**Reviewer #2 (R2):**

The authors thank the reviewer for providing constructive comments and insightful suggestions on the manuscript. We highly appreciate your time in reviewing the manuscript. The point-by-point response to all the comments and suggestions of reviewer #2 (R2) is provided in the following sections. For easy visualization, the reviewer's comments (R2 C) are provided in blue and the author's response (AR) is given in black color below the reviewer's comment. All the comments/suggestions were taken into consideration and incorporated in the revised manuscript which has improved the quality of the revised manuscript. The revised parts of the manuscript along with the references are shown in red.

**R2 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.

**AR:** Thank you for the useful and constructive general remarks. As suggested, the novelty of this work with reference to the previous studies on the topic is highlighted in a more prominent way in the revised manuscript. The discussion of results has been improved in the revised manuscript, and the details are given in the point-by-point response to the specific comments of the Reviewer below.

**Specific comments:**

**R2 C1:** 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.

**AR:** Thank you for the comment. We agree with the reviewer that the novelty of this work must be highlighted with reference to the past modeling studies on the optical properties of BC fractal aggregates and their comparisons to the modeled results with other observations. Before explaining and highlighting the novel research conducted in this study, we provide the context and highlights of the most relevant previous studies on the topic:

- Kahnert et al., 2010: The size-dependent empirical formula for the optical properties of BC aggregates was derived for the wavelength range from 200nm up to 12.2µm. The empirical formula derived from the optical properties modelled using the T-matrix method, can be used as an input to a radiative transfer model.

- Smith and Grainger., 2014: The radiative properties of pure BC fractal aggregate, i.e. without any external coating, were investigated, further developing a parametrization for the optical properties of pure BC fractal aggregates with respect to the number of primary particles ($N_s$).

- Luo et al., 2018: A method to estimate the optical properties of BC aggregates was proposed using a machine learning method, support vector machine.

- Liu et al., 2018: Empirical equations on the BC Ångstrom absorption exponent (AAE) was derived for different BC morphologies.

- Liu et al., 2019: A database was developed to calculate the optical properties of BC particles using the including numerical model, the multiple-sphere T-matrix method (MSTM). The database includes the aggregation structure, refractive index, and particle size of BC particles.

- Liu et al., 2020: It was emphasised that improved size-resolved datasets and models for the light absorbing carbon (LAC) is required that include information about the optical properties, OC/BC ratio, burning phase or fuel types.

Despite the numerous modeling-based studies, the developed databases/parametrisations lack information about the external coating of the BC fractal aggregate. The reason for this might be the computational load for such a task is substantial due to the time-consuming simulations. In this work, we covered the gap of the missing parameter of external coating. The external coating parameter is quantified through the fraction of organics ($f_{\mathrm{organics}}$). The computationally expensive task of investigating the optical properties of BC fractal aggregates as a function of the radius of the primary particle ($a_{\mathrm{o}}$), fractal dimension ($D_{\mathrm{f}}$), fraction of organics ($f_{\mathrm{organics}}$), wavelength ($\lambda$), and mobility diameter ($D_{\mathrm{mob}}$) was conducted. The size-resolved parametrization scheme for the optical properties of the coated BC fractal aggregates is provided at various fractal dimension ($D_{\mathrm{f}}$) and fraction of organics ($f_{\mathrm{organics}}$).

Accordingly, the lines 80-97 in 'Introduction' section of the preprint were rewritten in the revised manuscript as follows:

> Discrepancies due to Mie theory have caused an increasing interest in the simulation of the BC optical properties assuming a more realistic fractal morphology. The size-dependent empirical formula for the optical properties of BCFAs was derived for the wavelength range from 200nm up to 12.2µm (Kahnert et al., 2010). The optical properties of pure BCFAs, i.e., without any external coating, were investigated by Smith and Grainger (2014), further developing a parametrization for optical properties of pure BCFAs with respect to the number of primary particles ($N_{\mathrm{s}}$). A method to estimate the optical properties BCFAs was proposed using the machine learning method, support vector machine (Luo et al., 2018). Empirical equations on the BC Ångstrom absorption exponent (AAE) were derived for different BC morphologies (Liu et al., 2018). A database containing optical data was developed that includes the aggregation structure, refractive index, and particle size of BCFAs (Liu et al., 2019).
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> However, the previous modelling-based studies were not able to take into account the information about the parameter: external coating of the BCFAs. The reason for this could be that the time-consuming simulations make the computational load for such a task substantially large. Additionally, various ambient and laboratory studies have emphasized the role of organic external coating in influencing the BC absorption and scattering properties (Zhang et al., 2008, Ouf et al., 2016; Dong et al., 2018, Shiraiwa et al., 2010). It was also pointed out that improved size-resolved datasets and models for the light absorbing carbon (LAC) is required that includes observables like optical properties, OC/BC ratio, burning phase or fuel types (Liu et al., 2020). Therefore, a size-resolved parametrization scheme for optical properties of BCFAs including the external coating parameter is very important.
>
> This investigation involved computationally intensive modeling aimed at understanding and quantifying the changes that BCFAs and their optical properties undergo by simulating various cases of the BCFAs under an elaborated systematic approach that is designed to span a wide parameter space. The external coating parameter is quantified through the fraction of organics ($f_{\mathrm{organics}}$). The BCFAs cases are classified according to various $f_{\mathrm{organics}}$, morphologies, and wavelengths. This approach of categorization involving the $f_{\mathrm{organics}}$ of BCFAs is aimed to bridge the gaps that are present in the modeled optical data from the previous studies. The optical properties were calculated using the T-matrix code (Mackowski et al., 2013) and the findings are presented and discussed with respect to the equivalent mobility diameter ($\mathrm{D}_{\mathrm{mob}}$) making it more relevant and comparable for laboratory, and ambient studies in which mobility spectrometers are often used for size classification.
>
> The study highlights how modifications in the morphology and $f_{\mathrm{organics}}$ of BCFAs can further influence the BC radiative forcing. Finally, the parameterization scheme for optical properties (extinction, scattering, and absorption) of coated BCFAs was developed as a function of size for different morphologies, $f_{\mathrm{organics}}$, and wavelengths.

Liu, D., He, C., Schwarz, J. P., and Wang, X.: Lifecycle of light-absorbing carbonaceous aerosols in the atmosphere, npj Clim Atmos Sci, 3, 40, doi: 10.1038/s41612-020-00145-8, 2020.

Liu, C., Xu, X., Yin, Y., Schnaiter, M. and Yung, Y. L.: Black carbon aggregates: A database for optical properties, J. Quant. Spectrosc. Radiat. Transf., doi: 10.1016/j.jqsrt.2018.10.021, 2019.

**R2 C2:** Introduction section and later: Some key references should be included and discussed:

**AR:** Thank you for sharing some important and useful references. All the suggested references were included in the revised manuscript, further details given below for each sub-comment in blue.

- 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;

  **AR:** In the study by Liu et al. (2020), the requirement of incorporating observables such as OC/BC ratio, size distribution, degree of internal mixing, and hygroscopicity in the models and inventories of light absorbing carbon (LAC) has been highlighted. As suggested, the following lines about the study by Liu et al. (2020) have been added to the 'Introduction' section of the revised manuscript:

  It was also pointed out that improved size-resolved datasets and models for the light absorbing carbon (LAC) are required that includes observables like optical properties, OC/BC ratio, burning phase, or fuel types (Liu et al., 2020). Therefore, a size-resolved parametrization scheme for optical properties of BCFAs including different compositions is needed.

  Liu, D., He, C., Schwarz, J. P., and Wang, X.: Lifecycle of light-absorbing carbonaceous aerosols in the atmosphere, npj Clim Atmos Sci, 3, 40, doi: 10.1038/s41612-020-00145-8, 2020.

- 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;

  **AR:** In the theoretical study by Berry and Percival (1986), the optical cross-sections of absorbing spheres are computed as a function of number of spheres ($N_s$), fractal dimension ($D_f$), and complex refractive index. It is shown that the optics of clusters can be very different, strongly depending on whether the fractal dimension ($D_f$) is less than 2 or greater than 2. As suggested by the Reviewer, the following changes are made in the revised manuscript:

  After line 58 in the 'Introduction' section of the preprint:

  It was theoretically shown in clusters of absorbing spherules that the change in the optical cross-sections with an increasing number of spherules (aggregation) is strongly dependent on the morphology (Berry and Percival, 1986).

  After line 393 in the 'Results and discussion' section of the preprint:

  The dependency of the optical cross-section over the fractal dimension ($D_f$) was pointed out by Berry and Percival, 1986 where the change in the cross-sections depends on whether the fractal dimension ($D_f$) is less than 2 or greater than 2.

  Berry, M. V., and Percival, I. C.: Optics of fractal clusters such as smoke, Opt. Act., 33, 577-591, doi: 10.1080/713821987, 1986.

- 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.

  **AR:** Recent studies have reported a class of organic carbon (OC) with light absorbing properties, known as brown carbon (BrC). The imaginary part of the BrC refractive index increases towards shorter wavelengths, strongly absorbing solar radiation in the blue and near-ultraviolet spectrum. We agree with the reviewer that this aerosol plays an essential role in atmospheric light absorption. In this study, we focused on non-coated and organic coated BC aggregates. If BrC coated BC aggregates was included, the optical properties were to be computed for an addition set of refractive indices. Unfortunately, due to the time consuming nature of the simulations, we were not able to include BrC coatings. It will be very interesting to conduct a similar study for BC fractal aggregates with absorbing organic coating in the future.

As suggested by the Reviewer, the following changes are made in the revised manuscript:

After line 62, in the 'Introduction' section of the preprint, the concept was introduced:

Additionally, there exists a class of organic carbon (OC) with light absorbing properties, known as brown carbon, strongly absorbing solar radiation in the blue and near-ultraviolet spectrum (Fleming et al., 2020; Feng et al., 2004; Chakrabarty et al., 2010; Chen and Bond, 2010).

After line 166, in the 'Methods' section of the preprint:

It must be noted that the focus of this study is on BCFAs with coatings consisting of non-absorbing organics. If a brown carbon coating was to be included in the study, information and extra computational time regarding their refractive indices was needed. Unfortunately, due to the time-consuming nature of simulations, the generated database could not include BCFAs with brown carbon coating.

In 'Conclusion' section, a short outlook was given:

It is important to mention that the parametrisation schemes and databases based on realistic representation of BC like the one developed in this study is a successful step forward towards a more accurate estimation of the BC radiative forcing in climate models. Therefore, further studies must be conducted developing such databases that include more observables like varying refractive index, hygroscopicity, and light absorbing coating.

Chakrabarty, R. K., Moosmueller, H., Chen, L. W. A., Lewis, K., Arnott, W. P., Mazzoleni, C., Dubey, M. K., Wold, C. E., Hao, W. M., and Kreidenweis, S. M.: Brown carbon in tar balls from smoldering biomass combustion, Atmos. Chem. Phys., 10, 6363-6370, doi: 10.5194/acp-10-6363-2010

Fleming, L. T., Lin, P., Roberts, J. M., Selimovic, V., Yokelson, R., Laskin, J., Laskin, A., and Nizkorodov, S. A.: Molecular composition and photochemical lifetimes of brown carbon chromophores in biomass burning organic aerosol, Atmos. Chem. Phys., 20, 1105-1129, doi: 10.5194/acp-20-1105-2020, 2020.

Chen, Y., and Bond, T. C.: Light absorption by organic carbon from wood combustion, Atmos. Chem. Phys., 10, 1773-1787, doi: 10.5194/acp-10-1773-2010, 2010.

Feng, Y., Ramanathan, V., and Kotamarthi, V. R.: Brown carbon: a significant atmospheric absorber of solar radiation?, Atmos. Chem. Phys., 13, 8607-8621, doi: 10.5194/acp-13-8607-2013, 2013.

**R2 C3:** 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.

**AR:** Thank you for the reference to the important study related to our work. In Liu et al., 2017, the relative performance of various methods used for simulating the scattering cross-section and enhancement factors, with respect to the mass ratio, MR ($=M_{\text{non-BC}}/M_{\text{rBC}}$) was reported. Liu et al., 2017 compared four methods of mixing BC and non-BC components: externally mixed, Maxwell–Garnett homogeneously mixed, Bruggemann homogeneously mixed, and the Rayleigh–Debye–Gans (RDG) model. It was found that when $MR < 1$, the simulated results are best represented as having no absorption enhancement, i.e., external mixing is considered. Whereas, when $MR > 3$, it is necessary to consider methods assuming optical lensing effect like the core-shell method. A generalized hybrid model was also developed for estimating enhancement factors.

The findings of the Liu et al., 2017 were related to our study in the following ways:

In 'Introduction' section of the preprint, line 74, a brief reference to the study was given as:

It was shown that the ratio of non-BC to BC components plays an important role in determining the performance of different methods used for simulating the BC optical properties (Liu et al., 2017).

In section 3.4, the following was added:

The gradually decreasing impact of the fractal morphology over the scattering results of coated BC particles was shown by Liu et al., 2017. In this study, it is seen in the case of a non-coated BC particle (Fig .6c), the $C_{\mathrm{sca}}$ is more sensitive to the $D_{\mathrm{f}}$, whereases, when the BC particles are coated (Fig. 7c, Fig 8c), the $C_{\mathrm{sca}}$ is less sensitive towards $D_{\mathrm{f}}$ and $f_{\mathrm{organics}}$. It is observed that the $C_{\mathrm{sca}}$ and *SSA* (Fig. 8c, Fig. 8e) become more sensitive towards $D_{\mathrm{f}}$ when the BCFA grows in size, therefore, the impact of the fractal morphology over the scattering results is also a function of particle size. Moreover, it must be noted that even though there is a decreasing impact of the fractal morphology over the scattering results, optical parameters like $C_{\mathrm{abs}}$, $MAC_{\mathrm{BC}}$, and $g$ showed significant variability towards change in the $f_{\mathrm{organics}}$ (Fig 7a, 7b, 7e, and 7f).

In section 3.6, the following was added:

Liu et al., 2017 emphasized the role of mass ratio of non-BC to BC on the performance of various methods used for simulating the scattering cross-section and enhancement factors of BC particles. In this study, it is shown that the Ångstrom absorption exponent (AAE) calculated from just the MSTM method can show variability of up to a factor of 2 with an increase in the non-BC content ($f_{\mathrm{organics}} > 90\%$). Similarly, it can be seen that the difference in the enhancement factors calculated from the core-shell theory and fractal assuming MSTM method can be up to by a factor of 1.1 to 1.5.

Liu, D. T., Whitehead, J., Alfarra, M. R., Reyes-Villegas, E., Spracklen, D. V., Reddington, C. L., Kong, S. F., Williams, P. I., Ting, Y. C., Haslett, S., Taylor, J. W., Flynn, M. J., Morgan, W. T., McFiggans, G., Coe, H., and Allan, J. D.: Black-carbon absorption enhancement in the atmosphere determined by particle mixing state, Nat. Geosci., 10, 184-U132, doi: 10.1038/ngeo2901, 2017.

**R2 C4:** 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.

**AR:** Thank you for the suggestion. The previous studies have used the multiple reflection model (Haywood and Shine, 1995; Sheridan and Ogren, 1999) to calculate the top of the atmosphere TOA forcing for an optically thin partially absorbing aerosol. The expression by Chylek and Wong (1995) for the calculation of TOA forcing is a simplified version of the multiple reflection model with some implicit approximations. In our study, the simplified version was used since we highlighted the sensitivity of the TOA forcing towards morphology and composition of BC, and the aim was not to provide the exact radiative forcing estimates.

The information given above is added in section 2.4 of the revised manuscript as follows:

The model given by Chylek and Wong (1995) is for the calculation of TOA forcing is a simplified version of the multiple reflection model (Haywood and Shine, 1995; Sheridan and Ogren, 1999) with some implicit approximations. It is important to note that this is an analytical model which can be useful to understand the sensitivities of radiative forcing to various parameters (Chylek and Wong, 1995; Lesins et al., 2002). The simplified version was used in this study to highlight the sensitivity of the TOA forcing towards the morphology and composition of BC. However, the model cannot be used to replace the accurate direct radiative forcing calculations.

**Minor issues:**

1) 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.

**AR:** Thank you for the useful suggestion, we agree that the Title should reflect the novelty of the study.

Accordingly, the Title of the study is changed to: “Optical properties of coated black carbon aggregates: numerical simulations, radiative forcing estimates, and size-resolved parametrization scheme”

2) 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.

   **AR:** Thank you for the suggestion. The phrase “by-products of burning like organics” has been changed to “by-products of combustion like organic vapours” in the revised manuscript.

3) 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.

   **AR:** Thank you for highlighting this point. The concept early stage fractal morphology of BC has been introduced in the ‘Introduction’ section of the revised manuscript as:

   In the early stages of formation, BC particles consist of loosely bound agglomerates made of numerous small spherules, which collide to form strongly bound chain-like aggregates (Michelsen et al., 2017).

   Michelsen, H. A. Probing Soot Formation, Chemical and Physical Evolution, and Oxidation: A Review of In Situ Diagnostic Techniques and Needs. Proc. Combust. Inst. 2017, 36, 717−735.

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

   **AR:** Thank you for the correction. The change has been made in the revised manuscript.

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

   **AR:** Thank you for the suggestion. The phrase “as a function of size at various morphologies” has been changed to “as a function of size for various morphologies” in the revised manuscript .

6) 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.

   **AR:** Thanks for the important reference. The following addition/change has been made in the revised manuscript:

   Contrarily, Berry and Percival (1986) showed that light absorption measurements are insensitive to the radii of the primary particles. Additionally, Kahnert (2012b) pointed out that insensitivity is present when the radii of the primary particle fall in the range of 10 – 25nm.

   Berry, M. V., and Percival, I. C.: Optics of fractal clusters such as smoke, Opt. Act., 33, 577-591, doi: 10.1080/713821987, 1986.

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

   **AR:** Thank you for the correction. The change has been made throughout the revised manuscript.