

Review of “What drives the observed variability of HCN in the troposphere and lower stratosphere?” by Li et al.

This paper addresses the sources of the seasonal variability of HCN in the troposphere and lower stratosphere. The authors use a 3D CTM, GEOS-Chem, to compare with groundbased column measurements of HCN and also satellite HCN measurements from the ACE-FTS instrument on SCISat-I and also the MLS instrument on the Aura satellite. They do not address the source of shorter term (shorter than monthly) variability driven by the temporal and spatial variation of the main source, biomass burning. Their main finding is that the principal driver of variability of the variation in biomass burning emissions. They also use investigate the transmission of tropospheric signals/information into the stratosphere.

This is an interesting paper and should be published after the authors address a few comments that I have listed below. The main one is the authors focus on bias which can be misleading and perhaps in this case is misleading in terms of agreement as there are clearly areas of substantial disagreement of absolute amounts. Perhaps a more useful metric would be a standard deviation between models and measurements or even with as a self-measure of variability. Also the presentation of averages, particularly of the satellite data tends to obscure areas of disagreement (cf. for example Lupu et al. 2009, also quoted by the authors).

One lesser problem, but none-the less important is the display of satellite data used in a regime that the authors clearly identify as not being valid, I refer to the use of MLS/AURA data outside of the tropics.

Other comments

P10844, L26/27 “Before we can confidently use HCN to infer surface sources and sinks of trace gases”. I can see that HCN could assist with the inference of other BB gases using emission factors but I am unclear how this would assist with inference of BB-gas sinks not related to HCN.

P10888, L15: Pickett et al references are missing from reference list.

P10889, L27ff: Figure 3d: The ACE data are limb observations: there is no discussion of how the limb observations have been converted or scaled to vertical column observations taken on the ground. The text suggests that perhaps vertical columns between 7-20 km have been taken for the groundbased and for the model. Is this really vertical column? If so why? Why not calculate the slant column? Have the ACE slant column data been simply scaled to force agreement? It would seem unlikely since a relatively small bias has been indicated. However, a simple scaling between vertical and horizontal scaling is $\sqrt{2\pi RH} \sim 40$ where R is the radius of the Earth and H is the scale height. This is very confusing – and need not be.

Some details are necessary particularly if one makes statements regarding bias and as noted above bias alone can be quite misleading if one is talking about capturing variation of HCN. Also are the ACE observations in Figure 3d latitudinal averages appropriate to the latitude of the Jungfraujoch or within some predetermined distance from the Jungfraujoch?

P10890, L3/4. "Comparing to ground-based FTIR spectrometers, the ACE-FTS instrument has lower time resolution.." I think that perhaps this needs a slight rewording. To me time resolution suggests time taken for a measurement whereas temporal resolution suggests, to me, the frequency of observations.

P10891, L4; Figure 4, text suggests that only $\pm 10^\circ$ ACE-FTS data are shown while the data shown are from $\pm 45^\circ$. Better to be clearer here.

..... L7/8. The text indicates that the bias of GEOS-Chem is $\sim 15\%$. However, this does not address the issue of differences which can be quite large $\sim 50\%$ or more. Some more detailed description of differences other than bias would be useful to the reader. What about using something like standard deviation?

..... L11ff: The text claims that figure 4 shows a large UT asymmetry in both model and ACE-FTS mixing ratios. Yes the model does but the ACE data, as presented do not show a strong asymmetry at 100 mb, say. This may simply be a question of contour levels, but again it should be clarified (it is actually clear in the ACE data presented by Lupu et al, 2009 quoted by the authors.). Also the authors talk (line 12) of southern high latitudes but only between $\pm 45^\circ$ is shown.

Also in Figure 4 the ACE data appear to have quite a different character from those of GEOS-Chem in the lower stratosphere; certainly there appears to be a significant bias and from a tape-recorder perspective it would have been interesting to see the vertical profiles of HCN itself and not just the anomalies as shown in Figure 6. It is not just the anomalies that are transported, rather the total gas.

..... L13ff. Figure 4 shows the HCN MLS observations outside $\pm 10^\circ$ and this is really misleading and doesn't do justice to the MLS data. The authors themselves say that it is noisy and required averaging etc and note the impact of HNO_3 , so why show the data outside its limits.

P10892, L1 "with an atmospheric lifetime longer than the transit time from the tropopause to the mid-stratosphere, there is a clear upward transport of the signal from annual fluctuations, which has been called the 'atmospheric tape recorder'" Surely the CO lifetime is ~ 4 months and so doesn't fit the above, but it does exhibit the signature. It is more due to fluctuations in CH_4 (its source in the LS) and perhaps lower down emissions?

Figure 5, anomalies: Hmmm, I am not convinced by the superposed anomaly plots. There seems to be a lot of disagreement at 100 mb. It is certainly an interesting point but because I remain unconvinced is no reason not to explore the idea in a publication.

General: How well determined are the emission sources and sinks since the variability will also depend on the lifetime, in this case (HCN). For example if the sources were increased and one could justify an increase in the sink (ie decrease of the lifetime) this would also increase the variability (This been a addressed to some extent by an earlier paper by Li et al. (2003) but an appropriate summary if their findings would be useful.

Since this is a paper on variability there is no discussion of the standard deviation (SD) or some such metric. The use of bias is useful – but only to a point. The bias can be modified (somewhat) by tweaking emissions and deposition rates but the SD reveals (to some degree) how well the temporally (seasonal) variability has been captured.

Figure 1. Perhaps a table of monthly emissions would be better as it would allow the details in the time series of the various sources to be read, whereas now the details are lost in the bottom recess of the Figure for the lower sources, even though these can be important locally, eg. NA source is not important globally it is important locally. In fact, why not just add the monthly emissions to Table 1?

Figure 2. Same problem here as in Figure 1. This really requires 2 figures as the details of the individual sources are lost in the bottom of the Figure. Thus I would suggest one figure for the general comparison and one figure for the individual sources.

One other query here. In Figure 3d column amounts from ACE are shown but there is no discussion of how limb measurements were translated into vertical columns (see above).

Figure 3 has the same problem with Figure 2 in that the information located at the bottom is unreadable.

In general the labeling in the Figures is rather small. I know that they can be blown up electronically but even then some material is indecipherable such as the column units in Figure 3d.