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.

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:

$$I_{Dyn} = \text{Dyn\_tot} – \text{Dyn\_dir} – \text{Dyn\_ind}$$

or, using the original experiments,

$$I_{Dyn}=\text{Exp.8}–\text{Exp.6}–\text{Exp.7}+\text{Exp.5}.$$

Similarly, for the default-aging experiments:

$$I_{Def} = \text{Def\_tot} – \text{Def\_dir} – \text{Def\_ind} = \text{Exp.4} – \text{Exp.2} \_ \text{exp.3} + \text{exp.1}.$$

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 abour 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.

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.
3. **The assumed hygroscopicity of hydrophilic BC requires clearer justification and sensitivity framing.**
Section 2.3 assigns a hygroscopicity parameter of κ=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 5.0×10^{-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.

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?

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.

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.

7. The paper states a Student's t-testis 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.

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.

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)"?
2. Line 422 typo "angel" should be "angle"
3. Line 644 "clean aeras" should be "clean areas"
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? particulalry near the surface to PBL top evolution, where aging rates and semi-direct effects are sensitive to vertical mixing?
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:
    - Chracteristic effective diameter
    - Density
    - Geometric standard deviation
    - Hygroscopicity parameter
    - Soulubility or scavenging parameter
    - Wavelength depndent mass extinction or absorption coefficents
    - Refractive index and size distribution assumptions used to derive optical coefficencts
    - Hygroscopic growth factor or its parameterization.

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.