Prof. Tost,

Thank you very much for your efforts editing this article. We appreciate you accommodating our need for more time to complete the updated manuscript and response to reviewers. Please send my regards to Natascha Töpfer and the rest of the ACP support staff for their timely responses to our inquiries.

In your Editor Decision, you asked us to comment on a reviewer question about the length of our simulation and impact from meteorology in a coupled simulation. We hope that you will find our response to similar questions in the Response to Reviewers (following pages) satisfactory.

Thanks,

Brett Gantt et al.

Response to comments from Reviewer 1

Brett Gantt et al.

Reviewer 1: *This manuscript compares one-year simulations by the Community Earth System Model using its standard aerosol activation with simulations using several more advanced schemes. While several aspects of the clouds are evidently simulated more realistically with the more advanced scheme, the manuscript overextends itself by examining impacts on climate, which is inappropriate given the brevity of the simulation. I recommend limiting the analysis to the response of the cloud properties, which is interesting enough. This will require only minor revision. Other comments will also require special attention, but rerunning is not required.*

Reply:

We thank the reviewer for positive comments. Please see our point-by-point reply below.

1. Page 32292, Lines 22-23. Since SWCF is negative, an increase would make the value smaller in magnitude. You might say it is 13% more negative. Also, it's a 4% decrease in net surface downward solar.

Reply:

Since SWCF is a negative variable, more negative actually means an increase in SWCF, rather than decrease. For example, a SWCF of -47 W/m2 is greater than a SWCF of -41.6 W/m2 by 13%, although its numeric value is smaller. To avoid the confusion, we changed "an increase in shortwave cloud forcing of 13 %" to "an increase (more negative) in shortwave cloud forcing of 13%"

2. Page 32292, Line 29. A 0.9 C cooling is not slight. Why not focus on impact of activation changes on aerosol radiative forcing, with ocean surface temperature fixed?

Reply:

Although having a fixed ocean surface temperature would help to isolate the radiative feedbacks from aerosols, we chose to run the coupled model to accurately reflect the impact of aerosol activation on the Earth System. While a one-year simulation does not allow us to make conclusions about the impact of aerosol activation on climate, it does allow us to run several sensitivity studies to estimate the sign and potential magnitude of these impacts on climate forcing. Admittedly, simulations on the order of decades or longer would likely be required to allow all components of CESM to come closer to equilibrium even without changing anthropogenic forcing. Due to the limitations of a one-year simulation, we have remove discussion of precipitation and temperature in the updated manuscript and adjusted the tone to reflect the reality that these results are estimates of the potential magnitude to aerosol radiative forcing and that longer simulations are required for more definitive results.

3. Page 32293, line 10. Should use the AR5 nomenclature here.

Reply:

Corrected in the updated manuscript.

4. Page 32294, line 1. I would say the ARG scheme uses a semi-empirical treatment of supersaturation. It's not based on regressions, but coefficients on physically-based terms are adjusted to achieve agreement with numerical simulations.

Reply:

Corrected in the updated manuscript.

5. Page 32294, line 10. Insert "multiple" before "lognormal".

Reply:

Corrected in the updated manuscript.

6. Page 32294, lines 20-21. Replace "which" with "that".

Reply:

Corrected in the updated manuscript.

7. Page 32295, line 18. New paragraph here.

Reply:

Corrected in the updated manuscript.

8. Page 32296. Line 28. Instead of Neale et al., cite Liu et al. (2011a): Liu, X., R. C. Easter, S. J. Ghan, R. Zaveri, P. Rasch, J.-F. Lamarque, A. Gettelman, H. Morrison, F. Vitt, A. Conley, S. Park, R. Neale, C. Hannay, A. Ekman, P. Hess, N. Mahowald, W. Collins, M. Iacono, C. Bretherton, M. Flanner, D. Mitchell, 2012: Toward a minimal representation of aerosols in climate models: Description and evaluation in the Community Atmosphere Model CAM5. Geosci. Model Dev., 5, 709–739, doi:10.5194/gmd-5-709-2012.

Reply:

Corrected in the updated manuscript, except that the final revised paper is given as Liu et al., (2012).

9. Page 32298, lines 4-5. Why do you use the entrainment rate from deep convection to treat entrainment effects on activation? CAM5 only treats activation in stratiform clouds. Using entrainment from the deep convection scheme is inappropriate for stratiform clouds. If you want to treat entrainment effects, treat activation in shallow and deep convective clouds.

Reply:

We agree with the reviewer that entrainment from deep convection is not the appropriate parameter when examining activation in stratiform clouds. Because the inclusion of entrainment in shallow and deep convective clouds requires closer linkage of the activation and convection model processes than found in our current implementation, we have removed entrainment impacts from the text and figures.

10. Page 32298, line 22. A one year simulation seems very short for estimating effects on SWCF. How do you know it is long enough? What are the initial conditions? Why did you choose a coupled simulation?

Reply:

We agree that 1-yr is a very short period, however, our experiment is meaningful in examining the impact of different aerosol activation modules on predicted cloud/radiative variables.

To determine if the changes in model predictions such as SWCF due to changes in model configurations from the 1-yr simulation are statistically significant, the student's t-test analysis is performed between the runs pairs of 2001 simulations with different aerosol activation modules. A probability value from the student's t-test is 1×10^{-12}, which is less than 0.05 (i.e., 5%), indicating that the differences between the simulation pairs are statistically significant at the 95% confidence level. The results show that the changes in most cloud/radiative variables including SWCF due to changes in model configurations are statistically significant.

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 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. to provide initial conditions for chemical species that are treated in CB05_GE but not in MOZART. The initial conditions have been clarified in the updated manuscript.

We selected the coupled version of CESM to realistically simulate the impact of aerosol activation within an Earth Systems framework. This has been clarified in the updated manuscript.

11. Page 32300, line 6. NMB is bias normalized by the mean?

Reply:

Correct. The Normalized Mean Bias (NMB) is given by:

$NMB = [\sum_{i=1}^{N}(M_i - O_i)] / \sum_{i=1}^{N} O_i = (\frac{\overline{M}}{\overline{O}} - 1)$**, where** $\overline{M} = (1/N)\sum_{i=1}^{N} M_i$ **,** $\overline{O} = (1/N)\sum_{i=1}^{N} O_i$ **,** M_i **and** O_i **are values of model prediction and observation at time and location** i**, respectively.** N **is the number of samples (by time and/or location).**

12. Page 32302, line 16. Liu et al. (2011) should now be Liu et al. (2011b).

Reply:

We kept as Liu et al. (2011) because the other reference is Liu et al. (2012)

13. Page 32302, line 22. It's likely that the treatment of ice nucleation affects LWP in the arctic. See, e.g., Engstrom et al., J Climate, 2014.

Reply:

Reference to Engstrom et al. (2014) is included in the updated manuscript.

14. 32303, line 12. Changes of 4% for SWDOWN is not small in absolute terms. Note that CAM5 is highly tuned with the ARG scheme to produce a small NMB for SW flux. A variety of cloud parameters have been adjusted. Retuning with FN would be required to produce small NMB values again.

Reply:

We agree with the reviewer that the radiative changes occurring from the different aerosol activation parameterizations are not small in absolute terms. We consider retuning the model with a new activation scheme to be beyond of the scope of this work because our purpose is to examine the changes in model predictions (in particular cloud/radiative properties) caused by different aerosol activation parameterizations. Due to the potential tuning issues, we now mention model tuning in the discussion of SWDOWN and other radiation variables in the text of the updated manuscript.

15. Page 32303, lines 15-16. What is the basis for this suggestion? LWP increases considerably, so the increase in CDF can't explain all of the change in SWDOWN.

Reply:

We agree that the change in CF cannot explain changes in SWDOWN, which is caused by changes in several cloud variables. The statement has been changed to:

"The larger underprediction of SWDOWN in the FN05 series of simulations is likely associated in part with the overprediction in CF and in part with increases in CDNC, LWP, and COT."

16. Page 32303, lines 16-17. I really doubt this, as LW saturates quickly, and hence depends more on cloud altitude and CF than LWP.

Reply:

This statement has been removed.

17. Page 32303, lines 20-22. How can NMB be so large for T2? If the mean is 270 C, an NMB of 10% is 27 C! Doubling the NMB is NOT slightly larger.

Reply:

See response to comment #2 concerning temperature and precipitation evaluation.

18. Page 32304, lines 15-17. While relating CDNC biases to AOD biases is tempting given the ubiquity of AOD retrievals, it would be helpful to know if the simulated CCN is biased. There is CCN data available, albeit not nearly as pervasive as AOD data.

Reply:

We agree with the reviewer that evaluation of predicted CCN would be more informative to the attribution of CDNC biases. We now include global satellite-model CCN comparison in Tables 2 and 3 along with brief discussion within the text of the updated manuscript.

19. Page 32305, line 22. Why is the difference so much larger than that found by Ghan et al. (2011)? I really doubt the greater change is because Ghan et al. compared column droplet number rather than low-level droplet number concentration. Has the FN scheme changed? Please note and explain this change.

Reply:

The FN scheme used in our work was the latest version that contains several updates, although the FN05 remains the same as that used in Ghan et al. (2011).

To address the comments, we have included the following discussion in the updated manuscript: "This increase is substantially larger than the 20–50% increase reported by Ghan et al. (2011) for CAM5 but closer in magnitude (although larger) to the 100% increase reported by Zhang et al. (2012) for GU-WRF/Chem. Such differences can be attributed to differences in mass accommodation coefficients of water vapor used (1.0 in AR-G00 vs 0.06 in FN05), methods in solving max supersaturation, the temperature-dependence in the calculation of Kelvin effects (temperature dependence is neglected in AR-G00 but accounted for in FN05).

20. Page 32307, lines 17-20. A more likely explanation is that most clouds in the tropics are convective, which do not treat activation and hence are not dependent on the activation parameterization.

Reply:

This point has been added in the updated manuscript.

21. Page 32307, lines 25-28. The treatment of ice nucleation can have a large influence on LWP in low arctic clouds. You can cite Liu et al. (2011b). The following sentence notes this. The difference between MODIS retrievals and the simulated cloud properties in the arctic is much greater than the difference between the properties simulated by the different activation schemes. This suggests the sensitivity to the treatment of droplet number is not that important there.

Reply:

We agree with the reviewer that the properties of clouds in polar regions are not sensitive to treatment of droplet number and have cited Liu et al. (2011) and others in the updated manuscript.

22. Page 32308, lines 7-25. Since ocean temperature is allowed to respond to the changes in the cloud properties, one cannot ascribe all of the change in SWCF to the changes in aerosol activation. The feedback of the ocean temperature changes on SWCF must also be considered. It cannot be separated from the experiment design, but the feedback should at least be discussed. Better to have prescribed ocean surface conditions.

Reply:

We have included discussion of ocean-atmosphere feedbacks in the updated manuscript.

23. Page 32308, line 26 – page 32309, line 15. Why do you show and discuss changes in T2 and precipitation? The coupled model is far from being fully adjusted to the solar flux changes after just one year of simulated time. The reduction in precipitation is not simply due to inhibition of autoconversion, as the surface is cooling, thus suppressing evapotranspiration. I suggest you remove this entire paragraph.

Reply:

Discussion of temperature and precipitation changes due to the different activation schemes has been removed in the updated manuscript.

Response to comments from Reviewer 2

Brett Gantt et al.

Reviewer 2: *General: The manuscript describes the influence on the performance of the CESM/CAM5 model system of a new treatment of cloud droplet nucleation. Six different simulations are compared, one using the Abdul-Razzak & Ghan (2000) activation scheme, while the other 5 use the Fountoukis and Nenes (2005) [FN05] activation scheme. These latter 5 simulations differ in the degree of updates/modifications of the FN05 scheme, accounting for processes such as insoluble adsorption, the impact of giant CCN activation kinetics, the impact of dynamic entrainment, or all of those.*

This is definitely a worthwhile study. The uncertainty in current estimates of the aerosol indirect effect is still very large, and model improvements are urgently needed. To develop and implement new physically-based parameterizations for the underlying processes, and then to validate them, as done here, is a key step in moving forward in this area. Therefore, I would like to see this work published. However, before acceptance of the manuscript can be recommended, there are significant problems with the presentation of the results that need to be addressed. They are described under "Major comments" below.

Recommendation: Major revisions

Reply:

We thank the reviewer for constructive comments. Please see our point-by-point reply below.

Major comments:

1) The choice of data for validation A number of data sets of satellite-retrieved cloud properties is now available, with different strengths and weaknesses, e.g., CloudSat/CALIPSO (e.g., Su et al., 2013 in JGR), MODIS/CERES, SSM/I. Even for data from the same instrument, different algorithms give different results, especially for LWP, which is particularly high in MODIS data. A discussion of this is needed, e.g. in section 2.3, as well as in connection with the validation of the results in Table 2. In the discussion on page 32302, the authors give the impression that the underestimation of LWP is solely due to model deficiencies and 3-D effects in the MODIS retrievals, but if another data set had been used for LWP, these large underestimations might not have been present. The MODIS deficiencies are particularly severe over the polar regions, where very large values of LWP and COT are found. Such high values over the polar regions are inconsistent with in-situ measurements (e.g. McFarquhar et al., 2007 in JGR on M-PACE), and are clearly an artefact of the MODIS retrievals. That is not surprising given the fact that solar radiation is basically absent for half of the year in the Arctic, and in addition the wintertime atmosphere there is almost isothermal. This means that most of the MODIS channels in the visible, near-infrared and infrared are more or less useless for detecting clouds during the winter season (mid-September through mid-March). The most reasonable way to deal with that in Figure 1 would have been to only show the MODIS data at latitudes equatorward of 50 or 60 degrees.

Reply:

We agree with the reviewer that MODIS data is unreliable for polar regions, and have adjusted Figure 1 and the subsequent discussion in the updated manuscript to exclude polar comparisons.

*2) The selection of figures and their size The paper contains a large number of figures (45), and many of them are not informative. In addition, most of them are too small to be legible. Specifically: a) Figure 1 is OK as it is b) Figure 2 is too small, but otherwise OK. There is plenty of space on both sides, so it should be easy to expand the figures. c) Figure 3: Better to place the two panels side by side. d) Figure 4: Again the figures are too small. Another issue: Why is the difference in SWCF and surface incoming shortwave given in %? That is difficult to interpret. W / m**2 would be better. e) Figure 5: These figures are very noisy, and there is absolutely no need to show both absolute and percentwise changes for each of the four simulations. Please choose one of them and skip the other! With four panels, the figures can be made larger for clarity. f) Figure 6: Again, the 8 panels are too many and too small. I suggest skipping at least half of them, either showing 4 panels for only one of the quantities or only showing 1 panel from each of the quantities, from the combined experiment FN05/K09/B10/BN07. g) Figure 7: Too many and too small panels. In additions, the panels for precipitation are so noisy, that there's not much point in showing them. Solution: As in Figure 6.*

Reply:

We agree with the reviewer that the magnitude and clarity of figures could be improved. In the updated manuscript, all of the figures have either been rearranged to improve clarity or removed as suggested.

3) Figure captions The captions do not give enough information. For instance, in Fig.1, which variable is seen on what panel? Also, in Figure 4, "changes : : :. between the FN05 and AR-G00" could e.g. be reworded as "changes from AR-G00 to FN05" for added clarity.

Reply:

We have updated the figure captions in the updated manuscript to better describe the figures.

4) The conclusions drawn from the validation exercise The authors need to be more neutral and objective when they discuss the results. For instance, they argue (e.g. on lines 23-24 on page 32310) that the results are improved for CDNC, COT and LWP. However, looking at Figures 1 and 2, we see that concerning CDNC there are improvements in some areas (e.g. over the mid-latitude oceans), while the results have become worse in other areas (e.g. over SE Asia, Europe and N-Africa), where significant overestimations are evident with the new activation treatment. Globally, it is simply not true to claim, as the authors do, that simulations of CDNC have been improved.

Reply:

We agree with the reviewer that the neutrality of the results can be improved, and have made subsequent changes to the updated manuscript when describing the model validation.

5) Somewhere, the authors need to give information about the added computational cost of the FN05 schemes compared to AR-G00. This could e.g. be done in the introduction (cf. current text on lines 10-11 on page 32295).

Reply:

The updated manuscript now includes information about the computational cost of the FN05 scheme, which takes 10% more than that by AR-G00.

Minor comments: Line 8, page 32294: Typo: "signal" should be 'single'. Line 10, page 32294: Ambiguous sentence: "more consistent with that of : : :". Not clear what is meant. Please rephrase. Lines 11-15, page 32295: Were really all these aerosol nucleation formulations used simultaneously. Why? Line 19, page 32298: Right parenthesis should be moved to come right after "BN07". Lines 18-19, page 32298: "resulted" should be "resulting". Same place: What is meant by "chemistry feedbacks to meteorology through various direct and indirect effects"? Please rephrase. Line 10, page 32302: "underprediction" should be "overprediction". Line 11, pages 32302: "compensates" should be "compensate", "results" should be "result". Line 21, page 32305: "in the Tibetan plateau" should be "over the Tibetan plateau". Line 14, page 32306: "due to feedbacks from : : :". What is the nature of these feedbacks? Line 14, page 32307: "improved" should be "reduced". Lines 21-22, page 32307: "because of the influence of radiatively active snow on overlying cloud fraction". How do you know that this is the reason? One possible solution would be to precede with "possible partly" or something like that. Line 3, page 32308: "underpredictions of CF, COT, and LWP". In fact, there is no underprediction for CF by AR-G00. Line 10, page 32309: "where" should be "by which". Line 21, page 32309: In general, the text in the manuscript is very often too technical with widespread use of acronyms instead of words. This is one example where a reader that perhaps only has time to read the Abstract and the Conclusions will stumble over the unnecessarily cryptic language. Instead of "AR-G00" and "FN05", please explain in words. Lines 1-2, page 32310: "which may be explained by feedbacks ..". What is the nature of these feedbacks? Line 11, page 32310: "The more accurate prediction of CDNC : : :". The prediction of CDNC is not in general more accurate than before (see major comment #4). Line 12, page 32310: Again, the "acronym syndrome". Please spell out "NMB".

Reply:

All above suggested changes have been made in the updated manuscript.