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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Improvements in the profiles and distributions of nitric acid and nitrogen dioxide with the LIMS version 6 dataset" *by* E. Remsberg et al.

Anonymous Referee #2

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General comments:

The paper is a very valuable contribution in the area of stratospheric measurements. It presents an updated version of the LIMS HNO3 and NO2 observations which have been recorded as early as 1978/79. The global HNO3 and NO2 data set presented here covers the period of the Northern polar winter 78/79. It provides day and night observations including the poles up to 64° S and 84° N, and thus also covers the polar night. The improvements of the new version 6 data with respect to former version 5 are described, and the the version 6 data set is critically assessed regarding several aspects. I have a few minor concerns and one rather major one:

- My most severe concern is that the retrievals have been done with a rather outdated version of the HITRAN spectroscopic data base, namely with versions from 1991 to 1996. In particular for HNO3, many significant improvements on the spectroscopic line parameters have been provided in the last 10 to 15 years, and the matter is still ongoing. Without proper characterization of this issue, the LIMS data run the risk of being biased severely towards data sets from other contemporary satellite instruments like MLS, ODIN/SMR, HIRDLS, or MIPAS which were generated using more recent spectroscopic data. I suggest to add a paragraph on the impact of the spectroscopy on the accuracy of the data set and, in particular, on a potential bias of V6 LIMS data compared to a version which would use more recent HITRAN data.
- Since not all readers are familiar with the LIMS measurements, some more information on the instrument and data retrieval, giving at least information on the altitude sampling (tangent altitude step width), spectral range, and spectral resolution would be helpful.
- For the reader experienced in mid-infrared spectrometry it would be more helpful to first describe the retrieval approach, and later on the results in terms of zonal means. Understanding of section 2 depends quite much on this information.

Overall I recommend this manuscript for publication after the following points will have been properly taken into account:

Specific comments:

Abstract:
p 2770, l 17-20 (last sentence): Discuss this in the light of spectroscopic data versions used.

Background:
p 2771, l 7-11: Could you provide some examples (references)?

Section 2:
General: It was quite difficult for me to follow this section without knowing more about retrieval details - in particular which spectral resolution is provided by LIMS, and which spectral ranges have been used. This information should be provided before entering the discussion of zonal means.
p 2773, l 20-21: Could you provide a proof for this statement?
p 2773, l 22-24: It is very hard to judge on this hypothesis without information on the spectral resolution and the spectral range HNO3 has been retrieved from; from my experience with mid-infrared spectra, I am not quite convinced that CO2 and O3 contaminate very much the HNO3 signatures.
p 2774, l 7-11: An enhancement of HNO3 in the upper stratosphere to values around 2 ppmv as seen in Figure 1a should be accompanied by strong enhancements in NO2 in the same altitude range (see Stiller et al., 2005, Funke et al., 2005a, and Reddmann et al., 2010). However, no significant nighttime enhancements of NO2 are found for this day (Fig. 2a and Fig 11). Surprisingly, there are daytime enhancements (Fig. 2b). How many data points were used for the zonally averaged daytime NO2 profiles for 70 to 90 deg North on 15 Nov 1978?
p 2274, l 11-14: The existence of a Northern polar vortex in mid May is not very frequent; in general the break-down of the Northern polar vortex - at least in the last 20 years - occurs in March or April. Could you provide a proof, e.g. from potential vorticity maps that indeed diabatically descended polar vortex air was observed on 16 May 1978?

Section 3.1:
General: Some information on spectral range and spectral resolution should be

provided as an introduction to this chapter for all readers not familiar with the LIMS measurements.
p 2776, l 11 and 16: Although there is no information on the spectral regions used, I believe that the "hot band" of CO2 is usually called CO2 laser band.
p 2776, l 13 to p 2777, l 6: Why has the rather outdated HITRAN data version of 1991 been used for the retrievals? The most recent HITRAN data version for processing and archiving in 2002 was HITRAN2000 which contains significant improvements regarding HNO3 and NO2 spectral data (and those of other species). In particular HNO3 spectroscopic data have seen a major step forward in quality during the last 10 to 15 years. It is a pity that due to outdated spectroscopic data the comparison to and synergetic use of LIMS data with those of more recent instruments like MLS, ODIN/SMR, HIRDLS, or MIPAS has become spoiled to a certain degree. At least, an assessment of a potential bias introduced by using HITRAN91 instead of more recent HITRAN data versions should be provided.
p 2777, l 12-14: On which investigations, studies with synthetic data, comparisons to other instruments etc. is this statement based?
p 2777, l 14-17: Funke et al. (2005a and 200b) have provided improvements in the radiative transfer modeling of non-LTE affected NO2 emissions. Although I appreciate that processing and archiving of the LIMS V6 data was done in 2002, i.e. before Funke et al.'s work, some referencing of this recent work would be adequate.
p 2778, l 21 to p 2779, l 10: As I read this paragraph it is a rather complicated description of the fact that non-converged data points in the onion-peeling procedure have been filtered out and not archived. Could this para be shortened?

Section 3.2:
General: As pointed out earlier, I consider the systematic error contribution due to spectroscopic uncertainties as relevant. It should be included in the assessment of the accuracy. Further, to make the data set better comparable to more recent observational data sets, an assessment of the potential bias due to using HITRAN91-96

instead of more recent data versions should be quantified.
p 2779, l 19-26: How have precisions been calculated? As I read these sentences, the calculation of the precisions or standard deviations (are these the same?) are provided at the end of this para only. The text says that the precision estimates as shown in Fig.4 compare well with the standard deviations also shown in Fig. 4, but there is only one curve in Fig. 4 per ascending and descending orbit part, respectively. This is very confusing, please clarify! These sentences should be reorganized for better understanding.
p 2779, l 24: What does "**minima** of the variances" mean?

Section 4:
p 2782, l 24-25: Please could you provide some references?

Section 4.1:
p 2784, l 18-21: For confirmation/falsification of this hypothesis, I recommend to have a look into MIPAS data. They are measured at about 10 a.m. and 10 p.m. local time (equator crossing) and, thus are comparable in terms of observation mode to LIMS. To my knowledge, no HNO3 increase towards the upper stratosphere is found, besides in polar winter, which questions the hypothesis of delayed photolysis as reason for the higher upper stratosphere HNO3 values.

Section 4.2:
p 2785, l 16: Please could you provide some references?
p 2785, l 23/24: Some more description of the formulation of the heterogeneous chemistry in the two-dimensional model is required here. Is the use of a two-dimensional model appropriate?

Section 4.3:
p 2787, l 11: Please specify in more detail the diurnal photochemical model and

provide some description.
p 2787 l 26 to p 2788 l 1: The high bias of daytime LIMS NO2 is most pronounced between 1 and 0.5 hPa which is at and above 50 km. Therefore, the findings by Funke et al. (2005) that non-LTE is not an issue below 50 km do not really help here.

Section 4.4:
p 2788, l 15: Could you please specify on which data this statement is based, and provide a reference?

References:
Funke, B., et al. (2005b), Retrieval of stratospheric NOx from 5.3 and 6.2 μm nonlocal thermodynamic equilibrium emissions measured by Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on Envisat, J. Geophys. Res., 110, D09302, doi:10.1029/2004JD005225.

Funke, B., M. López-Puertas, S. Gil-López, T. von Clarmann, G. P. Stiller, H. Fischer, and S. Kellmann (2005b), Downward transport of upper atmospheric NOx into the polar stratosphere and lower mesosphere during the Antarctic 2003 and Arctic 2002/2003 winters, J. Geophys. Res., 110, D24308, doi:10.1029/2005JD006463.

Reddmann, T., R. Ruhnke, S. Versick, and W. Kouker (2010), Modeling disturbed stratospheric chemistry during solar-induced NOx enhancements observed with MIPAS/ENVISAT, J. Geophys. Res., 115, D00I11, doi:10.1029/2009JD012569.

Stiller, G. P., et al. (2005), An enhanced HNO3 second maximum in the Antarctic midwinter upper stratosphere 2003, J. Geophys. Res., 110, D20303, doi:10.1029/2005JD006011.

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