We thank the reviewers again for the time and effort that was dedicated to providing these suggestions. The reviewer comments are shown in black, with the author responses shown in blue and any edited manuscript language shown in *italicized blue font.*

Report #2 Response

Page 1065-1070: The SSA530 values in the Ascension Island MBL, between 0.75 and 0.83 (Section 3.2.4), are among the lowest in the world from BBA... In contrast, our results demonstrate that BBA in the MBL is more absorbing than that in the FT.

Remaining question:

The authors conclude that “our results demonstrate that BBA in the MBL is more absorbing than that in the FT”.

Authors compared LASIC MBL BBAs (8-10 days?) and ORACLES FT BBAs (4-6 days?) with different transport ages. However, Wu et al. (2020) compared MBL and FT BBAs over Ascension Island with close transport ages. The main distinction here, which leads to the different conclusions is BBAs’ transport ages.

I agree that “mixing with clean marine air should increase SSA530 in the MBL”, such as in Wu et al. (2020). However, the lower LASIC MBL SSA530 appears to be more associated with longer chemical aging and mass loss than ORACLES FT (such as discussions in Sect. 4.3), which leads to the lower OA/BC rations discussed later in Sect. 4.5.

These may require clearer clarification to avoid potential misleading

Wu et al. (2020) report a mean polluted-BL SSA$_{550}$ of 0.81. Their ‘period 1’ occurs at the tail end of our P8 and their ‘period 3’ coincides with our P9. For these 2 plumes, we report a mean SSA530 of ~0.793, with the standard deviation encompassing the mean CLARIFY value. Their Table 1 indicates a higher BC fraction in the BL during their period 3 than period 1, consistent with the slight reduction in SSA between the 2 periods shown in our Fig. 12. Although the SSA variability indicated in our Fig. 12 encompasses the CLARIFY values, the reviewer is correct that the CLARIFY values are likely even higher near the surface, as Wu et al. (2020) document a decrease in SSA with height within the boundary layer. The EXSCALABAR estimates of absorption and extinction are state-of-the-art and will provide a more routinely reliable measurement of SSA than the filter-based measurements applied during the LASIC campaign. The LASIC filter-based absorption measurements compare impressively well to the CAPS-SSA measurements (Fig. A1) while Fig. S6 within Barrett et al. (2020) shows good correspondence between the CAPS absorption and EXSCALABAR PAS absorption, for PM1.0. One explanation for the slightly lower LASIC SSA values than those measured by CLARIFY could be the cut size diameter of the aerosol inlet (Barrett et al. 2020). While the LASIC ARM site used a 1 micron aerodynamic impactor, the FAAM used a 1.3 micron aerodynamic impactor. The particles at the surface that are larger than 1 micron are most likely to be coming from sea spray (see Fig. 11 from the manuscript), which possess an SSA closer to 1.0. This is postulated within Barrett et al. (2022) to explain the differences in the scattering observations between the LASIC ARM site and the CLARIFY observations, and would be enough to explain the small differences in SSA noted here.

We have added the following text to Section 2.2, lines 159-163 to acknowledge the differences in the impactors used during LASIC and CLARIFY.
"EXSCALABAR sampled downstream of a 1.3 µm aerodynamic diameter impactor (Taylor et al., 2020), whereas a 1.0 µm aerodynamic impactor was positioned upstream of the instruments deployed in LASIC. Variations in impactor cutoff sizes could introduce biases in the measured aerosol optical properties, particularly the scattering coefficients. Nevertheless, data from both campaigns are utilized in this study to facilitate a comparative analysis."

We have revised the text to the following, beginning on line 1089, to acknowledge this:

"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, especially near the surface (Wu et al., 2020). For instance, our mean* SSA_{530} *value across P8–P9 of 0.79 compares relatively well with the polluted-MBL* SSA_{550} *value of ~0.81 measured during CLARIFY in the polluted MBL for the same time period. This slight difference can likely be explained by variations in impactor cutoff sizes (Section 2.2; Barrett et al., 2022), where larger aerosol sizes in surface-based measurements contribute to higher SSA values. The slight reduction in SSA from P8 to P9 (Fig. 12) is consistent with the observed increase in rBC fraction from Period 1 to Period 3 during CLARIFY (Wu et al., 2020). Our results further solidify that* SSA_{530} *is negatively correlated with FrBC (Fig. 17a) and positively correlated with OA:rBC (Fig. 17b) in both the MBL and in the FT between the African plateau and Ascension Island."*

Report #1 Response

There were a couple of additional issues that I noticed which suggest they should ensure the details of the revision are correct:

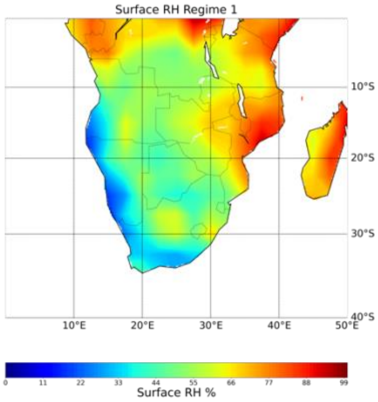
—The author response regarding comments about the efficient/inefficient classification by both reviewers indicates the authors have moved away from various aspects of this distinction, and I do understand the logic in choosing temporal classifications that are not limited to a specific, somewhat arbitrary calendar month. However, the stated main criterion (cutoff at 0.1 in the rBC:dCO ratio) does not seem to follow that for Regime 2? (Sec 2.2). The third plume has a mean <0.1 (and the range is quite large, besides), so I'm not following how that set can be uniformly considered "efficient," if this is in fact the single criterion being used. They have further clarified that the RH classification should be considered supplementary information only, but that seems a bit inconsistent with the language that says e.g. "RH>50% indicating efficient fires" rather than something less prescriptive like "generally associated with". All together, this distinction seems inconsistent and I think needs to be resolved before publication; the paper may be stronger without trying to fit so strongly these assumptions regarding burn conditions.

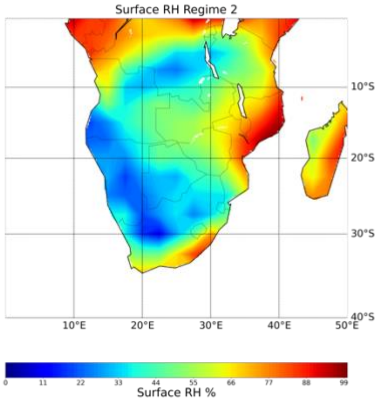
We have corrected the text in Section 2.2 to be more prescriptive. The text now reads *“Surface RH fields provided by the NOAA National Center for Environmental Prediction (NCEP) reanalysis are robustly used to assess the burning condition classification. RH values >50% are generally associated with efficient fires and <50% are generally associated with inefficient fires.”*

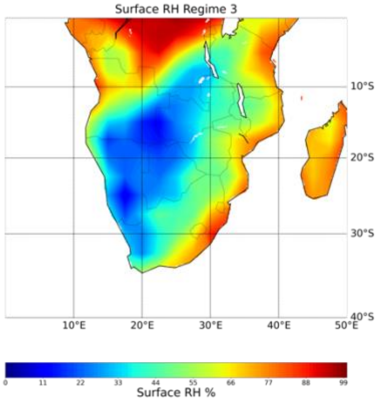
While there is variability in the rBC:ΔCO ratios, the mean values in Regime 2 are overall higher than both Regimes 1 and 3 which suggests that the fires in Regime 2 were more efficient that those in Regimes 1 and 3. Supplementary data sets (i.e., fire location, surface RH, and OA:rBC ratios) also support this classification. We have added the following sentence to Section 2.2 to further emphasize that the rBC:ΔCO is used as a general framework for this paper. *“The rBC:ΔCO classification is used as a general framework in this study, with supplementary context provided by additional factors such as surface RH, land use maps, fire locations, and the OA:rBC and SO4:rBC ratios. This approach is consistent with methodologies commonly used in other studies investigating biomass burning emissions and plume characteristics in this region* (Vakkari et al., 2018; Che et al., 2022a).*”*

—Figures 3-5 have been revised to include an extra week of observations in order to account for transportation time from fire origin; however, panels b) seem unaltered in terms of color contours and 5a) actually seems to show fewer total fire counts than the initial version, which I don’t see how that can be if an extra week of time is added. Regarding the latter, perhaps the authors also meant to change the ending dates for the time period?

The original 5a figure used the J1 VIIRS C1 data, instead of the SUOMI VIIRS C2. I have confirmed that all figures are using SUOMI VIIRS C2 data. All maps are now updated in the manuscript.







—I appreciate the authors clarifying the uncertainty bars on Fig 6, but the new, plume-specific standard deviations seem to be invisible on a few of the plumes; how many measurements went into these statistics and might another metric be more instructive, I wonder?

The uncertainty bars on Fig. 6 represent the standard deviation of the daily mean geometric peak diameters for each plume event. For some plumes, such as Plume 3, the daily mean geometric peak diameter remains consistent across multiple days (e.g., consistently at 125 nm over 7 days), resulting in a standard deviation of zero or near-zero. The time resolution of the rBC data is now included in the caption.

—Typo on Line 934 (Angloa)
This has been corrected.

—I'm still not sure whether Fig 15 (with comparison to data from the Arctic) adds much to the discussion, especially since all the SEA observations will be >>5h old, but I'll leave it to the editor and other reviewer to determine if this is crucial or not.
We have changed Figure 15 to only include ORACLES and LASIC data.

—Line 1322: suggest "for" rather than "from"
We have changed the caption to read as *"SSA_{530} calculated from the PSAP and nephelometer (black)"*.

—I do wish the questions posed at the end of Section 1 had been more explicitly answered/revisited in the conclusion, but I would consider this a more minor/stylistic issue (plus, 5 is a lot for a paper this size).
Given the length of the manuscript, we chose to focus the conclusions on the most useful and interesting takeaways from the study, while addressing each of the posed questions in greater detail within the main body of the manuscript. We believe this approach maintains clarity and ensures that the conclusions highlight the key findings of this work.