Response to comments from reviewers on "Modeling simulation of aerosol light absorption over the Beijing-Tianjin-Hebei region: the impact of mixing state and aging process" by Huiyun Du et al.

We thank the reviewer for their valuable comments and constructive suggestions. We have revised the manuscript according to the suggestions and responded to their concerns below (in blue).

Reviewer #1

This manuscript investigated the influences of mixing state on BC light absorption properties, mainly based on simulations. Different scenarios were designed, and the modeling results were constrained using field observational data. Although the topic of this manuscript is within the scope of ACP, I have substantial concerns on the methodologies as well as some results. It needs to be re-reviewed after major revisions.

First, the estimation of POC and SOC using the EC-tracer method. In the current manuscript, the minimum OC to EC ratio (1.16) was used to represent primary emissions (Page 4, equation 1). This approach needs to be refined, e.g., by using the lowest 10 % percentile of ambient OC/EC ratios (please refer to https://doi.org/10.1029/2008JD010902; Atmos. Chem. Phys., 15, 2969–2983, 2015; etc.).

Response: We acknowledge that this approach may require further refinement. To improve the accuracy of our methodology, we use the lowest 10% percentile of ambient OC/EC ratios, as suggested in the referenced study (Zheng et al., 2015; Lin et al., 2009). We appreciate your guidance on this matter and have revised our methods accordingly.

The figure below shows the estimated intercept and slope, i.e., values of $(OC/EC)_{pri}$, by using the lowest 10% percentile of ambient OC/EC ratios (about 70 data). Due to the change in the estimation of POC and SOC, corresponding modifications have been made to Figure 3 and Figure 5.

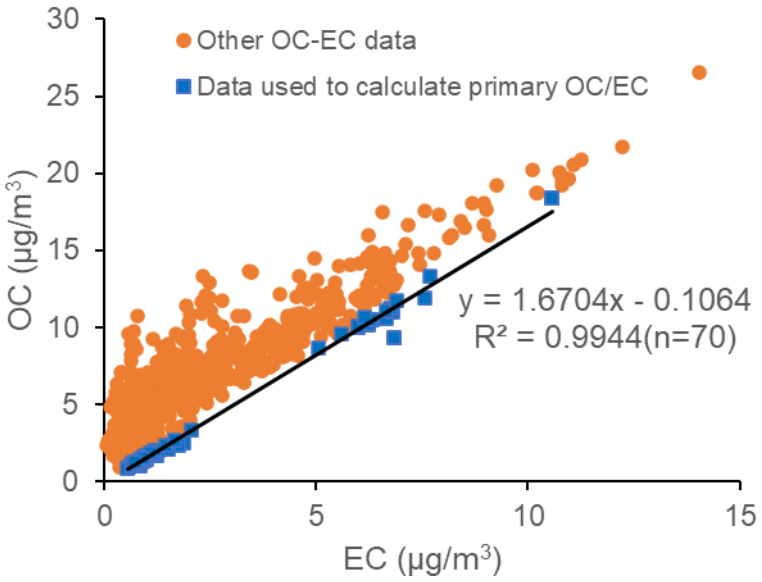


Figure 1. Estimation of SOC with EC-tracer method. Blue squares indicate data used to calculate primary OC/EC, while orange-filled circles indicate other OC/EC data.

Zheng, G. J., Duan, F. K., Su, H., Ma, Y. L., Cheng, Y., Zheng, B., Zhang, Q., Huang, T., Kimoto, T., Chang, D., Pöschl, U., Cheng, Y. F., and He, K. B.: Exploring the severe winter haze in Beijing: the impact of synoptic weather, regional transport and heterogeneous reactions, Atmos. Chem. Phys., 15, 2969-2983, 10.5194/acp-15-2969-2015, 2015.

Lin, P., Hu, M., Deng, Z., Slanina, J., Han, S., Kondo, Y., Takegawa, N., Miyazaki, Y., Zhao, Y., and Sugimoto, N.: Seasonal and diurnal variations of organic carbon in PM2.5 in Beijing and the estimation of secondary organic carbon, Journal of Geophysical Research: Atmospheres, 114, https://doi.org/10.1029/2008JD010902, 2009.

Change in the manuscript:

OC can further be classified into primary organic carbon (POC) and secondary organic carbon (SOC) using the elemental carbon (EC) tracer method (Castro et al., 1999; Zhao et al., 2013). A refined EC tracer method was proposed (Zheng et al., 2015; Lin et al., 2009), and data from the lowest 10% percentile of ambient OC/EC ratios was utilized to estimate the primary OC/EC ratio with the following equations (Figure S1).

$$POC = EC \times (OC/EC)_{pri} + N \quad (1)$$

$$SOC = OC - POC \quad (2)$$

Please refer to Lines 104-109, Line 222, Lines 253-255, Line 277.

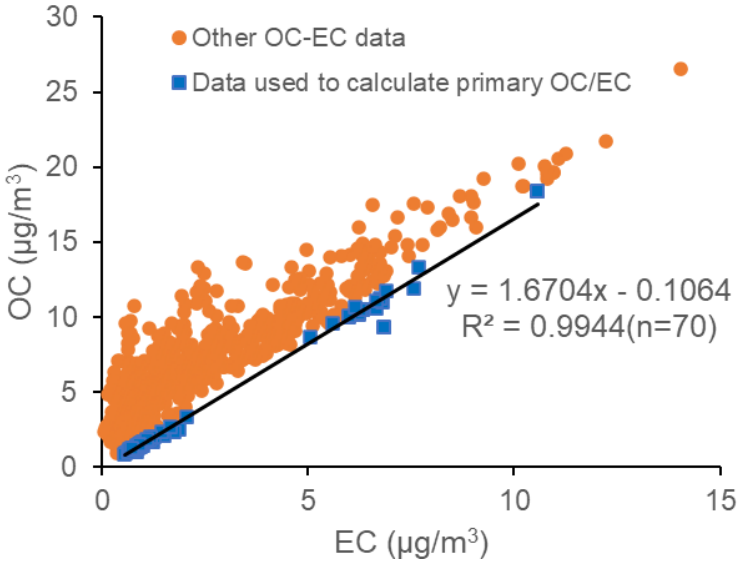


Figure S1. Estimation of SOC with EC-tracer method. Blue squares indicate data used to calculate primary OC/EC, while orange-filled circles indicate other OC/EC data.

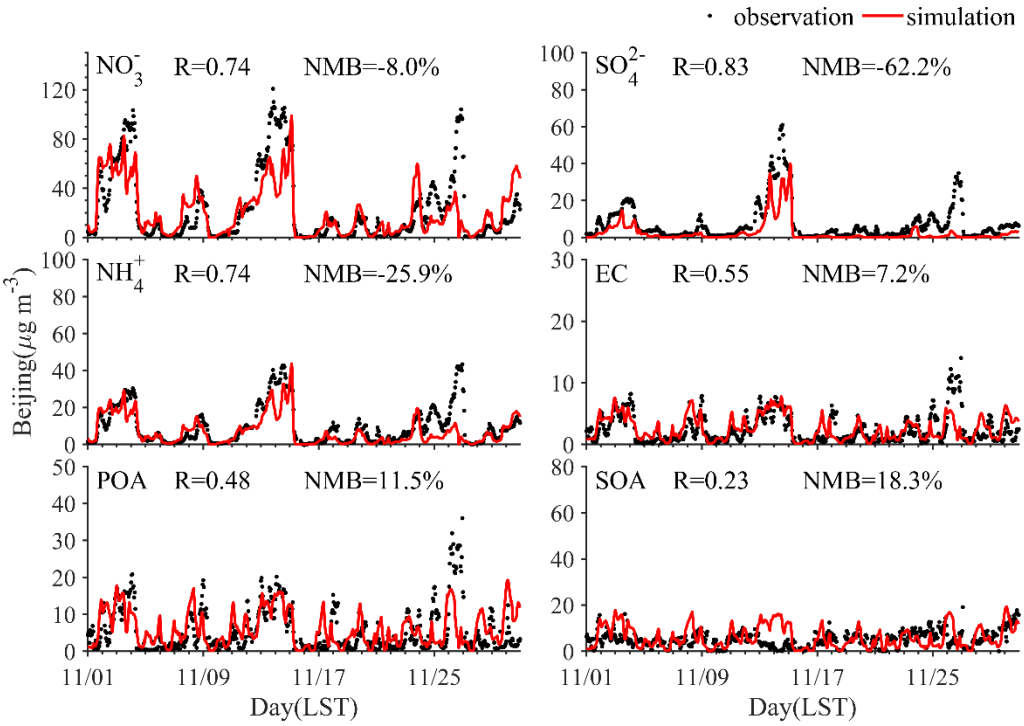


Figure 3. Simulated and observed $PM_{2.5}$ components in Beijing

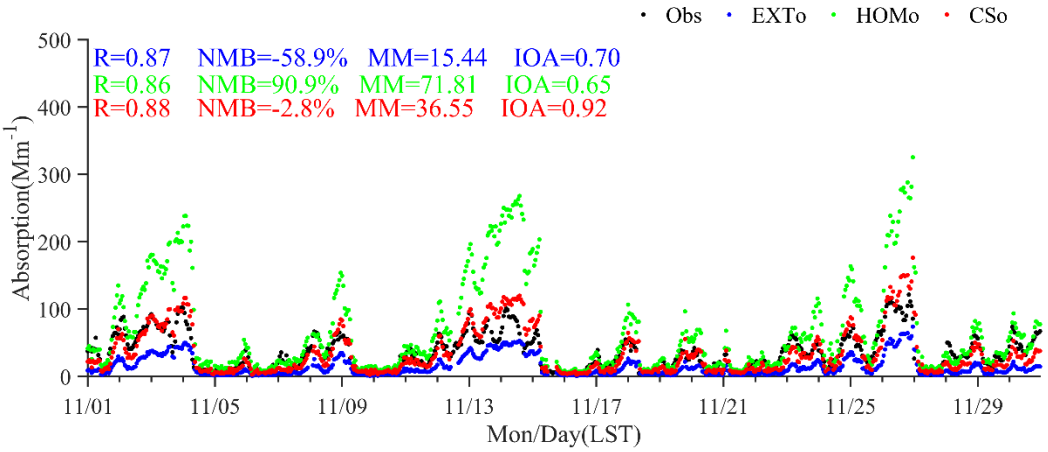


Figure 5. Observed and calculated absorption coefficients at 880 nm under external mixing (EXTo), homogeneous internal mixing (HOMo), and core-shell mixing (CSo) by FlexAOD based on observed components.

In addition, the POC and SOC results should be carefully evaluated, e.g., by examining the dependence of SOC on RH, and the relationship between POC and carbon monoxide.

Response: We thank the reviewer for constructive advice. I appreciate the emphasis on thoroughly evaluating the POC and SOC results.

Firstly, we plot the concentrations of SOC against different RH levels (Figure 2) and analyze the resulting trend. It reveals that SOC levels initially rise with increasing relative humidity but subsequently decline with RH, suggesting underlying chemical or physical processes. And the result is consistent with Wang et al., (2024, Fig.4) and Zheng et al., (2015, Fig. 9b).

We also examined the relationship between POC and carbon monoxide (Figure 3). It reveals that POC is highly correlated with CO, with an R^2 of 0.8.

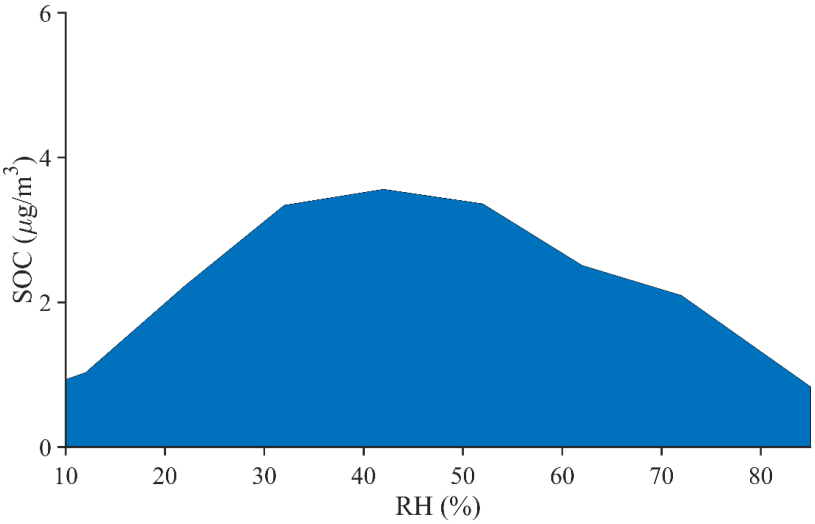


Figure 2 Change of SOC with RH

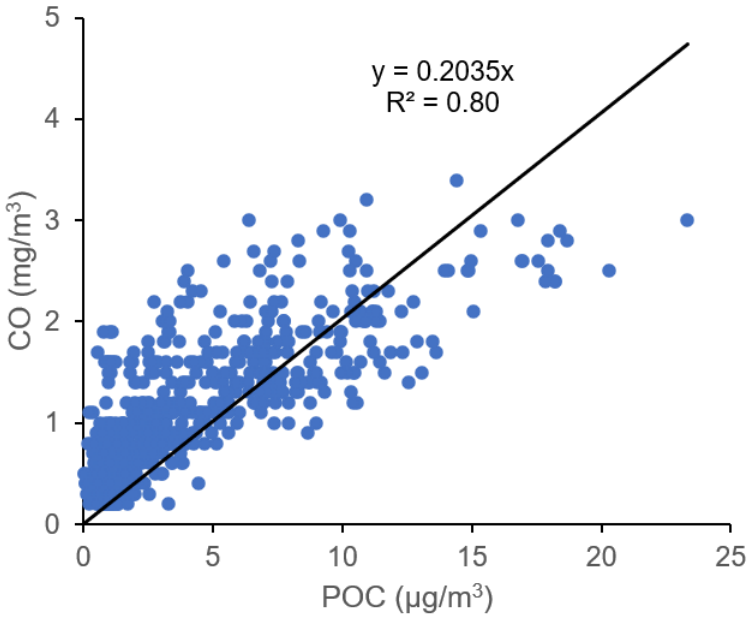


Figure 3 Relationship of POC with CO

Zheng, G. J., Duan, F. K., Su, H., Ma, Y. L., Cheng, Y., Zheng, B., Zhang, Q., Huang, T., Kimoto, T., Chang, D., Pöschl, U., Cheng, Y. F., and He, K. B.: Exploring the severe winter haze in Beijing: the impact of synoptic weather, regional transport15-2969-2015, 2015.

Wang, Q., Du, W., Zhou, W., Zhang, Y., Xie, C., Zhao, J., Xu, W., Tang, G., Fu, P., Wang, Z., Sun, Y., and Peng, L.: Characteristics of sub-micron aerosols above the urban canopy in Beijing during warm seasons, Science of The Total Environment, 926, 171989, https://doi.org/10.1016/j.scitotenv.2024.171989, 2024.

Change in the manuscript:

A refined EC tracer method was proposed (Zheng et al., 2015; Lin et al., 2009), and data from the lowest 10% percentile of ambient OC/EC ratios was utilized to estimate the primary OC/EC ratio with the following equations (Figure S1). Please refer to lines 105-107.

We have added the evaluation of SOC and POC in the supplement file.

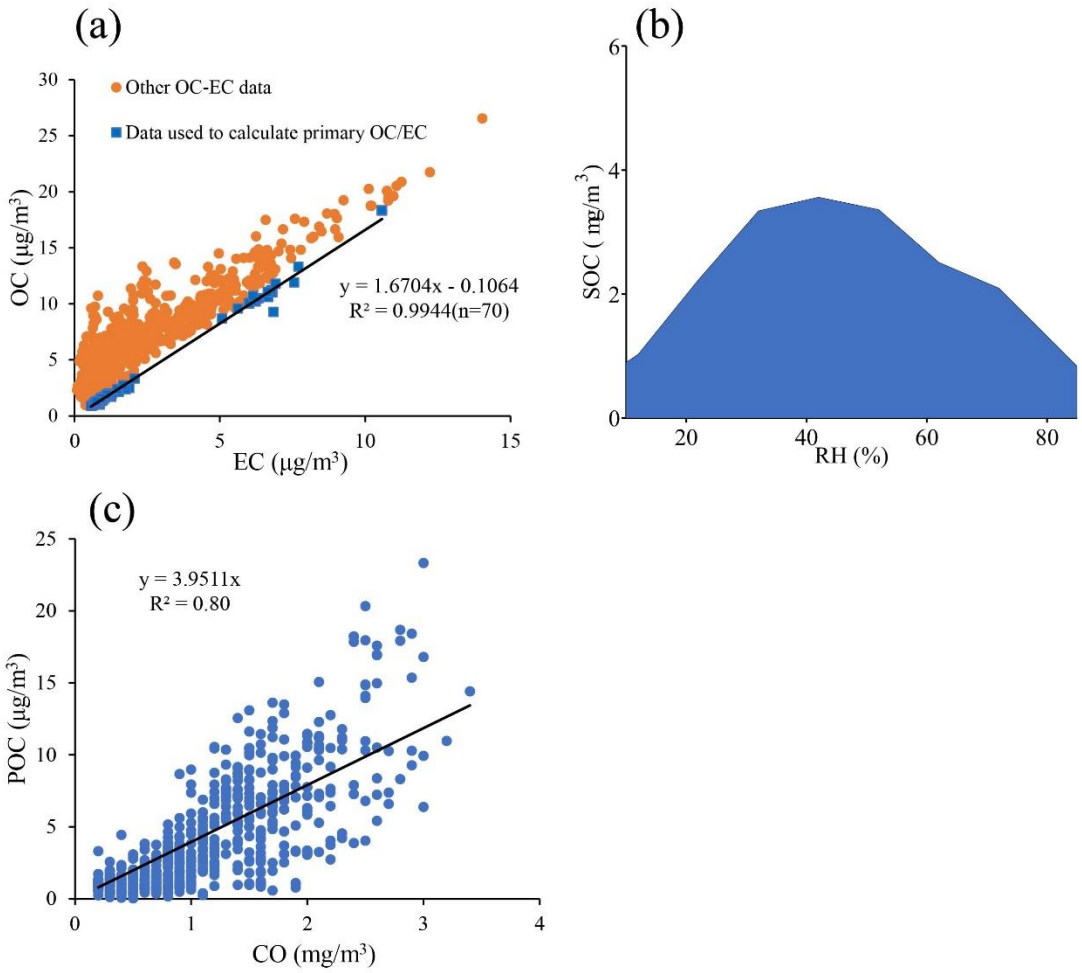


Figure S1 Evaluation of SOC formation and POC. (a) Estimation of SOC with EC-tracer method. Blue squares indicate data used to calculate primary OC/EC, while orange-filled circles indicate other OC/EC data. (b) Change of SOC with RH. (c) Relationship of POC with CO.

Second, the robustness of the measured babs, i.e., the observational constraint. For AE33, the correction factor for multiple scattering effect should be carefully determined (rather than simply using a reported value).

Response: Thank you very much for your valuable comments. Regarding your concern

about the determination of the correction factor for the multiple scattering effect in the AE33 Aethalometer, we would like to provide further clarification and response here:

Firstly, we acknowledge the significance of the multiple-scattering correction factor in deriving absorption coefficients from the AE33 measurements, as emphasized in various studies (Wu et al., 2024; Yus-Díez et al., 2021; Qin et al., 2018). Yus-Díez et al (2021) found that when the single-scattering albedo (SSA) of the collected particles is above a site-dependent threshold, neglecting the notable increase in the correction factor at high SSA levels can lead to a substantial overestimation of absorption coefficients obtained from Aethalometer instruments.

Secondly, regarding the observation in this study, the site-dependent experimentally multiple-scattering correction factor could not be obtained due to the lack of parallel observations. Consequently, the default multiple-scattering correction factor was utilized. As shown in the paper published before, an intercomparison of AE33 and CAPS PMssa was conducted and these results suggest that b_{abs} from different measurements agree reasonably well (Xie et al., 2019, Figure 2).

Finally, a note has been included in the text stating that there exists some uncertainty in the absorption measurement due to the use of the reported value of the multiple-scattering correction factor (Yus-Díez et al., 2021).

Thank you once again for your valuable comments.

Qin, Y. M., Tan, H. B., Li, Y. J., Li, Z. J., Schurman, M. I., Liu, L., Wu, C., and Chan, C. K.: Chemical characteristics of brown carbon in atmospheric particles at a suburban site near Guangzhou, China, Atmos. Chem. Phys., 18, 16409-16418, 10.5194/acp-18-16409-2018, 2018.

Wu, L., Wu, C., Deng, T., Wu, D., Li, M., Li, Y. J., and Zhou, Z.: Field comparison of dual- and single-spot Aethalometers: equivalent black carbon, light absorption, Ångström exponent and secondary brown carbon estimations, Atmos. Meas. Tech., 17, 2917-2936, 10.5194/amt-17-2917-2024, 2024.

Xie, C. H., Xu, W. Q., Wang, J. F., Wang, Q. Q., Liu, D. T., Tang, G. Q., Chen, P., Du, W., Zhao, J., Zhang, Y. J., Zhou, W., Han, T. T., Bian, Q. Y., Li, J., Fu, P. Q., Wang, Z. F., Ge, X. L., Allan, J., Coe, H., and Sun, Y. L.: Vertical characterization of aerosol optical properties and brown carbon in winter in urban Beijing, China, Atmospheric Chemistry and Physics, 19, 165-179, 10.5194/acp-19-165-2019, 2019.

Yus-Díez, J., Bernardoni, V., Močnik, G., Alastuey, A., Ciniglia, D., Ivančič, M., Querol, X., Perez,

N., Reche, C., Rigler, M., Vecchi, R., Valentini, S., and Pandolfi, M.: Determination of the multiple-scattering correction factor and its cross-sensitivity to scattering and wavelength dependence for different AE33 Aethalometer filter tapes: a multi-instrumental approach, Atmos. Meas. Tech., 14, 6335-6355, 10.5194/amt-14-6335-2021, 2021.

Change in the manuscript:

A new real-time loading effect compensation algorithm was adopted, which is based on a two-parallel spot measurement of optical absorption (Drinovec et al., 2015). It is worth noting that there may be some uncertainty in the absorption measurement due to the use of the reported multiple-scattering correction factor (Yus-Díez et al., 2021; Qin et al., 2018).

Please refer to Lines 112-115.

In addition, please confirm that similar to results from in-situ techniques (e.g., photoacoustic spectrometer), the AE33-based light absorption coefficients are "sensitive" to BC mixing state, e.g., by examining the relationship between babs and EC mass concentration.

Response: We thank the reviewer for helpful advice.

There are no parallel observations to measure absorption during the study period. However, a previous study showed that b_{abs} from different measurements agree reasonably well (Xie et al., 2019). Also, long-term changes in aerosol optical properties at IAP, Beijing have been investigated by AE33 in Sun et al. (2022).

Furthermore, we examine the relationship between AE33-based light absorption coefficients (b_{abs}) and EC mass concentration using linear regression analysis, as shown in the figure below. The b_{abs} measured by AE33 (880 nm) and EC mass concentration are highly correlated ($R^2 = 0.89$). This is consistent with the result of Wang et al. (2014).

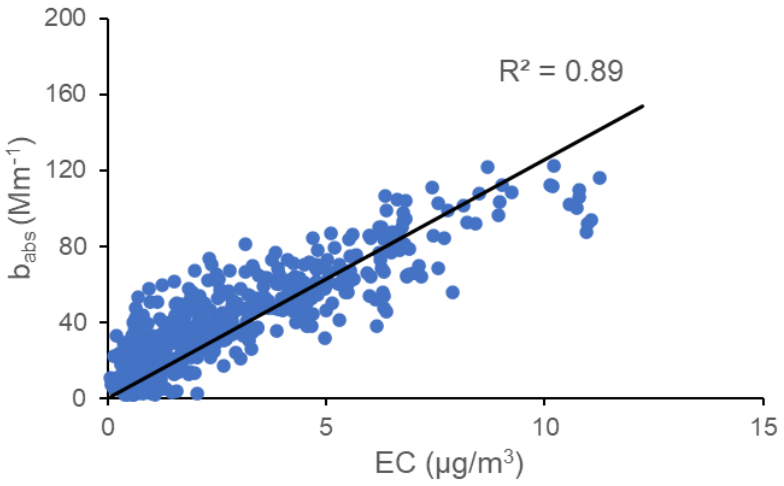


Figure 4 The linear relationship between b_{abs} at 880 nm from AE33 and EC mass concentration during the study period

Sun, J., Wang, Z., Zhou, W., Xie, C., Wu, C., Chen, C., Han, T., Wang, Q., Li, Z., Li, J., Fu, P., Wang, Z., and Sun, Y.: Measurement report: Long-term changes in black carbon and aerosol optical properties from 2012 to 2020 in Beijing, China, Atmos. Chem. Phys., 22, 561-575, 10.5194/acp-22-561-2022, 2022.

Wang, Q. Y., Huang, R. J., Cao, J. J., Han, Y. M., Wang, G. H., Li, G. H., Wang, Y. C., Dai, W. T., Zhang, R. J., and Zhou, Y. Q.: Mixing State of Black Carbon Aerosol in a Heavily Polluted Urban Area of China: Implications for Light Absorption Enhancement, Aerosol Science and Technology, 48, 689-697, 10.1080/02786826.2014.917758, 2014.

Change in the manuscript:

The b_{abs} at 880 nm and EC mass concentration are highly correlated (Fig. S3).
Please refer to Line 239.

3、Third, based on Figure 6 and Table 2, the performance of the "CSs" scenario (i.e., core-shell mixing) appeared best for reproducing the measured light absorption coefficients. Then I could not understand why the author argued that "Partial internal mixing and partial coating are the closest to reality". For the same reason, the logic of Sections 3.3 and 3.4 was confusing.

Response: We thank the reviewer for the careful review of our manuscript. We are very sorry that the manuscript confused the reviewer. To clarify this point, we would like to provide explanations from the following perspectives:

Firstly, we should admit that the performance of the "CSs" scenario appeared best in terms of reproducing the measured light absorption coefficients. However, there are uncertainties associated with the calculation. A good match between simulation and observation may be caused by unphysical reasons. As shown in Fig. 3, the concentration of secondary inorganic aerosols is underestimated by the NAQPMS model while the concentration of BC is overestimated. This could probably cause an underestimation in the coating of BC. Even if the model accurately captures the physical processes of aerosols, the fraction of embedded BC (F_{in}) and the coating on BC could be underestimated because of the representation uncertainties in chemical formation, potentially impacting the accuracy of the absorption calculation. What's more, uncertainty exists as other factors like the morphology of the BC core and the position of the BC core inside the coating are not considered.

The measured absorption coefficient was utilized to verify the rationality of the simulation results produced by the NAQPMS+APM and FlexAOD. Building upon this foundation, sensitivity experiments were designed based on the NAQPMS and optical module to investigate the impact of mixing state and aging processes on aerosol light absorption. As the reviewer comments before, there may exist some uncertainty in the absorption measurement in this study due to the use of the reported value of the multiple-scattering correction factor (Yus-Díez et al., 2021).

What's more, this study aims to investigate the impact of mixing state and aging processes on absorption based on the reasonable representation of the absorption coefficient.

Secondly, the CS scenario represents an idealized and simplified representation of the complex mixing state of black carbon in the atmosphere. In reality, BC particles often exist in a more complex mixing state, with varying degrees of internal mixing and coating by other aerosol components (Li et al., 2016; Reimer et al., 2019; Wang et al., 2021; Wu et al., 2021). As shown in published papers using the transmission electron microscope, only a fraction of BC is embedded in other aerosols (Li et al., 2016).

Our argument that "Partial internal mixing and partial coating are the closest to reality" is based on the recognition that the atmospheric environment is highly dynamic and heterogeneous, leading to a wide range of BC mixing states. These mixing states can vary depending on factors such as emission sources, atmospheric conditions, and transport processes. Therefore, while the "CSs" scenario may provide a good fit for certain datasets, it may not accurately represent the full range of BC mixing states observed in the atmosphere. Considering the fraction of embedded BC and secondary components coating on BC is a compromise and reasonable solution to represent the mixing state of BC in a three-dimensional model although uncertainties exist. The statement "Partial internal mixing and partial coating are the closest to reality" in Line 390 of the original manuscript is not that suitable and we have revised the description.

Finally, in Sections 3.3 and 3.4, we aimed to investigate the impact of the mixing state and aging processes on absorption and absorption enhancement. Our study provides a more comprehensive understanding of the BC mixing state by considering different scenarios, including considering the fraction of embedded BC (F_{in}), considering both the mass fraction of embedded BC and secondary components coating aerosols ($F_{in}F_c$) and the detailed microphysical process (CS-APM) calculated by advanced particle microphysics module coupled in NAQPMS. For example, comparing case CSs with

CS-$F_{in}F_c$, the impact of considering the fraction of embedded BC and secondary components coating on BC (aging process) can be investigated.

We hope that these revisions and explanations have addressed your concerns and clarified the logic of Sections 3.3 and 3.4. Thank you once again.

References

Li, W. J., Sun, J. X., Xu, L., Shi, Z. B., Riemer, N., Sun, Y. L., Fu, P. Q., Zhang, J. C., Lin, Y. T., Wang, X. F., Shao, L. Y., Chen, J. M., Zhang, X. Y., Wang, Z. F., and Wang, W. X.: A conceptual framework for mixing structures in individual aerosol particles, Journal of Geophysical Research-Atmospheres, 121, 13784-13798, 10.1002/2016jd025252, 2016.

Riemer, N., P. Ault, A., West, M., L. Craig, R., and H. Curtis, J.: Aerosol Mixing State: Measurements, Modeling, and Impacts, Reviews of Geophysics, 10.1029/2018RG000615, 2019.

Wang, Y. Y., Pang, Y. E., Huang, J., Bi, L., Che, H. Z., Zhang, X. Y., and Li, W. J.: Constructing Shapes and Mixing Structures of Black Carbon Particles with Applications to Optical Calculations, Journal of Geophysical Research-Atmospheres, 126, e2021JD034620, 10.1029/2021JD034620, 2021.

Wu, Y., Xia, Y., Zhou, C., Tian, P., Tao, J., Huang, R.-J., Liu, D., Wang, X., Xia, X., Han, Z., and Zhang, R.: Effect of source variation on the size and mixing state of black carbon aerosol in urban Beijing from 2013 to 2019: Implication on light absorption, Environmental pollution, 270, 116089, https://doi.org/10.1016/j.envpol.2020.116089, 2021.

Yus-Díez, J., Bernardoni, V., Močnik, G., Alastuey, A., Ciniglia, D., Ivančič, M., Querol, X., Perez, N., Reche, C., Rigler, M., Vecchi, R., Valentini, S., and Pandolfi, M.: Determination of the multiple-scattering correction factor and its cross-sensitivity to scattering and wavelength dependence for different AE33 Aethalometer filter tapes: a multi-instrumental approach, Atmos. Meas. Tech., 14, 6335-6355, 10.5194/amt-14-6335-2021, 2021.

Change in the manuscript:

Among the three ideal mixing state assumptions in this study, the result of the core-shell mixing state assumption is closest to the observed absorption coefficient. It should be noted that there are uncertainties associated with the calculation. As shown in Figure 3, the concentration of secondary inorganic aerosols is underestimated by the NAQPMS model while BC is overestimated; consequently, the coated thickness may be underestimated.

Please refer to Lines 271-275.

Considering the fraction of embedded BC and secondary components coating on BC is a compromise and reasonable solution to represent the mixing state of BC in a three-dimensional model although uncertainties exist.

Please refer to Lines 402-403.

Reviewer #2

This study uses the APM model combined with observations to discuss the impact of representative schemes of aerosols on optics. The whole study is meaningful and helpful for the experiment and model development. However, excessive use of concepts to represent aerosol mixing states lacks detailed and intuitive introductions, which reduces readability. A minor revision should be added before accepting.

1. Many excellent concept maps can be referenced to enhance readers' understanding of mixing states, such as Fig. 4 in 10.1038/s41467-018-05635-1, Fig. 1 in 10.1175/bams-d-16-0028.1

Response: Thank you for your constructive suggestion and for highlighting the valuable resources. Incorporating high-quality concept maps into our manuscript is important to enhance readers' comprehension of mixing states.

We reviewed the concept maps from the cited articles (Fig. 4 in 10.1038/s41467-018-05635-1 and Fig. 1 in 10.1175/bams-d-16-0028.1). Matsui et al. (2018, Fig. 4) showed the impact of resolving the mixing state on the direct radiative effect of black carbon. Fierce et al. (2017, Fig.1) showed the complex particle-resolved and reduced presentation of the mixing state.

We have added the references in the Introduction section to make it easy for the reader to understand the complex concepts. Also, we have added an abstract figure to the manuscript to clarify the mixing state considered in this study. Thank you again for your comments.

Changes in the manuscript:

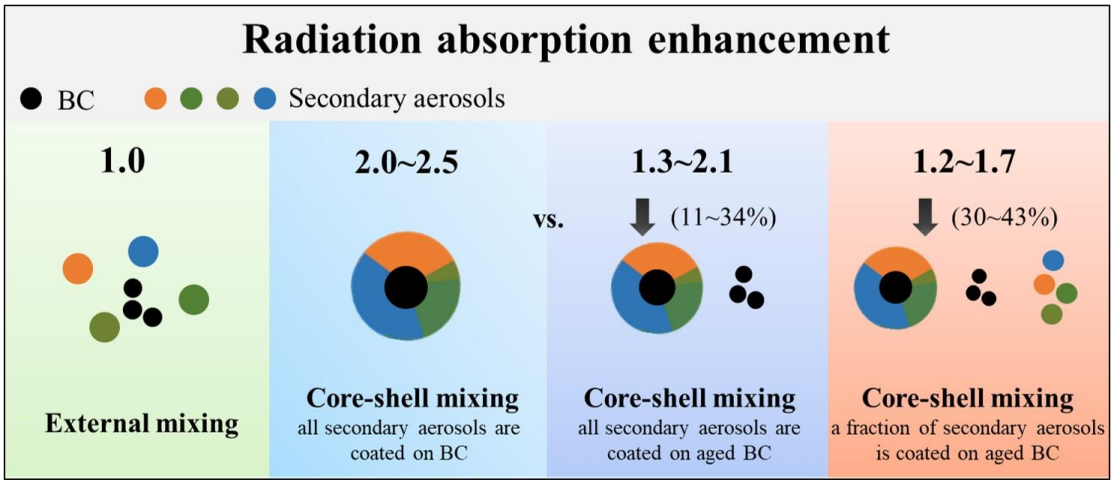
The concept maps illustrating the mixing state are shown in Matsui et al. (2018, Figure 4) and Fierce et al. (2017, Figure 1).

Please refer to Line 32, Lines 44-45, Line 573, Line 515.

Matsui, H., Hamilton, D. S., and Mahowald, N. M.: Black carbon radiative effects highly sensitive to emitted particle size when resolving mixing-state diversity, Nature Communications, 9, 3446, 10.1038/s41467-018-05635-1, 2018.

Fierce, L., Riemer, N., and Bond, T. C.: Toward Reduced Representation of Mixing State for Simulating Aerosol Effects on Climate, Bulletin of the American

Meteorological Society, 98, 971-980, 10.1175/bams-d-16-0028.1, 2017.



Abstract figure

2. Line 40: add references for condensation and coagulation processes: 10.1016/j.isci.2023.108125

Response: Thank you for your suggestion. We have added the reference in the manuscript.

Chen, X., Ye, C., Wang, Y., Wu, Z., Zhu, T., Zhang, F., Ding, X., Shi, Z., Zheng, Z., and Li, W.: Quantifying evolution of soot mixing state from transboundary transport of biomass burning emissions, iScience, 26, 108125, 10.1016/j.isci.2023.108125, 2023.

Changes in the manuscript:

The aerosol mixing state is dynamic and changes due to several processes, such as emission, new particle formation, transport, condensation, and coagulation processes (Chen et al., 2023). Please refer to Lines 47, 473.

3. Lines 190-192: Fin and Fc are not clear? Number fraction? Mass fraction?

Response: We are very sorry to make the reviewer confused. Fin and Fc are both mass fractions. Fin means the mass fraction of embedded BC. Fc is the mass fraction of coating aerosols (the secondary aerosols coated on BC). We have changed the description in the revised manuscript.

Changes in the manuscript:

Secondly, to investigate the impact of the aging process, simulations were designed using a partial core-shell mixing state in FlexAOD, encompassing two scenarios: CS-F_{in} (all secondary aerosols coating the F_{in} fraction of embedded BC, where "fraction" refers to mass fraction throughout this study) and CS-$F_{in}F_c$ (F_c fraction of secondary aerosols coating the F_{in} mass fraction of embedded BC), as illustrated in Fig. S2.

Please refer to Lines 198-201.

4. What are the differences between CS-Fin and CS-FinFc? You divided accumulation mode aerosols into 4 types (embedded, partly coated, bare-like BC and BC-free) or 3 types (embedded, bare-like BC and BC-free)? Detailed introductions should be added for mixing states in Table 1.

Response: Thank you for your constructive suggestion.

The differences between CS-F_{in} and CS-$F_{in}F_c$ lie in whether the fraction of secondary aerosols coated on BC is considered.

CS-F_{in} refers to a scenario where the F_{in} mass fraction of BC aerosols is embedded, with all secondary aerosols coating embedded BC (as illustrated in the third panel of Figure below).

CS-$F_{in}F_c$ refers to a scenario where the F_{in} mass fraction of BC is embedded, and the F_c mass fraction of secondary aerosols is coated onto the embedded BC (as illustrated in the fourth panel of the figure below).

In this study, aerosols can be classified into three types (embedded, bare-like BC, and BC-free) under the CS-$F_{in}F_c$ scenario. Furthermore, detailed introductions will be added to Table 1 in the manuscript.

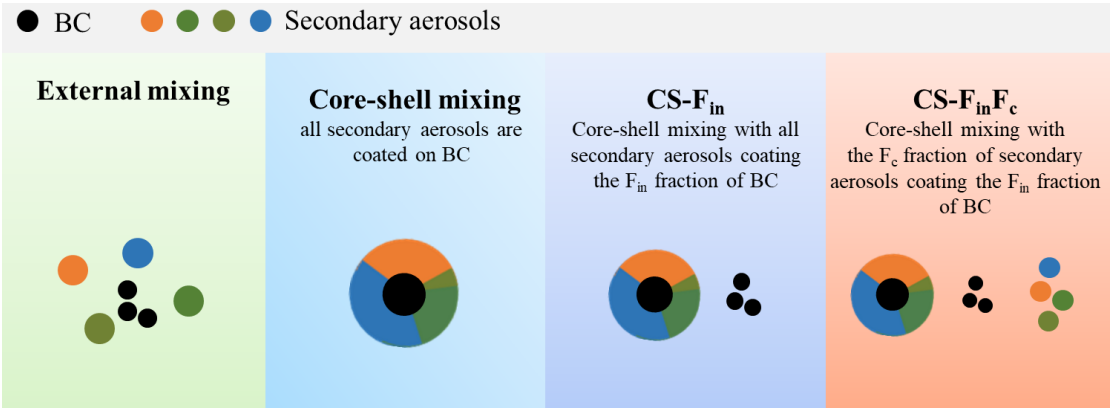


Figure S2 The concept of different mixing state assumptions

Changes in the manuscript:

Table 1 Simulation test design

Case	Method	Input	Size distribution	Mixing state
EXT_O	FlexAOD	observed	fixed	external
HOM_O	FlexAOD	observed	fixed	internal homogeneous
CS_O	FlexAOD	observed	fixed	core-shell

EXT_S	FlexAOD	simulated	fixed	external
HOM_S	FlexAOD	simulated	fixed	internal homogeneous
CS_S	FlexAOD	simulated	fixed	core-shell
CS-F_{in}	FlexAOD	simulated	fixed	partial core-shell and partial bare BC [a]
CS-$F_{in}F_c$	FlexAOD	simulated	fixed	partial core-shell, partial bare BC, and partial coating aerosols [b]
CS-APM	APM	simulated	simulated	semi-external (hourly) [c]

Impact	Description
EXT_O vs. HOMo vs. CSo	Impact of mixing state when inputting observed data
EXT_S vs. HOM_S vs. CS_S	Impact of mixing state when inputting simulated data
CSo vs. CSs	Impact of aerosol mass concentration
CSs vs. CS-F_{in}	Impact of aging process (fraction of embedded BC)
CSs vs. CS-$F_{in}F_c$	Impact of the aging process (fraction of embedded BC and coating shell)
CS-$F_{in}F_c$ vs. CS-APM	Impact of detailed microphysical process

[a] Aerosols are classified into two types: embedded, bare-like BC aerosols.

[b] Aerosols are classified into three types: embedded, bare-like BC, and BC-free aerosols.

[c] The concept map can be found in Chen et al. (2019, Figure 1)

Chen, X., Yang, W., Wang, Z., Li, J., Hu, M., An, J., Wu, Q., Wang, Z., Chen, H., Wei, Y., Du, H., and Wang, D.: Improving new particle formation simulation by coupling a volatility-basis set (VBS) organic aerosol module in NAQPMS+APM, Atmospheric Environment, 204, 1-11, 10.1016/j.atmosenv.2019.01.053, 2019.

Please refer to Lines 204-207, Lines 470-472.

5. How to define Partial internal mixing and partial coating?

Response: We apologize for this confusion caused by our terminology.

In this study, partial internal mixing and partial coating have the same meaning as CS-$F_{in}F_c$. Partial internal mixing means only part of the black carbon particles is core-shell mixed with the secondary component, and partial coating means only part of the secondary aerosols are coated on BC.

We appreciate your attention to detail. To avoid redundancy, we will uniformly adopt CS-$F_{in}F_c$ throughout the manuscript and omit unnecessary repetitions.

Changes in the manuscript:

We rewrite the sentences "Partial internal mixing and partial coating" to avoid redundancy and confusion.

"Considering the fraction of embedded BC and secondary components coating on BC is a compromise and reasonable solution to represent the mixing state of BC in a three-dimensional model although uncertainties exist".

Please refer to Line 402.

6. Line 313: How do you calculate Eabs? Add detailed calculation/inversion process

Response: Thank you for your constructive suggestion. The calculation of E_{abs} and the impact of the mixing state on E_{abs} were investigated in 3.4. We have added the detailed calculation process to this part.

In this study, the BC absorption enhancement is the ratio of the absorption coefficient calculated assuming core-shell mixing to that calculated using external mixing.

$$E_{abs} = \frac{b_{abs}(\lambda, core-shell\ mixing)}{b_{abs}(\lambda, external\ mixing)}$$

Therefore, the absorption enhancements in core-shell mixing, CS-F_{in}, and CS-$F_{in}F_c$ cases are the ratio of absorption under those cases to absorption under external mixing. In the CS-APM case, as described in Lines 337-340, the radiative absorption enhancement is the ratio of the absorption coefficient in the base simulation to that in the sensitivity test turning off the coating process.

Changes in the manuscript:

In this study, the BC absorption enhancement is calculated as the ratio of the absorption coefficient calculated assuming core-shell mixing (including CSs, CS-F_{in} and CS-$F_{in}F_c$ cases) to that calculated using external mixing.

$$E_{abs} = \frac{b_{abs}(\lambda, core-shell\ mixing)}{b_{abs}(\lambda, external\ mixing)}$$

Please refer to Lines 332-335.