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Comment on egusphere-2022-573

Anonymous Referee #2

Referee comment on "Importance of size representation and morphology in modelling optical properties of black carbon: comparison between laboratory measurements and model simulations" by Baseerat Romshoo et al., EGUsphere, https://doi.org/10.5194/egusphere-2022-573-RC2, 2022

Review of "Importance of size representation and morphology in modelling optical properties of black carbon: comparison between laboratory measurements and model simulations" by Romshoo et al.

In this paper, the authors describe various models of the optical properties of black carbon particles and compare them with laboratory measured values for soot generated with a mini-CAST. The work done is quite extensive and as such, it is important to disseminate it to the community. However, I think the paper could significantly be improved in terms of clarity and readability and would benefit from providing a clearer and more succinct overall message.

General comment

- This study compares models with laboratory measurements only. However, black particles in the atmosphere have been shown to be much more complex than those generated in the laboratory, for example, those emitted during biomass burning or those transported very far from the source. It is reasonable and already impressive to focus only on laboratory-generated particles. Still, the conclusions might need to be put more in view of this limitation/caveat to avoid the impression that the implications of the findings on describing the actual properties of atmospheric aerosols might be overinflated.
- The authors switch frequently (but mostly from the first part of the paper to the second) between the term "soot" and the term "black carbon". While this is understandable, this can cause confusion in view especially of a few papers discussing this terminology issue – an issue that can be problematic and is partially still unresolved (e.g., Buseck, P. R.; Adachi, K.; Gelencsér, A.; Tompa, É.; Pósfai, M., ns-Soot: A Material-Based Term for Strongly Light-Absorbing Carbonaceous Particles. Aerosol Science and Technology 2014, 48, (7), 777-788. And Petzold, A.; Ogren, J. A.;

Fiebig, M.; Laj, P.; Li, S. M.; Baltensperger, U.; Holzer-Popp, T.; Kinne, S.; Pappalardo, G.; Sugimoto, N.; Wehrli, C.; Wiedensohler, A.; Zhang, X. Y., Recommendations for reporting "black carbon" measurements. Atmospheric Chemistry and Physics 2013, 13, (16), 8365-8379.)

- A bit related to the first point, how representative is the Mini-CAST black carbon of ambient soot? Even for bare black carbon?
- The references used sometimes are a bit limited and maybe biased.
- Could removing the volatile organic compounds with the thermos-stripper change the morphology of the black carbon particles? For example, would the surface tension acting on the monomers while the coating is evaporating, or the heating cause partial collapse of the aggregate? The authors should comment on this potential issue.
- The conclusions seem to underline the need to account for polydispersity and accurate coating representation to improve the calculations of the optical properties of laboratory-generated black carbon, but the authors seem to gloss over some of the results previously discussed in the paper, for example, for the MAC of smaller particles where some of the spherical simulations seem better. So, I was left with a few mixed feelings about what is really the best representation.
- Maybe I missed it, but how was the SSA calculated? Using the CAPS or using the aethalometer + nephelometer combination, or the MAAP + nephelometer combination? How would the three calculations compare (after correcting for wavelength discrepancies?
- In general, I found the paper a bit difficult and dry to read; most of the second paper is a list-like tedious description of the results in the figures with little commentary or explanation (potential or not) of the findings. While I don't have a lot of detailed suggestions on how to improve this issue, I think the authors could try to make the reading more fluent focusing more on commenting and interpreting the figures than on describing them. It would also help to use more consistent figures formatting and style and more readable (larger size and thickness) markers for example, although this last point, I understand, could be mostly a personal preference.

Specific comments

Lines 58 – 61: black carbon aggregate compaction has also been associated with cloud (water and ice) processing alone (no organic coating).

Line 77: Many others published on this issue, for example, the following paper might be relevant here: Scarnato, B. V.; Vahidinia, S.; Richard, D. T.; Kirchstetter, T. W., Effects of internal mixing and aggregate morphology on optical properties of black carbon using a discrete dipole approximation model. Atmospheric Chemistry and Physics 2013, 13, (10), 5089-5101.

Line 135: remove "the" in front of "both"

Line 92: Many other papers before the one cited here discussed the nature of BC aggregates and monomers.

Table 1: specify the wavelength of the SSA data in the table or caption itself.

Line 182: Many studies report the mixing configurations and morphologies of black carbon aggregates in the laboratory or ambient samples using scanning electron microscopy or transmission electron microscopy. How do the properties of the particles generated from the mini-CAST compare with those previous results?

Line 184: "kept in mind" is a bit vague.

Figure 1: the mixed models all assume that the coating/mixing material did not affect the morphology of the bare aggregate underneath, correct? How good is that assumption? It is known that coating can cause very significant compaction of the initially bare black carbon particle.

Line 193 "Leaving some residuals" how much is "some" can the authors be more quantitative?

Line 225: How did the authors deal with issues induced by non-sphericity and multiple charges in the SMPS measurements?

Line 231: this approach assumes that the coating thickness is the same over each particle in the particle ensemble, correct? However, it has been shown that the amount of coating can be quite variable from particle to particle within an ensemble of ambient particles. Particle-resolved models suggest that this coating distribution can also induce significant deviations in simulated optical properties.

Line 248: It seems like this "first method" uses mass information, but in line 247, the authors mention that they did not use mass information (?).

Line 255: why 14 nm? And how good is this assumption? Does it matter?

Lines 263 – 265: These two sentences are not clear to me.

Lin 433: Maybe "in contrast" or "on the contrary" instead of "in contrary"?

Section 3.1.1 I am not sure I fully understand the rationale for calculating Npp from dp,V vs. dp,N, all the methods listed in appendix B seem to be using dp,N

Line 449: missing “to” in front of “the experimentally…”

Line 453: There are also numerical studies that show the effect of measured compaction on scattering.

Figures 5 and 6: I think it would be nice to have these figures be the same for both methods, meaning showing SSA and AAE in both, or maybe even better, showing SSA, AAE, and sigma abs in both.

Line 544: “the” in front of “nature”? In general, this sentence is a bit unclear to me.

Line 447: “the modelled results could not be validated with the modelled findings” maybe the authors mean “the modelled results could not be validated with measurements”?

Line 660: similarly, “modelled” or “measured”?

Line 689: “which pronounces” reads a bit awkward in the sense that the sentence is not very clear on what “which” refers to.

Appendix A: the authors mentioned the use of an Aurora4000 nephelometer, if I am not mistaken, that instrument is a polar nephelometer that should allow estimating the asymmetry parameter g using the backscatter fraction (e.g., Moosmüller, H.; Ogren, J. A., Parameterization of the Aerosol Upscatter Fraction as Function of the Backscatter Fraction and Their Relationships to the Asymmetry Parameter for Radiative Transfer Calculations. Atmosphere 2017, 8, (8), 133.). Therefore, I am unclear why the authors did not attempt comparing g measured with g simulated in figure 11.