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Interactive comment on "Interannual Variability of Ammonia Concentrations over the United States: Sources and Implications" *by* Luke D. Schiferl et al.

Anonymous Referee #2

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The article title, "Interannual Variability of Ammonia Concentrations over the United States: Sources and Implications", by Schiferl et al., is well written and timely given the limited knowledge of ammonia variability. Provided below are some review comments.

General Comments:

1) The paper talks about changes in the transfer of ammonia from the surface to the atmosphere due to temperature and windspeed (volatilization scaling), but does not put it in the context of bi-direction exchange and gas-aerosol phase transitions. The deposition and re-emission processes in the bidirectional exchange extends the spatial range of influence of the NH3 emissions, and hence the NH3 lifetime (e.g. Zhu et al., 2015). NH3 also contributes to the formation of atmospheric aerosols that can

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reside and be transported in the atmosphere for several days to a week releasing NH3 back into the atmosphere modifying the variability of ammonia concentrations. It would be good if the authors could provide insights on the impacts of the variability due to bi-directional flux. Zhu L., D.K. Henze, J.O. Bash, G.-R. Jeong, K.E. Cady-Pereira, M.W. Shephard, M. Luo, F. Paulot, and S. Capps, Global Evaluation of Ammonia Bi-Directional Exchange and Livestock Diurnal Variation Schemes, Atmos. Chem. Phys, 15, 12823-12843, doi:10.5194/acp-15-12823-2015, 2015.

2) The paper recognizes the limitation of the satellite observations due to lack of vertical information. It would be good to note that this is not general to satellite observations, but the particular IASI ammonia retrieval algorithm used in the study. For example, the new CrIS NH3 optimal estimation retrievals (Shephard et al., 2015) will be able to provide this type of information (e.g. averaging kernels and covariance matrics) allowing for more quantitative comparisons against the model simulations.

3) It is still not totally clear how the impact of the spatial sampling between the model and the observations impact the measurement variability. For a study over just North America, why was a global GEOS-Chem model used instead of a more regional model (i.e. CMAQ) to investigate the ammonia variability? A regional model would at least have a spatial sampling that is more representative for comparisons with the observations.

Minor Comments:

1) Page 5, line 27. Also should add in AIRS and CrIS.

2) Page5, lines 29-30. "...calculated from a wider spectral range than previous ammonia products,...". It is not clear if the point is to just state this fact, or imply that this is better. Using a wider spectral coverage does not necessarily produce a better retrieved product. For example, a robust spectral window selection approach can be based on the maximum information content by taking into consideration errors (e.g. interfering species, spectroscopic errors, measurement errors, etc.) (e.g. Echle et al., (2000) and

Worden et al. (2004)).

Echle, G., T. von Clarmann, A. Dudhia, J. M. Flaud, B. Funke, N. Glatthor, B. Kerridge, M. Lopez-Puertas, F. J. Martin-Torres, and G. P. Stiller (2000), Optimized spectral microwindows for data analysis of the Michelson Interferometer for Passive Atmospheric Sounding on the environmental satellite, Appl. Opt., 39(30), 5531–5540.

Worden J, S. Sund, M.W Shephard, S.A Clough, H. Worden, K Bowman, A Goldman. Predicted errors of tropospheric emission spectrometer nadir retrievals from spectral window selection. J Geophys Res. 2004;109:doi:10.1029/2004JD004522.

3) Page 6, line 1: Please state what forward radiative transfer model was used.

4) Page 6, line 5: Are these uncertainties relative, or absolute, or both?

5) Page 6, line 14: remove "present"

6) Page 6, line 16: Maybe also add to the line ending in "...distributed measurements" the additional "and the differences in measured quantities.", which leads nicely into the next sentence.

7) Page 9, lines 25-29: should mention in addition to vertical sensitivity, the last of the actual information content limits the comparison.

8) Page 10, lines 10-15. Could the lack of variability also be due to the fact that satellite total column values are being used, rather than information from only the parts of the profile where the satellite is sensitive (e.g. often limited information right at the surface).