Response to reviewer 3

We would like to thank Reviewer #3 for his/her thorough and useful comments. We have included in this response the original text (in italics) and our answers.

General statement: To answer this and the other reviews, we have considerably changed our figures to be more summarizing. This is in turn enables an easier side-by-side comparison of the various simulations, including the comparison of the two wet removal schemes. Regional aerosol optical depth and surface ozone diagnostics and discussions are also added. A better description of how CAM-chem relates to CAM4 and CESM is also included. Finally, a comparison of some meteorological fields is now in the paper.

A great point of interest of this paper is the presentation of 3 different simulations, with 3 different configurations of the model. The difference between on-line and off-line meteorology is clear. But, more details on the differences between the 3 runs, and what we should expect from these differences, are lacking :
• Even if the horizontal resolution is the same, the vertical resolution is quite different between the on-line (26) levels and the 2 off-line simulations (56). It would be very interesting to have the vertical repartition of the levels (PBL, Tropo and Strato). In a more general point of view, the difference between the vertical resolution and its potential impact is never discussed in the paper but should be.

We have included a figure describing the position of the vertical levels in the offline and online configurations. On the other hand, the impact of vertical resolution is very difficult to assess and there is no simple way to do this in this paper. It would require additional online simulations with varying vertical resolutions. While doable, we believe this is beyond the scope of this paper.

• The treatment of the stratosphere in the off-line simulations is not clear : no chemistry, but is there any transport ?

Yes there is transport, but many constituents are overwritten above 50 hPa. Added in section 6.3

And how is the tropopause defined (chemical or dynamic) ?

It is done through the temperature profile. Added.

• The differences between the 2 off-line simulations seem to lie on 2 key points. The first is of course that the forcing fields are not the same. An explanation of the principal differences between GEOS-5 and MERRA would hence be very helpful.

Some discussion is now included in sections 4 and 7.

The 2nd point is the different treatment of wet scavenging. A discussion on the impact of this new scavenging scheme would be interesting, in parallel with a brief comment on the differences on diagnosed precipitation.

We have majorly rewritten our analysis section and included more telling figures, including for the comparison of the new wet deposition scheme (for which an additional simulation was performed). This is done in section 7.2.

Section 3.3 Wet Deposition :
The sentence with Xi and Xiscav (p2205, line 15-16) is not clear.

There was a typo; the text should be clearer now.

The presentation of the Horowitz' scheme lets me understand that there is no below-cloud scavenging. Is it right?

It is actually present, but only for HNO3 and H2O2. Information added.

Section 3.5 PSC :
The treatment of TCly and TBry would deserve more explanation. How does this conservation impact the other chlorined and bromined species?

We have included a better description of TCly and TBry.

Section 4. Offline meteorology :
The time-step of the forcing as well as the horizontal and vertical resolutions of the forcing models would be interesting.

The vertical resolution is shown in Figure 3. The original horizontal resolution 1/5x2/3. This is added.

Section 6.1 Emissions :
Is it possible to have an estimation of the impact of the "simple linear interpolation" on the total emission?

There is no simple quantification since this will depend on the match between the horizontal grids and the overall regional structure of the emissions. It is at a maximum a few percent locally.

Section 7.
General : It is not always obvious to see in the figures presented which simulation better performs. Maybe a numeric summary with simple statistics would be helpful. For instance, the authors could give the mean bias profile for the whole stations in fig 3, or the mean correlation at a specific height for fig 4. This objectiveness would give more weight to the diagnostics.

We have included new figures to demonstrate more clearly the relative performance of the model configurations. In particular, the Taylor diagrams give a quantitative estimate of the annual mean bias and correlation to the seasonal cycle.

Section7.1 Ozone sondes
• Profiles : The question of the position of the tropopause is indeed crucial. Once again, it would be very useful to know how the tropopause (and stratosphere) is treated in the off-line simulations, to better analyse the STE.

See above.

• Seasonal cycle : The better performance of the GEOS5 is not obvious. If you compute the mean bias, then GEOS 5 gives always better results than MERRA, at every altitude. But MERRA gives a better average correlation. So what is the main feature to retain?

This is summarized in the Taylor diagrams. Based on those, GEOS5 gives a better simulation of ozone.

For the low bias in the SH Polar region, are you sure that you are not already in the stratosphere at 250hPa, or at least very close to the strong-gradient zone? That would explain why the cycle is so difficult to reproduce, and be coherent with the fig3 for South Pole.

It is indeed already sampling the lower stratosphere; this is now indicated in the text.

Do the authors have an idea of what happens in the low layers in North America in fall with the MERRA simulation?

We don't know, but it is clear that the seasonal cycle over North America at 900 hPa (see correlation #4 in Taylor diagram) is much worse than any other.

• Long-term change : Once again a numeric evaluation (of the correlation for instance) would help. The Edmonton case should be developed.

We are now providing (Table 8) bias and correlation. We have however removed the discussion of Edmonton as this was not central to our discussion.

• Ozone budget : How do the authors explain this huge difference of ozone net chemistry between online and off-line? Can the difference come from the fact that the chemical limit defined here as under 100ppbv may refer to different levels depending on the model? In addition, the lifetime of ozone for the online simulation is lacking.

We have redone our analysis to cover the exact same period in all simulations. This is listed in the revised Table 8 (now 9). The outlier is mostly MERRA, for which the enhanced STE leads to an increase ozone loss and overall smaller net chemical production. This is now mentioned in the paper.

Section 7.2 Aircrafts :
The "realistic meteorology" is a too drastic expression. The on-line version does not gives so unrealistic fields. A highlight on HNO3 and the difference between the two scavenging schemes would be valuable here.

The text was corrected. And we have now included a specific simulation to discuss the impact of the scavenging.

Section 7.3 Surface CO :
Can the author comment the exceptionally high values for mean surface CO at approx. 0_ and 30_ for the online simulation (related to the high value of the cycle at 0_)? However it is not clear whether the corresponding value for the offline simulation is in the plot.

The value for MERRA is actually higher and was not visible because of the maximum value plotted. We have updated both figures. The equatorial value is Mt Kenya and we might be seeing an effect of topography not captured by the model. Such statement in included in the paper.

Section 7.4 Ozone Total column :
The sentence 'Because the online model: : : only the mean and standard deviation is relevant in that case' is fundamental and true for the whole paper. It deserves to be at the beginning of the analysis.

We have now included such statement.

The figure 9 could be replaced by a numerical value of means and standard deviations.

For completeness we have included this information but have kept the plot.

Section 7.6 Aerosols :
Could the authors emphasize why the MERRA simulation is not presented here? For figure 12, once again, numerical value of the correlation would help.

We have now included MERRA; it was not included because the results are very similar to GEOS5. Numerical values were added as Table 11.

Typos
Pp 2215 line 13 "tropospheric ozone using ozone sonde measurements averaged on representative regions"

Done.

Pp 2218 lines 7 and 10 the figures are no well referenced

Corrected.

Pp 2219 line 18 : expect ! except

Done.

Pp 2219 line 23 : figure S5 ! S3

Corrected (note that the numbering and content of the supplement has changed).

Fig 5 : there seems to be a mistake in the caption of the corresponding figure, MERRA and GEOS-5 are switched

Corrected.

Fig 12 : the caption is wrong

Corrected.