Review of: Vertical variability of the properties of highly aged biomass burning aerosol transported over the southeast Atlantic during CLARIFY-2017
Authors: Wu et al.
Manuscript #: ACP-2020-197

The authors have provided a very comprehensive and thoughtful revision of their original manuscript. I appreciate the additional information that has been added. I only have a few minor comments. I was not an original reviewer for this manuscript, and after writing my review and revisiting the original comments, I came to a better understanding on some of the revisions. To me a few of the author revisions in response to the original reviewer comments did not always improve the writing. In places where my recommendations contradict those of the previous reviewers, I leave it to the authors and editor to arrive at a final decision. The line numbers below apply to the revised manuscript ("acp-2020-197-manuscript-version3.pdf").

Recommendation: Accept with Minor Revisions

Abstract, line 17-18: the comment "for the first time" doesn't seem quite right, as some members of this same team I believe documented a BBA plume at Ascension as part of SAFARI, and, the ORACLES measurements in 2016 preceeded the CLARIFY campaign. Some of that vertical structure is documented in Shinozuka et al., 2020 and Redemann et al. 2020. You could just leave out the phrase without loss of context, or, substitute something else here.

Page 1, line 48: Adebiyi and Zuidema 2016 might be the better reference here, as it focuses so strongly on the FT winds you mention.

Adebiyi, A., and Zuidema, P.: The Role of the Southern African Easterly Jet in Modifying the Southeast Atlantic Aerosol and Cloud Environments. Quarterly Journal of the Royal Meteorological Society, 142, 697, 1574–89. https://doi.org/10.1002/qj.2765, 2016

Page 2 line 56: I thought the point of Abel et al 2020 is that certain forms of mesoscale organization (POCs) don't support entrainment into the MBL so much. I'm not sure what to suggest here, but the sentence as written is mildly confusing.

Page 2 line 60: is there any evidence for the mechanism documented Fan et al., 2018, of ultra-fine particles enhancing shallow convection, at Ascension? We typically think of ultra-fine particle production over the marine ocean occurring over more pristine conditions (is my impression). The Koch and Del Genio reference is fine, but that is an overview document that highlights many other processes that are likely not relevant at Ascension. I would encourage the authors to include references here to processes that are more likely at Ascension. One mechanism I don't see mentioned is cloud dissipation in response to higher temperature/lower humidity. This effect was initially highlighted in Ackerman et al. 2000 and documented at Ascension in Zhang and Zuidema, 2019. The effect of enhancing convection formation through the additional heating is also used to explain a mid-morning cloudiness maximum at Ascension in Zhang and Zuidema, 2019. The free-tropospheric semi-direct effect also seems worth mentioning here, for which the authors could cite CLARIFY's very own Herbert et al., 2020 and Gordon et al., 2018 modeling papers. Under indirect effects, there are several pertinent papers that could be cited, e.g., Constantino and Breon, 2013; Kacarab et al., 2020; Gordon et al., 2020. Further ahead I do see you discussing a few of these studies further, e.g., Gordon et al., 2018, so that is good, but nevertheless this section here would be improved by tying it in better to that discussion on p. 2.

Ackerman, A. S., Toon, O. B., Stevens, D. E., Heymsfield, A. J., Ramanathan, V., and Welton, E. J.: Reduction of tropical cloudiness by soot, Science, 288, 1042-1047, 2000.

Costantino, L. and Bréon, F.-M.: Aerosol indirect effect on warm clouds over South-East Atlantic, from co-located MODIS and CALIPSO observations, Atmospheric Chemistry and Physics, 13, 69–88, doi:10.5194/acp-13-69-2013, https://www.atmos-chem-phys.net/13/69/ 2013/, 2013.

Gordon, H., Field, P. R., Abel, S. J., Dalvi, M., Grosvenor, D. P., Hill, A. A., Johnson, B. T., Miltenberger, A. K., Yoshioka, M., and Carslaw, K. S.: Large simulated radiative effects of smoke in the south-east Atlantic, Atmospheric Chemistry and Physics, 18, 15 261–15 289, doi:10.5194/acp-18-15261-2018, https://www.atmos-chem-phys.net/18/15261/2018/, 2018

Gordon, H., Field, P. R., Abel, S. J., Barrett, P., Bower, K., Crawford, I., Cui, Z., Grosvenor, D. P., Hill, A. A., Taylor, J., Wilkinson, J., Wu, H., and Carslaw, K. S.: Improving aerosol activation in the double-moment Unified Model with CLARIFY measurements, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-68, accepted, 2020

Herbert, R. J., Bellouin, N., Highwood, E. J., and Hill, A. A.: Diurnal cycle of the semi-direct effect from a persistent absorbing aerosol layer over marine stratocumulus in large-eddy simulations, Atmos. Chem. Phys., 20, 1317–1340, https://doi.org/10.5194/acp-20-1317-2020, 2020

Kacarab, M., et al. "Biomass burning aerosol as a modulator of the droplet number in the southeast Atlantic region." *Atmospheric Chemistry and Physics*, vol. 20, no. 5, 2020, p. 3029

Mallet, M., Solmon, F., Nabat, P., Elguindi, N., Waquet, F., Bouniol, D., Sayer, A. M., Meyer, K., Roehrig, R., Michou, M., Zuidema, P., Flamant, C., Redemann, J., and Formenti, P.: Direct and semi-direct radiative forcing of biomass burning aerosols over the Southeast Atlantic (SEA) and its sensitivity to absorbing properties: a regional climate modeling study, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-317, in review, 2020

Zhang, J. and Zuidema, P.: The diurnal cycle of the smoky marine boundary layer observed during August in the remote southeast Atlantic. Atmos. Chem. Phys., 19, 14493-14516, doi:acp-19-14493-2019, 2019.

Input on previous reviewer's comments: Upon reading the previous reviewers' comments, I came to understand the Fan et al 2018 paper was recommended by a previous reviewer. It does not seem to me that the Fan et al mechanism has support from the CLARIFY measurements nor other modeling studies. I am not clear what the reviewer's recommendation was based on.

Page 2 line 65: the idea that the large spatial coverage of aerosol will generate a regional forcing of increasing importance with time isn't well supported in this sentence. One useful reference here might be

Kloster, D. Bachelet, M. Forrest, G. Lasslop, F. Li, S. Mangeon, J. R. Melton, C. Yue and J. T. Randerson
N. Andela, D. C. Morton, L. Giglio, Y. Chen, G. R. van der Werf, P. S. Kasibhatla, R. S. DeFries, G. J. Collatz, S. Hantson, S.: A human-driven decline in global burned area. DOI: 10.1126/science.aal4108 Science 356 (6345), 1356-1362

Overall I would suggest the authors expend more time thinking critically on the previous reviewer comments, as opposed to simply accepting them.

p. 3 line 76: The Shinozuka et al 2020 paper would also be relevant to cite here, as it shows that there is too much BBA in the BL within most (all?) models, and, that the aerosol is located too low in altitude in most (all?) of the models (explaining the overestimate in the BL). This study also indicates a wide range of model SSA values, which is relevant to the subsequent sentence.

P. 3 line 84: the reason the Rajapakshe et al., 2017 paper reports an overestimate of the aerosol layer is primarily a reflection of the remote sensing algorithm, in which the aerosol layer base is overestimated (i.e. placed too high in altitude). I'm not sure I completely understand the point of this sentence or the previous sentence. Is it that we have little remote sensing information on the aerosol vertical structure from space that is known with confidence?

P. 3 line 90: I think the issue is that the lidar measurements are under constrained, and that the lidar perception of near- and far-field properties does not allow for a fully accurate extinction retrieval. It is not that they rely on assumptions on the aerosol properties. Maybe just say "…measurements due to retrieval limitations" ? I can't think of a great reference here, as I don't believe a thorough analysis of the LASIC lidar data has been published, but Delgadillo et al 2018 do discuss the issues for this type of lidar.

Delgadillo, R., K. Voss and P. Zuidema, 2018: Characteristics of optically-thin coastal Florida cumuli from surface-based lidar measurements. *J. Geophys. Res.*, **123**, p. 10,591-10,605, doi:10.1029/2018JD028867

P. 4 line 114: “the BL” -> “a BL”

p. 4 line 119: how did Hywood et al. 2020 infer an aerosol age? Some more specificity to the aerosol age statements would be nice.

P.7 line 205: “data was” -> “data were”

P. 7 line 225: would suggest defining PM1 on line 207, where it Is made clear that PCASP measurements are based on diameters < 1 micron (as opposed to the PAS/CRDS).

p. 8 line 252: should “were” be put in the present tense to be consistent with the prior “is”?

P. 10, end: I would suggest moving the sentence that is currently on p. 11, line 329-331, to the end of this paragraph. Currently the paragraph feels disconnected and it is not clear to the reader until the next page what the implication of the combustion efficiency is for this study.

p. 13, line 394-396: I’m not quite following this sentence, can it be clarified? I think it is meant to communicate that LASIC SSA values are lower than CLARIFY BL SSA values.

p. 13, lines 397-406: Isn’t the main finding here that CLARIFY FT SSA values agree with those from ORACLES, while the CLARIFY BL SSA values do not agree with those from LASIC, which are also measuring in the BL? what’s not coming through in this paragraph, or the previous sentence, is that the SSA values within Zuidema 2018 were filter-based. Recall that the LASIC PSAP-derived absorption coefficients compare well with those from CLARIFY, and the difference in the LASIC-CLARIFY SSA measurements is in the extinction. So I don’t think limitations with filter-based absorption explain the SSA differences, in contrast to what is written in the paragraph, with the Davies 2019 comparison only able to say something about the CLARIFY filter-derived values, not those from LASIC. Are the CLARIFY SSA values in the previous paragraph derived from the PAS/CRDS? I thought they were but either way might be good to restate here, to help the reader make more sense of the subsequent paragraph. If the CLARIFY SSA values are PAS/CRDS-derived, then the sentence focusing on Davies et al 2019 may not be that enlightening to this discussion. The LASIC and CLARIFY aerosol inlets have slightly different cut-offs (1.0 vs 1.3 aerodynamic diameter). Not clear if that explains it either. Perhaps the authors just want to provide an update on this comparison and say it is under investigation? Overall, please revisit this paragraph, to clarify further the distinction between the FT and BL comparisons.

P. 14 line 427: “could be also” -> “could also be” or “could be”
p. 14 line 431: “of well-mixed” -> “of a well-mixed”
p. 14 line 435: “C036, the” -> “C036, and the”
p. 14 line 459, 460: “”the increase in RH…..an increase in aerosol scattering”. grammatically, I think you need to either go with an “an” or a “the” in both places in front of “increase”
P. 16 line 491: “figure” - >”Figure”
p. 16 line 506: I don’t think the comma is needed.
p. 17 lines 539-540: I don’t understand how having an FT SSA that increases with altitude, and is lowest close to the cloud, enhances the direct radiative effect above what one expects from a column-mean SSA. At least, I think that is what the authors are implying.
P. 18 line 563: The new Mallet et al 2020 acpd manuscript would help to make this statement more concrete.
P. 18, line 578: Looks to me the sentence needs a “more” in front of the “likely” to be consistent with the “larger” at the beginning.

Figures:

Fig. 1: not a big deal but would be nice to see Ascension indicated in the lower 2 panels.
Fig. 2: would be good to spell out what conditions the 3 periods correspond to in the caption, for those readers who look at the figures first before reading the main text.
Fig. 10: would be good to include the date of the C036 flight in the caption, also for readers who look at figures prior to the text.