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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global peroxyacetyl nitrate (PAN) retrieval in the upper troposphere from limb emission spectra of the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS)" *by* N. Glatthor et al.

N. Glatthor et al.

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We also thank referee 3 for his helpful comments. With respect to his remarks we will perform the following changes:

Reply to general comments:

Reference to earlier PAN spectral detection:

The referee argues that "there is no reference to the fact that PAN spectral detection has been demonstrated for the MIPAS instrument on ENVISAT as well as MIPAS

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balloon." However, this is only partly correct. Indeed, there is a reference to the ACPD-paper by Remedios et al. (2006) describing PAN retrieval from MIPAS balloon spectra at the end of Section 1 (from pg. 1394, line 27, to pg. 1395, line 1). We will update the reference to the subsequent ACP-paper published most recently. To acknowledge the first PAN detection in spectra from MIPAS/ENVISAT we will cite the Ph.D. thesis of Allen (2005) and the paper of Remedios et al. (2006) contained in the Proceedings of the "First Conference on Atmospheric Science". Further, we will introduce the PAN values derived from MIPAS spectra by Remedios et al. and by Allen in Section 5.3.

Spectroscopic uncertainty of PAN:

The referee states that our spectroscopic PAN uncertainty of 3.2% is probably an underestimate. We adopted this value from the paper by Allen et al. (2005b), which in Table 2 gives a 2-sigma uncertainty of 6.4% for the PAN peak absorptivity at 250 K for the band at 794 cm^{-1}. To make this uncertainty comparable to the other error sources discussed in our paper (1-sigma estimates), we scaled it down by a factor 2. As emphasized in our paper as well as by the referee, a value of 3% is certainly an underestimate for temperatures well below 250 K, i.e. for higher altitudes. However, we do not want to introduce a mean of the spectroscopic error given by Allen et al. and of our own estimation of the temperature dependance into the general error calculation, because the temperature dependance is a purely systematic positive bias of the retrieved PAN amounts, whereas the contributions displayed in Figure 2 are generally random errors. We did not include any water vapor error because water vapor was joint-fitted along with PAN and related uncertainties are thus included in the noise error.

Large PAN values in the biomass burning plume:

The referee is concerned over large PAN values around 500 pptv in the biomass burning plume. However, in the paper we state, that these PAN amounts are maximum values and that average plume values are lower. In Figure 9 we show that the mean

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values are between 200 and 370 pptv at 8 km and around 200 pptv at 12 km altitude, which is well inside the range of previous measurements (Singh et al., 1996). To answer the referee's question for the number of profiles giving rise to large PAN values (400–500 pptv) in Figure 8: At 8 km altitude, the area of high PAN in the Southern subtropical Atlantic is represented by 34 profiles, the area between East Africa and Madagascar by 14 profiles, but the South African spot by 1 profile only. At the altitude of 12 km, the high PAN above North-East Brasil is the average of 4 profiles, the West-African spot contains 2 profiles, the East-African area 8 and the high-PAN area above the Indian Ocean 8 profiles.

C_2H_2 as an indication of biomass burning:

First and foremost we wanted to use the C_2H_2-PAN correlation to strengthen our PAN retrieval results regardless of the production process. However, Singh et al. (1996) indeed characterise C_2H_2 as "relative reliable, although not unique, tracer of biomass burning." We will add this reference into Section 5.2 to stress the relative suitablility of C_2H_2 as tracer for biomass burning.

Justification for elevated PAN in the northern hemisphere:

As justification for MIPAS observations of elevated PAN in the northern hemisphere we will refer to Kotchenruther et al. (2001) and Roberts et al. (2004), who report on PAN amounts of about 200 pptv in the altitude region 6–8 km off the westcoast of the United States and attribute these measurements to long-range transport of industrial pollution from East-Asia. The northern midlatitude PAN values at 8 km presented in this paper are of the same order, namely $\sim$125 pptv for the zonal average and $\sim$200 pptv above China and the northern West-Pacific.

Reply to specific minor comments:

1) Pg. 1394, line 1: The reference for PAN transport over more than 10000 km is also

Singh (1987). We will cite this reference again.

2) Pg. 1394, line 12: We also adopted this statement from the paper of Singh (1987) and will give this paper as reference.

3) Pg. 1396, line 20: To constrain our retrievals we use Tikhonov's regularization scheme along with a first derivative operator, which for low signal-to-noise ratio tends to drive the retrieved profile to the shape but not to the numerical value of the apriori profile. Therefore, the regularization does not bias the PAN vmrs towards low values. For a more extensive discussion of this issue, see Stiller, ACPD, 7, 3229–3268, 2007, SC S584.

4) Pg. 1396, line 20: Actually we do not have climatological PAN profiles from direct observations. We use a midlatitude mid-July PAN profile of the Model for OZone And Related chemical Tracers (MOZART) as first-guess for all latitudes and seasons. The respective wording will be changed in the paper. Since this profile is only used as first-guess but not as apriori and the retrieval is iterated to convergence, it has no influence on the retrieval.

5) Section 3, cloud flagging: Since the method of cloud flagging applied at IMK has been described in detail in previous publications, e.g. in Glatthor et al. (2006), we did not mention it here. We will add a sentence that "cloud-flagging was performed following the standard method applied at IMK, using a cloud-threshold of 4.0" and give a reference.

6) Pg. 1398, line 23: The sentence will be corrected.

7) Section 4, reference to already reported PAN spectral detection: We will repeat the references to Allen (2005) and to Remedios et al. (2006) from Section 1 (cf. general comments). With respect to the referee's objection that "it could be that another broad-band contaminant would do a similar job" we would like to note that our significance test is different from the preceding approach: In the test of a PAN-free atmosphere we

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performed a completely new retrieval of CCl_4, HCFC-22, H_2O, $ClONO_2$ CH_3CCl_3 and of C_2H_2, i.e. we allowed a fit by each possible broadband contaminant. It turned out that by use of these contaminants the measured spectrum could not be modelled as well as by inclusion of PAN (cf. Figure 4, left).

8) Pg. 1406, line 4: We do not completely understand the referee's problems with this sentence. We will change the wording into "ten days of the period 4 October to 1 December, 2003."

Reply to technical comments:

We will correct minor English errors as far as we are able to identify them.

References

[1] Glatthor, N., von Clarmann, T., Fischer, H., Funke, B., Gil-López, S., Grabowski, U., Höpfner, M., Kellmann, S., Linden, A., López-Puertas, M., Mengistu Tsidu, G., Milz, M., Steck, T., Stiller, G.P., and Wang, D.-Y: Retrieval of stratospheric ozone profiles from MIPAS/ENVISAT limb emission spectra: a sensitivity study, Atmos. Chem. Phys., 6, 2767–2781, 2006.

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