

Reply to the Review of Manuscript EGUSphere-2024-3975:

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 **blue**, with the authors' responses shown in **black** and any edited manuscript language shown in *italicized black font*.

Review of Wu et al., 2025: The transport history of African biomass burning aerosols arriving in the remote Southeast Atlantic marine boundary layer and their impacts on cloud properties. This paper combines in-situ data from the CLARIFY-2017 campaign to satellite data and backtrajectory analyses to investigate aerosol-low-level cloud interactions around the Ascension Island in the remote South-East Atlantic. The authors show that cloud droplet number concentrations are increased, and droplet radii are decreased by biomass burning aerosols, especially if the aerosols are in the sub-cloud layer. This agrees with published literature that highlights the important role of the respective positions of aerosol and cloud layers for ACI in this region, even though the exact functional relationship between aerosol and cloud properties seem to differ compared to other biomass burning regions. The authors then carry out backtrajectory analyses to estimate the exact timing of FT air entrainment into the marine boundary layer for 3 different case studies and identify efficient mixing regions 2-3 days upwind of the Ascension Island, which could be used to constrain climate models that struggle to accurately simulate ACI in the SEA. I think these results are interesting and can be published after major revisions.

Major comments:

1. The novelty of the paper is not immediately obvious to me, but I believe this could be fixed with some reorganization of its content.
 - 1) - Many of the figures in the first half of the paper are reminiscent of the figures in Haywood et al., 2021. Since there are a lot of figures in the supplementary material, maybe it would make sense to remove redundant figures and instead move some of the supplementary material to the main text.

We thank the reviewer for this suggestion. While Figures 1 (flight tracks) and 2 (aerosol vertical distributions) may appear similar to Figures 7 and 9a in Haywood et al. (2021), they serve a distinct purpose in our study. These panels provide essential context—defining the sampling region and summarizing biomass burning (BB) pollution conditions throughout the campaign—which underpin subsequent analyses. Therefore, we have retained Figures 1 and 2 in the revised manuscript.

Regarding this suggestion:

- a. We have added more information on Fig.2. Using backward dispersion simulations, we calculated fractional contributions of airmasses from the African continent (20°S – 5°N, 9°W – 35°E) arriving at Ascension Island as a function of time throughout the campaign period, differentiating between FT (Fig. 2b) and MBL (Fig. 2c) simulations. This is to evidence that the observed pollution events around Ascension Island originated from long-range transported African BB plumes.
 - b. We have moved the original Figs. S1 and S9 in the supplementary material to the main text.
- 2) - I also wonder whether the instrument descriptions have previously been published in data papers, in which case it might not be necessary to have such a long method section.

Thanks to the reviewer for this suggestion. Original Sections 2.1 and 2.2 have been combined into one section “Section 2.1 Aerosol and cloud measurements”. We have made the instrument descriptions more succinct.

- 3) Since it has been a few years since the CLARIFY 2017 campaign, it might be helpful to summarize for the reader the findings that have already been published using the campaign data, and mention questions that have yet to be answered, including the one(s) this study wants to address, which would make the novelty obvious.

Thanks to the reviewer’s suggestions on making novelty obvious. The authors have briefly summarized the related findings that have already been published using the campaign data and expanded on the novel research questions this study will address. The revised manuscript is as follows:

*To address the aforementioned issues, aircraft in-situ measurements are essential to provide unique constraints on the vertical distribution of transported African BB aerosols over the SEA for climate models. Aircraft observations with continuous vertical sampling are also the most reliable source for accurately characterizing the correlations between aerosols and clouds. This study presents airborne observations of BB aerosols and clouds collected during the CLARIFY campaign (CLOUD Aerosol-Radiation Interactions and Forcing; August–September 2017), which was based around Ascension Island (7.96° S, 14.35° W) (Haywood et al., 2021). **The CLARIFY campaign addresses a key observational gap over the remote SEA, contributing to a more comprehensive understanding of BB aerosol vertical distributions and interactions with clouds across the SEA.** When integrated with complementary campaigns such as ORACLES (ObseRvations of Aerosols above CLouds and their intERactionS), which primarily focused on regions westward of the African continent and eastward of 0° E (Redemann et al., 2021), **the CLARIFY campaign provides an integrated wide-scale assessment of BB aerosol transport and its impact on cloud microphysics over the SEA.** Observations from these collaborative projects have indicated that the physicochemical properties of BB aerosols continuously evolve during weeklong transport, influenced by aging, cloud processing, and MBL environments, implying different CCN activity after long-range transport (Wu et al., 2020; Dobracki et al., 2023). Additionally, CLARIFY observations have shown that there is often a complex vertical structure of BB aerosol relative to cloud layers, as well as vertical variability of aerosol chemical composition and size distributions (Wu et al., 2020; Haywood et al., 2021). **These complexities underscore the significance of investigating the mechanisms by which long-range transported BB aerosols modulate cloud microphysics over the remote SEA.***

*In this study, we **first present the pollution conditions over Ascension Island and trace the origins of air masses using backward-dispersion simulations** (Sect. 3.1). We then **characterize the vertical profiles of thermodynamic variables, aerosol properties, and cloud microphysics over Ascension Island** (Sect. 3.2). While the region is influenced by the transition from stratocumulus to cumulus clouds, associated with increasing sea surface temperatures (Gordon et al., 2018), this study **focuses on assessing the effects of BB aerosols on stratocumulus cloud microphysics** (Sect. 3.3). More details of cloud types (stratocumulus or cumulus clouds) collected during the CLARIFY are provided in Sect. 3.2. Finally, we integrate air parcel analysis with satellite observations (Spinning Enhanced Visible and Infrared Imager, SEVIRI), **to identify the efficient entrainment regions where FT air parcels from Africa are likely to enter the MBL over the SEA and to demonstrate their impact on cloud properties along transport** (Sect. 3.4).*

2. The paper could be more succinct if the authors removed some redundancy in the explanations. For example, there are long paragraphs dedicated to reviewing the literature in the results section, but the relevance of the cited studies to this study might be more convincing if summarized into 1 or 2 sentences only. - It might also be helpful to make the abstract more succinct.

Thanks to the reviewer for this suggestion. We have streamlined the literature review in the Results section to improve clarity and conciseness. We have also revised the abstract, following the guidelines provided by ACP. The revised abstract is:

African biomass burning (BB) aerosols transported over the southeast Atlantic (SEA) strongly influence cloud properties but remain a major source of uncertainty in regional climate assessment. This study characterizes vertical profiles of thermodynamic conditions, aerosol properties, and cloud microphysics around Ascension Island during an aircraft campaign (August–September 2017). Backward-dispersion simulations evidence that observed pollution originated from long-range transported African BB plumes. In BB-polluted marine boundary layers (MBL), aerosol number concentrations (N_a) were substantially elevated relative to the clean MBL, driving increased cloud droplet number concentrations (N_d) and reduced cloud effective radii (R_e). Cloud-layer mean N_d correlated strongly with aerosols below the cloud (sub- N_a) but weakly with free-tropospheric (FT) aerosols. Enhanced sub- N_a was due to BB aerosols entrained from the FT into the MBL along long-range transport and/or locally. Droplet activation fractions were similar in clean and moderately BB-polluted (sub- $N_a < 700 \text{ cm}^{-3}$) clouds, while a weaker N_d - N_a correlation was observed in more polluted clouds. Region-specific N_d - N_a parameterizations are necessary for representing BB aerosol-cloud interactions over the remote SEA. A robust inverse N_d - R_e relationship was observed, regardless of BB influence. By coupling backward simulations with satellite retrievals, this study indicates that FT-to-MBL entrainment of African BB aerosols over the SEA occurs several days before arrival at Ascension Island, predominantly west of 0° E for examined cases. These findings provide unique observational constraints for representing aerosol-cloud interactions and vertical transport of African BB aerosols in climate models, offering improved assessments of African BB impacts over the SEA.

3. On Ln 540, you mention that your study has important implications for the radiative effects of aerosols in the region, but have you looked at how TOA radiative fluxes from SEVIRI change along the study period, depending on the aerosol load and location?

We thank the reviewer for this insightful comment. We agree with the reviewer that analysis of TOA radiative fluxes (e.g., from SEVIRI) can provide valuable information on the radiative effects of aerosols in this region. The evolution of TOA radiative fluxes during the study period can reflect the combined direct and indirect radiative effects of transported African BB aerosols over the remote SEA but is also affected by the cloud optical thickness that is largely influenced by meteorology.

This study focuses specifically on the transport process of African BB aerosols and quantifying their impacts on cloud microphysical properties over the SEA. The extension of the work to a consideration of the TOA radiative fluxes would add further complexity to the paper and hence is not carried out here. These results establish the foundation needed for subsequent investigations of their indirect radiative forcing. Accordingly, planned follow-on work from the project

will integrate our aerosol–cloud interaction parameterizations developed here to evaluate the TOA radiative impacts of transported BB aerosols in this region.

To address the broader implications, the revised manuscript is:

Additionally, the modification of the cloud fields by high BB aerosol loadings, imply the important indirect radiative effects of transported African BB aerosols over the SEA. Future studies that integrate aerosol–cloud interaction parameterizations developed in this study will be conducted to improve the assessment of aerosol indirect radiative effects in this region.

4. Section 4.2: In my opinion, this section needs to be rewritten.

- I am concerned about the AOT analysis. Since AOT is a vertically integrated value, the analysis of AOT associated to either MBL or FT air parcels is misleading. For instance, an increase in AOT for the MBL air parcels could simply mean that the MBL is located under a polluted FT, but the MBL itself could still be pristine (and vice versa, for a clean FT over a polluted MBL). Could you address this caveat more explicitly?

We agree with the reviewer that AOT represents a vertically integrated quantity. In this study, the SEVIRI-retrieved AOT is integrated only for the column above the cloud, as the MBL is cloud-filled. Thus, it does not provide information about aerosol presence within the boundary layer. Our analysis focuses on assessing whether the above-cloud AOT is co-located with regions of efficient entrainment as identified by NAME simulations, and thereby whether aerosols are present in the FT within these efficient entrainment regions.

In the manuscript, we first highlight that the above-cloud AOT co-located with FT air parcels remained mostly high during efficient mixing periods indicated by NAME simulations (Cases 1 and 3). This indicates that FT BB aerosols existed over the efficient mixing area. BB aerosols may entrain from the FT into the MBL, if the bottom of FT BB layer is near the cloud top. Then, we highlight that the above-cloud AOT co-located with BL air parcels was enhanced when BL air parcels approached the efficient mixing area of FT air parcels (west of 0°E). This indicates that once FT BB aerosols could entrain into the MBL over efficient mixing periods, they could be subsequently advected by the MBL south-easterly winds toward Ascension Island area. Since the above-cloud AOT is a column integrated abundance and the aerosol layer may be vertically separated from the cloud top, this analysis does not unequivocally demonstrate entrainment of BB aerosols at these locations. Nevertheless, it is a necessary condition that elevated FT aerosol abundance is co-located with regions of efficient entrainment for the MBL to receive inputs of BB aerosols. Meteorological analysis of the SEA in August 2017 supports the connection between FT BB layers and the cloud top (Ryoo et al., 2022). The low-level easterly jet in early and late August allowed the transport of African BB plumes in the low FT and thus the possibility that BB aerosols were entrained into the MBL in efficient mixing regions.

5. Ln 490-510 is a very lengthy explanation mixing literature review and some interpretations. Some of the literature review could be moved to the introduction, which would allow the interpretations to stand out more convincingly.

We thank the reviewer for this suggestion. We have made the explanation succinct regarding literature. The revised manuscript is:

Figs. 9c and 9d show the $AOT_{weighted}$ along the FT and BL air parcel transport pathways respectively, for the three case

studies. High AOT_{weighted} values show the co-located and co-temporaneous abundance of African BB aerosols within the FT column above the cloud layer along the simulated transport pathway. For BB-impacted MBL cases (Cases 1 and 3), the AOT co-located with FT air parcels (black lines in Fig. 9c) remained mostly high during the periods of efficient mixing indicated by the NAME simulations. This indicates that FT BB aerosols existed over the efficient mixing area and so could be entrained from the FT into the MBL if the bottom of FT BB layer is near the cloud top. Along BL transport from southeast to northwest over the SEA, the AOT co-located with BL air parcels (black lines in Fig. 9d) was enhanced when approaching the region of efficient mixing of FT air parcels (west of 0°E). This further indicates that once FT BB aerosols could entrain into the MBL over efficient mixing periods, they could be subsequently advected by the MBL south-easterly winds toward Ascension Island area. Since the above-cloud AOT is a column integrated abundance and the aerosol layer may be vertically separated from the cloud top, this analysis does not unequivocally demonstrate entrainment of BB aerosols at these locations. Nevertheless, it is a necessary condition that elevated FT aerosol abundance is co-located with regions of efficient entrainment for the MBL to receive inputs of BB aerosols. **Meteorological analysis of the SEA in August 2017 supports the connection between FT BB layers and the cloud top (Ryoo et al., 2022). The low-level easterly jet in early and late August allowed the transport of African BB plumes in the low FT and thus the possibility of BB aerosols entrainment into the MBL in efficient mixing area.** In comparison, the AOT in the clean-MBL case (Case 2) was continuously low along both FT and BL transport (red lines in Fig. 9c and 9d), demonstrating a negligible contribution of BB pollution to the MBL over Ascension Island. **This is due to a strong mid-level easterly jet in mid-August (Ryoo et al., 2022), leading to a disconnection between FT BB layers and the BL top, thereby suppressing the entrainment of African BB aerosols into the MBL.**

Minor comments:

- some choices of words are a bit vague and confusing, for instance. For example, on Ln 28: « a greater variability was noted in more polluted clouds » is imprecise. Same on Ln 352 (« a greater variation »). Do you mean that the linear fit is weaker? Another example on Ln 295: « Larger ranges of N_d and LWC values ... a smaller range of R_e ». Do you mean that N_d and LWC values are larger on average and R_e values are smaller on average, or do you mean that there is a larger/smaller standard deviation in the observed distributions (to me, « range » suggests the latter). There are several other occasions where « range » is used in this way in the text (e.g., Ln 404, 572). Could you use more precise mathematical language to help readers better understand your point?

We thank the reviewer for the suggestions. We have rephrased the related descriptions to make the manuscript more precise. The revised manuscript includes e.g.

Droplet activation fractions were similar in clean and moderately BB-polluted ($\text{sub-}N_a < 700 \text{ cm}^{-3}$) clouds, while a weaker N_d - N_a correlation was noted in more polluted clouds.

The relationship between N_d and $\text{sub-}N_a$ follows a similar pattern in clean or moderately BB-impacted clouds ($\text{sub-}N_a < 700 \text{ cm}^{-3}$). The weaker correlation between N_d and $\text{sub-}N_a$ under more polluted conditions ($\text{sub-}N_a > 700 \text{ cm}^{-3}$) may be partly due to the variability in the MBL updraft velocity.

In-situ and SEVIRI observations consistently indicate higher N_d and smaller R_e values in the BB-impacted MBL compared

with the clean MBL.

Cloud layers with negligible entrainment mixing (average $LWC/aLWC$ values, ~ 1) present generally larger R_e values compared to those with greater entrainment mixing (lower average $LWC/aLWC$, < 0.83).

This resulted in increased N_d and LWC values but reduced R_e within BB-impacted clouds compared to clean clouds.

- Ln 50: affecting », not « affect ». The last part of the sentence (starting with « underscoring... ») sounds redundant and could probably be removed.

We have re-organized the introduction. The opening paragraph has been rewritten for conciseness and precision, and this requested sentence has been removed. The revised first paragraph now reads as follows:

Every year from July to October, seasonal wildfires across central and southern Africa account for about one-third of global carbon emissions from biomass burning (BB) (Roberts et al., 2009). The emitted smoke is frequently transported westward over the southeast Atlantic (SEA), where it often resides in the free troposphere (FT) and may entrain into the marine boundary layer (MBL) during its subsiding transport (Painemal et al., 2014; Adebiyi and Zuidema, 2016; Das et al., 2017). These transported BB aerosols exert complex radiative effects by absorbing and reflecting solar radiations, and by interacting with one of the world's largest semi-permanent stratocumulus cloud decks over the SEA. The SEA region is climatically important, due to a significant net cooling effect of these stratocumulus clouds through their strong reflection of solar radiation but a small positive longwave radiative effect (Wood, 2012).

- Ln 120: remove « within », correct « AfricaN continent »

The revised manuscript is:

*...which primarily focused on **regions westward of the African continent** and eastward of 0° E (Redemann et al., 2021)...*

- Ln 125: This study is obviously focused on stratocumulus clouds, and the study area is the Ascension Island, but traditionally, Sc are thought to form closer to the coast (e.g., areas defined by Klein and Hartmann, 1993), and then transition to cumulus clouds as they move westwards to Ascension Island, which might leave some readers confused. It would be good to address this. Since you actually already have a list of cloud types observed during the campaign in Table S1, it would be good to reference it in the main text.

Thanks to the reviewer's suggestions. More details regarding the suggestion have been added to the Introduction.

In this study, we characterize the vertical profiles of thermodynamics, aerosol properties, and cloud microphysics over Ascension Island. While the region is influenced by the transition from stratocumulus to cumulus clouds, associated with increasing sea surface temperatures (Gordon et al., 2018), this study focuses on assessing the effects of BB aerosols on stratocumulus cloud microphysics. More details of cloud types (stratocumulus or cumulus clouds) collected during the CLARIFY are provided in Sect. 3.2.

- Ln 138: «straight and level runs » not defined

We have added example flight patterns in Figure S1, to define “straight and level runs”.

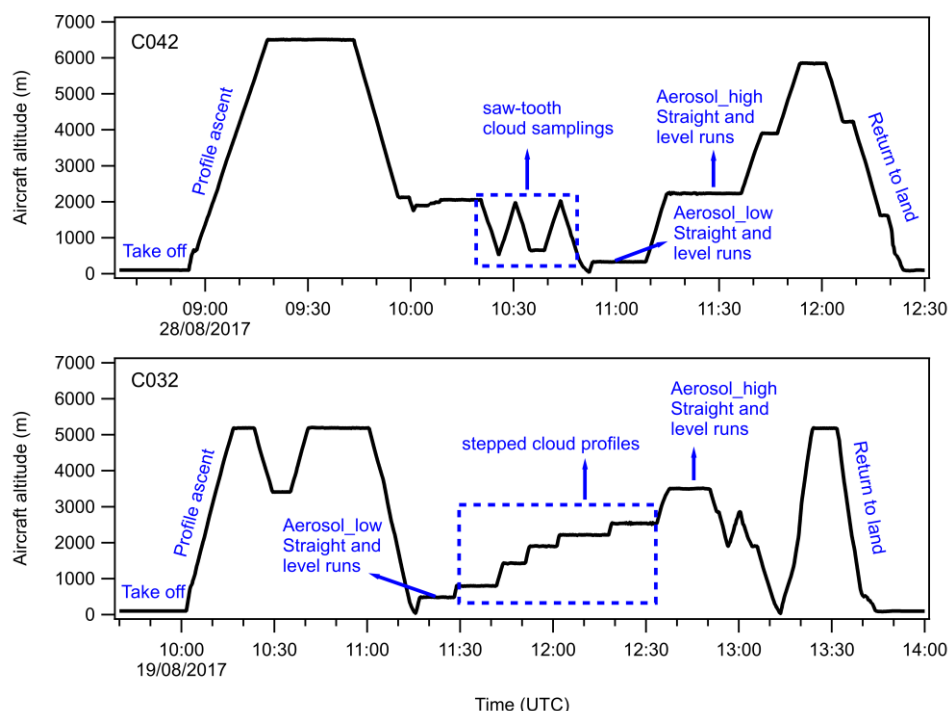


Figure S1. Example flight patterns on flight C042 (top) and C032 (bottom). The example flights provide an illustration of straight and level runs for aerosol characterization and saw-tooth and stepped profiles for cloud samplings.

- Ln 142: why would a POC event not be relevant to this study?

We have rephrased this part based on the suggestion.

Transit flights (C040-C041, predominately at high altitudes) and flights with mainly limited cloud samplings are not included in this study. Specific events (pocket of open cell, C052-C054) have been characterized in Abel et al. (2020), which are also excluded from this study.

- Ln 145: as written above, the instrument description could be shortened if the material is already published somewhere else. What would be interesting to add though is the typical size range of BBAs so that the readers can easily assess the suitability of the measurements.

Thanks to the reviewer for this suggestion. We have made the instrument descriptions more succinct.

The typical size range of BB aerosols has been added to the revised manuscript:

These aerosol instruments covered the typical size range of BB aerosols, primarily within the submicron range.

- Ln 182-185: the difference between the 0.1 g/m³ LWC and 0.2 g/m³ ‘bulk’ LWC thresholds was not clear to me

An LWC value over 0.01 g m⁻³ for 1 Hz measurements was used to define the low threshold for the presence of cloud.

Yes, there are minor differences between 0.1 g/m³ and 0.2 g/m³ of LWC. However, following previous studies (e.g. Lance et al., 2010; Bretherton et al., 2010), to eliminate the inclusion of optically thin clouds, a threshold of $N_d > 5 \text{ cm}^{-3}$ and bulk $\text{LWC} > 0.02 \text{ g m}^{-3}$ was used to perform statistically robust cloud sample analysis.

- Ln 190: (CTO) instead of (COT) - Ln 194: I believe there is an error in the equation, it should be $COT^{1/2}$ and not $COT^{-1/2}$

Thanks to the reviewer for pointing out the typo. The revised manuscript is:

*We also obtained the above-cloud aerosol optical thickness (AOT), cloud optical thickness (COT), R_e and **cloud-top height (CTH)** across the SEA region ($20^\circ W - 15^\circ W$; $30^\circ S - 0^\circ N$) ...*

The N_d was calculated assuming an adiabatic-like vertical stratification (Painemal et al., 2012):

$$N_d = 1.4067 \times 10^{-6} \left[cm^{-1} \right] COT^{\frac{1}{2}} R_e^{-\frac{5}{2}} \quad (4)$$

- Ln 196: Did you check the quality of your SEVIRI products close to sunrise and sunset times? There might be biases around those time, this could impact your results if sunrise/sunset points are colocated to the backtrajectories.

Yes, we have checked the quality of SEVIRI products close to UTC sunrise and sunset times. In addition, some of sunrise and sunset SEVIRI products were missing or unreliable when co-located with simulated transport, which were not included in the analysis.

- Ln 197: The NAME description section got me a bit confused. I did not understand why there seems to be 2 sets of backtrajectories, one at 3-hr resolution, and the second one at 15-min resolution. What is the point of having both sets of trajectories? Why use NAME backtrajectories vs. using the already produced HYSPLIT ones from the Haywood et al., 2021 paper?

Backward-dispersion and back-trajectory simulations both trace air parcel but differ fundamentally in how they treat transport and mixing:

	Back-trajectory	Backward-dispersion (what we did)
Purpose	Identify the origin and core transport path of air parcels arriving at a receptor	source region contributions to observed concentrations
Output	Trajectory lines	“footprint” fields showing where parcels most likely originated
Process	Advection only	Turbulence, deposition, chemistry

In this study, we conducted backward-dispersion simulations. To investigate vertical distributions and the transport of original air parcels, the model output instantaneous 3D air parcel footprints **every 3h** during the 7-days backward dispersion simulations. In the original manuscript, we also conducted Back-trajectory simulations (output resolution = 15min) to examine the entrainment rates. To avoid misleading the readers, we have deleted the back-trajectory simulations in the revised manuscript and focus on the dispersion results.

Haywood et al. (2021) employed HYSPLIT back trajectories to identify main source regions. However, this approach is not able to answer the vertical transport and mixing processes in this study, which is captured by our NAME backward-dispersion simulations.

- Ln 230: here you introduce one definition for the inversion height, but later (Ln 246) you use a different one, why is that?

Thanks to the reviewer for pointing out the repeated definition and typo. To avoid misleading the readers, more details

have been added to the manuscript. The revised parts in Sects. 2.2 and 3.1 are:

Sect. 2.2: *Here, we estimated the z_i over the SEA, using the outputs of 3D meteorological parameters. The z_i was quantified as the height at which the vertical gradient of liquid water potential temperature (θ_l) is the largest, and there is also a steep decrease in humidity (Jones et al., 2011). The θ_l was estimated following Eq. (5).*

$$\theta_l = \theta - \frac{L}{c_p} q_l \quad (5)$$

Where θ is the potential temperature, L is the latent heat of vaporization for water ($2.5 \times 10^6 \text{ J kg}^{-1}$), c_p is the specific heat of dry air at constant pressure ($1005 \text{ J kg}^{-1} \text{ K}^{-1}$), q_l is the liquid water mixing ratio (g kg^{-1}).

Sect. 3.1: *Figure 2a shows complex vertical distributions of aerosol number concentrations (N_a , $0.1 - 3 \mu\text{m}$) from PCASP measurements for the CLARIFY flights used in this study, alongside the estimated z_i . The z_i estimates were derived from airborne measurements of θ and q_l , using Eq. (5).*

-Ln 263: The FT humidity comparison between clean and BB-impacted FT cannot be inferred from Figure 3, can it?

-Ln 265: what is the interpretation for this positive correlation between BBA and humidity in the FT?

To differentiate the clean and BB-impacted FT, the vertical profiles of q_l (Fig. 3b) are now colored by three periods. In addition, more details about the positive correlation between BB plumes and humidity in the FT have been added to the manuscript:

Regarding the above two comments, the revised version is:

Figure 3b shows that FT humidity was generally higher on days with the presence of FT BB plumes (Periods 2 and 3, red and black profiles) compared to clean FT cases (Period 1, blue profiles). This pattern is consistent with previous studies, suggesting a positive correlation between BB plume strength and atmospheric water vapor content in the FT over the SEA (Pistone et al., 2021). The covariation between plume strength and humidity is attributed to the mixing between the moist, smoky continental boundary layer and the dry, clean FT. Consequently, humid BB plumes above the boundary layer can be advected to the SEA, enhancing FT humidity in the presence of transported smoke plumes (Pistone et al., 2021).

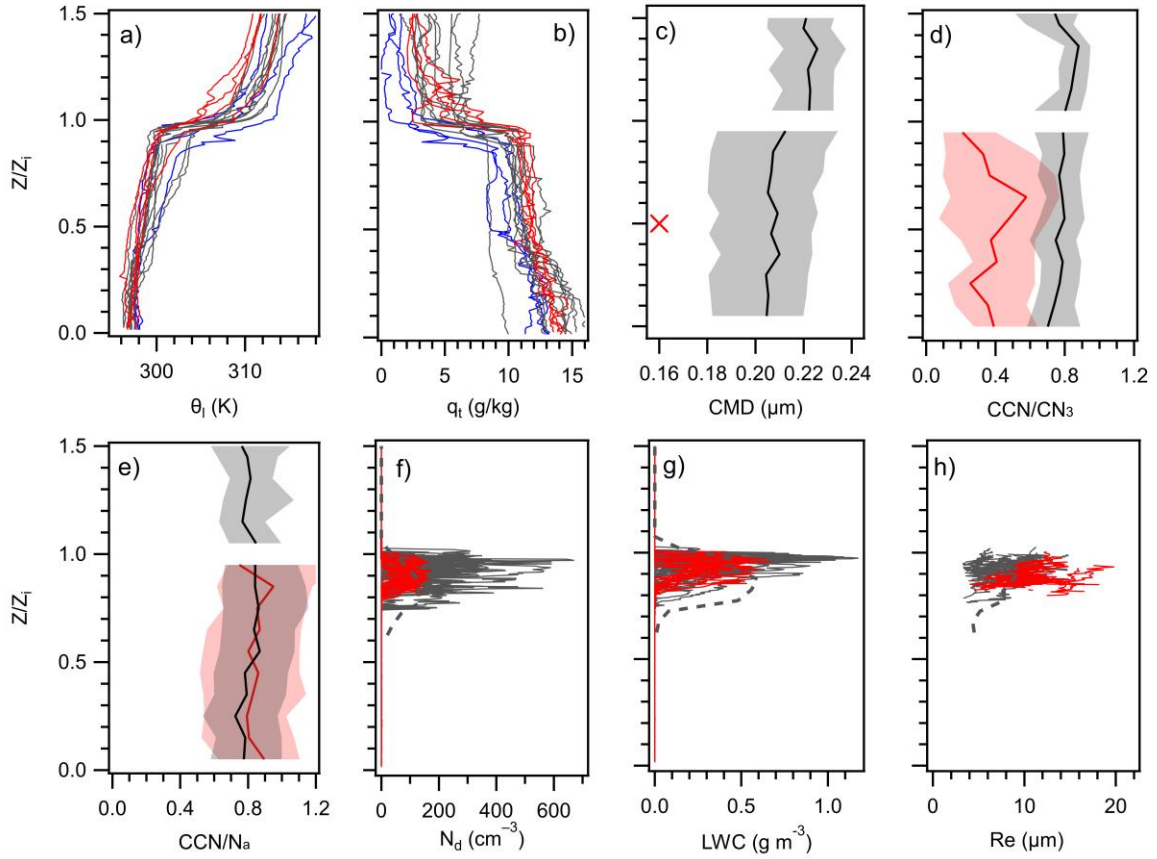


Figure 3. a, b) Average vertical profiles of a) liquid water potential temperature (θ_l , K), b) total water mixing ratio (q_t) for each flight used in this study. Blue, red and black lines represent measurements from Periods 1, 2 and 3 respectively. c-e) Summarized profiles of c) aerosol count median diameter derived from the PCASP (CMD, μm) and d,e) the ratio of CCN ($\sim 0.2\%$) to condensation nuclei ($D_a > 3 \text{ nm}$ from the CPC) (CCN/CN_3) and accumulation aerosol concentration ($D_a > 0.1 \mu\text{m}$ from the PCASP) (CCN/N_a) under polluted (black) and clean (red) conditions. Solid lines and shades represent median values and range from 10 to 90 percentiles. The red cross marker in Fig. 3c represents the average CMD in accumulation mode from measurements within the clean MBL. f-h) Vertical profiles of f) cloud droplet number concentration (N_d , $\# \text{cm}^{-3}$), g) liquid water content (LWC, g m^{-3}) and h) cloud effective radius (R_e , μm) in sampled continuous cloud layers. Red lines represent cloud measurements in the clean MBL, and grey lines represent cloud measurements in the BB-impacted MBL. It is noted that average vertical profiles of cloud properties from flight C032 are also provided in Figs. 3f-h (grey dashed lines). The y-axis uses a height scale normalized by inversion height (z_i).

- Ln 269: Could the MBL CMD simply be lower because the BBAs are mixed with smaller aerosols (e.g. sulfate, sea salt, etc.) in the MBL but not in the FT?

Thanks to the reviewer for the suggestion. The revised manuscript is:

Figure. 3c shows that the CMD of aerosols in the BB-impacted MBL was 10 – 15 % lower than in the FT BB plumes, which is probably attributable to some processes occurring in the MBL such as **mixing with marine emitted small particles** (i.e. marine sulfate) and aerosol removal by drizzle (Wu et al., 2020).

- Ln 279: CCN/CN₃ and not Na/CN₃

Here, we want to show that the majority of aerosols detected in the clean MBL were Aitken mode particles in the size

below 0.1 μm . Thus, it is **the ratio of N_a/CN_3** rather than CCN/CN_3 .

To make the description clearer, the revised manuscript is:

The aerosols in the clean MBL were smaller than in the BB-impacted MBL during CLARIFY. The average ratio of N_a/CN_3 in the clean MBL was calculated to be 0.40 ± 0.26 , also suggesting that the majority of submicron aerosols detected by the CPC (3 nm to 1 μm) in the clean MBL were Aitken mode particles ($< 0.1 \mu\text{m}$).

- Ln 288-291: as per my comment above, it would be helpful to already include a description of observed cloud types during CLARIFY in the introduction, to contextualize the method and results.

Thanks to the reviewer's suggestions. More details regarding the suggestion have been added to the Introduction.

While the region is influenced by the transition from stratocumulus to cumulus clouds, associated with increasing sea surface temperatures (Gordon et al., 2018), this study focuses on assessing the effects of BB aerosols on stratocumulus cloud microphysics. More details of cloud types (stratocumulus or cumulus clouds) collected during the CLARIFY are provided in Sect. 3.2.

- Ln 292: near *cloud* top (at the end of the line)

The revised manuscript is:

*The profiles of N_a (Fig. 3f) remain fairly constant, with few cases in the BB-impacted MBL (i.e. profiles in flights C029 and C038) presenting an increase **near cloud top**.*

- Ln 321: what does a « relatively » negative correlation mean? Relative to what?

The revised manuscript is:

*An **overall** negative correlation was observed between sub- N_a and CTtoAB.*

- Ln 345-346: could you provide the reference for these numbers again?

The Figures have been referred to for these numbers.

During CLARIFY, the BB-impacted MBL had substantially enhanced sub- N_a ($212 - 1183 \text{ cm}^{-3}$, black markers in Fig. 5a) compared to the clean MBL ($56 - 315 \text{ cm}^{-3}$, red markers in Fig. 5a).

- Ln 347: maybe « indicated » could be replaced by another verb, like « hypothesized »

The “indicated” has been replaced by “hypothesized”.

- Ln 353: remove « recent »

The “recent” has been deleted.

- Ln 361: how were these contact profiles selected?

To make the description clearer, the revised manuscript is:

When contact profiles had average $\text{LWC}/a\text{LWC}$ values close to 1, this suggests near-adiabatic profiles with negligible

mixing between cloudy and non-cloudy air. ... Notably, some of the near-adiabatic profiles (blue dashed box highlighted in Fig. 5a) displayed lower droplet activation fractions at similar sub- N_a levels, compared to other profiles.

- Ln 362: what does a « central » relationship mean?

To make the description clearer, the revised manuscript is:

Notably, some of the near-adiabatic profiles (blue dashed box highlighted in Fig. 5a) displayed lower droplet activation fractions at similar sub- N_a levels, compared to other profiles.

- Ln 365: just a note that entrainment of warmer and drier FT air could also lead to cloud droplet evaporation and decreases in Nd (cases of extreme inhomogeneous mixing, see Hill et al., 2009)

Thanks to the reviewer for the insightful comment. Yes, we agree that entrainment of warmer and drier FT air could also lead to cloud droplet evaporation and may decrease N_d . However, our observations suggest that contact profiles with greater entrainment generally promoted additional droplet nucleation and eventually enhanced N_d , compared to near-adiabatic profiles. We have added the reviewer's suggestion to the revised manuscript:

*When contact profiles had average $LWC/aLWC$ values close to 1, this suggests near-adiabatic profiles with negligible mixing between cloudy and non-cloudy air. In contrast, other contact profiles had lower average $LWC/aLWC$ values (0.34 – 0.83), indicating greater entrainment mixing of aerosols from above-cloud into the cloud layer at the place of observation. Notably, some of the near-adiabatic profiles (blue dashed box highlighted in Fig. 5a) displayed lower droplet activation fractions at similar sub- N_a levels, compared to other profiles. **Although cloud-top entrainment of warmer, drier FT air could cause cloud droplet evaporation, our observations suggest that contact profiles with greater entrainment generally promoted additional droplet activation and enhanced N_d , compared to near-adiabatic profiles.***

- Ln 371: Can you quantify the goodness of fit (for instance with the R^2 for the linear regression between $\log N_d$ and $\log N_a$)?

- Ln 372/373: from the confidence intervals, it looks like the fit is quite uncertain. How should these parameters be used if there are so uncertain ?

Thanks to the reviewer for the above two suggestions. The positive correlation between N_d and sub- N_a was strong ($r = 0.93$, $p < 0.01$) in clean or moderately BB-impacted clouds (sub- $N_a < 700 \text{ cm}^{-3}$), while a weaker correlation was observed in more polluted clouds (sub- $N_a > 700 \text{ cm}^{-3}$). The relationship between N_d and sub- N_a follows a similar pattern in clean or moderately BB-impacted clouds, which is regarded as an aerosol-limited regime. Following the reviewer's suggestions, we quantified the power law fit ($N_d \sim \alpha N_a^\beta$, with tight confidence intervals) and its goodness in aerosol-limited regime, for application in future studies.

Since droplet activation displayed similar behavior in clean and moderately BB-polluted clouds (sub- $N_a < 700 \text{ cm}^{-3}$) (Fig. 5a), a strong linear correlation ($r^2 = 0.87$) was observed between $\log(N_d)$ and $\log(\text{sub-}N_a)$. Data in this aerosol-limited droplet activation regime (sub- $N_a < 700 \text{ cm}^{-3}$), yielded $\alpha = 0.64$ (0.20 – 1.28) and $\beta = 0.93$ (0.79 – 1.04), which we recommend as representative parameters for application in future studies.

- Ln 382: Do you have a more detailed hypothesis of why SEA BBAs might have a better CCN ability compared to other

regions? (based on the literature?)

Regarding the comment, more discussions of CCN ability between different BB regions have been added to the manuscript. The revised manuscript is:

Our observations suggest a small difference in the response of N_d to N_a between BB-impacted and clean MBL profiles. This is likely due to their comparable CCN activation abilities of accumulation-mode aerosols under two MBL conditions (as discussed in Sect 3.2). In contrast, a previous study reported higher droplet activation fractions for the cleaner MBL compared to the BB-impacted MBL over the Pacific Ocean (Mardi et al., 2019). The discrepancy likely stems from differences in their droplet activation behaviors of transported BB aerosols between the studies. The β value for BB-impacted MBL cases in this study (0.71 (0.42 – 0.92)) is in a higher range than that reported for BB-impacted areas off the California coast of North America (0.26 (0.15 – 0.42)) in Mardi et al. (2019). A key factor contributing to the different droplet activation behaviors of transported BB aerosols, may be the variability in aerosol chemical composition and CCN activity, which depends on source combustion conditions and aging process (Wu et al., 2020; Farley et al., 2025). Submicron BB aerosols from western U.S. wildfires have been reported to be dominated by organic (~90%) with minimal inorganic content (<2%) from near-source to regional scales (0.5 hours to several days) (Farley et al., 2025). In contrast, highly aged African BB aerosols were reported to contain ~35% inorganic mass (Wu et al., 2020). This implies that highly aged African BB aerosols are more hygroscopic, as inorganics typically have higher hygroscopicity than organics on a global scale (Pöhlker et al., 2023). Additionally, transported BB aerosols from western US wildfire presented similar accumulation-mode aerosol size distributions to this study (Laing et al., 2016). Consequently, the CCN activation ability of transported African BB aerosols in this study is broadly higher than that reported for aged BB aerosols from Western/Northern American wildfires (CCN/CN = 0.11 – 0.62, at SS = 0.2 – 0.5%) (Pratt et al., 2011; Zheng et al., 2020), leading to the observed differences in their N_a - N_d relationship between these studies.

- Ln 410: in this paragraph, what can the given fit be interpreted, for instance does it tell us anything about the adiabaticity of the sampled clouds?

Thanks to the reviewer for the insightful comment. More discussions have been added to the manuscript:

Figure 7b shows the relationship between cloud-layer mean R_e versus the average ratio of LWC/N_d for all analyzed profiles. BB-impacted and clean MBL profiles yielded similar exponent (b) values of (0.34 ± 0.01) and (0.33 ± 0.01) respectively, which are close to the exponent (~0.33) validated in previous BB aerosol-cloud studies such as in Amazon (e.g. Reid et al., 1999). Previous studies have reported a theoretical exponent of $b = 1/3$ for adiabatic clouds, where droplet growth is dominated by condensation without entrainment (Burnet and Brenguier, 2007). In this study, the estimated ratios of $LWC/aLWC$ (~0.3 – 1, Fig. S5c) suggest the occurrence of entrainment mixing, nevertheless, the empirical b values near or slightly exceed ~1/3. This indicates that entrainment processes were likely dominated by homogeneous mixing, which could proportionally reduce LWC and N_d and thus preserve $b \sim 1/3$, particularly in clean cases (Burnet and Brenguier, 2007). In BB-impacted cases, entrainment of aerosol-rich air may also supply additional CCN, increasing N_d disproportionately to LWC reduction. This entrainment of additional CCN could yield cases with $b > 1/3$ in this study.

- Ln 428: maybe « linked » can be replaced with « co-located »

The “linked” has been replaced with “co-located”.

- Ln 450: in the methods, it is said that the trajectories are initiated from an altitude of 341m, yet on Fig. 7, it looks like they come from an altitude of 2 km, why is that?

As described in the Method section, for NAME MBL simulations, tracer particles were released within a height range in the MBL (341 ± 300 m) over Ascension Island. The original Fig. 7 (now Fig. 9) displays only the vertical distributions of source air parcels **originating from the FT region** along the backward-simulation time. Therefore, the figure specifically illustrates the vertical contribution of original FT air parcels along transport, before arriving at release area. This is designed to study **vertical transport and evolution of original FT air parcels**, and then the exchange or entrainment from the FT to the MBL. If the vertical distributions of original MBL air parcels were included in the figure, the starting point at time = 0 would appear at an altitude of $\sim (341 \pm 300$ m).

- Ln 478: Have you explained what the air-density-weight transformation means for AOT?

The definition of air-density-weight transformation has been added to the manuscript. The revised manuscript is:

The air-density-weighted AOT ($AOT_{weighted}$) was calculated for the co-located areas following Eg. (8).

$$AOT_{weighted} = \sum_{i=1}^N \left(\frac{Mass_i}{\sum_{i=1}^N Mass_i} \times AOT_i \right) \quad (8)$$

Where N is the total number of horizontal air parcel grids for co-located areas, $Mass_i$ is air parcel concentration ($g\ m^{-3}$) in each grid, $\frac{Mass_i}{\sum_{i=1}^N Mass_i}$ represents fractional air parcel concentration in each grid relative to the total air parcel density, AOT_i is the above-cloud AOT co-located with each air parcel grid.

- Ln 527: “observed larger droplets” is mostly true, but at -24h on Fig. 8, the droplet sizes are decreasing. Why is that?

Thanks to the reviewer for this insightful observation. When approaching Ascension Island, the MBL deepens and the deeper MBL also tends to be decoupled (Abel et al., 2020). The decoupled MBL is likely associated with an increased occurrence of drizzle due to the presence of larger droplets (Jones et al., 2011), which may result in slightly decreased N_d and R_e when approaching Ascension Island. The revised manuscript is:

*In Case 2 (red line in Fig. 10c), the CTH shows an increasing trend along the southeast to northwest transport path over the SEA. Concurrent MBL deepening and enhanced CTH could promote condensational growth, yielding larger droplets at the cloud top (Painemal et al., 2014). This agrees with the observed overall increase in R_e along the BL transport. **However, the deeper MBL also tends to be decoupled, and the decoupled MBL is likely associated with an increased occurrence of drizzle due to the presence of larger droplets (Jones et al., 2011). The occurrence of drizzle and associated deposition may result in reduced N_d and R_e , corresponding to the observed slightly reduction in the vicinity of Ascension Island.***

- Ln 573: “a stronger relationship” could be made more precise with “a stronger linear correlation”

The “a stronger relationship” has been rephrase to “a strong correlation”.

- Ln 576-577: Could it be interesting to add a quantification of relatively how much mass of aerosols is entrained along the transport vs at the place of observation? Or how much percent of the mass is entrained in the identified “efficient mixing regions”?

Thanks to the reviewer for this suggestion.

NAME simulations can provide the contribution and transport of original air parcels before arrival at the release location. However, it is not easy to distinguish the entrainment of aerosol at the place/time of observation, since the entrainment process is time dependent. Thus, we couldn’t quantify the relative contributions of aerosols entrained along the transport vs. at the place of observation from NAME simulations. However, in Sect. 3.3, using aircraft datasets, we have implied the influence of entrained BB aerosols to sub- N_a at the place of observation. In future work, it will be useful to quantify the relative contribution of long-range transport and local mixing.

In Fig. S8, we have provided cumulative exchange amounts of air parcels between the FT and the MBL along backward simulations, which could indicate the total entrained FT-to-MBL amounts.

- Ln 845: “Langitude” typo in the x-axis of Fig.1.

The typo has been corrected.

- Ln 845: why are the fire counts cumulated over August 2017 only, and not over the exact study period?

The fire counts have been cumulated over the campaign period, and Fig. 1 has been modified accordingly.

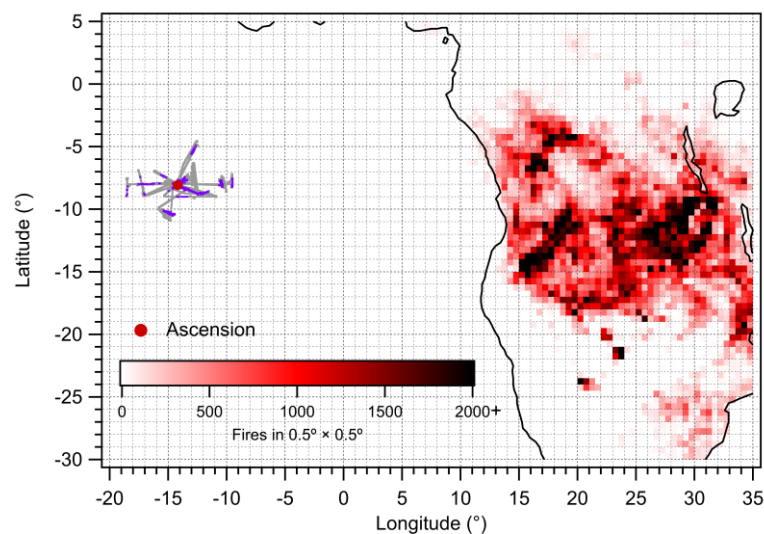


Figure 1. Tracks of the CLARIFY flights used in this study, cloud sampling periods during tracks ($N_d > 5 \text{ cm}^{-3}$ and bulk $LWC > 0.02 \text{ g m}^{-3}$) are highlighted in purple colour. Fire density maps are counted over the African continent, showing $2^\circ \times 2^\circ$ bins of the number of MODIS-detected fire during the campaign period.

- Ln 860: the f) and g) labels have been swapped compared to what is shown in the figure’s subplots

The labels have been revised.

f-h) Vertical profiles of 1-hz f) cloud droplet number concentration (N_d , $\# \text{ cm}^{-3}$), g) liquid water content (LWC , g m^{-3}) and h) cloud effective radius (R_e , μm) in sampled continuous cloud layers.

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