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Comment on acp-2020-1290

Anonymous Referee #2

Referee comment on "Optical properties of coated black carbon aggregates: numerical simulations, radiative forcing estimates, and size-resolved parameterization scheme" by Baseerat Romshoo et al., Atmos. Chem. Phys. Discuss.,
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GENERAL REMARKS

The manuscript presents results from modelling studies of the modification of optical properties of fractal-like black carbon (BC) particles, when the radius of the primary particles, fractal dimension, fraction of organics, wavelength, and mobility diameter are varied. The study uses the multiple sphere T-matrix (MSTM) method. Based on the results of the studies the authors estimate effects of the parameter variations on the radiative forcing by the fractal-like BC aggregates and develop a parametrisation scheme for radiative properties of BC fractal aggregates, which is applicable for modelling, ambient and laboratory-based BC studies. The study concludes with a detailed analysis of uncertainties when applying the proposed parameterisations.

The topic is of relevance for the modelling of the climate impact of BC containing aerosol particles. The study claims a significant improvement of results of climate model studies compared to the usually applied core-shell model for coated spheres using Mie theory. The parameter span used in the variation studies covers a wide range of BC properties from laboratory-generated aerosol to ambient BC containing particles. The study is well designed and systematically conducted. The presentation is concise and clear, and the topic fits well into the scope of the journal. Before being acceptable for publication, two major issues need to be tackled: the increase in knowledge compared to numerous previous studies on the radiative effects of fractal-like BC agglomerates is not clearly presented, and the discussion of results requires improvement. Details are specified in the next section.

SPECIFIC COMMENTS

1. As stated briefly in the General Remarks, a clearer presentation of the novelty of the

study and the gain in knowledge is requested. There have been numerous model studies published on the optical properties of fractal-like BC particles published, and various studies are available which conduct in-depth comparisons of model analyses with observations to quantify the discrepancy between applied theories and observations. It is recommended to explain the increase in knowledge triggered by this work in the introduction section.

2. Introduction section and later: Some key references should be included and discussed:

- Liu et al. (2020) discussed the life cycle of light-absorbing carbonaceous aerosols in the atmosphere, this paper should be included in the introduction section;
- one of the key papers on the optical properties of fractal-like aggregates by Berry and Percival (1986) is missing, here the authors discuss already that optical properties of fractal-like aggregates are determined by the primary spheres;
- the entire discussion of non-fractal light absorbing carbonaceous matter in the atmosphere from biomass burning is missing (Chakrabarty et al., 2010; Chen and Bond, 2010; Chung et al., 2012; Feng et al., 2013; Fleming et al., 2020). This aerosol type plays an important role in atmospheric light absorption but is not concerned by the proposed parameterisation. It needs to be clearly expressed that the proposed parameterisations do not apply to this aerosol type which, however, contributes significantly to the light absorbing carbonaceous aerosol.

3. The entire topic of the impact of atmospheric processing on particle shape and resulting optical properties requires reconsideration. There are numerous reports on the change in aerosol morphology during atmospheric processing. Particularly, Liu et al. (2017) presented a detailed study on the effect of coating of fractal-like particles on the absorption properties. This study is not mentioned here although it is of high relevance since it illustrates the gradually decreasing impact of the fractal-line structure of the BC particle when becoming more and more coated; see Sections 3.3 and 3.4 of the manuscript. This effect should also be considered when discussing the absorption Ångström exponent and enhancement factors in Section 3.6.

4. In Section 2.4, the authors use the expression by Chylek and Wong (1995) for the calculation of the radiative forcing at TOA. A brief discussion on the relationship to the more widely used radiative forcing efficiency according to Haywood and Shine (1995) and Sheridan and Ogren (1999) should be added.

MINOR ISSUES

Title: Suggested rephrasing: "Optical properties of ... "; the novelty of the study should also be reflected in the title. Otherwise the publication becomes indistinguishable from other papers with almost similar titles.

Line 55: Lab slang should be avoided and hence rephrasing of “by-products of burning like organics” to, e.g., “by-products of combustion like organic vapours” is suggested.

Line 57: Here the “reshaping of the BC particles into more spherical structures” by vapour deposition is mentioned but the fractal-like structure of fresh combustion aerosol has not been introduced before.

Line 61: Do the authors mean “is less absorbing by nature”?

Line 96: Suggested rephrasing: “as a function of size for various morphologies”.

Line 137: The effect that “light absorption measurements are insensitive to the radii of the primary particles” is already explained by Berry and Percival (1986); this should be mentioned here.

Line 218: The correct reference is Kim et al. (2015), this should be corrected throughout the manuscript.

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