Response to reviewers

Firstly, we sincerely thank the editor and referees for their contributions to this work.
We have addressed the comments below. The original comments are in black, our replies are in blue and the changes in original manuscript are in red.

Anonymous Referee #3

General Comments: 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

Detailed Comments:
1. 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.
We have removed "for the first time".

2. 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
We have added the reference of Adebiyi, A., and Zuidema, P. (2016).

3. 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.
We have removed the reference of Abel et al, (2020) to avoid misleading the reader.

4. 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.

We have extended the introduction of indirect and semi-direct effects, specifically for the southeast Atlantic studies.

These smoke layers typically over-lie vast stretches of marine stratocumulus clouds (Adebiyi et al., 2015), where they can exert a warming effect by absorbing both downwelling solar radiation and that scattered upwards from the low-lying clouds (Samset et al., 2013). This direct radiative effect is sensitive to the smoke's single-scattering albedo (SSA), which is a function of aerosol composition and size and evolves with particle age (Abel et al., 2005). Space-based and in-situ field observations also suggested that the smoke layers can be entrained into the marine boundary layer (MBL) during its transport from land over ocean (Painemal et al., 2014; Zuidema et al., 2018; Haslett et al., 2019a). *The entrained aerosols in the MBL can affect cloud microphysics by acting as cloud condensation nuclei (CCN), inducing indirect radiative effects over the southeast Atlantic by increasing cloud droplet number and reducing precipitation, thereby increasing cloud coverage and cloud albedo (e.g. Costantino and Bréon, 2013). In addition, BC immersed in cloud droplets absorbs light and may facilitate water evaporation. BC below clouds could enhance the formation of convection by providing additional heating within the sub-cloud layer. Zhang and Zuidema (2019) reported that shortwave absorption within the smoky MBL reduces the sub-cloud relative humidity due to raising the temperature, and so reduces daytime low-level cloud cover over the southeast Atlantic, which is opposite to the mechanism of increased aerosol increasing cloud droplet numbers. Furthermore, large eddy model studies have shown that marine stratocumulus clouds over the southeast Atlantic also adjust to the presence of overlying absorbing aerosol layers, depending on their properties and distance with low-cloud deck (e.g. Herbert et al., 2020). The above-cloud shortwave absorption can warm the FT, strengthening the temperature inversion and reducing the entrainment of warm and dry air from the FT into the MBL, thus influencing MBL humidity, temperature and dynamics. These effects described above, which perturb the temperature structure of the atmosphere and influence the cloud distribution, are collectively termed semi-direct effects.*

5. 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.

The referee has identified a lack of clarity in this sentence. We did not mean to discuss changes with time in this sentence, rather discuss the spatial extent of the aerosol influence. What we want to say is, the large spatial coverage of aerosol will generate an important regional forcing. We discussed the changes over time in the previous paragraph, "As controls continue to reduce aerosol emissions from fossil fuels and a changing climate potentially leads to more fires, the relative impact of BB on climate forcing is expected to increase (Fuzzi et al., 2015).", we also say "the contribution of transported BB aerosol to regional radiative forcing will also be increasingly important in the future".

We have removed "will be increasingly important" from the sentence highlighted by the reviewer to make sure the sentence is no longer misleading.

Although BB transport regions have lower aerosol concentrations than areas closer to the source, the large spatial coverage means that their contribution to the regional/global-average forcing is important.

6. 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.

We have added this information.

A recent study demonstrated that models generally underestimate the smoke base height over the southeast Atlantic and thus lead to an overestimation of aerosol loading in the MBL (Shinozuka et al., 2019). Uncertainty in SSA is also one of the largest sources of uncertainty in estimating the aerosol direct effects (McComiskey et al., 2008; Shinozuka et al., 2019).

7. 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?

We agree with the reviewer and that is what we wanted to say - the aerosol layer base using the remote sensing is overestimated, and BB layer is placed too high in altitude. We have rephrased this.

Satellite-based observations have been employed in this region, but satellite retrievals often detect the bottom of the aerosol layer too high and thereby overestimate the above-cloud aerosol height (e.g. Rajapakshe et al., 2017). The ability of satellites to quantify BBA amount and its microphysical and optical properties in the marine BL is also limited, since the presence of intervening cloud layers brings significant challenges to retrievals of aerosol properties. Due to the persistent stratocumulus cloud deck over the south Atlantic, most of the region is affected by clouds, and so MBL properties are hard to obtain from satellites.

8. 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.

We have rephrased this.

The vertically resolved retrievals obtained during the LASIC campaign using a co-located micropulse lidar also have retrieval limitations (Delgadillo et al., 2018).

9. P. 4 line 114: "the BL" -> "a BL"

We will keep "the BL" in manuscript, since we mean the specific BL over the southeast Atlantic.

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

We have added the information.

Haywood et al (2020) conducted back trajectories with particles released from Ascension Island at different altitudes from the MBL to FT, and they reported that air masses sampled in the CLARIFY operating area were of African BB origin and also indicated that the aerosol age was likely in a range of 4 to 10 days.

11. P.7 line 205: "data was" -> "data were" Accepted

12. 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).

Submicron aerosol (PM1) number concentrations from the PCASP were calculated using bins with diameter below 1 μm.

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

"the NH_{4mea}^{+} is the measured ammonium concentration from the AMS, the NH_{4neu}^{+} is the calculated ammunium concentration if all acids in the aerosol were neutralized" defines the terms NH_{4neu}^{+} and NH_{4mea}^{+}, so we used present tense.

"The linear fitted $NH_{4mea}^{+}/NH_{4neu}^{+}$ ratios in the BL were (0.86 ± 0.01) and (0.99 ± 0.02) for period 1 and 3 respectively" presents measurement results from CLARIFY, and we consistently used past tense to describe observation results in the manuscript. So, we will keep the sentence as was written in the original manuscript.

14. 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.

We have re-organized these two paragraphs.

The modified combustion efficiency (MCE) was calculated to indicate the combustion conditions at source (Yokelson et al., 2009). Details of the method of calculating MCE are listed in Supplementary S1. The MCEs of FT smoke were generally around 0.97 during CLARIFY, as shown in Fig. 5. An MCE > 0.9 is commonly used to indicate BB smoke predominantly influenced by combustion during the flaming phase whereas, MCE < 0.9 represents the smouldering phase (Reid et al., 2005). By this definition, CLARIFY smoke plumes transported from southern Africa are likely to be mostly controlled by flaming-phase combustion at source. The emission of BC is usually high during flaming combustion, while smouldering combustion tends to emit smoke high in CO and organic mass (e.g. Christian et al., 2003). The enhancement ratios of BC and OA with respect to CO (BC/ΔCO and OA/ΔCO, $\mu g\ m^{-3} / \mu g\ m^{-3}$) are generally used to indicate the emission conditions of fire at source. For example, BC/ΔCO values from 0.005 to 0.023 and OA/ΔCO values from 0.037 to 0.066 were observed for BB source in flaming combustion from previous measurements (May et al., 2014; Pratt et al., 2011), while a lower range of (0.0014 – 0.0072) for BC/ΔCO and a higher range of (0.080 – 0.096) for OA/ΔCO, were reported for BB source in smouldering combustion (Capes et al., 2008; Kondo et al., 2011; May et al., 2014).

For CLARIFY, the BC/ΔCO and OA/ΔCO ratios ($\mu g\ m^{-3} / \mu g\ m^{-3}$) were calculated in the FT by the unconstrained linear orthogonal distance regression (ODR) fit (Yokelson et al., 2013), and were calculated in the BL by dividing BC and OA by the excess concentration of CO, after background values had been removed (Lefer et al., 1994). The detailed calculation method is listed in Supplementary S1. The calculated enhancement ratios in FT and BL smoke for each flight are shown in Fig. 5.

15. 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.

We have rephrased this sentence.

Ground-based in-situ SSA measurements made on Ascension Island in 2017 (Zuidema et al., 2018) are lower than CLARIFY BL SSA values, and are the lowest values compared to all previously reported observations of southern African BBA.

16. 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.

We have rephrased this paragraph, emphasized the SSA consistence between CLARIFY and ORACLES FT values, and SSA difference between CLARIFY and LASIC BL values, and also the possible reason for the SSA difference between CLARIFY and LASIC.

These previous observations employed filter-based measurements, using the Particle Soot Absorption Photometer (PSAP) and nephelometer, in contrast to the PAS/CRD methods employed during CLARIFY. CLARIFY FT SSA values are similar to those measured from the FT during the ORACLES mission. It is also interesting to note that the radiometrically retrieved SSA from nine above-cloud flights performed during ORACLES in 2016 and 2017 (Cochrane et al., 2020; their Figure 4), which do not depend on in-situ measurements, yield average SSAs of (0.85 ± 0.02), (0.83 ± 0.03) and (0.82 ± 0.04) at wavelengths of 380, 550, and 660 nm respectively for FT BBA. These values are also in good agreement with our FT SSAs within the expected variability. However, CLARIFY BL SSA values do not agree with those from LASIC ground-based measurements. Although limitations with filter-based measurements of aerosol light absorption are known to introduce systematic measurement biases (Lack et al., 2008; Davies et al., 2019), the LASIC-derived aerosol absorption is comparable with those from the CLARIFY. The difference between CLARIFY and LASIC BL SSAs is possibly due to differences in the extinction measurements, which may be caused by the different inlet cut-offs (aerosol dynamic diameter of 1 μm for LASIC and 1.3 μm for CLARIFY).

17. P. 14 line 427: “could be also” -> “could also be” or “could be” Accepted

18. p. 14 line 431: “of well-mixed” -> “of a well-mixed” Accepted

19. p. 14 line 435: “C036, the” -> “C036, and the” Accepted

20. 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” Accepted

21. P. 16 line 491: “figure” - >”Figure” Accepted

22. p. 16 line 506: I don’t think the comma is needed. Accepted

23. 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.

We have added more explanation here. As the SSA of aged BBA used in climate models is generally higher than the SSA in this study, the positive radiative direct effects above clouds may be underestimated. Since there is also a vertical variation in SSA, with much lower values at lower levels within the FT, this underestimation maybe more important for the cases with lower and thinner smoke layers above clouds. We also extend the discussion to include the implications for semi-direct effects.

The relatively low dry SSA measured during CLARIFY, as determined by highly sensitive and accurate measurements that are not subject to the artefacts of filter-based methods, is an important result. The SSA of aged BBA used in climate models is generally higher than the SSA in this study (e.g. Randles and Ramaswamy, 2010; Johnson et al., 2016; Herbert et al., 2020). Furthermore, the vertical profiles of SSA show that the lowest values (averages: 0.82, 0.81 and 0.79 at 405, 550 and 658 nm) occur at low FT layer around 2000 m altitude, immediately above the stratiform cloud. The air is also relatively dry within these low FT layers, meaning that the measured dry SSA is analogous to ambient condition. This is important as the positive radiative feedback associated with the aerosol direct effects may be underestimated in current models, especially for the cases with low and thin smoke layers above clouds. Herbert et al. (2020) also found that both the cloud response and semi-direct radiative effects increase for thinner and denser overlying aerosol layers with lower SSA. The bias in modelled SSA values is likely to lead to mis-representation of semi-direct effects as may neglecting the vertical variation in SSA. These findings suggest that modelled climate effects of BBA in this region need reassessment in future studies and the variation in SSA values in different BB regions should be considered.

24. P. 18 line 563: The new Mallet et al 2020 acpd manuscript would help to make this statement more concrete.
Considering these BBAs have a long lifetime and their spatial range spans thousands of kilometres, and the direct and semi-direct radiative effects of smoke layers in the southeast Atlantic area are highly sensitive to the absorbing properties of BBA (Mallet et al., 2020), modelled climate effects need re-assessment over this region.

25. 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. Accepted

Figures:

26. Fig. 1: not a big deal but would be nice to see Ascension indicated in the lower 2 panels. Accepted

27. 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. Accepted

28. 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. Accepted