Atmos. Chem. Phys. Discuss., 14, C4726–C4735, 2014
www.atmos-chem-phys-discuss.net/14/C4726/2014/

Atmospheric Chemistry and Physics
Open Access
Discussions

Interactive comment on "Seasonal and interannual variations of HCN amounts in the upper troposphere and lower stratosphere observed by MIPAS" *by* N. Glatthor et al.

N. Glatthor et al.

norbert.glatthor@kit.edu

Received and published: 11 July 2014

We thank referee 3 for her/his helpful comments.

Reply to general comments:

The referee criticises that there are a "number of places throughout the manuscript, where the authors make unsupported assertions in explaining the behavior observed in their data." Therefore we will try to back up such assertions in the revised paper: We will add time series of GFED fire carbon emissions from different regions, which will facilitate the interpretation of the observed upper tropospheric HCN distribution. Sec-

ondly, we will show upper tropospheric windfields from the NCEP/NCAR reanalysis to confirm the postulated eastward transport of the observed African plume. Further, we will add plots of the outgoing longwave radiation (OLR) from the NCEP/NCAR reanalysis to explain the time lag between fire emissions and upper tropospheric HCN maxima observed by MIPAS.

Reply to specific substantive comments:

P8999, L25:

We will change the wording into "an almost unambiguous tracer" here and in the conclusions.

Section 2.2:

Contrary to the referee's impression, the retrieval results shown in this manuscript were not produced using a revised algorithm, but with the algorithm described in Wiegele et al. (2012) and in Section 2.2 of this publication (RR-mode retrievals). We will make this clearer in the updated manuscript by focusing the description of the retrieval to the setup actually used. Further the referee criticises that the bias between MIPAS and ACE-FTS HCN data is mentioned in the Conclusions, but not discussed in Subsection 2.2. However, this bias is discussed in the comparison to ACE-FTS in Subsection 3.2. We think, we should leave the discussion at this place. A dedicated comparison with MLS data was not performed, since the standard HCN product of MLS is restricted to the stratosphere and the focus of this paper is the UTLS region. The discussion of the retrieval error for a single MIPAS scan will be performed more precisely by giving error estimates for the UTLS region for enhanced and background HCN. In quoting stratospheric error estimates the impression will be avoided that HCN plumes extend into the middle stratosphere.

Section 3.1:

The referee feels it is not appropriate to call climatological features of enhanced HCN "plumes", because this term should be used only to describe particular events. We will introduce another term like "enhancements" or perform a wider definition of plumes.

P9004, L23:

We will add the suggested citations (Duncan et al., 2003, 2007; van der Werf et al., 2010) to show that northern hemispheric biomass burning typically occurs in this season.

P9005, L17-18:

We will add "boreal" in front of "summer" at the beginning of this paragraph and at other occasions. Further we will rewrite the sentence "Stratospheric HCN reflects the fully developed Antarctic vortex." as follows to clarify its meaning: "Due to subsidence of mesospheric air masses in the Antarctic vortex, this season exhibits the lowest stratospheric HCN amounts, observed at high southern latitudes."

P9005, L23-26:

We will cite, e.g., Edwards et al. (2006) and Glatthor et al. (2009) as reference for southern hemispheric biomass burning during this season and discuss elevated HCN

values at 40–60N.

P9006, L1-8:

We will reorder the paragraph as suggested.

P9006, L15-17:

We will explicitely refer to the bottom left graph of Fig. 7 (May 2005) and outline that the northern subtropical HCN anounts of May 2006 (bottom right graph) are untypically low.

P9007, L17-19:

The referee criticises the unsupported hypothesis of northward transport of northern hemispheric HCN in the discussion of Fig. 3. To support the hypothesis, we will refer to Fig. 4 or omit the statement at this place completely.

Section 3.2:

(1) The referee calls the qualitative comparison between the MIPAS and ACE-FTS by reference to plots in previously published papers instead of actually showing some comparisons not very convenient, but then suggests to add "(not shown)" at the end of the second sentence in this section. We will follow this suggestion.

(2) The referee asks, which Figure of Randel et al. (2010) confirms poleward transport of enhanced HCN following the southern hemispheric biomass burning season. This transport can be seen in Figure S2 of the supporting material. The confusion is caused by combined listing of references for two processes (low HCN amounts over the tropical oceans and poleward transport). We will separate the references to these two processes to make things clearer.

(3) The referee believes that the different underlying time periods will not lead to substantial discrepancies between the MIPAS and ACE-FTS HCN climatologies. But since some of the referenced ACE-FTS climatologies cover considerably shorter time periods (e.g. Lupu et al. (2008) analyse the period 2004–2006 only) this difference can cause discrepancies at least to a certain extent.

(4) She/he is puzzled about suggesting the use of different spectral regions as possible reason for systematic deviations of about 20% between HCN VMRs observed by MIPAS and ACE-FTS and feels that more explanation is needed. According to the error codes in the HITRAN database, the spectroscopic uncertainties of the strongest HCN lines in the spectral regions used for MIPAS and ACE-FTS retrieval are 5–10%, both for line intensity and for pressure broadening. These systematic error contributions have not been included in the error estimation presented in Section 2.2. If they are added to the upper tropospheric MIPAS HCN retrieval error of 5–15%, the deviations between MIPAS and ACE-FTS are within the error bars. Thus, both instruments capture the HCN distribution within the combined uncertainties. We will add some sentences explaining these issues both in Sections 2.2 and 3.2.

(5) The referee states that a systematic bias can not explain the differences in the vertical extent of the northern and southern hemispheric tropical and subtropical enhancements. She/he is right. This is a basic difference between HCN observations of ACE-FTS and of MIPAS.

(6) We agree that the difference in sampling of ACE-FTS and MIPAS is another explanation for differences and had already checked this issue. For example, the much sharper gradient of the autumnal (Sep–Nov) southern hemispheric plume observed by ACE-FTS towards high southern latitudes (Randel et al., 2010, Fig. S2) can at least partly be explained by sampling of regions south of 70°S in September, only. MIPAS HCN data restricted to September also show a sharper gradient than HCN data aver-

aged over the period September to November.

P9011, L10-13:

Â■The referee notes that the statement "entry of enhanced HCN into the lower stratosphere seems to be somewhat more effective in the northern than in the Southern Hemisphere, but the dominance of the Asian Monsoon is not as distinct as shown by Randel et al. (2010, Fig. 3)" needs more explication. First, with our statement we are not only referring to 22 km in Fig. 4 of our manuscript, but also to 18 km. For better comparison to the ACE-FTS/MLS-plot, we also averaged the MIPAS time series over the altitude region 16–23 km. But this does not lead to different conclusions. One reason for different interpretation of MIPAS HCN in the tropical UTLS region is analysis of a longer time series than in the reference papers. The MIPAS time series additionally contains the years 2010–2012 and especially 2003 with larger contributions from the Southern Hemisphere.

P9011, L24-27:

The referee criticises that our explanation for longer meridional transport times in the years 2002 and 2006 is not backed up by some references. We did not give any reference here, because we thought the statement is explained by the MIPAS measurements shown in Fig. 4 (10 km) itself. She/he wonders, why we did not look at GFED emissions. As mentioned above, we will introduce time series od monthly GFED fire emissions in relevant regions for better confirmation of our statements at the beginning of Section 3.

P9012, L11-17:

The referee criticises, that it is not meaningful to calculate HCN trends in the northern and southern hemispheres without presentation of error bars. Since referee 1 also criticises the discussion of trends, we will omit the presentation of trends.

P9012, L26 - 9013, L16:

(1) The referee is right. We will change the wording into "... in October most enhanced HCN amounts were observed above northern Australia and Indonesia, indicating strong biomass burning and effective upward transport in this region."

(2) The referee criticises that our discussion takes no account of the time lag between surface emissions and the appearance of HCN enhancements in the upper troposphere. We will address this problem by referencing the findings of Liu et al. (2010), by comparison of maxima in MIPAS HCN with GFED emission data and by investigating the seasonality of tropical deep convection.

(3) The referee criticises the wording in referencing the GFED fire emissions presented by Li et al. (2009). This sentence will be completely omitted in our revised manuscript, because we will refer to the GFED time series, which will be included in a revised paper.

P9015, L5:

We will change the citation to "(Randel et al., 2010, and references therein)".

Â■ P9015, L23:

We will change the wording as suggested.

P9017, L3-4:

Although the levels of 14, 17, 20 and 23 km are not completely separable, they were chosen to localize the transition from semi-annual to longer cycles and to track the time

lag of the tropical HCN variations with increasing altitude.

P9017, L3-21:

The referee misses the discussion (here and elsewhere) of the importance of seasonal variations in the strength and location of convection. To account for deep convection, we will introduce plots of the outgoing longwave radiation (OLR) from the NCEP/NCAR reanalysis and discuss the interplay of deep convection and enhancements in upper tropospheric HCN.

P9017, L20 and P9018, L9:

We will compare our time scales for vertical transport more closely to time scales for vertical transport of H_2O given by Mote et al. (1996) and Schoeberl et al. (2008), for vertical CO transport derived by Liu et al. (2013) and with HCN time scales estimated from plots presented in Pumphrey et al. (2008) and Li et al. (2009).

P9017, L26-29:

The referee thinks that our statement "... Randel et al. (2010) ... from analysis of ACE-FTS and MLS data conclude, that the HCN amounts at the tropical tropopause are too low for effective supply of stratospheric HCN" is too strong. We will reread the cited publication and update our statement. In case our statement was right, the requested implication is that according to observations of HCN by MIPAS troposphere-to-stratosphere transport via the AMA is not as important as with respect to HCN observed by ACE-FTS and MLS.

P9018, L3:

Right, the averages are slightly different, because they were calculated over the time periods covered by the respective plots.

p9018, L16-18:

We will change ”strong AMA of 2005" into ”strongly polluted AMA of 2005".

P9018, L26-29:

As pointed out be the referee, we will emphasise that low HCN was already observed at the beginning of 2008. The positive anomaly of autumn 2009 probably could not propagate into the stratosphere, because it was not followed by subsequent pulses in 2010. We will add this explanation in the revised paper.

P9019, L5 and 16:

The referee misses a reference for the biomass burning at the end of 2011. We will reference Fig. 4 (14 and 18 km) for the relatively strong biomass burning at the end of this year. Further, she/he states that the maximum of the tape recorder signal in 2011 is missing in L16. We will update the manuscript accordingly.

P9019, L26-28:

The referee states that the statement made here more or less agrees with the conclusions of Randel et al. (2010) and contradicts the statement made earlier that our findings are in opposition to those of Randel et al. But here we list three sources of positive HCN anomalies: Extensive southern hemispheric and Indonesian biomass burning followed by a strong AMA containing large amounts of HCN.

P9020, L6-9:

We will cite publications of the water vapor tape recorder by Mote et al. (1996) and

Schoeberl et al. (2008). Further, we will make the vertical propagation of HCN and H_2O anomalies better comparable by plotting the slope of the HCN anomalies into the H_2O time series.

P9021, L27-29:

We will give a concrete number for the time shift and weaken the differences to Randel et al. (2010).

P9022, L8-9:

Yes, the conclusion that the periodicities of the HCN tape recorder is similar to the findings of Pommrich et al. (2010). We will mention this fact in the revised manuscript.

P9022, L17-19:

As mentioned above, we will compare our ascent rates to rates of Mote et al. (1996), Schoeberl et al. (2008), Liu et al. (2013), Pumphrey et al. (2008) and Li et al. (2009).

Minor wording and grammar comments:

The suggested changes will be performed.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 8997, 2014.