

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

We would like to sincerely thank the editor and reviewer for their time, effort, and thoughtful feedback on our manuscript. The reviewer comments are shown in **blue**, with the authors' responses shown in **black** and any edited manuscript language shown in *italicized black font*.

Reply to the Reviewer #1

Wu et al. presented a study investigating the impacts of African biomass burning aerosols (BBAs) on liquid clouds over the southeast Atlantic region. This study combines both airborne observations on aerosol properties and cloud micro physics properties and numerical simulations for air mass sources and properties. It reveals that the presence of BBAs can increase the observed cloud droplet number but decrease cloud droplet size. In addition, this study evaluates the BBA-cloud interaction activities under both free troposphere and marine boundary layer conditions. Overall, the revised manuscript is improved compared with the original submission. If comments below can be addressed, we would like to suggest acceptance for publishing in ACP.

Major comments:

1. In addition to presenting NAME simulations for three cases (Figure S2) as representatives for three periods in Figure 2, the authors should provide one simulation result for each flight observation (e.g., releasing air parcels in the middle time of each flight). This will make the paper stronger. One would worry about whether the three cases presented in Figure S2 are the best cases. For example, one would wonder if one of the flight observations during period 2 with similar fractions of air mass from African continent to those of case 2 in Figure S2 also shows similar air mass back trajectories. In addition, rationales for selecting case 2 are not clearly provided in discussions on Figure 2. For example, it should be directly noted that it is a reference case as clean MBL for comparing with BBA polluted case 1 and 3.

Thanks to the reviewer's suggestions.

1) As described in the method section (Sect.2.2), we performed backward-dispersion simulations **at 3-hour (h) intervals** throughout the campaign period. In each simulation, a certain amount of hypothetical tracer particles was released from a $2^\circ \times 2^\circ$ grid box centered around the Ascension Island observation site (14.35°W , 7.96°S). To distinguish source origins of MBL and FT air mass over Ascension Island, we also performed simulations that released tracer particles within the MBL (341 ± 300 m) and FT (2.5–4 km) altitude ranges separately. In the original Sect. 3.1, we first present three representative backward-dispersion fields (Cases 1 to 3), that illustrate the major transport pathways of air parcels during the three defined periods. To assess the contributions of polluted air mass originating from the African continent to the Ascension Island area, we calculated the fractional contributions of original air mass over the African domain (20°S – 5°N , 9°W – 35°E) based on the outputs of 3-hourly NAME back-dispersion fields. This continental domain is closely associated with BB pollutions identified in satellite wildfire observations (Figure 1). The resulting analysis provides a quantitative indication of BB plume influence on the Ascension Island area throughout the campaign period, as shown in Fig. 2b (FT simulations) and Fig. 2c (MBL simulations). Therefore, the conclusions regarding pollution sources are supported not only by the three representative examples of dispersion fields shown in the original Fig. S2, but also by the statistical analysis of the overall African source contributions presented in Fig. 2b (FT simulations) and Fig. 2c (MBL simulations). To address the reviewer's concern, we have further included back-dispersion fields corresponding to each flight observation, which are now provided in Figure S3–4 of the revised Supplementary. The corresponding description in Sect 3.1 has also been rephrased as below:

“Example backward-dispersion fields corresponding to release times within the flight sampling period are presented, distinguishing between FT (Fig. S3) and MBL (Fig. S4) simulations. FT dispersion simulations (Fig. S3) indicate that most FT air parcels over Ascension Island originated from westerly flow across the SEA and African continent. Compared to clean FT cases from Period 1, polluted FT cases from Periods 2 and 3 show a substantially greater influence from African continental airmasses. MBL dispersion simulations (Fig. S4) suggest that MBL air parcels over Ascension Island mainly arose from clean oceanic flow that transported from the southeast to northwest over the SEA. Polluted MBL cases from Periods 1 and 3 also show contributions of westerly flow originated from the African continent, while negligible continental influence in clean MBL cases from Period 2. To assess the contributions of polluted air mass originating from the African continent to the Ascension Island area, we calculated the fractional contributions of original air mass over the African domain (20° S–5° N, 9° W–35° E) based on the outputs of 3-hourly NAME back-dispersion fields. This continental domain is closely associated with BB pollutions identified in satellite wildfire observations (Fig. 1). The resulting analysis provides a quantitative indication of BB plume influence on the Ascension Island area throughout the campaign period, as shown in Fig. 2b (FT simulations) and Fig. 2c (MBL simulations). The temporal evolution of continental contributions aligns well with the observed FT and MBL pollution patterns over Ascension Island. NAME simulations evidence that the FT and MBL pollutions observed during the campaign were attributable to long-range transport of African continental air mass which brought BB plumes.”

- 2) We have added in the manuscript: “Case 2 is a reference clean-MBL case for comparison with BB-polluted MBL cases (Cases 1 and 3).”.

2. This study focuses on the impacts of BBAs from Africa on SAE clouds. It is stated that ‘the presence of BB-pollution was defined using thresholds of carbon monoxide above 83 ppb and black carbon above $0.1\mu\text{g m}^{-3}$ (Line 223-225 in Section 3). However, Figure 2 and Figure S2 present results about all airmasses from African continent which may include all kinds of continental pollutions. To clearly indicate the presence and influence of BBAs, the abundance of carbon monoxide and black carbon as a function of time should also be provided in Figure 2 or in the supplement material.

Thanks to the reviewer’s suggestion.

We have added Figure S2 in the supplement material, including the measurements of carbon monoxide (CO) and black carbon (BC) from the CLARIFY campaign and surface observations on Ascension Island.

The revised manuscript includes:

Figure S2 presents vertical distributions of CO (Fig. S2a) and BC (Fig. S2b) for the CLARIFY flights (C028 to C051).

Concurrent surface observations of CO and BC on Ascension Island (Fig. S2) presented the same trend of MBL BB pollution as observed during the CLARIFY campaign (Zuidema et al., 2018).

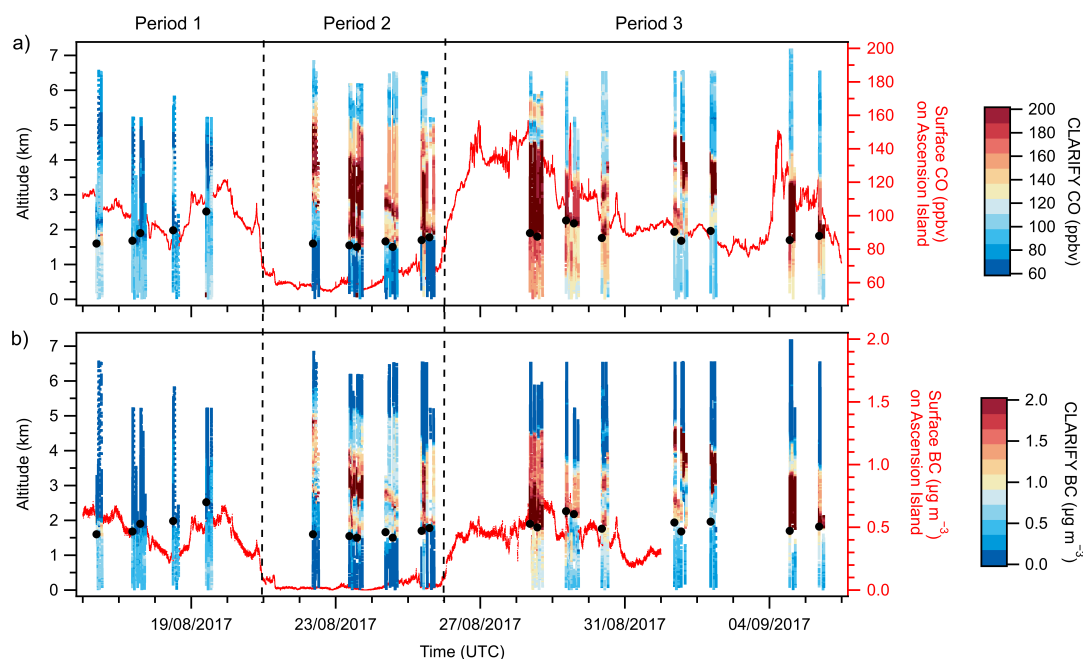


Figure S2. a) Vertical distributions of CO (ppbv) from the CLARIFY flights (C028 to C051), alongside the estimated z_i from each flight (black circles). The right axis is the surface CO (red line) observed on Ascension Island (Zuidema et al., 2018). a) Vertical distributions of BC ($\mu\text{g m}^{-3}$) from the CLARIFY flights (C028 to C051), alongside the estimated z_i from each flight (black circles). The right axis is the surface BC (red line) observed on Ascension Island (Zuidema et al., 2018).

3. Relevant text statements for introducing each figure and table in the Supplementary document should be provided.

Thanks to the reviewer's suggestion.

We have included text statements for introducing Figures/Table in the supplementary. In the revised supplementary, two sections are added: "S1 Observations of aerosol components and carbon monoxide" and "S2 NAME backward-dispersion fields".

Minor comments:

1. In the caption of Figure 8 and Figure S2, it should be clarified that Figure 8 is based on the output of 3D air parcel footprints whereas Figure S2 is based on the method introduced in the second paragraph in section 2.2.

We have added the related description to the captions.

2. Line 47: to reduce the relative humidity

Accepted.

3. Better to also provide the flight numbers for flights with mainly limited cloud samplings that are not included in this study.

The revised manuscript is:

Transit flights (C040-C041, predominately at high altitudes) and flights with mainly limited cloud samplings (C028, C030, C034, C035, and C043) are not included in this study.

4. Line 128-129 (and Line 133): PCASP is missing here for Passive Cavity Aerosol Spectrometer Probes.

Aerosol size distributions were measured at 1-Hz resolution via two wing-mounted Passive Cavity Aerosol Spectrometer

Probes (**PCASP**), which resolved number concentrations in 30 diameter bins between 0.1 and 3 μm .

5. Line 145-146: the general size range of BBAs observed in this region should be provided here (or refer to the literature).

The related description has been added to the manuscript.

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

6. Line 161: Why is 0.01 g m^{-3} used as the threshold LWC? Any rationale or reference?

A reference of (Heymsfield and McFarquhar, 2001) has been added to the manuscript.

Heymsfield, A. J. and McFarquhar, G. M.: Microphysics of INDOEX clean and polluted trade wind cumulus clouds, J. Geophys. Res., 106, 28653–28673, 2001.

7. Line 201: what is BL? Only MBL was defined before this.

The definition of “BL” has been added.

*Over the SEA, there is typically a strong thermodynamic capping inversion that inhibits turbulent mixing between the **boundary layer (BL)** and overlying FT air...*

8. Line 236: isn't case 1 at the middle of period 1?

*Case 1 released tracers in **the middle of Period 1** (12:00 UTC, August 18).*

9. Figure 2: Why not choose case 2 and case 3 at a point where flight observations are available?

As shown in the original Fig. 2 and the newly added Fig. S2, the MBL over Ascension Island became considerably clean from August 21 to 25 (Period 2), and became BB-polluted again from August 26 to September 5 (Period 3). Accordingly, we selected Cases 2 and 3 to release tracers near the start of Periods 2 (12:00 UTC, August 21) and 3 (12:00 UTC, August 26), respectively, aligning with these shifts in MBL pollution conditions.

The revised manuscript includes:

“Cases 2 and 3 released tracers near the start of Periods 2 (12:00 UTC, August 21) and 3 (12:00 UTC, August 26), respectively, coinciding with shifts in MBL pollution conditions observed over Ascension Island.”

10. Line 248-249: it might be too arbitrarily to use three cases to conclude the pollution source of the whole campaign. It is necessary to provide back trajectory results for each flight observation. Same as major comment 1.

Based on the outputs of 3-hourly NAME back-dispersion fields, we calculated the time series of the fractional contributions of air mass originating from the African continent (20°S–5°N, 9°W–35°E) to the Ascension Island area. The resulting analysis provides a quantitative indication of BB plume influence on the Ascension Island area throughout the campaign period, as shown in Fig. 2b (FT simulations) and Fig. 2c (MBL simulations). Therefore, the conclusions regarding pollution sources are supported not only by the three representative examples of dispersion fields shown in the original Fig. S2, but also by the statistical analysis of the overall African source contributions presented in Fig. 2b (FT simulations) and Fig. 2c (MBL simulations).

11. Line 276-278: please refer to Figure 3a. Also, please check through the manuscript to make sure relevant figures/tables are correctly and properly referred to in the discussion. Some of in-text references are missing, which makes it a bit hassle to follow the flow.

The in-text references of figures/tables have been checked and referred to relevant discussions throughout the manuscript.

12. Line 300: Also provide the value of the CCN activation fractions of these highly aged African BB aerosols

The fraction value has been added.

Due to atmospheric aging, the CCN activation fractions of these highly aged African BB aerosols ($CCN/N_a = 0.82 \pm 0.17$, at $SS = 0.2\%$) are generally higher than fresher BB aerosols sampled over the African continent ($CCN/N_a = 0.68$, at $SS = 0.3\%$)...

13. Line 304: A sentence for clear definition of clean MBL is missing in the manuscript. It should be already introduced in Section 1 or Section 2

The selection of clean BL air masses was added in the manuscript.

Clean BL air masses were selected when $CO < 66$ ppbv ($53 \mu g m^{-3}$), which corresponds to the lowest 5th percentile of all CO data collected in the MBL (Wu et al., 2020).

14. Line 305: “The aerosols in the clean MBL were smaller”, is the number or size of aerosols small?

The aerosol sizes in the clean MBL were smaller than in the BB-polluted MBL during CLARIFY.

15. Line 312-316: based on the results, the two individual statements seem right. However, it is awkward when putting them together

The description of CCN/CN_3 has been removed in the revised manuscript.

“However, average CCN/CN_3 in the clean MBL (0.38 ± 0.18) was much smaller than in the BB impacted MBL (0.76 ± 0.10), as the dominant Aitken mode particles ($< 0.1 \mu m$) in the clean MBL are not sufficiently large to act as CCN (Dusek et al., 2006).”

16. Figure 3: provide legend in each panel. A missing legend makes it not convenient to read it.

The legends have been added in Fig. 3.

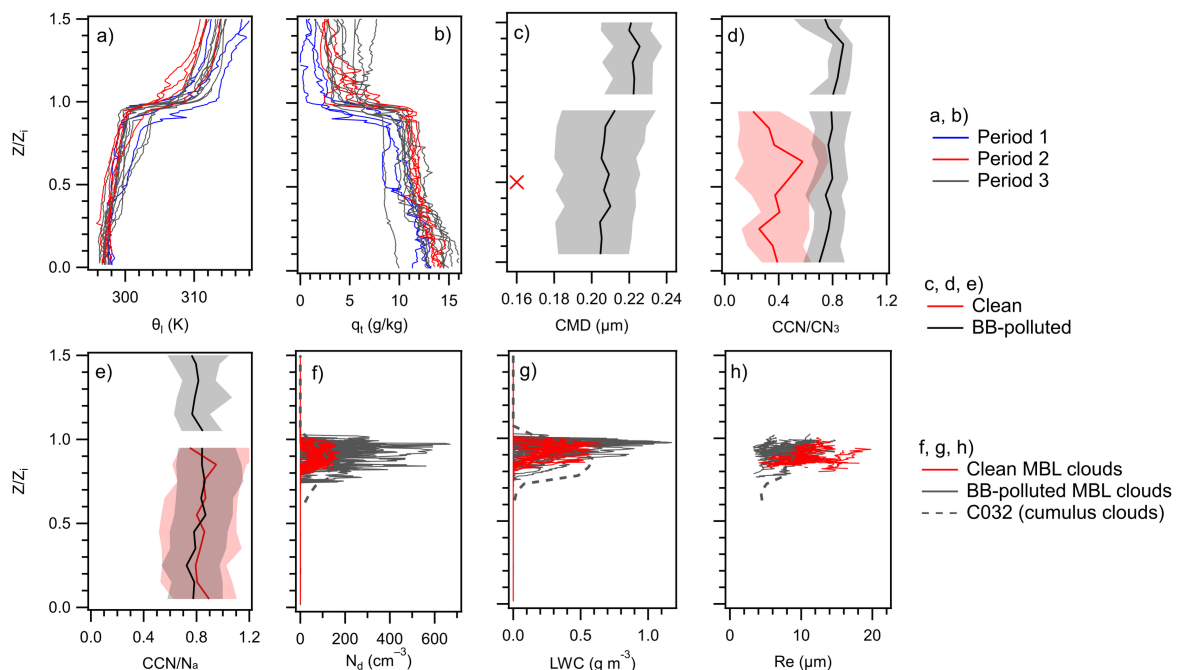


Figure 3. a, b) Average vertical profiles of a) liquid water potential temperature (θ , K), b) total water mixing ratio (q_t) for each flight used in this study. Blue, red and black lines represent measurements from Periods 1, 2 and 3, respectively. c-e) Summarized profiles of c) aerosol count median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

17. Line 406: ambiguous statement. Enhanced N_d value or larger N_d distribution range?

The revised manuscript is:

The enhanced values and broader range of sub- N_a in the BB-polluted MBL led to substantially higher and more variable N_d values as compared to the clean MBL (Fig. 5a).

18. Line 406-407: Figure 5a shows what kind of effects? It should be clearly stated out what the figure/result show but not only say ‘affect’.

The revised manuscript is:

*The enhanced values and broader range of sub- N_a in the BB-polluted MBL led to substantially higher and more variable N_d values as compared to the clean MBL (Fig. 5a), showing that **transported BB aerosols promote droplet activation and cloud formation in this region.***

19. Figure 8: indicate the atmospheric condition and case number in the legend for each panel.

The case numbers (Case 1, Case 2, and Case 3) and attribution column ranges (whole column, FT, and BL) of dispersion results have been added in Fig. 8.

20. Line 609-611: the continuous increase of CTH cannot explain why there is a decrease in R_e at -24 hours. Further details/results should be provided to explain this?

The revised manuscript has explained that: “Concurrent MBL deepening and enhanced CTH could promote condensational growth, yielding larger droplets at the cloud top (Painemal et al., 2014). This agrees with the observed overall increase in R_e along the BL transport. However, as the MBL continues to deepen and the entrainment of dry FT air strengthens, LWC may decrease, explaining the subsequent decline in R_e observed from -24 h (Wood, 2012; Ryoo et al., 2022).”

Reply to the Reviewer #3

General comments:

The authors sufficiently addressed the referee's comments. I recommend publication after minor edits (see below) and additional proofreading.

Minor edits:

Line 36. Change "radiations" to singular.

Accepted

Line 39. Add period. Also, I do not see a paper by Wood in 2012 in your references. Please add it and make sure he is the sole author on that paper, otherwise an "et al." should be added to the citation.

The citation has been checked:

Wood, R.: *Stratocumulus clouds*, *Mon. Weather Rev.*, 140, 2373– 2423, <https://doi.org/10.1175/MWR-D-11-00121.1>, 2012.

Line 48. Specify what about the semi-direct effect is "negative".

The revised manuscript is:

Modeling studies suggest that BB aerosols exert an overall negative semi-direct radiative forcing (cooling) over the SEA during the fire season.

Line 185. "e.g." is more appropriate than "i.e." because you are providing an example of a study. Other uses of "i.e." in the manuscript also do not seem appropriate and should be checked.

The uses of "i.e." have been checked and replaced by "e.g." throughout the manuscript.

Line 225. "Respectively" should be deleted.

Accepted

Line 238. When using "respectively" in a sentence, a comma must be added before. Please fix this issue throughout the manuscript. Also, double check the abbreviation "Figs." rather than "Fig." is correct under ACP guidelines.

A comma has been added before "respectively" in related sentences.

The use of "Figure" and its abbreviation have been checked throughout the manuscript. They now follow the ACP guidelines, e.g. "Figure 1", "Fig. 1", "Figure 3f–h", "Fig. 3f–h", "Figures 9c and 9d" and "Figs. 9c and 9d".

Lines 326 and 344. Change "samling" to "sampling".

Accepted

Line 346. Change "wiskers" to "whiskers".

Accepted

Line 464. "CTtoAB" is different that the variable on the x-axis in panel a.

The variable on the x-axis has been revised in Fig. 6a.

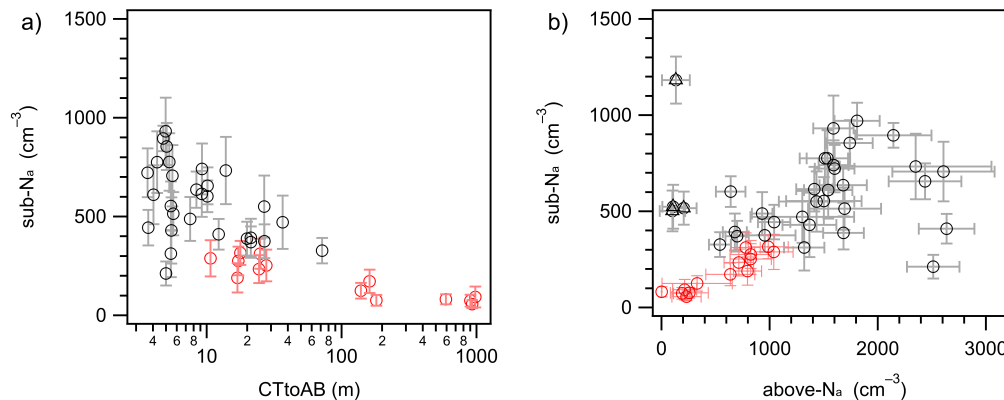


Figure 6. Relationships a) between sub- N_a and the distance from the top of cloud layers in the MBL to the bottom of BB layers in the FT (Cloud Top to Aerosol Base, CTtoAB); b) between sub- N_a and above- N_a . The markers and error bars represent the average values and standard deviation for each profile. Black triangle and circle markers are from the BB-polluted MBL in Periods 1 and 3 respectively, and red circle markers are from the clean MBL in Period 2.

Line 573. Change “abudance” to abundance”.

Accepted

Line 605. Change the word “Figures.” to correct abbreviation.

Accepted

References

- Heymsfield, A. J. and McFarquhar, G. M.: Microphysics of INDOEX clean and polluted trade wind cumulus clouds, *J. Geophys. Res.*, 106, 28653–28673, 2001.
- Painemal, D., Kato, S., and Minnis, P.: Boundary layer regulation in the southeast Atlantic cloud microphysics during the biomass burning season as seen by the A-train satellite constellation, *J. Geophys. Res.-Atmos.*, 119, 11288–11302, <https://doi.org/10.1002/2014JD022182>, 2014.
- Ryoo, J.-M., Pfister, L., Ueyama, R., Zuidema, P., Wood, R., Chang, I., and Redemann, J.: A meteorological overview of the ORACLES (ObseRvations of Aerosols above CLouds and their intEractionS) campaign over the southeastern Atlantic during 2016–2018: Part 2 – Daily and synoptic characteristics, *Atmos. Chem. Phys.*, 22, 14209–14241, <https://doi.org/10.5194/acp-22-14209-2022>, 2022.
- Wood, R.: Stratocumulus clouds, *Mon. Weather Rev.*, 140, 2373–2423, <https://doi.org/10.1175/MWR-D-11-00121.1>, 2012.
- Wu, H., Taylor, J. W., Szpek, K., Langridge, J. M., Williams, P. I., Flynn, M., Allan, J. D., Abel, S. J., Pitt, J., Cotterell, M. I., Fox, C., Davies, N. W., Haywood, J., and Coe, H.: Vertical variability of the properties of highly aged biomass burning aerosol transported over the southeast Atlantic during CLARIFY-2017, *Atmos. Chem. Phys.*, 20, 12697–12719, <https://doi.org/10.5194/acp-20-12697-2020>, 2020.
- Zuidema, P., Sedlacek III, A. J., Flynn, C., Springston, S., Delgadillo, R., Zhang, J., Aiken, A. C., Koontz, A., and Muradyan, P.: The Ascension Island Boundary Layer in the Remote Southeast Atlantic is Often Smoky, *Geophys. Res. Lett.*, 45, 4456–4465, <https://doi.org/10.1002/2017GL076926>, 2018.