

This manuscript presented a method to predict the optical properties of internally mixed black carbon. Based on this manuscript, we can use a single factor called the phase shift parameter to estimate the change of light absorption of internally mixed BC due to different morphology, mixing-state, mass, and coating composition. The method also can be used to predict the absorption properties of coating material and morphology of BC core. This study is significant for BC optical research since it can help improve uncertainties in the current climate model related to BC absorption. The manuscript is well written, and the presentation is clear. The topic also fits very well into the scope of the journal. However, I have some comments and questions about the paper. After considering, responding, and revising the manuscript based on them, this paper should be considered for publication.

Major Comments:

- 1) This manuscript discusses the influence of the mixing state of BC and coating on the absorption of the entire particle. Have you conducted any simulation for BC mixed with inorganics (e.g., dust, sea salt) then coated by other materials? How would that affect your model results?

We have not considered BC mixed with inorganics. This would alter the effective refractive index of the core, and subsequently alter the phase shift parameter and the absorption cross-section. The model developed in this work will not be accurate for aggregates whose properties vary significantly from those outlined in section 2.2. However, the properties given in section 2.2 are commonly used in large scale models.

- 2) For the ADDA model, could you provide a short description of it? I suggest adding a table of import parameters you used.

The most important input parameter for ADDA calculations is the number of dipoles per wavelength of incident light. We have added a description of this input to section 2.2 of the revised manuscript.

- 3) The first Section 3.1 is not clear to me.
 - a) Does ΔMAC_{BC} represent the MAC of the entire particle - MAC of BC core? Same question for $\Delta \kappa_{coat}$ and Δn_{coat} . Please clarify them.

$\Delta MAC_{BC} / \Delta \kappa_{coat}$ and $\Delta MAC_{BC} / \Delta n_{coat}$ represent partial derivatives of MAC_{BC} with respect to the refractive index. We have changed the notation to $\partial MAC_{BC} / \partial \kappa_{coat}$ and $\partial MAC_{BC} / \partial n_{coat}$, and clarified in the text.

- b) You used 30 BC aggregates. Does that mean for each aggregate, you used a different combination of n_{coat} and κ_{coat} ? Why do you only have 7 points in figure 3? Shouldn't you have 3 points for κ and 3 points for n at each R_{BC} ?

For each aggregate, we calculated the optical properties using 9 combinations of n_{coat} and κ_{coat} at 7 different values of R_{BC} . The 7 points in figure 3 are representative of the 7 values of R_{BC} , and the points and error bars are the average and standard deviation of the

partial derivative of total particle absorption normalized by BC mass with respect to n_{coat} and κ_{coat} .

- c) I also noticed that in Fig. 3, at $R_{\text{BC}} = 10$, the ratio decreases. Do you have any explanation for that?

We do not have any insights to the reason for this. However, since these aggregates are randomly selected this may be washed away if more aggregates are used in the averaging.

- d) I am not surprised to see $\Delta\text{MAC}_{\text{BC}}/\Delta\kappa_{\text{coat}}$ is always greater than $\Delta\text{MAC}_{\text{BC}}/\Delta n_{\text{coat}}$ since k changes over an order of magnitude, while n only changes within 0.2.

We wanted to show that changes in κ_{coat} that have been observed in ambient particles cause large changes in MAC_{BC} . However, the real part of the refractive index does not show as much variability. Therefore, we opted to constrain the sensitivity analysis to observations.

- e) You used k_{coat} between 0.00 and 0.05. What is the step size? Moreover, I think the upper limit of k might be too low to represent a highly absorbing coating. I am wondering why you choose 0.05 instead of 0.1? Also, a range of n instead of 1.55 can better represent ambient particles. I recommend adding sensitivity analysis with a broader range of k and n .

In figure 3, the step size for κ_{coat} is 0.05. This sensitivity analysis is not intended to give robust estimations of $\Delta\text{MAC}_{\text{BC}}/\Delta\kappa_{\text{coat}}$, but to demonstrate that MAC is more sensitive to κ_{coat} than to n_{coat} . We chose to limit our study to weakly absorbing coatings because highly absorbing coatings will cause accumulation of phase shift as light passes through the coating. It is not clear whether the scaling laws provide in this work will be accurate for highly absorbing coatings, but due to computation limits we have left this for future work.

- 4) For equations 3 a and b, how did you come out with this function? Is there any physical meaning? Moreover, does equation 3 only work with $k = 0, 0.01, \text{ and } 0.05$?

Equations 3a and 3b come from damped power law distributions of MAC_{BC} as a function of R_{BC} and ρ_{BC} . Other than $\rho_{\text{BC}} = 1$ representing the crossover from Rayleigh to geometric optics, equation 3 is an empirical fit based on previous work by Chakrabarty and Heinson 2018. Equation 3 is accurate for κ_{coat} between 0.00 and 0.05, but has not been tested for κ_{coat} outside of this range.

Specific comments

1. L83-85, "The mass of the BC ... (Bond and Bergstrom, 2006)." For the density you chose, I suggest using a range instead of single values so that you can represent the wide range of ambient particle density and uncertainties in the literature. Thus, I am also curious to see the dependence or uncertainties related to the density.

Uncertainty related to the density can be calculated by multiplying the calculated MAC_{BC} by the ratio of 1.8 g/cm^3 to the density in question. We have done this calculation and added a supplementary figure showing MAC_{BC} with densities between 1.6 and 2.0 g/cm^3 . We have left the main figure text showing BC with density of 1.8 g/cm^3 , as this is a commonly used value.

2. Figure 2 is very blurry. Please make sure the final version has a higher resolution.

This will be corrected upon revision.

3. Figure 3, What are these error bars represent?

They represent on standard deviation. This has been added to the caption.

4. In the manuscript, you used ρ to represent the phase shift parameter. However, ρ is usually used for density. I suggest using a different Greek letter.

We chose to stay with notation which has been used in previous publications (Sorenson 2011, <https://doi.org/10.1080/02786820117868>). While we understand that ρ is also used for density, we are hopeful that section 2.3 clarifies the notation.

5. L132-133, “The BC refractive ... ,2006).” Could you clarify whether you use this RI value for all wavelengths or just 550 nm ?

This has been clarified in the revised text.

6. Section number for sub-sections in Sect. 3 need to be corrected.

This has been corrected.

7. In figure 4, I can see the authors want to keep the same scale for all sub-figures, but there are too many white spaces in b, d, and f, making it very difficult to see the trend in the $\rho < 1$ regime. Please consider changing the y-axis limit. Moreover, in b and f, why the number of RBC dots is different from other RBC?

There were some issues in the orientation averaging of the optical properties of these particles, so they were excluded from figure 4. We have filled in data points where possible.

8. L232-234, “It has been ... fixed at 0.00.” Could it be that the coating is absorbing, but ρ is greater than 1?

This is possible, but unlikely given that these studies all use wavelengths $> 532 \text{ nm}$, where there would be very weak absorption by organic coatings.

9. Figure 7 b should be a table, not a figure.

We have moved figure 7b to a table.

10. L250, should not Figure 3(b) be Figure 7(b)?

Yes, this has been corrected.