

Interactive comment on “Vertical variability of the properties of highly aged biomass burning aerosol transported over the southeast Atlantic during CLARIFY-2017” by Huihui Wu et al.

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Title: Vertical variability of the properties of highly aged biomass burning aerosol transported over the southeast Atlantic during CLARIFY-2017

General Comments The authors present vertical information on long-range transport biomass burning (BB) aerosol from Southeastern Africa as sampled from aircraft measurements during CLARIFY-2017. The paper is focused on aerosol optical properties, chemical composition, size distributions, and emission factors. The work is thorough

C1

and of great interest in understanding how BB aerosol ages post emission within the boundary layer and free troposphere, especially the direct vertical profile measurements of single scattering albedo and chemical composition. This area of the globe is lacking detailed measurements such as the ones the authors present here, which are the first of their kind and done in a well-organized format. The information on the vertical structure will be very informative for understanding BB aging and aerosol-cloud interactions for this remote region of the globe to reduce the climate model uncertainties that are very high in this region. I have some comments and suggestions that are focused on improving the impact as well as some technical comments that should be addressed before final publication within the ACP Special Issue: New observations and related modelling studies of the aerosol-cloud-climate system in the Southeast Atlantic and southern Africa regions (ACP/AMT inter-journal SI). Overall, I support this work for publication in ACP.

Detailed Comments

1 – Page 1, Line 27 – Reporting the lowest SSA values in the FT and the location at 2 km is very noteworthy since as the authors mention, this means the BB aerosol in the region is more absorbing than what is currently used in climate models. Would it be possible to report an average column-weighted SSA as well? This would be useful for comparison with satellite data, other passive ground sampling and modeling efforts. If so, this could also be reported for the BL and FT separately as well in the following sentences. If not, then the ranges should be included for the BL and FT.

2 – Page 1, Line 30 – Recommend stating whether the SSA is highest in the BL or high in the FT since the earlier sentences say the SSA increases in the FT with altitude so it is not clear in the abstract where the highest SSA was observed.

3 – Page 2, Line 54 - What about the other effects on clouds besides CCN formation? Later on, two paragraphs following, the same paper is referenced having calculated direct, indirect and semi-direct effects. It seems disjointed to not mention these effects

C2

earlier.

4 – Page 2, Lines 54-55 – Another more recent publication also found significant enhanced convection over the Amazon region due to particles < 50 nm diameter containing BC - Fan et al., Science, 2018 - DOI: 10.1126/science.aan8461).

5 – Page 2, Line 59 – Suggest changing from “will be important.” to “is and will be increasingly important in the future.”

6 – Page 3, Line 75 – 77 - What about also the uncertainty in retrievals especially for vertically resolved information? Suggest including this in the manuscript text.

7 – Page 3, Lines 80-82 - This information is incomplete as written and should be modified. The paper referenced from the LASIC campaign reports the first results of BB aerosol measured during the campaign and focuses on the in situ. The full LASIC campaign included ground-based in situ aerosol measurements as well as column measurements on aerosols and clouds. Retrievals are being processed for column-weighted averages as well as to retrieve vertically-resolved information. While the vertically-resolved retrievals require assumptions and have limitations, there is more data to be analyzed that has yet to be reported in the peer-reviewed literature from LASIC.

8 - Page 3, Line 89 - Check that all references in the text are listed in the reference section. For example, the Zuidema et al. 2016 reference mentioned here does not appear to be listed in the References Section. - <https://journals.ametsoc.org/doi/pdf/10.1175/BAMS-D-15-00082.1>

9 - Page 4, Line 129 - Check and improve all reference formats as these 2 Trembath references appear incomplete in the Reference section. It is not clear how someone would find a tech report with no indication of who published it and/or without an associated doi. Also, the abbreviations should be spelled out, for example, SAES is not defined.

C3

10 – Page 5, Line 134 - 135 – Did the composition dependent CE that was determined based on Middlebrook et al. 2012 for the AMS data here include a mass comparison to the SMPS to validate the CE used?

11 – Page 5, Line 137 – Suggest including references to the early f44 and f60 work by Cubison et al., ACP, 2011 (already in your references), Ng et al., ACP, 2010 (<https://doi.org/10.5194/acp-10-4625-2010>), and Ortega et al., 2013 (doi:10.5194/acp-13-11551-2013) here where these factors are first mentioned in the text.

12 – Page 6, Lines 193 – 194 – How many flights were used out of how many total to make the temperature and specific humidity profiles? This information should be added to the main text and listed in the SI Figure caption.

13 – Page 7, Lines 199 – 200 – Can you state in the text how thick the typical inversion layer is between the BL and FT?

14 – Page 7, Line 225 – The higher SO₄ and lower BC are more striking than the 10% change in OA fraction. State here the % changes in these species for the BB-polluted BL Period 1 vs 3 comparison.

15 – Page 8, Line 227 - With as much as the mass fractions are mentioned in the text, a bar or pie chart with the % mass fractions indicated would be a good visual representation of this comparison that could be added to Figure 3.

16 – Page 8, Lines 226 – 227 – State the sulfate fractions in the BL and FT for completeness and a quick comparison in the text so the reader doesn't have to go to the Figures/Tables to get this information.

17 – Page 8, Lines 247 – 249 - This is an interesting topic that deserves more attention. A recent publication reviewed lab and field BBA aging of OA and should be included in this discussion is presented by Hodshire et al., ES&T, 2019 (<https://doi.org/10.1021/acs.est.9b02588>). How common is the observation of secondary inorganic aerosol formation due to aging in ambient and/or laboratory data?

C4

Is this unique to BBA?

18 – Page 9, Line 273 – Consider adding Ortega et al., ACP, 2013 (doi:10.5194/acp-13-11551-2013) here in addition to Cubison et al., ACP, 2011.

19 – Page 9, Line 275 – Is the ~5 days quoted the timeline of the less aged or more aged BBA? It is not clear in the way the sentence is currently written. Also, how does this relate to the earlier statement that mentions f60 is only a good tracer for BBA with aging timescales < 1 day?

20 – Page 9, Line 286 – An approximation of the average carbon oxidation state can also be presented using O/C and H/C and Equation 2 in Kroll et al., Nature Chemistry, 2011 - <https://www.nature.com/articles/nchem.948>

21 – Page 9, Line 291 – The aging and oxidation of organic aerosol in the atmosphere towards the formation of LV-OOA and high f44 should also include reference to the work done by Jimenez et al., 2010 - <https://science.sciencemag.org/lanl.idm.oclc.org/content/326/5959/1525>.

22 – Page 10, Lines 312 – 314 – How is it known that all “particles observed in the FT have not encountered cloud” during CLARIFY? Please explain, elaborate or reference previous work to substantiate the claim.

23 – Page 10, Lines 320 – 323 – The assumption seems to be that the BBA in the BL is the result of mixing between BBA that was lofted and transported in the FT via downdrafts into the BL. Is there relevant work that can be cited to support these claims for this region?

24 – Page 11, Line 332 – Suggest changing “indicates that new particle formation was occurring. . .” to “indicates that new particle formation and growth was likely occurring”.

25 - Page 14, Line 420 – Why are marine emissions and cloud processing considered one factor?

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26 – Page 14, Lines 420 – 421 – Does “removal processes” include wet and dry deposition?

27 – Page 14, Line 421 – Same as earlier comment – suggestion to restate to include “new particle formation and growth”.

28 – Page 14, Line 433 – Consider adding Heald et al., GRL, 2010 when referencing carboxylic acid content in addition to Duplissy et al., 2011). - <https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2010GL042737>

29 – Page 15, Lines 475 – 477 – State why enhanced MAC's are being attributed to coatings on BC and brown carbon is not being considered as another potential explanation.

FIGURES

Page 28, Figure 1 – A key should be provided for the wind speed range shown including units.

Page 30, Figure 4 – I strongly encourage the authors to contact the authors of the previous work so that the real data can be shown in this figure instead of circled approximations.

REFERENCES

Suggest adding a space between each citation. Also check that all references are complete, include the journal name or publisher, and that acronyms are spelled out. Specific examples are referenced in the comments above.

Technical Corrections

Page 2, Line 60 – Change “cloud” to “clouds”.

Page 4, Line 101 – Forgot degrees symbol before “S”

Page 4, Line 102 – Remove “be” from “The smoke is then be advected. . .”.

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Page 4, Line 110 – Change “was” to “were”

Page 6, Line 192 – Change “SMPS and PACSP” to “SMPS and PCASP”

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-197>, 2020.