

Interactive comment on "Accounting for the effects of non-ideal minor structures on the optical properties of black carbon aerosols" by Shiwen Teng et al.

Anonymous Referee #2

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General comment: This paper addressed the effects of minor geometric structures of the black carbon particles to their optical properties using DDA simulations, in a more detailed-and-comprehensive manner than any previous publications. I can recommend publication considering the huge efforts and the excellent quality of writing. However, I have some questions and critical comments on the methods and the result interpretations which should be taken into account in the paper before its publication.

Major comments: (1) The considered size range of BC particles in the DDA simulations seems to be Dv (volume equivalent diameter) < \sim 250 nm (at a=15nm, N=600), which could be a minor fraction of the whole size range of ambient BC particles. If the authors

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suggest the significance/usefulness of their results for radiative forcing estimations and aerosol remote sensings (page 1. line 15), they should provide appropriate rationale on the considered size-range. It is not evident whether the "correction ratio" defined and evaluated by the authors also applies to the BC particles not smaller than the wavelength.

(2) If the authors include any quantitative interpretations of the DDA-simulated optical properties with the accuracy of the order of several percents, they need to include careful evaluations of the absolute accuracy of their DDA results. The surface granularity inherent to the DDA model is known to cause systematic overestimation of the absorption cross-section for Rayleigh-sized particles [Draine 1988], which is persistent even in the long-wavelength limit. This is one of the major reasons for the difficulty of modeling the soot optical properties using the DDA [Yurkin 2007; Moteki 2016]. I can suggest a quick evaluation of the systematic error of DDA results presented in this paper by comparing the exact Mie solution and DDA calculation (using ~900 dipoles) for an isolated monomer. To my feeling, it might be difficult to separate the effects of minor structure M2 and the DDA-artifact unless each monomer is represented by a huge number of dipoles (> ~10000).

(3) In general, particle's orientation relative to the propagation direction of the incident wave substantially affects the optical properties (e.g., absorption cross-section) for a fractal-like cluster of spheres. Please clarify how the authors treat/assume the particle's orientation in their DDA simulations because the derived "correction ratio" might also change depending on the assumed orientation.

(4) The authors assumed "1.8+0.6i" for the refractive index of BC throughout this paper. However, this parameter is still highly uncertain [Bond and Bergstrom 2006] and might vary depending on the emission source. The authors need to provide appropriate reasons for choosing this value and explain the expected consequence of the assumption.

Minor/technical comments: (1) page 3 line 16: Please clarify that the "266 nm" means wavelength.

References Yurkin, Maxim A., and Alfons G. Hoekstra. "The discrete dipole approximation: an overview and recent developments." Journal of Quantitative Spectroscopy and Radiative Transfer 106.1-3 (2007): 558-589.

Draine, Bruce T. "The discrete-dipole approximation and its application to interstellar graphite grains." The Astrophysical Journal 333 (1988): 848-872.

Moteki, Nobuhiro. "Discrete dipole approximation for black carbon-containing aerosols in arbitrary mixing state: A hybrid discretization scheme." Journal of Quantitative Spectroscopy and Radiative Transfer 178 (2016): 306-314.

Bond, Tami C., and Robert W. Bergstrom. "Light absorption by carbonaceous particles: An investigative review." Aerosol science and technology 40.1 (2006): 27-67.

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