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Geoscientific Model Development Discussions

Interactive comment on "A size-composition resolved aerosol model for simulating the dynamics of externally mixed particles: SCRAM (v 1.0)" *by* S. Zhu et al.

Anonymous Referee #1

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In this manuscript, Zhu et al. present a new model for atmospheric aerosol microphysical processes that resolves both the size and mixing state of the aerosol particle population. The mixing state representation is quite general and in theory could provide a very detailed representation. The model description section focuses on the condensation/evaporation process component of the model, as the coagulation component is described in an earlier paper. The description includes a detailed derivation of the particle number and mass conservation equations in terms of the model's composition variables. Its integration with other components (nucleation, coagulation, emissions) and time-splitting are also described. Results for two "box model" scenarios are presented. The first is a rather simple idealized case that is used to validate the model

numerically. The second involves more realistic aerosols over the Paris metropolitan area.

Aerosol mixing state is an active area of atmospheric aerosol research. This paper presents new results, is appropriate for GMD, and should be of interest to other aerosol researchers. The manuscript should be acceptable for publication if the following comments can be addressed.

Major Comments

The new model has the ability to resolve aerosol mixing state in great detail, but at a significant computational cost. Thus the following two questions must be addressed. What aspects of mixing state are important to capture in a model or simulation? How should the mixing state representation (i.e., the specific species groups and bin boundaries for the composition fractions) be designed to efficiently capture this information? Unfortunately, the paper has almost nothing to say about these questions. For the second test case (Paris), there is no discussion of why the particular species groupings and composition bin boundaries were chosen. The abstract mentions investigating the importance of representing mixing state (P. 7938, L. 8-10), but the results in Section 4 simply demonstrate the external mixing of the aerosol and do not discuss why it is important. The size distributions and the mass concentrations of secondary aerosol species do not differ much between the internal and external mixing simulations. Aerosol properties that could be sensitive to mixing state (e.g., CCN and optical properties) are not discussed. Some discussion of these issues is needed, and would be of greater interest to readers than, for example, some of the material in Section 2.

In the model description section where the number and mass conservation equations are derived, the mass fraction of the last species (with index c) does not appear, since it is determined by the other (c-1) mass fractions. However, in the model implementation, when the composition bin boundaries are selected, it is possible to include this last mass fraction. Consider, for example, the first composition bin in Table 1, for which

the mass fractions of HLI, HLO, HBO, and BC are all 0-20%. This composition bin could be subdivided into bins with DU mass fractions of 0-20, 20-80, and 80-100%. This might be desirable, depending on the rationale for selecting the composition bins. Incorporating this last mass fraction into the conservations equations (5-8) might be difficult or impossible. However, as discussed below, the C/E solver and the moving center scheme used for redistribution of particles in composition space do not directly use the conservation equations, and subdividing the bins to also reflect the DU mass fraction should not cause any difficulties for either. The authors should at least discuss how this is possible, even if the current model does not have this capability.

The purpose of and need for the derivations and equations in Section 2.1.3 is not clear. These derivations are for a moving (lagrangian) size-composition bin structure. However, the model is designed for application in chemical transport models that require a fixed size-composition bin structure. Thus equations 9-16 do not seem relevant here. Even if these equations were replaced by their fixed bin equivalents, they would not seem very relevant to the SCRAM model for the following reason. The conservation/evaporation calculations for a time step involve solving (integrating) a set of ordinary differential equations for the time step. The particles in each size-composition bin are treated as uniform in size and composition, and the solver calculates their sizes and compositions some tens of seconds later, taking into account activity coefficients, particle phases (liquid/solid/mixed), equilibrium vapor pressures, mass transfer coefficients, etc. (The bulk equilibrium and hybrid approaches provide similar results using different assumptions and numerical methods.) The new size/composition information is then used by the size and composition redistribution algorithms to move particle number and species masses between bins, to reflect their new sizes and compositions. The conservation equations (5 and 8) have flux divergences on their right-hand sides, representing the next fluxes of number and mass into a bin. The moving diameter (or moving center) algorithm for composition (which numerically is probably the simplest algorithm that one could devise), does not use fluxes or flux divergences. The algorithm is consistent with the conservation equation (8) in some sense, but the algorithm

does not utilize the equation at all, and the equation is not needed to understand the model. If the authors feel that the discretized equations (fixed-bin versions) should remain the paper, then they should be in an appendix, although my recommendation is to remove them.

The condensation only test in Section 3 appears to be using a lagrangian bin structure, given the description of the test ("redistribution is not applied") and the near-exact agreement with the reference solution (which uses 500 lagrangian bins). Since the model is designed for CTMs that require a fixed bin structure, this is not a very appropriate test. A better test would use fixed bins, both with very high resolution for composition (e.g., 100 composition bins) and coarser resolution (10 and 3-5 bins). A high-resolution lagrangian-bin externally-mixed simulation could act as a reference for these fixed bin simulations. For comparison of the fixed bin and lagrangian bin results, plots like Figure 4 could be used, although they only provide a visually semi-quantitative comparison. Plotting the means and the standard deviations of the sulfate (i.e., species 1) mass fraction as functions of particle diameter would provide more quantitative comparison.

The discussion in Section 4 suffers in numerous places from insufficient details about actual compositions in the size-composition bins that are discussed. The composition ranges for many of the composition bins are very wide (e.g., 20-80%). Without stating actual compositions (i.e., actual mass fractions of relevant or dominant species groups), the discussions end up being qualitative and somewhat vague. Providing more quantitative information (where appropriate) would strengthen and clarify the explanations of various mixing state features and behaviors.

Other Comments

P. 7939, L. 14-18. Add that by influencing CCN, the mixing state also affects aerosol wet removal and thus the aerosol spatial/temporal distribution.

Paragraph starting on P. 7939, L. 21. For completeness, include some modal aerosol

models such at Stier et al. (2005, Atmos. Chem. Phys., p. 1125-1156) and Bauer et al. (2008, Atmos. Chem. Phys., p. 6003-6035).

Section 2.1. (a) Somewhere in this section, explain how aerosol water is treated. E. g., are “m” (unsubscripted) and “d” the particle dry mass and diameter, or the wet ones? Is water calculated using an equilibrium approach? (b) Somewhere in this section, note that the composition sections/bins can be based on mass fractions of individual species, or mass fractions of groups of species, or a combination of the two.

Sections 2.1.4, 2.2, and 2.3 should be reorganized somewhat. These all discuss the numerical implementation of condensation/evaporation. I suggest putting them all into a Section 2.2 (Numerical implementation of condensation/evaporation). Begin this section with a brief discussion of how gas-particle mass transfer is first calculated in an aerosol chemistry module, then redistribution in size and composition space is calculated. Then Section 2.2.1 (Gas-particle mass transfer) would contain the current 2.1.4 and 2.2), and Section 2.2.2 (Redistribution) would contain the current 2.3.

P. 7953, L. 24-27. The changes to the HEMEM algorithm should be presented in more detail (perhaps in appendix), especially since Devilliers et al. (2013) do not clearly describe how the algorithm works when aerosol number and species masses are both predicted.

Section 2.3, near the end. Add a brief description of ordering details for the size and composition redistribution. E.g., is it sequential, with size redistribution done first, along the size axis for each of the composition bins, then composition redistribution done second, using the compositions after size redistribution? Note that with the moving diameter method, the redistribution could be done in a single step.

Section 2.4, title. Something like “Overall time integration and operator splitting in SCRAM” might be better.

Section 2.4, paragraph 2. Describe more clearly how nucleation is treated. E.g., for

each cond./evap./nuc. time sub-step, first calculate condensation/evaporation, then calculate nucleation, then do redistribution at the end of the multiple sub-steps. (This seems problematic if new particles could grow out of their initial size bin over multiple sub-steps.) Also for nucleation, what is used for the H2SO4 vapor concentration? Is it a quasi steady state value that accounts for the simultaneous condensation loss and gas-phase chemistry production?

Section 2.4, paragraph 3. Split this into separate paragraphs for the bulk equilibrium and hybrid approaches. In each paragraph, give full details of the ordering and sub-stepping of emissions, coagulation, cond/evap mass transfer, redistribution after cond/evap, and nucleation.

P. 7955, L. 17-18. Provide a little more detail about the H2SO4 condensation. Do they specify a H2SO4 vapor source of about 0.46 umˆ3/cmˆ3/h, or an initial concentration of 5.5 umˆ3/cmˆ3, or something else?

P. 7956, L. 2-3. Figure 1 seems unnecessary. Just state in the text that for internal mixing, the initial particles are all 50% species 1 and 50% species 2; and for external mixing, half of the initial particles are 100% species 1 and the other half are 100% species 2.

Section 3, condensation plus coagulation test. It would be interesting to compare the performance with different numbers of composition bins. E.g., compare simulations with fewer bins (3-5 and 10) to a 100 bin “reference” simulation.

Section 4.1, first paragraph. How are the gas concentrations treated? Do they just specify initial concentrations, or do they somehow include gas-chemistry production of condensable species such as H2SO4, HNO3, and semi-volatile organics.

P. 7958, L. 24. Note in the text here that unmixed is used in an approximate sense, as all the composition bins allow some degree of mixing. This is especially true for bin 1 (unmixed DU), where the DU fraction can be as low as 20%.

P. 7959, L. 13-16 and Figures 7-8. (a) There is considerable discussion (much of it on P. 7960) about the contributions of emissions vs. background particles to unmixed and mixed mass and number. This would be much clearer to readers if an additional plot were added to Figures 7 and 8 showing background (initial conditions) only. (b) The emissions contributions to BC and dust mass at large sizes will be clearer with this addition, but the CI of these emissions should be noted in the text, and also the actual BC and DU mass fractions of this CI for one or two of the larger sizes. (For some CI's, the mass fraction ranges for BC and dust are very wide.)

P. 7959, L. 19-21. I question this migration explanation. In Figure 8a, size bin 3 is mostly CI 3. Thus the coagulation of size bin 3 particles with size bin 4 particles would be dominated by [size 3, CI 3] particles with [size 4, CI 14] particles that could produce the [size 4, CI 15] particles.

P. 7960, L. 3-4. Could be more specific here, and say that the [size 3, CI 5] particles come from condensation onto and transformation of [size 3, CI 3] particles

P. 7960, L. 8-10. State which pair of figures demonstrates this feature most clearly.

P. 7960, L. 13-17. This result (larger particles being better mixed) seems rather artificial, caused by (1) the assumption of all background particles being internally mixed and (2) the initial conditions dominating the large particles due to their low emissions and the short duration of the simulations. Large particles (especially coarse) are generally thought to be less internally mixed than fine particles. The authors should consider giving less emphasis to this result.

P. 7960, L. 19-21. This sentence seemed somewhat awkward to me. Maybe change to: "In scenario a, 42% (resp. 83%) of the particle number (resp. mass) originates from initial conditions and is mixed, while the remaining particles are due to emissions and are unmixed."

P. 7961, L. 14. The figure order (11 before 9 and 10) should be changed. If background

(initial conditions) only plots are added to Figures 7 and 8 (see earlier comment), then the Figure 11 plots could also be put into Figure 7 and 8. This would make visual comparison of external vs. internal mixing results easier.

P. 7961, L. 21. Typo: TI6.

Section 4.3, paragraph 2. This discussion seems not of much value unless more quantitative details are provided. (See the last major comment above.) Consider size bin 4, which is CI 4 in internal mixing and CI 4 and 6 in external mixing. The actuall mass fractions in the CI 6 could be quite close to those in the CI 4, in which case the internal/external mixing differences would not be very important.

Section 4.4. (a) The comparisons here should use the all processes simulations (with coagulation), since that is the most realistic, and turning off coagulation has a large impact (as seen in Fig. 7c vs. 7d, and 8c vs. 8d). (b) Since the focus of this paper is mixing state, the internally mixed results in this section should be removed, unless they are strongly needed to explain externally-mixed simulation differences between the dynamic, bulk equilibrium, and hybrid C/E methods.

P. 7963, L. 8-11. Point out that the speed of the hybrid C/E scheme is significantly degraded in the external mixing case. The number of size-composition bins increases by a factor of 20, but the hybrid C/E time increases by a factor of about 135. A brief explanation of why this happens would be of interest, although not necessary.

Section 5. Add some discussion of computational costs.

Table 1. Most of the numbers in the last column (DU) are incorrect. E.g., for composition bins 2-5, the DU fraction ranges are 0-80, 0-20, 0-80, 0-60, and 0-20%.

Figures 7-11. These should be improved in several ways. (a) The vertical bars should align better with the size section boundaries given on P. 7958, L. 9. (b) The density of the vertical or slanted lines in the vertical bars should match the line densities in the legends. (c) Some of the colors are difficult to distinguish. Since only about half of

the 20 composition indices are visible in the plots, the CI's with negligible contributions to mass and number should be grouped into an "other" class (or possibly mixed-other and unmixed-other). E.g., any CI whose maximum mass and number concentrations are less than about 2% of 40 ug/mˆ3 and 5e10 particles/mˆ3, respectively, is not really visible in the plots and should go into the other class. This will reduce the number of colors needed, and colors that are more easily distinguished can be used. (This would be done on an all plots basis, not an individual plot basis.) Note that this would further highlight which CI's are important and which are not. (d) Each caption should list briefly the simulations shown in the figure: external/internal mixing, dynamic/bulk-equilibrium/hybrid C/E solver, and which processes are active.

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