We would like to thank both referees for their constructive comments about our manuscript. We now address their comments individually. For clarity referee comments are coloured red and responses are in black.

Referee 1

Overview

This manuscript presents an analysis of cloud and aerosol measurements collected during the Microphysics of Antarctic Clouds field campaign. The main focus of the analysis presented is on extensive airborne observations from 24 flights, primarily in conditions containing stratiform cloud layers, and these are supplemented by measurements made at a surface site. The observations show that the clouds are dominated by liquid water with variable (and typically low) concentrations of ice particles, suggesting that there are limited sources of primary ice nuclei that are active in the temperature range of these clouds. The ice particle concentrations tended to increase in the H-M temperature zone, suggesting that secondary ice production can play a role in theseclouds. The main strength of the paper lies in the fact that there is a scarcity of in-situ observations of aerosol and clouds in the Antarctic, resulting in a very limited number of observational constraints that can be used to evaluate NWP and climate models in this region. The novel observations in this paper certainly have the potential to be useful for model evaluation studies and increase our knowledge of some cloud and aerosol microphysics parameters in the region. I do however think that some additional analysis of the data is required before the manuscript can be published in ACP (see comments below).

Main comments

1. Introduction: The authors give some background information on previous Antarctic INP measurements, but there is an absence of information on previous CCN measurements. The CCN are key to the liquid dominated clouds studied in this paper. The introduction should be expanded to include additional information on past results on Antarctic CCN data to put these new observations into context.

The description of previous aerosol measurements in Sect 3.2 has been moved to the introduction and extended to include references discussing aerosol hygroscopicity in the region.

2. Section 2.1: The information on the meteorological and cloud conditions that were present during the observation period needs to be strengthened significantly. I realise that there were a lot of flights and that it is not straightforward to summarise this information in a paper, but the very short bit of text on page 6 (lines 1 to 5) is inadequate. Perhaps including additional information in a supplement would be worthwhile, such as a surface analysis chart, a satellite image and the back-trajectories calculated in section 3.5 for each case.

As suggested we have added surface pressure charts and HYSPLIT back trajectories for each flight in the supplementary material. A table has been added to the manuscript (Table 1) detailing cloud base/top height and temperatures. Also included is information about whether multiple cloud layers were present.

3. Vertical profiles of thermodynamic and cloud data for each aircraft flight would also be extremely useful to include, which again perhaps could be included in a supplement. This would enable the reader to put the microphysical measurements into better context with the cloud and meteorological conditions for each case. It would also be extremely useful for model evaluation purposes.

The aim of this paper is to provide a statistical overview of all measurements. Showing thermodynamic and cloud data profiles for each individual flight would make the paper overly long and repetitive. The data used in this paper will be made publically available at the Centre for Environmental Analysis archive allowing direct comparisons to be made between measured and modelled microphysics.

4. Data from all flights are composited and summarised as a function of altitude (Figures 4, 11, 14), yet presumably there is significant day-to-day variability in the cloud top and cloud base heights. The main problem with this approach is that it is difficult to disentangle changes in the in-cloud, above cloud and below cloud measurements (e.g. location of ice and aerosol particles) with variability in the vertical location of the clouds. Have the authors considered normalising the data relative to the position in the cloud (at least for single-layer clouds), which would then be more comparable to previous studies e.g. McFarquhar et al. (2007)? This, in combination with vertical profile plots (comment 3) would give the reader a much clearer picture of the cases sampled by the aircraft

For the single layer clouds we have added a plot showing IMF (and liquid water content/effective radius) as a function of normalised position within clouds. The following text has been added to manuscript:

"Figure 3b, shows ice mass fraction measurements in single layer clouds as a function of the normalised position within the cloud, Zn.

$$Zn = \frac{Z - Z_B}{Z_T - Z_B},$$

Equation 2

where Z is the altitude, $Z_B$ and $Z_T$ are cloud base and cloud top altitude, respectively. We note that there is some uncertainty in determining cloud base/top due variability in the cloud and also incomplete sampling (this uncertainty is estimated in Table 1). The clouds were dominated by liquid drops throughout, while ice was more prevalent lower in the clouds. The relationship between ice mass fraction (IMF) and Zn over the range $0<Zn<1$ can be approximated by the equation:

$$IMF = 0.177 + 0.360Zn + 0.244Zn^2$$

Equation 3

This is shown as a red line in Fig. 3b. Figure 3c and d show that both liquid water content and cloud drop effective radius increase closer to cloud top. The effective radius increases from 4 ± 2 µm near cloud base to 8 ± 3 µm near cloud top.

Measurements in Arctic stratus/stratocumulus generally find these clouds to be similarly dominated by liquid drops (McFarquhar and Cober, 2004; McFarquhar et al., 2007; Lloyd et al., 2015a). A polynomial relationship derived during the Mixed-Phase Arctic Cloud Experiment (M-PACE) is also shown as a blue line in Fig. 3b (McFarquhar et al., 2007). McFarquhar et al. (2007) show a trend of increasing IMF with increasing distance from cloud top (and increasing temperature). Glaciated conditions were observed during 23% of their measurements. This is significantly more than during MAC, possibly due to lower INP concentrations available for primary ice development in the Antarctic compared to the Arctic, but differing sampling strategies may also contribute to this difference. ”

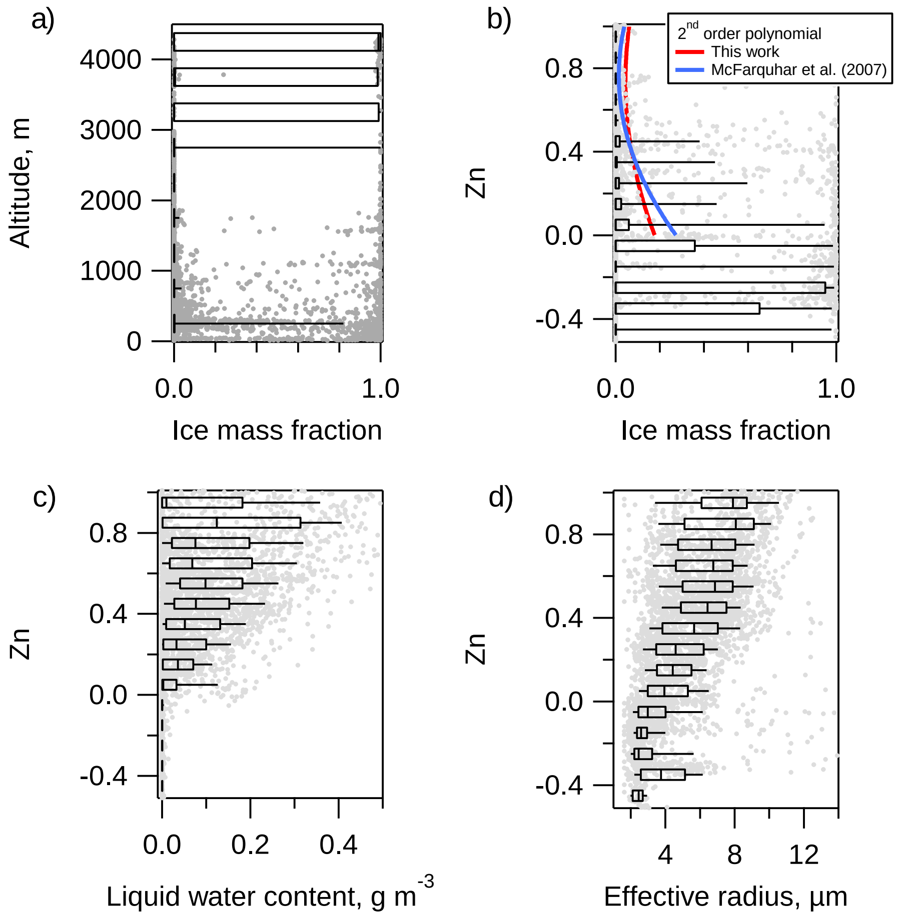



*Figure 3a. Ice mass fraction as a function of altitude and b) normalised position within the cloud (Zn). c) and d) show similar plots for liquid water content and effective radius from the CAS probe. Boxes are the 25th and 75th percentiles, the whiskers are the 10th and 90$^{th}$ percentiles.*

5. Page 9 line 22: The calculation of ice mass fraction does not include all particles. The ice mass is taken from the sum of the 2DS MI and HI categories, which cover particles larger than about 80 microns. The LWC is calculated from the CAS, which covers particles sizes less than about 50 microns. Furthermore, as discussed earlier in the manuscript the MI category may include large drizzle drops. The CAS would also be expected to measure small ice particles. Can the authors give some measure of the uncertainty in the calculated IMF that would result from defining liquid and ice water content in this way?

It is not possible to unambiguously determine the phase of particles smaller than about 80 µm. However, under mixed phase conditions ice will rapidly grow at the expense of liquid drops. Given a typical crystal growth rate of 1 µm $s^{-1}$, within a couple of minutes crystals will grow to a size where there phase can be determined by the 2DS. Therefore it is unlikely that the number of crystals smaller than 80 µm is significantly larger than those greater than 80 µm. As a consequence crystals smaller than 80 µm are only expected to make a small contribution to the total ice mass.

As described in the text the concentration of MI particles was generally significantly less than HI particles. The mean ratio HI:MI for the campaign was 7. If we assume that all MI particles are large drizzle droplets (inspection of the MI images suggests this is unlikely) then 5% of measurements are classified as fully glaciated (0.9<IMF<=1) compared to 6% if MI images are classified as ice.

6. Figure 4: It would be better if the authors showed the IMF data as a box and whisker plot rather than plot a mean profile which is pretty meaningless, especially above about 1 km altitude. At these altitudes the mean profile shows values between 0.2 and 0.8, yet the majority of the data points look to have values close to 0 or 1 i.e. the clouds appear to be dominated by liquid drops or they are almost completely glaciated at these altitudes.

As suggested this has been changed to a box and whisker plot.

7. All figures where you include box and whisker diagrams: I would encourage the authors to ensure that there are the same number of data points included in each bin, otherwise the plots can be misleading. For example, when referring to Figure 9 on page 18 line 6, the authors state that “there was a trend to higher ice concentrations in both updrafts and downdrafts”. It is unclear to me if the apparent increasing trend in the updrafts is simply the result of poor statistics. There appear to be very few data points with w > 2 ms-1, which are what I think the authors are using to justify the statement. If the bins were adjusted to include the same number of data points then I suspect that the trend may not be evident.

For each box and whisker plot the raw data is also shown so that the reader can see the strength of the relationship. Also note that Figure 9 has been removed from the manuscript (please see our response to comment 12).

8. Page 13 line 16: Can the authors use the CIP data for the flights where the 2DS was not operating? This would be of interest as it would extend the analysis to colder temperatures, where there is arguably larger differences in the INP parameterizations.

CIP data has been added to Fig. 4b (below). This shows that the INP parameterisations overestimate ice crystal concentrations at the lowest sampled temperatures (approximately -27 °C).

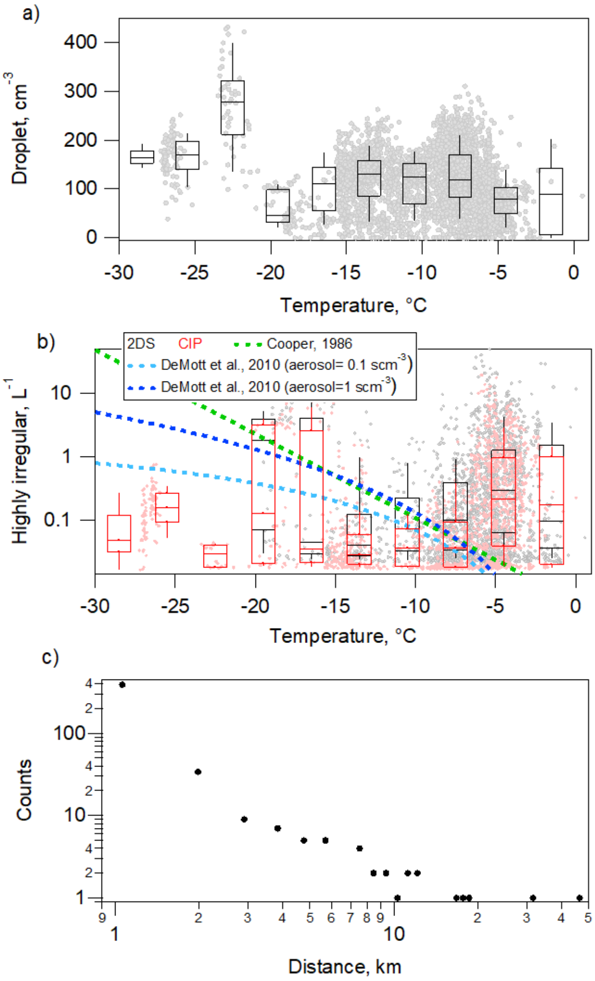


*Figure 4. Box and whisker plots summarising in cloud measurements (averaged over 10 s) as a function of temperature. Plate a) shows the concentration of cloud droplets ($cm^{-3}$), measured by CAS, while b) shows the concentration of ice particles measured by 2DS and CIP-25, based on those classified as highly irregular (see text for details). The concentration of ice nucleating particles predicted by the DeMott et al. (2010) parameterisation with a high (1 $scm^{3}$) and low (0.1 $scm^{3}$) aerosol input are shown as dark and light blue lines, respectively in b). The green line is the predicted ice particle concentration according to the Cooper (1986) parameterisation. c) a histogram of the flight distance while continuously sampling ice.*

9. Page 13 line 22: Again no clear trend is evident. There are very few data points at temperatures lower than -15 C and there is a lot of scatter in the data.

This refers to temperatures greater than -15C, which is where the majority of measurements during the campaign were made. This sentence has been clarified to read:

"At temperatures greater than -15 °C there is a trend of the ice crystal concentrations showing greater variability and higher median concentrations with increasing temperature."

10. Page 13 line 24: Is the figure showing the histogram missing? This is key to demonstrate that the ice occurs in small patches in the liquid clouds.

This figure has been added to the manuscript (Fig. 4c):

11. Page 15: Why do the authors only sub-select data in the H-M temperature range where secondary ice production might be expected to be enhanced? It would be beneficial to also look to see if there was a relationship outside of this temperature zone e.g. to see if there is any evidence of primary ice being formed from the freezing of drizzle drops (e.g. Rango and Hobbs 1991).

As discussed in the manuscript laboratory experiments have suggested that H-M production rates are proportional to the accumulation of large drops (>24 μm) (Mossop and Hallett, 1974). The aim of Fig. 5 was to examine if such a relationship could be identified in the MAC field measurements. As suggested similar plots have been added to the manuscript for temperatures <-8 C.

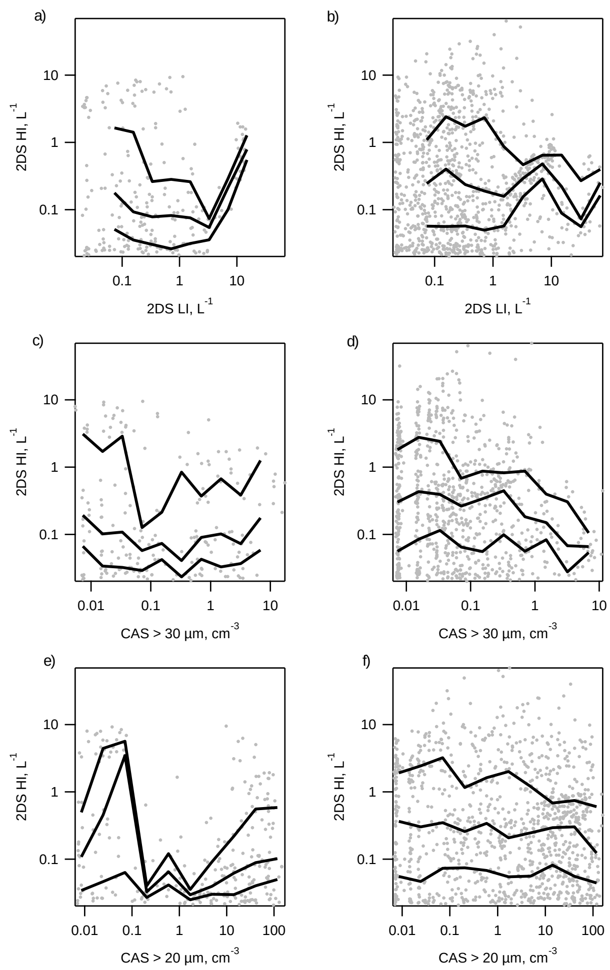



*Figure 5a,b. The relationship between the concentration of highly irregular (2DS HI) particles and low irregular particles (2DS LI) (low irregular particles greater than approximately 80 µm). Figures 6c,d and 6e,f show the relationship with the concentration of droplets larger than 30 and 20 µm, respectively. Panels on the left (a, c and e) show measurements at temperatures lower than -8°C and panels on the right (b, d and f) show* *those in the range -8 to 0 °C. The black lines are the 25th, 50th and 75th percentile of the 2DS HI concentration for each droplet concentration bin.*

12. Page 18 and figure 9: What is the mechanism by which glaciation would occur preferentially in downdrafts? 13. Figure 9: From the figure, it looks like the frequency of downdrafts measured by the aircraft is much larger than the updrafts. This was surprising to me and I would like the authors to confirm that there is no instrumental bias in the vertical wind measurement the xx. Assuming that there is no bias, can the authors explain the higher frequency of downdrafts in the measurements?

Given the uncertainty in the vertical wind measurements this has section has been removed from the paper. In anticipated that this will be incorporated into ongoing work examining eddy covariance flux measurements from the aircraft.

14. Page 19: Is the implication that the columns observed at warmer temperatures are generated by secondary ice processes? If so make that clear to the reader and link to figure 5 perhaps.

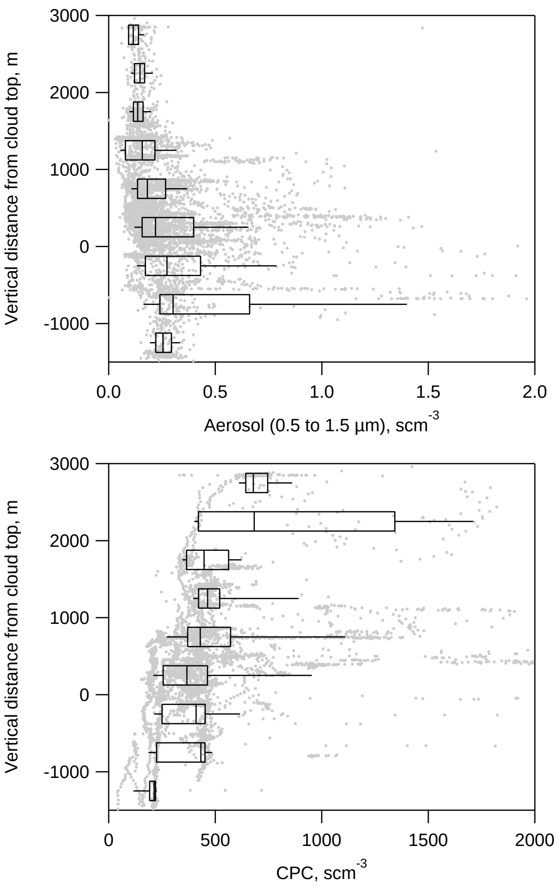
The paper is structured so that Sect. 3 presents the measurements with minimal interpretation, while Sect 4. provides more subjective discussion of the results. Section 4.2 makes the link between crystal habit and secondary production.

15. Figure 11: Can you include CPC data from the ground site? Also, how is number concentration derived from the aerosol scattering cross-section?

Measurements from the CPC at the ground site have been added to this figure. Particles are sized using Mie scattering theory assuming spherical particles of known refractive index.

16. Page 21 line 13: Presumably any surface generated aerosol is inhibited from being transported above the boundary layer as there are likely to be strong thermodynamic gradients co-incident with the top of single-layer clouds? If the data were normalised and plotted relative to cloud/boundary layer top (see point 4), you may expect to see sharper vertical changes in the profile of aerosol. These would be smeared out in figure 11 as all flight data are simply plotted relative to altitude and the data are therefore averaged over different boundary layer depths. Also, if the boundary layer was not well-mixed, then this could also result in a drop-off in the concentration of surface generated aerosol with altitude. Can you examine the aircraft thermodynamic data to examine if this was important?

The following figures show the aerosol concentrations plotted relative to the cloud top height. The concentration of aerosol in the size range 0.5 to 1.5 µm decrease above cloud top as would be expected from a surface source of aerosol. While the CPC shows layers above cloud with high concentrations. These features are the same as the original plot in the paper where the data was plotted against absolute altitude, so the manuscript is unchanged.




17. Page 23 line 8: Is there any evidence that the enhanced aerosol concentrations observed above cloud are being entrained into the cloud top? The suggested linkage between high aerosol concentrations above cloud top and the presence of clouds is rather tenuous.

The lack of correlation between above cloud aerosol and cloud concentrations across the flights suggests that the entrainment is relatively limited.

18. Page 24 lines 6 to 10: This short paragraph on the WIBS data (which is not very informative) should either be expanded to link to the aircraft measurements or other ground based data, or removed given that a subsequent paper on this data is planned.

Given this section discusses aerosol measurements it is appropriate to provide some information on the aerosol composition. Bioaerosol are particularly important since they have been identified as an important high temperature INP. However, these measurements are described in detail in a recently published ACPD paper, so are only briefly summarised here. A reference to the ACPD paper has been added to the manuscript.

19. Page 26 line 1: Can you use back-trajectories to see if there is a different source region for the case where the aerosol hygroscopicity increased between 28 and 29 Nov?

HYSPLIT trajectories indicate this airmass had passed over sea ice/open water regions. However after 30 November 2015 the hygroscopicity was relatively consistent and does not show a significant relationship with the wind direction/airmass history. This information has been added to the text.

20. Page 27 line 15: What about using the below-cloud aerosol concentration? This is more likely to be relevant, especially for cases were no elevated aerosol layers were observed to be in contact with cloud top.

No significant correlation was observed between below cloud aerosol and ice concentrations.

21. Figure 14: Is the data in the top right panel suggesting that the source of the elevated aerosol concentrations are from the southern Ocean? Are these above cloud?

Yes it does. Most of the measurements between 3 and 4 km had high clouds above.

22. Figure 15: It is somewhat surprising that given that there is evidence of a correlation between aerosol concentrations and source region (Fig 14) that this is not apparent in cloud drop concentration. I would have expected increases in the aerosol concentrations to result in elevated cloud drop concentrations in these liquid dominated layer clouds.

Yes this might be expected. The strongest relationship between aerosols and airmass history is for particles 0.5 to 1.5 µm this is only a small proportion of the total CCN. While the CPC covers the CCN but also smaller particles. Also, given that the majority of measurements were conducted over broken sea ice, it may be that the CCN origin may be more local and not show up in the far field trajectories. This discussion has been added to the text.

23. The discussion section focuses solely on ice production in the clouds. Given that these are liquid dominated clouds the authors should also include some discussion on the liquid phase.

The following paragraph has been added about the liquid phase:

"This section summarise the observations presented in the paper and discusses the important microphysical processes. The cloud types were generally stratus, both single and multiple layers, predominantly between -20 and -3 °C. These were dominated by super cooled liquid drops, with a median concentration of 113 cm-3. Droplet concentrations were relatively consistent during the campaign with an inter-quartile range of 86 cm-3. The exceptions to this were when the droplets were depleted by high ice concentrations and also flight 217 where anomalously high droplet concentrations were observed, which was associated with an enhanced aerosol layer below cloud. Similar to Arctic layer clouds (McFarquhar et al., 2007), liquid content and cloud drop effective radius both increased with distance from cloud base likely due to condensational growth. Collision coalescence may also have contributed to increase in effective radius. However, droplet number concentration was relatively invariant to position within the cloud."

24. Page 25 line 27: Can this not be estimated given that the aircraft was doing vertical profiles?

There isn't a line 27 on Page 25. Please could the reviewer clarify what this refers to?

Minor comments and technical corrections

1. Page 1 line 23: Additional clarification on what "key processes" means is required.

This refers to processes such as droplet activation, primary and secondary ice nucleation as well as the impact of dynamics and meteorology. To avoid the abstract becoming overly long these haven't been listed, but are discussed in the introduction and the remainder of the paper.

2. Page 1 line 27: Clarify that the size quoted is particle diameter.

Yes this is correct and clarified in the text.

3. Methods: It would seem more appropriate to introduce the NAME model in the methods section instead of in the results section.

This paragraph has been moved as suggested.

4. Page 6 line 22: I don't think the CAS part of a CAPS probe has anti-shatter tips. Please check. Also, was the CDP fitted with anti-shatter tips?

The CDP does not have anti-shatter tips. The leading edge of the CAS is fairly sharp and would have some anti-shatter properties, but we have removed the description of this as "anti-shatter" in the paper.

5. Are the aircraft altitudes relative to mean sea level?

Yes. This has been clarified in the text.

6. Page 6 line 28: Can you put a measure of uncertainty on your IWC estimate that results from assuming the Brown and Francis mass-diameter relation?

The Brown & Francis mass-diameter relation has been used by a number of previous mixed phase cloud studies (e.g. Lloyd et al., 2015, Crosier et al., 2011). However, several other M-D relationships exist. To estimate the uncertainty in Brown & Francis we re-calculate the IWC using the Heymsfield et al. (2004) M-D relationships. Differences between the two relationships are found to be approximately 20%.

Heymsfield, A. J., Bansemer, A., Schmitt, C., Twohy, C., and Poellot, M. R.: Effective ice particle densities derived from aircraft data, J. Atmos. Sci., 61, 982–1003, doi:10.1175/1520-0469(2004)061<0982:eipddf>2.0.co;2, 2004.

7. Page 8 line 5: The MI data is only shown in one small subset of data (Fig 8).

This line has been removed.

8. Figure 2: Is this a flight average or an in-cloud average PSD? Rather than showing one PSD, can you give a more quantitative measure of the agreement between instruments over all

flights e.g. compare drop number and LWC from CDP/CAS, IWC from 2DS/CIP, aerosol concentration from GRIMM/CAS. It is also worth describing why you choose certain probes in your analysis e.g. CAS appears to be used preferentially for LWC over the CDP.

This figure has been removed as suggested. The regression equations for all CAS/CDP and CIP/2DS measurements for the overlapping size ranges have now been included in the manuscript. They are as follows:

CDP = 0.87 × CAS + 1.7 $cm^{-3}$ ($R^2$ = 0.83) and CIP = 0.65 × 2DS + 0.7 $cm^{-3}$ ($R^2$ = 0.34)

A comparison between all CAS and GRIMM measurements can be found in Sect. 3.2. The CAS and 2DS were preferentially used over the CDP and CIP due to their higher resolution size bins.

9. Page 9 line 10 to 14: Suggest removing if data is not shown/discussed.

The paper describing these measurements is now in review in ACPD. This reference has been added to the paper. These results are also briefly presented in Section 3.2.

10. Figure 4: What is the point of figure 4b? Is it just to zoom in on the data closer to the surface? If so, why cut-off the x-axis at 0.3 when there are many data points above this as shown in figure 4a. Consider removing the lower panel.

The lower panel has been removed.

11. Figure 5: The continuous colorbar used does not adequately discriminate different flights. Suggest using an alternative way of plotting each flight or remove the colorbar.

The colour scale has been removed.

12. Page 12 line 10: The authors state the reason is not clear but then go on to say that higher aerosol concentrations were observed which could explain the higher drop numbers. Do you see any clear correlation between sub-cloud aerosol and cloud drop number across the different flights?

Cloud droplet number and below cloud CPC concentrations were not strongly correlated across the different flights ($R^2$ = 0.22).

13. Figure 10: The figure caption needs to include a lot more information.

A description of the image scale and times have been added to the caption.

14. Page 18 line 10: Replace 3:58 with 15:58 and 4:04 with 16:04 to be consistent with figure axis label.

Done.

15. Throughout the manuscript the authors use the abbreviation “ca”. Why not simply write “circa”? Or consider replacing with “approximately” or “about”, both of which are used in this context more extensively in the English language.

Done.

16. The authors switch between using cm-3 and scm-3 when referring to number concentrations of cloud and aerosol particles. Cloud drop number concentration data are shown in cm-3 (Figs 5, 6, 8, 12, 15), whereas aerosol number concentration data are mostly shown in scm-3 (Figs 11, 12, 14), except for CCN concentrations in Fig 13 which use cm-3. I would suggest being consistent in the use of units throughout andusing cm-3. It is the ambient aerosol number concentration on which the cloud drops form (not the value at STP) that is important for cloud drop number concentration for example.

When assessing whether any correlations exist between cloud droplet/ice concentrations, aerosol concentrations were converted from STP to concentrations at the same temperature and pressure as the cloud. In the figures we have left the concentrations in STP units so that comparisons can be more easily made with other studies in the literature.

Referee 2

The manuscript summarizes the characteristics of the Microphysics of Antarctic

Clouds, or MAC campaign conducted in 2015 over coastal Antarctica and the nearby ocean regions. The measurements, comprising cloud and aerosol retrievals from both ground-based and airborne measurements, provide a compelling source of information for this region, that will most likely be of high interest to the atmospheric modelling community for example. The Authors go further into analyzing the key features in the cloud and aerosol retrievals and the processes affecting them. Interesting features are revealed about the cloud ice mass fraction, ice particle types and the aerosol, even though conclusive explanations for many of the observed cloud features seem elusive. However, much of this can be rightfully attributed to the extremely challenging conditions as well as the so far quite low number of observations in this region. The descriptions of the instruments and retrieval techniques, as well as the presentation of data are for the most part adequate and very good.

My main concerns about the manuscript are related to the description of the surrounding conditions during these measurements. I do think the manuscript would greatly benefit from a bit more systematic description of the meteorological conditions as well as the structure of the clouds sampled during the campaign. This could potentially help with the interpretation of the results, many of which are now based simply on microphysical retrievals put together from all available flights. Below, I will try to summarize these points with more specific comments, followed by minor and technical comments.

1. The manuscript does not provide very detailed information about the height and depth of the sampled cloud layers. If there are considerable differences in the altitude and depth of the cloud layer, this would imply differences in the cloud dynamics as well as in the large-scale meteorological setting from one flight to the next, and would therefore be worth a look to support the subsequent analysis. It is also rather difficult to follow whether the analyzed data (in general but also in the few flight specific examples) represent the cloud base, in-cloud or cloud top conditions. For example, the vertical profile of ice mass fraction given in Fig 4 is interesting information, but it would make it far more interesting, if that data could be put in the context of sampling level with respect to the vertical extent of the sampled cloud.

A table (Table 1) has been added giving the temperature and height of cloud base/top. Figure 3 now shows how cloud properties vary relative to cloud base/top for single layer clouds. Please also see response to reviewer 1 comment 4 where this is discussed further.

2. Would it be possible to consider the existence of multiple cloud layers for all of the flights? It is commented in Section 4.1 that seeding effects were not detected apart from the frontal cloud case. However, could the possibly overlapping cloud layers affect the radiation budget of the sampled clouds, that might impact mixing and perhaps entrainment and thus the cloud properties?

Table 1 has been added to the manuscript and gives a description of whether multiple cloud layers were present. Discussing the cloud radiation budget in sufficient detail to be useful would likely make the paper overly long. It is anticipated that this will discussed in a separate paper where the radiation measurements will be compared with modelled values.

3. While the manuscript does consider the impact of airmass history on particle number concentrations in Section 3.5, it does not clearly outline how these shifts affect other aspects, such as large-scale meteorological forcing, which could impact the cloud properties. Generally, I think it would add great value to put these observations into context by including some more detailed information about the meteorological conditions during the campaign.

The information on the meteorological setting has been extended, we have included surface pressure charts and HYSPLIT back trajectories for each flight in the supplementary material.

A table has been added to the manuscript (Table 1) detailing cloud base/top height and temperatures. Also included is a description of whether multiple cloud layers were present.

4. Related to the above, it would be interesting to couple meteorological data (e.g. wind speed) with estimates of the cloud altitude (as per the comments no. 1) in order to estimate the potential of blowing snow to affect the measured cloud properties. This possibility is considered in Section 4.1 but explicitly only for one flight. A more detailed evaluation of the prevailing conditions would allow a broader consideration of at least the possibility of blowing snow contribution also for other flights.

The figure below shows histograms of the horizontal wind speed at different altitudes for measurements in cloud where ice is (blue lines) and is not (red lines) present. No strong relationship can be identified between the wind speed and presence of ice in the clouds. Though it should be noted that a high proportion of measurements are at wind speeds greater than 7 m $s^{-1}$, which is often considered a threshold for blowing snow.

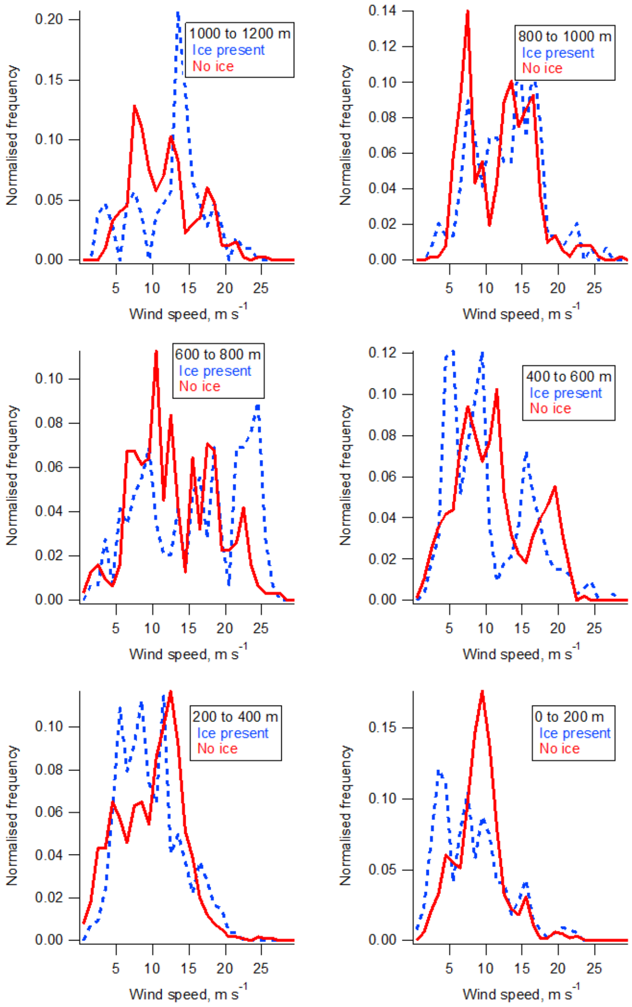



*Figure. Histograms of the horizontal wind speed at different altitudes for measurements in cloud where ice is (blue lines) and is not (red lines) present.*

The following text has been added to the manuscript discussing the relationship between surface type and the presence of ice precipitation:

"Evaluating the impact of these mechanisms during MAC is challenging since most of the in-cloud sampling was performed over snow covered sea ice, making it difficult to attribute local differences in the microphysics to the surface type. Figure 14 shows histograms of the surface albedo for out-of-cloud measurements (below 100 m) when there was (blue line) and was not (red line) ice observed. Here the surface albedo is used as a proxy for the surface type, since values near 0 correspond to overflying open water and the values near 1 correspond to a snow/ice covered surface. Figure 14 suggests that ice measured by the aircraft while out cloud almost exclusively occurred when overflying a snow/ice covered surface, implying a link between the surface type and the presence of ice in the clouds. The ice measured on the aircraft when out-of-cloud could either have originated from the surface or precipitated from clouds above. However, it should be noted that very few measurements were made over open water regions.

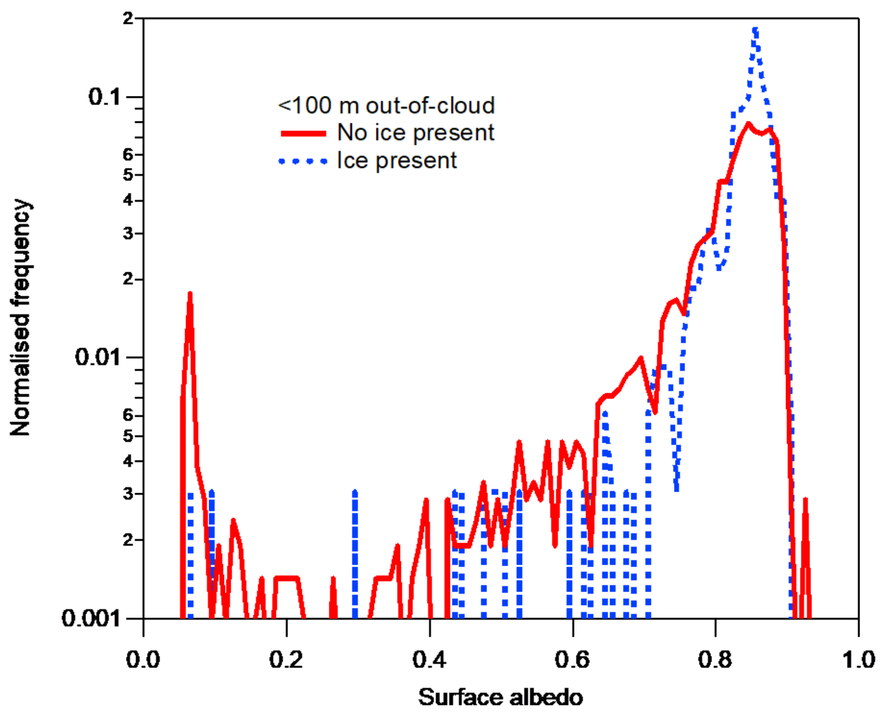



*Figure 14. Histograms of the surface albedo for of out-of-cloud measurements (below 100 m) when there was (blue line) and was not (red line) ice detected.”*

Other minor and technical comments

1. The notation scm-3 is used throughout the paper for the units of aerosol concentration, apparently referring to concentration defined in STP conditions, yet for cloud droplets the more commonplace cm-3 is used. I think the STP-related notation should be clearly defined and it should be made very clear where each of the notations are used to avoid confusion. However, I do think the best option by far would be to just use the same units (cm-3) for all concentrations.

We have clarified that this refers to standard temperature and pressure. In the figures we have left the concentrations in STP units so that comparisons can be more easily made with other studies in the literature.

2. Page 7, line 9: Please add a short definition for the circularity.

Done.

3. Fig 4, ”Radar altitude” vs ”altitude”, are the panels from different datasets? Please explain.

The radar altitude gives the height above the ground and “altitude” is above mean sea level (this is clarified in the text). However the trends are the same for both plots. For this reason, the radar altitude plot has been replaced with a plot showing normalised position within the cloud (please see our response to reviewer 1 comment 4)

4. Page 13 lines 24-27: Histogram of the spatial extent of ice: which figure should I be looking at here? If it is missing, please consider adding a figure showing this.

This figure has been added to the manuscript (Fig 4c).

5. Page 15, and fig 6: Why do you limit the analysis to the temperature range -8...-3 in this particular case? Please elaborate.

Please see our response to reviewer 1 comment 11, which answers this question.

6. The terminology regarding Fig 7 is confusing: page 16, line 9 ”particle size distributions”, page 17, line 2 ”droplet spectrum” and just ”size distributions” in the caption. Please try to use more consistent terminology here so it’s easier to read and to know exactly what you refer to.

“Particles size distribution” is now used throughout.

7. Fig 7: You don’t mention the dashed lines in the caption or elsewhere in the text. Please add an explanation for these or remove them from the figure.

Dashed lines show measurements from the CAS and solid lines are from the 2DS. This is now clarified in the text.

8. Page 17, line 5: Could you please add some numbers to make a more clear case for the droplet depletion?

Concentrations have been added to the manuscript.

9. Page 19: Please consider adding a dedicated subsection for the analysis of the cloud particle images. Coming directly after the quantitative results on ice fractions and number concentrations it feels a little out of place to me.

As suggested this has been given its own sub section.

# In situ measurements of cloud microphysics and aerosol over coastal Antarctica during the MAC campaign

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**Abstract**

During austral summer 2015 the Microphysics of Antarctic Clouds (MAC) field campaign collected unique and detailed airborne and ground based in situ measurements of cloud and aerosol properties over coastal Antarctica and the Weddell Sea. This paper presents the first results from the experiment and discusses the key processes important in this region, which is critical to predicting future climate change.

The sampling was predominantly of stratus cloud, at temperatures between -20 and 0 °C. These clouds were dominated by supercooled liquid water droplets, which had a median

concentration of 113 cm$^{-3}$ and an inter-quartile range of 86 cm$^{-3}$. Both cloud liquid water content and effective radius increased closer to cloud top. The cloud drop effective radius increased from 4 ± 2 µm near cloud base to 8 ± 3 µm near cloud top.

Cloud ice particle concentrations were highly variable with the ice tending to occur in small isolated patches. Below ca approximately 2000 m glaciated cloud regions were more common at higher temperatures; however the clouds were still predominantly liquid throughout. When ice was present at temperatures higher than -10 °C, secondary ice production most likely through the Hallet-Mossop mechanism lead to ice concentrations 1 to 3 orders of magnitude higher than the number predicted by commonly used primary ice nucleation parameterisations. The drivers of the ice crystal variability are investigated. No clear dependence on the droplet size distribution was found. However, higher ice concentrations were found in updrafts and downdrafts compared to quiescent regions. The source of first ice in the clouds remains uncertain, but may include contributions from biogenic particles, blowing snow or other surface ice production mechanisms.

The concentration of large aerosols (diameters 0.5 to 1.6 µm) decreased with altitude and were depleted in airmasses that originated over the Antarctic Continent compared to those more heavily influenced by the Southern Ocean and sea ice regions. The dominant aerosol in the region was hygroscopic in nature, with the hygroscopicity parameter, κ having a median value for the campaign of 0.64 (interquartile range = 0.34). This is consistent with other remote marine locations that are dominated by sea salt/sulphate.

# 1 Introduction

Antarctic clouds have a central role in the weather and climate at high southern latitudes (Lubin et al., 1998; Lawson and Gettelman, 2014). Through snow precipitation and their radiative effects they are key to the mass balance of the Antarctic ice sheet, which impacts on global sea levels (van den Broeke et al., 2011) and Southern Ocean circulation (Bromwich et al., 2012). In addition it has been suggested that changes in Antarctic clouds can influence weather patterns as far away as the tropics and even the extratropics of the Northern Hemisphere (Lubin et al., 1998).

Despite their importance Antarctic clouds are some of the least studied of any region around the globe (Bromwich et al., 2012). The remote location and harsh conditions cause significant logistical challenges for field projects in this region. As a consequence there is evidence that clouds and their radiative properties are poorly represented in weather and climate models over Antarctica (Bromwich et al., 2013; King et al., 2015; Listowski and Lachlan-Cope, 2017) and the Southern Ocean (Bodas-Salcedo et al., 2012; 2016).

Key uncertainties concern the aerosol in the region, in particular the number and sources of cloud condensation nuclei (CCN) and ice nucleating particles (INPs). Conventional parameterisations predicting INP concentrations have primarily been developed using measurements at mid-latitudes (e.g. Cooper, 1986; DeMott et al., 2010) and may not be appropriate for Antarctica. A number of intensive field campaigns have been conducted studying Arctic clouds (McFarquhar and Cober, 2004; McFarquhar et al., 2007; Verlinde et al., 2007; Lloyd et al., 2015a), however analogies between the polar regions may also not be appropriate. The Arctic receives significant anthropogenic aerosol input due to its proximity to industrial nations, and is therefore likely to have significantly different type and number of CCN/INP (Mauritsen et al., 2011; Lathem et al., 2013; Liu et al., 2015).

Previous, multi-year measurements of aerosol at the Neumayer coastal Antarctic research station had a median concentration of 258 $cm^{-3}$. Minimum values (less than 100 $cm^{-3}$) were typically observed in June/July, while concentrations increased in the austral summer to a maximum of approximately 1000 $cm^{-3}$ in March (Weller et al., 2011). In winter, aerosol number and mass were both dominated by sea salt particles (87% by mass, Weller et al., 2008). Although aerosol composition in summer is more variable, sea salt still accounts for a significant fraction (50% by mass) but now with a large contribution from non sea salt sulphate (27% by mass, Weller et al., 2008). Measurements at the coastal Antarctic station McMurdo show the persistent presence of sulphate aerosol throughout the year (Giordano et al., 2017). In the winter these particles are highly aged. Sulphate aerosol then increases through the austral spring/summer, due to enhanced emissions of dimethyl sulphide (DMS) and methanesulfonic acid (MSA) from phytoplankton in the Southern Ocean (Gibson et al., 1990; Giordano et al., 2017). Giordano et al. (2017) also report the presence of a sub-250 nm aerosol population of unknown composition during the winter to summer transition. In addition a study has observed a significant fraction of organic carbon (>10%) and lower contributions from sea salt (<10%) in summer marine Antarctic aerosol (Virkkula and Teinil, 2006). Measurements in the Antarctic have found that the aerosol is highly hygroscopic in marine airmasses (Mangold et al., 2017). While continental aerosol is less hygroscopic, which is consistent with a lower MSA fraction and the aging of marine organic components (Asmi et al., 2010).

To date, Antarctic INP measurements have mostly been made at surface sites. Measurements of snowflake residuals at the South Pole identified the long range transport of clays as the likely dominant source (Kumai, 1976). However, interpretation of these measurements is complicated due to secondary aerosol scavenging by the snowflakes and precipitation, meaning they contain particles in addition to the original nuclei. More recently, filter samples at the South Pole detected INPs that were active between -18 and -27°C, with concentrations of 1 $L^{-1}$ at -23 °C. Mineral dusts transported from the Patagonian deserts were identified as the likely source (Ardon-Dryer et al., 2011). A synthesis of INP measurements prior to 1988 from the high southern latitudes (> 60°S), found mean concentrations between $2x10^{-4}$ and 0.2 $L^{-1}$ at -15°C (Bigg, 1990). Given the general absence of other local INP sources, biogenic INPs may have a more important role in the Antarctic than in other regions. Biological species (pollen, bacteria, fungal spores and plankton) have been shown to act as INP at significantly higher temperatures than mineral dusts (> -15°C) (Möhler et al., 2007; Alpert et al., 2011; Murray et al., 2012; Amato et al., 2015; Wilson et al., 2015). However, Antarctic snowfall has been shown to be relatively depleted of biological INP (Christner et al., 2008) and bacteria commonly found in sea ice may not be effective INP (Junge and Swanson, 2007). The few in situ measurements of Antarctic clouds to date have suggested the importance of secondary ice processes (Grosvenor et al., 2012; Lachlan-Cope et al., 2016).

There is a clear need for more direct measurements to test and improve the representation of Antarctic clouds in climate/weather models. This paper presents both ground based and airborne measurements of cloud and aerosol properties during the 2015 Microphysics of Antarctic Clouds (MAC) field campaign aimed at addressing this. Section 2 provides an overview of the campaign and the measurement techniques used. Section 3 presents a statistical overview of the aerosol and cloud observations using all available measurements. Section 4 discusses the key microphysical processes. Conclusions are presented in Sect. 5.

# 2 Methods

## 2.1 Campaign and meteorological overview

The MAC experiment comprised both airborne and ground based measurements of cloud and aerosol properties. Ground based measurements were performed at the Clean Air Sector Laboratory (CASLab), which is located at the Halley research station. Halley is a coastal Antarctic base on the Brunt Ice shelf, approximately 30 km from the Weddell Sea (75.6° S, 26.7° W). The CASLab is located 1 km south of the main Halley buildings and receives minimal pollution from the base and vehicle traffic due to the prevailing easterly wind (Jones et al., 2008). All CASLab measurements were filtered using the wind direction to help remove any remaining influence from the base.

The airborne measurements were collected using the British Antarctic Survey's Twin Otter MASIN research aircraft (King et al., 2008). Twenty-four flights (a total of 80 hours) were performed during November and December 2015 from Halley. These flights have the nominal flight numbers 212 to 235. The flights were predominantly performed over the Weddell Sea (see Fig. 1), which at this time and location was covered by a mixture of broken sea ice and polynyas. This is shown in Fig. 1 together with the sea ice fraction (Maslanik and Stroeve, 1999). from the Nimbus-7 Multichannel Microwave Radiometer (SMMR) and Defense Meteorological Satellite Program (DMSP) SSM/I-SSMIS passive microwave data (Cavalieri et al., 1996.) One flight sampled clouds in-land over the Antarctic continent (Flight 233). In addition a transit took place from Rothera research station on the Antarctic Peninsula (Flights 212 to 215); however not all instruments were available during these transit flights. Since the aircraft was not pressurised, the measurements were restricted to altitudes below approximately 4000 m. As a consequence, the majority of clouds were sampled over the temperature range -11 and -3 °C (79%). Seventeen percent of in-cloud measurements were collected at temperatures below -11 °C and 4% at temperatures higher than -3 °C. In total 17 hours of sampling during the campaign was performed in-cloud.

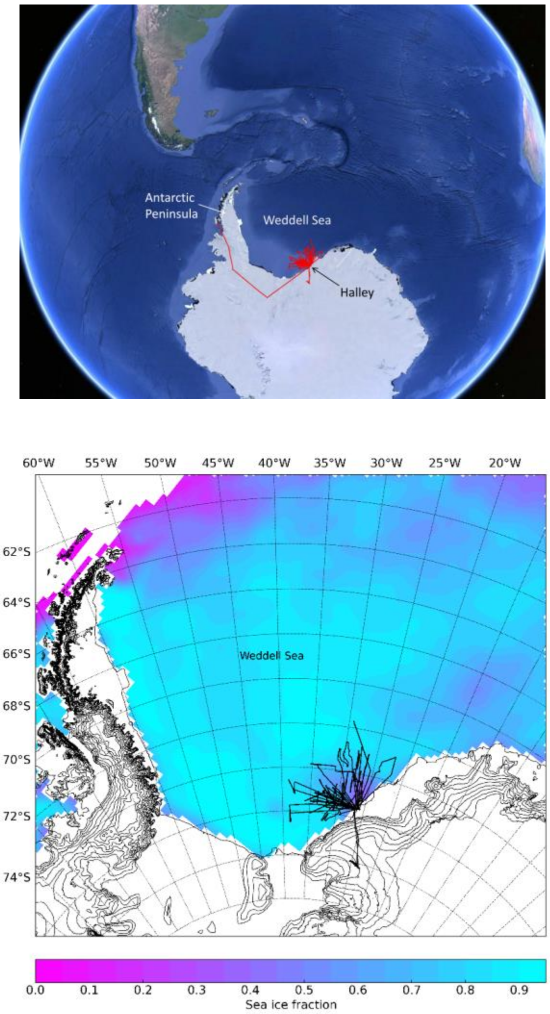


*Figure 1. Top panel: Flight tracks during the MAC field project (source Google Earth). Lower panel: shows the sea ice fraction on the Weddell Sea* (Maslanik and Stroeve, 1999)~~*(Cavalieri et al., 1996)*~~ *during the experimental period.*

The clouds sampled were generally stratiform, with strong temperature inversions at cloud top. The exception to this was Flight 224, which sampled frontal clouds. Table 1 shows the altitude and temperature of cloud base/–top for each flight. If multiple layers were present, unless otherwise noted, the height and temperatures given are of the layer where the majority of sampling took place. To show the meteorological setting for the campaign Figures S1 to S20 show surface pressure charts from the ERA-Interim reanalysis (at 12 UTC on the given day, Dee et al., 2011) and HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory, Stein et al., 2015) back trajectories for each flight. Back trajectory analysis showed that two broad regimes were present during the project. The earlier flights (up to Flight 223) generally sampled airmasses that had travelled south over the Southern Ocean and Weddell Sea. Later in the campaign there was a transition to airmasses with greater influence from the Antarctic continent.

| Number | Date | Base altitude (m) | Top altitude (m) | Base temperature (°C) | Top temperature (°C) | Comment |
|---|---|---|---|---|---|---|
| 216 | 21/11/2015 | 261 (246-283) | 951 (925-983) | -12.1 | -14.1 | Multiple layers. |
| 217 | 24/11/2015 | 330 (296-366) | 662 (621-700) | -9.8 | -12.3 | Multiple layers. |
| 218 | 27/11/2015 | 312 (298-327) | 554 (539-569) | -4.8 | -6.1 | Main layer with broken layers above. |
| 219 | 27/11/2015 | 375 (316-441) | 870 (847-890) | -4.7 | -7.8 | Single layer. |
| 220 | 28/11/2015 | 1143 (1129-1154) | 1303 (1289-1317) | -12.9 | -13.2 | Single layer. |
| 221 | 29/11/2015 | 157 (124-202) | 530 (499-564) | -6.0 | -6.6 | Single layer with high cloud above (3000 m). |
| 222 | 30/11/2015 | 170 (151-201) | 603 (573-635) | -6.8 | -8.5 | Predominately single layer, partial layer above. |
| 223 | 03/12/2015 | 262 (247-277) | 745 (712-771) | -7.1 | -9.5 | Multiple layers. |
| 224 | 06/12/2015 | 1056 (1022-1090) | 4278 (4253-4300) | -7.6 | -18.9 | Frontal cloud multiple layers. Cloud top not sampled. Height and temperature ranges are for all layers sampled. |
| 225 | 07/12/2015 | 694 (680-718) | 1010 (944-1066) | -5.0 | -5.7 | Single layer with high cloud above (4000 m). |
| 226 | 07/12/2015 | 1273 (1230-1319) | 1866 (1853-1873) | -5.4 | -6.8 | Single layer with high cloud above (4000 m). |

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|---|---|---|---|---|---|---|
| 227 | 08/12/2015 | 88 (68-107) | 417 (372-455) | -5.8 | -6.9 | Single layer. |
| 228 | 09/12/2015 | 76 (50-122) | 528 (493-567) | -6.7 | -5.9 | Single layer. 2nd partial layer at 1500m. |
| 229 | 09/12/2015 | | | | | No cloud sampled. |
| 230 | 10/12/2015 | 334 (304-362) | 574 (558-588) | -4.6 | -6.5 | Single layer. |
| 231 | 11/12/2015 | 293 (279-321) | 1171 (1158-1186) | -4.6 | -8.3 | Predominantly ~~S~~single layer~~-~~, partial layer above. |
| 232 | 11/12/2015 | 554 (516-601) | 1126 (1108-1148) | -6.3 | -10.1 | Single layer with high cloud above. |
| 233 | 12/12/2015 | 1630 (1600-1667) | 1857 (1852-1861) | -14.1 | -15.4 | Single broken layer. |
| 234 | 13/12/2015 | 409 (387-428) | 710 (700-720) | -5.9 | -7.1 | Lower layer. |
| | | 1489 (1479-1499) | 1785 (1764-1804) | -13.6 | -13.7 | Higher layer not directly above lower level. |
| 235 | 14/12/2015 | 954 (929-979) | 1432 (1404-1461) | -9.9 | -13.9 | Main layer sampled with broken layers below. |

Table 1. The height and temperature of cloud base and top for each flight. The range of altitudes in brackets are an estimate of the uncertainty in the cloud heights due to a combination of variability in the cloud and incomplete sampling. If multiple layers were present, unless noted otherwise, the height and temperatures given are for the main cloud layer sampled.

## 2.2 Aircraft

During MAC the Twin Otter MASIN research aircraft was fitted with a range of in situ aerosol and cloud microphysical instrumentation. Cloud particle size distributions were derived using the images from two optical array probes (OAP): a 2DS (2D-stereo, SPEC Inc., USA, see Lawson et al., 2006) with a nominal size range of 10 to 1280 µm (10 µm pixel resolution) and a CIP-25 (Cloud Imaging Probe, DMT Inc., USA, Baumgardner et al., 2001) with a size range of 25 to 1600 µm (25 µm pixel resolution). The 2DS was not operated on the flights before Flight 218.

Particle size distributions over the size range from 0.5 to 50 µm were recorded using a Cloud Aerosol Spectrometer (CAS, DMT Inc., USA, Baumgardner et al., 2001). The CAS sizing was calibrated by the manufacturer using polystyrene latex (PSL) spheres (< 2 µm) and glass beads (> 2 µm) (Baumgardner et al., 2014). During MAC the sizing of the CAS's larger bins (>10 µm) was also validated using reference glass calibration beads and show little instrument drift (see Fig 2.).

The aircraft was also fitted with a Cloud Droplet Probe (CDP-100, DMT Inc.) for observing cloud droplets between 3 and 50 µm (Lance et al., 2010). Following the method detailed by Rosenberg et al. (2012), glass beads were used to determine the CDP's size bin centres and widths. The 2DS and ~~,~~ CIP-25 ~~and CAS~~ were fitted with anti-shatter tips to minimise ice break-up on their leading edges (Korolev et al., 2011). For full details of the data processing and quality control of the 2DS and CIP-25 measurements see Crosier et al. (2011) and Taylor et al. (2016). It should be noted that in addition to the use of anti-shatter tips, an inter-arrival time algorithm was used to further reduce shattering artefacts on the 2DS and CIP-25 datasets. Ice mass content was determined from the 2DS and CIP-25 images using the Brown and Francis (1995) mass-diameter relationship. ~~As an example Fig. 2 shows a comparison between the CDP, CAS, 2DS, and CIP-25 size distributions for Flight 227.~~ Unless stated otherwise all flight data presented has been averaged to 10 second intervals. A linear fit to the number concentrations derived by the CDP and CAS where their size ranges overlap has equation CDP = 0.87 × CAS + 1.7 $cm^{-3}$ ($R^2$ = 0.83). Similarly, the regression equation for the CIP and 2DS is CIP = 0.65 × 2DS + 0.7 $cm^{-3}$ ($R^2$ = 0.34).

~~*Figure 2. Average size distribution for Flight 227 comparing the 2DS, CIP-25 CDP and CAS probes. The CAS and CDP shows the Flight 227 size distributions using results from the bead calibrations performed during the campaign in order to monitor instrument performance.*~~

Following Crosier et al. (2011), 2DS and CIP-25 images were classified based on a geometric analysis of their circularity, C:

$$C = \frac{P^2}{4\pi A}$$

Equation 1

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~~.~~ where P is the particles perimeter and A is its area. Particles containing less than 50 pixels (equivalent to a diameter of approximately 80 µm for the 2DS and 200 µm for the CIP-25) were not classified since they contain insufficient pixels to accurately determine their shape. Particles with circularity values less than 1.2 were classified as low irregular (LI) and are indicative of liquid drops. Circularity values greater than 1.4 are associated with ice crystals and are classified as high irregular (HI). Visual inspection of the LI and HI images confirmed that they were almost all liquid droplets and ice crystals, respectively. Circularities between 1.2 and 1.4 are classified as medium irregular (MI). Interpretation of the MI category with respect to the particle phase is more ambiguous than the other categories. In general, the MI images were of quasi-spherical ice crystals, such as recently frozen drops, however they may also include some poorly imaged liquid drops that should be classified as LI. During MAC the concentration of MI particles was generally significantly less than HI particles. The mean ratio HI:MI for the campaign was 7 (see also Sect. 3.1). This suggests that the HI concentration is likely a good proxy for the ice crystal concentration. ~~However to highlight the uncertainty in the phase separation, in Sect. 3 the MI concentration is also shown along with the HI concentration.~~

Aerosol instrumentation on the aircraft included a GRIMM optical particle counter (GRIMM Model 1.109) capable of detecting aerosol particles over the size range from 0.25 to 32 µm. The GRIMM sampled through a Brechtel Model 1200 isokinetic aerosol inlet with a >95% sampling efficiency for particles in the size range 0.01µm to 6 µm. Inlet losses only become significant for particles >6 µm and here we only consider the concentration of particles below 2µm. Total aerosol concentrations of particles >10 nm in size were determined using a Condensation Particle Counter (CPC, TSI Inc. Model 3772).

The aircraft was also fitted with instrumentation to measure temperature, turbulence, humidity, radiation and surface temperature. See King et al. (2008) for full details.

### 2.3 Ground site measurements

Aerosol instrumentation was installed at the CASLab sampling from its central aerosol stack (Jones et al., 2008) for the measurement period from 27 November 2015 to 15 December 2015. A Scanning Mobility Particle Sizer (SMPS, TSI) was used to generate a quasi-monodisperse aerosol flow. The SMPS performed 27 discrete steps over the aerosol size range from 30 to 500 nm. Downstream of the SMPS the flow (1 $L^{-1}$) was split isokinetically between a cloud condensation nuclei counter (CCNc, Droplet Measurement Technology Model CCN-100) and a condensation particle counter (CPC, TSI). The CCN concentration was measured at super saturations of 0.05%, 0.13%, 0.20%, 0.26% and 0.34%. The activated cloud droplet fraction was determined by the ratio of activated particles from the CCN to the total number of particles measured by the CPC. The dry diameter at which 50% of particles were activated ($D_{50}$) was determined by fitting a sigmoid curve to the activated fraction size spectrum (Whitehead et al., 2016). The total CCN concentration was determined by integrating the concentration of particles larger than $D_{50}$. The hygroscopicity parameter κ was derived from κ-Köhler theory using the $D_{50}$ and supersaturation values (Petters and Kreidenweis, 2007).

The SMPS and CCNc were calibrated at the beginning and end of the campaign (Good et al., 2010). The SMPS was size calibrated using NIST traceable polystyrene latex spheres (PSLs). Ammonium sulphate and sodium chloride were used to calibrate the CCNc supersaturations, by comparing measured values to theoretical ones from the Aerosol Diameter Dependent Equilibrium Model (ADDEM) (Topping et al., 2005).

Additional measurements were provided by an Aerodynamic Particle Sizer (TSI Model 3321) which provided aerodynamic particle size concentration measurements over the size range $0.5<D<20$ µm and in the size range $0.3<D<20$ µm from simultaneous aerosol scattering cross section measurements. Total aerosol concentrations (D >10 nm) were determined using a Condensation Particle Counter (CPC, TSI Inc. Model 3776).

Continuous measurements of airborne bio-fluorescent particle concentrations (primary biological and mixed biological and non-biological) were also made at CASLab using a Wideband Integrated Bioaerosol Spectrometer (WIBS Model Dstl-3)~~, Gabey et al. 2010, Crawford et al. 2014, 2015)~~. Measurements from this instrument ~~will be~~ are described in detail in ~~a separate paper~~Crawford et al. (2017).

## 2.4 Numerical Atmospheric Dispersion Modelling Environment (NAME)

To examine how aerosol and cloud properties vary with airmass history we perform back trajectory analysis using the UK Met. Office's NAME model (Numerical Atmospheric Dispersion Modelling Environment) (Jones et al., 2007) using Met Office Unified Model (UM) meteorological fields. Five-day retroplumes were determined by releasing 10000 particles in the model at locations coincident with the aircraft's position. Here we examine the relative sensitivity to surface emissions from the following regions; the Antarctic continent, sea ice, Southern Ocean, ice-shelf and South America. The numbers of particles near the surface (0 to 100 m) over each geographic region was summed every 15 minutes as the particles were dispersed five-days backwards in time. For each region, the time integration of particles over the region was divided by the total number of particles appearing in the whole domain to determine fractional contributions (see Fleming et al., 2012). Shape files representing the monthly averaged sea ice extent from Polarview and geographical contour files for the Antarctic plateau, the permanent sea ice (ice shelves and permanent sea ice) and the American continent were used to determine the passageway of the air masses at surface levels sampled by the aircraft. This analysis was repeated for particles released at 60s intervals along the flight track to determine a time series of contributions from each geographic region.

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# 3 Results

## 3.1 Cloud microphysics

The following section presents a broad overview of the microphysical measurements during the MAC field campaign. For this analysis "in-cloud" measurements were determined as periods when the liquid water content (LWC) was greater than 0.01 g $m^{-3}$ or when particles were detected by the 2DS. Flight 224 is excluded from this bulk analysis since this flight sampled frontal cloud, while the other flights sampled shallow layer cloud. The ice mass fraction (IMF) is calculated as the ratio of the ice mass to the total condensed water. Here the ice mass is taken as the sum of the HI and MI 2DS categories, while the liquid mass is taken as the sum of the CAS droplets (>3 μm) and the 2DS LI category. Ice mass fractions of 0 and 1 represent fully liquid and glaciated conditions, respectively. Figure ~~3~~ 2 (black line) shows the frequency distribution of ice mass fraction based on all 1 Hz measurements in layer clouds sampled during MAC. As can be seen in Fig. ~~3~~ 2 the clouds were dominated by liquid water. Ice mass fractions between 0 and 0.1 were observed 90% of the time, while only 6% of cases had values between 0.9 and 1. Figure ~~4~~ 3 shows the ice mass fraction as a function of height~~, the black line shows the mean for each altitude bin~~. For altitudes below ca. 2000 m (all altitudes given are meters above mean sea level) there is a general trend of glaciated conditions becoming more prevalent with decreasing altitude (and increasing temperature). At temperatures higher than -3 °C glaciated conditions (IMF greater than 0.9) were responsible for 15% of observations, compared to 7% at temperatures between -8 and -3 °C. Above 2000m glaciated regions become more frequent with increasing altitude, however this is based on comparatively few observations.

Figure 3b, shows ice mass fraction measurements in single layer clouds as a function of the normalised position within the cloud, Zn.

$$Zn = \frac{Z - Z_B}{Z_T - Z_B},$$

Equation 2

where Z is the altitude, $Z_B$ and $Z_T$ are cloud base and cloud top altitude, respectively. We note that there is some uncertainty in determining cloud base/top due variability in the cloud and also incomplete sampling (this uncertainty is estimated in Table 1). The clouds were dominated by liquid drops throughout, while ice was more prevalent lower in the clouds. The relationship between ice mass fraction (IMF) and Zn over the range 0<Zn<1 can be approximated by the equation:

$$IMF = 0.177 + 0.360Zn + 0.244Zn^2$$

Equation 3

This is shown as a red line in Fig. 3b. Figure 3c and d show that both liquid water content and cloud drop effective radius increased closer to cloud top. The effective radius increased from 4 ± 2 µm near cloud base to 8 ± 3 µm near cloud top.

Measurements in Arctic stratus/stratocumulus generally find these clouds to be similarly dominated by liquid drops (McFarquhar and Cober, 2004; McFarquhar et al., 2007; Lloyd et al., 2015a). A polynomial relationship derived during the Mixed-Phase Arctic Cloud Experiment (M-PACE) is shown as a blue line in Fig. 3b (McFarquhar et al., 2007). McFarquhar et al. (2007) show a trend of increasing IMF with increasing distance from cloud top (and increasing temperature). Glaciated conditions were observed during 23% of their measurements. This is significantly more than during MAC, possibly due to lower INP concentrations available for primary ice development in the Antarctic compared to the Arctic, but differing sampling strategies may also contribute to this difference.

Flight 224 sampled cloud layers at the rear of an occluded front that was associated with a low pressure system north of Halley. Several layers were observed between -19 °C and -1 °C with ice crystals precipitating between the layers. As shown in Fig. ~~3~~ 2 (red line) ice was more frequently observed in these clouds than during the flights where stratocumulus/stratus clouds were sampled. Twenty-four percent of measurements had ice mass fractions between 0.9 and 1, while 32% of observed ice mass fraction values were between 0.1 and 0.9. Droplet number concentrations were comparatively low with a mean of 40 (29 at 1σ) $cm^{-3}$.

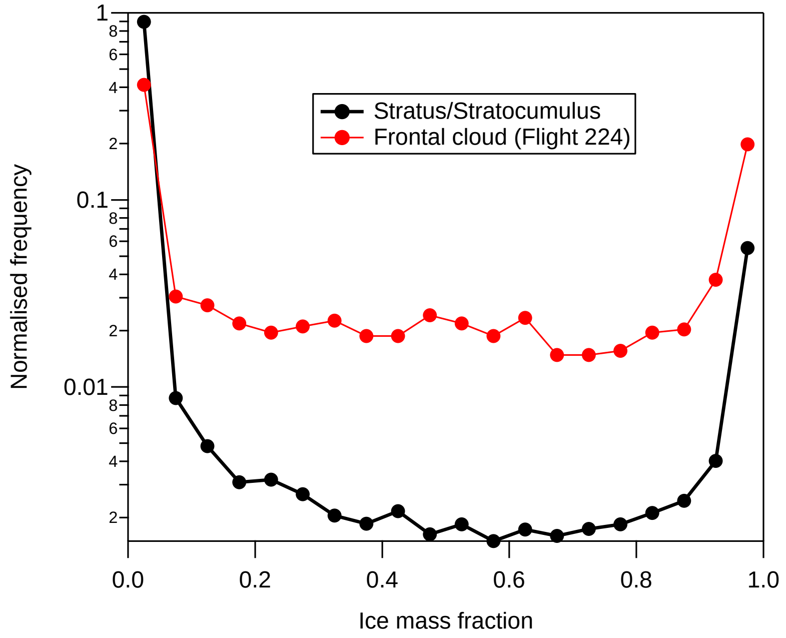


*Figure 32. Frequency distribution of the 1 Hz cloud ice mass fraction measurements.*

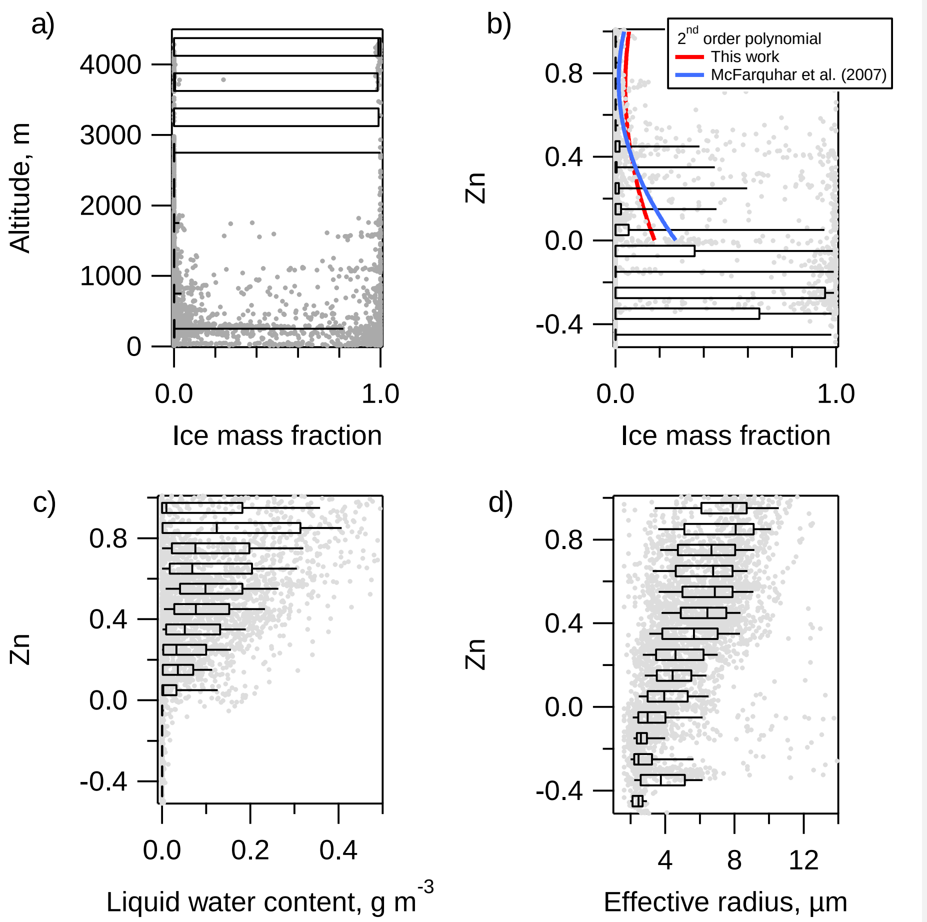


*Figure ~~4a~~3a. Ice mass fraction as a function of altitude and b)~~, black lines show the average ice mass fraction for each altitude bin.~~ normalised position within the cloud (Zn). c) and d) show similar plots for liquid water content and effective radius from the CAS probe. Boxes are the 25th and 75th percentiles, the whiskers are the 10th and 90$^{th}$ percentiles.*

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The droplet number concentration as a function of temperature is shown in Fig. ~~5a~~4a. This was found to be relatively consistent and temperature independent during the campaign with a median of 113 cm$^{-3}$ and an inter-quartile range of 86 cm$^{-3}$. An exception to this is Flight 217, when anomalously high droplet concentrations were observed at -23 °C (mean 310 cm$^{-3}$). The 2DS was not available during this flight but the CIP observations suggest that ice was not present in this cloud. The reason for the enhanced droplet concentrations is not clear, however the aerosol concentrations below the cloud layer was similarly elevated with the CPC recording concentrations of over 1200 scm$^{-3}$, compared to the median for the campaign of 408 scm$^{-3}$. Back trajectory analysis showed that in the previous days this airmass travelled over the Southern Ocean from South America.

The cloud droplet concentrations during MAC are found to be comparable with previous observations from the Antarctic Peninsula (Lachlan-Cope et al., 2016) and also Arctic summer stratocumulus (Lloyd et al., 2015a). Droplet concentrations over the Antarctic Peninsula varied between 60 and 200 cm$^{-3}$ (Lachlan-Cope et al., 2016). Concentrations on the eastern side of the Peninsula were moderately higher than on the west, which may be due to the greater sea ice coverage on the eastern side. It has been suggested that sea ice may provide a more efficient source of sea-salt aerosol, and therefore CCN, than open waters (Yang et al., 2008). Recent measurements and modelling found that sea ice made a significant contribution to the winter sea-salt aerosol loading at coastal (Dumont d'Urville) and central (Concordia) East Antarctic sites (Legrand et al., 2016).

The number of highly irregular particles observed by the 2DS/ CIP-25 can be used as a proxy for the number of ice crystals; it this is shown as a function of temperature in Fig. 5b4b. Box and whisker plots show statistics for those regions of the cloud where ice is present (i.e. excluding regions with only liquid cloud water). The 2DS was not operated during the flights previous to flight 218 so measurements are only available at temperatures higher than -20 °C. The two lowest temperature bins -21 to -15 °C in Fig. 5b 4b show the highest concentration of ice crystals. However these measurements come from only one flight (Flight 226) where the base (4000 m) of high cloud was sampled. These crystals (predominantly rosettes and aggregates) are highly likely to have been nucleated at lower temperatures higher up in the cloud which then sedimented down to be sampled by the aircraft. At temperatures greater thanAbove -15 °C there is a trend of the ice crystal concentrations showing greater variability and higher median concentrations with increasing temperature. Ice in the clouds tended to occur in small patches. A histogram of the spatial extent of ice patches shows that they increase in frequency with decreasing length up to the maximum resolvable by the 2DS measurements (a sampling frequency of 10s corresponds to a spatial scale of ~~ca~~ about 600m, Figure ~~5~~4c).

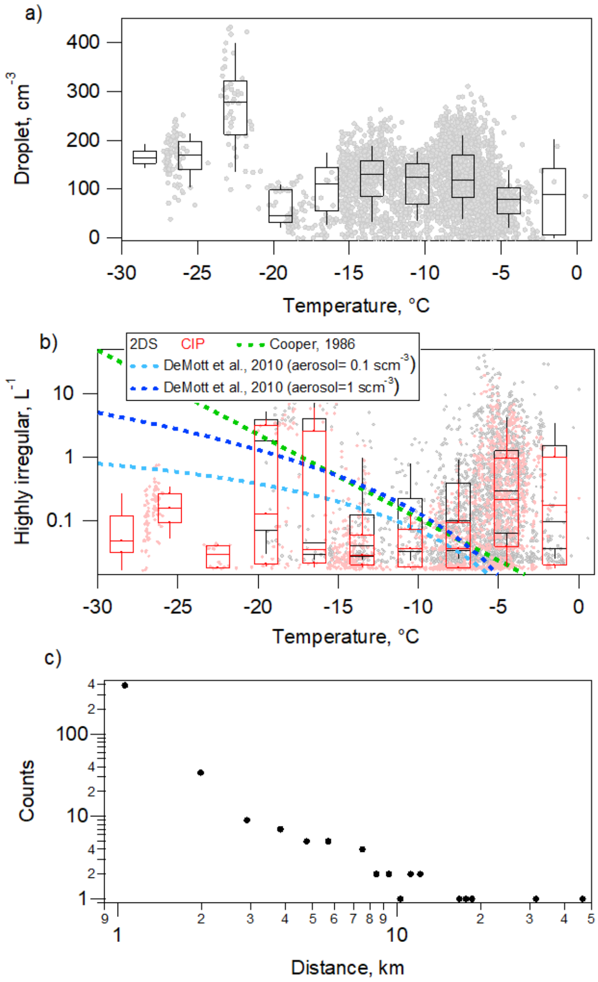


*Figure ~~5~~4. Box and whisker plots summarising in cloud measurements (averaged over 10 s) as a function of temperature. Plate a) shows the concentration of cloud droplets (cm$^{-3}$), measured by CAS, while b) shows the concentration of ice particles measured by 2DS and CIP-25, based on those classified as highly irregular (see text for details). The concentration* *of ice nucleating particles predicted by the DeMott et al. (2010) parameterisation with a high (1 scm$^{3}$) and low (0.1 scm$^{3}$) aerosol input are shown as dark and light blue lines, respectively in b). The ~~red~~ green line is the predicted ice particle concentration according to the Cooper (1986) parameterisation. c) a histogram of the flight distance while continuously sampling ice.*

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Previous observations of Arctic mixed phase clouds found that the presence of precipitating ice particles (> 400 µm) was associated with the number of large drops (>30 µm), however the precise nucleation mechanism through which this occurs is uncertain (Lance et al., 2011). To identify if a similar relationship was present during MAC Fig. ~~6a~~5a,b shows the relationship between the 2DS HI and the 2DS LI particles (droplets larger than approximately 80 µm). ~~over the temperature range -8 to -3 °C~~. Figures ~~6b~~~~6~~5c,d and ~~6c~~ ~~6~~5e,f show similar plots for the CAS measurements of droplets larger than 30 and 20 µm, respectively. Panels on the left (a, c and e) show measurements at temperatures lower than -8°C and panels on the right (b, d and f) show those in the range -8 to 0 °C. The HI concentrations are binned based on the droplet concentration and the 25, 50 and 75 percentiles are shown as black lines. When examining statistics for all stratus flights we find no evidence that the ice concentrations increase due to the presence of large drops. However, any relationship may be obscured as drops are depleted by ice crystal growth through riming and the Wegener-Bergeron-Findeisen process.

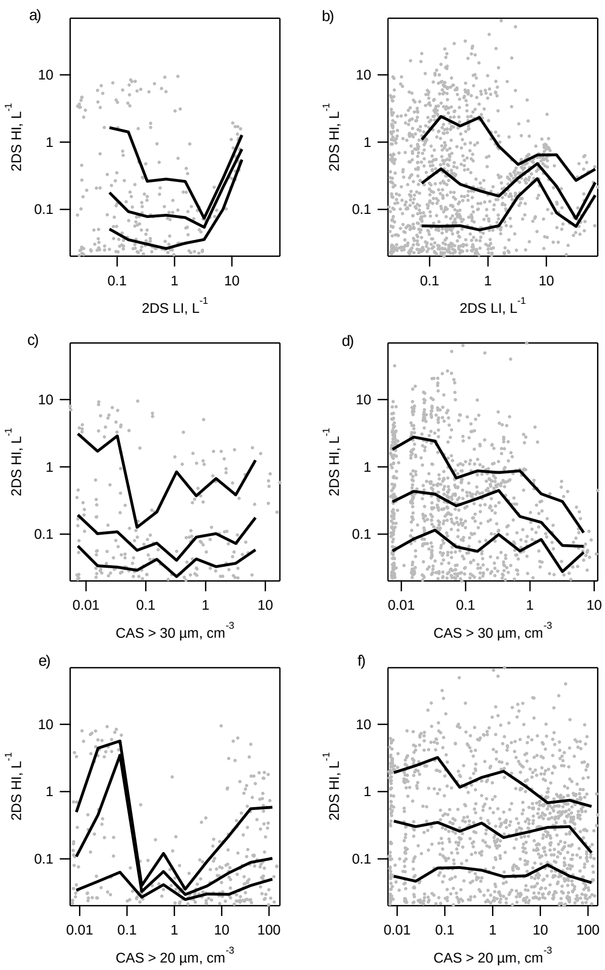


*Figure ~~6a~~5a,b. The relationship between the concentration of highly irregular (2DS HI) particles and low irregular particles (2DS LI) (low irregular particles greater than approximately 80 µm) ~~for the temperature range -8 to -3 °C~~. Figures ~~6b~~ 6c,d and ~~6c~~ 6e,f show the relationship with the concentration of droplets larger than 30 and 20 µm, respectively. Panels on the left (a, c and e) show measurements at temperatures lower than -8°C and panels on the right (b, d and f) show those in the range -8 to 0 °C. The black lines are the 25th, 50th and 75th percentile of the 2DS HI concentration for each droplet concentration bin.*

Similar results are found when case studies for individual flights are examined. Figure 7a 6a shows a comparison between the particle size distributions for three periods with quite different degrees of glaciation during a constant altitude run at -5 °C during Flight 218. Time series of the microphysical properties during this run are shown in Fig. 87. During this run there were patches of ice with concentrations of several per litre and regions where no ice was present. However, there are no distinct differences in the droplet spectrumparticle size distributions for particles <100 µm for these three cases. Figure 7b 6b shows a similar plot for a constant altitude run at -6 °C during Flight 219. During times with very high ice concentrations (2DS HI up to 50L$^{-1}$, blue line) the droplets (10s minimum of 11 cm$^{-3}$) are depleted compared to the cases when the 2DS HI concentration was 1 L$^{-1}$ and 0 L$^{-1}$ (approximately 100 cm$^{-3}$).

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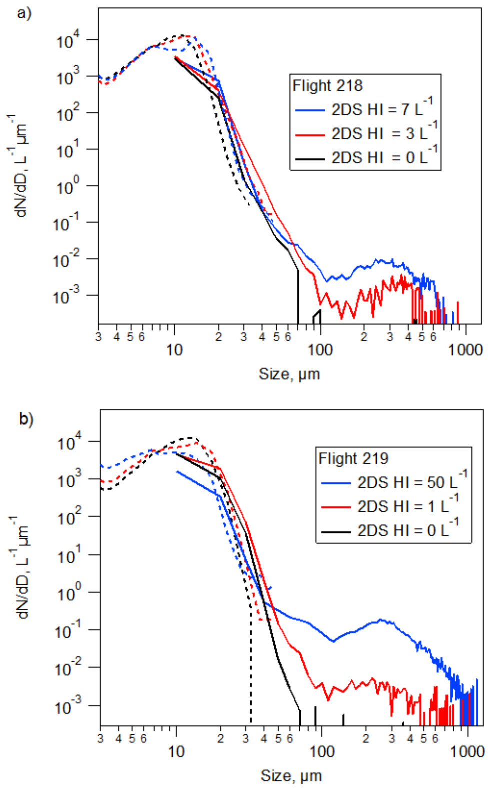


*Figure 7a6a. Comparison between the particle size distributions for 3 regions sampled in the constant altitude run at -5 °C during Flight 218, these are where the concentration of highly irregular particles (2DS HI) was 7 $L^{-1}$ (416:04 GMT), 3 $L^{-1}$ (315:58 GMT) and 0 $L^{-1}$ (315:52 GMT). Time series of the microphysical measurements during this run are shown in Figure 8. Figure 7b shows a similar plot for a run at -6 °C during Flight 219 when the 2DS highly irregular concentration was 50 $L^{-1}$, 1 $L^{-1}$ and 0 $L^{-1}$. Dashed lines show measurements from the CAS and solid lines are from the 2DS.*

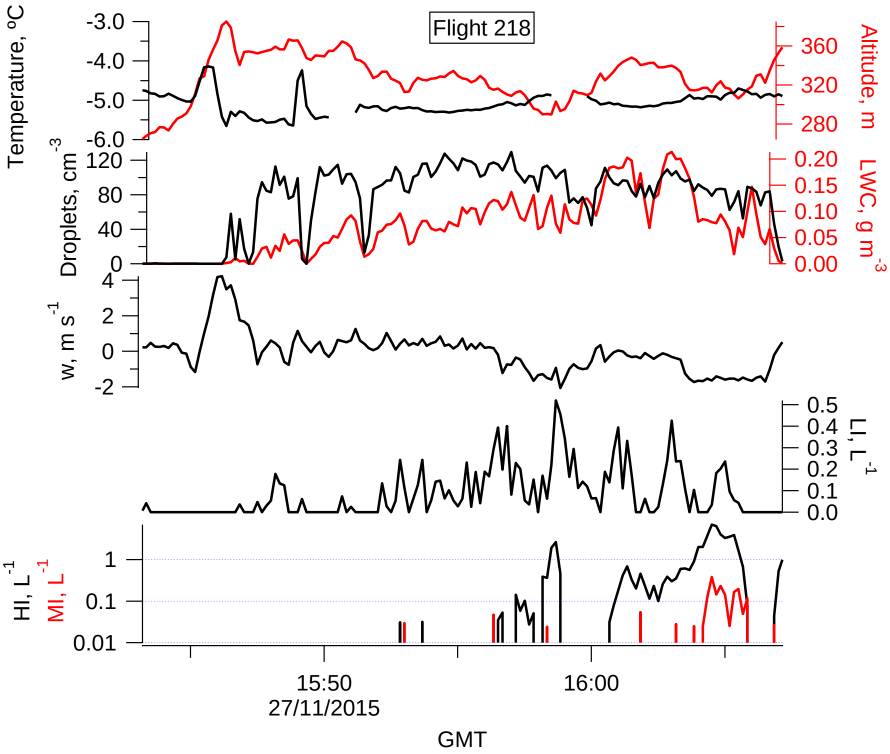


*Figure ~~8~~7. Time series of microphysical parameters during a constant altitude run at -5°C (400 m) during flight 218.*

~~During MAC there was a trend towards higher ice concentrations in both updrafts and downdrafts compared to quiescent regions of the clouds (see Fig. 9 for measurements during constant altitude runs). Previous measurements have observed secondary ice production in convective regions of mid-latitude stratus (Crosier et al., 2011). The run during Flight 218 at -5 °C (see Fig. 8) is an example of this where the two peaks at 3:58 (2DS HI maximum = 3 L$^{-1}$) and 4:04 (2DS HI maximum = 7 L$^{-1}$) in ice concentration occur in downdrafts of approximately 1 m s$^{-1}$. In contrast a similar run during Flight 219 (Fig. 7b) showed glaciated regions not to be associated with vertical motion.~~

~~*Figure 9. Box and whisker plots summarising the 1 Hz concentration of highly irregular particles (2DS HI) as a function of vertical velocity. Higher concentrations are observed in updrafts/downdrafts compared to quiescent regions.*~~

### 3.1.1 Ice Crystal Images

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Inspection of the cloud particle images shows that at temperatures higher than -10 °C columnar crystals appear as the dominant ice crystal habit, with irregular rimed crystals also widespread. This is illustrated by Fig. ~~10a~~ 8a showing example images from Flight 218 at -5 °C. Measurements in Arctic clouds at similar temperatures show that they are similarly dominated by columnar crystals (Lloyd et al., 2015a). Figure ~~10b~~8b. shows images at -15 °C collected in a single layer cloud over the Antarctic continent, approximately 300 km south of Halley (Flight 233). This cloud had some columns/needles, but also a high proportion of plates and stellar crystals. At the lowest sampled temperatures of – 20 °C (Fig. ~~10c~~8c, Flight 226) the ice mostly consists of rosettes and irregular crystals, which may be aggregates. However, measurements at these low temperatures were relatively infrequent, and the ice may have been nucleated at lower temperatures higher in the cloud.

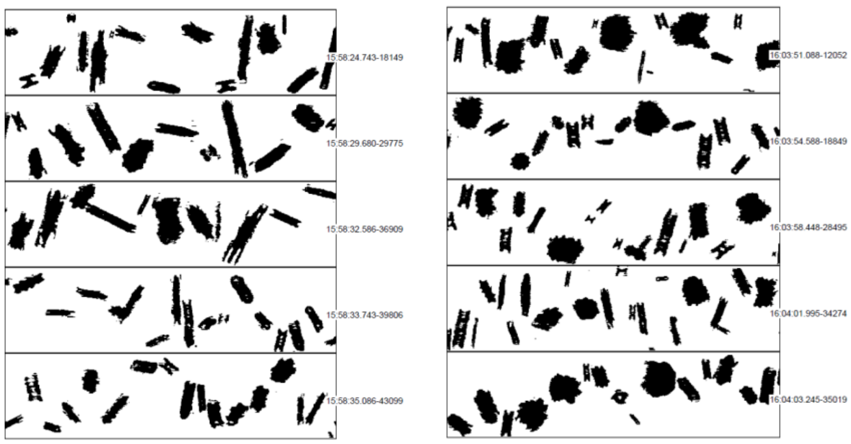


*Figure ~~10a~~8a. 2DS Images of highly irregular particles during a constant altitude run at -5°C (400 m) during flight 218.The times given are for the first crystal on each strip. The height of each strip corresponds to the 2DS array width of 1280 µm.*

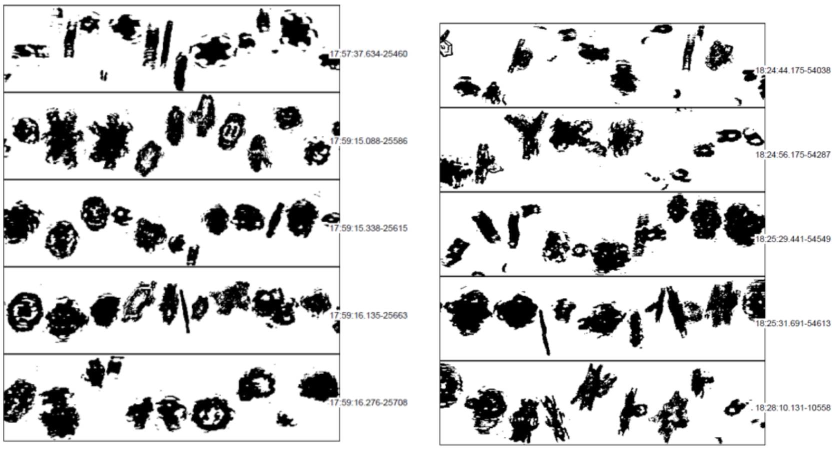

*Figure ~~10b~~8b. 2DS Images of highly irregular particles during a constant altitude run at -15°C during flight 233.*

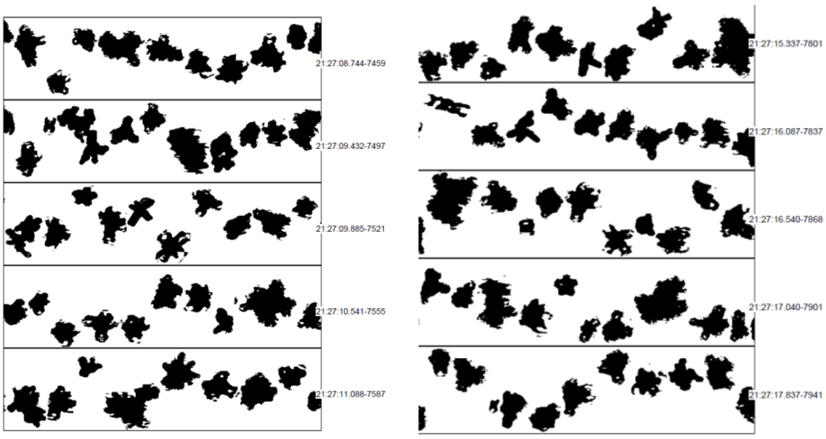

*Figure ~~10c~~8c. 2DS Images of highly irregular particles during a constant altitude run at -20°C during flight 226.*

### 3.2 Aerosol

Vertical profiles of the out-of-cloud aerosol measurements made by the aircraft are shown in Fig. ~~11~~9. Out-of-cloud measurements were selected as periods when the LWC was less than 0.001 g m$^{-3}$ and when the 2DS was not detecting particles. Contributions from large, swollen aerosol particles were also removed when the relative humidity was higher than 90%. Figure 11a shows aerosol concentrations over the size range from 0.5 to 1.5 µm as observed by the CAS and GRIMM probes. This size range of aerosols has been shown to best represent the concentration of INPs in many locations around the world (DeMott et al., 2010). Concentrations within this size range decrease significantly with increasing height, as would be expected, through sea spray aerosol being rapidly removed by cloud processing or sedimentation. Previous, measurements over the Antarctic Peninsula also found that aerosols in this size range decreased with height and ranged between 0.1 and 0.3 cm$^{-3}$ above approximately 2500m. Total aerosol concentrations, measured by the CPC during MAC, had a median value for the campaign of 408 scm$^{-3}$ (at standard temperature and pressure) and an inter-quartile range of 260 scm$^{-3}$.

~~Previous, multi-year measurements of aerosol at the Neumayer coastal Antarctic research station had a median concentration of 258 cm$^{-3}$. Minimum values (less than 100 cm$^{-3}$) were typically observed in June/July, while concentrations increased in the austral summer to a maximum of approximately 1000 cm$^{-3}$ in March (Weller et al., 2011). In winter, aerosol number and mass were both dominated by sea salt particles (87% by mass, Weller et al., 2008). Although aerosol composition in summer is more variable, sea salt still accounts for a significant fraction (50% by mass) but now with a large contribution from non sea salt sulphate (27% by mass, Weller et al., 2008). Measurements at the coastal Antarctic station McMurdo show the persistent presence of sulphate aerosol throughout the year (Giordano et al., 2017). In the winter these particles are highly aged. Sulphate aerosol then increases through the austral spring/summer, due to enhanced emissions of dimethyl sulphide (DMS) and methanesulfonic acid (MSA) from phytoplankton in the Southern Ocean (Gibson et al., 1990; Giordano et al., 2017). Giordano et al. (2017) also report the presence of a sub-250 nm aerosol population of unknown composition during the winter to summer transition. In addition a study has observed a significant fraction of organic carbon (>10%) and lower~~

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~~contributions from sea salt (<10%) in summer marine Antarctic aerosol (Virkkula and Teinil, 2006).~~

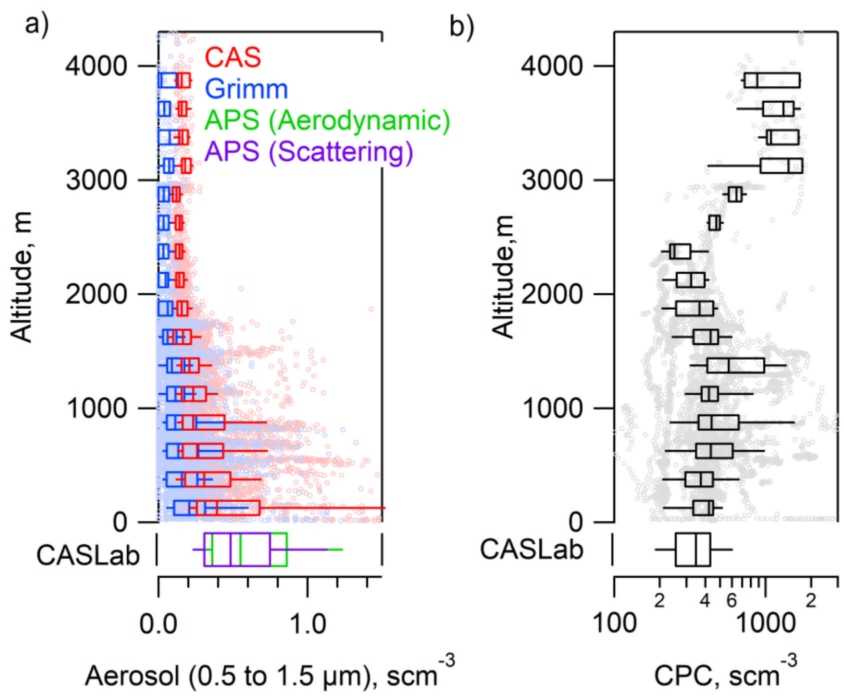


*Figure ~~11~~9. Aircraft clear sky aerosol concentrations (scm$^{-3}$) altitude profiles. Data are from: a) CAS and GRIMM instruments. Surface concentrations from CASLab are shown for comparison, from the APS; Green - aerodynamic particle size concentrations; Purple – scattering cross section derived particle size concentration measurements; b) Total fine aerosol concentration profiles, from CPCs on the aircraft and at the CASLab, (D>10 nm).*

During MAC episodic periods were observed with total aerosol concentrations in excess of 1000 scm$^{-3}$. These were often observed above cloud layers. The flights were designed to focus on cloud regions so may not represent a truly unbiased sample of the atmosphere, but the results do suggest a link between the observations of high aerosol concentrations and the presence of clouds. The limited spatial coverage of the aircraft measurements makes quantifying the extent of these layers uncertain, however they appear to extend over a few tens of kilometres to a hundred kilometres. At least two instances (flights 218, 219, see Fig. ~~12~~10) suggest a large layer extending beyond the cloud edge, pointing at the possibility of layers independent from clouds. The peak concentration usually occurred in the region up to 200 m above the cloud top (e.g. Flight 219). Some layers showed a clear drop in relative humidity (e.g. from 90% to 30%, e.g. during flight 220, 221, and 222) generally related to a clear temperature inversion, while other layers showed a much smaller decrease (by 10%) in relative humidity compared to the cloud underneath (e.g. flight 217, 218, 219). No clear systematic relationship was observed with respect to the vertical wind velocity (turbulence). The role of these particles as CCN/INPs is currently uncertain due to the lack of information about their composition.

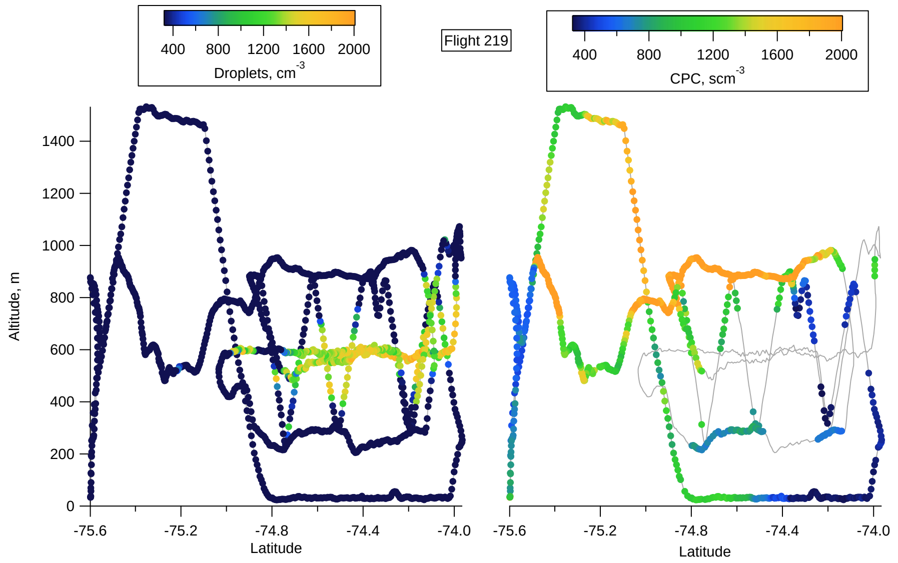


*Figure ~~12~~10. Latitudinal cross-sections of Flight 219 coloured by droplet concentration (left panel) and total aerosol concentrations out of cloud (right panel). Grey lines shows the flight track. These show a layer of high aerosol concentrations above the cloud top.*

Average total concentrations of UV-fluorescent aerosols (measured at CASLab with the WIBS) over the campaign period were ~ 1 $L^{-1}$, which was < 2% of the total particle concentration. Of these 0.01 $L^{-1}$ were identified as likely primary biological aerosols.~~, using the analysis described by Crawford et al. (2015)~~. During some Easterly and Westerly wind events, however, enhanced concentrations of the order of 5±7 $L^{-1}$ could be observed (Crawford et al., 2017).

### 3.3 Cloud condensation nuclei (CCN)

Figure ~~13~~ 11 (bottom panel) summarises the CCN measurements at the CASLab. The bottom panel shows the CCN at 5 different super saturations (0.05%, 0.13%, 0.20%, 0.26% and 0.34%). The hygroscopicity parameter κ is used to examine the effect chemical composition has on the CCN activity of aerosol particles. The derived κ values represent the average hygroscopicity of the volume-weighted fractions of the individual aerosol components. Non-hygroscopic components have a κ value of 0. Highly CCN active salts have κ values between 0.5 and 1.4, sodium chloride (NaCl) has a κ of 1.28 (measurement range 0.91 to 1.33). Organic species have values generally between 0.01 and 0.5 (Petters and Kreidenweis, 2007). The median κ value during MAC was 0.64 (inter-quartile range = 0.34, mean = 0.69), suggesting that this location is dominated by hygroscopic components, such as sea-salt and sulphate. Andreae and Rosenfeld (2008) review CCN measurements and find that κ values from marine locations generally cover a relatively narrow range of 0.7 ± 0.2, compared to 0.3 ± 0.1 for continental aerosols. A global model study subsequently presented a mean κ value of 0.92 (0.09 at 1σ) at the surface and 0.80 (0.17 at 1σ) within the boundary layer over the Southern Ocean (Pringle et al., 2010), only marginally higher than our MAC observations.

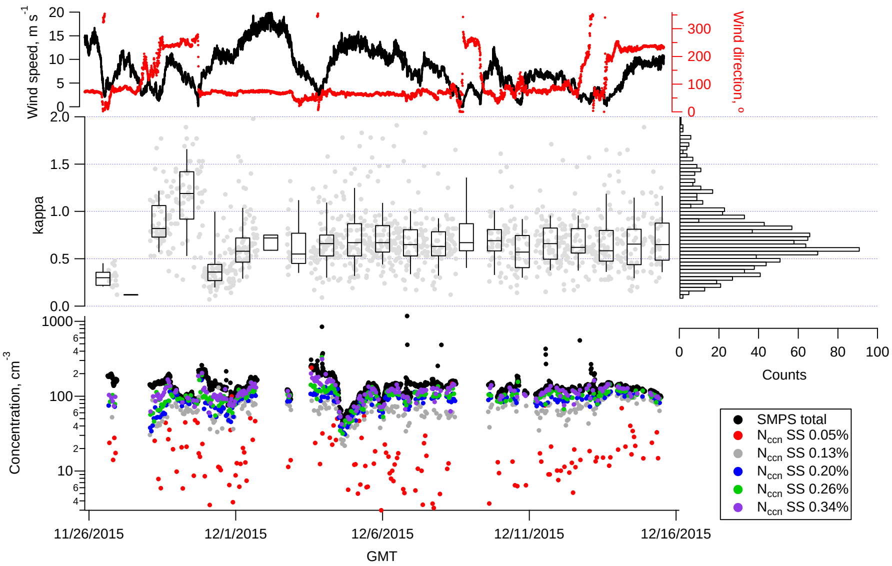


*Figure ~~13~~11. The top panel shows the time series of wind speed (black line) and direction (red markers) at the CASLab. The middle panel shows the time series of the hygroscopicity parameter κ. The box and whisker plots summarise the variability in κ for each day, while the right panel shows a histogram of κ for the whole measurement period. The bottom panel shows the total aerosol number from the integrated SMPS measurements (30 to 500 nm, black dots) and the CCN concentrations at 5 different supersaturations (SS, coloured dots from 0.05 to 0.34%).*

As shown in Fig. ~~13~~ 11 there was a period of increased hygroscopicity on 28 and 29 November 2015, with a median κ of 1.18 on 29 November. During this period there was a westerly wind. This changed to an easterly on 30 November 2015, which coincided with a decrease in hygroscopicity to a median κ for the 30 November of 0.36. Between the approximate headings 210° to 25° the CASLab lies between 30 and 60 km from the Weddell Sea. In contrast, within the sector 30° to 60° it lies several hundred km across the Brunt Ice Shelf from the Weddell Sea. To the south east of the CASLab lies the Antarctic Continent. HYSPLIT trajectories indicate over the past 5 days the airmass sampled on 28 and 29 November 2015 had passed over sea ice/open water regions. However after 30 November

2015 the hygroscopicity was relatively consistent and does not show a significant relationship with the wind direction or airmass history. For example, on the 14 and 15 December 2015 there was a westerly wind but the median κ for these days of 0.66 and 0.65, respectively, was similar to the campaign median (0.64).

### 3.4 Ice nucleating particles (INPs)

Ice nucleating particles (INPs) could not be directly measured on the aircraft during MAC. Instead we compare the cloud ice crystal concentrations with two parameterisations that are commonly used to predict INP concentrations. DeMott et al. (2010) compiled INP measurements from a range of locations around the world and derived a relationship using aerosol concentrations (within the size range 0.5 to 1.6 μm) and temperature that could explain the INP variability within their dataset to better than a factor of 10. For a broad comparison with the MAC dataset we evaluate DeMott et al. (2010) for a high (1 $scm^{-3}$, dark blue lines, Fig. ~~5b~~4b) and low (0.1 $scm^{-3}$, light blue lines, Fig. ~~5b~~4b) aerosol case. Cooper (1986) describes a simple INP parameterisation using only the ambient temperature, which is often used in the Weather Research Forecasting model (WRF) (Morrison et al., 2009). The concentration of INPs from Cooper (1986) is shown as a red line in Fig. ~~5b~~4b. It should be noted that neither of these parameterisations use Antarctic measurements. Given the marine location of the flights it is likely that these parameterisations may represent overestimates of the true INP concentration, since the number of INP in sea spray aerosol is generally several orders of magnitude lower than the number of INP in aerosol in the continental boundary layer (DeMott et al., 2015). The DeMott et al. (2010) parameterisation was derived using measurements at temperatures lower than -9°C, while Cooper (1986) used measurements below -5°C. For comparison they are extrapolated to higher temperatures and are therefore subject to increased uncertainty.

As shown in Fig. ~~5b~~4b, given the uncertainty in both parameterisations and the challenges with making a direct comparison with the measurements it is plausible that the observed ice concentrations at temperatures lower than ~~ca~~ about -10 °C could be explained by primary ice production. However above this temperature the measured ice concentrations diverge from the predicted INP by 1 to 3 orders of magnitude, suggesting that secondary ice production is becoming increasingly dominant.

Below -9 °C, where secondary ice production is likely to be less significant, Listowski and Lachlan-Cope (2017) found that the number of INP predicted by DeMott et al. (2010) gave better agreement with observed ice concentrations over the Antarctic Peninsula compared to INP parameterisations that only use the ambient temperature as input. For MAC, each in cloud data point was compared with the closest (in time) out-of-cloud aerosol measurement (1 minute average, RH < 90%). Data points were excluded from the comparison if no out-of-cloud aerosol measurements were made within 10 minutes of the in-cloud measurement. No clear relationship was found between the local aerosol concentrations and the ice concentrations ($R^2$=0.02 for the above cloud aerosol in the size range 0.5 to 1.6 μm). During MAC, the majority of cloud measurements showed no ice (see Fig. 3) suggesting that the Antarctic is a very low INP environment. As a result, all conventional INP schemes will likely overestimate the true concentrations.

### 3.5 Airmass history

To examine how aerosol and cloud properties vary with airmass history we perform back trajectory analysis using the UK Met. Office's NAME model (Numerical Atmospheric Dispersion Modelling Environment) (Jones et al., 2007) using Met Office Unified Model (UM) meteorological fields. Five day retroplumes were determined by releasing 10000 particles in the model at locations coincident with the aircraft's position. Here we examine the relative sensitivity to surface emissions from the following regions; the Antarctic continent, sea ice, Southern Ocean, ice shelf and South America. The numbers of particles near the surface (0 to 100 m) over each geographic region was summed every 15 minutes as the particles were dispersed five days backwards in time. For each region, the time integration of particles over the region was divided by the total number of particles appearing in the whole domain to determine fractional contributions (see Fleming et al., 2012). Shape files representing the monthly averaged sea ice extent from Polarview and geographical contour files for the Antarctic plateau, the permanent sea ice (ice shelves and permanent sea ice) and the American continent were used to determine the passageway of the air masses at surface levels sampled by the aircraft. This analysis was repeated for particles released at 60s intervals along the flight track to determine a time series of contributions from each geographic region.

The sampled airmasses were classified based on their time spent over different geographic regions (see Sect. 2.4). Figure 14 12 shows vertical profiles of the aerosol from the CAS (0.5 to 1.5 µm, relative humidity < 90%) when there was high (>50%, red markers) and low (<50%, blue markers) surface influence from the Southern Ocean, the sea ice and the Antarctic Continent. There is a broad trend of higher aerosol concentrations over this size range with greater contributions from the Ocean and sea ice, indicating significant emissions of sea salt/sulphate aerosol. Concentrations decrease with increased contributions from the continent, indicating a lack of sources in this region. These relationships are more distinct when the aircraft was sampling at low altitude, above approximately 1000 m the concentrations are less dependent on airmass origin due to their lower surface influence. This analysis was repeated using total aerosol concentrations from the CPC (Fig. 14). Similar to the CAS, higher concentrations were observed when there was greater influence from the Southern Ocean, with the differences again most distinct for the low altitude measurements. However, CPC concentrations are found to be less dependent on the influence of the sea ice and the Antarctic Continent.

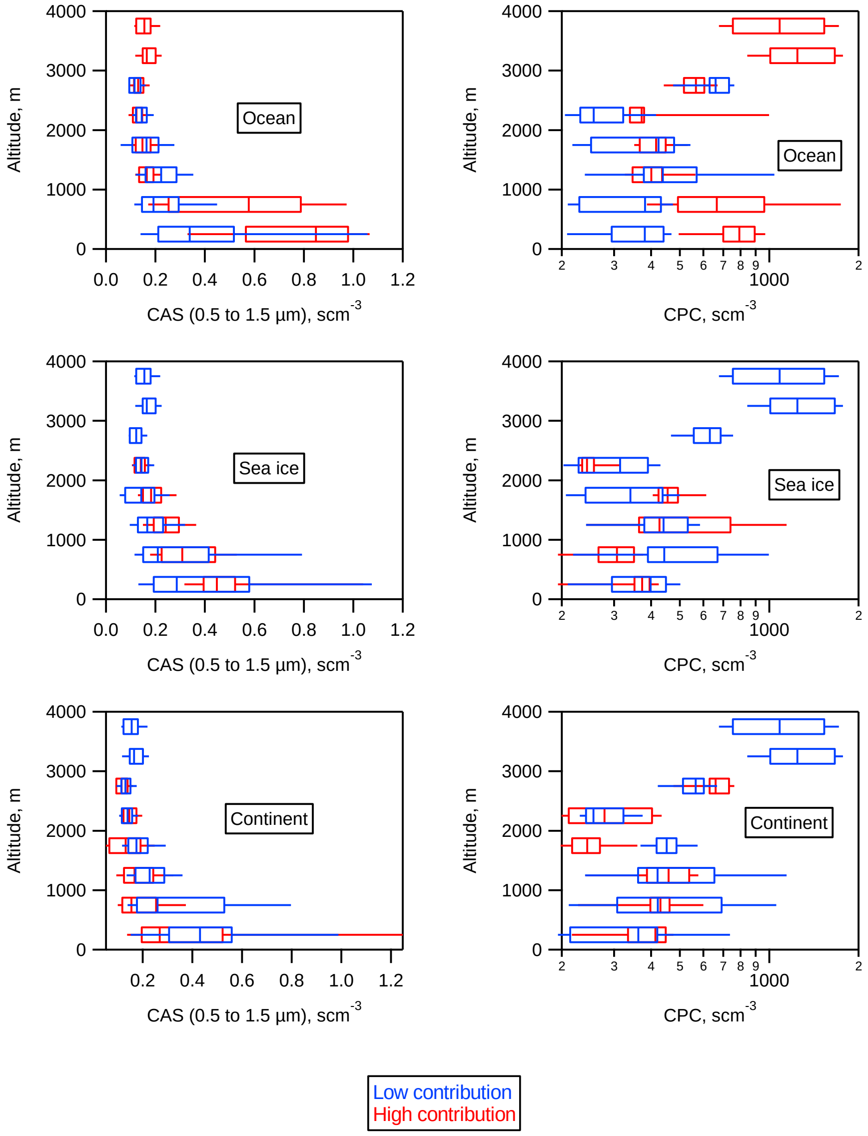


*Figure ~~14~~12. Altitude profiles of CAS aerosol over the size range 0.5 to 1.5 µm (left panels) and total aerosol, greater than 10nm from the CPC (right panels). The measurements have* *been partitioned into periods when the airmass had a high (red) and low (blue) contributions from different geographic regions (see text for details).*

Compared to the aerosol measurements the concentrations of cloud droplets and 2DS irregular particles are found to be less dependent on airmass history. Figure ~~15~~ 13 shows these variables as a function of the relative surface influence from the Southern Ocean, sea ice and the continent. The concentration of ice in the clouds is found to decrease for airmasses with increasing influence from the ocean. However, due to ice in the clouds being relatively infrequently observed the significance of this relationship cannot be determined. The effects of airmass history cannot easily be deconvolved from differences in sampling strategy or cloud properties (e.g. humidity, temperature, dynamics, and secondary ice production). The strongest relationship between aerosols and airmass history is for particles 0.5 to 1.5 µm this is only a small proportion of the total CCN. While the CPC will observe the CCN but also smaller particles. Also, given that the majority of measurements were conducted over broken sea ice, it may be that the CCN origin may be more local and not show up in the far field trajectories. ~~Most of the flights were conducted over sea ice, meaning that near field influences may be obscuring any relationship with airmass origin.~~

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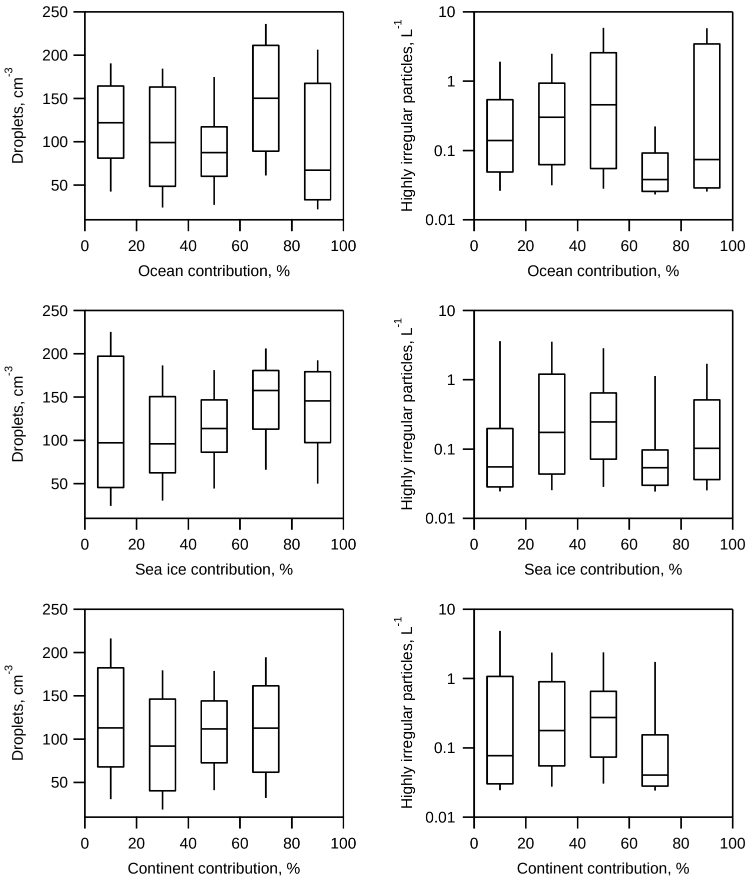


*Figure ~~15~~13. The concentration of cloud droplets and 2DS highly irregular particles as function of the airmass's contribution from the Southern Ocean, sea ice and the continent (see text for details). Boxes give the 25$^{th}$ and 75$^{th}$ and the whiskers are the 10$^{th}$ and 90$^{th}$ percentiles for each regional contribution bin.*

## 4 Discussion

This section summarise the observations presented in the paper and discusses the important microphysical processes. The cloud types were generally stratus, both single and multiple layers, predominantly between -20 and -3 °C. These were dominated by super cooled liquid drops, with a median concentration of 113 $cm^{-3}$. Droplet concentrations were relatively consistent during the campaign with an inter-quartile range of 86 $cm^{-3}$. The exceptions to this were when the droplets were depleted by high ice concentrations and also flight 217 where anomalously high droplet concentrations were observed, which was associated with an enhanced aerosol layer below cloud. Similar to Arctic layer clouds (McFarquhar et al., 2007), liquid content and cloud drop effective radius both increased with distance from cloud base likely due to condensational growth. Collision coalescence may also have contributed to this increase in effective radius. However, droplet number concentration was relatively invariant to position within the cloud.

Ice in the clouds exhibited a high degree of variability, occurring in small patches. Constant altitude runs by the aircraft through clouds at slightly supercooled temperatures (> -10°C) showed ice-free regions with patches of high ice concentrations (>1 $L^{-1}$). This variability is shown to exist over small spatial scales and may be a consequence of very low INP concentrations, where secondary processes may significantly amplify small differences in INP concentrations. This makes predicting in detail where ice will form in a given cloud extremely challenging. A detailed understanding of where the first ice will occur and also the conditions required for secondary production is needed. Here we examine this variability and discuss some of the potential controlling factors.

### 4.1 First Ice

First we examine the nature and sources of the INP. Global primary ice nucleation below approximately -15°C is thought to be dominated by soot and mineral dusts (Möhler et al., 2006; Murray et al., 2012; Niemand et al., 2012). However, this is colder than the cloud top temperatures generally observed during MAC. Biological species (pollen, bacteria, fungal spores and plankton) are the only INP that are known to be active at temperatures higher than approximately -15°C (Alpert et al., 2011; Murray et al., 2012; Wilson et al., 2015). Bioaerosol measurements at the CASLab show episodic high concentrations up to several per litre. This temporal variability in bioaerosol may be analogous to the spatial variability of the ice crystals observed in the clouds. Source apportionment of the bioaerosol at Halley is uncertain with the available dataset, but may include contributions from 1) the re-suspension of material from the local ice and snow surface, 2) coastal ice margin zones in Halley Bay where bird colonies are present and 3) long-range transport. The bioaerosol measurements will be presented and discussed in detail in a separate paper.

It is possible that the cloud layers sampled in MAC are seeded by precipitation from higher layers where the temperatures are low enough for dust to be active as an INP. During MAC the flights were designed so that measurements were performed between cloud layers to determine whether ice seeding from the upper layers was occurring. The frontal cloud sampled in flight 224 showed extensive ice precipitating between cloud layers and the cloud top temperature (below -20 °C) was sufficiently low for dust to be a potential source of ice nuclei. In the case of stratus clouds, those were not found to be seeded by layers at low enough temperatures for any dust to be active as an INP. Furthermore, single layer clouds such as those sampled in flights 219 and 227 still showed the patchy ice behaviour.

Detailed measurements of aerosol composition were not available on the aircraft. No clear relationship could be identified between the local aerosol concentrations (both above and below cloud) and the presence of ice in the clouds. However, only a small proportion of the total aerosol population are expected to be INP. Below ~~ca~~ approximately 2000 m (where most of MAC measurements were performed) there is a broad trend of ice being more frequent with decreasing altitude. A similar relationship is observed for the concentration of particles between 0.5 and 1.6 µm (Fig. ~~4~~9). However, this may in part be due to secondary ice production being efficient at these relatively high temperatures. Jackson et al. (2012) found a correlation (R=0.69) between the above cloud aerosol (0.1 < D < 3 µm) and ice concentrations in Arctic stratocumulus clouds. However these clouds were generally at lower temperatures (cloud top temperature < -10°C) than those during MAC and as a result are likely to have a higher proportion of primary ice production.

The surface may also be an ice crystal source either through blowing snow (Ardon-Dryer et al., 2011) or frost flowers (Gallet et al., 2014; Lloyd et al., 2015b). These will be most important for clouds in contact with the surface (Vali et al., 2012), but may also be relevant for low clouds when the humidity is sufficiently high that the crystals do not evaporate whilst being transported to the cloud base (Geerts et al., 2015). Space-borne lidar measurements of blowing snow over Antarctica found the thickness of these layers ranging between their detection limit (30 m) up to 1000 m, with an average thickness of 100 m. Approximately 71%

of these layers were less than 100 m thick and 25% were between 100 and 300 m thick (Palm et al., 2011). Similarly, lidar measurements at the South Pole found that layers were generally less than 400 m thick (63%), but could be up to 1000 m thick. Blowing snow is almost always constrained to the planetary boundary layer (Mahesh, 2003). The lofting of snow is complex; it is dependent on a range of variables, including: the snow type and surface meteorology (e.g. wind speed, turbulent mixing, temperature and humidity). A threshold wind speed of 7 to 10 m $s^{-1}$ is typically required (Dery and Yau, 1999). However, smaller crystals may show substantial fluxes at lower wind speeds. Aerosol fluxes from evaporated frost flowers have been estimated at $10^{-6}$ $m^{-2}$ $s^{-1}$ at wind speeds as low as 1 m $s^{-1}$ (Xu et al., 2013).

Evaluating the impact of these mechanisms during MAC is challenging since most of the in-cloud sampling was performed over snow covered sea ice, making it difficult to attribute local differences in the microphysics to the surface type. Figure 14 shows histograms of the surface albedo for out-of-cloud measurements (below 100 m) when there was (blue line) and was not (red line) ice observed. Here the surface albedo is used as a proxy for the surface type, since values near 0 correspond to overflying open water and the values near 1 correspond to a snow/ice covered surface. Figure 14 suggests that ice measured by the aircraft while out cloud (below 100 m) almost exclusively occurred when overflying a snow/ice covered surface, implying a link between the surface type and the presence of ice in the clouds. The ice measured on the aircraft when out-of-cloud could either have originated from the surface or precipitated from clouds above. However, it should be noted that very few measurements were made over open water regions. Evaluating the impact of these mechanisms during MAC is challenging since most of the in-cloud sampling was performed over snow covered sea ice, making it difficult to attribute local differences in the microphysics to the surface type.

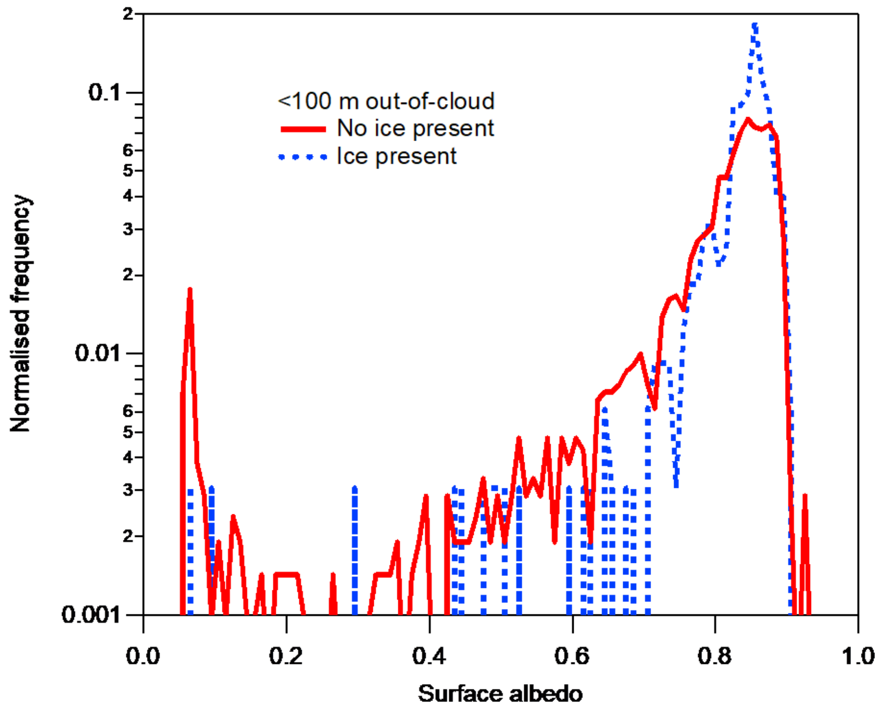


*Figure 14. Histograms of the surface albedo for of out-of-cloud measurements (below 100 m) when there was (blue line) and was not (red line) ice detected.*

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Flight 218 (Fig. 87) is one case where the first ice development may be due to surface ice crystals. During this flight ice was observed precipitating below cloud base. The majority of this ice precipitation was detected when flying over snow covered sea ice rather than open water. This was identified from the aircraft's forward facing camera and inspection of the surface albedo. Given the relatively low cloud base (300m), strong surface horizontal winds (5 to 10 m $s^{-1}$) and a relative humidity approaching 100% it is plausible that ice from the surface (e.g. from blowing snow) could mix up to cloud base, thus providing the first ice to the cloud. The sublimation rate of an ice crystal is largely dependent on the humidity. A 100 µm ice crystal at 0°C will have a lifetime of the order 100s at a relative humidity of 80%. At relative humilities of 90% and 95% the lifetime can be over 200 s and 400 s, respectively

(Thorpe and Mason, 1966). The ice crystals below cloud had similar habits to those observed in the cloud (a mixture of columns and rimed crystals) indicating they had not originated from the surface. However, only low concentrations of primary ice from the surface is needed if the ice is then able to multiply within the cloud due to secondary processes (Crawford et al., 2012).

### 4.2 Secondary Ice

Previous ice crystal observations over the Antarctic Peninsula show a similar behaviour to those during MAC with a peak in ice concentrations (> 1 $L^{-1}$) at approximately -5°C. Grosvenor et al. (2012) and Lachlan-Cope et al. (2016) attribute this to secondary ice production through the Hallett-Mossop process, where ice splinters are produced when a droplet freezes subsequent to colliding with an ice crystal (riming) (Hallett and Mossop, 1974). This can lead to rapid ice multiplication as the splinters freeze further drops, resulting in more splinters. Laboratory experiments suggest that this process is efficient over a narrow temperature range (-8 to -3 °C) with a peak at -5 °C (Mossop, 1976). Images from the 2DS probe at temperatures higher than -10°C generally show rimed crystals and small columns (Fig. 10a). These habits are generally observed when the Hallett-Mossop production mechanism is thought to be occurring (Crosier et al., 2011; Lloyd et al., 2015a).

A number of other secondary ice mechanisms have previously been identified, these include: large drops producing ice splinters when they freeze (Rangno and Hobbs, 2001; Lawson et al., 2015); and the break-up of ice crystals, generally either fragile dendrites due to sublimation, turbulence (Bacon et al., 1998) or because of collisions between crystals (Yano and Phillips, 2011). However, all these processes have generally only been observed to be efficient at temperatures lower than approximately -10 °C, which is lower than the temperature of the majority of clouds sampled during MAC. Taylor et al. (2015) suggest that the drop-freezing secondary ice production, identified by Lawson et al. (2015), may have occurred at temperatures higher than -10 °C in their measurements of cumulus clouds. However, they were not able to deconvolve its effects from the Hallett-Mossop mechanism. We have not performed automatic habit recognition on the 2DS images taken during MAC, however, inspecting the images "by-eye" suggests that the drop shattering events observed by Lawson et al. (2015) were not common during MAC.

The exact requirements for secondary ice production through Hallett-Mossop are still uncertain. It is thought that only a small of amount of primary ice is needed for it to be initiated, and recent model studies suggest this could be as low as 0.01 $L^{-1}$ (Crawford et al., 2012; Huang et al., 2017). Laboratory experiments suggest that production rates are proportional to the accumulation of large drops (>24 μm) (Mossop and Hallett, 1974). However, more recent field measurements found that estimated crystal production rates gave better agreement with observed ice concentrations if this constraint on drop diameter was removed (Crosier et al., 2011). Observations of Arctic mixed phase clouds found that the presence of precipitating ice particles (> 400 μm) was correlated with the number of large drops (>30 μm), however the precise nucleation mechanism through which this occurred was uncertain (Lance et al., 2011). During MAC both the analysis of individual case studies and the statistics for the whole campaign do not suggest that the concentration of large drops and ice crystals were related. However, any simple relationship is likely to be complicated as ice crystal growth will deplete the drops through riming and the Wegener-Bergeron-Findeisen process. This is shown in Fig. 6 5 and 7b 6b where the highest ice concentrations correspond to relatively low droplet concentrations.

Flights 226, 227 and 228 involved sequential vertical profiles to examine the dependency of ice on the clouds vertical structure. No link was identified between the presence of ice in the vertical profile and local variations in cloud top temperature. However, since the first ice occurs over small spatial scales, any relationship may be obscured by the aircraft's horizontal motion whilst changing altitude. As a result the precise cloud top temperature, and its variability, directly above the glaciated regions of the clouds is not known.

~~Higher ice concentrations were observed in updrafts/downdrafts compared to quiescent regions of the clouds. There are several possible explanations for this; first the more turbulent conditions may make more primary ice available through greater entrainment of aerosol and hence potentially more INP into the cloud. Second convective regions may indicate thicker regions of the cloud and lower cloud top temperature. This may lead to increased primary ice nucleation as the lower temperatures activate more INPs and the development of larger liquid droplets. Third, the more turbulent conditions could lead to more efficient ice production due~~

~~to ice being rapidly mixed to the Hallett-Mossop zone where concentrations would multiply. Finally, the riming rate may increase due to a greater number of ice-liquid collisions. More turbulent conditions may also indicate higher rimer velocity, however laboratory experiments suggest there is no lower cut-off rimer velocity for Hallett-Mossop to be active (Mossop, 1985).~~

## 5 Conclusions

Understanding the cloud in the Antarctic is essential to accurate predictions of future climate change. We have reported unique observations of cloud and aerosol properties over coastal Antarctica and the Weddell Sea. The aerosol was predominantly hygroscopic in nature, with κ being consistent with previous measurements and model predictions for remote locations dominated by marine emissions. The concentration of large aerosols (0.5 to 1.6 μm) decreased with altitude, as would be expected, through sea salt/sulphate aerosol being rapidly removed by cloud processing or sedimentation. Higher aerosol concentrations were observed in airmasses that travelled over the Southern Ocean/sea ice compared to those from the main Antarctic Continent.

In contrast to the aerosol concentrations, the droplet and ice concentrations showed minimal dependence on airmass origin, it may be that the CCN origin may be more local and not show up in the far field trajectories~~.~~ The cloud types were generally stratus, both single and multiple layers, at temperatures between -20 and -3 °C. These were dominated by super-cooled liquid drops, with a median concentration of 113 $cm^{-3}$. Droplet concentrations were relatively consistent throughout the campaign with an inter-quartile range of 86 $cm^{-3}$. The exceptions to this were cases when the concentrations became depleted by high ice concentrations, and also during Flight 217 when anomalously high droplet concentrations were observed; this was associated with an enhanced aerosol layer below the cloud layer. Both liquid water content and drop effective radius increased near cloud top.

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Ice in the clouds exhibited a high degree of inhomogeneity occurring in small patches. Below ~~ca~~ approximately 2000 m ice was more frequent at higher temperatures, however even within the -8 to -3 °C temperature range where Hallett-Mossop secondary production is most active, the clouds were predominantly liquid. When ice was present within the temperature range -8 to -3 °C it seems likely that secondary ice production, through the Hallett-Mossop process, resulted in concentrations that were 1 to 3 orders of magnitude higher than the number of INP predicted by conventional primary ice nucleation schemes. The source of first ice in the clouds is currently uncertain. First ice in the clouds often occurs at temperatures above -10 °C, this may be due to the presence of biogenic particles that are active INP at these temperatures or alternatively (or indeed simultaneously) ice from the surface (e.g. blowing snow or frost flowers) could be lofted into the clouds. The drivers of the ice crystal variability were investigated. No dependence on the droplet spectrum was found. However, higher ice concentrations were found in updrafts and downdrafts compared to quiescent zones, and therefore intermittent convective activity may explain the intermittent glaciation of clouds.

This paper has presented the most detailed in situ observations of coastal Antarctic clouds and their surrounding aerosol properties to date. Upcoming studies will use the MAC observations to test and improve the representation of Antarctic clouds in numerical weather/climate models in this particularly important region.

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