

Response to reviewer 4

We would like to thank Reviewer #4 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 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.

1. The terminology for the model needs to be clarified. In particular the use of "CAMchem" needs to be made more clear. Does "CAM-chem" refer to the chemistry component of CAM4 (which presumably can be turned on or off), or do you refer to the whole model, when chemistry is activated, as CAM-chem. Does the terminology differ between "on-line" and "off-line" versions? I know this is a bit nitpicky of a distinction, but it leads to considerable awkwardness throughout the paper, where some features are described as being done by "CAM4" and others by "CAM-chem". A clear definition of what is meant by "CAM-chem" would clarify these issues tremendously.

We have now included such description in section 2.

2. There are a lot of figures (and particularly figure panels) in Section 7, the model evaluation section. This is typical for evaluation papers of new atmospheric chemistry models, and provides a welcome glimpse into how the model performs with regard to distributions of a number of different atmospheric species. However, the large number of figure panels makes it hard for a reader to absorb all of the information presented and distill an understanding of how well the model is performing. The text needs to do a better job of synthesizing and summarizing model performance, both in an absolute sense and for the relative differences across model versions.

We have considerably revised our figures to bring out quantitative information. In particular, we have now make use of Taylor diagrams. And section 8 was expanded to reflect the additional information.

Specific Comments

p.2200, Abstract – Add one sentence to the abstract describing (qualitatively) the degree of similarity/difference in results across the three model configurations presented.

We have added such a statement.

p.2200, l.24 – As mentioned in General Comment above, define here what is meant by "CAM-chem".

Done in section 2.

p.2201, l.18 - "(iv)" -> "(4)"

Done

p.2202, l.9 - Clarify the distinction between "CCSM4" (mentioned here) and "CESM" (used earlier).

Also see section 2.

pp.2202-2203 - Are the aerosols and ozone concentrations simulated from CAM-chem radiatively active in CAM4? Is this true in both online and offline versions? If the aerosols are radiatively active, some description of how the aerosol optics are treated needs to be included in the model description. Does this model include aerosol-cloud interactions?

All these are now answered in Section 2.

p.2205, l.9 - Remove "gas-phase", since you describe below both gas-phase and aerosol scavenging.

It is actually only used for gas-phase. Aerosol removal is described at the end of the section.

p.2205, l.15-16 - Is this a typo? Do you mean "Xi is the species mass (in kg), and Xiscav is the amount of the species scavenged in time step delta-t"?

This was a typo. It is corrected.

p.2205, l.24 - Clarify that each of the two separate parameterizations includes both in-cloud and below-cloud scavenging.

Done.

p.2205, l.28 - Delete "the Neu and Prather scheme". Change "accounts" to "accounting".

Done.

p.2207, l.23-26 - It is unclear from the description provided here how the TCly and TBry tracers are implemented. Does the TCly tracer include both organic Cl and (inorganic) Cl? How is the conservation enforced. Are all Cl-containing species rescaled uniformly each timestep to match TCly?

The summed mass is forced to be the same as the TCly/TBry mass; scaling is applied to all species. This is now described in the text.

p.2208, l.19 - Does the "calculation of photolysis rates for wavelengths shorter than

200 nm" still use the lookup table, or is this calculation done online?

It uses the online calculation. This is now clarified.

pp.2209-2211 – Are any of the CAM4 physics parameterizations modified to account for the different vertical resolution when running with offline fields? It would be helpful to add to Table 6 (or the text) some indication of where the extra levels are located (e.g., how many levels below 2km? how many levels in the stratosphere?).

This is discussed in section 4, with figure 3 added.

Are there any systematic changes (e.g., in clouds, PBL, precipitation) resulting from the resolution change, distinct from the changes from the use of offline meteorology? (I assume it would be hard to determine this without running the model online with 56 levels. Has this been done?)

No it hasn't, and we haven't found any major problem in the representation of the physical climate (see section 7).

pp.2209-2211 – What is the time frequency at which the offline GEOS5 and MERRA data is archived? (every 3 hours? every 6 hours?) Are input fields interpolated to each model timestep, or are values just read in once every 3-6 hours and the model allowed to advance the fields for 3-6 hours?

6 hours and linear interpolation is included. This is now described in section 4.

p.2211, l.18-20 – Does the choice of explicit/implicit species vary in your model configurations when stratospheric chemistry is introduced? (i.e., do some species have long lifetimes in the troposphere, but sufficiently short lifetimes in the stratosphere?)

It can be, but it is not the case. Because the implicit solver is quite efficient, it is recommended to use this one unless the chemical species has a long lifetime everywhere. Text added.

p.2213, l.6 – Change to "Neu and Prather" to be consistent with earlier usage.

Done.

p.2213, l.24-25 – Clarify that this sentence refers only to anthropogenic emissions. (You previously stated that biomass burning emissions vary interannually.)

Done.

p.2214, l.16-25 – Over what altitude/pressure range are these concentration boundary conditions imposed? All the way down to the local tropopause, or just in the upper stratosphere?

The text was updated to provide this information.

*p.2215, l.13 – This sentence is garbled, rewrite as "... *using* ozone sonde measurements averaged *over*"*

Sorry about that. Corrected.

p.2215, l.17-18 – Is the tropopause position better matched in the online or offline simulations? Why? Be explicit about what you mean here. You are (presumably) referring to the chemical (dynamical) tropopause location, not just the thermal (or lapse rate) tropopause. Are you just seeing the effects of excessive exchange of ozone across the thermal tropopause?

It is indeed the chemical tropopause (and have updated the text). Based on the Taylor diagrams, there is no clear distinction between the various models for 250 hPa. However, the online is clearly deficient in the 500 hPa, most likely due to misrepresentation of transport. The text was changed to reflect that.

p.2215, l.21-26 – Include some quantification in the text of the ozone biases versus ozonesonde profiles (Fig.3) in the various model configurations.

This is done by the Taylor diagrams.

p.2216, l.1-2 – MERRA fields seem to provide the worst match to ozonesonde observations in Figure 4. Can you provide any information on why the differences among model configurations are so large?

MERRA and GEOS5 have different assimilation schemes and assimilated data (information added to the text). We have included a discussion of various climate variables in section 7. But we don't have a clear answer on why they are different.

p.2216, l.6-11 – Are the biases slightly better in the online version with stratospheric chemistry? In particular, does the inclusion of stratospheric chemistry affect the magnitude of the UT/LS ozone biases? (Presumably this answer depends on how far down the concentration upper boundary condition for ozone is imposed when stratospheric chemistry is not used.)

See discussion of Taylor diagrams and Table 8.

p.2216, l.7 – Change "especially true" to "with the largest biases".

Done.

p.2217, l.6 – STE in offline simulations is given here as 520 Tg/yr, but in Table 8 as 497.5 Tg/yr.

Sorry for the mismatch. This is now corrected; numbers were recomputed over the same period for all simulations and for ozone < 100 ppbv; they are therefore slightly different.

p.2217, l.9 – If you calculate ozone lifetime (as reported here), I assume that you have diagnostics for ozone production and loss (separately). If so, it would be much more informative to include those in Table 8, rather than just including the net chemical tendency.

Table 8 (now 9) was expanded accordingly.

p.2217, l.13-14 – Discuss any systematic differences between wet deposition schemes. It looks like H₂O₂ and HNO₃ are generally lower with the Neu and Prather scheme (Figure S3).

We have added a figure and a simulation to more explicitly discuss the wet removal schemes.

p.2217, l.27 – Note that the Lin et al. (2008) paper discusses results from MOZART-2, not CAM-chem.

Added.

p.2217, l.26-28 – The discussion of surface ozone biases should be split off into a separate sub-section. The surface ozone bias would be better diagnosed by comparisons with surface observations rather than aircraft profiles. Such a comparison could be included, at least as a supplemental figure.

We have now included a specific discussion (in expanded section 7.1) on European and US surface ozone stations.

p.2218, l.4-6 – This discussion of possible biases in the lightning NO_x source should at least reference some of the recent papers on this topic (e.g., Hudman et al., 2007).

Reference added to section 3.4.

p.2218, l.15 – Change "distribution" to "concentration".

Done.

p.2218, l.11-16 – Note again here that the offline CAM-chem simulations being discussed here use the same wet deposition scheme as in MOZART-2 and MOZART-4.

For simplicity, we have removed this comparison with the MOZART-4 results.

p.2218, l.19 – Define how the tropospheric methane lifetime is calculated, presumably

the usual definition of (total atmospheric burden)/(tropospheric OH loss).

That is correct; this is added.

What tropopause is used for this calculation – the same 100 ppb O3 tropopause as for the O3 budget?

Yes; information added to updated Table 8 (now 9).

Reconcile the same CH4 lifetime with earlier statement regarding larger OH concentrations in CAM-chem versus MOZART. Are the OH concentrations different in regions that don't strongly impact CH4 lifetime?

Done.

p.2218, l.22-27 – Provide more details about the simulated surface O3 concentrations, and differences between MOZART-4 and CAM-chem. (See earlier comment, p.2217, l.26-28, about moving surface ozone discussion into a separate section.)

We have considerably expanded this analysis of surface ozone but could not include MOZART-4 since hourly data was not discussed in E2010.

p.2219, l.20-28 – The discussion of OH concentrations (and CO/CH4 lifetimes) is currently split up across different sections. It would be preferable to consolidate this discussion in a separate sub-section on OH.

There is no real good way to do this as OH is relevant to many species. To avoid duplication of discussion, we have kept the structure as is, focusing on species for which we have extensive measurements instead of focusing on OH.

p.2219, l.25-28 – Discuss the OH differences across model versions. The global mean CO and CH4 lifetimes are similar, but figure S5 shows significant regional differences in OH.

This is discussed in more detail, especially in the light of the relative humidity and isoprene differences (see section 7).

p.2220, l.8 – Change "extension" to "extent".

Done.

pp.2220-2221 – Given the brevity of the discussion of stratospheric results, it seems excessive to include 3 figures (10a, 10b, 11) on stratospheric evaluation in the main paper. Could some be moved to supplementary material.

We have removed this material.

p.2221, l.20-22 – Mention that the IMPROVE dataset is over the US only.

Done.

pp.2221-2222 – The discussion and evaluation of aerosol results should be expanded, given their potentially large climate impact. (See earlier question, pp.2202-2203, about whether simulated aerosols have a radiative impact in this model.) Some evaluation of free tropospheric aerosols (or column AOD) needs to be included. Do the INTEX campaign datasets include any aerosol measurements that could be used for evaluation? Ideally, some observations in regions outside of North America would also be used for aerosol observation. Global datasets (e.g., AOD from satellite retrievals or AERONET) would be useful.

We have now included a discussion of MODIS and MISR AOD.

p.2222, l.6 – It looks like only the one highest point is underestimated. Others appear to be overestimated (cf. Figure 13).

We redrew those figures to include all points.

p.2222, l.8-18 – Are there any systematic differences across model configurations related to the use of different wet deposition schemes?

This is now discussed more specifically with the addition of a separate MERRA simulation discussed in section 7.2.

p.2222, l.10-13 – Do you have any possible explanations for these biases in the sulfate PDF? Do SO₂ emissions in the model all go into the surface layer, or are they emitted into an elevated layer?

They are all in the surface layer and no SO₄ is emitted, only SO₂. The high resolution in the MERRA/GEOS5 might be partly responsible for this bias. Text was added.

p.2223, l.4-7 – Can you rule out transport problems as contributing to the CO underestimate?

Not completely, but this is a fairly prevalent bias that is present in other models and in both online and SD configurations.

Quantify the magnitude of the bias versus observations.

This is already indicated in Figure 7a. We have included a Table 9.

p.2223, l.12-14 – Is the surface ozone bias present only over the NE U.S. or also the SE U.S.? (See also earlier comment, p.2217, l.26-28, about a separate section on surface ozone.)

Both.

pp.2222-2223 – This section needs to include a summary of the major differences among model versions. Are there notable systematic differences between online versus offline versions, stratospheric chemistry versus no stratospheric chemistry, varying vertical resolutions, different wet deposition schemes, etc.? Which simulated quantities are most sensitive? You mentioned the chemical tropopause position and STE as being sensitive to model configuration. Concentrations of H₂O₂ and HNO₃ appeared to be sensitive to the wet deposition scheme. This paper contains a wealth of information, but the reader needs some summary in this conclusion section to help make sense of all of the data presented throughout the paper.

We have expanded the discussion section to include specific statements about model configuration performance.

Table 8 – Are the ozone budgets identical (to 4 significant figures) in both offline versions of the model?

Table 8 was redone (now Table 9).

Figure 2 – Define BrO_y, ClO_y. Presumably these are intended to refer to total organic Br, Cl. I don't think that this figure is important enough to justify its inclusion in the main paper (or maybe even as supplementary material).

This figure is removed in the revised version.

Figures 3-6 (and 10-11) – These figures include a lot of small panels. This is useful for conveying a lot of information concisely, but it is hard for a reader to interpret and distill all of this information. The text needs to do a better job of summarizing the bias patterns and the differences across model configurations.

We have moved all the panel figures to the supplementary material and replaced them with summary figures.

Figure 12 – Include the model-obs correlation coefficients and regression slopes in this figure.

Done.

Figure 13 – You should also include some evaluation of aerosols globally (e.g., AOD versus MODIS or AERONET).

Done.

Supplementary material, p.1 – Change "isoprene" to "biogenic" since Fig. S1 shows other species as well. Figure S1 – Give global totals.

We have included the totals in Table 9 and removed the Figure.

Figure S3 – Give the methane lifetime for each model version as part of this figure.

See new Table 8 and associated text.

Figure S4a – This figure includes results from a new simulation not previously mentioned anywhere in the paper. This simulation should be described briefly in the text. Also, add "with MOZART wet removal scheme" to description of red curve in figure caption.

This figure is now removed and replaced by the discussion in the main text (section 7.2). The additional simulation is listed in updated Table 3.

