

Interactive comment on “Atmospheric ammonia (NH₃) over the Paris megacity: 9 years of total column observations from ground-based infrared remote sensing” by B. Tournadre et al.

Anonymous Referee #1

Received and published: 3 October 2019

Review of Tournadre et al.,

The paper presents an extensive and highly usable data record of FTIR-NH₃. Without any doubt it will be very helpful for future air quality evaluation and model and satellite validations. There are not many locations in the world with such an extensive and long term NH₃ record, and only a few with instruments with the capability to measure the total column of NH₃ at high temporal resolution. The paper is easy to read but could use some restructuring and editing of the text. The sections on the FTIR retrieval and the comparison with IASI are interesting, but section 3.3 seems added on and could be removed without too much impact to the manuscript. For example PM_{2.5} is barely

Printer-friendly version

Discussion paper



mentioned in the introduction. While comparing FTIR-NH₃ to pm_{2.5} is interesting, a more complete analysis and interpretation using a model will be needed if the authors want to keep the section.

Major comments.

1. The retrieval fits are performed over a very wide window. While the authors claim that this is needed, have tests been performed for smaller windows? Past results with the more high resolution FTIR have shown problems with very wide windows, which was one of the reasons to use smaller micro windows (Dammers et al., 2015). The FTIR used by the UNAM team in Mexico City is also a VERTEX and they have reported succesfull fits with smaller windows (Dammers et al., 2017). A comparison of Figure 2 and 3 (maybe merge the figure?) shows that the strongest signatures in the residual correlate well with the location of the strongest NH₃ lines. While the fits with an SD of 2% are excellent, compared to the weak absorption feature of NH₃ this can still result in a large offset of the NH₃ total columns. If possible add a % based fit and take a look at the % deviation around the NH₃ lines (maybe mark the locations like in Figure 2). In the text the authors mentioned that HITRAN 2008 was used. Dammers et al 2015 and most of the NDACC FTIR teams used HITRAN 2012 in combination with a few CO₂ line adjustments. This can potentially improve the spectral fits.

2. The PROFFIT retrieval seems to be based on a scaling method instead of a full physical retrieval (although I can be mistaken, but as far as I can see it is not mentioned in the text) therefore the choice of the NH₃ apriori profile shape is quite essential. The authors mention in section 2.3 (this should be moved into 2.2 probably) that they use a climatological ammonia profile. Does this profile vary monthly? Furthermore, can some more information (or a figure with the shape) be provided on how it compares to profiles used in other studies/products?, for example the profile used in the IASI-NNv2.X product (Van Damme et al., 2017), the CrIS-NH₃ product (Shephard and Cady-Pereira, 2015), and the NDACC-FTIR retrievals (Dammers et al., 2015). As mentiond by Van Damme et al. (2014) the choice of profile shape in a column based

retrieval can easily vary the results by a factor 2. A similar result seems to be found by the authors as they mention on P6/line 22-24 with a relative different of +20%. What makes the MIPAS profile optimal in this sense? Did the other tested apriori produce worse fits?

3. The averaging kernel or observational operator are an essential piece of information but are completely missing in the text. The OASIS-NH3 instrument should be superior in its sensitivity to the lower boundary layer compared to satellite measurements. A figure and short discussion of the (total column) averaging kernel can go a long way in helping us understand where the sensitivity of the retrieval lies and why there are differences compared to IASI.

4. This brings us to the comparison of OASIS-NH3 to IASI-NH3. The authors reference the results in Dammers et al., 2015 but that study focussed on an older version of IASI, IASI-LUT. Dammers et al., 2017 reports the results using a more recent version of IASI-NH3, IASI-NNv1 (Figure A1). The slope of $S=0.96$ for that product is a lot better than the reported $S=0.6$ for the older product. Van Damme et al., 2018 also state that the most recent version of IASI-NN shows even better results and a lower bias for higher total columns, which would mean we can expect a better comparison. One of the reasons can be found in the absence of the use of an averaging kernel to adjust the IASI total columns to the same playing field. The current comparison can be seen as incomplete as its uncertain where the sensitivity of both instruments lie, and potentially we're comparing the NH3 in the mixing layer to half the boundary layer or the effect of a different apriori (shape).

5. The authors show a initial comparison of OASIS-Nh3 to nearby pm2.5 measurements. While this is interesting it feels somewhat out of place. PM2.5 is barely mentioned in the introduction and only pops up at the end of section 3. Furthermore, most facts are referenced from other studies and the improvement that this study brings, both the high temporal resolution of the FTIR and the vertical total column, are not really used in the analysis. If the authors want to keep the section on PM2.5, an improved

[Printer-friendly version](#)[Discussion paper](#)

comparison will be needed, with for example the help of a model for interpretation. The in review study by Viatte et al., 2019 for example, shows similar results with a more extensive analysis of the Ile de France region.

6. Something that the author could add instead (but not essential to the text!) is an initial analysis of the diurnal variability, which should not take too long to produce. The authors did excellent work on getting such a long dataset and have around 5000 measurements spread over 9 years, which accounting for overcast days would mean around 5-10 measurements a day. Spread out every 15 minutes this must show some diurnal variability of the NH₃ total column concentrations (for example split by season) and I for one would be very interested to see that instead of a comparison to PM_{2.5}.

Minor comments and edits.

1. Split section 2 in 2.1 for FTIR, 2.2 with a description of IASI, 2.3 with a description on PM_{2.5}. This will improve the readability and is easier for reference of retrieval characteristics, uncertainties etc. 2. Maybe move section 3.2 up before the comparison with IASI. First completely describe the dataset and variabilities before moving to the comparison with IASI. This can help in the interpretation of any differences between the two. 3. Section 3.1: The authors choose a collocation criteria of 15 km and 30 min while the study that they compare their results with (Dammers et al., 2016) uses 50 km and 90 minutes. Do your results change a lot when using those criteria? Using wider criteria should increase the number of observations, as only 50 measurements out of 5000 initial measurements remain.

Some smaller edits:

1. P2 L21, there have been several studies recently covering the lifetime of NH₃. If possible reference Lutsch et al., 2016, Van Damme 2018 and Dammers et al., 2019.
2. P3. L 13: add some examples of networks with high temporal resolution measurements (for example LML in the Netherlands, Volten et al., 2013)

3. P3. L20: the correct reference for CrIS would be Shephard and Cady-Pereira 2015. GOSAT also has a NH_3 product: Someya et al., 2019.

4. P3. L28-31, not important for the intro, move to dataset section.

5. P8. L20-21. Although I somewhat agree with the statement, the underestimation can also be caused by other sources. Also the averaging kernel/observational operator has not been applied therefore the results can not be directly compared to the results in Dammers et al., 2016. Explore some further causes of the underestimation (apriori choice) or show some supporting proof that the sensitivity is the cause (which should somewhat be resolved by the use of the averaging kernel).

References:

Dammers, E., Vigouroux, C., Palm, M., Mahieu, E., Warneke, T., Smale, D., Langerock, B., Franco, B., Van Damme, M., Schaap, M., Notholt, J., and Erisman, J. W.: Retrieval of ammonia from ground-based FTIR solar spectra, *Atmos. Chem. Phys.*, 15, 12789–12803, <https://doi.org/10.5194/acp-15-12789-2015>, 2015.

Dammers, E., Shephard, M. W., Palm, M., Cady-Pereira, K., Capps, S., Lutsch, E., Strong, K., Hannigan, J. W., Ortega, I., Toon, G. C., Stremme, W., Grutter, M., Jones, N., Smale, D., Siemons, J., Hrpcek, K., Tremblay, D., Schaap, M., Notholt, J., and Erisman, J. W.: Validation of the CrIS fast physical NH_3 retrieval with ground-based FTIR, *Atmos. Meas. Tech.*, 10, 2645–2667, <https://doi.org/10.5194/amt-10-2645-2017>, 2017.

Dammers, E., McLinden, C. A., Griffin, D., Shephard, M. W., Van Der Graaf, S., Lutsch, E., Schaap, M., Gainairu-Matz, Y., Fioletov, V., Van Damme, M., Whitburn, S., Clarisse, L., Cady-Pereira, K., Clerbaux, C., Coheur, P. F., and Erisman, J. W.: NH_3 emissions from large point sources derived from CrIS and IASI satellite observations, *Atmos. Chem. Phys.*, 19, 12261–12293, <https://doi.org/10.5194/acp-19-12261-2019>, 2019.

Lutsch, E., Dammers, E., Conway, S., & Strong, K. (2016). Long-range Transport of

[Printer-friendly version](#)[Discussion paper](#)

NH₃, CO, HCN and C₂H₆ from the 2014 Canadian Wildfires. *Geophysical Research Letters*, (43), 8286-8297. <https://doi.org/10.1002/2016GL070114>.

Shephard, M. W. and Cady-Pereira, K. E.: Cross-track Infrared Sounder (CrIS) satellite observations of tropospheric ammonia, *Atmos. Meas. Tech.*, 8, 1323–1336, <https://doi.org/10.5194/amt-8-1323-2015>, 2015.

Someya, Y., Imasu, R., Shiomi, K., and Saitoh, N.: Atmospheric ammonia retrieval from the TANSO-FTS/GOSAT thermal infrared sounder, *Atmos. Meas. Tech. Discuss.*, <https://doi.org/10.5194/amt-2019-49>, in review, 2019.

Van Damme, M., Clarisse, L., Heald, C. L., Hurtmans, D., Ngadi, Y., Clerbaux, C., Dolman, A. J., Erisman, J. W., and Coheur, P. F.: Global distributions, time series and error characterization of atmospheric ammonia (NH₃) from IASI satellite observations, *Atmos. Chem. Phys.*, 14, 2905–2922, doi:10.5194/acp-14-2905-2014, 2014.

Van Damme, M., Whitburn, S., Clarisse, L., Clerbaux, C., Hurtmans, D., and Coheur, P. F.: Version 2 of the IASI NH₃ neural network retrieval algorithm: near-real-time and re-analysed datasets, *Atmos. Meas. Tech.*, 10, 4905–4914, <https://doi.org/10.5194/amt-10-4905-2017>, 2017.

Van Damme, M., Clarisse, L., Whitburn, S., Hadji-Lazaro, J., Hurtmans, D., Clerbaux, C., & Coheur, P. F. (2018). Industrial and agricultural ammonia point sources exposed. *Nature*, 564(7734), 99.

Viatte, C., Wang, T., Van Damme, M., Dammers, E., Meleux, F., Clarisse, L., Shephard, M. W., Whitburn, S., Coheur, P. F., Cady-Pereira, K. E., and Clerbaux, C.: Atmospheric ammonia variability and link with PM formation: a case study over the Paris area, *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2019-138>, in review, 2019.

Interactive comment on *Atmos. Meas. Tech. Discuss.*, doi:10.5194/amt-2019-301, 2019.