Reply to Referee #1’s Comments

The authors have made a substantial effort to revise the manuscript. Specifically, they have conducted three additional 5-year integrations using prescribed SST. The results of these runs provide a much more solid base comparing to those previous 1-year runs for the performance evaluation of the new model, especially in simulating atmospheric chemistry and aerosol life cycle. I also admire the authors’ effort to reemphasize their discussions on science rather than model development related topics, and also to accommodate many suggestions from the reviewers. The revised manuscript appears to be a significant improvement from the original one. In their responses and also revisions, the authors have addressed many of my concerns raised in previous review. There are, however, still a few remaining issues for the authors to improve in my opinion. The following comments are provided for the authors to consider when making their revision.

Reply:

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

The first issue I see a room for improvement is regarding the discussions on modeled changes of meteorological/climate variables. Specifically speaking, these include the changes of T2, Q2, WS10, precip, and PBLH. Discussions on radiative variables and cloud forcing are fine. The reason is rather simple and actually explained in my previous comment regarding the 1-year transient configuration, that is the changes of these meteorological/climate variables mostly represent exaggerated transient responses (mostly noise in other words) due to the model configuration (mainly ocean response). The adoption of climatological SST or the so-called AMIP (Atmospheric Model Intercomparison Project) configuration is an excellent choice because ocean controls to a great extent the climate responses, and observed SST largely reflects actual responses of the climate system to anthropogenic forcings. This is why such a configuration is commonly used when modeled physical or chemical features rather than dynamical feedbacks to these features are concerned. To interpret the changes of climate/meteorological variables in this configuration, however, should be constrained or simply avoided particularly when dealing with water cycle because of the stiff atmosphere-ocean interface due to prescribed SST. Again, the purpose of this paper is to demonstrate the difference in modeling chemistry and aerosol features of the new model components brought to CESM, for discussion on climate consequences, more sophisticated configuration and much longer integration time would be needed. Papers that do not follow the correct procedure might make to the “news” but would not stand long.

Reply:

We agree with the reviewer and have removed the information on changes in meteorological/climate variables in text discussions, Tables, and Figures in the revised paper.

Several specific comments.

1. Line 23, suggest adding “thus could” before “improve model performance”; the same applies to Line 27, suggest using “could further improve” instead of “further improves”.

Reply:

We have modified this in the revised paper.

2. Line 32, suggest deleting “as well as near-surface … precipitation”.

Reply:

We have modified this in the revised paper.

3. Line 76, “…of these aerosols”, perhaps should say “of these aerosol constituents” instead.

Reply:

We have modified this in the revised paper.

4. Line 101-135, a very brief (1~2 sentences) summary of the major features and differences of these various schemes might be very useful.

Reply:

We have modified this in the revised paper.

5. Line 198-217 and other places: please make it clear enough to a wide reader group that this “J value” is referring to “nucleation rate”, not “photolysis rate”. The latter is commonly used in atmospheric chemistry literature.

Reply:

We have made it clear that J is referring to new particle formation rate in the revised paper.

6. Line 237-239, “given their…” this sentence could be revised, e.g., “giving the high computational cost due to …”.

Reply:

We have modified this in the revised paper.

7. Line 325-326, “All production… December 31, 2005” duplicates the previous sentence.

Reply:

We have modified this in the revised paper.

8. Line 387, “discussed below. That” to “discussed below, that”.

Reply:

We have modified this in the revised paper.

9. 4. Model Evaluation: again, here and in many other places, I'd suggest the authors to remove "meteorological" and discussions of changes of T2, Q2, and WS10 from the text and Tables. The authors could make a clear statement that the major interest of this paper is to evaluate the model performance in atmospheric chemistry and aerosol simulations.

Reply:

All information regarding meteorological variables has been removed in the revised paper.

10. Line 445: the authors might want to use a commonly used unit for OH in molecules/cm^3.

Reply:

We have modified this in the revised paper.

11. General comment on "5. Impacts …2001 Predictions", perhaps more adequate terms than "2001 predictions" are "sensitivity simulations". If prescribed 5-year SST run is computational demanding, the discussion in this section should emphasize more on the nature of these sensitivity tests exploring the differences brought by various schemes. Also, wondering if the authors want to add a few comments to compare 5.5 and later 6 (i.e., 5-year integration), because the latter demonstrates a better evaluation base for the model performance.

Reply:

We have modified this in the revised paper.

12. Line 945, "increases over global with a…", indicate increases from what. There are many other similar sentences.

Reply:

In lines 944-946, we have indicated the comparison is between MAM_SIM_5Y and MAM_NEW_5YA: "Compared with MAM_SIM_5Y, J from MAM_NEW_5YA increases over globe with a global mean difference of 0.066 cm^{-3} s^{-1}, due to the use of a lower mass accommodation coefficient of H_2SO_4 in MAM_NEW_5YA".

13. Line 950-955, again these should be avoided.

Reply:

These discussions have been removed in the revised paper.

14. Line 971, "most gas species", did the authors mean "gaseous aerosol precursors"? Line 972, "more accurate aerosol thermodynamic treatments", is this referring to the extremely low accommodation coefficients (actually discussed a few lines later)?

Reply:

Yes, "most gas species" refers to "most gaseous precursors of aerosol". We have modified this in

the revised paper.

"more accurate aerosol thermodynamic treatments" refers to the incorporation of ISORROPIA II in MAM_NEW_5YA. We have indicated this in the revised paper.

15. Line 976, "previous works", the authors might want to provide references for this, either here or in later discussion.

Reply:

We have listed references below the Table 8. We also provide references here in the revised paper.

16. Line 979-981, "SO4= burden higher…due to greater SO2 oxidation" seems inconsistent with higher H2SO4 "due to lower mass accommodation coefficient". Is "oxidation" here referring to aqueous phase oxidation? If so, please make it clear.

Reply:

The oxidation here refers to gas phase oxidation. SO_4^{2-} burden here only includes aerosol phase not cloud phase. Higher SO_4^{2-} burden results from higher SO_2 burden. Higher SO_2 burden leads to more SO_2 to be oxidized to produce SO_4^{2-}, which overweighs the impacts from less H_2SO_4 condensation due to lower mass accommodation coefficient.

17. Line 983-984, it seems that dry deposition change is less likely to cause such a change in carbonaceous aerosol burden, also "slower … aging rate" seems inconsistent with higher H2SO4.

Reply:

Condensation onto the primary carbon mode produces aging of the particles in this mode. A lower accommodation coefficient is used in MAM_NEW_5YA, which results in less condensation. Therefore, the fraction of aged particles has decreased. This has been clarified in the revised paper.

18, Line 1025-1040, it's better to delete "T2", "Q2", "WS10", "precip", "meteorological" from the discussion, here and elsewhere.

Reply:

These discussions have been removed in the revised paper.

19. Tables. It is odd not listing the mean values of modeled variables while listing their observed values in Table 3 – Table 7. Table 1, purpose column of MAM_NEW_5YB, "Earth's components" reads odd.

Reply:

Mean modeled values can be estimated from mean observed values and model mean bias. We have added mean values of modeled variables in the revised paper and revised purpose column of MAM_NEW_5YB.

Reply to Referee #3's Comments

The original manuscript did not provide adequate details on how the authors have implemented ISORROPIA II (a bulk thermodynamic equilibrium model) in CAM5 to perform size (mode) resolved gas-particle partitioning of sulfate, nitrate, chloride, and ammonium. The authors now state that they actually use a kinetic approach, but unfortunately offer no details on how exactly this is done. Below are some questions that need to be answered to better understand their approach:

Reply:

We thank the reviewer for the second-round review. The default treatment for gas to particle mass transfer uses a kinetic approach (although it assumes non-volatile for H_2SO_4 and NH_3, see our reply to specific comment 2). ISORROPIA II was implemented as part of the kinetic mass transfer treatments. The sequence for the model to simulate aerosol processes is condensation, nucleation, thermodynamics (using ISORROPIA II), and coagulation.

We provide below our point-by-point replies.

1) How exactly do the authors calculate the driving forces for mass transfer for each species to each mode?

Reply:

This kinetic mass transfer treatment was included by the original CAM5 code developers at NCAR/PNNL (see Liu et al., 2012). We did not modify this part of the code. In the released version of CAM5, the following equation is used to calculate the driving forces for mass transfer for each species to each mode,

$$I_{net} = \int dx \frac{dN}{dx} I_{cond} \quad (1)$$

$$I_{cond} = 2 \times \pi \times D_g \times D_p \times F(Kn, \alpha) \quad (2)$$

$$F(Kn, \alpha) = \frac{0.75 \times (1 + Kn)}{Kn \times \left(\frac{1 + Kn}{\alpha} + 0.283 \right) + 0.75} \quad (3)$$

where D_p is the particle diameter; x is the logrithemic diameter of particle, = $\ln(D_p)$; dN/dx is the log-normal particle number density distribution; I_{net} is the gas condensation rate; Kn is the Knudsen number; α is the accommodation coefficient of condensable vapor; D_g is the gas diffusivity, and F(Kn, α) is the Fuchs-Sutugin correction factor that describes the resistance to uptake caused by gas-phase diffusion. Equation (1) is solved using the Gauss-Hermite quadrature of order 2. Based on equation (3), as α approaches zero, F(Kn, α) approaches zero. Consequently, I_{cond} (i.e., the uptake rate) approaches zero in equation (1).

To address the reviewer's comment, the above equations have been added in Section 2.1 that describes the model treatments in the released version of CESM/CAM5.

2) What do the authors mean by "Since by default the model treats the condensation of inorganic

volatile gas species as irreversible process (no evaporation), …"? What is this so-called "default," and why does it treat condensation of volatile species as if they are non-volatile? This basic foundation of their kinetic approach appears to be questionable, but I cannot comment on it further until I have more details on it.

Reply:

The "default" refers to the officially public released CESM1.0.5/CAM5.1 (without any modifications by us). In this released version of the model, the condensation of inorganic gases (i.e., H_2SO_4 and NH_3) is treated to be irreversible. Assuming irreversible condensation for H_2SO_4 has been commonly accepted in 3-D atmospheric models due to its extremely low saturation vapor pressure. Although NH_3 is volatile, the acidity in the particulate caused by the condensation of H_2SO_4 provides a driving force for it to neutralize sulfate in the particulate phase to form ammonium salts (CAM5 assumes this salt to beammonium bisulfate (NH_4HSO_4)). NH_3 condensation stops when the NH_4/SO_4 molar ratio of a particle mode reaches 2 (i.e., fully neutralized by SO_4 ions). While it is more accurate to treat volatile species such as NH_3 a truly dynamic manner (i.e., allowing it evaporates and forms all possible salts instead of just NH_4HSO_4), the non-volatile (no evaporation) assumption used in the default CAM5 code greatly simplifies the thermodynamic calculation so that this model can be applied for long-term climate modeling. Following this approach, we incorporated HNO_3/NO_3^- and HCl/Cl^- in MAM7 by using a lower limit value of accommodation coefficient for the condensation process to compensate the loss of the evaporation process. Such a treatment can be further improved in the future when resources (both human and computer) are available.

3) How does the kinetic approach ensure electroneutrality in each mode after kinetically condensing H2SO4, NH3, HNO3, and HCl at different rates (using different accommodation coefficients)?

Reply:

In this case, we followed the default gas/particle mass transfer approach to assume that the condensation of NH_3 will stop when the mole concentration of cations (i.e., NH_4^+, in this case) is equal to the sum of those of anions (i.e., $[NH_4^+] = 2*[SO_4^{2-}]+[NO_3^-]+[Cl^-]$).

To address the reviewer's comment, the above treatment has been clarified in Section 2.2.4.

4) The authors say that evaporation is not treated in the model, so then how would HCl evaporate from sea salt when H_2SO_4 and HNO_3 condense on it?

Reply:

This evaporation process for HCl is treated by ISORRPIA II through thermodynamic partitioning, strictly speaking, "thermodynamic partitioning" is more accurate than "evaporation". We have corrected this in the revised paper.

5) The authors need to provide mathematical proof for their claim that the lower limit of mass accommodation coefficients correspond to uptake coefficients, which represent the net fluxes.

Reply:

Please see the fuchs_sutugin equation in (3) in our reply to the specific comment 1.

As the accommodation coefficient (α) approaches zero (low limit), the denominator approaches infinite, and F(Kn, α) approaches zero (low limit) consequently, the gas condensation rate, I_{cond} (i.e., the uptake rate) approaches zero in (1) (low limit). This approximation is consistent with the physical meanings of the accommodation and uptake coefficients. While α represents the incoming flux of the condensing species, an uptake coefficient represents the net fluxes and is generally smaller than α. Therefore, the lower limit of α can be considered to correspond to uptake coefficients.

To address the reviewer's comment, the above treatment has been clarified in Section 2.1.

6) Again, ignoring coarse mode sea salt and dust from participating in gas-particle partitioning of H2SO4, HNO3, and HCl is a gross oversimplification. Since the authors are using some sort of a kinetic approach, why cannot the coarse modes be also included in the gas-particle partitioning calculation?

Reply:

As explained in the revised paper, the dynamic gas/particle mass transfer approach for coarse modes is computationally expensive (by a factor of 3 or more, depending on the model and configuration) and may be important in areas where dry coarse partiles are abundant (e.g., deserts). As such, such a treatment has not been implemented in many 3-D atmospheric models. To maintain the computational efficiency for CESM/CAM5 that is designed for long-term climate modeling, this is not considered in the initial implementation of ISORROPIA II. The dynamic approach for coarse modes can be implemented in the future if the advancement of computational powers enables such a detailed treatment in long-term climate models.

To address the reviewer's comment, we acknowledge this as a possible reason causing large bias in simulated Cl^- ion concentration in the coarse mode, See Section 6.1 and a limitation of this work, see Section 7.

7) Why was $NH_3(g)/NH_4^+$ not added to cloud water?

Reply:

In default MAM7, $NH_3(g)/NH_4^+$ are already included in cloud water. We then added HNO_3/NO_3^- and HCl/Cl^- in cloud water.

This has been clarified in the revised paper.

Again, these issues must be clearly addressed before the present work can be considered an improvement to the existing CAM5.1 model. I cannot recommend that the manuscript be accepted or rejected until the authors clearly explain their approach with enough detail such that the reader can reproduce the kinetic approach (using ISORROPIA II) on their own. The authors are requested to not simply cite some other reference(s) for details on the present approach to circumvent the issues outlined above.

Reply:

To address the reviewer's comments, we have provided a more described description on the original model treatments for condensation and our modified treatments to add the condensations of additional species such as HNO_3 and HCl.

Reference cited in this reply:

Liu, X., Easter, R. C., Ghan, S. J., Zaveri, R., Rasch, P., Shi, X., Lamarque, J.-F., Gettleman, A., Morrison, H., Vitt, F., Conley, A., Park, S., Neale, R., Hannay, C., Ekman, A. M. L., Hess, P., Mahowald, N., Collins, W., Iacono, M.J., Bretherton, C. S., Flanner, M. G., and Mitchell, D.L.: Toward a minimal representation of aerosols in climate models: description and evaluation in the Community Atmosphere Model CAM5, Geosci. Model Dev., 5, 709-739, 2012.