Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-449-AC2, 2020

Atmospheric Chemistry and Physics Discussions

Interactive comment on "An overview of the ORACLES (ObseRvations of Aerosols above CLouds and their intEractionS) project: aerosol-cloud-radiation interactions in the Southeast Atlantic basin" *by* Jens Redemann et al.

Jens Redemann et al.

jredemann@ou.edu

Received and published: 9 October 2020

Authors' response to interactive comments on "An overview of the ORACLES (ObseRvations of Aerosols above CLouds and their intEractionS) project: aerosol-cloud-radiation interactions in the Southeast Atlantic basin" by Jens Redemann et al.

Anonymous Referee #2 Received and published: 13 August 2020

We greatly appreciated this reviewer's scrutiny of our paper. We feel that their comments stem from great knowledge and experience with investigations similar to OR-

ACLES. We have taken the liberty of numbering their comments and attempted to respond with the diligence that the thoughtfulness of the comments inspired.

Detailed comments:

1. The first four sections of the paper are lengthy, but very informative. I wish that the background and context for more of these large field campaigns were so thoroughly documented in the literature. However, the conclusions drawn from figure 9, which are very important to the broader context of the experiment, could be more concretely supported. The notions that the "fire counts in the three deployment years are very similar to the climatologies" and that the analysis "supports the conclusion of an earlier and possibly prolonged presence of the biomass burning plume ...in recent years" are only supported with small color/contour maps, when the quantitative data presented in those charts could actually be interpreted with proper statistics to determine the magnitudes of any differences between them and how the relative differences or similarities compare to the magnitude of interannual variability. Rather than asking the reader to eye-ball the differences and interpret subtle differences in shading or contour shapes, why not actually reduce the data to statistics that support the conclusion?

Response: A more careful analysis indicated that the statement about "... an earlier and possibly prolonged presence of the biomass burning plume ...in recent years" is overly simplistic and that it depends strongly on the observational period considered. As the analysis of historical context is beyond the scope of this overview paper and somewhat tangential, we decided to just remove the sentence on burning season length from the discussion following figure 9 and leave the analysis to a future publication. Our added text in response to this reviewer's suggestion 2 below partially addresses the context issue.

2. At the very end of section 4.3 is a brief summary of the broad conclusions for how the aerosol and cloud properties during the months of the field campaign relate to climatologies and interannual variability, but no conclusion is drawn for what this means

for the outcome of the field campaign. Do the authors feel they captured representative conditions from their sampling? Do the differences from climatologies noted for August 2017 and October 2018 have any implications for the resulting dataset in terms of whether the results drawn from the data can be thought of as broadly representative of aerosol/cloud relationships in the region?

Response: We have added the following text to the end of section 4.3: "Taking all three years together, mean ACAOD values were slightly lower than their climatological mean values for August-October and Nc values slightly higher than climatology. These deviations are substantially smaller than the day-to-day variability sampled in the three campaigns, and we therefore consider the ORACLES measurements to have captured representative conditions overall. Detailed assessment of the representativeness of the actual aircraft observations, which only sampled a fraction of the days within each of the three measurement months, has been undertaken in the model-observation intercomparison studies (Shinozuka et al., 2020). This assessment indicates that the airborne sampling provides averages sufficiently representative of the monthly means to be able to characterize and test model skill at representing geographical gradients in climatological mean plume structure. The wide range of varying aerosol-cloud vertical structures sampled are sufficient for addressing all of the originally-postulated objectives."

3. The discussion in the paragraph starting in line 820 raises the question of whether any future planned satellite lidar instruments will have the signal-to-noise ratio to provide a better view of the frequency of clear air between aerosol and cloud layers? Can the results of ORACLES help constrain a future mission so that past inferences about the relationships between aerosol and cloud layers can be improved upon with large statistics from satellites in the future?

Response: The ORACLES results can indeed be used to study the information content and capabilities of future satellite missions. A manuscript led by co-author F. Xu that studies joint lidar+polarimeter retrieval capabilities on the basis of ORACLES RSP and

HSRL-2 observations in the context of the NASA observable study ACCP (Aerosols, Clouds, Convection and Precipitation) is close to submission and will be added to the reference list if it is submitted before this manuscript needs to be finalized. The question on required SNR to unambiguously detect the clear-air layers in the SE Atlantic depends on very specific instrument characteristics. For now, we have added the following text to the manuscript at the end of section 5.2.1: "ORACLES data provide a useful testbed for algorithm development in support of future satellite missions, for example NASA's ACCP (Aerosols, Clouds, Convection and Precipitation) mission. For instance, ORACLES observations are currently being used to develop joint polarimeter+lidar retrievals of aerosol and cloud properties. Whether such observations will successfully detect features such as the clear-air layers in the SE Atlantic will depend on specific instrument characteristics, but the ORACLES measurements should provide useful benchmarks for the testing of candidate observing concepts."

4. The discussion between lines 861 and 865 is confusing and seems to be missing some key elements to the interpretation of the observed SSA and BC:OA ratio. From figure 14, Aug. and Sept. appear similar with Oct. being the outlier, but the discussion draws a distinction, in terms of 4 km winds, between Aug. and the other months. Furthermore, I was not clear on exactly what the higher winds in Sept./Oct. are responsible for. Finally, if I am understanding the argument correctly, the aerosol lower in the column is older and also exhibits lower SSA and higher BC:OA ratio. However, the discussion does not link these aerosol properties to age? Am I to conclude that aging depletes the organic fraction of the aerosol, and that is why the SSA decreases with altitude?

Response: The discussion surrounding Fig. 14 in lines 840-865 has been modified to more clearly reflect what is shown in the figure and the inferences we make from it, with the rewritten paragraphs included here: "The lidar-derived increase in extinction with height for September (2016) in Fig. 13 is accompanied by a similar increase with height of the mean in-situ SSA (derived from the in-situ PSAP absorption paired with nephelometer scattering at 530 nm) from 0.84 to 0.87 (Fig. 14, middle of top row). The SSA values are in reasonable agreement with values of ∼0.83 for dry conditions in Davies et al. (2019) derived using state-of-the-art photoacoustic and cavity ring-down instrumentation. Pistone et al., (2019) compare the ORACLES absorption+scattering measurements with SSA derived by several different airborne remote-sensing methods at wavelengths between 400 and 995nm and found reasonable agreement both for specific case studies and for the range of measured spectral SSA over the full ORACLES-2016 deployment. Black carbon is the primary absorber of sunlight within BB aerosol (e.g., Bond et al., 2013). Although a corresponding decrease with height of the refractory black carbon (BC) mass concentration relative to the mean organic aerosol (OA) mass concentration is not clearly apparent in Fig. 14, (middle of middle row), an example from an individual profile from the September 24, 2016 flight indicates more nitrate and organic aerosol above 3.5km than below it, relative to the black carbon mass concentration (Fig. 14, middle of bottom row). This is consistent with an increase in SSA with height. Examples of individual profiles are shown from each year, broken down by aerosol species [black carbon, organic aerosol, nitrate (NO3), ammonium (NH4) and sulfates (SO4)], indicate distinct vertical structures (Fig. 14 bottom row). Profiles from August 2017 also indicate some vertical structure to the SSA without a clear mapping to the BC/OA ratio, while the mean SSA from the October, 2018 deployment increases even more sharply with altitude than does the mean SSA from September (2016). For October, 2018, the increase of SSA with height is clearly consistent with the proportional increase in organic aerosol relative to black carbon. Work is ongoing to attribute changes in SSA to both thermodynamically-driven changes in gas-particle phase partitioning (e.g., Wu et al., 2020) and more irreversibly driven changes related to forms of photodegradation in this near-equatorial, sun-exposed environment. Overall the SSA values are less than has been previously assumed, with that implication further explored within the modeling study of Mallet et al., (2020).

Some of the differences between the individual profiles in Fig. 14, bottom row, can be related to differences in the prevailing meteorology shown in Fig. 8. The distinc-

tive two-layer aerosol structure profiled on 24 September, 2016 reflects the ability of strong winds at 4 km (see also Fig. 6) to disperse aerosol westward, with the aerosol lower down, at 2-3 km, resulting from an anticyclonic circulation (Fig. 8). The strong 4km zonal winds are much less apparent in August, consistent with a lower-lying, less layered aerosol vertical structure. The free-tropospheric winds remain strong into October, but by then the fire emissions have reduced considerably and less aerosol appears to reach the altitude at which the zonal winds are strongest. “

5. The results presented in figure 18 are intriguing and certainly demand further study. I understand that there is a paper in progress to do just that (Gupta et al. 2020). However, it is rather disappointing to read that there are hypotheses that might explain the results, but not be able to read what they are. Is it not possible to share the hypotheses and then note that they are to be evaluated in the other paper?

Response: This point is well taken; after considerable discussion among co-authors and mild disagreement on the hypotheses that best explain the findings, we would prefer to eliminate any mention of specific hypotheses and instead refer the reader to Gupta et al., 2020. Consequently, we have made the following changes to the paper:

a) We have changed the sentence “...it experiences significant increase with ZN for the Contact/Na>250 cm-3 case, possibly for reasons hypothesized by Gupta et al. (2020).” from lines 950-951 of the original manuscript with “...it experiences a significant and unexpected increase with ZN for the Contact/Na>250 cm-3 case.”

b) We have replaced “"These findings, and hypotheses for the processes responsible, are further described by Gupta et al. (2020)." with "These findings are further described by Gupta et al. (2020)." on lines 957-958 of the original manuscript;

c) For the bullet point in the conclusion section (lines 1179-1180) we replaced "there was ample evidence for aerosol-induced modifications of Sc cloud properties (Gupta et al., in prep.), and some evidence for the suppression of drizzle (Dzambo et al., 2019);" with "there was ample evidence for aerosol-induced modifications of Sc cloud properties (Fig. 18) and those of mid-level clouds (Adebiyi et al., 2020); a new cloud/rain dataset produced by a joint multi-wavelength cloud radar and multi-angle/multi-wavelength polarimeter will allow for further investigation for the suppression of drizzle by aerosol (Dzambo et al. 2019, 2020);"

6. Conversely, there are a number of declarative statements in the bulleted list in section 7 of the paper that are not supported in the paper and the supporting citations are to papers that are not yet published. While I understand that the notion here is to summarize some of the findings from the campaign as they presently stand, this does raise a question of whether it would be proper to have this paper on the record citing some declarative results that may either not end up in the peer-reviewed literature, or be altered somewhat after peer-review.

Response: See our response to point 18 raised by reviewer #1 - We removed the bullet point with the conclusion about the kappa parameter (which was referred to with the numerical value of 0.2) and eliminated all references to papers that have not yet been submitted. Fortunately, except for the bullet point with the kappa parameters, all other conclusions are supported by figures in this paper; we added references to sections 4.2, 4.3, 5.2.2 and 5.2.3 and Figure 18, where necessary.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-449, 2020.