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

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.

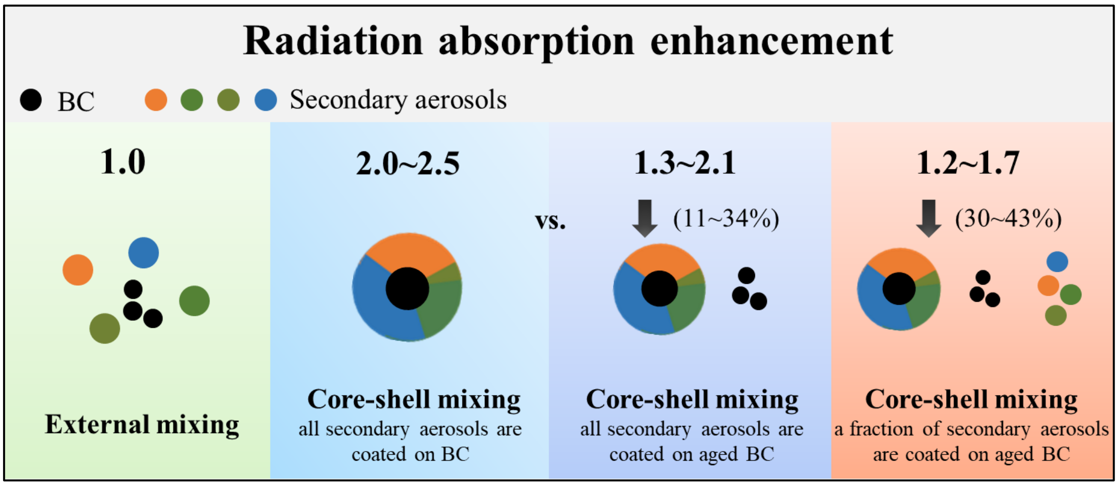
We have added the references in the Introduction section to make it easy for the reader to understand the complex concepts. Also, we have added an abstract figure to the manuscript to clarify the mixing state considered in this study. Thank you again for your comments.

Changes in the manuscript:

The concept maps illustrating the mixing state can be found in Matsui et al. (2018, Fig. 4) and Fierce et al. (2017, Fig.1).

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

Please refer to Line 29, Lines 40-41, Line 508, Line 563.

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 43, 469.

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 see the impact of the aging process, simulations were designed using a partial core-shell mixing state in FlexAOD, including CS-Fin (Fin mass fraction of embedded BC core) and CS-FinFc (Fc mass fraction of coating aerosols coating on Fin mass fraction of embedded BC) (Fig. S3). Please refer to Lines 194-196.

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 on 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 on embedded BC (as illustrated in the fourth panel of Figure below).

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

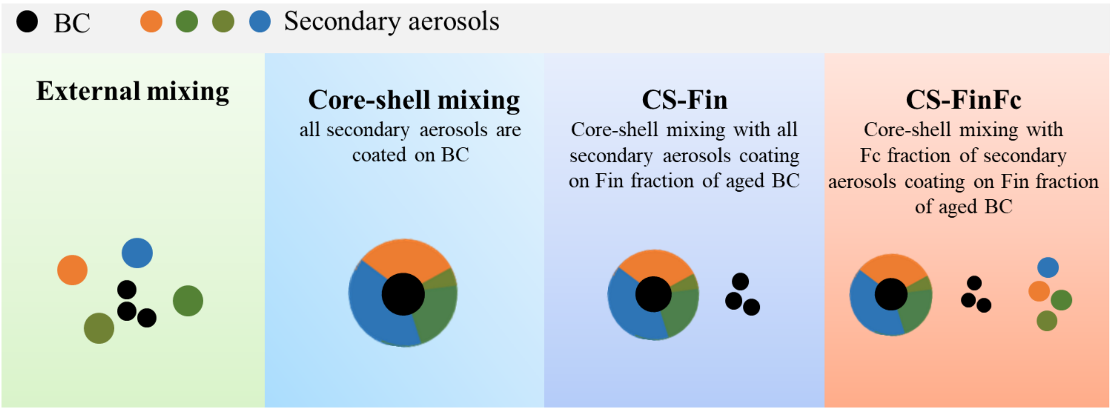


Figure S3 The concept of different mixing state assumption

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
$CS\text{-}F_{in}$	FlexAOD	simulated	fixed	partial core-shell and partial bare BC [a]
$CS\text{-}F_{in}F_c$	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. HOM_O vs. CS_O	Impact of mixing state when inputting observed data
EXT_S vs. HOM_S vs. CS_S	Impact of mixing state when inputting simulated data
CS_O vs. CS_S	Impact of aerosol mass concentration
CS_S vs. $CS\text{-}F_{in}$	Impact of aging process (fraction of embedded BC)
CS_S vs. $CS\text{-}F_{in}F_c$	Impact of the aging process (fraction of embedded BC and coating shell)
$CS\text{-}F_{in}F_c$ vs. CS-APM	Impact of detailed microphysical process

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

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

[c] Concept map can be referred to Chen et al. (2019, Fig1)

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 199-202, Lines 466-468.

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 $CS\text{-}F_{in}F_c$. 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 398.

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 Eabs 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.

$$E_{abs} = \frac{b_{abs}(\lambda, coreshell\ mixing)}{b_{abs}(\lambda, external\ mixing)}$$

Therefore, the absorption enhancements in core-shell mixing, CS-Fin, and CS-FinFc cases are the ratio of absorption under those cases to absorption under external mixing. In the CS-APM case, as described in Line 330-334, 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:

The BC absorption enhancement is calculated as the ratio of the absorption coefficient calculated assuming core-shell mixing to that calculated using external mixing.

$$E_{abs} = \frac{b_{abs}(\lambda, coreshell\ mixing)}{b_{abs}(\lambda, external\ mixing)}$$

Please refer to Lines 328-331.