Response to the comments of Anonymous Referee #2 (egusphere-2024-2372)

In this study, a BC aging parameterization was implemented into the WRF-CMAQ model. BC aging processes are important for accurately estimating the spatial distribution, atmospheric lifetime, optical properties, and activation to cloud particles of BC. While BC has been treated as internally mixed particles in CMAQ, the authors have classified BC into externally mixed and internally mixed particles and introduced an aging parameterization for converting externally mixed particles to internally mixed particles, as well as a scheme for calculating the differences in cloud activation and optical properties between externally mixed and internally mixed BC particles. This study contains interesting aspects as a BC modeling study. However, many similar studies have been published in the last 15 years, and this study lacks scientific novelty. Considering this point, I cannot recommend this study as an ACP paper.

Response: Thank you so much for your thorough review and agreement on the importance of this study. We believe our manuscript is not only technically valuable but also scientifically novel for the following reasons:

1. This is the first time the BC aging process has been considered and tracked within the framework of the CMAQ model and WRF-CMAQ coupled model. The CMAQ model, a widely accepted chemistry transport model, recognized for regulatory applications in the US. No doubt BC aging is a worthwhile process (which the reviewer also noted), and this work acquired the BC aging in our focal model, WRF-CMAQ coupled model. Thus, our model development reflects a sophisticated and state-of-the-art integration of different physical processes. Although some of the parametrizations are adopted from previous works for other models, our work is path breaking in the following two aspects:

(1) We introduced a new scheme using two additional species: Bare BC and Coated BC, to represent different aging states. Significant differences in BC properties drove us to modify wet deposition and aerosol optics algorithms. Unlike other methods, which added new modes in modal models, in previous studies, our approach not only reduced overall complexity, but also enhanced the model's functionality and accuracy with minimal computational demand.

(2) Our in-cloud scavenging calculation method accounts for evolving hydrophilicity of BC aerosol, improving the accuracy of wet deposition simulations. Specifically, Bare BC, being hydrophobic, does not engage in nucleation scavenging but is subject to impact scavenging. After impact scavenging, Bare BC becomes hydrophilic and is subsequently removed as Coated BC.

2. Besides the model development itself, this manuscript showed some new findings and conclusions as well. We are regretted that we didn't present them clearly in the original submission, and have improved them in the revision.

(1) Previous studies have primarily focused on the effects of the BC aging process on mixing state and optical properties, with less emphasis on its impact on concentration. In this work, we found that the

changes in wet deposition caused by BC aging process can significantly impact BC concentrations. In the US CONUS domain, considering the aging process results in a 17.7% reduction in simulated wet deposition, accompanied by a 10.5% increase in BC column concentration.

(2) We have explicitly provided the spatial and temporal distribution characteristics of BC aerosol mixing states: Bare BC is prevalent near emission sources, while Coated BC is more common farther from sources, and the Number Fraction of Coated BC increases during the daytime.

(3) The influence of BC aging on its optical properties has been a long-standing uncertainty, although with increasingly detailed characterizations of BC mixing states (Wang et al., 2018; Chen et al., 2023). The CMAQ model, as a well-established air quality model, accounts for over a hundred aerosol species and hundreds of chemical reactions, enabling more accurate aerosol information. Within the WRF-CMAQ model framework, we found that incorporating differences in BC mixing states due to aging alone improves the alignment of simulation results with observational data. The median MAC (mass absorption cross-section) simulated by our new model is 8.77 m^2/g, approximately 30% lower than the original model's value of 12.45 m^2/g, bringing it closer to the observed value of 9.75 m^2/g. This result was achieved without accounting for aerosols scattering separately. This implies in models with comprehensive physical-chemical processes, optical calculation process could be simplified in future.

Thus, we do believe our work offers both technical contributions and scientific advancements, and think it is suitable for both ACP and specialized journals on model development. We are considering ACP due to its high impact and profound influence within the communities that focused on both model development and scientific researches.

Please find our point-by-point responses listed below. The reviewer’s comments are in *Italic* followed by our responses and revisions (in blue). The modifications in the manuscript are highlighted in red.

Major comments:

The introduction is generally well structured and covers the important previous studies. However, the model developed in this study has already been developed and used in the papers listed in the introduction, indicating that this study lacks scientific originality. It can be said that the optical property part is relatively new, but there have already been many studies focusing on the differences in light absorption efficiency caused by the mixing state of BC particles.

Is it considered that this research is not new scientifically, but new to the CMAQ model? Or does it contain some new scientific findings? I think this research falls into the former category. In that case, this study is not appropriate for ACP. I strongly recommend that the authors submit this study to another model development journal (e.g., GMD).

Response: Thank you so much for such a candid comment. Through collaboration with the US EPA CMAQ development team, this is the first time that BC aging process has been considered and tracked within the framework of the CMAQ and WRF-CMAQ models. As discussed in our previous response, our work contributes significantly to both model development and scientific understanding, and not only the optical properties are new. Given the comprehensive nature of this work, we believe it provides valuable advancements beyond previous studies. Therefore, we believe that ACP is an appropriate journal for this publication.

2) Related to the comment above, the BC aging parameterization used in this study is the same as that developed and used in previous studies, and there is nothing new about it. In addition, the methods section needs substantial revision because there are many things that are not adequately described. For example, does the model treat aerosols that do not contain BC (BC-free particles) in the accumulation mode? Considering BC-free particles is important for estimating the mixing state and optical properties of BC, but it is not clear from the text how the model distinguishes non-BC species between BC-containing particles (used for coating) and BC-free particles (see comment 4 below).

Response: Thank you for your comments. As mentioned in my previous response, our approach to incorporating the BC aging process differs from the methods summarized in the third paragraph of the introduction regarding previous models.

We also appreciate your suggestion to expand the methods section. We apologize for not providing a more detailed explanation earlier and have revised the manuscript accordingly (in red) to strengthen the methodology description.

(1) In Section 2.1, “New Model”, we added a new Table 2 (please see comment 4 below) and an explanation highlighting the differences in the treatment of BC aerosol between the original model and the new model.

"In the original model, BC aerosol did not include the aging process and was treated as being in a completely internally mixed state. This approach neglected the presence and impact of externally mixed BC aerosol in the real atmosphere, leading to inaccuracy in the numerical simulation of BC aerosol. In our new model, the differences of BC aerosol across major processes under different aging states are considered. Freshly emitted BC aerosol (Bare BC) enters the BC aging module from the emission module and gradually converts into Coated BC through the aging process. When Bare BC and Coated BC enter the wet deposition module, hydrophobic Bare BC cannot undergo nucleation scavenging during in-cloud scavenging, in turns affecting the wet deposition of BC aerosol. Bare BC exists in an external mixing state, whereas Coated BC has stronger light absorption due to the lensing effect. Therefore, Bare BC and Coated BC need to be considered separately when calculating aerosol optics."

(2) In Section 2.2, "BC Aging Module", we revised the description of the method for the virtual chemical reaction.

"Based on the selected OH aging scheme, the dynamic process of BC aging is represented by setting a virtual reaction, wherein Bare BC progressively converts into Coated BC. The aging rate is used as the reaction rate for this virtual chemical reaction."

(3) In Section 2.3, "Cloud Chemistry Module", we included a clarification that this study does not consider precipitation scavenging and focuses mainly on in-cloud scavenging processes.

"In the WRF-CMAQ model, the wet deposition module carries the following tasks: calculates in-cloud scavenging and precipitation scavenging of BC aerosol, performs aqueous chemistry calculations, and accumulates wet deposition. Precipitation scavenging associated with precipitation forms such as rain, snow, and graupel, and is minimally affected by changes in BC aerosol properties before and after aging. Therefore, this study does not consider the differences in precipitation scavenging process. In contrast, in-cloud scavenging (includes nucleation scavenging and impact scavenging) is strongly associated with BC aging process, reflecting the hydrophobicity differences between Bare BC and Coated BC."

(4) In Section 2.4, "Aerosol Optics Module", we revised potentially misleading description related to coarse mode scattering aerosols and clarified the explanation of the aerosol optics algorithm modification.

"For aerosols containing BC in the Aitken and accumulation modes, the Core-shell Mie theory is employed to calculate their optical characteristics (coarse mode aerosols without BC component are not considered in this study). The particle structure information cannot be fully represented in the current WRF-CMAQ model, as the spherical shape assumption is used for calculations. This simplification may introduce biases in the results (He et al., 2015; Wang et al., 2017, 2021). The potential impacts of such structural simplifications are not addressed in this study. In our WRF-CMAQ-BCG model, we calculated the optics of Bare BC and Coated BC separately. We introduced a variable, the number fraction of Coated BC (NF_{coated}). The NF_{coated} variable was brought into the aerosol optics module for translating BC core back to Bare BC and Coated BC. Only Coated BC can be a core surrounded by a shell. Once encapsulated, Coated BC becomes a BC-containing particle, represented as a core-shell sphere, and its particle size information is recalculated based on the volume of the Coated BC core and the shell. Its optical properties are calculated using Core-shell Mie theory. In contrast, Bare BC is represented as a homogeneous sphere, with the particle size recalculated using the volume of Bare BC, and its optics properties are calculated using standard Mie theory. By apportioning the BC core, the overestimation of aerosol light absorption can be corrected."

Regarding your comment on BC-free particles in the accumulation mode, the current version of the model does not treat these particles (for specific details, please refer to comment 4 below).

Specific comments:

3) L41: formation or altering -> formation and altering.

Response: Thanks for your suggestion. We have changed “formation or altering” to “formation and altering” in revised manuscript.

4) L144: Figure 2: Are BC-free particles considered in the accumulation mode? In Figure 2, it appears that BC-free particles are treated as scattering aerosols. If so, how are scattering aerosols in coated BC and scattering aerosols (BC-free particles) treated in the model? Are they treated as separate aerosol variables?

Response: Thanks for your comments. We did not consider BC-free particles in the accumulation mode in our model. Apologies for any confusion caused. The WRF-CMAQ model operates under the assumption of complete internal mixing, and calculating aerosol optics on a modal basis. The Aitken mode and accumulation mode containing BC aerosol both utilize the Core-shell Mie method for optical calculations, while coarse mode aerosols that lack BC content are treated as BC-free particles and calculated using the standard Mie method. In this study, to account for the effects of the BC aging process, we specifically considered the Aitken and accumulation modes with BC, with a distinguishing mixing state of BC aerosol without including BC-free particles. We have replaced Figure 2 with Table 2 and revised the text in Section 2.4 to clarify this point.

Table 2. Comparison of BC aerosol in major processes (Aitken and accumulation modes).

Species Processes		BC (Original)	Bare BC (New)	Coated BC (New)
Emission		Yes	Yes	No
BC Aging		No Aging process	Bare BC is aged to Coated BC	
Wet Deposition	Impact scavenging	Yes	Yes	Yes
	Nucleation scavenging	Yes	No	Yes
Aerosol Optics		Core-shell sphere	Homogeneous sphere	Core-shell sphere

“In the WRF-CMAQ model, the light absorption of aerosols is entirely attributed to BC aerosol. BC aerosol is considered the core, with water-soluble aerosols, insoluble aerosols, aerosol water, and sea salt as the shell. … For aerosols containing BC in the Aitken and accumulation modes, the Core-shell Mie theory is employed to calculate their optical characteristics (coarse mode aerosols without BC component are not considered in this study). The particle structure information cannot be fully represented in the current WRF-CMAQ model, as the spherical shape assumption is used for calculations. This simplification may introduce biases in the results (He et al., 2015; Wang et al., 2017, 2021). The potential impacts of such structural simplifications are not addressed in this study. In our WRF-CMAQ-BCG model, we calculated the optics of Bare BC and Coated BC separately. We introduced a variable, the number fraction of Coated BC (NF_{coated}). The NF_{coated} variable was brought into the aerosol optics module for translating BC core back to Bare BC and Coated BC. Only Coated BC can be a core surrounded by a shell. Once encapsulated, Coated BC becomes a BC-containing particle, represented as a core-shell sphere, and its particle size information is recalculated based on the volume of the Coated BC core and the shell. Its optical properties are calculated using Core-shell Mie theory. In contrast, Bare BC is represented as a homogeneous sphere, with the particle size recalculated using the volume of Bare BC, and its optics properties are calculated using standard Mie theory. By apportioning the BC core, the overestimation of aerosol light absorption can be corrected."

5) L167-168: It is not correct to assume that the aging speed of coagulation is constant. Coagulation occurs very fast near sources and has a large contribution to BC aging. The speed of coagulation aging is highly dependent on aerosol concentrations.

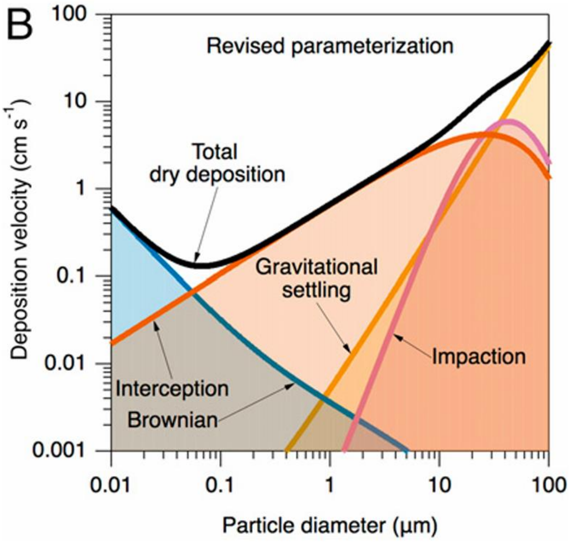
Response: Thank you for your suggestion. We agree that coagulation can occur rapidly near sources. However, coagulation primarily manifests as aggregation between BC particles forming chain-like structures (Bond et al., 2013). Collisions between externally mixed black carbon particles do not significantly alter their mixing state. Additionally, we ran CMAQ which is a regional air quality model, with a spatial resolution of 12 km, therefore, it was not able capturing rapid coagulation that took place close to emission sources. In our simulation, the daily average concentration of $PM_{2.5}$ reached a maximum value of 33.48 μg m^{-3}. Within this range, Coagulation does not play a dominant role compared to Condensation (particularly pronounced in the accumulation mode, where BC predominantly present). Based on these considerations, we have followed other studies (Liu et al., 2011; Huang et al., 2013; Oshima and Koike, 2013) to employ a constant coagulation rate.

6) L178: What are the particle size distributions of Bare BC and Coated BC? They should have different dry deposition speed because their particle size distributions are different due to the coating species and water uptake.

Response: Thank you for your comment. In this study, Bare BC and Coated BC are assumed to have the same particle size distribution because both consist of elemental carbon (EC) but have different aging properties. When calculating aerosol optical properties, the volumes of other species (as shell) are added to the volume of Coated BC for recalculating the particle size. We have added a relevant explanation in Section 2.4 in the revised manuscript as follows.

"In our WRF-CMAQ-BCG model, we calculated the optics of Bare BC and Coated BC separately. We introduced a variable, the number fraction of Coated BC (NF_{coated}). The NF_{coated} variable was brought into the aerosol optics module for translating BC core back to Bare BC and Coated BC. Only Coated BC can be a core surrounded by a shell. Once encapsulated, Coated BC becomes a BC-containing particle, represented as a core-shell sphere, and its particle size information is recalculated based on the volume of the Coated BC core and the shell. Its optical properties are calculated using Core-shell Mie theory. In contrast, Bare BC is represented as a homogeneous sphere, with the particle size recalculated using the volume of Bare BC, and its optics properties are calculated using standard Mie theory. By apportioning the BC core, the overestimation of aerosol light absorption can be corrected."

Regarding dry deposition, as mentioned in Section 2.3, primary influencing factors are meteorological conditions and land surface types. This implies there is only a minor difference in the dry deposition rates of Bare BC and Coated BC. As shown in Emerson et al. (2020), the dry deposition velocities for both Bare BC and Coated BC are approximately 0.2 cm s^{-1}, indicating minimal differences. Moreover, research by Textor et al. (2006) indicates that wet deposition accounts for 79% of the total removal of BC aerosol. Therefore, we mainly emphasized the wet deposition, while the dry deposition settings remain consistent with the original model.



(Figure in Emerson et al., 2020, PNAS)

7) L181: Do you use "precipitation scavenging" and "impact scavenging" with different meanings or the same meaning?

Response: Thank you for your comment. They have different meanings. Precipitation scavenging refers to removal caused by precipitation, such as below-cloud washout, while impact scavenging refers to removal resulting from the collision between cloud water and aerosols within the cloud (Barrett et al., 2019; Choi et al., 2020). Scavenging includes in-cloud scavenging and precipitation scavenging, while in-cloud scavenging includes nucleation scavenging and impact scavenging. The primary difference between Bare BC and Coated BC lies in in-cloud scavenging, whereas differences in the precipitation scavenging process are negligible. Therefore, we modified the in-cloud scavenging algorithm. Apologies for any confusion caused. We have re-drawn Fig.1 and replaced Fig. 2 with a clearer Table 2, without mentioning precipitation scavenging.

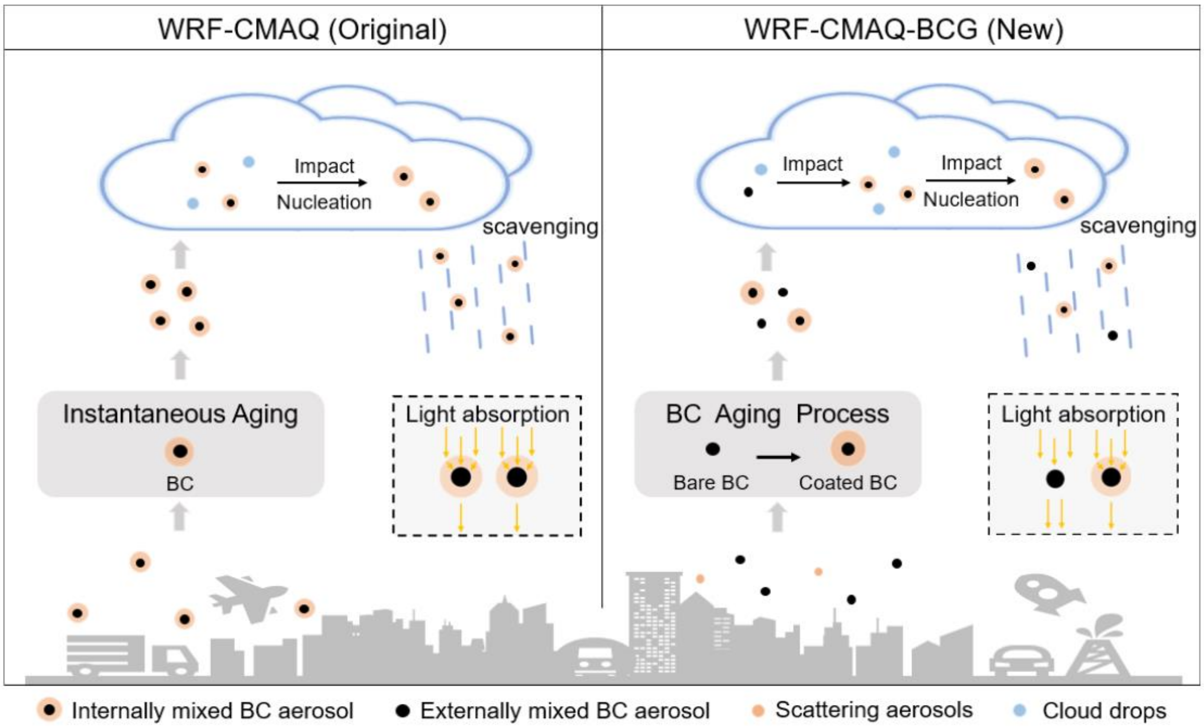


Figure 1: The BC mixing state in the WRF-CMAQ model and the WRF-CMAQ-BCG model.

Table 2. Comparison of BC aerosol in major processes (Aitken and accumulation modes).

Species Processes		BC (Original)	Bare BC (New)	Coated BC (New)
Emission		Yes	Yes	No
BC Aging		No Aging process	Bare BC is aged to Coated BC	
Wet Deposition	Impact scavenging	Yes	Yes	Yes
	Nucleation scavenging	Yes	No	Yes
Aerosol Optics		Core-shell sphere	Homogeneous sphere	Core-shell sphere

8) L194: in direct radiative forcing -> in estimating direct radiative effect

Response: Thanks for your suggestion. We have changed “in direct radiative forcing” to “in estimating direct radiative effect” in the revised manuscript.

9) L221: Are all BC emissions treated as externally mixed particles?

Response: Thank you for your comment. Yes, we consider freshly emitted BC to be externally mixed, consistent with the approach adopted by most atmospheric models. In our model, this external mixing assumption is used to represent the initial stage of BC aging, where BC particles are treated as separate entities. Over time, these particles may undergo processes like condensation, coagulation, and other interactions that lead to their final internal mixing state with other components. Even though we recognize that BC can form chain-like structures through aggregation, the interaction between BC and other aerosol species remains relatively limited, especially when they are freshly emitted. Therefore, we kept this simplification in our model, as it sufficiently represents the typical behavior of BC emissions without bringing in unnecessary complexity or large uncertainties.

10) L267-269: Figure 6a: Why is this spatial distribution obtained?

Response: Thank you for your comment. This is related to the distribution of OH concentration. The areas with higher values reflect higher atmospheric oxidizing capacity and more active photochemical reactions, leading to a higher aging rate. We have added relevant explanations in the revised manuscript.

"The aging rate (k) and the aging timescale (τ) are important variables to quantify the aging process. The standard WRF-CMAQ model lacks the capability to generate BC aging-related variables. In contrast, in the WRF-CMAQ-BCG model, spatiotemporal variations of the aging-associated variables in the BC aging process are related to the concentration of OH radicals. The areas with higher values reflect higher atmospheric oxidizing capacity and more active photochemical reactions, leading to a higher aging rate. Fig. 5(a) shows the aging rate…"

11) L278-279: Again, are all BC emissions treated as externally mixed particles?

Response: Thank you. Yes, please refer to our response to Comment 9 for the details.

12) Figure 8: The unit of the vertical axis is unclear. This is only a qualitative evaluation, and a quantitative evaluation is needed.

Response: Thanks for your suggestion. We have changed the unit of the vertical axis to "Normalized NF_{coated}". Due to the variability of BC mass concentrations and mixing states across different regions and times, a direct quantitative comparison is not feasible. Therefore, we normalized NF_{coated} to facilitate quantitative comparison and updated the relevant text in the revised manuscript.

"Due to the variability of BC mass concentrations and mixing states across different regions and times, we normalized NF_{coated} to facilitate quantitative comparison, as shown in Eq. (5). By comparing the simulated and observed Normalized NF_{coated}, it is evident that the BC mixing state reflects a temporal variation characteristic, with the proportion of Coated BC significantly increasing during the daytime.

$$\text{Normalized } NF_{\text{coated}} = \frac{NF_{\text{coated}} - NF_{\text{coated\_min}}}{NF_{\text{coated\_max}} - NF_{\text{coated\_min}}}, \qquad (5)$$

Where $NF_{\text{coated\_min}}$ is the minimum value of NF_{coated}, $NF_{\text{coated\_max}}$ is the maximum value of NF_{coated}."

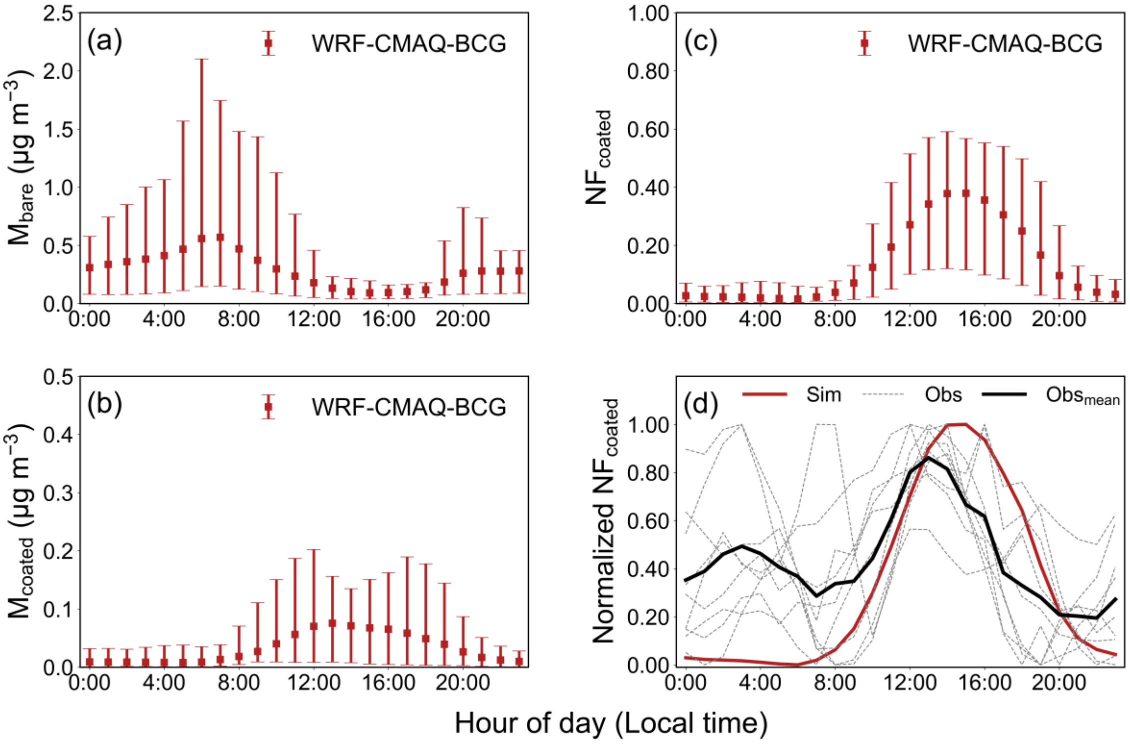


Figure 8: Daily average variation in simulated results: (a) Bare BC mass concentration (M_{bare}), (b) Coated BC mass concentration (M_{coated}), (c) Number fraction of Coated BC (NF_{coated}) and (d) Normalized NF_{coated} compared with other observational studies.

13) L319-320: This description is probably incorrect. Aerosols are transported over long distances in a few days, so I think the speed of aging is not related to Fig. 9a. I think this is because in-cloud scavenging is not considered (only below-cloud scavenging is considered).

Response: Thanks for your suggestion. We have revised the text to: "This distinctive distribution is attributed to the spatial distribution of Bare BC concentration, and the fact that Bare BC cannot be removed by nucleation scavenging."

14) Figure 13: Why don't you show the time series plot for MAC?

Response: We have added the time series plot for MAC in Figure 12. The original Figure 12 shows the time series of the absorption coefficient (b_{abs}) and BC mass concentration (M_{BC}). Since the MAC value is directly derived from the ratio of these two variables (b_{abs}/M_{BC}), we included it in Figure 12 and updated the relevant text in the revised manuscript.

"We compared the time series plot of b_{abs}, M_{BC} and MAC values simulated by the WRF-CMAQ model and the WRF-CMAQ-BCG model, with the observed values at the T0 site (Fig. 11). For b_{abs}, the Mean Bias Error (*MBE*) value for the WRF-CMAQ model is 0.34 × 10^{-5} m^{-1}, while for the WRF-CMAQ-BCG model is 0.19 × 10^{-5} m^{-1}, representing a 44% reduction. The simulated M_{BC} values for both models show little difference, with both *MBE* values of 0.30 μg m^{-3}. For MAC, the *MBE* value for the WRF-CMAQ model is 2.63 m^2 g^{-1}, whereas the WRF-CMAQ-BCG model yields an MBE of -1.24 m^2 g^{-1}, demonstrating a significant reduction. The new model, which incorporated the aging process, demonstrates accuracy improvement in simulating b_{abs} values with respect to observation data. However, the discrepancies in BC mass concentration

(M_{BC}) between the two models are minimal, suggesting that variations in M_{BC} play a minor role in the observed changes in the MAC values. This indicates that the main contributing factor to the optical impact of BC aging is the alteration in BC mixing state during the aging process, rather than BC concentration."

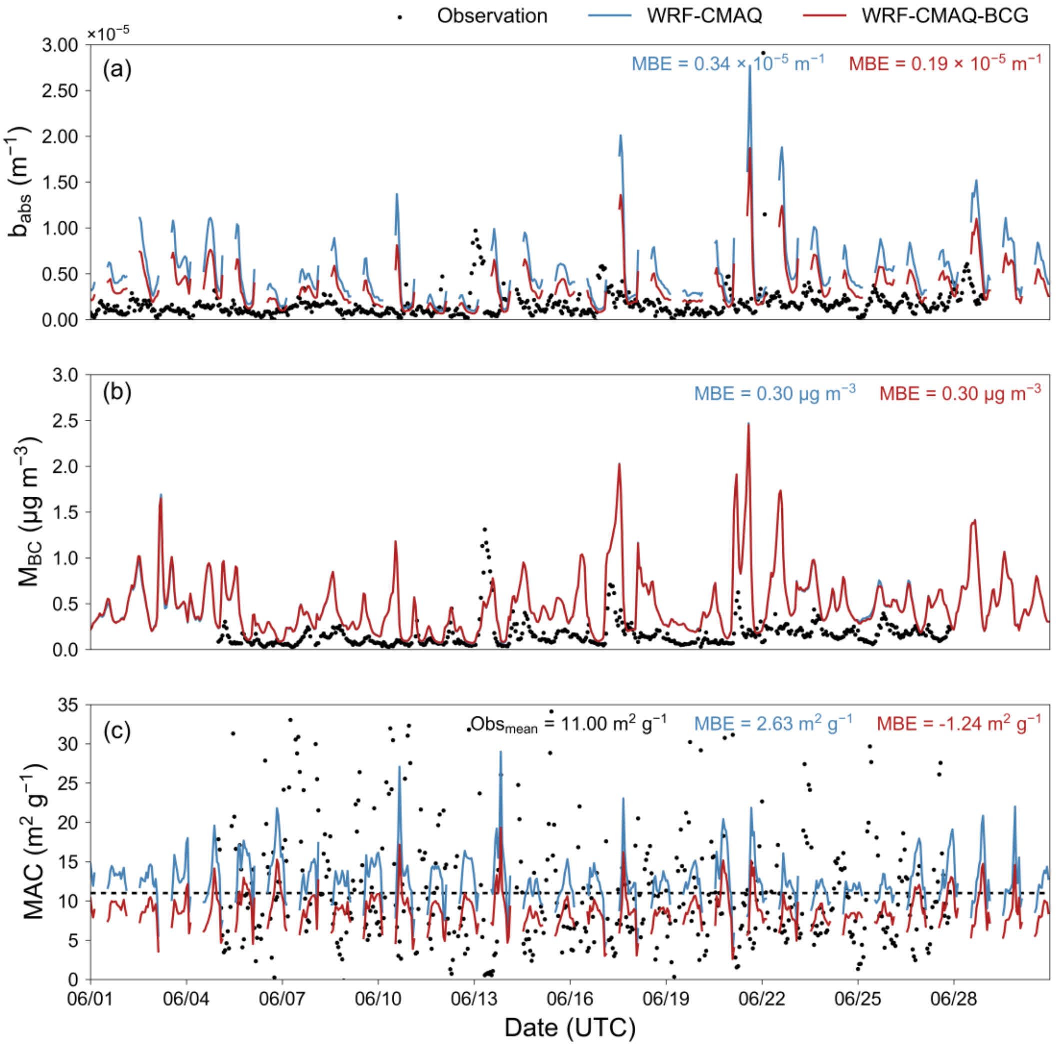


Figure 12: Time series comparison of simulated and observed (a) absorption coefficient (b_{abs}), (b) BC mass concentration (M_{BC}) and (c) BC mass absorption cross-section (MAC).

Given the significant oscillations in the observational data, making it challenging to analyze, we originally chose the more intuitive violin plot (Figure 13). In the revised manuscript, we have now retained both the time series (Figure 12) and the statistical plots (Figure 13).

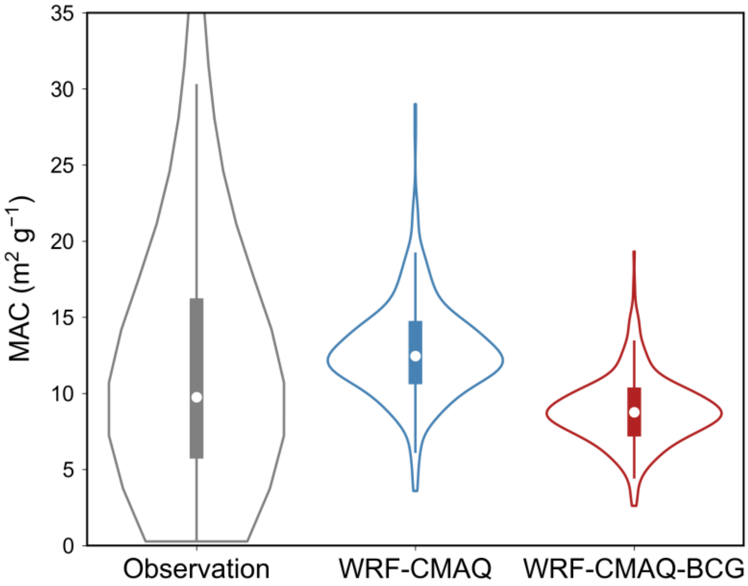


Figure 13: Statistical comparison of simulated and observed BC mass absorption cross-section (MAC). The width of the violin plot reflects the distribution of the MAC values, while the rectangular shapes inside represent miniature box-and-whisker plots. The white dots denote the median values of the results for that category.

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