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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global distributions of nitric acid from IASI/MetOP measurements" *by* C. Wespes et al.

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First of all, we thank the referee for his useful comments on the study, which have helped us clarifying some points and improving the manuscript. In particular, the retrieval errors have been more closely discussed to clarify the analysis of the daily variability in the revised manuscript. Below are our point-by-point responses to his comments and suggestions. Before each response, the reviewer comments have been quoted between [. . .]. Information and corrections have been added to the revised version of the manuscript.

Response to the Anonymous Referee #1:

General comments:

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[At present however, it is very difficult to evaluate really the precision of these data and hence, the extent to which the data set will be useful to investigate particular chemical processes, and to estimate seasonal and interannual variabilities quantitatively. The authors pay very little attention to the uncertainties associated with the data. From Figs. 6 and 8, one sees that the 3sigma variability within a grid box (1_lat, 2_ lon) is of order 100%. It is not clear what is real spatial and daily variability and what comes from the measurement uncertainty.]

As it has been explained in the paper, what we consider as the retrieval errors (error bars associated to the retrieved total columns) are directly derived from the retrieval error covariance and are thus the statistical errors. In most cases, they vary between 5-10 percent at mid-latitudes and polar regions to the chosen threshold value of 32% at equatorial belt. Higher errors found in the equatorial belt are due to the strong water vapor lines interfering with HNO3 lines. High errors are also found at polar regions over very cold or icy surfaces. They are due to particularly weak radiances or emissivity issues over such surfaces. For instance, figure 1 attached present the total errors at the global scale for several days during August. From Figs. 6 and 8 of the paper, the grey areas (3 sigma around the daily mean values) represent the daily variability of the total columns observed for the different selected regions. The total errors associated to the HNO3 total columns are lower than the observed daily variability of HNO3 total columns. We are very confident that we capture real daily variabilities in mid and high latitudes. For instance, the retrieval errors (error bars) associated to the total columns are shown for one selected regions in the Fig. 2 attached. For that particular region, the 3-sigma retrieval error is on average 26% whereas the daily variability, mainly associated to the rapid zonal transport of air masses in polar region, is on average 52% and can be as high as 170%. Numbers have been added in the text to clarify the discussion. In the equatorial belt, on other hand, the 3-sigma retrieval error are indeed much closer to the variability (which is also less than in the polar regions) and the variations should be looked at with more caution. This has also been better explained in the text. We agree with the referee that the errors should have been better referred to in this part of the discussion. This has now been done in the revised manuscript (page 8048).

[The comparison with MLS (Fig. 6) is rather good at first sight but again, at some stations and for some time periods, the differences between both data sets are large and they are not discussed at all.]

We would like to point out that we compare in this Figure the total columns measured by IASI and the VMR measured at 25 km by MLS. The comparison is thus fully qualitative and differences are expected for this reason. Another important source of discrepancies in the comparison lies in the different number of data within the selected boxes. For the MLS observations, we had in the best cases, just one measurement per day in the selected small box regions. Considering the possible daily variability, we cannot expect perfect comparisons between the two datasets. The result that we wanted to highlight here was the similar spatial and seasonal variations between IASI and MLS measurement. Some words of caution have been added in the manuscript to insist on the qualitative character of the comparison.

[The authors claim that it is impossible to make a quantitative validation ‘due to the absence of archived data from ground-based measurements’. Even if data from 2008 are not fully archived yet, it should be possible to get valid ground-based total column data for HNO3 from many NDACC PIs individually.]

At the time the paper was written, there were indeed very few archived data on the NDAAC website. Of course we could have taken contact with individual PI’s to have a first validation dataset but this would have require an additional workload, and probably wouldn’t have been extremely representative (in terms of common methodology, spatial representativeness . . .). We think that a more thorough validation would be preferable. This will obviously be an important next step of the research activities with IASI.

[Figures 3 to 8 (especially 3, 4, and 7) are really too small. It is impossible to distinguish the PV and T contours, and to get a good view on the variabilities and gradients.]

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Figures have been enlarged. Note also that this is an online paper and the electronic images have been provided in high quality so that they can be enlarged to see the details.

Specific comments:

[Abstract, line 15: I do not like the word ‘trends’ to indicate seasonal variations. Trends rather refers to long-term evolutions.]

The sentence has been changed according to the referee’s suggestion.

[Introduction, pg. 8039, line 5: in order to really discern the full seasonal variation, one would need at least a 12 months data set. I wonder why the paper shows no data for January and February?]

During January and February 2008, the HNO3-FORLI was still under development. It is unfortunately operational only since March 2008. If the referee thinks it is needed, we could add the distribution of January and February 2009 which have now been processed.

[Section 2.1, pg. 8040, line 4: IASI provides global coverage twice daily because of its high temporal sampling ’and large swath width’. The swath width is very important to achieve the coverage.]

This has been added in the revised version.

[Section 2.2: pg. 8040, line 17: please explicitly say that this is the HITRAN2004 database. Have the database updates beyond 2004 (before the release of HITRAN2008) been included?]

Indeed, updates beyond 2004 have been included in the HITRAN2004 database which is used for the retrievals. This has been specified in the revised version.

[FORLI-HNO3: the present paper does not give any reference to a previous paper that provides more details about the performances of this software. How do the results from

FORLI-HNO3 compare to those from the line-by-line code atmosphit? Have detailed comparisons been made? Has the reliability of FORLI-HNO3 been demonstrated?]

We did not perform systematic and detailed comparisons between FORLI and Atmosphit. There are several reasons for this: 1. FORLI-HNO3 is a dedicated algorithm for IASI. It was optimized such as to provide HNO3 distributions globally in near-real-time. Performing Atmosphit retrievals on a representative number of locations/days, which would be needed for a proper comparison, would be extremely lengthy. The Figure 3 attached, given only for two days and several locations (spanning a range of HNO3 concentrations) is however comforting in that it shows a fairly good agreement between the two procedures. We hope the referee would agree with this. We have not included the figure in the revised manuscript in order to avoid further lengthy technical descriptions. 2. Atmosphit has not been validated before for HNO3. Moreover, FORLI-HNO3 has a number of upgraded features in the inversion code so that the results are not less secure with this software than for Atmosphit. Accordingly there is not much to be gained from the comparisons. 3. Atmosphit has been used here only to provide insight onto the IASI information content (not directly accessible from FORLI because only a column is retrieved). It should not be considered beyond this purpose for the paper. FORLI-HNO3 provides the reference product from our group for further work, including validation. This part of the future research activities should at best enable to gauge the reliability of the product, rather than an algorithm intercomparison.

[In particular, Section 2.2 pg. 8040, lines 20 to 25: the steps in T and relative humidity seem rather large taking into account that the dependence of the absorption cross sections on these parameters is not linear, and that a linear interpolation in the tables (LUT) is performed. This joins the previous questions about the verification of the performances of FORLI-HNO3.]

Indeed, there is not linear dependence between T (or relative humidity) and the absorption cross sections. However, the steps have been carefully chosen so that the difference between Ainterpolated (P,T) and A (P,T) is around 0.1%. Even by consider-

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ing this error for each level, one remains under the instrumental noise, thus justifying the procedure.

[Section 2.2, pg. 8041, line 7: does the contribution from the downward flux include both the thermal and solar radiation? Please specify.]

The downward flux includes only the thermal radiation since the solar radiation does not contribute in the emission source below 2200 cm-1.

[Section 2.2, pg. 8041, line 15: pixels with a cloud coverage of less than 25% are kept. If there are clouds (within the 25% limit), these will mostly mask the tropospheric column. Can you give an estimate of the possible uncertainty this induces on the total column?]

This threshold has been chosen after several tests. It does not seem to impact significantly on the columns. A similar threshold was selected for the CO retrievals (e.g. George et al. in this issue) while for ozone, which is extremely sensitive to baseline variations, it had to be decreased to about 10 % (ongoing research activity). It is at this stage not possible to better quantify the impact of partially cloudy scenes on the retrievals. Again more validation would be desirable to assess this.

[Section 2.3, pg. 8041, line 19: 'The retrieval scheme used here is the Optimal Estimation Method: the word 'is' is missing.]

This has been corrected.

[Section 2.3, pg. 8042, line 10: 'by limited strong overlapping other trace gases absorption features': do you mean: 'by a limited number of strong overlapping absorption lines from interfering trace gases' ? The statement should be written more comprehensively.]

The sentence has been rewritten according to the referee's suggestion.

[Table 1 and Section 3.1: It looks like you have adopted another Sa matrix in the atmo-

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sphit evaluation than in the FORLI approach. In particular, Sa seems to be diagonal in the FORLI approach with constant values (100%) at all altitudes, while these diagonal elements vary in the Sa adopted for the atmosphit evaluation, and the latter Sa has non-zero off-diagonal elements. Please clarify. In that case, is the atmosphit evaluation then representative of the FORLI results ?]

In FORLI-HNO3, a total column is retrieved by scaling the a priori profile by a single scalar value. The variability for this single parameter was chosen to be 100% in order to allow sufficiently variability on the column; this value is within the range of the diagonal elements of the full Sa matrix. Both referees seem to have misunderstood this; we have rewritten it more explicitly in the paper (page 8042 and 8045).

[Section 3.1, pg. 8044, line 9: do the spectral fit (Fig. 2) show results from the atmosphit fit or from a FORLI fit ? If Fig. 2 represents atmosphit results, then you should comment on the differences between an atmosphit and a FORLI result.]

The spectral fit on Fig. 2 belongs to an Atmosphit retrieval. However, we agree that a FORLI reference should be added here to guide the rest of the discussion. This has been archived by adding two horizontal lines in the residual figure, that show the average RMS at a global scale after FORLI retrievals. This is then referenced in the text (page 8045) when one comes in discussing the global distributions.

[Section 2.3, pg. 8043, line 4: by accounting for the smoothing error, the measurement noise error and the errors from the fitted model parameters, you do not have the complete error budget. For example, you miss the systematic errors from the spectroscopic uncertainties. You also miss the errors from the not-fitted model parameters like the T profiles, the adopted emissivity, etc. Therefore, the error budget is underestimated. Please provide a complete error budget and discuss the results taking the error budget into account.]

We agree with the referee that the calculated total error does not take into account for the spectroscopic uncertainties and errors from the not-fitted parameters. The error

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that we provide is the retrieval error, which places only a lower bound onto the total error. However, from previous studies, the smoothing error is usually the principal source of uncertainty on the HNO3 retrievals and other error sources, such as those on emissivity (assuming a 5 % uncertainty) or the temperature profile (assuming 1K each km) were found not to contribute significantly. This level of detailed and technical characterization has not been repeated here but we have made explicit reference to this previous work in the revised manuscript (page 8043). Regarding the emissivity we would furthermore like to point out that weak emissivity variations are fairly well accounted for in the retrievals by adjusting the source function through surface temperature while sharp emissivity features characterizing sandy surfaces, where rejected based on adequate filtering of the spectra.

[Section 2.3: no information is given about the emissivity adopted in the model: please provide enough information and indicate the impact of these emissivities on the retrieval results and associated uncertainties.]

We agree with the referee that the treatment of surface emissivity was not sufficiently explained in the manuscript. We used surface emissivity taken from climatology based on MODIS/Terra satellite observations at 908 cm-1, which is close to the absorption bands of HNO3 used for the retrievals (860 to 900 cm-1). In the TIR nadir retrieval, it is well known that surface emissivities and temperatures are strongly correlated. We have chosen not to adjust the emissivity but well the surface temperature, which is much more variable. Hence small-scale variations in the former parameter will be compensated by an adjustment of the surface temperature. This has been clarified in the revised manuscript (page 8041 and 8042). It is at this stage extremely difficult to quantify the error on this (as surface temperature is also not perfectly known) but it should remain fairly low, especially for HNO3 where the sensitivity is essentially in the highest layers. As stated above however, over deserts, some sharp emissivity features are found in the retrieval spectral range, causing a strong overestimation of the HNO3 total columns. These overestimated columns have been filtered out based on specific channels associated to these emissivity features. In the future, we hope that such sharp emissivity structures will be better known and can then be taken into account in the radiative transfer.

[Section 4, pg. 8050, line 23: the authors claim that the daily variations of the total columns in the grid boxes are relatively important. As already expressed in my general comments, it is difficult to believe that these variations are real variations of HNO3. I suspect that part of the variability comes from measurement/ retrieval uncertainties. Similarly, I am not convinced from the actually presented data set that 'in-depth analysis of chemical processes' (Section 4, pg. 8051, line 11) is already possible. But I agree that these 'preliminary' data indicate a high potential of IASI.]

Please, see Fig. 2 of this commentary paper and the response above in the general comment for this point on the daily variations. The statement highlighted by the referee on the possibility of conducting “in-depth chemistry analysis” has been less firmly made in the revised manuscript. The sentence was changed to “further global and small-scale analyses of chemical processes”

[Fig. 2 caption: 'dark grey line' should be 'the blue line'?]

This has been corrected in the revised version

[Fig. 8, high latitude plots: why is the ratio HNO3/O3 columns almost 'symmetric'around the denitrification period? I would expect higher values just after that period (early October) because of the ozone depletion at that time?]

Indeed, after the denitrification period, because of the ozone depletion, the HNO3/O3 ratios is increasing and higher values than before the denitrification period are found as it can be observed on figure 7 of the manuscript. The figure 8 of the manuscript did not exhibit this as high as on figure 7 due to the fact that the error filter was wrongly not applied to generate this figure. We thank the referee for pointing this. The figure has been corrected in the revised manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 8035, 2009.

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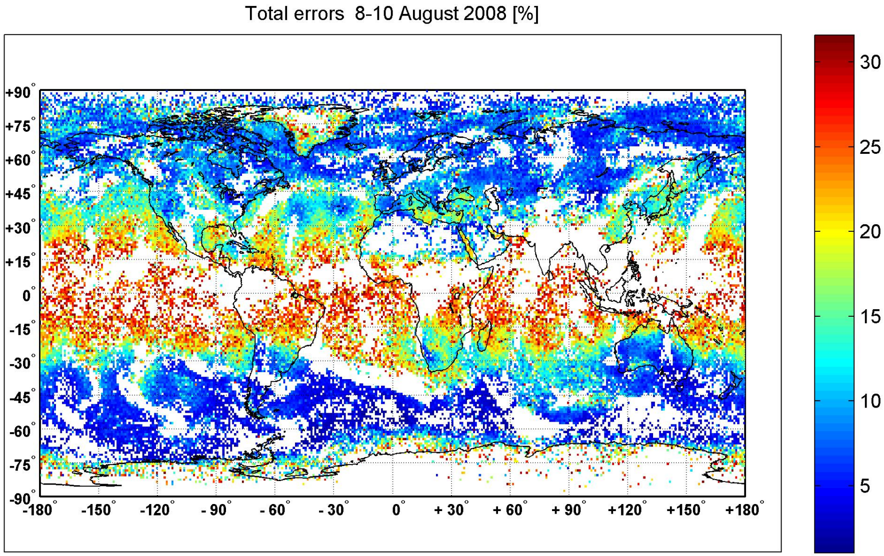

Total errors 8-10 August 2008 [%]

Fig. 1.

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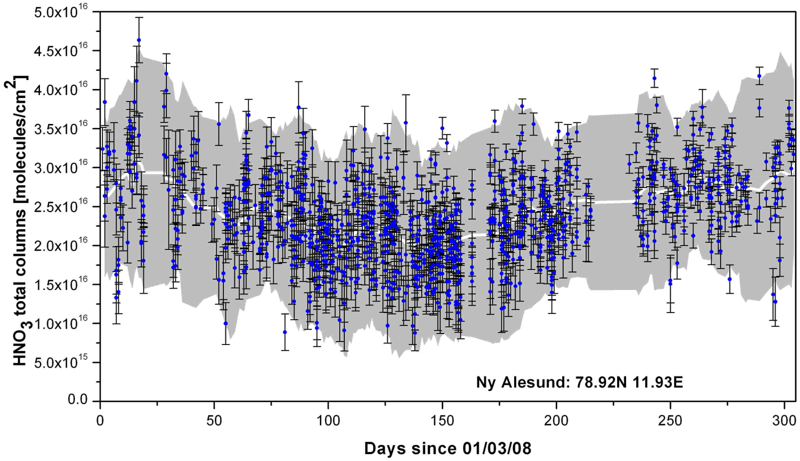
5.0x10^{16}
4.5x10^{16}
4.0x10^{16}
3.5x10^{16}
3.0x10^{16}
2.5x10^{16}
2.0x10^{16}
1.5x10^{16}
1.0x10^{16}
5.0x10^{15}
0.0
HNO$_3$ total columns [molecules/cm^2]
0
50
100
150
200
250
300
Days since 01/03/08
Ny Alesund: 78.92N 11.93E

Fig. 2.

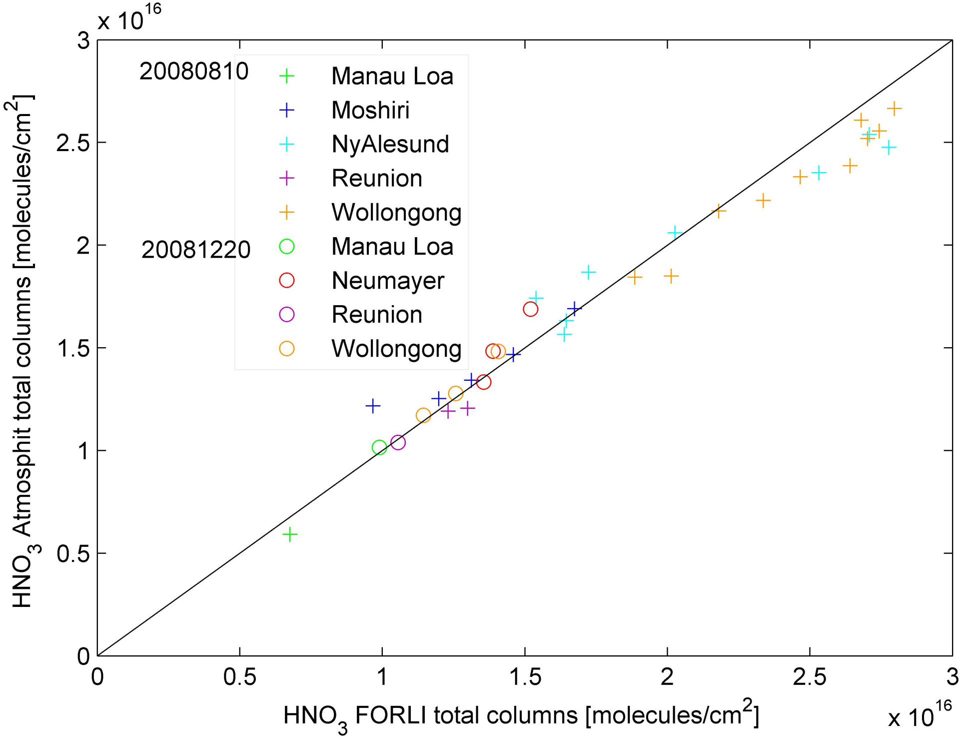

Fig. 3.

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