Review of "Natural Marine Cloud Brightening in the Southern Ocean" by Mace et al. (acp-2022-571)

The presented study analyzes strong gradients in the cloud droplet concentration found in the Southern Ocean, using five years of satellite observations. The authors show that these stark differences in the cloud microphysical composition can be traced back to biological primary production at the Antarctic Shelf, from where airmasses with high cloud droplet concentrations are moved to the north, while low cloud droplet concentrations originate from the open sea equatorward.

Despite many technical issues and a few minor comments, I enjoyed reading this manuscript. In a concise way, the article informs about the aerosol and cloud microphysics of an important region of the Earth. Thus, I support publication in *Atmospheric Chemistry and Physics* once my concerns are addressed. However, this article is also submitted to be published as an *Atmospheric Chemistry and Physics Letter*. In the current form, I cannot support the publication in this format, as I will outline in my only major comment below, but I am willing to be convinced otherwise.

Major Comment

Does the article meet the requirements for an Atmospheric Chemistry and Physics Letter? It is stated that an *Atmospheric Chemistry and Physics Letter* must fulfill the following requirements (see www.atmospheric-chemistry-and-physics.net/about/manuscript_types/acp_letters.html):

- Important discoveries and research highlights in atmospheric chemistry and physics.
- Solutions to or progress with long-standing and important questions in atmospheric research.

In its current form, the manuscript does not give substantial hints on how these requirements are fulfilled. I understand that the Southern Ocean is one of the least understood parts of the climate system, with inherent problems in modeling it. But how does the presented study contribute to improving its understanding? What are the important discoveries made? While I admit that the presented research is very interesting, the authors should use the opportunity to frame their work and highlight the advancements made through their work.

Response: The manuscript builds upon prior research in several important ways that I think elevate the findings to the level of "important discoveries and research highlights". Prior work (D. McCoy et al., 2015) illustrate the correlations among various MODIS-derived cloud parameters and the processes associated with biogenic aerosol production while I. McCoy et al., (2020) documents the latitudinal variability in Nd in the Southern Ocean and claims that this ocean basin is the last vestige of the preindustrial Earth. In the present manuscript we find that the gradient in Nd associated with the production of biogenic aerosol in the high latitude Southern Ocean results in clouds that are significantly more reflective than their lower latitude counterparts. The higher albedo of these clouds with lower overall liquid water is a significant finding (discovery, if you will) that implicates biological processes in modulating the surface radiative balance of the high latitude Southern Ocean. While cloud property and aerosol sensitivities to biology have been documented in this region, a direct connection to radiative effects has not been documented until now. The higher albedos for lower liquid water path along the Antarctic Shelf now firmly establish that the surface solar radiation along the Antarctic shelf within the highly productive zone is modulated by the biology. This represents a forcing. The CLAW hypothesis (which we do not mention) describes a feedback – that the biology will change to keep the environment conducive to itself. The forcing that we identify is a necessary but not sufficient condition to establish the CLAW feedback.

Minor Comments

Ll. 47 – 50: What cloud process is sensible to the CCN concentration? In non-precipitating clouds, an increase in CCN will not change the precipitation efficiency. I guess the authors refer to the cloud albedo.

Response: Yes, something like the albedo susceptibility is what I refer to although we do not

quantify that derivative specifically in this study. While I think such a quantitative analysis would be interesting, it would put the present paper out of scope of a letter.

Ll. 56 – 58: Define what "primary production" is. This will also help to frame the importance of the work (major comment). Do ll. 79 – 84 also refer to primary production?

Response: Primary productivity refers to the net organic matter, mostly produced by phytoplankton, that is suspended in the ocean. The definition has been added to the text.

Ll. 105 – 108: What is the purpose of this sentence? N_d and r_e are in opposite phase, not out of phase. So there is a very tight relationship.

Response: I was simply pointing out how Nd varies with effective radius due to equation A1. Wording changed.

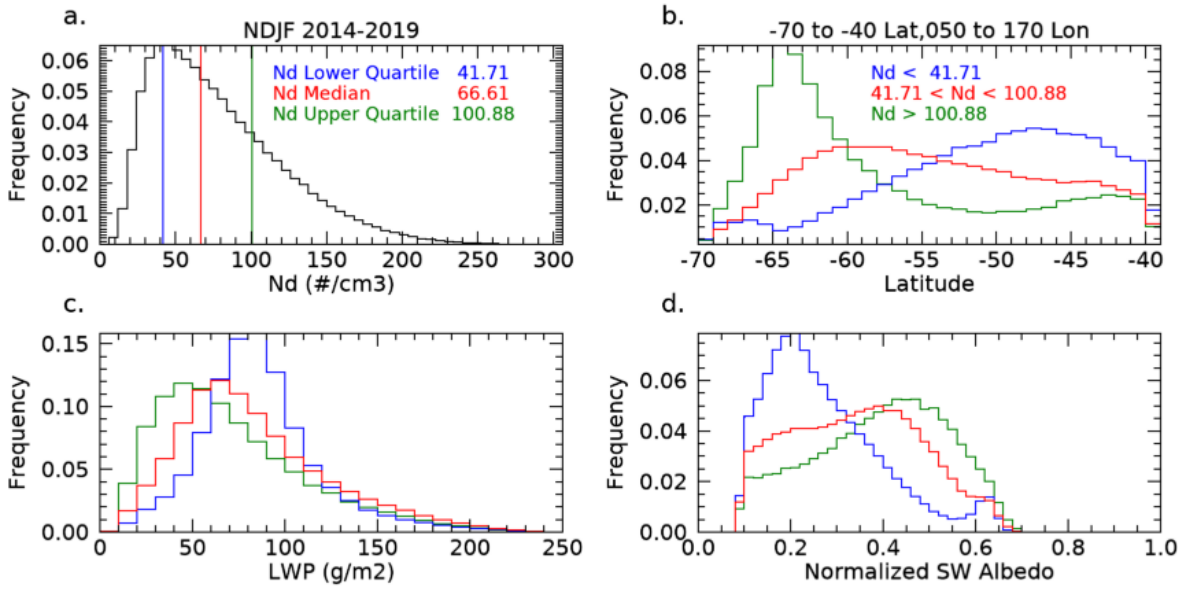
L. 108: Replace "cycle" with "variability"?

Response: Done.

Fig. 2b: Add labels. The blue line represents locations with $N_d > 101$ cm^{-3} and the green line $N_d < 42$ cm^{-3}. What about adding a red line for 42 cm^{-3} < N_d < 101 cm^{-3}?

Response: We have clarified this in the caption. The colors of the histograms in the b-d are as defined in the inset of panel a.

Here is a plot with the middle tercile. I don't think it adds much to the discussion to include this since the histograms sit in between the upper and lower as one might expect.



Ll. 113 – 117: The LWP changes non-monotonically with N_d. It increases with N_d for precipitation stratocumulus, but decreases for non-precipitating stratocumulus due to increasing entrainment rates (Glassmeier et al. 2021).

Response: Thank-you for pointing this out. I have mentioned this in the revision and cite the Glassmeier paper.

Ll. 131 – 134: Why is N_d only large at the ACFA and not below it?

Response: I think the peak near 65S is just an artifact of the analysis. Figure 3 shows that the high Nd quartiles occurrence peaks immediately adjacent to Antarctica. However, when compiling the frequency distribution in Figure 2, the higher Nd occurrence at maximum latitude is minimized due to the decreased ocean surface area along the continental margin. I've removed this potentially confusing statement in the revision.

Ll. 135 – 139: With a mean r_e of 13 µm north of the ACFA, droplet coalescence might decrease N_d. See, e.g., Freud and Rosenfeld (2012), who showed that at 14 µm surface precipitation occurs, i.e., there is probably some drizzle at a slightly smaller r_e. This should be discussed.

Response: I address this in the response to the comment below.

Ll. 275 – 278: The significantly lower adiabaticity north of the ACFA could be due to precipitation, triggering the transition of closed- to open-cell stratocumulus. Discuss the possibility of precipitation.

Response: I address the previous two comments together. The Freud and Rosenfeld paper analyzes in situ data collected in cumulus clouds. We are analyzing mostly stratocumulus clouds where the threshold relationships found in Freud and Rosenfeld might be different although it is likely that such a threshold exists. We do note in the revision the increased likelihood of precipitation in the lower latitude clouds given the larger droplet sizes and potential that precipitation is responsible for the lower adiabaticity of the lower latitude clouds.

Ll. 297 – 300: Clouds with r_e > 14 µm are usually precipitating (Freud and Rosenfeld 2012). Declaring all pixels with r_e < 50 µm to non-precipitating clouds will cause a substantial bias. Please elaborate.

Response: Our objective is to have cloud scenes that are mostly non precipitating (note we have changed the characterization to weakly precipitating in the methods section). A requirement for weakly precipitating clouds is because of the potential biases in cloud effective radius that occurs when pixels have significant precipitation water coexisting with a cloud droplet mode (Xu et al., 2022). We have found that the water path filter is the most important and that the effective radius criteria is irrelevant. To avoid confusion, I've removed reference to an upper effective radius bound.

Z. Xu, G. G. Mace and D. J. Posselt, "Impact of Rain on Retrieved Warm Cloud Properties Using Visible and Near-Infrared Reflectances Using Markov Chain Monte Carlo Techniques," in IEEE Transactions on Geoscience and Remote Sensing*, vol. 60, pp. 1-10, 2022, Art no. 4110110, doi: 10.1109/TGRS.2022.3208007.*

Technical Comments

Response: All Technical comments addressed as suggested

L. 32: "Both" usually refers to two objects. Here, it refers to three (latitudinal, longitudinal, and temporal). Revise.

L. 45: SO for Southern Ocean is already defined. Use it.

L. 55: To what is "respectively" referring to?

L. 66: Why is "TOF" defined? It is never used.

L. 88: "ACF" is not defined. Only "ACFA".

Ll. 91, 109, 182: "N_d", not "Nd".

L. 96: See Shaw et al. (1988) for what?

Fig. 1, l. 122: Define "Chl-a".

Fig. 1: What are "MOD" and "MYD"?

Ll. 128 – 129: Switch "upper" an "lower"?

L. 189: Define G18.

L. 213: Use "PP" instead of "primary production".

L. 269: "A1", not 1.

References

Freud, E., & Rosenfeld, D. (2012). Linear relation between convective cloud drop number concentration and depth for rain initiation. *Journal of Geophysical Research: Atmospheres*, *117*(D2).

Glassmeier, F., Hoffmann, F., Johnson, J. S., Yamaguchi, T., Carslaw, K. S., & Feingold, G. (2021). Aerosol-cloud-climate cooling overestimated by ship-track data. *Science*, *371*(6528), 485-489.