

## Responses to the comments of the reviewer #1

(The responses are highlighted in blue)

First of all, we would like to thank the three anonymous reviewers for their thoughtful review and valuable comments to the manuscript. In the revision, we have accommodated all the suggested changes into consideration and revised the manuscript accordingly. All changes are highlighted in the revised manuscript in **BLUE** in the revision.

In this response, the questions and comments of reviewers are in black font, and responses are highlighted in **BLUE**. The changes made in the revised manuscript are marked in **RED** font.

This is a solid contribution on an important subject. I appreciate the authors' use of state-of-the-art modeling techniques for the study of soot-containing aerosols with highly complex morphologies. The technical content of the paper appears to be correct, and the conclusions are well justified. I have only three minor comments.

1. In Section 2.3, an appropriate generic reference for the DDA would be J. Quant. Spectrosc. Radiat. Transfer 106, 558-589 (2007).

**Response:** Thanks for pointing it out. We have modified it in the revised manuscript.

2. The authors model randomly oriented non-spherical aerosols. The use of the model of randomly oriented particles has two aspects (see the recent rigorous analysis in Optics Letters 42, 494-497 (2017)). First, the orientation distribution function must have a specific mathematical form, so I wonder whether this is the case with the computer program used to calculate light scattering. Second, technically speaking, the computation for a non-spherical particle must be supplemented by the computation for its mirror image. I wonder whether this was done, or it was found that the two computations yield very close results. These two issues need to be clarified.

**Response:** Thanks for your comments. First of all, we agree that the calculations for randomly oriented non-spherical aerosols should be clarified. The non-spherical particles that we considered in this manuscript are atmospheric aerosols. It is reasonable to assume the orientations of black carbon (BC) particles in the atmosphere is completely random, that is to say that the probability of every orientation is identical. As a result, the normalized probability density function of particle directions is nearly a constant. Therefore, Eqs. (3) in the study of Mishchenko and Yurkin, (2017) is satisfied. We have added some text and related reference in the revised manuscript:

**“In this study, all the radiative properties of BC were calculated based on the assumption that BC**

particles and their mirror counterparts are present in equal numbers in ensemble of randomly oriented particles. In the atmosphere, it is reasonable to assume that the possibility of each particle direction is identical, which mathematically satisfies the definition of random orientation (Mishchenko and Yurkin, 2017).”

Rigorously, the computation for a non-spherical particle should indeed be supplemented by the computation for its mirror image. However, Kahnert (2017) has demonstrated that the calculations for closed-cell model calculated using DDA by numerically averaging over each particle direction and those using MSTM don't deviate largely. This indirectly verifies that the results for randomly oriented non-spherical aerosols are close to their mirror counterparts. In addition, we found little changes by altering the option of `target_euler_angles_deg` in MSTM. Therefore, we didn't provide the computation for their mirror counterparts.

3. The authors' analysis of the differences between the effects of absorbing and non-absorbing shells is quite interesting. It would be instructive to compare their observations with those in Optics Letters 39, 2607-2610 (2014).

Response: Thanks for your suggestion. We have compare the results in present study with the results presented in Mishchenko et al. (2014) in the section 3.1 of revised manuscript:

“For aged BC with thick coatings, BC absorption is underestimated at the UV, visible, and IR wavelengths (Kahnert et al., 2012). Mishchenko et al. (2014) has also demonstrated that the  $C_{abs}$  of thickly coated with non-absorbing coatings is significantly underestimated by a core-shell sphere, and investigated the effects of off-center of BC. Their results indicated that the  $C_{abs}$  of aged BC covered with thickly non-absorbing coatings are approximately 1.44 times higher than those calculated with a core-shell sphere model. Nevertheless, the effects of coating absorption on the applicability of the core-shell sphere model have not been evaluated”.

Kahnert, M.: Optical properties of black carbon aerosols encapsulated in a shell of sulfate: comparison of the closed cell model with a coated aggregate model, Opt Express, 25, 24579-24593, 2017.

Mishchenko, M. I., and Yurkin, M. A.: On the concept of random orientation in far-field electromagnetic scattering by nonspherical particles, Opt Lett, 42, 494-497, 2017.

Mishchenko, M. I., Liu, L., Cairns, B., and Mackowski, D. W.: Optics of water cloud droplets mixed with black-carbon aerosols, Opt Lett, 39, 2607-2610, 2014.