RESPONSE LETTER (amt-2023-53)

Title: Numerical investigation on measurement errors of mixing states of fractal black carbon aerosols using single-particle soot photometer and the effects on radiative forcing estimation

Dear Joshua Schwarz:

We have revised our manuscript based on your comments. 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.

Comments:

Unfortunately, I think another revision is necessary, as it appears that your new calculations don't combine forward and back-scattering. The SP2 uses a single intra-cavity laser - meaning that a single scattering detector measures the combined forward- and back-scattering from each particle. If you were already combining them for your main figures, this can merely be corrected/clarified in the text. In the Supplemental figure 1, however, it doesn't make sense to separate forward and back scattering, as the SP2 is not capable of doing this. Here better to just show combine forward/back into a single scalar measure of scattering for the comparison.

Response:

Thanks a lot for reviewing our revised manuscript again and for the valuable constructive comments. We also noticed the point you mentioned above during our retrieval study.

As shown in the last column of Figure 1, the dashed lines represent the sum of forward and backward differential scattering cross-sections for different fractal models (closed-cell model and coated-aggregate model) with all the preset morphology parameters at fixed BC core diameters (D_c). The curves indicate the variation of the sum of the forward and backward differential scattering cross sections with coated particle diameter D_p for the spherical core-shell model with fixed D_c. The horizontal coordinate corresponding to the point where the dashed line and the curve intersect is the retrieved result of the optical equivalent D_p. It can be seen from Figure 1 that the curves corresponding to the sum of forward and backward differential scattering cross-sections become more and more flat with the increase of D_c, which means that less optical equivalent D_p can be retrieved based on Mie theory. Therefore, we infer that the mixing state of some coated soot particles may be ignored by SP2 in the laboratory and field observations.

Therefore, we further explore the performance of the BC mixing state retrieve based on forward and backward scattering separately. Through the comprehensive analyses of all the simulated optical properties, we found that compared to retrieval using backward scattering and a combination of both forward and backward scattering, the retrieval using forward scattering can avoid the coated BC particles being missed by SP2 to the greatest extent. For example, for BC particles with D_c=371.3 nm, the mixing states of soot particles missed by using forward scattering is half of that missed by using a combination of both forward and backward scattering. Therefore, we think retrieving the mixing state using forward scattering only may be the most efficient for coated BC particles which can be roughly represented by the closed-cell model and the coated aggregate model.

Our results are meaningful in improving the optical cavity structure of the SP2 instrument. However, it should be noted that we only focused on the closed-cell model and coated-aggregate model, mixing state retrieval performance comparisons of retrieval using forward scattering, backward scattering, and a combination of both forward and backward scattering should be further conducted in future based on more kinds of complex morphological models of coated soot particles.

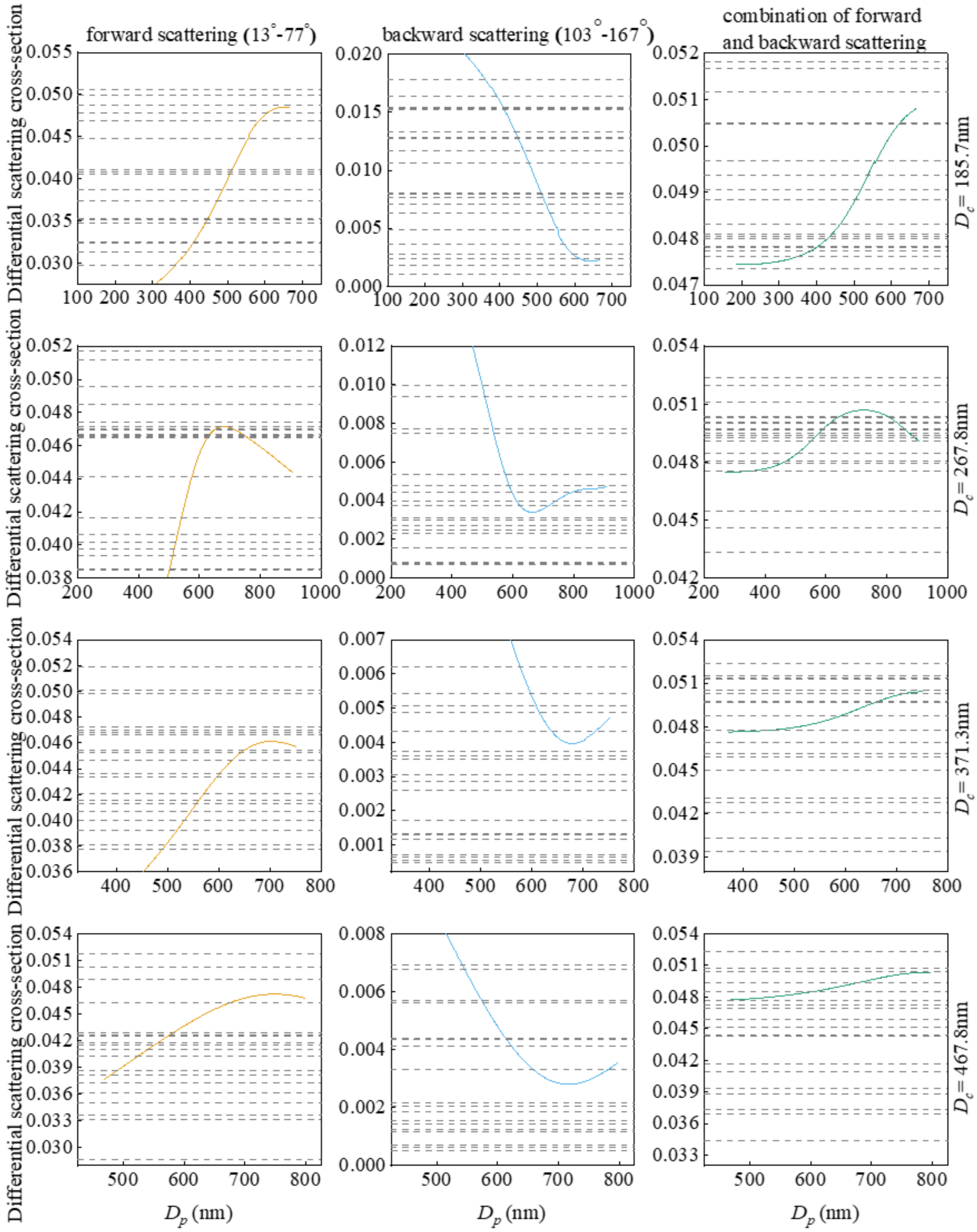

Figure 1. The variations of differential scattering cross-sections with coated particle diameter D_p of core-shell models for soot core diameter D_c=185.7 nm, 267.8 nm, 371.3 nm, 467.8 nm. The dashed lines indicate the differential scattering cross-sections of the fractal model with different morphology parameters. The first column represents the forward differential scattering cross-sections, the second column represents the backward differential scattering cross-sections, and the third column represents the sum of the differential scattering cross-sections integrated in both directions.

For the sake of rigor, related descriptions in the L169, L205, and L375 in the firstly revised manuscript have been modified as follows in the second version of the revised manuscript:

"The two scattering signal detectors of the SP2 are distributed at forward scattering and backward scattering directions to simultaneously measure the forward and backward scattering of each particle, which have view angles (θ) range in 13-77° and 103-167°, respectively (Wu et al., 2023)."

"When the combination of forward and backward differential scattering cross-sections is employed to retrieve BC mixing state, D_p/D_c for a noteworthy amount of coated soot particles cannot be retrieved based on Mie scattering theory due to the different optical properties between spherical and fractal models, which means that the mixing states of some coated soot particles may be ignored. Therefore, the retrieval performances of the BC mixing state based on forward and backward scattering are further explored, separately. Comprehensive comparisons showed that the retrieval based on forward scattering can avoid the coated BC particles being missed by SP2 to the greatest extent. Therefore, the retrieved results of mixing states based on forward scattering are selected for further discussion in the following sections. **Figure 3** shows the retrieval results and relative errors (*RE*) of mixing states of thinly coated soot under different fractal dimensions (D_f) and volume equivalent particle diameter ratio shell/core ($D_{p,v}/D_{c,v}$), the colored lines and bars stand for retrieved D_p/D_c and *RE*, respectively."

"For future work, more morphological models that are suitable for modeling the microstructure of coated soot aerosols should be considered, the retrieval performance of mixing state based on forward scattering, backward scattering, and their combination should be explored, and unified parameterization schemes of retrieval errors are much needed. Furthermore, comprehensive field or laboratory studies for the validation of the possible errors in mixing states are also future directions worth the effort. "