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Interactive Comment

# *Interactive comment on* "Variation of the radiative properties during black carbon aging: theoretical and experimental intercomparison" *by* C. He et al.

**Anonymous Referee #2**

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Review comments on

Variation of the radiative properties during black carbon aging: theoretical and experimental intercomparison by He et al.

General comments 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.

Especially, I raise two major comments that should be taken into account in revision:

(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".

(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.

Specific comments

Comment (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 andAckerman, 1981) for a concentric core-shell shape."

In these referred papers, I could not find any results supporting the accuracy of GOS



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for BC-containing particles with morphologies “off-center core-shell”, “closed-cell aggregate”, “partially encapsulated”, “pen-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.

Comment (2) P.19843, L3$\sim$ “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 effciency, including very large particles (e.g., $\sim$100–1000 $\mu$m 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.

Comment (3) P.19843, L13$\sim$ “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.

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 19835, 2015.