Interactive comment on "Global distributions and trends of atmospheric ammonia (NH3) from IASI satellite observations" by M. Van Damme et al.

Anonymous Referee #2

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We would like to thank the anonymous referee for his/her positive and encouraging remarks and for his/her interesting highlighted points. We have carefully addressed all of them and the response to each comment is provided below (in blue).

1 Overview
2 Comments and questions

The manuscript by Van Damme et al., "Global distributions and trends of atmospheric ammonia (NH3) from IASI satellite observations," marks a significant step forward in mapping observations of NH3 from space. This new dataset includes comprehensive spatial coverage with quantitative error estimation that will be a valuable resource for reducing uncertainties in our understanding of the sources and fate of reactive nitrogen. The only aspect that is sorely missing is some discussion of the vertical sensitivity of the retrieval, which is critical for validation of the IASI NH3 product and application of this product for constraining model simulations. Overall the manuscript is fairly clear and well organized; it will be suitable for publication in ACP following consideration of this point in addition to the comments below.

It is clear that the vertical sensitivity of the infrared satellite measurements in the lower troposphere is key to be able to interpret the observations. Unfortunately, infrared measurements from space are not the most suitable to retrieve trace gases profiles. To build this new and improved dataset based on IASI observations, we have chosen to use only two profiles to retrieve NH_3 columns: one for land and one for sea. In the absence of more accurate data, this we believe is a sensible approach, which still enables to take into account the large land/sea differences and allows us to provide one coherent and multiple years dataset.

To test the dependency on the vertical profile used to build the LUTs, we have taken the observations over land on the 15 August 2010 and used the LUT made for sea observations - built with a transported profile - to retrieve the concentrations. Confronting the two set of retrieved values and using the same type of fit than the one used in fig. 9, we obtain a regression line ($y = 0.5\,x + 2.9\ 10^{15}$) presenting a factor 2 between the retrieval with a profile having its maximum at the surface and one having its maximum at 1.4 km. The Pearson's r coefficient for this regression is equal to 0.93. Given the fact that the two different profiles correspond to two extreme scenario's (ammonia profile directly above the emission source vs long range transport profile), it is reasonable to conclude that in the worst case scenario errors related to the usage of a fixed NH_3 profile will be inferior to a factor two. We agree that the profile dependency is a drawback of the HRI based processing scheme, but this is something which at present all NH_3 retrievals from space suffer from. We have now introduced and discussed this sensitivity study on the vertical profiles in the paper.

- A significant motivating factor for this work is improving NH3 emissions estimates and Nr model simulations. In order to compare the values presented here with models, it is necessary to know how the retrievals depend upon the vertical distribution of NH3. Can the authors provide representative averaging kernels for their data, or at least discuss their findings in light of the sensitivity of the retrieval to NH3 concentrations at different altitudes? Further, have values from IASI ever been directly compared to NH3 observations, from air craft or in situ measurements? Could the authors provide the necessary information regarding vertical

sensitivity of the retrievals to make such comparisons possible?

(See also our response to the previous comment)
This improved method is not providing averaging kernels (AVK), as it is not fully based on the Optimal Estimation Method for the inversion step. In fact, the previous processing chain FORLI was providing AVK but their uses have shown difficulties. They were too low and not representative of the information available inside the spectra. It is why the FORLI dataset has been mainly used qualitatively (Heald et al., 2012; Kharol et al., 2013). We are confident that the errors associated to each retrieved NH3 column with the new product can be adequately used for model comparisons, especially as above land the NH3 will be mainly confined at near-surface level.

Regarding the comparison of the IASI-NH$_3$ dataset with independent observations, this would indeed be very important but this is definitively beyond the scope of this paper. First comparisons with correlative measurements have been made and are promising. We expect a dedicated publication of this validation exercise, which is complicated due to the poor spatial and temporal representativity of the in situ surface measurements. . The error associated with each retrieved column will be essential information to perform the comparison adequately.

> • Introduction: IASI NH3 was also compared to GEOS-Chem in Kharol et al., ES&T, 2013, and TES NH3 measurements were used to constrain emissions with GEOS-Chem in Zhu et al., JGR, 2013.

The references to Kharol et al., 2013 and Zhu et al., 2013 have been included at the end of the introduction.

> • 24307.9: It might be nice to include in Fig 1 a line indicating the region used by TES that is discussed here in the text.

We have included in Fig. 1 the microwindows used by TES.

> • 24310.15 - 20: It's not clear why model profiles are scaled by this amount. Did the LUT start with only a single mean profile over each land-type based on GEOSChem? Were there not actually profiles in the GEOS-Chem simulations spanning the necessary range of concentrations to include in the LUT? If so, is this indicative of a shortcoming with the model simulations, either owing to underestimated emissions or to coarse model resolution?

To obtain the modelled profiles, polluted/transported profiles from one year of GEOS-Chem simulations were averaged. The method is thus indeed based on the same profiles for each land/sea pixel. For simplicity, instead of using the complete set of NH3 profiles from the model, these single profiles have been scaled at various NH$_3$ concentrations to account the variability of columns observed in the atmosphere. This method was used to provide a coherent and multiple year dataset as the IR measurement.

> • 24311.2: It wasn't clear to me why artificially enhanced thermal contrasts are necessary – wouldn't these be present in the data already if they were important? I'm probably just missing something here, but maybe it could be explained a bit more in depth.

We artificially increased the thermal contrast to be able to retrieve NH$_3$ columns for all encountered situations. We had to do that as the atmospheric profiles provided by Chevallier (2001) did not sufficiently cover the observed variability for the large thermal contrast cases.

> • 24312.6: It might be worth mentioning some typical column values here to put these numbers in context.

We agree that a comparison with a typical value was missing. As suggested, we have added a sentence at the end of section 3.2.2, which now reads: "For the more favorable values of TC the IASI measurements should be able to measure NH_3 down to the 10^{16} molec/cm^2 level. As an illustration, this detection limit would allow measuring NH_3 columns year round in the San Joaquin Valley, where we measure columns varying above 10^{16} to 4.3×10^{17} molec/cm^2 for 81 % of the observations (median at 3.2×10^{16} molec/cm^2)."

> • The prominence of the biomass burning regions stemming from single-year events in the five-year mean is striking, and a bit odd. My expectation would be to see signals over persistent source regions (e.g., India) to be much larger than any singe-year event. Is this just an artifact of saturating the color scales at 3 1016, an order of magnitude smaller than the peak values? Could the authors indicate this somehow in the plots?

Both of these hypotheses are right. First of all, columns over India are still higher than in any other place of the Earth, up to $6.4.10^{17}$ molec/cm². The importance of fire events in the five years distribution comes largely from the weighting of the mean by the relative errors. As we have more sensitivity for fire plumes observations, where the thermal contrast is higher and the NH_3 emitted generally more concentrated, the relative error for such observations is lower. That implies a high weight in the averaging following eq. 3. The opportunity to use another averaging method is largely discussed in the response to Referee #1. We have added in the paper an additional figure (Figure 11) presenting averaged distributions weighted using absolute errors and discuss the challenges of averaging data with such large variability in the errors (Section 4.2).

> • Why is the outflow from West Africa so much more pronounced than other regions with much larger hotspots? Is this owing to the meteorology in the region or a lack (relative to more industrialized areas) of NOx and SO2 to react with NH3?

We also expected to see more transport above Bohai and Yellow sea, for instance. Comparing the NH_3 distributions with NO_x and SO_2 distributions is subject of future research but from initial comparison, it seems that the lower concentrations of other reactive species could definitely be one of the explanations for large transport from the western-African coast. The meteorology and especially the wind direction are also responsible for the importance of the transport from West Africa.

> • 24318.12: What is the nature of the source in southeast Calgary?

It is likely agricultural, but industrial sources could also play a role, as some emitters are situated in the south of Alberta province (Environment Canada, 2013a). This hotspot area is reported in national NH_3 inventories as shown in figure 1 (Environment Canada, 2013b).
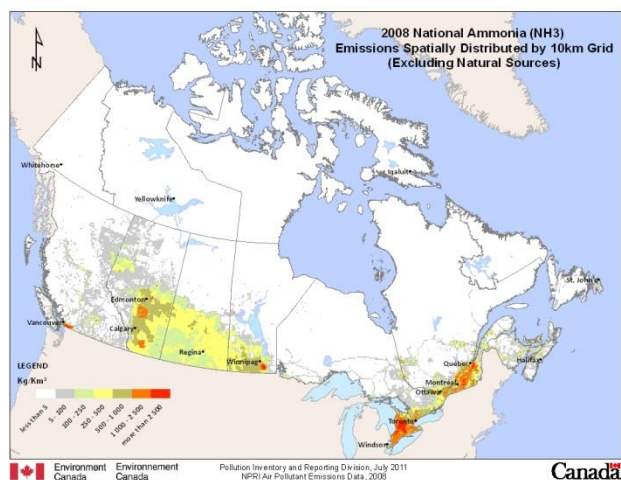
Figure 1: 2008 National NH₃ emissions inventory from Environment Canada (Environment Canada, 2013b).

> • The writing contains some awkward phrases (e.g., "emissions in the atmosphere" or "marked emissions") and grammatical errors (usage of commas). It would be nice if any of the native english speaking authors would be willing to spend an hour smoothing out the language prior to final publication.

The text was checked once again for the English.

<u>References</u>

− Environment Canada: Ammonia emissions by facility, Canada, 2011, http://maps-cartes.ec.gc.ca/indicators-indicateurs/default.aspx?lang=en&mapId=9&xMin=-13662737.90269002&yMin=6270340.489049873&xMax=-11368404.06168278&yMax=7737931.432124867&sr=102100, 2013a.
− Environment Canada: Ammonia (NH3) Emission Density in 2008, http://www.ec.gc.ca/inrp-npri/default.asp?lang=En&n=E788969F-1, 2013b.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 24301, 2013.