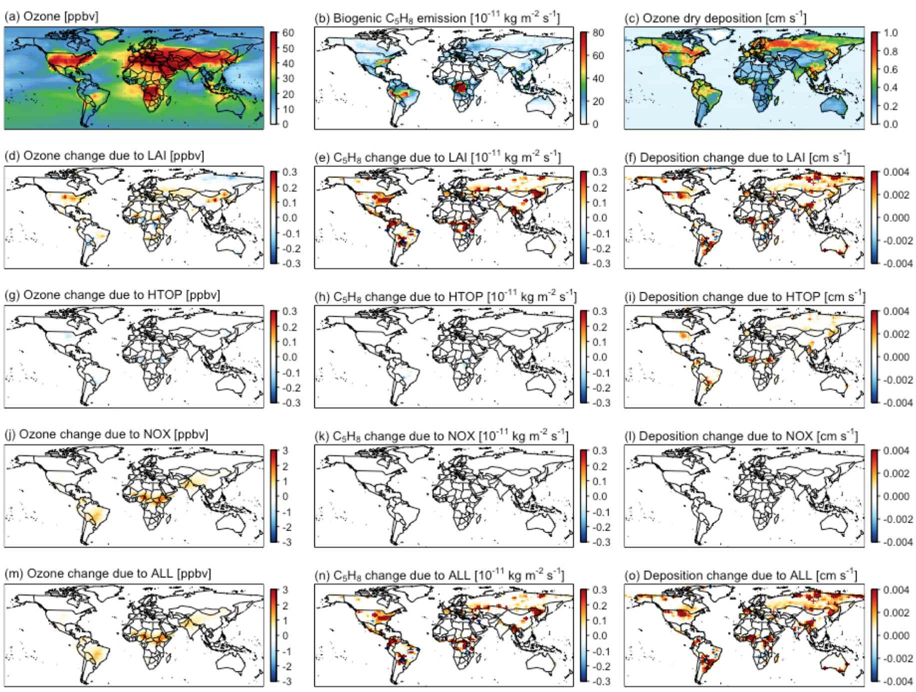

Figure 5. Year-2000 summertime (June-July-August; JJA) level of **(a)** surface ozone concentration, **(b)** biogenic isoprene emission, **(c)** ozone dry deposition, and their corresponding changes due to nitrogen-mediated increases in LAI only **(d, e, f)**, canopy height only **(g, h, i)** and soil NO_x emission only **(j, k, l)**, and the combination increases of all **(m, n, o)** with prescribed meteorology.

5.2 Surface ozone changes with dynamic meteorology

To evaluate the relative importance of regional terrestrial changes vs. terrestrial changes with meteorological changes in regulating surface ozone concentration, we also conducted simulations with dynamic meteorology (i.e., overlying boundary-layer meteorology and large-scale circulation could respond to terrestrial changes). The ozone changes with dynamic meteorology are the combined results from regional terrestrial changes and associated meteorological changes, an integration over both biogeochemical and biogeophysical effects. Figure 6 shows that the changes in summertime surface ozone are within

±2–3 ppbv with dynamic meteorology. Overall ozone change with dynamic meteorology (Fig. 6m) are the combined results from the integrated effects of vegetation changes (Fig. 6d, g) as well as biogeochemical effects of soil NO_x changes (Fig. 6j).

Ozone changes in response to vegetation changes with dynamic meteorology (Fig. 6d, g) are much higher than those with prescribed meteorology (Fig. 5d, g) as vegetation changes could modify boundary-layer meteorology, shift circulation patterns and moisture flows, and thus shape ozone concentrations. In contrast to the clear, localized signals in ozone changes through the biogeochemical pathways, both local and remote surface ozone changes are found when biogeophysical pathways are involved (Wang et al., 2020). For example, changes in biogenic VOC emission with dynamic meteorology correlate with air temperature changes (Fig. S3, S4) apart from local vegetation changes. Changes in dry deposition also correlate to meteorological changes; stomatal resistance can respond to atmospheric dryness and soil water stress (Fig. S3, S4). Ozone changes in response to soil NO_x changes with dynamic meteorology (Fig. 6j) are within the same magnitude as those with prescribed meteorology (Fig. 5j), as soil NO_x emissions only change photochemical production of surface ozone, but do not affect biogenic VOC emission and ozone dry deposition directly (Fig.5 k, l) or via meteorological changes indirectly (Fig.6 k, l).

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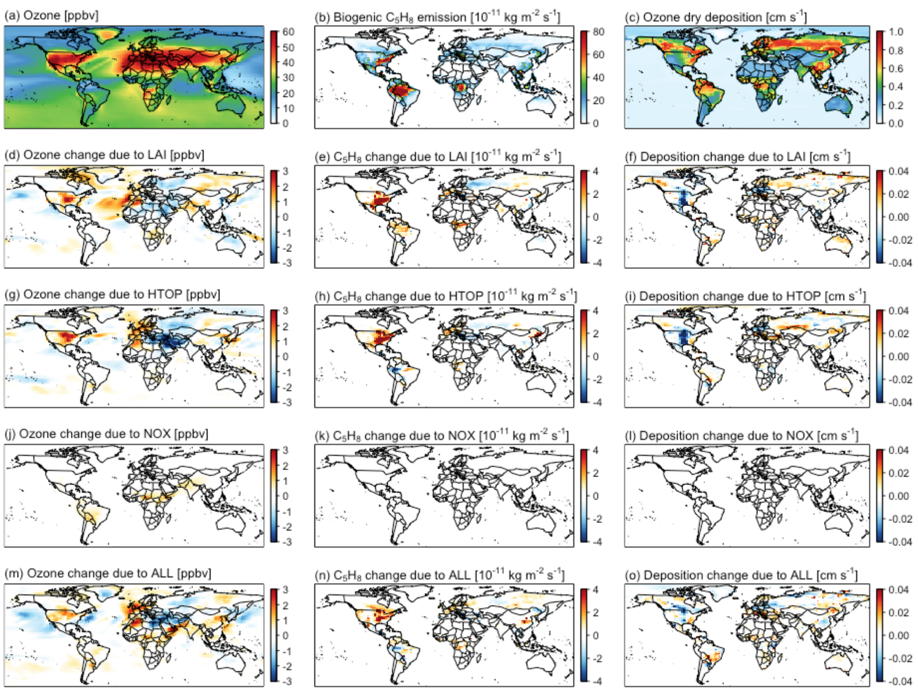

Figure 6. Same as Fig. 5 but with dynamic meteorology.

The greatest vegetation enhancements in response to future nitrogen deposition in this study are found over tropical savannas and grasslands, which are less capable of affecting local and pan-regional climate than forests, and our forest structural changes are only mild. Therefore, here we choose the US, which shows obvious ozone enhancement following vegetation changes, as an example to illustrate the biogeophysical effects further. Figure 7 shows that in the forest regions in the eastern US where

LAI and canopy height changes are relatively large following higher nitrogen deposition, albedo decreases, absorbed radiation increases, latent heat flux increases, and such changes appear to have shifted the surface energy balance and circulation patterns in a way that enhances moisture convergence, precipitation and soil moisture in the originally wetter places (i.e., the forested eastern US), but reduces the moisture convergence in the originally drier places (i.e., the grassland regions in the central US). This constitutes a feedback loop in these grassland regions that reduces transpiration, increases temperature, increases aridity and thus the plant stomata close more, all leading to the relatively large enhancements in surface ozone there. Our mild vegetation changes only have modest local impacts in places with dense vegetation to begin with (e.g., the eastern US). We found that vegetation changes shift the circulation patterns and moisture convergence such that it is the adjacent places that are the most affected, which was also found by Wang et al. (2020), who found obvious temperature increases in the central US after reforestation in the eastern US under RCP4.5 land use and land cover change. High temperature and reduced stomatal conductance in the central US further cause reduced ozone deposition (Fig. 6f), while increased temperature and LAI in the eastern US enhances biogenic emissions, both of which increase surface ozone in the central-eastern US (Fig. 6d).

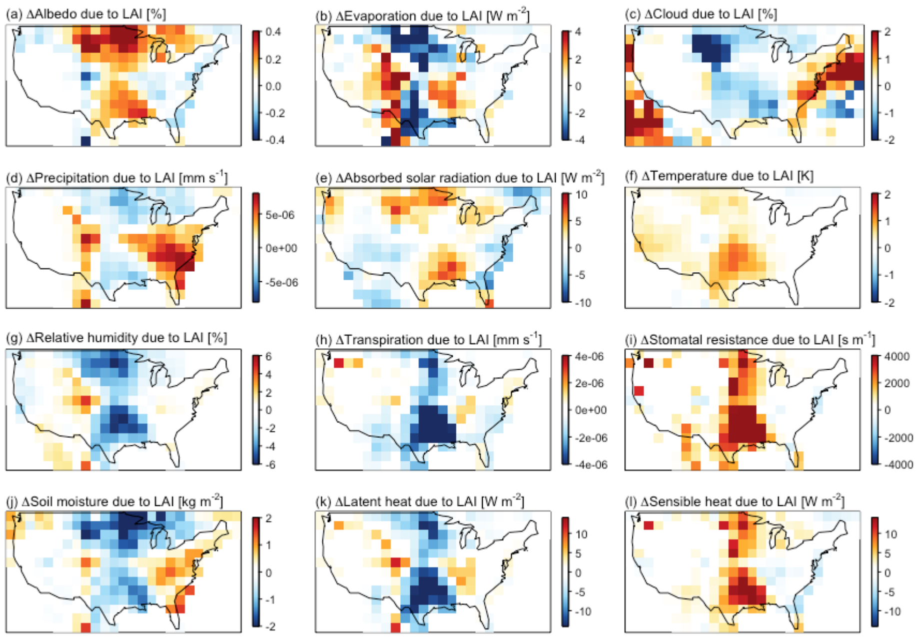

Figure 7. Summertime changes in **(a)** albedo, **(b)** ground evaporation, **(c)** cloud cover, **(d)** precipitation, **(e)** absorbed solar radiation, **(f)** surface temperature, **(g)** relative humidity, **(h)** vegetation transpiration, **(i)** stomatal resistance, **(j)** soil moisture, **(k)** latent heat flux, and **(l)** sensible heat flux driven by LAI increase with dynamic meteorology.

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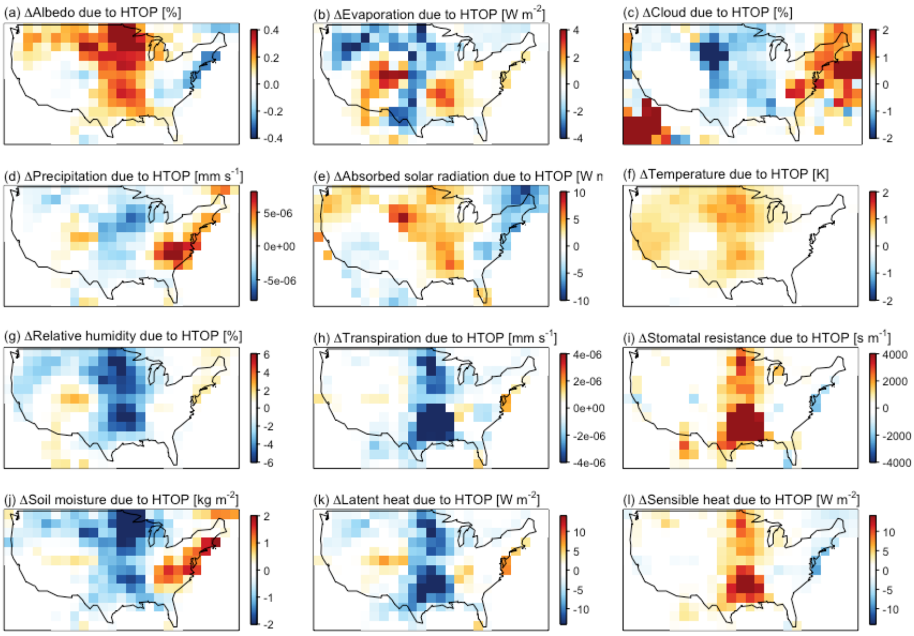

Figure 8. Same as Fig.7 but driven by canopy height increase.

Changes in canopy height show similar trends in modulating meteorological conditions (Fig. 8). The effects of meteorological variations induced by vegetation changes can be as important as or even more important than the direct biogeochemical effects of vegetation structural changes per se in terms of modulating surface ozone, and are of similar magnitude to the biogeochemical effects of soil NO_x changes. We note specifically that temperature changes resulted from vegetation-meteorology coupling are more important than LAI changes per se in regulating biogenic isoprene emission, especially in regions where obvious warming or cooling occurs. It is noteworthy that unlike with prescribed meteorology, individual effects may not add up linearly with dynamic meteorology for a given location due to the complex and far-reaching changes in atmospheric circulation and the associated cascade of local and nonlocal changes in climate that are dynamically simulated following terrestrial changes.

6 Conclusions

With the rising food need for the future world population, more intense agricultural activities are expected to cause substantial perturbations to the global nitrogen cycle, aggravating surface air pollution and imposing stress on terrestrial ecosystems. Much less studied, however, is how the ecosystem changes induced by agricultural nitrogen deposition may modify biosphere-atmosphere exchange and further exert secondary effects on global air quality. In this paper we present a study to quantify the response of surface ozone air quality to vegetation structural (LAI and canopy height) and soil NO_x emission changes under year-2000 vs. year-2050 agricultural ammonia emissions over centurial timescales by using an asynchronously coupled framework.

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unlike prescribed meteorology, individual effects may not add up with dynamic meteorology. The meteorology and climate are actively simulated following terrestrial changes, and thus will lead to a cascade of changes in local and regional climate. For instance, it's possible that moisture transport from the Gulf of Mexico can be strengthened when the moderate temperature increases are combined into a larger increase in central US, such that regional temperature increases can be alleviated and thus ozone deposition and concentrations are lower that individual effect (Fig. 6 and Fig. S3–S5). Wang et al. (2020) also find greater moisture convergence from the Gulf of Mexico into central US following temperature increases in central US under future land use/cover changes.

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Agricultural ammonia emission in the coming decades is destined to increase. We estimated year-2050 NH_3 emission to be 71 Tg N yr^{-1}, a 34% increase compared to year-2000 emission. Our estimate is comparable to 71 Tg N yr^{-1} made by RCP8.5 as both studies assumed a business-as-usual scenario where future NUE in agroecosystems is not expected to be improved much. However, it should be acknowledged that increases in food production may also be obtained with a less-than-proportionate increase in fertilizer use as countries are developing greater awareness of agriculture-related environmental impacts, and adopting more efficient nutrient use practices in the coming decades. Gu et al. (2015) reported that reasonable changes in diet, NUE, and N recycling could reduce year-2050 N losses and anthropogenic reactive nitrogen creation to 52% and 64% of 2010 levels, respectively, in China. Fung et al. (2019) showed that the maize-soybean intercropping improves NUE by easing fertilizer application and NH_3 volatilization in agricultural soils in China. Therefore, we acknowledge that the future paths of agricultural NH_3 emission and nitrogen deposition may differ from what we projected as a worst-case scenario in this study, but we do not expect the nature of the mechanisms and conclusions in this study to be altered significantly.

Atmospheric nitrogen deposition increases carbon uptake by terrestrial biosphere in nitrogen-limited areas, and also stimulates release of NO_x, nitrous oxide (N_2O) and NH_3 from soils (Reis et al., 2009; Zaehle et al., 2011). We found that nitrogen deposition increases by 20% from year 2000 to 2050 due to rising agricultural NH_3 emission, and this enhances global GPP by 2.1 Pg C yr^{-1}. LAI was simulated to be higher by up to 0.3–0.4 m^2 m^{-2} in tropical grasslands and croplands, and 0.1–0.2 m^2 m^{-2} in midlatitude boreal and temperate forests. Canopy height increases were found in boreal and temperate forests (by 0.1–0.4 m), as well as in tropical grasslands and croplands (by ~0.1 m). Soil NO_x emission budget rises to 8.7 Tg N yr^{-1} with year-2050 nitrogen deposition because of intensive nitrification and denitrification processes. Due to decreasing trends of anthropogenic NO_x emission throughout this century (IPCC, 2013), soil NO_x is expected to play an increasingly important role in global NO_x budget. Therefore, the inclusion of effects of soil NO_x emission to surface ozone is essential. These estimates are based on carbon and nitrogen interactions in CLM4.5 biogeochemistry (CLM4.5-BGC), which are widely used in estimating long-term trajectory of terrestrial variations (Lombardozzi et al., 2012; Val Martin et al., 2014; Sadiq et al., 2017; Zhou et al., 2018). However, the internal soil nitrogen cycle, its coupling with the atmosphere and reactive nitrogen gas emissions other than N_2O are not fully represented in default CLM4.5-BGC. The soil NO_x emission module that we added, which allows soil NO_x to respond to nitrogen deposition from the atmosphere, partly improved the representation (Fung et al., 2021), but the NH_3 emission we used was still based on inventories and scaling with future crop production and thus did not respond to nitrogen deposition. We expect, however, that the secondary effect of nitrogen deposition on NH_3 should be much smaller than any perturbations due to agricultural changes (Fung et al., 2021). Moreover, fully coupled bidirectional nitrogen fluxes were not enabled in our model setting. Future work is needed to examine the overall downstream biogeochemical and biogeophysical effects in an Earth system model with a closed nitrogen cycle where soil NO_x and NH_3 emissions to the atmosphere and nitrogen deposition from the atmosphere are fully coupled dynamically.

With only the biogeochemical effects of nitrogen-induced terrestrial changes (with prescribed meteorology where meteorological changes are not included), surface ozone is elevated by 1–3 ppbv in certain low-NO_x equatorial regions due to increased soil NO_x emission, while LAI and canopy height only modulate surface ozone by ±0.5 and 0.2 ppbv, respectively. With both the biogeochemical and biogeophysical effects under dynamic meteorology, changes in summertime surface ozone are within ±2–3 ppbv. Ozone responses due to vegetation changes are much higher with dynamic meteorology than prescribed meteorology, as vegetation changes shift surface energy balance, circulation patterns, moisture flow, and thus shape ozone concentrations. Local meteorological variations induced by vegetation structural changes are generally more important than the vegetation changes per se in terms of modulating surface ozone concentration, and appear to be as important as biogeochemical soil NO_x effect. Furthermore, biogeophysical pathways related to canopy height changes have not been accounted for by most previous studies of ozone-vegetation interactions, which usually only considered LAI and other ecophysiological changes (Wang et al, 2020; Wong et al., 2018; Zhao et al., 2017; Fu et al., 2015). Global vegetation growth is altered by land use and land cover change, warming, CO_2 fertilization, nitrogen deposition and ozone damage, etc., but the associated canopy height changes have usually been ignored, rendering an incomplete representation of terrestrial effects on surface air quality predictions. Here, we found that the effects of canopy height changes on surface ozone through the biogeophysical pathways are noticeable and can be as much as the effects associated with LAI changes alone.

One limitation of this study is that we did not consider ozone damage on stomatal conductance and photosynthesis as in the study by Sadiq et al. (2017). If ozone damage on stomatal conductance is considered, higher ozone concentrations could have positive feedbacks on ozone itself via reduced dry deposition and enhanced isoprene emission. Meanwhile, ozone damage on plant productivity may also diminish the fertilization effect of nitrogen and foliar nitrogen content, which is itself vital for photosynthetic capacity (Franz and Zaehle, 2021). Therefore, if ozone damage is considered, lower LAI and canopy height are expected, compensating some of the enhanced LAI and canopy height induced by higher nitrogen deposition found in this study. These changes in LAI and canopy height could further affect ozone via various biogeochemical and biogeophysical pathways, but such a secondary feedback effect is expected to be relatively minor (Zhou et al., 2018). More work is warranted to investigate the individual and combined effects of nitrogen deposition and ozone damage on plant growth and terrestrial carbon uptake, especially in light of the possible nonlinear interactions between ozone and nitrogen in plants (e.g., Shang et al., 2021).

Overall, our study demonstrates a novel linkage between agricultural activities and ozone air quality via the modulation of vegetation and soil biogeochemistry by nitrogen deposition, and highlights the particular importance of considering meteorological changes following vegetation structural changes including those in canopy height, as well as soil NO_x changes, in studying the effects of ozone-nitrogen-vegetation interactions in the future.

Data availability

Model output data used for analysis and plotting can be made available in RData format by contacting the corresponding author (Amos P. K. Tai: amostai@cuhk.edu.hk).

Author contributions

A.P.K.T. devised the overall methodology and supervised the writing of the manuscript. X.L. conducted model simulation, analyzed results and drafted the manuscript. K.M.F. implemented soil NO_x and NH_3 emission in the model.

Competing interests

The authors declare that they have no conflict of interest.

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