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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Agricultural ammonia emissions in China: reconciling bottom-up and top-down estimates" *by* Lin Zhang et al.

Lin Zhang et al.

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Comment: Zhang et al have done a terrific job using models and observations to improve understanding of ammonia emissions in China. Not only do they do a top-down analysis, using the GEOS-CHEM adjoint constrained by TES column NH3 measurements, to improve the seasonal and spatial variability in NH3 emissions, they then do a very thorough job improving past bottom-up inventories through careful analysis of fertilization practices and animal emissions. Combined, these make for a very strong paper – one of the best I have reviewed in some time. I recommend the authors attend to a few comments in revising the manuscript:
Response: We thank Prof. Collett for the valuable comments. We have addressed all of them in the revised manuscript, and please see the itemized responses below.

Comment: 1. One of the main challenges in accurately simulating ammonia concentrations in chemical transport models is the treatment of dry deposition. Considerable attention has been paid recently to including more realistic, bi-directional flux parameterizations and this seems to help quite a lot in some regional simulations. Without a bidirectional treatment, NH3 loss rates by dry deposition can be biased high. While I am OK with the authors not including a bidi treatment in their model simulations here, I do think they should add some discussion how its absence might influence their results. This is relevant to the top-down NH3 emissions estimates and to the comparison of model vs. surface concentration and wet deposition estimates.
Response: Thank you for pointing it out. We now add the following text in Sect. 5.3 to discuss the bi-directional NH3 flux: "Furthermore, while land-atmosphere exchange of NH3 is bi-directional, the model here treats it as one-way emission and dry deposition processes. Zhu et al. (2015) previously implemented a bi-directional NH3 exchange algorithm in GEOS-Chem, and they found that it led to small changes in wet deposition fluxes but had large impacts on emission estimates and surface concentrations over eastern China. Future work is required to improve the bi-directional exchange processes in the model."
Added Reference: Zhu, L., Henze, D., Bash, J., Jeong, G.-R., Cady-Pereira, K., Shephard, M., Luo, M., Paulot, F., and Capps, S.: Global evaluation of ammonia bidirectional exchange and livestock diurnal variation schemes, Atmos. Chem. Phys., 15, 12823-12843, https://doi.org/10.5194/acp-15-12823-2015, 2015.

Comment: 2. Line 77: I suggest changing "together contribute" to "together are estimated to contribute"
Response: Changed as suggested.

Comment: 3. Lines 150-157: the authors should discuss the Streets emission inventory here in the text. It is included in the figure and shows the strongest season-

ality.
Response: We now plot the spatial distribution of NH3 emissions from the Streets inventory on Figure 1. We also state here: “NH3 emission estimates of Streets et al. (2003) have a strong peak in June, and are much higher than Huang et al. (2012) and Paulot et al. (2014) in winter.”

Comment: 4. Line 173: I suggest changing “NH3 prefers to combine” to “NH3 is thermodynamically favored to combine”
Response: Changed as suggested.

Comment: 5. Line 182: change “mixed clouds” to “mixed-phase clouds”
Response: Changed as suggested.

Comment: 6. Lines 182-184: please explain and justify the retention efficiencies chosen for mixed-phase and cold clouds
Response: Here we add the reference: “Wang, J., Hoffmann, A. A., Park, R. J., Jacob, D. J., and Martin, S. T.: Global distribution of solid and aqueous sulfate aerosols: Effect of the hysteresis of particle phase transitions, J. Geophys. Res., 113, D11206, doi:10.1029/2007jd009367, 2008”.

Comment: 7. Lines 248-249: are NH3 concentrations possibly also higher here because there are fewer NOx and SO2 emissions to generate acids that tie NH3 up in aerosols?
Response: Thanks for pointing it out. We now state here “High NH3 concentrations are also observed over Xinjiang province in Northwest China, which are likely emitted from animal grazing and remain mainly in gas phase due to fewer NOx and SO2 emissions to generate acids there.”

Comment: 8. Lines 336-338: How accurate/representative for China are the

authors’ assumptions here re: frequency of application of injection and broadcast fertilization methods?
Response: We now state here “Based on fertilizer application practices, the first fertilizer application at plant is typically through injection (for rice and tobacco the first applications at both seeding and transplanting fields), and the rest by broadcast.”

Comment: 9. While the manuscript is generally quite well written, there are several small grammatical errors that should be corrected. The most significant are
a. Line 52: change “have” to “has” and “cause” to “causes”
b. Line 66: change “in the eastern China” to “in eastern China”
c. Line 129: change “human” to “humans”
d. Line 268: change “while overestimate” to “while they overestimate”
e. Line 276: change “increases in” to “increases are noted in”
f. Line 319: change “need to consider” to “requires considering”
g. Line 394: change “spending” to “spent”
h. Line 400: change “while only” to “while we only”
i. Line 442: change “needs to address” to “requires addressing”
j. Line 443: change “layer centered” to “layer is centered”
Response: Thank you for pointing them out. We have corrected them and a few other errors in the manuscript.

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