

Interactive comment on “Acetone variability in the upper troposphere: analysis of CARIBIC observations and LMDz-INCA chemistry-climate model simulations” by T. Elias et al.

Please find my answer to the comments made by referee #1. Referee comments are in italic.

Anonymous Referee #1

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General comments:

- The authors present a detailed analysis of the LMDz-INCA simulation of acetone, through comparisons to aircraft and (some selected) surface observations. It is obvious that a lot of effort has gone into this work. However, without any disrespect to the authors, I found it hard to keep interest in their results despite what is quite a decent model-observational comparison. I would therefore urge the authors to really make an effort to shorten the manuscript and make it much more ‘punchier’. For example, during the comparisons there are lots of absolute quantities/values given in the text, when sometimes a mere percentage bias is more intuitive of the model performance.

We thank the referee for the useful comments. The manuscript has been shortened and some absolute values have been replaced by percentages to facilitate comparisons.

- Similarly, it is hard to know what conclusions can be drawn when differences to the observations are found. In such instances (e.g., top left panel of Fig 2) are the model emissions, chemistry or transport incorrect? Therefore, after a good 3 hours of solid reading, I’m still left wondering what this analysis tells us about the acetone budget that we didn’t already know from previous studies. That said, I still recommend publication in ACP, but only once the manuscript has been made more concise.

First of all, we would like to insist on the originality of the CARIBIC dataset, which is not often used for model evaluation. Global chemistry models cruelly suffer of the lack of global coverage dataset. Furthermore their evaluation is often focussed on ozone or carbon monoxide and far less on oxygenated compounds. However such compounds, partly secondary, are of interest to assess our ability to simulate ozone, but for good reasons. Hence, such comparisons confirm the general validity of the information and hypothesis, all integrated in the current climate models, using a new observation data set.

Specifically CARIBIC gives the opportunity to study the model efficiency and to reproduce the acetone annual cycle in UT and at regional scales. Agreement on annual global values confirms the global budget computation, which suggests stronger acetone sources (and a higher acetone burden) than previously estimated, due to increased primary terrestrial biogenic production. To our knowledge, previous studies did not present regional budget analysis, showing that South Asia and Europe are net source regions, contributing up to 40% to the NH acetone chemical

production, and up to 25% to the NH primary emissions. However, the acetone burden is proportional to the covered surface area, suggesting efficient acetone transport beyond these regions.

Comparison with CARIBIC indicates satisfactory representation of acetone source and transport to UT over South China, and underestimation of acetone in air masses subject to long range transport in UT over the Atlantic Ocean to Europe. Comparison with CARIBIC could even validate the modelled annual cycle of acetone vmr in UT over South China.

Another conclusion come precisely from this complexity: since acetone shows large spatial and temporal variability, it is hard (or even impossible), to produce climatology ready to use for model benchmarking. From a methodological point of view it's important to highlight.

Specific comments:

- Why are flight numbers inserted in the main text at certain points, when details of each flight are not given anywhere?

Flight numbers were erased, dates are kept to identify the flight when necessary.

- Page 9168, line 16: properly define OH and HO₂.

Now written as hydroxyl radical OH and hydroperoxy radical HO₂.

- Page 9168, line 17: Provide the reader with a suitable reference that explains tropospheric ozone production from CO and VOC oxidation.

We have added this reference:

Brasseur et al. Atmospheric Chemistry and Global Change, Oxford University Press, USA, 1999.

- Page 9168, line 23: The authors casually write ‘...considering 300 pptv acetone in a photochemical box model was sufficient to reach near agreement between measured and simulated OH concentration in the UT.’ Without an in depth read of the cited Wennberg et al. (1998) paper, I could assume the difference between the measured and observed OH could be due to any number of issues; please explain this work in more detail.

A sentence has been added to express that acetone may contribute to improve agreement but not in all cases:

“Indeed, Wennberg *et al.* [1998] could reach near agreement between measured and simulated OH concentration in the UT by considering 300 pptv acetone in a photochemical box model, under specific conditions. For example the OH production is more sensitive to acetone at low solar zenith angle, and differences reaching factor 5 are still observed”

- Page 9171, line 18: What does ‘. . .)’ imply? Page 9171, line 19: Put the model resolution in

degrees rather than the number of grid cells.

We removed ‘...’.

Model resolution converted to degrees: “from 3.75° lon x 2.5° lat to 3.75° lon x 1.875° lat spatial resolution”

- Page 9171, line 21: Does the chemical scheme include the latest findings from the Peeters et al (2010) work which allows extra HO_x generation from isoprene oxidation?

No, the chemical scheme of LMDzINCA does not include the latest findings by Peeters et al. (2010). Thank you for the reference. However, even if such results are of high interest in particular to understand the high OH levels measured in tropical canopy, their implementation in 3D model chemical mechanism are not trivial as shown by Archibald et al. (2010), and would require other laboratory study to confirm their robustness.

A. T. Archibald, M. C. Cooke, S. R. Utembe, D. E. Shallcross, R. G. Derwent, and M. E. Jenkin, Impacts of mechanistic changes on HO_x formation and recycling in the oxidation of isoprene Atmos. Chem. Phys., 10, 8097–8118, 2010 www.atmos-chem-phys.net/10/8097/2010/ doi:10.5194/acp-10-8097-2010

- Page 9172, line 8: What are ‘primitive equations’ – explain. Page 9172, line 18-19: What were the sizes of these positive/negative biases from the Folberth et al. 2006 paper?

Primitive equations are: Equations of motion, hydrostatic equation, equation of continuity and thermodynamic equation.

Few absolute values are given by Folberth et al. [2006] about comparison of acetone vmr (other species are also discussed). These indications are given:

“The comparison of acetone profiles with observations show a fairly good model-to-data agreement over the entire troposphere.”

“It generally also reproduces the acetone mixing ratios observed in the Asian outflow during PEM-WEST B, but significantly overestimates acetone at the near continental sites measured during the SONEX campaign.”, The figure showing i.e. 900 pptv simulated for 500 pptv observed, as largest difference.

- Page 9176, line 10: The vertical profile of the global budget also depends on transport, and from a modelling perspective our ability to correctly simulate boundary-layer mixing (to vent from the surface to higher altitudes) and full atmospheric mixing processes e.g., convection.

Sentence corrected as:

“Because primary emissions and dry deposition occur exclusively at surface level, the only source/sink terms impacting the vertical profile of the global budget are the chemical terms”

- Comment on section 2.2.2: There are lots of numbers being thrown at the reader, which

truthfully are not absorbed. Please consider isolating the truly important points/budget terms and make the section more concise.

The section has been made more concise.

- Page 9177, line 13 to Page 9178, line 19: I appreciate the thoroughness of the authors, but I think this section of text could be made significantly shorter.

This part has been made shorter.

- Page 9180, line 13: Shouldn't 'Fig. 5' actually be labelled as 'Fig 3' since this is the third figure referenced in the text.

This reference to figure 5 in section 3 has been removed.

- Section 4.1 and Table 2: Maybe more useful metrics like the normalised mean bias and error would make it easier for the reader to diagnose the model performance.

One column has been added in Table 2 with normalised mean bias.

- Page 9184, line 28: Why wasn't the model compared with the MIPAS (or ACE) data? Surely the satellite observations would give a further opportunity to evaluate the model.

Satellite data are highly valuable to provide temporal variability of the spatial distribution and inter-annual variability. However, such data also have large uncertainty for oxygenated compounds compared with in situ measurement and sparse sampling. We aim to use such data when they will be widely distributed but it should be the subject of a full paper. Furthermore, observation data sets acquired aboard aircrafts and satellites should all be compiled together to serve as reference for modelling results. We chose to not deal in detail with another data set.

- Caption to Table 2: Should 'bolt police' mean bold face.

This is corrected.

Reference:

Peeters, J., and J.-F. Müller, HO_x radical regeneration in isoprene oxidation via peroxy radical isomerisations. II: Experimental evidence and global impact, Phys. Chem. Chem. Phys., 12(42), 14227-14235, DOI:10.1039/C0CP00811G, 2010.