

## Responses to Reviewer 2

### Comment:

*This paper presents a parameterization for evaluating ammonia (NH<sub>3</sub>) volatilization from soils within the Community Earth System Model (CESM). The authors couple both the emission and deposition fluxes between the land and atmospheric components of the CESM, and use this capability to evaluate the emission of NH<sub>3</sub> following fertilizer applications and the transport, deposition and possible re-emission of the emitted NH<sub>3</sub>. They perform further simulations to evaluate the effect of increased fertilizer usage on modeled ammonia emissions and crop harvests and the role of atmospheric feedbacks in these responses.*

*The two-way land-atmosphere exchange of ammonia has been simulated in a number of other models, although not within CESM. However, introducing the bi-directional NH<sub>3</sub> exchange into the nitrogen cycle of an Earth system model does open up new possibilities for assessing the role of atmospheric transport of NH<sub>3</sub> in the global nitrogen cascade. The coupled simulations in this manuscript present an interesting step towards this direction.*

*Thus, I find the manuscript in principle suitable for publication Biogeosciences. However, I also have several concerns related to the model formulation and the experiments, as detailed below. Addressing these will probably require major revisions before the paper can be published.*

### Response:

We thank the reviewer for their comments and suggestions to improve the manuscript. We have revised the manuscript to address the reviewer's feedback as below. The revised manuscript (with subsequent changes highlighted) and the updated **Supplementary Information** are also attached in the Supplement files.

Comment:

1. Model formulation and evaluation

*The model equations (1-9) are supposed to be derived from the DNDC model. However, except for the chemical equilibria in Eqs (3)-(6), which are fairly standard, I haven't found the equations in the DNDC literature cited. It is possible that I have missed something, since there exist multiple versions of DNDC. But if the DNDC is only a source of inspiration, then the authors should not write that their model is "derived from" the DNDC. In the current form, the model equations look like plausible but rather ad-hoc parameterizations for the volatilization process.*

Response:

As suggested also by reviewer 1 we have improved the description of the NH<sub>3</sub> volatilization scheme.

**Eq. (1) – (8)** are directly implemented from DNDC, corresponding citations are added to the manuscripts.

**Eq. (7)** is obtained from the source code of DNDCv9.5 shared by Changsheng Li with us via personal communication on Jun 18<sup>th</sup>, 2015.

**Eq. (9)** in our paper is from the Eq. (17) from the DNDC v9.5 Scientific Basis and Processes (Institute for the Study of Earth, Oceans, and Space, University of New Hampshire, 2017), with additional terms of  $b(h_{top} - h_{bot})$  to account for the height effect of the plant canopies, referring to Eq. (11) in (Pleim et al., 2013) for calculating the in-canopy aerodynamic resistance.

Comment:

*Most of the existing models for NH<sub>3</sub> volatilization and exchange have been verified with at least some field data. Here, the model is evaluated by comparing with existing inventories and by comparing the simulated NH<sub>3</sub> columns with the IASI data on yearly level. This makes sense, but as the only source of empirical evaluation, it has the issue that the geographic variation predicted by the model becomes conflated with the variation in the N input. In other words, some of the resolved variation is likely to originate in the geographic distribution of fertilizer use. Only a fraction of the global NH<sub>3</sub> emission is from synthetic fertilizers, which further weakens the signal.*

*I understand that there is no easy way around this, but it would be good to see how the model predicts the geographic distribution of the ratio between NH<sub>3</sub>-N emitted and fertilizer-N applied.*

Response:

We welcome the reviewer's suggestion and have calculated two ratios, nitrogen leakage ratio (NLR) = NH<sub>3</sub> emission / fertilizer N input, and nitrogen use efficiency = grain N harvested / fertilizer N input, to evaluate the loss and conversion of fertilizer N to NH<sub>3</sub> and grain production. We have added this discussion in **Section 3.3** and **Table 4**:

“**Table 4** summarizes the changes in annual-total fertilizer-induced NH<sub>3</sub> emission estimated by these simulations when the global synthetic fertilizer use rises to 130% of the 2000 level. The total fertilization rate at the 2050 level was 117 Tg-N yr<sup>-1</sup> (or +21% from the present-day total fertilization rate). We also computed the nitrogen leakage ratio (NLR) and nitrogen use efficiency (NUE) for each case. NLR maintains at ~15% for [CAM4\_CLM5\_2000] and [CAM4\_CLM5\_2050] while NUE decreases from 23% to 22%, respectively, indicating that the crops are under nitrogen surplus under this future fertilization scenario. This is also confirmed by the reduced ratio of crop uptake to fertilization from ~130% to ~115% (**Table S3**).

We also note that the DNDC NH<sub>3</sub> emission algorithm itself has been well validated with field observations, as explained in the first paragraph of **Section 2.2** in the revised manuscript.

Comment:

*Also, for easier comparison with existing inventories, it would be useful to provide the regional emission totals.*

Response:

Regional emission totals are used to compute the statistics in **Figure 2**. The information is now included in the manuscript as **Table 3**.

Comment:

*General comments*

*Eq. (1): How do you define the potential emission rate? The left hand side is a time derivative of a NH<sub>3</sub> concentration (?), which is generally not the same as the NH<sub>3</sub> flux to the surface. The factor 1/(delta t) on the right hand side does not make sense: the flux per time unit cannot depend on the timestep of the model.*

Response:

As mentioned above, we have revised the notations of the variables in the equations to clarify the physical meaning of each term in the NH<sub>3</sub> volatilization scheme.

Comment:

*How does the deposited  $\text{NH}_4^+$  enter the soil pools? Is 100 % of the wet deposition assumed to enter soil, or do you consider surface runoff? Figure 1 indicates that  $\text{NO}_x$  deposition goes to the  $\text{NH}_4^+$  pool, is this really the case?*

Response:

In CLM5 (and in our parameterization), the depositional nitrogen from dry and wet deposition of nitrate and  $\text{NH}_4^+$  both enter the soil  $\text{NH}_4^+$  pools (Lawrence et al., 2019). Loss of soil nitrogen via runoff/leaching only happen to nitrate in the model.

Comment:

*Eq. (2): What is the source of this formula? Does it mean that  $f_{ads}$  is greater than 1 when  $f_{clay}$  is very small? Why?*

Response:

**Eq (2)** is from the source code of DNDCv9.5 (Li et al., 1992; Nömmik, 1965). We implemented upper and lower bounds to  $f_{abs}$  such that its value is within 0 and 1 so  $f_{ads}$  will be close to 1 but will not be larger than unity when  $f_{clay}$  is small. We added the information regarding the upper and lower limit of  $f_{ads}$  to the revised manuscript.

Comment:

*Eq. (7): Please give a rationale for this equation. What is the role of  $T_{\text{soil}}$  here, given that it already appears in Eqs. (4) and (5)? Why does the flux from a given layer depend on the thickness of the soil column below? If you evaluate the emission from deeper layers, shouldn't there be also exchange between the layers? Finally, why parameterize the exchange between the soil and the atmosphere using the wind speed instead of using the resistance formulation already present in CLM for calculating dry deposition?*

Response:

According to DNDCv9.5, **Eq. (7)** encapsulates the effects of soil temperature, wind speed, and depth on gas diffusion along the depth of the soil. While DNDC does not directly compute the gas exchange between layers, the last term in **Eq. (7)** provides an estimation of how much gaseous  $\text{NH}_3$  from a layer at depth  $l$  will reach the surface air immediately above the soil column, i.e., quantifying the tendency of  $\text{NH}_3$  vapor in the soil to break the interface to enter the atmosphere.  $T_{\text{soil}}$  in Eq. (4) and Eq. (5) is used to determine the reaction rates that govern  $\text{NH}_4^+/\text{NH}_3$  equilibrium in the soil.

We adopt the parameterization in DNDC for the soil-atmosphere exchange instead of the CLM dry deposition formulation so that our scheme was as more consistent as possible with DNDC. It is now explained in greater detail in the 4<sup>th</sup> paragraph in **Section 2.2**.

Comment:

*How does the evaluation of the volatilization flux relate to the other NH<sub>4</sub>-consuming processes? Does it contribute to the N demand similar to the plants and microbial immobilization?*

Response:

The model distributes available soil NH<sub>4</sub><sup>+</sup> 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 processes are: nitrification (flux size = ~500 Tg-N yr<sup>-1</sup>), immobilization (~1600 Tg-N yr<sup>-1</sup>), plant uptake (~900 Tg-N yr<sup>-1</sup>) and NH<sub>3</sub> volatilization (14 Tg N/yr). Thus, when NH<sub>3</sub> emission is introduced, the 14 Tg-N yr<sup>-1</sup> is taken partially from plant uptake and partially from microbial immobilization. These are now explained in the 5<sup>th</sup> paragraph in **Section 2.2**.

Comment:

*Eqs (8) and (9): Which equations in the DNDC document do you refer to? Also, I'm not sure of what Eq. (8) means – it gives a linear relation between the concentration in soil and in the canopy, but what about the flux? What is the rationale for the  $1/s$  factor? What if the wind is calm? Finally, the constant  $14 \text{ m}^{-1}$  seems to go back to the paper of Erisman et al. (1994), where it is given as an empirical constant in a certain resistance component. How does it correct for the effect of canopy thickness?*

Response:

As mentioned above, we have revised the notations of the variables in the equations to clarify the physical meaning of each term for the  $\text{NH}_3$  volatilization scheme in **Section 2.2**.

**Eq. (8)** ( $F_{\text{atm}}$ ) is the portion of soil  $\text{NH}_3$  emission flux that is not captured by the canopy and is released to the atmosphere. We followed the DNDC scheme and discussed in **Section 2.2** that “dividing soil  $\text{NH}_3$  emission rate by  $s_{10}$  gives an approximate in-canopy  $\text{NH}_3$  concentration”. Hence, when the wind speed is low, the in-canopy  $\text{NH}_3$  concentration will be higher (i.e., slower dispersion) under a constant soil  $\text{NH}_3$  emission rate.

Our scheme takes into account the effect of canopy thickness in **Eq. (9)**, which is based on the Eq. (17) from the DNDC v9.5 Scientific Basis and Processes (Institute for the Study of Earth, Oceans, and Space, University of New Hampshire, 2017). The additional terms  $b(h_{\text{top}} - h_{\text{bot}})$  account for the height effect of the plant canopies, i.e., canopy thickness. We used Eq. (11) in Pleim et al., (2013) for calculating the in-canopy aerodynamic resistance.

Comment:

*2. Experiment setup*

*More details are needed about the model setup.*

*First, describe how the aerosol-radiation interaction was evaluated. Direct and/or indirect effects, nitrates, sulfates?*

Response:

As indicated by the reviewer, we extended the model setup description and added the following information to the manuscript:

“Atmospheric NH<sub>y</sub> does not directly interact with radiative transfer in CAM4-chem. Instead, its radiative implications are manifested in the radiative effect of changes in sulfate formation (direct) and the sequential sulfate-induced changes in cloud optical properties (indirect). Detailed description of the radiative transfer processes in CAM4-chem is provided in [Lamarque et al. \(2012\)](#) and the model manual (CAM Reference Manual, 2021).

Comment:

*Second, please describe whether the runs used some kind of nudging of the meteorological fields. This would be very important for understanding the comparisons between the runs which are presented later.*

Response:

The simulations were run with free dynamics. We revised the manuscript to provide more information about this in **Section 2.3**:

“CAM4-chem was run with free dynamics in the standard spatial resolution of 1.9° by 2.5° horizontally with 27 vertical layers (from surface to ~40 km). CLM5 was run in the same horizontal resolution with 25 soil layers down to ~50 m below ground. Sea surface temperature (SST) and sea ice conditions (Hurrell et al., 2008), as well as the mixing ratios of greenhouse gases (Meinshausen et al., 2017) were all fixed at the 2000-levels.”

Comment:

*Are CAM and CLM the only active components in the simulation?*

Response:

We clarified this point in **Section 2.3** as:

“Only the atmosphere (CAM4-chem) and the land (CLM5) components were active.”

Comment:

*If the model is driven or nudged by atmospheric reanalysis data (I think this is called the “offline” configuration in Lamarque et al. (2012)), the different simulations will share the same meteorological variability. However, if the simulations are run with fully prognostic atmospheric dynamics, the simulations will develop chaotic variations, and in this case, five years is unlikely to be long enough to obtain statistically significant differences. If the results shown are indeed from a free-running CAM simulation, all comparisons of means between the configurations should be tested for statistical significance to rule out the effect of the internal variability. The large differences in parameters like surface temperature over remote areas (Fig. S6) suggest that this might be an issue.*

Response:

Our simulations were run under “free dynamics” and not “offline” nor “specific dynamics”. To address the concern of long-term interannual meteorological variability, we extended our simulation to 30 years, as also suggested by reviewer 1. We provided details in **Section 2.3** in the revised manuscript:

“All simulations were run for 30 years using the spun-up year-2000 initial conditions with the corresponding land cover data provided out-of-the-box by CLM5. The first 10 years of outputs were used to further stabilize the model (such that the change in annual emission fluxes  $< \pm 10\%$ ) after our ammonia scheme was implemented. Our analysis in the next section focuses on the averages of last 20 years of simulated results to minimize influence from any long-term meteorological variability.”

We also conducted a series of two-sample t-tests to determine where the meteorological changes are significant and included the results in corresponding figures.

Comment:

*The experiments using modified fertilization rates (Section 3.3) should be introduced in the methods (Section 2.4). It might be worth noting that the future increases in agricultural production might involve also expansion of agricultural land area, and thus, the fertilizer application rate might on some areas change differently from the total fertilizer use. This would affect the response of nonlinear processes.*

Response:

We welcome the reviewer's point. We have now focused our analysis on the feedback effect of increased fertilizer use, as extended now in the last paragraph of **Section 2.3**, including a note to highlight the potential influence of cropland expansion in our discussion:

“We note that future increases in agricultural production might also involve cropland expansion, but such practice is not included in this study.”

Comment:

3. Results

*Regardless of the statistical aspects, some of the findings seem non-trivial and should be backed up with more analysis.*

*First, fairly large differences between the configurations are attributed to aerosol radiative effects. Please show the differences in the aerosol load (e.g. AOD) and in the aerosol radiative forcing, perhaps split by aerosol type if relevant. Are you able to rule out other atmospheric feedbacks, for example due to changed evapotranspiration?*

Response:

We appreciate the reviewer's suggestion and have added the contrast of  $\text{NH}_4^+$  and sulfate burden, net downward radiation fluxes of our simulations in **Figure S11** to **Figure S13** in the **Supplementary Information** and discussed the details in the last paragraph in **Section 3.3** of the revised manuscript.

Comment:

*Second, the results for grain production under increased N fertilization seem surprising. Generally increasing N fertilization would be expected increase harvest yield, even if not linearly. Here, the effect is negative for many regions, especially those in the southern hemisphere. What causes this? The authors should verify that the fertilization response in the CLM crop model is realistic (perhaps on a regional or per-crop basis) because otherwise the discussion of atmospheric feedbacks on crop production is not very meaningful.*

Response:

Lombardozzi et al. (2020) has extensively studied the crop response to fertilization in CLM5. Though increased N input would likely raise grain yield (Lombardozzi et al., 2020), a warmer temperature can also shorten crop growth period as the grain is harvested immediately when crop growing-degree-day (GDD) reach the maturity threshold, which may shorten the grain-filling period and result in smaller grain mass at harvest (Levis et al., 2012). We see that effect in our results as, for example, grain production decreases in South America (**Figure 8b**) coinciding with warmer surface temperatures (**Figure S9b**). We have now extended our discussion of these results in the second last paragraph in **Section 3.3**.

Comment:

*Finally, I'm a bit surprised to see such a big difference in crop growth (Fig. 6) between CAM4\_CLM and CAM4\_CLM\_CLIM (i.e. due to deposition) over areas like China or the U.S. corn belt, where N fertilization rates are known to be high. How large is the difference in the annual N deposition flux, and how does it compare to the annual N fertilization per crop area?*

Response:

The annual total global N deposition rate ranges from 15 to 16 Tg-N yr<sup>-1</sup>, which is ~16% relative to the model “present-day” fertilization rate (96.5 Tg-N yr<sup>-1</sup>) or ~13% relative to the “future” rate (117 Tg-N yr<sup>-1</sup>). The difference in grain production associated with the increased N deposition is revealed in **Figure 8** and **Figure S8** of the revised manuscript. The figures show that the increased N deposition are substantial, specially over China and US Corn Belt, and likely enhanced grain yield.

Comment:

*Specific comments*

*Introduction: the intro is not bad, but could be shortened to give a stronger focus on the present work. For example, the paragraph about in-situ observations seems excessive, since none of those data are used here.*

Response:

As indicated by the reviewer as well as reviewer 1, we have revised the Introduction to focus more on the relevant content.

Comment:

*L53: is the spending in USD a global total?*

Response:

Yes, it is referring to a global total. We revised the sentence to reflect this:

“**The global** public health system may have to spend 20–290 billion USD more each year to compensate for the NH<sub>3</sub>-derived detrimental effects on air quality and health (Gu et al., 2012; Paulot and Jacob, 2014; Guthrie et al., 2018).”

Comment:

*L142-145: there have been many (non-CESM) modeling studies using the resistance framework to simulate NH<sub>3</sub> exchange, including the canopy capture.*

Response:

We thank the reviewer to point out to other non-CESM studies. We are aware of such studies and revised the manuscript accordingly:

“We also developed a prognostic parameterization for canopy capture of NH<sub>3</sub>, instead of using a fixed generic value (e.g., one constant canopy reduction factor for all plants as used in some other studies (e.g., Riddick et al., 2016; Bouwman et al., 1997).”

Comment:

*L175-180: Lombardozzi et al., (2020) could be a useful reference about the CLM crop model.*

Response:

The suggested citation is added.

Comment:

*Figure 1: Much of the litter N is first assimilated to the microbial biomass and then remains in the soil organic matter (SOM) before becoming mineralized to  $\text{NH}_4^+$ . Having a SOM N pool in the figure would make sense, perhaps instead of the microbial N, which is anyway only implicitly represented in CLM (see e.g. [https://escomp.github.io/ctsm-docs/versions/master/html/tech\\_note/Decomposition/CLM50\\_Tech\\_Note\\_Decomposition.html](https://escomp.github.io/ctsm-docs/versions/master/html/tech_note/Decomposition/CLM50_Tech_Note_Decomposition.html)). Also,  $\text{N}_2\text{O}$  and  $\text{NO}_x$  are produced by both nitrification and denitrification. Denitrification also produces  $\text{N}_2$ .*

Response:

The microbe N pool is shown to highlight the two processes, namely, N fixation and immobilization associated with the microbes. We renamed the “Litter N” to “Litter/SOM N” to reflect the existence of SOM N in the model. We decided not to show  $\text{N}_2$  in the diagram as it is an inert species but mentioned it in the figure caption.

Comment:

*Section 2.3: the purpose of this section is unclear, since the rest of the paper is only about NH<sub>3</sub>.*

Response:

This section has been moved to **Supplementary Information**.

Comment:

*L297: Is this the setup described in Lamarque et al., (2012)?*

Response:

The reviewer is correct: The basic setup is largely similar to Lamarque et al., (2012), which is now cited in the introduction of that section.

Comment:

*Section 2.4: do you include any biomass burning emissions of NH<sub>3</sub>? If not, aren't you missing part of the N deposition in some regions?*

Response:

Yes, emission of NH<sub>3</sub> from biomass burning is prescribed in the CMIP6/CEDS inventory. We supplemented the information in the revised manuscript as:

“CAM4-chem employs a bulk aerosol approach and predicts the formation of PM<sub>2.5</sub> components including SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>, where the injection rates of precursors – sulfur dioxide (SO<sub>2</sub>), NO<sub>x</sub>, and NH<sub>3</sub> – are prescribed by Coupled Model Intercomparison Project phase 6 (CMIP6)/Community Emissions Data System (CEDS) emission inventory for anthropogenic activities as well as biomass burning in the default configuration (Hoesly et al., 2018).”

Comment:

*L303: biogenic emissions...of isoprene?*

Response:

Isoprene emission is one of the biogenic emissions handled by MEGAN2.1. We added isoprene as an example in Section 2.3:

“Biogenic emissions, e.g., of isoprene, are updated online from CLM5 using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 (Guenther et al., 2012).”

Comment:

*L315: by boundary layer, do you mean the quasi-laminar layer resistance  $R_b$ ?*

Response:

Yes, it refers to the laminar sublayer. We hence revised the sentence as:

“For NH<sub>3</sub> vapor, the model calculates the aerodynamic and the boundary-layer (laminar sublayer) resistance based on the online atmospheric dynamics, ...”

Comment:

*L321: the Henry's law applies to NH<sub>3</sub>, right?*

Response:

Yes, as stated in the manuscript in Section 2.3:

“For NH<sub>3</sub> vapor, the model calculates the aerodynamic and the boundary-layer (laminar sublayer) resistance based on the online atmospheric dynamics, while the surface resistance over land is determined according to the online CLM5 surface variables, e.g., canopy height and LAI, as well as species-specific reactivity factor for oxidation and effective Henry's Law coefficients.”

Comment:

*L343: Manure N is a significant N source in many areas. What is the reason for omitting it, and how does this affect the model results?*

Response:

The current version of CLM5, manure is assumed to be applied to crop land at a constant rate of  $2 \text{ g-N m}^{-2} \text{ yr}^{-1}$  (Lombardozzi et al., 2020), which is ~30% of total fertilizer input. Since the model is yet capable of tracing the source of the soil  $\text{NH}_4^+$  which then is responsible for the  $\text{NH}_3$  emission, we originally decided to focus our estimation of the soil  $\text{NH}_3$  emission that was solely from synthetic fertilizers by omitting the manure fertilizer. In the revised manuscript, we have re-run our simulations to include both synthetic and manure fertilizers and updated our results discussed in **Section 3** in the manuscript substantially accordingly.

Comment:

*L387: Boyland and Russell discuss a certain type of air quality models. I don't think that their conclusion can be used as a universal standard for a quite different application.*

Response:

We agree with the reviewer and revised the sentence to refrain from referring to the "acceptable range".

Comment:

*L410: perhaps even more importantly, some fertilizers have typically much lower NH<sub>3</sub> emission factor than urea (e.g. Bouwman et al., 2002). This includes for example anhydrous ammonia, which is common in the US.*

Response:

We agree that MASAGE has included fertilizers which can be more or less prone to NH<sub>3</sub> than urea. We revised the sentence in **Section 3.1** to reflect such view:

“MASAGE considers multiple-type fertilizers that can be more or less prone to NH<sub>3</sub> loss than urea (Bouwman et al., 2002), and assumes a three-stage fertilization at sowing, growth, and harvesting (Paulot et al., 2014).”

Comment:

*L421: “high spatiotemporal correlation is” unclear, I guess you just mean the temporal correlation. Though, how interesting is it to correlate the model to the inventories, given that the inventories usually prescribe the monthly variation? Why not compare to the IASI data on a monthly basis?*

Response:

We updated the analysis to include monthly IASA data and find that our updated model can reduce model low-biases on a monthly basis compared to the simulation using the CEDS emission inventory. We included this results in **Figure 4** and added relevant discussion in **Section 3.2** of the revised manuscript.

Comment:

*L464: did Hu et al. really use CESM?*

Response:

Thank you for catching this typo. The correct citation shall be He et al., (2015)

He, J., Zhang, Y., Glotfelty, T., He, R., Bennartz, R., Rausch, J., and Sartelet, K.: Decadal simulation and comprehensive evaluation of CESM/CAM5.1 with advanced chemistry, aerosol microphysics, and aerosol-cloud interactions, *J. Adv. Model. Earth Syst.*, 7, 110–141, <https://doi.org/10.1002/2014MS000360>, 2015.

*Figure 4: the IASI heatmap is saturated over large areas. Can you add more color levels to better show the variability of high-NH3 regions?*

We revised **Figure 4** to address the saturation issue.

Comment:

*L515: “ammonium salts” maybe better “secondary aerosols”*

Response:

We changed “ammonium salts” to “secondary ammonium aerosols”.

Comment:

*L529: I'm not sure if this kind of nonlinearity is expected, since the NH<sub>3</sub> emission is usually evaluated with constant emission factors. The emission has sometimes been suggested to increase faster than linearly (Jiang et al., 2017). Slower than linear increase seems to imply that either plant or microbial N demand increases nonlinearly to the input. Have you tried to analyze this?*

Response:

We reanalyzed the simulations and have updated the results in **Section 3.3**:

“The super-linear increase in NH<sub>3</sub> emission (+24%) relative to total fertilizer (+21%) is associated with sub-linear rise in nitrification (+17%), crop uptake (+5.8%) and other loss processes of soil NH<sub>4</sub><sup>+</sup>.”

Comment:

*L540: I'm not sure if I follow the logic here. What drives such a gradient in plant uptake?*

Response:

We have removed the sentence and updated the results in **Section 3.3**.

Comment:

*L555: is there any further evidence to show that this is a causal connection?*

Response:

The enhanced evapotranspiration due to better vegetation growth tends to shift surface energy balance to latent heat flux from sensible heat flux (Bonan, 2019). This sentence has been removed in the revised manuscript.

Comment:

*L566: is this in Tg of C, dry matter, or something else?*

Response:

Grain production is measured in Tg-(dry matter). Notes on this are added to the same line and the caption of **Figure 6**.

Comment:

*L589: more reliable...than what?*

Response:

Based on our results, we found that the dynamic estimation of NH<sub>3</sub> is more reliable than using constant emission inventory values under dynamic climate and environmental conditions. Hence, we revised the sentence as:

“These new features enabled CESM2 to perform, for the first time, a more reliable estimation of soil NH<sub>3</sub> emission and atmospheric NH<sub>3</sub> concentration than using constant emission inventory values under dynamic climate and environmental conditions.”

Comment:

*L610-615: Are the manure management emissions really easier to track? There is a huge diversity in manure management systems around the world. Not all facilities are confined. In many regions, collecting accurate information about farming practices is certainly not a trivial task.*

Response:

We intended to bring up that NH<sub>3</sub> emissions from manure management and other sources can be estimated and validated more efficiently using different approaches from ours. We rewrote the part to better convey such message:

“Unlike soil emission whereby the volatilization of NH<sub>3</sub> depends on a series of biogeochemical processes, emissions associated with manure management are typically estimated differently, e.g., collecting activity data and emission factors from factory managers, and installing monitoring instruments at outlets of confined facilities, e.g., animal factories (Bouwman et al., 1997; Paulot et al., 2014).”

Comment:

*L620: what data for validation do you have for fertilizer but not manure?*

Response:

For example, we do not have the amounts of manure fertilizers applied for each crop in each region and the NH<sub>3</sub> emission attributed to such fertilizers. We have now made this point clearer in the manuscript.

Comment:

*L621: By manure fertilizer, do you mean manure application on crops, or all manure-related sources? But Riddick et al (2016) only considered agricultural emissions, and did not consider different manure management processes.*

Response:

We revised the sentence to reflect the correct information:

“It is noteworthy that manure is attributable up to ~60% of total soil NH<sub>3</sub> emission (Vira et al., 2020) and hence shall warrant further research efforts in terms of its downstream impact on ecosystems via nitrogen deposition and aerosol radiative effect.”

Comment:

*L629-632: Does the effect of the initial NH<sub>4</sub> pools still remain after five years of spinup? What do you mean with a “soil nitrogen map”? I thought that the soil N is evaluated prognostically in CLM.*

Response:

Though the key metric variables in this study, e.g., NH<sub>3</sub> emission flux and grain production, have reached a quasi-equilibrium (interannual changes <±10%) in a few simulation years, different initial conditions may result in different steady states of a model simulation. For this, a more realistic “soil nitrogen map” – which we refer to the information regarding the global spatial distribution of soil N content – may help to constrain the modeled N concentration in the soil N pools by providing a more accurate initial conditions for model simulations. We have clarified this in the **Section 4** accordingly:

“The overestimation by CLM5 in this study may point to the more-fertile-than-reality soil conditions in the model, highlighting the need for a more realistic soil nitrogen map compiled by field surveys to better constrain the initial conditions for the model.”

Comment:

*L640: note that the N fertilization rate soybean is usually low, since it is a leguminous crop.*

Response:

Thank you for providing this information. We updated the fertilization rate used in the model in **Section 4**:

“A chamber study suggested that soybean can absorb up to 20 kg-N ha<sup>-1</sup> of NH<sub>3</sub> via leaf capturing (Hutchinson et al., 1972), which is a significant amount compared to average fertilizer use for soybean of 13–45 kg-N ha<sup>-1</sup> in CLM5”