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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Validation of HNO_3, $ClONO_2$, and N_2O_5 from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS)" *by* M. A. Wolff et al.

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We thank the reviewer for his or her comments. In the following we present the original comments (in italics) and our responses below.

Data quality against the presence of polar stratospheric clouds (PSCs) in a line of sight.

Since the intercomparison was made even for winter vortex periods, PSCs would present in a line of sight. Do not they affect the retrieval process and hence the retrieved values? This is not only for ACE-FTS, but also for all other instruments. This should be mentioned. If they does not affect the retrieval, the authors should state it in a quantitative sense. The ACE-MAESTRO measures the aerosol extinction (AE) coefficient. Do the ACE-FTS HNO3/ClONO2/N2O5 values depend on the magnitude of AE

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coefficient? In regard to this, if temporal uptake of HNO3 in the PSC particle occurred, the HNO3 value should be lowered. This effect should also be carefully treated in the comparison. ClONO2 and N2O5 values would also be lowered by heterogeneous reactions on the particle.

The ACE-FTS is a high-resolution instrument with sufficient resolution to resolve individual lines for molecules in the gas phase. Spectral signatures for PSC particles (and for other particles in general) are broad. For HNO_3, contributions to the spectrum from PSC particles in the microwindows used (the microwindows have a width of 3 cm^{-1} or less) will simply give an apparent change in baseline. With no spectral structure in the microwindows, there can be no impact on the retrievals for the gas phase molecule. The same is true for $ClONO_2$, with microwindows of width 5 cm^{-1} or less. It is perhaps worth noting that a relatively sharp feature (typical width of 10-15 cm^{-1}) for NAT near 820 cm^{-1} lies at lower wavenumber than microwindows used for HNO_3 (870 cm^{-1} and up) and at higher wavenumber than a microwindow used for $ClONO_2$ (near 780 cm^{-1}).

The spectral window used to retrieve N_2O_5 is wider (30 cm^{-1} wide, from 1230 to 1260 cm^{-1}). If there is curvature (as a function of wavenumber) to the spectral contribution of PSC particles in this window, there could be some impact on the retrieved N_2O_5. Parameters describing the baseline are fitted during the retrieval process, but these parameters only allow for a linear variation of the baseline as a function of wavenumber. The curvature of potential PSC spectral contributions (e.g., from NAT, STS, or NAD) has not been assessed for this window.

For this, a more detailed study of the correlation between the ACE MAESTRO Aeorosol extinction coefficient and the quality of VMR profiles, as the reviewer suggests, would be of some interest. This has not been done yet and would need a more thorough approach than possible within the scope of this individual validation paper.

Comparison with SPIRALE

For altitudes between 19.3 and 20.7 km (hereafter referred to as region A), SPIRALE

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measured PSCs with enhanced values of a conjunction aerosol counter. Above these altitudes between 20.7 and 22 km (region B), SPIRALE measured reduced values of HNO3. The authors hence speculates that HNO3-containing particles were fallen from B to A, resulting in permanent denitrification in B. But my concern is that temporary uptake would continue to take place in A, lowering the HNO3 values also in A. The authors do not discuss temperature information at the measurements. How about this? "Particle greater than 1 micron." Is this diameter or radius? The particle size distribution plots are welcome here in order to provide a more roust discussion. A falling speed of the particle should also be discussed, compared to the geo-graphical locations of backward air parcel trajectories both from A and B.

Thanks to the reviewer for his/her interesting comment on possible processes in the PSC seen during SPIRALE measurements and the suggestion to have a closer look on temperature profiles and particle sizes. The SPIRALE team is currently working on a detailed analysis of all their measurements, including modelling investigations and the verification of the temperature of NAT saturation in the mentioned layer (region A) in order to determine if NAT particles are still growing, are at equilibrium or evaporating. That will result in a complete description of the PSC event seen during the balloon-launch in another work. We have included a figure showing the measured temperature and the NAT equilibrium temperature for the SPIRALE flight in order to give some background information to our explanation of the HNO_3 distribution connected with the PSC. We also added two references giving more information about the STAC aerosol counter which detected the PSC.

For this ACE-FTS validation paper, however, the main interest is to explain differences between the SPIRALE measurement and the 13-h later ACE-FTS measurement. We added a paragraph in order to spotlight these differences. In our opinion, the ACE-FTS HNO_3 profile differs from the SPIRALE profile, because it was measured in a situation where a PSC was not (longer) existant. That is indicated by ACE MAESTRO aerosol optical depth profile and a ACE-FTS temperature profile well above TNAT. Since we

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used the MAESTRO data we added C.T. McElroy as co-author.

Thanks for pointing out the inaccurate size description. Particles with a diameter greater than 1 μm was meant. We changed the manuscript accordingly.

Comparison with FTIR

Discussion on comparisons with the Arctic stations can be shortened. The largest relative differences reported here are less important information. The authors only say that there are inhomogeneities in the distribution of chemical species under coincidence criteria (1000 km in separation and 24 hours in time difference) used in their study. This is not a surprising thing. I would suggest that the authors only show the differences excluding the winter vortex period.

We agree with the reviewer that comparisons during the winter polar vortex period should be separated from the others. Unfortunately, for two of the polar stations, we only have winter measurements and for the two remaining, the number of comparisons would significantly drop. In order to have a representative number of comparisons, we still show and analyze all comparisons, but have now marked the measurements performed under winter conditions in Figures 14 and 16. Consequently, we still discuss the clearly larger differences found in the comparisons made under polar vortex conditions. Following the suggestions of the reviewer, we have shortened that paragraph.

2.1, Line 6, It is unclear to me what an earlier retrieval is. Give a more explanation for this. The OCS is retrieved prior the HNO3 retrieval and fixed at that value. We have added the explanation in the manuscript.

2.2, Line 13, How do the standard profiles of H2O2 determine? The VMR profile for H_2O_2 is fixed to a standard profile taken from the ATMOS experiment. We added that information to the manuscript.

2.3, Line 2, What are the previous retrievals for HDO, O3, HNO3, and COF2? Is it v1.0? The authors should explain the relation among earlier retrievals, standard profiles, and

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previous retrievals. We added the explanation to the mansucript.

In Fig. 2, Eq. (3) is used for calculating mean relative differences, not Eq. (2). We changed the figure caption accordingly.

4.2, Line 19, A more careful analysis is needed to discuss large values of the mean relative difference and its standard deviation found in the southern high latitudes. At least, a distinction should be made for data obtained inside and outside the vortex. Are the low values of HNO3 found mainly in temperatures below the existence temperature for NAT (TNAT)? A data separation using some threshold temperature might also improve the large values of the statistics. Comparisons should also be made separately by season, since the permanent denitrification is seen in spring to early summer when T above TNAT.

We performed an additional separate comparison between ACE-FTS and Aura-MLS excluding all profiles with temperatures below PSC formation temperature (T_{NAT}=196 K) to support our argument that the large standard deviation on the mean relative differences is caused by the measurements performed during high PSC activity. A paragraph describing these results has been added to Section 4.2.

4.3.3, Last paragraph, Comparisons were made for a wide latitude band between 30 and 90 degrees N. Is there any latitudinal difference in comparison results? The magnitude of the low bias at 30 km is smaller in the comparison with the daytime MIPAS (-30%) than in that with the nighttime (-50%). I assume that the daytime comparisons were made for lower latitudes, since the values of ACE-FTS (ss) are lager than those of the nighttime comparison.

Different latitude binning (30°-60°N; 60°-90°N) did not remove the day-night discrepancy, nor did it clarify a potential cause of the discrepancy. The mean latitude is 75°N for the day measurements and 72°N for the night measurements. PV-values at the 485K level are 30.8×10^{-6} Km^2 kg^{-1} s^{-1} for day measurements and 29.5×10^{-6} Km^2 kg^{-1} s^{-1} for night measurements. The day measurements are slightly further north

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and slightly more inside the vortex, which possibly explains the higher N_2O_5 values seen during the day comparisons, resulting from an enhancement of N_2O_5 inside the vortex, due to the previous strong downwelling of no_y-rich air, as can be seen from MIPAS cross sections of N_2O_5 along orbits. The differences between ACE-FTS and MIPAS at lower altitudes are partly due to the difference in vertical resultion between the two instruments there. These are strongly reduced when MIPAS averaging kernels are taken into account. We added that comparison to Figure 9, discussed it in Section 4.3.3 and added a summarizing sentence to the conclusions.

5.2, What is the spectral resolution of SPIRALE? The spectral resolution of SPIRALE is 0.001 cm^{-1}, the information has been added to the manuscript.

5.3, Also add discussion on a difference seen in HNO_3 at 26-30 km (Fig. 12).

Since PSCs are typically limited to altitudes below 26 km, we can not explain this feature with the existing PSC. The FIRS-2 HNO_3 retrieval is very robust with a good signal-to-noise ratio and does not show any obvious errors. Therefore, the enhanced HNO_3 values seen by FIRS-2 above 26 km remain unexplained at this point. We added that to the manuscript.

6, Line 28, I think that "Atmospheric density profiles were calculated based on ..." is grammatically incorrect. We changed the sentence.

In Fig. 18, How about showing the total number of comparison in the legend? Such as SMR(1571). We have implemented the total number of comparisons in the legend, as suggested.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 2429, 2008.