Atmos. Chem. Phys. Discuss., 10, C15266–C15272, 2011
www.atmos-chem-phys-discuss.net/10/C15266/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global distributions of acetone in the upper troposphere from MIPAS–E spectra" *by* D. P. Moore et al.

D. P. Moore et al.

dpm9@le.ac.uk

Received and published: 21 October 2011

We thank the referee for their valuable comments which helped improve the paper. One of the main concerns raised in the general comments was that there was not a very convincing discussion of the results. The main aim of the paper is to present the first reported, global, dataset of acetone in the upper troposphere from spaceborne instrumentation and we have now included an extended geophysical discussion of the results. The retrieval set-up is further expanded to include discussion of the retrieval approach.

General comments:

1)"The paper is in parts not written very clearly, and cannot be understood without first

reading Moore and Remedios (2010). While all the details on the retrieval do not need to be repeated here, a brief summary of the analysis is needed to make the paper more readable. Also, the error discussion should follow after the method was described."

We have rewritten the measurements section to make the retrieval scheme and error sections clearer to the reader. We do not want to completely reproduce the background theory from Moore and Remedios (2010), but have rewritten this section to include all aspects of the retrieval set-up including a priori information and covariance set-up. What we do aim to clarify in the article text are the retrieval inputs (i.e. spectroscopy, a priori assumptions, treatment of interfering species). The error discussion follows the method.

2)"Reference is made to the detection of acetone in MIPAS-B balloon data, but another fitting window is used and the fit results look much less convincing than in remedios et al. 2007. Please explain a) why only a small windows was used, b) what the background correction is and why you only apply it only for the red curve shown in figure 3 and c) how you can exclude interference by other species."

Point a) – There are two reasons. The first is instrument-determined as unfortunately the MIPAS spectral band coverage is 1020 to 1170 cm-1 in band AB and 1215 to 1500 cm-1 in band B. Hence measurements aren't made between 1170 and 1215 cm-1 and hence MIPAS misses a large portion of the acetone P-branch. We note this point in the revised paper. Secondly, we tested the use of more microwindows over a larger spectral range and found that this greatly increased the time taken for processing. From a test on several orbits, the results between the small and large windows gave a similar global distribution. This particular microwindow (1216.75 to 1217.5 cm-1) also had the highest information content for acetone from simulations. We have added comments in the text to clarify the point.

Point b) we are not sure what the reviewer means by the background correction. The acetone was jointly retrieved with continuum which leads to point c) as we retrieve all

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known interfering species before we attempt acetone retrievals (rather than joint-fit). This approach does not attempt to exclude interfering species, rather to give us the best estimate of these gases and their contribution to the measured spectrum. Since this step is performed before acetone retrievals and using other spectral regions, what are left with in the acetone microwindow is assumed to be the acetone signal (+MIPAS noise).

3)"Why are the acetone results not shown also on the lowest (and arguably most interesting) layer in figure 4?"

Actually, the lowest levels are poorly sampled and we have unintentionally overclaimed on the retrieval range. Hence we have changed the retrieval range for good data noted in the paper and altered the zonal mean plot which was Figure 5 in the original paper. In fact it is not at all clear that the most interesting layer is the lowest one, since the largest acetone effects on ozone occur at the higher upper tropospheric levels where one also finds significant inter-continental transport.

4)"The zonal distribution in Fig 5. Looks odd to me – there is no clear separation between troposphere and stratosphere in the acetone distribution which needs to be explained or at least discussed. Why should acetone which is released at the surface show a smooth transition into the lower stratosphere? Could this be linked to averaging kernels of the measurements?"

It is not quite clear what the reviewer means by a “clear separation” so we clarify what is expected.

Briefly, from a physical retrieval point-of-view, it is most certainly the case that averaging kernels will smooth the vertical gradient (see main answer below) . Secondly the plot is a gridded monthly mean data set which will smooth across the tropopause also, averaging both tropospheric and stratospheric air. Our altered plot removes the smoothing element and “sharp changes” with altitude are more apparent. Finally, in the stratospheric overworld, there is little sensitivity in our retrieval but profiles are assumed

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to have very low values of 50 pptv commensurate with literature (see next paragraph).

Perhaps the most important point, however, is that from an atmospheric process point-of-view, it is understandable but naive of the reviewer to assume an “a priori” discontinuity at the tropopause. We refer the reviewer to the excellent paper of Sprung and Zahn, ACP, 2010 which uses CARIBIC aircraft acetone and ozone data to examine acetone in the lowermost stratosphere (LMS). In the summer time, acetone is shown to propagate into the extra-tropical (mid-latitude) LMS with 150 to 350 pptv present in the 2 km above the tropopause. Given the spacing of MIPAS retrieval levels, this corresponds to one MIPAS level above the tropopause. So there will be a strong vertical gradient which MIPAS will observe in a smoothed way. This idea is entirely consistent with our plot and indeed “LMS”-type values are observed close to the tropopause in our data. We comment on this in the revised paper.

One should also note that acetone is not produced just at the surface but there is also a potentially strong but uncertain secondary chemical production source.

5)"There is no discussion of the vertical resolution and information content – please add typical averaging kernels and a short discussion."

The averaging kernel aspect is relevant to both points 4 and point 5 which we answer in more detail together as the issues raised overlap. We thank the reviewer for the opportunity to clarify. We have included an example in the text of a “typical” acetone averaging kernel from MIPAS. What we find is that the upper tropospheric averaging kernels have a typical width (or vertical resolution) of 3 to 4 km. The stratospheric averaging kernels are slightly broader with a resolution of 4-6 km and are likely to smooth over the upper part of the troposphere. This is the most likely explanation for the smooth troposphere to stratosphere transition, but also noting the evidence from aircraft data from CARIBIC as described above.

6)"In summary, it is stated that acetone enhancements “linked to biomass burning” are observed in several regions – this is not discussed in the paper and I don’t see any

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evidence for that in the figures. Please remove or provide support for the statement."

We thank the reviewer for noting this. The aim of this work is to show the first global observations of acetone in the upper troposphere as measured from space rather than apportion the sources (which is to be done next in a coupled paper linking data with a chemical transport model). So we had reduced the geophysical discussion which we are happy to improve. The addition of wind field information (and examination of trajectories) clearly shows larger levels of acetone in the biomass burning outflow regions of Africa and South America. There is particularly good agreement for Africa with 150 hPa MOPITT V3 data and MIPAS C2H2 (see Parker et al, ACP, 2011). We have removed the statement from the paper.

7)"In several places, the paper reads like a draft with repetitions and mixed up sentences."

We have removed repetitions and mixed-up sentences from the text.

Detailed comments:

Here we outline the agreements to the reviewers suggestions. Where further clarification is needed we have addressed the point specifically.

a) "Page 23540 l4: in August"

Changed as requested"

b) "Page 23540 l9: you report “up to 3000 ppt in the upper troposphere” but in line 11, you give 20000 ppt as upper limit."

The text has been altered to represent the fact that upper troposphere values reach up to 2000 ppt. (We believe that the reviewer meant 2000 ppt as stated elsewhere in the paper, not 20000 ppt as written in the comments)

c) "Page 23540, l9: What are high distributions?"

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This has been altered to read “high vmrs” – high volume mixing ratios, with vmrs being defined earlier in the paragraph.

d) "Page 23540, l11: reversible transport between troposphere and stratosphere not discussed in the paper."

We have removed this comment from the paper.

e) "Page 23542, l5; “during the nearly continuous first two years of flight” – I assume that ENVISAT flight was not only “near continuous” – do you mean continuous measurement."

We thank the reviewer for pointing this out, this is indeed what was meant. The text has been altered to read “during the first two years of measurements.”

f) "Page 23542, l23; gives confidence."

Text changed accordingly.

g) "Page 23542, l26; mention that these were balloon measurements."

The text has been rewritten to reflect this and reads “The presence of acetone spectral signatures was demonstrated by Remedios et al. 2007 from a balloon-borne version of the MIPAS.”

h) "Page 23545, l13; retrieval is performed."

Text changed accordingly.

i) "Page 23545, l15; of the remaining"

Text changed accordingly.

j) "Page 23545, l20; within the MIPAS-E noise"

Text changed accordingly.

k) "Page 23546, l9; this sentence is odd, please check."

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We agree with this statement. The text has been edited to remove “and suggesting that if the ocean. . .” until the end of the sentence.

l) "Page 23543, l27; designed to be able"

This has been removed in the author response to general comment (1).

m) "Caption Fig. 5; zonal distribution of acetone"

We have removed the extra occurrence of the word “acetone” from the text.

n) "Caption Fig. 6; why are there only 6 points if daily averages are shown."

The text should have pointed to the fact that these data are 5-day averages of acetone. The word “daily” has been replaced with “five-day” in the article.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 23539, 2010.