Referee Report #1.1:

The authors have satisfactorily addressed most of the previous comments, and the additional MERRA-2, MODIS fire, and HYSPLIT analyses improve the manuscript. I recommend publication after the following minor revisions.

1. Lines 261–272 and 449–451: conclusions regarding BrC remain somewhat stronger than the available evidence. The statement that a nearly constant MERRA-2 BC:OC ratio indicates "little change in BrC concentration" is not sufficiently justified, because MERRA-2 OC is not equivalent to optically absorbing brown carbon and does not separate BrC from non-absorbing organic aerosol. Please revise this interpretation. It would be more appropriate to state that the BC:OC ratio suggests limited change in the bulk carbonaceous aerosol partitioning, but cannot independently constrain BrC. For the same reason, expressions such as "lack of BrC contributions" and "domination by fine BC aerosols" should be softened to "no clear optical evidence for a substantial BrC contribution within the measured wavelength range."

Author's Response #1.1:

Thank you for highlighting the fact that changes in BrC concentration are not directly inferable from the OC concentrations. As such, we have further softened our BrC conclusions and have adopted your suggested language.

Manuscript Changes #1.1:

Rearranged and softened our language regarding BrC contributions in Lines 271-283:

The AAE interquartile ranges are about 1.05 - 1.35 for August and September, which drops to 0.8 - 1.1 in October, remaining within the typical AAE range of BC throughout the season (Liu et al., 2018). If the aerosol brightening in October were caused by increased BrC contributions, then we would expect the AAE to increase, rather than decrease (Russell et al., 2010). This is complemented by BC and OC columnar mass densities from MERRA-2 (Modern-Era Retrospective analysis for Research and Applications, version 2) reanalysis (Fig. S1), where the BC:OC ratio remains near 1:10 throughout the ORACLES campaign months, suggesting limited change in bulk carbonaceous aerosol partitioning.

We cannot fully discount the presence of BrC, as this study was unable to directly examine the UV wavelengths best absorbed by BrC (Alexander et al., 2008). Previous studies were also unable to conclusively identify BrC over the Southeast Atlantic through optical methods (Denjean et al., 2020; Taylor et al., 2020; Wu et al., 2020; Dobracki et al., 2023; Dobracki et al., 2025), although Zhang et al. (2022) noted that electron microscopy yields greater BrC contributions than optical methods. Since there does not appear to be a significant shift in aerosol type during the BBA emission season, such as the processes described by Bond et al. (2013), we posit that the aerosol brightening is instead due to changes in aerosol composition (Eck et al., 2013).

Also softened our conclusions regarding BrC contributions in Lines 462-467:

EAE has little subseasonal variation, indicating that fine aerosols dominate throughout the season. The AAE values remain within the typical range for BC during the season. If the aerosol brightening in October were caused by an increase in BrC contributions, then we would expect to see an increase in AAE. However, we instead see a decrease in AAE, so there is no clear optical evidence for substantial BrC contributions within the retrieved wavelength range, in alignment with previous studies. This suggests that the aerosol brightening is due to changes in aerosol composition during the season, rather than a change in aerosol type, which had been the subject of ongoing debate.

Referee Report #1.2:

2. Lines 212–218: please clarify how the reduced independent sample size was implemented in the Wilcoxon tests. A Wilcoxon rank-sum test normally receives the observations themselves rather than an independently specified effective sample size. Did the authors subsample the 1 Hz data at 10 s intervals, average the data into non-overlapping 10 s blocks, or adjust the significance calculation using an effective sample size? This should be stated explicitly and applied consistently. In addition, the rank-sum test does not generally test equality of medians unless the distributions have comparable shapes; the parenthetical definition in Line 212 should therefore be revised.

Author's Response #1.2:

We subsampled the 1 Hz data at a 10 s interval to yield the desired independent sample size for input into the Wilcoxon rank-sum test, so we have clarified that in the text. We have also revised the Wilcoxon rank-sum test definition so that it specifies that the distributions have comparable shapes for median equality testing.

Manuscript Changes #1.2:

Revised the parenthetical definition in Lines 222-223:

(which determines the equality of medians for samples from continuous distributions of comparable shapes)

Clarified how in situ SSA was reduced to the independent sample size in Line 223:

(reduced to the independent sample size by subsampling every 10 s after the first value)

Referee Report #1.3:

3. Lines 219 and 295–296: distinguish increased SSA from increased absolute scattering.
Line 219 interprets increasing SSA as "an increase in aerosol scattering," whereas Lines 295–296 conclude that the change is primarily caused by decreasing absorptivity rather than increased scattering. Since SSA is a relative quantity, an increase in SSA alone does not demonstrate an increase in the scattering coefficient. Please use wording such as "an increase in scattering relative to extinction" and keep this interpretation consistent throughout the manuscript and abstract.

Author's Response #1.3:

We have clarified that the increase in scattering is relative to total extinction and ensured that it is maintained throughout the manuscript.

Manuscript Changes #1.3:

Added "relative to total extinction" to Lines 19, 230, 393, and 490.

Referee Report #1.4:

4. Lines 62–70: retain a more explicit interannual limitation. The added literature supports relatively modest interannual variability, but a regional burned-area coefficient of variation below 0.5 does not by itself demonstrate that interannual effects are negligible for the sampled air masses.

Author's Response #1.4:

We have expanded our discussion to include above-cloud AOD within the smoke plume, which does show greater interannual variability than biomass burning fire location or Southeast Atlantic meteorology. As such, we have also made the interannual variability limitations more explicit.

Manuscript Changes #1.4:

Further expanded our interannual variability discussion and retained a more explicit limitation in Lines 75-79:

A type of interannual variability that is less constrained is above-cloud Aerosol Optical Depth (AOD), with the August 2017 plume being moderately weaker, the September 2016 plume being slightly weaker, and the October 2018 plume being slightly stronger than climatological averages (Redemann et al., 2021). As such, the results of this study are dependent upon the differing strengths of the smoke plume observed during each ORACLES campaign year.

Editor Decision #2.1:

Dear Mitchell et al.,
many thanks for submitting your manuscript to ACP and responding to the concerns of two anonymous referees. The paper is now substantially improved with respect to the previous version. I encourage you to address a few minor issues raised by Anonymous Referee #1.
In addition, please also address some major issues I outline below.
Kind regards,
Marco Gaetani

Abstract: please add a few lines at the beginning to add some context to your research, and a few lines at the end to highlight the importance and implications of the results. You can refer to the ACP Guidelines for authors (https://www.atmospheric-chemistry-and-physics.net/policies/guidelines_for_authors.html) to improve the abstract.

Author's Response #2.1:

Thank you for recommending ways that we can improve the abstract. We have rewritten the abstract to better align with ACP guidelines, adding to the background context and implications of the research.

Manuscript Changes #2.1:

Rewrote the abstract in Lines 13-28:

Biomass Burning Aerosols (BBA) are a source of climate uncertainty over the Southeast Atlantic. Following emission in Southern Africa, the BBA smoke plume overlies a stratocumulus cloud deck, resulting in aerosol-cloud-radiation interactions that are difficult to observe. During 2016-2018, NASA conducted the ORACLES (ObseRvations of Aerosols above CLouds and their intEractionS) airborne campaigns to constrain BBA uncertainty, employing 4STAR (Spectrometer for Sky-Scanning, Sun-Tracking Atmospheric Research) to retrieve aerosol properties. Here, we use all three months of ORACLES 4STAR sky-scans to investigate the subseasonal variability of BBA radiative properties. Changes in Single Scattering Albedo (SSA) indicate increased scattering relative to extinction as the BBA emission season progresses. We attribute this aerosol brightening to changes in composition, rather than aerosol type, as our Absorption Ångström Exponent (AAE) analysis provides no evidence for Brown Carbon (BrC) contributions. SSA from our 4STAR sky-scans is compared against a 30-year climatology from 31 AERONET (AErosol RObotic NETwork) stations in Southern Africa, showing consistent changes in aerosol scattering across land and sea. ORACLES in situ SSA is also examined spatially. Our latitudinal analysis indicates that the September decrease in SSA from 4STAR is affected by the southerly sampling of ORACLES 2016 compared to the other two campaigns. Westward gradual increases and sharper decreases in SSA are attributed to late-transport aging processes identified by recent studies. These processes start further eastward in October, in conjunction with the southeastward shift in fires. These subseasonal and spatial changes in BBA radiative properties have implications for aerosol modelling and satellite validation.

Editor Decision #2.2:

Conclusions: this section should also be improved, in particular regarding the comparison and context of your results with respect the existing knowledge and the implications of your findings for our understanding of the state and/or behaviour of the atmosphere and climate. You can refer to the ACP Guidelines for authors to improve the section.

Author's Response #2.2:

Thank you for suggesting ways for us to improve the conclusions. We have increased discussion regarding our agreement with previous findings and added to the implications of our findings for our understanding of the atmosphere.

Manuscript Changes #2.2:

Added more context relating our results to previous studies in Line 466:

in alignment with previous studies.

And in Line 467:

which had been the subject of ongoing debate.

And in Line 498:

noted by previous studies,

Increased discussion regarding the implications of our findings for our understanding of the atmosphere in Lines 504-508:

A subseasonal SSA increase of about 0.05 is significant enough to affect physical processes involving DARE, heating rates, and atmospheric stability. As such, this subseasonal variability needs to be properly accounted for in climate models. The airborne 4STAR sky-scans are an invaluable tool for validating above-cloud aerosol products from satellites and other methods, which will in turn reduce aerosol uncertainties within this critical study region.

Editor Decision #2.3:

Minor: Figure S4 is not mentioned in the manuscript.

Author's Response #2.3:

We have added a reference to Figure S4.

Manuscript Changes #2.3:

Added a reference to Figure S4 in Lines 291-294:

The relationships between AAE, EAE, and SSA are shown per 4STAR sky-scan as scatterplots in Fig. S4, displaying a shift away from the greater AAE values expected with increased BrC contributions as the BBA emission season progresses, for the retrieved wavelength range.