Responses to Reviewer #1

We thank the reviewer for taking the time to review our paper and for the constructive comments. The page and line numbers that we quote for indicating where we changed the manuscript refer to the revised marked-up version.

(1.1) The proposed scheme is based on interpreting the fluxes coming from a numerical advection scheme as probabilities. This is valid only if the "probability" given by (14) is between 0 and 1. However, this may not always be the case. It seems to me that (14) will usually be very close numerically to the local Courant number. Some advection schemes are stable with Courant numbers greater than 1. This includes the WRF schemes used in this paper. Interestingly, in 1D the same condition that guarantees the probability to be less than 1 ($\frac{\Delta t}{\Delta x} f_{i+1/2} < n_i$) also guarantees positivity preservation. Yet, most advection schemes aren't positivity-preserving without additional limiting. Can the authors comment on this ? In the provided reproducibility notebook for the 1D test case there is code that clips the probability value, but there is no mention of this in the paper. Was this necessary and was a similar limiter used in the other test cases ?

> Thank you for pointing out this issue. In the notebook example for the 1D test case, the clipping is indeed unnessessary since the Courant numbers are sufficiently low (we have chosen 0.4). We removed the corresponding lines of code from the notebook to avoid confusion.
>
> More generally, in the current implementation, we chose a time step that is small enough for the sum of the probabilities remaining smaller than or equal to 1 (i.e., we do not use any clipping). This requires WRF-PartMC to take somewhat smaller time steps than the finite-difference advection in WRF would require.
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> We have made the following changes to the manuscript:
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> - Line 160: "Since probabilities larger than 1 are not meaningful, the time step needs to be chosen such that the probability (14) is less than or equal to 1. As a result, WRF-PartMC may need to take somewhat smaller time steps than required by the finite-difference advection in WRF."
> - Line 170: "The time step should be chosen so that the sum of these probabilities is at most 1."
> - Line 395: "The model time step was set to $\Delta t = 20$ s, ensuring that the sum of the particle cell transfer probabilities does not exceed 1."

(1.2) The authors show that the stochastic transport algorithm injects energy at high spatial frequencies and analyze this process in considerable detail, including an approximate Fourier analysis. This analysis is very similar to von Neumann stability analysis of finite-difference schemes. Based on this the authors say that odd-order advection schemes are preferable, as they damp high spatial frequencies. I am wondering if a stronger statement could be made: that the stochastic algorithm based on even-order energy-conserving advection schemes is unconditionally unstable in a periodic domain, since it leads to unbounded growth of energy? In general, are the stochastic algorithms less stable than their finite-volume base schemes ?

> The reviewer is correct. The even-order energy-conserving is unconditionally unstable and in general, the energy injection will make stochastic methods less stable than the equivalent finite-volume based schemes. The following changes were made:
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> - Line 355: "In general, stochastic methods are less stable than their finite volume counterparts as the stochastic noise injects energy on average. Conservative even-order methods are unconditionally unstable due to this noise injection, because the scheme itself will never damp any of this additional energy."

(1.3) One of the motivations for using the proposed stochastic transport algorithm instead of Lagrangian advection is computational performance. Would it be possible to add to the article some performance numbers showing how much slower the stochastic algorithm is compared to its base finite-volume scheme ?

We agree that there is benefit to adding some general discussion of the computational costs of the various methods, including finite volume, stochastic sampling and particle-tracking. At the same time, we feel that we placed too much emphasis on the computational cost advantages of our methods and too little emphasis on the main motivation for this paper, namely to create a stochastic transport algorithm that closely mimics the finite volume advection scheme (including its numerical diffusion). From our perspective, following the finite volume method closely is the main goal while the computational advantages of the stochastic sampling is more of a potentially helpful incidental benefit. To communicate this more clearly, we made the following changes:

- Beginning at Line 73, we made the following change to better emphasize the goals: "First, a stochastic algorithm can be constructed analogously to the finite volume transport schemes used in numerical weather models and chemical transport models, as we will show in this paper. This is beneficial for direct comparisons of different aerosol representations, which is one of our main motivations for developing particle-resolved aerosol models on the regional scale. Second, stochastic methods are more easily implemented in models that rely on different numerical grid structures, because they are based on the discretizations of the host model on the host grid. Lastly, stochastic methods for transport are computationally less expensive than tracking and updating particle positions throughout the simulation. However, stochastic transport algorithms have the disadvantage of numerical diffusion, similar to finite volume methods. This is in contrast to Lagrangian particle tracking methods that are inherently free of numerical diffusion."
- We added a new section 2.8, beginning at Line 224: "Regarding the computational costs of the finite volume, stochastic sampling, and Lagrangian particle tracking approaches, we consider a domain consisting of N_g grid cells and N_p computational particles per grid cell. The finite volume method, which only depends on the number of grid cells, has a cost $\mathcal{O}(N_\mathrm{g})$. In contrast, the Lagrangian particle tracking and stochastic methods depend on both number of grid cells and the number of particles. Therefore these methods scale as $\mathcal{O}(N_\mathrm{g} \times N_\mathrm{p})$ but the Lagrangian method has a higher cost as each particle must be checked and updated. In contrast, the cost of the stochastic method depends on the number of particles that actually move from one grid cell to another, which is frequently only a small fraction of the total number."

(1.4) Line 157 "However, we now have three different probabilities for each boundary, ..." : If I understood the extension to three dimensions correctly, this sentence can be misleading. Maybe it would be better to say: "However, we now have three different probabilities, one for each boundary, ..."

Thank you for pointing this out. We have improved the wording as suggested:

- Line 170: "However, we now have three different probabilities, one for each boundary, corresponding to the three different directions in which particles can move."

(1.5) Line 161: I think it is not (16), but a multi-dimensional extension of it, that needs to be used in three-dimensional simulations.

Thank you for catching this. It is indeed correct that Equation (16) must be extended to three dimensions by considering all possible fluxes. As a result, we made the following correction:

- Line 173: "Finally, the number of particles in each grid cell is updated by extending Eq. (16) from one dimension (i) to three dimensions (i, j, k)."

(1.6) Section 2.5: Please provide more information on the new monotonic third-order advection scheme. There are many approaches to constructing limiters for advection schemes. If the approach is something standard, like FCT, then I don't think it is necessary to provide every detail, but indicating which method was used and adding a citation would be helpful.

The third-order method is based directly off the limiter used for the fifth-order presented in Skamarock (2006) and further evaluated in Wang et al. (2009). This monotonic advection scheme uses a low-order monotonic flux limiter. We added the following to the manuscript:

- Line 190: "This implementation utilized the existing third-order positive-definite scheme in WRF and applied the same limiter as used in the fifth-order monotonic scheme (Skamarock, 2006; Wang et al., 2009)."

(1.7) Subsection 2.8 feels out of place to me in Section 2, since it details the computation of error metrics in numerical experiments. Maybe put it at the beginning of Section 3 ?

We followed the reviewer's suggestion and shifted the content of Section 2.8 to be at the end of the text that starts Section 3 (just before Section 3.1).

(1.8) In all numerical examples: Please indicate which experiments used the monotonic versions of the schemes and which the unlimited ones.

We followed the reviewer's suggestion. The 1D Python notebook example contains no limiter while both WRF cases use monotonic limiters. We made the following additions to the manuscript to clearly denote this:

- Line 276 (Section 3.1): "Simulation results were produced for first- through sixth-order advection schemes with no limiters applied."
- Line 345 (Section 3.2): "... all simulations were run without limiters."
- Line 376 (Section 3.3): "Simulations were conducted using third- and fifth-order monotonic advection schemes."
- Line 408 (Section 3.4): "Simulations were conducted using third- and fifth-order monotonic advection."

(1.9) Figure 2 caption: "at $T = 2$" should be "at $t = 2$".

The reviewer is correct that it should be time t and not total time T. We have made the following correction:

- In caption of Figure 2: "...the analytical solution for first- to sixth-order advection with varying number of computational particles per grid cell at $t = 2$. "

(1.10) Lines 304-305 "The initial number concentration is given as ...": In subsection 2.6 it is stated that q refers to the mixing ratio. Figures 6 and 7 are also labeled as mixing ratios. I realize that in this simple advection example the values are probably numerically equal. However, a similar issue is present in the subsequent realistic meteorology test case, where the text sometimes refers to the number concentration field, but, according to their labels, the figures are showing mixing ratios. For example, in line 328. It would be good if the language was consistent with the symbols and labels.

The reviewer is correct that often number concentration and mixing ratio are numerically equal when density is uniform (such as in the 1D case and 2D case). However, we have made corrections throughout the manuscript in the cases where mixing ratio is truly the correct term to use.

(1.11) Section 3.4 (WRF meteorology test case) : It would be helpful to provide more information about the setup of this test case. At which geographic location was the computational domain centered ? What was the time step ? What were the boundary conditions ?

We made the following changes to the manuscript to more clearly define the simulation:

- Line 391: "We prescribed an idealized initial condition of particle mixing ratio and gas tracer mixing ratio for the model domain of Northern California."
- Line 393: "The domain comprised $170 \times 160 \times 40$ grid cells, with $\Delta x = \Delta y = 4$ km and Δz increasing logarithmically from an average value of 55 m near the surface to 650 m near the top of the model domain."
- Line 402: "... beginning at 0 UTC on 7 June 2010 using a time step $\Delta t = 20$s."
- Line 403: "Meteorological initial and boundary conditions were based on analyses from the National Center for Environmental Predictions North American Mesoscale (NAM) model."
- Line 406: "Gas and aerosol boundary conditions were prescribed from initial values given in Eq. (26). When flow enters the domain at a boundary grid cell, the prescribed value is applied. Conversely, when flow exits the domain, the boundary grid cell assumes a zero gradient condition, consistent with the host model WRF."

(1.12) (E10): Shouldn't the conditions on the right be $A_k < 0$ and $A_k = 0$ since a in (E7) corresponds to $\exp(A_k)$?

Thank you for catching this. We have made the correction to Equation (E10).

References

Skamarock, W. C.: Positive-definite and monotonic limiters for unrestricted-time-step transport schemes, Mon. Weather Rev., 134, 2241–2250, https://doi.org/10.1175/MWR3170.1, 2006.

Wang, H., Skamarock, W. C., and Feingold, G.: Evaluation of scalar advection schemes in the Advanced Research WRF model using large-eddy simulations of aerosol–cloud interactions, Mon. Weather Rev., 137, 2547–2558, https://doi.org/10.1175/2009MWR2820.1, 2009.