Responses to the Editor

Comment:

Please also carefully check the level of N fertiliser application rate used in the simulations. From the top of my head, the number given 68 TgN/yr is low biased compared to other estimtates.

Response:

We thank the editor's comments. Our new estimates are that annual fertilization is 96.5 Tg-N/yr in the present day (2000) and 117 Tg-N/yr in our future scenarios (2050). These numbers are more comparable to the values suggested by FAO than the old estimate of 68 Tg-N/yr (when manure was excluded).

We have edited the text to reflect this update. In 3rd paragraph of the revised manuscript:

"The total fertilization rate at the 2050 level was 117 Tg-N yr^{-1} (or +21% from the present-day total fertilization rate at 96.5 Tg-N yr^{-1}, **which is comparable to ~100 Tg-N yr^{-1} suggested by FAO (2008)**)."

Reference:

Food and Agriculture Organization of the United Nations: Current world fertilizer trends and outlook to 2011/12, 2008.

Responses to Reviewer 1

Comment:

This paper explores some of the interactions and feedbacks of a fully coupled nitrogen cycle by examining the interactive coupling between nitrogen emissions from synthetic fertilizer, the resulting nitrogen deposition, the impact on climate, terrestrial ecosystems and crops. The paper first introduces a parameterization for nitrogen emissions from synthetic fertilizer, then evaluates it, and then finally examines various feedbacks with interactive nitrogen emissions.

While the topic is interesting, the paper requires substantial improvement in all aspects prior to publication as detailed below.

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. The explanation of the scheme for nitrogen emissions (section 2.2) needs to be more complete.

While there is a general reference to the DNDC model at the beginning of the derivation of NH3 emissions from synthetic fertilizer, it is somewhat of a mystery where the specific equations come from. Please explicitly include a rationale for the formulation of the specific equations, especially equations 1, 2 and 7.

Response:

Eq. 1 – **Eq. 7** are borrowed directly from DNDCv9.5 (Li et al., 2012; Gilhespy et al., 2014; source code of DNDC v9.5 provided by Changsheng Li via personal communication on Jun 18th, 2015, “the source code” hereinafter). Specifically, **Eq. 1** is from Li et al. (2012); **Eq. 2** is from Li et al. (1992) and Dutta et al. (2016); **Eq. 7** is based on the source code (Li et al., 1992; Gardner, 1965).

We have extended the description of the NH3 scheme in **Section 2.2** as suggested by the reviewer.

Comment:

Equation (1) is in terms of f_{vol}, the fraction of the non-adsorbed aqueous NH3 that volatilizes into NH3 gas. This does not seem to include an explicit term for the partitioning between NH3(aq) and NH3(g). If not, why not? Other formulations include this term.

Response:

Eq. 1 provides information to calculate the concentration of gaseous and aqueous NH_3, i.e. the partitioning between $NH_{3(aq)}$ and $NH_{3\ (g)}$: the non-adsorbed $[NH_4{}^+{}_{(aq)}]$ is given by $[NH4^+{}_{(soil)}](1 - f_{ads})$, $[NH_{3(aq)}]$ by $[NH_4{}^+{}_{(soil)}](1 - f_{abs})f_{dis}$, and $[NH_{3(g)}]$ by $[NH_4{}^+{}_{(soil)}](1 - f_{ads})f_{dis}f_{vol}$.

Comment:

Where does the formulation for f_{vol} *come from (equation 7)? This equation seems to have the peculiar property that for a one layer model (l_max=l) there are no emissions, while for a very thin layer this fraction will be maximum.*

Response:

Eq. 7 is obtained directly from the source code of DNDC v9.5. The variables *l* and *l*_max refer to the depth of a particular soil layer and the maximum depth of a soil column, but not their thickness. In other words, the last term in **Eq. 7** of a shallower soil layer is larger than that of a deeper layer, reflecting the fact that NH_3 gaseous from a shallower layer has a higher tendency to be emitted to the surface than that from a deeper layer.

We have made this discussion clearer in the description of **Eq. 7** in **Section 2.2**.

Comment:

Are the emissions sensitive to the vertical depth profile of fertilizer application? If yes, what is the depth profile of application?

Response:

Yes, emissions are sensitive to the vertical soil profile. Details of the soil profile structure is tabulated below, which is now added as **Table S1** in the revised Supplementary Information. Soil NH_4^+ pool exists in the first 20 layers, which are all prone to volatilization based on our scheme. Fertilizer N and depositional N are added to each soil layer based on a N Input Distribution Fraction, which is a fraction contribution of the predefined weighting factor, $e^{(-10l)/\Delta l}$, where l is the depth of a soil layer and Δl the layer thickness, both in meters.

Table A1. Soil layer structure.

Layer#	Layer Node Depth l (m)	Layer Thickness Δl (m)	Weighting Factor $e^{(-10l)/\Delta l}$	N Input Distribution Fraction
1	0.01	0.02	45.2	62.8%
2	0.04	0.04	16.8	23.3%
3	0.09	0.06	6.8	9.4%
4	0.16	0.08	2.5	3.5%
5	0.26	0.12	0.6	0.9%
6	0.40	0.16	0.1	0.2%
7	0.58	0.20	0.0	-
8	0.80	0.24	0.0	-
9	1.06	0.28	0.0	-
10	1.36	0.32	0.0	-
11	1.70	0.36	0.0	-
12	2.08	0.40	0.0	-
13	2.50	0.44	0.0	-
14	2.99	0.54	0.0	-
15	3.58	0.64	0.0	-
16	4.27	0.74	0.0	-
17	5.06	0.84	0.0	-
18	5.95	0.94	0.0	-
19	6.94	1.04	0.0	-
20	8.03	1.14	0.0	-
21	9.80	2.39	0.0	-
22	13.33	4.68	0.0	-
23	19.48	7.64	0.0	-
24	28.87	11.14	0.0	-
25	42.00	15.12	0.0	-

Reference:

- Section 2.2.2 in CLM5 technical notes, https://escomp.github.io/ctsm-docs/versions/release-clm5.0/html/tech_note/Ecosystem/CLM50_Tech_Note_Ecosystem.html, accessed on Jul 29, 2021
- Source code of CLM5: https://github.com/ESCOMP/CTSM/blob/master/src/soilbiogeochem/SoilBiogeochemVerticalProfileMod.F90, accessed on Jul 29, 2021

Comment:

Most formulations use the resistance approach for emissions from the surface which seems appropriate. The formulation presented here appears not to include resistances either for emissions into the canopy or from the canopy to the atmosphere. What is the evidence that this type of approach is valid? It also appears that NH3 from any soil layer is emitted directly into the atmosphere. What is the justification or rationale for this?

Response:

In our scheme, the term f_{vol} encapsulate wind speed, temperature of soil, and soil layer thickness to quantify the tendency of soil $NH_{3(aq)}$ to break the soil-air interface and vaporize to $NH_{3(g)}$ at the ground surface. The variables considered in this approach are similar to the boundary-layer resistance in several resistance methods (e.g., Pleim et al., 2013).

Our scheme assumes that there is vertical diffusion of $NH_3(g)$ from a deeper soil layer to the surface, but does not explicitly simulate it. Instead, this is represented in the last term in **Eq. 7** as a ratio of $(l_{max} - l)/l$ for the $NH_{3(g)}$ contained in each soil layer. As a result, NH_4^+ in a deeper layer is also subject to loss to NH_3 volatilization, but at much slower rate than the upper layers. We have added this information in the revised description of **Eq. 7** in **Section 2.2**.

Comment:

A number of models have incorporated a bidirectional flux of ammonia emissions. The results from the model presented here are indeed more complex than some of the simplest schemes used, but they seem somewhat less complex than the bidirectional approach in Pleim et al (2019) or Zhu et al (2015). Comparisons to these other more complex schemes should also be made in the text and these additional papers should be referenced.

Response:

We acknowledge that there are limitations in the NH_3 volatilization scheme in DNDC v9.5. We decided to implement this scheme as it is one of the models of intermediate complexity that have been developed and validated in multiple studies against field observations. The process-based nature of this scheme can also allow us to evaluate the response of NH_3 emission to soil climate, soil nitrogen content, fertilization, deposition, competition against other soil biogeochemical processes (nitrification, microbial uptake, etc.), and vegetation growth. Comparing to other approaches, the DNDC scheme requires variables that are mostly already modeled in CLM5, allowing us to largely capture the dynamic nature of NH_3 emission.

We added the citations in the revised manuscript in the last paragraph of **Introduction** and compared our approach to other more complex schemes mentioned by the reviewer.

Comment:

2. I find the results from the model are rather minimally evaluated and in some cases the evaluation is questionable. This is a bit strange as in lines 72-86 the paper outlines various measurement techniques for evaluation of NH3 emissions, but these are not used in the paper. This seems a little incongruous, as certainly the paper could have used more detailed model-measurement analysis, particularly the N deposition. Better evaluation is needed. While I could understand a minimalistic evaluation if the section on the feedbacks in the system were presented with more depth (see comments below) this is not the case.

Response:

We thank the reviewer for this note and have revised the introduction to focus on more relevant content.

We decided to use IASI satellite observations and the emission inventories because these datasets provide a global coverage, which allow us to widely compare the results of our global simulations in a consistent manner, including regions with few observations, e.g., South America and Africa. The emission inventories also better match the spatial resolution of our model simulations.

As pointed out by the reviewer, a key result in this study is to evaluate the sensitivity of NH_3 and grain production to the dynamic N cycle under intensified fertilization. Hence, we decided to focus our analysis on the benchmarking exercise to qualitatively compare the model performance with our scheme against observations and emission inventories, but not to exhaustively evaluate or improve the model-observation mismatch. We have also extended our analysis and discussion on the feedbacks in the system, as done in **Section 3.3**.

Comment:

-The model evaluation is in part against other established inventories EDGAR, CMIP and MASAGE (although arguably not with state of the art inventories such as the HTAP.v2.2 inventory and the CEDS inventory, which include significant local information into their emission estimates). The trouble is the EDGAR and CMIP inventories do not separate out manure and synthetic fertilizer emissions from agricultural soils. The paper assumes 1/3 of these emissions are "fertilizer associated" (line 364) where I assume the authors mean synthetic fertilizers. This number may be roughly valid globally, but certainly not regionally valid. Regionally, there may be very different apportionments between manure and fertilizer emissions from soils. Consequently, the geographic comparisons and statistical analysis between the CAM4_CLM5 EDGAR and CMIP6 are likely subject to significant local errors and are therefore not suitable for a quantitative comparison of the inventories. The temporal comparison between these emission inventories is also suspect as NH3 emissions of manure from agricultural soils may have a different seasonality than those from synthetic fertilizer.

Response:

We agree with the reviewer that excluding manure from our analysis make our model-emission inventory comparison a bit inconsistent. We have re-run our simulations to include manure and modified the manuscript substantially accordingly for the results presented in **Section 3**. The addition of manure did not change the main results of this paper, but simply improved the model-inventory comparison. We would like to note however that model-inventory comparisons are not exact given that our runs are performed using free-running dynamics and thus do not match with the meteorological year of the inventories, while the synthetic fertilizer use is not identical to the ones assumed when inventories were compiled. Thus, these results are presented as qualitative comparisons to indicate where our estimation is consistent with the inventories and where it is not. We also added this clarification to the revised manuscript in **Section 2.4**.

We also note that our named CMIP6 emission inventory is actually the CEDS emission inventory and made that clearer in the revised manuscript.

Comment:

Two model simulations are compared against the IASI satellite measurements: CAM4_CLM5 and CAM4_CMIP6. It is not clear to me (although maybe I missed it) if the CAM4_CLM5 and the CAM4_CMIP6 emissions are identical except for the synthetic fertilizer NH3 emissions. Are the other sources of NH3 emissions identical so that the only difference in these simulations is from differences in the synthetic fertilizer emissions? Differences in the simulations can only be attributed to the simulated ammonia emissions if the inventories are identical except for the emissions from agricultural soils. Not only do the NH3 emissions from other sectors besides synthetic fertilizer need to be the same between the simulations, but also the NOx and sulfate emissions as the partitioning of NH3 into the aerosol phase depends sensitively on these emissions also. Thus, unless the emission inventories are identical except for the ammonia emissions from fertilizer, it seems difficult, without more analysis, to quantitatively compare CAM4_CLM5 and CAM4_CMIP6.

Response:

The emission inventories are identical, except for the difference in the fertilize-induced NH_3 estimated by CLM5. We have made this point clearer in **Section 3.2** stating that:

"Source of non-fertilizer related NH_3 and other reactive gases were identical in these two cases."

Comment:

3. The model simulations are not well explained.

A number of simulations were made with various feedbacks enabled. However, it is unclear how long any of these simulations were run for, whether ensemble simulations were made and the statistical significance of any difference between the simulations. In these coupled simulations meteorological variability can result in apparent differences. The regional changes in temperature (S6, for example), are quite large, probably larger than can be expected from rather small changes in radiative forcing.

Response:

We took the advice of the reviewer and decided to extend the duration of our simulations.

We agree with the reviewer that meteorological variability might affect our coupled simulations as only 5-year averages were used originally. We therefore decided to extend the duration of our simulations to 30 years to minimize that influence. We detailed our new set up as well as made clearer our simulation description in Section 2.3:

"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 < ±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 included results of two-sample t-tests to highlight where differences are statistically significant in our figures.

Comment:

Biogeochemical models of soils are notoriously difficult to deal with, as they are notoriously difficult to spin up to equilibrium. In these simulations this is not discussed. Please elaborate on the spinup of the biogeochemical part of these simulations including the extent to which the coupled system was spun up to equilibrium. What state was the model initialized from? How did this state change with the introduction of the new parameterization? Was the model spun up to equilibrium after making changes to the parameterization of ammonia emissions?

Response:

We used the out-of-the-box spun-up initial condition provided by CLM5. We identified the annual-total ammonia emission fluxes fluctuate mildly after the first few years of simulations (<10% yr^{-1}). Hence, we set to use the first 10 years to stabilize the simulations for our modifications.

We provided this information in the revised manuscript, **Section 2.3**, as cited in our responses to the comment above.

Comment:

There are some feedbacks with the crop model which need to be addressed. It appears that only synthetic fertilizer is added to the crops whereas in reality manure is also be used to fertilize crops. Consequently, the crops are likely significantly under-fertilized in the model simulations. The apparent under fertilization in these simulations would suggest that the crops take up a greater fraction of added nitrogen in the model-world than they would in reality. This would suggest the fraction of applied fertilizer volatilized is underestimated in the model. (In reality surplus nitrogen is added to agricultural systems, a fraction of which is lost.) Please address the extent to which this might impact the simulations.

Response:

In the revised manuscript, we have included manure (at +2 g-N m^{-2} s^{-1} in addition to synthetic fertilizer as in default CLM5) in our simulations. Our present-day fully-coupled case [CAM4_CLM5_2000] estimates that 68 Tg-N yr^{-1} of synthetic fertilizer is applied annually, and it is increased to 96 Tg-N yr (+41%) when manure is included. The corresponding grain-N produced increases from 18.5 Tg-N yr^{-1} to 22.1 Tg-N yr^{-1} (+19%). As suggested by reviewer 2, in this study, we have also determined the nitrogen-use efficient (NUE) as the ratio of grain-N produced to fertilizer input, and the nitrogen leakage ratio (NLR) as the ratio of NH_3 emission rate to fertilization rate. The NUE is reduced from 27% (synthetic fertilizer only) to 23% (synthetic and manure fertilizer), indicating that the crops are not under-fertilized.

The ammonia emission is correspondingly increased from 10.5 Tg-N yr^{-1} to 14.4 Tg-N yr^{-1} (+38%). We also find that NLR is slightly reduced from 15.1% to 15.0%, implying that NH_3 volatilization is likely not underestimated in the cases with and without manure fertilizers.

Details are provided in **Table 3**, **Section 3.1**.

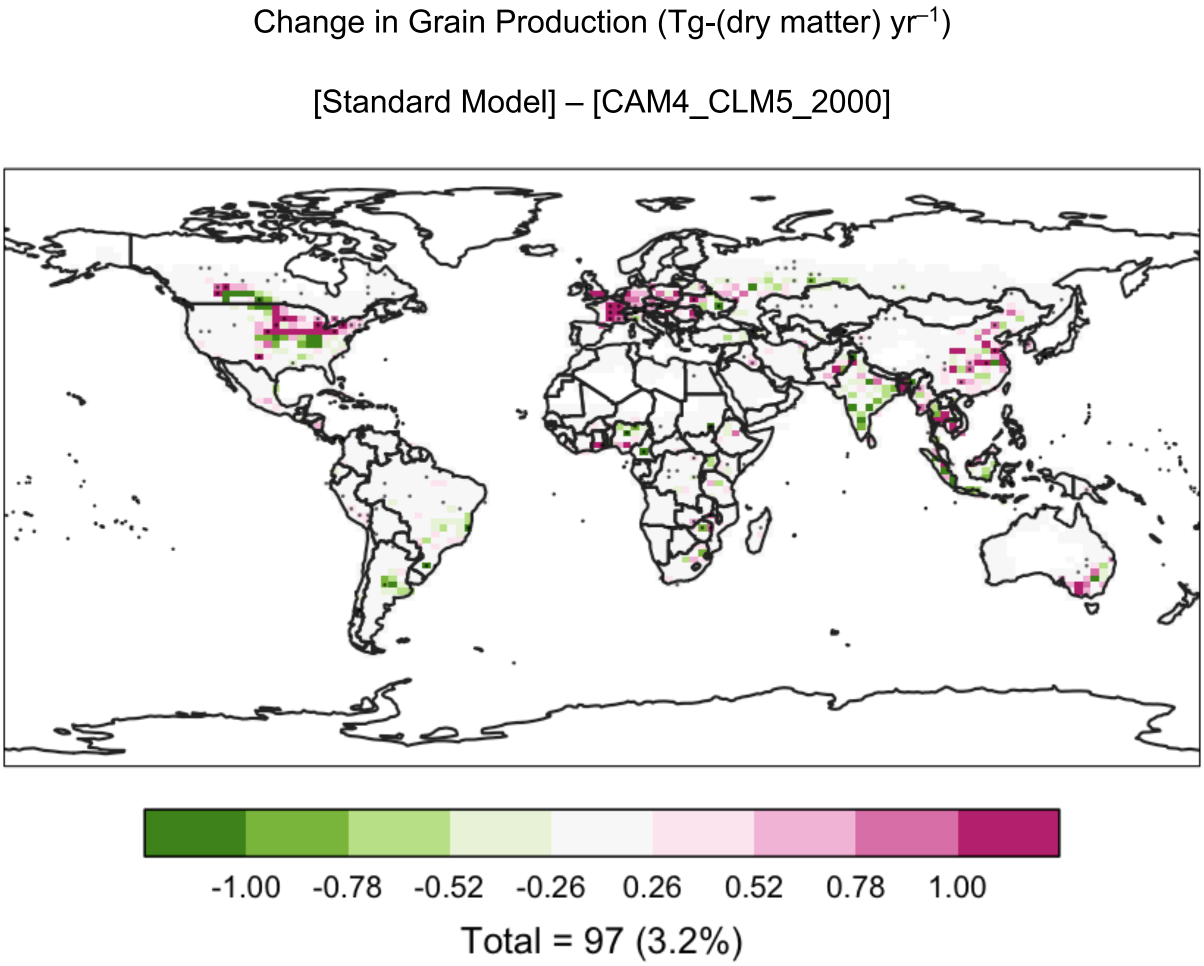
Comment:

It seems likely that with the under-fertilization the simulated crops may not show sufficient growth. This is likely to have climate impacts including changes in latent and sensible heat flux and changes in albedo. How does crop growth in the scheme documented here compare with that in the standard CLM model without the ammonia emissions? How does this impact the radiative budget?

Response:

There are some differences in crop growth, which is partly manifested in the change in crop yield/production. Without our NH_3 scheme and the dynamic land-atmosphere nitrogen cycle, the standard model estimates 3.2% more annual total grain production than the fully coupled case. This difference is expected as NH_3 volatilization is a substantial competitor to soil ammonium and implementing our NH_3 scheme would reduce soil NH_4^+ available for plant uptake.

Spatially, larger differences are seen in the northern US and Europe as shown in the figure below:



Change in Grain Production (Tg-(dry matter) yr^{-1})

[Standard Model] – [CAM4_CLM5_2000]

Regarding the radiative budget, we find the following results and added to Section 3.3:

“Compared to the default model, our fully coupled simulation estimated a 0.13 W m^{-2} increase in global downward radiative flux, which is substantial compared to the total aerosol radiative forcing of +1.0 Wm^{-2} (Myhre et al., 2013).”

Comment:

4. I found the section regarding model sensitivities needs significant more in depth analysis. It seems to me this section could be the real novelty of the paper (schemes with bidirectional fluxes have been implemented previously as have prognostic equations for NH3 emissions). This is particularly true as the model evaluation is not comprehensive.

In section 3.3 the different responses of the system are simulated after a change in forcing (i.e., a change in added fertilizer). If I understand correctly the authors are comparing the [CAM4_CLM5] with 2000-level fertilization with: (i) [CAM4_CLM5] with a 30% increase in fertilization, with (ii) CAM4_CLM5_CLIM with a 30% increase in fertilization but constant nitrogen deposition and with (iii) CAM4_CLM5_NDEP with a 30% increase in fertilization but constant aerosol forcing. I found the section somewhat confusing, perhaps in part due to the notation. It would probably be clearer if the authors distinguished in their notation the simulations with different emissions (e.g., the CAM4_CLM5 with standard emissions versus that with a 30% increase in fertilization).

Response:

We appreciate the reviewer's suggestion and have updated the notations of our simulations with different fertilization levels with corresponding suffixes, i.e., "_2000: and "_2050". As mentioned above, we also extended substantially the discussion in **Section 3.3** to strengthen the results from our model sensitivity analysis.

Comment:

Also, how was the aerosol forcing kept constant in CAM4_CLM5_NDEP?

Response:

We chose a configuration in CAM4-chem that makes the atmospheric chemistry inactive to the radiative transfer module. We have made this point clearer in the manuscript, **Section 3.3**:

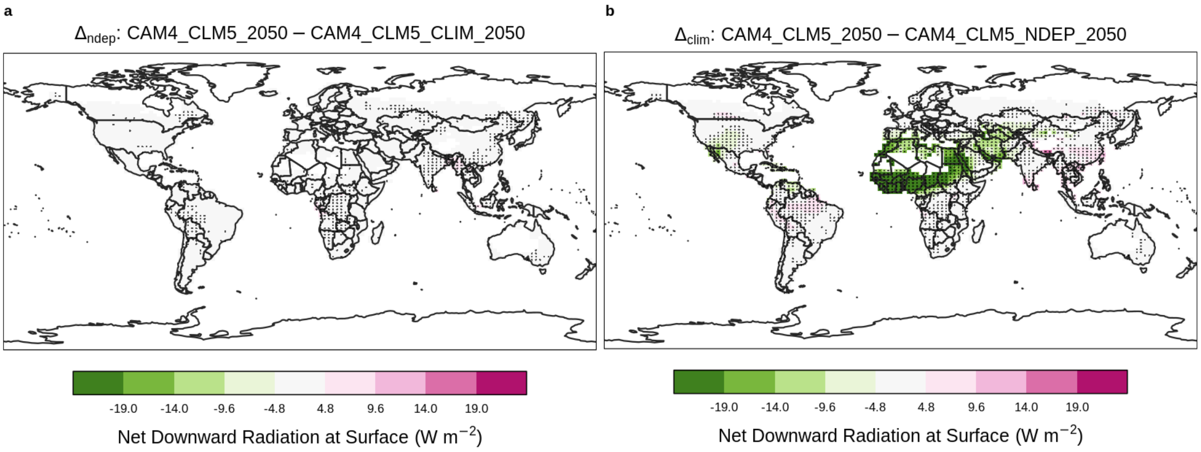
"Similarly, [CAM4_CLM5_NDEP_2050] was set up such that addition/reduction of NH_3-induced aerosols would be inactive to the radiative transfer module, i.e., would not induce changes in aerosol-climate interactions, so that we could isolate the impact of NH_y deposition on NH_3 emission and crop growth."

Comment:

In CAM4_CLM5 emissions increase by 27% or to 2.4 Tg N/year. In CAM4_CLM5_CLIM with constant year 2000 deposition fluxes the emissions increase to 2.5 Tg N/year; in CAM4_CLM5_NDEP and constant aerosol forcing they increase to 2.7 Tg N/year. It is hard to interpret the significance of these differences as they seem small on the face of it. Are these differences really significant? What is the difference in radiative forcing? Substantial more analysis could be conducted here. As just one example the paper states some changes are "likely a consequence of better vegetation growth driven by increased NHy deposition following higher NH3 emissions". This can be evaluated by examining the model.

Response:

The reviewer raises an important point with respect to the significance of the results. We have conducted a series of two-sample t-tests when comparing variables and added indications for statistically significant results in all relevant figures as suggested. For example, in Supplementary Information, we provide **Figure S11** to illustrate the changes in annual-mean net downward radiation flux at the Earth's surface:



a Δ_{ndep}: CAM4_CLM5_2050 – CAM4_CLM5_CLIM_2050

b Δ_{clim}: CAM4_CLM5_2050 – CAM4_CLM5_NDEP_2050

To examine vegetation growth, we used crop grain production as the increase in grain biomass during the grain filling period is a direct measure of crop growth as well as the nitrogen use efficient for crop food products. As in CLM, crop grain production is one key indicator of crop growth (Levis et al., 2012) and farming efficiency. We decided to focus our results on the reported grain production.

Comment:

I am rather puzzled by the statement (Lines 601, 602): "We estimated that the effect of nitrogen deposition on NH3 emission is +2.7 Tg-N yr–1 globally" with a reference to Figure 5. Figure 5 shows the changes in emissions when fertilizer is increased by 30% compared to the case with no change in emissions. Shouldn't the 2.7 Tg-N yr–1 increase in emissions in CAM4-CLM-NDEP be, to a large extent, attributable to the increase in fertilizer, not to the effect of nitrogen deposition. Maybe I have completely missed something here.

Response:

The statement was referring to the change in NH_3 from [CAM4_CLM5] with present-day level fertilizer to [CAM_CLM5_NDEP] with fertilizer at the future level. We have clarified our statement and rewrote that sentence. It reads now as:

"We also estimated that if the synthetic fertilizer use was to increase by 30% from 2000's level, NH_3 emission would rise by 3.3 Tg-N yr^{-1} globally (see **Figure 5**)."

Comment:

Other points:

-Lines 104-105: "Recent inventories…..", but then the paper quotes Sutton (2013). There are in fact much more recent inventories than that. Other more recent inventories include the HTAP_v2.2 inventory and the CEDS inventory which are not mentioned.

Response:

We welcome the reviewer's suggestion to include results from more recent inventories and have revised it accordingly. Our CMIP6 emission inventory is actually the CEDS inventory, and we have updated the manuscript accordingly to clarify this information:

"CAM4-chem employs a bulk aerosol approach and predicts the formation of $PM_{2.5}$ components including SO_4^{2-}, NO_3^-, and NH_4^+, where the injection rates of precursors – sulfur dioxide (SO_2), NO_x, and NH_3 – 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:

-Lines 274- 295 Emissions of other reactive nitrogen compounds. As far as I can the emissions of species other than NH3 are not discussed in the paper or evaluated. This section can then be omitted. It seems to me what is pertinent here is the loss of ammonia through nitrification.

Response:

We moved this section to **Supplementary Information** to improve the readability.

Comment:

- Line 212: I assume that equation (1) should also include other loss terms: washout and nitrification for example. Please clarify.

The potential soil NH_3 emission rate determined by **Eq. (1)** is used by the model to compute the competition for available soil NH_4^+ with other processes, namely, plant uptake, microbial immobilization, and nitrification. We expanded the sentence to describe this treatment more explicitly. The model assumes leaching (including "washout") occurs for soil nitrate only.

We added the addition information to the 2nd and the 5th paragraphs of **Section 2.2** in the revised manuscript.

Comment:

-Lines 325-327: In equation (9) why is Vc set to a fixed deposition velocity instead of the deposition used in the chemistry model?

Response:

We wanted to be as consistent as possible with the DNDC configuration and kept the V_c for in-canopy NH_3 constant as provided in the DNDC scheme (Li et al., 1992; Nõmmik, 1965).

Comment:

- The constants in a number of the equations in section 2.2 do not have defined units (e.g., equations 4 and 5). Please give explicitly where these equations come from as appropriate and the units for the constants.

Response:

We added the references and units of constants to the equations in **Section 2.2** as suggested.

Comment:

-The change in ammonia is apparently calculated for each soil layer (equation 1), but I assume that equation (8) is in terms of all soil layers. Please clarify.

Response:

In **Eq. (8)**, f_{can} is a column-level variable. It is applied to the column-total actual NH_3 emission flux (g-N m^{-2} s^{-1}). We clarified this point as suggested.

Comment:

-Line 305: "In our coupled simulations, we omitted the portion of NH3 emission associated with synthetic fertilizer from the inventory input for CAM4-chem."It is not clear where the inventory for this input comes from in CAM4-chem.

Response:

We revised the sentence to emphasize that the portion of fertilizer-induced NH3 is omitted from the CMIP6/CEDS emission inventory: (**Section 2.3**)

"In our coupled simulations, we substituted the portion of NH_3 emission associated with synthetic fertilizer from the CAM4-chem inventory input (CESD) for our online simulated emission rates from CLM5. Atmospheric NH_3 and NH_4^+ formed sequentially return to CLM5 through deposition."

Comment:

-Line 409: Some synthetic fertilizers have a much smaller ammonia volatilization loss than urea.

Response:

We agree with the reviewer's comment. MASAGE includes fertilizers that can be more or less prone to NH_3 volatilization than urea. We revised the sentence to reflect such view:

"For example, MASAGE considers multiple-type fertilizers that can be more or less prone to NH_3 loss than urea (Bouwman et al., 2002), and assumes a three-stage fertilization at sowing, growth, and harvesting (Paulot et al., 2014)."

Comment:

-Figure 5. Please include figure captions for the figure components: a, b, c and d. Also the caption on Figure 5b is misleading. It would be helpful if the two cases were distinguished.

Response:

Captions of **Figure 5** and **Figure 6** are edited according to the reviewer's suggestions.

Responses to Reviewer 2

Comment:

This paper presents a parameterization for evaluating ammonia (NH3) volatilization from soils within the Commonity Eath 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 NH3 following fertilizer applications and the transport, deposition and possible re-emission of the emitted NH3. 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 NH3 exchange into the nitrogen cycle of an Earth system model does open up new possibilities for assessing the role of atmospheric transport of NH3 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 NH3 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 18th, 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 NH3 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 NH3 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 NH3 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 NH3-N emitted and fertilizer-N applied.

Response:

We welcome the reviewer's suggestion and have calculated two ratios, nitrogen leakage ratio (NLR) = NH_3 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_3 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_3 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^{-1} (or +21% from the present-day total fertilization rate, which is comparable to ~100 Tg-N yr^{-1} suggested by FAO (2008)). 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_3 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 NH3 concentration (?), which is generally not the same as the NH3 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_3 volatilization scheme.

Comment:

How does the deposited NH4+ enter the soil pools? Is 100 % of the wet deposition assumed to enter soil, or do you consider surface runoff? Figure 1 indicates that NOx deposition goes to the NH4+ pool, is this really the case?

Response:

In CLM5 (and in our parameterization), the depositional nitrogen from dry and wet deposition of nitrate and NH_4^+ both enter the soil 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; Nõmmik, 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_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 NH_3 from a layer at depth *l* will reach the surface air immediately above the soil column, i.e., quantifying the tendency of NH_3 vapor in the soil to break the interface to enter the atmosphere. T_{soil} in Eq. (4) and Eq. (5) is used to determine the reaction rates that govern NH_4^+/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 4th paragraph in **Section 2.2**.

Comment:

How does the evaluation of the volatilization flux relate to the other NH4-consuming processes? Does it contribute to the N demand similar to the plants and microbial immobilization?

Response:

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 processes are: nitrification (flux size = ~500 Tg-N yr^{-1}), immobilization (~1600 Tg-N yr^{-1}), plant uptake (~900 Tg-N yr^{-1}) and NH_3 volatilization (14 Tg N/yr). Thus, when NH_3 emission is introduced, the 14 Tg-N yr^{-1} is taken partially from plant uptake and partially from microbial immobilization. These are now explained in the 5th 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 m-1 seems to go back to the 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 NH_3 volatilization scheme in **Section 2.2**.

Eq. (8) (F_{atm}) is the portion of soil NH_3 emission flux that is not capture by the canopy and is released to the atmosphere. We followed the DNDC scheme and discussed in **Section 2.2** that "dividing soil NH_3 emission rate by s_{10} gives an approximate in-canopy NH_3 concentration". Hence, when the wind speed is low, the in-canopy NH_3 concentration will be higher (i.e., slower dispersion) under a constant soil 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 $b(h_{top} - h_{bot})$ 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_y 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 1.9º by 2.5º 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 < ±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 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^{-1}, which is ~16% relative to the model “present-day” fertilization rate (96.5 Tg-N yr^{-1}) or ~13% relative to the “future” rate (117 Tg-N yr^{-1}). 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_3-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 NH3 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_3, 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 NH4+. 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). Also, N2O and NOx are produced by both nitrification and denitrification. Denitrification also produces N2.

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 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 NH3.

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 NH3?. If not, aren't you missing part of the N deposition in some regions?

Response:

Yes, emission of NH3 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 $PM_{2.5}$ components including SO_4^{2-}, NO_3^-, and NH_4^+, where the injection rates of precursors – sulfur dioxide (SO_2), NO_x, and NH_3 – 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 Rb?

Response:

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

"For NH3 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 NH3, right?

Response:

Yes, as stated in the manuscript in Section 2.3:

"For NH_3 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 g-N m^{-2} 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 NH_4^+ which then is responsible for the NH_3 emission, we originally decided to focus our estimation of the soil 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 NH3 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_3 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_3 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 excepted, since the NH3 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_3 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_4^+."

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_3 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_3 emission and atmospheric NH_3 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 NH3 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_3 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_3 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_3 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 NH4 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_3 emission flux and grain production, have reached a quasi-equilibrium (interannual changes <±10%) in a few simulation years, different initial conditions may result in difference 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^{-1} of NH_3 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^{-1} in CLM5"