Response to the comments of Reviewer #2 (EGUSPHERE-2024-1924)

Reviewer #2: The manuscript "Steady-State Mixing State of Black Carbon Aerosols from a Particle-Resolved Model" by Zhang et al., investigates the mixing state of black carbon aerosols. Their results indicate, based on both a particle-resolved model and observations in Nanjing that the mixing state of BC aerosol reaches a steady state in few hours. The results of the paper are interesting and can be useful for improving the treatment of mixing of BC with scattering compounds in atmospheric models. However, the current study does not provide yet the means to apply this finding in models. The paper in within the scope of ACP, it presents novel ideas, reaches substantial conclusions. The paper is well written and I can recommend accepting it for publication after the following issues are addressed.

Response: Thanks to reviewer #2 for the insightful suggestions and comments. In response, we have restructured the manuscript and have included detailed, point-by-point responses to all comments and questions raised. The reviewer's comments are presented in italics, followed by our responses and revisions highlighted in blue. In the revised manuscript, we have elaborated on the application methods and potential fields of application for the study's results in response to Main Comment 1. Additionally, we have clarified the description of the gas setup on Page 4, Lines 105-108, to provide a clearer introduction to the setup, addressing Main Comment 2. Furthermore, we have added legends explanations for curves and points in the caption of Figures S2 and S3, in response to Technical Comment 1. We contend that these modifications, prompted by your valuable feedback, have significantly enhanced the coherence and readability of the article. Here are our point-to-point responses.

Main Comments:

1. My main comment is related to how to apply this knowledge in atmospheric modelling. I assume that since this holds only near emission sources, parameterizing the mixing state should be embedded in emission schemes, right? Away from the sources and higher up in the atmosphere where there are no emissions and surface removal, such steady state assumption may not hold. It would be good to add some discussion about this.

Response: Thank you for your comment. We discuss the potential utility of steady-state theory for modeling efforts and its application scopes in the "4. Discussion" of the revised manuscript. To be brief, the characteristic timescale for BC approaching a steady-state mixing state is significantly shorter than its atmospheric lifetime, suggesting that the properties of BC under the steady state can effectively represent the averaged properties of BC across extensive spatial and temporal scales. The parameter k denotes the distribution of coating thickness ($n(\mathrm{CT}) = kN \cdot e^{-k \cdot \mathrm{CT}}$) under steady state, obtained from the growth rate and removal rate, thus efficiently helping calculate the light absorption capacity of BC aerosols. Regarding the application of the slope k in models, we propose utilizing existing state values in models, such as the total mass concentration of each component combined with the distribution of coating thickness and diameter of BC-core, to build a parameterized framework of the k value based on the studies by Chen et al. (2023, 2024), which is an avenue for future research. Furthermore, we plan to employ machine learning techniques to develop an emulator for the k value based on training data obtained from PartMC-MOSAIC. In this approach, the k value will serve as the "label", while emissions, initial conditions, and meteorological conditions will be treated as "features". These tasks are avenues for our future work.

We appreciate your interpretation of the applicability of the steady-state assumption, particularly the point that it 'holds only near emission sources, and parameterizing the mixing state should be embedded in emission schemes.' In the revised manuscript, we have provided a detailed interpretation to fully capture the nuances of the assumption's applicability in various conditions. To be more precise, the steady-state assumption can be applied to the analysis of the average properties of BC across extensive spatial and temporal scales. According to the region away from the sources and higher up in the atmosphere where there are no emissions and surface removal, t the BC mixing state is considered to have reached a steady state. In determining the k value, the emissions and removal of BC aerosols can be represented by their transport dynamics, including both inflow and outflow, within the region.

We have added the description of the application of our findings in models in the revised manuscript (Lines 348-370):

"We discuss the potential utility of steady-state theory for modeling efforts and its application scopes. The slope parameter k denotes the CT distribution of BC aerosols $n(CT) = kN \cdot e^{-k \cdot CT}$ under steady state. According to the derivation presented by Wang et al. (2023), the value of 1/k was

determined to be equivalent to the average CT of BC aerosols (Table S3). The relationship between E_{abs} and CT is approximately linear when CT is less than 200 nm ($k > 0.005\ nm^{-1}$) (demonstrated in the Fig. S3 of Wang et al., 2023). Therefore, a monodisperse CT, 1/k, can replace the BC coating thickness distribution when calculating the BC absorption. Fig. 5 in this paper shows that the E_{abs} based on the steady-state theory can serve as a characterization of the light absorption enhancement of BC aerosols under the steady-state condition. The k-value can be obtained from the growth rate and removal rate (k = Dep/GR), thus efficiently helping evaluate the BC absorption. Regarding the application of the slope k in models, we propose utilizing existing state values in models, such as the total mass concentration of each component combined with the distribution of coating thickness and diameter of BC-core, to build a parameterized framework of the k value based on the studies by Chen et al. (2023, 2024), which is an avenue for future research. Furthermore, we plan to employ machine learning techniques to develop an emulator for the k value based on training data obtained from PartMC-MOSAIC. In this approach, the k value will serve as the 'label', while emissions, initial conditions, and meteorological conditions will be treated as 'features'. This method is inspired by the study by Zheng et al. (2021), which used the matrix χ as the 'label' for their emulator. Further, the characteristic timescale for BC reaching a steady-state mixing state ranges from 1.9 to 9.7 hours, which is considerably shorter than their atmospheric lifetime, typically around 7 days. Thus, the steady-state assumption may effectively be applied to climate models, which are generally concerned with the characteristics of BC aerosols across extensive spatial and temporal scales. While it may function less well for high-resolution modeling studies of urban areas wherein a larger percent of BC emissions are fresh, in which the steady state may be influenced. In these conditions, we propose categorizing BC-containing particles into two distinct types: fresh BC, which lacks a coating layer, and aged BC, which possesses a coating layer. The slope parameter k is capable of characterizing the distribution of the latter. Hence, the k-value method can be applied in models for BC absorption under the steady-state condition."

2. Page 3, Line 97: "In this study, the initial gas concentration and emission rate were slightly adjusted based on Riemer et al. (2009) according to Wang et al. (2017)." The meaning of the sentence is unclear. What was the initial assumption for gas concentrations and emissions and how were they adjusted? Were these assumption tuned to match the model with observations?

Response: Thank you for your reminder. The previous sentence may be deemed somewhat misleading. We employed the gas setup of Riemer et al. (2009) within PartMC, as Riemer et al., being the developers and experienced users of PartMC, have established a gas setup that is both authoritative and widely adopted in subsequent. To better reflect the aerosol composition characteristic of Chinese conditions, we subsequently adjusted the gas setup based on the field observations in China by Wang et al. (2017). To more precisely communicate our intended message, we have revised the original statement as follows: [Page 4, Lines 105-108]

"In this study, the initial gas concentration and emission rate were established in accordance with the parameters set by Riemer et al. (2009). To reflect the typical composition of aerosols in China, the setup for the baseline case was subsequently adjusted based on the observations reported by Wang et al. (2017), as detailed in Table S1 of the Supplement."

*"**Table S1.** Input variables assigned in the baseline case"*

Environmental variable	**Value**
Temperature [K]	289
Relative humidity	0
Boundary layer height [m]	293.14
Mass loss (deposition)	constant
Latitude	0 °N
Day of year	July 19
Aerosol characteristic	**Value**
Emission rate [m^{-2} s^{-1}]	1.8×10^{7}
Fraction Bare-BC emissions	24.6%
Fraction Mix-BC emissions	2.4%
Fraction BC-free emissions	72.7%
Aerosol type	**Geo. mean dia. [nm]**
Bare-BC	89
Mix-BC	109
BC-free	110
Aerosol type	**Geo. standard dev.**
Bare-BC	1.60
Mix-BC	1.60
BC-free	1.70
Aerosol type	**Mass composition**
Bare-BC	100% BC
Mix-BC	68.3% BC, 31.7% OC
BC-free	100% OC
Emitted gas species	**Rate [mol · m^{-2} · s^{-1}]** with a total multiplication factor of 25 %
Sulfur dioxide	2.51×10^{-8}
Nitrogen dioxide	1.20×10^{-9}
Nitric oxide	2.50×10^{-8}
Ammonia	6.11×10^{-9}
Carbon monoxide	2.91×10^{-7}
Acetaldehyde	6.80×10^{-10}
Formaldehyde	1.68×10^{-9}
Ethene	7.20×10^{-9}
Internal olefin carbons	2.42×10^{-9}
Terminal olefin carbons	2.42×10^{-9}
Toluene	4.04×10^{-9}
Xylene	2.41×10^{-9}

Acetone	1.23×10^{-9}
Paraffin carbon	9.60×10^{-8}
Isoprene	2.30×10^{-10}
Methanol	2.80×10^{-10}
Alcohols	3.45×10^{-9}

3. Was the motivation for the additional cases to show that for all conditions, the exponential linear distribution occurs?

Response: Thank you for your question. Yes, we aim to convey that the exponential linear distribution universally applies (occurs in all conditions), although the additional cases may not encompass all conditions. To improve the generalizability of our findings, we varied the temperature, the size distribution of freshly emitted BC, and the particle-gas emission ratio, constructing additional cases. We can simplify the calculation of BC absorption by applying the exponential linear distribution. The exponential linear distribution of CT can be represented by a single slope parameter k ($n(\mathrm{CT}) = kN \cdot e^{-k \cdot \mathrm{CT}}$). According to the derivation presented by Wang et al. (2023), the value of $1/k$ was determined to be equivalent to the average CT of BC aerosols (Table S3). The relationship between E_{abs} and CT is approximately linear when CT is less than 200 nm ($k > 0.005$ nm^{-1}). This linear behavior is clearly illustrated in Fig. 1, which corresponds to Figure S3 in Wang et al. (2023). Therefore, a monodisperse CT, $1/k$, can replace the BC coating thickness distribution when calculating the BC absorption.

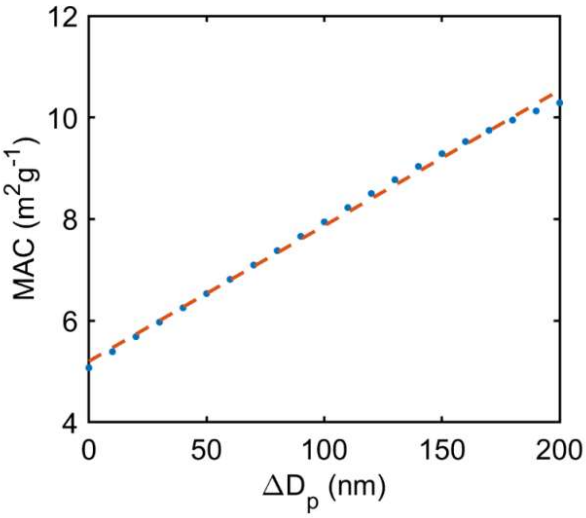


Figure 1. Change of mass absorption cross-section (MAC) of black carbon (BC) with coating thickness (ΔDp). Blue dots represent the calculated MAC based on core-shell Mie theory with 5 the linear fit shown as the red line.

Besides, we have incorporated three additional cases into the revised manuscript to address the previously insufficient representation of natural biomass burning black carbon, as pointed out by Reviewer #1.

The geometric mean diameter of freshly emitted BC was set as 150 nm based on Fig. 2, which corresponds to Figure 4 of Bond et al.(2013), and varying particle-gas emission ratios. In these three new scenarios, the CT distribution follows an exponential linear distribution, with a steady-state timescale ranging from 2.1 to 2.8 hours.

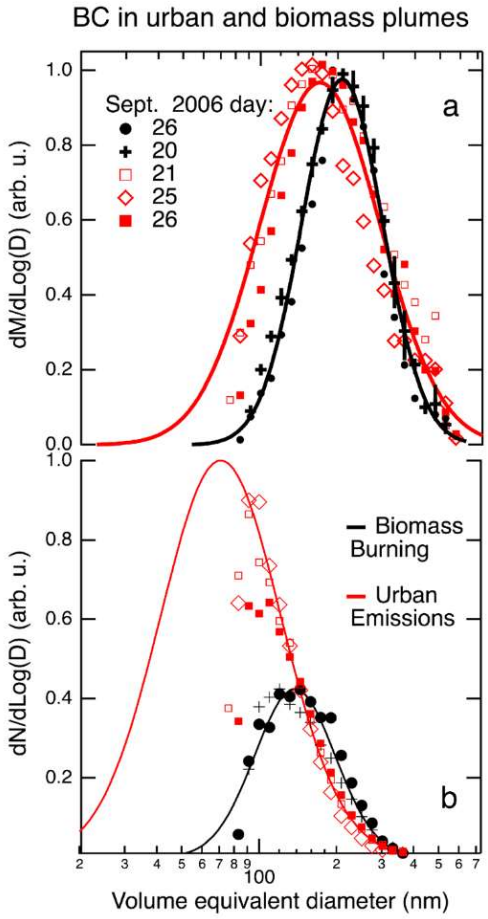
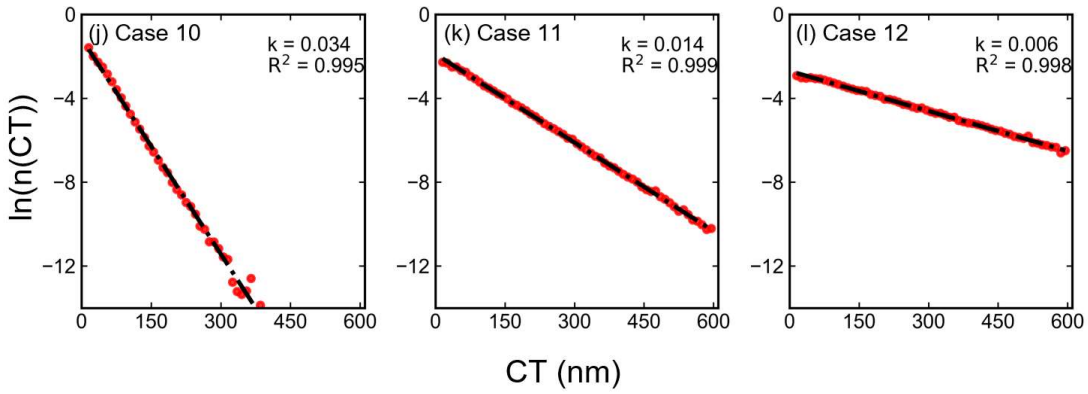


Figure 2. Mass and number size distributions of BC particles observed in three fresh urban (red) and two fresh biomass burning (black) plumes as identified in the legend. The measurements are made on board an aircraft using an in situ, single-particle detection instrument (SP2). The variable coatings on the BC particles are not shown. The observed (a) mass and (b) number amounts are plotted as symbols versus volume equivalent diameter based on assuming a spherical particle shape. The mass distributions are normalized to the same peak value. The observations are fit by a lognormal function between 90 and 600 nm (solid lines). The number distribution fits are those consistent with the fit to the respective mass distribution and are scaled to represent the same BC mass. From Schwarz et al. [2008b].

The three new cases are detailed in Sect. 1.2 of the Supplement, with their CT distributions shown in Figures S3 (j), (k), and (l), and steady-state characteristic timescales listed in Table S3 of Supplement.



"Part of Fig S3. Results of CT distribution of BC aerosols in 12 simulation cases (excluding the Baseline case)."

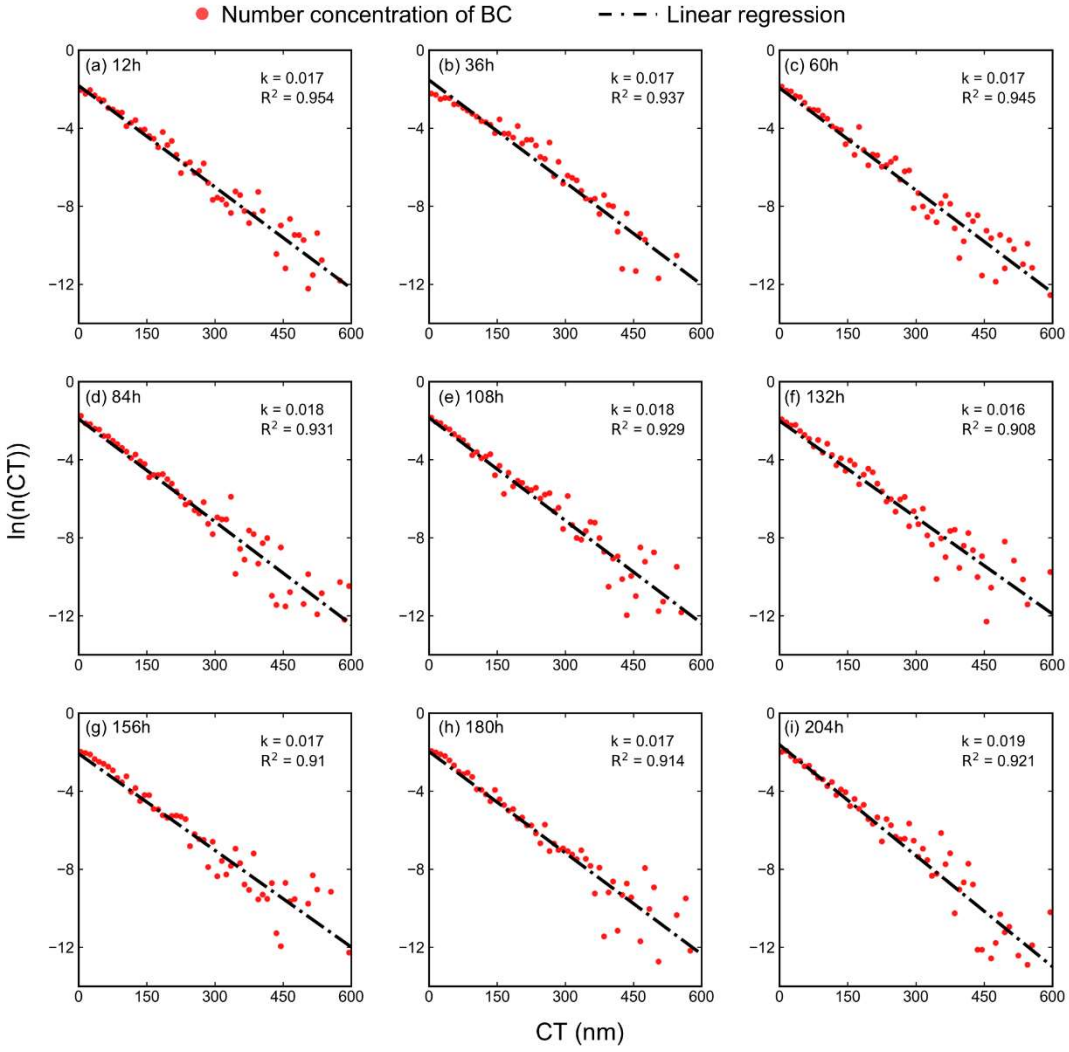
"Part of Table S3. The characteristic time τ, equivalent CT (1/k), coefficient of determination of linear regression, and the average true CT value for ten different cases."

Scenario	***Characteristic time (hour)***	***Equivalent CT (nm)***	***Coefficient of determination***	***Average true CT (nm)***
Baseline case	*3.2*	*62*	*0.999*	*63*
Case 10	*2.8*	*29*	*0.995*	*29*
Case 11	*2.1*	*71*	*0.999*	*76*
Case 12	*2.2*	*157*	*0.998*	*162*

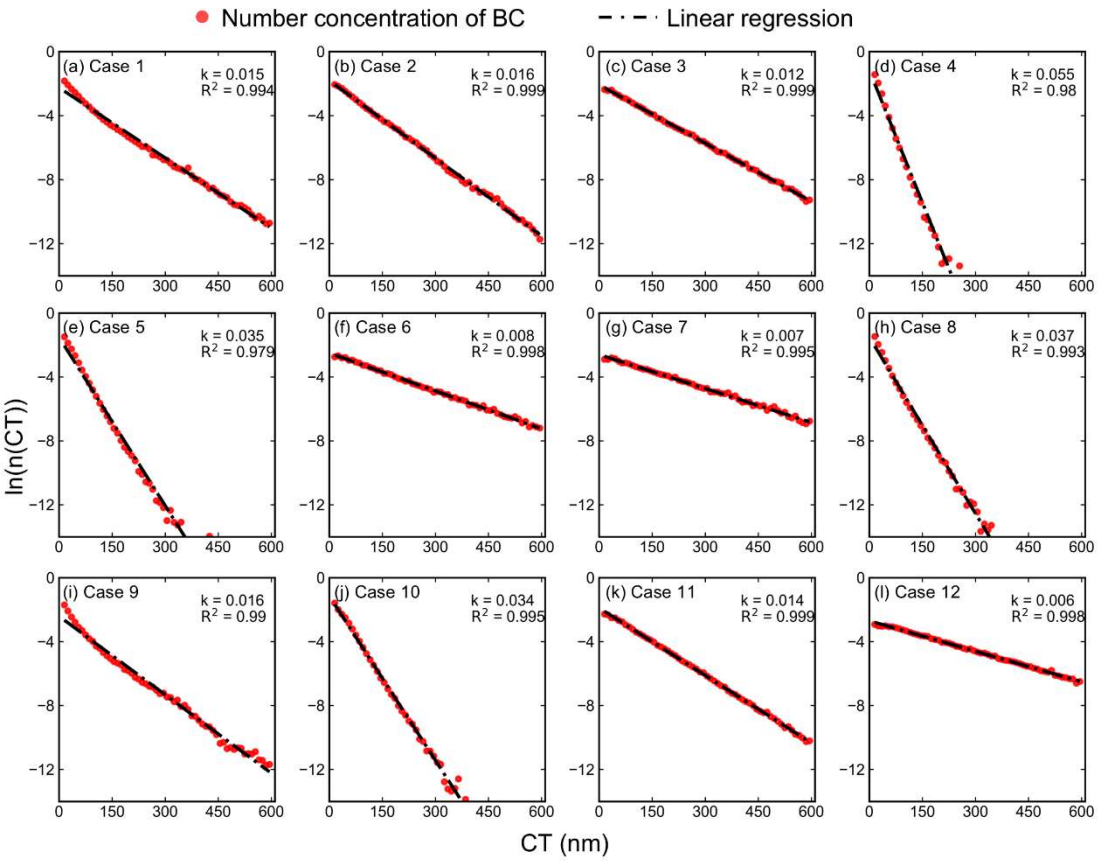
Technical comment:

1. Please add legends or explanations for curves and points in Figures S2 and S3

Response: Thank you for your reminder. We have added legends explanations for curves and points in the caption of Figures S2 and S3. [Page 5 and 7]



"Figure S2. *Coating thickness (CT) distribution of black carbon (BC) aerosols in the baseline case during simulation progress. Each subfigure shows the CT distribution for different simulation times (from 12 hours to 204 hours, with an interval of 24 hours). Each dot indicates the total number of particles that are in the CT interval and the linear regression of each distribution is represented by dashed lines. Under the combined influence of multiple atmospheric processes, the CT distribution rapidly exhibits an exponential linear distribution. As the simulation progresses, the BC mixing state remains steady (the overall correlation of linear fitting maintains a high value)."*



*"**Figure S3.** Results of CT distribution of BC aerosols in 12 simulation cases (excluding the Baseline case). The subfigures correspond to the results of cases 1 to 12. Each subfigure shows that the statistical CT distribution of BC aerosols follows the exponential linear distribution during the steady-state period (after 48 hours) simulated by PartMC-MOSAIC. Each dot indicates the total number of particles that are in the CT interval and the linear regression of each distribution is represented by dashed lines. The slope k = Dep/GR is determined by the deposition rate (Dep) and the growth rate (GR) (Wang et al., 2023). Temperature, aerosol and gas emissions can affect the GR, thereby indirectly affecting the k value. In the setup of the simulation cases, we fixed the deposition rate and altered temperature, emissions of particles and gas to change the growth rate. By comparing Case 1 (a) and Case 2 (b), we found that temperature has little effect on the k value (the higher the temperature, the smaller the k value); by comparing Case 3 (c) and Case 4 (d), we discovered that the larger median diameter of emitted BC-core brings a larger k value. Through the comparison of Case 5 to 8, we observed that a higher gas emission rate or a lower particle emission rate leads to a smaller k value (thicker coating). In Cases 10 to 12, which involve natural biomass BC emissions, the CT distribution also followed an exponential linear distribution."*

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