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Comment on bg-2021-63

Anonymous Referee #2

Referee comment on "Modeling the interinfluence of fertilizer-induced NH₃ emission, nitrogen deposition, and aerosol radiative effects using modified CESM2" by Ka Ming Fung et al., Biogeosciences Discuss., <https://doi.org/10.5194/bg-2021-63-RC2>, 2021

This paper presents a parameterization for evaluating ammonia (NH₃) 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 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.

General comments

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.

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

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.

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 NO_x deposition goes to the NH₄⁺ pool, is this really the case?

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?

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?

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?

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 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?

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

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.

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.

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?

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.

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?

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.

L53: is the spending in USD a global total?

L142-145: there have been many (non-CESM) modeling studies using the resistance framework to simulate NH₃ exchange, including the canopy capture.

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

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 NO_x are produced by both nitrification and denitrification. Denitrification also produces N₂.

Section 2.3: the purpose of this section is unclear, since the rest of the paper is only about NH₃.

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

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?

L303: biogenic emissions...of isoprene?

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

L321: the Henry's law applies to NH₃, right?r.

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?

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.

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.

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?

L464: did Hu et al. really use CESM?

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?

L515: "ammonium salts" maybe better "secondary aerosols"

L529: I'm not sure if this kind of nonlinearity is expected, 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?

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

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

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

L589: more reliable...than what?

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.

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

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.

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.

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

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