"A detailed aerosol mixing state model for investigating interactions between mixing state, semivolatile partitioning, and coagulation", Response to the reviewers

We appreciate the comments and suggestions from the anonymous referees and provide our responses below.

Response to Reviewer #1

A more detailed and precise model description should be given

The revised manuscript has updated the Model Formulation section to give a clearer and more complete description of the model. Model variables, mathematical equations, and solution procedures are more precisely defined.

How useful would this model framework be for treating more realistic scenarios with more species?

For more complex scenarios, the external mixture definitions need to be adjusted to maintain a reasonable number of compositional populations. This can be accomplished by grouping individual compounds together into a smaller number of components that define the mixture or with other customized mixture definitions as long as all possible compositions can be assigned to a unique population. Details about computational costs are included in a new section added to the revised manuscript as described in the response to comment 8 and the responses to reviewer 2.

1- Which variables and equations are used and solved by the model?

The primary variables defining aerosol particles in the computer model are the particle diameter and mass concentration of individual compounds within each compositional/size bin. When needed for model subroutine calculations the particle number concentration and compound volume concentrations within bins are calculated from the diameter and mass concentration variables. The revised manuscript has new text explaining and better defining the variables used in the model. The specific equations solved by the model for coagulation and condensation processes have also been more clearly described and defined within the text.

2- page 427, lines 5-10: what happens to the mass of B - is component mass conserved?

The text has been revised to clarify that when a very small AB particle coagulates with a larger A particle, the mass of both A and B from the AB particle is moved to the A population. The model equations for coagulation and condensation are formulated to conserve total, component, and compound mass.

4- Figure 3: add information to caption regarding the simulation time for which results are shown.

Captions for Figures 3-5 and titles for Tables 3-6 have been updated to specify that the results shown are for a simulation time of 188 hours. As explained in section 3, a diurnal temperature

cycle causes gas and aerosol concentrations to change throughout the day, but after several days the model reaches a repeating steady state. Additional simulation time did not noticeably change the daily cycles of bulk aerosol mass, number, and composition or its distribution among the different size and composition bins.

5 - Figure 2 and 3 legend uses "INORG" instead of "INERT"

The legend for these figures has been corrected to use INERT, consistent with the text.

6- Table 1: what particle sizes are assigned to emissions?

The emissions of INERT, PO1, and PO2 have the same size distribution as the initial size distribution shown in Figure 2. Text to explain this has been added to the description of Table 1 and the caption for Figure 2. Existing text discussing Figure 2 also notes that emissions have the same composition and size distribution as the initial distribution.

7- Figure 3, Figure 4, Table 3: Add another figure that shows the size distributions of the individual particle populations.

A new figure, Figure 5, has been added that shows the size distribution of each of the compositional populations in the base case simulation both with and without coagulation. Relatively unmixed populations have mean particle diameters of approximately 0.1 μm that are only slightly larger than the emitted populations. Highly mixed populations, which are formed by coagulation, exhibit growth to a mean diameter of about 0.3 μm. Discussion of this figure has been added to sections 4.1 and 4.2. Together with the total composition and size distributions shown in Figure 3, the average compositions of each population shown in Figure 4, and the mass concentration values listed in Table 3, this new figure helps provide a more complete picture of the mixing state of the entire aerosol system.

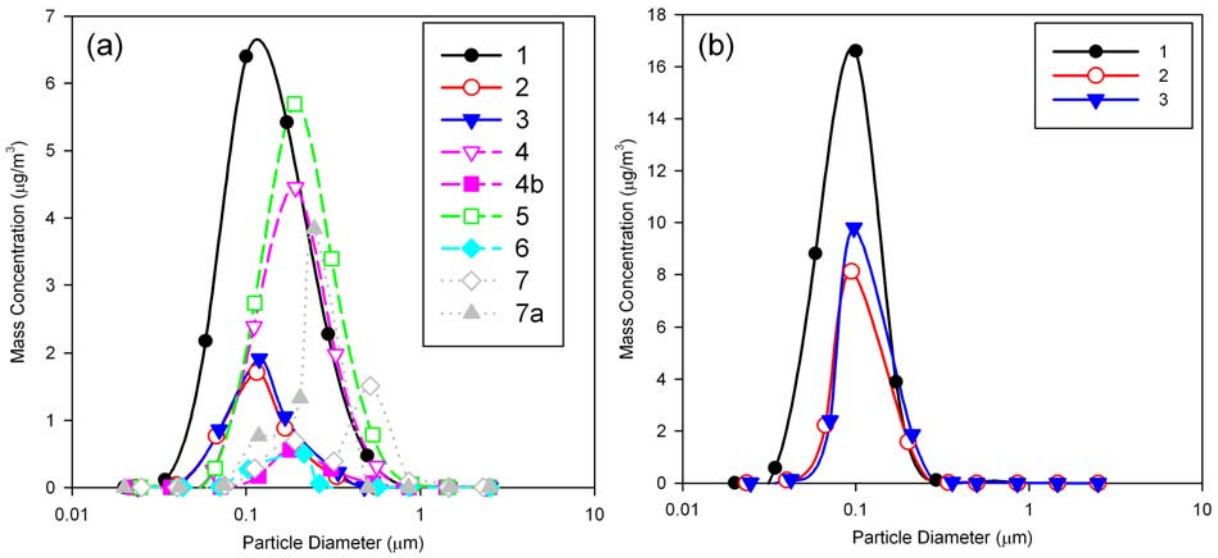

Fig. 5. Size distributions for external mixture aerosol populations formed at t=188 hours in base case scenario simulations. (a) with coagulation, (b) without coagulation.

8- For how many components is the method expected to be computationally feasible?

A new section discussing computational costs and methods to improve computational efficiency has been included in the revised manuscript. A new table detailing computational time requirements for different model configurations has also been added. The current model formulation has undergone only limited optimization to minimize cpu times and has the potential for significant improvements. However, even with more efficient solvers and coding, the computational demands for external mixture simulations will remain high. Cpu time roughly scales with the square of the number of occupied particle bins. As suggested by reviewer #2, the number of compositional populations can be reduced by grouping individual compounds together into a smaller number of components that define the mixture.

Response to Reviewer #2

Agree with Referee 1 that the paper would be improved with a more detailed model description and an extra figure showing the mass distribution of the populations in Figure 4 and Table 3.

As described in the response to reviewer #1, the revised manuscript includes each of these suggested changes.

Would like to see more discussion of computational costs.

As noted in the response to reviewer #1, a new section discussing computational costs and methods to improve computational efficiency has been added to the revised manuscript. It includes a table detailing computational time requirements for different model configurations.

Specific Comments

1a) What is the computational cost of the externally-mixed scheme compared to the internally mixed scheme? Is it dependent on the number of populations present? How does the choice of mixing criteria influence this cost?

In the current model formulation, simulations with the 5 component base case external mixture require approximately 500 times more cpu time than the corresponding internal mixture. Computational costs are roughly proportional to the square of the number of occupied particle bins, and the number of particle bins depends directly on the number of compositional populations. The mixing criteria parameter has little effect on computation time because while it influences how mass is distributed between populations, it does not change the total number of populations for which calculations are performed. Other changes in the mixture definition that reduce the number of populations have a much more significant effect.

1b) For the aerosol system in this paper there are three obvious component populations: INERT, POLAR (PO2 and SV2) and NON-POLAR (PO1 and SV1). Using these component populations would reduce the number of separate mixture populations to 7, with only a little information lost, and probably a large saving in computational cost. Does the model have the flexibility to

change the basis of the mixing criteria in this manner? If so, it would be informative to see the effects of such a change.

This is a valuable suggestion for reducing the computational cost of model simulations. The model does allow flexibility in how external mixtures are defined. New model simulations have been performed using the suggested 3-component mixture definition (INERT, POLAR, NONPOLAR) and have been added to the revised manuscript. Because it requires only 7 populations, instead of 31, computational time is reduced by a factor of 30. Model results with this simplified mixture are extremely similar to the base case simulations. Total and compound mass concentrations, total mass concentrations, and overall size distribution are nearly identical to the base case. The simplified mixture combines base case populations 4 and 4b together and 7 and 7a together, and mass is slightly higher in population 5 and lower in population 1, but otherwise the simulations results are very similar for all populations. Other mixture definitions besides the DAMS representation could also be used as long as all possible compositions can be assigned to a unique population.

Technical Comments

1) Equation 4 would be clearer if radius (R_{pj}) was used rather than diameter (D_{pj}).

The revised manuscript uses radius in this equation, now equation (7), instead. Additionally, the subscript has been updated to R_{pNk} to be consistent with other equations in the manuscript.

2) Page 423 (and elsewhere): usage of compound/component could be made more consistent.

Usage of "compound" and "component" has been reviewed and updated throughout the paper. In the revised manuscript "compound" refers to distinct model compounds and "component" refers to the categories on which compositional populations are based. Components may be either an individual compound, as in the base case scenario where each of the five compounds (INERT, PO1, PO2, SV1, SV2) has a corresponding component, or they may include multiple compounds that are grouped together, as in the new simplified mixture scenario where compounds PO1 and SV1 form component NONPOLAR, and compounds PO2 and SV2 form component POLAR.

3) Page 433, line 14: "Tabl" should be "Table".

This error has been corrected.