

Response: We thank the reviewers for reviewing our manuscript and providing feedback. Below we provide responses to comments and suggestions in blue font.

Reviewer 1: Accepted as is

Response: Thanks for the support.

Reviewer 2:

I appreciate the consideration the authors gave to comments from Reviewer 2 and myself, and changes they made in response. In particular I am pleased by the additional attention given to establishing that the observed enrichment of organics in residual particles is very unlikely to be a measurement artifact, and the expanded discussion of the lack of evidence for a clear impact of cloud processing on the composition of aerosol sampled just above or below clouds.

As noted in my first review, it seems misleading or ill-advised to suggest that measuring the composition of residual particles provides insight into the composition of the CCN that activated to form the cloud drop, especially when the main point of the paper is to suggest that aqueous processing in cloud drops significantly modifies the composition of the residual particles. I urge the authors to consider reworking 2 sentences in the introduction to address this concern. Specifically I would change the sentence in lines 84-87 to something like: "Furthermore chemical analysis of droplet residuals should lend insight into the properties of the aerosol that will be released after the droplet evaporates which could control its propensity to activate in a subsequent passage through cloud, with past work.....(ref as they are)". Likewise, I would suggest deleting the last sentence of the introduction to remove this misleading assertion (any modification that I might suggest for this sentence would be largely redundant with those outlined for lines 84-87.)

Response: We modified Lines 84-87 as suggested: "Furthermore, chemical analysis of droplet residuals should lend insight into the properties of aerosol particles that will be released after droplets evaporate, which could control their propensity to activate in a subsequent passage through cloud, with past work showing an important role for organics (Russell et al., 2000; Drewnick et al., 2007; Mertes et al., 2007; Hawkins et al., 2008; Asa-Awuku et al., 2015)."

As suggested, we also deleted the last sentence of the introduction.

When I read the response to reviewers I was mildly alarmed by the assertion that "Others have shown a similar type of phenomenon for other regions....." (First paragraph in the third block of blue text/author response). Issue is that the Sorooshian group apparently made all of the other measurements in other regions using other techniques that are cited in this sentence, making this not truly independent confirmation. As a result I read section 4 quite closely, and found that the language describing prior work is much more careful in the revised text than in the response document.

Response: We interpreted this comment as saying that the manuscript text was written carefully and is thus adequate, whereas they felt the response file text was "mildly alarming". Either way, we are confident the manuscript text is fine as is in Section 4 in terms of addressing past work in relation to results of this study. As a result, we make no further changes for this comment.

Below are a small number of editorial suggestions to consider.

lines 145-146 For consistency it seems that the manufacturer of the 2DS-V probe should be provided.

Response: Done – added “(SPEC, Inc.)”.

lines 236-238 Is this sentence needed? No attempt is made anywhere in the paper to explain why eastern North Atlantic should be so different than the western part focused on in ACTIVATE. Just pointing out such a major difference raises a lot of questions in some readers' minds that beg answers. If you are not going to try to give the answers why throw a possibly troublesome bit of trivia onto the table?

Response: That sentence is certainly not needed so we removed it.

line 245 is it important that chloride is low? Much later the fact that the AMS is not very sensitive to SS chloride is acknowledged; should that be mentioned here instead (if you do not choose to stop the sentence at the comma and delete any mention of Cl)?

Response: We just removed the part in question from that line: “~~with chloride being much lower.~~”

lines 363-375 I like Figure 4, but have to point out that this figure makes it much easier for the reader to see a point I raised last time; i.e the organic mass fraction is generally higher in cloud free air than it is in the residual particles sampled during the same season. I feel it would be a good idea to point this out and explain it (as done in the response) rather than hope readers do not notice.

Response: Sure, good point. We added the following text to that section: “The higher organic mass fractions in the BBL/ABL legs of clear ensembles relative to BCB/ACT legs of cloud ensembles can be explained by how most of the clear ensemble data were collected closer to land where there are greater organic levels in the continental outflow relative to farther offshore where sulfate presumably becomes more important due to marine emissions of precursors such as dimethylsulfide. The region’s synoptic flow is not always strictly offshore from west-to-east. Thus, the higher organic content near the coast often could just be due to local emissions that are confined to the coast and are not advected any farther east.”

lines 402-403 change to "Comparing CVI-AMS data to the closest ACT leg in the same ensemble gives similar trend (not shown)."

Response: Change made.

line 471 delete "with", or change "with conducting" to "to conduct"

Response: Change made: “A way to test this is to conduct CVI...”

line 617 Not sure you know that the aerosols resulting from cloud processing will shift in "size" compared to the precloud CCN. Very plausible that they will gain organic mass, so a dry aerosol might be larger, but if the extra organics make the particle less hygroscopic it would take up less water and might

be smaller.

Response: Change made to mention that there is a possible shift in size but not guaranteed: “That the droplet residuals shift to a more organic-rich signature with more oxygenated organics has implications for the aerosol particle properties remaining after droplet evaporation as they shift in composition and possibly size.”

line 643 change "another" to "one other"

Response: Change made.