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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Validation of ACE-FTS N_2O measurements" *by* K. Strong et al.

K. Strong et al.

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We thank the reviewer for his helpful comments. In the following we present his original comments in italics and our responses in plain text.

General comments
This paper deals with the validation of a new atmospheric chemistry satellite mission (ACE) for an important atmospheric species (N2O). This theme is of high significance for the atmospheric chemistry and physics science community and fits well into the ACP journal scope. The adopted validation concepts are of excellent scientific quality, and the study is based upon a full variety of high quality correlative data sets. The paper is presented well and should definitely be published.

On going carefully through this paper, I encountered a few minor issues which are listed as <Specific comments> thereafter. While consideration of these issues might help to further improve science quality and readability of the paper, I would like to

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render the decision on whether and how to deal with which of these minor points to the responsibility of the authors, because of the already high quality of the discussion paper at hand.

Specific comments
<Abstract>
>The abstract summarizes validation results/numbers mainly in terms of <mean absolute differences>. However, the validation results shown in the discussion, contain in addition some nice information on natural variability of N2O as a function of altitude as well as some information on precision of the individually measured profiles or profile differences (sigma, sigma over sqrt N, of profiles and differences, see panels a-d of the validation figures). Why is this valuable information not exploited, leading to some final statement within the abstract? I would be personally interested in questions like
i) in which altitude domain is data quality sufficient to detect the natural variability of N2O from individual measurements?
ii) up to which altitude is data quality found to be sufficient to measure the absolute VMR from an individual ACE profile?

The following discussion of these issues has been added to Section 7, and mentioned in the Abstract.

"To assess the altitude range over which the ACE-FTS data quality is sufficient to detect the natural variability of N_2O, we can compare the ACE-FTS VMR statistical fitting errors (i.e., random errors) to the natural variability represented by the relative standard deviations on the mean profiles plotted in panel (d) of Figures 1 to 7. As noted in Section 2, the fitting errors have a median value of $<$3% from 5–45 km, increasing to 17% at 60 km, with a mean of $<$4% from 5–35 km, oscillating above this due to some outliers in the individual percent fitting errors. Examining Figures 1d and 3d, as these show results for the largest data sets (1099 and 6876 coincidences respectively), the

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ACE-FTS relative standard deviations are greater than the median fitting errors at all altitudes. This indicates that the data quality should be sufficient to detect the natural variability of N_2O over the entire altitude range examined, 5–60 km.

To assess the altitude range over which the ACE-FTS data quality is sufficient to measure the absolute VMR, we must rely on the differences relative to the other instruments, as there is no systematic error budget for the v2.2 data product. However, work is underway to produce an error budget for the next version to be released (v3.0). The mean absolute and relative differences given in Table 4 and Figure 12 are therefore our best estimates of the absolute data quality."

3599/13-14 <Overall, the quality of the ACE-FTS version 2.2 N2O VMR profiles is good over the entire altitude range from 5 to 60 km.>
>qualitative statement within an Abstract - maybe you skip this sentence?

This sentence has been removed.

3559/19-20 <..., again excluding the aircraft and balloon and aircraft comparisons.>
>i) I do not see to what exactly <again> is referring; and ii) why are balloon and aircraft validation results excluded?

The numbers in the preceding sentence ("Between 6 and 30 km") referred only to the satellite comparisons, and this sentence is discussing a subset of that data from 18 to 30 km. The results of the satellite comparisons are emphasized here as they provide larger datasets for statistical comparisons. A full discussion of all the results is given in the respective sections and in Section 7 (Conclusions). The text in the Abstract has been clarified.

<Introduction>
>It gives mainly a very nice science overview on the role of N2O, and on the satellite measurements/missions that dealt with it before. But at the end of the third paragraph I would expect a geophysical science paper to follow, and not a validation paper. I.e.,

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paragraph 4 shows up then rather surprisingly.
To solve this the thirst 3 paragraphs might be shortened, and it would be interesting to get in addition here in the introduction some information on the state of the art (achievements/problems) of previous satellite validation studies that dealt with N2O: Paragraph 3605/8-13 might be shifted from Section 2 to the Introduction for this purpose.

We prefer to leave the Introduction as is (and note that Reviewer 2 described it as a "Very nice and complete introduction"). The first three paragraphs serve to introduce the origin, role, and importance of the N_2O in the atmosphere and are already concise. It is not clear what is meant by a validation paper at the end of the third paragraph 8211; the last reference in this paragraph is to Brasseur and Solomon (2005). Paragraphs four and five then briefly review previous and current space-based measurements of N_2O, citing references that describe these measurements and their validation, thus setting the scene for the work described in this paper, which is outlined in paragraph seven.

Furthermore, I would be interested in a brief discussion of the natural variability of the N2O profile as a function of altitude and the mechanisms behind that. (E.g., we constructed a covariance of the N2O profile limited to 3-9 km a.s.l., see Fig. 9 of Sussmann and Borsdorff, Atmos. Chem. Phys., 7, 3537-3557, 2007). This would be a basis then to the later question whether ACE is able retrieve the natural variability of N2O.

We think that a meaningful discussion on the natural variability of N_2O and the underlying physical and chemical mechanisms is beyond the scope of this paper. We are currently working on two related papers, one examining the climatology of ACE-FTS NO_y and N_2O, and another concerning methods of assessing the precision and variability of ACE-FTS data products. We have added some discussion regarding the ability of ACE-FTS to retrieve the natural variability of N_2O, as noted above.

Finally, I feel that the paragraph on the ACE mission and science goals given in Section

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2 (3603/9-13 plus 20-27) would better fit to the Introduction than to the <retrievals> Section.

We have changed the title of Section 2 to better reflect its content. We prefer to keep the description of the mission, instrument, and N_2O retrievals together, as well as paragraph 3605/8-13 listing previous publications that looked at ACE-FTS N_2O data.

<3 Validation approach>
>Very good, especially the type of plots with panels a-d. But, as said before under <Abstract> (see above): These validation plots contain some nice information on natural variability of N2O as a function of altitude as well some information on precision of the individually measured profiles or profile differences. Why is this valuable information not further discussed/exploited, leading to some final statement within the Abstract, in addition to the numbers on the <mean absolute differences>? My overall interest would be to get a picture on all what is known a priori on the true N2O profile and its variability, and, then, in which altitude domains ACE is able to measure this profile and its variability with sufficient quality.

See the response above regarding this point.

3607/13 <..., along with the standard deviations on each of these two profiles>
>I would more easily understand <..., along with the standard deviations calculated from the individual profiles for each altitude>.

This change has been made.

3607/20-21 <..., and the standard deviation of the distribution of this mean difference.>
> I would more easily understand <..., and the standard deviation of the individual differences of all coincident pairs as a function of altitude.>

This change has been made.

<eq. (2) versus eq. (3)>
>It is certainly good to use eq. (3) and not eq. (2). But nevertheless, it is a rather trivial

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point, in a sense that it can indeed be easily understood by a scientist from 1 short explanatory statement like <use averages in case of small denominators>. To avoid overstatement and to improve readability of the paper, I would therefore recommend to cancel eq. (2) cancel all cyan solid lines in all Figures cancel 3611/9-24 cancel 3614/13-18 (<Dividing ... behaviour>)

We respectfully disagree with this recommendation. The decision to include both equation 2 and equation 3, along with the corresponding results, was made after extensive discussion amongst the co-authors on how best to calculate and show differences between different data sets. It is not always clearly explained in validation papers exactly how relative differences are calculated, resulting in ambiguous results. We prefer to include both, clearly illustrating the usefulness of equation 3, but also showing, with the figures, why it is appropriate in this case.

<Table 1>; <100-1 hPa>
>change for consistency to altitude units: <x-y km> (same with 3613/10, 3613/18, 3613/21, and 3613/23)

We have changed the vertical range entry for MLS to altitude units in Table 1 for consistency, as this is the altitude range used for the comparisons with ACE-FTS. However, the text on page 3613 summarizes the results of other papers describing the Aura/MLS N_2O data product. As those results are provided as a function of pressure, not altitude, we prefer to present them as published, rather than making assumptions to convert to a different vertical grid.

<Table 1>; <Coincidence criteria>
>Different coincidence criteria are chosen for every correlative technique, and even different ones for MIPAS-ESA versus MIPAS-IMK.
Why are they different and what is the reason behind that? Are this ad hoc assumptions of the different validation groups? Or are all the different coincidence criteria the result of one common strategy to find them, e.g., some kind of tradeoff?

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I know this is a difficult question, and it is not answered in many validation papers, but it is an important question of general interest. One possible way around is to discuss the effect on the validation results from changing the selection criteria by, e.g., a factor of 2 or 0.5: you may find an example for this approach via Table 1 of Sussmann et al., Atmos. Chem. Phys., 5, 2419-2429, 2005.

There is some discussion of the choice of coincidence criteria in the first paragraph of Section 3. As noted, they were chosen in consultation with each of the teams involved, striving for consistency where possible. Two truly global datasets were available for comparison (SMR and MLS), and for these, we did use the same criteria, consistent with criteria previously used (and published) for N_2O. The two MIPAS teams worked independently and chose tighter criteria as they deemed appropriate for the two-month period of overlap with ACE at northern mid- and high latitudes. For the ASUR aircraft measurements, we used the same temporal criterion ($\pm$12 hours) as was used for SMR and MLS, with a spatial criterion of 1000 km to ensure coincidences. For the ground-based FTIR measurements, the temporal coincidence was extended to $\pm$24 hours to yield a reasonable number of data points for comparison. Only two balloon flights anywhere near ACE occultations were available, and we included both. The choice of coincidence criteria is always a balance between making them large enough in time and space to provide a useful number of co-located measurements, but not so large as to introduce biases due to temporal and spatial inhomogeneities in the distribution of the species of interest. We have not been able to perform a rigorous trade-off study, but for a long-lived, well-mixed species like N_2O, this should not be as critical as for more rapidly varying constituents.

3612/1-3 <These larger values are consistent with the noisier data, particularly from SMR, above 40 km, as seen in the relative standard deviations on the mean profiles plotted in Fig. 1d.>
>I do not understand this sentence.

This sentence has been revised as follows:

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"These large values of the relative deviation from the mean at high altitudes are due to the noisy data in this region, particularly for SMR, as can be seen in the large relative standard deviations on the mean profiles plotted in Fig. 1d."

3612/23 <... relative standard deviations on the mean profiles>
>I would more easily understand <... relative standard deviations of the individual profiles>

Figure 2d shows the ratio (times 100%) of the standard deviation calculated for the ensemble of data used to generate the mean in Figure 2a, to that mean. The text has been changed to: "The relative deviations from the mean increase above 40 km, where the relative standard deviations for the individual mean profiles are also seen to reach values of 100% and larger."

3616/7 versus 3617/23 <25 March> versus <26 March>?
>Typo or intended difference?

These two different dates are correct. However, MIPAS full-resolution measurements ended on 26 March, not 25 March as was stated. This has been corrected.

3616/8-9 <... horizontal resolution is 300-500 km along-track (Vigouroux et al., 2007).>
> Is it really appropriate to make reference to a validation paper to document the horizontal resolution of MIPAS?

This reference has been changed to Fischer et al., ACP, 2008.

<Section 5.2 and Section 5.3>
>Individual profile time mismatch is 13 h (SPIRALE) and 26 h (FIRS-2), respectively. I would be interested in some discussion on the magnitude of the differences seen in the profile comparisons relative to the expected natural variability of the N2O profile for the given time mismatch. Is it significant errors of the measurement systems or might the differences seen just reflect natural variability?

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We do not have information on the natural variability of N_2O as a function of time period, and the sparseness of the ACE solar occultation data set makes this difficult to quantify. Both the SPIRALE and FIRS-2 balloon flights were launched from Kiruna in January, when the movement of the polar vortex can cause descent and can therefore affect the vertical profile of N_2O on relatively rapid time-scales. As noted in the paper, SPIRALE and ACE-FTS sampled similar air masses within the vortex, while FIRS-2 sampled inside the vortex and ACE-FTS the vortex edge. There are clearly large measurement errors in the ACE-FTS – FIRS-2 comparison above 20 km, which probably explains the large relative differences seen in this region.

3625/24-25 <... determined by the sensitivity of the FTIR measurements, which 25 was required to be 0.5 or greater, ...>
>I can only guess what you really mean by <sensitivity>. Could you give a hard explanation in terms of retrieval theory: Is it the peak-height of averaging kernels, in which unit (VMR or normed), or is it the area of the kernels, or maybe the peak of the partial column averaging kernels ...?

The sensitivity, as used here, follows the definition given in Vigouroux et al., ACP, 2007, as referenced in the paper. Some additional information has been provided in the paper, as follows:
"The lower limit of the altitude range of the partial columns at each station was determined by the ACE-FTS altitudes and the upper limit was determined by the sensitivity of the FTIR measurements, defined for a given altitude as the sum of the elements of the corresponding averaging kernel (Vigouroux et al., 2007). This sensitivity was required to be 0.5 or greater, indicating that the measurement contributes at least 50% to the retrieved profile, with the remainder coming from the a priori information (Vigouroux et al., 2007)."

3626/4-5 <... the state space interference error (due to unphysical correlations between different parameters in the state space), ...>
Finally, here I become personally very interested and curious:

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-which interfering species do you consider?
-how big are the interference errors you get in real numbers?
-did you use the simple initial 1-parameter approach by Rodgers and Connor (J. Geophys. Res., 2003, eq. 8 therein), or did you use the corrected and extended approach by Sussmann and Borsdorff, Atmos. Chem. Phys., 7, 3537-3557, 2007, which gives more realistic values and thus shows much higher interference errors for ground-based FTIR, using scaling retrieval for the interfering species?
-did you try to minimize the interference errors as we suggested?

For the Toronto FTIR N_2O retrievals, we included the interfering species CO_2, CH_4, and O_3. We used the standard approach of Rodgers and Connor to calculate the interference error. For N_2O, we find that the state space parameter interference error is significantly smaller than that of the retrieval noise error at all altitudes. Although implementing the approach of Sussman and Borsdorff (2007) may yield larger values for the interference error, the total error given here is only intended as a typical value. None of the FTIR sites that provided error information provided interference errors using this extended approach.

We revisited the partial column errors for the mean altitude range for the FTIR N_2O partial columns (14–27 km) using Toronto data and found that the retrieval noise error was 1%, the state space interference error was $<$0.25%, the temperature error was 3%, and the smoothing error was 3.5%. Combining these in quadrature yields a total error of approximately 3% without, and approximately 5% with, smoothing error, not 10% as stated in the manuscript. This has been corrected in the text. Since the ACE profiles are smoothed by the FTIR averaging kernels, the smoothing error should not be included in the total error budget. We also checked that these values were consistent with errors provided for other FTIR sites. For example, Jungfraujoch N_2O partial columns (18-28 km) have a total error of 4.07%, of which 3.83% is smoothing error, and Wollongong total errors are 4-10%.

I enjoyed reading this paper.

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Ralf Sussmann

Thank-you.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 3597, 2008.

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