The authors would like to thank the anonymous reviewer for their time and efforts in reviewing the manuscript. This document contains author responses to reviewer comments. Reviewer comments are in red and author responses are in black. Removed text is in "*quotes and italicized*" and added/replacement text is in "**quotes and in bold**".

Review of "Impact of the Variability in Vertical Separation between Biomass-Burning Aerosols and Marine Stratocumulus on Cloud Microphysical Properties over the Southeast Atlantic" by Gupta et al.

This study reports on the important issue of smoke-cloud interactions with a focus on the vertical separation of aerosol layers from cloud tops, which is of importance in the southeast Atlantic Ocean region where there can be large smoke plumes above and within the boundary layer. This study makes use of ORACLES data, specifically from six research flights. Statistics are provided about the number of cases where theaerosol layers (> 500 cm-3) are within 100 m of cloud tops ("contact") or in excess of 100 m from tops (called "separated"). Subsequently, cloud properties and free tropospheric humidity are compared for these two categories of cases. A finding was that droplet evaporation (from entrainment drying at cloud top) was enhanced in cases where plumes were above 100 m from cloud tops (called "separated"); this was coincident with greater reductions in cloud drop number concentration and liquid water content near cloud tops. Another finding was that sub-cloud aerosol number concentrations were typically higher for "contact" cases (> 350 cm-3). Also, the "contact" cases with high aerosol concentrations in the boundary layer had higher drop concentrations as compared to "separated" cases. The paper was well written and easy to follow. At least one of the tables was difficult for me to digest but in general the tables and figures were clear too. The results are important and I am fully supportive of publication after the comments below are addressed.

The authors thank the reviewer for their comments and support for publication. Specific reviewer comments are addressed below.

Specific Comments:

Table 5: Took me several time to read the caption to try to understand the table and I am still not sure I understand it.

The caption for Table 5 was changed for clarity.

"*Differences between the average below- and above-cloud N_a, and the average N_c and R_e measured in the cloud layer for contact profiles relative to separated profiles. The differences are classified by the maximum below-cloud N_a within the boundary layer and correspond to 95% confidence intervals based on a two-sample t-test (not reported when $p > 0.05$).*" to

"**Aerosol and cloud properties were averaged across all *contact/separated* profiles flown in low N_a and high N_a boundary layers. These averages were compared between *contact* and *separated* profiles. The values listed below represent the 95% confidence intervals (from a two-sample t-test) when the differences were statistically significant. Positive values indicate the average for *contact* profiles was higher and "insignificant" denotes the differences were statistically insignificant.**"

Line 76-85: I suspect it may be important to refer to this study somewhere here or elsewhere in the paper owing to its high relevance:
Rajapakshe, C., et al. 2017. Seasonally transported aerosol layers over southeast Atlantic are closer to underlying clouds than previously reported. Geophysical Research Letters, 44, 5818–5825. https://doi.org/10.1002/2017GL073559

The authors note the relevance of this study here. The following text was added in section 1:
"**Rajapakshe et al. (2017) found the aerosol layer was located within 360 m above the cloud layer for about 60% of the Cloud-Aerosol Transport System (CATS) lidar night-time scenes over the southeast Atlantic.**"

Line 118-119: Give a brief description of how the collection efficiency was computed and handled for the data presented.

A time- and composition-dependent collection efficiency (CE) was applied to correct for the incomplete vaporization of mixed phase particles. As discussed in Middlebrook et al., (2012), the CE is primarily determined by the efficiency with which a particle's impaction upon the vaporizer is detected. This is turn is mostly explained by the phase of the particle, with liquid aerosol collected more efficiently than neutralized aerosol because it is less likely to bounce off the heater and escape detection. Liquid aerosol is primarily acidic, and the acidity of the free-tropospheric aerosol is assessed by comparing the molar ratio of NH_4 to $2xSO_4$. The use of the $NH_4/(2SO_4)$ ratio is a simplification of the $NH_{4,measured}/NH_{4,predicted}$ relationship. $NH_{4,predicted}$ is the amount of ammonium required to neutralize the inorganic anions observed by the AMS. The collection efficiency is then determined from $CE=max(0.5, 1- NH_4/(2xSO_4))$, with a value of 0.5 serving as the lower limit, consistent with the default value applied within most field campaigns (Middlebrook et al., 2012).

The following text was added after line 119:
"**A time- and composition-dependent collection efficiency (CE) was applied to AMS data. The molar ratio of ammonium to sulphate ($NH_4/(2xSO_4)$) was calculated to assess the acidity of liquid aerosol which are collected more efficiently compared to neutralized aerosol. Thus, CE**

was determined as the maximum between 0.5 and (1- $NH_4/(2xSO_4)$), with a value of 0.5 serving as the lower limit, consistent with estimates from most previous field campaigns (Middlebrook et al., 2012)."

Middlebrook, A. M., Bahreini, R., Jimenez, J. L., and Canagaratna, M. R.: Evaluation of composition-dependent collection efficiencies for the aerodyne aerosol mass spectrometer using field data, Aerosol Sci. Technol., 46, 258–271, doi:10.1080/02786826.2011.620041s, 2012.

Line 182: Are the authors sure they mean LWC > 10 g m-3? That seems too high (by 2 orders of magnitude).

"*LWC > 10*" has been corrected to "**LWC > 0.05**" to reflect the correct value for the LWC threshold used.

Throughout the paper I suggest the authors consult with 3 other recent references toat least mention them for the sake of comparison and contrast. The Mardi et al. (2018) paper quantifies in detail smoke layer separation from stratocumulus cloud top heights, while their 2019 paper digs into cloud-smoke interactions that are related to results from this study. The Diamond et al. (2018) examines smoke-cloud interactions too over the same region as that of this study. In particular I find that the threshold to use for what constitutes a smoke plume (i.e., its base altitude) to be quite important, for which results of studies like this can be sensitive to; I found it interesting that the criteria in this study seemed to be Na > 500 cm-3, whereas that in the Mardi et al. papers was 1000 cm-3.

The authors note the importance of referencing these studies and comparing their observations with the results presented here. These studies are referenced at appropriate points within the manuscript. In addition to discussions within subsection 4.4 and section 5, the following additions were made:

The following text was added after Line 199 of the old manuscript:
"**This is also likely to be associated with the history of entrainment mixing of polluted free tropospheric air into the boundary layer prior to these observations (Diamond et al., 2018).**"
The following text was added after Line 208 of the old manuscript:
"**In a previous study, a significantly higher threshold (PCASP N_a = 1000 cm^{-3}) was used to identify the BBA layer above stratocumulus clouds off the coast of California (Mardi et al., 2018). The sensitivity of the threshold chosen in this study is examined in Appendix-A and using a threshold of 1000 cm^{-3} would have no significant impact on the results presented in this study.**"

The following text was added within subsection 4.4:

"**Previous studies have argued the changes in N_c due to the impact of BBA are more strongly correlated with below-cloud N_a compared to above-cloud N_a (Diamond et al., 2018; Mardi et al., 2019). However, these results suggest that although the differences in N_c were lower than the differences in above-cloud N_a, significant changes in N_c and R_e were associated with contact with above-cloud BBA, and these changes were independent of the below-cloud aerosol loading.**"

References:

Mardi, A.H., et al. 2019. Effects of Biomass Burning on Stratocumulus Droplet Characteristics, Drizzle Rate, and Composition. J Geophys Res-Atmos 124, 12301-12318.

Mardi, A.H., et al. 2018. Biomass Burning Plumes in the Vicinity of the California Coast: Airborne Characterization of Physicochemical Properties, Heating Rates, and Spatiotemporal Features. J Geophys Res-Atmos 123, 13560-13582.

Diamond, M. S., et al. 2018. Time-dependent entrainment of smoke presents an observational challenge for assessing aerosol-cloud interactions over the southeast Atlantic Ocean. Atmospheric Chemistry and Physics, 18(19), 14623–14636. https://doi.org/10.5194/acp-18-14623-2018

Line 374-375: Are the authors sure they have unambiguous evidence of these causal relationships? This is always a tricky thing with aircraft data and I caution the authors to reconsider if they want to use this strong language.

The authors acknowledge the caveats presented by aircraft data. These are snapshots in space and time and may not reflect the conditions of the entire domain.

The sentence has been moved to follow the next sentence starting "In-situ measurements" was changed:

"*The presence of biomass-burning aerosols immediately above cloud tops impacts N_c, R_e, and LWC through cloud-top entrainment and increases the free tropospheric temperature and humidity*"

to

"**These observations suggest the presence of biomass-burning aerosols immediately above cloud tops was associated with changes in vertical profiles of N_c, R_e, and LWC due to cloud-top entrainment and increases in the free tropospheric temperature and humidity.**"

The authors made the following changes to the manuscript in addition to reviewer comments:

1. Citations for Adebiyi and Zuidema (2016) and Wilcox (2010) were added.
2. "*BL*" was changed to "**boundary layer**" throughout the manuscript.
- Lines 167, 172, 199, 246, captions for Fig. 2 and 4
3. "*Figure*" was abbreviated as "**Fig.**"
- Lines 161, 163, 166, 167, 169, 171, 172, 177, 178, 183, 186, 191, 193, 194, 204, 212, 216, 217, 224, 228, 233, 261, 277, 284, 289, 292, 313, 318, 341, and 351.
4. Minor formatting changes were made to Table 2 and 3 for clarity.
5. A legend was added in Fig. 3 to represent in-cloud altitudes.