Response to Reviewer #1 (EGUSPHERE-2023-2122)

First of all, we would like to thank the editor and reviewers for their valuable comments. We have taken all the suggested changes into consideration and revised the manuscript accordingly. The reviewers' comments are copied here as texts in BLACK, our responses are followed in BLUE, and the major corrections are marked in RED in the manuscript.

Brown carbon (BrC) is an important component of aerosols in the atmosphere, and there are still significant uncertainties on their chemistry and physical properties as well as their influences on the atmosphere. This manuscript presents an observationally-constrained approach to estimate the radiative effects of BrC aerosols using routine ground-based measurements, and offers a convenient method to assess the climate impacts of BrC. This study effectively integrates observations, optical computations, and radiative transfer models. By employing optical closure techniques, the radiative effects of black carbon were isolated. This approach proves to be both straightforward and efficacious. Meanwhile, by considering only the conventional observations and numerical models, the framework of the proposed method shows great potential for further studies. Overall, the study is well motivated and adds to our understanding of BrC effects. There are several areas that need clarification or revision prior to publication.

Response: Thanks so much for your constructive comments. We have implemented all suggestions for improvement in the revised manuscript. Please find our point-by-point responses listed below.

1. More details could be provided on the observations used to constrain the analysis. ln particular, the authors should specify details such as the sampling time period and measurement frequency for each instrument.

Response: Thanks. We have added much more descriptions on the observations, such as sampling time period, measurement frequency, and so on (Lines 88 in the revised manuscript).

2. From the perspective of content relevance, it appears more appropriate to position Figure 1 and its associated description with in the Section 2 rather than the third section.

Response: Thanks for the suggestion, we have reorganized the manuscript by moving Figure 1 and its associated description to Section 2.

3. Figure 2 serves as a comprehensive overview of the proposed methodology, playing a pivotal role in the exposition of this paper. To provide a clearer understanding, additional space to elucidate the details within Figure 2 is suggested, including the calculation methods for parameters such as MAC (Mass Absorption Coefficient). Alternatively, to manage space constraints, specific algorithms for each subsection of Figure 2 can be referenced in the subsequent sections of this paper.

Response: Thanks for the suggestion. We added detailed descriptions of the flowchart in the figure caption, in which the important components in the figure are referred to the subsections of the paper. This will ensure the readers have a clearer understanding of our methodology and calculations.

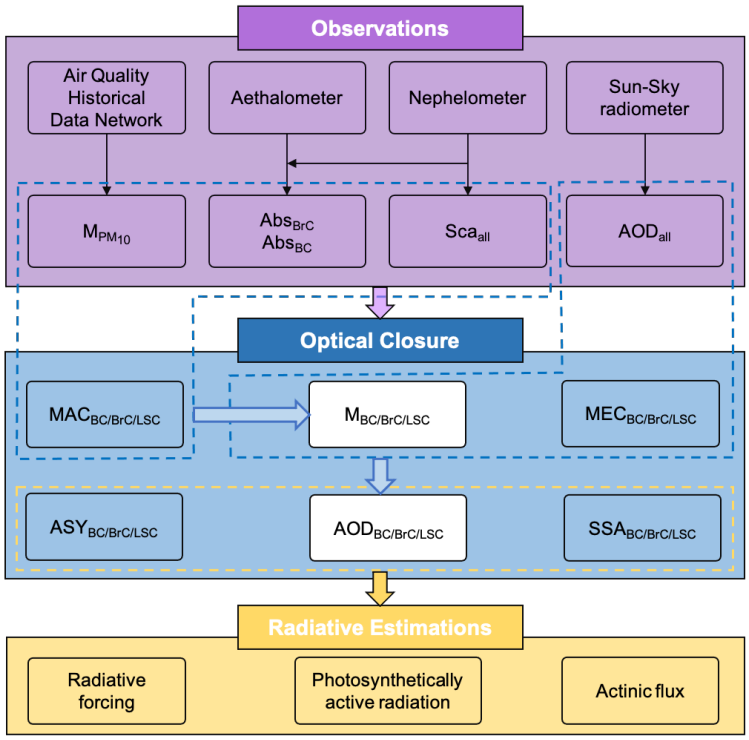

Figure 2. Flowchart of estimation of BrC radiative effect. The part with purple background corresponds to the direct observations used, which are detailed in Sections 2 and 3.1. The optical closure part, which uses the direct observations to separate the properties of each type of aerosol (i.e., AOD, SSA, ASY, surrounded by the yellow dotted line), is illustrated by the part with blue background. The bottom yellow part indicates the output for radiative estimations. We adopted a three-component aerosol model (BrC, BC, and pure light-scattering components, i.e., LSC). More details are available in Section. 3.2. After clarifying the properties of each type of aerosol (i.e., AOD, SSA, ASY), the LibRadTran Model is used to estimate the BC and BrC radiative effects.

4. Figure 4 indicates that the imaginary part of BrC refractive indices may differ over two orders of magnitudes. Would such variation introduce additional uncertainties on the results of this study?

Response: Yes, there are significant uncertainties on the refractive indices of BrC. As suggested by both reviewers, we added a sensitive study on the influences of BrC refractive indices (imaginary part) on our BC and BrC radiative effect estimation. We found that BrC refractive index variations may introduce uncertainties up to over 50% on the BrC TOA RF, and more details were added in Section 5.

Page 16 line 364-366 in the main text,
"The right panel shows the influences deriving from uncertainties of the absorptivity of BrC. Except the RF (TOA) with a relatively larger difference of 62%, the rest of the BrC radiative effects (average absolute values) were all below 30%, and, as expected, all the differences were close to zero."

5. In Figure 5, the label "LSC/10" is confusion.

Response: To avoid confusion, we redesigned the figure, and the new figure in the form of a double Y-axis becomes much clearer.

6. More discussions on regarding the applicability of the method and the generalizability of the results are suggested, and the limitations of the method could also be discussed.

Response: In the article, we use observational data from the Nanjing site as an example to verify the feasibility of the method, but the method can be applied to other regions. Additionally, we acknowledge that the current method still entails a certain degree of uncertainty. We added a new section (Section 5) to discuss and analyze the uncertainties of our method, including the imaginary part of the BrC refractive index, BC particle geometries, and AAE. Furthermore, some discussions on the further works were added in Section 6.

7. It is advisable to further improve the figures qualities. For instance, the tick labels in Figure 4 appear relatively small and could benefit from a consistent font size.

Response: Thanks for the suggestions. We have improved all figures and kept a relatively consistent format for them in the revision.

Some minor comments:

8. Line 17, To enhance clarity, you can split the sentence into two as follows: “To constrain the total and other aerosol contents, we conducted an optical closure study. Subsequently, the optical properties and concentrations were estimated.”

Response: Thanks for the suggestion, we have corrected it in the revised manuscript (Line 17 – Line 18).

9. Line 46, “currently, materials such as humic-like substances, polycyclic aromatic hydrocarbons, and lignin are all considered BrC” should be “Currently, materials such as humic-like substances, polycyclic aromatic hydrocarbons, and lignin are all considered as BrC”.

Response: We have corrected it in the revised manuscript (Line 47).

10. Line 106,"LT" should be "local time".

Response: Thanks, and it is corrected (Line 108).

11. The label (a) and (b) in figure 5 is missed.

Response: Sorry for the mistake and we have added it to the revised manuscript.

12. Line 337, "that of BC" should be "that caused by BC".

Response: We have corrected it in the revised manuscript (Line 383).