Response to the comments of reviewers #2

The authors are very grateful to the editors and reviewers for their valuable comments and constructive suggestions. The reviewers' questions and comments are highlighted in **BLACK font**, and the answers in **BLUE**. The changes made in the revised manuscript are highlighted in **RED**.

Comments: This manuscript quantifies the uncertainty of the mixing state and absorption of partially-coated BC based on SP2 with numerical simulation, and points out that quantification of the mixing states of partially-coated black carbon based on the single-particle soot, as well as its impacts on BC absorption and radiative forcing. Based on the simulation, the authors suggest adding a parameter F to model the radiative effect of BC in climate modeling. In general, the manuscript is not rigorously organized and the figures are not clear. The following issues should be taken into consideration for improvement.

The manuscript highlights the importance of partially-coated BC, and there is large uncertainty when assuming BC is fully coated. The title is "Numerical quantification of the mixing states of partially-coated black carbon based on the single-particle soot photometer", but there is no measurement data from SP2. In fact, it is just the difference between Mie theory and MSTM. This problem is common in current regional or climate model, not just for SP2. Zhang et al (2018) did similar work on numerical simulation of partially-coated BC absorption. In the introduction part, the progress of studying on partially-coated BC should be summarized and the novelty of this study should be pointed out.

Response: Thanks very much for your comments. There is indeed no SP2 measurements. We combined the calculations from the MSTM and the SP2 measurement principle to represents the "pseudo measurement", and then retrieved the mixing states based on the Mie theory like many SP2 user done. By exploring the effects of the microphysical properties on the calculations, we can provide some insights on the uncertainties of the mixing states based on the SP2 measurement and Mie retrievals, and this is one of the main difference from Zhang et al. (2018).

Indeed, like Reviewer #1 says, SP2 just detects scattering, does not assume core-shell structure, and some techniques that don't use the core-shell Mie theory was also proposed. However, these techniques commonly needs the combinations of different instruments, and it's difficult for many people to own so many expensive instruments at the same time. Therefore, The measurement based on SP2 using Mie scattering to retrieve the mixing state of black carbon remains a preferred choice for many researchers. This study primarily focuses on SP2 users who employ Mie scattering for measuring the mixing state, and we have clarified this in the revised manuscript.

In addition, another distinction from Zhang et al. (2018) is the exploration of using multiple spheres to improve the calculation of absorption enhancement via Mie scattering. This is also a significant demand for many SP2 users when measuring the mixing state. While the uncertainty of bare black carbon absorption has been relatively well studied through the efforts of many researchers, the mixing state can significantly affect the absorption enhancement of black carbon. One of the main purposes of SP2 mixing state measurement is to reduce the uncertainty of absorption enhancement through the understanding of particle mixing states. For most SP2 users, Mie scattering is often used to calculate the absorption enhancement of particle ensembles after measuring Dp/Dc, while ignoring the influence of F. Zhang et al. (2018) demonstrated through the Multi-Sphere T-Matrix (MSTM) method that F can affect absorption enhancement. However, it is difficult to apply an MSTM model to SP2 measurements and climate models due to the variations in F among individual particles.

Another contribution of this paper is to explore the improvement in absorption enhancement calculations by considering the influence of F based on Mie scattering. This is a highly application-oriented approach. If Mie scattering is feasible, we can simultaneously retrieve F and Dp/Dc based on SP2 measurements and absorption measurements, obtaining F and Dp/Dc values in different regions. This will further constrain the radiative effects of black carbon at the single-particle level, which has significant practical value for the future. We will also conduct further research in the near future. We have re-written the introduction in the revised manuscript:

"The single particle soot photometer (SP2), an instrument for measuring the mass of individual BC particles, has recently been widely used to measure mixing states (Schwarz et al., 2006; R. S. Gao and Worsnop, 2007). The SP2 measures scattering of individual particles reflected from a 1064 nm laser, and the mass of the BC core is estimated from the incandescence signal (Moteki and Kondo, 2008; Wu et al., 2023). Based on an assumed BC mass density, we can calculate the mass-equivalent diameter of the BC cores. To obtain the mixing state of single-particle black carbon (BC), many researchers have attempted to develop methods for simultaneously measuring the total particle size online. A significant advancement in this field has been achieved by combining SP2 with the Centrifugal Particle Mass Analyzer (CPMA) (Olfert and Collings, 2005; Liu et al., 2017b; Yu et al., 2020; Naseri et al., 2024). In this process, CPMA measures the mass of individual particles to infer particle size, without simplifying morphological features. Some techniques based on the differential mobility analyzer (DMA)-SP2 system was also developed (Andrew R. Metcalf and Seinfeld, 2013; Zhao et al., 2022; Huang et al., 2024). However, due to the high cost of these instruments, many researchers find it challenging to own them simultaneously. In addition, in some cases (such as unmanned aerial vehicle detection), it is very inconvenient to measure by combining so many instruments. Consequently, alternative methods for measuring mixing states have been adopted (R. S. Gao and Worsnop, 2007; Naseri et al., 2024; Lee et al., 2022). The SP2 can simultaneously provide information on the scattering properties of particles, and the leading-edge-only (LEO) technique enables the extraction of particle scattering signals. Many researchers thus invert the total particle size based on Mie theory, utilizing the scattering signals measured by the SP2 (R. S. Gao and Worsnop, 2007). Nevertheless, this approach has limitations: coated BC often does not exhibit a perfect core-shell structure. Despite the limitations of instruments and costs, this method, subsequently referred to as SP2-Mie, is still widely used by researchers. A primary target audience of this paper is researchers who employ the SP2 for measuring BC mixing states based on Mie scattering. Nevertheless, current research on explaining the uncertainties associated with the SP2-Mie method remains limited.

SP2 users often explain the measured mixed state with Mie scattering (Moteki and Kondo, 2008; Schwarz et al., 2008; Naseri et al., 2024). However, Moteki et al. (2014) found that the discrepancy between the calculated results of Mie scattering and the scattering cross-section measured by SP2 can reach up to 40% in some cases. One of the important reasons is that the morphology of BC is often complex and frequently partially-coated (Adachi et al., 2007; China et al., 2013; Wang et al., 2017). Although previous studies have recognized that simplifying the microphysical properties of BC aerosols can lead to inaccurate determination of mixing states (Schwarz et al., 2015), there is still a lack of quantification of the effects of microphysical properties.

Previous studies have compared the scattering cross-section of core-shell BC and more morphologically realistic BC, and they found that the scattering properties of BC is significantly affected by morphologies (Schwarz et al., 2008). However, the direct comparison of the mixing states retrieved based on the SP2-Mie and the volume-mean are very limited. A recent study by Wu et al. (2023) has studied the impact of adopting the core-shell model on the inversion of optical particle size using the SP2 based on the the multiple-sphere T-matrix (MSTM), but their study assumed fully coated BC, neglecting partially coated BC. However, BC is often partially coated, and the absorption of partially coated BC is more complex than that of fully coated BC, determined not only by the ratio of the core size to the total particle (D_p/D_c) but also by the ratio of the volume of coated BC cores to the total volume of BC cores (F). Furthermore, D_p/D_c has broader applications in climate research, yet Wu et al. (2023) did not explore the influence of microphysical properties on the inversion of mixed states or assess their implications for climate effects. Liu et al. (2023) attempted to use a similar model to Wu et al. (2023) to evaluate errors in the mixed state due to BC morphology, but their inversion parameters are based on the differential scattering cross section which differ from the measurement principle of the SP2, incompletely reflecting the SP2's measurement process. The scattered signal measured by SP2 should be proportional to the scattering cross-section within the measurement angle range, rather than the differential scattering cross section (Moteki and Kondo, 2008; Wu et al., 2023; Naseri et al., 2024) Additionally, Liu et al. (2023) also failed to investigate the impact of partially coated BC aerosols. The aim of this paper is not to discredit the use of the SP2 for measuring mixing states but rather to theoretically investigate the influence of BC microphysical properties on the accuracy of the SP2-Mie method, assisting researchers in analyzing the sources of measurement uncertainty in the SP2-Mie method during actual measurements.

Another important concern for SP2 users is the absorption enhancement of coated BC. When BC is mixed with other components, its total absorption can be enhanced due to the "lensing effect." In reality, the mixing state of BC significantly influences absorption enhancement, making the mixing state measured by SP2 crucial for absorption enhancement calculations and climate predictions. However, as mentioned above, when using the mixing state measured by SP2 to calculate absorption enhancement, it is still common to assume a core-shell structure and use Mie scattering calculations. Another objective of this paper is to assess the uncertainties in calculating absorption enhancement using the mixing state retrieved by SP2-Mie and to explore methods for improvement. Previous researchers have conducted a series of studies comparing the absorption enhancement of BC with complex morphologies and their Mie scattering results, including studies on partially-coated BC (Wang et al., 2021b). However, there is a lack of evaluation specifically for SP2-Mie users. In practical measurements, the mixing state measured by SP2-Mie is often used to calculate absorption enhancement, which may differ from the "true" mixing state. Therefore, it is necessary to evaluate the mixing state retrieved by SP2-Mie. Furthermore, previous studies have found that the absorption enhancement of partially-coated BC is simultaneously influenced by both F and Dp/Dc. However, in real-world situations, it is difficult to obtain F using realistic morphology models. Developing a simplified model that considers both F and Dp/Dc is significant for retrieving F during measurements and for statistical analysis of F in different regions, thereby improving the accuracy of climate simulations."

Zhang, X., Mao, M., Yin, Y., and Wang, B.: Numerical investigation on absorption enhancement of black carbon aerosols partially coated with nonabsorbing organics, Journal of Geophysical Research: Atmospheres, 123, 1297–1308, https://doi.org/https://doi.org/10.1002/2017JD027833, 2018.

Comments: As stated in the manuscript, the model calculated MACBC is inconsistent with the measured MACBC, and most models underestimate MAC_{BC} based on the measured mass density and refractive index, why did the authors choose a MACBC of 7.5 m^2g-1 in this study? Both fluffy and compact BC aggregates were considered in this study, and MACBC also varies for both BC shapes. How does MACBC affect the calculated absorption and radiative forcing?

Response: Thanks for your comments. The mass absorption cross-section (MAC) of bare black carbon is indeed affected by its morphology, but numerous studies have conducted extensive measurements and simulation research on this topic, leading to a clearer understanding of its uncertainty. This current research primarily focuses on the understanding of the impact of mixing characteristics on SP2 users' measurements. Since mixing characteristics mainly affect absorption enhancement, this study primarily concentrates on the influence of absorption enhancement. For the effects of morphology on the black carbon core, please refer to other literature. Furthermore, in the process of model application, due to the current underestimation of model predictions for the MAC of bare black carbon, many researchers in climate modeling studies have adopted a MAC value of 7.5 m^2/g, as it is derived from measurements. To focus on SP2 users and the impact of black carbon absorption enhancement, this paper adopts a black carbon core MAC of 7.5 m^2/g. We have made clarifications in the revised manuscript:

"It should be noted that in this work we assume that the mass absorption cross-section (MAC) of the BC core is fixed. In reality, however, the MAC of the BC core is also influenced by its morphology. Nevertheless, this study primarily aims to understand the influence of mixture properties on the measurements performed by SP2 users. Since mixing states primarily affect absorption enhancement, we focus primarily on the effects of absorption enhancement. Besides, many climate modeling studies have also used a fixed MAC from measurements because model predictions for the MAC of BC core are currently underestimated, and the total absorption is estimated by multiplying the absorption enhancement (Bey et al., 2001; Eastham et al., 2018; Wang et al., 2014; Zhang et al., 2021). In this process, the absorption enhancement of BC is the main influencing parameter (Wang et al., 2014; Zhang et al., 2021). To focus on SP2 users and the effects of BC absorption enhancement, a BC core MAC of 7.5 ± 1.2 m^2g^{-1} is assumed in this work. Regarding the influence of morphology on the MAC of the BC core, previous studies have performed extensive measurements and simulation studies that provide a clearer picture of the uncertainties (Liu and Mishchenko, 2005; Kahnert, 2010; Luo et al., 2018; Fengshan Liu and Corbin, 2020). Our study is therefore primarily concerned with the effects on absorption enhancement."

Comments: In the fourth paragraph, it is mentioned twice that "BC is often partially-coated". If it was written carefully, I think the authors want to emphasize the importance of partially-coated BC. However, the exact fraction of partially-coated BC in the real atmosphere is more convincing than current expression. In addition, the fraction of partially-coated BC in the real atmosphere also impacts on the uncertainties of climate model, so the uncertainty of climate model is not reliable when consider partially-coated BC alone.

Response: Thanks very much for your comments. Thank you very much for your comment. Indeed, in order to conduct a more accurate assessment, we should consider the proportion of partially covered black carbon. However, the calculations in this paper are merely for sensitivity analysis, aiming to demonstrate the significance of the microphysical properties of partially covered black carbon in global climate simulations. More precise simulations in the future will need to take into account the proportions of BC with different morphologies. We have described this in the results of the revised manuscript:

"It is worth noting that all the BC considered in this study is assumed to be partially-coated, while in reality, there exist various types of BC. The sensitivity analysis conducted in this study is solely to illustrate the importance of the microphysical properties of partially covered BC in SP2-Mie inversion and global climate assessment. For more accurate climate simulations in the future, it will be necessary to consider the proportions of BC with different morphologies."

However, considering that your opinion is very reasonable, we only keep the bar chart of the global average AAOD, and the spatial distribution map of DRF is moved to the appendix for reference only.

Comments: The authors propose to consider not only the effects of mixing states (Dp/Dc) but also the effects of the proportion of the coated BC core (F) in climate model. So how to determine F value in climate model? Any suggestions?

Response: Thanks for your comments. This paper discovers that in addition to D_p/D_c, F also significantly impacts climate predictions. Since it is challenging to directly simulate F in climate models, statistical analysis of F in different regions through observations is required for practical applications. However, current observation methods also face difficulties in directly observing F, so inversion methods can be leveraged for F measurement. In the future, SP2 can be used to measure D_p/D_c, and optical measurements can also be employed to obtain black carbon absorption characteristics (such as absorption enhancement). With the Mie scattering model proposed in this paper that considers F, F can be retrieved through inversion. By adopting such methods to measure F under various conditions, including different regions and pollution environments, we can obtain F values under different conditions. Finally, these F statistical values under different conditions can be utilized in climate model simulations. We have clarified this in the revised manuscript:

"Since it is difficult to simulate F directly in climate models, a statistical analysis of F in different regions by observations is required for practical applications. However, current observational methods also have difficulty in observing F directly, so inversion methods can be used for F measurement. In the future, SP2 can be used to measure D_p/D_c, and optical measurements can also be used to obtain BC absorption properties (such as absorption enhancement). With the Mie scattering model proposed in this paper, which takes F into account, F can be determined by inversion. By applying such methods to measure F under different conditions, including different regions and pollution environments, we can obtain F values under different conditions. Finally, these statistical F values under different conditions can be used in climate model simulations. Recent studies have shown that Mie-based estimates of absorption enhancement can still overestimate measured values even when accounting for non-uniform mixing states, underlining the importance of our proposed refinement (Huang et al., 2024; Fierce et al., 2020)."

Comments: Lines 39: please add the corresponding references that point out that that simplifying the microphysical properties of BC aerosols can lead to inaccurate determination of mixing states.

Response: Thanks for your comments. We have re-written this paragraph and added some references in the revised manuscript.

Comments: Line 75: the second "of" should be changed as "and".

Response: Thanks for your comments. We have corrected it in the revised manuscript.

Comments: The font size varies a lot in different figures. For figure 2, 5, 6, 7,10, 11 and 12, the font size should be enlarged.

Response: Thanks for your comments and suggestions. We have enlarged the font size in the revised manuscript.

Comments: In figure 2, 5 and 10, it is hard to figure out the results, because there are too many legends and the colors are hard to distinguish. Please replot and improve the quality.

Response: Thanks for your comments and suggestions. To make the figures clearer, we have divided one figure to different figures in the revised manuscript.

Comments: What is the difference between "BC" and "BCs"? Why do the authors use these two expressions?

Response: Thanks for your comments. All "BCs" are uniformly expressed as "BC" in the revised manuscript.