Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
https://doi.org/10.5194/acp-2022-473-RC2, 2022

Comment on acp-2022-473

Anonymous Referee #2

Referee comment on "Aerosol first indirect effect of African smoke at the cloud base of marine cumulus clouds over Ascension Island, southern Atlantic Ocean" by Martin de Graaf et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-473-RC2, 2022

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.

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 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).

- "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.

- 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?

- 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.

- 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.

- 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 (~ 14 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?

Minor comments:

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

- 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).

- Line 39: Please provide a definition of "SNR" prior to using the acronym.

- Eq (3): what is r_{atm}? Is this supposed to be r_{air} as in Eq(4)? Please be consistent with these variable names.

- Eq(4): Based on the units of $r_{dry\ air}$ (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 $R_{dry\ air}$, i.e. the universal gas constant for dry air, not the gas density of dry air.

- 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?

- 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?

- Line 131: Please clarify the name of G_l. Is this an adiabatic lapse rate?

- Figure 3,4,5: Do these figures use data from both years or has 2017 data been excluded? Please clarify.

- 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)).

- 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?

- 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).

- 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.

- 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?

- 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.

- Line 229-230: What is the meaning of a "saturated Twomey effect"?

- 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.

- Line 234: please clarify "various parameters and instrument noise".

- 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?

- Line 250: Can the authors provide a statistical significance value for the AOT vs AERONET correlation coefficient of 0.76?

- 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?

- 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?

- Line 293: Are the authors referring to liquid water path or the cloud droplet number concentration 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}.

- 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.

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

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

- 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).

- 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.

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

- 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 10^{th} of September are difficult to distinguish.

- 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.

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

- Figure 11: Is the dashed line in this figure the 1:1 line or the regression? Please clarify.

- 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.

Technical Corrections:

- 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

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