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Atmospheric Chemistry and Physics
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.

Anonymous Referee #3

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This manuscript presents a climatology of HCN in the UTLS based on 10 years of MIPAS measurements and explores spatial, seasonal, and interannual variations in the HCN distribution. In particular, variability in biomass burning signatures is investigated and used to study the tropical tape recorder and horizontal transport timescales. In general I think that applying the MIPAS HCN measurements to the study of these topics makes a valuable contribution that will be of interest to the ACP readership. However, in my opinion there are a few weaknesses in the analysis, or the description thereof, that should be corrected before the paper can be published. Most notably, there are a number of places throughout the manuscript where the authors make unsupported

assertions in explaining the behavior observed in their data. Although the mechanisms they assume are plausible, the paper would be strengthened considerably if the authors backed up these assertions with more rigorous analysis or even, in many cases, merely more diligent referencing of previous work. These and other issues are described in detail below.

Specific substantive comments:

– p8999, L25: "an almost unique tracer of biomass burning" is an ambiguous statement that could be interpreted to imply that no other species are biomass burning tracers. I believe the authors meant that HCN has few other sources, but the wording should be clarified. A similar phrase ("a nearly unique tracer of biomass burning") in the Conclusions (p9020, L20) should also be changed. I note that the wording used in the Abstract ("an almost unambiguous tracer of biomass burning") is much clearer.

– Section 2.2: It is mentioned that retrieval of MIPAS HCN was described previously by Wiegele et al. [2012] (for RR data) and Glatthor et al. [2009] (for HR data). However, my impression is that the results shown in this manuscript were produced using a revised algorithm and are thus not exactly comparable to those presented previously. This point needs to be clarified. If it is indeed the case that the data presented in this manuscript were produced with a different algorithm, then more validation of the data is warranted. In any case, since the MIPAS HCN climatology is discussed in connection with those from ACE-FTS and MLS, some information on how all of these data sets intercompare would be appropriate here. It is mentioned in the Conclusions that MIPAS HCN values generally exceed those from ACE-FTS by 50 pptv, so it seems odd to me that no such statement is made in the "Retrieval method and error estimation" section. Furthermore, the characterization of the error is confusing. It is stated that "the retrieval error for a single MIPAS scan observing a tropospheric biomass plume burning plume is 12-15% in the upper troposphere and 18-22% in the lower and middle stratosphere" – do the authors really mean to quote a value for a tropospheric biomass burning plume in the middle stratosphere? This sentence needs to be rewritten for clarity. What is the error in background air masses in which HCN is not enhanced (I presume the accuracy is lower (worse) in these cases)?

– Section 3.1: This is purely a matter of terminology, but I do not feel that it is appropriate to refer to the climatological features based on seasonal averages over a 10-year period being discussed in this section (e.g., persistent biomass burning signatures or the AMA) as "plumes", a term that to me should be used only to describe particular events.

– p9004, L23: I don't doubt that the enhanced HCN observed by MIPAS is related to biomass burning, but some general citations showing that burning typically occurs in this season at these latitudes is needed (e.g., Duncan et al. 2003, 2007; van der Werf et al., 2010; others).

– p9005, L17-18: I have no idea what the sentence "Stratospheric HCN reflects the fully developed Antarctic vortex." is referring to. I'm guessing that the authors may have intended to describe the effects of unmixed descent inside the vortex bringing down air poor in HCN from above, but that needs to be clarified. Also, it is somewhat jarring to start the paragraph discussing the "summer" period and end it with the Antarctic vortex. Either add "boreal" in front of "summer" or refer to the specific months covered in the panel.

– p9005, L23-26: As above, general references for southern hemispheric biomass burning during this season should be given here. In addition, the northward extension of the HCN enhancement does not terminate at 20N – elevated HCN values appear to be present up to 60N, and they should also be discussed.

– p9006, L1-8: I suggest slightly reordering this paragraph, moving the last sentence up to after the first sentence, so that all of the northern hemisphere material is discussed together, and then going on to the horizontal transport in the southern hemisphere.

– p9006, L15-17: "This feature becomes even more obvious in a climatology restricted

to the month of May, where enhanced amounts of HCN prevail in the whole northern subtropics (cf. Fig. 7)." One problem with pointing to Fig. 7 here is that it shows May averages for two years, during one of which (2006) HCN values were very low throughout the northern subtropics.

– p9006, L25-26: "A smaller part of the polluted air masses is obviously transported northeastward from Africa over South Asia to the northern Pacific." In my opinion, the authors tend to overuse the word "obviously", which by my count occurs 11 times in this manuscript. This is the first instance of it, and it typifies their approach. I agree that the results in Fig. 2 suggest the occurrence of pollution transport as they have described, but a definitive conclusion cannot be drawn on the basis of this plot alone. At the least such an assertion needs to be substantiated through referencing of detailed modeling studies that might support it (e.g., perhaps Duncan et al. [2007], Liu et al. [2010], or Barret et al. [2008] might be appropriate references – I did not go back and check these myself).

– p9007, L17-19: "The summer maxima at 10 km at northern mid- and high latitudes are caused by boreal biomass burning and partly by northward transport of pollutants released at lower latitudes." Again, I'm sure that this is true, but it is poor form to make such an assertion with no attempt to support it. Ideally tagged model runs or sensitivity tests would be used to confirm this hypothesis, but I accept that such efforts are beyond the scope of this paper. However, as noted above, inferences for HCN can be drawn by analogy to other species for which such modeling and comparison to measurements has been performed, such as CO.

– Section 3.2:

(1) The discussion of the qualitative comparison between the MIPAS and ACE-FTS HCN distributions is somewhat awkward, as the way it is presented the reader expects to actually see some of the comparisons. Instead, the results are reported through reference to plots in previously published papers, which is not very convenient. I suggest

adding “(not shown)” at the end of the second sentence in this section.

(2) I am not completely confident that all referenced plots really show what the authors state. They point to Fig. 1 of Randel et al. [2010] for the poleward transport of enhanced HCN following the southern hemispheric biomass burning season, but Fig. 1 of that paper is a map that cuts off at 30S. Perhaps they mean the time series of Fig. 3, although that plot is cut off at 40S?

(3) They attribute the discrepancies between the MIPAS and ACE-FTS HCN climatologies in part to the different underlying time periods they are based on. However, since both climatologies cover multi-year periods, the specific years included should not make that substantial a difference. Moreover, I believe that the time periods used for each climatology do not differ by that much.

(4) Another possible cause suggested for the discrepancy is the use of different spectral regions in the MIPAS and ACE-FTS retrievals. This statement puzzles me, and I feel that more explanation is needed. After all, there is only one HCN distribution in the atmosphere, and if the measurements are reliable then they should capture that distribution no matter which wavelengths are being used. The differences should be within the combined accuracies of the respective data sets.

(5) The possibility of a systematic bias between the two data sets is also mentioned. But such an offset could not explain the differences in the vertical extent of the northern and southern hemisphere enhancements between the two sets of observations that is reported here.

(6) In my opinion, the most likely explanation for the disagreement between the MIPAS and ACE-FTS distributions is the difference in the sampling of the two instruments. Not only does ACE-FTS, as a solar occultation instrument, lack global daily coverage, but also its orbit is such that coverage of the tropics is particularly poor. In fact, ACE-FTS averages only a few hundred occultations intermittently during a few months of the year in the tropics and subtropics, as Fig. 3 of Randel et al. [2010] shows. The authors

should explore this point in more detail.

– p9011, L10-13: The authors state that “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)”. I feel that this conclusion needs more explication. I assume that they are referring to the fact that at 22 km the MIPAS peak mixing ratios are more centered around the equator and stand out less against the background than those plotted in Randel’s figure, but this should be explained more clearly. It should not be assumed that all readers will have the Randel paper in front of them when they read this paper. Moreover, it seems to me that the MIPAS data at 18 km exhibit patterns that are more similar to those in the 2010 paper than do the data at 22 km, and an average of those two levels might provide a fairer comparison. It might also be noted that Randel’s Fig. 3 includes both ACE-FTS and MLS HCN data, and that those two data sets show fairly good agreement, which perhaps calls into question somewhat the MIPAS results.

– p9011, L24-27: This is another example of the authors making statements (in this case about interannual variations in the location of biomass burning hotspots) that are not backed up in any way. Their explanation is certainly plausible but would be much stronger if supported, at least by some references. When I first read this section, I wondered why they had not looked at GFED emissions to substantiate some of their claims. They do at least cite some papers that used GFED emissions in the following subsection, but at this point in the manuscript the reader does not know that. In fact, this entire subsection (3.3) would benefit from some words at the beginning telling readers that some of the variations noted in presenting the time series will be discussed in greater detail in subsequent sections.

– p9012, L11-17: Trends in HCN in the northern and southern hemispheres are calculated from the MIPAS data set, but no error bars on the reported values are given. The statement is made that “Future analysis will address the significance of MIPAS HCN trends in more detail". If the statistical significance of the derived trends is not known, is it really worth mentioning them? I think it is potentially misleading to report trend estimates that may turn out not to be significant.

– p9012, L26 - 9013, L16:

(1) First, a minor comment about wording: it is not quite correct to state that "the focal points of biomass burning were above northern Australia and Indonesia". In fact, the burning took place on the ground; it is only in the satellite data that the burning products appear above these locations.

(2) More importantly, this discussion takes no account of the time lag between emissions at the surface and when HCN enhancements appear in the UT. Beyond the fundamental transport timescales involved, a delay between the peak in fire emissions and the peak in UT CO abundances over southern Africa in November has been attributed to subsidence temporarily inhibiting convection and trapping pollution in the boundary layer [e.g., Liu et al., 2010].

(3) Here (L9-11) the authors do confirm the above-average strength of the burning activity in Indonesia in 2002 through referencing Li et al. [2009]. But they need to be a little more precise in their wording. GFED does not directly provide HCN emissions; rather, Li et al. calculated them by scaling from an observed HCN:CO emission ratio over the western Pacific.

– p9015, L5: Randel et al. [2010] is by no means the only relevant reference for the AMA, so please change the citation to "(e.g., Randel et al., 2010, and references therein)".

– p9015, L23: Again, GFED does not provide HCN emission fluxes directly. I suggest changing this wording to "… HCN emission fluxes calculated from the GFEDv2 biomass *burning* inventory".

– p9017, L3-4: Unlike Figure 3, which showed results at 10, 14, 18, and 22 km, Figure

9 looks at 14, 17, 20, and 23 km. But how separable are these different levels, given the vertical resolution of the MIPAS HCN data (4-5 km in the troposphere, 6-8 km in the stratosphere)?

– p9017, L3-21: Another factor that I think is missing from the discussion (both here and elsewhere in the manuscript) is the importance of seasonal variations in the strength and location of convection. It is not only the variations in biomass burning that force a tape recorder signature, but also those in tropical deep convection.

– p9017, L20: Is the time scale for the transport from 20 to 23 km inferred from the MIPAS HCN data consistent with that estimated in other studies using observations of the tape recorder to calculate ascent rates (e.g., Mote et al., 1996; Schoeberl et al., 2008; Flury et al., 2013)? A similar question can be asked about the statement on p9018, L9 about transport from 17 to 25 km.

– p9017, L26-29: The authors state that "This result is different to the findings of Randel et al. (2010), who 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". I am not sure that I agree with this statement. Randel et al. [2010] certainly argue strongly for the importance of the Asian monsoon circulation, but do they really explicitly draw the conclusion that is attributed to them here? Even if so, what are the implications of the distinction between Randel's conclusion and that based on the MIPAS data?

– p9018, L3: The anomalies plotted in Figs. 8 and 10 were calculated by subtracting the average value at each altitude. I assume that the averages were calculated over the time periods covered by the respective plots, that is, the average used in Fig. 10 was different from that used for Fig. 8?

– p9018, L16-18: "The second positive anomaly … seems to be caused by the strong AMA of 2005". As written this sentence seems to imply that the meteorology of the 2005 Asian monsoon was different from that of other years, whereas I believe that the authors mean to refer to the severity of the pollution trapped inside the AMA in that year.

– p9018, L26-29: Actually, the beginning of 2008 was also characterized by low HCN. In addition, is there any explanation for why the "positive anomaly of autumn 2009 could not propagate into the stratosphere"?

– p9019, L5 and 16: Is there a reference for the biomass burning at the end of 2011? Again, the explanation is plausible, but 2011 has not been discussed previously, so some support for this statement is needed. In addition, the weak maximum in the tape recorder signal in 2011 should be mentioned in L16.

– p9019, L26-28: It seems to me that the statement "Effective upward propagation of positive HCN anomalies into the stratosphere obviously occurs during periods of extensive southern hemispheric and Indonesian biomass burning followed by a strong AMA containing large amounts of HCN" more or less agrees with the conclusions of Randel et al. [2010] and thus apparently contradicts the statement made earlier in the manuscript that these findings are in opposition to those of Randel et al..

– p9020, L6-9: It would be appropriate to cite some references on the water vapor tape recorder here. In addition, it is not possible to gauge "by eye" that "the temporal shift of the H2O anomalies with increasing altitude obviously agrees quite well with the stratospheric vertical speed observed in HCN", especially given the size of the two panels in Fig. 10. Overlaying (maybe in light grey) a contour of HCN on the H2O plot would facilitate the comparison.

– p9021, L27-29: The first sentence is rather vague (a time shift of "some months"), and as noted earlier I am not convinced that their results are notably different from those of Randel et al. [2010], as stated in the second sentence.

– p9022, L8-9: "These periodicities . . . are introduced by source strength variations". Is this not the same basic conclusion that Pommrich et al. [2010] reached?

– p9022, L17-19: Again, comparison of the ascent rates implied here with previous estimates would be appropriate.

Minor wording and grammar comments:

– p9000, L23-24: "MLS" was already defined in L14 above

– p9001, L12: replace "shortly" with "briefly"

– p9001, L16: delete comma after "show"

– p9001, L23: delete "has been"

– p9002, L4 and L6: replace "have been" with "were" and "has been" with "was"

– p9002, L23: "IMK" not defined until the following page

– p9003, L17: I think that the hyphen in "joint-fitting" should be deleted

– p9003, L21: I think that "ozone was joint-fitted" would be better as "ozone was fitted jointly"

– p9003, L24-25: replace "were" with "included" and add a comma after "height-independent"

– p9004, L1: delete "plume"

– p9004, L12: delete hyphen in "upward-shift"

– p9004, L13: replace "less" with "fewer"

– p9004, L14: replace "some dozens" with "a few dozen"

– p9004, L25: "during *the* previous"

– p9005, L4: "as dynamic tracer" would be better as "as a transport tracer"

– p9005, L8: replace "northern" with "Arctic"

– p9005, L19-20: add “boreal” in front of “spring” and “autumn”

– p9005, L24: “stretches”

– p9006, L16: replace “where” by “when”

– p9006, L19: adding “Fig. 2, ” in front of “top right”, would make the discussion easier to follow

– p9007, L13: replace “nearly” with “essentially”

– p9008, L9-10: “ACE-FTS” was already defined on p9000

– p9008, L28: “different to” should be “different from”

– p9009, L11: replace “little” with “limited”

– p9009, L19: “a thousand”

– p9009, L20: replace “less values were binned” with “were fewer values binned”

– p9011, L27: replace “nearly no” with “very little”

– p9014, L5: “MLS” has already been defined

– p9015, L19: “bottom”

– p9017, L15: add a comma after “2006”

– p9017, L26: “different to” should be “different from”

– p9018, L7: delete “, respectively”

– p9018, L18: replace “a region” with “an interval”

– p9018, L20, 22, 27, and 29: add “boreal” in front of “autumn” or write specific months

– p9019, L1-2: as in previous comment

– p9021, L20: delete the second “and”

– Fig. 5 and Fig. 9: the titles of the plots will not be meaningful to most readers, especially as 221 was not even mentioned in Section 2.2

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