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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Simulation of nitrate, sulfate, and ammonium aerosols over the United States" *by* J. M. Walker et al.

Anonymous Referee #1

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This is a review of the submitted article:

Journal: ACP Title: Simulation of nitrate, sulfate, and ammonium aerosols over the United States Author(s): J. M. Walker et al. MS No.: acp-2012-568 MS Type: Research Article

The submitted manuscript titled, " Simulation of nitrate, sulfate, and ammonium aerosols over the United States", is concise and very well written. There is still significant uncertainty in the model-simulated concentrations of nitrate, sulfate, and ammonium aerosols over the United States (and even more so globally). Surface and satellite observations of NH3 provide valuable information on the performance of the chemical transport models, especially over California. There are many advantages of utilizing the recently available satellite NH3 observations, but due to the nature of the

infrared retrievals they can be challenging to evaluate. There are many advantages of utilizing the recently available satellite NH3 observations, but due to the nature of the infrared retrievals further details and explanations are needed. This research is very relevant, but it does not appear to be as consistent with recent similar analysis in as simple of a way as currently presented for this CA case study and will need to be addressed (see comments below).

Main Comments:

1. There are a couple IASI data oddities that would benefit from further explanation:

a) There are two peaks in WA that are greater than any values in the Midwest. The one on the right might be plausible – it's over a farming valley. The one on the left is not – it's over the North Cascade National Park. Not sure what could be emitting NH3 in this region.

b) IASI doesn't appear to pick up the hotspot in North Carolina, which according to EPA has the highest density of hog farms of any county in the country, and thus high NH3.

2. NH3 lifetime:

a) According to Seinfeld and Pandis, NH3 lifetime is as long as 10 days (page 38), although most people would estimate it's lifetime to be much shorter, as short as a few hours. Regardless, Turner et al. (2012) showed that columns of NH3 in GEOS-Chem can be influenced by NH3 emissions several grid cells away. Thus, directly relating model vs. IASA NH3 concentrations is challenging so please provide justification.

3. IASA and TES indicate that NH3 concentrations are also underestimated throughout much of the country. However, in the Midwest / east, nitrate is overestimated. So there appears to be a conflict here as I don't think this CA case study presented is as consistent with the Lye et al., 2012, Heald et al., 2012, and Zhang et al., 2012 in as simple way as is presented here. Not that I am doubting that the CA NH3 emissions are underestimated – all signs (recent studies) do point in that direction. It's just there may

be more happening to explain nitrate nation wide. For example, if NH3 is increased by 300% in CA, that leads to more NH4NO3, which then downwind will exacerbate overestimates of nitrate in other parts of the country.

4. Observational errors.

a) It would be helpful if the authors please provide typical estimated observational errors.

5. In regards to the satellite observations in Section 4.2.

a) Please provide the reader with more details on the exact IASI inverse retrieval methodology used in this study and its characteristics (i.e. apriori, sensitivity, assumptions, and estimated errors, etc.).

b) Several limitations in the utilization of the satellite observations are mentioned (i.e. "reliable satellite averaging kernels were not available. . .", ". . .ammonia columns are not available during the winter months due to insufficient ammonia sensitivity. . .". Could the IASI observations with great spatial coverage not be supplemented with satellite observations from the Aura TES, which operationally produces reliable averaging kernels, error estimates, and better sensitivity (but less dense spatial coverage), to provide greater insight over the U.S?

c) It should be noted in the manuscript (maybe just an additional sentence or so with a references) that a more robust and quantitative assessment of GEOS-Chem emissions using satellite observations would be obtained through more detailed inverse modeling using the satellite observational operator (averaging kernel, apriori, errors).

d) Page 19, lines:19-23. In order to include these lines more supporting evidence needs to be presented for the statement about the vertical sensitivity from IASI from 0-2km being uniform. Even though the temperature contrast and elevated amounts of NH3 over California can significantly improve the vertical sensitivity of the infrared retrievals, it is still generally not the case that the vertical sensitivity is uniform. For

example, routine observational Aura TES NH3 retrievals (which typically has greater sensitivity) over California show a non-linear vertical sensitivity (see attached Fig 1. of a typical TES averaging kernel over CA). I don't think this will change the resulting qualitative scientific conclusions based on the IASI observations in the manuscript as California is a region with the greatest infrared satellite vertical sensitivity in the boundary layer. Therefore, I would suggest just changing lines 19-23 to something along the lines that the linear assumption is made, and that it is realized that the sensitivity in the boundary layer changes from profile-to-profile and not in general equally sensitive in the bottom 0-2 km (provide a reference), but that in the California region the satellite observations do in general have good boundary layer sensitivity.

6. Sensitivity sampling errors between GEOS-CHEM and IASI.

a) This is an active area of research and it is not expected or the goal of this manuscript to address this, but it should be noted in the manuscript that some differences in the IASI GEOS-Chem comparisons could be due to the different sampling and sensitivity. For example, IASI has a much smaller $\sim$15 km footprint, and is mostly sensitive to elevated NH3 values, which could skew mean values compared with the model.

References:

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Zhang, L., Jacob, D. J., Knipping, E. M., Kumar, N., Munger, J. W., Carouge, C. C.,

van Donkelaar, A., Wang, Y. X., and Chen, D.: Nitrogen deposition to the United States: distribution, sources, and processes, Atmos. Chem. Phys., 12, 4539–4554, doi:10.5194/acp-12-4539-2012, 2012

Zhu L., D.K. Henze, K.E. Cady-Pereira, M.W. Shephard, M. Luo, R.W., Pinder, J.O. Bash, G. Jeong, Constraining U.S. ammonia emissions using TES remote sensing observations and the GEOS-Chem adjoint model, accepted to J. of Geophys. Res., 2012.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/12/C7961/2012/acpd-12-C7961-2012-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 19499, 2012.

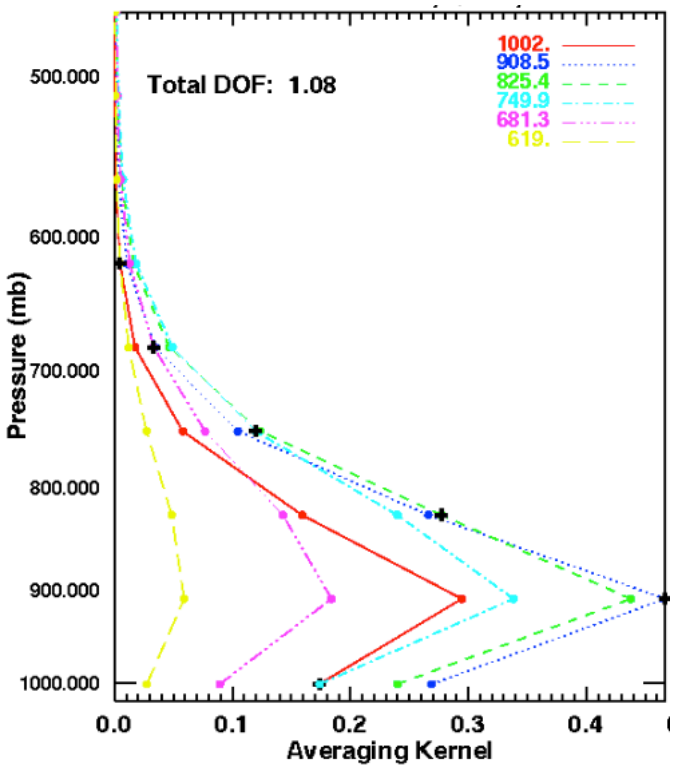

Fig. 1.