Response to referee comments

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Title: Exploring the Aerosol Activation Properties in Coastal Shallow Convection Using Cloud and Particle-resolving Models

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We thank the reviewers for the comments and suggestions, following which we revised the manuscript. Below are our responses to the reviewers' comments. The text in black is the original comments from reviewers, and our **responses** are the **text in blue**. Some text from the revised manuscript is quoted in this response letter for the reviewers' convenience, and the *quoted text* is in *italic font*.

Referee 1:

The authors have addressed the reviewers' questions well, and the paper is improved in many ways. There are a few issues left that require another round of revisions. They are minor in nature, so I suggest minor revisions.

1. Motivation of this study: in the introduction, the authors now say: "Currently, large-scale meteorological simulations generally employ relatively simple aerosol parameterization methods when considering the contribution of CCN to cloud microphysical processes (Thompson and Eidhammer, 2014; Morrison and Milbrandt, 2011; Hazra et al., 2020), such as directly prescribing aerosol activation rates. These simplifications may introduce errors in the prediction of cloud behavior and associated atmospheric processes. To this end, this study integrates the meteorological Cloud Model 1 (CM1) (Bryan and Fritsch, 2002) with the aerosol evolution model PartMC-MOSAIC for the first time." I agree with you that CCN in large-scale models are represented overly simplified, but this paper does not directly address the impact of these simplifications. Could you add something to the paper that addresses the question how directly prescribing aerosol activation rates would introduce error? (This is even more simplified than using composition-averaged information, which is what the paper does investigate.) This would be very useful for large-scale modelers.

Thank you for your suggestions. We have added further explanations regarding the error mechanisms associated with simplified parameterizations.

The phrase "such as directly prescribing aerosol activation rates" refers to a method for estimating the relationship between CCN number concentration and ambient supersaturation. A widely used approach is the power-law parameterization proposed by Twomey (1959), expressed as $N_{CCN} = C \cdot S^k$, where S

represents supersaturation, and C and k are coefficients derived from observed CCN characteristics, typically prescribed as parameters in simulations. In observational data, particles that do not reach the critical activation diameter are sometimes misidentified as CCN, introducing errors in real-time CCN estimated by the formula. Due to its simplicity, this approach remains widely used to date. However, it undeniably yields coarse CCN estimates that deviate from real-world conditions. This deviation further impacts the calculation of critical properties such as cloud droplet number concentration and droplet radius, which may affect the treatment of cloud water conversion processes in models (Fan et al., 2012). Therefore, improving the accuracy of CCN concentration estimates is essential to reduce model uncertainties. When simulating a case of shallow cumulus clouds, Wang et al. (2025) revised the basic relationship, revealing that the uncorrected power-law parameterization for CCN suppresses precipitation formation and overestimates cloud radiative cooling. Notably, the effectiveness of the correction decreases under high aerosol loading conditions. Additionally, Hazra et al. (2020) compared different microphysics schemes within the WRF model for the same case, highlighting significant variability in results. These simplifications in CCN estimation can lead to errors in hydrometeor predictions, thereby affecting the accuracy of cloud behavior and associated atmospheric processes.

We have expanded the description in this section to improve clarity as suggested. The updated content is provided below.

'Currently, large-scale meteorological simulations typically employ simplified aerosol parameterization methods to represent the contribution of cloud condensation nuclei (CCN) to cloud microphysical processes (Thompson and Eidhammer, 2014; Morrison and Milbrandt, 2011). For instance, a common approach directly relates CCN concentration to ambient supersaturation through power-law parameterization. As proposed by Twomey (1959), the function is expressed as $N_{CCN} = C \cdot S^k$*, where* S *represents supersaturation, and* C *and* k *are coefficients derived from observed CCN characteristics, typically prescribed as parameters in simulations. Due to its simplicity, this approach remains widely used in many microphysical schemes (Hong et al., 2010; Mansell et al., 2010; Morrison et al., 2009). However, it undeniably yields coarse CCN estimates that deviate from real-world conditions. This deviation further impacts the calculation of critical properties such as cloud droplet number and droplet radius, which may affect the cloud-rain conversion processes in models (Fan et al., 2012). When simulating a case of shallow cumulus clouds, Wang et al. (2025) corrected the basic power-law, revealing that the uncorrected power-law parameterization for CCN suppresses precipitation formation and overestimates cloud radiative cooling. Notably, the effectiveness of the correction decreases under high aerosol loading conditions. Comparative studies, such as Hazra et al. (2020), further demonstrate significant variability in results across different microphysical schemes within the WRF model for the same case. The simplifications can affect CCN concentrations, potentially introducing errors in hydrometeor predictions and impacting cloud behavior and related atmospheric processes. Therefore, the accuracy of CCN concentration is essential to reduce model uncertainties. Given the distinct characteristics of convective cloud processes across different vertical regions, the influence of vertical CCN distribution differences on precipitation is also a*

promising topic. Improving the estimation of CCN variation in cloud-forming locations is crucial for enhancing cloud microphysical schemes. To this end, this study integrates the meteorological Cloud Model 1 (CM1) (Bryan and Fritsch, 2002) with the aerosol evolution model PartMC-MOSAIC for the first time. The CM1 is applied to initially investigate the movement characteristics of air parcels under a shallow cumulus convection condition. Key parameters obtained from the CM1 experiments and observations are then introduced into the PartMC-MOSAIC model to investigate the evolution of aerosol populations across different air parcel scenarios. By analyzing key indicators such as hygroscopicity, critical supersaturation, and mixing state index, this study explores the cloud-forming abilities of aerosols within different parcels.'

2. Re: Comment 3: Figure S1: Average over the entire x-y plane not meaningful. Suggest to only average over the core of the cloud? In general, it'd be helpful to visualize the parcel trajectories better, i.e. include a 3D plot of a few example trajectories. I'm not sure how feasible this is, but it would illustrate the purpose (and novelty) of this study well.

Thanks for your comments. Comment 3 raised questions about the cloud water and ice content subplot: the horizontally averaged qc values in the figure might be too small and potentially confusing to readers. As suggested, we have added a subplot that shows the average of cloud water where its value exceeds 10^{-5} kg/kg (i.e., the cloud core average). The revised figure and captions are presented below.

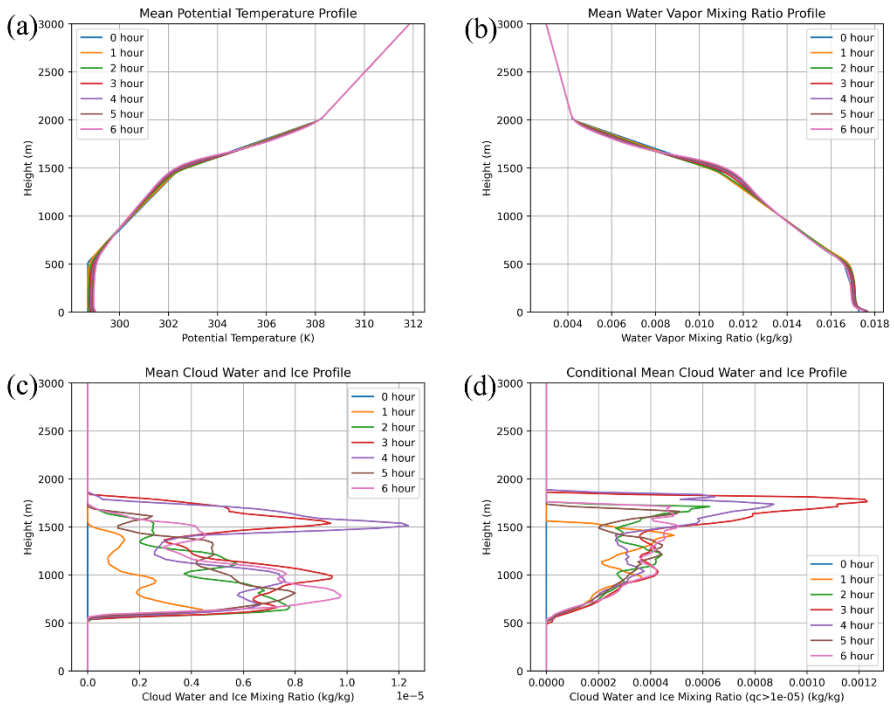

Figure S1. Temporal evolution of horizontal-averaged (a) potential temperature, (b) water vapor mixing ratio, and (c) cloud water and ice mixing ratio with the height in the preliminary CM1 experiment. Panel (d) depicts the conditional mean cloud water and ice mixing ratio over the cloud core (defined as regions where the cloud water mixing ratio exceeds 10^{-5} kg/kg).

We have also added a 3D plot featuring representative trajectory examples in this section. The plot includes both rapidly ascending air parcels (different ascent timings) and a parcel that remains predominantly within the boundary layer. The corresponding figure and modified descriptions are presented below.

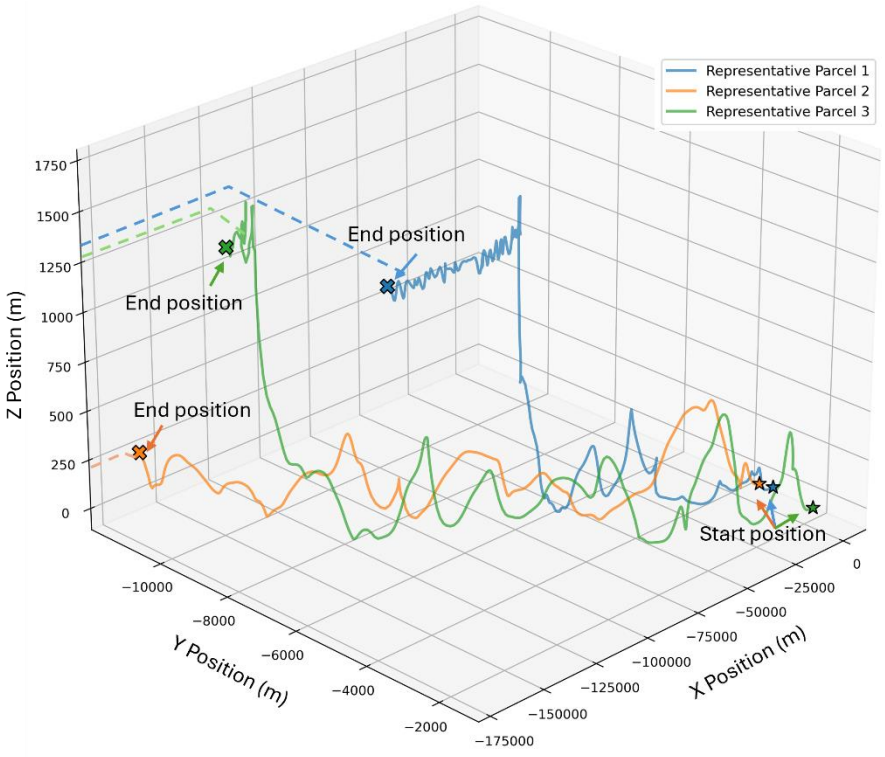

Figure S2. Three-dimensional trajectories of three representative air parcels simulated by the CM1 model. The x and y positions of the parcels have been adjusted to account for the horizontal periodic boundary conditions of the simulation domain, ensuring continuous trajectories across domain boundaries.

'The presentation of Fig. S1 is to illustrate the potential cloud formation heights under the typical shallow cumulus convection conditions. As stated in the main text, approximately one-third of the air parcels ascend to altitudes exceeding 1000 meters, where clouds could form. Figure S2 shows three representative parcel trajectories simulated using CM1. In this case, there is an initial background wind speed of approximately 8 m/s in the -x direction. The x and y positions of the air parcels have been adjusted to eliminate discontinuities arising from the horizontal periodic boundary conditions of the simulation domain. The trajectory plot includes both rapidly ascending parcels (with varying ascent timings) and a parcel that remains predominantly within the boundary layer.'

3. Re: Comment 9: Thanks for adding information about the MERRA-2 dataset. However, it is still not clear to me how the mass concentrations of sulfate, black carbon, organic carbon and sea salt from MERRA are converted into the data that PartMC can use. I believe, you needed to apply some assumptions about size distributions and mixing state. What are these assumptions?

We thank the reviewer for the comments. Based on ground observations, size distributions were reported and aerosol particles were classified into three modes. Lognormal size distribution for particles and internal mixing assumption in every mode were adopted to ensure compatibility with the model.

After calculating the new mass concentrations of species at higher altitudes, we applied the same size distribution and internal mixing assumptions for the three modes as those used at ground level (as described in lines 150 ~ 156). That is, these assumptions and transformation methods were identical for both high-altitude and surface aerosols. As reminded by the comment, we have also revised some descriptions. The relevant content is pasted below:

'For cloud-altitude aerosol background conditions, we extracted the reanalysis aerosol data of Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2, DOI: 10.5067/LTVB4GPCOTK2) in the Hong Kong region. The data of the lowest atmospheric layers (from 880 to 1000 hPa) in summer was processed and the vertical proportionality relationships between the ground-level aerosol species and cloud-altitude species were estimated. Subsequent calculations determined the mass concentrations of different species at elevated altitudes. The same size distribution and mixing state assumptions as those used for ground-level conditions were applied, and the cloud-altitude background aerosol for the simulation was ultimately generated.'

4. Table S1: Instead of "ground fraction"/ "high-altitude fraction", I suggest using "mixing ratio at ground level" and "mixing ratio at elevated altitude" (I would not consider 880 hPa as "high-altitude")

We have revised the descriptions as suggested. Additionally, a detailed explanation of more modifications made to Table S1 has been provided in our response to Question 5.

Following the reviewer's suggestions, we have replaced the term "high altitude" with either "cloud altitude" or "elevated altitude" throughout the manuscript to enhance the precision and clarity of the expression.

5. Furthermore, the following sentences are unclear (in section S5): “For elevated atmospheric conditions, we implemented the ideal gas Clapeyron equation with the following boundary conditions”. There is the ideal gas law, and there is the Clausius-Clapeyron equation. I assume you mean the ideal gas law here, but how did you use it in this context to obtain the numbers in the column for “high-altitude fraction”?

Yes, we quite mean the ideal gas law here. Thank you for raising this issue. Initially, we mistakenly assumed the required ‘gas concentration’ in the model to be mass volume fraction. Thus, we applied the ideal gas law and calculated the ratio of mass concentration between the elevated altitude and the surface, which was incorrect for the input conditions. Upon revisiting the model’s framework, we recognized that it employs the mixing ratio (unit: ppb) for gas species. Reapplying the ideal gas law demonstrated that the volume mixing ratios of the trace gases remain constant with altitude.

To fix this problem, we have revised the elevated-altitude background gas conditions in Scenarios B, C, and D, which marginally influence diffusion processes after ascent. After re-running the simulations and updating the data analyses, we have updated all relevant figures and descriptions accordingly. While the revised figures show minor differences (the comparison is visible in the tracked-changes PDF), these adjustments do not affect the study’s conclusions, as the patterns and quantitative relationships remain virtually unchanged and consistent with our previous results. Additionally, we updated Supplementary S5, including the table stating the background gas conditions and associated explanations. The modified Table S1 and descriptions are presented below:

Table S1. The Background and Emission Conditions for Gases

Gas Species	Symbol	Mixing Ratio of the Background Gas (ppb)	Emissions (nmol m^2 s^{-1})
Nitric oxide	NO	0.1	31.8
Nitrogen dioxide	NO_2	1.0	1.67
Nitric acid	HNO_3	1.0	
Ozone	O_3	50.0	
Hydrogen peroxide	H_2O_2	1.1	
Carbon monoxide	CO	21.0	291.3
Sulfur dioxide	SO_2	0.8	2.51
Ammonia	NH_3	0.5	6.11
Hydrogen chloride	HCl	0.7	
Methane	CH_4	2200.0	
Ethane	C_2H_6	1.0	
Formaldehyde	HCHO	1.2	1.68
Methanol	CH_3OH	0.12	0.28
Methyl hydroperoxide	CH_3OOH	0.5	
Acetaldehyde	ALD_2	1.0	0.68
Paraffin carbon	PAR	2.0	96.0

Acetone	AONE	1.0	1.23
Ethene	ETH	0.2	7.2
Terminal olefin carbons	OLET	0.023	2.42
Internal olefin carbons	OLEI	0.00031	2.42
Toluene	TOL	0.1	4.04
Xylene	XYL	0.1	2.41
Lumped organic nitrate	ONIT	0.1	
Peroxyacetyl nitrate	PAN	0.8	
Higher organic acid	RCOOH	0.2	
Higher organic peroxide	ROOH	0.025	
Isoprene	ISOP	0.5	0.23
Alcohols	ANOL		3.45

'The gaseous species inputs across all simulation scenarios are systematically cataloged in Table S1. These chemical species align with those documented in Zaveri and Peter (1999), while surface background concentrations and emission parameters were adopted from Riemer et al. (2009). For the background gas concentration at the elevated altitude, we assume no additional substances are introduced into the system. Based on the ideal gas law, the volume mixing ratios of these background trace gases remain constant between the surface and elevated altitudes.'

6. Equation (5): Why is the denominator the total aerosol concentration? Usually when defining the error in a quantity, one would divide by the reference value (N_CCN in this case).

Thank you for your question. Dividing N_CCN by the total aerosol concentration allows us to calculate the CCN activation ratio. We aimed to directly compare differences in the CCN activation ratio across different scenarios after performing composition averaging.

Additionally, as discussed in Section 4.2 and illustrated in Figure 6, N_CCN may occasionally equal zero in smaller size groups, which introduces complications when used as a denominator. To maintain consistency in error calculations throughout Chapter 4, we adopted the formula specified in Equation (5).

7. New figure 6: I suggest labelling the x-axis with the size ranges of the bins that the particles were grouped into.

We have revised Figure 6 according to the suggestions provided. The modified figure and its captions are presented below.

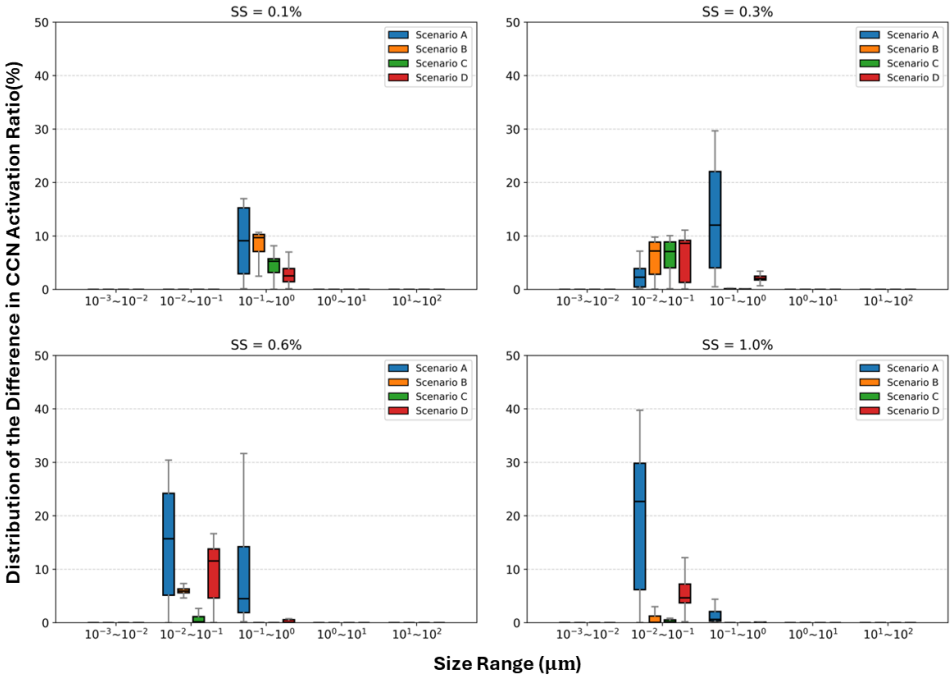

Figure 6. The difference in CCN activation between composition-averaged and particle-resolved approaches within 5 logarithmic size bins (10^{-3} ~ 10^{2} μm) at various environmental supersaturations for four scenarios. Boxplots show 25th–75th percentiles (with median line); data comes from ten-minute intervals over the simulation.

8. Nomenclature: The authors use “hydrophilic” and “hygroscopic” interchangeably. The two terms relate to how substances interact with water, but they’re not exactly interchangeable. When referring to substances like sulfate, ammonium, and nitrate, I suggest calling them hygroscopic species, because this is directly related to water uptake and CCN activity.

We appreciate the reviewer's useful comments. We have replaced all instances of ‘hydrophilic’ with ‘hygroscopic’ in the manuscript and figures. For convenience, the modified Figure 1 is pasted below.

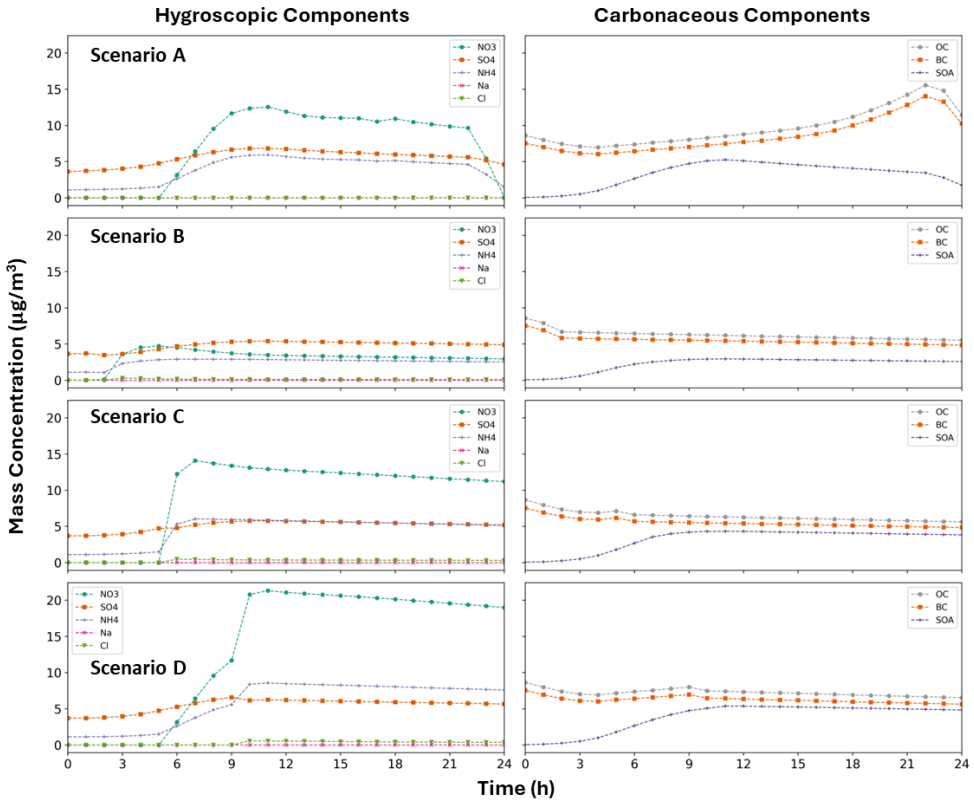

Figure 1. Mass concentration variations over time of hygroscopic and carbonaceous components in Scenario A, B, C, and D. For Scenario A: the parcel remains near the ground; B, C and D: the parcel ascends after 2 hours, 6 hours and 10 hours respectively.

9. Choice of title: I think the title is too generic. In my opinion, this paper is really about different pathways of aerosol aging, depending on how quickly the air parcel is cut off from the emissions at the ground. Suggest something like: "Modeling Aerosol Aging and Cloud Activation During Air Parcel Ascent in Coastal Shallow Convection"

Thank you for your suggestions. After careful consideration, we agree that the title should be more specific, as you noted, to better reflect the shallow convection condition. Additionally, we aim to highlight that this work utilizes the cloud model and the PartMC-MOSAIC model. Accordingly, we have revised the title to: "Exploring Aerosol Activation Properties in Coastal Shallow Convection Using Cloud and Particle-Resolving Models."

10. Last sentence: "Additionally, the mixing state index has the potential to be accounted for in future climate models as it is still challenging to represent some microscale aerosol-cloud interactions and reduce the microphysical uncertainties in large-scale models." I'm not clear on how this is supposed to work.

Thanks for your comments. The mixing state index has been implemented in several climate models. Shen et al.(2024) divided an aerosol mode (previously assumed as internal mixing) into two parts in the Community Atmosphere Model Version 6 (CAM6): one containing BC and one without BC. By

prescribing the ratio of these two parts, the real-time mixing state index χ, can be calculated. They employed a machine learning model to predict the χ in advance and continuously adjusted the mentioned proportion in CAM6 to align the real-time calculated χ with the machine learning predictions. Through this χ-based adjustment process, the new predicted global average ratio of BC-containing particles to BC-free particles was found to be closer to observations (the error is 52% lower than that from traditional CAM6 simulations).

Current global climate models still exhibit biases in representing aerosol mixing states, and improvements in this area could help reduce uncertainties in aerosol-cloud interactions. Further research on the impact of aerosol mixing states on aerosol activation in convective clouds is also of great interest (Shan et al., 2021; Wang et al., 2013).

To enhance the manuscript clarity, we have revised the original text as follows:

'Additionally, the mixing state index can be incorporated into future climate models to better constrain aerosol-cloud interactions. Shen et al.(2024) developed a machine learning model coupled in the Community Atmosphere Model Version 6 (CAM6) that can online correct biases in the mixing states. The χ-guided adjustments provide a more accurate treatment of BC-related processes, including mixing state and coating status. In the future, χ has the potential to dynamically adjust other critical processes, offering a pathway to reduce microphysical uncertainties without requiring explicit microscale resolution.'

New mentioned references:

Shan, Y., Liu, X., Lin, L., Ke, Z., & Lu, Z. (2021). An Improved Representation of Aerosol Wet Removal by Deep Convection and Impacts on Simulated Aerosol Vertical Profiles. *Journal of Geophysical Research: Atmospheres*, *126*(13), e2020JD034173. https://doi.org/10.1029/2020JD034173

Shen, W., Wang, M., Riemer, N., Zheng, Z., Liu, Y., and Dong, X.: Improving BC Mixing State and CCN Activity Representation With Machine Learning in the Community Atmosphere Model Version 6 (CAM6), Journal of Advances in Modeling Earth Systems, 16, e2023MS003889, https://doi.org/10.1029/2023MS003889, 2024.

Wang, H., Easter, R. C., Rasch, P. J., Wang, M., Liu, X., Ghan, S. J., Qian, Y., Yoon, J.-H., Ma, P.-L., & Vinoj, V. (2013). Sensitivity of remote aerosol distributions to representation of cloud–aerosol interactions in a global climate model. *Geoscientific Model Development*, *6*(3), 765–782. https://doi.org/10.5194/gmd-6-765-2013

Referee 2:

1. I would recommend the authors to provide more details about the exchange (of aerosol species an gases) between the parcel and the environment, i.e. due to diffusion in both vertical and horizontal directions (related to my original comments 1.7 and 1.8), especially the numerical values of dilution coefficients input to PartMC-MOSAIC. Besides, the times series of eddy diffusivity of scenarios A-D would be helpful for readers to understand the impact of parcel motion (driven by meteorology) on aging of aerosol particles.

Thank you for your suggestions. In subsection 2.3, we have expanded the explanation of the PartMC model's diffusion algorithm, including separate treatments of vertical and horizontal components. For the vertical diffusion influenced by diurnal mixing height variation and the surface-level horizontal diffusion coefficient, we adopted the specific values from Riemer et al. (2009). The ground coefficient equals 1.5×10^{-5} s^{-1} and the elevated- altitude one equals 3×10^{-6} s^{-1}. The modified description is as follows:

'The PartMC model incorporates a diffusion algorithm to simulate material exchange between the Lagrangian parcel and its surrounding environment, handling both gas and aerosol particle exchanges. Additionally, the algorithm separates horizontal and vertical diffusion coefficients. The vertical diffusion rate depends on both the magnitude and the variation rate of mixing height, with an increase in the mixing height leading to positive values. The horizontal coefficient requires user-defined input and represents the effects of horizontal turbulent diffusion. The diurnal variation of the mixing height and the surface-level horizontal coefficient in this study are consistent with Riemer et al. (2009). When the parcel is at ground level, the horizontal diffusion coefficient is set as 1.5×10^{-5} s^{-1}*. After the parcel rises, the value of mixing height is constant to achieve zero vertical diffusion rate, simulating free tropospheric conditions. The horizontal coefficient at the elevated altitude is set as* 3×10^{-6} s^{-1}*, corresponding to the one-fifth ratio relative to the surface coefficient as described in subsection 2.1. The temporal evolution of horizontal coefficients for Scenarios A~D is documented in* ***Supplementary Information S5****.'*

Besides, the time series of the horizontal diffusion coefficients for Scenarios A-D have been added to Table S2 in the Supplementary Information S5. The specific contents are as follows:

Table S2. The Time Series of the Horizontal Dilution Coefficients in Different Scenarios

Scenario	Time (h)	Coefficient (s^{-1})
A	0 ~ 24	1.5×10^{-5}
B	0 ~ 2	1.5×10^{-5}
B	2 ~ 24	3×10^{-6}
C	0 ~ 6	1.5×10^{-5}

C	6 ~ 24	3×10^{-6}
D	0 ~ 10	1.5×10^{-5}
D	10 ~ 24	3×10^{-6}

'In Table S2, the displayed time aligns with the temporal scale used for result analysis in the main text, excluding the 6-hour spin-up period. In the 6-hour spin-up PartMC-MOSAIC simulation, the horizontal diffusion coefficients were maintained at 1.5 × 10⁻⁵ s⁻¹ since the parcels were near the surface throughout the initialization phase.'