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Interactive comment on “Improvement and further development in CESM/CAM5: gas-phase chemistry and inorganic aerosol treatments” by J. He and Y. Zhang

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[Reply to Reviewer 1's Comments](#)

This paper discusses the outcome of an improvement by the authors to the aerosol model (the Modal Aerosol Model 7 or MAM7) of the Community Earth System Model (CESM). The authors have successfully incorporated several sophisticated components to replace the more simplified counterparts in the current MAM7/CESM. These include a gas chemistry package, an aerosol thermodynamics module, and a new ion-based aerosol nucleation module. Besides, the authors have also performed a sensi-

[Full Screen / Esc](#)

[Printer-friendly Version](#)

[Interactive Discussion](#)

[Discussion Paper](#)



tivity simulation using altered emissions. The effort is to assess the performance of the CESM model after the introduction of these new aerosol and chemistry components.

I admire this effort that covers many detailed aspects of model prediction of aerosols and tropospheric chemistry. On the other hand, however, the paper is mostly about model development rather than science findings. Noticeably, all the model components introduced into the CESM have been developed previously and mostly used in other frameworks, thus what the paper describes is the outcome from coupling these components with a different model framework. This would still be invaluable. The work has provided an arguably better platform than the current CESM in modeling aerosol and chemistry interaction with climate. The evaluation of such a model improvement needs to be recorded in literature. A relevant question here is whether the authors would want to consider journals that specifically solicit works dealing with model techniques and evaluations such as Geoscientific Model Development. If not, I would suggest the authors reconsider the scope of the paper and focus more on science features in discussion. My (most general) comments here are for the authors to consider when making their revision, either for ACP or resubmitting to another journal.

Reply:

We thank the reviewer for positive comments. It is true that the model components such as ISORROPIA II and YU10 ion-mediated nucleation parameterization were developed previously and have been used in some other models. However, incorporating them into CESM/CAM5 is not a trivial effort, as it involves a substantial amount of recoding of some of those model components, developing new interface between them and the host model, and testing and evaluating against a large set of observational data. We agree with the reviewer that such work is invaluable and should be recorded in literature.

We also thank the reviewer for a suggestion to submit our paper to Geoscientific Model Development (GMD). However, prior to our submission to ACPD, we have considered

[Full Screen / Esc](#)

[Printer-friendly Version](#)

[Interactive Discussion](#)

[Discussion Paper](#)



several possible journals including GMD and believe that ACP is the best journal for our paper, as our work fits well into the scope of work published by ACP.

The authors used the fully coupled CESM configuration to run their simulations. This is a configuration includes a full ocean GCM coupled with the atmospheric model along with other components in a transient mode. Surprisingly, with this configuration, all the simulations were integrated for only one year. In the discussion of modeled results, the authors spent quite an effort on changes of many climate variables from surface air temperature to wind speed. Arguably, this is not adequate because of the short integration time and very long ocean response. Note that the model was cold started and forced by an introduced new and different forcings than the standard 1850-2000 run (it is understood that the last output of this run was used as initial condition for the simulations). In my opinion, the emphasis of the effort should be, as stated by the authors, to assess the aerosol and chemistry predictions introduced by the new model components. Such an effort, however, would likely suffer from the first year response of modeled transient climate after a cold start, which involves interactions between aerosol, air chemistry and meteorology/climate as well. It would be much better if the authors use a configuration using prescribed SST, run simulations for at least three years, and then remove all the irrelevant discussions of climate variables (the changes are hardly to identify anyway from Table 3) from the paper but those of chemistry and aerosol features.

Reply:

While the initial conditions for ice model and ocean model are from CESM default settings, the initial conditions for land model are the output from NCAR CESM B_1850-2000_CN run. The initial conditions for CAM5 are derived from a 10-yr (1990-2000) CAM5 standalone simulation with the MOZART chemistry provided by NCAR. A 1-year (January 1-December 31, 2000) CESM/CAM5 simulation using NCAR's CESM B_1850-2000_CAM5_CN component set is performed as spinup to provide the initial conditions for meteorological variables and chemical species that are treated in

C13031

ACPD

13, C13029–C13040,
2014

[Interactive
Comment](#)

[Full Screen / Esc](#)

[Printer-friendly Version](#)

[Interactive Discussion](#)

[Discussion Paper](#)



both MOZART and CB05_GE. An additional 3-month (October 1-December 31, 2000) CESM/CAM5 simulation based on a 10-month (January-October, 2000) CESM/CAM5 output using initial conditions from NCAR's CESM B_1850-2000_CAM5_CN is performed as spinup to provide initial conditions for chemical species that are treated in CB05_GE but not in MOZART. So, the model was not cold started for atmospheric and land models. We have clarified this in the revised paper.

We agree with the reviewer that the 1-yr simulation time is a bit too short for global model simulations and the results may be more robust with longer simulations and prescribed SST. However, we believe that the changes in most radiation, aerosol, and cloud-related variables are caused mainly by changes in the model representations. We have performed the student's t-test, which verified this. Therefore, we kept Table 3, but we provided info from our t-test in Table A1 in the supplementary material.

To address the reviewer's comment, we have performed additional simulations for 2001-2005 with prescribed SST and analyzed the results in the revised paper. To compare predictions with prescribed SST and in a fully-coupled mode, we also performed a 5-yr simulation for 2001-2005 using fully-coupled CESM. These new results are shown in Tables 6-7, Figures 9-10, and a new discussion section (i.e., Section 6).

The improvements in terms of aerosol and chemistry model predictions are mostly limited in comparison to observations (see Table 4), while the conclusion drawn in abstract and Conclusion section about this is much too optimistic to me. Realizing the very high demanding in computation of this new model, the best purpose this effort could serve is to provide certain information for the improvement of simplified schemes, because practically it would come to the usage of the latter types to conduct aerosol chemistry-climate interaction simulations. Therefore, the emphasis perhaps should be on the relative differences between various model configurations.

Reply:

We agree that it is interesting to compare the relative differences between various

model configurations, our submitted paper actually includes extensive discussions on such differences (e.g., most of the original Section 5 describes such differences along with all original Figures 1, and 3-9). The evaluation with observations would attest if the improved model representations improve the model performance, and the performance statistics shown in the original Tables 3 and 4 also illustrated the differences in model predictions using different model configurations. Such differences continued to be the main focus of our discussions in the revised paper, as shown in the revised Section 5 and newly added Section 6, Figures 1, and 3-10, and Tables 3, 4, 6, and 7.

Given rapid advancement of computer powers from Teraflops to Petaflops, we believe that in near future, long-term aerosol chemistry-climate interaction simulations can be performed using CESM with more comprehensive aerosol and chemistry treatments. Therefore, the evaluation presented in this work would provide a benchmark to assess the capability of the improved model vs the original model with the simplified schemes and/or to provide rationale for any further improvement in the future.

There is an argument that the emission uncertainty could be the reason for much of the remaining problem of the new model. This is also too simplistic to me. By introducing those sophisticated model components, the uncertainty is multi-dimensional (a good example is the scale inconsistency of fast chemistry in the current model). In addition, the sensitivity simulation using alternative emissions were not clearly explained in the paper.

Reply:

We agree with the reviewer that the uncertainty is multi-dimensional when introducing these sophisticated model components. We did not state or imply in the manuscript that “the emission uncertainty could be the reason for much of the remaining problem of the new model.” What we mentioned was “some large biases are caused by inaccuracies in the emissions of CO, SO₂, BC, OC, and NH₃”. As a matter of fact, we indeed indicated that “Additional uncertainties exist in the model treatments” in the conclusion

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

section (last paragraph) in our submitted paper, in addition to the uncertainty in the anthropogenic emissions. We provided several examples in that section, e.g., uncertainty in the dust emission treatment, nucleation parameterizations, SOA module, cloud microphysics schemes and aerosol-cloud interaction parameterizations. The uncertainty may also stem from other model settings such as the use of a coarse grid resolution and a large time step for solving chemical ODEs. Emission uncertainty could be one of the reasons for model bias, which can be estimated through the sensitivity simulation using alternative emission inputs.

To address the reviewer's comment, we have added a few more factors that may contribute to model uncertainties such as the use of a coarse grid resolution and a large model time step for solving the chemical system in the last paragraph of the conclusion section.

The current discussions appear to have too many leads, while detailed connections among various chemical or physical processes were not clearly analyzed. For example, simulations of tropospheric chemical features seem being improved much significantly over Europe than other places, the reasons behind this, however, has not been explained or stated clearly. More quantitative comparisons between precursors and products in different continents (could be selective) would help (note that the information in Table 4 is incomplete, see specific comment).

Reply:

Given the comprehensive evaluation that we performed, it is not possible to do quantitative comparisons between precursors and products in different continents. However, we indeed selected representative continents for such discussions. For example, in Section 5.5 we discussed the main reason for the large NMB of O₃ predictions over Europe in MAM_NEW_A and showed additional seasonal performance statistics over Europe in Table 5.

The chemical species included in the observational networks in East Asia are not as

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

many as in Europe and CONUS. We have included major chemical species whose observations are available from East Asia in Tables 4 and 6.

To address the reviewer's comment, we included additional evaluations for NO₂ and O₃ over East Asia using limited observations in those tables.

Several Specific Comments.

1. How many variables are prognostic and included in tracer advection? This would be useful for the reader to estimate the workload of the new model.

Reply:

There are a total of 139 prognostic species included in tracer advection. This information has been added in the revised paper.

2. How were the photolysis rates calculated, using any J-table (note, not the nucleation rate)?

Reply:

The photolytic rates are calculated based on Lamarque et al. (2012) which used a combination of a lookup table and online calculation. Photolytic rates for wavelengths larger than 200 nm are calculated using a lookup table, which is based on the Stratosphere, Troposphere, Ultraviolet (STUV) radiative transfer model. At wavelengths less than 200 nm, the wavelength-dependent cross section and quantum yields for each species are specified and the transmission function is calculated explicitly for each wavelength interval.

The Lamarque et al. (2012) reference has been added in the revised paper.

3. P. 27720, L17-19, “: : mass accommodation coefficient: : can be measured: :”, the authors might want to indicate the high uncertainty in this type of measurements, as being demonstrated later in the paper by a difference in orders of magnitude in the adopted values of sulfuric acid and others.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Reply:

The reviewer is correct that such measurements are highly uncertain. This uncertainty has been indicated in the revised paper.

4. P.27728, it may help if the authors could further explain the derivation of the initial condition for the model, was the ending result of B1850-2000 run used?

Reply:

Please see our reply to the general comment on this.

5. Section 3.3, although the evaluation procedure might have been explained in detail in the cited publication, it would still be useful if the authors could explain briefly about this procedure, for instance, whether the data of 2001 or multi-year average data were used in the comparison, etc..

Reply:

All observational data used for evaluating 2001 simulations are based on 2001 only except for particle formation rates that are based on different years during the period of 1998-2002 compiled from Kulmala et al. (2004) and Yu et al. (2008). All observational data used for evaluating 2001-2005 simulations are based on 2001-2005.

This information has been added in the revised paper.

6. P.27732, Section 4, how did the modeled aerosol results in the MAM_SIM differ from those in Liu et al. (2012)?

Reply:

The results from MAM_SIM in Section 4 are not comparable to those in Liu et al. (2012) for several reasons. First the datasets used to generate initial conditions are different in our work and Liu et al. (2012). We use the B_1850-2000_CAM5_CN configuration for MAM_SIM, whereas Liu et al. (2012) used CAM standalone configuration. Second,

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



our MAM_SIM was performed only for 1-yr period, i.e., 2001 whereas the simulation results in Liu et al. (2012) were based on 5-year average. Finally, the emissions used in our simulations are different from those in Liu et al. (2012). Those differences in model configurations and inputs would result in different aerosol predictions.

To address the reviewer's comment, we compared the global burden of major gaseous and aerosol species from MAM_SIM_5Y with those from Liu et al. (2012) since similar configurations and time period are used for both simulations, although they used different emission inventories. This comparison has been added in the revised paper in Section 6 and Table 8.

7. Many of the gas chemistry comparisons were done by comparing the new model results with those of MAM_SIM, this is not informative in my opinion. Many of the chemical fields in MAM7 are prescribed using climatological data derived from MOZART model or alike based on my understanding, and this is done for reason. The authors should compare the chemistry features to the “mother” model of chemical fields used in MAM7, such information would be useful for the aerosol modelers to decide whether they should use alternative climatology, and if not, which aspect in the current climatology need to be improved.

Reply:

We believe that comparisons of results from the improved model with those of MAM_SIM are useful, as they show differences in model predictions due to different gas-phase chemical mechanisms.

We agree that comparison of results from the improved model with those of the “mother” model of chemical fields used in MAM7 would also be useful for the reason stated by reviewer. We have therefore compared prescribed OH, HO₂, NO₃, and O₃ in MAM_SIM with their prognostic predictions from the improved model and added a new Figure (Figure 1b) for this comparison in the revised paper.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

8. P.27733, the last paragraph and other places, is SO₄= here in aerosol or aqueous phase?

Reply:

SO₄²⁻ is in the particulate phase. This has been indicated clearly when we define it for the first time in Section 1 in the following statement:

Inorganic aerosols comprise 25-50% of fine aerosol mass (Heintzenberg, 1989), which mainly includes sulfate (SO₄²⁻), ammonium (NH₄⁺), nitrate (NO₃⁻), chloride (Cl⁻), and sodium (Na⁺).

9. P.27735, L20, “The aforementioned changes : :”, changes due to what?

Reply:

The changes are due to new gas-phase chemistry implemented in the model and the feedbacks to meteorology and radiation through the climate system. This has been indicated in the revised paper.

10. P.27746, L25, “CCN: : : cm⁻³(-2)”, is this referred to the column loading?

Reply:

Yes, this is referred to the column loading. To clarify this, we have changed “CCN at a supersaturation of 0.5% by . . .” to “Column CCN at a supersaturation of 0.5% by . . .”

11. P.27747, last two lines and following, “Large biases : : : due to uncertainties in model input (e.g., meteorology and emissions): : :”, this deserves a thorough analysis, or at least citations to support this statement.

Reply:

To address the reviewer’s comment, we have added more discussions on the emission impacts in Section 5.6 in the revised paper.

12. P.27749, 5.6; it is not quite clear that what emission inventory the authors had

used in this simulation, and what the differences are between the alternative emission inventory and the default one. If the authors selectively adjusted emissions for certain species, what are these species then and how was the adjustment done and based on which works?

Reply:

As indicated in lines 23-29, P 27728 and lines 1-2, P 27729 of our ACPD paper, the default emission inventory used in all simulations except for MAM_NEW/EMIS is the 2001 emission file of Zhang et al. (2012). Table 2 of Zhang et al. (2012) showed the sources of this 2001 detailed emission inventory. Those adjusted emissions used in MAM_NEW/EMIS are adjusted based on the comparison with the emission inventories from the Representative Concentration Pathways (RCPs), the MOZART version 4 (MOZART-4), the Reanalysis of the TROpospheric chemical composition (RETRO), the Global Fire Emissions Database (GFED) version 2, and preliminary evaluation of CESM/CAM5.1 with modified and new gas and aerosol treatments using available observations.

To address the reviewer's comment, we have referred the readers to Table 2 of Zhang et al. (2012) for details in this 2001 emission inventory (This Table 2 is provided below for the reviewer's convenience).

13. Table 4. Noticeably, the listed regions for each variable are not always the same, for example, East Asia numbers were only listed for CO and SO₂, especially not for ozone and many other gaseous and aerosol species. The authors might want to list the results from a same collection of regions.

Reply:

The chemical observations over East Asia are very limited, and they only include surface concentrations of CO, SO₂, NO₂, O₃, and PM₁₀.

To avoid confusion, we have indicated this in the revised paper.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Figures. The color scale in several figures was not always selected to show the necessary details, e.g., Fig. 7 and 8.

Reply:

We have modified the color scale used in Figures 7 and 9 in the revised paper. Note that the original Figure 8 from 1-yr (2001) simulation was replaced by Figure 9 from the 5-yr (2001-2005) simulation, in which appropriate scales were used for all plots.

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ACPD

13, C13029–C13040,
2014

[Interactive
Comment](#)

[Full Screen / Esc](#)

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[Discussion Paper](#)

C13040

