This short letter describes an analysis that combines MODIS satellite estimates of cloud droplet concentration in liquid-dominated marine low clouds with trajectory analysis over the Southern Ocean. The findings indicate that high concentrations of cloud droplets (Nd) tend to occur to the south (poleward) of a boundary previously identified as a "compositional front" that rings Antarctica. South of the "atmosphere compositional front of Antarctica (ACFA)" at roughly 60S comprises extremely biologically rich ocean waters that are copious sources of aerosol precursor gases (in particular dimethyl sulfide). Air mass back trajectories from high Nd clouds tend originate more frequently south of the ACFA. The high Nd south of 60S are associated with smaller effective radii and higher cloud optical thickness, but only marginally higher LWP, indicating that the cloud optical depth increase is largely driven by higher Nd, i.e., Twomey brightening.

The results presented here are interesting and important and I think very relevant to the ACP readership. I recommend publication subject to some minor revisions.

The main question I would like to raise is that I believe that the latitudinal gradient of light precipitation may also play an important role in setting the Nd latitudinal gradient through coalescence scavenging, in addition to the consideration of aerosol sources. We know from spaceborne 94 GHz radar that light precipitation maximizes at around 55S and decreases southward of this (see e.g., McCoy et al., 2020), so the reducing precipitation south of the ACFA may also be partly responsible for high Nd there. Another paper by Kang et al. (2022) illustrates the significant role that precipitation sinks may play. I wonder if the authors have tried to use any of the ship or aircraft measurements associated with CAPRICORN/MARCUS/SOCRATES to explore how precipitation sinks may change across the ACFA.

Response: There is no question that precipitation plays a key role in controlling the concentration of liquid cloud droplets in shallow boundary layer clouds. We noted the role of drizzle in the original manuscript near line 220. In the revised manuscript, we also add the important finding from Kang et al. and, we note also in the conclusions that the latitudinal gradient in Nd has many influences beyond a simple aerosol explanation. Both Kang et al. (2022) and McCoy et al. (2020) are referenced in the revised manuscript.

Other points

1. Line 35. Albedo increases with solar zenith angle, so how is this accounted for? Also, I didn't see any albedo measurements in the paper.

Response: We have added CERES-derived albedo measurements in the revised manuscript. To account for the variation of cloud albedo with solar zenith angle, we developed and implemented an empirical correction for latitude using calculations presented in Minnis et al., 1998. The method for normalizing the CERES albedos for latitude are described in the methods section (Appendix) of the revised manuscript. The results of the albedo analysis are supportive of the original conclusions and make the paper much more compelling.

2. Line 47-50: Why does a lack of precipitation make clouds more sensitive to CCN? Shiptracks in precipitating boundary layers tend to more visually apparent than those forming in non-precipitating clouds.

Response: The analysis of Kang et al., 2022 that uses the simple empirical model of Wood et al. (2012) is based on the sources and sinks of Nd in liquid clouds. We reason that when precipitation is weak or absent, one of the sinks of Nd is also absent and, therefore, the sensitivity of Nd to CCN (a source of Nd) would be enhanced.

3. Figure 1b does not seem important. Can't the essence of this simply be stated in the text?

Response: Agree. Figure 1b has been removed and we have adapted the text to describe the main points.

4. Line 95 and several studies point out the importance of air masses moving from interior Antarctica over the ocean as being the source of new particles. I do not understand why the Antarctic continent would be a good source of aerosol or aerosol precursor gases. It seems as though the highly productive ocean waters south of the ACFA are the main sources of aerosol. Can the authors comment on this?

Response: I believe this statement to be accurate. The reason that trajectories that pass over Antarctica seem to have a higher CCN concentration is not yet definitively established in the observational literature. However, we know from prior studies that one of the pathways for nucleation of new aerosol and eventual growth to CCN from precursor gasses requires ultraviolet sunlight and that this process often happens in the free troposphere above low-level clouds. Air masses that pass over Antarctica with sufficient precursor gasses certainly get a higher dose of UV because of the high albedo of the ice-covered continent. The glaciated surface of Antarctica is also elevated and has low overall cloud cover. We speculate that trajectories passing over the elevated, high-albedo surface (especially in summer with long days) would encounter conditions conducive to new particle formation. We have attempted to suggest this process in the revised manuscript although we would like to be somewhat conservative in promoting the idea because of the lack of definitive observational evidence.

5. Figure 1a: why not provide the correlation coefficients between cloud variables to make the points quantitatively?

Response: McCoy et al. (2015) do an extensive regression analysis of the relationship between MODIS and factors responsible for Chl-a variations. Our results are consistent with their results. The correlation coefficients of the various quantities in Figure 1 are now noted in the revised manuscript.

6. Line 116: LWP can remove aerosol, suppressing Nd (Wood et al., 2012). Nd can suppress precipitation, but the LWP response to this is bidirectional, and depends upon whether the background clouds were precipitating and up the dryness of the free troposphere. I don't think you can necessarily conclude that the seasonal cycle of LWP is dominated by meteorology (i.e. is NOT driven by aerosol, at least in part).

Response: I don't think that LWP, per se, is the cause of N_d change. Coalescence scavenging would increase as LWP (and likely r_e) increases. As N_d increases, r_e would decrease and LWP increase because of drizzle suppression. I assume that the entrainment drying of the MBL would tend to increase in summer as the free troposphere dries due to warming and a lower frequency of deep storms thereby decreasing LWP. All these factors are, of course, inter-related and saying that N_d and r_e are independent of LWP is hard to justify without a more thorough analysis that would be beyond the scope of this letter. So, I have softened the language here a bit and removed the statement that LWP is independent of N_d.

7. Line 122: Provide evidence of the one month lag between Chl-a and Nd. Is this at all locations across the SO?

Response: I say that this is the case in 4 of the 5 years. It can be seen by inspection of Figure 2 that Chl-a is rising about a month ahead of Nd (and decrease in re). Figure 2 is the result of averaging the MODIS and Chl-a retrievals over the entire analysis domain in each month. This result becomes much noisier when examined on finer spatial scales although, as shown by McCoy et al., (2015) for lower latitudes, there is a statistically significant relationship in broad regions of the Southern Ocean. A 1-month lag correlation increases the correlation between Nd and Chl-a from 0.27 to 0.60. However, because of the break in the time series (recall that we are examining November through February of each year) interpreting this quantitatively should be avoided. I think what the lag correlation captures is just what can be seen visually in the time series where Chl-a tends to rise about a month ahead of Nd in 4 of the 5 years.

8. Fig 2/Line 134: This Nd gradient is documented and discussed in McCoy et al. (2020).

Response. True. Appropriately noted in the revised manuscript in the paragraph starting around line 200.

9. Line 156: Cite Korhonen et al. (2008), who established the pathway through the free troposphere. I would have expected the need for transport to the FT and nucleation of new particles to effectively reduce the sharpness of the Nd gradient driven by the gradient in surface-emitted precursor gases. Sources will lose their identity through the mid-deep tropospheric mixing and latitudinal displacement related to cyclonic systems. I would appreciate if the authors can comment on this issue. Line 179 seems to partly challenge the Korhonen transport pathway being primarily through the free troposphere.

Response. I did not mean to challenge the idea that transport of aerosol through the free troposphere is unimportant. I do address the importance of new particle formation in the free troposphere and transport there in the paragraph around line 220. While we do not address it here in detail because we only examine the summer, the entire Southern Ocean undergoes a large seasonal oscillation in CCN (Gras and Keywood, 2017) and Nd (McCoy et al., 2015; Mace and Avey, 2017) that has been documented although not fully explained. It is my opinion (and only something of an hypothesis at this point) that CCN formed by new particle formation in the deep southern latitudes seeds much of the rest of the SO through northward transport through the free troposphere. We hint at this in the last paragraph of the introduction and expand further upon it in the revised manuscript and in the conclusions. Glen Shaw hints at this process in his early 1988 paper and again in 2007. We also cite Korhonen et al., 2008 in the last paragraph of the introduction and again in the paragraph around line 220.

10. Line 169-171: Are these 3D trajectories, or 2D? What method was used to determine the vertical ascent (model vertical velocity, isentropic….)?

Response: The HYSPLIT trajectory model is described in Stein et al. (2015). The trajectory model uses the 3d model grids. The vertical motions and horizontal winds are as predicted in the GDAS model.

11. 4: The differences between the latitudes crossed by high Nd and low Nd trajectories shown here are quite modest yet are described as "overwhelming" (line 179). Does this statement pertain to clouds only south of the ACFA? It certainly does not pertain to high Nd cloud north of 60S since the majority of trajectories ending north of 60S never go below 60S.

Response: No argument. I have removed the "overwhelming" adjective.

12. Line 245: No shortwave measurements are presented in the paper, so I'm not sure that the term "brightening" is appropriate unless said measurements are presented.

Response: We have added the CERES albedo in the revision to support the brightening claim.

All papers cited in our response are listed in the revision with the following exception.

Wood, R., Leon, D., Lebsock, M., Snider, J., & Clarke, A. D. (2012). Precipitation driving of droplet concentration variability in marine lowclouds. Journal of Geophysical Research, 117(D19). https://doi.org/10.1029/2012jd018305

References

Kang, L., Marchand, R. T., Wood, R., & McCoy, I. L. (2022). Coalescence scavenging drives droplet number concentration in Southern Ocean low clouds. Geophysical Research Letters, 49, e2022GL097819.

Korhonen, H., Carslaw, K. S., Spracklen, D. V., Mann, G. W., & Woodhouse, M. T. (2008). Influence of oceanic dimethyl sulfide emissions on cloud condensation nuclei concentrations and seasonality over the remote Southern Hemisphere oceans: A global model study. Journal of Geophysical Research, 113(D15). https://doi.org/10.1029/2007JD009718

McCoy, I. L., McCoy, D. T., Wood, R., Regayre, L., Watson-Parris, D., Grosvenor, D. P., Mulcahy, J. P., Hu, Y., Bender, F. A.-M., Field, P. R., Carslaw, K. S., & Gordon, H. (2020). The hemispheric contrast in cloud microphysical properties constrains aerosol forcing. Proceedings of the National Academy of Sciences, 117(32), 18998–19006. https://doi.org/10.1073/pnas.1922502117