

Response to referee # 2:

We thank the reviewer for a useful examination of our paper. As the reviewer suggested, we have significantly increased the length of data used for this study, including model simulations. Also, we have changed to use the less biased MLS weekly zonal mean products (see below), to validate the model HCN. Below are our responses to specific comments (shown in italics, numbers have been added here).

*1) 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.*

We agree with the reviewer that biomass burning variability occurs on timescales much less a month but we acknowledge that UTLS variability of HCN will likely be dominated by larger timescales e.g. months. We acknowledge this in the revised manuscript.

*2) 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).*

The bias represents the systematic errors, indicating the difference between two datasets which is an efficient method in comparisons of model and observations. We have also included the standard deviation analysis as the reviewer suggested. Although the averages may obscure some area of disagreements, we found the averages more useful to reveal the general statement of the vertical and latitudinal distribution of HCN mixing ratio.

*3) 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.*

The MLS version 2 standard daily products of HCN used in our previous analysis are not valid. However, in the revised manuscript we have changed to use the MLS offline weekly zonal mean products of HCN (Pumphrey et al., 2008) which are valid not only in tropics but also outside of the tropics.

*4) 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.*

As the reviewer suggested, we have reworded this sentence to “Before we can confidently use HCN to infer its surface sources and sinks ...”.

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

We have added the missed references in the reference list.

6) 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  $2RH\pi \sim 40$  where  $R$  is the radius of the Earth and  $H$  is the scale height. This is very confusing – and need not be.*

The ACE data used in this paper are level 2 vmr profiles data interpolated onto a 1 km vertical grid. In Figure 3, we calculated the vertical columns using the same equations for both the ACE data and the model simulation, taking into account the pressure and temperature data available from the different data sets. The total and partial vertical columns for the ground-based data are retrieved from the FTIR observations, supplied by colleagues from University of Liege.

7) *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?*

In Figure 3d, we compute a latitudinal average of ACE data that fall within  $\sim \pm 5^\circ$  from the Jungfraujoch station (46.6°N, 8.0°E). We have added these details to the Fig.3 legend and the corresponding text.

8) 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.*

We have reworded the sentence as suggested by the reviewer.

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

Figure 4 shows  $\pm 45^\circ$  (in the revised manuscript we used  $\pm 60^\circ$ ) while we use the  $\pm 10^\circ$  data to calculate the tape recorder. We have clarified this.

10) ..... 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?*

We have added the analysis of standard deviation in the comparison between the model and the satellite observations.

11) ..... 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.*

As the reviewer suggested, we have re-arranged the color/contour interval and extended the latitudes to  $\pm 70^\circ$  in Figure 4. We have also corrected the text about the hemispheric asymmetry of HCN.

12) *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.*

We have added plots of profiles of HCN mixing ratio and standard deviation along with the anomalies in Fig.6.

13) ..... 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<sub>3</sub>, so why show the data outside its limits.*

In Figure 4, we have changed to use the MLS offline weekly zonal mean products of HCN with smaller biases (Pumphrey et al., 2008), to validate the model HCN mixing ratio.

14) 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<sub>4</sub> (its source in the LS) and perhaps lower down emissions?

Using the MLS measurements, the signal of annual tape recorder of CO can only be observed below 20 km (Schoeberl et al., 2006, 2008). However, CH<sub>4</sub> has a strong QBO signal in the stratosphere rather than an annual cycle (Randel et al., 1998). The annual variability of CO in the stratosphere is unlikely due to fluctuations in CH<sub>4</sub>. In this case, CO<sub>2</sub> might be a better choice than CO: see Andrews et al. (1999, JGR).

15) 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.

We used the superposition in Figure 5 to compare the temporal variability of HCN between the observations and the model. This superposition clearly shows a good agreement between satellite observations and model, including the timing and magnitudes of HCN anomalies.

16) 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.

As the reviewer suggested, in section 4.1 we have included a discussion of the sources and sinks of HCN and their determining role on the lifetime of HCN in our model.

17) 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.

We have added the standard deviation analysis along with the bias analysis between the model and the observations.

18) 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?*

The monthly emission of individual regions including NA can be found in Figure 2.

*19) 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.*

As suggested by the reviewer, we have changed Figure 2 from a single figure to two figures, one for the general/global comparison and one for the individual/regional sources.

*20) 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).*

The ACE columns are vertical columns, calculated directly from the ACE level 2 vmr, temperature and pressure profiles. Please see our response to the 6<sup>th</sup> question.

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

We have changed Figure 3.

*22) 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.*

We have modulated the labeling in the Figures accordingly.