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***.

General comments:

Wu et al. presented aircraft observations on aerosol-cloud interactions over Ascension Island in the Southeast Atlantic, focusing on biomass burning aerosols from Africa. In-situ measurements of aerosol and cloud droplet properties, along with simulation experiments using the UK Met Office's Numerical Atmospheric Modelling Environment (NAME) and Unified Model, were conducted. This study also considered the effects of atmospheric conditions, such as in the marine boundary layer or free troposphere, on data analysis. Despite many language and presentation issues, the key findings are presented. In general, this study provides insights into cloud-aerosol interactions over remote oceans and shows the role of biomass burning aerosols under different atmospheric conditions. However, the manuscript requires substantial revision to improve the quality of data analysis and presentation. The main text should be better structured, and the results (see detailed comments below) need to be reanalyzed and better interpreted. Once these issues are addressed, the paper could be further considered for publication in Atmospheric Chemistry and Physics (ACP).

1. The abstract is overly redundant and needs revision for clarity and conciseness. It should be carefully restructured to avoid repetition and follow the guidelines provided by ACP, particularly regarding word limit and structure. Please refer to the ACP author guidelines for the required format https://www.atmospheric-chemistry-and-physics.net/policies/guidelines_for_authors.html.

   Thanks to the reviewer's suggestions on the abstract. The authors have revised the abstract for clarity and conciseness, 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 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 Arican BB aerosols in climate models, offering improved assessments of African BB impacts over the SEA.

2. The introduction is poorly developed and lacks a clear logical flow. In the first paragraph, the authors attempt to discuss the effects of biomass burning aerosols (BBAs) on cloud properties under different scenarios based on their vertical location relative to the cloud layer. However, the points are disorganized, and the lack of a clear leading sentence makes it difficult for readers, especially for those outside the field of cloud-aerosol interactions, to follow. The second and third paragraphs discuss the limitations of space-borne observations and model simulations respectively, but the authors fail to introduce specific research questions for the study. The statements in Lines 123-131 are too general and do not contribute enough detail. We would like to suggest the authors streamline the introduction and clarify the research questions to improve clarity and readability.

Thanks to the reviewer's suggestions on the introduction section. The authors have re-organized the introduction and added leading or/and conclusion sentences in each paragraph. For example:

"Most previous studies have focused on assessing the direct effect (solar absorption and scattering by BB aerosols) and the semi-direct effect (cloud adjustments in response to the direct radiative effect) of transported African BB aerosols."

"In contrast, the effect of BB aerosols on clouds by acting as cloud condensation nuclei (CCN) (known as the indirect effect) has received less attention compared with their direct and semi-direct effects over the SEA."

"However, quantifying the cloud response to BB aerosols over the SEA remains uncertain due to observational limitations." "More accurate in-situ measurements of aerosol-cloud relationship are needed to quantify the response of clouds to BB aerosols in this region."

"Another uncertainty in assessing BB aerosol-cloud interactions over the SEA arises from model biases in simulating the vertical distribution of transported BB aerosols."

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).*

3. The Cloud Droplet Probe detects cloud droplets down to 2.0 µm. How does overlooking droplets smaller than 2.0 µm affect the investigation and conclusions?

Thanks to the reviewer for raising the potential that droplets smaller than 2.0 µm may be overlooked. Typically cloud droplet diameters are much greater than this size. However, to check the potential influence of cloud droplets smaller than 2.0 µm—below the detection limit of the Cloud Droplet Probe (CDP), the authors did a statistical analysis of measured cloud droplet number and volume size distributions obtained from CDP measurements. Figure R1 shows the number (left axis) and volume (right axis) size distributions of cloud droplets sampled during the CLARIFY flights used in this study. The size distributions indicate that droplets smaller than 2.0 µm contributed minimally to cloud droplet volume concentrations and liquid water content.

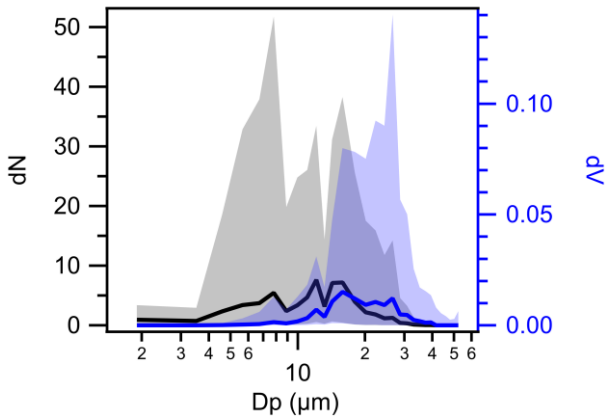

Figure R1. Droplet number (left axis) and volume (right axis) size distributions of clouds sampled during the CLARIFY flights used in this study. The line and shades represent medians, and ranges from 10 to 90 percentiles.

4. For Figure 3, the authors need to provide evidence or results to clearly classify biomass burning (BB)-impacted cases and those free of BB impacts. The manuscript mentions this classification, stating that "From 21st to 25th Aug, the MBL

became cleaner, with BB pollution predominantly in the FT. From 26th to 31st Aug, the MBL was BB-impacted again, with BBAs observed in both the MBL and FT." However, more detailed results and clear classification criteria should be unambiguously provided.

Thanks to the reviewer's suggestions on classification criteria. More details of the classification criteria for BB pollution have been added in the manuscript:

Following established criteria used in previous CLARIFY studies, the presence of BB-pollution was defined using thresholds of carbon monoxide above 83 ppb and black carbon above 0.1μg m^{-3} (Wu et al., 2020; Haywood et al., 2021).

5. For the results in Figure 4, further discussion is needed to gain more insights. For example, some above-N_a values (2000-2600 cm^{-3}) are significantly higher than sub-N_a. Are these high above-N_a data points due to N_d activation being limited by updraft velocity or supersaturation? For lower N_a values, both above- and below-cloud cases may be limited by aerosol particle concentrations. Thus, the differing correlation coefficients between N_d and N_a for sub-cloud and above-cloud cases may result from different N_a ranges. What happens if the same N_a range is compared for both cases? The coefficient difference may be smaller, and this should be further investigated.

Thanks to the reviewer's suggestion to further investigate N_d–N_a relationships for above- and sub-cloud cases. The objective of comparing N_d–N_a relationships between above- and sub-cloud cases is to investigate the activation mechanism (cloud-base activation and cloud-top activation through entrainment). For below-cloud cases, the N_d exhibited a stronger positive correlation with sub-N_a in the clean or moderately BB-impacted clouds (sub-N_a < 700 cm^{-3}), compared to more polluted clouds (sub-N_a > 700 cm^{-3}). Following the reviewer's suggestion, we have discussed correlation coefficients from different N_a ranges for below-cloud cases. We have also discussed the possible reasons why the correlation between N_d and sub-N_a was weaker in more-polluted clouds, e.g., differences in updraft velocity and the strength of cloud-top entrainment processes. The overall correlation of N_d with above-N_a was weaker and more complex than sub-N_a at the place/time of observation, indicating that cloud base activation of N_a played a greater role as compared to cloud-top activation of N_a through entrainment. Following the reviewer's suggestion, we have discussed correlation coefficients from different N_a ranges for above-cloud cases. For polluted FT cases. the N_d exhibited a fairly positive correlation with above-N_a ($r = 0.74$, $p < 0.01$) when above-N_a was below 2000 cm^{-3}, while the correlation became weaker when above-N_a was above 2000 cm^{-3}. We have also discussed the possible reasons for the weak correlation between N_d and above-N_a at the place/time of observation, e.g., entrainment rate and time duration. The variation in correlations between N_d and above-N_a, e.g. a weaker correlation when above-N_a was above 2000 cm^{-3}, may be due to differences in the strength of cloud-top entrainment.

We have rephrased the description of correlation coefficients as below:

Figures 5a and 5b show the relationship between cloud-layer mean N_d and N_a concentrations for all the analyzed profiles, in terms of sub-N_a and above-N_a respectively The N_d exhibited a strongly positive correlation with sub-N_a, with a Pearson correlation coefficient (r) of 0.89 ($p < 0.01$, statistically significant) for all analyzed profiles. The positive correlation between N_d and sub-N_a was stronger ($r = 0.93$, $p < 0.01$) in the clean or moderately BB-impacted clouds (sub-N_a < 700 cm^{-3}), while a weaker correlation was observed in more polluted clouds (sub-N_a > 700 cm^{-3}). In contrast, the overall correlation between N_d and above-N_a was weak but statistically significant when considering all analyzed

profiles (r = 0.41, p < 0.01). For polluted FT cases (circle markers in Fig. 5b), the N_d exhibited a fairly positive correlation with above-N_a (r = 0.74, p <0.01) when above-N_a was <2000 cm^{-3}, while the correlation became weaker when above-N_a was >2000 cm^{-3}. These results suggest that the influence of above-N_a on cloud properties was weaker than sub-N_a at the place/time of observation, indicating that cloud base activation of N_a played a considerably greater role as compared to cloud-top activation of N_a through entrainment. Particularly, the cloud base activation of N_a could play a dominant role, on observation days characterized by a clean FT but BB impacted MBL (triangle markers in Fig. 5).

6. The results section lacks coherence and a clear storyline. A more organized presentation would improve readability. For example, first classify conditions with and without BBA influences, providing evidence of BBA presence. Air mass footprint and trajectory results should be presented earlier to highlight differences in aerosol sources. Then, present the properties (aerosol and cloud) of each classified case. Next, show the relationship between cloud-layer mean R_e and the average LWC/N_d ratio, followed by the development of the parameterization and comparisons with the literature. This is just a suggestion; the authors may find other ways to better present the results.

Thanks to the reviewer's suggestion on the result section. The authors have re-organized the structure of result sections. We first added one section to present BB pollution conditions during the campaign period and to provide original airmass sources and evidence of BB presence by employing NAME simulations. The added section is as follows:

3.1 BB pollution conditions and source analysis

Figure 2a shows complex vertical distributions of aerosol number concentrations (N_a, 0.1 – 3 μm) 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). Following established criteria used in previous CLARIFY studies, the presence of BB-pollution was defined using thresholds of carbon monoxide above 83 ppb and black carbon above 0.1μg m^{-3} (Wu et al., 2020; Haywood et al., 2021). Table S1 summarizes the BB pollution conditions in the FT and MBL respectively, for the 17 flights used in this study. During the CLARIFY campaign, BB pollution events were observed at different times to be solely in the MBL, or solely in the FT, or in both layers. Concurrent surface observations on Ascension Island (Zuidema et al., 2018) presented the same trend of MBL BB pollution as observed during the CLARIFY campaign. Based on the BB pollution conditions, the CLARIFY campaign can be divided into three periods: Period 1 from August 16 to 20, BB aerosols were observed to exist predominantly in the MBL, and the FT was mainly clean; Period 2 from August 21 to 25, the MBL became considerably clean, while the BB pollution existed predominantly in the FT; Period 3 from August 26 to 31, the MBL was BB-impacted again, and the BB aerosols were observed in both the MBL and FT.

NAME backward-dispersion fields are used to represent the horizontal footprint of air parcels transported over the past 7 days before arriving at the sampling area over Ascension Island. Figure S2 provides three examples of backward-dispersion fields (cases 1 to 3), with release times indicated by red markers in Fig. 2a. These cases illustrate distinct and representative transport pathways of original air parcels for three periods. Case 1 released tracers near the end of Period 1 (12:00 UTC, August 18). Cases 2 and 3 released tracers at the beginning of Periods 2 (12:00 UTC, August 21) and 3 (12:00 UTC, August 26) respectively, coinciding with shifts in MBL pollution conditions. FT dispersion

simulations (Figs. S2a–c) indicate that most FT air parcels over Ascension Island originated from westerly flow across the SEA and African continent. Polluted FT cases (Figs. S2b, c) show a substantially greater influence from African continental airmasses compared to the clean FT case (Fig. S2a). MBL dispersion simulations (Figs. S2d–f) suggest that MBL air parcels over Ascension Island mainly arose from clean oceanic flow that transported from the southeast to northwest over the SEA. Polluted MBL cases (Figs. S2d, f) also show contributions of westerly flow originated from African continent, while negligible continental influence in the clean MBL case (Fig. S2e). We calculated fractional contributions of airmass from African continent (20°S – 5°N, 9°W – 35°E) to Ascension Island area throughout the campaign period, differentiating between FT (Fig. 2b) and MBL (Fig. 2c) simulations. These contributions closely tracked the influence of BB plumes from African wildfire regions (Fig. 1, fire maps) throughout the campaign period. The temporal evolution of continental contributions aligns well with the observed FT and MBL pollutions over Ascension Island. NAME simulations evidence that the FT and MBL pollutions observed during the campaign were attributable to long-range transport of African BB plumes.

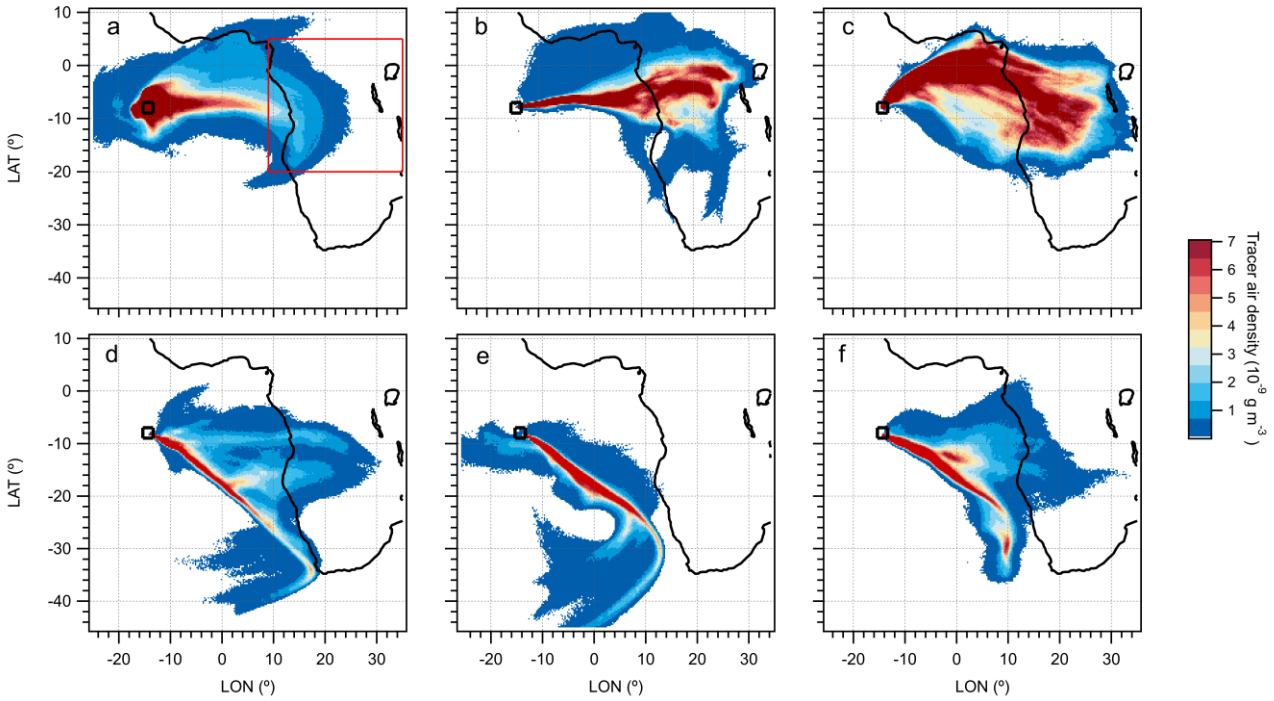

Figure S2. Examples of backward-dispersion fields from NAME simulations, illustrating representative horizontal footprints of original air parcels transported over the past 7 days before reaching Ascension Island area. a-c) Top and d-f) bottom panels are results from FT and MBL simulations respectively, for three example cases. Left two panels (a, d) are Case 1 released on 18 August 2017 at 12:00 UTC, middle two panels (b, e) are Case 2 released on 21 August 2017 at 12:00 UTC, and right two panels (c, f) are Case 3 released on 26 August 2017 at 12:00 UTC. The black boxes represent the release area around Ascension Island. The red box in a) represent the horizontal grids (20°S – 5°N, 9°W – 35°E) used for calculating fractional contributions of airmass from African continent to Ascension Island.

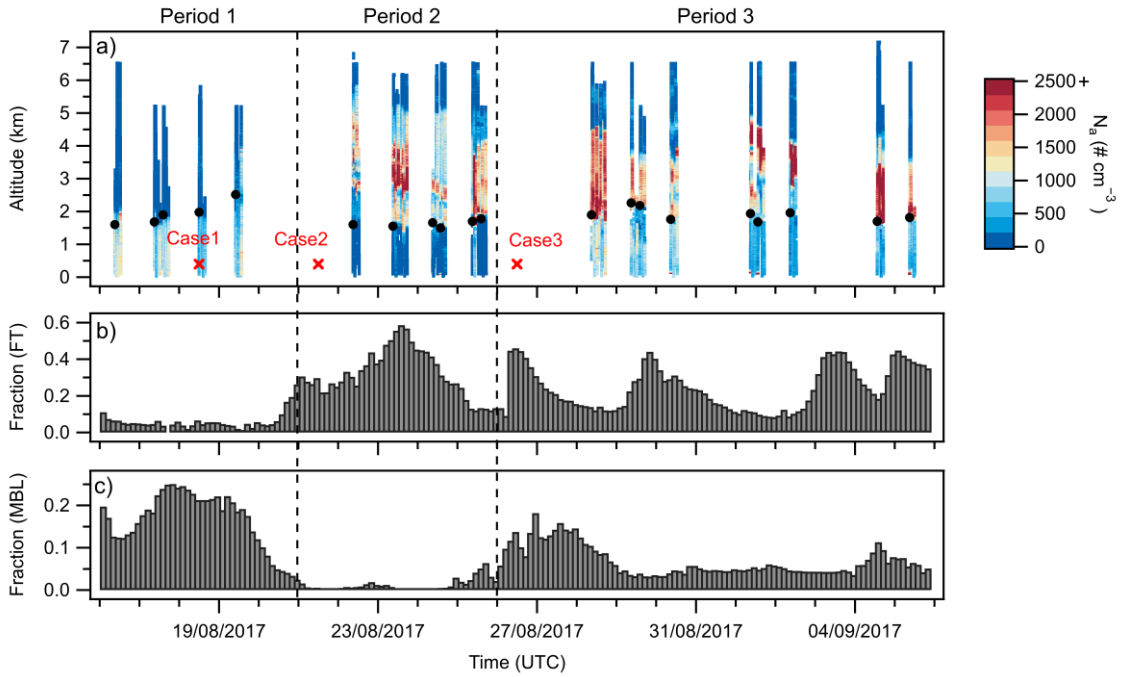

Figure 2. a) Vertical distributions of aerosol number concentrations (N_a, 0.1 – 3 μm) from PCASP measurements for the CLARIFY flights used in this study, alongside the estimated z_i. The red markers represent the release times for three NAME example cases. b, c) Fractional contributions of airmass from African continent (20°S – 5°N, 9°W – 35°E) to Ascension Island area throughout the campaign period, differentiating between b) FT and c) MBL simulations.

Then, as in the previous manuscript, we focused on observational results.

3.2 Vertical profiles of thermodynamics, aerosol and cloud

3.3 Aerosol-cloud interaction around Ascension

3.3.1 Aerosol-N_d relationship

3.3.2 R_e relationship with N_a and N_d

In the last section, we focused on the investigation of vertical transport and entrainment processes. We integrate air parcel analysis with satellite observations, 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.

3.4 Vertical transport history of observed MBL pollutants

3.4.1 Entrainment area

3.4.2 Cloud fields along transport

7. As shown in Figure 2, the flight altitude can reach 6 km, where ice crystals may be sampled, meaning that the aircraft could fly through mixed-phase clouds. What is the impact of sampling ice crystals? Were measures taken for the sampling inlet to account for this or to evaluate the influence? The authors refer to previous studies for details on instrumentation and experiments, but this may not provide enough information for new readers.

Thanks to the reviewer for raising this useful point. To interpret the potential influence of ice crystals, the authors did a statistical analysis of measured ice particle number concentrations obtained from a Two- Dimensional Stereo Optical Array Probe (2DS). The 2D-S (SPEC Inc, 2024) provided high sample volume images of cloud droplets and ice particles

ranging in diameter from 10 to 1280 μm. Ice particle concentrations were derived and categorized according to morphology using circularity definitions as described by SPEC Inc (2024). Figure R2 shows the vertical profile of a) ice particle number concentrations (N_{ice}) and b) cloud droplet number concentrations (N_d) sampled from the flights used in this study. At high altitudes, there were negligible amounts of ice crystals and cloud droplets. At low altitudes, the amounts of ice crystals were also minor compared to cloud droplets. Additionally, aerosol measurements inside clouds are suggested to be unreliable and in-cloud aerosol data have been removed in this study. Overall, the influence of ice crystals on PCASP results is suggested to be negligible.

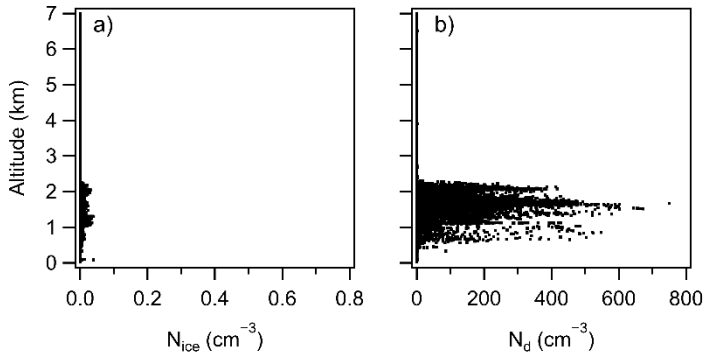

Figure R2. Vertical profile of a) ice particle number concentrations (N_{ice}) and b) cloud droplet number concentrations (N_d) sampled from the flights used in this study.

8. For high-speed aircraft in-situ sampling, correcting the sampled volume to the ambient volume (or standard conditions) is challenging due to air compression, which can cause significant volume differences. Did the authors account for the effects of air compression at high speeds? If so, how was it corrected or calculated?

Thanks to the reviewer for raising this important point. We have accounted for the effects of air compression during high-speed in situ sampling aboard the FAAM BAE-146 research aircraft. The ambient static pressure and temperature were obtained from the Rosemount pitot-static system, and stagnation (inlet) conditions were recorded near the aerosol sampling inlet. To correct the sampled volume to ambient atmospheric conditions, we applied the volumetric correction a described in in previous studies (Trembath et al., 2013).

We have added the brief description in the method section: "*Aerosol concentrations measured aboard the FAAM BAe-146 were corrected for ram air compression at inlet speed using the volumetric correction as described in previous studies (Trembath et al., 2013).*"

9. This study used a water condensation particle counter (CPC, model: 3786-LP). It is reported that water CPC undercounts hydrophobic organic-rich soot particles (Keller et al., 2013), potentially a component of BBAs. How does this undercounting affect the results and analysis in the study?

Thanks to the reviewer for highlighting the potential limitations of water-based CPC in detecting hydrophobic organic-rich soot particles (Keller et al., 2013). The BB aerosols observed in this study have undergone week-long transport and were highly aged. As shown in previous studies, aging processes during transport can reduce the hydrophobicity of BB organics due to their oxidation and internally mixing with secondary species such as sulfates (i.e. Pósfai et al., 2003). Thus, aged BB aerosols in this study are likely to be more easily detected than fresh BB aerosols by water CPCs.

Additionally, CPC measurements were used primarily to show general aerosol loading and the CCN activation fractions of submicron aerosols (CCN/CN_3), possible undercounting would not significantly affect the core conclusions.

We have added the brief description in the manuscript: "*The CPC used in this study is water-based and may undercount hydrophobic particles, particularly soot-rich BB aerosols (Keller et al., 2013). However, most of the sampled BB plumes have undergone week-long transport. Aging processes may have reduced the hydrophobicity of BB organics due to their oxidation and internal mixing with secondary species such as sulfates (Pósfai et al., 2003). Therefore, undercounting is expected to be minimal for the conditions of this study.*"

10. Relevant statements are needed to introduce and briefly discuss the results in the supplementary tables and figures. Simply presenting a figure or table without context is too abrupt.

    Thanks to the reviewer for the suggestion. The authors have added more descriptions regarding the supplementary tables and figures.

11. There are way too many abbreviations, which does not help the presentation but makes the reading more difficult. Please check through the manuscript and reduce the use of abbreviations.

    Thanks to the reviewer for the suggestion. The authors have reduced the use of abbreviations throughout the manuscript.

12. The language needs revision to improve readability. Many statements are too long, increasing the likelihood of errors and making them hard to follow.

    Thanks to the reviewer for the suggestion. The authors have reduced the use of long statements throughout the manuscript.

Specific comments:

Line 17: "vertical structures of thermodynamics, aerosol properties and cloud microphysics" sounds awkward

The revised manuscript is:

This study characterizes vertical profiles of thermodynamic conditions, aerosol properties, and cloud microphysics around Ascension Island during an aircraft campaign (August–September 2017).

Line 20-23: This sentence is too long and it is redundant.

The revised manuscript is:

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).

Line 23-24: The last part of the sentence can be more concise. And 'immediately' is not an appropriate adverb here.

The "immediately" has been deleted.

Line 47: It is stated that 'stratocumulus cloud (Sc) has been extensively studied'. However, the authors later on tried to bring

out the research question about the impacts of biomass burning aerosols (BBAs) from African wildfires on the Sc deck over the southeast Atlantic (SEA) Ocean. Isn't it self-contradictory?

The introduction has been revised. This sentence has been deleted.

Line 47-48: The statement in 'These clouds are climatically important, as they reflect a significant amount of solar radiation and exert only a small radiative effect in the longwave, leading to a net cooling effect.' Is not very clear. Is the small longwave radiative effect positive or negative? And relevant references are missing.

The revised manuscript is:

The SEA region is climatically important, due to the significant net cooling effect of these stratocumulus clouds through their strong reflection of solar radiation but small positive longwave radiative effect (Wood, 2012).

Line 68: should it be 'BB' or 'BBA layers'? Is it really necessary to abbreviate both BB and BBA?

The abbreviation of "BBA" has been replaced by "BB aerosols".

Line 69: What is 'sub-cloud relative humidity (RH)'? Please make a clear definition since it is not a standard or regular terminology and it is used quite often through the manuscript.

Line 69-70: The statement in 'In the smoky MBL, BBA heating tends to reduce the sub-cloud relative humidity (RH) and liquid water content, thereby decreasing the cloud cover Zhang and Zuidema (2019) may lead to misunderstanding that BBAs heat the cloud and thus decrease liquid water path and cloud cover. It would be better to specify it as BBA induced solar radiation absorption.

Regarding the above two comments, the revised manuscript is:

Conversely, in the smoky MBL, ***BB aerosol heating induced by absorbing solar radiation*** *tends to* ***reduce the humidity below the cloud****, thereby decreasing the cloud cover (Zhang and Zuidema, 2019).*

Line 74: The authors have defined the semi-direct effect in Line 67. It would be good to also define the indirect effect and shortly explain what is the difference between semi-direct effect and indirect effect.

The revised manuscript is:

Most previous studies have focused on assessing the direct effect (solar absorption and scattering by BB aerosols) and the semi-direct effect (cloud adjustments in response to the direct radiative effect) of transported African BB aerosols.

Line 79: how could the indirect effect of BBAs affect the cloud lifetime mentioned here? Is it a decrease or increase?

The revised manuscript is:

Recent modeling studies show that BB aerosols can increase cloud droplet number concentrations (N_d) and reduce droplet sizes, which may enhance cloud lifetime and coverage by reducing drizzle formation (Gordon et al., 2018; Lu et al., 2018).

Line 83: Specify the region since it is at the beginning of the paragraph.

The revised manuscript is:

However, quantifying the cloud response to BB aerosols over the SEA remains uncertain due to observational limitations. Satellite-based observations have been widely used to examine the vertical distance between the smoke layer base and the cloud top, which plays a role in understanding the impact of BB aerosol layers on cloud properties (Costantino and Bréon, 2010, 2013; Painemal et al., 2014).

Line 98-99: 'Recent models tend to show BBA layers descending rapidly when off the western coast of the African continent, resulting in BBAs layers that are too low in altitude over the SEA (Das et al., 2017; Gordon et al., 2018).' needs to be revised.
Line 100: 'the models'. Which models? They are not introduced beforehand.

Regarding the above two comments, the revised manuscript is:

Many simulations using Earth System models show that smoke layers descend too rapidly as they advect away from the African coast, thus the smoke layer heights were underestimated over the SEA (i.e. Das et al., 2017; Gordon et al., 2018; Shinozuka et al., 2020).

Line 122-123: Is it only because aircraft observations over SEA region is lacking? What is advantages or advancements for aircraft observations in this study in comparison to those conducted before?

The revised manuscript is:

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.

Line 129-130: It should be Spinning 130 Enhanced Visible and Infrared Imager (SEVIRI), instead of 'SEVIRI (Spinning 130 Enhanced Visible and Infrared Imager)'.

The revised manuscript is:

We combine air parcel analysis with Spinning Enhanced Visible and Infrared Imager (SEVIRI) satellite observations…

Line 136: It should be 7 September 2017 (British English) or September 7, 2017 (American English).

The time format consistently follows American English standards.

Line 137: What is the difference of 'temperature (T)' compared to 'potential temperature (θ)'? Is it ambient temperature?

The revised manuscript is:

The aircraft was equipped with a range of instruments to derive aerosol and cloud properties, as well as meteorological variables (e.g. ***ambient temperature (T, K)****, potential temperature (θ, K), and total water mixing ratio (q_t, g kg^{-1}, the total mass of vapor, liquid, and ice forms of water per unit mass of dry air)).*

Line 137: What is 'total water mixing ratio'? Definition is needed.

The revised manuscript is:

The aircraft was equipped with a range of instruments to derive aerosol and cloud properties, as well as meteorological variables (e.g. ambient temperature (T, K), potential temperature (θ, K), and ***total water mixing ratio (q_t, g kg^{-1}, the total mass of vapor, liquid, and ice forms of water per unit mass of dry air)).***

Line 138: Does it really help to abbreviate straight and level runs as SLRs? Maybe not very helpful for readers but one more abbreviation to keep in mind.

We have deleted the abbreviation of "SLRs".

Line 142: Again, is the abbreviation of 'POC' helpful and necessary?

We have deleted the abbreviation of "POC".

Line 151: Better delete 'accumulation mode' because some studies also define particles between 0.1 and 2.5 µm or between 0.1 and 1.0 µm as accumulation mode particles.

The "accumulation mode" has been deleted. The revised manuscript is:

Aerosol number concentration in the size range of 0.1 – 3 µm (N_a) was obtained by integrating the PCASP distribution.

Line 155: Manufacturer for the water CPC is missing.

The manufacturer for the water CPC has been added.

A model 3786-LP water-filled condensation particle counter (CPC, TSI Inc.)…

Line 161: Will the abbreviated 'STP' frequently used below? Please check through the manuscript for abbreviations and make sure only use it when it is really necessary.

We have deleted the abbreviation of "STP".

Line 166: Is the period the same for all observations on different days during the campaign?

Yes, the period for all SEVIRI observations on different days is the same during the campaign.

Line 176: To 'STP' or to the condition? Please use more scientific and formal language.

The revised manuscript is:

Airborne aerosol and cloud measurements reported here were corrected to the standard temperature and pressure condition (273.15 K and 1013.25 hPa).

Line 176-184: The introduction statements of Nd, Re, and liquid water content (LWC) using the three equations should be better organized. Simply putting all variables and parameters together seems too cursory.

The revised manuscript is:

From the CDP's cloud droplet spectrum, three key cloud microphysical parameters were derived: N_d, R_e, and liquid water content (LWC). The N_d was calculated by integrating the droplet size distribution, as described by Eq. (1).

$$N_d = \int n(r)\, dr \approx \sum_1^m n\,(r_i) \quad (1)$$

where $n(r_i)$ is the droplet number concentration in a particular size bin, and r_i is the middle radius for each of the size bins.

The R_e was calculated based on the Eq. (2).

$$R_e = \frac{\int r^3\, n(r)\, dr}{\int r^2\, n(r)\, dr} \approx \frac{\sum_1^m r_i^3\, n(r_i)}{\sum_1^m r_i^2\, n(r_i)} \quad (2)$$

The LWC was calculated based on the Eq. (3), assuming spherical droplets and a liquid water density (ρ_{water}) of 1 g cm^{-3}.

$$LWC = \frac{4\pi}{3}\, \rho_{water} \int r^3\, n(r)\, dr \approx \frac{4\pi}{3}\, \rho_{water} \sum_1^m r_i^3\, n(r_i) \quad (3)$$

Line 190: How could there be two 'COT's? 'cloud optical thickness (COT)' and 'cloud-top height (COT)'

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° W − 15° W; 30° S − 0° N) …*

Line 195-196: This sentence is repeating the one in Lines 165-166.

The repeating part has been deleted.

Section 2.2: A clear definition for sub-cloud and above-cloud regions may be necessary.

The revised manuscript is: *Of relevance to this work are the saw-tooths and stepped profiles to characterize cloud regions.* The definition for sub- and above-cloud are provided in Sect. 3.3.1. *In each profile with a continuous cloud layer, we averaged the N_a* ***within 200m above the cloud top*** *to obtain the* ***above-****N_a, and* ***within 200m below the cloud base*** *to obtain the* ***sub-****N_a.*

Line 206: How can people understand the 'reasonably well' performance of NWP if people do not go through the referred paper? It is too cursory to make such a statement.

The manuscript has been revised:

NAME was chosen as an appropriate model for this study because it uses high-resolution meteorological fields (~10 km × 10 km). It can predict transport over distances ranging from a few kilometers to the whole globe, and it has been used successfully in similar studies (i.e. Panagi et al., 2020).

Line 210-211: Isn't this sentence a repetition of the content introduced in the previous paragraph?

The NAME method section has been revised, and the repeated sentence has been deleted.

Line 225: Didn't the stratocumulus cloud defined as Sc in Line 46-47?

We have deleted the abbreviation of "Sc".

Line 230: What are the FT and BL products?

The revised manuscript is:

Instead of using the BL depth in NAME, the estimated z_i was employed to divide the column-integrated horizontal footprints into the FT and BL separately, distinguishing original air parcels as either from the FT or BL.

Section 2.3 can be shortened and more concise. It can be better structured. The introduction for 3D NAME experiments with Numerical Weather Prediction (NWP) model and the Unified Model (MetUM) is not clear. Which model is for what products? What is the difference between the two models? This is not easy for readers not from the field.

The Unified Model (MetUM) is a Numerical Weather Prediction (NWP) model developed by the UK Met Office. We have made the description more concise. The revised manuscript is:

The transport of released tracer particles was tracked backward over the past 7-days in the NAME, driven by the three-dimensional gridded (3D) meteorological fields from the UK Met Office's Unified Model (Brown et al., 2012).

Line 235-236: It is not clear enough by just mentioning the classification was introduced in a previous study. How? and what are the criteria?

More details have been added to the manuscript:

Following established criteria used in previous CLARIFY studies, the presence of BB-pollution was defined using thresholds of carbon monoxide above 83 ppb and black carbon above 0.1μg m^{-3} (Wu et al., 2020; Haywood et al., 2021). Table S1 summarizes the BB pollution conditions in the FT and MBL respectively, for the 17 flights used in this study.

Line 236: It should be 'Figure 2' but not 'Fig. 2' at the starting of a sentence. This is clearly specified in the journal guidelines. Please follow the guideline carefully and check through the manuscript carefully.

The error has been corrected throughout the manuscript.

Line 236: Does the number concentration of aerosol particles have a structure? Or it means the vertical distribution?

The revised manuscript is:

Figure 2a shows complex vertical distributions of aerosol concentrations (N_a, 0.1 – 3 μm) from PCASP measurements for the CLARIFY flights used in this study, alongside the estimated z_i.

Line 238: How could field observations be consistent with the dates? Such a statement does not make sense.

The revised manuscript is:

Concurrent surface-based observations on Ascension Island (Zuidema et al., 2018) presented the same trend of MBL BB pollution as observed during the CLARIFY campaign.

Line 248: An equation is needed to show how 'the θl was calculated from the T, θ and water mixing ratios'.

More details have been added to the manuscript:

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 (2.5×10^6 J kg^{-1}), c_p is the specific heat of dry air at constant pressure (1005 J kg^{-1} K^{-1}), q_l is the liquid water mixing ratio (g kg^{-1}).

Line 248: 'Fig. 3', the same comment as for Line 236.

The error has been corrected throughout the manuscript.

Line 251: What is lifting condensation level (LCL)? A clear definition is necessary since it shows up in the following discussions. It can also be clearly indicated in Fig.3a or 3b. What does it mean a surface layer? Whose surface? Or if it is a language issue, maybe better a flat layer or a layer with rather constant z/z_i.

More details have been added to the manuscript following the reviewer's suggestions:

The MBL generally showed a well-mixed layer with near-constant θ_l and q_t between the sea surface and the lifting condensation level (LCL, the height at which the relative humidity of an air parcel reaches 100% when lifted adiabatically), which is at an altitude of ~ 300 to 800 m (z/z_i = ~ 0.2 – 0.5). Above the LCL, there was another layer that extended to the base of the inversion.

Line 254: 'relatively' means the comparison to what?

The "relatively well-mixed" has been revised to "well-mixed".

Line 256: How can the decoupling parameters α_q and α_θ be derived? It would be good to provide the equations and explain it with more details somewhere (e.g., in the supplement but properly referred in the main text).

More details have been added in the manuscript regarding the estimation of the decoupling parameters:

The BL decoupling parameters α_q and α_θ, were also calculated from the respective profiles following Eq. (6) and (7) respectively, which represent the relative differences of the q_t and θ_l between the surface and the upper part of the BL respectively (Zheng et al., 2011).

$$\alpha_q = \frac{q_t(z_i^-) - q_t(0)}{q_t(z_i^+) - q_t(0)} \quad (6)$$

$$\alpha_\theta = \frac{\theta_l(z_i^-) - \theta_l(0)}{\theta_l(z_i^+) - \theta_l(0)} \quad (7)$$

Where $\theta_l(z_i^+)$ and $q_t(z_i^+)$ are θ_l and q_t at the level 50m above the inversion height, $\theta_l(z_i^-)$ and $q_t(z_i^-)$ are at the level 50m below the inversion height, θ_l (0) and q_t (0) are at the lowest measured level near sea surface.

Line 269: 'become' can be better replaced by 'act as'

The "become" has been revised to "act as".

Line 274: What is the definition of 'highly aged African BBAs in this study' and what are their property differences compared

to fresh ones?

More details have been added in the manuscript regarding the "'highly aged African BB aerosols":

In this study, the sampled African BB aerosols have undergone week-long transport and are defined as highly aged African BB aerosols (Wu et al., 2020). Compared to freshly emitted African BB aerosols, these highly aged BB aerosols exhibited larger particle sizes, enhanced fractions of inorganics and the loss of organics after atmospheric aging such as chemical oxidation and cloud processing (Wu et al., 2020; Dobracki et al., 2023). Due to atmospheric aging, the CCN activation fractions of these highly aged African BB aerosols are generally higher than fresher BB aerosols sampled over the African continent (CCN/N_a = 0.68, at SS = 0.3%) (Ross et al., 2003), but are comparable to transatlantic African BB aerosols observed over Amazon area during the dry season (the average ratio of CCN to aerosol concentrations in a range of 20 nm to 1μm is 0.83 ± 0.06 at SS = 0.5%) (Holanda et al., 2020).

Line 275: 'CN_{20}' was not defined in the main text and only appeared once.

The revised manuscript is:

the average ratio of CCN to aerosol concentrations in a range of 20 nm to 1μm is 0.83 ± 0.06 at SS = 0.5%.

Line 280: 'the majority of particles were in the size below 0.1 μm' refer to which kind of particles? Aerosol particles larger than 3 nm or CCN particles?

The revised manuscript is:

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 μm).

Line 284: 'more sulfate'. Yes, it was stated before that 'The submicron aerosols from marine emissions in the clean MBL were previously reported to be dominated by sulfate (~ 60%) and organic (~ 24%).' However, there is no evidence provided to show that aerosols in the clean MBL contain more sulfate.

The revised manuscript is:

*Although the clean MBL aerosols contained **enhanced fractions** of sulfate with high hygroscopicity, the average CCN/N_a (0.83 ± 0.20) was close to those in the BB-impacted MBL (0.82 ± 0.17), suggesting that the CCN activated fractions of accumulation-mode aerosols were similar between the clean and BB-impacted MBL conditions.*

Section 3.1:

1) It seems the second paragraph is mainly citing a previous study from Wu et al. (2020) but not presenting the results in Figure 3 in this study.

To avoid redundancy with Wu et al. (2020), vertical profiles of aerosol chemical composition under BB-impacted conditions have been provided in Supplementary Figure S2. In the main text, information of aerosol chemical composition in the clean MBL is referenced succinctly as "*The submicron aerosols from marine emissions in the clean MBL were previously reported to be dominated by sulfate (~ 60%) and organic (~ 24%) (Wu et al., 2020).*".

2) Figure 3d clearly shows that CCN/CN_3 under clean MBL conditions is lower than that under BB-impacted MBL

conditions and such a difference becomes smaller when it is in the FT. However, the CCN/N_a in Fig. 3e did not show such a dependence on the atmospheric conditions (in the MBL or FT). Discussions on this finding is missing.

Since background aerosols in the clean FT during Period 1 mainly fell below instrumental detection limits, FT analysis of aerosol properties (CMD, CCN/CN_3, and CCN/N_a ratios) only included those sampled during BB-impacted conditions. The red and black lines and shades in original Figs. 3c-e were summarized based on MBL pollution conditions, rather than indicating FT pollution conditions. To avoid misleading the reader, the authors have modified Figs. 3c-e and revised the manuscript.

In this study, vertical profiles of aerosol properties (CMD, CCN/CN_3 and CCN/N_a ratios) were summarized under clean (red line and shades in Figs. 3c-e) and BB-impacted (black line and shades in Figs. 3c-e) conditions separately. MBL analysis included both the clean and BB-impacted conditions. However, due to background aerosols in the clean FT during Period 1 being mainly below instrumental detection limits, the FT analysis (CMD, CCN/CN_3, and CCN/N_a ratios) was restricted to BB-impacted conditions.

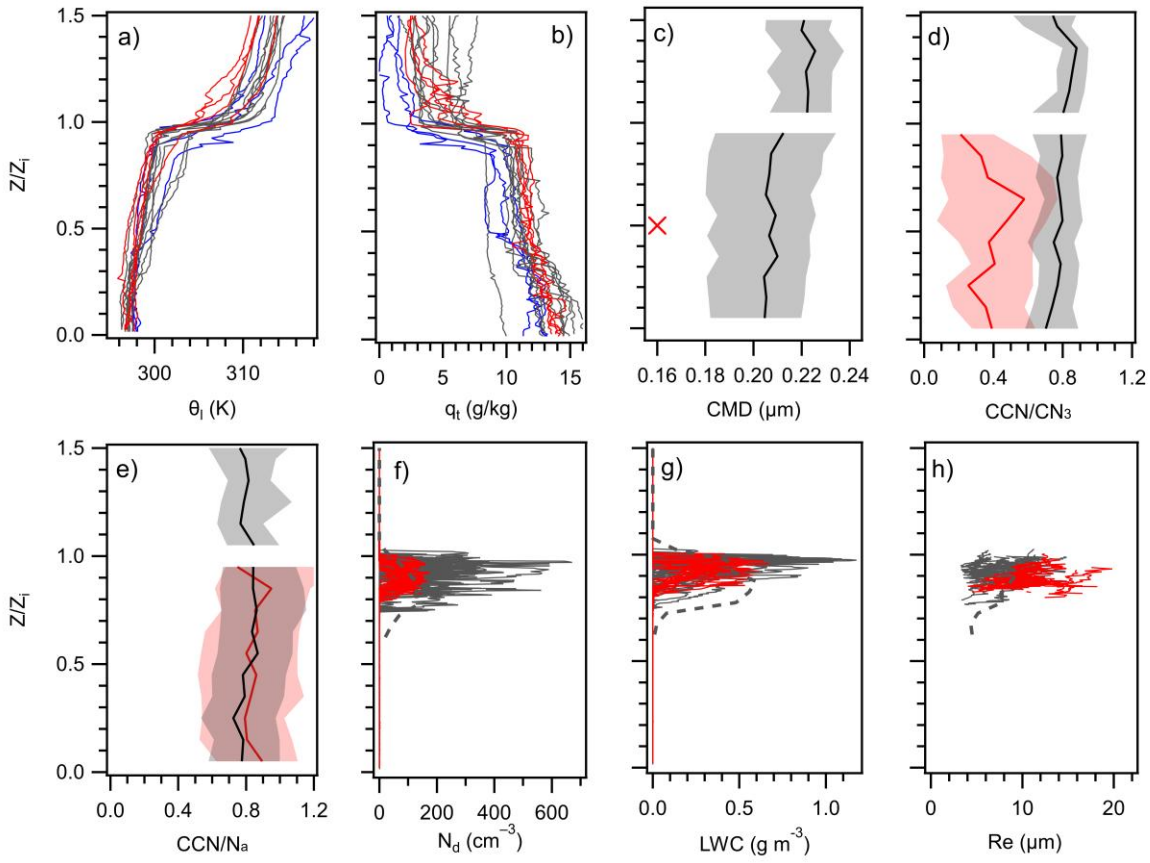

Figure 3. a, b) Average vertical profiles of a) liquid water potential temperature (θ_l, K), b) total water mixing ratio (q_t) for each flight used in this study. Blue, red and black lines represent measurements from Periods 1, 2 and 3 respectively. c-e) Summarized profiles of c) aerosol count median diameter derived from the PCASP (CMD, μm) and d,e) the ratio of CCN (~ 0.2%) to condensation nuclei (D_a > 3 nm from the CPC) (CCN/CN_3) and accumulation aerosol concentration (D_a > 0.1 μm 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 1-hz, f) liquid water content (LWC, g m^{-3}), g) cloud droplet number concentration (N_d, # cm^{-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).

3) The role of particles between 3 and 100 nm can also be investigated, which is also overlooked.

We thank the reviewer for pointing out the potential importance of the 3–100 nm size range.

The revised manuscript is: *Under both clean and BB-impacted conditions, the CCN/CN_3 ratios remained consistently below unity, whereas CCN/N_a occasionally exceeded unity, implying that a subset of Aitken mode particles (< 0.1 μm) contributed to CCN.*

4) In addition, why some data around z/z_i in Figs. 3c, 3d and 3e is missing?

Since observations near top of the MBL (z/z_i ~0.9 to 1) were sampling predominantly cloudy conditions rather than clear-air aerosols, the aerosol data was deleted. Consequently, there are missing gaps in Figs. 3c–e.

5) Moreover, frequently using 'some days' (Line 253 and 255) or 'sometimes' (Line 292) makes the statement less convincible. Please use solid (clearly presented and referred) results to support the argument.

Related description in the manuscript has been revised following the reviewer's suggestions, to make the statement clearer and more convincing:

During 6 flight cases (marked as type-d in Table S1), profiles above the LCL exhibited near-constant θ_l and q_t, indicating a fairly well-mixed upper layer and the presence of a decoupled stratocumulus deck. By contrast, in the remaining cases, the upper layer above the LCL was conditionally unstable, suggesting the presence of a stratocumulus-over-cumulus MBL (marked as type-e in Table S1) or a cumulus-capped MBL (marked as type-f in Table S1) structure.

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

Line 288-290: 'In Flight C032 with deep cumulus clouds, stepped profiles and SLRs were carried out and the average vertical profiles of cloud properties are provided.' Where was the result provided? Please refer to a figure or table.

The average vertical profiles of cloud properties for Flight C032 were provided in the original Figs. 3f-h. To make these profiles clearer, they are now shown as grey dashed lines in those panels.

The revised manuscript is: "*In Flight C032 with cumulus clouds, stepped profiles and SLRs were carried out. The average vertical profiles of cloud properties from flight C032 are also provided in Figs. 3f-h (grey dashed lines).*"

Line 299-301: Cloud condensation nuclei concentrations depend on cloud supersaturations. For the focus of this study, it is the contribution of N_a to cloud condensation nuclei numbers that impacts cloud microphysics.

The revised manuscript is:

Here, we use N_a as the metric instead of CCN concentrations to establish the aerosol impact on cloud microphysics, since CCN concentrations depend on cloud supersaturations. In this study, we focus on aerosols in the size above 0.1 μm that behaved as the dominant CCN (as discussed in Sect. 3.2) and impacted cloud microphysics.

Line 302: Should be subsection 3.2.1

The subsection has been added.

Line 310-311: What is the p value for the calculated Pearson correlation coefficients? It would be good to indicate the value in Figure 4 accordingly.

The p values have been added.

Line 312: Regarding the statement 'especially for the flights with clean FT but high Na in the BB-impacted MBL (dashed black box in Fig. 4b)', the data point with N_d close to 400 cm^{-3} in the black box looks like an outliner. There are only three data points in the box. It is far-fetched to treat it as a special case to investigate the correlations.

The dashed black box in the previous Fig. 4b highlights profiles sampled during Period 1, characterized by a clean FT but BB impacted MBL. In this case, activation of N_a at the cloud base played a dominant role in determining N_d. Therefore, the weak correlation was observed between N_d and above-N_a in these profiles, which should not be considered as outliers.

Regarding the above two comments, the revised manuscript is:

Figures 5a and 5b show the relationship between cloud-layer mean N_d and N_a concentrations for all the analyzed profiles, in terms of sub-N_a and above-N_a respectively The N_d exhibited a strongly positive correlation with sub-N_a, with a Pearson correlation coefficient (r) of 0.89 ($p < 0.01$, statistically significant) for all analyzed profiles. The positive correlation between N_d and sub-N_a was stronger ($r = 0.93$, $p < 0.01$) in the clean or moderately BB-impacted clouds (sub-N_a < 700 cm^{-3}), while a weaker correlation was observed in more polluted clouds (sub-N_a > 700 cm^{-3}). In contrast, the overall correlation between N_d and above-N_a was weak but statistically significant when considering all analyzed profiles ($r = 0.41$, $p < 0.01$). For polluted FT cases (circle markers in Fig. 5b), the N_d exhibited a fairly positive correlation with above-N_a ($r = 0.74$, $p < 0.01$) when above-N_a was <2000 cm^{-3}, while the correlation became weaker when above-N_a was >2000 cm^{-3}. These results suggest that the influence of above-N_a on cloud properties was weaker than sub-N_a at the place/time of observation, indicating that cloud base activation of N_a played a greater role as compared to cloud-top activation of N_a through entrainment. Particularly, the cloud base activation of N_a could play a dominant role, on observation days characterized by a clean FT but BB impacted MBL (triangle markers in Fig. 5).

Line 318-321: What is the relative position of FT, BB layer and the cloud? What is the cloud vertical extent? Without a clear statement or a schematic to show their relative altitudes, one can image different scenarios and can misunderstand the results.

The revised manuscript is:

The bottom of the BB layer in the FT was defined as the lowest altitude of the plume where observed N_a exceeded 500 cm^{-3} (Gupta et al., 2021). The distance from the top of cloud layers in the MBL to the bottom of BB layers in the FT is referred to as Cloud Top to Aerosol Base (CTtoAB). Figure 6a shows the relationship between sub-N_a and the CTtoAB. An overall negative correlation was observed between sub-N_a and CTtoAB.

Line 345-346: Please refer to the results in which Figure and which panel.

The revised manuscript is:

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

Line 377-379: why there is a difference? Please shortly explain it. Is it because of limitations in this study?

Line 381-382: What aerosol properties are responsible for the differences? Please clarify it. Simply leaving a very general (well-known) statement does not contribute to the advances of the field.

Regarding the above two comments, more details 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.

Line 387: Should be subsection 3.2.2

The subsection has been added.

Line 394-396: References are needed. And corresponding figures for the results should be referred.

The reference has been added to the manuscript. The corresponding figures for the results have been referred to.

The revised manuscript is:

The relationship between R_e and sub-N_a (Fig. 7a) shows a weaker correlation than that between N_d and sub-N_a (Fig. 5a), likely due to the influence of additional atmospheric factors such as the MBL thermodynamic structure, cloud depth, cloud-top entrainment process, etc (Wood, 2012; Herbert et al., 2020).

Line 426-427: Why select these three cases but not others? Is there a reason? Or was it randomly picked up?

Based on the BB pollution conditions, the CLARIFY campaign can be divided into three periods: Period 1 from August 16 to 20, BB aerosols were observed to exist predominantly in the MBL, and the FT was mainly clean; Period 2 from August 21 to 25, the MBL became considerably clean, while the BB pollution existed predominantly in the FT; Period 3 from August

26 to the end of the campaign, the MBL was again impacted by BB, and the BB aerosols were observed in both the MBL and FT. The three cases chosen were typical and representative examples for the three periods classified in this study. The revised manuscript is:

These cases illustrate distinct and representative transport pathways of original air parcels for three periods. *Case 1 released tracers near the end of Period 1 (12:00 UTC, August 18). Cases 2 and 3 released tracers at the beginning of Periods 2 (12:00 UTC, August 21) and 3 (12:00 UTC, August 26) respectively, coinciding with shifts in MBL pollution conditions.*

Line 431: The journal does not allow the use of sub-panels like a1, a2, and a3. Instead, all panels should be listed following the alphabetic table.

The sub-panel labels have been corrected, following the alphabetic table.

Line 435: Why start with case 2 first but not case 1?

In the revised manuscript, cases 1 and 3 (BB polluted MBL) are described before the case2.

Line 439-440: the results in which previous figure supports the statement 'and the MBL was observed to be BB-impacted over Ascension Island' ? Please refer to it. If not, please provide relevant results to support it (for case 1 and 3).

The authors have reorganized the structure of this manuscript. The relevant description of this comment has been relocated to Section 3.1. The corresponding figures that can support this statement have been referred to.

Line 511-512: Why present the results in Fig. S9 but not in the main text? Since it is discussed in the main text.

Figure S9 has been moved to Sect. 3.2 (Fig. 4) in the main text.

Line 575: Better not to use 'primary activation' and 'secondary activation'. Instead, please directly state-out the activation pathway. This helps the readability.

The "primary activation" has been revised to "cloud-base activation" via updrafts carrying aerosols to the cloud base. The "secondary activation" has been revised to "cloud-top activation" via entrainment through turbulent mixing at the cloud top.

Technical corrections:

1. The comments on language and statements in the above specific comments are not exhausted. There are also many long sentences which generally have more grammatic errors and make it hard to digest. Please carefully go through the manuscript and improve the presentation.

   The authors have reduced the use of long statements and checked grammar throughout the manuscript.

2. Figure 1: It will be helpful to show the grid lines to guide naked eyes for reading flight paths.

   The grid lines have been added in Fig. 1.

3. Equations should be numbered. This is missing.

The numbers of Equations have been added.

4. English for dates need to be checked through the manuscript. Please use either British or American style.

   The date format now consistently follows American English standards.

5. Table S1: Full spelling for MBL and FT is necessary.

   The full spelling for MBL (marine boundary layer) and FT (free troposphere) has been added to Table S1.

6. Figure 2: Axis ticks should be indicated for the ease of reading. Please also show the height for MBL as a function of time, which will help the discussions in Line 239-242.

   The axis ticks have been added on Fig.2. The inversion height (z_i) as a function of time has also been added in Fig. 2.

7. Figure 3: Line 857 'shades represent 10% and 90% value'. The shading area represents a range.

   The revised manuscript is: *Solid lines and shades represent median values and ranges from 10 to 90 percentiles.*

8. Figure 6: Revise the panel label and specify which panel corresponds to which case.

   The labels have been revised in the previous Fig. 6 (now Fig. 8), and the cases have been correlated to the labels.

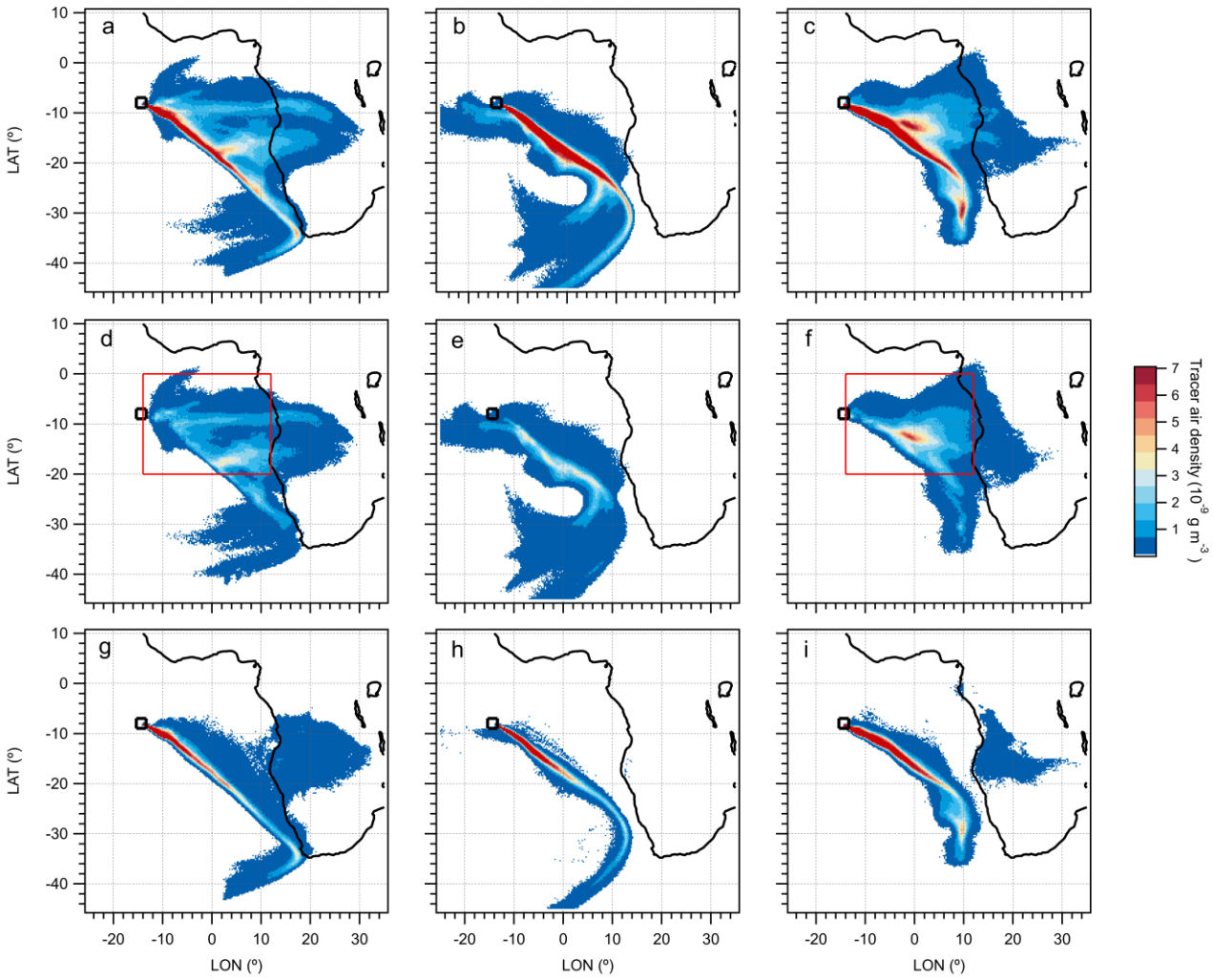

Figure 8. The 7-days backward-dispersion of three cases from NAME simulations. Panels a-c) are column-integrated horizontal footprints for a) Case 1, b) Case 2 and c) Case 3. Panels d-f) are dispersion results attributed to FT for d) Case 1, e) Case 2 and f) Case 3. Panels g-i) are dispersion results attributed to BL for g) Case 1, h) Case 2 and i) Case 3. All plots are shown in the same colour scale. The black boxes represent the release locations of Ascension Island. The red boxes in Fig. e and g represent the horizontal grids (covering the area of 20°S – 0°N, 15°W – 12°E) used for integrating within each vertical layer to derive the vertical distribution of dispersed air parcels in Fig. 9.

9. Figure 7: The legend for cases should come earlier in panel (c). The two AOT for y-axis in panel (c) and (d) should be differentiated (further specified).

   The legend for cases has been moved to panel (c). The two AOT for y-axis in panel (c) and (d) have been differentiated ($AOT_{weighted\text{-}FT}$ and $AOT_{weighted\text{-}BL}$).

10. Figure 8: X-axis label is missing.

    The label of x-axis has been added in the previous Fig. 8 (now Fig. 10).

References:

Brown, A., Milton, S., Cullen, M., Golding, B., Mitchell, J., and Shelly, A.: Unified Modeling and Prediction of Weather and Climate: A 25-Year Journey, B. Am. Meteorol. Soc., 93, 1865–1877, https://doi.org/10.1175/bams-d-12-00018.1, 2012.

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