Review #2 of “Aerosol first indirect effect of African smoke in marine stratocumulus clouds over Ascension Island, south Atlantic Ocean” by M. de Graaf et al.

In this work, the authors report on cloud microphysical properties of low-level marine clouds inferred from UV-polarization lidar. The lidar was deployed during the dry season months of 2016 and 2017 on a remote south Atlantic island. A new technique developed in an earlier work (Donovan et al., 2015) was applied to infer microphysical parameters (aerosol optical depth, cloud droplet effective radius, and cloud droplet number concentration) and compared with in situ measurements from AERONET and instruments deployed during the ARM LASIC campaign. Although the work provides valuable insights into the complex ACI at Ascension Island, the authors have contributed some preliminary understanding to processes contributing to the observed interactions due to smoke intrusions into the cloud deck, environmental and instrumental effects on measured uncertainties, but they do not relate their findings to the growing body of literature in this region for comparison. I believe this paper is worthy of publication after these components have been more clearly addressed for compliance with ACP criteria, therefore major revision is recommended.
The reviewer is thanked for the extensive and helpful review of the paper. We agree that the paper should be related to large body of existing studies. The reviewer is acknowledged for the many suggestions for accompanying papers, which are now cited in the new manuscript. The manuscript was completely rewritten, especially the introduction and conclusion sections, to reflect the contributions to this field.
Below, the suggestions and comments raised by the reviewer are all answered in detail, and the changes to the manuscript are indicated.

Major comments:

This paper would benefit from a more complete description of the context of the work and its motivation. To this end, the introduction should be expanded. Particularly, the authors provide no description of the first indirect / Twomey effect in the introduction and only offer a vague claim that drizzle accompanying low-level marine clouds can be modulated by an interaction with aerosol. Many modeling and observational studies have conducted examinations of aerosol effects on low-level marine clouds (e.g. McComiskey et al. (2009), Yamaguchi et al. (2017)), and this work should be explicitly placed in that context. Specific focus on absorbing aerosol, such as the biomass burning smoke that impact the cloud deck that reaches Ascension Island has also been investigated (e.g. Ajoku et al. (2021), Diamond et al. (2018), Kacarab et al. (2020), Painemal et al. (2014)). These and related works should be cited to give context for the aerosol expected to drive changes in the Ascension Island microphysics and the potential environmental, compositional, and physical factors contributing to these changes. The authors should also describe what makes UV-polarization lidar advantageous over other commonly applied methods as well as its limitations.

The introduction has been completely rewritten, following the recommendations of the reviewer. The recommended works have all been added and cited to provide a proper context of the paper, for which the reviewer is thanked. The advantage of limitations of both the lidar and the radar methods used are described in more detail, a discussion section was added in which the results are compared with existing studies on retrieval of cloud parameters.

- The authors should consider restructuring the paper's outline of sections, namely the order of the theory, measurements, and methods, as these sections appear to be interspersed throughout the paper rather than contained within specifically focused sections. It would benefit understanding and context of the work if "Section 3: Measurement" campaign was placed before "Section 2: Theory" as some of the discussion in Section 2 references data described in Section 3 (Fig. 1).
  The paper has been restructured following the recommendation of both reviewers: The measurement campaign section now follows the introduction, and the theory section was moved to the appendix. The paper now clearly describes the aerosol-cloud interactions using a variety of methods, all with their merits and drawbacks. The technical description of the measurements was moved from the main story.

- "Section 5.3 Cloud Base Height validation" does not report on any aerosol-cloud interaction results and only gives a comparison between lidar-estimated cloud base height and two external estimates of cloud base. For this reason, it may be appropriate for this section to be moved to the supplement.
  The section was moved to the Appendix. We feel this is an important aspect of the assumptions used in the paper. In order to show the robustness of the method, the assumption of a proper cloud base height is essential. However, it was removed from the main story.

- More detail about the UV-polarization lidar used in this work should be provided. Specific details about the instrument itself, measurement frequency, uncertainties, and calibration should be included before introducing the theory equations in Section 2. Are the main results shown as daily averages? Were specific filtering techniques applied during averaging?
  The description of the lidar and measurements have been expanded. Calibration details and other details that are necessary for reproducing the results, but not necessary for interpretation of the results, are cited.
  The main results are differentiated into three main cases, clearly described in the manuscript. Some are daily averages, some are cases by case from selected cloudy intervals.
  The filtering techniques are described in the manuscript and the cited theses.

- As the authors have stated, it is customary to examine aerosol indirect effects by controlling for macrophysical (McComiskey et al., 2009; M. Miller et al., 2022) or meteorological (Scott et al., 2020) This was not done in

this work. The authors should speak more to how a lack of factor control on these measurements may impact the interpretation of the results.
Agreed. A section about the meteorological conditions during the campaign was added. A sidcussion of the impact of the meteorological conditions and its impact on the ACI results was added.

- Ascension Island lies at the terminating stage of the Southeast Atlantic stratocumulus-to-cumulus transition in the quiescent trade wind cumulus region. Zhang and Zuidema (2019) reported that the cloud types at Ascension are predominantly cumulus clouds with little vertical extent or cumulus clouds overlain by stratocumulus (two-layers), with single stratocumulus contributing less than 3% during the smoky season (August 2016 & 2017). The authors should describe how the specific cloud scenes were selected for the measurement comparisons and note, as in the title, that stratocumulus were the predominant cloud types observed and analyzed.
Agreed. The clouds that were selected were actually broken clouds over Ascension, so more likely cumulus clouds instead of stratocumulus. A paragraph on the measurement selection was added, showing a measurement sample with various cloud conditions and how the cloud selection was performed. The introduction has been changed to describe the paper in the context of the SCT and the title was changed.

- A broadened discussion comparing the retrieved microphysical parameters and computed aerosol indirect effects is necessary to provide more scientific basis to the report and interpretation of results. The authors should aim to answer specific questions about these results and their relation to measurements from other studies in relevant and related environments. How do the cloud droplet number and size inferred from this lidar technique compare to these parameters in other open ocean environments that are clean and impacted by smoke aerosol? The relative magnitude of the droplet number change appears to be much larger than that of the size change. Was this expected and consistent with previous work? If not, why? Additionally, the clean effective radius appears to be much smaller than the global average for warm clouds ($\tilde{1}4$ μm). Can the authors ascribe this low value to a property of the observed clouds or environment? How do the computed indirect effects compare to other regionally and globally estimated aerosol indirect effects? Are the magnitudes of these results consistent with other pristine environments perturbed by strong pollution signals?
The introduction, discussion and conclusion sections were rewritten to describe the measurements in the context of the SCT. The ACI_r found from the lidar measurements are at the high end of the ranges found in other papers, but consistent. The effective radius from the lidar is much smaller than the global average. This is mainly due to the sensitivity of the lidar to the cloud base. A comparison with radar retrievals at the cloud base

are consistent within the measurement uncertainty and comparative with previous lidar, in-situ and radar retrievals of the cloud droplet effective radius, if the strong dependence on height is taken into account. This is now discussed more extensively.
The indirect effects are at the high end of magnitudes found in other environments, but consistent with strong pollution events.

Minor comments:

The reader would benefit from having the aerosol indirect effect slopes summarized in the abstract.
Done

- Several cited papers in the main text are missing from the list of references, including: Bennartz (2007), Albrecht et al. (1998), Paluch et al. (1991). Done
- Line 39: Please provide a definition of "SNR" prior to using the acronym. Done
- Eq (3): what is r_{atm}? Is this supposed to be r_{air} as in Eq(4)? Please be consistent with these variable names. Yes, done
- Eq(4): Based on the units of r_{dryair} (J kg-1 K-1) and the fact that this equation is solving for the atmospheric density using ideal gas law, I believe this variable should be R_{dryair}, i.e. the universal gas constant for dry air, not the gas density of dry air. Correct, changed
- Line 87-89: What did the tests in which S_{marine} and S_{dark} were varied reveal about the sensitivity of the lidar ratio choices used in this work? A five percent change in AOT was found for changes in the lidar ratio within reasonable values. This was added to the manuscript.
- Figure 1: There is a discrepancy between the title label of this plot and the caption: the title shows 20170826, but the caption reads 27 Aug. 2017. Is there a reason for this discrepancy? It's 26 Aug. This was changed.
- Line 131: Please clarify the name of Γ_l. Is this an adiabatic lapse rate? Yes, this was added.
- Figure 3,4,5: Do these figures use data from both years or has 2017 data been excluded? Please clarify.

  Yes, only 2016 data were used to determine aerosol-cloud interactions. The section now opens with this statement.
- Line 167-169: Boundary layer and free tropospheric aerosol composition during the dry monsoonal season in the Southeast Atlantic has been characterized in previous work and should be cited (see (Dang et al., 2022; R.

Miller et al., 2021; Swap et al., 1996)).
The introduction and the measurement campaign section were rewritten and the references added.

- Line 193-194: How were the atmospheric layers (850 – 2150 m and 2150 – 5000 m) selected. Was the lidar backscatter or radiosonde profiles used to distinguish between cloud base – top and free troposphere?
The lidar range was used to determine the altitude. The backscatter lidar quickviews were investigated by eye to determine a rough estimate of the vertical layers. An example of the quickview and selection process was added to the manuscript.

- Line 193-196: The authors should use consistent terminology when referring to the above-cloud atmospheric layer as either the "free troposphere" (as in Line 194) or "upper air" (as in Line 196).
Done

- Line 205 – 207: The authors state: "It is assumed that aerosols between these levels have a significant impact on cloud forming." This statement is a bit vague and should provide evidence as to why it is believed that aerosol at these levels are most significant for cloud formation in this region.
This was rephrased to state that the cloud base is the lidar-sensitive region, and the aerosol are sampled in this region as well.

- Line 225-227: The statement about "other meteorological conditions" contributing to retrievals with large numbers and uncertainties is vague. Can the authors point to specific meteorological conditions relevant to Ascension Island and the Southeast Atlantic Ocean that would contribute to such results? I would expect that meteorological conditions are fairly persistent and unchanged at this tropical site. Have the authors fully exhausted their assessment of uncertainty in the retrievals that could potentially lead to large numbers or uncertainty not explained by the meteorology?
The meteorological conditions were checked by inspection of backtrajectories during the campaign and afterwards in the analyses, showing stable MBL conditions and variable upper air transport. In the manuscript a discussion is added citing new recent references describing the meteorological and climatological circumstances during the various measurement campaigns.

- Line 229-231: Shouldn't the months of discussion be September not August if referencing Figs. 6,7? The text states August in these passages.
Yes, this was corrected.

- Line 229-230:What is the meaning of a "saturated Twomey effect"?
  This statement was deleted. No change is observed if all aerosols are activated.

- Line 230-231: The authors state they observe "elevated AOT" in Sept. 12-15 leading to near zero indirect effect (cloud drop number). This is a bit difficult to glean from Fig. 7 given that near zero indirect effect (cloud drop number) is observed for Sept. 9-10, which also had low AOT. Is this AOT elevation relative to the month observed, and what is the magnitude of this "elevation" relative to the seasonal or annual average in AOT? Zuidema et al. (2018) report on the boundary smoke aerosol loading during these periods, which may help the authors attain insight into the aerosol impact on the observed indirect effects.
  A new paragraph was introduced to discuss the AOT and aerosol concentrations during the campaign in relationship with climatological means. The AOT values are high due to smoke incursions, as described by Ryoo et al. (2022), but not extremely high values compared to August 2016 values, as described by Zuidema et al. (2018). This is now described more clearly in the manuscript.

- Line 234: please clarify "various parameters and instrument noise".
  This line was removed

- Line 234-235: Although a reference is provided for the 2017 indirect results being inconclusive, please provide a brief summary of how these results lead to an "inconclusive effect." In the context of the computed indirect effects, what does inconclusive mean?
  In 2016 the lidar was just been serviced by Leosphere which made that the alignment was better than in 2017 and thus the SNR was higher in 2016 than in 2017. Therefore, retrieval error in 2016 was 19.75% and in 2017 39.05%, due to the calibration, retrieval and measurement errors and the 2017 results provide no statistical significant ACI due to the large uncertainties. This referenced statement has now been added to the manuscript.

- Line 250: Can the authors provide a statistical significance value for the AOT vs AERONET correlation coefficient of 0.76?
  Pearson's statistical correlation coefficient was 0.76, showing strong correlation. The figure was changed to include a scatterplot of the measurements, and a linear fit was drawn to show the relation.

- Line 284-285: What is a typical cloud droplet size estimate and range for marine low-level clouds? Are these typical values consistent with having large cloud drop concentrations as observed in this study?
  Typical numbers range from a few microns at the cloud base to several tens of microns at cloud top for well developed clouds. Numbers from

various references have now been added to the manuscript and the results are discussed in the light of previous studies.

- Line 285-290: How was the Reff100 derived using LWP measurements from the MWR? The authors note, MWR-retrieved Reff100 was much more wildly varying than the lidar and cloud radars followed by a reference to Fig. 8, however, a comparison of lidar, cloud radar, and MWR retrievals is not shown. Why have the authors not shown the MWR-retrieved Reff100?

  The R_{eff}^{100} was derived following a method described by Frisch et al. 2002. A MWR-derived R_{eff}^{100} is discussed in one of the theses which are the basis for this paper, but the results ($R_{\text{eff}}^{100} > 15$ μm) were strongly inconsistent with non-drizzling clouds. The reason for this is unclear, but may point to biases in the LWP data used or an error in the implementation. Therefore the results are not shown in the paper. This statement was added to the manuscript.

- Line 293: Are the authors referring to liquid water path or the cloud droplet number density when it is stated that "this parameter was more than 5 times higher than the assumed 100 g m-2"? I assume this is the cloud droplet number concentration and the units should be cm-3.
  Yes, it should be cloud droplet number density. This and the unit were changed.

- Line 296-297: Zhang et al. (2011) is later referenced as a citation for the statement that cloud radii are strongly dependent on height in the cloud (Line 303-304). Please consolidate these statements or provide the citation the first time the statement is mentioned.
  Done

- Line 299-300: Please provide a citation describing higher radar measurement sensitivity to drizzle than lidar measurements.
  Several references have been added.

- Line 307-308:Please provide the correlation and statistical significance of the CBH correlation in these lines of text.
  Done

- Line 318-321: These lines do not contribute to a summary of the results of the paper and instead provide theory of the measurements used in this work. It is recommended that this material be moved to the theory section (Section 2).
  Done

- Line 334: Based on the results previously described, the indirect effect for cloud droplet effective radius should be negative, i.e. -0.18 ±06 μm, not positive.
  Corrected

- Data availability statement: Can the authors please provide a source to locate the freely available lidar data?
  Yes, done.

- Figure 6: Please extend the ticks of the x-axis and labels in both panels so that the dates can be clearly read. The numbers following the 10th of September are difficult to distinguish.
  The figure was updated and improved.

- Figure 7: There is a discrepancy between terminology in the figure and caption. The y-label shows AOD, while the caption references Aerosol Optical Thickness and AOT. Please choose a consistent terminology.
  Done. All changed into AOT.

- Figure 10: Can the authors provide the elevation of the main ARM site and airport site in the caption?
  Yes, done.

- Figure 11: Is the dashed line in this figure the 1:1 line or the regression? Please clarify.
  It's the 1:1 line, added.

- Where the authors have discussed or shown time series between measurements (lidar vs. radar, AERONET vs lidar, MWR vs lidar / radar), comparison plots (e.g. Figure 11) should also be provided with acknowledgement of the slope or bias in these comparisons.
  Done, all those figures have been revisited and now contain scatter plots with linear fits, showing slopes and biases.

Technical Corrections

All of the following technical correction were implemented as suggested:

- Line 48: Please correct "devided" to "divided".
- Line 202: Please correct "garantueed" to "guaranteed".
- Line 206: Please correct "forming" to "formation".
- Figure 5 caption: Please correct "daioly" to "daily".

References

1. Ajoku, O., Miller, A., & Norris, J. (2021). Impacts of aerosols produced by biomass burning on the stratocumulus-to-cumulus transition in the equatorial Atlantic. Atmospheric Science Letters, 22(4). Article.

2. Dang, C., Segal-Rozenhaimer, M., Che, H., Zhang, L., Formenti, P., Taylor, J., et al. (2022). Biomass burning and marine aerosol processing over the southeast Atlantic Ocean: a TEM single-particle analysis. Atmospheric Chemistry and Physics, 22(14), 9389-9412. Article.

3. Diamond, M., Dobracki, A., Freitag, S., Griswold, J., Heikkila, A., Howell, S., et al. (2018). Time-dependent entrainment of smoke presents an observational challenge for assessing aerosol-cloud interactions over the southeast Atlantic Ocean. Atmospheric Chemistry and Physics, 18(19), 14623-14636. Article.

4. Kacarab, M., Thornhill, K., Dobracki, A., Howell, S., O'Brien, J., Freitag, S., et al. (2020). Biomass burning aerosol as a modulator of the droplet number in the southeast Atlantic region. Atmospheric Chemistry and Physics, 20(5), 3029-3040. Article.

5. McComiskey, A., Feingold, G., Frisch, A., Turner, D., Miller, M., Chiu, J., et al. (2009). An assessment of aerosol-cloud interactions in marine stratus clouds based on surface remote sensing. Journal of Geophysical Research-Atmospheres, 114. Article.

6. Miller, M., Mages, Z., Zheng, Q., Trabachino, L., Russell, L., Shilling, J., & Zawadowicz, M. (2022). Observed Relationships Between Cloud Droplet Effective Radius and Biogenic Gas Concentrations in Summertime Marine Stratocumulus Over the Eastern North Atlantic. Earth and Space Science, 9(2). Article.

7. Miller, R., McFarquhar, G., Rauber, R., O'Brien, J., Gupta, S., Segal-Rozenhaimer, M., et al. (2021). Observations of supermicron-sized aerosols originating from biomass burning in southern Central Africa. Atmospheric Chemistry and Physics, 21(19), 14815-14831. Article.

8. Painemal, D., Kato, S., & Minnis, P. (2014). Boundary layer regulation in the southeast Atlantic cloud microphysics during the biomass burning season as seen by the A-train satellite constellation. Journal of Geophysical Research-Atmospheres, 119(19), 11288-11302. Article.

9. Scott, R., Myers, T., Norris, J., Zelinka, M., Klein, S., Sun, M., & Doelling, D. (2020). Observed Sensitivity of Low-Cloud Radiative Effects to Meteorological Perturbations over the Global Oceans. Journal of Climate, 33(18), 7717-7734. Article.

10. Swap, R., Garstang, M., Macko, S., Tyson, P., Maenhaut, W., Artaxo, P., et al. (1996). The long-range transport of southern African aerosols the tropical South Atlantic. Journal of Geophysical Research-Atmospheres, 101(D19), 23777-23791. Article.

11. Yamaguchi, T., Feingold, G., & Kazil, J. (2017). Stratocumulus to Cumulus Transition by Drizzle. Journal of Advances in Modeling Earth Systems, 9(6), 2333-2349. Article.

12. Zhang, J., & Zuidema, P. (2019). The diurnal cycle of the smoky marine boundary layer observed during August in the remote southeast Atlantic. Atmospheric Chemistry and Physics, 19(23), 14493-14516. Article.

13. Zuidema, P., Sedlacek, A., Flynn, C., Springston, S., Delgadillo, R., Zhang, J., et al. (2018). The Ascension Island Boundary Layer in the Remote Southeast Atlantic is Often Smoky. Geophysical Research Letters, 45(9), 4456-4465. Article. Citation: https://doi.org/10.5194/acp-2022-473-RC2