1 Responses to Reviewer #2

We greatly appreciate the reviewer's comments. The page and line numbers that we quote for indicating where we changed the manuscript refer to the revised marked-up version. Our responses are as follows:

(2.1) Vertical transport is more generally used to represent vertical movement and distribution of aerosols. But not only turbulent diffusion and dry deposition affect vertical distribution, why only they are considered?

> In the single column model, we only consider vertical advection due to density changes, which is handled in the WRF timestep component. The grid in the WRF model is constant in its native hydrostatic-pressure coordinate η but not constant in height z. Any pressure-induced changes therefore cause the z grid to shift and this is entirely handled by the WRF model grid transformation. It is important to note that particles will not shift grid cells during this process.
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> In the case of considering large scale vertical advection, similar equations could be derived to create the corresponding transport probabilities. This aspect will be important in the future as the model is extended to be fully three-dimensional where large-scale vertical advection will exist. To explain the treatment of the vertical advection term, we made the following change:
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> - In Section 3, page 6, line 8, we added: "The WRF model is discretized using a terrain-following hydrostatic-pressure coordinate system η which is constant in time. The aerosols and gas species are evolved in the transport step ($\Phi^{\text{Trans}}_{\Delta t}$) on a geometric height coordinate system z that is computed from the geopotential field and changes over time due to column pressure changes. The vertical advection terms found in Eqs. (2) and (3) are entirely due to pressure-induced grid changes and do not cause particles to be transported across grid cell edges. At every time step the z grid is moved by WRF, and this accounts for vertical advection."

(2.2) WRF already has vertical transport schemes. Why this study uses new and different equations when WRF is coupled?

> The WRF vertical transport scheme operates on scalar quantities such as mass mixing ratios. The method we presented is similar to WRF but includes an additional step where we discretize the equations to finite particle number. This is necessary because WRF-PartMC-MOSAIC uses a particle-resolved approach where individual particles, and not scalars, are transported. Equations are presented in terms of sampling and probabilities as the stochastic approach avoids the computational cost of tracking the exact vertical position of each particle. We added the following to the manuscript:
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> - In Section 3.1, page 7, line 18, we added the following to clearly explain the difference between WRF/WRF-Chem and our model: "This is in contrast to conventional models which transport scalar variables such as mass mixing ratios. Therefore, the discretization process requires an additional step to transport particles."

(2.3) WRF has Asymmetric Convective Model, version 2, (ACM2) to include both an eddy diffusion scheme and the nonlocal scheme to better represent the rise and fall of the convective boundary layer. Has this been considered in this model?

> When we started the work that is presented in our manuscript, the ACM2 scheme was not yet supported in WRF-Chem [Pleim, 2011], and therefore we did not consider it.
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> For simplicity, our current model formulation requires local K theory so a local PBL scheme is required as particles may only move to nearest neighbors within a sub cycle time step. This

constraint could be removed in the future for use with the ACM2 scheme or any other non-local scheme. This will require rewriting the transport equations to determine particle transfer probabilities from any grid cell to any other grid cell such as by using a transilient matrix. For example, the non-local term in ACM2 involves the upward transport of the mass mixing ratio from the lowest grid cell C_1 to all over grid cells above. This would involve sampling for the particle population of the lowest grid to other grid cells above it.

In the future, we will look to include ACM2 because of its good performance, to be consistent with other chemical transport models (e.g., CMAQ), and to allow for additional flexibility in the selection of boundary layer schemes. We made the following comments in the manuscript about PBL schemes:

- In Section 5.1, page 20, line 16, we added to clarify the valid choices of boundary layer schemes: "The presented model formulation requires the use of local boundary layer schemes such as MYJ and Mellor-Yamada-Nakanishi-Niino (MYNN) [Nakanishi and Niino, 2006, 2009]. Non-local schemes such as Asymmetric Convection Model 2 Scheme (ACM2) [Pleim, 2007] may be included in future work."
- In Section 7, page 27, line 14, we added a comment to emphasize that the methods presented in this paper could be extend to include ACM2 in the future: "Potential future model development includes the implementation of other numerical methods for turbulent diffusion, such as higher-order and/or semi-implicit schemes, and non-local boundary layer schemes such as ACM2."

(2.4) The abstract is rather simple. Only what have been done were presented but no results were shown.

We submitted this paper under GMD's "model development paper" category where the main goal is to "describe both the underlying scientific basis and purpose of the model and overview the numerical solutions employed." (see https://www.geosci-model-dev.net/by_ms_types.html). The contribution of this paper therefore consists of the model development in terms of the numerical methods and the verification of the model algorithms, which is the reason why the abstract focuses on these aspects.

(2.5) If aerosol mixing state is used to refer distribution of chemical species. Then, all current models are able to and predicting aerosol mixing state. What makes this study different? Mixing state is better used for how particle components are distributed in each particle, homogeneous, core-shell or else. But it is not discussed in this study.

The reviewer points out an important source of confusion. The uniqueness of the particle-resolved approach used in PartMC-MOSAIC is that we track the composition of individual particles, i.e., we do not need to make an assumption about how chemical species are distributed amongst different particles. We are using the term mixing state in the sense of Winkler [1973] who noted that "the same net composition of an aerosol can be caused by an infinite variety of different internal distributions of the various compounds." In this sense, mixing state is a property of the aerosol *population*, not of individual particles. Traditional aerosol models that use sections or modes are based on the assumption that within one bin or mode all particles are internally mixed, and hence do not resolve (or not fully resolve) mixing state. Importantly, this is distinct from the issue of how the chemical species are arranged *within* the particles (e.g., homogeneous, core-shell, or else). With PartMC-MOSAIC, we do not predict the arrangement of chemical species within the particles.

To clarify this we made the following changes in the paper (page 1, line 17): "For the purposes of this paper we use the term "aerosol mixing state" to refer to the distribution of chemical species across the aerosol *population* [Riemer and West, 2013, Winkler, 1973]. This is distinct

> from the use of the term "mixing state" for the arrangement of components *within* a particle (e.g., homogeneous mixture or core-shell arrangements)."

(2.6) Too much detailed information in sections 2 and 3. They should be greatly reduced by put information to appendix. Very less readers would be interested in the algorithms.

> We agree with this comment and reorganized Sections 2 and 3 to follow the suggestions of the reviewer:
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> - The finite volume discretization derivation (Equations 14–21 in the original manuscript) was moved to Appendix B. These equations are important steps to arrive at the final model expression (Equation 22 in the original manuscript) but may not be of primary interest to readers.
> - We combined Section 3.1.2 and 3.1.3.
> - We moved the stochastic algorithms for transport and dry deposition to Appendix C.

(2.7) Point source emissions are important in vertical distribution calculation of particle? Why this study did not consider that? How would that change the results?

> Point source emissions are indeed important, and will be included in our framework once we extended it to 3D. However, for the 1D column model, our underlying assumption is horizontal homogeneity, and hence we consider emissions at the surface, representing an extended area source, but not a point source. We added the following text to clarify this:
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> - In Section 5.1, page 20, line 5, we added: "The model allows for the inclusion of aerosol emissions within any grid cell in the column and has flexibility in the choice of the parameters of the size distribution as well as the particle composition of the emitted particles."
> - In Section 5.1, page 20, line 12, we added: "We consider this set of simplified surface emissions, as the underlying assumption of the 1D column setup is horizontal homogeneity. "

(2.8) The tested case only shows the concentrations of PM components. It is not clear how mixing state is changed or simulated as the title emphasizes it.

> Our response to this comment is related to the response to comment (2.5). Since we define mixing state as detailed in our response to (2.5), the key figure that illustrates mixing state is Figure 12. This figure shows two-dimensional number distributions in terms of particle diameter and particle BC mass fraction for three different altitudes in the boundary layer. It conveys the information of how BC is distributed over the populations. We see that for a given size (e.g. 100 nm), the particles can have a wide range of different BC mass fractions. It is important to note that we know these distributions for all aerosol components, not only BC. To clarify this, we made the following change to the manuscript:
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> - At the very end of Section 5.3 we added a new final paragraph: "While Figure 12 only shows the BC-diameter distribution of the aerosol, the simulation results contain the full high-dimensional distribution over all constituent species, thus permitting the calculation of any desired mixing state measures or visualizations."

References

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