Atmos. Chem. Phys. Discuss., 9, C6796–C6802, 2009
www.atmos-chem-phys-discuss.net/9/C6796/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source classification of upper tropospheric pollution by MIPAS HCN and C_2H_6 global distributions" *by* N. Glatthor et al.

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Received and published: 6 November 2009

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

Specific comments:

P16198, L5, Here and elsewhere:

The ENVISAT satellite is still in orbit and MIPAS is currently operating with 100% duty-cycle. The measurements presented here consist of 54 days from September 2003 to March 2004 only, because at the time the paper was written no further HCN and

C_2H_6 data had been analyzed at IMK. A lot more MIPAS spectral data are available for the time periods September 2002 to March 2004 and from 2005 until present and we intend to expand our HCN and C_2H_6 datasets.

P16198, L18, Here and elsewhere:

The referee states a "lack of HCN" in the northern hemispheric tropics and subtropics in March 2004, which falls into the local biomass burning season. We will include his statement that generally there is a strong biomass burning season in the northern tropics from February to March in the manuscript with reference to his paper. He is right that there is a much lower HCN biomass burning signal than in the preceding fall in the southern hemisphere. However at least some biomass burning activity can be seen above North-East Africa and South-Arabia. As he already mentions the reason for lower northern hemispheric HCN can be lower burning activity in spring 2004 or stay of the polluted air masses in the boundary layer. For comparison, preliminary inspection of MIPAS HCN data of the preceding spring (March to May 2003), which we are currently analysing, indicates a more distinct northern hemispheric biomass burning signal.

P16198, L22:

As suggested, we will change "agreement" into "spatial overlap".

P16199, L5:

We assume, here the referee offers an explanation for the high C_2H_6/HCN ratios measured in March 2004. According to the reference he provides (Bertschi et al., 2003) African open wood cooking fires are characterised by a C_2H_6/CO emission ratio of 0.0145 ± 0.0031. If the respective HCN/CO emission ratio is 0.0085 ± 0.0029 as published by Yokelson et al. (2003) for open savanna fires, the C_2H_6/HCN emission ratio for cooking fires would be $\sim$1.7. This might explain the high C_2H_6/HCN ratios measured

ACPD

9, C6796–C6802, 2009

by MIPAS in March 2004 around Southern Africa. We will add a respective sentence in the manuscript.

P16199, L22-23:

We will add additional references to Yokelson et al. (2007) and Crounse et al. (2009) after the references to Li et al., 2003 and Singh et al., 2003.

P16200, L12:

We agree, that according to more recent publications of Singh et al. (2001) and Jacob et al. (2005) methanol, and not C_2H_6, is the second most abundant organic trace gas in the troposphere. Singh et al. (2001) state that C_2H_6 is the most important tropospheric NMHC. Therefore we will change the respective sentence into "C_2H_6 is the most important non-methane hydrocarbon (NMHC) in the troposphere (Singh et al., 2001)."

P16202, L6:

For the time being we assume our HCN and C_2H_6 datasets as fully developed and the datasets discussed in the paper are the most recent ones. We will clarify this by inclusion of the passage "..., which are the most recent ones" in the manuscript. However, the HCN and C_2H_6 data product is under development for the reduced resolution mode of MIPAS, operated since 2005.

P16202, bottom half:

We are well aware that there are other biomass burning species in the region 715-783 cm^{-1} and, e.g., also retrieve C_2H_2 profiles using microwindows in the spectral region 730–799 cm^{-1}. Generally, the HCN microwindows are custom-tailored such that they as far as possible do not contain C_2H_2 or CH_3Cl lines and there was no indication that we have missed certain species in the fit. We can not easily estimate the influence of fu-

ACPD

9, C6796–C6802, 2009

ran and phenol since we do not have spectroscopic data of these species. However, according to bibliographical references (Cabanas et al., 2005; http://www.scorecard.org) the atmospheric lifetime of these species is around 13 hours only, which makes upper tropospheric concentrations visible by MIPAS rather unrealistic. We will update Figure 2 containing the HCN fits to enable a better impression on the quality of the fits.

P16203, L14–P16204, L15:

As already illustrated in the text (P16203, L18-21) the total error shown in Figure 1c (note: NOT Figure 2) is total HCN precision, which is the sum of the squares of instrumental noise and of the random-like parameter uncertainties also plotted in Figure 1c. The height resolution shown in Figure 1b is not treated as error component but a quantity to estimate the vertical smearing of the signal measured at a certain altitude. Thus it is not included in total HCN precision. The same applies to uncertainties in HCN line intensity and half-width, which are systematic errors and have to be considered to estimate absolute HCN accuracy. We will try to make the error discussion better understandable by changing the figure caption of Figure 1c into “Bottom left: Total HCN precision ...”, by removing the not discussed “param” curve (dashed) from Figure 1c, by moving the sentence about the estimated standard deviation (P16293, L14-15) after the description of the vertical resolution and by inserting the sentence “To estimate the total HCN accuracy, systematic errors have also be taken into account” before the sentence “The major systematic error source ...”.

P16204, L23:

The phrase “analyzed so far” does not mean that the results are preliminary with respect to their quality, but that this was the data available at the time, when the paper was written. Since there are much more MIPAS spectral data available, we plan to expand our HCN and C_2H_6 datasets. We will replace the phrase “temporarily non-averaged

ACPD
9, C6796–C6802, 2009

data" by "single-scan data" as suggested by the referee.

P16205, L13:

Yes, we agree that the October average of HCN in Figure 4 (top left) possibly shows pollution by northern boreal fires peaking earlier in the year in August (cf. MODIS fire counts, http://firefly.geog.umd.edu/firemap). This assumption is supported by the fact that Arctic HCN in September 2003 is even higher (cf Figure 5, top left). We will add a statement on this source of pollution in the paper.

P16205, L16-19:

We regard both the HCN amounts in the Southern Hemisphere (red) as well as those in high northern latitudes (yellow) as enhanced above background values (green) and wanted to express this accordingly in the paper. We try to make this clearer by changing the sentence into "... whereas HCN in northern high latitudes was moderately enhanced to $\sim$270 pptv."

P16207, L12:

We changed "Caribic" into "Caribbean Sea".

P16208, L16-25:

We will include the referee's ideas in the lifetime discussion.

P16210, L1:

In the meantime tropospheric CO was also retrieved from MIPAS measurements (Funke et al., 2009). Comparison of the MIPAS CO values and of the MOPITT CO amounts published by Edwards et al. (2006) shows that both datasets are in reasonable agreement. E.g., both the MIPAS (270 hPa, September/October 2003) and MOPITT CO amounts (250 hPa, end of September 2003) are between 100 and 150

ACPD

9, C6796–C6802, 2009

ppbv in the southern biomass burning plume. Thus, due to easier access, we used MIPAS CO to calculate HCN/CO emission ratios. For the lowermost MIPAS altitudes (8-10 km) the September/October ratios are between 0.003 and 0.004 for the South-African region, which is somewhat lower than the range of HCN/CO ratios published by Yokelson et al. (2003) for South-African savanna fires, namely 0.0085 ± 0.0029. We consider inclusion of a HCN/CO discussion, as suggested by the referee, in the final version of the manuscript. However, this requires some deeper look into the data. An effort to estimate total global biomass burning from our data might be possible after we have produced a more comprehensive dataset.

P16212, 3 general comments:

We concede to the referee that the trajectory plots are "hard to understand", but unfortunately we do not see the possibility to simplify them without removing neccessary information. Further, we will repeat the lifetime of 50–60 days of C_2H_6 and quote the annual magnitude of the industrial sources of C_2H_6 presumed to cause the enhancements discussed on P16212, L21, P16213, L9 and P16214, L15.

P16215, L28:

We will replace the expression "sample of representative air masses" by "selected".

P16216, L11-13:

Yes, we agree that commonly papers describe long range transport from China and Japan to the West Coast of North America. Thus the possibility that the West Coast also receives pollution from Indonesia and northern South America, as indicated by Figure 11, might be a new finding. We will also point this out in the manuscript.

P16216, L18-19:

We will also discuss open wood cooking fires as possible reason for the high C_2H_6/HCN ratio (cf comment to P16199, L5).

ACPD

9, C6796–C6802, 2009

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ACPD

9, C6796–C6802, 2009