Author Response to Reviewers

We thank all of the reviewers for their helpful comments on our manuscript. Our response to each of these points, including any relevant revisions, is detailed below. Referee comments are written in black font color, while the author responses are written in red. Any page and line numbers in our response refer to the revised manuscript.

Referee #1

This manuscript uses date from LASIC campaign and data from Ascension Island between June 2016 and October 2017. They catalogue ultra-clean events (41 in number) over this time period and compare them to smoky cases and background conditions. They find that there is evidence for seasonality in the occurrence of the ultra-clean layers and that they occur during the same time as smoky conditions. They propose mechanisms and ways that these ultra-clean days are related to cloud properties and precipitation. The most interesting aspect of this work is the comparison between the smoky and ultra-clean days and how it related to drizzle amount and occurrence. This work is well written, easy to read and to follow, the results follow clearly from their analysis and the figures are well chosen and presented. My recommendation to accept his work with minor revisions

1) It would be helpful to justify why the authors chose < 50 cm-3 as ultra-clean. Is this arbitrary or is there some other reason for this choice?
In response to this and other similar reviewer comments, we have added the following clarification to the text (P3, L15-26): "While admittedly somewhat subjective, this 50 cm^{-3} threshold is consistent with the upper bound of near-surface and below-cloud observations in MBL environments routinely featuring exceptionally low N_A such as subtropical pockets of open cells (Abel et al., 2019; Sharon et al., 2006; Terai et al., 2014), mid-latitude open-cellular convection (Abel et al., 2017; Pennypacker and Wood, 2017) and across the trade wind stratocumulus-to-cumulus transition (Bretherton et al., 2019). It is also well situated within the typical range (~30 – 60 cm^{-3}) of number concentrations used for the lowest aerosol cases in large eddy simulation studies of MBL aerosol-cloud interactions (Wang et al., 2010; Wang and Feingold, 2009; Yamaguchi and Feingold, 2015; Zhou et al., 2017). Prior work defined ultra-clean layers near the top of the MBL, often observed in the stratocumulus-to-cumulus transition, with $N_A < 10$ cm^{-3} (Wood et al., 2018). We argue it is reasonable to set a higher threshold near the surface, where aerosol number concentrations are generally higher due to proximity to the sea spray source. Furthermore, Wood et al. (2018) focused on these layers primarily as a mesoscale feature within larger cloud systems, whereas our interest is in studying ultra-clean conditions as daily-scale events. Defining ultra-clean conditions using daily median $N_A < 50$ cm^{-3} balances the need to reasonably capture conditions with exceptionally low near-surface N_A in the remote MBL while maintaining a robust sample of cases to study."

2) The record is quite short. Is there any way to extrapolate information about ultraclean days from satellite data sets to get an idea of how often these ultra-clean days occur in a longer-term record? This could be brought up in the discussion section, perhaps, as future work.
We have added a brief mention of this point to our Discussion (P8, L16-20):

“Satellite retrievals of cloud droplet number concentration (Grosvenor et al., 2018) may provide a tool for extending our analysis with both a longer temporal record and greater spatial context of extreme depletion events in the SEATL MBL. However, these retrievals remain far more uncertain in the broken and/or heavily drizzling cloud scenes that often coincide with ultra-clean conditions.”

3) For the figures, the tick marks are hard to read and the numbers bleed into the figure space. Also, for Figure 2 there should be a few more markers (or at least tick marks) on (d).
Thank you for suggesting changes to make sure our figures are clear. We have increased the thickness of the grid lines on our plots, added grid lines for the minor axis ticks in Figure 2d and increased the padding for the axis tick labels to avoid any bleeding.

4) For Figure 2, what do the PDFs for non-UC days look like for CO and rBC from the SP2? It’d be interesting to see the comparison for the non-clean days in these PDFs.
We have updated Figure 2 to include the PDFs for the non-burning background rather than just presenting the statistics.

Referee #2

This manuscript presents a really interesting observation using data from a recent field campaign (LASIC) in the southeast Atlantic, such that both the highest and lowest accumulation mode aerosol particle concentrations have been observed in the same season, i.e. the biomass burning season, highlighting the high daily variability of MBL aerosol loading and the level of complexity between aerosol-cloud interactions in the region. In the study, comparisons between the ultra-clean days and the background non-burning days are made, in terms of CO and BC concentrations, LWP, and precipitation intensity and frequency, to highlight the role of coalescence scavenging in creating these ultra-clean days in between highly polluted days. I find the manuscript well-written and easy to follow, with well-organized sections and clear questions to address.

Major comments:

1. Medians with inter-quartiles are used throughout the manuscript, favored over means with standard deviations. I wonder if there is a reason for such preference, and suggest specifying why medians better represent the characteristics of these variables at the daily time scale?
Thank you for your question about this choice. This decision is primarily motivated by the fact that means will generally be more sensitive to being biased by any outlier observations than medians. Furthermore, medians and inter-quartile ranges will also generally be more robust and illustrative (or at least as illustrative) if the distribution of observations is skewed (e.g. Figs 2c,d). We have updated the text in several locations to specifically address this:

- In reference to the use of daily median accumulation mode number concentration to identify ultra-clean events, we have added (P3, L26-28) the following clarification: "We take daily medians as a better indication of the aerosol number concentration over the course of a day since they are more robust to any outlier observations than daily means, though this choice only leads to a discrepancy over one day identified as ultra-clean."
- In reference to CO and rBC concentrations (P4, L8-10): "Again, we report median concentrations in order to minimize the potential impact of any outlier observations. We also report and compare inter-quartile ranges since a long tail often skews the variability about these medians."

2. The polluted days used for the back-trajectory comparison raised a concern. As most of the ultra-clean days are from the biomass burning season, would it make more sense to compare directly to the polluted days within the BB season, i.e. excluding the non-burning season? Besides, even the upper 5 percentile of daily median Na from December to April aren't really "polluted," are they?
Thank you for raising the need to clarify this point. The polluted back trajectories are only from the months where there are also ultra-clean days (i.e. during the biomass burning season). We have added text in the Abstract (P1, L16) and Section 2.2 (P4, L20-21), where the back trajectories are introduced, to ensure that this is clear. We have also added a supplemental table (Table S1) listing all of the ultra-clean and polluted dates from those same months that are included in the analysis.

Minor comments:

P2-13, only one set of parentheses is needed here.
Thank you, this has been fixed.

P3-9, Is there a reason or any reference suggesting 50 as the threshold for ultra-clean condition? What is the instrument sensitivity of the UHSAS measurements?
In response to this and other similar reviewer comments, we have added the following clarification to the text (P3, L15-26): "While admittedly somewhat subjective, this 50 cm^{-3} threshold is consistent with the upper bound of near-surface and below-cloud observations in MBL environments routinely featuring exceptionally low N_A such as subtropical pockets of open cells (Abel et al., 2019; Sharon et al., 2006; Terai et al., 2014), mid-latitude open-cellular convection (Abel et al., 2017; Pennypacker and Wood, 2017) and across the trade wind stratocumulus-to-cumulus transition (Bretherton et al., 2019). It is also well situated within the typical range (~30 – 60 cm^{-3}) of number concentrations used for the lowest aerosol cases in large eddy simulation studies of MBL aerosol-cloud interactions (Wang et al., 2010; Wang and Feingold, 2009; Yamaguchi and Feingold, 2015; Zhou et al., 2017). Prior work defined ultra-clean layers near the top of the MBL, often observed in the stratocumulus-to-cumulus transition, with $N_A < 10$ cm^{-3} (Wood et al., 2018). We argue it is reasonable to set a higher threshold near the surface, where aerosol number concentrations are generally higher due to proximity to the sea spray source. Furthermore, Wood et al. (2018) focused on these layers primarily as a mesoscale feature within larger cloud systems, whereas our interest is in studying ultra-clean conditions as daily-scale events. Defining ultra-clean conditions using daily median $N_A < 50$ cm^{-3} balances the need to reasonably capture conditions with exceptionally low near-surface N_A in the remote MBL while maintaining a robust sample of cases to study."

P3-14, I suggest including the sampling frequency as well as the instrument sensitivity for CO and rBC measurements.
We have added (P4, L2) the sampling frequency for the CO measurements (1 Hz). According to the ARM documentation for the CO gas analyzer measurements, sensitivity is "not meaningful" for the CO measurements because tropospheric background values are "well above" minimum detectable levels. The sampling frequency and sensitivity of the SP2 rBC measurements were included in the original manuscript.

P3-28, the full name of HYSPLIT should be introduced here, and details on the HYSPLIT runs should be given as well, e.g. what meteorological dataset is used, at what spatial resolution.
The text has been updated to address these points: "We take a complimentary approach by analysing 7-day isobaric boundary layer back-trajectories initialized at approximately 500 m over ASI at 12:00 UTC as computed by the NOAA Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) with Global Data Assimilation System meteorology on a 0.5 degree by 0.5 degree grid (Stein et al., 2015)."

Section 2.3, sampling frequency of the laser disdrometer and the microwave radiometer should be given here, and I assume the 2-channel MWR is the one at the AMF1 C2, not the one at the airport, correct?
The text has been updated (P5, L1-3) to include the relevant sampling/averaging windows, as well as to indicate that all data is from the AMF site (not the airport).

P4-24, "available LASIC data," I am curious to know how many available days there are in total, i.e. 41 out of how many days?
There are 460 available days of UHSAS data between June 1 2016 and October 30 2017. We have added a note of this in the text (P3, L12).

P5-5~7, could you label the r^2 values on Fig. 1 c and d?
Done.

P5-10, could you provide r^2 values for the hourly correlations in Fig. S1 (on the figure as well)?
Done.

P5-16, this sentence is unclear to me, are you saying r^2>0.65 is for the early biomass burning season, which is a subset of your data? Please define the early biomass burning season, i.e. which month?
Thank you for noting the need to clarify this. We now reference which months these statistics refer to (P6, L4-5).

P5-17, "day-to-day the correlation" → "the day-to-day correlation", and you are talking about CO to Na correlation here, right?
The text has been clarified to read: "the day-to-day N_A-CO correlation."

P5-18, "rBC generally follows . . ." suggest adding r^2 values here.
Done.

P5-22, agreed, for future work, maybe check with the SP2 instrument mentor, as SP2 is an optical device, and things that are not smoke can still trigger it.
We agree. This was outside the scope of our current analysis, but hopefully will get addressed in future work.

P5-26, "median hourly median", how about "median of hourly median"?
Thank you for the suggestion. Fixed.

P5-27, I suggest putting these statistics on the figure as well, maybe in the same color as the ultra-clean days, but in dashed lines, this will help me a lot to visualize the shift.
In response to the comments from Reviewer One, we decided to just show the background PDFs instead of plotting just the statistics. This will also hopefully help visualize the shift.

P6-9~10, see the second comment in Major comments, and how many polluted days are there, based on your criteria?
See response to the Major Comment 2 above.

P6-25, I agree with you on the use of CDF, but I find PDFs are useful to see as well, have you considered showing both of them on the same plot? Well, this could make a really busy plot, I will leave the decision up to you.

We appreciate that both data presentations can be useful, but we agree that having both on the same plot could be confusing. Our main goal here is to highlight the general differences and shifts in the distributions, which the CDF plots nicely demonstrate. To us, they also more clearly represent the parts of the distribution that contribute most to those shifts and by how much.

P6-29, Why the mean is shown here instead of median, what are the median values of these?

Thank you for pointing out this inconsistency. We now report the median and inter-quartile spread for each (P7, L27-29). This does not change any results qualitatively, but the means are higher than the medians by about 60 – 70 gm^{-2}. This correction actually further highlights why we generally elected to report medians in this analysis (see response to Major Comment 1).

Figure 4 a, suggest adding those vertical dashed lines in the background when log scale is used, just like you did in figure 5 a.

Thank you for the suggestion, we have added those axis lines to Figure 4a.

Section 4, In the discussion, you proposed reasons for the fact that these ultra-clean days are prone to appear during the BB season. Besides the fact that the seasonal peaks in LWP and CF coincide with this time period, I think of the buffering system introduced by Stevens and Feingold, Nature, 2009, i.e. high loading of Na -> strong indirect effect -> higher LWP -> strong scavenging -> remove Na. As you mentioned a two-way aerosol-cloud interactions in the abstract, have you considered this as another possibility?

Our observations point to the importance of enhanced coalescence scavenging for driving and maintaining ultra-clean conditions in the SEATL MBL, but cannot indicate what might have caused the initial enhancement. However, given the evidence in our analysis for previous air mass contact with smoke even with low N_A, we certainly agree that the mechanism described in Stevens and Feingold (2009) may be at work at some point in the evolution of these boundary layers. The seasonal cycles of cloud cover and LWP in the region still seem as likely backdrops for generating these enhancements to enough of a point where extreme aerosol number depletions can develop. We have added mention in the Discussion of the Stevens and Feingold (2009) feedback as something for possible consideration in the Lagrangian LES studies we hope our results will motivate (P10, L3).

Referee #3

This is an interesting and mostly clearly-written manuscript. However, I was startled to learn that 'ultra-clean', as the authors have defined it, can still apply to boundary layers with elevated CO and rBC concentrations. It invites the question: what is a boundary layer with low Na, and non-elevated CO and rBC concentrations? 'extreme-clean'? I suggest that the authors rephrase 'ultra-clean' as 'ultra-low-aerosol' (or something along those lines), to be more specific. The term 'ultra-clean' appears to have been defined from northern hemisphere studies in which this distinction was not relevant, but I think for a new reader that the term 'ultra-clean' is confusing. The authors themselves touch on this on p. 8, second paragraph.

p. 2, line ~18: this is where the term 'ultra-clean' is introduced, in parentheses, with the paragraph providing detail on the prior studies that have used this term. I suggest including a subsequent paragraph that discusses how this term may or may not apply well to the southeast Atlantic, and use that to define 'ultra-low-aerosol'.
Thank you for your comments. We agree that the potential for influence from overlying smoke makes the SEATL a unique environment relative to many of the other regions where 'ultra-clean' conditions have been studied before. In fact, that's why we decided to extend analysis of these conditions into this region and shaped how we framed our three guiding research questions from the start. We feel that it is reasonable, at least for this study, to continue using the terminology that is consistent with the literature so far in order to directly connect to that prior work. We have made a few changes to the text to address this point:

- In the Abstract, we have added a new sentence (P1, L18-20) that more specifically addresses this potential discrepancy we had already mentioned in our Discussion and you raise in your comment: "Since exceptionally low accumulation mode aerosol numbers at ASI do not necessarily indicate the relative lack of other trace pollutants, this suggests the importance of regional variations in what constitutes an 'ultra-clean' marine boundary layer."
- We added "…as broadly defined for other regions in prior work noted above" when introducing our goal of expanding considerations of ultra-clean conditions to the SEATL at the end of the Introduction (P2, L31-32).
- We have added a sentence in Section 3.2 noting that we will return to the implications of the CO/rBC observations for 'ultra-clean' characterization in the Discussion (P6, L30-32).
- Added further emphasis on this distinction and the need for future work on this in the Discussion (P9, L15-18): "The wide range of trace pollutant concentrations observed over our sample of 41 days at ASI with exceptionally low N_A highlights the importance of carefully considering what constitutes an 'ultra-clean' MBL in a particular region. More work is needed on systematically comparing the variability of pollutants like CO and rBC during periods of otherwise low accumulation mode aerosol number both within and between regions"

P. 3, line 9: definition of 'ultra-clean' ('ultra-low-aerosol') needs more justification. Likely this follows that in prior studies, given the importance of this definition would suggest mentioning the definition within that paragraph on p.2 (and using a different term).

In response to this and other similar reviewer comments, we have added the following clarification to the text (P3, L15-26): "While admittedly somewhat subjective, this 50 cm^{-3} threshold is consistent with the upper bound of near-surface and below-cloud observations in MBL environments routinely featuring exceptionally low N_A such as subtropical pockets of open cells (Abel et al., 2019; Sharon et al., 2006; Terai et al., 2014), mid-latitude open-cellular convection (Abel et al., 2017; Pennypacker and Wood, 2017) and across the trade wind stratocumulus-to-cumulus transition (Bretherton et al., 2019). It is also well situated within the typical range (~30 – 60 cm^{-3}) of number concentrations used for the lowest aerosol cases in large eddy simulation studies of MBL aerosol-cloud interactions (Wang et al., 2010; Wang and Feingold, 2009; Yamaguchi and Feingold, 2015; Zhou et al., 2017). Prior work defined ultra-clean layers near the top of the MBL, often observed in the stratocumulus-to-cumulus transition, with $N_A < 10$ cm^{-3} (Wood et al., 2018). We argue it is reasonable to set a higher threshold near the surface, where aerosol number concentrations are generally higher due to proximity to the sea spray source. Furthermore, Wood et al. (2018) focused on these layers primarily as a mesoscale feature within larger cloud systems, whereas our interest is in studying ultra-clean conditions as daily-scale events. Defining ultra-clean conditions using daily median $N_A < 50$ cm^{-3} balances the need to reasonably capture conditions with exceptionally low near-surface N_A in the remote MBL while maintaining a robust sample of cases to study."

P.3 line 30: Would high aerosol counts but low CO/rBC qualify as 'polluted' ? The authors suggest this might occur during February. Overall a bit more description of the high-Na days would be helpful. Are they all from the months when smoke is clearly present?

Thank you for raising the need to clarify this point. The polluted back trajectories are only from the months where there are also ultra-clean days (i.e. during the biomass burning season). We have added text in the Abstract (P1, L16) and Section 2.2 (P4, L20-21), where the back trajectories are introduced, to ensure that this is clear. We have also added a supplemental table (Table S1) listing all of the ultra-clean and polluted dates from those same months that are included in the analysis.

Section 3.2, fig. 3: It would be interesting to also discriminate further those days that are more truly pristine. Do those correspond to the back-trajectories that more clearly go back to the southern oceans? There may not be many days with daily median CO values <~ 60 ppb and rBC values within the sensitivity limit, but there should be some, and it would add interest to hear about those as well.

We have expanded Section 3.2 (P7, L4-9), including the addition of a new supplemental figure, with the following: "However, trajectory latitude at seven days back from ASI only explains 25% of the variance in daily median CO concentrations across ultra-clean days. Trajectories from days with daily median CO ≤ 59 ppb (n = 6), the non-burning background median concentration, can be anywhere between 40° - 60°S at seven days back from ASI (Figure S2). Overall, boundary layer air mass origin is a relatively weak predictor of downwind variability in CO concentration on ultra-clean days." So while there is a weak suggestion that if a boundary layer trajectory originates farther toward the mid-latitudes/the Southern Ocean it ends up with lower CO around ASI, this is still not the dominant driver of variability across ultra-clean days.

This continues to point to the importance of variability above the boundary layer rather than to some systematic difference in boundary layer origin and/or trajectory.

Figures:

Fig. 1: It's hard to tell how many UC days occur per month from panel a and b. One idea would be to mention how many occur each month near the top of the figure.
In response to this and other reviewer comments, we have added a supplemental table (Table S1) that lists all the ultra-clean days for reference.

Fig. 2 panels c and d: I suppose this is saying something about temporal variability as well, with hourly values being shown for a given daily median threshold on Na. For completeness it would be nice to see a similar plot for the pdf of the hourly median Na. It would be a fifth panel. Not sure what to suggest for a 6th panel to balance it visually.
We had previously examined hourly N_A on ultra-clean days as a consistency check on the representativeness of the daily median concentrations (see box plots below for data by month, for example). There is certainly finer scale variability in N_A within ultra-clean days, but this is not the focus of our analysis. Figure 2 highlights the burning tracer results that more directly inform the conclusions of the paper.

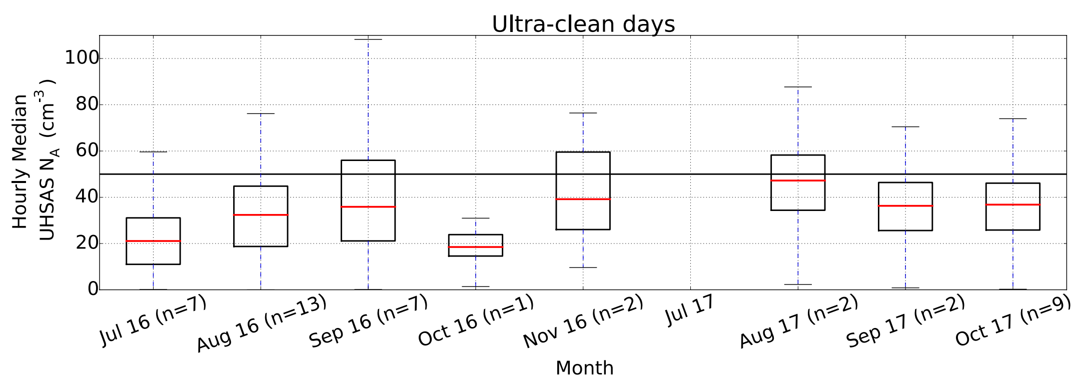


Fig. 4: the cumulative distributions take some study to interpret. Have the authors considered a normalized frequency distribution instead? Same for Fig. 5a.
We appreciate that CDF and PDF plots can both be useful. Our main goal here is to highlight the general differences and shifts in the distributions, which the CDF plots nicely demonstrate. To us, they also more clearly represent the parts of the distribution that contribute most to those shifts and by how much.

Fig. 6: I don't see a clear correspondence between LWP and UC days through this figure. I wonder if the MWR LWP data are simply too local.
Figure 6 shows the monthly boxplot distributions and seasonality for all of the LASIC TSI cloud fraction and MWR LWP data. This provides context for our observations, but is not just for any particular period featuring ultra-clean days. Ultra-clean days are only observed in months around the seasonal peak in cloud cover and LWP in this local dataset. The enhancements in coalescence scavenging, which would require cloudiness and high LWP, that we argue are likely responsible for driving ultra-clean conditions observed at ASI occur against the backdrop this broader seasonal pattern. These local observations will certainly be more variable than those over an

entire region or with a longer temporal record. We agree that local cloud properties will not be the only factor in driving ultra-clean conditions, and we point to the need to consider the detailed Lagrangian evolution of these boundary layers over several days in future work. We also reference prior work with satellite observations from the region that are largely consistent with the smaller-scale ASI observations (O'Dell et al., 2008; Zuidema et al., 2016). Nevertheless, even the local observations available from LASIC are consistent with a shift in cloud & precipitation properties (situated around seasonal maxima in cloud fraction and LWP) that could drive and maintain strong aerosol loss rates on ultra-clean days.

O'Dell, C. W., Wentz, F. J. and Bennartz, R.: Cloud liquid water path from satellite-based passive microwave observations: A new climatology over the global oceans, J. Clim., 21, 1721–1739, doi:10.1175/2007JCLI1958.1, 2008.

Zuidema, P., Chang, P., Medeiros, B., Kirtman, B. P., Mechoso, R., Schneider, E. K., Toniazzo, T., Richter, I., Small, R. J., Bellomo, K., Brandt, P., De Szoeke, S., Farrar, J. T., Jung, E., Kato, S., Li, M., Patricola, C., Wang, Z., Wood, R. and Xu, Z.: Challenges and prospects for reducing coupled climate model SST biases in the eastern tropical atlantic and pacific oceans: The U.S. Clivar eastern tropical oceans synthesis working group., 2016.

Referee #4

Manuscript Summary

The authors analyze surface observations of aerosol, gas phase composition, and cloud properties at Ascension Island over a period of 16 months, acquired during the Layered Atlantic Smoke Interactions with Clouds (LASIC) campaign. Back-trajectory calculations support the analysis. The authors distinguish three aerosol states at Ascension Island: Background conditions, polluted conditions, and ultraclean conditions. Ultraclean conditions are defined based on a daily median concentration of aerosol particles (CCN) with dry diameters between 60 nm and 1 µm < 50 cm−3 . The authors find 41 days with ultraclean conditions at Ascension Island. All of these occur during the South-West African biomass burning season. A portion of the ultraclean days also exhibits carbon monoxide and refractory black carbon levels above background. Apart from ultraclean days, boundary layer CCN concentrations at Ascension Island are significantly elevated above background levels. No days with ultraclean conditions are found outside the biomass burning season, which defines background conditions. The authors conclude, based on analysis of carbon monoxide and refractory black carbon levels, statistics of precipitation and liquid water path at Ascension Island, and back-trajectory calculations that CCN concentrations are low on the ultraclean days not because originally clean air has been advected to Ascension Island, but because enhanced coalescence scavenging in low clouds has strongly reduced CCN in polluted air masses. This is an interesting result because it points to a more complex interaction between (anthropogenic) aerosol and cloud properties in the region, with causal links in both directions.

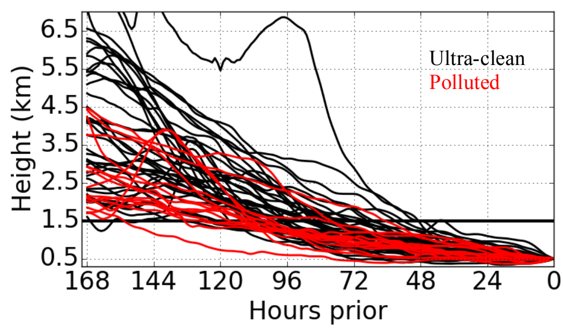
Review Summary

In their analysis of the observations the authors accumulated a good amount of circumstantial evidence to render their hypothesis plausible, although the analyzed data are specific to conditions at Ascension Island only and hence do not establish a causal connection between conditions on ultraclean days and processes that may give rise to them. Although not quantitative, the back-trajectory analysis is helpful. The study is, as the authors point out in their closing statements, a good motivation and starting point for subsequent investigations. There are a few points that I would ask the authors to look after, listed below. Otherwise, the manuscript is in good shape.

Major comments –

Could there be other explanations for the ultraclean days than enhanced coalescence scavenging in low clouds with higher liquid water content? E.g., is it possible that on the ultraclean days, the polluted air has entrained earlier into the boundary layer, hence spent a more time there compared to other days during the biomass burning season? A longer sojourn in the boundary layer would give coalescence scavenging more time to deplete the aerosol. Please comment and if applicable, discuss in the manuscript.

We agree that the time spent experiencing coalescence scavenging could also play a role in N_A variability sampled at ASI. Addressing when particular air masses entrained into the MBL wasn't the focus of this analysis since we are studying isobaric boundary layer trajectories. However, we have previously examined standard 3D trajectories (only for 2016) and did not find an immediately apparent systematic difference in the timing of when the ultra-clean day (black) and polluted (red) trajectories crossed below a 1.5 km threshold taken as roughly indicative of MBL height in the region. There is a fair amount of spread in height across the trajectories 3-5 days back from ASI, but both the trajectory and cloud/precipitation results presented still point to the importance of sources (smoke structure above the MBL) and sinks (scavenging) encountered along the way in driving the bulk of variability downwind rather than any consistent differences in the trajectories themselves. For future work, we do note the importance of considering the detailed evolution of a range of these cases in a Lagrangian modeling framework in the Discussion. This will hopefully provide more insight into the relative importance of different processes and their associated time scales along trajectories that cannot be addressed with the observations here.



Please calculate the average speed of the trajectories between 35 S and Ascension Island. Is there a difference in advection velocity between the ultraclean and nonultraclean days during the biomass burning season? If yes, discuss what this could mean for the processes that cause ultraclean conditions.
Thank you for raising this possibility. We do not observe any systematic differences in advection velocity between ultra-clean and polluted trajectories. Almost all boundary trajectories converge to within 2-3 lat/lon degrees of each other approximately 3 days back, leading to minimal differences in the speed with which they approach ASI. The large-scale horizontal circulation in the boundary layer is largely consistent once in the SEATL trades. We have now made note of this fact in the text (P7, L10-11).

The criterion for what makes ultraclean conditions varies between works. Albrecht et al. (doi:10.1175/BAMS-D-17-0180.1), e.g., define ultraclean conditions as having aerosol concentrations of less than 10 cm−3 in the nominal range between 0.06 - 1.0 μm., while in the present work it is < 50 cm−3 . Please add a passage mentioning the different criteria and explain why in the present work the criterion of < 50 cm−3 was chosen.
In response to this and other similar reviewer comments, we have added the following clarification to the text (P3, L15-26): "While admittedly somewhat subjective, this 50 cm^{-3} threshold is consistent with the upper bound of near-surface and below-cloud observations in MBL environments routinely featuring exceptionally low N_A such as subtropical pockets of open cells (Abel et al., 2019; Sharon et al., 2006; Terai et al., 2014), mid-latitude open-cellular convection (Abel et al., 2017; Pennypacker and Wood, 2017) and across the trade wind stratocumulus-to-cumulus transition (Bretherton et al., 2019). It is also well situated within the typical range (~30 – 60 cm^{-3}) of number concentrations used for the lowest aerosol cases in large eddy simulation studies of MBL aerosol-cloud interactions (Wang et al., 2010; Wang and

Feingold, 2009; Yamaguchi and Feingold, 2015; Zhou et al., 2017). Prior work defined ultra-clean layers near the top of the MBL, often observed in the stratocumulus-to-cumulus transition, with $N_A < 10$ cm^{-3} (Wood et al., 2018). We argue it is reasonable to set a higher threshold near the surface, where aerosol number concentrations are generally higher due to proximity to the sea spray source. Furthermore, Wood et al. (2018) focused on these layers primarily as a mesoscale feature within larger cloud systems, whereas our interest is in studying ultra-clean conditions as daily-scale events. Defining ultra-clean conditions using daily median $N_A < 50$ cm^{-3} balances the need to reasonably capture conditions with exceptionally low near-surface N_A in the remote MBL while maintaining a robust sample of cases to study."

How robust is the number of ultraclean days to the UHSAS < 50 cm−3 criterion?
See the comment above for our discussion of how we arrived at the threshold of 50 cm^{-3}. If you change the cutoff, you would of course increase or decrease the number of days in your sample (60 cm^{-3} puts you at 56 days, 30 cm^{-3} puts you at 19) but either start including conditions that deviate more from the observations and modeling of MBL environments typically characterized by exceptionally low N_A or limit the number of available days to study. The updated text above includes a consideration of that balance (see comment above).

"... with the correlation statistically indistinguishable from zero (r2 = 0.06), ..." To make this statement you /must/ calculate the p-value of the linear regression/correlation. Without the p-value, there is no way of telling whether a correlation coefficient/coefficient of determination is statistically indistinguishable from zero, regardless of its numerical value.
Thank you for raising the need to clarify this point. Our statement that this correlation (N_A vs. N_{CN3} on ultra-clean days) is statistically indistinguishable from zero is based on the fact that the correlation coefficient's calculated 95% confidence interval includes zero. We have updated the text to make this clear (P5, L27-28): "This relationship is substantially weaker ($r^2 = 0.06$), with the 95% confidence interval for this correlation including zero, on ultra-clean days (Figure 1d)."

Other comments

Please check the text for sentences that can be simplified; some are hard to understand. For example, "The relative invariance of isobaric boundary layer back trajectories between ultraclean and the most polluted days at ASI suggests that the potential for BBA entrainment set by the vertical separation of a smoke layer and the evolution of the boundary layer cloud field plays a larger role in upwind (e.g. at ASI) aerosol variability than a systematic difference in large-scale horizontal circulation in the boundary layer." is rather difficult to decipher.
Thank you for the suggestion to clear up the language here, we have separated this into two more straightforward sentences.

Please mention the meteorological input that you used to drive the HYSPLIT model.
Thank you for pointing out the need to include this information. The text has been updated to address this point: "We take a complimentary approach by analysing 7-day isobaric boundary layer back-trajectories initialized at approximately 500 m over ASI at 12:00 UTC as computed by the NOAA Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) with Global Data Assimilation System meteorology on a 0.5 degree by 0.5 degree grid (Stein et al., 2015)."

500 m trajectories are not isobaric.
In the updated text, we further clarify that the trajectories are *initialized* at approximately 500 m over ASI.

Please consider if the labeling of the abscissa in the plots that show data as a function of the month is precise enough to inform the reader on the actual point in time (are the vertical lines the 1st of the month or the 15th?)
The full time series (Figures 1a,b; 2a,b) are intended to give a general sense of how ultra-clean days fit into the larger pattern and seasonal cycle of aerosol and trace-gas observations at ASI, rather than for tracking the details around any one event or even month.

- Furthermore, in response to several reviewer comments we have added a supplemental table (Table S1) that lists all of the ultra-clean (and polluted) days for reference so individual events can be identified quickly if needed.
- We have updated the captions in Figures 1 and 2 to clarify that the vertical grid lines mark the first of each month labeled on the tick.

Ultra-clean and smoky marine boundary layers frequently occur in the same season over the southeast Atlantic

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Abstract. We study forty-one days with daily median surface accumulation mode aerosol particle concentrations below 50 cm^{-3} (ultra-clean conditions) observed at Ascension Island (ASI; 7.9°S, 14.4°W) between June 2016 and October 2017 as part of the Layered Atlantic Smoke Interactions with Clouds (LASIC) campaign. Interestingly, these days occur during a period of great relevance for aerosol-cloud-radiation interactions, the southeast Atlantic (SEATL) biomass-burning season (approximately June - October). That means that these critical months can feature both the highest surface aerosol numbers, from smoke intrusion into the marine boundary layer, as well as the lowest. While carbon monoxide and refractory black carbon concentrations on ultra-clean days do not approach those on days with heavy smoke, they also frequently exceed background concentrations calculated in the non-burning season from December 2016 - April 2017. This is evidence that even what become ultra-clean boundary layers can make contact with and entrain from an overlying SEATL smoke layer before undergoing a process of rapid aerosol removal. Because many ultra-clean and polluted boundary layers observed at Ascension Island during the biomass burning season follow similar isobaric back-trajectories, the variability in this entrainment is likely more closely tied to the variability in the overlying smoke rather than large-scale horizontal circulation through the boundary layer. Since exceptionally low accumulation mode aerosol numbers at ASI do not necessarily indicate the relative lack of other trace pollutants, this suggests the importance of regional variations in what constitutes an 'ultra-clean' marine boundary layer. Finally, surface drizzle rates, frequencies and accumulation – as well as retrievals of liquid water path – all consistently tend toward higher values on ultra-clean days. This implicates enhanced coalescence scavenging in low clouds as the key driver of ultra-clean events in the southeast Atlantic marine boundary layer. These enhancements occur against and are likely mediated by the backdrop of a seasonal increase in daily mean cloud fraction and daily median liquid water path over ASI, peaking in September and October in both LASIC years. Therefore the seasonality in ultra-clean day occurrence seems directly linked to the seasonality in SEATL cloud properties. These results highlight the importance of two-way aerosol-cloud interactions in the region.

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Comment [1]: Added in response to comments from Reviewers #2 and #3

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Comment [2]: Added in response to comments from Reviewer 3.

1 Introduction

Cloud-mediated aerosol radiative effects remain a significant source of uncertainty in our understanding of the climate system (Boucher et al., 2013; Rosenfeld et al., 2014). The Southeast Atlantic (SEATL) is a focal point for studying these effects because biomass-burning aerosol (BBA) particles transported from central and southern Africa frequently overly a major stratocumulus deck between approximately July and October (Devasthale and Thomas, 2011; Zuidema et al., 2016c). The regional peak in satellite-retrieved cloud fraction and aerosol optical depth, as well as vertical overlap between the smoke layer and clouds, tends to occur between September and October (Adebiyi et al., 2015; Zuidema et al., 2016a). This establishes the potential for a complex web of aerosol-cloud-radiation interactions on seasonal and regional scales.

By absorbing solar radiation, BBA can alter the thermodynamic structure of the lower troposphere, leading to changes in low cloud cover (Gordon et al., 2018; Johnson et al., 2004; Sakaeda et al., 2011; Tummon et al., 2010; Yamaguchi et al., 2015; Zhou et al., 2017). If smoke entrains into the marine boundary layer (MBL) and activates into a cloud droplet, BBA may also induce indirect effects (Costantino and Bréon, 2013; Diamond et al., 2018; Zhou et al., 2017). However, contact between the base of smoke layers and the cloud-topped MBL is highly variable and difficult to constrain with satellite remote sensing (e.g. (Rajapakshe et al., 2017)). At Ascension Island (ASI, details below), there is frequently heavy smoke intrusion into the MBL earlier in the burning season (June-August) than expected given the later (September-October) peak in aerosol optical depth (Zuidema et al., 2018). We should note that ASI is situated further to the west of the 'classically' defined (Klein and Hartmann, 1993) SEATL stratocumulus region. The full role of BBA in the SEATL MBL particle budget and its subsequent interactions with low clouds remains under investigation.

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Comment [3]: Double parentheses fixed in response to comment from Reviewer #2.

Generally, aerosol particle number concentrations in the remote MBL exhibit significant spatio-temporal variability (Allen et al., 2011; Anderson et al., 2003; Mohrmann et al., 2017). One feature of this variability observed at the surface is periods of extremely low (ultra-clean) accumulation mode (approximately 100 nm – 1 μm) number concentrations (Pennypacker and Wood, 2017; Wood et al., 2017). Wood et al. (2017) noted relative enhancements in satellite retrievals of cloud liquid water path (LWP), a crucial driver of MBL coalescence scavenging (Wood, 2006), in ultra-clean air mass back trajectories several days before arriving over the Azores in the North Atlantic. Pennypacker and Wood (2017) further explored the properties of the post-frontal open cellular clouds typically associated with these ultra-clean conditions over the North Atlantic. Other studies have noted ultra-clean layers near the top of the MBL, in subtropical pockets of open cells and during the stratocumulus-to-cumulus transition (Petters et al., 2006; Terai et al., 2014; Wood et al., 2018). These examples from both mid-latitude and subtropical MBLs point to heavy drizzle-driven coalescence scavenging in regions of changing low cloud morphology as key for driving this particular feature of MBL aerosol variability. Drizzle also plays an important role in setting the mean MBL aerosol state under subtropical stratocumulus (Wood et al., 2012).

Our goal is to expand these analyses of ultra-clean conditions, as broadly defined for other regions in prior work noted above, into the SEATL, especially given the unique potential for influence from BBA. Our study is structured around the following three questions:

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Comment [4]: Added in response to comments from Reviewer #3

1. Do ultra-clean conditions occur at the surface in the SEATL, and what is their place in aerosol variability?

2. How do concentrations of biomass burning tracers during any ultra-clean conditions compare to background values from the non-burning season?
3. Are ultra-clean conditions associated with enhancements in precipitation?

To address these questions, we employ observations from the first Atmospheric Radiation Measurement (ARM) Mobile Facility (AMF1) deployed to Ascension Island (7.9333°S, 14.41667°W) as part of the Layered Atlantic Smoke Interactions with Clouds (LASIC) campaign (Zuidema et al., 2016c, 2016b).

2 Data and Methods

2.1 Aerosol and Trace Gas Observations from AMF 1

A Droplet Measurement Technologies (DMT) Ultra-High Sensitivity Aerosol Spectrometer (UHSAS; Uin, 2016; DOI: 10.5439/1333828) provides aerosol number concentrations (N_A) at 0.1 Hz for particles with dry diameters between 60 nm and 1 µm. We define any day in the June 1, 2016 – October 30, 2017 LASIC UHSAS observational record (460 available days) as ultra-clean if the daily median N_A falls below 50 cm^{-3}. UHSAS data is currently unavailable for July 2017 due to unresolved quality control issues. While admittedly somewhat subjective, this 50 cm^{-3} threshold is consistent with the upper bound of near-surface and below-cloud observations in MBL environments routinely featuring exceptionally low N_A such as subtropical pockets of open cells (Abel et al., 2019; Sharon et al., 2006; Terai et al., 2014), mid-latitude open-cellular convection (Abel et al., 2017; Pennypacker and Wood, 2017) and across the trade wind stratocumulus-to-cumulus transition (Bretherton et al., 2019). It is also well situated within the typical range (~30 – 60 cm^{-3}) of number concentrations used for the lowest aerosol cases in large eddy simulation studies of MBL aerosol-cloud interactions (Wang et al., 2010; Wang and Feingold, 2009; Yamaguchi and Feingold, 2015; Zhou et al., 2017). Prior work defined ultra-clean layers near the top of the MBL, often observed in the stratocumulus-to-cumulus transition, with N_A < 10 cm^{-3} (Wood et al., 2018). We argue it is reasonable to set a higher threshold near the surface, where aerosol number concentrations are generally higher due to proximity to the sea spray source. Furthermore, Wood et al. (2018) focused on these layers primarily as a mesoscale feature within larger cloud systems, whereas our interest is in studying ultra-clean conditions as daily-scale events. Defining ultra-clean conditions using daily median N_A < 50 cm^{-3} balances the need to reasonably capture conditions with exceptionally low near-surface N_A in the remote MBL while maintaining a robust sample of cases to study. We take daily medians as a better indication of the aerosol number concentration over the course of a day since they are more robust to any outlier observations than daily means, though this choice only leads to a discrepancy over one day identified as ultra-clean. Observations of total particle concentrations from a TSI Incorporated Ultrafine (>3 nm, N_{CN3}) Condensation Particle Counter (CPC) compliment the UHSAS observations (Kuang, 2016; DOI: 10.5439/1046186).

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Comment [5]: Added in response to comments from Reviewer #2.

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Comment [6]: Added in response to comments from all reviewers.

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We also consider measurements of carbon monoxide (CO) and refractory black carbon (rBC) mass concentrations from AMF1. CO concentrations are measured at 1 Hz by a Los Gatos Research trace gas analyser, while a DMT Single Particle Soot Photometer (SP2; (Sedlacek, 2017)) measures the rBC. The black carbon concentrations are calculated on 10-second intervals with a sensitivity of 10 ng m^{-3}. Our primary goal with these data (Question 2) is to determine whether ultra-clean days represent the absence of any biomass burning influence in the MBL, relative to the regional background. This background is calculated from the non-burning season from December 2016 – April 2017. Both CO and black carbon act as biomass burning signatures, but since precipitation scavenging does not impact CO, it can reveal prior smoke contact even if aerosol concentrations are low. Again, we report median concentrations in order to minimize the potential impact of any outlier observations. We also report and compare inter-quartile ranges since a long tail on the distribution often skews the variability about these medians.

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Comment [8]: Added in response to comments from Reviewer #2.

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2.2 Back Trajectories

Systematic differences in surface aerosol concentrations and composition at ASI, like those between ultra-clean and smoky days, could be explained by upwind differences in MBL entrainment from the free troposphere. The frequency of contact between the smoke base and MBL top over the SEATL is notoriously difficult to constrain because the aerosol often significantly attenuates lidar beams (Rajapakshe et al., 2017). Zuidema et al. (2018) also posited that changes in transport pathway from the African continent, illuminated by three-dimensional back-trajectories, were key to explaining the smokiest conditions in the MBL near ASI. We take a complimentary approach by analysing 7-day isobaric boundary layer back-trajectories initialized at approximately 500 m over ASI at 12:00 UTC as computed by the NOAA Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) with Global Data Assimilation System meteorology on a 0.5 degree by 0.5 degree grid (Stein et al., 2015). We compare the behaviour of these trajectories on ultra-clean days and days within the same months that exceed their monthly 90th percentile of daily median N_A, which we label as polluted. See Table S1 for a complete listing of the specific dates. Isobaric trajectories specifically reveal the origins and paths of the boundary layers that would be entraining smoke from the free troposphere.

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Comment [10]: Edited to include details of meteorological forcing in response to comments from Reviewers #2 and #4.

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Comment [11]: Added in response to comments from Reviewers #2 and #3

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Comment [12]: Added in response to comments from Reviewer #2.

2.3 Clouds and Precipitation

Based on prior analysis of ultra-clean conditions in the mid-latitude (Pennypacker and Wood, 2017; Wood et al., 2017) and subtropical (Petters et al., 2006; Wood et al., 2018) MBL, we hypothesize that enhanced drizzle also plays an important role in driving aerosol variability over the SEATL. Local surface precipitation rates at ASI are measured over a one minute averaging period using a Parsivel2 Laser Disdrometer (Delamere et al., 2016; DOI: 10.5439/1150252). We calculate a daily precipitation frequency as the ratio of these averaging periods with a detected precipitation rate to the total within a day. This metric will of course not be a total drizzle frequency because it cannot include periods when precipitation evaporates before reaching the ground. We also examine differences in nonzero (i.e. only when clouds are detected) best-estimate retrievals of liquid water path from the AMF 2-channel microwave radiometer (Cadeddu et al., 2013; Gaustad et al., 2016; DOI: 10.5439/1027369) between ultra-clean and all other days in the LASIC record. These retrievals are reported in 40 minute averaging windows. LWP is a key driver of MBL aerosol loss through coalescence scavenging even when drizzle doesn't reach the surface. In particular, we examine the statistics of retrieved LWP across bins of daily median N_A (by 50 cm^{-3} from 0 - 400 cm^{-3}, by 100 cm^{-3} from 400 - 700 and then by 300 cm^{-3} from 700 - 1000 cm^{-3}). Finally, we place all of our observations in the context of the full LASIC record of both daily median MWR-retrieved LWP and cloud fraction as estimated by the ASI Total Sky Imager (Morris, 2005; DOI: 10.5439/1025308).

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3 Results

3.1 Ultra-clean days

Forty-one days meet our criteria for ultra-clean conditions (daily median $N_A < 50$ cm^{-3}) in the available LASIC data. The 28 events from 2016 and the 13 events from 2017 all occur between July and November (Figure 1a, Table S1). The distribution of events within these months varies, with August 2016 (12 days) and October 2017 (9 days) having the highest number of ultra-clean days in their respective years. As expected from Zuidema et al. (2018), we observe the highest daily N_A peaks in the early 2016 and 2017 biomass burning seasons (June - August). Understanding SEATL MBL aerosol variability in this crucial period thus requires an understanding of both smoke intrusions and ultra-clean conditions. In months with few or none of these extremes (October 2016 - April 2017), the daily and monthly median particle concentrations vary more consistently around 200 cm^{-3}.

Median N_{CN3} mostly follows the same seasonal pattern as N_A across the LASIC record (Figure 1b). This leads us to expect that the accumulation mode is generally an important driver of the variability in the total particle population. On ultra-clean days, however, the accumulation is by definition mostly depleted, while daily median N_{CN3} ranges from a 115 cm^{-3} minimum to a 374 cm^{-3} maximum. The range of total particle concentrations is therefore much higher than the range within the accumulation mode. On all except ultra-clean days, daily median N_A explains more than half ($r^2 = 0.65$) of the variance in the total particle concentration, as expected (Figure 1c). This relationship is substantially weaker ($r^2 = 0.06$), with the 95% confidence interval for this correlation including zero, on ultra-clean days (Figure 1d). While ultra-clean days tend to have lower N_{CN3} than other days, certainly those with smoke intrusions, the weakened correlation with N_A further confirms that different processes are responsible for governing the range of total particle concentrations outside of the accumulation mode. A similar difference in correlation strength between ultra-clean and other days holds at hourly time scales as well (Figure S1).

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Comment [15]: Added in response to Reviewer #4

3.2 Biomass Burning Signatures

Perhaps unsurprisingly, CO generally tracks the accumulation mode aerosol number concentrations in Figure 1, correlating with daily median N_A most strongly ($r^2 > 0.65$) in the early biomass burning seasons (June – August 2016 & 2017) when smoke influence in the boundary layer is highest (Figure 2a). Outside of the primary burning season (December 2016 – April 2017), the day-to-day N_A-CO correlation strength varies with r^2 values between 0.04 and 0.49, depending on the month. rBC also generally follows the same patterns as aerosol number and CO (Figure 2b), with day-to-day N_A-rBC correlation again strongest ($r^2 > 0.55$) in the early burning season. The 2017 observations again confirm the analysis of Zuidema et al. (2018), which based on the 2016 data, found that the signature of smoke in the ASI MBL is strongest earlier in the traditional SEATL biomass burning season. There is a smaller but noticeable peak in black carbon in January/February 2017 (Fig. 2b) that is oddly not evident in the CO observations. We leave a full diagnosis of this secondary peak for future work.

Of primary interest to this study is the range of BBA signature observations during ultra-clean events, relative to a background value. Prior observations in the subtropical Southern Hemisphere have put background CO concentrations between 50 - 60 ppb (Allen et al., 2008, 2011; Shank et al., 2012). The median of hourly median CO concentration on ultra-clean days is 69 ppb, with an inter-quartile range of 62 - 74 ppb, and the full distribution of ultra-clean CO concentrations exhibits some moderate bi-modality (Figure 2c). In the non-burning season (December 2016 - April 2017), the distribution shifts to generally lower CO concentrations. The background median CO concentration is 59 ppb and the inter-quartile range is between 55 and 65 ppb, consistent with the prior estimates noted above. The first mode of ultra-clean CO concentrations (Fig. 2c) overlaps more with the background distribution and is consistent with the background statistics. However, the second mode and longer tail of the distribution highlight the larger range of possible concentrations on ultra-clean days. This pulls the overall statistics toward higher concentrations on ultra-clean days relative to the non-burning background.

There is also some overlap in the distributions of ultra-clean and the non-burning background SP2 rBC (Dec. 2016, March-April 2017, Figure 2d). However, as with CO, the statistics do indicate a shift toward overall higher concentrations on ultra-clean days. The median of hourly median SP2 rBC is 51 ng m^{-3} with an inter-quartile range of 23 - 120 ng m^{-3} on ultra-clean days, compared to the background median of 20 ng m^{-3} and inter-quartile range of 12 - 45 ng m^{-3}. Even the hourly extremes captured by the 5th and 95th percentiles are higher on ultra-clean days (12 and 312 ng m^{-3}) than across the non-burning background (10 and 135 ng m^{-3}). In summary, there is no indication that ultra-clean days are devoid of BBA signatures or even exhibit the same distribution of smoke tracer concentrations as the non-burning season background at ASI. We will return to the implication of these results for the characterization of extremely low aerosol number events as 'ultra-clean' in the Discussion.

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Comment [16]: Months added in response to comments from Reviewer #2.

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Comment [19]: Edited in response to comments from Reviewer #2.

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Comment [20]: Added in response to comments from Reviewer #3.

Relative to the polluted extremes (recall these are defined by daily median N_A above monthly 95th percentile), there are somewhat more ultra-clean boundary layer isobaric back-trajectories that originate farther toward the mid-latitudes and the Southern Ocean (Figure 3a). We might expect lower background aerosol concentrations and weaker influence from African biomass burning in these air masses than in those spending more time in the subtropics, helping explain the subset of ultra-clean days with burning tracer concentrations closer to background levels. However, trajectory latitude at seven days back from ASI only explains 25% of the variance in daily median CO concentrations across ultra-clean days. Trajectories from days with daily median CO ≤ 59 ppb (n = 6), the non-burning background median concentration, can be anywhere between 40° - 60°S at seven days back from ASI (Figure S2). Overall, boundary layer trajectory origin is a relatively weak predictor of downwind variability in CO concentration on ultra-clean days. Furthermore, there are many polluted and ultra-clean boundary layers that follow similar isobaric trajectories on their way toward ASI (Figure 3b). By three days away from ASI, most trajectories have converged to within three to four degrees latitude and longitude of each other. In other words, the boundary layers that would be entraining smoke from the tree troposphere often follow very similar horizontal circulation patterns for both the highest and lowest upstream extremes of N_A. This all points to a smaller role for variations in large-scale horizontal circulation in the SEATL MBL in driving aerosol and trace gas variability observed at ASI.

3.3 Precipitation and Cloud Liquid Water

Ultra-clean days exhibit markedly different surface precipitation characteristics, as measured by the ASI Parsivel2. The distribution of precipitation rates shifts toward higher intensities on ultra-clean days (Figure 4a). Precipitation is also much more common on ultra-clean days (Figure 4b), with almost 90% of non-UC days having a precipitation frequency of less than 0.05. The tendency for more frequent and more intense precipitation inevitably leads to higher total accumulation on ultra-clean days (Figure 4c). The difference mostly stems from the shift toward more frequent drizzle conditions in ultra-clean conditions. These data are all presented with cumulative distributions in order to concisely highlight the generally different behaviour of precipitation across ultra-clean days. However, the increase in drizzle intensity, frequency and accumulation also holds for ultra-clean days relative only to the distribution within their respective months (not shown).

The median LWP retrieved by MWR measurements is higher on ultra-clean days (110 g m^{-2}) compared to other days (76 g m^{-2}). The inter-quartile spreads are actually larger than the median LWP whether within ultra-clean days (41 – 235 g m^{-2}) or not (26 – 192 g m^{-2}). These statistics are further illustrated by the difference in the LWP cumulative distributions (Figure 5a). The shift is noted across most of the sampled range of LWP, though the distributions do overlap at the very highest values. While the shift toward higher LWP on ultra-clean days may not appear substantial, recall that coalescence scavenging is non-linearly dependent on LWP (Wood, 2006). The approximately 35% increase in median LWP on ultra-clean days would strengthen the coalescence scavenging aerosol sink by 70%.

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Comment [21]: Added in response to comments from Reviewer #3.

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Comment [22]: Added in response to comments from Reviewer #4

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Comment [23]: Edited to include medians and inter-quartile ranges, rather than means, in response to comments from Reviewer #2.

Below a daily median N_A of about 150 cm^{-3}, daily median LWP generally increases with decreasing N_A (Figure 5b), still accompanied by high variability. This is indicative of higher LWP driving reductions in accumulation mode aerosol through drizzle production and scavenging. Over a wide range of intermediate daily median N_A (~150 - 500 cm^{-3}) there is no discernible variation in binned LWP that would point to a dominant process. At higher number concentrations, however, daily median LWP continues to drop with increasing N_A, implicating at least some role for a relative lack of thick, drizzling clouds in sustaining the highest accumulation mode number concentrations.

4 Discussion and Conclusions

The SEATL remains the focus of intensive study because of the potential for direct, indirect and semi-direct radiative effects arising from extensive biomass burning aerosol layers overlying a major stratocumulus deck. We utilize data collected from an ARM Mobile Facility deployed (June 2016 - October 2017) to Ascension Island during the LASIC campaign to study 41 days with daily median accumulation mode aerosol concentrations below 50 cm^{-3}. Perhaps counter-intuitively, all of these observed ultra-clean days occur between July and November, the season where BBA concentrations in the SEATL region generally peak. In the 2016 observations, ultra-clean days are particularly prevalent in July and August, and frequently both precede and follow the periods of heavy smoke intrusion into the MBL around ASI examined in Zuidema et al. (2018). In 2017, most of the ultra-clean days occur in October, but we hesitate to comment on the robustness of any interannual variability given the relatively infrequent sampling of these events and the two-year observational record. Satellite retrievals of cloud droplet number concentration (Grosvenor et al., 2018) may provide a tool for extending our analysis with both a longer temporal record and greater spatial context of extreme depletion events in the SEATL MBL. However, these retrievals remain far more uncertain in the broken and/or heavily drizzling cloud scenes that often coincide with ultra-clean conditions. Nonetheless, both years of the LASIC deployment situate ultra-clean days as a feature of surface aerosol variability at ASI during the broader SEATL biomass burning season. This naturally leads to the question of what might drive this seasonality in the occurrence of ultra-clean days.

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Comment [24]: Added in response to comments from Reviewer #1.

Surface precipitation rates, frequency and accumulation, as well as retrieved cloud LWP, are all systematically enhanced on ultra-clean days relative to non-ultra clean days. These observations are indicative of vigorous coalescence scavenging being a key driver of ultra-clean days at ASI. Clouds capable of initiating and sustaining this scavenging process are therefore likely precursors for these conditions. The months featuring ultra-clean days in the LASIC record are also the months leading up to and including the seasonal maximum in daily mean cloud fraction recorded by the ASI Total Sky Imager (Figure 6a). These same months also tend to be associated with the seasonal peak in daily median MWR best-estimate LWP values, though there is substantial spread in the monthly distributions in both 2016 and 2017 (Figure 6b). While limited to the two-year LASIC deployment period, these local observations are broadly consistent with prior work that has noted a seasonal maximum in satellite-retrieved SEATL regional cloud fraction (Zuidema et al., 2016a) and LWP (O'Dell et al., 2008; Zuidema et al., 2016a) between August and October. And though these satellite-based analyses tend to consider data from an area to the southeast of ASI, scavenging upwind of our observations is also likely important. Thus, the seasonality in ultra-clean day occurrence appears broadly tied to the seasonality of SEATL clouds. The increase toward the seasonal maximum in cloud cover and LWP likely provides the necessary backdrop for enhancements in coalescence scavenging needed to nearly fully deplete the accumulation mode in the MBL around ASI.

However, our results further shows that using the term "ultra-clean" incompletely describes conditions with extremely low accumulation mode particle number concentrations over the SEATL. Accumulation mode and total particle concentrations are generally well correlated at ASI, though much less so on ultra-clean days, on both daily and hourly scales. Even when N_A < 50 cm^{-3}, smaller particles can have 2 – 4 times the number concentrations than in the accumulation mode. The variability of Aitken and nucleation mode particle concentrations deserves more attention in future work, including any possibility of contributions from new particle formation. Carbon monoxide and refractory black carbon mass are also not necessarily at non-burning season (December – April) background levels despite the depletion of the accumulation mode. This points to the possibility of more frequent but subtler influence of smoke in the ASI MBL outside of the most extreme intrusions like those examined by Zuidema et al. (2018). The wide range of trace pollutant concentrations observed over our sample of 41 days at ASI with exceptionally low N_A highlights the importance of carefully considering what constitutes an 'ultra-clean' MBL in a particular region. More work is needed on systematically comparing the variability of pollutants like CO and rBC during periods of otherwise low accumulation mode aerosol number both within and between regions.

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This analysis highlights an additional layer of complexity in the SEATL aerosol-cloud system. The months featuring the highest daily concentrations of aerosol particles in the MBL around Ascension Island also feature the lowest, likely due to multi-day time scale enhancements in coalescence scavenging on top of a pre-existing seasonal cycle. Ultra-clean MBL conditions present an important test for large eddy simulation (LES) physics and provide a tool for further probing underlying processes and their associated timescales. The initiation, evolution and persistence of these conditions could make particularly interesting case studies for LES modelling of the Lagrangian evolution of the SEATL MBL. More broadly, air mass history is an important factor in the interpretation of aerosol-cloud interactions over the SEATL given the typical time scales associated with entrainment of free tropospheric aerosol into the MBL and loss from precipitation (Diamond et al., 2018). The broad similarities in isobaric boundary layer back trajectories even between ultra-clean and the most polluted days at ASI suggest that systematic differences in large-scale horizontal circulation in the boundary layer may play less of a role in downwind (e.g. at ASI) aerosol variability. Instead, the vertical separation between smoke and the MBL along air mass trajectories, in addition to the co-evolution of clouds and precipitation, could set a balance between entrainment and scavenging. The transport and consequent three dimensional structure of BBA layers certainly varies with circulation patterns above the boundary layer (Adebiyi and Zuidema, 2016). Abel et al. (2019) also noted relative reductions in the entrainment of overlying smoke tracers into the MBL in a pocket of heavily drizzling open cells near ASI, potentially

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Comment [26]: Sentences separated and edited in response to comments from Reviewer #4.

driven by cloud dynamical differences noted in other previous work (Berner et al., 2011). The progression of clouds and precipitation along trajectories in the SEATL will depend on a number of factors, including potential influence of overlying smoke layers (Yamaguchi et al., 2015; Zhou et al., 2017) and buffering feedbacks (Stevens and Feingold, 2009). The detailed evolution of how all of this might lead to downwind ultra-clean conditions and the variations in other trace pollutant concentrations observed during these events should be further explored in a Lagrangian LES framework. Coarser resolution models used to study aerosol-cloud interactions across the broader SEATL region should also test their capability of reproducing these events and their place in the aerosol, cloud and meteorological seasonal cycles.

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Comment [27]: Added in response to comments from Reviewer #2.

Acknowledgements

Sam Pennypacker's work was supported by the United States Department of Energy (DOE) award DE-SC0013489 (ENA Site Science). Michael Diamond's work was supported by NASA headquarters under the NASA Earth and Space Science Fellowship Program, grant NNX-80NSSC17K0404. Robert Wood's work was supported by NASA Earth Venture Suborbital-2 grant NNX-15AF98G. The U.S. Department of Energy, Office of Science, Office of Biological and Environmental Research, Climate and Environmental Sciences Division sponsors the Atmospheric Radiation Measurement (ARM) program. We are indebted to the scientists and staff who make these data possible by taking and quality controlling the measurements. The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and/or READY website (http://www.ready.noaa.gov) used in this publication. Particular thanks to Paquita Zuidema for helpful discussions about the LASIC campaign and to Arthur Sedlacek for providing access to and discussing the SP2 dataset.

Data Availability

All LASIC ARM data are publicly available at https://www.archive.arm.gov/discovery/. ARM dataset DOI references are included in the text.

Author Contributions

SP and RW conceived the study design and analysis. SP performed the analysis of the LASIC data. MD ran the HYSPLIT transport and dispersion model to produce the back-trajectories. SP wrote the paper and all authors reviewed the manuscript.

Competing interests

The authors declare that they have no conflict of interest.

Special Issue Statement

This article is part of the special issue “New observations and related modelling studies of the aerosol–cloud–climate system in the Southeast Atlantic and southern Africa regions (ACP/AMT inter-journal SI)”. It is not associated with a conference.

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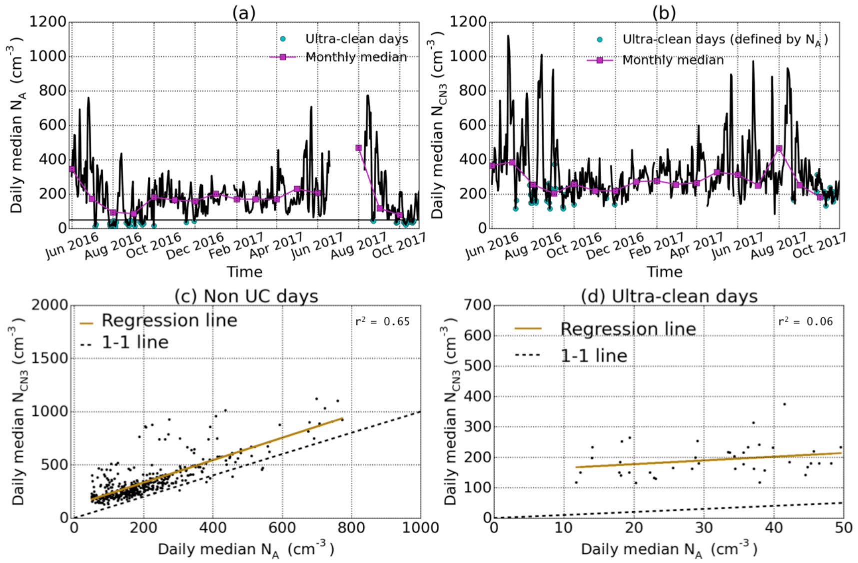

Figure 1: Time series of daily and monthly median (a) N_A and (b) N_{CN3} measured during LASIC, with ultra-clean days marked in cyan. In (a) and (b), vertical grid lines mark the first of each month labeled on the tick. Daily median N_{CN3} is then regressed against daily median N_A for both (c) non ultra-clean days and (d) ultra-clean days.

Author
Comment [28]: All figures have been updated with thicker grid spacing and more spacing for the tick labels to improve readability.

Author
Comment [29]: r^2 values added in response to comments from Reviewer #2.

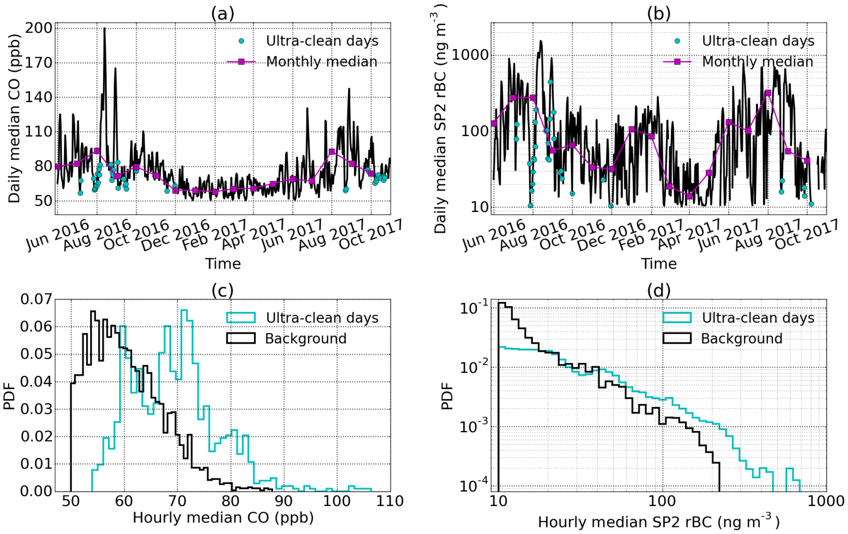

Figure 2: Time series of daily and monthly median (a) CO and (b) rBC measured during LASIC, with ultra-clean days marked in cyan. In (a) and (b), vertical grid lines mark the first of each month labelled on the tick. We then compare the PDF of hourly median (c) CO and (d) rBC from ultra-clean days to the PDF of hourly median concentrations from each tracers' respective non-burning background.

Author

Comment [30]: Figure updated to include PDFs in response to comments from Reviewer #1.

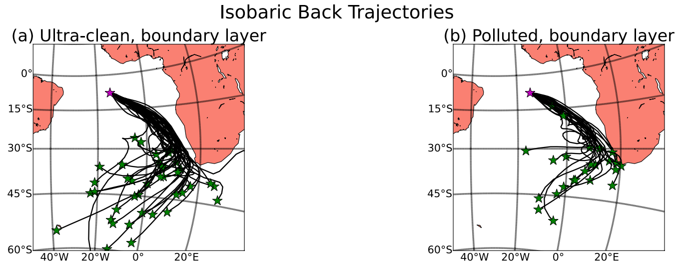

Figure 3: Isobaric 7-day HYSPLIT back trajectories at 500 m for (a) ultra-clean and (b) polluted days from ASI.

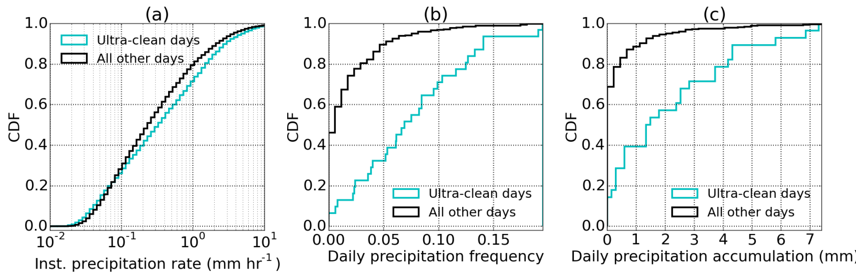

Figure 4: Cumulative distributions of (a) instantaneous precipitation rate, (b) daily precipitation frequency and (c) daily precipitation accumulation as measured by the ASI Parsivel2 laser disdrometer.

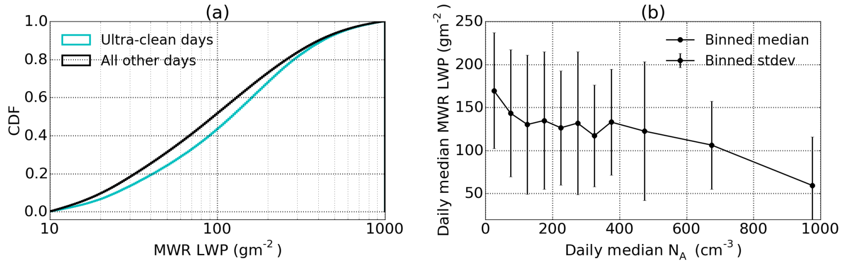

Figure 5: (a) Comparison of the cumulative distributions of best-estimate LWP retrieval from the ASI MWR between ultra-clean and all other days and (b) medians/standard deviations of daily median MWR LWP across bins of daily median accumulation mode aerosol for the entire LASIC record. In (b), bin widths were selected to account for varying density of days across the range of aerosol concentrations while still visualizing the broader pattern.

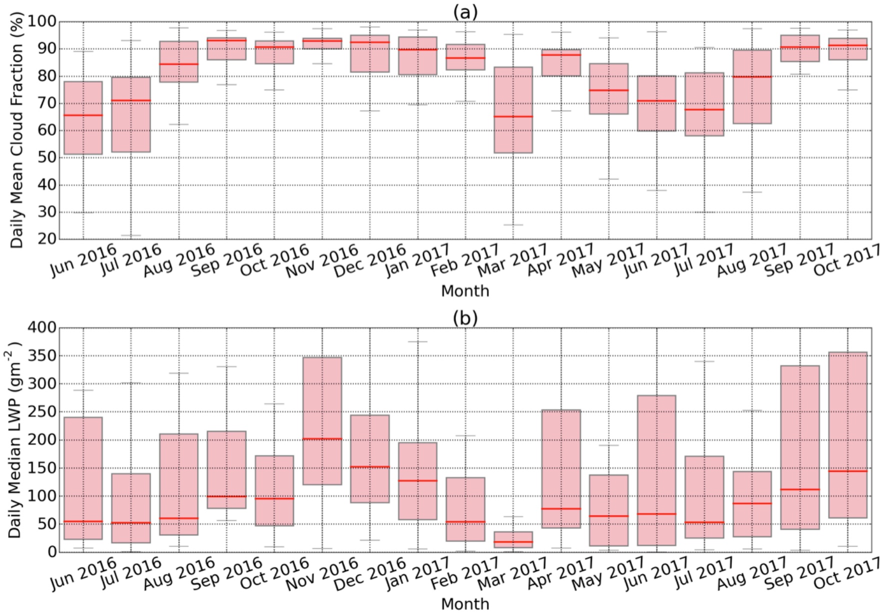

Figure 6: Monthly boxplots of (a) daily mean Total Sky Imager cloud fraction and (b) daily median MWR best-estimate LWP for each month in the LASIC record.