

Review of 'Light Absorption by Brown Carbon over the South-East Atlantic Ocean', by L. Zhang, *et al.*, submitted to *Atmospheric Chemistry and Physics*.

Summary and general comments

The authors report spectroscopy measurements of aerosol optical properties from a research aircraft platform for biomass burning aerosols over the South East Atlantic Ocean. A modelling framework is developed for attributing the contribution of light absorbing organics (brown carbon, BrC) to the overall aerosol light absorption. This framework exploits either Core-Shell or 'grey sphere' Mie models, with the latter utilising one of three different effective medium approximations to treat the refractive index. The manuscript is interesting, uses established measurement methods, and adds to the debate surrounding the suitability of Mie models for treating aerosol optical properties. However, the authors assert conclusions that are not reinforced by their analysis, not least because there is scant treatment of systematic uncertainties in their measurements nor appreciation of the limitations imposed by their modelling assumptions. Considerable moderation of some of the claims of key outcomes is needed. Specific Comments are provided below in addition to Technical Corrections that need to be addressed.

Specific comments (referring to the page "P" and line "L" numbers)

P1 L32. ' $0.25 \pm 0.34 \text{ m}^2 \text{ g}^{-1}$ ': The error is larger than the mean value. The MAC cannot physically be negative, so this uncertainty is nonsensical. Either this uncertainty needs reconsidering, or some explanation of the significance of a negative MAC is needed.

P5 L11-12. '...except for the Neph, whose humidity is not controlled.' The lack of control of the humidity inside the nephelometer is a big issue in terms of the potential for large systematic uncertainties in measured optical properties that then impact on the later retrievals of m_{eBC} and attribution of absorption to brown carbon. There is no discussion in manuscript of humidity impacts on scattering coefficients and subsequent optical closure calculations. Indeed, no details on the RH in the Neph are provided for the different flights. The Supporting Information just states that the RH is less than 40% (apart from RF05_1 and RF05_2). Samples with an RH of 40% could have considerable mass concentrations of water. This would introduce bias in optical closure calculations, but this bias is not assessed. This limitation of the study should be spelled out clearly to the reader.

P5 L15. I would add the text ", including descriptions and validation data for the conversion of UHSAS and APS data to particle number size distributions". On first reading, I was left thinking that a description was missing of how the APS data were processed to give a geometric or mobility diameter (rather than the raw APS-measured aerodynamic diameter) as well as that for how the optical size was corrected for refractive index. It was only on second reading and double-checking the SI that I realized these descriptions are given.

P5 L23-24. This sentence ('particles were separated into BC-containing particles and BC-free particles') leads to confusion because, at the end of the previous paragraph, you state that you do not investigate external mixing. Please could the authors bring some clarity to these seemingly contradictory statements.

P6 L5-7. $m_{\text{BC-free}}$ is calculated using a volume fraction weighting mixing rule. What was the reasoning for constraining the choice of mixing rule for $m_{\text{BC-free}}$ to the volume fraction mixing? Why not try MG and BG mixing rules also?

P9 L5-6. 'if other absorbing components are present, the imaginary part (k_{eBC}) of m_{eBC} would be greater...'. Not necessarily - if those additional components are absorbing, they could have a lower absorption than BC, and therefore lower k_{eBC} .

P9 L26-27. 'magnetite, whose refractive index is generally invariant between wavelengths 470 and 660 nm.' Reference is needed.

P10 L16-17. ‘The MR_{100} is the MR for particles with 100 nm BC core only, i.e. D_c equals to 100 nm.’ It is not clear why you calculated MR_{100} . Can you provide the reader with your motivations for calculating this quantity? After twice reading this manuscript, I do not understand the utility of this parameter.

P10 L18-19. ‘To justify its feasibility, we derived m_{eBC} for other flights using Eq. 9 and found the differences are less than 5% for both the real and imaginary parts’. This is very surprising as I would expect absorption coefficients to provide a very poor constraint on the real refractive index. Please could the authors provide more evidence, perhaps in the Supplement, to convince the reader that this discrepancy is <5%.

P10 L20. ‘no m_{eBC} value has been achieved in the retrieval.’ What does this statement mean? Do the authors mean to say that no minimum location in the merit function defined by Eqn. 9 could be located within the prescribed search bounds of n_{eBC} and k_{eBC} ?

P12 L6. ‘otherwise, higher k_{eBC} values indicate larger contributions of absorbers other than BC at 660 nm.’ This is not correct. First, other absorbing species may have a lower absorption strength than BC. Second, the choice of optical closure model affects the accuracy of the derived k and it is not clear that these grey sphere models represent the particles in question. Third, no assessment of measurement biases is provided here, which would impact on the derived k_{eBC} ; at the forefront of my mind are the known (up to ~50%) biases in absorption from filter-based absorption techniques, the impact of humidity on the scattering measurements, and the accuracy of the particle size distributions using the authors’ chosen techniques and subsequent impacts on retrievals of m .

P13 L3-4. ‘no value was attained with the CS model.’ Why was no value attained? It is not clear why this is the case.

P13 L11. ‘however, these values are still smaller than those for RF05_3.’ This statement is misleading and is not comparing like-for-like. The value from Taylor et al. (2020) is a campaign average value rather than a single leg of a single flight (as is quantified by the value for RF05_3). Moreover, if the reported uncertainties for RF05_3 and Taylor *et al.* values are considered, then the E_{Abs} match to within statistical uncertainty.

P13 L14. ‘ MR_{100} was equal to 5.3’. I do not follow the utility of this parameter.

P13 L16. ‘implying that the remaining 41% of absorption was contributed by other absorbers’. This statement is not suitably justified by the discussion, because no assessment of the impact of measurement biases is provided (which I suspect are very large indeed), while the optical properties may not be described well for these particles by a grey sphere or core-shell Mie model. More justification/discussion of these confounding factors is needed, along with moderation of language used to reflect that the role of other absorbers is a hypothesis.

P13 L16-17. ‘This huge absorption from other absorbers...’. This is strong language; now the authors are asserting with certainty that the model-measurement discrepancy is real and is attributed to other absorbers. The authors must moderate the language here to ensure that it is clear that the role of other absorbers is only a hypothesis.

P14 L8. The error in the quoted $MAC_{OA,470}$ is larger than the mean value, allowing for a negative MAC that is nonsensical. In addition to rectifying this issue, the authors should state what this uncertainty represents. Is it the precision in the retrieved $MAC_{OA,470}$? Or perhaps the standard deviation across the different model treatments?

P14 L9. ‘slightly lower...’. I do not agree that this not lower. They are identical within the stated uncertainties.

P14 L10-11. ‘suggesting BrC bleaching during transport considering our result is at a much longer wavelength’. Following my last comment above, this statement cannot be inferred from the reported MAC values, with the values reported in this paper highly uncertain. Indeed, the quoted uncertainty values are large even though the contributions from some error sources are ignored, such as model assumptions (i.e., grey sphere, invariant m_{eBC} with wavelength) and measurement biases. This statement must be removed.

P14 L13. Uncertainty larger than value, giving nonsensical negative values for MAC_{OA} .

P14 L23-24. 'however, to the best of our knowledge, such high MAC for secondary BrC have not been documented'. This statement must be removed. It is at odds with the last sentence of the last paragraph, where you show that the MAC_{OA} from your measurements are, within statistical uncertainty, the same as those reported by Taylor et al. (2020).

P17 L3. 'within the range expected for BC.' Please state the expected range and provide references.

P17 L11-12. 'The extreme underestimation from the AAE attribution method is mainly due to the fact that the AAE and absorption coefficients used in this method are not derived from BC alone...' This statement is not justifiable. It is not clear that the CS or homogeneous "grey sphere" models used by the authors are superior in their approach compared to the AAE-attribution model. For example, the "grey sphere" Mie models assume sphericity and homogeneous mixing, ignoring lensing effects as well as internal multiple scattering interactions. If I look at the Taylor (2020) paper that the authors refer to heavily, Taylor et al. show that the grey sphere models (using volume mixing, Bruggeman, and MG models) all overestimate the absorption. Instead, Taylor et al. show that two different semi-empirical models for MAC (parameterisations fitted to comprehensive numerical simulations of optical properties for coated aggregated using either T-Matrix or discrete dipole approximation models) perform very well. It is important that the authors recognise the limitations of their framework and they need to moderate their language; it is not supported by the current paper that the AAE method is inferior in predicting the true BrC contribution to absorption.

P18 L15: Error in MAC is bigger than mean value, which is nonsensical.

P18 L26. Authors assert that brown carbon attribution from AAE approach is underestimated, i.e. implying that the AAE attribution approach is inferior. This is not justified by current manuscript, which ignores major limitations and uncertainty estimates in the optical closure approach.

Technical corrections

P4 L7. The authors use the acronym 'FRP'. This acronym is not used again, so please remove.

P6 Eqn 6. The "2" in the chemical symbol for potassium sulfate needs to be subscripted.

P6 L10. The minus sign in "1.8 g cm⁻³" needs to be superscripted.

P8 Figure 3. The tick marks on the vertical axes for the adjacent absorption cross section plots (i.e., the columns corresponding to $m_{BC}=1.95+0.79i$, $2.26+1.26i$) do not match up. Also, what are the diamond symbols on these plots?

P8 L12. "note" should be "noted".

P9 L14. 'range' should be 'ranges'.

P11 Figure 4. The positioning of tick marks on the lower horizontal scale confusing. Perhaps simply mark every interval of 0.2, because I think it is clear from the legend what the upper limit of the grey shaded region is.

P11 L4-7. This long sentence is difficult to read and does not make grammatical sense in places. Please revise.

P14 L7. The statement "and other flights from homogeneous models" is nonsensical and needs revising.

P16 L20. The authors state "This AAE" but do not provide the value. Please state the value for the AAE of BrC referred to in this sentence.

P18 L29. Please remove "in the meanwhile".