Response to the comments of Anonymous Referee #1 (egusphere-2024-2372)

The authors implemented a BC aging scheme into the WRF-CMAQ model and found that accounting for BC aging process improves simulated BC optical properties. They also found that adding the aging process significantly affect BC concentration distribution and wet deposition. Overall, the manuscript is well organized and the study fits well into the journal scope. However, there are a few places that require further descriptions and clarifications. I have a few comments and suggestions below for the authors to consider.

Response: Thank you for your thoughtful and constructive feedback. We are grateful for your recognition of our work's organization and relevance to the journal's scope. Below, we address each of the comments and suggestions in detail. We have implemented all suggestions for improving our manuscript. 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 major concern I have is the insufficient model evaluation. Currently, the authors only evaluated the model simulation at one measurement sites, while all their modeling analysis and conclusions come from regional results. Thus, regional-scale evaluation is needed, such as evaluations using IMPROVE aerosol measurement network, AERONET AOD/AAOD measurement network, EPA AirNow network, and/or MODIS AOD data.

Response: Thanks for your suggestion. We have conducted a regional-scale evaluation of BC mass concentration (M_{BC}), O_3, SO_2, other gas concentration, and Aerosol Optical Depth (AOD). The data sources and evaluation results have been included in the newly added Supplementary Material, and a relevant explanation has been added to the main text.

"Figure 3 illustrates the spatial distribution of BC emission on the simulation domain in red and the blue star represents the ground observation station T0 of the Carbonaceous Aerosols and Radiative Effects Study (CARES) campaign, located in Sacramento, California. The observation dataset is available from the Atmospheric Radiation Measurement (ARM) program of the US Department of Energy (DOE) (https://adc.arm.gov/discovery/#/results/s::CARES). This comprehensive campaign collected a diverse array of data, encompassing aerosols, atmospheric conditions, cloud properties, and radiation data. The accuracy of the dataset has been widely recognized (Zaveri et al., 2012; Cahill et al., 2012). Data collected from this site is used as the benchmark for site evaluation and comparison, and the data used in our study are presented in Table 3. The data sources for the model regional-scale evaluation and analysis of results are provided in the supplementary material."

"To evaluate accuracy of the models' regional-scale results, we compared several common variables, as shown in Table S1. These include BC mass concentration (M_{BC}) data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) measurement network, O_3, SO_2, and other gas concentration data from the Air Quality System (AQS) surface dataset, and Aerosol Optical Depth (AOD) data obtained from the Polarization and Directionality of the Earth's Reflectances (POLDER) satellite observations."

Table S1. Performance Evaluation of WRF-CMAQ and WRF-CMAQ-BCG Models.

Results Variables	Observation	WRF-CMAQ				WRF-CMAQ-BCG			
	Mean	Mean	MBE	RMSE	NMB	Mean	MBE	RMSE	NMB
M_{BC} (μg m^{-3})	0.199	0.117	-0.082	0.139	-0.412	0.123	-0.077	0.137	-0.382
O_3 (ppb)	33.391	35.048	1.657	6.370	0.050	35.063	1.672	6.367	0.050
SO_2 (ppb)	1.934	1.740	-0.194	4.202	-0.100	1.740	-0.194	4.202	-0.100
NO (ppb)	1.910	1.952	0.042	2.573	0.022	1.947	0.037	2.573	0.019
NO_2 (ppb)	6.539	7.795	1.256	4.290	0.192	7.787	1.248	4.277	0.191
AOD_{533}	0.143	0.0586	-0.0844	0.146	-0.590	0.0585	-0.0845	0.146	-0.591

"With the Mean, Mean Bias Error (*MBE*), Root Mean Square Error (*RMSE*), and Normalized Mean Bias (*NMB*) metrics presented in Table S1, it can be seen that both the WRF-CMAQ and WRF-CMAQ-BCG models simulated accurately. In addition, the mean error of M_{BC} simulated by the new model is -0.077 μg m^{-3}, which is closer to the observations compared to the original model's values of -0.082 μg m^{-3}, indicating a slight improvement in alignment with the observation (Fig. S1). Overall, the new model, while adding new functionalities and enhancing the accuracy of optical calculations, does not compromise the simulation of other variables and slightly improves the simulation of BC mass concentration."

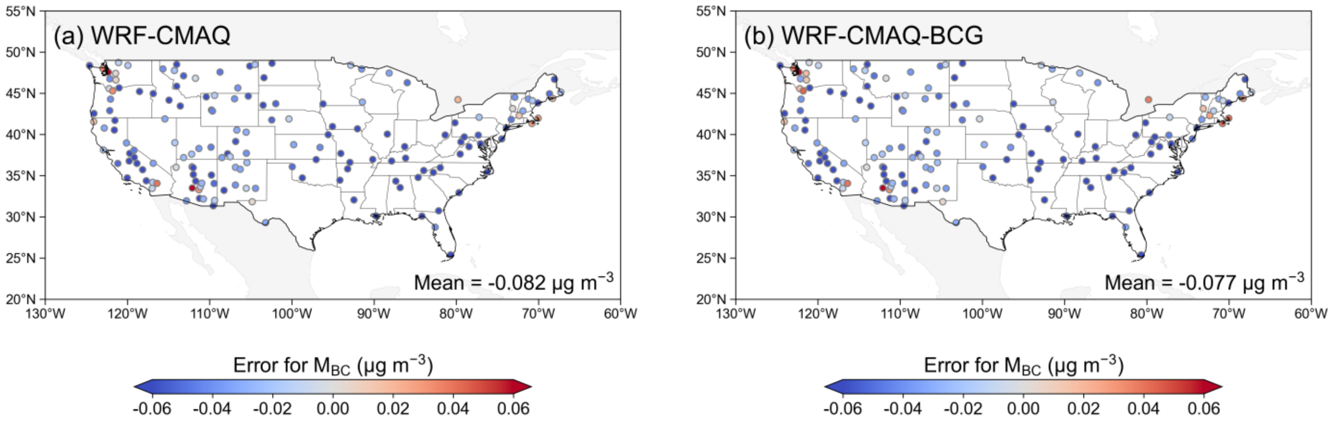

Figure S1: Comparison of BC mass concentration (M_{BC}) errors between simulations and observations: (a) the WRF-CMAQ model, (b) the WRF-CMAQ-BCG model.

Specific comments:

1) Lines 87-88: This "limited impact" is not accurate, since different BC mixing states affect aerosol hygroscopicity and hence wet deposition, which subsequently change the mass concentration.

Response: Thanks for your suggestion, and we apologize for the inaccurate statement. We have revised it to provide a clearer explanation.

"The Community Multiscale Air Quality (CMAQ) model, developed by the US Environmental Protection Agency (EPA), is widely used in the research community as well as in the US government as a regulatory model, and it continues to evolve. To account for the interactive two-way feedback between aerosols and meteorological conditions, Wong et al. (2013) developed the Weather Research and Forecasting - Community Multiscale Air Quality (WRF-CMAQ) two-way coupled model. The mixing state of BC aerosol is simplified to a fully internally mixed state in the CMAQ model. When considering the influence of aerosols on meteorology, this simplification has a more pronounced impact. Therefore, incorporating the BC aging process in the WRF-CMAQ model is essential, as it significantly influences hydrophobicity and light absorption."

2) Line 103: "... coated with scattering aerosol component". This is not very accurate since BC can also be coated by some absorbing organics.

Response: Thank you for your suggestion. We have changed "scattering aerosol component" to "other aerosol component" and added the explanation "(some absorbing organics that coat BC are not considered in this study)" in the second paragraph of the introduction in the revised manuscript.

"The BC component exhibits very low chemical reactivity and is refractory (Bond et al., 2013). Consequently, previous studies have typically considered BC aerosol as chemically inert, primarily serving as a reaction interface for other chemical reactions due to its unique morphology (Monge et al., 2010). However, freshly emitted BC aerosol experiences condensation, coagulation, and heterogeneous oxidation processes during atmospheric transport, becoming coated by scattering aerosol components and converted into aged BC aerosol (some absorbing organics that coat BC are not considered in this study)."

3) Line 108: Even for two-way coupled WRF-CMAQ model, there is no aerosol indirect effect considered? Did the authors mean the standard EPA-version of WRF-CMAQ? There might be a WRF-CMAQ version from individual research group that may already have this capability. Please double check.

Response: Thanks for your suggestion. Yes, we are referring to "the standard EPA-version of WRF-CMAQ." While Yu et al. (2014) studied the indirect effects in WRF-CMAQ, these have not been incorporated into the standard version of the model. This is also an area of ongoing work for us. We have revised the sentence in the manuscript as: "(the EPA's publicly released WRF-CMAQ model does not yet have the indirect radiative effect capability)".

4) In Figures 1 and 2, the authors showed that the model includes aerosol-cloud interaction, but in the description of Section 2.1, it seems that the model does not account for the aerosol indirect effect. This needs further clarification.

Response: Thanks for your comment. We emphasized the impact of clouds on BC aerosol in the figures, particularly the wet deposition caused by in-cloud scavenging. The role of BC aerosol as CCN is indeed not considered in CMAQ. We apologize for any confusion this may have caused and have updated Figures 1 and 2 accordingly (Figure 1 and Table 2).

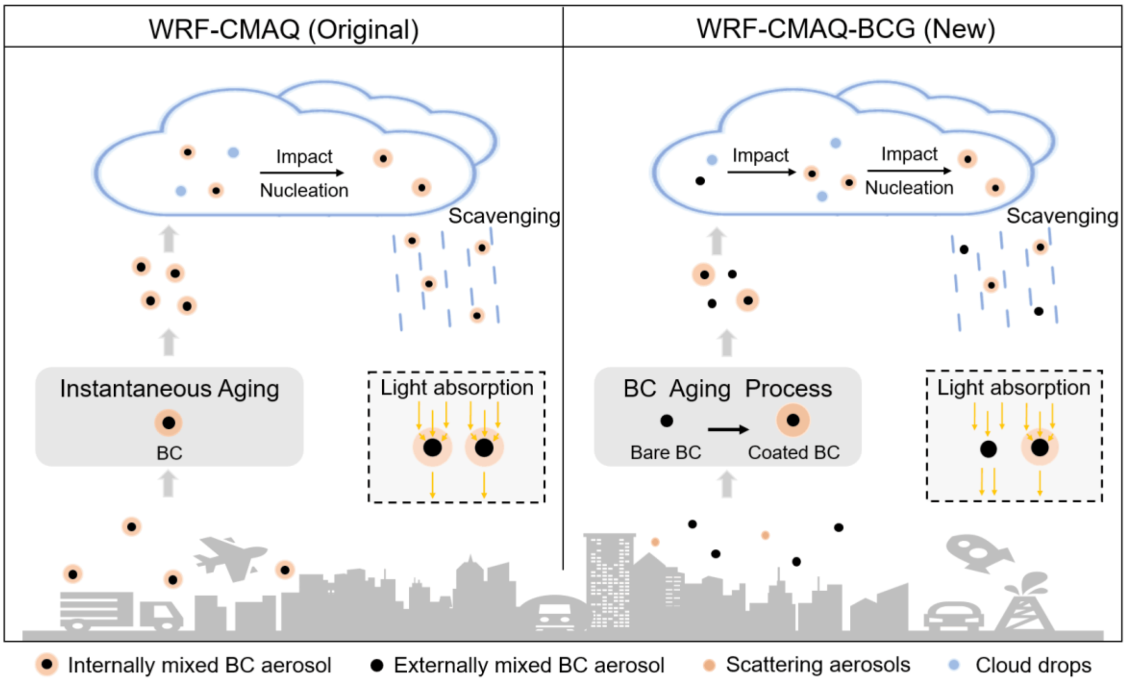

Figure 1: The BC mixing state in the WRF-CMAQ model and the WRF-CMAQ-BCG model.

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

5) Equation (3): Which term is for condensation (fast-aging)? It seems that the first beta[OH] term represents chemical aging and alpha represents coagulation.*

Response: Thanks for your comment. The "β" is for condensation (fast-aging), we have updated the description of the equation in the revised manuscript.

"β is estimated by assuming an e-folding aging timescale of 2.5 days for condensation, and α is estimated by assuming a 20 days e-folding lifetime for coagulation (Liu et al., 2011; Huang et al., 2013; Oshima and Koike, 2013)."

6) Section 2.3: the use of "cloud chemistry" is confusing since BC does not undergo any chemical process in the cloud droplets in the model. Also, the description of this part is not very clear. How did the authors *set the hydrophobicity of coated BC? What scheme did the authors use to compute CCN from aerosol number concentration and hygroscopicity? What equations did the authors use to compute the impact scavenging of BC aerosol? More details are needed.*

Response: Thanks for your suggestions. The term “cloud chemistry module” was used in the CMAQ Users Guide. To avoid confusion, it has been changed to “wet deposition module” in the revised manuscript.

We apologize for the ambiguous description that led to your confusion. We defined “Coated BC” species as hydrophilic and “Bare BC” species as hydrophobic. When these two new species enter the wet deposition module, we modified the primary in-cloud scavenging algorithm based on their differences in hydrophobicity. Since the WRF-CMAQ model does not yet account for aerosol indirect effects, CCN is calculated based on assumptions, and removal rates are computed using cloud water content and precipitation rate. BC in the accumulation mode is removed by nucleation scavenging, while BC in the Aitken mode is treated as interstitial aerosols subjected to impact scavenging (Binkowski and Roselle, 2003). In the CMAQ model, the method for calculating impact scavenging is derived from Binkowski and Roselle (2003), as shown below:

$$E_s = 2\pi\, m_{1c}\, \langle D \rangle\ (1 + 0.5\, Pe^{1/3})$$

where E_s is the scavenging efficiency, m_{1c} is the first moment of the cloud droplet distribution, $\langle D \rangle$ is the polydisperse diffusivity, and Pe is Peclet number.

Within the framework of the WRF-CMAQ model, we modified the wet deposition algorithm to ensure that hydrophobic Bare BC in the accumulation mode is not removed by nucleation scavenging, while Aitken-mode Bare BC undergoes impact scavenging. After impact scavenging, Bare BC is converted to Coated BC due to water envelopment, which continues its removal from the atmosphere. The above additional description has been added in Section 2.3 in the revision.

“In the WRF-CMAQ model, removal rates are computed using cloud water content and precipitation rate. The original model further differentiates scavenging mechanisms based on particle size: BC aerosol in the accumulation mode undergoes nucleation scavenging, while BC aerosol in the Aitken mode experiences impact scavenging as interstitial aerosol (Binkowski and Roselle, 2003). The hydrophobicity changes caused by BC aging process mainly affect nucleation scavenging in the way that hydrophobic Bare BC cannot act as CCN, while hydrophilic Coated BC can. In other words, Bare BC cannot undergo nucleation scavenging. Within the framework of the WRF-CMAQ model, we modified the wet deposition algorithm to ensure that hydrophobic Bare BC in the accumulation mode is not removed by nucleation scavenging, while Aitken-mode Bare BC undergoes impact scavenging. After impact scavenging, Bare BC is converted to Coated BC due to water envelopment, which continues its removal from the atmosphere, as illustrated in right panel of Fig. 1. Overall, these updates in the wet deposition module in this work enhance the representation of BC aerosol in various aspects.”

7) Section 2.4: It is not clear that how the authors compute the number concentrations of bare and coated BC. (1) Are these number concentrations two new prognostic variables tracked by the model? What are the size distributions of bare and coated BC particles used during the mass-to-number conversion? More *clarifications are needed. (2) Another key uncertainty factor related to the calculation of BC optics is the particle structure. Many previous studies have shown that using core-shell assumption for coated BC and spherical shape for bare BC cannot realistically represent BC particle optics (e.g., https://doi.org/10.1029/2021GL096437; https://doi-org.cuucar.idm.oclc.org/10.1021/acs.estlett.7b00418; https://doi.org/10.5194/acp-15-11967-2015). It may be challenging to add the particle structure info into the model, but some discussions on this uncertainty factor will be helpful.*

Response: Thank you for your suggestions.

(1) Bare BC and coated BC are assumed to have the same size distribution, have the same density and volume but are merely in different aging states. Therefore, the number fraction of Coated BC can be calculated from the mass fraction of Coated BC, as shown in Eq.(4). When calculating aerosol optical properties, BC particle size information is highly sensitive. For aerosol optics calculation, the volumes of other species (as the shell) are added to the volume of Coated BC to recalculate the particle size. We have revised the manuscript to reflect this point of view.

"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 properties are calculated using the standard Mie theory. By apportioning the BC core, the overestimation of aerosol light absorption can be corrected.

$$NF_{\text{coated}} = \frac{N_{\text{coated}}}{N_{\text{bare}} + N_{\text{coated}}} = \frac{V_{\text{coated}}}{V_{\text{bare}} + V_{\text{coated}}} = \frac{M_{\text{coated}}}{M_{\text{bare}} + M_{\text{coated}}}, \tag{4}$$

where NF_{coated} represents the number fraction of Coated BC, N_{bare} and N_{coated} are the number concentration of Bare BC and Coated BC aerosol, respectively. V_{bare} and V_{coated} are the volume of Bare BC and Coated BC aerosol, respectively. M_{bare} and M_{coated} are the mass concentration of Bare BC and Coated BC aerosol, respectively. Bare BC and Coated BC are merely in different aging states, they are essentially BC aerosol with the same density and volume. Therefore, the number fraction of Coated BC can be calculated from the mass fraction of Coated BC. "

(2) Thank you for suggesting the addition of a discussion on particle structure. The CMAQ model currently cannot incorporate particle structure information and still uses the spherical shape assumption (Core-shell sphere and Homogeneous sphere), which may introduce some biases. We have included relevant discussion in the revised manuscript.

"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. Each substance has its corresponding refractive index across 14 wavelengths under the Rapid Radiative Transfer Model (RRTM) for global climate model (GCM) applications (RRTMG) scheme. 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."

8)Lines 220-221: How sensitive the model results are to the assumption of equal fraction of bare and coated BC in the initial and boundary conditions?

Response: Thank you for your comment. We used the entire month of May as the spin-up period. With such a sufficiently long spin-up time, the model results are not sensitive to variations in the fraction of bare and coated BC in the initial condition. Similarly, BC aerosol transported over long distances undergo significant aging, justifying the assumption of equal fraction of bare and coated BC in the boundary condition.

9)Line 223: "the model assimilated data" Did the authors mean they also used data assimilation in their model simulations? Did the authors conduct 3 different simulations by using these 3 datasets (FNL, NAM, and NARR), respectively?

Response: Thank you for your comment. The sentence has been corrected in the revised manuscript. We used the Weather Research and Forecasting Data Assimilation (WRFDA) method for data assimilation, with the assimilated data sourced from the North American Mesoscale Forecast System (NAM), provided by the National Weather Service's National Centers for Environmental Prediction (NCEP).

"The WRF model parameterization schemes selected for the simulation case are listed in Table 3. The CMAQ model employs the "cb6r5_AERO7" chemical mechanism and utilizes the Rosenbrock solver. Additionally, we used the Weather Research and Forecasting Data Assimilation (WRFDA) method for data assimilation, with the assimilated data sourced from the North American Mesoscale Forecast System (NAM), provided by the National Weather Service's National Centers for Environmental Prediction (NCEP)."

10)Figures 4-5: It seems that including BC aging only has negligible benefits on model performance. How to better justify the need to include this aging scheme?

Response: Thanks for your comment. The results in Figures 4-5 are intended to demonstrate that the new model, like the original model, can provide accurate meteorological and chemical conditions for considering the BC aging process in this study. We apologize for any confusion caused by our ambiguous explanation, and we have provided a clearer expression in the revised manuscript.

"To evaluate the accuracy of meteorological and chemical conditions for considering the BC aging process in this study, we compared the simulation results of the WRF-CMAQ model and the WRF-CMAQ-BCG model with various meteorological observations, as well as the volumetric concentrations of several gases at the T0 site in the CARES campaign. Figure 3 illustrates…Clearly, the inclusion of the BC aging process does not degrade the original model's accuracy in simulating these common variables and the models can provide reasonably accurate meteorological and chemical conditions for the aging process."

The inclusion of the BC aging process does not degrade the original model's accuracy in simulating some common variables. Instead, it adds new functionalities to the WRF-CMAQ model, such as simulating BC mixing state distributions, tracking BC aging timescales, and separately analyzing the wet deposition of BC with different mixing states. Additionally, it enhances the model's performance in simulating BC aerosols, including their mass concentration and optical properties. These points illustrate the necessity of considering the BC aging process in the WRF-CMAQ model.

References

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