We thank the reviewer for their thoughtful comments and insights on our manuscript. We appreciate the time and effort that was dedicated to providing these suggestions. These revisions have enhanced the clarity and impact of our work. The reviewer comments are shown in black, with the author responses shown in blue and any edited manuscript language shown in *italicized blue font*.

The main major comment I have on this paper is regarding the Regime separation and its justification and significance. The authors state that the division is supported by differing burn conditions (fire density, surface RH, land use, and fuel type) over the season (Line ~228+, Table 3), but it is difficult to see how these differences are supported in the analysis (Figs 2; 3-5).

Due to limited availability of conserved tracer data (i.e., CO_2 and other gaseous tracers) and airspace restrictions, we included reanalysis data to aid in interpreting the burning conditions over land. This approach aligns with Che et al. (2022), who similarly used soil moisture content as a supplementary metric to characterize burning conditions throughout the season. We have revised the text in Section 2.2 to clarify that the reanalysis data serve as supplementary information for interpreting the rBC:ΔCO ratios rather than as a primary classification criterion.

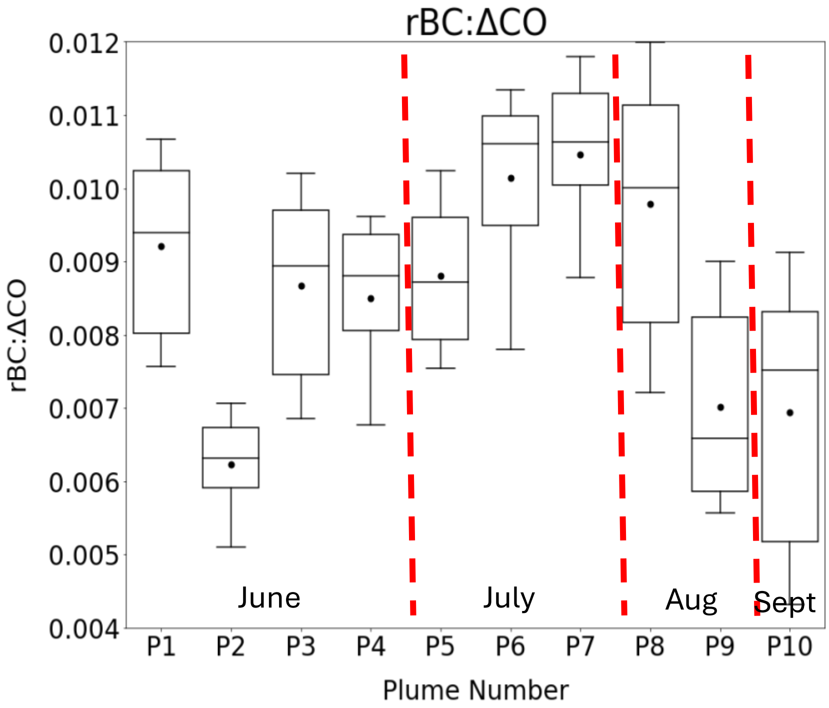
"Surface RH fields provided by the NOAA National Center for Environmental Prediction (NCEP) reanalysis are robustly used to assess the burning condition classification, with RH values >50% indicating efficient fires and <50% indicating inefficient fires. The surface RH analysis serves as supplementary information for interpreting the rBC:ΔCO ratios, rather than as a primary classification criterion, similar to the approach used in Che et al. (2022a). Inefficient fires typically produce relatively more OA and SO_4 *and relatively less rBC than efficient fires (Collier et al., 2016; Rickly et al., 2022). Although modified combustion efficiency (MCE) may be a better determinant of burning conditions (Collier et al., 2016; Dobracki et al., 2023), this quantity could not be calculated because CO2 was not sampled at Ascension Island during the LASIC campaign."*

The rBC:delCO distributions are described in the text (e.g. Line 16) as being used to determine the different fuel and burn conditions, but as these values are shown in Fig 2, they are not obviously subdivided according to a particular criterion

We note here that the classification of the 10 plumes into the 3 regimes is mostly meant to aid interpretation. The regime differentiation is not statistically significant. Indeed, one remarkable feature of the long-range transport BBA is how little it changes, with PMF analyses only identifying two major components that mostly resemble LVOOA. Previous studies from LASIC have often classified BBA using monthly-means (Zuidema et al., 2018; Carter et al., 2021; Che et al., 2022) However, smoke loading at Ascension is strongly synoptically modulated (evident in Fig. 1 of this manuscript) with very clean time periods interspersed with time periods experiencing significant loading. Thus, monthly-means (which are inherently another arbitrary time unit separately by only one day) may not adequately capture the BBA properties in the Ascension Island marine boundary layer (MBL).

We have added the following text to Section 3. "*Previous studies from LASIC have often classified BBA using monthly means (Zuidema et al., 2018; Carter et al., 2021; Che et al., 2022). However, due to the variability in aerosol loading across the four months, monthly means may not adequately capture the BBA properties in the Ascension Island MBL. Here we explore ten plume events…*"

This is evident simply by reconsidering the grouping of rBC:Delta_CO ratios by month, (shown below) with the red dashed lines now distinguishing the June, July, August and September smoke plumes.



As shown, the rBC:Delta_CO ratios indicate the first plume event in July (P5) more closely resembles the June BBA characteristics, the first plume event in August is more similar to late July BBA, and the last plume in August was more similar to the September BBA. Given that we wanted to move away from monthly-means to a more individual analysis, when differences in the BBA characteristics are nevertheless subtle compared to more complex northern hemisphere urban environments, yet also wanted to reduce the complexity of 10 individual events to a fewer number, we came up with the regime discrimination we used throughout the manuscript.

In response to the reviewer's feedback, we did revise the manuscript to reduce the emphasis on reanalysis data (RH, fire density, land use) while retaining the regime classification based on the rBC:ΔCO ratio.

In Section 2.2, we have included the following text, "*Surface RH fields provided by the NOAA National Center for Environmental Prediction (NCEP) reanalysis are robustly used to assess the burning condition classification, with RH values >50% indicating efficient fires and <50% indicating inefficient fires. The surface RH analysis serves as supplementary information for interpreting the rBC:ΔCO ratios, rather than as a primary classification criterion, similar to the approach used in Che et al. (2022a). Inefficient fires typically produce relatively more OA and SO_4 and relatively less rBC than efficient fires (Collier et al., 2016; Rickly et al., 2022). Although modified combustion efficiency (MCE) may be a better determinant of burning conditions (Collier et al., 2016; Dobracki et al., 2023), this quantity could not be calculated because CO2 was not sampled at Ascension Island during the LASIC campaign.*"

And in Section 3, we have included the following text, "*Previous studies from LASIC have often classified BBA using monthly means (Zuidema et al., 2018; Carter et al., 2021; Che et al., 2022). However, due to the variability in aerosol loading across the four months, monthly means may not adequately capture the BBA properties in the Ascension Island MBL. Here we explore ten plume events…*"

Please note, these are the same responses from the first two comments above.

Particularly looking at plume P2, it doesn't seem to "suggest burning conditions remained mostly homogeneous over the six weeks" (Line 281). I wonder if there's any evidence to suggest that P2 is more in line with P9-10 in terms of origins? Figs 9, 10, and 12 also don't clearly show 3 distinct sets of properties, so I would like to see a more concrete justification for that division, if you choose to keep it. (and actually, P2 is an obvious outlier in Fig 12 as well).

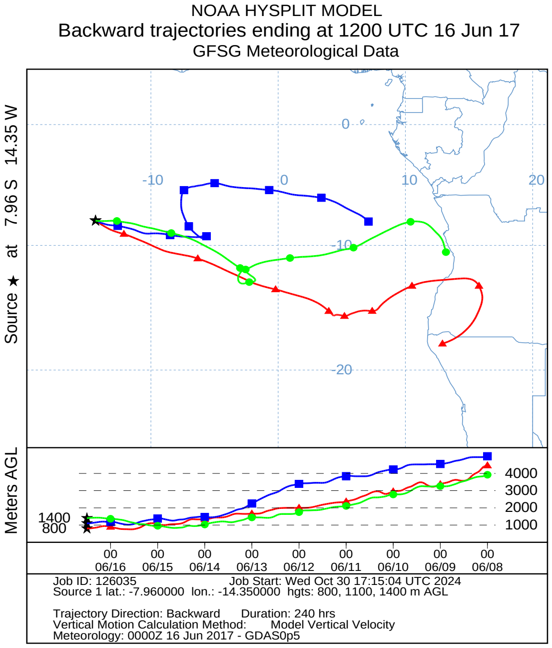
As also in the above figure, P2 is clearly an outlier compared to its neighboring-in-time smoke plumes. We have provided additional text in Section 3.1 to highlight this anomalous plume.

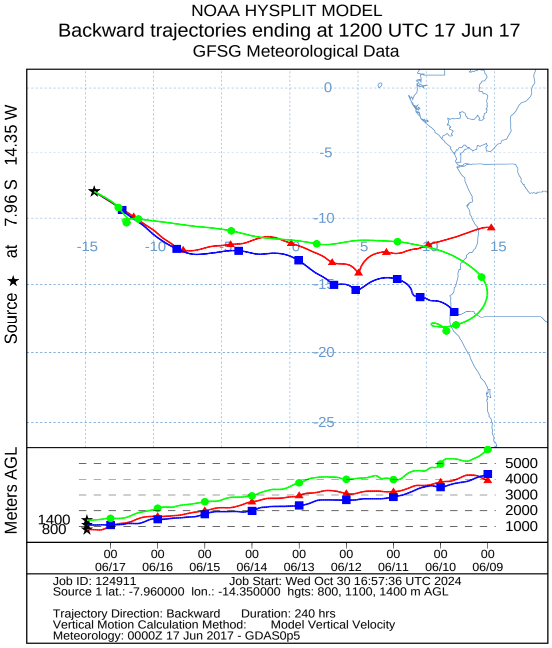
Lines 353-354 "*Despite the anomalously low rBC:ΔCO value of 0.006±0.001 in P2, the rBC:ΔCO ratios of the other four plumes suggest that burning conditions remained mostly homogeneous over the six weeks.*"

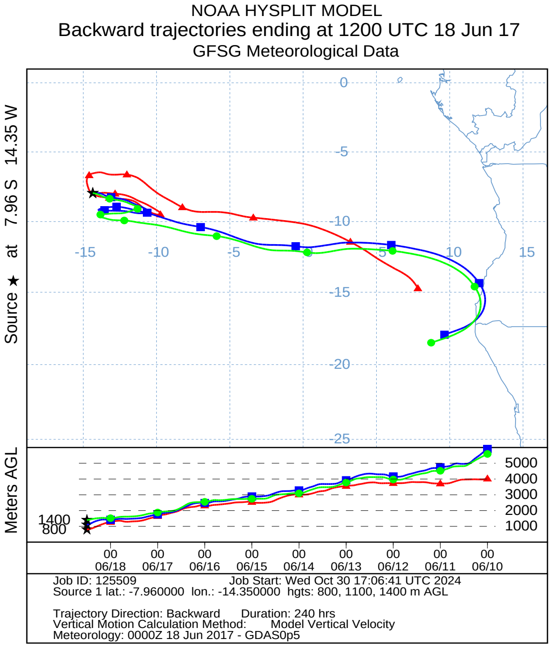
Lines 362-380 "*The final two plume events, spanning August 24 to September 11, have the lowest overall rBC:ΔCO, with a mean of 0.0071±0.0004. These low rBC:ΔCO values indicate that the fires were inefficient, however, most fires occurred east of 30˚E and south of 10˚S (Fig. 5a, in northeast Zambia, 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 insinuate that the two plumes in this regime may have originated from fires that are both efficient and inefficient. We further describe these* *conflicting 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).*

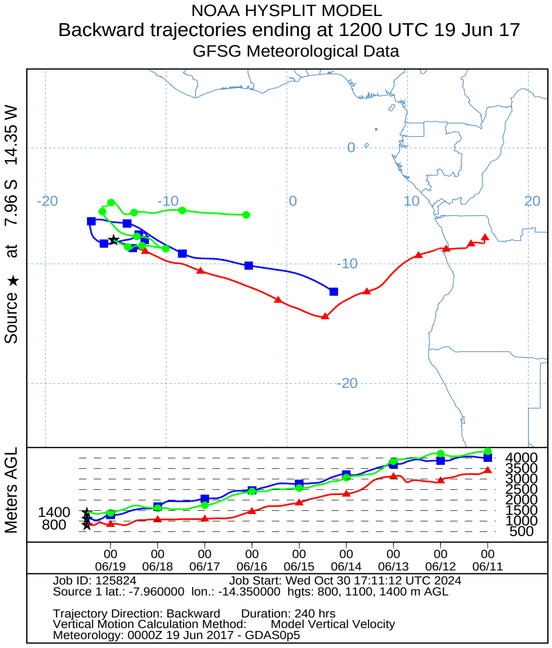
Additional daily HYSPLIT back trajectories reveal that this plume likely originated from northwestern Angola and little to no precipitation occurred as the plume was transported to Ascension Island. This is now discussed in Section 3.3.

"Additional HYSPLIT back trajectories (not shown) highlight that P2 with the anomalously low rBC:ΔCO ratio (0.006±0.001) also originated from northern Angola, which suggests that the source of this plume was likely similar to those from the other four plume events in this regime."









The source origin of P2 is similar to P1, P3-P5, and there is no evidence that precipitation removed rBC (while maintaining the CO). While the rBC:ΔCO ratio aligns more closely with that from Regime 3, the source region of P2 aligns more closely with that from Regime 1. The rBC core size and the aerosol number size distributions from P2 also more closely align with the other four plumes in Regime 1 (Figs. 6, 11a). Overall, we can only conclude that perhaps this BBA originated from a late-stage fire, when the most intense flaming was over, and was developing characteristics of a more smoldering fire. This would explain why this plume had lower rBC mass concentrations than the other plumes in Regime 1 relative to OA, SO_4, and CO, likely due to less-efficient combustion processes. This conjecture must remain speculative but is consistent with the measurements we do have. The reanalysis and HYSPLIT back-trajectories indicate P2 should be viewed in the context of the neighboring-in-time plumes, and not grouped with P9-P10.

We also respond to the further concern raised by the reviewer, that the transition between Regime 1 and 2, Fig 3 vs Fig 4 show averages over two periods of 4-6 weeks each, with only one day of separation between them. It's not at all obvious that e.g. the spatial distribution of fire density is meaningfully different between these two periods (Table 4 also seems to indicate that many parameters may be statistically indistinguishable between the two of them); instead, these figures' panels (a) seem to show that there is greater density in the latter period, but that the fires occur over largely the same spatial domain. A distinction of surface RH <50% or >50% (p. 10) also seems a bit arbitrary a cutoff, and e.g. it's not actually clear that the locations of the fires in Fig 3 correspond to RH>50% (line 283).

As mentioned above, we have revised the text in Section 2.2 and placed less emphasis on the reanalysis data.

"*Surface RH fields provided by the NOAA National Center for Environmental Prediction (NCEP) reanalysis are robustly used to assess the burning condition classification, with RH values >50% indicating efficient fires and <50% indicating inefficient fires. The surface RH analysis serves as supplementary information for interpreting the rBC:ΔCO ratios, rather than as a primary classification criterion, similar to the approach used in Che et al. (2022a). Inefficient fires typically produce relatively more OA and SO_4 and relatively less rBC than efficient fires (Collier et al., 2016; Rickly et al., 2022). Although modified combustion efficiency (MCE) may be a better determinant of burning conditions (Collier et al., 2016; Dobracki et al., 2023), this quantity could not be calculated because CO2 was not sampled at Ascension Island during the LASIC campaign.*"

Please note this is the same response from the first comment above.

The reanalysis data was used as a tool to help with interpretation of the large LASIC data set. We wanted to move away from a monthly mean analysis, which also would have had 1 day of separation between observational periods. Previous studies have shown that as the biomass burning season progresses, the grass (moisture) content decreases (Hoffa et al., 1999; Korontzi et al., 2003) and the fires shift toward central and southeast Africa

(Redemann et al., 2022). The surface RH and fire density maps in Figures 3-5 corroborate this.

In the same vein, the time intervals averaged in Figures 3-5, if I’m reading it correctly, show the continental conditions directly coincident with the observed conditions at ASI. Yet, obviously, airmasses don’t arrive at ASI instantaneously; according to Figs 13 and 19, the transport time to reach ASI is ~5 days minimum and may be even greater than 10 days, so do the continental conditions during the exact same time actually indicate changes in conditions? And if so, shouldn’t the conditions (and Regime definition) be lead/lagged relative to the ASI observations?
We have updated figures 3-5 to include fire density and surface RH data 7 days prior to the start of each regime. We had already accounted for this in Figures 12 and 13. And no, we are basing the regime definition on the aerosol characteristics perceived at Ascension by design because the rBC:ΔCO ratio is conserved on these timescales.

This is also complicated by Table 2 vs Table 3; the Regimes are defined as these larger periods, and yet there are also “clean periods” within those periods. So do Figs 3-5 show the average including “clean” times? From Fig 1 it’s about half clean, half plume over a given “regime,” so even if transport time lag is taken into account, did the fires vary within these longer periods?
The clean periods at Ascension Island are due to the presence of a sea-level pressure high between 0˚ and 20˚W, which promoted the advection of pristine air from the southern oceans and forced the BBA to north of Ascension Island. Though the smoke plumes did not reach Ascension Island, there were still fires over these time periods which is why they are included in Figures 3-5. The values in Table 3 only include data from the plume. We have added a footnote on Table 3 to clarify this.

The “efficient”/”inefficient” burning condition distinction also was not clear: the low rBC:delCO mass ratios = inefficient combustion (Line 280, 806+); higher values in the second period = efficient combustion; and the third period is either “inefficient combustion” (Line 292) or a combination of the two (Table 3), despite being lower values than either of the other two which are supposedly distinct? If anything, it seems to me that Regime 1 with the one outlier P2 plume should be the mix of burning conditions.
Every plume in Regime 1 had a mean rBC:ΔCO ratio < 0.1. P2 is an outlier; however, it still fits into the classification of inefficient fires. The confusion with Regime 3 was also brought to our attention by the first reviewer, and we have revised the text in Sections 3.1 and 4.1 to explain why the plumes in Regime 3 are likely from a mix of inefficient and efficient despite the overall low rBC:ΔCO ratios.

In Section 3.1, the text 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 insinuate that the two plumes in this regime may have originated from fires that are both efficient and inefficient. We further describe these conflicting 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).”*

In Section 4, the text now reads, *“The mean rBC:ΔCO values in Regime 3 were less than 0.01, indicating that the fires across the DRC and Mozambique were inefficient, as concluded above (Figs. 5a, 13c-d). However, the low OA:rBC values, low ΔSO_4:rBC values, and high FrBC observed during P9 (late August) suggest that BBA during this plume event originated from efficient fires. A similar discrepancy is seen in P10 (early September), where high OA:rBC, large rBC coating-to-core mass ratios, and high ΔSO_4:rBC would suggest that the fires were inefficient, while the high FrBC would indicate that the fires were efficient. These conflicting results imply that despite the overall low rBC:ΔCO values in Regime 3, the BBA from earlier in this regime likely resulted from efficient fires across central African grasslands, whereas the BBA from later in this regime likley resulted from both efficient fires across the grasslands and inefficient fires near the eastern coast (Jiang et al., 2020). These intriguing BBA properties observed in early September are further investigated in Sect. 5.1. Overall, the efficient fires in late August and the combination of efficient and inefficient fires in September are consistent with observations reported by Che et al. (2022a), who concluded that burning conditions in this region become less efficient as cloud cover increases, precipitation increases, surface wind speed decreases, and soil moisture increases from August to October.”*

This also gets a bit muddled with the discussion of oxidation and evaporation, transport, and other processes (Section 4, throughout), since it’s not clear to start how these plume events are similar to one another. If the transport pathways in Fig 13 vary from t~5 to 9 days in these examples (just from when it exited the continent, it’s difficult to follow how this can be used to state that the OA:rBC etc ratios vary at the time of emission, if they’re then oxidized at different rates over different times, if I’m following Sec 4. It’s not clear how “faster transport pathways (Line ~974+) are definitive in one regime over another. Yes, the reviewer makes a good point here, this is an assumption that we make but did not clearly state. In Regime 1, the OA:rBC ratios are the highest, and transport also took the longest to get to Ascension Island. Whereas in Regime 2, OA:rBC ratios were lower and transport time was the fastest. We assume that OA:rBC values were higher initially due to the inefficient fires and even with 10 days of transport time and that the OA concentrations were still higher when the plume was sampled at Ascension Island. We have added text throughout Section 4.1 to clarify that our interpretation of the OA:rBC ratios measured at Ascension Island are consistent with the burning conditions at the source. We have also revised the following text in the beginning of Section 4.1 to add more transparency to our interpretation.

“In this section, we discuss how burning conditions and fuel types change across the three temporal regimes, and how these changes may affect BBA properties such as FrBC,

rBC coating-to-core mass ratios, and OA:rBC and ΔSO_4:rBC mass ratios. These analyses offer further BBA characterization than what has typically been presented for biomass-burning events. However, given the limited data set, we cannot definitively describe conditions over land and instead rely on supporting data from previous studies in this region to interpret the observed patterns."

Figure 1: caption states "Pink boxes indicate selected plume events; blue boxes indicate selected clean periods" but I'm only seeing grey boxes edged in orange. It might also be nice to label the different plume events on this figure (P1-10).
This has been corrected. We have also labeled the 10 plume events on the figure.

Check that subpanels for Figs 3-5 are labeled properly in the caption and in the text; the caption seems to describe a, c, b rather than a, b, c.
This has been corrected.

A minor note: VIIRS is an instrument on the Suomi NPP satellite (e.g. Fig 3 caption)
This has been corrected. We have also corrected the figure caption to read *"...fires detected by the NASA SUOMI NPP satellite..." for each of the captions.*

Line 51: suggest to report lat/lon in S, W coordinates, rather than negative N,E.
This has been adjusted to S, W coordinates.

Discussion ~Line 47-58: the context might benefit from discussing Eck et al 2013 (doi:10.1002/jgrd.50500), which saw an increase in SSA through the BB season at one site, i.e., changes in optical properties likely from similar geographical regions.
Eck et al., 2013 is a relevant study to this work. We agree with the reviewer that their work should be mentioned in this section. We have edited and added the following text to Section 1, *"The BBA in the FT is highly absorbing of shortwave radiation (Pistone et al., 2019; Denjean et al., 2020a; Taylor et al., 2020; Wu et al., 2020; Dobracki et al., 2023) with single scattering albedo (the ratio of the aerosol scattering coefficient to the sum of the aerosol scattering and absorption coefficients) at wavelength 530 nm (SSA_{530}) values increasing from 0.80 to 0.95 between June and October in the continental boundary layer across southern Africa (Eck et al., 2013). Measurements in the FT over the SEA show August-September mean SSA_{530} values near 0.84 (Pistone et al., 2019; Wu et al., 2020, Dobracki et al., 2023) and measurements in the MBL at Ascension Island (7.95 ˚S, 14.36 ˚W), a remote location in the tropical Atlantic, yield an even lower SSA_{530}, with June-August monthly-mean values near 0.80 (Zuidema et al., 2018; Che et al., 2022a). The lower SSA_{530} values at Ascension Island in the MBL compared to that in the FT above the island has not been previously explained (Barrett et al., 2022; Sedlacek et al., 2022)."*

Line 181: surely there aren't many fires at 5.7W, 3.2 N? Typo?
These were just the selected box parameters for the NASA FIRMS data selection. We wanted to encompass the entire region. Figures 3c-5c show that there were no fires above 0˚N and of course none over the southeast Atlantic Ocean. We have adjusted the text to

read, *"The locations of the fires between 12.0 ˚W-52.0 ˚E, and 0 ˚N-34.5 ˚S, encompass the sources…"*

Fig 6: I'm curious what the bars are on this plot— is it some standard deviation rather than the percentile distribution over a given event? I ask because in contrast to many of the other figures, these ones seem to be uniform rather than varying from plume to plume, but I would imagine that the range in diameters would vary between plumes as well?
The error bars on the original figure were the standard deviation of the data set. We have updated the figure to include error bars for the standard deviation of each plume.

Sentence starting on Line 205: sentence fragment or missing a verb, I think
We have rewritten the sentence as, *"Oxalate, an organic acid that is a well-known tracer of aqueous-phase OA oxidation contributes to f44 and can indicate that the aerosol has interacted with cloud (Sorooshian et al., 2010; Ervens et al., 2011)."*

Line ~224: rBC units switch back and forth between micrograms and nanograms; I'd pick one. Also it might be good to show the rBC<20ng/m3 threshold in Fig 1, which has 200ng/m3 as the lowest tick mark, if I'm reading it correctly
We have converted the ng m^{-3} to µg m^{-3} in the text and have added blue color bars to Figure 1 to denote clean time periods.

Table 3: I presume these values are means/standard deviations, but it would be good to confirm that in the caption, if you stick with the Regime construction.
We have rewritten the table captions to include "mean ± standard deviation" when relevant.

Table 4: last row might be missing a +/-
This has been corrected.

Fig 8: it might be nice to show the plume events here, as in Fig1, or perhaps as the ratios between parameters as described in the text.
This is a good idea. We have added the colored boxes to the figure to show the plume events. The boxes are also labeled.
Line 479: typo in "mass"
This has been corrected.

Line 584: it's a bit difficult to follow what this sentence is saying.
We have rewritten the sentence as *"Only 4 out of the 12 polluted days in Regime 3 exhibited a bimodal number size distribution (Fig. S4c), which is fewer than in both Regimes 1 and 2. The monomodal number size distributions only contained the accumulation mode, with a number concentrations between 400 and 800 cm^{-3} and a modal diameter near 200 nm, the largest of the three regimes (Fig. 11a)."*

Line 651/Figs 13-14: I'm curious why the 850hPa winds are shown (I think; would be nice to specify in the caption). Especially later in the season, the south African Easterly Jet for continental transport is 700 or 600hPa in Aug or Sep.
The 850 hPa winds are used to demonstrate the boundary layer transport to Ascension Island, as also shown in Haywood et al. (2021), Zhang and Zuidema, (2021) and Che et al. (2022).

Table 5: I'm a bit curious about the ATTO transport pathway that gets African biomass burning smoke to the Amazon sooner than to Ascension in June; is that just based on the season? (I might add that to the header, then). But this is a different value than stated in Fig 16.
The similar transport time of 10 days for the BBA to reach both Ascension Island and Brazil can be attributed to the differences in transport pathways between the MBL and FT. The BBA reaching Ascension Island traveled via slower boundary layer transport, as described in Section 3.3. In contrast, Holanda et al. (2020) reported that BBA sampled at ATTO in Brazil was transported primarily within the FT, facilitated by the African Easterly Jet south, resulting in more rapid transport at higher altitudes. Notably, the lower pollution layer (LPL) in their study corresponds to the lower FT, rather than the MBL. We do agree that this description would be helpful to the reader and provide a clearer explanation as to why the BBA is aged similarly, yet the *f44* values at ATTO were higher than those at Ascension Island. We have added the following text to Section 4.5.

"The BBA sampled at ATTO was transported primarily within the FT, facilitated by the southern African Easterly Jet, resulting in rapid transport at higher altitudes. As a result, the BBA reached Brazil in a similar time frame to Ascension Island, which likely explains the similarities in f44 and OA:rBC values between the two studies. Notably, the lower pollution layer (LPL) in their study corresponds to the lower FT and not the MBL."

Line 863: what is the relevance of Arctic observations to the present study? I presume they were also of BB aerosol? But Figure 15 suggests that perhaps the present study and the past Arctic observations were not comparable. I presume the aging/transport time was much longer for Arctic aerosol?
The use of the Arctic data was meant to demonstrate how *f44* and *f60* evolve as fresh BBOA ages. While it is not directly comparable to the African BBOA, it illustrates how different the aged African BBOA is from the fresh Arctic BBOA since there are no studies that report *f44* and *f60* values of fresh African BBOA. We do have language in the text about how the Arctic BBOA is only aged 5 hours and the ORACLES and LASIC BBOA is aged 4-10 days. Figure 15 also highlights that the Arctic BBA is fresh BBA. We have added the following text to Section 4.3, *"Although Arctic BBA likely differs from African BBA in composition, this data is useful to illustrate how the BBA evolves in the initial 0 to 5 hours after emission, for which direct data is not available in our study."*

Line 902 and 946: seem to be conflicted as to whether wet deposition can happen in this region?
The text on line 902 refers specifically to the FT, where precipitation was unlikely to occur. In contrast, the text on line 948 addresses the MBL, where precipitation, though

not a major removal mechanism, is possible due to the presence of marine stratocumulus clouds. Additionally, the text following line 948 highlights that precipitation was not a dominant process in the MBL, as there is no indication of large particles being removed in the aerosol size distributions.

Line 968: missing a word?
We have corrected this sentence. It now reads as, "*These conditions allow more time for interaction with clouds. The highest fraction of activated aerosol particles occurred during this time (Zuidema et al., 2018), consistent with larger accumulation-mode aerosol (Dedrick et al. 2024).*"

Line 1079: wrong verb?
We have corrected this sentence. It now reads as, *"Although these chemical properties are consistent with inefficient combustion, the optical properties are not."*

Fig A1: what averaging is shown in this figure (I don't think this is the 1Hz data)? Also, the caption and the legend seem to be saying two different things re: which line/color is the CAPS?
The data presented in the figure is the 30-minute average of the ARM VAP SSA_{530}, the CAPS SSA_{530}, and rBC. The caps data were interpolated onto the 30-minute average time scale to match the ARM VAP SSA and rBC values. The ARM VAP SSA_{530} mean for the time period shown is 0.80±0.02 and the CAPS SSA_{530} mean is 0.79±0.06.The mean rBC concentration was 0.55±0.2 to indicate these means were compared during a plume event. The figure caption text has been corrected to agree with the legend on Figure A1.