Responses to Review of Anonymous Referee #2

We would like to acknowledge the anonymous referee for his/her useful remarks and comments which have helped to improve the manuscript. All comments have been addressed as detailed hereafter in blue.

Interactive comment on "Towards validation of ammonia (NH_3) measurements from the IASI satellite" by M. Van Damme et al.

Anonymous Referee #2

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The following is an review of the paper titled, "Towards validation of ammonia (NH3) measurements from the IASI satellite", authored by Van Damme et al.. The paper is well suited for AMT and provides valuable insight on the quality of the IASI NH3 retrievals through various and extensive comparison datasets. This information is very valuable to the expanding satellite NH3 user community (i.e. air quality monitoring and modeling). Finding coincident observations that are representative of the satellite observations that can be used to "validate" satellite retrievals can be very challenging. I recommend that the paper be published in AMT providing that more clarity is provided on the capability of the IASI NH3 measurements in order to better interpret the comparison results, especially at the surface and under "clearer" conditions where IASI has limited sensitivity.

A) General Comments

1) The main goal of this comparison paper is to provide the user with insight on the uncertainty and biases of the Van Damme (2014) IASI-NH3 retrieval approach given the limitation of the direct validation observations. Often in the paper the comparison results are provided only for retrievals that have errors less than 100%. This brings up the question of how well the IASI-NH3 retrievals performs when all the possible data is included as the filtering can create biases, and users will often like to utilize all available retrievals set for a given purpose. I don't suggest that the authors need to perform any additional analysis (like was nicely done in Table 3 that showed the filtered and unfiltered results) as this might be a large effort, but suggest that it would be very valuable to at least add in the percentage of all the possible IASI pixels that were used (filtered) for each statistic that is reported. Also, please justify why a 100% error criteria was selected as a filter.

The main goal of this paper is to explore the challenge that represents the validation of satellite NH3 measurements given the limited independent NH3 measurements available and their associated issues of representativeness for the satellite observations. This is achieved by extracting the largest information on uncertainty and possible biases of the IASI-NH3 measurements by comparing them with airborne and ground-based data sets.

The criteria at 100% has in fact been arbitrarily chosen to post-filter the non-reliable satellite data on a monthly basis, but after careful check that this was not biasing the distributions.

The percentage of monthly means with error below to 100% for Europe, China and Africa for each period under consideration is respectively 85% (1337 observations with a relative monthly mean error below 100% over 1577 observations collocated with NEU network), 96% (1149/1196), 95% (367/387).

On a single observation basis (non-averaged) and at global scale for all 2011 spectra over land, we obtain 88% of the IASI-NH3 measurements associated with a relative error below 100% (12 399 037 observations over 14 067 142).

So in general, the 100% criteria does not exclude too many data points, and probably those that correspond to bad retrievals.

2) In Section 2.1 it would be very helpful in interpreting the comparisons if additional IASI-NH3 measurement characteristics were provided in the article in addition to just

“The detection limit of NH3 depends on both thermal contrast and vertical distribution of NH3, an illustration of this can be found in Fig.5 of Van Damme et al. (2014)”. It is well known that the sensitivity of infrared observations depends on the condition of the atmospheric state being observed (i.e. temperature, thermal contrast, atmospheric concentrations, clouds, etc.). However, what are the common conditions under which IASI would not be able to detect NH3 (i.e. NH3<??, thermal contrast < ??), and what part of the column is IASI typically sensitive. For example, Clarrise et al. (2010) noted that, “Based on this analysis we can put a lower bound on the detection of ammonia to surface concentrations of about 3ppb for large thermal contrasts.” Since both the vertical sensitivity in the column and the minimum detectability are fundamentally governed by the line-by-line radiative transfer, and not the specific inversion retrieval approach, simple forward model Jacobians (either analytic or finite difference) show the general lack of sensitivity at the surface, and when there is typically no detectable signal in the IASI spectra. This is important as it indicates the dependence on additional information not directly provided by the satellite observations themselves (i.e. profile shape, etc.) in the inversion. For example, satellite comparisons at the surface with ground-based observations under conditions when IASI is not sensitive to surface concentrations provides some insight on the assumed profile shape (correlation in height) utilized in the retrieval, but does not validate the actual information provided by IASI observations themselves. Thus, in order to help explain the potential differences with other observations the IASI sensitivity components should be explicitly stated. For example, this type of information would also help support statements such as (P12139:line 5-7) “. . .for such small concentrations in a region associated with low thermal contrast, cannot be reliably detected by IASI.” Also, the smaller variations in the IASI-NH3 observations can be due to a number of potential reasons as noted in the article. However, it should also be noted that the inherited minimum detectability of satellite infrared observations under cleaner conditions, and the lack of sensitivity to surface concentrations might help explain the reduction in the variation in the satellite observations due to non-detects, especially if the data is filtered.

In this paper, we have decided to not repeat too much the content of previous publications by referring to them. In Figure R3 we provide figure 5 from Van Damme et al., ACP 2014, but converted into surface concentrations (red, µg NH3/m³) and vmr (green, ppb). We made the choice to not add this figure in the publication as it is not the most relevant quantity to be extracted using the Hyperspectral Range Index (HRI) retrieval method applied on the IASI spectra. It can be seen that these minimum detectability values (significant below 2 sigma in HRI) are lower than the 3 ppb provided by Clarisse et al. (2010) for large thermal contrast.

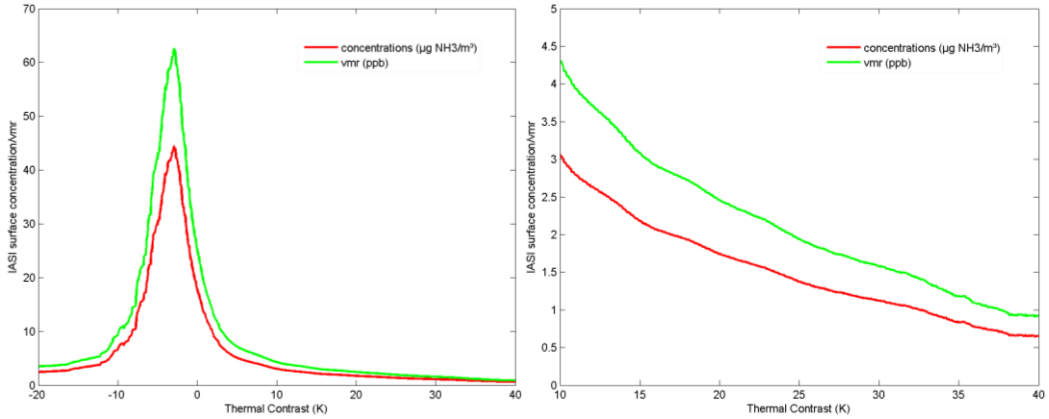

Figure R1: Lowest possible detectable NH3 surface concentration (red, µg NH_3/m^3) and vmr (green, ppb) for all thermal contrast taken into account in the look-up tables (left) and for larger thermal contrast (right). The values are those for which the retrieved column would be significant below 2 sigma in HRI, converted into surface abundance using the land profile taken into account for the forward simulations.

To clarify this point, we have added at L229: “As an example of detection limits on individual observation, a NH_3 retrieved column is considered detectable when the column is above

$9.68x10^{15}$ molec/cm² (1.74 µg NH3/m³) given a thermal contrast of 20K while the column should be larger than 1.69 $x10^{16}$ molec/cm² (3.05 µgNH3/m³) for 10K."
It is worth noting that the error estimate calculation includes the dependency on the thermal contrast and therefore the sensitivity of the retrieval method. It is also important to note that we do not retrieve a vertical profile and that we are therefore always depending on the assumed profile shape.

B) Specific Comments and Technical Edits

1) P12127:line 13: comma before "but" and remove "that"
We agree with the referee and made the suggested change.

2) P12128:line 26 to P12129:line 5: There are recent global GEOS-Chem results that contain both bi-directional and diurnal nh3 profiles, so these recent improvements are not limited to regional models as implied in these lines.
For example: Liye Zhu, Daven K. Henze, Jesse O. Bash, Karen E. Cady-Pereira, Mark W. Shephard, Gill-Ran Jeong, Robert W. Pinder, Mingzhao Luo. (2012) Evaluation of bi-directional ammonia exchange with the GEOS-Chem model. AGU Fall Meeting, San Francisco, CA.
There is also new ACPD article submitted on this work.
We were not aware of this work and based our introduction taking into account published work (with very recent references presented in this paragraph). We therefore think that the original statements are currently correct. We are looking forward to being able to take into account these global results when they will be available to us.

3) P12129: line 19: The sentence might read a little better to put the "Therefore" at the start of the sentence. For example, "Therefore, the spatial coverage of these networks and campaigns is strongly . . ."
We agree with the referee and made this change.

4) P12130: line 12: should also include the NPP CrIS as an instrument with high density of observations (i.e. Shephard and Cady-Pereira, 2014). Shephard, M. W. and Cady-Pereira, K. E.: Cross-track Infrared Sounder (CrIS) satellite observations of tropospheric ammonia, Atmos. Meas. Tech. Discuss., 7, 11379-11413, doi:10.5194/amtd-7-11379-2014, 2014.
We agree with the referee (noting that this work was not published (even in discussions) when we submitted this manuscript) and have change the sentence which now read (L133-139): "Satellite sounders with a high spatiotemporal resolution, such as the Infrared Atmospheric Sounding Interferometer (IASI) or the Cross-track Infrared Sounder (CrIS, Shephard and Cady-Pereira, 2014), also offer the opportunity to identify area-specific and time-dependent emission profiles (Van Damme et al., 2014b)."

5) P12130: line 18-19: "While NH3 satellite measurements have started to be used by models, their validation has yet to be performed." Although there has been very little "validation" analysis, this general statement is not completely true. Since satellite "validation" in this paper includes ground-based surface comparisons, there are satellite NH3 comparisons with ground-based observations (i.e. Pinder et al., 2011; Shephard and Cady-Pereira, 2014). Thus, either change the sentence to be specifically". . .IASI-NH3 satellite measurements. . ." or state that there are very few "validations" and reference previous examples.
Pinder, R. W., J. T. Walker, J. O. Bash, K. E. Cady-Pereira, D. K. Henze, M. Luo, G. B. Osterman, andM. W. Shephard (2011), Quantifying spatial and seasonal variability in atmospheric ammonia with in situ and space-based observations, Geophys. Res. Lett., 38, L04802, doi:10.1029/2010GL046146.

Again and similarly to the answer of the previous referee review, we do not claim to validate IASI-NH_3 columns with this study. As far as we know, TES and CrIS satellite based NH3 data set have not been validated yet neither.
In order to avoid misunderstanding we have followed the referee's suggestion and have added to the sentence (L147): "even if sparse comparisons have already shown their consistency (e.g. Pinder 2011; Shephard and Cady-Pereira 2014)."

6) P12132: lines 17-19: What impact does pixels with 25% cloud cover have on the NH3 retrievals? For example, depending on the optical depth and altitude of the clouds this can significantly impact the weak spectral signature of NH3, especially if the cloud radiances are taken into account in the forward model/retrieval.
There is no doubt that signal of pixels with 25% cloud coverage might be converted into underestimated columns. However, as the clouds mainly impact the baseline, the HRI-based retrieval method will be weakly impacted. It is worth noting also that the look-up-tables used to convert the IASI signal into IASI-NH_3 total columns are based on forward simulations made at clear conditions.

7) P12133: line 26: suggest rewording sentence ". . .this network which have started to provide. . ." to something like, ". . .this network, which started providing. . ."
We agree with the referee that this sentence was not clear. We have changed the sentence to (L275): "From this network, we use the 28 sites which have started to provide…". It is not exactly what the referee proposed but we think it express better the fact that we use, from all sites, the 28 sites having started to provide measurements no later than 1 March 2011.

8) P12138: line 10: suggest rewording "Overall, however, IASI observes. . .." to something like "However, overall IASI observes. . .."
We agree with the referee and made that change.

9) P12114: lines 16-26: Again the underlying reason, which incompasses some of the reasons provided for the high bias at surface under "clean" conditions, is the simple fact that no infrared satellite instrument is not sensitive to NH3 under these conditions, especially at the surface.
We agree with the referee. However, when IASI has low sensitivity, we retrieve NH3 columns with large associated errors, which are then taken into account in the weighted averaging procedure. In order to deal with the comment of the referee, we have changed the sentence to (L588-592): "Applying weighted averaging of IASI columns using the relative retrieval error tends to favor the largest concentrations as the lower values are associated with higher error estimates for the same value of thermal contrast (see Van Damme et al. (2014a))."

10) P12142 lines 2-3: suggest putting a column before the two "but" in these sentences.
We agree that these sentences read better with a comma before the two "but" and add them.

11) P12142 line /8: The sentence might read better to mover the "however" to put it at the start of the sentence with a comma after it.
We agree with the referee and made that change.

12) P12146 lines 14-15: As mentioned before, not just "weak signal", but also cases with no nh3 signal at all, which will bias the statistics.
It is not really what we mean. Our intention with this sentence is to underline the fact that with the same thermal contrast, as the retrieval error is depending on the NH_3 abundance, more weight will be given to the higher NH_3 columns. We agree of course with the referee that in the case of no NH_3 signal at all, the error will be high.

13) P12147 lines 4-10: ". . .-the most relevant quantity available from the satellite measurements. . ." This is not true in general. This is likely the case for this Van Damme (2014) IASI-NH3 retrieval approach, but is not the case for other NH3 retrieval approaches. For example, an optimal estimation retrieval approach with a quality forward model can directly utilize aircraft profiles in comparisons (i.e. Rodgers, 2000) and explicitly take into consideration the satellite vertical resolution and the information content provided by the satellite observations themselves. Since optimal estimation retrievals are commonly applied (even to IASI), these lines should be removed or modified to explicitly state that column data are require for the Van Damme (2014) NH3 type of retrieval. Rodgers, C. D.: Inverse Methods For Atmospheric Sounding: Theory and Practice, World Sci., Hackensack, N. J., 2000.

We thought that it was clear that this was applying to the retrieval method used here as the whole sentence read as: "All statistical results were shown for concentrations (or vmr) and do not allow the validity of the IASI derived columns to be assessed, which is – considering the limited vertical sensitivity achievable – the most relevant quantity available from the satellite measurements."

In order to avoid confusion we have changed the last part of the sentence to (L820) "the most relevant quantity available from these satellite measurements."