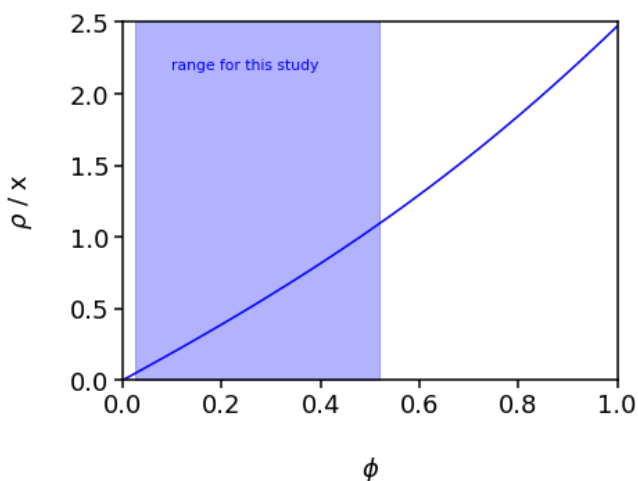


- 1) Could the authors add a more quantitative discussion of ϕ ? As stated, the entire discussion of the BC-core ρ comes down to ϕ , which can be constrained as about 0.1 to 0.4 (Zangmeister et al., 2018, <http://www.pnas.org/cgi/doi/10.1073/pnas.1403768111>; also Schnitzler et al. 2017 is relevant <http://dx.doi.org/10.1016/j.jaerosci.2017.01.005>). If there was some reason the authors did not discuss ϕ directly, could they please comment? If not,
- a) can the authors calculate ϕ for their model aggregates, and discuss whether the upper-limit packing density identified by Zangmeister et al. 2018 allows the literature trends to be fully explained by ϕ ?

The ϕ of our model aggregates ranged from 0.026 to 0.52. Full explanation of some literature trends requires packing densities that are larger than the upper limit observed by Zangmeister et al. in their 2017 paper. While it is not impossible for monomers to arrange in a way that achieves these higher packing fractions (face-centered cubic arrangement with $\phi = 0.74$, for example). However, based on recent findings we think it is unlikely that BC monomer would compact to this degree. Therefore, we think that it is extremely likely that other mechanisms are also playing a role in the decreased MAC observed by some studies, most notably heterogeneity in R_{BC} . The intention of this work and subsequent discussion is to demonstrate that BC morphology affects light absorption properties, and to provide a tool which eliminates reliance on models which make assumption about BC morphology. We will add further discussion on the packing fraction of our modeled aggregates and the limits of ambient BC packing fraction upon editing.

- b) Also, can the authors provide more information about the ϕ of their modelled particles, for example by plotting ϕ versus ρ or MAC?

Since the phase shift parameter is dependent on the size parameter and ϕ , we will include the following supplemental figure, which shows the size parameter normalized phase shift parameter as a function of ϕ . This plot also shows the range of ϕ which was included in this study.



- 2) The authors state that “Our results indicate that studies which find little to no increase in MACBC with increased RBC are measuring BC aggregates which have undergone

significant coating-induced restructuring, while studies that find significant increases in MAC_{BC} are measuring aggregates which have undergone little to no restructuring.” How confident are the authors that alternatives have been excluded, and that this statement is the most likely given all available evidence? For example:

- a) Based on a recent review of soot-restructuring studies ([Corbin, Modini, and Gysel-Beer, arXiv.2206.03646](#)) I believe this statement should be reconsidered or discussed in terms of the fundamental physics it implies. To briefly summarize that review, we identified multiple studies that demonstrated unequivocally that liquid condensation typically induces restructuring. These studies used various materials including organics of varying polarity and sulfuric acid. Our review of these studies and complementary laboratory demonstration, showed that condensation-compaction coatings can only be avoided when solid coatings or liquids with very high contact angles (which may result in heterogeneous nanodroplet activation and therefore avoid compacting surface tension forces) were used. Examples of such solids include SOA formed at low RH or anthracene (relevant only to the laboratory). Compaction can also be avoided by coagulation. So, if internally mixed BC has not undergone extensive restructuring, it must have mixed with solids (including highly viscous glasses) or by coagulation.

My impression from the recent studies by Fierce et al. (cited by the authors) is that while night-time coagulation can be significant, it is unlikely that most soot particles mix by coagulation. But, perhaps the authors’ work implies that my impression was inaccurate. My impression is also that solid organic coatings form only rarely, since they require very low RH or low temperatures, while organic vapours are emitted most often at higher temperatures. It seems to me less likely that solid coatings explain the field data on absorption enhancement.

So, the authors’ conclusions can be reconciled with the known mechanisms of soot restructuring by arguing that some studies primarily observe liquid-condensation coatings while other primarily observe solid-like coatings or coagulation coatings.

We agree with all points given above, and that our previous statement needs to be altered. The original intention of our statement was not to imply that some studies are measuring soot which has not restructured, but that some studies are measuring soot which has achieved $\rho_{BC} > 1$ and others are measuring soot with $\rho_{BC} < 1$. We agree with the conclusions of the above comment that it is unlikely for solid coatings to form in ambient conditions, and therefore unlikely that BC morphology would remain unaltered during coating uptake. We believe that studies which find significant absorption enhancement could be measuring particles which have undergone restructuring, but ρ_{BC} has not increased past unity. This could happen when the product of size parameter and packing fraction is small. For example, small soot particles which undergo significant restructuring (large ϕ) may still have $\rho_{BC} < 1$, and based on our results would show significant absorption enhancement with increasing coating amounts. We will rephrase our previous statement as follows:

“Our results indicate that studies which find little to no increase in MAC_{BC} with increased R_{BC} may be measuring BC aggregates which have undergone significant

coating-induced restructuring, leading to $\rho_{BC} > 1$. On the other hand, studies that find significant increases in MAC_{BC} may be measuring aggregates which have $\rho_{BC} < 1$, and may also be measuring particles which have significant heterogeneity in R_{BC} . This does not imply that these studies are measuring BC which has not restructured, only that the product of the size parameter and core packing fraction of BC is not large enough such that $\rho_{BC} < 1$.”

- b) On Line 71, the authors state, “Recent studies have found that the non-sphericity of BC containing particles (partial encapsulation of BC) can decrease absorption enhancement (Hu et al., 2022, 2021). While these findings are notable, previous studies have not observed a prevalence of partially-encapsulated BC, yet decreased light absorption is still observed”. Is it possible that the authors have too readily rejected the hypothesis of H1) partially encapsulated, or off-center mixing states, in favour of H2) condensation-without-compaction? Given the abovementioned review, I believe H1 is plausible while H2 is extremely unlikely. I would consider the entire manuscript to remain valid and valuable if H1 is rejected over H2. The only change is that ϕ becomes ϕ_{eff} . (Would the same trends in MAC be observed?)

We do not consider here the effects of off-center mixing states, as it is outside the scope of this work. We agree that we should not reject H1 altogether, but the effects of off-center coatings will be more prevalent at small R_{BC} , and we do not believe that H1 can fully explain observations of low E_{abs} at large R_{BC} . Since our definition of ϕ only includes the BC monomers, the ϕ of modeled aggregates will be unaffected by coating location for modeled aggregates.

3. This is more of an editorial comment. The highest ρ_{BC} in Figure 7b were measured at the shortest wavelengths and the two highest studies were both first-authored by Cappa. Some readers may wonder whether there was a systematic effect here (for wavelength, it is expected by definition; and for the Cappa group, the question is whether they use a unique experimental approach that caused a bias relative to other data sets). I do not personally believe that these are real issues, but they deserve may a brief comment for the reader’s benefit.

We believe that it is possible that the estimated ρ_{BC} for the Cappa studies may be slightly higher than the true ρ_{BC} of the measured particles. This is because the Cappa studies both made use of thermodenuders. Thermodenuders may not completely remove low-volatility organics from BC, causing overestimation of absorption by BC, and subsequently lower E_{abs} (Shetty et al., 2021 <https://doi.org/10.1080/02786826.2021.1873909>). It is unclear whether low-volatility organics in fact did bias the absorption measurements in these studies, but is a possible explanation for the elevated ρ_{BC} .

While reading, I also made various minor notes. I will list them below as suggestions for the authors.

- a) I'd add a row showing partially encapsulated/collapsed examples in Figure 2.
[We will move the partially collapsed BC example shown in figure S1 to the main text.](#)
- b) Line 105-109 may need clarifying. Why would someone use the RDG MAC when estimating Eabs? To me, a "literature value" would be a measured MAC of mature, open-structured BC (Liu et al., linked below). Text may not convey your intention here.
[This section will be altered to reflect the intention that consideration of fractal morphology can affect the MAC of pure BC.](#)
- c) What is the role of ϕ in Figure 3? No effect?
[The change in MAC for changing coating refractive index was calculated with constant \$\phi\$, so there is no effect. This will be clarified in the text.](#)
- d) Line 161, there may be a better citation for the imaginary refractive index (Sun and Bond?).
[This study will be cited upon revision.](#)
- e) Line 163, add SOA after pinene.
[This will be incorporated upon revision.](#)
- f) Line 164, consider citing Lu et al <http://dx.doi.org/10.1021/acs.est.5b00211>
[This will be incorporated upon revision.](#)
- g) Line 180, are the four digits of precision meaningful in 6.819 m²/g? What is the corresponding standard deviation? Also, it may be worth discussing this value in comparison to the measured mean value of 8.0 ± 0.7 m²/g (Liu et al. <https://doi.org/10.1080/02786826.2019.1676878>)
[The number of significant digits in the calculated MAC₀ is limited by the density of BC, therefore it will be changed to 6.8 m²/g upon revision. We will also discuss this finding in comparison with Liu et al.](#)