

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

Anonymous Referee #1

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The paper on acetone retrievals from MIPAS spectra by Moore et al. provides new information which - once proven that the results are robust - might be useful to the scientific community. Numerous methodical problems and major scientific and presentation issues, however, will have to be clarified before this paper can be recommended for publication in ACP.

Major Issues

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.

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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.

p 23543 | 15: 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 abundance. It is not essential to see an acetone signal but to know that one would see it if it was there. Averaging kernels are needed to judge if the result is significant or if the priori information is reproduced.

p23543/4: The description of the retrieval is inaccurate: How are background continuum and zero-level calibration treated? 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? Is there any prior information on elevation pointing used? 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.

p 23544: The data characterization is incomplete. Besides the lack of dedicated error estimation information, averaging kernels, estimates of altitude resolution, horizontal resolution are missing. The data characterization is far below the state of the art, and some of the results are not supported unless the data are carefully analyzed. Error analysis has to be performed for the retrieval presented; it is not sufficient to report errors 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.

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p 23545 l15/16: 15% convergence failure is a lot. Is there some explanation why so many retrievals do not converge?

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 plots seem 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 the background continuum: A major part of the improvement of the fit is caused by 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.

p 23546 l5 ff: 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 source region is actually uplifted into altitudes where MIPAS can see it.

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? Averaging kernels will certainly look different for full profiles and such where the lowermost measurements are discarded. This effect could also be an issue at other longitudes, where still large cloud coverage is detected but a few valid spectra remain.

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

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tropopause altitude may have been higher. This analysis requires to investigate each single measurement along with the relevant tropopause altitude instead of monthly means to be conclusive.

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 end of August?

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?

In summary, the description of the retrieval is incomplete, data characterization is far below the 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.

Minor Issues

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.

p 23541 l 5 Why mentioning the acronym “MPIK”? It is never used in the following.

p 23541 l 15/16: There are not only limb FTIR instruments but also nadir-looking ones (TES), thus the restriction to the UPPER troposphere is inappropriate.

p 23541 l 16 “One such instrument...” (if restricted to limb emission) are there any other?

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p 23542 l 9: “ $3 \times 30 \times 400$ 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’.

p 23542 l9/10: “nominal mode”: This term is ambiguous because there are a high resolution nominal mode and a reduced resolution nominal mode. Judging by the date of the measurements it must be the high resolution nominal mode but this needs explicit mentioning.

p 23542 l14: “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 the first time, I understood “Here (in the discussion paper as opposed to the Waterfall paper) a set of ...” which is most probably not meant.

p 23542 l 21 “infrared absorption cross section” infrared is obsolete.

p23543 l 10 and elsewhere: Pers. Comm references need Initial(s) and year.

p 23543 l 20 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.

p 23544 l 7: What are “model parameter errors”? I suspect these are temperature and trace species profiles used in the forward model. Why are these errors systematic? Don't they change with time?

Figure 3: RFM this acronym is undefined; if the Reference Forward Model is meant, a reference is needed.

p23545, Results: This Section contains a lot of technical information which does not belong into the results section.

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Fig 6 The figure caption mentions “daily averages”, while related text says “5-day average”; please clarify.

p 23547 l 2: Why is the chemical notation C_3H_6O presented only here?

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