RESPONSE LETTER OF REVISION (EGUSPHERE-2024-3781)

Title: Numerical quantitation on the effect of coating materials on the mixing state retrieval accuracy of fractal black carbon based on single particle soot photometer

Dear Editor:

We have revised our manuscript based on the comments of Referees. The corrections and modifications have been included in the revised manuscript and the details are listed as follows. The responses are highlighted in blue font. The changes made in the revised manuscript are marked in red font.

Referee #4

The author has adequately addressed the reviewers' suggestions, and the manuscript is now close to a generally publishable state. Below are my comments for further refinement before publication:

Response:

Thank you very much for reviewing out manuscript and the comments. We have responded to all the comments point by point, and the related descriptions have been modified in the revised manuscript.

(1) Expressions and discussions requiring correction: SP2 is not the only instrument capable of measuring BC mixing states. Other methods include single particle aerosol mass spectrometry (SPAMS) and single-particle soot mass spectrometry (SP-AMS).

Response:

Thanks a lot for the suggestions. We have modified the related descriptions in the revised manuscript as follows:

"The "mixing state" is a key microphysical property for aged BC, describing the mixing structure of BC and its coating. It can be characterized through different principles and instruments, such as the single particle soot photometer (SP2), single

particle aerosol mass spectrometry (SPAMS), and single-particle soot mass spectrometry (SP-AMS) (Liu et al., 2023b; Liu et al., 2025). The SP2 (Droplet Measurement Technology, Inc.), as one of the most effective online instruments that measures the mixing state of coated BC based on the combination of laser-induced incandescence and light scattering measurement, has been widely employed in laboratory and field observations (Liu et al., 2020a; Liu et al., 2022; Schwarz, 2019)."

(2) Conclusion section is overly verbose, repeating many minor results. It is recommended to focus on the core findings and implications directly related to the paper's theme, providing concise conclusions.

Response:

Thank you very much for the constructive comments. We have reduced some descriptions in the Conclusion section to further focus on the main findings of our study. The modified Conclusion is shown in the revised manuscript.

(3) Numerical precision issues: The manuscript contains numerous measured and simulated values with insufficient significant figures. These should be adjusted to reflect the precision of the measurements and simulations.

Response:

Thank you for the valuable comments. To be consistent with the measurement results of the SP2, we have modified the numerical precision in the revised manuscript. All these values of morphological parameters, optical properties, radiative effects, and the preset and retrieved mixing states have been retained to two decimal places. Except for one of the values of the BC volume fraction, 0.075.

Referee #5

General comments:

This paper analyses the effect of the simplified hypothesis underlying the SP2 data processing used to determine the mixing state of black carbon. In particular, the authors tested the impact of using different coating materials with different refractive indices (sulphates, organic carbon and brown carbon) and of giving the coated BC particles different shapes (ranging from more fractal-like to a core-shell model) on the mixing state. The mixing state is defined as the diameter ratio between the BC core and the whole BC particle, and is calculated from SP2 measurements using the usual retrieval method. This method uses Mie theory and assumes a core-shell morphology and a fixed refractive index. The authors identified biases resulting from these simplifications, particularly for thinly coated BC. BC coated with BrC also exhibited higher inaccuracies due to its absorbing properties. The authors further investigated subsequent errors in radiative forcing and absorption enhancement. Since the SP2 is the only online instrument that can measure the coating thickness of BC-containing particles at a particle-by-particle level, the methodology to process its mixing state data is crucial for interpreting the results. This study provides valuable insights into errors in the BC mixing state resulting from core-shell and fixed refractive index assumptions. Furthermore, the results suggest that a more accurate characterization of BC optical and radiative properties would be achieved by considering more complex morphologies and variable refractive indices. The tests and experiments conducted in this study are robust and well described. However, the good scientific quality of the paper is still suffering from a significant number of grammatical errors. Some of them are listed bellow. I would strongly recommend that the authors carefully review the entire manuscript in terms of English writing, before publication.

Response:

Thanks a lot for reviewing our manuscript and your recognition of our study. We have tried our best to carefully review the entire manuscript to correct the grammatical errors and improve the English writing. All these related modifications are given in the revised manuscript.

Specific comments:

l 14. I would add particles: coated black carbon particles (BC)

l 14. "optical properties"

l. 16. "the coating material"

l. 16 "diverse"

l. 21. "are used to study optical properties…"

l. 27 "be up to 6.15 times higher than"

l 33-34 This sentence is not clear since human = anthropogenic. Please reformulate.

L 36 "optical absorption properties"

l. 39 "BC particles are usually"

l. 40 "BC particles can be coated by various"

l. 42: "through condensation and coagulation"

l. 47 "one of the most effective online instrument that can measure"

l. 57 It would be useful to precise that the change in the Gaussian pattern is due to the different refractive indices between the coating and the core.

l. 58 : Please provide references for the refractive indices mentioned here.

l.59 : I would start a new sentence from the word "since" to add clarity.

l.60 "by assuming spherical particles and using BC density …"

l.64 "into bare-to-thinly coated"

l. 69 The work package is a bit weird here, please reformulate

l 74. "sulfate or organics"

l. 79 "the measurements of the SP2 are usually…"

l. 83 "are built to represent"

l. 84 "can be classified using the lag time"

l. 100 "is the gyration radius"

l. 100 formatting of the ith

l. 109 Please don't start a sentence with "and"

l. 112 "are built". That would be nice to add a schematic of the typical shapes of these two different models.

l. 135 "For the quantification"

Formula 6: Please define what is X^2. More generally, it would be better to introduce each before its corresponding definition (equations 9, 10, 11,12,13), as the authors did for i.e. equation 7.

L 150-151 This sentence is not grammatically correct, or something is missing maybe?

L 152 "the precision in the estimated typical optical"

l. 163-164 Keep the same structure, either first variable and then definition as the beginning of the sentence or the other way around

l. 176 "the variation of complex refractive"

l. 178-180 The sentence starting with "while" is not grammatically correct.

l. 184-187 The sentence starting with "as can be" is not grammatically correct.

l.189 "more compact and the retrieval"

l. 189 "BC volume fraction due to a smaller amount"

l. 192 "SP2 has a better performance"

l. 193 This sentence is not clear to me. Please reformulate.

l.198 "from about 200 to 400 nm, as shown by Liu et al."

l.201 "missing data points of retrieved"

l.202-203 Not only that but it will also lead to a bias toward size ranges of coated BC that the SP2 is able to characterize when considering the ensemble of BC particle.

Figure 1: This is good to include a schematic to show the corresponding morphologies of BC but I don't really understand what values of Dc/Dp and Dc,v each of the representation refers to. Maybe move the images more to the corners, ie. Maximum value of Dc/Dp?

Figure 3 : The Dp,v/Dc,v in vertical on the right hand side is a bit difficult to understand. Maybe move to the left, add a space between it and the y-label, and increase the font? Same for the column titles, increase font and space between title and figures

Figure 4: Describe what is the dashed line on the violins

Caption of Figure 5: "…and the corresponding core-shell model, , represented for different values of Dp,v/Dc,v."

Caption of Figure 6: Same as previous comment

l. 207 "to the coating components, thus the"

l. 211 move the "compared with the BC refractive index" to the end of the sentence

l. 212 "on the other hand, when the soot"

l. 215 Do you mean Cappa study, by "this study"? Then citation needs to be done properly: Cappa et al found ...

l. 215: The sentence starting with "The enlargement" is not clear to me. Please reformulate.

l. 220 This was already said l193, so maybe change to "that confirms..."

l. 230 "between sulfate and OC could explain their similar Dp/Dc"

l. 231: If "could" is used for the first part of the sentence, then one would use "may have caused" or so

l. 240 A higher accuracy than for other coating materials?

l. 242 "The coupling effect […] would lose": not grammatically correct. Would lead to …

l. 247 "Three distinct regions coloured in"

l. 248-250 : It would be nice to put that in the Figure caption

l. 254-256: If last part of the sentence refers to the distribution width, to which characteristic of the distribution is the beginning about?

l. 273: Don't start a sentence by "And"

l. 279: "by the higher number of missed data points"

l. 283: Maybe introduce the lensing effect earlier when the authors first mention it.

l. 295-296. I am not sure to really understand this sentence.

l. 304: "Eab values"

l. 306 "about 6.14 times larger than"?

l. 321: Don't start a sentence by "And"

l. 322: It seems to me that Compact and fractal don't match together

l.357: "overestimated or underestimated"

l.366: "B C" remove space

l. 371: Maybe introduce which coating material have been tested before using "these three components"

l. 373: Is there a reason to use "As" in this sentence?

l. 377: "thickly coated by BrC are more numerous" maybe?

l. 381: Please introduce RE in the beginning of the sentence

l. 387: Need to precise that Eab was studied as a function of the diameter Dc to understand the "at first", and the numbers that were cited in this sentence.

l. 396: "The oversimplification […] is inappropriate" This part of the sentence is not very clear, could you reformulate please?

Response:

Thank you very much for the responsible review on our manuscripts. The related descriptions about all these above 75 valuable suggestions have been rewritten or modified in the revised manuscript, and the Figures have also been made necessary modifications to improve clarity.