Editor's Report:

Three reviewers have provided comments, with reviewer 1 suggesting minor revisions that account for the known variation in aerosol vertical structure from July-September, reviewer 2 also suggesting minor clarifications, primarily of the model and how it is used, and reviewer 3 also suggesting minor revisions, primarily related to the language.

After reviewing the comments, the authors' response, and the overall manuscript, it is my determination that the authors have not sufficiently responded to the intent of the reviewer comments. I suggest the authors give this another go-through. Reading through the manuscript, I also have several specific comments, listed below in primarily chronological order, that I would like to see addressed before the paper is finalized.

Abstract: Please read this over more carefully. Keep in mind many readers will not look past the abstract.

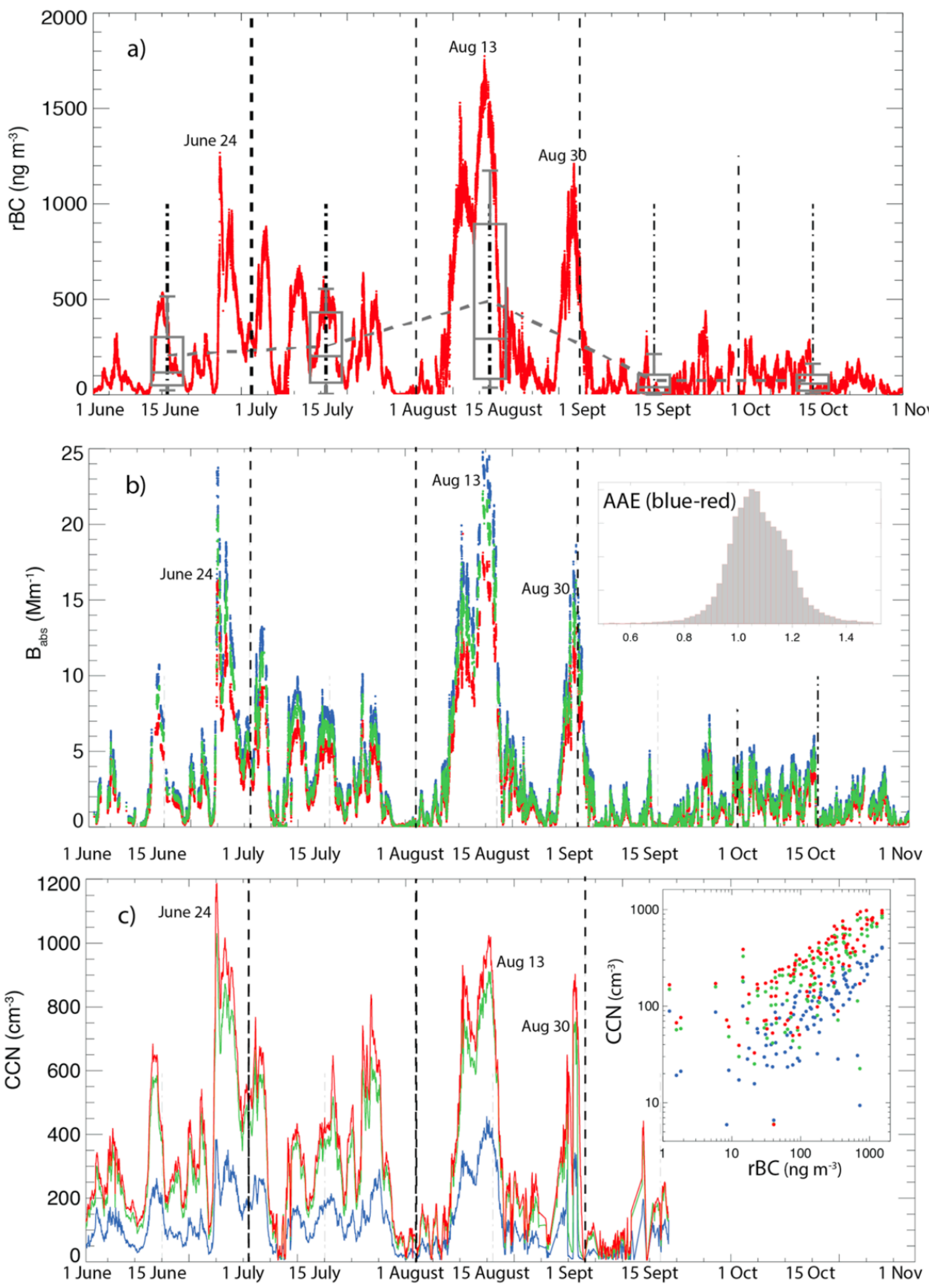
1. The description of 'total nucleation' in the abstract as 'tropospheric and stratospheric nucleation' is confusing. This is the first time the reader encounters this term. It's worth including an additional sentence here to define the term.
2. Mention the time period you are looking into.
3. Lines 21-23 seem to include two contradictory phrases - perhaps the authors mean to say most of the BL CCN is introduced from above through entrainment? Wouldn't BB then be the dominant BL CCN source?
4. Line 25: the reader doesn't yet know the model simulation places most of the aerosol above the BL. Keep reviewer 3's comments in mind here and rewrite.
5. State something about the model aerosol vertical structure in the abstract - results are highly dependent on the aerosol being located above the cloud.
6. The reader might be reasonably surprised to read that non-BB anthro emissions are the largest contributors to MBL CCN_0.2%, above BB and sea spray. On p. 12 you clarify the CCN in the cloud layer - thus those most likely to form cloud droplets - are more likely to be BB. This would be worth mentioning in the abstract.

Overall:
The authors conclude, that because in the UKEMS1 model, the BBA is mainly in the FT for July-September, and has a poor hygroscopicity, that BBA is less important as a BL CCN, and instead, primarily serves to strengthen the inversion top. This is valuable to know about the UKESM1 model behavior. What is less clear is how well the UKESM1 simulations are capturing the observations. The reviewer comments relate to this: Reviewer 1 mentions the seasonal cycle. Rev 2 and 3 mention model characteristics.

For example: the LASIC campaign has shown that there can be significant CCN in the BL, for example, Zuidema et al 2018 Fig. 1, reproduced below, shows CCN_0.2% reaching 10^3 /cc. The temporal variation indicates it is primarily modulated by BC. These values I believe exceed those shown in Fig. R1, though Fig. R1 is difficult to interpret; absolute values for the CCN depicted would have helped, or at least an explanatory caption. The July-September model means shown in Fig. 2 are difficult to interpret for the BL, and a 3-month model mean doesn't communicate the range.

In addition, in several portions of the manuscript, the authors refer to Che 2021, as a model validation paper. That paper only compared aerosol extinctions along CLARIFY and ORACLES flight tracks, with the ORACLES flight tracks spending little time in the boundary layer. During September, the LASIC values also indicate a clean MBL, consistent with ORACLES-2016, but a BL lacking smoke in September does not mean the BL is also non-smoky in July and August. The comparison to the aircraft flight track data isn't a sufficient validation for the 3 months, in contrast to the statement on p. 8, line14. Is the model genuinely capturing the boundary layer smoke in July and August? Can the authors create a figure from their model simulation that is comparable to the LASIC data? The authors also refer to several other papers as a form of validation (e.g., p. 2, line28): Deaconu 2019 and Wilcox 2010 rely on satellite datasets that have difficulty distinguishing BB within the BL from sea-spray, Gordon 2018 is a modeling study focusing on a 10-day August time period only that produced an unrealistic 8K warming in the free troposphere, and Ackerman 2004 is for a different location. These references ignore the new information we have thanks to LASIC, ORACLES, and CLARIFY. Besides the studies mentioned by Rev 1, there are also more detailed StCU-to-Cu transition papers indicating the BB can also have a radiative impact in the BL.

Similarly, Kacarab et al. 2020 is also relevant, indicating a kappa of 0.4 for smoke based on oracles observations. This study should be referenced and discussed somewhere, as it does not support the low hygroscopicity for smoke assumed here. https://acp.copernicus.org/articles/20/3029/2020/.



The manuscript should also be read over again by a native English speaker, to clarify some of the language. I mention a few specific comments below:

Line 12 p 3: of ->from
Line 13 p 3: to the -> to that of
Line 16: after -> by
etc.

p. 3 line 20-21: Kalahari dust doesn't advect far according to https://acp.copernicus.org/articles/21/8169/2021/acp-21-8169-2021.pdf, and it certainly wasn't one of the most observed aerosol at Ascension Island during the LASIC/CLARIFY campaigns. The Begue result for the Netherlands isn't relevant here. The first author could draw on their own work assessing kappa using LASIC measurements. Overall this paragraph and its emphasis on dust lacks support and is misleading.

p. 5: are the BC aerosols internally mixed? Results don't acknowledge that a kappa of 0 for BC doesn't reflect that all of it is likely internally mixed (e.g., Dang et al., 2021), with the BC particle size helping cloud nucleation. How well does the Petters and Kreidenweis internal mixing rule work for this region based on what we know so far from the observations?

P. 5: clarify that anthropogenic does not include BB. You could consider calling it 'non-BB anthropogenic'.

Language on nucleation confusing - authors use the same term for gas to particle production of aerosols, and for cloud activation. Here it might be worth adding additional detail to the naming, meaning, to use the longer term of 'aerosol nucleation'. Mention how boundary nucleation differs from total nucleation in the boundary layer for caption of fig. 3. Also, the comment from Rev 2 that the Hadley center models lend more emphasis on nucleation driven CDNC than other models should be mentioned, including the citation to Bellouin et al 2013.

p. 7 line 11: Kacarab et al. 2019 is not consistent with the low model hygroscopicity.

p. 8: Note Redemann 2021 shows satellite-derived Nd that are clearly elevated

p. 16, line 7: to say that 'nucleation is important to CDNC' is very unclear - if you're talking about cloud nucleation, it's overstating the obvious. You don't mean cloud nucleation I recognize, but this is simply not clear writing. Explain what nucleation means in these sentences.

More detail on the non-BB anthro emissions would also be useful. I recognize these are difficult to validate - do the sulfate contributions from the non-BB anthro+sea spray match what was measured at Ascension Island?

A bit more effort could be made in the last section mentioning how the new observations can be used to assess and/or improve the model, including using the enhanced resolution of the seasonal cycle.