We would like to sincerely thank the editor and reviewer for their time, effort, and thoughtful feedback on our manuscript. The reviewer comments are shown in black, with the author responses shown in blue and any edited manuscript language shown in *italicized blue font*.

General Comments

1. Marine background environments over the Ascension Island have been considered when presenting chemical compositions such as $\Delta SO4$. However, the marine background environments should be given more consideration when presenting the BBAs results such as FrBC and SSA.

    - In this manuscript, the FrBC in the BBA was "calculated by dividing the number concentrations of rBC-containing particles measured by the SP2 by the total particle number concentration measured by the CPC" (lines 211-212). In the MBL over Ascension Island, the FrBC during BB-impacted periods is not only related to BBAs, but also likely affected by marine emissions. The concentration levels between background marine aerosols and mixed BBAs, as well as the mixing state (internally or externally) between BBAs and marine aerosols will affect the FrBC values.

    - Similar to FrBC, the SSA given in this manuscript is also likely affected by such as marine sulfates in the MBL.

The marine backgrounds may have small effects on these BBA properties, and the temporal trends shown in this manuscript may still depend on primarily difference in source conditions and transport processes. However, including the aforementioned details would provide a more comprehensive picture of BBAs in the MBL.

The reviewer brings up a good point here. The aerosol size distributions in Figure 11 nicely demonstrate how the accumulation mode number concentration differs between clean and polluted time periods. We have added additional language to inform the reader that background aerosol concentrations in the accumulation mode are a fraction of those during the biomass burning plume events. We are only focusing on accumulation mode aerosols here because the SP2 cannot sample particles < 80 nm. SSA_{530} values are also not likely affected by the small contribution of background aerosol particles in either the accumulation or Aitken mode during the plume events. We have added additional language to Section 4.5. The mean SSA_{530} values during clean events was 0.98±0.01 and highlight how the BBA substantially lowers the SSA.

We have added the following text to Section 2.2, *"The FrBC values are not likely influenced by the background aerosol emissions because aerosol number concentrations in the accumulation mode during these clean periods is only a fraction of the BBA aerosol number concentrations sampled during plume events. We further highlight these differences in aerosol number concentration between the clean and polluted periods in Sect. 3.2.3."*

We have added the following text to Section 4.5, *"This result is surprising, as the mean background* SSA_{530} *value was 0.98±0.01, and mixing with clean marine air should increase* SSA_{530} *in the MBL. Previous studies, such as Wu et al. (2020), concluded that BBA in the MBL is* *less absorbing due to this mixing. In contrast, our results demonstrate that BBA in the MBL is more absorbing than that in the FT."*

2. In Sect 4.1, 4.4 and 5.1, the term of "SO4:rBC" is used for discussions. However, based on descriptions in Sect 3, the term of SO4:rBC should be changed to Δ*SO4:rBC* (refer to above-background ΔSO4 mass concentrations). For example, in lines 804, 809, 816 etc.

The text in Sections 4.1, 4.4. and 5.1 where SO_4 and SO_4:rBC are referenced has been corrected to ΔSO_4 and ΔSO4:rBC throughout. Thank you for catching this typo.

3. In Sect. 4.1, the authors state that (lines 824-825) "high OA:rBC, large rBC coating-to-core mass ratios, and high SO4:rBC would suggest that the fires were inefficient, *while the large values of rBCgpd and rBCmpd and high FrBC would indicate that the fires were efficient*". This assumes that efficient burning is related to larger BC core sizes. However, in the second paragraph of Sect. 4.2, the authors discuss that "the relationship between BC core size and source burning conditions are not sure from this study and previous studies".

These present conflicting clarifications to some extent. I suggest rephrasing this part.

We have adjusted the text in Section 4.1 to exclude any discussion of the rBC size distributions. The reviewer makes a good point that the previous language was confusing and conflicted with Section 4.2. We kept Section 4.2 as is because it provides a more thorough discussion on the rBC size distributions.

4. In Sect. 4 (line 755-757), the authors state that "Lastly, we examine the dependence of SSA530 on the chemical properties of the BBA and explain why the BBA in the MBL has a lower SSA530 than that in the FT", and further explanations follows in Sect. 4.5. I agree that the authors provide a detailed explanation on the relationship between SSA530 and OA:rBC, combining the LASIC MBL and ORACLES FT. However, more details should be provided regarding the discussion of low SSA530 in the Ascension Island MBL and the combination of different datasets:
   - In line 998, the authors state that "The SSA530 values in the Ascension Island MBL even lower than those in the nearby FT by about 0.07". However, it is not clearly demonstrated that what this "nearby FT" data is here. Does this "nearby FT data" refer to ORACLES FT? I suggest clarifying it clearer here.

We have adjusted the following text in Section 4.5 to clarify what region we are referring to. *"These values are even lower (by about 0.07) than those sampled in the FT between the African plateau and Ascension Island during the September ORACLES 2016 campaign."*

   - Following the above, the authors state that "This result is surprising, as mixing with clean marine air should increase SSA530 in the MBL all else equal (Wu et al., 2020)".

Wu et al., 2020 conclude that the higher SSA values in the MBL are due to the BBA mixing with the clean marine air, especially near the surface layer. The results presented here, show that during biomass burning events, the BBA dominates the aerosol optical properties and not submicron sea salt.

From Comment 1 above: We have added the following text to Section 4.5, *"This result is surprising, as the mean background SSA_{530} value was 0.98±0.01, and mixing with clean marine air should increase SSA_{530} in the MBL. Previous studies, such as Wu et al. (2020), concluded that BBA in the MBL is less absorbing due to this mixing. In contrast, our results demonstrate that BBA in the MBL is more absorbing than that in the FT."*

In Barrett et al., (2022), LASIC ARM observed much less scatterings than CLARIFY FAAM during intercomparison flight legs, while the absorptions measurements were relatively consistent. This discrepancy in scattering measurements is attributed to the differences in the upper cut sizes of the impactors. LASIC scattering measurements are likely less affected by submicron coarse mode particles from marine sea sprays, due to its lower cut size (0.78 µm) compared to CLARIFY (1.0 µm). If the authors compare the LASIC-MBL/ORACLES-FT SSA trends to CLARIFY-MBL/FT SSA trends, the inconsistency in scattering measurements should be considered.

- Is there any consistency or inconsistency in optical/chemical measurements between the LASIC and ORACLES platforms, if the authors combined two dataset to establish the correlation between SSA530 and OA:rBC? E.g.

The aerosol optical instruments deployed during LASIC were behind a 1µm impactor (Uin et al., 2020) which is identical to the CLARIFY 1 µm cut off. The SSA_{530} measurements calculated in the ARM VAP (from the PSAP and the nephelometer) compare well to those from the $CAPS_{SSA}$ monitor that was also deployed to Ascension Island (Fig. A2). The consistency between the ARM VAP SSA_{530} values and the $CAPS_{SSA}$ values when BBA is present provides confidence that the scattering and absorption coefficients measured at Ascension Island during LASIC are trustworthy. However, we cannot explain the discrepancies in the scattering coefficient between the LASIC and CLARIFY described in Barrett et al. (2022).

The submicron SSA_{530} calculations from ORACLES were performed with the same set of corrections (Pistone et al., 2019; Dobracki et al., 2023) that were used in the ARM VAP (Flynn et al., 2020; Kassianov et al., 2023). The ORACLES campaign also deployed the same models of the PSAP (Radiance Research) and Nephelometer (TSI 3563). Therefore, we expect consistency between the ORACLES and LASIC optical property measurements.

We have added the following text to Section 2.1, *"The aerosol optical properties measured in this study were also compared to those from the ORACLES and CLARIFY campaigns. During ORACLES, the aerosol optical properties were measured using the same instruments as in the LASIC campaign. Specifically, scattering coefficients at wavelengths of 450, 550, and 700 nm were measured using the TSI 3563 nephelometer, while absorption coefficients at 465, 530, and 650 nm were measured with the PSAP (Radiance Research) (Dobracki et al., 2023). Corrections for the scattering coefficients were applied using both Virkkula and Bond & Ogren methods (Pistone et al., 2019; Dobracki et al., 2023). In contrast, during the CLARIFY campaign, absorption coefficients at 405, 515, and 660 nm, along with aerosol dry extinction at 405 and 658 nm, were measured using the EXtinction SCattering and Absorption of Light for Airborne Aerosol Research (EXSCALABAR) (Wu et al., 2020; Barrett et al., 2022). Despite these* *differences in measurement techniques, we aim to utilize the data from both campaigns for comparative analysis.”*

Regarding the chemical properties, there were slight differences between the mass spectrometer and the SP2 models. The ACSM at Ascension Island is the lower resolution model of the Aerosol Mass Spectrometer (AMS) that was used during ORACLES. However, because we are using the bulk chemical composition and comparing the OA:rBC ratios (not absolute concentrations of OA μg m^{-3}), there should be no major inconsistencies due to collection methods or instrument abilities. The ACSM and AMS data were both corrected with the composition dependence collection efficiency (Dobracki et al., 2023).

The SP2 at Ascension Island was the 8-channel model from DMT, whereas the SP2 on ORACLES was the 4-channel model from DMT. The advantage of the 8-channel model is that it can provide the coating thickness. Aside from that feature, the rBC mass measurements from both LASIC and ORACLES should be consistent as the data was collected and processed by Arthur J. Sedlacek (BNL).

We have added the following text to Section 2.1, *“Non-refractory aerosols during the ORACLES 2016 campaign were sampled using a High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS, Aerodyne Inc.)(Dobracki et al., 2023). During the CLARIFY campaign, the same type of aerosol was sampled with a Compact Time-of-Flight Aerosol Mass Spectrometer (C-ToF-AMS) (Taylor et al., 2020; Barrett et al., 2022). rBC sampled during the ORACLES 2016 campaign and CLARIFY campaign was measured with a four-channel Single Particle Soot Photometer (SP2, DMT) (Barrett et al., 2022; Sedlacek et al., 2022). Despite these slight differences in instrumentation and sampling strategies, we utilize the data from both campaigns in this study to ensure a comprehensive analysis.”*

5. The uncertainties of calculated values from different instruments, such as SSA, should be provided in this manuscript.

We have added a column to Table 2 listing the instrument or calculation uncertainty when applicable.

6. When getting to Ascension Island, the clouds tend to be decoupled from the surface mixed layer (e.g. Abel et al., 2020), and the upper part of the MBL is relatively separated to the lower surface layer. Under this condition, surface layer measurements from LASIC may not directly represent BBAs properties below the cloud. This should be taken into account when interpreting the implications of LASIC datasets.

This is a fair point, however the aircraft measurements bias the conditions. Zhang and Zuidema (2019) report that the clouds at Ascension Island are coupled with surface during the early morning and evening, especially in August. The decoupling typically occurred during the day, when the aircrafts sampled near Ascension Island. It would be more appropriate to say that the clouds and the surface were intermittently coupled. We do agree that additional language should be added to Section 2.1. We have added the following text, *“While the LASIC data are constrained to surface based measurements, they are predominantly representative of the* *boundary layer surrounding Ascension Island. It is crucial to consider that the clouds intermittently coupled with the surface during the early morning and evening. The observed intermittent coupling of clouds with the surface during early morning and evening hours highlights a more complex interaction than the constant decoupling suggested by aircraft measurements (Zhang and Zuidema., 2019). Despite these complexities, our findings provide a reliable representation of the prevailing boundary layer conditions (Zhang and Zuidema, 2019; Abel et al., 2020; Wu et al., 2020)."*

Specific Comments

1. Page 1, Line 23: The authors state that "OA to rBC mass ratios (OA:rBC) in the MBL between 2 and 5 contrast to higher values of 5 to 15 observed in the nearby FT". I suggest adding the ages of BBAs for this MBL and "nearby FT".

We have adjusted the text to read *"OA to rBC mass ratios (OA:rBC) in the MBL between 2 and 5 for BBA aged over 10 days contrast to higher values of 5 to 15 for BBA aged between 4 and 8 days observed in the nearby FT."*

2. Page 7, Line 200: What is the supporting material (reference or else) for using BC < 20 ng/m3 as the criteria to calculate background SO4 mass concentration?

This was the 5^{th} percentile of the rBC mass concentrations for June, July, August, and September. This method was adopted from Che et al., (2022). This detail has been added to the text the sentence now reads *"…when rBC mass concentrations were less than 20 ng m^{-3}, the bottom 5^{th} percentile…"*

3. Page 7, Table 2: Error with captions, is it ΔSO4 or SO4? And the same question for CO, is it clean period CO or ΔCO?

The table caption and the text contained within the table have been corrected to ΔSO_4 and ΔCO.

4. Page 8, Lines 225-226: What is the supporting material (reference or else) for using BC > 150 ng/m3 as the threshold to define plume events?

This was the 70^{th} percentile rBC mass concentration for June, July, August, and September. This value is double the threshold that Che et al., (2020) used to define plume events. This threshold was to ensure that BBA dominated the plume events.

5. Page 9: In Table 3, Regime 3 burning condition is described as "Inefficient and efficient". Page 10, line 292-293: Regime 3 is described as "Although fires are still highly active in grassland regions during this time, these values indicate *inefficient combustion*". Page 25: The reasons why Regime 3 had a mixture of inefficient and efficient combustion sources is explained in Sect 4.1. The description of Regime 3 burning condition is not consistent in Sect. 3 and 4, which may confuse the readers. I suggest rephrasing this part.

We have corrected the text in Section 3.1 to avoid confusion between the two sections. The paragraph in Section 3.1 now reads, *"The final two plume events, spanning August 24 to September 11, have the lowest overall rBC:ΔCO, with a mean of 0.0071±0.0004. These low rBC:ΔCO values indicate that the fires were inefficient, however, most fires occurred east of*

30˚E and south of 10˚S (Fig. 5a, in northeast Zambia, southwest Tanzania, Mozambique, and Zimbabwe), with surface RH ranging between 30 and 60 % (Fig. 5b), over vegetation types varying from grasslands to woody savannas (Fig. 5c). Also, most of the fires occurred over dry central Africa and many also occurred on the eastern African coast where precipitation was greater in September (Ryoo et al., 2021). The variation in surface RH, and vegetation suggest that the two plumes in this regime originated from fires that are both efficient and inefficient. We further describe these conditions in Section 4. A notable feature of this regime is that the strong free-tropospheric winds known as the African Easterly Jet-South became active around August 20, at approximately 700 hPa (Ryoo et al., 2022).”

6. Pages 11-13, Figures 3-5: There are errors with figure caption, (b) should be surface relative humidity, and (c) should be land use map.

This has been corrected.

7. Page 14, Table 4: the last line of SSA530, “0.81 0.01”, a “±” is missing.

This has been corrected.

8. Page 16, Lines 499-502: The authors state that “the exception f44 values of P5 and P10… These differences possibly result from regime transitions and/or more complex fuel source mixtures”. Could you provide more explanations why more complex fuel source mixtures may lead to the exception f44?

The BBA over the SEA is some of the most oxidized aerosol that has been intensively sampled. During the PMF analysis, we could barely distinguish two factors, as the organic aerosol was mostly homogeneous by the time it reached Ascension Island because it was so aged. Typically, PMF analyses apportion up to 5 aerosol sources because most field campaigns study more heterogeneous organic aerosols. We lack sufficient information to determine the causes of these slight differences in mass spectrums due to the limited amount of PMF data on highly oxidized organic aerosol. Ultimately, we took the average of the *f44* values to make help ease the interpretation of the PMF analysis.

9. Page 18, Line 557: May need a reference for Hoppel minimum.

We have added the reference (Hoppel et al., 1986) here.

10. Page 18, Line 567-568: “The average of the bimodal size distributions during BBA-laden times had an Aitken mode with a peak 200 cm-3 *at a larger diameter* near 45 nm”. It should be clear to specify, “at a larger diameter” is larger than pristine period or?

We have rewritten the sentence as, “*The average of the bimodal size distributions during BBA-laden times had an Aitken mode with a peak 200 cm^{-3} at a larger diameter than that during the pristine period, near 45 nm, and an accumulation mode with a much larger peak than that during the pristine period ranging from 400-700 cm^{-3} at a diameter near 165 nm (Fig. 11a), with a Hoppel minimum diameter near 70 nm.”*

11. Page 18, Line 584: "A lower fraction of the BBA-laden days (4 of 12) than *Regimes 1 and 2*".

We have rewritten the sentence as, *"A lower fraction of the BBA-laden days (4 out of 12) than in Regimes 1 and 2 had bimodal number size distributions (Fig. S4c)…"*

12. Page 20, Line 628: "For example, the lowest SSA530, 0.75□0.01, in P6, also had the highest FrBC". The highest FrBC is in P9, rather than the P6. Please rephrase this.

We have rewritten the sentence as, *"Lower SSA_{530} values were typically associated with higher FrBC and lower OA:rBC and ΔSO_4:rBC. For example, the lowest SSA_{530}, 0.75±0.01, in P6, also had the second highest FrBC (0.32±0.04)…"*

13. Page 20, Line 653: There is no Table S1 in the supplement. Please add it if it is needed.

The sentence was from a previous draft and has been removed. The current draft provides greater detail about the transportation pathways than the previous draft. Therefore, a supplemental table is no longer needed.

14. Page 23, Line 744-745: There is a redundant reference of "(Ryoo et al., 2022)" in one sentence.

We have corrected the sentence. *"The African Easterly Jet South became active after Aug. 20 causing a dramatic switch in the BBA transport to much higher in the FT, near 700 hPa (Ryoo et al., 2022)."*

15. Page 24, Table 5: Wu et al. (2020) provided the SSA values at green channel. I suggest replacing SSA658 with the blue channel SSA from Wu et al. (2020) in Table 5.

We chose to use the SSA values at 658 nm because those are shown in Figure 7 for the polluted boundary layer (Wu et al., 2020) and that time period (Period 3) is the most relevant to the results we are presenting in this study. We acknowledge that Wu et al., (2020) states that the SSA values at 550 nm ranged from 0.88 to 0.97; however, this range appears to be the range for the entire study, not just the polluted periods. We would prefer to keep the values in the table as is. We have added text to Section 2.5 briefly describing differences in optical property measurements (see Comment 4 above).

16. Page 26, Line 837: "(although still within instrument uncertainty)". What is the uncertainty of "rBCmpd"?

The uncertainty of the SP2 was ~20% (line 256).

17. Page 26, Lines 847-849: The authors state that "However, large rBCgpd and rBCmpd values of 135 and 205 nm, respectively, were also observed during Regime 3, in late August and early September when burning conditions were less efficient (rBC:ΔCO = 0.008)". The values (rBC:ΔCO = 0.008) for Regime 3 is not consistent with the average value (0.007) presented in Table 4.

The text has been corrected to agree with the table. The rBC:ΔCO value of 0.007 is correct.

18. Page 26, Lines 850-852: The authors state that "This contradicts this study's findings that rBCgpd and rBCmpd values were smaller (125 and 195 nm) when fires were also less efficient (rBC:ΔCO = 0.009)". Are these BC core size and rBC:ΔCO values from Regime 1 or which single plume event? I suggest clarifying this clearer.

We have rewritten the sentence as *"Yet, smaller rBC_{gpd} and rBC_{mpd} values in Regime 1 (125 and 195 nm) occurred when fires were also less efficient (rBC:ΔCO=0.008), which is contradictory to the higher values of rBC_{gpd} and rBC_{mpd} in Regime 3 that support Holder et al., (2016). These contrasting results suggest that further research is needed on the dependence of rBC core diameters on burning conditions and fuel types."*

19. Page 27-28, Lines 900-901: The authors state that "f44 values were greater than 0.22 and OA:rBC values were less than 5 for BBA sampled in the FT and MBL near Ascension Island". In Fig. 16, only the FT f44 values are presented. Is "MBL" near Ascension Island needed here?

The green marker on Figure 16 shows the LASIC MBL f44 and OA:rBC values. We have added an orange marker for CLARIFY data in the MBL based on the results from Wu et al., (2020).

The figure caption now reads *"Figure 16. OA:rBC versus f44 for the FT during ORACLES (2016-2018)(red), CLARIFY FT (2017) (blue) (Wu et al., 2020), CLARIFY 2017 MBL (Wu et al., 2020)(orange), ATTO (2014)(black) (Holanda et al., 2020), and for the LASIC MBL (2017) (green dash). Error bars represent the standard deviation of the data set."*

20. Page 31, Line 1040: Error with the figure caption. a) SSA 530 nm versus "*FrBC*".

The figure caption now reads *" a) SSA_{530} versus FrBC"*