

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

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We would like to thank referee for their comments and we look to address the issues raised.

Major comments:

a) "Wide parts of the paper are speculative. Vague phrases like 'are possibly linked', 'show the likely strong influence', 'we may expect', 'give confidence that', 'we believe that', 'inaccuracies were set conservatively at... ', 'it is likely' are used at many places where clear or quantitative statements are needed instead."

The phrases are not intended to be speculative. We should have been more assertive and are more so in the revised paper where this is justifiable. The reviewer needs to

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bear in mind that acetone distributions are only just being explored as an observational-led activity and a principal activity is to characterise the distributions since these are quite different to the models. In fact, some aspects of acetone behaviour are quite different to the models and this is not explained as yet but needs to be articulated and evidenced. It is worth the reviewer examining the recent paper by Elias et al, ACP, 2011, describing CARIBIC observations of acetone.

b)"Abstract: The decrease of acetone mixing ratios with altitude is reported in the abstract but not in the results section. Is this decrease significant? Without any specific diagnostics like averaging kernels it is not possible to judge if there is any significant altitude information at all."

To answer this point we have included a representative averaging kernel figure in the text. Most averaging kernels for our data show the highest vertical resolution at lower altitudes (3-4 km vertical resolution at FWHM at ~9 km altitude) and decreasing to between 4 and 6 km resolution at the higher altitudes. The averaging kernels tend to zero at and above 18 km and so all information comes from the a priori data for these data points. From these data we do conclude that the decrease with altitude is significant. Compare the maps at 185 hPa and 277 hPa where the scales have changed between the 2 levels (185 hPa scale maximum is nearly a factor of 3 smaller than at the lower level). The mid-latitude values of 700 pptv max. at 185 hPa are consistent with tropopause values of 650 pptv median at the tropopause from Sprung and Zahn, ACP, 2010.

c)"P 23543 I15: A low signal does NOT mean that the sensitivity is low. The sensitivity depends on the jacobian, not on the signal. Low signal combined with large Jacobian allows to significantly retrieve zero abundances. It is not essential to see an acetone signal but to know that one would see it if it were there. Averaging kernels are needed to judge if the result is significant or if the priori information is reproduced."

Similar to b) above, we have shown a typical example of an acetone averaging kernel

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and discussed the implications for gas concentration sensitivity and vertical sensitivity.

d)"P23543/4: The description of the retrieval is inaccurate: i) How are background continuum and zero-level calibration treated? ii) In the retrieval section these quantities and related strategies are not mentioned at all but in the caption of figure 3 continuum is mentioned – is the continuum constrained? iii) Is there any prior information on elevation pointing used? iv) Does the retrieved water vapour abundance really represent the water vapour signal in the acetone spectral region? Spectroscopy might be inconsistent between the spectral range used for water vapour retrievals and the spectral range used for acetone retrievals."

We have significantly re-written the retrieval set-up section to address these points and should have provided this in the first version. We address each issue in turn: i) The background continuum is jointly retrieved in the retrieval microwindow for each species independently. Since cloud filtering is performed, the background continuum is constrained to low values commensurate with this cloud index and typically provide radiance contributions less than the noise. We do not retrieve a zero-level calibration but assume that the level 1b calibration is correct to the order of 2-3 nW (Kleinert et al, ACP, 2007). ii) we now discuss the continuum retrieval (and non zero-level calibration retrieval) in the section explicitly. For iii) we use the engineering altitude from MIPAS level 1b (V4) data as a prior and use this to retrieve and altitude during the pressure and temperature retrieval step. This retrieved altitude is then used for the retrieval of each other species in turn. For iv) we retrieve water vapour using the four operational microwindows (and masks) used for the full-resolution retrieval. We therefore use the best estimate of retrieved water vapour in the acetone retrieval and residuals seem to fit quite well. We agree that spectroscopy might be inconsistent but find no evidence for this from either the retrieval residuals plotted in Figure 4 or the new figure showing global statistics for the residuals (fits within the expected noise envelope)

e)"The data characterisation is incomplete. Besides the lack of dedicated error estimation information, averaging kernels, estimates of altitude resolution, horizontal resolu-

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tion are missing. The data characterization is far below state of the art, and some of the results are not supported unless the data are carefully analysed. Error analysis has to be performed for the retrieval presented; it is not sufficient to report error evaluated for another data set generated with another retrieval scheme. Further it is not clear why the biases of the interfering species are used to estimate the single profile error of acetone. Single profile total errors (precision and bias) are needed instead and the use of biases keeps the estimated error components of the acetone retrieval artificially small."

We have presented an analysis of the error calculation for individual acetone profiles. To address the first point, we have now included averaging kernels along with altitude and horizontal resolution. The retrieval scheme used has been in development since 2004 and has been compared by ourselves to the operational ESA MIPAS products with no significant bias between the datasets. We should have made this clear in the paper. The errors were calculated based on a climatology of acetone derived from model data in the presented latitude bands. Because of the influence of several interfering species (figure 1 of original paper), errors based on the contribution of each of these are presented. If we had performed a joint retrieval then we would not have had to include these errors but our retrieval scheme is sequential.

Due to tests we performed to check that the difference between the MORSE derived products and the ESA operational product were small, we used biases, from published results, based on the ESA data products. We also show monthly averages in the paper and so the random component of the retrieval error will be reduced. The systematic error will remain unchanged (model parameter bias and spectroscopy chiefly) and so becomes the most important error in our results. The instrument gain, shift and its errors were calculated but were below the 1% level.

f)"P 23545 I15/16: 15% convergence failure is a lot. Is there some explanation why so many retrievals do not converge"

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We thank the reviewer for pointing this out. This was an error in that these data were from the lowest level of the retrieval (not used in this paper). Of these data at the lowest level, 8.0% had a chisq of between 2 and 10 and only 1.7% a chisq of greater than 10. The remainder were actually flagged as “bad quality spectra” from the MIPAS data quality information. We have rewritten the section to include the 277 hPa convergence success of 97.3% (with 32% of data classed as cloud-free) and the 185 hPa convergence success of 99.9% (with 59% of data classed as cloud-free)

g)"Fig 3 and related text: The purpose of Figure 3 seems to be to convince the reader that there is indeed substantial information on acetone in the measured spectra and that inclusion of acetone in the retrieval improves the fit. At first sight the plot seems to support this: The red residual line (fit under consideration of acetone) is actually much closer to zero than the fit without consideration of acetone. This appears to support the robustness of the retrieval. Closer inspection of the figure and related text, however reveals that not only the treatment of acetone was different but also that of inclusion of the background continuum: A major part of the improvement of the fit is caused by the inclusion of the background continuum. Thus this plot fails to prove that inclusion of acetone indeed actually improves the fit and the reviewer is left wondering what the intention of the authors might have been to manipulate this sensitivity study as they have done."

We object strongly to the wording used by the referee in this point. No attempt was made to “manipulate” this sensitivity study and the inclusion of other supporting evidence through averaging kernels and residual fits shows the sensitivity we achieve for acetone. As we state clearly in the text we perform a joint-retrieval of acetone and background continuum, as it good practise for atmospheric retrievals, and so we showed that by inclusion of the joint retrieval data, acetone and continuum, that we fit to the MIPAS spectrum within the noise equivalent spectral radiance. We do accept that there was an unintentional ambiguity in the figure and have included continuum effects explicitly in the revised figure which will clarify the referee’s concern.

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h)"P 23546 I5: The authors attribute enhanced acetone values in the upper troposphere to particular source regions but no trajectory calculations are presented to support these speculations. Particularly it has to be clarified if gas emitted in the candidate region is actually uplifted into altitudes where MIPAS can see it."

Trajectory models often struggle to capture vertical transport on the small scales associated with convection. We have, however, performed trajectory calculations for a number of regions which we will use to comment in the revised paper. However, we also believe that we have over-stated the “source” argument for acetone and are grateful for the opportunity to rectify this. There actually seems to be increasing evidence of secondary acetone production and we will comment on this. The presence of this factor as well as the chemical loss terms means that tying together surface emissions from a single source with observations at 9 km to 12 km is a challenge. We place clear limits on the implications that can be drawn but state where the direction of evidence lies as also shown by aircraft data. Indeed we find surprisingly good agreement with findings from Elias et al, ACP, 2011, particularly in the region of Europe.

i)"Fig. 4: From comparison of the distribution at 277 hPa with that at 185 hPa we see that above clouds there are particularly low acetone values. Can this be an artefact at 185 hPa caused by discarding cloud-contaminated spectra at 277 hPa tangent altitudes?."

We performed further analysis on this with 1) only data where the measurement level directly below the retrieval level was clear (i.e. cloud-index greater than 4) and 2) as 1) but with data “cloudy” at the level below (i.e. data which weren’t used in the retrieval). The differences in global median value at the 185 hPa level were reduced from 240 pptv to 182 pptv when ONLY data clear at 9 km were included but this change in the average was due to the removal of the lower acetone vmr data over the Western Pacific. Other plots also do not clearly indicate the effect that the referee is looking for. As it is unclear whether the persistent cloud at 277 hPa (~9 km) causes a bias in the data at 185 hPa (~12 km) we believe the plot should remain unchanged.

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j) "p 23546, l13 ff: From monthly averages of acetone distributions in combination with monthly averages of meteorological data no conclusion on mixing into the "middle-world" can be drawn: At days when acetone was enhanced at higher altitudes, the tropopause altitude may have been higher. This analysis requires to investigate each single measurement along with a relevant altitude instead of monthly means to be conclusive."

We agree that is not possible to draw conclusions about the likely mixing in single events, in tropospheric folds for example, based on observation of monthly averages of data. The average tropopause level was put on as a guide to the reader. This is perhaps mis-leading and so we propose to resubmit the plot without the PV levels and do not draw conclusions about vertical mixing. We also determine this as the averaging kernels have a vertical resolution of 3-4 km it is difficult to draw firm conclusions about the mixing from these data.

k) "p 23546 l26: analysis of the temporal development of acetone at 277 hPa again is based on the assumption that all emitted acetone immediately is uplifted to the 277 hPa level. Couldn't the same time series be explained by reduced uplifting due to reduced convective activity towards the end of August."

We thank the referee for this observation. We agree that reduced convection over the later period of the month could be a contributing factor to the trend, not just reduced biogenic activity. We also note the influence of secondary chemical production. We have added this to the text.

l) "fig 5. If spots of enhanced acetone in the tropics were indeed caused by biomass burning as larger source, one would expect enhanced mixing ratios also at lower altitudes. A maximum at 300 hPa (c.f. fig 5) with lower values below can hardly be explained by a local source. How shall lower mixing ratios below the maximum be explained."

It is important to note that this plot is a monthly average, rather than a weekly, syn-

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optic scale, plot. It is clear from the trend plot that we're seeing changes in acetone throughout the course of the month. The referee is also making an assumption here about the nature of biomass burning plumes and convective outflow. CALIPSO data, for example, shows that there are well defined plume altitudes. Some of the confusion is also caused by the zonal mean plot which we have now corrected so this point is no longer relevant in any case.

m) "In summary, the description of the retrieval is incomplete, data characterization is far below state of the art, the robustness test is , well, suspicious, and the explanation of results in terms of atmospheric sciences is neither convincing nor conclusive but pure speculation."

In taking this final comment in parts we would like to state the following. The retrieval was not fully described in the paper although it is accepted that additional explanation (which was presented in a previous paper by this author) should be included to explain the approach to a general audience and provided necessary details. For the data characterization, we have included example averaging kernels so that the general reader can see where MIPAS is sensitive to measurement information. Being a limb-sounding instrument, these averaging kernels are sharply peaked in the upper troposphere, broadening in the stratosphere where the sensitivity (and gas concentration) decreases with altitude. It is also a standard approach to look at the fit of the residual in the way we present. We show that the microwindow used to retrieve acetone shows a marked improvement to the spectral fit, improving to within the MIPAS noise levels (which must be remembered are around 15 nW for this band). As acetone microwindow is relatively small, we show an extended wavenumber range to see the change to the fit but emphasising that the best fit occurs with higher acetone concentrations. We are sorry that this has made the referee "suspicious" but are confident of our own integrity. Finally, we have taken on board the comments kindly made by the referee about the explanation of the results in terms of atmospheric science and improved where necessary. We are particularly pleased to be able to better utilise

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the evidence from the CARIBIC aircraft data which have been published at the same time as this paper. We note again that the aim of this paper is to present a very new dataset of acetone and show the first global distributions of this species in the UTLS. These data will be invaluable to modellers (for example) to test the robustness of chemistry/transport schemes. They strongly make the very important point, in conjunction, with recent aircraft data of the need to understand the acetone distributions which do not fit well in some regions with model results.

Minor issues:

a)"Title and elsewhere: The established acronym for the spaceborne instrument is just "MIPAS". Although the term "MIPAS-E" may be more accurate because there do exist also other MIPAS instruments, one should comply with the established naming convention in order to avoid confusion."

We use the term MIPAS-E to distinguish it from the also well- established balloon-borne version of the MIPAS (MIPAS-B) and aircraft versions (MIPAS-STR). We agree that the "MIPAS-E" is not commonly used in the literature so have removed the "E" from the article.

b)"P 23541 I5 Why mentioning the acronym MPIK? It is never used in the following"

This acronym has been removed as requested.

c)"P 23541, I15/16 There are not only limb FTIR instruments but also nadir-looking ones (TES), thus the restriction to the UPPER troposphere is inappropriate."

We do not dispute that nadir-viewing instruments such as TES and more recently IASI provide information for some gas species in the upper troposphere; note however this is often limited to major uplifts into the upper troposphere (Clarisse et al, GRL, 2011). The main advantage of some of the limb-viewing instruments, which should have been made here, is the high spectral resolution offered by instruments such as MIPAS (and ACE on SCISAT) so that it is possible to resolve line structure in gases with lower at-

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mospheric concentrations from stronger absorbers such as water vapour and carbon dioxide. We have adjusted the text to read "The advent of spaceborne limb-viewing Fourier transform spectrometers, with high spectral resolution, has allowed the possibility to distinguish lines of weakly absorbing species from stronger absorbers such as carbon dioxide in the upper troposphere and lower stratosphere." This is of course in addition to the well-known vertical profiling abilities of limb sounding instruments.

d)"P23541, I16 "One such instrument. . ." (if restricted to limb emission) are there any other?"

We have made it clearer in the text that MIPAS measures limb-emission. Other limb-viewing instruments are capable of measuring trace gases, such as the ACE on SCISAT which uses solar occultation.

e)"P 23542 I9: "3X30x400 km": The fields of view is 2D, the third dimension is unclear. If the horizontal resolution (information smearing) is meant, how was this number derived? Or is the horizontal sampling meant? Then the number (400km) is incorrect. Anyway, it is not appropriate to call this quantity which seems to refer to the along-track dimension field of view."

We apologise for the confusion. The field-of-view should be defined as the 3 km (vertical) x 30 km (horizontal) view of MIPAS. The text has been altered to talk only about this instantaneous field-of-view. The 400 km referred to the approximate along-track resolution due to the long path view of the FTS. We agree that this figure is incorrect and should be closer to 510 km.

f)"P 23534 I9/10: "nominal mode": This term is ambiguous because there are a high resolution nominal mode and a reduced nominal mode. Judging by the date of the measurements it must be the high resolution nominal mode but this needs explicit mentioning."

We agree that this may cause some confusion. We initially left this out as only data

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from the "original" mode were considered in the study before the problems with the slide mirror of the FTS. For completeness, we agree that this problems and subsequent change in data mode should be discussed and is something we have put into the text. We make it clear that data from the full-resolution nominal mode are used in this work.

g)"P 23542 I14: "Here" does this refer to this (Moore et al.) or to the Waterfall paper? This seems a little ambiguous to me. When reading this part for the first time, I understood "Here (in the discussion paper as opposed to the Waterfall paper) a set of ..." which is probably not meant."

We apologise for the ambiguity. The text has been altered to state that we mean the work of Waterfall replacing "here" with "In their work"

h)"P 23542 I 21 "infrared absorption cross section" infrared is obsolete."

We have removed the word "infrared"

i)"P 23543, I10 and elsewhere: Pers. Comm references need initial(s) and year."

We have made these changes.

j)"P 23543, I20 The Rothman reference is older than the updates used, thus there is actually no reference to the updates used. A web address may help, or references to the original data sources"

We have added a link to the HITRAN website which lists updates to the line data

k)"P 23544 I7: What are "model parameter errors"? I suspect that these are temperature and trace species profiles used in the forward model. Why are these errors systematic? Don't they change with time?"

That is correct, these are temperature and pressure and gases in the microwindow. We have made this clearer in the text. These errors are systematic in the sense that various validation campaigns have shown a systematic bias of MIPAS retrieved data compared to in-situ data. We only use data only for a single month in the analysis,

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shorter than timescales over which these errors are likely to vary.

l)"Figure 3: RFM this acronym is undefined; if the Reference forward model is meant a reference is needed"

This has been amended in the text

m)"P 23545 Results: This section contains a lot of technical information which does not belong in the results section."

We have made changes to the results section which now does not include this sort of technical information

n)"Fig 6 The figure caption mentions "daily averages", while related text says "5-day averages"."

These data are 5-day averages. The figure caption has been modified.

o)"p 23547 I2: Why is the chemical notation C<sub>3</sub>H<sub>6</sub>O presented only here."

This has been moved to where we first mention acetone in the text.

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

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