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Interactive comment on “Estimate of changes in agricultural terrestrial nitrogen pathways and ammonia emissions from 1850 to present in the Community Earth System Model” by S. N. Riddick et al.

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Received and published: 21 October 2015

I acknowledge the comments of my colleague Dr. Andrej Ceglar to this review.

General Comments

To our knowledge, this is a first attempt to calculate global NH₃ emissions into air with a land-model, including nitrogen cycling in soil. This study represents an important step forward in earth system modeling, using a process based model for NH₃ emissions due to the nitrogen applied to the surface of agricultural land, therefore incorporating

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an important agricultural component. The process-based model is well described, as well as the coupling with boundary conditions provided by output from the atmospheric compartment of the earth system model.

The authors evaluate the sensitivity to inputs of manure and synthetic fertilizer of the various nitrogen pathways on grassland and agricultural crops, considering cycling in soils, and release to water- and the main theme of the paper- to air. The latter is of high importance in view of the increasing importance of NH_3 in determining atmospheric aerosol, as well as a range of other environmental issues, and also in the context of increasing nitrogen use to feed a growing population in the next decades. Probably one of the interesting potential outcomes of this work could be a real investigation of the climate dependency of NH_3 emissions, speculated to be important by Sutton et al.; Geels et al. (cited in the paper).

While it is in principle possible to estimate the climate dependency of NH_3 emissions using future temperature precipitation scenarios, the authors have chosen to leave the evaluation to a follow-up paper. However it would be valuable to provide an estimate of the pre-industrial and present day impacts of this temperature signal (in the order of 0.7 C world average, and higher in the major agricultural production areas) on NH_3 emissions.

As indicated also by the authors- the calculated NH_3 emissions are highly uncertain, since the actual emissions will depend on a host of parameters, including management practices on fairly local scales, and highly simplifying assumptions. The estimated global NH_3 emissions have been compared with a number of alternative more static inventories (e.g. Beusen, EDGAR4). Although the comparison looks reasonable (note here the static inventories partly make similar assumptions), I am left with the impression that this agreement is dependent on rather arbitrary choice of key-parameters such as soil pH and canopy capture factor. Especially the assumption that on average 60 % of nitrogen is captured by plants is rather unconstrained and critical. As far as I understand CLM4.5 does consider plant functional types that include also nitrogen

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pools in the various tissue. In my view this represent a missed opportunity to get more solid insights in the role of soil-canopy exchange in determining emissions.

Another issue that can be probably relatively easily addressed is the run-off of N and comparison with other estimates. Once NH₃ is emitted in the atmosphere it doesn't mean it is gone- it will probably deposit for a large part not very far from the sources. Also NO_y deposition will contribute to nitrogen inputs into the watersheds. It would be good if the figures and text dealing with this correct for this phenomenon, or clearly mention that run-off only partly includes the full picture.

The authors do a fairly good job in providing single-parameter sensitivity studies, but of course it should be recognized that some of the parameters may co-vary and can not be seen in isolation.

The following 2 comments are perhaps oriented more towards the future work in this field, which could be clarified as well within the manuscript.

You mention that simulation was run in “decoupled mode” with atmosphere, using re-analysis from Quian et al (2006). As you mention, several important processes of exchange between canopy and atmosphere are not explicitly simulated. This is an important consideration, given that the canopy “health” status and biomass accumulation depends critically on fertilizer input. Nitrogen affects also the other processes in crop canopy, such as transpiration rates, which might have the cooling effect over the cropland. Therefore, fully coupled model should be explored in the future to observe and understand better also the feedback mechanisms related with the atmosphere. Are you planning to perform these simulations?

The resolution of your simulations is quite coarse (1.9 x 2.5 degrees). This makes the results of your study applicable from regional to global level and importantly contribute to understanding the NH₃ emissions globally. As you mentioned, agricultural practices are regionally highly variable and can substantially change also within the resolution of your simulation runs, especially so over the regions with heterogeneous soils. Prob-

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bly, model results are therefore better over more homogeneous agricultural areas, such as grasslands. Important source of uncertainty in this study is associated with simplifying farming methods (e.g. a single date for fertilizer application is used, only urea fertilizer used, etc.). I suppose a next important step will be to incorporate agricultural management model, trying to capture these processes better. Nevertheless, due to high spatial variability of agricultural management practices related to fertilization, the results of such simulations might have limited value for regional applications when run on coarse resolution. How do you foresee in the future this problem might be tackled? Perhaps a kind of up-scaling method could be applied to bridge the gap between local (management practices) to coarser resolution of earth system models?

As the paper is in a way very conceptual- a more structured analysis in the discussion section of the next key-steps to come to more robust analysis of N-cycling and NH₃ emissions in the land and ESM model would be valuable.

Despite our criticisms, we nevertheless think that the study is very valuable since it provides a first framework on top of which in a later stage modifications, improvements and extensive testing will be possible. I think the authors did a good job in describing assumptions, but the manuscript could include some more detailed insights on the next steps to be taken. Unfortunately the manuscript doesn't read very smoothly, any improvement of the readability would be welcome.

All in all I recommend publication of this manuscript after taking into consideration my general and detailed comments.

Detailed Comments.

p. 15949 l. 13,14,20. It would be useful to have the numbers matching the overall N-inputs. Providing soil N_r formation would be useful. Please keep the same units.

p. 15949 l. 20 Why do the earlier mentioned emissions in 2000 not match? In case of all nitrogen I would expect $21+12=33$ Tg N (40 Tg NH₃) if only organic nitrogen: 25.5

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Tg NH3.

p. 15951 l. 13 provide year of reference since numbers can change rapidly over time.

p. 15951 l. 28 in future

p. 15952 l. 14 degree C.

p. 15952 l. 13-15 it should probably be recognized that also other soil microbiological processes may change with climate, as well as agro-management practice (beyond this study).

p. 15959 l. 28 l. 182 check the chronology of the publications, hard to imagine that results that were published in 2010 (and not available in 2007) were used in 2007.

p. 15957 l. 14 I agree, but a correct mix of the two concepts is a challenge. This could be further elucidated in the discussion.

p. 15957 I recommend to use the wording synthetic fertilizer (or inorganic fertilizer) as manure is also a fertilizer. Throughout the manuscript.

p 15959 l. 17 at least in Europe import of feed is at the core of many nitrogen-related environmental issues. I propose to delete the 'generally' statement, unless the authors are certain that this is not the case elsewhere.

P 15959 l. 25 I would like to see this discussion on simplifying assumptions such as not explicitly using crops, but rather what is in the land-model, earlier in the manuscript, possibly in the introduction. It is not very clear to me whether there is a consistency between the gridded manure database, and the CLM land-use. How much of the manure is on place where CLM thinks there is a forest, desert or similar? It would be good to know that indeed manure is placed where grasslands and crops grow (+fractions).

p. 15961 l. 16: it is quite well known that net-losses from urea (NH₃ emissions), can be much higher (15-20 %) than for most other nitrogen components (a few %). See Bouwman et al., (1997) Is it correct to assume that the numbers in this manuscript are

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from this point of view an upper limit? I am surprised by the sensitivity study which contradicts these general view, and I don't understand why.

p. 15961 I. 24; corn (grain maize) is an important crop. However it is not representative for instance for winter wheat, another widely used crop. Some discussion on the implications could be given.

p. 15964 I. 12 There are a number of modeling papers, and satellite retrievals that could provide a more insight on the range of concentrations. See recent review in Zhu and Henze Curr Pollution Rep (2015) 1:95–116.

p. 15964 I. 20 Can the authors comment on the physical evidence for such 're-capture' of NH₃ emissions? It is well known that nitrogen in the plants tissues will also tend to maintain an NH₃ partial pressure. If stomata are closed this will probably less so play a role. Is this mainly a tuning factor? And how sensitive is the model to it?

p. 15971 I. 18 Descriptions of global mapsetc

p. 15973 One aspect which should be mentioned here is that in reality the NH₃ emitted, will probably not travel very far and on the time scale of hours (NH₃) or days(NH₄) deposition near the emission regions. On the gridscale considered, it can be safely assumed that most of the NH₃ also enters in run-off. It was not clear how run-off simulations of this model compare to observations were taken into account (see general remark)

p. 15973 I. 25 See previous remark.

p. 15973 confusing use of the word deposited – I assume the autors talk about atmospheric deposition.

p. 15974 I do think the hypothesis of some fraction of NH₃ being readily available to plants could be tested using the information available in CLM. It is a missed opportunity.

p. 15976 I. 5 EDGAR4.2 output contains output separating manure management

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and manure directly on the field. It seems that the authors have not included manure management (sector B2), which at least to a great extent finds its way in to the fields as well. This would make the overall EDGAR estimates from agriculture larger, but the ratios more consistent. Beusen et al. 2008 estimate does include emissions from housing/storage.

p. 15978 I am somewhat surprised by the low sensitivity to the background concentration. It would be good to include in the figures a visualization of the $\text{NH}_3(\text{g})$ the NH_3 concentration in equilibrium with the TAN pool. 15970, line 5: Does this imply that other factors, such as wind and water availability lead to increased NH_3 emissions, even though the temperature is lower?

15970, line 12: Can you clarify why in your opinion the performance over grassland is better? In addition, differences in the distribution of manure over grassland and arable land can affect the NH_3 emissions, since the emission factors differ between slurry application methods on grassland and arable land. Is arable land foreseen as category of land use in your simulations? Perhaps, some clarification would be needed.

15973, line 23: Did Bouwman take into account soil nitrogen pools, which could describe the difference between the two studies?

p. 15979/10. On page 15965/19 the authors declare that soil pH is simply set to a value of $\text{pH}=7$. The sensitivity tests suggest that pH is one of the more determining factors for NH_3 emissions (as expected). Can the authors explain why they have not used one of the global soil pH databases around to try to get more robust results?

l. 15979/15 As argued above it would actually make sense to dedicate a sensitivity study to this- as this assumption (no influence on soil pools) doesn't seem very correct.

l. 15980 l. 8 I think this needs some further exploration as to the underlying reasons. Can the reason be related to different management practices related to synthetic fertilizer?

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