Response to the comments of Editor (egusphere-2024-2372)

The authors revised their manuscript substantially addressing most reviewers concerns. While the BC aging topic has been extensively studied in the past decade and some of the scientific knowledge and processes included in this study is not new (which are some of the remaining major concerns from reviewers), it is new to the WRF-CMAQ coupled model. It is important to advance the WRF-CMAQ modeling capability on this aspect given the wide use of this model and thus I agree to continue considering the manuscript for publication on ACP. Authors, please revise the manuscript for technical correction according to some of the remaining reviewers comments:

Response: We sincerely thank the editor for recognizing and supporting our work. Please find our point-by-point responses listed below. The reviewers' comments are in *Italic* followed by our responses and revisions (in blue). The modifications in the manuscript are highlighted in red.

The reviewers' comments:

 # The only one minor thing that requires a further clarification is their response to my previous specific comment #9: The authors mentioned that they used WRFDA to assimilate NAM data. What variables did they assimilate? How frequently did they assimilate (e.g., every 6 hours)? Why did they do the DA using NAM instead of observations? Did the authors mean "meteorological nudging" instead of DA? Some clarification will be useful.

Response: Thank you for pointing out the need for clarification regarding our data assimilation approach. FDDA algorithm is part of the EPA physics package incorporated into the WRF structure. This algorithm is based on NAM data with a 3-hour frequency. NAM data were chosen for their spatial and temporal continuity across the simulation domain, ensuring consistent and high-quality boundary and initial conditions to better constrain the model dynamics. The control of nudging as well as the strength of nudging is done in the namelist. In the WRF-CMAQ coupled model scenario, the strength is set to very low value (Gilliam et al., 2012) to let the dynamics do its work. Horizontal winds (U and V), temperature (T), and water vapor mixing ratio (Q) are the nudging variables.

"Additionally, we applied the Four-Dimensional Data Assimilation (FDDA) technique within the WRF model framework to improve the meteorological fields during the simulation. Specifically, we used analysis nudging to assimilate with a 3-hour frequency. The assimilated data were sourced from the North American Mesoscale Forecast System (NAM), provided by the National Weather Service's National Centers for Environmental Prediction (NCEP). The nudging variables included horizontal winds (U and V), temperature (T), and water vapor mixing ratio (Q). In the WRF-CMAQ coupled model scenario, the nudging strength was set to a very low value (Gilliam et al., 2012) to minimize interference with the model dynamics while constraining the meteorological fields."

 # The sentence '(coarse mode aerosols without BC component are not considered in this study)' has been added. Are particles without BC explicitly considered in the accumulation and Aitken modes? As I pointed out in my previous comment, considering particles without BC is important. If particles without BC are not considered, BC coating is overestimated, and this has a large impact on the lifetime and optical properties of BC.

Response: Thank you for your suggestions. This work primarily investigates the impact of BC aging process on BC aerosol. Consequently, non-BC particles (particles without BC) in the Aitken and accumulation modes are not included in our analysis. We acknowledge that this omission may introduce some bias into our results. In our recent work, we have explored the impact of non-BC aerosols on aerosol optics in the CAM6-MAM4 model (Chen et al., 2023). As illustrated in Fig. R1, the Mass Absorption Cross-section (MAC) values of aged BC simulated by CAM6-ABC have undergone significant changes. These modifications, which include adjusting the BC core size and distinguishing non-BC particles, among other effects, have resulted in a substantial decrease in MAC values from approximately 13 $m^2 g^{-1}$ in the original model to about 8 $m^2 g^{-1}$ in the revised version. Extrapolating from this trend (see Fig. R2), we estimate that the MAC values simulated by our aging model could potentially decrease further, from around 9 $m^2 g^{-1}$ to approximately 8 $m^2 g^{-1}$. It is important to note that WRF-CMAQ is a two-way coupled meteorology - air quality model with complete internal mixing of aerosols assumption. Consequently, the presence of non-BC particles may significantly influence various atmospheric processes beyond those directly related to BC aging. In light of these considerations, we intend to conduct further investigations into the role of non-BC particles within the WRF-CMAQ framework in our future research. We sincerely appreciate your invaluable input, which has prompted us to incorporate a relevant discussion in the revised manuscript.

"In our WRF-CMAQ-BCG model, we calculated the optics of Bare BC and Coated BC separately in the Aitken and accumulation modes, without accounting for the presence of scattering aerosols independently. This omission may result in an overestimation of the coating thickness, potentially introducing some bias into the optical simulation results (Chen et al., 2023)."

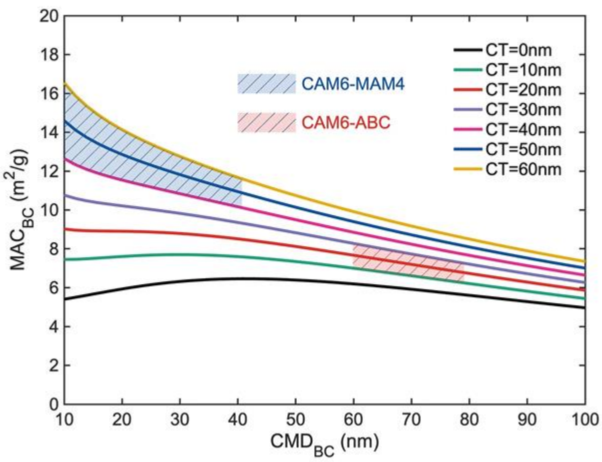

Figure R1: Comparisons of BC MAC (MAC_{BC}) between the default model (CAM6-MAM4) and the new model (CAM6-ABC) in different BC size distributions (coating thickness (CT)). (cited from Chen et al., 2023)

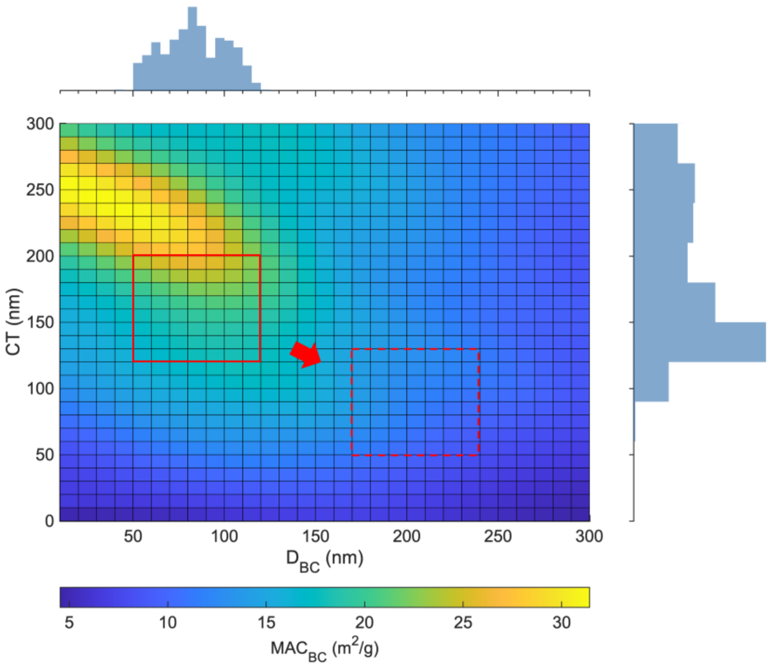

Figure R2: Schematic of the trend in the change of MAC values after considering non-BC particles (Note: Unlike the MAC results presented in the paper, this figure does not include Bare BC and Aitken mode).

#Coagulation is important, so I do not recommend using a constant aging value with no reasons/discussion.(Please see Riemer et al., 2019 and references therein).

Response: Thank you for your suggestion. As we mentioned in our previous response, we agree that coagulation is an important process in BC aging near sources. However, within the context of mesoscale modeling, the effects of coagulation on aerosol dynamics are generally considered to be of minor importance compared to condensation.

Riemer et al. (2010) demonstrated that even under extreme conditions highly conducive to coagulation in a supersaturated environment (with supersaturation levels ranging from 0.1% to 1%) with high concentration of aerosols, the impact of coagulation on BC aging remained less significant than that of condensation. This was evidenced by the stark contrast in aging time-scales: in condensation-dominated daytime environments, aging occurred much more rapidly (11 to 0.068 h) compared to coagulation-dominated nighttime environments (54 to 6.4 h). These findings underscore that condensation plays a more substantial role in accelerating the BC aging process. Zaveri et al. (2010) used the particle-resolved PartMC-MOSAIC model to study aerosol mixing state evolution in an idealized urban plume. Their results (Fig. R3) showed slightly higher mass concentrations without coagulation, indicating that coagulation only has a minor effect. Oshima et al. (2009) and Doran et al. (2008) also support this conclusion.

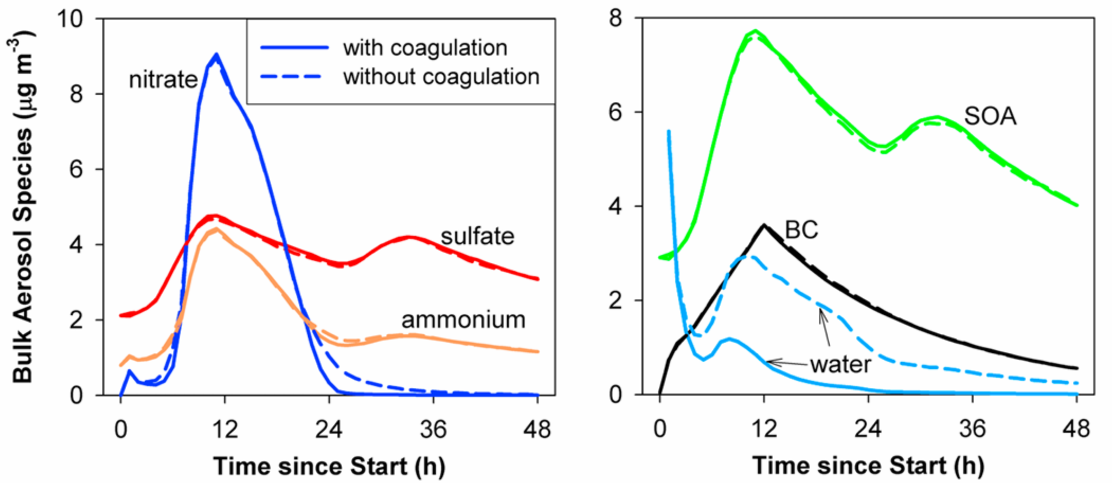

Figure R3: Comparison of aerosol mass concentration with coagulation (solid) and without coagulation (dashed). Evolution of bulk aerosol species in the urban air parcel as it is advected downwind over a period of 2 days. (cited form Zaveri et al., 2010)

Given the above studies, we have treated the condensation-related terms in the aging rate as fast-aging term and coagulation as a slow-aging term, simplifying it to a constant, as done in studies by Liu et al. (2011), Huang et al. (2013), Oshima and Koike (2013), and others. In the future, we will explore using variables to represent the rates of coagulation and condensation processes to obtain a more accurate BC aging rate. We have added the requested reasons/discussion in the description of Eq. (3) and cited the relevant papers.

"This study employs a BC aging module that quantifies the BC aging rate using an equation dependent on the concentration of OH radicals, as shown in Eq. (3). Condensation is considered through the setting of a fast-aging term, while coagulation is considered through a slow-aging term. Although coagulation can occur rapidly near sources, it does not play a dominant role within the context of mesoscale modeling compared to condensation (Doran et al., 2008; Oshima et al. 2009; Riemer et al., 2010; Zaveri et al., 2010). …

$$k = \beta\,[\mathrm{OH}] + \alpha\,, \qquad (3)$$

where k represents the aging rate, [OH] represents OH radical concentration. β and α are assumed to be constant, with values 4.6×10^{-12} $\mathrm{cm^3\ molecule^{-1}\ s^{-1}}$ and 5.8×10^{-7} $\mathrm{s^{-1}}$, 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). Indeed, this constant assumption does not account for the dynamic variations of the coagulation and condensation processes, which could introduce some bias."

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