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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 #2. Referee comments are in bolt italic.

Anonymous Referee #2
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- This paper presents a detailed comparison of the LMDz-INCA CTM simulation of acetone with CARIBIC observations in the upper troposphere. The authors state that the goals of the paper are to describe the variability of acetone, define constraints to improve tropospheric modelling, and investigate how representative the dataset is. The authors do address the first and third goals, finding significant seasonal and geographical variability in the CARIBIC observations, which might be difficult to capture on a monthly basis. However, I find that the most interesting goal, related to actually improving understanding of the acetone budget and processes is not really addressed. In that I agree with the first referee that one is left a little bit unclear as to what was actually learned in terms of the budget of acetone and how could the model simulation be improved.

We thank the referee for the useful advice.
The purpose of the paper is to provide information on acetone variability in UT using two complementary techniques, modelling and observation. The approach is to perform a detailed comparison of both extensive data sets. To prevent confusion, we propose to rewrite the second objective as "proposing benchmarks deduced from the observation data set". Improving model simulation, as by testing different parameterisations, was not the subject of our paper, but we propose a perspective to do so in future work: CARIBIC data acquired over the Atlantic Ocean in summer could be analysed to understand why acetone vmr is highly underestimated in air masses transported to Europe over Atlantic Ocean.

There are numerous language issues in the text, and I highly recommend that the authors have a native english speaker carefully read and edit their manuscript. This would greatly increase the readability of the results.

We have had a colleague check the paper and we have made a range of improvements accordingly.

- Given that most of the previous CTM evaluations relied on the dataset compiled by Emmons et al. (as noted by the authors), it would be useful to repeat the comparison with the LMDz-INCA model and see whether it presents an improved simulation relative to previous studies (including one with the same model). It would also be useful to address how information from this previous dataset (collected mostly over oceanic regions) and the CARIBIC dataset (with

more info over continental regions) actually complement each other and are consistent with each other.

We agree that it would be really interesting. However it is important to understand how large is already the CARIBIC dataset. We can do such recommendation about the need to gather all the data in a common climatology, however, one conclusion of our work is the difficulty, due to the large variability of acetone sampled by CARIBIC, to average data for different year in climatological fields. The comparisons between LMDz-INCA and the Emmons datasets are presented in Folberth et al. 2006. However, our work is focused on the comparison of the CARIBIC data and co-located INCA results for recent year. The comparisons with data in other database (Emmons, or more recent campaigns) is beyond the purpose of this current paper. In a further step it would indeed be interesting and useful to compile all complementary observation data sets: field campaigns compiled by Emmons, providing many vertical profiles in the troposphere over oceans; the CARIBIC data set continuously extending, and sounding mainly the upper troposphere over continents and over the Atlantic Ocean; new satellite products, as from MIPAS or ACE. When a reference acetone pattern would be designed, it would be important to compare with different chemistry models.

Minor comments

1) Could the authors elaborate on the actual differences between the Folberth study using the same model and their own? It seems from the description in the text (section 2.1) that the only changes relate to a slight increase in resolution and in the number of chemical species. It is unclear whether the acetone quantum yield was also updated in Folberth et al. or not. Table 1 shows that the emissions used in the present study and this previous study are actually different (biogenic + biomass burning emissions). A more in depth discussion of the differences would be useful. Why aren't the residence time and global atmospheric burden listed for the Folberth et al. study?

Except Acetone quantum yield, which has been updated since Folberth et al. [2006], the chemical scheme corresponds to the detailed description available in Folberth et al. [2006]. The difference between the two versions mainly concern emissions. However, a more in depth discussion on budget terms is difficult as few values are given by Folberth et al. [2006].

2*) page 9179 line 25 (section 3.2). "the height above the tropopause defined by Sprung and Zahn (2010)" a bit more detail on this would be useful. How are stratospheric measurements identified? Does the observed tropopause correspond well with the model tropopause on a flight by flight basis?*

The sentence is modified to be more informative:
"Sprung and Zahn [2010] discriminated tropospheric data from stratospheric data, and also defined a (mixing-based) height above the thermal tropopause, by translating ozone concentrations measured on flights, using data collected at 12 ozonesonde stations."

No comparison was effectively made with the modelled height of the tropopause. But both the

Sprung and Zahn and the Koppe *et al.* [2009] criteria seem appropriate enough to avoid observation made in the stratosphere. Indeed acetone decreases sharply above the tropopause:
- according Sprung and Zahn classification, 530±290 pptv in the upper troposphere, 350±250 pptv in the stratosphere, 230±150 pptv 0.5 km above the tropopause;
- according to Koppe et al; [2009] in summer from 780 to 1280 pptv in troposphere, 510 pptv at tropopause, 270 pptv in stratosphere; in winter from 450 to 700 pptv in troposphere, 330 pptv at tropopause, 150 pptv in stratosphere.
- from modelling, acetone decreases significantly from 200 hPa upwards (Figure 7), while 95% of measurements were made between 200 and 300 hPa.
Regional annual observed values discussed in the paper are clearly representative of troposphere, varying from 360±120 pptv (winter NAO) to 865±500 pptv (EurMed 2007), suggesting no impact of stratosphere, as well as for modelling results varying from 445±55 to 1150±580 pptv.

Moreover, in case the tropopause is simulated below the flight level, the underestimation would be significant, and not such has been observed (spanning from 20% underestimation to 180% overestimation).

3) Figure 7. This figure shows only the model simulation. It would be useful to show comparisons of observed vertical profiles.

We agree that vertical profiles of acetone vmr acquired over the airports are highly valuable. However vertical profiling over large cities (populated regions) is expected to generate high variability which can not be resolved by the relatively raw spatial resolution of climate models. Then we chose to avoid such variability by screening data made during changing aircraft altitude. Also, for each long distance flight of 10 hours only 2 vertical profiles can be obtained.