Referee Comment #1.1:

This study investigates the subseasonal and spatial variability of biomass burning aerosol (BBA) radiative properties over the Southeast Atlantic (SEA). By utilizing airborne 4STAR measurements from the NASA ORACLES campaigns (2016-2018) alongside long-term AERONET climatological data, the manuscript provides valuable insights into the evolution of Single Scattering Albedo (SSA), Aerosol Optical Depth (AOD), and Absorption Aerosol Optical Depth (AAOD). The findings regarding the "aerosol brightening" effect as the burning season progresses are particularly interesting.

Overall, the paper is well-structured, and the dataset is highly valuable for improving aerosol-radiation interactions in climate models. However, there are several areas where the analysis requires deeper attribution, particularly regarding interannual variability, aerosol composition, and the justification of certain methodological constraints. I recommend the manuscript for publication after addressing the following major revisions.

Major Comments:

1. The data representing the three different months in this study (August, September, and October) were collected across three completely different years (2017, 2016, and 2018, respectively). Consequently, the deduced subseasonal trends (e.g., from August to October) might be heavily influenced or obscured by the interannual characteristics of aerosol emissions and meteorological conditions. The authors need to explicitly state this limitation in the text and discuss how interannual emission variations might impact their subseasonal conclusions.

Author's Response #1.1:

Thank you for raising the issue of interannual variability. We agree that better addressing the interannual variability will strengthen our conclusions regarding the subseasonal variability and improve our manuscript. In alignment with your comment and that of RC2, we have expanded the discussion (Lines 62-70) regarding the interannual variability of Southern African aerosol emissions and meteorological conditions over the Southeast Atlantic, with literature suggesting that it is small enough in both cases to not significantly affect our results or major conclusions.

Manuscript Changes #1.1:

Discussion of interannual variability was added to Lines 62-70:

There is remarkably little interannual variability in Southern African fires due to the consistency of agricultural burning practices in the region (van der Werf et al., 2010; Redemann et al., 2021). Using the 18-year (2001-2018) European Space Agency Fire Climate Change Initiative, version 5.1 (FireCCI51) dataset derived from MODIS aboard the NASA Terra satellite, Chuvieco et al. (2021) found that Southern Africa had the smallest interannual variability in burned area in the world, with a coefficient of variation < 0.5 across the region. Ryoo et al. (2021) documented the

interannual variability in meteorological conditions over the Southeast Atlantic, finding that wind velocity, relative humidity, and precipitation were near climatological averages for August 2017, September 2016, and October 2018. They also found that despite sea surface temperatures during the ORACLES campaign months being 0.2 – 0.4 K above climatological averages, they did not have a systematic effect on low-level cloud fraction.

Referee Comment #1.2:

2. The current discussion regarding the characteristics of SSA, AOD, and AAOD in Sections 3.1 and 3.2 is somewhat descriptive and one-sided. Could the authors provide a more in-depth attribution linking these optical properties to the proportional changes and concentrations of aerosol compositions during the biomass burning period? It is highly recommended to acquire observational data or reanalysis datasets (such as MERRA-2) on aerosol composition and concentration, and add these to the supplementary materials. This would intuitively demonstrate how variations in aerosol composition and concentration drive the observed changes in SSA, AOD, etc.

Author's Response #1.2:

Thank you for suggesting an additional way that we can tie the subseasonal variability of aerosol radiative properties to changes in aerosol composition and concentration. In alignment with your comment, we have created maps of BC and OC columnar mass densities from MERRA-2 reanalysis (Fig. S1). The BC:OC ratio remains near 1:10 throughout the ORACLES campaign months, which is consistent with our finding that the subseasonal increase in SSA is not attributable to an increase in BrC concentration. August has the greatest BC and OC densities, although the maximums are close together and located over land. By September, higher BC and OC densities are more widespread across the Southeast Atlantic, followed by decreases in October, aligning with the September peak in AOD and AAOD noted by 4STAR. The BC and OC densities also shift southeastward over the course of the BBA emission season.

Manuscript Changes #1.2:

Added MERRA-2 reanalysis maps via Supplementary Figure 1.

Discussion of MERRA-2 reanalysis added to Lines 269-275:

This is supported 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, indicating little change in BrC concentration. August has the greatest BC and OC densities, although the maximums are close together and located over land. By September, higher BC and OC densities are more widespread across the Southeast Atlantic, followed by decreases in October, aligning with the September peak in AOD and AAOD noted by 4STAR.

And added to Lines 280-281:

A similar southeastward shift over the course of the BBA emission season is seen in the BC and OC columnar mass densities (Fig. S1),

Referee Comment #1.3:

3. To further support the explanations for the variations in parameters like SSA and AOD, the authors should consider adding statistical charts or maps that represent the intensity of biomass burning (e.g., Fire Radiative Power (FRP) or fire counts) to the supplementary materials. Illustrating the spatial and temporal variation characteristics of the emission sources would greatly assist in interpreting the downstream aerosol properties.

Author's Response #1.3:

In alignment with your comment, we have created maps of Fire Radiative Power (FRP) from Terra and Aqua MODIS thermal anomalies (Fig. S2). Fire counts peak in August and decrease over the course of the BBA emission season, with far fewer fires in October, aligning with the October decrease in AOD. The fires shift southeastward over the season, which is also visible from the movement in the FRP-weighted mean coordinates, aligning with our hypothesis that the geographic shift in fires contributes to the October increase in SSA.

Manuscript Changes #1.3:

Added MODIS-detected fire maps via Supplementary Figure 2.

Discussion of MODIS-detected fire maps added to Lines 280-281:

A similar southeastward shift over the course of the BBA emission season is seen in the BC and OC columnar mass densities (Fig. S1), as well as in MODIS-detected vegetation fires (Fig. S2),

Referee Comment #1.4:

4. The manuscript demonstrates that the lower SSA observed by 4STAR in 2016 (September) is largely due to the campaign base being in Namibia, meaning the sampling region was further south and did not fully capture the central smoke plume near the equator. This implies that the subseasonal trend shown in Figures 2 and 3 (the drop in September and massive rise in October) is largely driven by a spatial sampling bias rather than pure temporal evolution. This spatial caveat should be stated more explicitly in the Abstract and Conclusions to avoid misleading readers who might interpret this purely as a temporal shift.

Author's Response #1.4:

We agree that the more southerly location of ORACLES 2016 affected our results, which was the primary focus of the latitudinal analysis in our discussion section. We have further reiterated its effects in our abstract (Lines 23-24) and conclusions (Lines 469-473).

Manuscript Changes #1.4:

Discussion of the southerly effect of the ORACLES campaign was added to the abstract (Lines 23-24):

The latitudinal analysis indicates that the September decrease in SSA noted by 4STAR is affected by the southerly sampling location of ORACLES 2016 compared to the other two campaigns.

And clarified in the conclusions (Lines 469-473):

A latitudinal analysis of in situ measurements and subsampling of 4STAR sky-scans indicates that the September decrease in SSA noted by 4STAR is affected by the more southerly sampling location of ORACLES 2016 relative to the other two campaigns, rather than from a temporal evolution, such that ORACLES 2016 would have observed relatively greater scattering if it had similarly targeted the central smoke plume from the equator, bringing the 4STAR SSA medians more in line with the subseasonal increase observed by AERONET.

Referee Comment #1.5:

5. The study restricts the 4STAR sky scans to altitudes of < 3 km. Is the selection of this specific boundary reasonable and fully justified? Is it possible that a significant portion of the biomass burning aerosols is transported to altitudes above 3 km in this region? A brief justification or a reference regarding the vertical distribution of the aerosol plume during the campaign should be added to clarify why 3 km is an appropriate cutoff.

Author's Response #1.5:

The altitudinal restriction requires that the 4STAR sky-scans occurred below 3 km, but it does not remove any measurements above that altitude. The 4STAR sky-scans are total columnar measurements, so all above-aircraft values are included. The altitudinal restriction was selected to best ensure that the 4STAR sky-scans have a full columnar view below the smoke plume (Mitchell et al., 2025) and is the same criterion used by Pistone et al. (2019).

Manuscript Changes #1.5:

The 4STAR altitude restriction was clarified in Lines 147-149:

For this study, we additionally required that all sky-scans were taken from flight altitudes below 3 km, as to best ensure the full column of the smoke plume was observed.

Referee Comment #1.6:

6. In Lines 427-430, the authors mention the potential to extrapolate 4STAR data to the UV band. Given this possibility, why wasn't this step performed in the current study? Relying solely on visible and near-infrared Absorption Angstrom Exponent (AAE) to completely exclude the role of Brown Carbon (BrC) might not be rigorous enough. Combining optical parameters in the UV band with the AAE numerical characteristics would provide a much more convincing argument when explaining the statistical features and relative contributions of Black Carbon (BC) versus BrC. If there are technical limitations preventing this extrapolation currently, please explain them explicitly and slightly soften the conclusions regarding the absence of BrC.

Author's Response #1.6:

Expansion into the UV is complicated by the presence of an instrument artifact near 420 nm and stray light scattering at 360 and 380 nm. In alignment with your comment and that of RC2, we have increased discussion of the technical limitations and softened our conclusions regarding BrC (Lines 449-455).

Manuscript Changes #1.6:

The technical limitations for 4STAR expansion into the UV spectrum was clarified in Lines 449-455:

EAE and AAE have little subseasonal variation, indicating domination by fine BC aerosols throughout the season, suggesting that the aerosol brightening is due to a change in aerosol composition through the season, rather than a change in aerosol type, although 4STAR did not directly observe the UV wavelengths at which BrC absorption is the strongest. Improving 4STAR's ability to identify BrC absorption via expansion into the UV spectrum is currently complicated by an instrument artifact near 420 nm and stray light scattering at 360 and 380 nm (Mitchell et al., 2025), but could be overcome with the use of hyperspectral GRASP (Generalized Retrieval of Atmosphere and Surface Properties) code (Román et al., 2018), which is the subject of ongoing work.

Referee Comment #1.7:

Minor Comments:

Lines 165-166: The authors mention using a 10-second rolling average for in situ data and dividing the total observations by 10 to estimate the number of independent samples. While statistically reasonable, please briefly state the typical cruising speed of the aircraft and translate this 10-second window into an approximate spatial resolution (in kilometers). This will help readers understand the spatial scale of an "independent sample."

Author's Response #1.7:

Thank you for recommending a way that we can illustrate the spatial scale of an independent sample of our in situ measurements.

Manuscript Changes #1.7:

The spatial resolution of an in situ independent sample was added to Lines 180-182:

Given that the typical ORACLES sampling speed of the NASA P-3 Orion aircraft was ~382 km per hour, this results in a spatial resolution of about 1.1 km per independent sample.

Referee Comment #1.8:

Lines 196-202： When utilizing the two-sample Wilcoxon rank sum test, strong autocorrelation in continuous flight data can artificially lower p-values (overestimating significance). Please explicitly confirm in the text that the sample size ( ) input into these statistical tests is the reduced independent sample size (divided by 10), rather than the raw number of data points.

Author's Response #1.8:

It is correct that we are using the reduced independent sample sizes for the statistical tests, so we have reaffirmed that in the text.

Manuscript Changes #1.8:

A reference to the independent sample size was added to Line 213:

(reduced to the independent sample size)

Referee Comment #1.9:

Lines 245-248：The authors cite several papers that similarly found a lack of BrC. Since the presence and transport of BrC in this region can be a debated topic, it would provide a more balanced literature review to briefly mention 1-2 studies that have identified BrC contributions in the SEA or its source regions (if applicable), before explaining why this study's findings differ.

Author's Response #1.9:

We agree that discussing an article that identified greater BrC contributions over the Southeast Atlantic would further strengthen our manuscript, so we have added discussion regarding Zhang et al. (2022) to the introduction (Lines 85-89) and results (Lines 264-266).

Manuscript Changes #1.9:

Discussion of Zhang et al. (2022), which found greater BrC contributions over the Southeast Atlantic, was added to the introduction (Lines 85-89):

Using TEM-EDX (Transmission Electron Microscopy - Energy Dispersive X-ray spectroscopy) and an assumed BC refractive index, Zhang et al. (2022) determined that BrC could contribute 8 – 22 % of total aerosol absorption at 470 nm. However, their BrC estimate is much lower using the AAE attribution method, at only 2 – 6 % of total aerosol absorption at 470 nm, indicating that spectroscopy could reveal a greater BrC presence over the Southeast Atlantic than suggested by optical methods.

And added to the results (Lines 264-266):

The lack of BrC contributions from our AAE analysis also aligns with the findings of Zhang et al. (2022), although their results indicate that electron microscopy would yield greater BrC contributions than optical methods.

Referee Comment #2.1:

The manuscript documents the subseasonal and spatial variability of biomass burning (BB) aerosols properties over the Southeast Atlantic (SEA) region during the NASA ORACLES campaigns (2016-2018). It combines airborne 4STAR and long-term AERONET measurements of single scattering albedo (SSA), aerosol optical depth (AOD), and absorption aerosol optical depth (AAOD). The results investigate the distinction between BC and BrC during the biomass burning season.

My recommendation is that the manuscript can be published after major revisions. The dataset and the manuscript concept are strong, but several central methodological choices are not yet documented or stress-tested enough to support some of the stronger interpretations.

Major Comments:

While the manuscript effectively identifies subseasonal and spatial trends, it lacks atmospheric trajectory modeling (e.g., HYSPLIT or FLEXPART) to confirm the specific origins of the aerosols. I suggest including an analysis of back-trajectories for the study periods, this would provide vital context for the transport pathways, provide a more robust validation of the proposed source fire shifts, and strengthen the manuscript.

Author's Response #2.1:

Thank you for suggesting an additional way that we can tie the subseasonal variability of aerosol radiative properties to changes in emissions. To address your comment, we have created maps of HYSPLIT back-trajectories (Fig. S3) for ORACLES research flight dates near the beginning, middle, and end of each campaign. HYSPLIT back-trajectories suggest aerosol transport from northern Angola in August, shifting southward from southern Angola in September, and even further southeastward from Botswana, Zambia, and Zimbabwe in October. This aligns with our hypothesis that the geographic shift in fires contributes to the subseasonal aerosol brightening noted in October.

Manuscript Changes #2.1:

Added HYSPLIT back-trajectory maps via Supplementary Figure 3.

Discussion of HYSPLIT back-trajectory maps added to Lines 280-282:

A similar southeastward shift over the course of the BBA emission season is seen in the BC and OC columnar mass densities (Fig. S1), as well as in MODIS-detected vegetation fires (Fig. S2), and in HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) back-trajectories (Fig. S3).

Referee Comment #2.2:

The study covers three months (August–October) over a three-year period. I recommend adding a discussion on how interannual differences in meteorological conditions might have affected the finding.

Author's Response #2.2:

Thank you for raising the issue of interannual variability. We agree that better addressing the interannual variability will strengthen our conclusions regarding the subseasonal variability and improve our manuscript. In alignment with your comment and that of RC1, we have increased discussion (Lines 62-70) regarding the interannual variability of Southern African aerosol emissions and meteorological conditions over the Southeast Atlantic, with literature suggesting that it is small enough in both cases to not significantly affect our results or major conclusions.

Manuscript Changes #2.2:

Discussion of interannual variability was added to Lines 62-70:

There is remarkably little interannual variability in Southern African fires due to the consistency of agricultural burning practices in the region (van der Werf et al., 2010; Redemann et al., 2021). Using the 18-year (2001-2018) European Space Agency Fire Climate Change Initiative, version 5.1 (FireCCI51) dataset derived from MODIS aboard the NASA Terra satellite, Chuvieco et al. (2021) found that Southern Africa had the smallest interannual variability in burned area in the world, with a coefficient of variation < 0.5 across the region. Ryoo et al. (2021) documented the interannual variability in meteorological conditions over the Southeast Atlantic, finding that wind velocity, relative humidity, and precipitation were near climatological averages for August 2017, September 2016, and October 2018. They also found that despite sea surface temperatures during the ORACLES campaign months being 0.2 – 0.4 K above climatological averages, it did not have a systematic effect on low-level cloud fraction.

Referee Comment #2.3:

Lines 255–258: The authors attribute regional differences in SSA to a shift in source fires toward Botswana, Zimbabwe, and Mozambique, referencing trajectory analyses from Dobracki et al. (2023). However, Dobracki et al. primarily analyzed data from 2016. Given that this manuscript includes ORACLES data from 2017 and 2018, it is unclear whether this source shift is consistent across the entire study period. I recommend that the authors either demonstrate that their own data (2017–2018) exhibits similar transport patterns or explicitly discuss the interannual consistency of this source fire shift.

To better support the claims regarding the aerosol brightening process and the distinction between BrC and BC, I suggest adding a scatter plot showing the relationship between AAE and SSA (or another identification space). This would allow the reader to see if the aerosol population shifts from a cluster associated with BrC absorption toward a less absorbing regime over time.

Author's Response #2.3:

Dobracki et al. (2023) indeed primarily focused on back-trajectories from 2016, so we hope that the HYSPLIT back-trajectories from all three ORACLES campaigns that we added via Fig. S3 are more illustrative of the subseasonal shift in aerosol emissions, with Chuvieco et al. (2021) indicating that there is little interannual variability in source fires.

Thank you for suggesting an additional way that we can visualize the relationship between different aerosol radiative properties. In response to your comment, we have created scatterplots showing the relationships between AAE, EAE, and SSA over time (Fig. S4). We would expect an increase in AAE if the change in SSA was due to increasing BrC concentration, but instead we see a decrease in AAE over the course of the BBA emission season, displaying a population shift even further away from BrC absorption, supporting our hypothesis that the aerosol brightening is not due to a change in aerosol type.

Manuscript Changes #2.3:

Added scatterplots linking AAE, EAE, and SSA via Supplementary Figure 4.

Referee Comment #2.4:

Lines 427–429: The authors note that expanding the 4STAR dataset into the UV spectrum using the hyperspectral GRASP code could better isolate BrC absorption. Given that the manuscript's primary focus is the aerosol brightening process and the distinction between BrC and BC, this would be a great addition. I recommend that the authors either incorporate these results or provide a more detailed justification for why this methodology was not applied in the conclusions.

Author's Response #2.4:

Expansion into the UV is complicated by the presence of an instrument artifact near 420 nm and stray light scattering at 360 and 380 nm. In alignment with your comment and that of RC1, we have increased discussion of the technical limitations (Lines 449-455).

Manuscript Changes #2.4:

The technical limitations for 4STAR expansion into the UV spectrum were clarified in Lines 449-455:

EAE and AAE have little subseasonal variation, indicating domination by fine BC aerosols throughout the season, suggesting that the aerosol brightening is due to a change in aerosol composition through the season, rather than a change in aerosol type, although 4STAR did not directly observe the UV wavelengths at which BrC absorption is the strongest. Improving 4STAR's ability to identify BrC absorption via expansion into the UV spectrum is currently complicated by an instrument artifact near 420 nm and stray light scattering at 360 and 380 nm (Mitchell et al., 2025), but could be overcome with the use of hyperspectral GRASP (Generalized Retrieval of Atmosphere and Surface Properties) code (Román et al., 2018), which is the subject of ongoing work.

Referee Comment #2.5:

Minor Comments:

Line 19: First use of acronym “AERONET” is here so add "AErosol RObotic NETwork".

Lines 112-113: SSA, AOD, AAAOD, EAE, AAE are already stated in the introduction so you can use freely the acronym.

Line 146: First use of acronym “PSAP”, so you need to define the acronym.

Author’s Response #2.5:

We have corrected the acronym introduction errors.

Manuscript Changes #2.5:

AERONET is now expanded at first use in Lines 19-20, with the repeated expansions of SSA, AOD, AAOD, EAE, and AAE in Line 123 removed, and the PSAP and Neph definitions added to Line 157:

PSAP (Particle Soot Absorption Photometer) & Neph (Nephelometer)

Referee Comment #2.6:

Line 146: You mention that both PSAP & Neph measure in the same wavelengths, consider correcting the table caption (Since in the table you mention a difference of 20 and 40 nm between two instruments).

Author's Response #2.6:

The table is correct, as the PSAP and Neph do have slightly different RGB reporting wavelengths. We have corrected the table caption (Lines 157-159) such that the observation counts are for SSA calculated from paired PSAP absorption and Neph scattering coefficients, following the interpolation of the scattering coefficients from the Neph wavelengths (450, 550, 700 nm) to the PSAP wavelengths (470, 530, 660 nm).

Manuscript Changes #2.6:

The wavelengths of the paired PSAP and Neph SSA values are clarified in Lines 157-159:

The PSAP (Particle Soot Absorption Photometer) & Neph (Nephelometer) SSA observation counts are following the interpolation of scattering coefficients from the Neph wavelengths (450, 550, 700 nm) to the PSAP wavelengths (470, 530, 660 nm). The given PSAP & Neph observation count is for 530 nm, with slightly more measurements at 470 nm, and slightly fewer at 660 nm.