

Responses to the Review 1

Comments:

The authors have rerun the simulations and extended them over several decades, which should indeed be enough to obtain statistically significant differences. The model description is also improved, although the references to the DNDC source code are problematic because the code is not publicly available.

The longer simulations seem to have resolved my biggest concern (statistical significance) in the initial manuscript. There are still some puzzling results which require attention. Unless there is a simple explanation which I'm missing, addressing these issues may still require more revisions.

Reponses:

We again thank the reviewer's constructive feedback. We have revised the manuscript to address the reviewer's comments/suggestions as follows. The revised manuscript (with changes highlighted) is attached.

Comments:

First, it is unclear to me how the runs with aerosol-radiation (apparently also aerosol-cloud) interactions disabled are set up. I understand that in the “interactive” runs, the prognostic aerosol fields are used in the radiative transfer and cloud microphysical calculations. What aerosol distributions are used in the two non-interactive (NDEP) runs? If these are somehow prescribed, then how is it ensured that the differences attributed to the aerosol-climate interactions are really due feedback effects and not just because the prescribed forcing is different from the prognostic aerosol forcing? It would be helpful if the authors would formulate their hypotheses regarding the climate effects more explicitly and then explain how the model experiments test these hypotheses.

Responses:

We thank the reviewer for the opportunity to further explain our experimental design. In our non-interactive runs, the aerosol fields are prescribed. Hence, when comparing with the interactive cases, the differences in radiative budgets would include the effects of both the N deposition, the corresponding feedbacks, and the inherent spatial differences between the prescribed and prognostic aerosols for radiative transfer calculations. We note that the prescribed-prognostic differences for [CAM4_CLM5_NDEP_2050] and [CAM4_CLM5_2050] are substantial for sulfate (up to 4% in zonal mean mass ratio; mostly inland) and dust (up to 30% zonally) and rather negligible for organic carbon, black carbon, and sea salt. Since intense loading of dusts are mostly found in the desert regions, we would be able to characterize the impacts induced by these two kinds of aerosols.

We now provided more details about our configurations and experimental design in the second last paragraph of **Section 2.3**:

“**Table 1** provides configuration details of our experiments. [CAM4_CLM5_2000] and [CAM4_CLM5_2050] encapsulated the full functionality of our implementation, i.e., CAM4-chem receives the online CLM5 NH₃ emission rates as input to predict atmospheric NH₃ concentration, the subsequent formation of secondary ammonium aerosols (modeled as changes in sulfate aerosols in the model), and the corresponding instantaneous sulfate aerosol radiative effect, whilst CLM5 obtains the online CAM4-chem dry and wet deposition rates of NH_y and NO_x to calculate the addition of soil NH₄⁺ via deposition. The deposited nitrogen will eventually enrich soil fertility and fuel the re-emission of soil NH₃ while the aforementioned aerosol radiative effect can cool the Earth’s surface and suppress NH₃ volatilization. **The [CAM4 CLM5 NDEP] cases were set to isolate the impact of NH_y deposition on NH₃ emission and crop growth. In this setup, CAM4-chem used prescribed gases (except for water vapor) and aerosols in the radiation transfer calculation (i.e., aerosol-radiation interaction is disabled). Hence, any changes in the atmospheric sulfate aerosol loading induced by the addition/reduction of NH₃ would not affect radiative transfer. We note that the differences in radiative budget between the [CAM4 CLM5 NDEP] and other configurations with aerosol-radiation interaction enabled would include the effects attributable to both NH₃-induced sulfate changes as well as the differences in spatial distribution between the prescribed and prognostic aerosols. For instance, the differences**

between [CAM4 CLM5 NDEP] and [CAM4 CLM5] are substantial for sulfate (up to 4% in zonal mean mass ratio; mostly inland) and dust (up to 30% zonally; mostly in Sub-Saharan Africa and other desert regions), and are unlikely related to NH₃ changes; meanwhile, the differences are rather negligible for organic carbon, black carbon, and sea salt. This configuration was intended to isolate the enhanced fertilization effect of N deposition. Similarly, [CAM4_CLM5_CLIM] cases were prescribed with constant nitrogen deposition fluxes so that we could quantify the impacts of the changes in instantaneous aerosol radiative effects. We hypothesized that an increased NH₃ emission would promote the formation of sulfate aerosols, and the subsequent aerosol cooling effect would be observed in this setup. Finally, we further evaluated the impacts of intensive fertilizer use to promote agricultural production in the future as projected by FAO (2007) by repeating the first three simulations with fertilization at present-day (2000; model default) and future (2050; assuming 30% more synthetic fertilizers while manure fertilizer is kept at 2000-level) rates. We note that future increases in agricultural production might also involve cropland expansion, but such practice was not included in this study.”

Comments:

Second, the radiative forcing due to NH₃ emissions has been thought to be small, negative and mainly due to nitrates (see e.g., IPCC AR5 or AR6). But the differences between the CAM4_CLM5_2050 and CAM4_CLM5_NDEP_2050 (runs with and without aerosol feedbacks) in Figs. S9 and S11 are large, with about 1 K change in surface temperature and down to 20 W m⁻² difference in net surface radiation over large areas. Even more surprisingly, the differences occur over areas like the Sub-Saharan Africa where the simulated NH₃ emissions seem quite small. The areas affected partly coincide with a large change in the sulfate concentration, but the increase of sulfates over Africa seems to coincide with a higher downward radiation flux and higher surface temperature. Wouldn't we expect a cooling effect from the sulfates? What is the mechanism here? I don't remember ever seeing such a big effect attributed emissions of ammonia.

Responses:

We would like to clarify that

- 1) **Figure S11** is illustrating the local annual mean of the radiative forcing at the surface level for the N deposition and for the aerosol-radiation interaction, both measured by local changes in net (SW + LW) downward radiation at the model surface level, over pixels with croplands. These metrics are not directly comparable to the radiative forcing reported in, e.g., IPCC AR5 or AR6, which is usually the difference in area-weighted global mean (direct) radiative effects of aerosols (measured as net downward SW + LW radiation) at the top of atmosphere, under present-day and preindustrial emissions.

For comparison, the global (over all land types and oceans) annual mean radiative forcing induced by the increased fertilization, i.e., [CAM4_CLM5_2050] vs [CAM4_CLM5_2000], is -0.005 W m^{-2} .

- 2) The large differences in downward radiation over the Sub-Saharan Africa in **Figure S11(b)** are largely due to the differences between the prescribed and prognostic dusts, rather than the reduced emission of NH₃. The more negative values (greener) in **Figure S11(b)** indeed indicate stronger cooling effects, where coincide in the lower surface temperature (greener) in **Figure S9(b)**. The cooler condition over the Sub-Saharan Africa from 3) has resulted in reduced NH₃ emission (greener) in **Figure 7(b)** as well as less sulfate aerosols (greener) in **Figure S12(b)**.

To make these points clearer in the manuscript, we added these descriptions and results as follows:

In the last paragraph of **Section 3.3**:

“Comparing the 2000- and 2050-fertilization levels, our fully coupled simulation estimated a -0.005 W m^{-2} of global downward radiative flux (i.e., cooling), which is

virtually negligible compared to the 16-model mean total anthropogenic aerosol radiative forcing of -0.27 W m^{-2} reported in Myhre et al. (2013).

In the last paragraph of **Section 3.3**:

“Though the global impact is also negligible ($+0.004 \text{ W m}^{-2}$), Δ_{clim} reveals a substantial regional cooling at the surface level (**Figure S11(b)**) largely because there are more prognostic dusts in [CAM4_CLM5_2050] over Sub-Saharan Africa than prescribed in [CAM4_CLM5_NDEP_2050]. Such cooling effect results in lower surface temperature (**Figure S9(b)**) and also suppressing the formation of particulate sulfate (**Figure S12(b)**).”

We also provided more details in the second last paragraph of **Section 2.3**, which helps clarify what the differences in sulfate and dust between the simulations actually entail. See our revisions and responses above.

Comments:

The differences caused by the nitrogen deposition (CAM4_CLM5_2050 vs CAM4_CLM5_CLIM_2050) seem smaller, as expected. But even here, some of the regional differences seem large. For example, in Fig. 6, CAM4_CLM5_CLIM_2050 shows only 2 % increase in grain production for Europe, while CAM4_CLM5_2050 with interactive N deposition has a 17 % increase, as if the effect of deposition was greater than the fertilization itself. Is this difference realistic and statistically robust?

Responses:

The changes concerned were checked and found to be insignificant, and were removed from our discussion; these did not affect the major conclusions of our paper.

Comments:

Specific comments (line numbers refer to the track changes version)

Eq. 1: It's still unclear how $F(\text{soil, pot})$ is converted to a flux (per unit of time). If Eq. (1) gives the maximum flux per timestep, then what happens if the length of timestep is changed?

Responses:

This equation was originally developed in DNDC to estimate the potential amount of soil NH_3 to be emitted per day. In this study, we converted the unit of this daily value from per day to per second, so that we can get the total amount of NH_3 emitted in each time step (30 min in this study). Then, this daily value is recalculated in the next time step. A longer timestep would reduce the frequency of such recalculation.

Comments:

Eq. 2 is actually OK, I misread it on the first go.

Responses:

Thank you for your feedback.

Comments:

Eq. 7: Is l_{\max} 42 m (per Table S1)? We know that even relatively shallow (5-10 cm) incorporation of fertilizers reduces the NH_3 emissions by 50 % or more, so it could be expected that the depth-dependent factor would be close to zero for all but the top 5-10 layers or so.

Responses:

Yes, the reviewer is correct, l_{\max} corresponds to the bottom layer of soil. NH_3 emission from the deeper layers contributes very little to the total column emission.

Comments:

L318: Hoesly et al. (2018) state that “CEDS data do not include open burning, e.g., forest and grassland fires, and agricultural waste burning on fields, which was developed by van Marle et al. (2017).” These are important sources of NH₃. Please check.

Responses:

We have revised the sentence to clarify that biomass burning in our model are prescribed using the emission inventories described by von Marle et al. The updated sentence now reads:

“CAM4-chem employs a bulk aerosol approach and predicts the formation of PM_{2.5} components including SO₄²⁻, NO₃⁻, and NH₄⁺, where the injection rates of precursors – sulfur dioxide (SO₂), NO_x, and NH₃ – are prescribed by the Coupled Model Intercomparison Project phase 6 (CMIP6)/Community Emissions Data System (CEDS) emission inventory (CMIP6 hereinafter) for anthropogenic activities (Hoesly et al., 2018). **The biomass burning emissions used for our simulations are described by von Marle et al. (2016, 2017) and are all assumed as surface emissions without plume-rise nor predefined vertical distribution.**”

Comments:

L346: I actually do not find a detailed description of the radiative transfer in CAM4 in Lamarque et al. (2012) or in Collins et al. (2006). However, Lamarque et al. states that no cloud-aerosol interaction is available in CAM4, which appears to contradict with what is stated here. A better description of the simulated aerosol-radiation and aerosol-cloud effects is needed, since they are critical for some of the model experiments.

Responses:

We thank the reviewer for noting that our description of the CAM4 radiate transfer was not fully correct. We updated the description in the 3rd paragraph in **Section 2.3** and provided a more appropriate reference:

“In the default configuration, atmospheric chemistry interacts with the climate solely through radiation in CAM4-chem (Lamarque et al., 2012). Furthermore, atmospheric reactive nitrogen (NH₄⁺ or NO₃⁻) does not directly interact with radiative transfer in the model. Instead, its radiative implications are manifested via altering the gas-aqueous partitioning of sulfate (Emmons et al., 2010; Metzger, 2002) and the subsequent changes in direct radiative effect due to any changes in sulfate aerosols. The subsequent sulfate-induced changes in cloud optical properties (indirect radiative effect) were not considered in this work. Detailed description of the radiative transfer processes in CAM4-chem is provided in Neale et al. (2010).”

Neale et al (2010) Description of the NCAR Community Atmosphere Model (CAM 4.0), https://www.cesm.ucar.edu/models/ccsm4.0/cam/docs/description/cam4_desc.pdf

We also corrected the description in the last paragraph of **Section 3.3**:

“On the other hand, we expected the **sulfate aerosols** induced by agricultural NH₃, which directly increases aerosol albedo ~~or enhanced cloud brightness and influences cloud lifetime (Twomey, 1977; Albrecht, 1989)~~, would reduce the amount of insolation reaching the Earth’s surface.”

Comments:

L618: So why is it warmer in the 2050 fertilization scenario?

Responses:

After considering the reviewer's comments and reexamining the results, we found that the Asian warmer surface temperature and grain production increases are not uniformly statistically significant and decided that temperature may not be the important factor. We hence removed the concerning discussion from the main text.

Comments:

L647: The overall aerosol radiative forcing is thought to be negative. It seems that the best estimate for total aerosol effect in AR5 (p. 662) was -0.9 W m^{-2} .

Responses:

We thank the reviewer for rising this point. As indicated above we added the following discussion:

“Comparing the 2000- and 2050-fertilization levels, our fully coupled simulation estimated a -0.005 W m^{-2} in global downward radiative flux (i.e., cooling), which is virtually neglectable compared to **the 16-model mean total anthropogenic aerosol radiative forcing of -0.27 W m^{-2} reported in Myhre et al. (2013).**”

Responses to Reviewer 2

Comments:

I found the revised paper improved over the previous version, but still have some concerns. The introduction reads quite well and except for a few minor points looks good. The model description also well. The description of the model simulations and results were much clearer.

All line numbers refer to the version with author tracked comments.

Responses:

We thank the reviewer for their constructive feedback. We have revised the manuscript to address the reviewer's comments/suggestions as below. The revised manuscript (with changes highlighted) is attached.

Comments:

Major Comments

1. The authors should carefully distinguish between fertilizer (referring to synthetic fertilizer and manure) and synthetic fertilizer. They should clearly state the difference. In some places it was rather confusing as to what the authors were referring. Instances are referred to below. This should be easily remedied.

It appears that Fig. 2 is probably only from synthetic fertilizers, yet the caption says “fertilizer induced” which would imply manure also? The numbers from MESSAGE look like they are from synthetic fertilizer, but it is not clear how one obtains synthetic fertilizer emissions from EDGAR and CMIP6. To my knowledge those latter inventories give agricultural soil manure emissions which include manure, fertilizer and grazing emissions. To what extent does the added manure in the present manuscript represent the grazing component?

Responses:

As suggested by the reviewer, we have clarified throughout the manuscript what type of fertilizer our study refers to.

In **Section 3.1**, 1st paragraph:

“We extracted the monthly fertilizer-induced NH₃ emission estimates from MASAGE, and assumed that one-third of the total agricultural NH₃ emission reported by CMIP6 and EDGAR are **associated with synthetic fertilizer**, which is consistent with the apportionment reported in previous studies and environmental reports (Paulot et al., 2014; Riddick et al., 2016; National Oceanic and Atmospheric Administration, 2000; European Environment Agency, 2010; Gu et al., 2012; Paulot et al., 2015; Zheng et al., 2017).”

In **Section 3.1**, 3rd paragraph:

“Our estimate is higher than all three inventories of NH₃ emissions associated with synthetic fertilizers, which are 10 Tg-N yr⁻¹ for CMIP6 and EDGAR, and 9.1 Tg-N yr⁻¹ for MASAGE.”

Caption of **Figure 2**:

“Fertilizer-induced NH₃ emission estimated by CLM5 (**synthetic and manure**) and other emission inventories (**synthetic only**).”

We also wanted to note that NH₃ emission due to grazing is not considered as the emission associated with manure applications in this study, and clarified this in the text too:.

In **Section 2.3**, 1st paragraph:

“...for our online simulated emission rates from CLM5. **This study did not consider manure spreading on pastures and grazing animals.** Atmospheric NH₃ and NH₄⁺ formed sequentially return...”

Comments:

2. Using a constant manure fertilization rate is somewhat surprising as there are global distributions of manure application (e.g., Zhang et al., 2017). The rate incorporated (2 g-N s^{-1}) was presumably used in the CLM so crops would grow even in regions where synthetic fertilizer was lacking. This rate does not seem appropriate to represent the regional distribution of manure fertilizer for emissions. Presumably using a constant rate implies that crops in regions largely fertilized largely with synthetic fertilizer the added nitrogen is much higher than reality? Thus the detailed comparison between the various inventories and CAM4_CLM5_2000 is difficult to interpret. Is this due to the emission scheme or due to a simplistic estimate of applied manure? You might compare the manure N rates used here to those from an established inventory. The units here don't make sense for added manure don't makes sense: 2 g-N s^{-1} .

Responses:

Thank you for pointing this out, and we have corrected the unit as $\text{g-N m}^{-2} \text{ yr}^{-1}$. Now the relevant sentence in **Section 2.1**, 2nd paragraph, reads:

“Manure fertilizer application rate is assumed constant for all crops at $2 \text{ g-N m}^{-2} \text{ yr}^{-1}$, same as the model default (Lombardozzi et al., 2020).”

This application rate is a default value of CLM5 (Lombardozzi et al., 2020). Though we agree a more realistic development of manure application would be a big improvement, expanding the capacity of CLM5 to include spatial and temporal varying manure application rates is beyond the scope of this study.

Comments:

3. It is difficult to understand how the authors set up the sensitivity simulations. It is very important to clearly define the simulation one is assessing the sensitivity against so as to interpret the results.

For the deposition rates the paper states: “The deposition rates were prescribed in the default configuration and dynamically computed by CAM4-chem in our version.” (1199-1201). It appears then in both CAM4_CLM5_CLIM_2000 and CAM4_CLM5_CLIM_2050 the deposition rates are the same and taken from the CAM4_CLM5 assuming 2000- level fertilization (Table 1), where I take it that CAM4_CLM5 has fixed deposition. Since the lifetime of NH₃ and NH₄⁺ in the atmosphere is short the deposition should be equal to the emissions on an annual basis. So by comparing the depositions in these model runs the authors are essentially comparing the emissions in the interactive model (CAM4_CLM5_CLIM) and the implied emissions in CAM4_CLM5. This comparison does not seem to get at the importance of interactive emissions, but at the difference between the different emissions, which would not be expected to be the same. A quantification of the effect of interactive emissions in the CAM4_CLM5_CLIM simulations have on the emissions themselves (through deposition) seems difficult. At any rate from the explanations in the paper I don't see how it was done.

Likewise for aerosols it appears that the “NH₃-induced aerosols would be inactive” (1578). This seems to imply that this aspect is just examining the importance of a simulation with NH₃-induced aerosols to one without. The differences, then, between these simulations have little to do with interactive emissions, but with including ammonia emissions at all. This could have been done, just as well, with fixed emission inventories. Moreover, it is unclear what is assumed with regard to sulfate aerosols – I assume it is only the ammonium nitrate aerosols that are impacted.

Responses:

To make the explanation of our experimental design clearer, we expanded the description of our modeling experimental design in Table 1. For that, we moved an introductory paragraph from **Section 3.3** to **Section 2.3** to explain our experimental designs earlier in the paper. We also rewrote the paragraph to provide more design details, as also suggested by Reviewer 1.

The second last paragraph in **Section 2.3** now reads:

“Table 1 provides configuration details of our experiments. [CAM4 CLM5 2000] and [CAM4 CLM5 2050] encapsulated the full functionality of our implementation, i.e., CAM4-chem receives the online CLM5 NH₃ emission rates as input to predict atmospheric NH₃ concentration, the subsequent formation of secondary ammonium aerosols (modeled as changes in sulfate aerosols in the model), and the corresponding

instantaneous sulfate aerosol radiative effect, whilst CLM5 obtains the online CAM4-chem dry and wet deposition rates of NH_y and NO_x to calculate the addition of soil NH_4^+ via deposition. The deposited nitrogen will eventually enrich soil fertility and fuel the re-emission of soil NH_3 while the aforementioned aerosol radiative effect can cool the Earth's surface and suppress NH_3 volatilization. The [CAM4 CLM5 NDEP] cases were set to isolate the impact of NH_y deposition on NH_3 emission and crop growth. In this setup, CAM4-chem used prescribed gases (except for water vapor) and aerosols in the radiation transfer calculation (i.e., aerosol-radiation interaction is disabled). Hence, any changes in the atmospheric sulfate aerosol loading induced by the addition/reduction of NH_3 would not affect radiative transfer. We note that the differences in radiative budget between the [CAM4 CLM5 NDEP] and other configurations with aerosol-radiation interaction enabled would include the effects attributable to both NH_3 -induced sulfate changes as well as the differences in spatial distribution between the prescribed and prognostic aerosols. For instance, the differences between [CAM4 CLM5 NDEP] and [CAM4 CLM5] are substantial for sulfate (up to 4% in zonal mean mass ratio; mostly inland) and dust (up to 30% zonally; mostly in Sub-Saharan Africa and other desert regions), and are unlikely related to NH_3 changes; meanwhile, the differences are rather negligible for organic carbon, black carbon, and sea salt. This configuration was intended to isolate the enhanced fertilization effect of N deposition. Similarly, [CAM4 CLM5 CLIM] cases were prescribed with constant nitrogen deposition fluxes so that we could quantify the impacts of the changes in instantaneous aerosol radiative effects. We hypothesized that an increased NH_3 emission would promote the formation of sulfate aerosols, and the subsequent aerosol cooling effect would be observed in this setup. Finally, we further evaluated the impacts of intensive fertilizer use to promote agricultural production in the future as projected by FAO (2007) by repeating the first three simulations with fertilization at present-day (2000; model default) and future (2050; assuming 30% more synthetic fertilizers while manure fertilizer is kept at 2000-level) rates. We note that future increases in agricultural production might also involve cropland expansion, but such practice was not included in this study.”

We also provided more information about how NH_3 emission would affect aerosols and its radiative effects in the third paragraph in **Section 2.3**:

“In the default configuration, atmospheric chemistry interacts with the climate solely through radiation in CAM4-chem (Lamarque et al., 2012). Furthermore, atmospheric reactive nitrogen (NH_4^+ or NO_3^-) does not directly interact with radiative transfer in the model. Instead, its radiative implications are manifested via altering the gas-aqueous partitioning of sulfate (Emmons et al., 2010; Metzger, 2002) and the subsequent changes in direct radiative effect due to any changes in sulfate aerosols. The subsequent sulfate-induced changes in cloud optical properties (indirect radiative effect) were not considered in this work. Detailed description of the radiative transfer processes in CAM4-chem is provided in Neale et al. (2010).”

Comments:

Minor points.

-130 “disrupts” – not sure what this means in the current context

Reponses:

We replaced “disrupts” to “influences”. Now the sentence reads:

“Interactively coupling terrestrial NH₃ emissions to atmospheric chemistry simulations by the Community Atmospheric Model version 4 with chemistry (CAM4-chem), we found that such emissions favor the formation and deposition of NH₄⁺ aerosol, which in turn **influences** the aerosol radiative effect and enhances soil NH₃ volatilization in regions downwind of fertilized croplands.”

Comments:

-197-99 “near real time high-resolution maps of atmospheric NH₃”. The maps shown in Van Damme et al., 2018 are time averaged over a rather long periods. Are near real time maps of satellite NH₃ really accurate?

Reponses:

We agree with the reviewer’s comment and have deleted “near real-time”. The revised sentence now reads:

“It enables the **creation of high-resolution maps** of atmospheric NH₃ and the possibility of pinpointing industrial and agricultural emission hotspots with diameters smaller than 50 km (Van Damme et al., 2018).”

Comments:

-1108 “agricultural emissions”. It would be good to define these precisely. As used does this term include emissions from both manure management (mostly from barns and storage) and from agricultural soils (which includes grazing animals, manure spreading onto pastures and other cropland and synthetic fertilizer emissions)?

Reponses:

We expanded the sentence to provide more information regarding the “agricultural emissions” here.

“Recent inventories adjusted the estimated agricultural emissions (**including manure management, and both synthetic and manure fertilizers**) in 2000–2010 to 33–37 Tg-N yr⁻¹ (Sutton et al., 2013; Janssens-Maenhout et al., 2015; Hoesly et al., 2018).”

Comments:

-I139-I144 Vira et al., 2020 and Vira et al. 2021 also used the CESM.

Responses:

We added the two citations as suggested:

“Many studies have employed CESM for studying processes in both the atmospheric and terrestrial nitrogen cycles, e.g., NO_x and N₂O emission (Saikawa et al., 2013, 2014; Zhao et al., 2017), deposition (Lamarque et al., 2013), denitrification and nitrate leaching (Nevison et al., 2016), crop nitrogen uptake (Levis et al., 2018), and reactive nitrogen input to ecosystem associated with synthetic and manure fertilizers (Riddick et al., 2016; **Vira et al., 2020, 2021**)”

Comments:

-1153 'DNDC' is just kind of thrown in there. Please give the abbreviation, plus maybe a short introduction to the model

Responses:

We agree with the reviewer's suggestion and have edited the sentence as:

“Comparing to other approaches, **our scheme, which borrowed from a standalone biogeochemical model, the DeNitrification-DeComposition (DNDC)**, requires variables that are mostly already modeled in CLM5, allowing us to largely capture the dynamic nature of NH₃ emission.”

Comments:

-I200 All deposition N is added as NH₄⁺. Is there a reference for this? Lawrence et al. (2020) does not seem to document this (although maybe I missed it).

Responses:

The model treatment of N deposition is explained in **Section 22.3** of Lawrence *et al.* (2018) Technical Description of version 5.0 of the Community Land Model (CLM) (https://www.cesm.ucar.edu/models/cesm2/land/CLM50_Tech_Note.pdf).

We added this citation in our manuscript:

“All added depositional and fertilizers N are added to the soil NH₄⁺ pool of each layer from ground surface to 0.4 m underground according to a model-defined soil profile (**Table S1**) (Lawrence et al., 2018).”

Comments:

-Were interactive soil NO_x emissions included in these simulations? Or were they fixed.

Responses:

Interactive soil NO_x emissions were not included as inputs for the atmospheric model in our simulations. All NO_x emissions for CAM4-chem were from emission inventories.

We added a sentence to the 1st paragraph in **Section 2.3** to clarify this:

“We note that the NO_x emission inputs for CAM4-chem were solely from the emission inventories and did not include those from our modified denitrification and nitrification schemes.”

Comments:

-1258 “ evaluated the uncertainty” to pH. Please provide a brief summary of the findings in the main part of the paper

Responses:

We added a summary there in the revise manuscript to improve clarity as suggested by the reviewer:

“Briefly, a higher pH would promote model NH₃ emission rate exponentially as the emission rate is of the order of 10^{pH}. This high sensitivity warrants the need to include crucial chemical processes in the model for accurately determining soil pH online.”

Comments:

-1260-265 this explanation should probably be included after equation (7), not before it.

Responses:

We agree with the reviewer and have rearranged the paragraph as:

“Lastly, we used this equation to calculate f_{vol} (Li et al., 1992; Gardner, 1965; source code of DNDC v9.5):

$$f_{\text{vol}} = \left(\frac{1.5s}{1 [\text{m s}^{-1}] + s} \right) \left(\frac{T_{\text{soil}}}{50 [\text{°C}] + T_{\text{soil}}} \right) \left(\frac{l_{\text{max}} - l}{l_{\text{max}}} \right) \quad (1)$$

where s (m s^{-1}) is surface wind speed; T_{soil} ($^{\circ}\text{C}$) is soil temperature; l and l_{max} (both in m) are the depth of each soil layer and the maximum depth of a soil column, respectively. **Our scheme assumes that vaporized soil NH_3 in a deeper layer diffuse upward to the surface, but does not explicitly simulate the process. Instead, it is represented in the last term in Eq. (7) as a ratio of $(l_{\text{max}} - l)/l$ for the $\text{NH}_{3(\text{g})}$ contained in each soil layer. Hence, soil NH_4^+ in deeper layers is also subject to loss to NH_3 volatilization but at much slower rates than that in the upper layers.** Details of the soil profile are provided in **Table S1**. The actual soil NH_3 to be emitted ($F_{\text{soil,act}}$; g-N m^{-3}) from each soil layer is then determined by the lower of the $F_{\text{soil,pot}}$ or the available soil NH_4^+ after competition with other processes, namely, plant uptake, microbial immobilization, and nitrification. The model distributes available soil NH_4^+ to all competing processes according to their relative demands (individual potential flux to sum of all four potential fluxes) without bias toward any process (Lawrence et al., 2019). The column-level actual soil NH_3 emission flux (F_{soil} ; $\text{g-N m}^{-2} \text{s}^{-1}$) is computed as the sum of the product of the $F_{\text{soil,act}}$ and layer thickness (m) at each layer, and assumed to emit to the atmosphere constantly over a model time step size ($\Delta t = 1800$ s in this study).”

Comments:

-I301-303 “dividing soil NH₃ emission rate by s_{10} gives an approximate in-canopy NH₃ concentration, and multiplying the latter with v_c and L produces an estimated quantity of NH₃ retained by the canopy”. This is not obvious to me – is there a reference or can you clarify?

Responses:

We appreciate the opportunity to explain this.

Consider the in-canopy volume as a rectangular box with a base area of 1 m by 1 m. Assume also that there is a constant inflow of NH₃ at F g-N m⁻² s⁻¹ from below into the box. On the other hand, some wind (at a speed s_{10} m s⁻¹ from an arbitrary direction) replaces the air inside the box with some fresh air (assuming no or very little NH₃). Dividing F by s_{10} would give us an equilibrium concentration of NH₃, say C g-N m⁻³, that still remains in the box. This C g-N m⁻³ of NH₃ is what the canopy would be exposed to. Multiplying this C g-N m⁻³ of NH₃ with the depositional velocity (v_c m s⁻¹) and the LAI (L m² m⁻²) would give us the NH₃ being captured/retained by the canopy (in g-N m⁻² s⁻¹).

Comments:

-1322 “we substituted the portion of NH₃ emission associated with synthetic fertilizer”. This is confusing. Why just synthetic fertilizer when you are considering both synthetic fertilizer and manure. And according to Hoesly et al. (2018) sectors in CEDS includes Agricultural Manure-management and Soil-emissions. I don’t see how you can subtract out synthetic fertilizer. Moreover, soil emissions usually include manure spreading on pastures and grazing animals. The latter two are not included in the author’s estimate, correct? Additional clarification is necessary here.

Responses:

Thank you for catching this. We meant to say fertilizer, and not synthetic fertilizer.

Regarding the CEDS, we made an assumption as explained in the first paragraph of **Section 3.1**:

“We ... assumed that one-third of the total agricultural NH₃ emission reported by CMIP6 and EDGAR are associated with synthetic fertilizer, which is consistent with the apportionment reported in previous studies and environmental reports (Paulot et al., 2014; Riddick et al., 2016; National Oceanic and Atmospheric Administration, 2000; European Environment Agency, 2010; Gu et al., 2012; Paulot et al., 2015; Zheng et al., 2017).”

The reviewer is correct that this study did not consider manure spreading on pastures and grazing animals.

Hence, we clarified these points in the paragraph and it now reads :

“In our coupled simulations, we substituted the portion of NH₃ emission associated with **fertilizers** from the CAM4-chem inventory input (CESD) for our online simulated emission rates from CLM5. Atmospheric NH₃ and NH₄⁺ formed sequentially return to CLM5 through deposition. **This study did not consider manure spreading on pastures and grazing animals.**”

Comments:

-1342, are the effects of atmospheric reactive nitrogen on ozone or methane included?

Reponses:

We did not assess the changes in ozone or methane, as it is beyond the scope of our study. Our speculation is that, as NH_3 reacts with atmospheric NO_2 , changes in the spatial distribution in NO_2 associated with the new NH_3 scheme might affect the ozone chemistry and likely also methane lifetime accordingly.

Comments:

-1358 “change in annual emission fluxes $\leq 10\%$ ” - does this mean the long-term trend is less than 10% or the interannual variability. A long term trend of 10% seems rather significant.

Reponses:

Here the 10% refer to interannual variability of the emission fluxes. We have revised the sentence to clarify this:

“The first 10 years of outputs were used to further stabilize the model (such that **the interannual variability of the emission fluxes could be $< \pm 10\%$**) after our ammonia scheme was implemented.”

Comments:

-1401 “one-third are fertilizer-associated”. First, please clarify what is meant by fertilizer here: synthetic fertilizer or synthetic and manure fertilizers. It might be helpful to explain the emission sectors from the inventories.

Responses:

We agree with the reviewer’s suggestion and have updated the sentence to reflect that it’s associated with synthetic fertilizers:

“We extracted the monthly fertilizer-induced NH₃ emission estimates from MASAGE, and assumed that one-third of the total agricultural NH₃ emission reported by CMIP6 and EDGAR are **associated with synthetic fertilizer**, which is consistent with the apportionment reported in previous studies and environmental reports (Paulot et al., 2014; Riddick et al., 2016; National Oceanic and Atmospheric Administration, 2000; European Environment Agency, 2010; Gu et al., 2012; Paulot et al., 2015; Zheng et al., 2017).”

Comments:

-Table 3 should clearly state where the numbers are estimated and where they are straight from the inventory. It should be possible to put some error bounds on these numbers from the literature.

Responses:

We edited the caption of **Table 3** to clarify that the simulated NH₃ are estimated in this study while other values are reported by the inventories. The caption now reads:

“Regional fertilizer-induced NH₃ emission totals estimated by our model and reported by other inventories.”

As state in the manuscript that these model-inventory comparisons are not meant to be exact but shall be considered as qualitative ones. We decided to include 1-SD's for the multi-year estimations / inventory values while maintain how other values are presented in **Table 3**:

Table 3. Regional fertilizer-induced NH₃ emission totals (Tg-N yr⁻¹) estimated by our model and reported by other inventories.

	Global	USA	Europe	India	China
CAM4_CLM5_2000^a	<u>14.2 (± 0.60)</u>	<u>2.1 (± 0.35)</u>	<u>0.6 (± 0.07)</u>	<u>2.8 (± 0.28)</u>	<u>1.2 (± 0.08)</u>
CMIP6^b	<u>10.9 (± 0.65)</u>	<u>0.9 (± 0.05)</u>	<u>0.7 (± 0.16)</u>	<u>1.9 (± 0.15)</u>	<u>2.6 (± 0.30)</u>
EDGAR^c	10.5	0.7	0.5	1.6	3.0
MASAGE	9.1	0.5	0.4	1.7	2.8

a 20-year 1-SD are shown in the brackets

b 16-year 1-SD are shown in the brackets (2000–2015)

c Variation in 2012 of both cultural statistics and emission factors ranged from 186% to 294% (Crippa et al., 2018)

Comments:

-1421 Vira et al (2020) gives the contribution from synthetic fertilizer which I think you are comparing against.

Responses:

We made our comparison with Vira et al (2020) clearer and the text reads now:

“Our estimation is close to the 12 Tg-N yr⁻¹ (from synthetic fertilizer only) and 18 Tg-N yr⁻¹ (**11 Tg-N yr⁻¹ from both synthetic fertilizer and 6.5 Tg-N yr⁻¹ from manure application**) reported by two similar studies, Riddick et al. (2016) and Vira et al. (2020), respectively.”

Comments:

-Since ammonia emissions from manure dominate over those from synthetic fertilizer, and the inventories give emissions from agricultural soils why aren't the combined manure, synthetic fertilizer emissions being compared.

Responses:

The manure NH₃ reported in the emission inventories includes both from manure fertilizer and management, but accurately disaggregating the reported emission rates is not straightforward and out of the scope of this study. Such comparison would also require further simulations with exact meteorology inputs matching with the inventories. We hence decided to present the qualitative comparison in this study, as stated in **Section 2.4** of the manuscript.

Comments:

-1480-483. This is confusing. EDGAR contains manure management.

Responses:

We meant here that the source data of EDGAR we used in this study accounted for NH₃ emission from both synthetic & manure fertilizers, while CMIP6's NH₃ emission included also manure management.

We have revised the sentence to clarify this:

“EDGAR and CMIP6 have higher background levels than MASAGE because the original estimates **used in this study** accounted for not only synthetic fertilizer but also manure application (for both) and management (for CMIP6 only)...”

Comments:

-1618 It is unclear how "the warmer temperature allows crops to reach maturity sooner, hence, shortening their grain filling periods" leading to reduced N uptake. The relation between the processes is not clear to me. It is unclear to me the extent to which this hypothesis is supported.

Responses:

After considering the reviewer's comments and reexamining the results, we found that the Asian warmer surface temperature and grain production increases are not uniformly statistically significant and decided that temperature may not be the important factor. We hence removed the concerning discussion from the main text.

Comments:

-1671-672 “These new features enabled CESM2 to perform, for the first time, a more reliable estimation of soil NH₃ emission and atmospheric NH₃ concentration than using constant emission inventory values under dynamic climate and environmental conditions.” Vira et al. (2021) also included the coupling between soil and atmosphere.

Responses:

Vira et al. (2021) was not published when this paper was written and submitted for discussion. We agree to remove the phrase “for the first time” in the sentence.

Comments:

-I706: “We did not include manure application”. I thought manure application was indeed included.

Responses:

Thank you. We have corrected the typo:

“We did not include manure **management** in our study due to the high uncertainty and data insufficiency for validation.”

Comments:

-1720 There are global pH datasets that can be used!

Responses:

Thank you for pointing this out. We are aware of some of the coarse resolution options, but we would still like to advocate for the necessary of field surveys, especially for places with low data coverage. We revised the sentence to emphasize that:

“We also note that such field surveys, **especially in underrepresented regions with low data coverage**, would also be useful to infer a soil pH map that constraints the uncertainty in simulations using a constant pH, like those reported in this study.”