



## ***Interactive comment on “CAM-chem: description and evaluation of interactive atmospheric chemistry in CESM” by J.-F. Lamarque et al.***

### **Anonymous Referee #4**

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Review of "CAM-chem: description and evaluation of interactive atmospheric chemistry in CESM" by J.-F. Lamarque et al.

This manuscript describes and evaluates the atmospheric chemistry and aerosol simulation by CAM-chem in the CAM4 global atmospheric GCM. This is an important paper documenting the chemistry component of this widely used model that serves as the atmospheric component of the CESM coupled climate model. As such, I expect this paper to attract considerable interest from the atmospheric chemistry and climate modeling communities.

The paper provides results from three separate configurations of CAM-chem: a free-running "on-line" version of the GCM driven by prescribed SST and sea ice; and two "off-line" versions driven by meteorological fields prescribed from GEOS-5 analyses or

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reanalyses (MERRA).

This paper is generally well-written and attempts to cover a lot of material. There are a few shortcomings to the paper as currently written, as detailed in my comments below. After addressing these comments, I believe that this paper would be suitable for publication in GMD and would be a valuable contribution to the literature.

#### General Comments

1. The terminology for the model needs to be clarified. In particular the use of "CAM-chem" 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.

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.

#### 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.

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

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"CAM-chem".

p.2201, l.18 – "(iv)" -> "(4)"

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

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?

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

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  $\Delta t$ "?

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

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

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) Cly? How is the conservation enforced. Are all Cl-containing species rescaled uniformly each timestep to match TCly?

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?

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

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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?). 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?)

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?

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?)

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

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

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?

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

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?

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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.

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?

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.)

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

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

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.

p.2217, l.13-14 – Discuss any systematic differences between wet deposition schemes. It looks like H<sub>2</sub>O<sub>2</sub> and HNO<sub>3</sub> are generally lower with the Neu and Prather scheme (Figure S3).

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

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.

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p.2218, l.4-6 – This discussion of possible biases in the lightning NO<sub>x</sub> source should at least reference some of the recent papers on this topic (e.g., Hudman et al., 2007).

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

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.

p.2218, l.19 – Define how the tropospheric methane lifetime is calculated, presumably the usual definition of (total atmospheric burden)/(tropospheric OH loss). What tropopause is used for this calculation – the same 100 ppb O<sub>3</sub> tropopause as for the O<sub>3</sub> budget? Reconcile the same CH<sub>4</sub> 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 CH<sub>4</sub> lifetime?

p.2218, l.22-27 – Provide more details about the simulated surface O<sub>3</sub> 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.)

p.2219, l.20-28 – The discussion of OH concentrations (and CO/CH<sub>4</sub> lifetimes) is currently split up across different sections. It would be preferable to consolidate this discussion in a separate sub-section on OH.

p.2219, l.25-28 – Discuss the OH differences across model versions. The global mean CO and CH<sub>4</sub> lifetimes are similar, but figure S5 shows significant regional differences in OH.

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

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.

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

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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.

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

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

p.2222, l.10-13 – Do you have any possible explanations for these biases in the sulfate PDF? Do SO<sub>2</sub> emissions in the model all go into the surface layer, or are they emitted into an elevated layer?

p.2223, l.4-7 – Can you rule out transport problems as contributing to the CO underestimate? Quantify the magnitude of the bias versus observations.

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.)

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 H<sub>2</sub>O<sub>2</sub> and HNO<sub>3</sub> appeared to

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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.

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

Figure 2 – Define BrO<sub>y</sub>, ClO<sub>y</sub>. 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).

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.

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

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

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

Figure S1 – Give global totals.

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

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.

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