

Interactive comment on “Global evaluation of ammonia bi-directional exchange” by L. Zhu et al.

Anonymous Referee #3

Received and published: 10 June 2015

The manuscript reports an ambitious attempt to evaluate improvements in the representation of ammonia surface-atmosphere exchange in chemical transport models. A significant portion of the manuscript is devoted to discussing the implementation of a new representation of diurnal variability for agricultural emissions (referencing an in prep publication) and a new bottom-up agricultural inventory (MASAGE_NH3). However, the title of the manuscript only reflects the second aspect of the paper, which examines the implementation of a bidirectional exchange scheme. In the description of GEOS-Chem in 2.1, the authors should emphasize that the description of the NH3 emissions is the base case and that the variants will be explained in the following sections.

A pervasive problem with the manuscript is that the comparisons between different model runs and between model runs and observations are difficult to digest. Partly

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this is because the figures are so small and the information is all over the continents, and partly it is because the text reports the extremes of differences, rather than more general metrics. Below I provide general and detailed comments for the two portions of the manuscript separately:

Sections 3 and 4

The evaluation of the updated dynamic emissions scheme is quite haphazard. It is challenging to extract quantitative information from Figures 2, 3, and 4 and the related text which describes comparisons with ground sites and remotely sensed representative volumetric mixing ratios reports biases or improvements in a handful of regions, but the approach lacks consistency. While the cases reported may be representative, the reader is left wondering about regions and times that aren't mentioned.

Section 3.1 Surface measurements – what is the impact of having observational constraints at such differing time resolutions? What is the impact of including urban and rural sites in some regions, and only rural sites in others?

P4828, L22-25 This sentence reads as though soil pH and fertilizer application influence livestock emissions. Is that correct?

P4829, L11-10, This paragraph is hard to follow. How many SEARCH sites are used, and do they all provide observations of NH3 and wet deposited NH4+? Are the three sites with high time resolution data combined because they fall in the same model grid cell?

P4831 – L10-14 If I understand correctly, the fraction of anthropogenic emissions that are due to livestock are estimated for all regions of the world based on the NEI for the U.S. Is this fraction likely to be the same in other parts of the world?

P4832, L6-23 This section describes a comparison with modelled RVMR and those retrieved from TES, but is very hard to follow. Why not include a figure, or a table of statistics, rather than quoting differences from a few regions?

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A larger question from this section is: If the total livestock emissions are staying constant, how can the deposition be decreasing?

Sections 5-7

The manuscript provides significant detail on the representation of the soil ammonium pool, which responds to atmospheric deposition. On the other hand, there is no mention of the ammonium pool in the vegetation, which one assumes would influence the stomatal compensation point. Is there a reason this is not addressed in a similar online manner?

What is the rationale for looking at the adjoint sensitivity with respect to soil pH rather than soil $[H^+]$? It seems like the log scale might skew the perception of the emission potential.

A more general question is whether the adjoint of GEOS-Chem been sufficiently validated for a species like NH_3 with significant non-linearities in its behavior? Presumably, one would need to have met fields very accurate and also abundance of SO_4 , HNO_3 . As stated by the authors, the HNO_3 in the model is likely biased, and one would assume that the sensitivity of NH_3 concentrations to emissions depends on the model HNO_3 .

An issue with many of the comparisons between models runs in Section 6 is that the value that is typically quoted in the text is the largest difference, which may not provide much insight on typical behaviour. I recommend quoting the median difference, as well as the maximum.

In section 6.1, it would be interesting to know if the annual gross emissions are lower or higher across the US in the base vs BIDI cases.

In section 6.1.1, the AMoN comparison suggests that the BIDI parameterization degrades the ability of the model to represent the variability in two-week integrated measurements in the spring and fall. Can the authors speculate if this would also be the

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case for higher time resolution? Additionally, what fraction of the gross emissions are from bidirectional exchange as a function of space and time?

In Section 6.1.3, it is not really clear what the authors are trying to demonstrate with this comparison. Are the 'uni-directional' emissions from Zhu et al., 2013, replacing the MASAGE inventory? I think the hybrid, piecemeal nature of the comparison makes it difficult to interpret the results.

Section 6.2 – again, it would be interesting to know how the annual emissions change for each model run.

P4845, L17-23 I find this section confusing. On some spatial scale, there ought to be mass balance between the changes in emission and deposition. Obviously, there could be some change to the amount of dry deposition, so one cannot expect the emissions and wet deposition to change in exactly the same way, but they should be close. In comparing the changes in wet deposition to the changes in emissions, why quote one change in absolute terms and the other as a percent. It makes in challenging to compare them.

Section 6.4.3 – I think this spot sensitivity analysis is one of the more interesting parts of the manuscript, as it provides one of the more robust and digestible results of implement bi-directional flux.

Technical corrections:

Figure 3 is missing a colour scale.

P4826, L26-27 – missing word in sentence

P 4836, L8 Gaussian is misspelled

Fig 9 shows R_2 whereas Fig 10 shows R , it would be better to be consistent

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 4823, 2015.

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