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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.

Anonymous Referee #1

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Referee Comment to the paper 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., ACPD 7, 1391-1420, 2007

General Comments

The paper presents global distributions of upper tropospheric PAN on a global scale, derived from MIPAS limb observations in the mid-infrared spectral region, during a 10 day period between October 4 and December 1, 2003 (150 orbits). The plumes

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of PAN are detected mostly in the southern hemisphere tropics and subtropics (from tropical South America over the southern Atlantic, Africa, eastward to Australia) and are less strong in the northern hemisphere, with largest concentrations over East Asia (East of China). The strong PAN signals seem to be correlated with signatures of biomass burning. The measured concentrations appear to be in agreement with values observed in earlier airborne campaigns in these regions (South Atlantic and South Pacific).

The paper is written very well: it is well structured, the English is good, and precise enough information is provided to understand the retrieval procedures, the sensitivity tests and the interpretation of the results. Only the reference to MOPITT observations - to underline the correlation with other signatures of biomass burning - might have been somewhat more elaborated and better documented. To my knowledge, the results presented here are indeed the first published observations of PAN on a global scale from satellite measurements. It will be interesting to see the announced follow-on paper to elaborate more in depth the geophysical phenomena that are underlying the observations. In this context, the question comes to me what has driven the selection of the period Oct. 4-Dec. 1, 2004. Fire maps show that there have been important boreal fires between May and August over Russia and parts of Northern Asia: has MIPAS observed PAN also for these conditions? I am also a little surprised that the signal over Australia is much weaker than over S. America and Africa, whereas fire maps show many hot spots also in Australia in the Oct-Dec period. So it will be useful to examine the PAN observations as a function of the amount and type of burning activities and to verify the observed differences.

Still, the paper raises some questions as to the reliability of some retrieval results that are discussed in the paper - see "Specific comments" hereinafter. Are the retrieved distributions really reliable on the global scale or are they below the detection limit for a large part of the globe?

Specific Comments

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- Pg. 1393, line 23-24: the thermal decomposition rate is given in units of time (1h) so in fact you're giving the lifetime associated with thermal decomposition instead of a rate. - Pg. 1394, line 12: you're focusing on the upper troposphere, so I don't understand why you are talking about the natural sources in the stratosphere ? - Pg. 1396, first paragraph: is it possible to cite a reference for the IMK MIPAS data processor ? - Pg. 1396, climatological PAN profiles: you say that the present paper provides the first measurements of upper tropo- PAN on a global scale. Where then do the climatologies come from ? - pg. 1397, line 8: What do the microwindow-dependent continuum radiation profiles represent? Is this the same feature that is referred to on on pg. 1398, line 8 with 'a continuum profile' ? - pg. 1398, lines 20-25: how do you explain that you have more information (larger number of degrees of freedom, better height resolution) in the northern midlatitude case, where the PAN signal is much smaller ?? - The lower plots in Fig. 2 are really too small to appreciate. The yellow colour is hardly visible. The legend is not always understood: for example: 'spectro' is spectroscopic errors ? 'tgra' = ? , 'gain' refers to what ? The curve 'param' represents the error due to model parameters, others than the ones explicitly mentioned, or to the total error associated with the mentioned parameters ? More explanation would be welcome, either in the figure caption or in the text. It would also be preferable to use the same colours for the same error contributions in both plots; e.g., "los" is presented in red on the LHS, and in blue on the RHS.

Besides these technical comments, there are some more essential questions raised by the figure and the text about error estimation: Why do the left and right plots not have the same error components? E.g., 'tgra' does not appear in the left plot. Is it not significant ? Also the interfering species are not the same ones, although the same microwindow has been used in both retrieval cases? Are LOS errors and temperature uncertainties not somehow correlated errors, in the sense that an uncertainty in the tangent height (LOS error) will inevitably also induce an error in the adopted temperature profile? How much LOS uncertainty have you assumed to calculate the LOS contribution to the uncertainty budget ? Have you derived this from the dispersion in the fitted

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tangent heights ? The similar question also applies to the temperature uncertainties: are these also derived from the fits ? Why, in the African situation (Fig. 2, LHS), are all errors minimal at 13 km: is there a clear physical reason for that ? Can you comment on the spiky behaviour of the error components in the northern midlatitude case (RHS plot). To better appreciate the total error, and therefore the geophysical usage of the results, it would be interesting to comment the errors in the perspective of the natural variability of the target species. - Pg. 1400, line 5: Following the reasoning about the linear dependence of the retrieval on the temperature of the PAN X-sections, I believe that the 30% overestimation of the true PAN amount at 13 km is an erroneous value and should be 23%. Is it not possible to inter- or extrapolate the PAN X-sections from the given temperatures (from laboratory experiments) to other ones? - Pg. 1400, lines 18-22. If the PAN signature at 19 km is a factor 2 smaller than the NESR, can you trust the retrievals at this altitude ? I am surprised that the error is not larger than 100%? This remark joins my following question. - Pg. 1401, line 20: RMS ratio is close to unity => would this not imply that the error on the target is as large as 100% ? When I look at Figure 4b, I have the feeling that you have no sensitivity at all to the amount of PAN, (at least not for a single spectrum analysis), and therefore, I am skeptical towards all statements in the paper concerning PAN with concentrations of order 100 pptv and smaller (see also Fig. 5). I am not convinced that the retrieved negative values for CH3CCl3 (which has even smaller signatures than PAN) in a PAN-free retrieval are a good proof of the sensitivity to PAN. Do all retrievals including PAN provide positive values for the other interfering species like C2H2, CH3CCl3 and ClONO2? I have the impression from Fig.10 that this is not the case. For example, Fig. 8 bottom right: can we believe the patchy behaviour at this altitude, or is what we see a problem of trying to pull out information below the limit of detection ? Also in Fig. 10: whatever is red below 100 pptv does not show a clear correlation, and includes many negative values for C2H2. So my question is: how can you really demonstrate that you are still sensitive to PAN if the concentrations drop below something like 100 pptv. - Pg. 1403, line 1, industrial pollution is suggested as the reason for having higher PAN in northern

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mid and high latitudes compared to southern latitudes: do you expect that industrial pollution has an impact up to ? 80°N due to meridional mixing? - Pg. 1406, in part. Lines 18-19: I would be more cautious and be more specific as to the conditions in which the fits degrade clearly if PAN is not consideredĚ.

Typographic / technical corrections

- pg. 1395, line 21: measurement - pg. 1398, line 23: eliminate one of both 'and' - pg. 1398, line 29: don't you mean 'in which' instead of 'whereas' ? - Fig. 2: see with "Specific comments" - Fig. 3: it is very hard to distinguish the C2H2 contribution in the LHS plot. It doesn't make much sense to me to (try to) show it in the LHS plot while you omit it in the RHS plot. The RHS plot includes the total modeled spectrum - see traces in the top part of the plot: these should not be shown because they do not bring any information, they are just causing confusion. Typo errors in the caption, last line: 'shown' - Fig. 4: to avoid any ambiguity, it should be mentioned that the residuals are in the same units as the radiance plots themselves. - Fig. 8: I would appreciate having latitude ticks on the left vertical axis, in order to ease the comparison with Fig. 7.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 1391, 2007.