

Interactive comment on “TES ammonia retrieval strategy and global observations of the spatial and seasonal variability of ammonia” by M. W. Shephard et al.

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Author's response to Referee's #2 comments for “TES ammonia retrieval strategy and global observations of the spatial and seasonal variability of ammonia” by M. W. Shephard et al.

Anonymous Referee #2

The authors are again very grateful to the referee for providing very helpful comments that improved the manuscript. Below are detail responses to Referee #2 comments.

1. Page 16026, Line 25, 'initial comparisons results' should be 'results of initial compar-

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isons'

Response: Agreed (Referee #1 also caught this). We changed “initial comparisons results” to “initial comparison results”.

2. Page 16028, Lines 15-21 The authors state that the retrieval for NH₃ is carried out after the retrievals for temperature, water vapor, ozone, methane, carbon dioxide, clouds, and surface temperature and emissivity. Is it ever the case that the ammonia signal is significant enough that it turns out to have interfered with the retrievals of the other trace gases?

Response: This is a very insightful comment. This is not an issue for the TES ammonia retrievals due to the fact that TES retrieval strategy utilizes its high spectral resolution (0.06 cm⁻¹) and selects retrieval parameter specific microwindows for each retrieval. This greatly minimizes the impact of interfering species (systematic errors) on the retrievals. For example, the spectral region used for these TES ammonia retrievals is not used by any other TES retrieval and thus will not impact any prior retrievals.

3. Page 16028, Line 27, 'ozone), the' should be 'ozone, and the'

Response: Agreed. We added the “and” after the “ozone),” as suggested by the referee.

4. Figure 1 – Have the 10% perturbations to H₂O, CO₂ and O₃ been applied to the entire vertical profile of each gas? And is this in contrast to the NH₃ perturbation, which has presumably changed the relative vertical distribution? I'm not sure if this matters but perhaps another sentence in the main body of the paper would help to clarify.

Response: The referee's assumptions are correct. The 10% perturbations to H₂O, CO₂, and O₃ were applied to the entire vertical profile of each gas. Since NH₃ has a much weaker signal the perturbation is actually from an atmosphere with no NH₃ to an atmosphere containing a polluted NH₃ profile. Note that an unpolluted NH₃ profile

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with limited thermal contrast often has very little signal, which is very similar to the zero case.

To make this clearer in the paper we used the Referee's suggestion to add after the line: "Figure 1 shows a simulated sensitivity analysis depicting the NH₃ microwindows and interfering species." the following sentences : " The perturbation in Figure 1 for the interfering species were applied to the entire vertical profile of each gas. Since NH₃ has a weak atmospheric signal the perturbation is created by going from an atmosphere without any ammonia to an atmosphere with a polluted NH₃ profile."

As the Referee commented, this is not of great importance as these perturbations are mostly just used to identify the spectral regions associated with the various gases in this spectral region.

5. Figures 5 and 6 and sections 2.1.4 and 2.2.1 – why does data only extend along the x-axis in Figure 5 down to -7 K, but down to -25 K in Figure 6, if the analyses are based on the same simulated data (Page 16032 Line 11)?

Response : In Figure 6 we adjusted the thermal contrast to account for the surface emissivity, which we did not do in Figure 5. We have recreated Figure 6 without the emissivity adjustment: both figures now have the same x-axis range. This surface emissivity adjustment was removed from Figure 6 as it was operationally infeasible.

6. Figures 6 and 7 – remove information about who created the figure

Response: Yes, we noticed this just after the final submission to ACPD. This has been fixed.

7. Section 2.2.2 The effect of clouds is described here, but not the potential effect of aerosol on the retrieval. Given that agricultural regions and biomass burning areas are both high emission areas for NH₃ and also aerosol, it would be interesting to know if the presence of particles will influence the retrieval.

Response: This is a good and frequently asked question. The portion of the mid-
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infrared spectrum used for ammonia retrievals (960-970 cm⁻¹) is not sensitive to aerosols. If the retrievals were performed at larger wavenumbers (closer to the near-infrared) then the presence of particles would likely start influencing the retrievals.

8. Page 16036, Lines –The authors state that, on average, the representative volume mixing ratio (RVMR) is expected to be about 30% of the in situ mixing ratio that would be measured at the ground. Is this based on GEOS-Chem output, or are there vertically resolved observations that confirm this falloff with altitude. Given that future evaluations and comparisons of the TES product are likely to rely on surface-based observations, it would be useful to have a slightly more detailed discussion about the factors that might influence this ratio or scaling factor.

Response : This is a really good and important point. First of all we should not have said surface, but rather the peak value in the profile (we had it correct in the abstract, but not in this section of the text). Our purpose when stating the factor was to provide readers a rough estimate of the RVMR value corresponding to the 1 ppbv sensitivity threshold. We analyzed this factor again from a set of retrievals from simulated spectra obtained from GEOS_Chem profiles and a good "rule of thumb" estimator is closer to 40%. Note that the true factor is highly dependent on the profile shape, and thus using this number to convert from TES RVMR to surface values is not advisable. As mentioned by the referee if a relationship with a surface value could be attained that would be very useful. This is very difficult, but we are presently working on the issue using EPA surface data.

We rewrote the following paragraph:

From: "The RVMR is a "weighted" average over the region of high TES sensitivity and thus is significantly lower than the surface values. By comparing RVMR and surface values from a set of simulated cases (see next section) we determined that the RVMR in general can be roughly estimated as 30% of the surface value; thus in terms of RVMR, the detectability level is 0.3 ppbv, though as stated earlier, this level is influ-

enced by the thermal contrast and other atmospheric parameters.” To: “The RVMR is a “weighted” average over the region of the profile where TES is sensitive and thus is significantly lower than the maximum value. By comparing the RVMR and peak profile values from a set of simulated cases (see next section) we determined that the RVMR for cases with NH₃ near the detectability level of 1 ppbv, can in general be roughly estimated as 40% of the maximum value; thus in terms of RVMR, the detectability level is 0.4 ppbv, though as stated earlier, this level is influenced by the thermal contrast and other atmospheric parameters.”

Note that this 0.4 threshold was applied to update Figures 15 and 16.

Also, more description is added on line 1 on page 16039.

Changed “. . .Fig. 15” To “. . .Fig. 15 for retrievals with DOFS \geq 0.5 and RVMR \geq 0.4.”

9. Page 16038, Lines 4-5 “where TES is sensitive” Can the authors be more specific? Is it restricted the places where GEOS-Chem predicts surface mixing ratios > 1 ppb, a cut off that was identified earlier in the manuscript?

Response: We agree that we were not explicit enough here. Since at this stage we have performed retrievals, the sensitivity is directly computed from the retrievals as the DOFS we used the DOFS \geq 0.5

We changed, “where TES is sensitive” To: “where TES is sensitive with a DOFS \geq 0.5”

10. Page 16038, Lines 12-13 Why are the emission databases used here different from those described in Section 2.1.4?

Response: The a priori profiles and constraint matrices of section 2 were estimated several years earlier than the more recent effort described in section 3.3. The earlier version of the model relied on the GEIA inventory for values throughout the globe. More recent versions overwrite these values in certain areas, such as Europe and China. As the initial calculation was solely for the purpose of describing the range of NH₃ profile values and variability, this is probably sufficient.

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11. Page 16038 Lines 20-25, Is it possible that GEOS-Chem is converting too much of the ammonia to particle phase ammonium?

Response: The reviewer is correct that errors in partitioning could contribute to the low modeled values of NH₃ compared to TES, but this seems unlikely to be a dominant source of error for a few reasons. First, some comparisons of the model to surface observations in the US of total ammonium (gas-phase NH₃ + particle phase NH₄) indicate that the model values of total ammonium are still too low in many regions. Additional calculations over areas such as India indicate that the model is already saturated with NH₃, and ammonium formation is limited by lack of nitric acid rather than NH₃. Levels of sulfate aerosol in the model could also modulate the amount of ammonium, however further studies comparing GEOS-Chem sulfate concentrations to surface observations, while far from being in perfect agreement, were not broadly inconsistent in a way which would lead to the under-estimating of NH₃ in the model noted here. Still, the incorporation of additional aerosol-phase observations to assess these possibilities will be the subject of a subsequent paper in progress.

12. Page 16041, Lines 2-3 ‘or some strong sources have less seasonal dependence.’ should read ‘or some strong sources that have less seasonal dependence.’

Response: Agreed and modified as suggested by Referee.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 16023, 2011.

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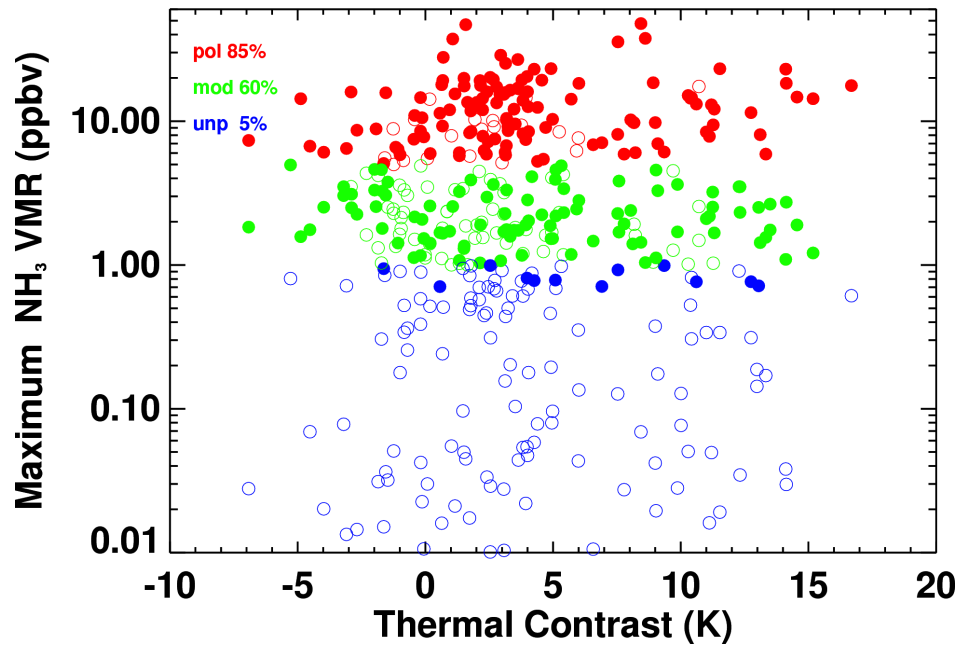


Fig. 1. Figure 6

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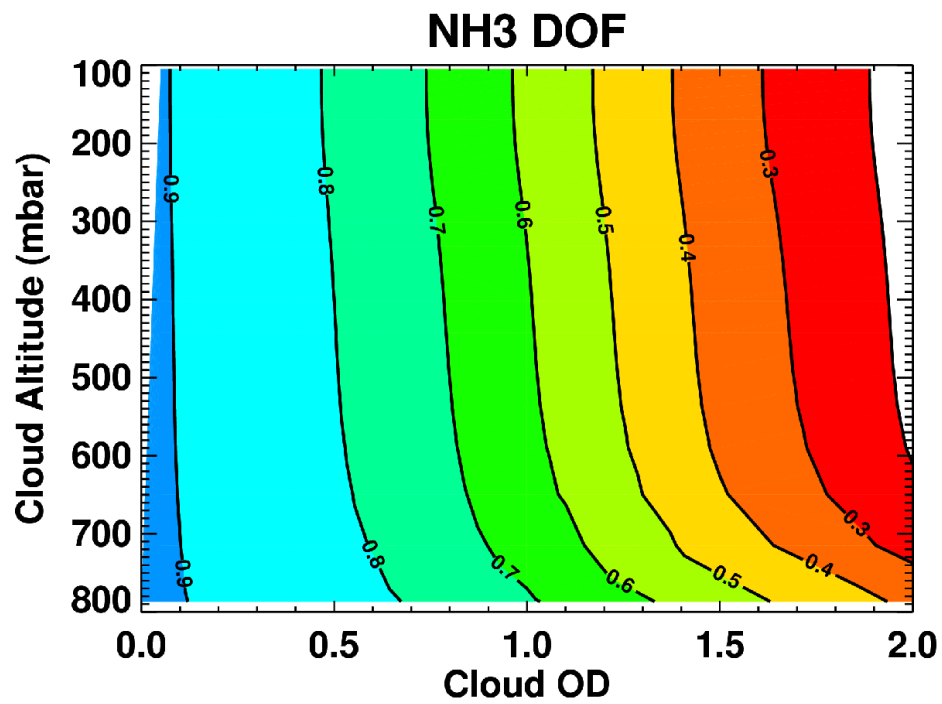


Fig. 2. Figure 7

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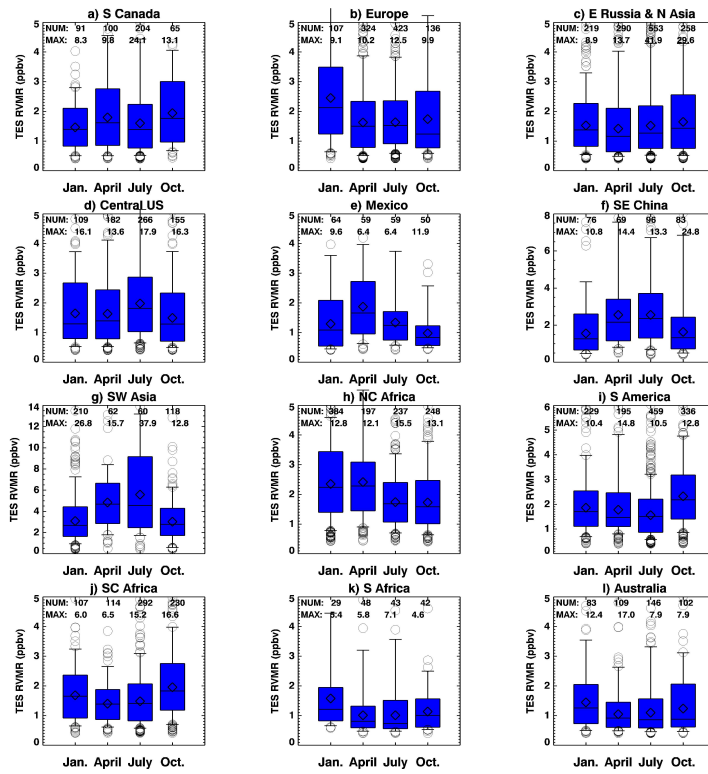


Fig. 3. Figure 15

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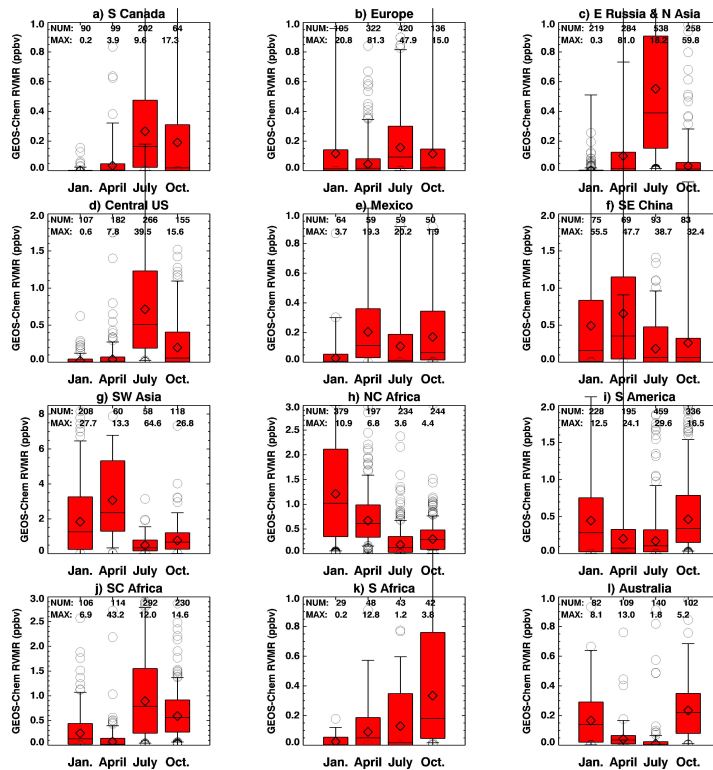


Fig. 4. Figure 15

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