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.

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.

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. 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 \( (l_{max}=1) \) there are no emissions, while for a very thin layer this fraction will be maximum.

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

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.

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.

- 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.

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.

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.

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?

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.

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?

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

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 NH3 deposition following higher NH3 emissions”. This can be evaluated by examining the model.

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.

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.

- 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.

- Line 212: I assume that equation (1) should also include other loss terms: washout and nitrification for example. Please clarify.
In equation (9) why is $V_c$ set to a fixed deposition velocity instead of the deposition used in the chemistry model?

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.

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.

"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.

Some synthetic fertilizers have a much smaller ammonia volatilization loss than urea.

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.

REFERENCES
