

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2021-1000

Anonymous Referee #1

Referee comment on "Light absorption by brown carbon over the South-East Atlantic Ocean" by Lu Zhang et al., Atmos. Chem. Phys. Discuss.,
<https://doi.org/10.5194/acp-2021-1000-RC2>, 2022

This study used comprehensive aircraft measurements to investigate the aged biomass burning plumes from south African transported to the Atlantic Ocean. The authors used optical closure between iterated refractive indices measured scattering-absorption at $\lambda=660\text{nm}$, to derive a proposed " effective refractive index of BC " in order to account for all BC absorption at multiple wavelengths. This study had a valuable dataset and could potentially contribute to the understanding on the aerosol absorption at this climatically important region. However I have a few concerns about the methods of this study, before it can be considered for publication.

Major:

1) My main concern is about the nil absorption of brown carbon at 660nm. There are many studies stating OA could be absorbing at relatively long visible wavelength, particularly for less-volatile organics from biomass burning (Saleh et al. ES&T letters, 2018). These OA has a large molecular weight and more functionalized (hereby lower volatility) and may survive after transport, when more volatile species were evaporated with remaining more absorbing component to be transported to a longer distance, as evidenced by a recent field study (Liu et al., ACP 2021).

So it is very possible that the long visible absorption (e.g. 660nm) in your results contained some BrC. I would suggest it may not be necessary to really assume a nil absorption of BrC but using a combined positive k value for OA to feed the optical closure. See a study to derive the k_{OA} by assuming some absorptivity for both externally and internally mixed OA in BC-containing particles (Liu et al., 2021 ES&T). This may lead to a more solid conclusion.

2) Figure 4 is crucial and needs large improvement. Are these from all the straight-level

runs from all flights? Error bars for each dot are required. It said only 6 dots had potential additional absorbing component, if so, why there were some "magnetite" but some without, these need to be discussed.

There are still no solid evidences that the magnetite did exist and contributed to the absorption at long visible. I may suggest having a plot like "additional absorption besides BC" vs magnetite fraction, the former is from Fig. 4 and the latter is from Fig. 5. Additionally, the origin of magnetite is speculated but not evidenced, where did they come from? This needs exterior support.

3) The optical closure of the partial dataset is performed on absorption-only and others are absorption-scattering. My concern is this may induce some discrepancies, especially when presenting both datasets on the same table. Why not using consistent absorption-only approach, at least they are consistent.

Others:

There should be more details discussing about the determination of bulk mass ratio of coating over rBC core from the SP2 measurement.

The title itself only includes brown carbon, though the authors also largely discussed about the possible absorption by iron.

It would be useful to briefly mention what effective refractive index of BC is in the abstract.

Conclusions should include some discussions about evolution.

Page 2, line 3, what is the commonly used AAE? To what extent lower?

The last sentence in the abstract needs rewriting, it is too long and a bit confusing.

Page 8, line 9, no need italic font.

Page 10, line 5, MAC is mass absorption cross section, not coefficient.

Equation (9) why using sigma, but only one wavelength is used.

References

Liu, Q., et al.: Reduced volatility of aerosols from surface emissions to the top of the planetary boundary layer, *Atmos. Chem. Phys.*, 21, 14749–14760, <https://doi.org/10.5194/acp-21-14749-2021>, 2021.

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Saleh, R., Z. Cheng, and K. Atwi (2018), The Brown–Black Continuum of Light-Absorbing Combustion Aerosols, *Environmental Science & Technology Letters*, doi: 10.1021/acs.estlett.8b00305.