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General comments Sanchez et al. (Cloud Processes and the Transport of Biological Emissions Regulate Southern Ocean Particle and Cloud Condensation Nuclei Concentrations) provides a very good summary of airborne and marine boundary layer aerosol and CCN measurements over the Southern Ocean. These are extremely valuable measurements and provide excellent information on aerosol sources, atmospheric processing and the resulting CCN/CDNC. I have outlined some minor changes to the manuscript. A little more detail on the calculation and interpretation of the hygroscopicity, and on the uncertainties would improve the clarity of the manuscript. Specific comments are outlined below.

Specific comments
L 41: What are the conclusions regarding the latitudinal gradient in particle composition and hygroscopicity based on? Are there measurements for this?
The text has been updated to indicate the measurement hygroscopicity is based on combining CCN spectra and particle size distributions. See updated text in response to the next comment.
L 41: Isn't there biogenic/coastal emissions in the lower latitude Southern Ocean, and therefore lower sea salt fraction?
Yes, there are biogenic/coastal emissions near the Antarctic coast that are transported northward, but the main reason for the lower sea salt fraction in these lower latitudes is a result of less wind and the presence of sea ice (consistent with findings from Schmale et al. (2019)).
We believe the reviewer is referring to the Australian coastal/biogenic emissions. Almost all of our measurements are at latitudes higher than 50S, putting them well away from the Australian coastal influence. Furthermore, the back trajectories are all from the west/south and therefore, are not heavily influenced by the Australian coast. We have added that the gradient is derived from measurements south of the Australian coast:

> "In addition, a latitudinal gradient in the particle composition, south of the Australian and Tasmanian coast, is apparent in aerosol hygroscopicity derived from CCN spectra and aerosol particle size distribution. The particles are more hygroscopic to the north, consistent with a greater fraction of sea salt from PMA, and less hygroscopic to the south as there is more sulfate and organic particles originating from biogenic sources in coastal Antarctica."

L 54: Which particle sources?
We have changed "these particle sources" to "Southern Ocean particle sources".
L 84: Alroe et al. doesn't show such dramatic increase in number concentrations, and was a summertime voyage.
We agree, Alroe et al. showed a less dramatic increase. We have discovered that the Alroe et al. manuscript has been updated to its final published form, which no longer contains plots showing the particle concentration as a function of latitude. For this reason, we have removed the reference to Alroe et al. here, but left the reference in other sentences where it is still appropriate. The following reference has also been added to the results section:

> "The storm track frequency peaks around 60°S (Patoux et al., 2009), suggesting parcels of air entering the storm track from the south have also been influenced by coastal Antarctic biogenic DMS and other VOC emissions, eventually leading to increases in CCN concentrations via cloud processing and in the absence of precipitation. Schmale et al, 2019 and Alroe et al. 2020 also

find that the higher fraction of particles serving as CCN near the coast of Antarctica, are also from biologically derived particles."

L 92: This paragraph could be simplified. The discussion of PMA (e.g. from "In a recent study.." onward) could be separated out from the discussion of seasonality.

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 FT 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)."

L 93: Biological particles is an unclear term, perhaps something like secondary particles from biological sources. Biogenic particles is used on L95.
"biological particles" has been changed to "secondary particles from biological emissions"

L123: The sentence beginning "In contrast, the inclusion..." would be better placed in a paragraph about PMA.
We agree and have moved this sentence to the paragraph related to PMA.

L125: The sentence beginning "SO satellite-derived....", begins the discussion of cloud properties and could be the start of a new paragraph.
We agree and have started a new paragraph here.

L120: Which sensitivity? McCoy et al. doesn't discuss aerosol growth via cloud processing.
McCoy et al. did not specifically discuss aerosol growth via cloud processing, but they did discuss the general complexity of aerosol processes that lead to natural aerosols influencing clouds, CDNC and overall radiative forcing.
We have updated the text as follows to prevent confusion.

"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. 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). Results from McCoy et al. (2015) emphasized that, despite the ambiguous results from focused modeling 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."

L149: Do we have any idea to what RH the sample was dried?
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."

L159: Some detail on how kappa was calculated is required. I assume the supersaturation at which the CCNc = UHSAS # conc (>0.07um) was taken as the Scrit. Then the Scrit/Dcrit were used to compute the kappa. Although I'm not sure how it was computed, I don't it was measured at 0.07 um (i.e. pre-selected). Therefore, perhaps the expression "the 0.07 _m diameter hygroscopicity parameter (_70)" is a bit misleading.

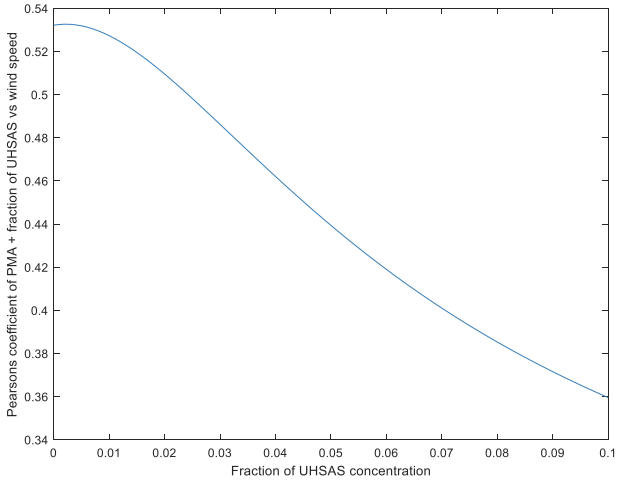
The UHSAS distribution lower cut off size of 0.07 µm diameter particles was used as the critical diameter in the kappa derivation; hence, the 70 notation. We have updated the text to explain how the critical supersaturation and diameter were determined.

Updated text:

"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."

L195: How do the authors know that the fitted PMA distribution isn't a subset of the total PMA? Particularly the deliquesced mode at _0.6 um, since the GSD _1.44 is on the lower end of that observed by Saliba et al. 2019. Does the sensitivity testing that was done in Saliba et al. 2019 necessarily transfer across to these data?

To test if the fitted PMA distribution is not only a subset of the total PMA distribution, we added various fractions of the remaining UHSAS distribution to see if the correlation with wind speed would improve (figure below), similar to the sensitivity test performed by Saliba et al. 2019. The correlation only decreased suggesting the fitted PMA distribution is representative of the total PMA.



Reviewer Figure 1

Since this deliquesced mode exists, it suggests the particles have not reached efflorescence, which is based on the relative humidity and particle composition independent of size for larger particles if at equilibrium (Cheng et al., 2015). However, if the particles are limited by mass or heat transfer rates and are not at equilibrium, then it may be that larger particles are slower to dry out than smaller particles. While typically observed in cloud droplet distributions, condensational growth leads to narrower and larger distributions (Pinsky et al., 2014; Yum and Hudson, 2005), so it is not surprising that the GSD is on the lower end of what was observed by Saliba et al. (2019).

Note the efflorescence RH of sea salt (~47%) is higher than the estimated drying RH (<40%) suggested by Strapp et al. (1992). We hypothesize that the low residence time in the PCASP (~0.2 seconds) was not enough for the larger hygroscopic sea salt to fully dry before being measured. In addition, the fact that we do not see this mode in the free tropospheric measurements supports that this is a PMA mode.

We added the following text to address this concern:

> "This deliquesced mode was present despite the findings by Strapp et al. (1992), suggesting the deicing 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."

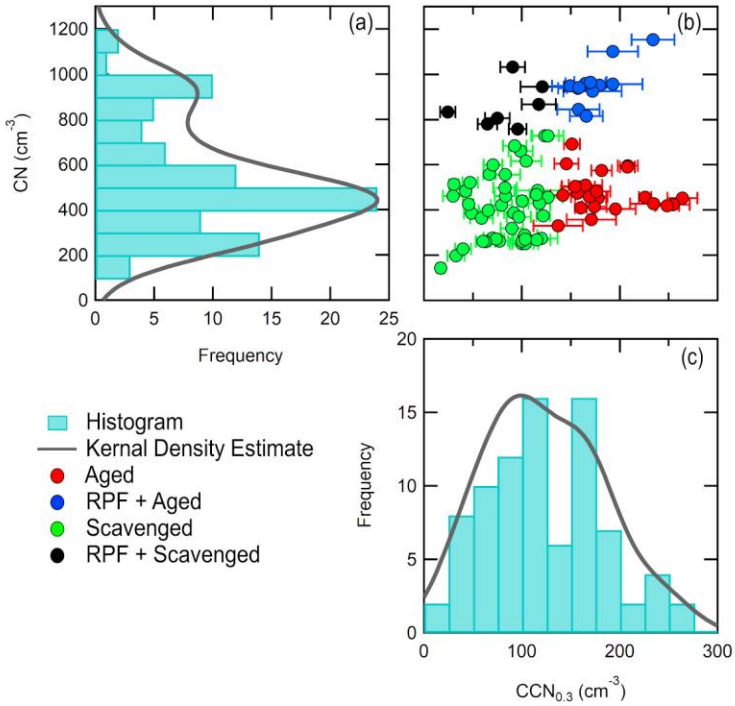
L199: What are the mode (and sd) for the fitted PMA distribution? and how do they vary? Only N is reported here.

The following sentence has been updated to include the mode diameter and the sd of the fitted PMA distributions.

> "The estimated PMA mode diameter and geometric width (0.59 ± 0.04 µm, 1.44±0.25, respectively) are consistent with sea salt distributions (from PMA) observed on size-resolved particles collected in the marine boundary layer during SOCRATES and analyzed with transmission electron microscopy (TEM)."

L208/Fig 1b: What is the uncertainty/variability in these averaged CN/CCN? Error bars would be helpful.

The figure has been updated to include error bars representing the standard error. The standard error in CN concentration is sufficiently small so that the error bars are within the size of the marker.



L228: "Aitken-mode particles (CN)" is unclear and suggests CN excludes the accumulation mode.
We agree and have changed the text to "Aitken + accumulation-mode particles (CN)"

L249: "(RPF + Aged and RPF + Scavenged)"
Fixed.

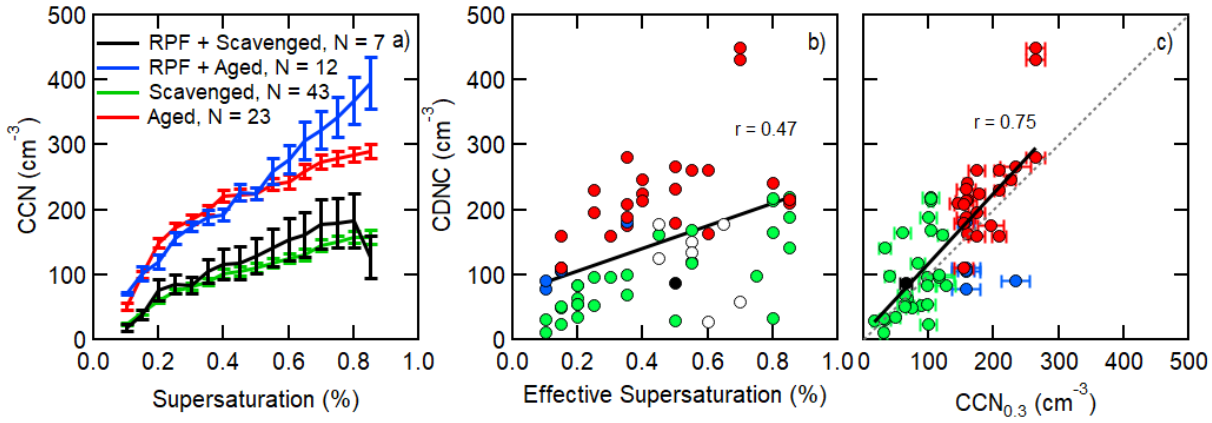
L256: What was the rationale for taking the 90th percentile? and how wasa this implemented? the 90th percentile for each vertical profile?
The 90th percentile was used to consistently exclude cloud droplet measurements that were heavily influenced by entrainment drying. The 100th percentile was not used to avoid outliers. The text has been modified and is shown below to clearly state our reasoning:

> "For this comparison, the 90th percentile of CDNC from each vertical profile is matched to the nearest below-cloud MBL leg CCN concentration. The use of the 90th percentile of CDNC excludes measurements that are heavily influenced by entrainment drying and also excludes outliers."

L256/Fig 3c: It would be good to represent the variability CDNC, and how this variability influences the relationship between CCN and CDNC.

We agree this would be ideal; however the CDNC were generally measured during vertical profiles and are susceptible to variations at cloud top/cloud base. Horizontal legs through cloud layers would be better for identifying the variability in CDNC, but there were few such legs during SOCRATES.

Fig 3: Error bars are required in Figure 3b and c.
Since we used the 90th percentile rather than an average (and since we measured CDNC during vertical profiles), we do not have a good representation of the CDNC horizontal variability. We have updated the figure to include the horizontal variability of CCN during low level legs.

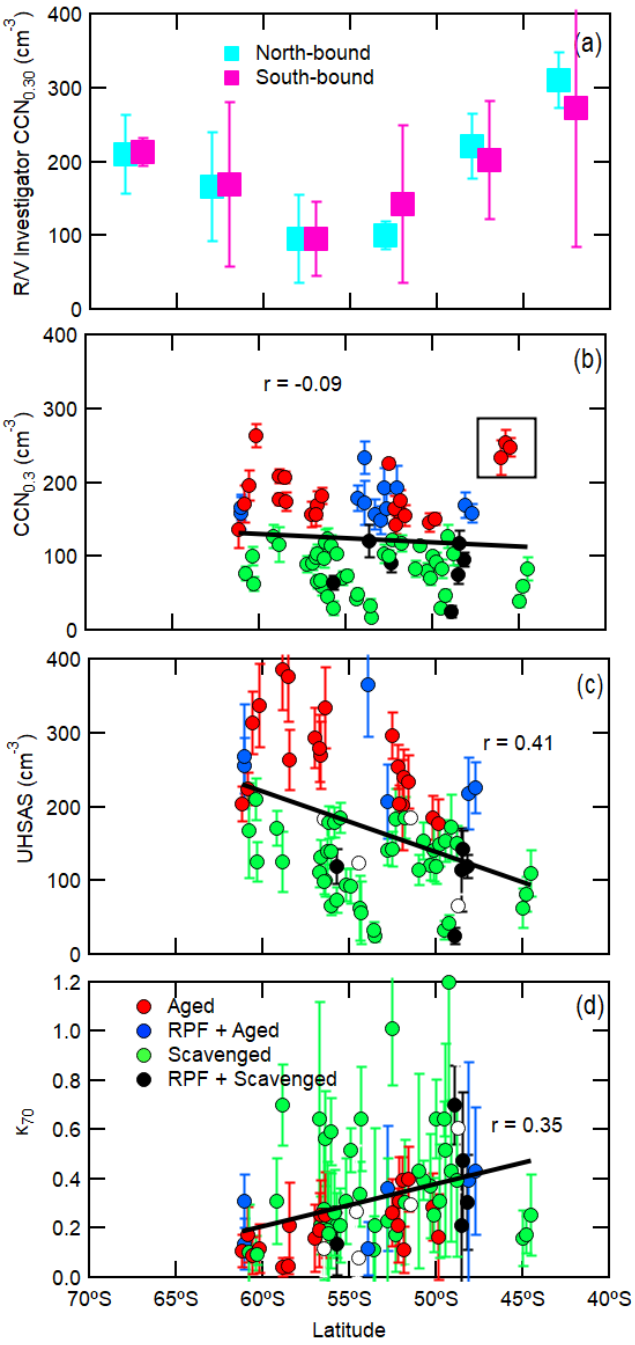


L266: The sentence starting "The two regimes.." should be revised. The figure shows that the CDNC for aged is greater than CDNC for scavenged, there is too little data to draw conclusions for the RPF cases.
We agree and have revised the sentence to the following:

> "The two regimes with aged particles (high CCN) consistently had higher CDNCs than the scavenged regime, highlighting the role of CCN concentrations as CDNC."

L310: Does removing these transported outliers result in statistical significance in the trend ? to what p-value?
Removing the outliers results in a weak correlation ($r = -0.26$; p-value < 0.05).

Fig 5: Could error bars be added to the CCN, UHSAS and kappa?
1-sigma error bars have been added to the figure and is shown below:



L315: It is not clear how the CCN and CN (>0.07 um) concentrations by themselves imply anything about composition/hygroscopicity.

As mentioned earlier in the text, the CCN spectra and UHSAS number concentrations were used to estimate the hygroscopicity parameter at (κ_{70}) by relating the UHSAS concentration at 0.07 µm to a CCN concentration at a given supersaturation. The lower cut size of the UHSAS (0.07 µm diameter) serves as the critical diameter in determining kappa.We have updated the text to better explain how the data indicates there is a north-south variation in particle composition:

> "A latitudinal gradient is observed in both the 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) for Dp > 0.07 µm (Figure 6d)."

L319: The computed kappa values are lower than that expected for sulfates, particularly at high SO latitudes. At lower latitudes the kappa values are consistent with sulfates. This should be noted/discussed.

The sentence has been revised:

"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."

L332: Seawater biogenics (e.g. chl-a) increase in the low latitude SO, north of the subantarct ic front. This isn't necessarily associated with the Aus continent. McCoy et al. 2015 pointed out that secondary aerosols played a more important role in CCN/CDNC in the lower latitude SO, with the contribution from PMA increasing with latitude up to 60S.

We agree with this statement have updated the text and cited McCoy et al. 2015:

"There are also elevated CCN concentrations north of 50°S measured on the R/V Investigator, probably related to continental emissions from Australia, elevated biomass emissions of VOCs (aerosol precursors), as suggested by increasing chlorophyll-a concentrations north of the subantarctic front (McCoy et al. 2015), and even long-range transport of Antarctic coastal emissions (Ayers and Gillett, 2000; Twohy et al., submitted)."

L346: In general the variability using this method is quite high. How does the variability compare with that in Saliba et al.

The variability in the mode diameter is actually low compared to Saliba et al. 2019. Please see updated text:

"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)."

L414/L41: The hygroscopicity values are low and aren't necessarily consistent with sea salt in the north and sulfates (+organics) in the south.

We agree that the hygroscopicity values are not "consistent" with sea salt but are "consistent with a greater fraction of sea salt".

Technical questions

L29: "coarse-mode" not "course-mode" Fixed.

L101: "though" is not necessary Fixed.

L235: RPF regimes exhibit Fixed.

L370: I think it should be "< 0.7" not "> 0.7" Fixed.