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

Peng Gao et al.

**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.

RC1: 'Comment on egusphere-2026-2776', Anonymous Referee #1, 22 Jul 2026

**Synopsis**

The manuscript "*Sensitivity of dynamic aging on the climate effects of black carbon aerosols over East Asia in summer*" by Gao et al. couples a Fierce et al. (2017)/Ghosh et al. (2021)-style dynamic BC aging parameterization (condensation + coagulation dependent e-folding timescale) into RegCM-Chem, replacing the conventional fixed 1.15-day aging timescale, and uses an 8-experiment factorial design to isolate the direct, indirect, and total climate effects of BC over East Asia in summer, under both aging schemes. The core findings, shorter aging timescales in polluted regions (<10 h in the NCP/Sichuan Basin), a modest regional-mean decrease in BC column burden (-0.12 mg $m^{-2}$) driven by enhanced wet removal of hydrophilic BC, but a local increase in surface concentration/AOD from weakened dry deposition, and downstream effects on ERF, circulation, and precipitation are physically coherent and represent a genuine advance over the fixed-timescale treatment used in most regional/global models. The topic is well suited for ACP, the writing is generally clear, and the experimental design (Table 1) is sensible wau to decompose direct/indirect/total effects with and without dynamic aging.

However, several important methodological and interpretive issues require further attention. Most importantly, the combined climate responses are strongly non-additive relative to the separately diagnosed direct and indirect responses, but the magnitude and physical origin of this interaction are not quantified. Additional concerns relate to the externally mixed optical representation of aging, the sensitivity of BC–cloud effects to assumed hygroscopicity and clean-background conditions, the treatment of emissions over the full domain, the evaluation of the dynamic-aging scheme, and the statistical significance testing. I therefore recommend major revision.

**R:** Thank you for your comprehensive review of our manuscript. We appreciate the opportunity to enhance the quality of our work based on your insightful feedback. We understand the necessity of a major revision and are committed to addressing each of your concerns meticulously. Detailed point-by-point responses to the specific issues are provided below.

**Major Comments**

1. **The non-additivity of the direct, indirect, and total climate responses requires explicit quantification and mechanistic analysis.**

The experimental design provides separate estimates of the BC-direct, BC-indirect, and combined direct+indirect responses. However, Table 2 shows that the combined response differs substantially from the sum of the separately diagnosed direct and indirect responses. For example, in Table 2 (East Asia column): Dyn_Dir TOA ERF = +3.60 W $m^{-2}$, Dyn_Ind TOA ERF = −0.577 W $m^{-2}$, sum = +3.02 W $m^{-2}$, but Dyn_Tot = +0.901 W $m^{-2}$ — the "total" is less than a third of the naive sum. At the surface the mismatch is worse: Dyn_Dir + Dyn_Ind = −0.676 W $m^{-2}$, but Dyn_Tot = −2.68 W $m^{-2}$ (roughly 4× larger in magnitude). The same pattern recurs in T2m and precipitation in Table 2 (e.g., NC precipitation: Dyn_Dir −0.0108, Dyn_Ind −0.145, sum ≈ −0.156, vs. Dyn_Tot −0.0917 - sign-consistent but magnitude off by ~40%; EA T2m: Dyn_Dir +0.0285, Dyn_Ind +0.0100, sum +0.0385, vs. Dyn_Tot −0.0373, a sign flip). Although the manuscript qualitatively relates this non-additivity to climate feedbacks, circulation changes, and cloud adjustments in several sections, the magnitude and physical origin of the interaction are not quantified.

Given that pronounced nonlinearity is one of the manuscript's principal conclusions, the authors should provide a more explicit physical diagnosis of why the combined simulation produces such strong cancellation or amplification. For example, does the cloud suppression associated with the BC-radiation interaction modify circulation and moisture transport in a way that changes the BC-cloud response when both processes are activated simultaneously? Given the factorial experimental design, the nonlinear interaction term can be calculated explicitly as:

$$I\_Dyn = Dyn\_tot - Dyn\_dir - Dyn\_ind$$

or, using the original experiments,

$$I\_Dyn = Exp.8 - Exp.6 - Exp.7 + Exp.5.$$

Similarly, for the default-aging experiments:

$$I\_Def = Def\_tot - Def\_dir - Def\_ind = Exp.4 - Exp.2 - Exp.3 + Exp.1.$$

Based on the East Asian means in Table 2, this interaction residual is approximately -2.12 W $m^{-2}$ at the TOA, -2.00 W $m^{-2}$ at the surface, −0.0758 K for T2m, and +0.103 mm $day^{-1}$ for precipitation. These residuals are sufficiently large that they should be diagnosed directly rather than discussed only qualitatively.

Please calculate the interaction term for both the default- and dynamic-aging experiment families and report its spatial distribution, regional means, interannual variability, and statistical significance. It would also be useful to identify whether the interaction is primarily associated with cloud-fraction changes, cloud optical depth, semi-direct adjustments, circulation responses, or moisture-transport feedbacks. The manuscript should clarify that the "direct" and "indirect" experiments represent isolated pathway sensitivities and should not be interpreted as additive components of the total response.

This also adds another query about the semi direct effects, how do the author account for the fact that this heating and stability if drives away water vapor, which in turn will reduce clouds as well as change the size of the coated BC particles, and in turn change their heating rate? If not, can they make some estimate of this effect? If so, how do they calculate this loop?

Finally, please compare the magnitude of the nonlinear residual with those reported in the cited studies of Zhuang et al. (2019), Chen et al. (2020), and Gao et al. (2024). This would help establish

whether the magnitude found here is typical of previous regional simulations or represents a particularly strong and potentially novel interaction.

**R:** Thank you for your constructive comment. We agree that the pronounced non-additivity among the direct, indirect and total climate responses should be explicitly quantified. Following your comment, we have therefore introduced the nonlinear interaction terms for both the dynamic and default aging experiments in the revised manuscript, which are defined as:

$$\mathrm{Dyn\_I} = \mathrm{Dyn\_tot} - \mathrm{Dyn\_dir} - \mathrm{Dyn\_ind},$$

and

$$\mathrm{Def\_I} = \mathrm{Def\_tot} - \mathrm{Def\_dir} - \mathrm{Def\_ind}.$$

A two-sided paired Student's *t*-test is performed between the total response and the sum of the isolated BC–radiation and BC–cloud interaction responses based on the summer means for each individual year. The regional means of the nonlinear interaction terms for the dynamic and default aging experiments have been added to Table 3. The spatial distributions of the Dyn_I and Def_I in TOA and surface ERF, CF, wind field, specific humidity, $T_{2m}$ and precipitation are further presented in Figure 11 of the revised manuscript and Figure S4 in the Supplement. Significant nonlinear residuals in major climate variables over East Asia are generally consistent with the changes in circulation and cloud cover. For the dynamic aging experiments, this interaction enhances moisture convergence and CF over northeastern China (Fig. 11c and d), leading to substantial negative residuals in ERF ($-2.12$ W m$^{-2}$ at the TOA and $-2.00$ W m$^{-2}$ at the surface) and $T_{2m}$ ($-0.076$ K) over East Asia (Table 3). In southern China, the anomalous cyclonic flow is conducive to cloud formation. Meanwhile, the upward motion induced by BC–radiation interactions may also promote increases in $N_d$ and strengthen the CF response (Figs. 7c and 11c). These coupled moisture and cloud adjustments subsequently contribute to increased precipitation over most areas of East Asia, resulting in the Dyn_I of 0.10 mm day$^{-1}$ in precipitation (Fig. 11). Similar relationships are also evident for Def_I. For example, opposite changes in CF over northern and southern China correspond to different signs in ERF and surface temperature residuals (Fig S4). Over the low-latitude South China Sea, the reduction in water vapor associated with descending airflow weakens atmospheric absorption of solar shortwave radiation during summer (Kim et al., 2022; Harris et al., 2025), thereby increasing the radiative forcing at the sea surface and contributing to reduced precipitation there. These results indicate that the pronounced nonlinear residuals arise primarily from coupled circulation–moisture–cloud adjustments (Sadiq et al., 2015).

In the experiment considering only BC–radiation interactions, BC direct heating induces thermodynamic and semi-direct cloud responses, whereas in the experiment for only BC–cloud interactions, perturbations to cloud microphysical processes by BC trigger adjustments in cloud radiation and dynamics. When the two pathways are activated simultaneously, processes that are separated in the individual pathway experiments become dynamically coupled through complex climate and aerosol feedbacks, and their combined responses cannot be reconstructed as a linear sum. Therefore, the results diagnosed from the BC direct or indirect effects are interpreted as sensitivities of isolated pathways rather than as additive components of the combined climate responses in this study. Strongly nonlinear climate responses have also been reported in previous studies (Zhuang et al., 2019; Chen et al., 2020; Gao et al., 2024; Gao et al., 2025), as aerosol-induced changes in atmospheric circulation and clouds can feed back

onto the initial radiative perturbation, although their experimental designs differ from that employed here. Chen et al. (2020) and Zhuang et al. (2019) both found that the combined direct effect of BC emissions from multiple source regions or sectors could not be reproduced by a simple linear summation of the effects caused by individual source region or sector. Gao et al. (2024) and Gao et al. (2025) demonstrated that regional climate responses to future aerosol emission reductions also exhibited pronounced nonlinear behavior. Therefore, different from source-region contributions or emission changes, the nonlinearity reported here is more appropriately represented as a strong pathway-dependent interaction (coupling between BC–radiation and BC–cloud interactions) within the summertime East Asian climate system. Notably, the magnitude of this interaction can be comparable to, or even exceed, isolated pathway responses at the regional scale (Table 3). These revisions have been incorporated into Table 3, Figs. 11 and S4, and the corresponding discussion in the revised manuscript.

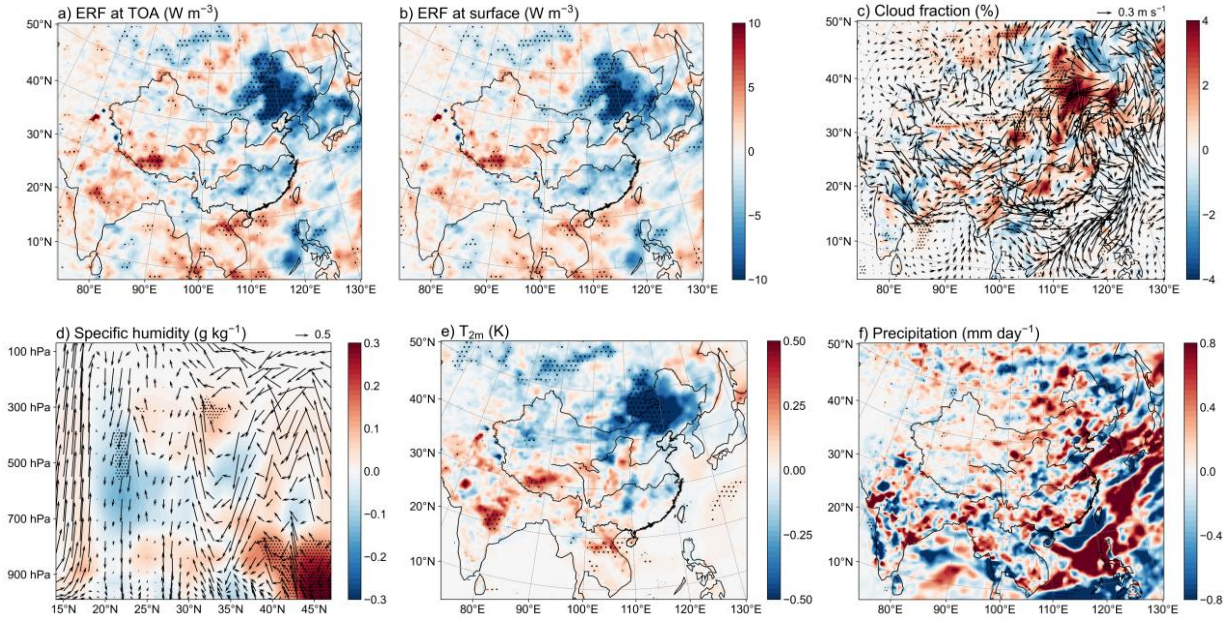


Figure 11 in the revised manuscript. Spatial distributions of the nonlinear interaction term under the dynamic aging scheme (Dyn_I) for the BC effective radiative forcing (ERF) at TOA and surface (a and b, W $m^{-2}$), wind field (arrow, m $s^{-1}$) and cloud fraction (shaded, %) (c) near 850 hPa, meridional circulation (arrow) and specific humidity (shaded, g $kg^{-1}$) (d) in the altitude–latitude section averaged from 112° to 122°E, surface temperature at 2 m ($T_{2m}$) (e, K) and precipitation (f, mm $day^{-1}$) over East Asia in summer during 2008–2021. Black-dotted regions denote statistically significant differences at the 90% confidence level based on a two-sided paired Student's *t*-test.

Regarding the feedback loop between BC direct heating and the meteorological fields, these processes are represented interactively during the simulations in RegCM-Chem. For example, BC-induced changes in atmospheric stability, circulation and precipitation can modify the transport and concentration of aerosol species and their precursors. These changes in turn affect the BC aging timescale that depends on the sulfate condensation rate and the total aerosol number concentration. The resulting redistribution between hydrophobic and hydrophilic BC further influence BC optical properties and

consequently the radiative heating. In addition, changes in relative humidity (RH) directly affect aerosol extinction through the species-dependent hygroscopic growth factor, $(1 - RH)^{\alpha}$, used in the AOD calculation (Section 2.3 in the revised manuscript). However, these feedbacks are represented online within the coupled RegCM-Chem framework rather than being prescribed independently. A rigorous separation and quantification of this individual feedback pathway would require additional targeted sensitivity experiments, which are beyond the scope of the present study. Nevertheless, we agree that explicitly quantifying this feedback loop would be valuable and consider it an important direction for future work.

Reference

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Gao, P., Gao, Y., Zhou, Y., Cao, H., Hu, Y., Li, S., Liang, S., Wang, T., Xie, M., Li, M., and Zhuang, B.: Changes in the Direct Climate Effect of Black Carbon Aerosols in East Asia Under the "Dual Carbon" Goal of China, J. Geophys. Res.: Atmos., 129, e2024JD040874, https://doi.org10.1029/2024jd040874, 2024.

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Sadiq, M., Tao, W., Liu, J., and Tao, S.: Air quality and climate responses to anthropogenic black carbon emission changes from East Asia, North America and Europe, Atmos. Environ., 120, 262-276, https://doi.org10.1016/j.atmosenv.2015.07.001, 2015.

Zhuang, B. L., Chen, H. M., Li, S., Wang, T. J., Liu, J., Zhang, L. J., Liu, H. N., Xie, M., Chen, P. L., Li, M. M., and Zhao, M.: The direct effects of black carbon aerosols from different source sectors in East Asia in summer, Clim. Dyn., 53, 5293-5310, https://doi.org10.1007/s00382-019-04863-5, 2019.

**2. The relationship between dynamic aging, external mixing, and BC optical enhancement needs clearer definition.**

The Introduction emphasizes that aging can lead to internal mixing and coating-driven absorption enhancement through the lensing effect, noting that this process is important for BC optical properties and direct radiative forcing. However, Section 2.1 states that the aerosol tracers are treated as externally mixed, while Eq. (5) calculates AOD using prescribed tracer-specific mass-extinction

coefficients and hygroscopic growth factors.

It is therefore unclear to what extent the implemented dynamic-aging scheme represents the optical consequences of aging. Please clarify precisely what changes when BC is transferred from the hydrophobic to the hydrophilic tracer in the radiation calculation. In particular:

- Are different mass-extinction, mass-absorption, single-scattering-albedo, or asymmetry parameters assigned to hydrophobic and hydrophilic BC?
- Does the prescribed optical coefficient for hydrophilic BC implicitly include any coating-related absorption enhancement?
- Does aging affect absorption explicitly, or does it affect only tracer abundance, particle size, hygroscopic growth, deposition, and cloud activation?

If coating-dependent absorption enhancement is not represented explicitly, the manuscript should state more clearly that the current implementation captures the bulk partitioning, deposition, hygroscopicity, and cloud-activation consequences of aging, but not the continuous evolution of BC mixing state, coating morphology, or lensing enhancement. Although this limitation is briefly acknowledged in the concluding discussion of future model development, its possible implications for the simulated BC AOD, IDRF, and direct ERF should be discussed more explicitly.

Missing absorption enhancement may contribute to an underestimate of BC optical depth or radiative forcing for a given BC mass, but it should be distinguished from the separate underestimate of surface BC mass concentration, which is controlled primarily by emissions, transport, mixing, and removal processes.

**R:** Thank you for your valuable suggestions. Hydrophobic and hydrophilic BC are assigned different extinction coefficients and single-scattering albedos in this study. To account for coating-related absorption enhancement due to aging, hydrophilic BC is implicitly assigned a larger characteristic diameter and extinction coefficient for solar radiation. Therefore, aging also has a pronounced impact on BC absorption.

In the RegCM-Chem model, the treatment of external mixing means that hydrophobic and hydrophilic BC tracers, as well as other aerosol species, are considered independent aerosol populations in both concentration tracking and radiation calculations, which does not resolve the continuous evolution of the mixing state of individual BC particles. Nevertheless, the optical consequences of BC aging are represented implicitly through the distinct effective physical properties prescribed for the two BC tracers. We have added Table 1 to the Method section to summarize these parameters prescribed for hydrophobic and hydrophilic BC.

Specifically, when BC are transferred from the hydrophobic to the hydrophilic state through the dynamic aging scheme, their size distribution shifts toward larger particle, accompanied by increases in the effective diameter and extinction coefficient. In addition, different single-scattering albedos (SSA) are used for hydrophobic and hydrophilic BC. For hydrophilic BC, its extinction coefficient calculation relative to hydrophobic BC further considers an absorption humidification factor associated with aging (Huang et al., 2007). Specifically, in the shortwave spectral range, the extinction coefficient of hydrophilic BC is approximately 1.3 times that of hydrophobic BC (Solmon et al., 2006). Therefore, absorption enhancement during BC aging is partially represented in the prescribed optical treatment of

hydrophilic BC. The increase in solubility and hygroscopicity also represents the greater wet removal and cloud activation abilities of aged BC. Overall, the current implementation captures the bulk transition of BC from a freshly emitted, weakly hygroscopic state to a more hygroscopic and optically enhanced state. The above statement has been revised accordingly in the manuscript.

Table 1 in the revised manuscript. Physical and optical parameters of hydrophobic and hydrophilic BC used in this study, which are adopted from Solmon et al. (2006) and Wu et al. (2019). $r_0$ and $s$ are the geometric mean diameter and standard deviation of the particle lognormal size distribution. D, $\rho$ and $\alpha$ are the characteristic effective diameter, density and hygroscopic growth factor, respectively.

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

Reference

Huang, Y., Chameides, W. L., and Dickinson, R. E.: Direct and indirect effects of anthropogenic aerosols on regional precipitation over east Asia, J. Geophys. Res.: Atmos., 112, https://doi.org10.1029/2006jd007114, 2007.

Jacobson, M. Z.: A physically-based treatment of elemental carbon optics: Implications for global direct forcing of aerosols, Geophys. Res. Lett., 27, 217-220, https://doi.org10.1029/1999gl010968, 2000.

Redemann, J., Russell, P. B., and Hamill, P.: Dependence of aerosol light absorption and single-scattering albedo on ambient relative humidity for sulfate aerosols with black carbon cores, J. Geophys. Res.: Atmos., 106, 27485-27495, https://doi.org10.1029/2001jd900231, 2001.

**3. The assumed hygroscopicity of hydrophilic BC requires clearer justification and sensitivity framing.**

Section 2.3 assigns a hygroscopicity parameter of k=0.22 to hydrophilic BC, based on Wu et al. (2019), while noting that some previous modeling studies used a nearly non-hygroscopic value of $5.0\times10^{-7}$ for BC. These values appear to represent physically different particle states—fresh or uncoated BC versus aged BC-containing particles and therefore their numerical difference alone does not demonstrate that k=0.22 is inappropriate.

Nevertheless, the assumed hygroscopicity directly affects critical supersaturation, cloud-droplet activation, Nd, cloud optical depth, and consequently the magnitude of the diagnosed BC–cloud ERF. Please clarify:

- What value of k is assigned to hydrophobic BC?
- Are the same hydrophobic and hydrophilic BC hygroscopicity values applied in both the default- and dynamic-aging experiment families?
- Is a single value of k=0.22 considered representative across the entire East Asian domain and across the range of BC coating compositions?

If the same hygroscopicity values are used in the Def and Dyn experiments, then the comparison between the two experiment families can reasonably isolate the influence of the aging timescale. However, the absolute magnitudes of Dyn_Ind, Def_Ind, and the total ERFs may remain sensitive to the selected hydrophilic-BC hygroscopicity.

A sensitivity experiment using a plausible range of hygroscopicity values for aged BC-containing particles would substantially strengthen the analysis. If an additional coupled simulation is not feasible, an offline cloud-activation sensitivity calculation or a quantitative uncertainty discussion should be provided. Comparisons of the absolute indirect ERF with previous literature should also acknowledge this parameter dependence.

**R:** Thank you for your comment. We have clarified the hygroscopicity settings of BC in the revised manuscript. Specifically, the hygroscopicity parameter of hydrophobic BC is set to $5.0\times10^{-7}$, whereas that of hydrophilic BC is set to 0.22. To make these clearer, we have added Table 1 to the Method section to summarize the physical and optical parameters prescribed for hydrophobic and hydrophilic BC. We also clarify that exactly the same parameters for hydrophobic and hydrophilic BC are applied in both the default and dynamic aging experiments. The only difference between the two series of experiments is the treatment of the BC aging timescale. This information has now been explicitly stated in Section 2.4.

Wu et al. (2019) indicated that the hygroscopicity of ambient BC-containing particles varies substantially with particle size, coating thickness and coating composition. However, the simplified aerosol scheme currently implemented in RegCM-Chem tracks BC using only two tracer states, namely hydrophobic and hydrophilic BC. Therefore, it does not explicitly resolve the continuous evolution of individual BC-containing particles in particle size and coating thickness during aging. Such a detailed representation would in principle require a particle-resolved model (Riemer et al., 2009; Riemer et al., 2019), which is currently impractical for long-term and large-domain climate simulations due to the extremely high computational and storage costs. Therefore, within the present two-state BC framework, we adopted a hygroscopicity parameter of 0.22, based on the bulk hygroscopicity range (0.11–0.34) recommended by Wu et al. (2019) from their observations, as a representative value for the bulk hydrophilic BC tracer across the entire East Asian domain. We fully agree with you that prescribing a single hygroscopicity value for hydrophilic BC introduces uncertainty into the simulated cloud activation and consequently into the absolute magnitude of BC–cloud interactions and ERF. However, a more detailed treatment (size-resolved hygroscopicity parameter) is currently constrained by the simplified aerosol framework of the model and by the limited observational information over the entire East Asian domain.

To further assess the sensitivity of the modeled BC–cloud interactions to the hygroscopicity parameter, additional sensitivity experiments are performed based on Exp. 7, in which the hygroscopicity value of hydrophilic BC (0.22) is replaced by 0.11 and 0.34, respectively. These values were selected to represent a plausible range constrained by observations of ambient BC-containing particles (Wu et al., 2019). The formation of $N_d$ induced by BC exhibits limited sensitivity to variations in the assumed hygroscopicity. Specifically, as the value increases from 0.11 to 0.34 (about a threefold increase), the BC-induced $N_d$ ranges from −13.1% to 8.1% relative to the results in Exp. 7. Similar results have also been reported in the observational study by Wu et al. (2019), who found the BC activation fraction was

33 ± 16% at a critical supersaturation of 0.1% within the aforementioned hygroscopicity range. In contrast, we find a stronger sensitivity of the BC-induced cloud radiative forcing and regional climate response to the assumed hygroscopicity. For example, the surface ERF over East Asia varied from −0.25 to −0.93 W $m^{-2}$ across the tested hygroscopicity values. This indicates that, although the changes in $N_d$ are relatively weak, subsequent cloud radiative and dynamical adjustments can amplify the regional radiative forcing and climate response to BC hygroscopicity, which is also reflected in the sensitivity of the BC indirect effect to dynamic aging (Figs 7 and 8). Previous studies have reported a wide range of BC indirect radiative forcing in liquid clouds from −0.64 to +0.23 W $m^{-2}$ (Kristjánsson, 2002; Chen et al., 2010; Koch et al., 2011; Spracklen et al., 2011; Storelvmo, 2012), with considerable uncertainty largely attributed to challenges in accurately assessing the hygroscopicity of BC-containing particles (Zhang et al., 2025). Therefore, it should be clarified that the absolute climate responses here depend on the single parameterization scheme selected in this study. Future work could further improve the representation of BC by incorporating more detailed improvements in size distribution, mixing state and coating composition. The above descriptions have been added to the revised manuscript accordingly.

Reference

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

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**4. The "clean atmosphere" idealization for the indirect effect needs a clearer statement of scope, since its stated caveat (Section 3.5, L479-489) undersells its relevance to the polluted regions that are the paper's main focus.**

The authors acknowledge that "other aerosols are not considered" and that this "represents the ideal conditions against the background of a clean atmosphere," explaining that this inflates Nd sensitivity in remote/oceanic regions where competition for water vapor is otherwise weak. But this same competition-suppression mechanism should also inflate the indirect effect over the polluted source regions (NCP, Sichuan Basin) that anchor most of the paper's claims. Please clarify explicitly what "other aerosols" means here - are sulfate and OC (both of which RegCM-Chem's aerosol module explicitly tracks per Section 2.1) included as background CCN in every experiment, with only additional aerosol types (dust, sea salt, nitrate) excluded? Or is BC literally the only species contributing to Nd throughout? If BC is the only aerosol population permitted to affect cloud-droplet activation, the resulting indirect and total ERFs should be framed as idealized BC-only, clean-background susceptibility experiments rather than realistic estimates of regional aerosol-cloud forcing.

Specifically please elucidate on:

- Do sulfate and organic aerosol contribute to the background CCN population and Nd?
- Are they included in the competition for water vapor and the calculation of maximum supersaturation?
- Or is BC the only aerosol species permitted to influence cloud-droplet activation in these experiments?

**R:** Thank you for your question. We agree that our previous description could lead to misunderstanding regarding the treatment of the cloud activation calculation. We have therefore clarified both the model configuration and the scope of the indirect effect experiments in the revised manuscript.

The aerosol module in RegCM-Chem tracks BC, OC and sulfate aerosols. However, in the aerosol–cloud interaction treatment adopted in the present experiments, only BC is explicitly coupled to the cloud-droplet activation parameterization. The simulated sulfate and OC concentrations are not included as additional aerosol populations in the calculation of cloud droplet number concentration ($N_d$). Background cloud droplet concentrations are employed in this study because this model does not include background aerosols. Wang and Penner (2009) suggested that it is reasonable to set a background droplet number concentration to prevent situations in which the simulated aerosol loadings are too low. Following the parameterization adopted in Kristjánsson (2002), the background $N_d$ is prescribed as 400 $cm^{-3}$ over land (decreasing with height) and 150 $cm^{-3}$ over ocean. More precisely, the results in this study should be regarded as idealized BC-only activation experiments superimposed on a prescribed background $N_d$, rather than as simulations of cloud activation by the complete ambient aerosol population. The above statements have been added in the revised manuscript.

Reference

Kristjánsson, J. E.: Studies of the aerosol indirect effect from sulfate and black carbon aerosols, J. Geophys. Res.: Atmos., 107, https://doi.org10.1029/2001jd000887, 2002.

Wang, M. and Penner, J. E.: Aerosol indirect forcing in a global model with particle nucleation, Atmos. Chem. Phys., 9, 239-260, https://doi.org10.5194/acp-9-239-2009, 2009.

**5. The spatial coverage and temporal treatment of the emission inventories need to be documented more completely.**

The manuscript states that anthropogenic emissions are provided by MEIC. However, MEIC is primarily a China-focused inventory, whereas the simulation domain includes South and Southeast Asia and the results show substantial BC emissions and loading over northern India.

Please specify:

- Which inventory provides anthropogenic emissions outside China, particularly over India and Southeast Asia?
- Which inventory provides open biomass-burning emissions?
- Whether the emissions include monthly or seasonal variability
- Whether anthropogenic and biomass-burning emissions vary annually throughout the simulation or whether a single inventory year is repeated.

- Which inventory year or years are used for China and for the non-China portion of the domain.

Also, given the well-documented decline in Chinese anthropogenic BC/SO2 emissions from ~2013 onward under China's clean air actions, it matters a great deal whether MEIC is applied as a fixed year (e.g., 2007 or 2020) or as a time-evolving inventory across the 14-year simulation period. If fixed, the reported "summertime aging timescale" and BC loading climatologies represent averaging over a period during which real-world BC loadings/composition were changing substantially, and interannual variability in the analysis (used for the t-tests) would then reflect only meteorological variability, not emission trends.

**R:** Thank you for your valuable comment. We have revised the Methods section to explicitly describe the spatial coverage and temporal treatment of the emission inventories. Details are as follows.

Anthropogenic emissions over China are prescribed using the Multi-resolution Emission Inventory for China (MEIC), a bottom-up emission inventory model developed by Tsinghua University (Li et al., 2017a; Zheng et al., 2018). The resolution of the MEIC employed is 0.25°, which includes monthly and interannual variations during 2008–2021. Therefore, both the seasonal variability and interannual evolution of anthropogenic emissions over China are represented in the simulations. In particular, the substantial reductions in Chinese anthropogenic emissions associated with the implementation of clean air actions after 2013 included rather than represented by a fixed inventory year.

The emission outside China in Asia are taken from the 2008 MIX Asian anthropogenic emission inventory (Li et al., 2017b). MIX provides monthly gridded emissions, but the 12 monthly emission fields for 2008 are repeated annually throughout the simulation period. Thus, seasonal variability is retained outside China, whereas interannual changes in anthropogenic emissions are not represented. However, in contrast to the substantial emission reductions over China, primary anthropogenic particulate emissions over India exhibited relatively weak temporal changes during the 2010s (Li et al., 2024). Additionally, this study mainly focuses on the climate response in East Asia, especially China. Therefore, although this treatment represents a simplification of the temporal evolution of emissions outside China, its influence on the main conclusions of this study is expected to be limited. Nevertheless, we agree that the spatial coverage and temporal treatment of the emission inventories should be documented more explicitly, and the corresponding description has now been added to the revised Methods section.

It should be acknowledged that the open biomass-burning emissions are not included in the present simulations, which may partly contribute to the underestimation of BC concentrations and AOD identified in the model evaluation. This limitation has also been explicitly stated in the Conclusions section of the revised manuscript.

Reference

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**6. The manuscript should distinguish more clearly between evaluation of the general model climatology and evaluation of the dynamic BC-aging treatment.**

Figure 1 shows that Exp. 8 reproduces the broad summertime meteorological and BC AOD patterns over East Asia when compared with ERA5, GPM, and MERRA-2. This is useful for evaluating the overall RegCM-Chem configuration. However, the large-scale meteorological state is strongly influenced by the prescribed ERA-Interim lateral boundary conditions and OISST, while regional precipitation and circulation also depend on the model's convection, PBL, land-surface, and cloud parameterizations. Consequently, agreement in these broad fields does not by itself validate the dynamic BC-aging parameterization.

Figure S1 compares observed surface BC concentrations with Exp. 4 and Exp. 8 over 33 CBNET sites during 2012-2018, while Table S1 provides the corresponding station-level values. Both simulations reproduce the observed decreasing tendency, and Exp. 8 appears to reproduce the trend slightly better. However, Exp. 4 and Exp. 8 remain very similar, and both show substantial site-dependent biases.

Please quantify this comparison using consistent evaluation metrics for both simulations, including mean bias, MAE, RMSE, and spatial and temporal correlations. A simple calculation from the values in Table S1 suggests that Exp. 8 slightly reduces the domain-mean negative bias but does not clearly improve MAE, RMSE, or spatial correlation relative to Exp. 4. The manuscript should therefore clarify whether the dynamic-aging treatment provides a statistically or practically meaningful improvement in surface BC simulation, rather than relying primarily on visual comparison.

It would also be useful to provide the equivalent comparison of BC AOD between Exp. 4 and Exp. 8. Because MERRA-2 BC AOD is itself a reanalysis/model-derived product rather than a direct observation of BC AOD, this comparison should be described as a cross-model or reanalysis evaluation rather than independent observational validation.

More generally, the manuscript should distinguish among: (i) evaluation of the background meteorological climatology, (ii) evaluation of BC concentration and optical depth, (iii) evaluation of the default versus dynamic aging treatment, and (iv) evaluation of the resulting BC-induced climate perturbations. The existing results support the physical consistency of the dynamic-aging mechanism,

particularly the conversion from hydrophobic to hydrophilic BC shown in Fig. S2, but they do not yet demonstrate that the new treatment substantially improves the fidelity of the simulated climate response.

**R:** Thank you for your constructive comment. Accordingly, we have revised Section 3.1 to separately discuss the background meteorological climatology, BC surface concentration and optical depth, and the relative performance of the default and dynamic aging treatments.

The simulated meteorological variables, including thermodynamic fields, humidity and precipitation, are first compared with ERA5 and GPM datasets. The results show that RegCM-Chem successfully reproduces the regional climatic features of East Asia in summer. However, we agree that the large-scale meteorological state is constrained by the boundary conditions and physical parameterizations used in RegCM-Chem. Therefore, agreement in these broad meteorological fields provides confidence in the background climate simulation but does not independently demonstrate the validity of the dynamic aging treatment.

Following your suggestion, 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).

We have also added an equivalent quantitative evaluation of BC AOD from Exp. 4 and Exp. 8 against MERRA-2 AOD. We agree that the MERRA-2 AOD product is a reanalysis/model-derived dataset rather than a direct observation of BC AOD. However, long-term and spatially extensive direct observations of BC AOD over China remain very limited. Therefore, we employ MERRA-2 BC AOD as a reference dataset for this evaluation. It is also worth noting that the aerosol AOD products from MERRA-2 have been extensively evaluated against multiple independent observational datasets in previous studies (Buchard et al., 2017; Che et al., 2019; Sun et al., 2019; Ou et al., 2022). These evaluations generally demonstrate reasonable agreement in the magnitude and spatiotemporal variability of MEAAR-2 AOD. Meanwhile, in the revised manuscript and Supplement, this comparison is now explicitly described as a reanalysis-based evaluation rather than an independent observational validation. 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.

In addition, we 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

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**7. The paper states a Student's t-tests performed "on the seasonal summer means of each year" (n~13-14 years) to assess significance of differences between experiments.**

Since each experiment pair shares identical lateral boundary/SST forcing for a given year, a paired comparison (differencing before testing) would be appropriate and more powerful than an unpaired test; please confirm this is what was done. With only ~13 independent summers and known interannual autocorrelation from monsoon phase/ENSO state, it would help to state how autocorrelation or limited sample size was handled. Please confirm whether a paired, two-sided t-test was used. If an unpaired test was applied, the analysis should be repeated using the paired annual differences. The authors should also state whether the annual differences were examined for temporal autocorrelation and whether the effective sample size was adjusted if significant serial dependence was present.

**R:** Thank you for your important comment. We agree that a paired comparison is more appropriate

because each pair of experiments was driven by identical lateral boundary conditions and SST forcing for the same year. Following your suggestions, we have repeated the significance analysis 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.

**8. The paper associates condensation contribution to BC aging using the sulfate condensation mass flux.**

However, the model description states that RegCM-Chem also includes a volatility-basis-set treatment of secondary organic aerosol, and the Introduction describes BC aging through condensation more generally. Please clarify whether condensation of secondary organic material contributes to I_cond elsewhere in the model.

**R:** Thank you for your important comment. We agree that the condensation of secondary organic material can contribute to BC aging. Although RegCM-Chem includes a volatility-basis-set treatment of secondary organic aerosol, the organic-vapor condensation is not currently coupled to the BC aging parameterization due to the simplification of the aerosol module in current RegCM-Chem configuration, which does not explicitly resolve secondary organic aerosol formation and volatility-resolved organic vapor. Therefore, an explicit organic-condensation contribution cannot be robustly represented in the present model framework. For this reason, we use the sulfate condensation flux as the primary proxy for the condensational aging term because sulfate is a major hygroscopic secondary aerosol component and an efficient contributor to the conversion of freshly emitted hydrophobic BC toward a more hydrophilic state, particularly in polluted East Asian environments where sulfate formation is important (Miyakawa et al., 2017; Lim et al., 2018; Cui et al., 2022). Nevertheless, we recognize that neglecting organic condensation may lead to uncertainties in regions or seasons with substantial secondary organic aerosol formation. We have added this point to the limitation section and clarified that future work should improve the representation of BC condensational aging by incorporating more complete aerosol chemistry, including secondary organic aerosol formation and other aerosol species.

Reference

Cui, S., Huang, D. D., Wu, Y., Wang, J., Shen, F., Xian, J., Zhang, Y., Wang, H., Huang, C., Liao, H., and Ge, X.: Chemical properties, sources and size-resolved hygroscopicity of submicron black-carbon-containing aerosols in urban Shanghai, Atmos. Chem. Phys., 22, 8073-8096, https://doi.org10.5194/acp-22-8073-2022, 2022.

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**Minor Comments:**

**1.** Fig. 8 caption (Line500-507) uses the same panel labels "(b) and (e)" twice for two different comparisons (Dyn_Ind vs. Def_Ind, and Dyn_Tot vs. Dyn_Ind) - the second instance should presumably be "(c) and (f)"?

**R:** Thank you for your comment. We have corrected the panel labels in the caption of Fig. 8 accordingly.

**2.** Line 422 typo “angel” should be “angle”

Corrected

**3.** Line 644 “clean aeras” should be “clean areas”

Corrected

**4.** The model uses 18 -sigma layers from the surface to 5 hPa, just a question out of curiosity, are is this resolution sufficently fine? particularly near the surface to PBL top evolution, where aging rates and semi-direct effects are sensitive to vertical mixing?

**R:** Thank you for your important comment. Although the model employs only 18 terrain-following sigma levels from the surface to 5 hPa, the vertical levels are not uniformly distributed. Approximately six model levels are located between the surface and 850 hPa in our configuration. Therefore, the vertical resolution is considerably finer near the surface than in the middle and upper troposphere, allowing the model to represent the major vertical gradients and evolution of the lower troposphere and PBL.

Additionally, the University of Washington (UW) PBL scheme is applied in RegCM-Chem. Compared with purely empirical diffusion approaches, the UW scheme explicitly represents turbulent mixing processes and vertical transport associated with boundary-layer evolution and entrainment near the PBL top, which has also been demonstrated to more appropriately capture regional aerosol distributions and their climate impacts over East Asia (Cao et al., 2026). These provide an appropriate framework for representing the principal evolution and mixing processes of the PBL at the regional climate scale considered in this study.

Reference

Cao, H., Zhuang, B., Zhou, Y., Gao, P., Hu, Y., Wang, T., Li, S., Li, M., Xie, M., and Liu, Q.: Impact of boundary layer schemes in RegCM-Chem on East Asian climate and its response to the aerosol-radiation interaction, Atmos. Res., 337, https://doi.org10.1016/j.atmosres.2026.108951, 2026.

**5.** Please provide the physical and optical parameters assigned to the hydrophobic and hydrophilic BC tracers. In particular, the assumed effective diameter D and density ρ used for hydrophobic vs. hydrophilic BC tracers, since these directly control both the aerosol number concentration N feeding into the aging timescale and the AOD/extinction calculation. For reproducibility, a compact table should list, for each BC tracer:

- Characteristic effective diameter
- Density
- Geometric standard deviation
- Hygroscopicity parameter
- Solubility or scavenging parameter
- Wavelength dependent mass extinction or absorption coefficients
- Refractive index and size distribution assumptions used to derive optical coefficients
- Hygroscopic growth factor or its parameterization.

**R:** Thank you for your useful suggestions. 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, $\rho$ and $\alpha$ are the characteristic effective diameter, density and hygroscopic growth factor, respectively.

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

Overall, I think this is a solid, well-motivated contribution to a genuinely important process-representation gap in regional aerosol-climate modeling, and the core result (aging timescale inversely related to pollution, decoupling of column burden and surface-level responses via the dry/wet deposition compensation mechanism) is useful.