

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.

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The authors thank the referee for providing a thorough review. We modified and clarified the paper as suggested. Each item starts with the reviewer's comment in italics.

1 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.*

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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.

This paper aims at presenting the model. Therefore, we tried to keep the model description as general as possible, so that the model can easily be applied to different applications. An example of application is given here, focusing on showing that aerosols may not be totally mixed in urban areas and how different aerosol processes (condensation/evaporation, coagulation) impact the mixing state. The mixing state representation is flexible enough to be modified by users and it would particularly affect CCN and optical properties. A discussion about the importance of the mixing state for CCN and optical properties is given in the introduction “The mixing state assumption may strongly influence aerosol chemistry and the hygroscopic characteristics of particles. Particles from different origins may not be well mixed, and their chemical composition may vary with their origins, leading to variations in their hygroscopic characteristics. This chemical identity of particles is gradually lost as the degree of mixing increases (or completely lost under the internal mixing assumption). By influencing the hygroscopic characteristics of particles, the mixing state also influences the formation of secondary organic aerosols (SOA), because condensation/evaporation differs for species that are hydrophilic and/or hydrophobic (Couvidat et al., 2012). As

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the particle wet diameter is strongly related to the hygroscopic properties of particles, the mixing state also impacts particle wet diameters and the number of particles that become Cloud Condensation Nuclei (CCN), because the activation of particles into CCN is strongly related to the particle wet diameter (Leck and Svensson, 2014). By influencing CCN, the mixing state also affects aerosol wet removal and thus the aerosol spatial/temporal distribution. Besides, the mixing state influences the particle optical properties, which depend on both the particle size distribution (wet diameters) and composition (different chemical species possess different absorption/scattering properties). Lesins et al. (2002) found that the percentage difference in the optical properties between an internal mixture and external mixture of black carbon and ammonium sulphate can be over 50% for wet aerosols. The mixing state may also influence radiative forcing, as shown by Jacobson (2001) who obtained different direct forcing results between external and internal mixing simulations of black carbon.”

Concerning the particular species grouping, a simple explanation is given in (P.7958,L.12): these species were grouped into 5 groups based on their chemical nature. Additional explanations are now added. “these species were grouped into 5 groups based on their chemical nature, which influences the formation of particles and their optical properties. Black carbon, organic species, inorganic species and dust are separated. Although sulphate could be separated from nitrate and ammonium for optical properties or for comparisons to observations of mixing state (Healy et al., 2012), and although chloride and sodium could be grouped together in a marine environment, all inorganic species are grouped together here for the sake of simplicity. However, because the hydrophilic properties of the particles strongly influence their formation and cloud condensation nuclei, hydrophilic and hydrophobic organic species are separated.” The composition bin boundaries were chosen in order to balance the computational cost and the comprehensive representation of mixing state. These sections are designed to represent poor, medium and high abundance of one group within a particle.

The sentence in the abstract “The importance of representing the mixing state

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when modelling atmospheric aerosol concentrations is investigated in a box model simulation using data representative of air pollution in Greater Paris.” was poorly formulated and is therefore rewritten as “The degree of mixing of particles is investigated in a box model simulation using data representative of air pollution in Greater Paris. The relative influence on the mixing state of the different aerosol processes (condensation/evaporation, coagulation) and of the algorithm used to model condensation/evaporation (bulk equilibrium, dynamic) is studied.”

As the application presented in this paper is a 0D test, the information obtained from the simulation is insufficient to precisely discuss the importance of mixing state on AOD or CNN. We are currently coupling SCRAM to a 3D eulerian chemical transport model, and such discussion will be based on our future 3D simulation results.

•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 conservation 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.

In our model, we chose to compute the mass fraction of one of the group by mass conservation, i.e. the mass fraction of one group is not treated explicitly in the

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subdivision of the mass composition bins. In our example, we chose dust to be the group for which mass fraction is not treated explicitly, but if it is desirable to include the mass fraction of dust explicitly into the mass composition bins, another group could be chosen as the group for which mass fraction is not treated explicitly. This is now added in the paper. "Note that although as an example we chose dust to be the group for which mass fraction is not treated explicitly, another group could be chosen as the group for which mass fraction is not treated explicitly." Sentences are also added to the paper to explain how it would be possible to subdivide the mass fraction of every group. "If all groups need to have their mass fraction treated explicitly, additional composition bins for the last group should be added to the current composition list without any modification to the main structure of the program. The mass fraction of the last group would still be obtained by mass conservation, and the composition bin of the particles would be chosen depending on this mass fraction."

●2993-10:*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 vapour 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.*

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The conservation equations (5 and 8) have flux divergences on their right-hand sides, representing the net 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 in the paper, then they should be in an appendix, although my recommendation is to remove them.

Either eulerian (fixed bins) or lagrangian (moving bins) approaches can be used to solve condensation/evaporation equations. However, the drawback of the eulerian approaches is numerical diffusion (see Kim and Seinfeld (1990)). That is why lagrangian approaches are commonly used, as in our model (instead of a fixed-bin eulerian approach). Equations 9-16 are crucial as they explain the derivation of the equations which are actually solved: equations (14) and (17). Because we use a lagrangian approach, a redistribution scheme onto a fixed-sections grid is necessary for 3D applications. The redistribution is applied after solving condensation/evaporation. The redistribution does not solve equations (5) and (8), it simply redistributes mass and number onto fixed sections. The redistribution algorithm is completely independent from the lagrangian solver, and the 0D model is operational without it. Equations (5) and (8) are continuous equations. They cannot be solved as they are: they have to be discretized. We use a sectional discretization. The discretization is explained in this paper as well as the derivation of the discretized equations from the continuous ones. As suggested by the reviewer, the derivation of the discretized equations is now put in an appendix. But the discretized equations (now 11 and 12) are kept, as those are the equations solved in the model. For clarity, the following explanation is added before section 2.1.1: "A lagrangian approach is used to solve the equations of change for the mass and number concentrations, which are redistributed onto fixed sections through a redistribution algorithm (moving diameter)."

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Section 2.3 is moved to 2.1.5, so that all the algorithm for solving dynamic C/E are now detailed in section 2.1. Section 2.2 presents the simplified approaches that may be used to solve condensation/evaporation and to gain CPU time (bulk equilibrium, hybrid approaches).

•2994-5: *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 sulphate (i.e., species 1) mass fraction as functions of particle diameter would provide more quantitative comparison.*

Because this paper presents an algorithm to solve condensation/evaporation for externally mixed particles, it is important to show the behaviour of this algorithm without redistribution. The impact of using different redistribution algorithms on mass and number concentrations has been investigated for example by Devilliers et al. (2013) who compared different algorithms for size section redistribution. They found that the moving diameter approach is one of the best performing algorithms. Furthermore, for mass fraction redistribution, a moving mass fraction approach similar to the moving diameter approach is the most appropriate one to use. Therefore, we do not need to compare different algorithms for mass fraction redistribution and we feel that it is appropriate to use the moving diameter approach. We agree that it is interesting to investigate the impact of different composition resolutions to external mixing results. So we conducted

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additional tests with 2 and 100 composition sections and compared their size distributions using the mean and standard deviation of sulphate mass fractions as suggested. The following discussion was added to the second paragraph of section 3:

"In order to investigate the influence of the composition resolution on simulation results, two additional tests are conducted using 2 and 100 composition bins. The mean mass fraction of species 1 is computed for all particles within each size section, as well as their standard deviations. Figure 3 shows the size distribution of these statistics. The mean mass fraction is barely affected by the different composition resolutions as the condensation rate of sulphate is independent of the particle compositions. However, a different composition resolution does lead to different standard deviation distributions, as only particles with larger fraction difference ($d > 0.2\mu m$ for 2 compositions and $d > 0.09\mu m$ for 10 compositions) can be distinguished from each other under coarser composition resolutions."

We also tried to plot the result composition distribution of the test with 100 composition sections using the style of figure 4. However the overall trend of the distribution is very similar to the 10 composition plot. Therefore, this figure would not add any interesting information to the paper and we think that it is better not to include it in the paper.

•2994-19: *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 behaviours.*

The composition range is chosen in order to balance the computational cost and the comprehensive representation of mixing state. These sections are designed simply to represent poor, medium and high abundance of one group within a particle. As

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suggested by the reviewer, we have added the actual mass fraction of each species groups during the discussion of section 4, which helps improve the presentation of mixing state.

2 Other Comments

●2994-25: 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.

Thank you for your suggestion, we have added this description into the discussion of the mixing state importance in the fourth paragraph of the introduction.

●Paragraph starting on P. 7939, L. 21. For completeness, include some modal aerosol models such as Stier et al. (2005, *Atmos. Chem. Phys.*, p. 1125-1156) and Bauer et al. (2008, *Atmos. Chem. Phys.*, p. 6003-6035).

We have added the following description in the fifth paragraph of the introduction: "On the other hand, Stier et al. (2005) and Bauer et al. (2008) simulate externally mixed particles using modal aerosol models, where aerosol populations with different mixing states are represented by modes of different compositions (soluble/mixed or insoluble/not mixed). Although these models may be computationally cheap, they may not model accurately the dynamics of mixing."

●2995-3: 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.

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For question (a), water is treated separately from other species, the water content within each bin is computed using ISORROPIA before each loop of condensation/evaporation process. As specified on P.7942-L.16, d_p is the particle wet diameter while "m" stands for particle dry mass.

For question (b), the following sentence was added P.7943-L.19: "As $f_i = m_i/m$ is the mass fraction of species (or group of species) X_i , we may write:"

●2995-8: 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.

We reorganised the sections. Section 2.1 is now dedicated to the dynamic model of condensation/evaporation. This section presents the equations, their discretizations, the numerical implementation and the redistribution scheme. We have moved section 2.3 into section 2.1.5, so that all algorithms for solving dynamically condensation/evaporation are detailed in section 2.1. Section 2.2 is dedicated to the simplified algorithms that may be used instead of the dynamic approach to speed up the computation of condensation/evaporation (bulk equilibrium and hybrid approaches). Finally, section 2.3 explains the overall numerical structure of SCRAM, including all the processes: not only condensation/evaporation but also coagulation and nucleation.

●2995-15: 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

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masses are both predicted.

Devilliers et al. (2013) studied the influence of redistribution on aerosol number and mass treated jointly, whereas previous papers had only studied the influence on either aerosol mass or number. The algorithm was not modified, except to allow the diameter of a section to grow larger than the upper bound diameter of the next section, as detailed in paragraph 2.1.5. For clarity, the following sentence was added to the paper: "In that case, the mean diameter of the section after condensation/evaporation is used to diagnose in which fixed-diameter sections the redistribution is performed."

●2995-9: *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.*

The following paragraph is added at the end of the redistribution section:

"The composition redistribution is applied first, followed by the size redistribution for each of the composition sections."

●2995-14: *Section 2.4, title. Something like "Overall time integration and operator splitting in SCRAM" might be better.*

We have changed the section title as you suggested.

●2995-16: *Section 2.4, paragraph 2. Describe more clearly how nucleation is treated E.g., for each cond./evap./nucl. time sub-step, first calculate condensation/evaporation, then calculate nucleation, then do redistribution at the end of the multiple sub-steps.*

Section 2.3 now describes in greater detail of the time integration. The following

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sentence is added to the paper in Section 2.4, paragraph 2 before "Redistribution" on P.7955, L.4: "Condensation/evaporation/nucleation are solved simultaneously."

●2996-2: *This seems problematic if new particles could grow out of their initial size bin over multiple sub-steps.*

As we are using a lagrangian approach for solving condensation/evaporation, size bin boundaries are not fixed and they can grow freely with the particle within multiple sub-steps. However, the redistribution algorithm is systematically applied after condensation/evaporation, and it redistributes mass and number onto fixed size section boundaries.

●2996-4: *Also for nucleation, what is used for the H₂SO₄ vapour concentration? Is it a quasi steady state value that accounts for the simultaneous condensation loss and gas-phase chemistry production?*

Gas-phase chemistry is not considered here in this 0D module. The H₂SO₄ vapour concentration available for condensation and nucleation is obtained from either emission or initial background concentration. Because condensation and nucleation are solved simultaneously, the same value of H₂SO₄ concentration is used for both processes.

●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.*

We replaced section 2.4, paragraph 3. with the following two paragraphs: "When the bulk thermodynamic equilibrium approach is used to solve condensation/evaporation,

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coagulation then nucleation are solved after each emission time step. The resolution is done as previously explained, except that the dynamic condensation/evaporation solver is disabled: sub time steps are used to solve coagulation and nucleation during one emission time step. Condensation/evaporation is then solved using the bulk equilibrium approach and the redistribution process is applied after the bulk equilibrium algorithm.”

“When the hybrid approach is used to solve condensation/evaporation, a time loop is added with a fixed time step of 600 s outside the emission time loop to compute bulk equilibrium condensation/evaporation for equilibrium sections. This additional time loop is designed to ensure that bulk equilibrium condensation/evaporation of equilibrium sections is not applied too often, so that the dynamic condensation/evaporation of dynamic sections has time to evolve. Redistribution is applied after the bulk equilibrium algorithm. Within this time loop, the aerosol dynamics is solved as previously explained using the dynamic condensation/evaporation algorithm for dynamic size sections: emissions are solved followed by coagulation and condensation/evaporation/nucleation. As in the fully dynamic approach, redistribution is applied after dynamic condensation/evaporation.”

●2996-11: P. 7955, L. 17-18. *Provide a little more detail about the H₂SO₄ condensation. Do they specify a H₂SO₄ vapour source of about 0.46 μm³/cm³/h, or an initial concentration of 5.5 μm³/cm³, or something else?*

We are using a H₂SO₄ vapour source, so the P. 7955, L. 17-18. are updated to: “sulphuric acid vapour source of 0.46 μm³cm⁻³ per hour”

●2996-14: *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.*

We removed figure 1 and replaced it with the suggested description.

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●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.*

As detailed in the major comment replies, following the reviewer’s advice, the performance with different numbers of composition sections was added when testing the condensation algorithm. We do not think that it would be useful to add a similar test for condensation plus coagulation.

●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 condensible species such as H₂SO₄, HNO₃, and semi-volatile organics.*

As previously explained, gas-phase-chemistry is not included in this 0-D model. Gas-phase concentrations are specified with initial concentrations and emissions (for H₂SO₄ and POA). They may also evolve because of condensation/evaporation.

●2996-24: 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.*

We added this note in P.7958-L.24:

“Among them, there are 5 unmixed particles and 15 mixed particles. Here unmixed is used in an approximate sense: it means that the mass fraction of one chemical component is high (between 0.8 and 1), while the mass fraction of the other chemical components is low (between 0 and 0.2).”

●2997-1: 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*

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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.)

We have added the distribution of initial conditions to figures 7 and 8. We also agree it is better to clarify the nature of emissions in the text, so the following sentences have been added at the end of the second paragraph of 4.2: "All emitted particles are unmixed: CI 1 (100% DU) into size section (4-6), CI 3 (100% BC) into size section (3-6). Emissions also involve POA and H₂SO₄ gas-phase emissions."

●2997-9: *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.*

Thanks for the insightful argument, your explanation provides another possible path to the generation of CI 15 in size 4. Nevertheless, coagulation between [size 3, CI 15] particles themselves is also capable to produce [size 4, CI 15] particles. So we rewrote the analyse in P. 7959, L. 21.

"Also, due to coagulation, small particles migrated to higher sections. For example, Fig.8 shows the mixed particles CI 15 migrated from the third size section to the fourth size section, this might be a result of the coagulation between CI 3 particles in the third size section and the CI 14 in the fourth size section. Besides, coagulation between CI 5 particles in the third size section may also produce some part of CI 15 in the fourth size section."

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●2997-9: *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*

Thank you for your suggestion, we rewrote P. 7960, L. 3-4. :

"The condensation of organic matter on freshly emitted BC particles (CI 3) also occurs, as shown by the mixed BC and HBO particles (CI 5) which appear in the third and fourth size sections."

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

We rewrote P. 7960, L. 8-10. :

"For example, as demonstrated by the difference between scenarios (C) and (D), newly mixed particles of CI 4 (between 20% and 80% of HBO) are formed by the coagulation of unmixed particles from CI 6 with others within the fourth and fifth size sections."

●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.*

Yes, indeed this result is rather artificial, and it is a consequence of the set up of the simulation. The purpose of this work is not to provide realistic based conclusions, which will be the purpose of future 3-D simulations. P. 7960, L. 13-17. is only trying to demonstrate how mixing occurs and to which extent. It is also an illustration of the basic informations that can be obtained from simulations. We rewrote this paragraph as follows:

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“Table 2 shows the percentage of mixed particles for each scenario based on both particle number and mass concentrations. It seems that large particles are better mixed than small particles as the mixing percentages of mass are always higher than those of number. However, this phenomenon is specific to this case study; it is caused by the assumption of all initial particles being internally mixed and the initial conditions dominating for large particles due to their low emissions and the short duration of the simulations.”

●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."*

We rewrote the sentence accordingly.

●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.*

We added both the initial conditions and internal mixing result distributions in figures 7 and 8 as suggested by the reviewer.

●P. 7961, L. 21. *Typo: TI6.*

We corrected TI6 into CI 6.

●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*

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bin 4, which is CI 4 in internal mixing and CI 4 and 6 in external mixing. The actual 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.

More quantitative details about actual mass fraction of dominated species group were added in this paragraph. Indeed, the actual mass fraction in the CI 6 (81% HBO) is not so different from the one in the CI 4 (75.6%). However, it is interesting to keep the discussion in the paper, because it demonstrates the additional information provided by the external mixing results. Of course, we have to keep in mind that the difference between internally and externally mixed particles depends on the thresholds used to classify the particles.

●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.*

(a) Here, we focus on the impact of the different algorithms on mixing state, we want to study the impact of the simplified assumption of C/E computation on mixing state. Therefore, these studies are crucial. Adding coagulation would make the interpretation difficult and less accurate. These results are more process-oriented, rather than realistic. Realistic simulations will be performed with 3-D model coupled with SCRAM. (b) Comparisons to internally mixed results is also very important, because we want to compare the results obtained with different simplified assumptions (internally mixed assumption, bulk equilibrium C/E, hybrid C/E). For example, as stated in the paper "Although internal and external compositions are different with the dynamic approach, they are quite similar with the bulk equilibrium approach.". This is important to know, because it means that we should not bother with the externally mixed approach if we

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make the bulk equilibrium assumption.

•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.

For external mixing, the C/E hybrid scheme does not significantly improve the CPU time compared to the C/E dynamic scheme (whereas it does for internal mixing), probably because the redistribution after each time step between the different compositions leads to small time steps in the solver ROS2.

We added the following discussion into the end of 4.4 " This significant speed degradation of the hybrid C/E scheme in the external mixing case is probably a consequence of small time steps used in the ROS2 solver because of the redistribution among the different composition sections performed after each time step. In other words, it takes CPU time to compute the dynamic distribution among the different composition sections."

•Section 5. Add some discussion of computational costs.

The following discussion about computational cost was added in section 5.

"Although the simulation of externally mixed particles increases the computational cost, SCRAM offers the possibility to investigate particle mixing state in a comprehensive manner. Besides, its mixing state representation is flexible enough to be modified by users. Better computational performance could be reached with fewer, yet appropriately specified species groups and more optimised composition discretisations. For example, about half of the 20 compositions designed in this work have really low mass concentrations (e.g. see Figures 7, 8, 9 and 10). Those compositions might be

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dynamically deactivated in the future version of SCRAM to lower computational cost by using an algorithm to skip empty sections during coagulation and C/E processing."

•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%.

We updated the table accordingly.

•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 and $5e10$, 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.

We improved the figures based on the reviewer's suggestions.

References

Kim, Y. P. and Seinfeld, J. H.: Simulation of multicomponent aerosol condensation by the moving sectional method, Journal of Colloid and Interface Science, 135, 185–199, 1990.

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