Reply on RC1

Payton Beeler and Rajan K. Chakrabarty

Author comment on "Constraining the particle-scale diversity of black carbon light absorption using a unified framework" by Payton Beeler and Rajan K. Chakrabarty, EGUsphere, https://doi.org/10.5194/egusphere-2022-163-AC1, 2022

The manuscript "Constraining the particle-scale diversity of black carbon light absorption using a unified framework" by Payton Beeler et al. reveals the effect of morphology of BC core on light absorption enhancement of BC due to the "lensing effect". The study used the Amsterdam Discrete Dipole Approximation (ADDA) method to calculate the particle absorption with varying BC core morphology and different coating compositions. Based on the results of the studies the authors found a factor (phase shift parameter) to describe the increase and decrease of absorption enhancements of BC caused by its morphology, and formulate universal scaling laws centered on the phase shift parameter. This study also provides physics-based insights regarding core-shell approximation overestimating BC light absorption. The presentation is concise and clear, and the topic fits well into the scope of the journal. The manuscript could be considered for publication with the following concerns being addressed.

COMMENT: The novelty of the manuscript is not well presented. There have been a number of numerical studies on optical properties of BC with complex morphology using DDA and T-matrix method. Either the fractal aggregate model or coating scheme has been considered before. It is recommended to explain the advance specifically for this work at least in the introduction section.

RESPONSE: While T-matrix and DDA methods have been used extensively to investigate light absorption by BC, this study is the first to provide physics-based scaling laws for quick calculation of BC optical properties. It is also one of the first studies to provide insight into causes of discrepancies in modeled and measured light absorption enhancement due to internal mixing of BC. Without the use of the scaling laws developed by this work, one would need to rely on computationally expensive methods (T-matrix or DDA) or incomplete models (such as core-shell Mie theory or scaling laws given by Chakrabarty and Heinson, 2018). Additionally, this study is the first to show (per our knowledge) that measurements of BC light absorption using contemporary instruments can be used to infer the morphology of fractal BC aggregates. We have emphasized these points in the introduction of the revised manuscript.

COMMENT: In this study, ρ_{BC} (the phase shift parameter of BC core) shows the influence of BC core morphology on its light enhancement, but ρ_{BC} was determined not only by the morphology, but the size of BC can also influence ρ_{BC} according to formula (1). In 2.1

section, the authors state to calculate with BC core masses between 1 fg and 70 fg, but the BC size calculation was missing in the results. In addition, previous fractal aggregate studies used the fractal dimension (D_f) to represent the morphology, what is the D_f for the freshly emitted, partially collapsed, and collapsed aggregate in this study?

RESPONSE: The gyration radius of BC aggregates in this study ranged from 50 – 300 nm. The D_f of freshly emitted, partially collapsed, and fully collapsed BC was 1.832 ± 0.089, 2.105 ± 0.223, and 3.0, respectively. This has been added to section 2.1 of the revised manuscript.

COMMENT: The absorption enhancement of BC core through the "lensing effect" was also investigated for light-absorbing coating materials like BrC, and the author notices that the total particle absorption is very sensitive to the image refractive index of the coating material. The increase of particles absorption with coating increase was a competition between the increase of BrC absorption and the decrease of the BC enhancement due to less light on the BC core. However, MAEBC in this study shows the total absorption of the particle (e.g. in Fig. 4). It is recommended to subtract the absorption by the BrC shell in order to investigate the "lensing effect" of BC.

RESPONSE: The choice was made to focus on total particle absorption as opposed to separating BC absorption due to lensing and BrC absorption because common methods for measuring light absorption by BC will measure total particle absorption, and will likely not be able to parse whether absorption enhancement is the result of absorbing coatings or increased lensing. Therefore, we chose to develop scaling laws based on total particle absorption in order to make the results applicable for experimentalists. The contribution of BrC absorption to absorption enhancement, as well as the change in BC absorption due to the so-called "sunglass effect" is left for future work (Luo et al., 2021).

COMMENT: Section 2.1: The discussion about the influence of spherical monomer of BC aggregates on its optical properties is missing. Berry and Percival (1986) discussed that optical properties of fractal-like aggregates were determined by the primary spheres. In this study the primary sphere was chosen to be 20nm, Shetty et al., (2021) used 40nm. (https://doi.org/10.1080/02786826.2021.1873909).

RESPONSE: The radius of BC monomers will affect the radius of gyration, and eventually the phase shift parameter. Therefore, the developed framework is able to account for changing BC monomer size. Shetty et al., (2021) utilized BC spheres with diameter of 40 nm, we have chosen the same monomer size, with radius of 20 nm.

COMMENT: Section 2.2: The settings about the ADDA are not well described. The accuracy of ADDA depended on the size of the sub-volume compared to the wavelength of the incident light. What's the resolution of dipoles per wave-length in this study?

RESPONSE: ADDA recommends 10 dipoles per wavelength for accurate estimation of absorption and scattering properties. Our study utilizes > 100 dipoles per wavelength. This has been added to Section 2.2, for clarity.

COMMENT: Fig. 2: Y axis label is missing.

RESPONSE: This has been corrected.