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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The global tropospheric ammonia distribution as seen in the 13 year AIRS measurement record" *by* J. X. Warner et al.

J. X. Warner et al.

juying@atmos.umd.edu

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Reviewer #1 (Comments to Author):

Authors: We are grateful to the reviewers for their constructive comments that we believe have helped us to strengthen the manuscript. Below we include the original review, and we respond to each comment line-by-line.

Reviewer: In this manuscript, observations made from the Atmospheric Infrared Sounder (AIRS) in the spectral region between 860 and 967 cm-1 are used in a forward model to deduce NH3 concentrations, averaging kernels and degrees of freedom for signal. As the authors note, other satellite-borne IR spectrometers, including IASI and TES, also provide retrievals for ammonia on a global scale. The challenge for all

such measurements is that NH3 mixing ratios are typically at a maximum within tens or hundreds of meters of the Earth's surface, whereas the retrieval is most sensitive at higher altitudes. Further compounding the challenge is that chemical transport models often struggle to reproduce the diel behaviour of NH3, and its vertical distribution in the atmosphere in the few regions where in situ measurements are available for comparison.

Authors: We agree with the reviewer that it is challenging to measure NH3 on a global scale, which makes it more valuable to use satellite products. We do not agree with the statement that the retrieval is most sensitive at higher altitudes, because the spectral range we use is in the atmospheric window region, and with NH3 concentration typically at a maximum within tens or hundreds of meters of the Earth's surface (as stated by the reviewer), we can see through the "clean" atmospheric column into the lower troposphere with best sensitivity at 918 hPa.

Reviewer: Throughout the paper, the authors conflate high observed volume mixing ratios (VMRs) retrieved at 918 hPa with high emissions at that location. It is not necessarily the case that high VMRs observed aloft correspond to high emissions directly below the retrieval, especially given the importance of wildfires to high signal at 918 hPa. Even if the VMR is related to local emissions, the retrieved quantity will also depend on the degree of vertical mixing and the impacts of sinks such as deposition and gas-particle partitioning. I think the language used in the manuscript is somewhat misleading since it implies that elevated VMRs at 918 hPa are uniquely associated with elevated emissions. Since the authors restrict themselves to only three pollution scenarios to serve as a priori profiles, it may be a convenient shorthand but it can lead to misleading statements about the interpretation of the retrievals.

Authors: This is our oversight. We have carried out model studies over China (not shown) that demonstrate the NH3 emissions and the concentrations are linearly correlated, however, not at the same rate. We have made correction to change the term 'emission' into 'concentration' in all the relevant locations in the manuscript.

Reviewer: In Section 3, the authors use in situ measurements from the DISCOVER-AQ field campaign in California during the winter of 2013 to evaluate retrieval profiles. These aircraft profiles are useful, in that they occur in a significant source region, which may be expected to correspond to the 'high pollution' a priori. On the other hand, there is significant heterogeneity, with individual aircraft profiles corresponding to the same satellite retrieval measuring mixing ratios that differ by an order of magnitude close to the surface. This section ends without a clear statement about the quality of the retrieval methodology, as evaluated using this comparison.

Authors: We added the following sentence at the end of the session: "Nonetheless, the vertical profiles show good agreement ($\sim$5 – 15%) between AIRS NH3 and the in situ profiles in the examples given above."

Reviewer: In Section 4, the authors apply the methodology to the globe from 2002-2015. As they state, interpreting the analysis requires not only consideration of the average VMR at 918 hPa, but also the frequency of elevated 'emissions' (actually VMRs > 1.0 ppbv at 918 hPa), and also the retrieval DOFs. It appears that many of the regions with the highest average VMRs are in places with infrequent occurrences of high emissions, probably related to episodic wildfires. Given that the relationship between VMR at 918 hPa and emission is likely very different for agricultural and wildfire emissions, it becomes challenging to use the retrievals to constrain the global budget of NH3. Furthermore, the authors state that regions where DOFs are never above 0.1 are excluded from the analysis. But what about regions where DOFs are < 0.1 the majority of the time, but are occasionally impacted by wildfire. Is the average VMR reported for that pixel simply the average of the high signal episodes, or the average of the entire time period, in which case a significant fraction of the time the retrieval is probably indistinguishable from the a priori?

Authors: This approach with related thresholds was applied to illustrate where the major global NH3 sources are; was not intended to constrain the global budget of NH3. Also once a region is identified as being persistent sources using the frequent

occurrences thresholds, we use all data (not just high signal episodes) in the average in that region.

Reviewer: I did not find Figure 6, and the associated discussion to add significantly to the manuscript. It is to be expected that fertilizer use and animal husbandry will dominate 'persistent sources' of NH3 on a global scale, so what new information is gained here?

Authors: We intend to show here that our observations are consistent with prior knowledge.

Reviewer: Specific Comments Lines 45-47 – References would be good for these statements, particularly that idea that NH3 deposition increases emissions of CH4.

Authors: We have added references and modified into the following sentences: "Ammonia deposition modifies the transport lifetimes, and deposition patterns of sulfur dioxide (SO2) and nitrogen dioxide (NOx) (Wang et al., 2008; Henze et al., 2012). Additionally, ammonia increases the concentrations of the greenhouse gases nitrous oxide (N2O) (EPA, 2011) and, together with NH4+ content in soils, NH3 is involved in CH4 production and release (Fowler et al., 2009)."

Reviewer: Lines 52-72 – In the second paragraph of the introduction, it's difficult to tell if the authors are stating that they used the NH3 fields generated in the Park et al., 2004 study, or whether they ran GEOS-Chem themselves using the methods described in Park et al., 2004. Later, it is mentioned that v9-02 was used – this information should be clarified in the introductory paragraph.

Authors: We moved the sentence "We used the simulated NH3 fields from GEOS-Chem as the retrieval a priori for this study." to the front of the paragraph to clarify the purpose of this model discussion.

Reviewer: Lines 9-99 – This sentence is a bit confusing. Does the 'both' in 'if both are large enough' refer to the concentrations and thermal contrast, or retrieval and radiative

transfer model runs. One assumes the former, but it's hard to tell from the sentence.

Authors: We changed the sentence to "..., if both of the NH3 concentrations and thermal contrast are large enough, ..."

Reviewer: Line 142 – Sentence refers to the contributions of co-authors Strow and Hannon, but Hannon does not appear in the author list of this manuscript

Authors: Changed to co-author Strow and team.

Reviewer: L185-187 Why is the a priori developed for 2003-2012 when it is applied for observations between 2002-2015?

Authors: The a priori was developed at the beginning of algorithm development (in 2012), but we extended our data product to current time. A priori information shows our current knowledge of the data range, which has not changed significantly between 2012 and 2015. Therefore, there is no need to update the a priori information as we continue processing new measurements.

Reviewer: L186 and Figure 1. I find the use of 'level' for the three different versions of the a priori somewhat confusing because it makes me think of vertical levels. Perhaps using the term 'emission scenarios' instead of 'emission levels' would be more clear? Also, it would be interesting to know if the three scenarios differ in terms of shape or just overall levels. This could be shown with an additional panel in which each scenario is shown normalized to the surface concentration.

Authors: We changed "levels" to "scenarios". We used a large number of NH3 profiles from GEOS-Chem model output to derive the statistic properties of these a priori profiles. The shapes of the three scenarios are different but follow the model property.

Reviewer: L196-198 How significant are the adjustments and extrapolations mentioned here?

Authors: It is not possible to measure the near surface high NH3 values as described

by the model, so we modified the a priori profile shape to suit satellite measurements better.

Reviewer: Figure 3 – it's too difficult to distinguish between the solid and dashed green lines in the figure panels

Authors: We changed the green dashed lines to blue.

Reviewer: L 318-320 Can the authors clarify why they excluded the nighttime retrievals carried out at 01:30? Measurements of NH3 in the residual layer would be valuable.

Authors: Additional studies are needed to specifically address NH3 retrievals at nighttime and will be included in a future publication.

Reviewer: Section 5 - Can the seasonality in the retrievals be uniquely attributed to seasonality in column NH3 or the VMR at 918 hPa?

Authors: The seasonality is for the VMR at 918 hPa.

Reviewer: Technical comments L79 remove 'the' before Beijing'âĂĺ

Authors: Corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 35823, 2015.