This document includes the reviewer comments (red) and author responses (black).

- Does the paper address relevant scientific questions within the scope of ACP?
- The paper addresses "Precipitation Susceptibility of Marine Stratocumulus" to variations in aerosol concentrations. This is an important topic within the scope of ACP.
- Does the paper present novel concepts, ideas, tools, or data?
- The novel part of this research is the aircraft dataset. No new concepts, ideas or tools are introduced.
- Does the title clearly reflect the contents of the paper?
- The title places emphasis on precipitation susceptibility, which is only a small component of the paper, has questionable validity, and lacks proper discussion.

This comment is addressed by modifying the title to include "Factors affecting" at the beginning. The discussion of precipitation susceptibility (S_o) in Section 6.3 has been edited for clarity. The data analysis before quantifying S_o in Section 6 supported the discussion of S_o for different aerosol regimes. Figure 9 was added to make the link more explicitly with a summary in Section 6.4. The reviewer's concerns about methodology, data interpretation and validity of the results are addressed through this document.

- Does the abstract provide a concise and complete summary?
- The title emphasises "Precipitation Susceptibility". This is not mentioned at all in the abstract, but it is discussed in the conclusions.

The reviewer is directed to Lines 22-24 from the abstract. These lines mention S_o and provide a quantitative estimate of the aerosol-induced change in S_o.

- Are substantial conclusions reached?
- The main results stem from figures 3 and 7, which demonstrate that there is some difference in the cloud microphysical properties between two subsets of the data. This is attributed to biomass burning aerosols entrained into the boundary layer from above. The paper also explores potential impacts of aerosols on precipitation formation, and the role of meteorological factors also affecting the microphysical properties. The conclusions are rather limited in scope, and in part unsubstantiated. I understand observational based research is important and also difficult to publish in its own right without complex modelling, so maybe a "Measurement Report" is more suitable format?

Modelling and observational studies must complement each other. Quantification of cloud microphysical properties, precipitation formation process rates, and S_o for different aerosol regimes using in situ data can aid modelling efforts. For example, the autoconversion and accretion rates were based on a commonly used model parameterization. The aerosol-induced changes could be compared with model output. Given the size of the dataset used, model simulations were beyond the scope of the study. The authors agree with the reviewer that most observational studies of aerosol effects on clouds and precipitation would benefit from model support since in situ observations essentially provide snapshots of a particular cloud.

The data analysis throughout this study quantified perturbations in droplet concentration (N_c) and precipitation rate (R_p) due to droplet growth or increasing aerosol concentration (N_a). Figure 9 was added to link the data analysis to changes in S_o as a function of H under different aerosol regimes. The study addressed hypotheses about variations in S_o under different above- or below-cloud N_a (Duong et al., 2011; Jung et al., 2016). We do not know of a prior study that did this using in situ measurements. Observational analysis of variables/processes affecting S_o for the aerosol regimes should justify the current format as a research article. Further, this study is similar in scope to many other papers that used observational data to evaluate process-based hypotheses without inclusion of modeling results.

- Are the scientific methods and assumptions valid and clearly outlined?
- Are the results sufficient to support the interpretations and conclusions?
- The paper uses specific statistical terminology (e.g. "95% Confidence Interval") which inherently implies parameters are known to exhibit normal distributions when properly sampled. Is this valid? Is this approach needed?

95% confidence intervals were used to provide statistical confidence to the comparisons between aerosol and cloud properties from different aerosol regimes. The average values for the parameters were provided throughout the study (Table 5 was updated to add the averages). Since every variable may not be normally distributed, the addition of average values allows the reader to directly compare the average values rather than using 95% confidence intervals. It is noted that the 95% confidence intervals include this direct difference between the average values regardless of the shape of the distribution.

- Adiabatic approximations of Brenguier are used. Limitations should be discussed/quantified.

The adiabatic value of liquid water content (LWC) was used as a maximum threshold to select the cloud probe to be used for analysis of data from ORACLES 2016. The impact of choosing the CAS over the PDI was discussed in detail within author responses to Reviewer Comment 1. Other discussions based on the adiabatic model did not affect data analysis or the conclusions. These discussions (Lines 244 to 252 and Lines 256 to 271) were removed for brevity.

The following sentence was added to the paper: "LWC_{ad} can be used to compare LWC from different probes since it is derived using environmental conditions without any input from the cloud probes."

- Measurement uncertainties are not presented alongside observations. E.g. What is the estimated uncertainty in measurements of droplet effective radius, and how does this relate to changes between cloud base/top, and also between the aerosol regimes studied?

The uncertainties associated with cloud probes were discussed in the supplement. Estimates of LWC from cloud droplet size distributions were validated against an independent measurement of LWC from a hot-wire. Previous work has shown a 15 to 20 % uncertainty in N_c can result in up to a 50 % uncertainty in LWC (Lance, 2012). These estimates were consistent with the differences in N_c and LWC between cloud probes used during ORACLES (see supplement).

A sizing uncertainty within 20 % could be expected for droplets larger than 5 μm from CAS and CDP, 50 μm from 2D-S, and 750 μm from HVPS-3 (Baumgardner et al., 2017). For differences between R_e across aerosol regimes (below 2 μm) and changes in R_e from cloud base to top (below 3 μm), the uncertainty can be assumed to be constant. Since relative changes in cloud properties were quantified, measurement uncertainties would have a minor impact on the results. The following sentences were added to the paper:

"Measurement uncertainties in droplet sizes were expected to be within 20 % for droplets with D > 5 μm from the CAS and the CDP, D > 50 μm from the 2D-S, and D > 750 μm from the HVPS-3 (Baumgardner et al., 2017)."

"The relative differences between the LWC_{ad} and the LWC estimates from cloud probes provide a measure of the uncertainty associated with using one probe over the other for data analysis."

- Calculations in the paper suggest the thinnest clouds have large precipitation sensitivity to aerosols. This seems odd given these thin clouds have nominally the same droplet concentrations as thicker clouds, but only have the smaller droplets. This raises concerns with how data are handled and the overall validity of conclusions drawn from the analysis. From the text I don't fully understanding what was done here with the data to determine the precipitation susceptibility, so maybe my interpretation is wrong, but I speculate it is a result of using outputs from regression analysis which are statistically meaningless. If this is true, the paper is presenting misleading results which is very undesirable. If this is not true it needs making clearer.

The concerns are addressed by providing specific justifications for the methodology used, data interpretation, and observations of high S_o for thin clouds:

Methodology: The best fit slope from a regression between ln(N_c) and - ln(R_p) was used to quantify S_o following Eq. 8. S_o was quantified for different populations of cloud profiles classified based on H to quantify S_o as a function of H. This is consistent with previous studies of S_o using airborne measurements (e.g., Jung et al., 2016).

Data interpretation: The authors assume the reviewer is referring to low correlation coefficient (R) values between ln(N_c) and ln(R_p) when they say regression outputs are "statistically meaningless". The values of R between 0.3 to 0.6 were consistent with previous studies (e.g., Jung et al., 2016) and were statistically significant. Low values of R were observed because N_c was calculated for the entire droplet size distribution while R_p was calculated for drizzle drops (diameter D > 50 μm). If R_p was calculated for the entire size distribution, the values for R increased. However, including smaller droplets within R_p is not useful since the smaller droplets would have little chance of precipitating.

Thin clouds having high S_o: The quantification of high S_o for thin clouds in the cleanest conditions and S_o close to zero for thin clouds in polluted conditions is consistent with previous studies (discussed in Section 6.4). The reviewer may be referring to cloud profiles with H < 129 m from the Separated-low N_a regime (Fig. 8b) since these profiles contributed to the high S_o for thin separated profiles (Fig. 8a). Figure 7 (c, d) shows these profiles had very low N_c and the highest R_p, on average, compared to thin clouds from other aerosol regimes. These profiles had fewer droplets and these droplets more frequently had D > 50 μm. It is reasonable to see high values of S_o in these conditions (see Figure 9 which was added to the text).

- The stratiform clouds are shown to be around 200m thick, and often occur in the vicinity of convective clouds. Is direct comparison of high resolution in-situ datasets with relatively coarse resolution ERA5 reanalysis data sufficient to untangle effects of meteorology? What small-scale/local variations in SST could you expect based on other studies? What are the actual sizes/resolutions of ERA5 grid boxes in units relatable to the observations? Can ERA5 resolve the inversions etc? The correlations in Fig10b between LWP from in-situ and ERA5 are poor, which casts a large doubt over the validity over the in-situ LWP vs SST/LTS/EIS from ERA5. Why aren't in-situ observations of inversion strength analysed?

Reanalysis data have been used in recent studies to constrain environmental conditions and their impact on LWP and/or aerosol-cloud interactions (Douglas and L'Ecuyer 2019; 2020). The advantage of using reanalysis data was that calculation of LTS and EIS was consistent across all profiles. This was desirable given the main purpose of LTS and EIS was comparisons between aerosol regimes.

The horizontal resolution of ERA5 reanalysis was 0.25 degrees latitude and longitude (Hersbach et al., 2020) which is about 20 km. For closed cell marine stratocumulus, horizontal heterogeneity over this distance can be assumed to be low. Based on the reanalysis temperature at different pressure levels, the model was able to resolve the inversion near cloud tops for the co-located in situ profiles (Fig. 1).

Low correlation between in situ LWP and ERA5 LWP did not have a dependence on the thermodynamic parameters used to determine EIS or LTS. Ahlgrimm et al. (2009) showed biases in cloud properties from the model were due to assumptions within the model parameterization They found improved correlation between model LWP and ground-based LWP when the autoconversion-accretion parameterization was updated.

A sawtooth pattern followed for cloud sampling during ORACLES meant the aircraft frequently flew only about 100 m above or below the cloud layer (Fig. 2) Further, there were concerns with airborne measurements of thermodynamic parameters during descents into cloud (Gupta et al., 2021). This meant that thermodynamic parameters needed to calculate LTS or EIS were not available from in situ measurements near every profile.

- From the very beginning this paper places emphasis on the role of aerosols from above cloud and their ability to modify clouds via entrainment etc. There is no discussion of the potential for the boundary layer being polluted with Biomass Burning aerosols in its own right, without the requirement for entrainment from above the BL inversion. Is there data showing the transition of the BL from clean to polluted as aerosol mix downwards? If so it would be very useful to show it.

It is unlikely there were sources of biomass-burning aerosols over the southeast Atlantic Ocean. Continental aerosols reached the marine boundary layer through cloud-top entrainment or entrainment into a clear boundary layer. Evidence to support this is provided:

Biomass burning aerosols are lofted into the free troposphere over the continent (Gui et al., 2021). The aerosol layer is transported over the southeast Atlantic by mid-tropospheric winds (Adebiyi and Zuidema, 2016). Back-trajectory analysis has shown polluted above-cloud airmasses originate from high altitudes over the continent while clean below-cloud airmasses originating from the boundary layer in the southeast (Gupta et al., 2021; Miller et al., 2021). The altitude of the aerosol layer near Ascension Island also tends to increase from July to October (Zhang and Zuidema, 2021).

The following text was added to the introduction section: "Biomass-burning aerosols from southern Africa are lofted into the free troposphere (Gui et al., 2021) and transported over the southeast Atlantic by mid-tropospheric winds where the aerosols overlay an extensive MSC deck that exists off the coast of Namibia and Angola (Adebiyi and Zuidema, 2016; Devasthale and Thomas, 2011)."

The boundary layer could be polluted due to entrainment prior to in situ observations (Diamond et al., 2018). Ground-based observations from Ascension Island have shown clean boundary layers with elevated biomass burning trace gas concentrations during the burning season (Pennypacker et al., 2020). This suggests precipitation scavenging can lead to clean boundary layers in terms of N_a despite the entrainment of biomass-burning aerosols into the boundary layer.

The following text was added to Section 6.1: "Ground-based observations from Ascension Island have shown clean boundary layers can have elevated biomass burning trace gas concentrations during the burning season (Pennypacker et al., 2020). This suggests boundary layers could be clean in terms of N_a despite the entrainment of biomass-burning aerosols into the boundary layer due to precipitation scavenging of below-cloud aerosols."

- The paper filters data according to aerosol concentrations above cloud (“contact” vs “separate” using a 500cm^{-3} threshold) and below cloud (“high” and “low” Na with a threshold of 350cm^{-3}). However, the cloud droplet concentrations in Fig 6 do not show evidence of enhancement due to above cloud aerosols for the “clean” BL cases. The only strong response in droplet concentration is when there are lots of aerosols also in the boundary layer. It seems impossible to disentangle the below and above cloud aerosols and therefore the role of entrainment and above cloud aerosols is ambiguous.

The reviewer is directed to Table 5 where the increase in N_c for low boundary layers was quantified and compared with the corresponding increase in N_c for polluted boundary layers. While the increase in N_c was relatively smaller compared to polluted boundary layers, it was statistically significant. We agree with the reviewer’s statement that it is difficult to disentangle the relative impact of above- and below-cloud aerosols. Thus, the study did not distinguish the impact of above-cloud versus below-cloud aerosols. Instead, the combined impact of above- and below-cloud aerosols was compared with the impact of above-cloud aerosols alone (Lines 437 to 443 in the original manuscript).

- There is no contextualisation of the results. For instance, are the calculated changes in re, or values of S_o “large” or “small”? Are changes in these clouds due to the Biomass Burning aerosols having any meaningful impact? What have other studies found?

As stated in Section 6.3, S_o for observational datasets should only be compared with observational datasets given the dependence of S_o on data analysis techniques and aerosol analogues used in satellite studies (e.g., Sorooshian et al., 2009). Previous studies using observational data did not quantify S_o under different aerosol regimes within a similar domain. Instead, S_o has been quantified for different cloud types or regions. Therefore, it is difficult to contextualize the changes in S_o presented in this study in terms of previous studies. The changes in R_e and N_c were consistent with previous studies which are referenced throughout the text.

- Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?
- Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?
- Undefined formulae: Z_N

The following text was added (Line 304) to address this: “Figure 3 shows violin plots for cloud properties as a function of normalized height (Z_N), defined as $Z_N = Z - Z_B / Z_T - Z_B$.”

- Confusing presentation of $\Gamma^{850}_{ð???}$, in eqn 10

This part of the equation was removed to avoid confusion along with the equation that defined Γ_m (Eq. 11). Instead, the reader was directed to Wood and Bretherton (2006) consistent with the approach followed by Douglas and L’Ecuyer (2021).

- Description of LCL is confusing and the equation is poorly formatted

LCL has been defined using the appropriate formatting.

- Some of the technical details of data processing are in figure captions, but should be included in the text.

Assuming this comment was directed at Figure 3, the caption is improved for clarity and the following description was added to the text: "The violin plots include box plots and illustrate the distribution of the data (Hintze and Nelson, 1998)." The details for data presented in every figure are provided during the corresponding discussion in the text.

- What are the "kernel density estimates" mentioned in caption for Figure 3? They are not mentioned anywhere else in the paper.

The figure shows violin plots where the width of the shaded area represents the proportion of data there (Hintze and Nelson, 1998). The description was added to the text: "The violin plots include box plots and illustrate the distribution of the data (Hintze and Nelson, 1998)."

- Is the overall presentation well structured and clear?
- Is the language fluent and precise?
- Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?
- The paper was difficult to follow. I feel the paper is too long and lacked coherence. It is very ambitious, and the authors have covered lots of areas which are all important and related, but the balance is not quite right. The paper has lots of useful data but in its current form does not provide concrete outputs which can be used by the broader community.

The concerns with balance/coherence, presentation, and paper length were addressed:

The title of the paper was edited to emphasize the role of factors affecting S_o. Figure 9 was added to relate the preceding data analysis with the discussion of S_o in Sections 6.3 and 6.4. The impact of perturbations in N_c and R_p on S_o (due to droplet growth processes with H or increasing aerosols) was further illustrated in a mathematical framework. Recommendations for future work were added. The following text was removed to reduce the paper length:

- Line 214 to 218: Comment on relationship between cloud top height and liquid water path adjustments associated with aerosol-cloud interactions.
- Line 244 to 252, 256 to 271: The discussion of parameters associated with adiabatic cloud optical thickness.
- Line 315, 318 to 320: Comment on aerosol influence on cloud water.
- Line 576 to 579: Definition of Γ_m.

- Most figures should be improved and are poorly rendered, and some do not have proper legends etc (e.g. Fig 10b has a 1:1 line listed as “x=y”, wrong coloured text in legends).

Every figure was updated and rendered following journal guidelines (300 dpi resolution). The legends were updated: text color corrected for Fig. 3, 4, and 9 and “x=y” replaced by “1:1 line” for Fig. 10b.

- Do the authors give proper credit to related work and clearly indicate their own new/original contribution?
- Are the number and quality of references appropriate?
- Yes there are a good number of quality references. Some references are missing (e.g. description of instruments in section 2) but nothing major.

The appropriate references were added to Section 2.

- Is the amount and quality of supplementary material appropriate?
- Yes, supplementary material is of good quality and is a useful addition.

FIGURES:

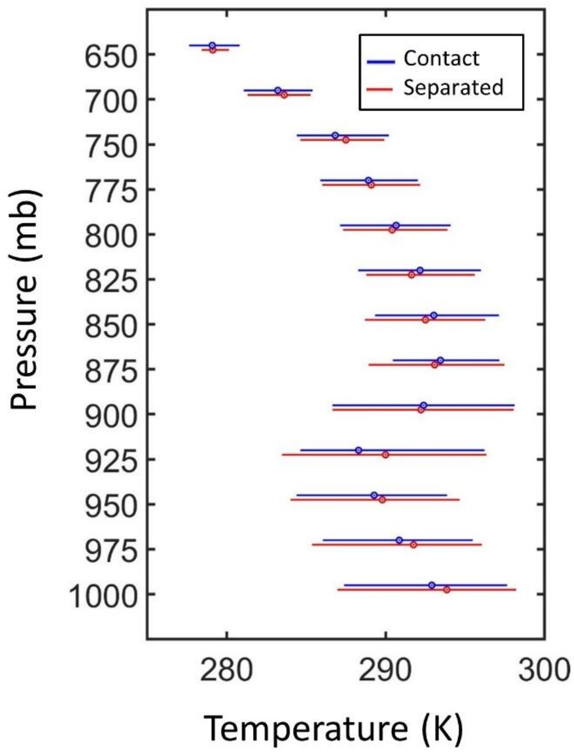

Figure 1: Box plots of temperature from the ERA5 reanalysis at model pressure levels. The data correspond to grid boxes co-located with an in situ cloud profile used in the study.

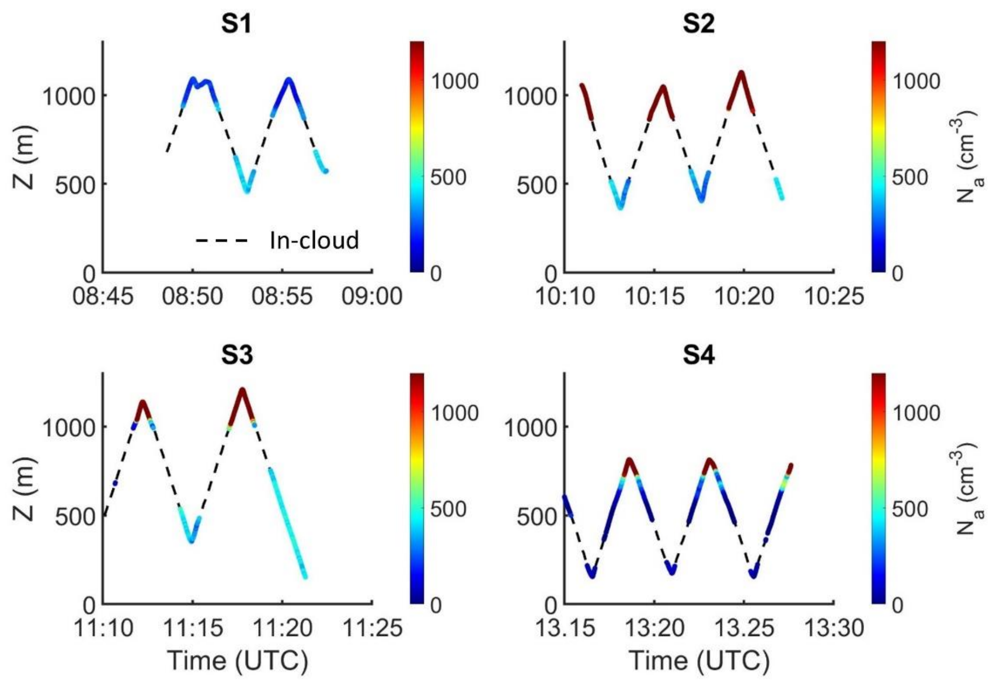

Figure 2: P-3 aircraft altitude as a function of time during sawtooth flight patterns. Data are colored by accumulation mode aerosol concentration (taken from Gupta et al., 2021).

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