Response to reviewers on egusphere-2026-2776: Sensitivity of dynamic aging on the climate effects of black carbon aerosols over East Asia in summer

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R: We sincerely thank the reviewers for their constructive comments and valuable suggestions on our manuscript. These suggestions have greatly helped us improve the quality and scientific rigor of our manuscript. We have carefully considered and addressed each comment, and corresponding revisions have been made throughout the manuscript. For clarity, the reviewers' comments are presented in black, while our responses are highlighted in blue. We hope that these revisions will meet with reviewers' approval.

RC2: 'Comment on egusphere-2026-2776', Anonymous Referee #2, 01 Aug 2026

Black carbon aerosols (BC) substantially affect regional and global climate and environmental systems through interacting with radiation and cloud. Since BC aging strongly modulates aerosol lifetime and aerosol–climate feedbacks, this study further explores how aging processes alter the climatic impacts of BC over East Asia during summer by implementing a dynamic aging scheme within the regional climate model RegCM-Chem. A set of numerical experiments is conducted to quantify the direct, indirect, and combined (total) climatic effects of BC over the East Asian domain. This work effectively addresses a major uncertainty in current climate simulations. The manuscript is well structured with a solid experimental framework and systematic analyses. The results underscore the vital importance of BC aging representations in regional climate modeling and offer valuable implications for constraining uncertainties in BC climatic effect assessments. Overall, this is a rigorous and scientifically valuable study. I therefore recommend the publication of this manuscript following minor revisions, with detailed comments provided below.

R: Thank you for your instructive comment and suggestions. In response to your helpful recommendations, we have made necessary revisions to our previous manuscript. The detailed revisions are provided below.

1. The manuscript implemented the dynamic BC aging scheme based on the parameterization developed by Fierce et al. (2017) and Ghosh et al. (2021). As noted in the Introduction, several aging schemes have been developed and applied in previous modeling studies. I suggest that the authors should clarify why the schemes is more suitable for the study and strengthen the scientific justification of the methodology.

R: Thank you for your valuable comment. Accurately resolving the evolution of complex particle populations at high spatial and temporal resolutions remains a major challenge for Earth system modeling. In principle, such a treatment would require tracking the size and chemical composition of individual particles throughout the whole process, as done in particle-resolved models (Riemer et al., 2009; Riemer et al., 2019). However, this approach is currently impractical for long-term and large-domain climate simulations due to the extremely high computational and storage costs. Therefore, most climate models necessarily employ simplified aerosol representations. The default aerosol module in RegCM-Chem simplifies the representation by tracking distinct bulk aerosol species (hydrophobic and hydrophilic BC) and uses a fixed *e*-folding aging timescale (1.15 days) to convert hydrophobic BC into hydrophilic BC. Although this approach is computationally efficient, it cannot represent the strong dependence of BC aging on local atmospheric conditions. Since East Asia contains both highly polluted and relatively clean background regions, a spatially and temporally variable aging timescale is necessary for this work. The dynamic aging scheme of Fierce et al. (2017) and Ghosh et al. (2021) provides a physically based yet computationally efficient way to represent this variability. The scheme expresses the aging timescale as a function of coagulation and condensation growth, which are two major microphysical pathways controlling BC aging (Riemer et al., 2010; Fierce et al., 2015; Liu et al., 2020). These controlling variables can be diagnosed from the bulk aerosol fields available in RegCM-Chem, such as aerosol number concentration and sulfate condensation flux. The reliability of the dynamic aging scheme is supported by its derivation from particle-resolved PartMC-MOSAIC benchmark simulations (Fierce et al., 2017), in which the evolution of individual particle mixing states was explicitly solved under a wide range of atmospheric conditions. Its regression-based parameterization has a clear physical basis and has been statistically validated against comprehensive physical simulations, making it suitable for long-term regional climate applications where particle-resolved modeling is computationally prohibitive.

We also agree that other schemes may emphasize different aspects of BC aging, such as particle-resolved composition or more detailed coating chemistry. These approaches are valuable, but they generally require additional prognostic variables or substantially higher computational cost, which are beyond the current RegCM-Chem framework and the scope of this study. Therefore, the selected schemes should be viewed as a practical and physically constrained approach with intermediate complexity rather than as a complete representation of all BC aging processes. These clarifications have been incorporated into the revised manuscript to better explain the reason for selecting these schemes.

Reference

Fierce, L., Riemer, N., and Bond, T. C.: Explaining variance in black carbon's aging timescale, Atmos. Chem. Phys., 15, 3173-3191, https://doi.org10.5194/acp-15-3173-2015, 2015.

Fierce, L., Riemer, N., and Bond, T. C.: Toward reduced representation of mixing state for simulating aerosol effects on climate, Bull. Am. Meteorol. Soc., 98, 971-980, https://doi.org10.1175/bams-d-16-0028.1, 2017.

Ghosh, S., Riemer, N., Giuliani, G., Giorgi, F., Ganguly, D., and Dey, S.: Sensitivity of Carbonaceous Aerosol Properties to the Implementation of a Dynamic Aging Parameterization in the Regional Climate Model RegCM, J. Geophys. Res.: Atmos., 126, https://doi.org10.1029/2020jd033613, 2021.

Liu, D., He, C., Schwarz, J. P., and Wang, X.: Lifecycle of light-absorbing carbonaceous aerosols in the atmosphere, npj Clim. Atmos. Sci., 3, https://doi.org10.1038/s41612-020-00145-8, 2020.

Riemer, N., West, M., Zaveri, R., and Easter, R.: Estimating black carbon aging time-scales with a particle-resolved aerosol model, J. Aerosol Sci., 41, 143-158, https://doi.org10.1016/j.jaerosci.2009.08.009, 2010.

Riemer, N., West, M., Zaveri, R. A., and Easter, R. C.: Simulating the evolution of soot mixing state with a particle-resolved aerosol model, Journal of Geophysical Research-Atmospheres, 114, https://doi.org10.1029/2008jd011073, 2009.

Riemer, N., Ault, A. P., West, M., Craig, R. L., and Curtis, J. H.: Aerosol Mixing State: Measurements, Modeling, and Impacts, Rev. Geophys., 57, 187-249, https://doi.org10.1029/2018rg000615, 2019.

2. The reliability of the adopted dynamic aging scheme requires further statement. In Eq. (2), the two parameters (kcond and kcoag) related to condensation and coagulation processes appear to be prescribed constants. The authors should provide more details regarding how these parameters were derived and whether their applicability under East Asian atmospheric conditions has been evaluated.

R: Thank you for your comment. The two parameters (k_{cond} and k_{coag}) used in this study are derived from a rigorous statistical regression based on benchmark simulations with an advanced particle-resolved aerosol model (Fierce et al., 2017).

Specifically, Fierce et al. (2017) used the particle-resolved model PartMC-MOSAIC as a benchmark framework, in which the exact chemical composition of individual aerosol particles was explicitly tracked (about 14000 populations, each containing 5000 particles), together with multiple microphysical processes. The concept of τ_{mix} represents the characteristic *e*-folding timescale over which the error between the true particle-resolved state and a simplified fully mixed approximation decays with time. To construct this parameterization, Latin Hypercube Sampling was employed to vary 28 input parameters across 100 diverse sensitivity scenarios, spanning extreme environmental regimes from heavily polluted megacities to pristine Arctic conditions. The results revealed that τ_{mix} is fundamentally constrained by two independent atmospheric control variables, namely the condensational growth rate (I_{cond}) and the overall aerosol number concentration (*N*). The regression coefficients (k_{cond} and k_{coag}) were rigorously obtained from separate condensation-only and coagulation-only simulations and subsequently validated against the full-physics simulations. This first-order decay model captures the explicit physical decay mechanism with exceptionally high explanatory power, yielding an R^2 of 85% for the variance in the mixing-state metrics across the entire ensemble. Additionally, a 60%/40% training-testing data split was used to avoid the overfitting. Therefore, this scheme has both a clear physical basis and statistical validation, which has also been applied in global and regional modeling studies (Ghosh et al., 2021; Fierce et al., 2025). We apologize for not providing these essential details in our manuscript to fully substantiate the reliability of the dynamic aging scheme. We have comprehensively integrated these descriptions in the revised manuscript.

Reference

Fierce, L., Riemer, N., and Bond, T. C.: Toward reduced representation of mixing state for simulating aerosol effects on climate, Bull. Am. Meteorol. Soc., 98, 971-980, https://doi.org10.1175/bams-d-16-0028.1, 2017.

Fierce, L., Li, Y., Feng, Y., Riemer, N., Schutgens, N. A. J., Aiken, A. C., Dubey, M. K., Ma, P.-L., and Wuebbles, D.: Constraining Black Carbon Aging in Global Models to Reflect Timescales for Internal Mixing, J. Adv. Model. Earth Syst., 17, https://doi.org10.1029/2024ms004471, 2025.

Ghosh, S., Riemer, N., Giuliani, G., Giorgi, F., Ganguly, D., and Dey, S.: Sensitivity of Carbonaceous Aerosol Properties to the Implementation of a Dynamic Aging Parameterization in the Regional Climate Model RegCM, J. Geophys. Res.: Atmos., 126, https://doi.org10.1029/2020jd033613, 2021.

3. The study evaluates the simulated BC concentrations under the default aging scheme and the dynamic aging scheme (Exp. 4 and Exp. 8) and shows the comparison with observations in Fig. S1. It seems that introducing the dynamic aging scheme leads to only limited improvement in BC concentration simulations and the authors also acknowledge this. However, considering that improving BC aging timescales is one of the motivations in this study, the authors should provide a more comprehensive evaluation of simulated BC using statistical metrics both spatially and temporally.

R: Thank you for your constructive comment. We have added a more comprehensive evaluation of simulated BC using statistical metrics both spatially and temporally in the Section 3.1

First, we have quantitatively evaluated the surface BC concentrations simulated by both Exp. 4 and Exp. 8 against observations from 33 CBNET sites during 2012–2018. Figure S1 shows the observed and simulated annual mean BC surface concentrations and their decreasing trends, and the corresponding statistical metrics are summarized in Table S1. The model reasonably reproduces the distribution of BC surface concentration, with correlation coefficients of 0.97 and 0.70 in the temporal and spatial dimensions, respectively. Nevertheless, the model still substantially underestimates BC surface concentration. This underestimation is consistent with the findings of most previous climate modeling studies, which have generally reported negative biases of approximately 30–60% in simulated surface BC concentrations across China (Li et al., 2016; Yang et al., 2017; Fang et al., 2020; Liu et al., 2022). Compared with Exp. 4, Exp. 8 slightly reduces the temporal RMSE and NMB and better captures the declining trend under the context of emission reductions, whereas the influence in the spatial distribution is not significant. These results indicate that although the dynamic aging treatment modestly reduces the overall negative bias in simulated BC concentration to some extent, this improvement remains limited, consistent with the findings of Ghosh et al. (2021) and Shen et al. (2023). This bias may stem from other processes in the climate model, such as grid resolution, dry and wet deposition, and aerosol transport (Wang et al., 2013; Liu et al., 2016).

Additionally, we have added an equivalent quantitative evaluation of BC AOD from Exp. 4 and Exp. 8 against MERRA-2 AOD. The results show that, compared with the relatively modest improvement in BC concentration, Exp. 8 more substantially reduces the model underestimation of BC AOD (Table S1), although the low bias still remains. This performance reflects the physically consistent response of BC optical properties to dynamic aging. As shown in Fig. S2, accelerated aging increases the fraction of hydrophilic BC, which is characterized by a larger extinction coefficient and therefore makes a greater contribution to AOD. The associated mechanisms are discussed in detail in Section 3.4.

We have also evaluated the ability of the dynamic aging scheme to represent the BC aging timescale using evidence from other observational and modeling studies, and the simulated aging timescale was found to be within a reasonable range in Section 3.2. Overall, these analyses provide additional support for the physical consistency of the dynamic aging scheme. The corresponding descriptions and discussions have been revised accordingly in the manuscript and Supplement.

Table S2 in the Supplement. Statistical metrics for the temporal and spatial evaluation of simulated surface BC concentrations and AOD from Exps. 4 and 8 against reference datasets. Observed surface

BC concentrations are derived from Shen et al. (2023), whereas the AOD reference is obtained from the MERRA-2 reanalysis. Therefore, the AOD comparison should be regarded as a reanalysis-based evaluation rather than an independent observational validation. R, RMSE and NMB denote the correlation coefficient, root-mean-square error and normalized mean bias, respectively.

Variables	**Experiment**	**Temporal**			**Spatial**		
		R	RMSE	NMB	R	RMSE	NMB
Surface concentration	Exp. 4	0.97	0.69	−23%	0.70	1.71	−24%
	Exp. 8	0.97	0.65	−21%	0.70	1.73	−22%
AOD	Exp. 4	0.95	0.014	−49%	0.96	0.016	−50%
	Exp. 8	0.95	0.012	−42%	0.96	0.013	−43%

Reference

Fang, C., Zhu, B., Pan, C., Yun, X., Ding, D., and Tao, S.: Regional and Sectoral Sources for Black Carbon Over South China in Spring and Their Sensitivity to East Asian Summer Monsoon Onset, J. Geophys. Res.: Atmos., 125, https://doi.org10.1029/2020jd033219, 2020.

Ghosh, S., Riemer, N., Giuliani, G., Giorgi, F., Ganguly, D., and Dey, S.: Sensitivity of Carbonaceous Aerosol Properties to the Implementation of a Dynamic Aging Parameterization in the Regional Climate Model RegCM, J. Geophys. Res.: Atmos., 126, https://doi.org10.1029/2020jd033613, 2021.

Li, K., Liao, H., Mao, Y., and Ridley, D. A.: Source sector and region contributions to concentration and direct radiative forcing of black carbon in China, Atmos. Environ., 124, 351-366, https://doi.org10.1016/j.atmosenv.2015.06.014, 2016.

Liu, X., Ma, P. L., Wang, H., Tilmes, S., Singh, B., Easter, R. C., Ghan, S. J., and Rasch, P. J.: Description and evaluation of a new four-mode version of the Modal Aerosol Module (MAM4) within version 5.3 of the Community Atmosphere Model, Geosci. Model Dev., 9, 505-522, https://doi.org10.5194/gmd-9-505-2016, 2016.

Liu, Y., Wang, M., Qian, Y., and Ding, A.: A Strong Anthropogenic Black Carbon Forcing Constrained by Pollution Trends Over China, Geophys. Res. Lett., 49, https://doi.org10.1029/2022gl098965, 2022.

Shen, W., Wang, M., Liu, Y., Dong, X., Zhao, D., Yue, M., Tian, P., and Ding, D.: Evaluating BC Aging Processes in the Community Atmosphere Model Version 6 (CAM6), J. Geophys. Res.: Atmos., 128, https://doi.org10.1029/2022jd037427, 2023.

Wang, H., Easter, R. C., Rasch, P. J., Wang, M., Liu, X., Ghan, S. J., Qian, Y., Yoon, J. H., Ma, P. L., and Vinoj, V.: Sensitivity of remote aerosol distributions to representation of cloud-aerosol interactions in a global climate model, Geosci. Model Dev., 6, 765-782, https://doi.org10.5194/gmd-6-765-2013, 2013.

Yang, Y., Wang, H., Smith, S. J., Ma, P.-L., and Rasch, P. J.: Source attribution of black carbon and its direct radiative forcing in China, Atmos. Chem. Phys., 17, 4319-4336, https://doi.org10.5194/acp-17-4319-2017, 2017.

4. The impacts of BC aging on aerosol concentration, column burden, optical properties and cloud activation capability strongly depend on how hydrophobic and hydrophilic BC are represented in the model. Detailed information about the prescribed parameters of these two BC categories should be provided.

R: Thank you for your suggestion. Accordingly, we have added Table 1 to the Methods section to summarize the physical and optical parameters prescribed for the two BC tracers in this study.

Table 1 in the revised manuscript. Physical and optical parameters of hydrophobic and hydrophilic BC used in this study. r_0 and s are the geometric mean diameter and standard deviation of the particle lognormal size distribution. D, ρ and α are the characteristic effective diameter, density and hygroscopic growth factor, respectively.

Tracers	r_0 (μm)	s	D (μm)	ρ (g cm^{-3})	Refractive index	α	Solubility	Hygroscopicity
Hydrophobic BC	0.0118	1.7	0.05	1.5	1.87 + 0.569i	0	0.05	5.0×10^{-7}
Hydrophilic BC	0.03	1.9	0.17	1.5	1.87 + 0.569i	0.25	0.95	0.22

5. More detail on the statistical testing methodology would strengthen confidence in the reported significance of the results (e.g., treatment of temporal autocorrelation and effective sample size).

R: Thank you for your important comment. We have clarified that the significance analysis is assessed using a two-sided paired Student's t-test based on the season summer means of each year (n = 14). Specifically, the summer-mean difference between the two experiments was calculated separately for each year, and the resulting annual difference series was tested against a null mean of zero.

We also agree that temporal autocorrelation may reduce the number of independent samples and lead to an overestimation of statistical significance if the original sample size is used directly. Therefore, the lag-1 autocorrelation coefficient (r_1) is calculated for the time series of the paired annual differences. The effective sample size is estimated following (Zwiers and Vonstorch, 1995):

$$n_{eff} = n\frac{1 - r_1}{1 + r_1},$$

where n is the original sample size (14 years) and r_1 is the lag-1 autocorrelation coefficient. The calculated n_{eff} is subsequently used to adjust the degrees of freedom ($df = n_{eff} - 1$), and to determine the corresponding critical t-value for the two-sided significance test.

Reference

Zwiers, F. W. and Vonstorch, H.: Taking serial correlation into account in tests of the mean, J. Clim., 8, 336-351, https://doi.org10.1175/1520-0442(1995)008<0336:Tsciai>2.0.Co;2, 1995.

6. Eq. (4), how the sulfate condensation flux is addressed in the model?

R: Thank you for your helpful comment. In RegCM-Chem, the sulfate condensation flux is represented as the local mass production rate of secondary sulfate aerosol. Specifically, sulfate formation in the model is mainly driven by the oxidation of SO_2 following the sulfate chemistry scheme of Qian et al. (2001). The sulfate mass newly formed during each chemistry time step is treated as the condensable secondary aerosol mass that contributes to aerosol growth. Therefore, the condensation mass flux used in the dynamic aging scheme is diagnosed from the simulated sulfate production tendency. We have revised the manuscript to clarify that the condensation mass flux used here refers to the sulfate production rate associated with SO_2 oxidation in RegCM-Chem.

Reference

Qian, Y., Giorgi, F., Huang, Y., Chameides, W., and Luo, C.: Regional simulation of anthropogenic sulfur over East Asia and its sensitivity to model parameters, Tellus Series B-Chemical and Physical Meteorology, 53, 171-191, https://doi.org10.1034/j.1600-0889.2001.d01-14.x, 2001.

7. Section 3.4, please explicitly state what baseline state the radiative forcing in this study is compared against (e.g., relative to preindustrial era?).

R: Thank you for your comment. In the revised manuscript, we have supplemented the explanation that the radiative forcing calculated is relative to zero BC effect in this study.

8. Some writing errors. Line 296, “hotpots” should be “hotspots”. Line 305, “polluter” should be “polluted”. Line 644, “aera” should be “area”.

Corrected