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.

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

The manuscript by Zhu et al. presents the simulation results from a size - composition resolved aerosol box model SCRAM. After providing the detailed description regarding the model treatments, the authors conduct a couple of ideal case studies to demonstrate the ability of this box model in reproducing the reference results of Zhang et al. (1999). Finally additional case simulations are conducted by using a real case of model inputs from previous 3-D simulation results to show the impacts of different

aerosol dynamic processes, mixing states (i.e., internal vs external mixing), and different assumptions for condensation/evaporation approaches on simulated aerosol mass and number distribution. This manuscript is generally well written with many interesting analyses. It's definitely of scientific interest to the atmospheric science community and I would recommend it to be accepted after a minor revision.

1 Specific Comments

•*One of the main reasons for majority of current 3-D atmospheric chemical transport or air quality models to use the assumption of internally-mixed aerosol treatment is the large computational cost, which could even make 3-D simulations impossible without sacrificing accuracy of aerosol representation (e.g., without reducing the number of aerosol species and size bins). The past studies have also found the overall aerosol mass and number concentrations could be very comparable for different aerosol size distributions between internal- and external-mixing. This point has also been verified in many occasions (in both text and figures) in Section 3 and 4 by this study. So I would really like to see some discussions in the conclusion section on how the current 3-D models can take advantage of and benefit from the treatment of external-mixing aerosols (such as the ability to predict the mixed states of aerosols) in SCRAM to compensate the loss of computational efficiency.*

We added the following discussion about mixing state and CPU time before the last paragraph of the conclusion section.

"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 4). Those compositions might be 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."

We also added discussions about the importance of the mixing state 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 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 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 aerosol. 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."

C3297

● *Page 7938, line 12: "thresholds of 12"; this is for primary aerosol only. There is another standard for secondary aerosol. May need to explicitly mention it here.*

We rewrote the sentence into:

"For example, regulatory concentration thresholds of 12 and 20 μg m^{-3} have been set for PM$_{2.5}$ annual mass concentrations of primary aerosol in the United States and Europe, respectively."

● *Page 7940, lines 9-18: I don't suggest putting such detailed information in the introduction. If it's essential information that authors would like to deliver, they should move it into somewhere else such as the methodology section.*

We replaced the sentences by:

"Dergaoui et al. (2013) further expanded on these modelling approaches by discretising the mass fraction of any chemical species into sections, as well as the size distribution (see Section 2.1.3 for details)."

● *Page 7942, line 19: How do you select values of accommodation coefficient?*

The accommodation coefficient is set at 0.5, as now mentioned in the paper. There is a large uncertainty about the accommodation coefficient, which could vary between 0 and 1.

● *Page 7943, line 6: which version of ISORROPIA here?*

We are using ISORROPIA v1.7 here, as now stated.

● *Page 7956, lines 11-12: I wouldn't say it's a perfect match since there is a little difference for the peak values of number distribution.*

C3298

We replaced "perfect" by "good".

• *Page 7957, Section 4: It seems the SOA formation is not included in SCRAM and it has to rely on the other SOA modules if incorporated into 3-D models? It should be indicated in the manuscript.*

In SCRAM, SOA originate either from initial conditions or they are emitted as semi-volatile organic compounds during the simulation. They are not oxidized, as the gas chemistry is not included in SCRAM. Yes, the gas chemistry will be solved in 3-D simulation with other modules. We updated our manuscript to specify this at the end of the second paragraph of 4.1:

"Besides, gas chemistry (such as SOA formation) is not included in SCRAM, and is expected to be solved separately using a gas chemistry scheme. In the simulations of this paper, organics originate either from initial conditions or they are emitted as semi-volatile organic compounds during the simulation. They do partition between the gas and the aerosol phases by condensation/evaporation."

• *Page 7957, line 25: deposition is also ignored here and should be mentioned.*

We updated the description into:

"As our simulations are 0D, the transport of gases and particles and the deposition processes are not taken into account."

• *Page 7958, line 8: why choose 7 sections? Many existing aerosol modules typically use 4/8/12-bin structures.*

We chose 7 sections here because we obtained the initial condition data from existing and published 3D simulation results (of SIREAM) with 7 size sections.

• *Page 7958, line 12: When you grouped them, does the model still be able to track the concentrations of individual species (this is very important!). How do you treat individual species within each group for mass fraction sections? Such information is expected here.*

Yes the model can still track the concentration of individual species. The model memorizes the relationship between each species index and group index, and stores the mass concentration separately for each species within each size-composition bins. The total mass concentration of each group is computed from the mass concentration of each species based on the species-group relations, allowing the computation of the mass fraction of each group. We added this explanation at the end of this paragraph: Page 7958, line 27.

• *Page 7959, line 3: change all the scenario names to upper cases (e.g., a to A and b to B).*

We updated the scenario names accordingly.

• *Page 7960, lines 13-15: this seems to be conflicted with our general understanding that fine-mode particles are much easier internally-mixed than coarse-mode particles.*

It is caused by the internal mixing assumption we made for the initial particles. The following sentences were added to explain this:

" 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."

●*Page 7961, line 14: I didn't see that Figures 9 and 10 were mentioned before this. Please don't jump the figure numbers.*

We rearranged the figure orders.

●*Page 7963, line 8: The information provided in this table indicates that the CUP time required by the external-mixing for the current box model could significantly slow down the simulation. I would expect some discussion on how this box model could be incorporate in a feasible way into 3-D models without reducing the number of size bins and mass fraction sections (which are keys for accurate simulations of externally-mixed aerosol processes). Are there any rooms for authors to optimize the code to further reduce he computational time, because I am not sure if the current performance in terms of CPU time is acceptable for the 3-D implementation?*

We are currently working on the 3D implementation, which will be presented in another paper. We just managed to successfully run 3-D simulations with SCRAM with a reasonable CPU time (about 8 times slower than the internal mixing case for a dynamic C/E + coagulation simulation) without compromising any size and mass fraction sections. Some discussion about computation costs and optimization has been added in the conclusion as mentioned earlier in the reply.

●*Page 7963, line 11: C/E; this acronym should be defined much earlier in Section 4.2.*

We moved up this definition to the first paragraph of Section 4.2.

●*Page 7963, lines 24-25: I was looking for this information when I read Section 2. I would suggest adding this information where it is appropriate.*

Thank you for your suggestion, we have added this information at the beginning of Section 2.1.3:

"As SCRAM is a size-composition resolved model, both particle size and composition are discretised into sections, while the numbers and bounds of both size and composition sections can be customised by the user."

●*Page 7964, lines 23-24: Again I am really not sure how feasible it could be to incorporate the current box model into 3-D considering the huge increase of computation cost between the internal- and external-mixed results.*

see reply above.

●*Figure 2: Can you replace the lines for reference and internal cases with markers?*

Thank you for your advice, we have replaced the lines for reference and internal cases with markers as you suggested.

●*Figure 6: Is it UTC or local time?*

It is UTC, we updated figure 6 to specify that.

Technical notes:

Thank you for your detailed technical notes, all of them have been taken into account accordingly.