

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2021-414

Anonymous Referee #2

Referee comment on "Observations of supermicron-sized aerosols originating from biomass burning in southern Central Africa" by Rose M. Miller et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-414-RC2>, 2021

This work reports observations of supermicron-sized aerosol particles (SAPs) using a wing-mounted imaging probe during the ORACLES airborne measurement campaign. Coincident measurements of aerosol composition and corroborating back trajectories support a biomass burning source for these SAPs. The authors hypothesize that the SAPs are unburned vegetative matter (i.e., grass) that is convectively lifted and transported by fires in south-central Africa.

Major Critiques

The number of individual SAP particles identified during extensive smoke-plume sampling is extremely low, and the authors opt to not even calculate their concentrations and rather simply report counts. Thus, it feels that the paper lacks sufficient 1) presentation of the actual data, 2) second-level analysis of SAP properties, and 3) justification for the vegetative source.

- With so few of the SAP particles identified, it would be very informative to show more 2DS images in the paper. My suggestion would be to show a full-page figure of all SAPs for each of the case studies. This will allow readers to get a better sense of the variation in particle shape and would help to corroborate the unburned-grass hypothesis. Second, the paper is focused solely on observations in the African smoke plume, but are there any SAPs that were identified outside of the plume? A comparison of the BB data with data in background boundary layer or for free-tropospheric conditions would strengthen the argument that the SAPs are truly being emitted with smoke.
- There is some qualitative discussion of the SAP particle shape in the text, but a more systematic and quantitative analysis strengthen the paper. First, it is not clear how many of the SAPs are identified in only one 10-um pixel and whether those single pixel detections can be confidently counted as real particles and not a sampling artifact biasing the results. The number of single-pixel counts need to be explicitly presented in the text. Second, can the SAPs be grouped into similar shapes to quantify those that are grass-like (i.e., rods, elongated in one dimension) compared to other more spherical shapes? With so few particles, this grouping could be done manually and

likely would not necessitate a mathematical clustering approach similar to cloud morphology studies. Some form of shape clustering would be helpful, especially to justify the grass source theory.

- The source of the SAPs is purely speculative and needs some further justification. Shape analysis (above) would help, but at least providing a more rigorous literature feasibility study is necessary. Has vegetative material been identified using cloud imaging probes before? Are there images of these particles in the literature that could be compared to the measurements presented here? Can any of the variability in counts flight-to-flight be related to the presence or type of vegetation in the source area?

Minor Edits

Page-Line

2-50 Please clarify this sentence. I'm not sure what you mean by "still contributes to uncertainty estimates of RF".

2-56 A more recent reference is Shingler et al. [2016] for soot restructuring (<https://agupubs.onlinelibrary.wiley.com/doi/full/10.1002/2015JD024498>).

2-58 Please provide a reference for the KCl age statement. Also this should be "KCl" not "KCL".

2-76 Can you further comment here, or more appropriately in the discussion section, about why no SAPs were identified in 2016?

2-85 change "difference" to "differences"

3-104 Can you comment in the 2DS section about instrument noise? What is the false-count rate in clear air? Given that the number of SAPs is so low, I think it's worth the effort to quantify the false-count rate in order to trust the SAP observations. Also for this section, can you include a sentence or two on calibration of the probe?

3-126 The AMS vaporizer is typically operated at a nominal 600C temperature, see the cited DeCarlo [2006] reference. Can you comment on why it was run at 650C for these flights and if that affects the AMS data presented?

3-129 Since you state that the AMS measurements are quantitative, please include a statement about the collection efficiency that was used to calculate final mass concentrations.

4-177 I don't think the Eloranta [2008] reference is appropriate for the HSRL-2 instrument. I suggest Hair et al. [2008] for a general reference for the instrument (<https://www.osapublishing.org/ao/fulltext.cfm?uri=ao-47-36-6734&id=175351>) or just use Burton et al. [2018] (<https://www.osapublishing.org/ao/fulltext.cfm?uri=ao-57-21-6061&id=395340>)

5-199 For this section, I highly suggest adding time-series plots for the case study. This should show, at a minimum, altitude, 2DS counts for SAPs, LWC to confirm a lack of cloud, CO and BC to illustrate the location of the smoke plume. Also, please comment on the following: Are the SAP detections randomly observed or do they cluster in time? Are there SAP detections outside of the smoke plume? Are the SAPs more frequently observed at a specific altitude? As stated above, I recommend that all 72 + 12 SAP images should be shown and discussed for this section.

6-209 The text states that RF11 had 71 SAPs measured but TABLE-1 shows only 12. TABLE-1 also does not have RF12. Please check the text and table and confirm this discrepancy.

8-230 I do not understand the following sentence, "The CAS did not report...". Please clarify.

8-232 What is the conclusion drawn from the CAS data? There seems to be two orders of magnitude difference in concentrations of particles > 10um diameter for RF11 and RF1, but these flights have nearly the same number of SAPs observed (12 and 15 SAPs, respectively, from Table-1). Please comment on this discrepancy. Also, why were these 5 flights chosen for the plot, and what about 2018 flights? In general, I suggest inclusion of a more succinct description of the utility of the CAS data for this plot to be useful and included in the final paper.

9-256 Are the soot, organic, and dust particles that you reference here shown in Figure-6?

9-255 "It is therefore likely that the SAPs are unburnt plant material." I do not follow this argument. Please provide justification.

11-263 As with the first case study, I highly suggest addition of a time series to address the questions posed above. Even though there are more SAP observations, showing each image would be beneficial.

12-278 "SAPs could be correlated with fire intensity". This statement is very speculative and needs to be justified or removed. I do not know what "copious amount" of material means, please explain.

13-292 "... were always observed within the BB aerosol plume..." As written, there is no evidence for this statement. Time series of the case studies showing both BB and non-BB sampling would be helpful. Statistics for BB and non-BB sampling would also help.

13-303 "This could have resulted from an increase in fire intensity." This statement is very speculative and needs to be justified or removed.

14-322 "Aerosol plumes where SAPs were observed...". I do not understand this statement. Why is the "spread" in the AMS data indicative of more intense fires? Please explain.

15-325 The caption states that data are shown for "5 minute average when SAPs are observed within the BB plume", but for many of the flights the SAP count was zero and data are still shown? I'm confused. Please clarify. Also, RF5 (2017) has more sulfate than organics and therefore does not appear to be from BB. Please comment. Same argument for RF4 (2018).

18-355 What is the bin width of the histograms? How many of the counts are for 10um particles (i.e., for a single pixel). Please comment on if you think these are real or a possible artifact.