Anonymous Referee #1

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The manuscript analyzes measurements of particle number concentrations (CN) and cloud condensation nuclei (CCN) obtained by airborne measurements during SOCRATES and by ship-borne observations during CAPRICORN-2 in the Australian sector of the Southern Ocean. The study comprehensively shows the effects of cloud processing, precipitation and air mass origin on particle size and number, and on CCN. To this end the authors combine direct observations and re-analyses data. They also show nicely that in most cases new particles are formed in the free troposphere and not in the marine boundary layer. These measurements make an important contribution to our understanding of CN and CCN processes in the Southern Ocean. I recommend that the paper be published, however, only after considering below points, which I believe will help improve the study. Also, if possible, I recommend that this manuscript be published as a normal research article rather than a measurement report. The depth of analysis is not untypical of that in research articles.

We thank the reviewer for the detailed and thoughtful comments, which have improved the overall quality of the manuscript. For convenience, specific comments that the reviewer included in the supplement document have all been copied and responded to at the end of this review. The article was initially submitted with the intent of a normal research article; however, at the editor's request, we resubmitted the article as a measurement report. Based on the reviews, the decision of how to publish will be left for the editor.

General comments:

A map with the cruise and flights tracks is needed.

We have added the following figure with the cruise and flight tracks.

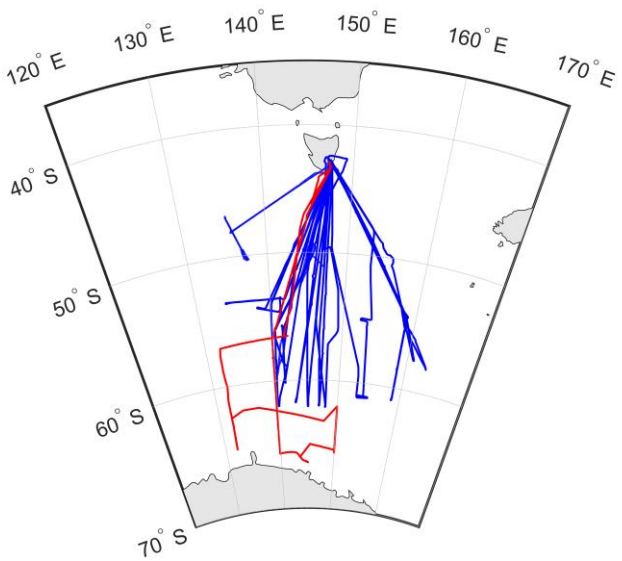


Figure 1. SOCRATES and CAPRICORN-2 study region. Blue and red lines represent the SOCRATES flight tracks and CAPRICORN-2 *R/V Investigator* tracks, respectively.

The discussion of NPF in the Southern Ocean (SO) boundary layer is partly incorrect, because it is said that the condensation sink (CS) is low. See specific comments in the attachment and also further below.
We have addressed all of the comments on condensation sink in detail in the responses below.

Some statements about the Southern Ocean are too general, e.g., the claim that microbial activity is low compared to the Antarctic coastal region. There are hotspots, like South Georgia, and if measurements had been taken in that region of the Southern Ocean the paper would report different observations. Hence acknowledging the regional variability of the SO is very important. Otherwise incorrect messages about this large region are published. This comment is also true for the conclusions. See comments in the attached.

We acknowledge that there are hotspots and have addressed all comments about the region of the Southern Ocean SOCRATES and CAPRICORN-2 measurements below.

The introduction jumps between topics, particularly ll. 92 – 110. The main message is not clear. I suggest to structure this part of the introduction as follows: observations of NPF, CN and CCN near the Antarctic coast, observations of NPF, CN and CCN over the open southern ocean (and not only between Australia and Antarctica), then discuss how the coastal and open ocean regions are connected, then go deeper into cloud processing.
This introduction has been reorganized based on suggestions by multiple reviewers:

> "The remote mid-latitude SO contains much less biological activity near the ocean surface relative to the Antarctic continental coast, which creates a latitudinal gradient in the contribution of particles from biogenic sources, with the exception of some biological hotspots such as near South Georgia (Alroe et al., 2020; Humphries et al., 2016; Kim et al., 2019; O'Dowd et al., 1997; O'Shea et al., 2017; Schmale et al., 2019; Weller et al., 2018). Shipborne observations in the region south of Australia show a distinct increase in aerosol concentrations south of 64°S, where CN concentrations are about five times higher during the austral spring months (Humphries et al., 2016). The seasonal variability of biogenically derived particles is linked to seasonal variations in SO biological activity (Ayers and Gras, 1991; Korhonen et al., 2008). On the Antarctic peninsula, NPF events occurred mostly during the austral summer, with CCN concentrations (at 0.4% supersaturation) increasing on average by 11% (Kim et al., 2019). Similarly, higher average concentrations of cloud droplet number concentrations (CDNC) are observed in the austral summer (Mace and Avey, 2017; McCoy et al., 2015). Some studies suggest biologically productive waters enhance PMA production (Fuentes et al., 2010), while other studies find that biogenic content has little to no influence on PMA production (Bates et al., 2020; Collins et al., 2016). In any case, PMA CCN is found to have little seasonal variability relative to biogenic CCN (Vallina et al., 2006), likely driven by small seasonal differences in wind speed (Saliba et al., 2019). Organic enrichment of PMA in biologically productive waters may further reduce their hygroscopicity (Burrows et al., 2018; Cravigan et al., 2019; Law et al., 2017; Meskhidze and Nenes, 2010).
>
> Long-range transport of aerosol and gaseous precursors in the MBL and free troposphere from the Antarctic continental coast plays a significant role in increasing CN, CCN, and CDNC concentrations in the SO (Bates et al., 1998a; Clarke et al., 1998, 2013; Dzepina et al., 2015; Korhonen et al., 2008; Woodhouse et al., 2010). With substantial growth of newly formed particles by the uptake of VOC oxidation products through cloud processing, particles from

biogenic sources may grow CCN larger and subsequently increase CDNC (Hoppel et al., 1986; Hudson et al., 2015; Pirjola et al., 2004; Russell et al., 2007; Sanchez et al., 2018). Cloud processing occurs when small particles activate to form cloud droplets, leading to enhanced condensation of VOC oxidation products onto the droplet because the droplet surface area is larger than that of the unactivated particles. Aqueous phase oxidation of absorbed VOCs also results in the formation of less volatile compounds, which remain in the particle phase upon evaporation of the water (Hoppel et al., 1986). In the event that the cloud droplets do not precipitate, the evaporated particles are larger than their original size since aqueous oxidation of volatile compounds (i.e., DMS, MSA, SO2 and nitric acid) have formed non-volatile sulfates and nitrates that remain in the particle phase. This added mass eventually shifts Aitken mode particles to the accumulation mode (Hoppel et al., 1986; Hudson et al., 2015; Kaufman and Tanré, 1994; Sanchez et al., 2017; Schmale et al., 2019). Results from McCoy et al. (2015) show that, despite the ambiguous results from focused modelling and observational studies of such aerosol processes, their general global model simulations of natural aerosol account for more than half the spatial and temporal variability in the satellite-derived CDNC over the SO. These areas of enhanced CDNC also correlate with areas of high chlorophyll-a, a tracer for phytoplankton activity, which increases secondary sulfate and organic aerosol concentrations (Krüger and Grabßl, 2011; McCoy et al., 2015). SO satellite-derived cloud properties such as liquid water content (LWC), effective radius, and cloud fraction showed seasonal variations that resulted in a difference in cloud radiative forcing (i.e., surface cooling) between 14 and 23 W m-2 (Mccoy et al., 2014). Increased CDNC is also shown to correlate with enhanced cloud fraction, significantly increasing overall cloud shortwave forcing (Rosenfeld et al., 2019). If cloud droplets precipitate, CN and CCN concentrations are reduced through precipitation scavenging (Croft et al., 2010; Stevens and Feingold, 2009)."

Some more details in the methodology section are needed, in particular regarding the mini CCNCs and the calculation of kappa. Also the inlet system and position of the CCNC on R/V Investigator is not described. In addition, it is unclear why the CCN data were not compared at the same supersaturation. It is possible to interpolate from the spectrum. See attachment for more specific comments.

We have added the following text to describe the calculation of kappa:

"The CCN spectra and UHSAS number concentrations on the GV were used to estimate the hygroscopicity parameter at 0.07 µm diameter (κ_{70}) for each MBL leg. For this calculation, the critical supersaturation is derived from the CCN spectra, where the UHSAS concentration at 0.07 µm diameter is equivalent to the CCN concentration."

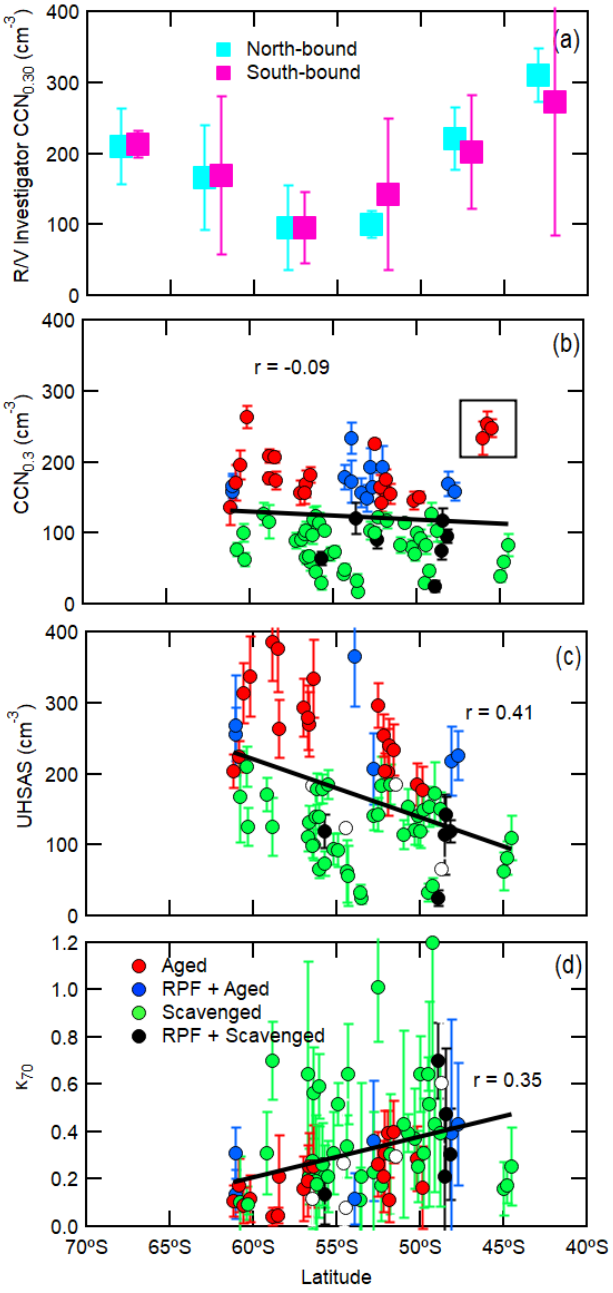
We have also added the following text to clearly identify details of the mini CCNc can be found:

"The miniature CCN counters are custom-made and operate with the same physical principles described by Roberts and Nenes (2005)."

The following text was added to address inlet system on the R/V Investigator.

"Details of the aerosol sampling system on board the RV Investigator are presented in Humphries et al. (2019) and Alroe et al., (2020). In short, aerosol sampling occurred via a common sampling inlet mounted on a mast at the bow of the ship, located 18 m above sea level. The CCN counter sampled from a manifold located 8 m below the mast in the ship's bow."

We have updated Figure 5a (now figure 6a) to show the R/V Investigator CCN concentration at 0.30% SS by interpolating the concentrations measured at 0.25% and 0.35% (shown below).



The calculation of back trajectories is not well described. If it is really the case that only one location per leg was used, the results will be highly uncertain. Some more clarification is needed, particularly a better description of flight legs.

We have updated the text as follows:

> "The latitude, longitude and altitude (50-500 m) averaged for each CCN spectra (~150 seconds, ~15-20 km horizontal distance) collected during the MBL legs on the GV HIAPER were used as starting points for the back trajectories."

Specific comments:

l. 193: A quantification or at least better approximation of the RH in the sample flow is needed to make this study comparable to previous and future studies.

Strapp et al. 1992 showed that the PCASP-100X dries aerosol almost completely (<40% RH) with the deicing heaters. The UHSAS and PCASP are made by the same company (DMT) and have identical deicing front ends. On a GV, there is additional heating related to ram air, consequently, we consider that the UHSAS air as completely dried (also <40% RH).

We added the following text:

> "This deliquesced mode was present despite the findings by Strapp et al. (1992), suggesting the de-icing heaters of the PCASP-100X (which are identical to those used for the UHSAS) is expected to dry the particles to less than 40% relative humidity. We hypothesize that the low residence time of the aerosol in the instrument (~0.2 seconds) prevented the large hygroscopic sea salt from fully drying before being measured."

l. 202: the description of the fraction of PMA to particles > 0.2 _m is inconsistent (see attached comments).

This statement has been simplified to improve clarity:

> "The TEM analysis showed that ~70-95% of marine boundary layer particles > 0.5 um optical diameter are PMA sea spray (Twohy et al., submitted)."

l. 229: An explanation of how the Aitken mode was derived is missing. The UHSAS was only used for particles greater 70 nm, so cannot have been used for that purpose. Was the CPC data used? If yes, how were CPC and UHSAS intercompared?

The Aitken mode was not directly derived, and the following text has been modified to prevent confusion:

> "The classification of each regime is based on the relative concentration of Aitken + accumulation-mode particles (CN) and accumulation-mode particles (CCN sizes), with a naming convention that describes the corresponding airmass history. Similar to analyses in previous studies, the relative contribution of the accumulation-mode to the total particle concentration is used to identify recent particle formation (RPF) events and growth of small (<70 nm diameter) particles to accumulation-mode or CCN sizes (Kalivitis et al., 2015; Kleinman et al., 2012; Williamson et al., 2019). The Scavenged regime is named based on evidence indicating the removal of CCN-sized particles through precipitation scavenging (Section 3.3). The Aged regime represents cases in which accumulation-mode is prominent and CCN particle concentrations are relatively high, likely due to atmospheric processes that increase particle size over time such as the condensation of VOC oxidation products or cloud processing (Section 3.2 and 3.3, respectively). The RPF regimes exhibit a high CN concentration (>10 nm diameter), indicative of recent particle formation (Section 3.2)."

Section 3.3 on cloud processing relies strongly on ERA 5 data. Some discussion on the representation of clouds and particularly precipitation in the reanalysis product is needed. Over the SO there are not many observations that would constrain the reanalysis.

We agree that discussion of the ERA5 data set is needed, specifically on the lack of observations and have added the following text to section 3.3:

"Manton et al. (2020) showed the ERA5 annual cycle of precipitation across the SO is consistent with in-situ data, but it is important to note that there is large uncertainty because of the low number of observations to constrain the ERA5."

"Similar to ERA5 precipitation, there are also a low number of observations to constrain the ERA5 cloud fraction product. Ship measurements in the region south of Australia were recently shown to be consistent with daily averaged observations and ERA5 cloud fraction values of 0.75±0.23 and 0.71±0.27, respectively, providing some confidence in the ERA5 (Wang et al. 2020)."

Section 3.4 Latitudinal Gradient: Recent observations by Schmale et al. (2019) also highlight the higher concentrations of CCN near the Antarctic coast. See also their discussion of kappa for MSA and the role of particle size to activate as CCN. I recommend referring to their work in section 3.4, since they already came to similar conclusions presented in section 3.4.

We have modified this section based on comments from all the reviewers. Below is the relevant updated text:

"A latitudinal gradient is observed in both the GV HIAPER UHSAS particle (Dp > 0.07 µm) and CCN concentrations; however, the differences in their slopes imply a north-south gradient in particle composition (i.e., hygroscopicity) across the SO, as identified by the hygroscopicity parameter (κ_{70}) for Dp > 0.07 µm (Figure 6d). The presence of a latitudinal gradient in aerosol concentrations (Dp > 0.07 µm) and a weak gradient in the GV HIAPER CCN implies a north-south gradient in particle composition (i.e., hygroscopicity) across the SO. Figure 6d shows the hygroscopicity parameter (κ) for Dp > 0.07 µm derived at each MBL leg. The lower κ (less hygroscopic aerosol) at high latitudes is consistent with sulfates and organic aerosol from biogenic emissions, which have relatively low κ values (κ = 0.6-0.9 and κ < 0.2, respectively) compared to PMA (κ ~ 1.0 (Quinn et al., 2014)) present in primary emissions at the lower latitudes (Kreidenweis and Asa-Awuku, 2013; Petters and Kreidenweis, 2007). These results are consistent with findings of Schmale et al. (2019) showing MSA, an aerosol component associated with biogenic emissions, contributed about 2.5 times more mass in the Antarctic coastal region compared to the remote SO. Furthermore, the elevated CCN near the Antarctic coast is also consistent with a higher incidence of cloud processing in the region, despite the lower particle hygroscopicity (Alroe et al., 2020; Schmale et al., 2019)."

l. 346: Do the authors means the low variability +/- 0.04? Why would a wet diameter lead to a lower variability?

We have updated the text to more clearly indicate that a wet diameter could lead to a low geometric width. While typically observed in cloud droplet distributions, condensational growth leads to narrower distributions (Pinsky et al., 2014; Yum and Hudson, 2005).

"The mode diameter of the retrieved PMA number size distribution was 0.59 ± 0.04 µm, which is consistent with the average mode diameter observed in the North Atlantic of 0.54 ± 0.21 µm (Saliba et al., 2019). The low geometric width (1.44±0.25) of the PMA mode, relative to Saliba et al., (2019) (ranging from 1.5-4.0) likely reflects the available statistics (N = 74), and the possibility that the PMA particles were not completely dry (section 2.3)."

In section 3.5 PMA Marine Aerosol, again it would be useful to put the results into context with recent publication from other sectors of the SO. Schmale et al. (2019) show in their table 3 the contribution of their similarly identified sea spray mode to CCN and find between 20 and 30 % for SS = 0.15 %.

We have added this relevant finding in the following text:

> "These results are consistent with Twohy et al. (submitted) who found sea-spray aerosol comprised a minority of cloud droplet residual number in three SOCRATES cases. Similarly, Quinn et al. (2017) who found that PMA contributed to less than 30% of CCN number concentrations (at 0.3% supersaturation) from measurements collected during other field campaigns conducted between 130°E (near Tasmania) and eastward to 60°W (near South America) . Also, Schmale et al. (2020) showed that over three measurement legs that spanned the entire longitudinal range of the SO, the average PMA contribution to CCN ranged from 19-32% at a supersaturation of 0.15%."

l. 363 The low condensation sink (CS) is not really true because the presence of sea spray leads to such a high condensation sink that new particle formation in the marine boundary layer is rather an exception (also due to other factors). Compared to the Arctic Ocean (Baccarini et al. (2020), https://doi.org/10.1038/s41467-020-18551-0), the CS in the SO will be a factor four, or even more, higher. The authors have the necessary data to actually calculate the CS. Compared to other oceans (except polar oceans) the CS might be lower, but given the low new particle formation occurrence, saying low CS is not completely correct.

We agree that the presence of sea spray leads to a high condensation sink. However, we are pointing out that new particle formation often happens in the FT and not the MBL, even in the SO, despite the remoteness of the SO to continental and anthropogenic influences. In a recently published article (Sanchez et al. 2020; https://doi.org/10.5194/acp-2020-702) this exact calculation was performed using sea spray concentrations in the North Atlantic to show that under high wind conditions (> 10 m s^{-1}), the sea spray particles account for a significant fraction of the total particle surface area, and less so under low wind speed conditions. It is unclear as to why the reviewer states that the CS in the SO will be a factor of four or more higher compared to the arctic. There seems to be no mention of the SO in Baccarini et al. 2020, and little on sea spray specifically as a CS.

To better describe the CS in the SO we have updated the text as followed:

> "During this study, the CN_{Inv} is generally greater than CN_{MBL}, which suggests particle formation occurs more frequently above the MBL inversion, either in the free troposphere or a decoupled layer above the marine boundary layer. Despite the lack of influence from continental and anthropogenic particles as condensational sinks in the SO, the presence of a small concentration of PMA particles can lead to a high total particle surface area (Sanchez et al. 2020; Cainey and Harvey, 2002; Yoon and Brimblecombe, 2002) and prevent new particle formation in the MBL."

l. 366, which trend?

We have updated the statement as follows:

> "To determine if the SO MBL truly is an exception to the trend of NPF typically occurring in the FT, we compare the concentrations of FT and BL CN and UHSAS concentrations across the MBL."

Section 3.6, please provide the number of data points per vertical profile. It is difficult to understand how representative the six profiles from figure 7 are and why particularly those were chosen. How many were there? How was the histogram in Fig. 9 calculated, is there one ratio per profile?
Each data point in Figure 7 (now figure 8) represents a 4 ± 2 minute vertical profile of measurements made at 1 Hz. Consequently, the number of data points per vertical profile differ depending on the range of the vertical profile and presence of a cloud as shown in figure 7a-c (now figure 8a-c). Showing the vertical profiles are not necessary to make the point we have made about comparisons between CN concentration in the MBL and above inversion layers. The six profiles are provided as examples from each section of Figure 7d (now figure 8d). Overall, there were 110 profiles with measurements in both layers (MBL and above the inversion). The histograms in Figure 8 and 9 (now figure 9 and 10) are derived directly from the points in Figure 7 and Figure 8 (now figure 8 and 9) as indicated in the Figure 9 (now figure 10) description (one ratio per profile).

L. 400: The explanation of long-range transported CCN from the Antarctic coast is in contradiction to the minimum near 60_S. If the higher concentrations near the coast of Australia are due to specific long-range transport events, this should be said explicitly.

The text has been updated to indicate that a specific case of higher concentrations near Australia is found to be due to long-range transport from near the coast of Australia.

> ”Enhanced ship-based CCN concentrations north of 50°S are likely from Australia. In one case enhanced CCN concentration measured on the GV near the Australian coast is shown to be from long-range transport from Antarctic coastal emissions. Elevated CCN concentrations to the south of 60°S originate from biogenic emissions from the Antarctic coastal area.”

l. 392 f: The information on the four regimes is repeated in l. 404ff. Consider removing some redundancy from the conclusions.

The sentence on line 404 defining the four regimes has been removed.

Reviewer’s comments copied from attached supplement document with responses:

L. 23 “a location with elevated phytoplankton emissions relative to the rest of the SO.” … There are regions in the Southern Ocean, e.g. around South Georgia, which are known for the productivity. This statement is too strong. Rather "relative to most other locations in the Southern Ocean".

We have updated this statement as follows:

> “In 5-day HYSPLIT back trajectories, air parcels with elevated CCN concentrations were almost always shown to have crossed the Antarctic coast, a location with elevated phytoplankton emissions relative to the rest of the SO in the region south of Australia.”

L. 37 “The Antarctic coastal source of CCN from the south as well as CCN sources from the mid-latitudes create a latitudinal gradient in CCN concentration with an observed minimum in the SO between 55°S and 60°S.”... Is the minimum only a question of sources? Between 55 and 60 °S many cyclones lead to precipitation. So the minimum can also be an effect of enhanced removal processes.

We agree and have updated the statement as follows:

"The Antarctic coastal source of CCN from the south, CCN sources from the mid-latitudes and enhanced precipitation sink in the cyclonic circulation between the Ferrel and Polar cells (around 60 °S) create opposing latitudinal gradients in the CCN concentration with an observed minimum in the SO between 55°S and 60°S."

L. 51 "Schmale, 2019;"... incomplete citation.
Fixed

L. 54 A new publication on that topic is by Efraim et al. (2020): DOI : 10.1029/2020JD032409.
The citation has been added.

L. 70 "New particle formation (NPF) from the oxidation of marine biologically emitted VOCs occurs when the particle condensational 70 sink is low and temperature is low, both of which are prevalent conditions over the SO (Raes et al., 1997; Yue and Deepak, 1982)." … This is incorrect. The sea spray particles are large and actually produce a large condensation sink.

The text has been updated as follows:
"New particle formation (NPF) from the oxidation of marine biologically emitted VOCs occurs mostly in the FT where the particle condensational sink and temperature are lower than in the MBL, which are prevalent conditions over the SO (Raes et al., 1997; Yue and Deepak, 1982)"

L. 72 "While new particle formation has been observed in the SO marine boundary layer (Covert et al., 1992; Humphries et al., 2015; Kyrö et al., 2013; Pirjola et al., 2000; Weller et al., 2015)" ... I am not familiar with all paper listed here, but the Weller et al. 2015 paper does not say that NPF occurred over the ocean. They report rather on NPF at a coastal Antarctic site which has very different characteristics from the open Southern Ocean.
See updated text in response to the next comment.

L. 75 " owing to the absence of PMA in the SO MBL"... This statement has to be revised or different literature needs to be cited. This does not make sense. Did the authors want to say owing to the absence of PMA in the SO free troposphere?

The text has been updated as follows:
"While NPF has been observed in the marine boundary layer, often at coastal locations (Covert et al., 1992; Humphries et al., 2015; Kyrö et al., 2013; Pirjola et al., 2000; Weller et al., 2015), it occurs more commonly in the FT (Bates et al., 1998b; Clarke et al., 1998; Humphries et al., 2016; Odowd et al., 1997; Reus et al., 2000; Sanchez et al., 2018; Yoon and Brimblecombe, 2002) owing to the absence of PMA in the FT (McCoy et al., 2015)."

L. 81 "SO contains much less biological activity near the ocean surface relative to the Antarctic continental coast,"...This is only partly corrected. The area around South Georgia is highly productive. See https://doi.org/10.1002/jgrc.20270 for a climatology. So if measurements had been taken over that transect, the observations would be very different. Given the vast extent of the Southern Ocean it is important to consider regional variability.

We agree and have updated the statement to:
"The remote mid-latitude SO south of Australia contains much less biological activity near the ocean surface relative to the Antarctic continental coast, which creates a latitudinal gradient in

aerosol concentrations driven by biogenic particle formation with the exception of some biological hotspots such as near South Georgia (Alroe et al., 2019; Humphries et al., 2016; Kim et al., 2019; O'shea et al., 2017; Odowd et al., 1997; Weller et al., 2018)."

L. 84 "This trend in biology is linked to observations showing a distinct transition in aerosol properties around 64°S where CN concentrations"... Again, this is a certain longitude band which is not necessarily representative of all SO. Be specific.

The statement has been updated as follows:

"Shipborne observations in the region south of Australia show a distinct increase in aerosol concentrations south of 64°S, where CN concentrations are about five times higher during the austral spring months" …

L. 85 "Regions of sea ice melt on the Antarctic coast have been observed to be a significant source of methanesulfonic acid (MSA) as well as DMS and organic nitrogen (Dallósto et al., 2017; Dowd et al., 1997; Vana et al., 2007),"... The organic nitrogen observations from the Dall'Osto (note spelling) paper have not been made in conjunction with NPF. We do not know if Norg plays a role over the Southern Ocean for NPF.

We have omitted organic nitrogen and the Dall'Osto et al., 2017 reference from this sentence.

L. 116 "In the event that the cloud droplets do not precipitate, the evaporated particles are larger than their original size since non-volatile compounds (i.e., sulfate and MSA) that condensed onto the cloud droplet remain in the particle phase." … For aqueous chemistry, one cannot really speak of condensation.

We agree and have updated the sentence to state:

"In the event that the cloud droplets do not precipitate, the evaporated particles are larger than their original size since aqueous oxidation of volatile compounds (i.e., DMS, MSA, SO2 and nitric acid) have formed non-volatile sulfates and nitrates that remain in the particle phase."

L. 118 "This added mass shifts Aitken mode particles to the accumulation mode (Hoppel et al., 1986; Hudson et al., 2015; Kaufman and Tanré, 1994; Sanchez et al., 2017)." … Schmale et al. (2019) describe this as well in their overview article. This would also be a good resource for measurements in sectors of the SO other than South of Australia and South of South America.

We agree that we should indicate that our measurements are representative of the region south of Australia; however, we have mentioned properties of other areas of the Southern Ocean as needed. We have added Schmale et al. (2019) as a reference in this statement.

L. 130 "In this study, we discuss airborne CN and CCN measurements from the SOCRATES campaign."… What about CAPRICORN?

We have added the following text to the end of the statement:

" and briefly discuss shipborne CCN measurements made on the *R/V Investigator* for the CAPRICORN2 campaign, which was conducted in the same timeframe and region as SOCRATES"

L. 143 “research flights (RF)”... inconvenient abbreviation, because it also means radiative forcing, and this is certainly the more common use. Might confuse readers.

We agree that acronyms can be confusing, but it is standard NCAR protocol to use RF for ‘research flights’. We will keep RF in the manuscript.

L. 149 “deiced components”... This is not understandable to me.

We have updated the sentence as follows:
“Ambient subsaturated particles collected with the UHSAS were dried through a de-icing system (designed to vary the temperature and pressure of sampled air to prevent ice formation in the inlet).”

L. 150 The miniature version is not described in the 2005 reference. Either provide a relevant reference or describe the instrument.

The exact method described in Roberts and Nenes 2005 is used for the miniature CCN counter.

We have added the following sentence for clarity:

> “The miniature CCN counters are custom-made and operate with the same physical principles as described by Roberts and Nenes (2005). “

L.159 “hygroscopicity parameter (κ_{70}) ” add information on how exactly this was done

The text has been updated and a sentence has been added to explain the calculation of the hygroscopicity parameter.

> “The CCN spectra and UHSAS number concentrations on the GV were used to estimate the hygroscopicity parameter at 0.07 µm diameter (κ70) for each MBL leg. For this calculation, the critical supersaturation is derived from the CCN spectra, where the UHSAS concentration at 0.07 um diameter is equivalent to the CCN concentration.”

L. 171 “R/V Investigator CCN at 0.35% are analyzed and compared to the GV HIAPER CCN0.3 measurements.” … Why are they not interpolated to 0.3 % SS? Description of the inlet and position of the instruments on the ship is missing.

The R/V investigator CCN measurements have now been linearly interpolated to obtain an estimate of the 0.3% SS CCN concentration.

The following text has been added to address the ship inlet and position of instruments on:

> “Details of the aerosol sampling system on board the RV Investigator are presented in Humphries et al. (2019) and Alroe et al., (2020). In short, aerosol sampling occurred via a common sampling inlet mounted on a mast at the bow of the ship, located 18 m above sea level. The CCN counter sampled from a manifold located 8 m below the mast in the ships bow.”

L. 177 “The average latitude, longitude and altitude (50-500 m) of the MBL legs on the GV HIAPER were used as starting points for the back trajectories.” … Unclear. To me this sounds that for each leg there was only one coordinate from which the trajectories were released? If this is the case, large

uncertainties are linked to this method in terms of meteorological properties obtained along the trajectories. This makes the results less trustworthy.

We have updated the text as follows:

> "The latitude, longitude and altitude (50-500 m) averaged for each CCN spectra (~150 seconds, ~15-20 km horizontal distance) collected during the MBL legs on the GV HIAPER were used as starting points for the back trajectories."

L. 193 "UHSAS particles in the SOCRATES campaign were not fully dried"...A quantification would be helpful to make this study comparable.

This has been answered in a previous comment.

L. 201 "The TEM analysis shows almost all particles > 0.2 µm in diameter consist of sea salt; and sea salt particles account for 25% to 100% of particle number concentrations at particle diameters > 0.4 µm (Saliba et al. submitted)." … This is inconsistent."almost all" means to me near 100 %. So how can only 25 % of particles > 0.4 um be sea salt?

This has been answered in a previous comment.

L. 207 "Figure 1b shows" ... should reference 1a first.
Note figure 1 is now figure 2.
We believe Figure 2b should be referenced first, Figure 2 is designed so that figure 2a and 2c align with the y and x-axis respectively, of figure 2b and contain histograms of the data shown in figure 2b.

L. 212 "The regime thresholds were selected based 210 on the bimodality of observed CN and CCN0.3 concentrations shown by the histograms and kernel density functions in Figure 1a,c. Figure 1a,c also shows the kernel density estimate based on a normal kernel function."..... I only see one kernel density function. Why "also"?

The statement was repetitive, and the second sentence has been removed.

L. 213 "each measurement is represented by a normal distribution"... Why not log-normal?

A kernel density is a non-parametric (no assumptions involved) way to estimate a probability distribution. For the range of concentrations encountered here, there is also no compelling reason to represent measurements of concentration as log-normal.

L. 229 "the relative contribution of the accumulation-mode and the Aitken-mode are used to identify recent particle formation (RPF) events"... An explanation of how the Aitken-mode was estimated is missing.

This comment has been answered in the previous reference to L. 229.