Response to Reviewers' comments on "Variation of the radiative properties during black carbon aging: theoretical and experimental intercomparison" by He et al.

Reviewer #2

"The authors theoretically investigate the effects of morphology on optical properties of black carbon using the GOS approximation and compare the theoretical results with laboratory experiments. After that, the developed optical model was implemented to the radiative transfer simulation for evaluating the evolution of direct radiative forcing of BC with aging. Overall, this paper is well written and I recommend its publication in ACP, provided the following comments/questions are taken into account in revision."

Response: We thank the reviewer for kind words. We have provided itemized responses to the following comments.

General Comments:

1. *"Although the observed shape of internally-mixed compounds with BC (e.g., sulfate, organics) is usually dissimilar to composite of spheres (e.g., Fig.5 of Adachi et al. 2010), they are simply represented by a cluster/composite of spheres or core-shell spheres throughout this work. I hardly imagine physical processes (i.e., condensation, coagulation, cloud processing) that can produce "closed-cell" and "open-cell" structures (Fig.1) in the atmosphere. It seems to me that use of these two morphological models is too artificial and only of theoretical interest. It is more plausible to adopt nonspherical morphological models (e.g., "thinly-coated" or "partial thin-coating") without restriction to cluster/composite of spheres, instead of unrealistic "closed-cell" and "open-cell"."*

Response: We thank the reviewer for this comment. To clarify, the "closed-cell" model shown in Figure 1 is to mimic the observed structure presented in Strawa et al. (1999), while the "open-cell" model is to mimic the structure generated in the laboratory by Stratmann et al. (2010). Thus, "closed-cell" and "open-cell" structures are physical models to approximately represent actual observations. We have included discussions on the limitation of using "closed-cell" and "open-cell" structures in the following:
Lines 157-159:
"In this study, six typical BC coating structures (Fig. 1) have been considered for Stages II and III to approximately represent observations in the real atmosphere or laboratory, …"
Lines 170-173:
"We wish to note that the six coating structures used in this study, including closed-cell and open-cell structures, are theoretical models and as such, they may not completely

capture detailed BC coating structures from aircraft and ground-based observations."

We also agree with the reviewer that the nonspherical morphology models (e.g., partial thin-coating) without restriction to composite of spheres are more realistic, which, however, could be much more complex in terms of representing the structure with geometric parameters compared with primary spherules used in the present study. This will be investigated in our future work. We have added discussions on the limitation of assuming composite of spheres in Lines 289-292 as follows:

"We note that assuming a cluster of spheres for the above-mentioned coating structures may not be sufficiently realistic and that nonspherical morphology models without restrictions to composite of spheres appear to be more plausible (Adachi et al., 2010), a challenging subject to be investigated in future work."

2. *"If the authors restrict the model particles to clusters/composites of spheres, why don't you use numerically-exact superposition T-matrix method (e.g., MSTM code developed by Mackowski) instead of the GOS approximation. MSTM version 3 can be applied to any internal and external composites of spheres without surface cut, encompassing all morphological models assumed in this paper. The authors need to explain the advantages of GOS compared to MSTM (or DDA or FDTD) under the ranges of size parameter and refractive index considered in this work."*

Response: The main reason that we used the GOS approach is that it is computationally efficient and sufficiently accurate for the simulations involved in this manuscript. Validation of GOS has been reported by Takano et al. (2013). In particular, we showed that the GOS and superposition T-matrix results are both close to the observed specific absorption of BC aggregates for the size range used in the present work. Also, in Liou et al. (2011), we compared GOS results for specific absorption with superposition T-matrix results published in Liu et al. (2008) and demonstrated close comparison for the mass absorption coefficient measured by Sheridan et al. (2005). Furthermore, we respectfully submit that the comparison between GOS and experimental results conducted in this study constitutes a significant verification of the GOS approach. Also, GOS does not pose a limit on the number of spherical monomers of an aggregate from the perspective of practical light scattering computations. Some numerical methods such as DDA and FDTD require a substantial amount of computational resources and are not practical for moderate and large size parameters. Because the GOS approach is quite accurate, using either GOS or MSTM will not alter the major findings presented in this study. We have incorporated additional discussions in the track-change manuscript to reflect the above-mentioned points:

Lines 242-245:

"Takano et al. (2013) showed that the coupled GOS-RGD and superposition T-matrix results are both close to the observed specific absorption of BC aggregates for the range of size parameter considered in the present study."

Lines 268-269:
"The comparison between GOS and experimental results in this study provides an additional dimension of validation/cross-check of the GOS approach."

Specific Comments:

1. *"P.19842, L.25~ 'Liou et al. (2010, 2011) and Takano et al. (2013) demonstrated that the single scattering properties of aerosols with different sizes and shapes determined from the GOS approach compare reasonably well with those determined from the Finite Difference Time Domain (FDTD) method (Yang and Liou, 1996) and DDA (Draine and Flatau, 1994) for column and plate ice crystals, the superposition T-matrix method (Mackowski and Mishchenko, 1996) for fractal aggregates, and the Lorenz–Mie model (Toon and Ackerman, 1981) for a concentric core-shell shape.' In these referred papers, I could not find any results supporting the accuracy of GOS for BC-containing particles with morphologies "off-center core-shell", "closed-cell aggregate", "partially encapsulated", "open-cell aggregate", and "externally attached", by mean of comparison with numerically-exact techniques (e.g., superposition T-matrix). The authors need to show results (or refer specific part of some publications) supporting the accuracy of GOS/RDG approach for all the morphologies assumed here, because it seems difficult to quantify the error of GOS/RDG approximation without comparisons with some numerically-exact solver of Maxwell equation."*

Response: Please see the Responses to general comment #2. In addition, we would like to point out that every light-scattering approach has its limitation, including the superposition T-matrix methods. Moreover, the GOS approach has been cross-validated with both observations and T-matrix/DDA/FDTD (Liou et al., 2011; Takano et al., 2013) for various particle structures as stated in the text (Lines 225-231), which shows the accuracy and capability of GOS in dealing with different BC coating structures to certain extent. Lastly, we submit that the current comparison of GOS results with laboratory measurements for extinction, absorption, and scattering cross sections provides an additional dimension of validation/cross-check of the GOS approach. In response to the reviewer comments, we have incorporated additional discussions to reflect the preceding points (see also the response to general comment #2).

2. *"P.19843, L3~ 'Moreover, compared with other numerical methods, the GOS approach can be applied to a wider range of particle sizes, shapes, and coating morphology with a high computational efficiency, including very large particles (e.g., ~100–1000 μm snowflakes) and complex multiple inclusions of aerosols within irregular snow grains (Liou et al., 2014; He et al., 2014), in which the FDTD, DDA, and T-matrix methods have not been able to apply.' As far as I read the references cited in this paper, the accuracy of GOS has been investigated only limited number of particle shapes: sphere, core-shell sphere, hexagonal ice-crystals, and aggregate of spheres. The authors* *need to show (or refer) the direct evidences on the accuracy of GOS in other shapes considering in this work. Computational efficiency seems to be of secondary importance at least for the purpose of this paper."*

Response: Thank you. Please see the Responses to general comment #2 and specific comment #1.

3. *"P.19843, L13～ 'To supplement GOS, we have developed the Rayleigh–Gan–Debye (RGD) approximation coupled with GOS for very small particles, which has been cross-validated with the superposition T-matrix method (Takano et al., 2013). The combined GOS/RGD approach can be applied to size parameters covering 0.1 to 1000.' Please clarify which of GOS and RGD was assumed in each theoretical calculation in this paper."*

Response: We thank the reviewer for this comment. To clarify, we have included the following statement in Lines 246-248:
"In the present study, the coupled GOS-RGD approach is used for fresh BC aggregates (Stage I), while the GOS approach without RGD coupling is used for coated BC particles (Stages II and III)."