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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Hydrogen cyanide in the upper troposphere: GEM-AQ simulation and comparison with ACE-FTS observations" *by* A. Lupu et al.

A. Lupu et al.

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We thank the anonymous reviewer and Andreas Richter for their comments.

Anonymous Referee #2

1) Page 2170, line 13: "The fires in Africa exhibit..."; would avoid possible misunderstanding.

Reply: "The fires exhibit seasonal variation..." will be changed to "The fires in Africa exhibit seasonal variation..."

2) Page 2171, line 13: All sources quantities are provided in Tg N yr-1. What the 41% variabil-

ity refer to is unclear in that respect and should be specified.

Reply: The PMF method was applied to the measured mixing ratios. The text will be changed from "...41% of the HCN variability could be explained..." to "...41% of the HCN variability in the measured volume mixing ratios could be explained..."

3) Page 2173, line 26: Although it is true that SCISAT-1 orbit provides coverage from tropical to higher latitudes, the measurement distribution is very uneven. The authors should say that the orbit was optimized for high latitudes, also providing some coverage of the tropical regions.

Reply: Will be added to text: "The orbit was optimized to provide the largest coverage at high latitudes in winter and spring."

4) Page 2174, lines 10 through 13: Labeling heavier molecular isotopologues with numbers may not sound familiar for many readers. The authors should indicate which isotopologues these number refer to.

Reply: Isotopologues will be explicitly listed in the revised text: "Interferers in the microwindows (retrieved simultaneously with HCN) were H_2O isotopologues $H_2{}^{16}O$, $H_2{}^{18}O$, $H_2{}^{17}O$ and $HD^{16}O$, CO_2 isotopologues $^{16}O^{12}C^{16}O$ and $^{16}O^{12}C^{18}O$, N_2O isotopologues $^{14}N^{14}N^{16}O$ and $^{15}N^{14}N^{16}O$, CH_4 and C_2H_2,..."

5) Page 2174, line 26: The authors should add the extent of signal to noise reduction between 3300 cm-1 and 1400 cm-1.

Reply: The SNR values will be added to the revised text: "Even in the absence of ice, errors above 10 km are smaller than errors below 10 km since the signal-to-noise ratio near 1400 cm^{-1}, being slightly over 300:1, is much larger than it is near 3300 cm^{-1}, *i.e.*, 150:1."

6) Page 2175, lines 24-25. The author state here that the time series where separated between sunrises and sunsets for clarity. However, I don't see any strong argument for the separation

ACPD

9, S2031–S2037, 2009

Interactive Comment

and would think the discussion would even be simplified if there was no distinction between the two datasets. In fact, the seasonal behaviors and in particular the peak concentrations would likely appear better. The authors could consider revising the Figure and the associated text on page 2176.

Reply: It is always challenging to summarize in a single plot this kind of unevenly distributed 4-D data. We have experimented, as suggested, with plotting the time series of vertical profiles for Northern and Southern Hemispheres separately in order to make the seasonal variations easier to visualize, but this resulted sometimes (*e.g.*, in August in Southern Hemisphere) in alternating sunrise and sunset profiles next to one another that are difficult to tell apart as to their spatial location (latitude). As a consequence, we have decided to keep the original plot in the manuscript. The advantage of having a sunrise/sunset separation is that profiles adjacent in time will not be spatially too far apart, thus giving a quasi-continuity to the plot. Moreover, the seasonal variation is discussed later in a quantitative way in the text describing Figure 4 (zonal monthly mean time series).

7) Page 2176, line 3: It is said that "the high UT values in October are the result of peak fire activity in South America, South Africa, insular SE Asia and Northern Australia, as shown in Figure 1". This is difficult to apprehend without discussing simultaneously Figure 3, which shows the actual measured and modeled zonal distributions. It would probably be more sensible to say the "the results are consistent with peak fire activity"; but that even is not fully clear. Indeed from Figure 1, wouldn't one expect larger concentrations in these regions (South America and South Africa) earlier in the summer? In light of this the authors could consider revising somewhat the text around Figure 2 and maybe the Figure itself (see above).

Reply: The temporal variation of the mixing ratio at a given location depends not only on the temporal and spatial distribution of fire emissions, but also on meteorology and on the lifetime of the tracer. The calculated zonal mean is also prone to bias because of uneven (unrepresentative) sampling. For example, August was the month with peak HCN emissions in Southern Hemisphere in 2005, with most of emissions (about 60% according to our estimation; see Fig-

ACPD

9, S2031–S2037, 2009

ure 1) coming out of tropical South America. In September, emissions in S America decreased by about 40%, and in October they became relatively small. Emissions from Africa (the second largest source) also decreased over the same three months. Looking at Figure 4 (zonal monthly mean time series), we see that, in S Hemisphere, the monthly zonal means at 8.5 km increased from August to October. Now, the region with the largest emissions (tropical S America) experiences in August the last month of the dry season, which is characterized by subsidence that tends to inhibit deep convection. Thus, the emissions are rather localized around the source areas and confined to the lower troposphere more often than not. This results in relatively small mixing ratios of HCN in terms of zonal means at our altitude of interest. As the dry season breaks down in September, synoptic activity starts to loft the emissions, transport them quasi-isentropically to the middle to upper troposphere and towards extratropical latitudes, and spread them zonally. At the same time, the atmospheric burden of HCN increases over months, due to the long lifetime of the tracer. This picture is consistent with the pattern of variation seen in Figure 4.

Thus, our statement that "[t]his is a result of peak fire activity... as shown in Figure 1" is indeed misleading if not followed by a clarification. We will remove it from the text describing Figure 1 and add a paragraph explaining the occurrence of the UT mixing ratio peaks when commenting on Figure 4.

8) Page 2177, line 10: How is the subtropical megaplume characterized and where exactly is it to be seen?

Reply: The "megaplume" is characterized by large HCN mixing ratios (>500 ppt) between about 15 and 40 degrees South at 8.5 km altitude during autumn (SON). Since this is not readily visible in Figure 3, where only the occultation data have been plotted, we will add and comment on a 2-D or 3-D plot showing the modelled plume as monthly-averaged HCN volume mixing ratios in October 2005 and 2006.

9) Page 2179, line 6: For the ground-based measurements the HITRAN 1996 database has been

ACPD

9, S2031–S2037, 2009

used. Does it differ from more recent versions for HCN and does that have any impact on the retrieval?

Reply: We have recalculated the columns at Moshiri and Rikubetsu by using spectroscopic parameters from HITRAN 2004 (that were also used for the ACE retrievals). The new columns are larger by up to 7%. This results in an improved agreement between GEM-AQ and retrieved columns for the period August to October 2004, and no dramatic change overall. Figure 6 will be replotted with the recalculated columns and the associated text will be changed accordingly.

10) References: Some Journal abbreviations are probably to be corrected (Journal of Quantitative Spectroscopy and Radiative Transfer)

Reply: We will ask the production office to check the abbreviations.

A. Richter (Editor)

1) The uncertainties of the ACE measurements are only addressed by stating their random errors. Are there no systematic errors, e.g. from spectroscopy? The same question applies for the FTS data.

Reply: The systematic error on the retrieved volume mixing ratios from the spectroscopic constants is 2 to 5%. For the ground-based FTS, the total uncertainty was estimated to be 8% (Zhao *et al.*, 2000, Geophys. Res. Lett., 27, 2085–2087). This will be added to the text.

2) I was surprised that uncertainties in model transport are not considered at all - couldn't that also explain part of the observed differences?

Reply: Uncertainties in biomass burning emissions and their temporal and spatial distribution, in injection heights and in the loss term (as mentioned in the manuscript in sections 2.1 and 2.2)

ACPD

9, S2031–S2037, 2009

are far larger than those in model transport (with the possible exception of subgrid convective transport). An assessment of transport errors in the model is a non-trivial task worth a study of its own.

3) The correlations between measurements and model data in individual latitude bands are not very large - more than half of them are below 0.5. Do you have any explanation for that? Could uncertainties in model transport be part of the reason?

Reply: Indeed, but see our reply to comment 2. We will add a short paragraph summarizing the uncertainties and explaining the correlation values.

4) Have you applied the averaging kernels of the measurements to the model data before comparison or is that not necessary, and if not, why?

Reply: The ACE averaging kernel was not applied to the model, since the kernel for limb-viewing geometry is sharply peaked (*e.g.*, see Clerbaux *et al.*, 2005, Geophys. Res. Lett., 32, L16S01, doi:10.1029/2005GL022394). As for the ground-based columns, SFIT1 is a least squares fitting and averaging kernels are not produced.

5) Abstract: I'd suggest adding the comparison to ground-based FTS data

Reply: Will be added to abstract.

6) Abstract: "Our model results show..." I'd rather say that the comparison to the satellite data shows that the model performs well

Reply: Text will be changed to: "The comparison with the satellite data shows that GEM-AQ performs well..."

7) Fig. 5: It would be good if you could reduce the y-scale to 0-600 to make the figures more clear

ACPD

9, S2031–S2037, 2009

Reply: We used the same *y* scale in both Figs 4 and 5 for easier comparison. In the revised manuscript we will place these two plots side by side in one figure.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 2165, 2009.

ACPD

9, S2031–S2037, 2009