Atmos. Chem. Phys. Discuss., 7, S1283–S1288, 2007
www.atmos-chem-phys-discuss.net/7/S1283/2007/


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.**

Received and published: 20 April 2007

We thank referee 1 for his helpful and comprehensive comments. With respect to his suggestions we will perform the following changes:

**Reply to general comments:**

1) More elaborated and better documented reference to MOPITT observations:

The comparison with MOPITT CO measurements from the end of September 2003 will be extended.

2) Motivation to select the period October 4 to December 1, 2003, for analysis:

Due to the considerable computation time needed for PAN retrieval we had to focus on a restricted dataset for a first effort to evaluate PAN. The period October 4 to December 1, 2003 (note: 2003, not 2004), was chosen, because it covers a significant part of the southern hemispheric biomass burning season, which is generally characterized by a strong tropospheric pollution plume ranging from South America over Africa as far as Australia and persisting for weeks. This plume often extends to the upper troposphere, which favours its detection by MIPAS. Further, this southern hemispheric biomass burning signal is not smeared by potential ubiquitous industrial pollution like in the northern hemisphere. Of course we plan to extend our PAN analysis to other seasons. Then we will also be able to answer the question, if MIPAS was able to observe PAN arising from Northern Asian boreal fires during the period May to August.

3) Reason for the weaker PAN signal over Australia:

We agree that there were also hot spots in Australia during the investigated period. But from our inspection of maps of TRMM fire count clusters we conclude that there were more hot spots in South America and Africa. Further, model calculations of the Goddart Space flight center show a rather dense coverage of the uppper troposphere over South America and Africa by trajectories originating at fire count clusters, whereas the trajectories originating above the Australian hot spots disperse relatively fast over the Southern Pacific. We assume that these are the reasons, why comparable PAN amounts like over South America and Africa did not accumulate over Australia. An examination of the PAN observations as function of the amount and type of burning would be out of the frame of the present paper. Beside presentation of the retrieval approach, the paper is mainly focused on the description of the global PAN distribution inclusive the biomass burning plume averaged over the whole period analysed. The referee's suggestion, however, would additionally require a detailed investigation of uplift processes and of the temporal and spatial development of the plume, which we would like to postpone to a follow-on paper.

**Reply to the specific comments:**

Pg. 1393, line 23-24: We agree and will use the expression "lifetime".

Pg. 1394, line 12: Here we compare PAN and ozone as indicators of tropospheric pollution: Stratospheric ozone transported into the upper troposphere can mock tropospheric pollution. This confusion is not possible with PAN, since it has no stratospheric source. We will change the wording in the text to make this clearer.

Pg. 1396, first paragraph: The reference for the IMK MIPAS processor (von Clarmann et al., 2003b) is already cited in the preceding sentence. To make this clearer, we will add a subordinate clause "..., amongst others one at the Institut für Meteorologie und Klimaforschung (IMK)," to this sentence.

Pg. 1396, climatological PAN profiles: 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.

Pg. 1397, line 8: The "microwindow-dependent continuum radiation profiles" represent the atmospheric particle continuum, which is weakly wavenumber dependent. For this reason one continuum profile is retrieved for each microwindow. Since PAN is evaluated in one contiguous analysis window, only one continuum profile is fitted in this case. Thus the "continuum profile" referred to on page 1398, line 8, is the same feature. We will make this clearer in the paper.

Pg. 1398, lines 20-25: The number of degrees of freedom is larger for the northern midlatitude case, because the available MIPAS scan contains more tangent altitudes



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free of cloud-contamination. Viewed top down, it extends until 7.1 km into the troposphere, whereas due to cloud contamination the useable tropical scan ends at 13.0 km altitude. Further, the temperature and thus the PAN spectral signal in the altitude range 15–20 km is much lower at the tropical than at the midlatitude geolocation.

Figure 2: Figure 2 will be enlarged and the yellow colour will be changed into brown. Each individual error source displayed in Figure 2 will be explained in the figure caption or in the text. The left and right plots of the lower panel of Figure 2 do not have the same error components, because for the reason of clarity only the major components (contribution to total error at any altitude larger than 2% (left) or 4% (right), respectively) are shown. Due to different atmospheric conditions and altitude coverage, slightly different components, e.g. interfering species, proved important. The errors in temperature and LOS were assumed to be 1 K and 150 m, respectively, which is a conservative estimate in comparison to the ESD resulting from the preceding retrieval of temperature and LOS. Both errors are indeed correlated. This correlation is not considered in the PAN error estimation, which causes a slight overestimation of the related errors. Referring to the referee's questions for the reason of the shape of the error profiles (minimum at 13 km in the tropical or spiky behaviour of the midlatitude profile), we first want to stress (as also outlined in the paper) that the PAN error assessment was performed for two individual profiles, using the actual atmospheric conditions, in particular the retrieved temperature profiles. Thus, any structure in the retrieved temperature profile is mapped via the gain function onto the estimated PAN uncertainty. At the African geolocation all errors are minimal at the altitude of the lowest useable tangent height (13 km), because here the sensitivity of the retrieval is maximal, which is expressed by the peak of the averaging kernel at 13 km. The error is larger at 11 km altitude, because here the averaging kernel is considerably smaller. Towards higher altitudes than 13 km the relative PAN error increases because of strongly decreasing PAN amounts. For the same reason as in the African case the error at the midlatitude geolocation is minimal at 10 km and increases towards lower and higher altitudes. The distinct spike at 8 km is due to a sharp local minimum in the retrieved temperature profile. As a measure for the



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natural variability of PAN we refer to Figure 2 in Roberts et al. (2004, see paper), where upper troposperic PAN values in marine air masses measured by various instruments range from 20 to 700 pptv. This variation is considerably larger than the estimated PAN retrieval error.

Pg. 1400, line 5: We agree and will give the correct value of 23% for the overestimation of the "true" PAN amount in the paper. In this context we noticed that some of the values given on pg. 1400, line 10 are also wrong. The correct numbers are "2, 8, 14, 19 and 23%". Note that these values are estimates for the reduction of overestimated vmrs, whereas the values given on pg. 1400, lines 2 and 5 are estimates for the increase of the "true" vmrs. It would certainly be possible to extrapolate the available PAN cross-sections to lower temperatures, as suggested by the referee. However, we prefer not to do this, but to use the data as given and to document the related shortcomings.

Pg. 1400, lines 18-22: As discussed in the following point, we really can not trust the retrieval very much at 19 km. We will make this clearer in the paper.

Pg. 1401, line 20: Figure 5 shows that the significance of PAN values below 100 pptv in single profiles is altitude dependent. There is no sensitivity to low PAN-vmrs (below $\sim$80 pptv) at 17 and 18 km. However, there are other points between 8 and 14 km altitude (violet to green), which exhibit PAN vmrs below 100 pptv but more significant RMS-ratios of up to 1.1. Further one has to take into account that the later discussion deals with PAN averages, for which generally 10–20 profiles have been added up. With regard to the referee's doubts about the significance of negative $CH_3CCl_3$ values, we admit that the $CH_3CCl_3$ amount at 19 km is only slightly negative in case of a PAN-free retrieval. However, the $CH_3CCl_3$ value obtained with the PAN-free retrieval is distinctly negative at 13 km, namely -180 pptv, whereas it is only -10 pptv in case of a retrieval inclusive PAN. We admit that not all retrievals including PAN do always provide positive values for $ClONO_2$, $CH_3CCl_3$ and $C_2H_2$, but much more negative values of $ClONO_2$, $CH_3CCl_3$ were fitted in case of a PAN-free retrieval for an atmosphere obviously containing PAN.



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Figure 8, bottom right: In the first sentence describing the bottom right graph of Figure 8 (pg. 1403, lines 24–27) we nearly come to the same conclusion as the referee. Moreover, considering the referee's doubts and the above new inspection of Figure 5, we will withdraw the bottom right PAN distribution at 20 km altitude from the paper. Further we will also restrict Figure 7 to the altitude region 7–18 km.

Figure 10: The referee is right that the red points (15–16 km altitude) of the $C_2H_2$-PAN correlation include many negative $C_2H_2$-values. But there are also a lot of red points as well as yellow, green and blue points (9–14 km) with low PAN amounts and positive $C_2H_2$ values. Further, the correlation coefficient for data from the altitude of 15 km only is 0.62, which is not as bad as the referee states. Moreover, $C_2H_2$ has no large influence on the retrieved PAN amounts, because contrary to PAN it has sharp spectral signatures. The joint-fit of $C_2H_2$ was mainly performed to avoid spikes in the residual spectra and because it was a good opportunity to retrieve another tropospheric pollutant on low cost.

Pg. 1403, line 1: Yes, we think that industrially polluted air with enhanced PAN vmrs can be transported to high northern latitudes by meridional transport. There are a number of papers discussing the occurence of "arctic haze" containing sulphuric acid and organic compounds, which are assumed to originate from industrial regions. We will refer to Bottenheim and Galland (1989), who report measurement of PAN-vmrs of up to 300 pptv at 7 km altitude at Arctic latitudes of 70°N. However, we cannot exclude any other pollution source like boreal forest fires (cf. Stohl et al., 2006) without further analysis.

Pg. 1406, lines 18-19: We agree and will use a more specific wording.



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