**We would like to thank both referees for taking the time to read and comment on the paper. Your comments are much appreciated and have led to improvements in the paper. We address each of the comments below.**

## Referee #2

*This manuscript, describing a unique data set, is a welcome addition to the literature since there are few or no cloud observations reported for the Antarctica region. The measurements and analyses are all quite good. I recommend some modest reorganization that would improve readability, namely to revise section 2 as a broader "Methods" section so that it can include not only discussion of such issues as aircraft produced ice particles and ice particle data analysis, but to introduce the ice nucleation parameterizations that data are compared to later in the paper.*

**We briefly introduced the IN parameterizations in the introduction as it was relevant to the discussion there. However, we decided to keep the discussion of the details of them contained within section 4 since we feel that such details are not needed before the main results of the in-situ observations and would therefore interrupt the flow of the paper.**

*Additionally, I think that some explicit discussion of expected Hallett-Mossop ice enhancement factors would be useful, to know if these are consistent with the measured ice concentrations attributed to primary nucleation and the peak values inferred to be caused by secondary ice formation. Otherwise, many improvements in organization were made already from the pre-press version of the paper, and it is reasonably well written, if a little long.*

**Some quantitative Hallet-Mossop splinter production calculations have now been made (see below).**

## Specific Points:-

*Page 17300, lines 23-24: The statement here gives the impression that ice nucleation activity in bacteria is a common trait. This is not so, as it is quite rare. Please modify, easily done by removing the "contrary" part of the statement.*

**Done.**

*Page 17300, end of Section 1.1: It seems appropriate here to mention other recent studies to investigate biological ice nuclei from oceans, such as diatoms (e.g., Alpert et al., 2011, Phys. Chem. Chem. Phys., 13, 19882–19894; Knopf et al., 2011, Nature Geoscience, 4, 88–90)*

**Added these to section 1.1:-**

In such a pristine environment as Antarctica it is possible that biogenic IN could play a relatively more important role, particularly on a seasonal basis. Alpert et al. (2011) and Knopf et al. (2011) showed that the presence of certain marine phytoplankton caused droplets to freeze at temperatures warmer than homogeneous freezing temperatures and it has been suggested that there are some bacteria that can nucleate ice at temperatures as warm as $-2\,^{\circ}$C (see Moehler et al., 2007; Hoose et al., 2010). However, concentrations of biological IN in worldwide snowfall have been found to be lowest in Antarctica compared to elsewhere (Christner et al., 2008) and also Junge and Swanson (2008) found that bacteria common in sea ice were not particularly efficient at nucleating ice at relevant temperatures.

*Page 17303, line 6: Aircraft exhaust acting as IN, at modestly supercooled temperatures? I think one needs a reference to suggest such a possibility as in the realm of believability. There has never been any laboratory or observational evidence for IN produced by aircraft exhaust at modest supercoolings that I am aware of. I have only ever previously seen the alternate hypothesis involving the cooling around propeller tips, which seems clearly justified.*

**The reference to exhaust aerosol has been removed.**

*Page 17307, line 10: Is there a way to know if clouds were above the flight level at any time? I gather no, but this should be stated somewhere as a potential weakness in clearly identifying the source of ice crystals at different levels.*

**Some description of the MODIS cloud top temperature field (Fig. 2) has been added to section 3.2.1 since this shows the presence of a layer at around -20 $^{o}$C over the ridge, extending over the Larsen C. The plane climbed from below this level and so was below the cloud top at this time. There was also cloud above the flight level whilst over Larsen C:-**

80 %). It is possible that there was a liquid water containing cloud above and that the ice was precipitation from this, which would be consistent with the relatively large size of the ice (mode size of 350 µm, but with an upper size limit of ~1000 µm). This is supported by MODIS cloud top temperature and cloud phase information from 14:15 and 18:15 UTC (1.5 hours before and after the period in question, respectively; not shown), which revealed the presence of bands of mixed phase cloud caused by lee waves forced by the northwesterly flow over Adelaide Island and the AP. This cloud was at temperatures similar to those sampled by the aircraft according to the earlier satellite image, but around 5-7 K cooler according to the later image.

*Page 17307, end of Section 3.2.1: There is mention here of the existence of supercooled rain in these clouds. This is a highly unusual observation that seems to beg better evidence than shown in Fig. 6c. Can it be pointed out which images are supercooled rain drops? Are there better examples available to support this claim?*

**The imaged particles in question have been highlighted in Fig. 6b. Some references for observations of supercooled precipitation formation have been provided:-**

b) and then to mostly supercooled water cloud, Fig. 6c. A very few large, smooth ellipsoidal particles (e.g. Fig. 6b) that were likely rain drops were also observed. The presence of precipitation sized liquid particles at supercooled temperatures without the presence of a warm layer above (i.e. ruling out melted ice) has been observed several times before and is generally associated with low IN and ice crystal concentrations (Huffman and Norman, 1988; Cober et al., 1996; Kajikawa et al., 2000).

**It generally seems to be the case that these occur when ice concentrations are low (as in the Antarctic clouds studied here).**

*Page 17310, lines 17-20: Is there any evidence for the seeds of the HM process? In other words, what was the initiation mechanism? Graupel? Frozen drops? Or can this not be determined?*

**Unfortunately, this may be difficult to determine since the frozen droplets may have been too small to detect. No large clearly frozen drops or graupel were directly observed, perhaps because they were too low in number. Additionally it is worth mentioning that Crosier et al. (2011) showed that snow crystals can also act as seeds for the HM process. However, we did not observe these in this region either.**

*Page 17310, line 21: Please take care to be clear as to what the "two regions" are. The two HM regions perhaps? The word "region" is used liberally throughout the paper, so sometimes it is necessary to be more explicit.*
**This has been made clearer in the text.**

*Page 17311, line 24: Suggest this sentence should end with "once primary ice has formed."*
**Done.**

*Page 17312, end of Section 3.2: Is there a better word than "complicated" to describe the ice ultimate formation process? It seems somewhat chaotic or at least heterogeneous, driven by the availability of ice nuclei and the conditions for secondary ice formation, neither of which are always assured. Furthermore, it is at this point of the paper that one wishes for some discussion of the likelihood of producing the observed ice concentrations in the HM regime on the basis of the inferred primary ice crystal concentrations potentially triggering the process, and the cloud droplet spectra. Can any kind of quantitative statement be made absent a complete modeling treatment of the clouds, such as performed by Phillips et al. (2003, Q. J. R. Meteorol. Soc., 129, 1351–1371) for convective clouds?*
**We have replaced this with, "but also demonstrates the somewhat chaotic and inhomogeneous nature of this process.". Since the process depends on a few different difficult-to-capture factors we wanted to get across the difficulties in representing this in GCMs.**

**We have made some quantitative estimates of the likely maximum concentrations of ice crystals that would be produced given the concentration of large droplets, based upon Mossop (1985). This has**

**also been done for flights 99 and 100 (see below). Unfortunately, using the primary ice concentrations to estimate the likely fraction of large droplets that are frozen is difficult since it would depend on accretion rates and on how far HM splinters are ejected, i.e. likely requiring an estimate of the area of cloud that can be frozen by each ice crystal falling from above. Since this is a multiplication mechanism, it is feasible that a single crystal falling from above could cause the glaciation of a large area of cloud. Detailed microphysical modeling would likely be required in order to do this.**

**The amended text is as follows:-**

> >25 µm in order to become active. The CAPS CAS instrument measured droplet size distributions in the supercooled liquid cloud region during the descent (to −2 °C) and revealed that at −5 °C, where the HM process has been shown to be the most active, small droplets (<12.5 µm) comprised ~80 % of the total droplet concentration, whereas droplets >25 µm represented ~2 % of the population. Mossop (1985) gives estimates of the number of ice crystals produced in the HM process for every large droplet that is frozen. This number varied between $\sim 1 \times 10^{-3}$ and $12 \times 10^{-3}$ and was found to be mainly dependent (although not monotonically) on the accretion velocity between the water and ice hydrometeors. The maximum concentration of large droplets observed in the descent was ~4 $cm^{-3}$, which, if they all were to freeze, would give a splinter concentration of between 4 and 48 $l^{-1}$. This likely represents an upper limit to this range since all of the large droplets may not freeze in reality. The lower end of this
>
> range agrees with the maximum ice concentrations seen in the precipitating region below and suggests that the number of large droplets would likely have been enough to allow the HM process to produce significant concentrations of ice once droplet freezing was initiated. Mossop (1985) also suggests that the HM process is most efficient when the ratio of small to large droplets is high, as in this case.

*Page 17313, lines15-16: In this case where no ice was observed, were the drop sizes requisite for HM? This seems relevant to document explicitly considering the discussion previous to this point. That is, primary ice is needed and appropriate cloud droplet conditions.*

**Similar calculations to those above for this region suggest that only low concentrations of ice would be produced in this region due to the presence of few large droplets. This has been added to the text:-**

> pled less than 8 km from the nearest edge of the sample run in this layer. However, it is likely that this ice evaporated before reaching the layer since the relative humidity w.r.t. ice reached as low as 60 % in the air above. Besides this, the number of large droplets (D >25 µm) was much smaller in this case (~ 0.025 $cm^{-3}$), which using the method described in Section 3.1.3 would give maximum ice concentrations in the range of only 0.025-0.3 $l^{-1}$. Whilst likely detectable, such ice concentrations are low compared to those seen in the HM regions of flight 104 and might suggest that a lack of large droplets also played a role in suppressing the HM process in this area.

*Page 17313-17314, Section 3.3.2: Here a cloud case is presented with stated top temperatures of only -6 _C . Is this case an exception to the stated likelihood of primary ice sedimenting from above (temperatures below -12 _C) in order to trigger the HM process in local regions, a point that is reiterated in the last few lines of the paper?*

**Examination of a MODIS cloud top temperature plot from 14:05 UTC reveals that there was a layer of cloud above the sampled cloud with a cloud top temperature of -35 to -40 °C. Thus it is possible that this provided seeding. We also added estimates of the splinter production rates, as just described for the other flights. The text has been modified to read:-**

**"**

In this temperature range the number of large droplets (D >25 µm) was ~ 0.9 $cm^{-3}$). Using the method described in Section 3.1.3 this would allow maximum HM ice concentrations in the range of 0.9-10.8 $l^{-1}$, which is comparable with the maximum ice concentrations actually observed.

Given the low concentrations of primary ice particles observed in the flights presented in this manuscript it might seem unlikely that in-cloud nucleation would be sufficient to initiate the HM process given that the cloud top temperature of the sampled cloud was only around −7 °C. However, a MODIS satellite image from 14:05 UTC (not shown) reveals that around the time at which the HM region was sampled higher altitude cloud with top temperatures of ~ −35 to −40 °C was present above the flight region. Thus, it is possible that this may have provided some ice seeding from above. However, it is unknown whether the relative humidity in the air between the clouds (estimated to be a layer around 3 km thick) was high enough to prevent ice evaporation.

Ice-free liquid-only regions were frequently present in the HM temperature zone dur-

**…"**

*Page 17316, lines 10-11: The parenthetical statement could use "activated" before "ice residues," as one could confuse the fact that the noted study examined the ice nuclei from freshly activated tiny ice crystals versus the residues of larger cloud ice particles.*

**This has been changed to:"**

($N_{0.5}$) since D10 argues that IN particles are usually in this size range (residue measurements from ice particles nucleated in their diffusion chamber suggested a mode size of 0.5 µm). There are likely to be some IN that are smaller than this, but the choice

**"**

*Page 17316, lines 25-26: Can it be clarified here if these measurements do or do not include periods in the marine boundary layer? It is unclear on the basis of discussion.*

**Except for the Larsen cloud sampling in Flight 99, the ice measurements made in the orographic and cloud layers over Larsen categories were all demonstrably above the marine boundary layer as judged from profiles of equivalent potential temperature near the measurement regions. Since the Flight 99 cloud was above the ice shelf it is perhaps unlikely that it would be strongly affected by sea salt. The Hallett Mossop clouds were close to the surface and were likely in the MBL – however, these were not used to characterize heterogeneous ice nucleation. The text has been modified to read:-**

One likely exception is in marine boundary layers where sea salt aerosol have been observed to contribute significantly to the total aerosol numbers for sizes >0.5 µm (O'Dowd et al., 1997). Sea salt has little or no ice nucleation ability (D10) and so its presence would be likely to disrupt the correlation between total aerosol and IN concentrations. All of the cloud in the "orographic" and "cloud layers over Larsen C" categories were demonstrably above the boundary layer with the exception of clouds

in the latter category in Flight 99. Sea salt is usually quickly removed because of its efficiency as CCN and so it is likely that the aerosols measured in these clouds contained little of it. The clouds in Flight 99 were above the ice shelf surface and thus also were unlikely to be contaminated by sea salt considering its short atmospheric lifetime. The HM flights were closer to the surface and thus were more likely affected (especially flight 100, which was over open ocean). However, these types of clouds were generally not used to characterize heterogeneous ice nucleation.

*It should also be made clear that the total CAS concentrations are used as the >0.5 micron surrogate concentrations. This discussion spreads over two pages, but could be greatly simplified by stating the assumptions used and then justifying them.*

**The text has been modified and shortened to address these points:-**

The CAS instrument has a lower size limit of 0.62 µm and thus we use the aerosol concentration above this size as a surrogate for the $N_{0.5}$ parameter required for D10. This would tend towards the underestimation of $N_{0.5}$. Also, in D10 there was a maximum possible sampled aerosol size of 1.6 µm. Here we impose an upper limit of 2 µm for approximate consistency with this. An exact match is not possible given the channels available from the CAS instrument. Thus, along with the uncertainty associated with high RH and the inherent instrument uncertainty, the precise measurement of $N_{0.5}$ is difficult. However, we will demonstrate shortly that the D10 parameterization is not especially sensitive to its exact value in the parameter space in question here.

*Page 17317-17318: I think it should be stated at the point of introducing the Cooper, Meyers, and Fletcher schemes that these have no inherent dependence on aerosol concentrations. Also, Figure 10 begs the question as to whether the D10 values are given for STP conditions (requires aerosol concentrations at STP as well), and if conversions for this factor have been made in all of the tabulated data as well. Has this been accounted for? As stated in my overview comments, these parameterizations could be introduced earlier in the paper.*

**We have noted that the schemes mentioned do not depend on aerosol concentration. These parameterizations were already introduced in section 1.**

**We did scale all quoted data to STP – this was mentioned at the end of section 2, except that we only mentioned it for ice concentrations. This has been changed to say that aerosol and LWC measurements were also scaled to STP.**

*Page 17321, lines 11-13: I suggest revising to note that not only IN profiles and cloud microphysical data are needed. Aerosol profiles and thermodynamic characterization of the cloud environment would be useful so that numerical simulations could be performed*

*using IN parameterizations and consideration of mixing processes to better determine if there is consistency or not between predicted and observed ice formation in these clouds.*

**We have modified this to read:-**

> These difficulties highlight the need in Antarctica for accurate measurements of IN and aerosol concentration data alongside in-situ cloud microphysical and thermodynamic data to allow a more accurate assessment of cloud parameterizations. Such data would also allow detailed cloud and microphysical modelling, which may give answers to some of the problems just mentioned (i.e. determining the details of the processes that likely occurred to produce the sampled ice).

*Page 17321-17322: The sentence straddling these two pages is the only quantitative statement regarding the efficiency of the Hallett-Mosspop process made in this paper. Just wondering if there is any way to determine the consistency of observations made in these flights with quantitative expectations for the HM process?*

**We have mentioned in the conclusions the quantitative analysis performed in response to the comments earlier :-**

> range. Such differences are consistent with previous studies. For example, Crawford et al. (2011) showed that in the presence of drizzle droplets, secondary ice particle production by the Hallett-Mossop process was able to increase the ice crystal concentrations by up to 4 orders of magnitude in timescales of up to 40 min. It has also been demonstrated here that the observed ice concentrations and droplet spectra are roughly consistent with the number of ice splinters produced by the riming of each large droplet (D >25 μm) in the laboratory studies of Mossop (1985), if a reasonably high fraction of the large droplets were to freeze.

## Referee #1

*1) While the DeMott et al. (2010) parameterization gives better agreement with the data than the older, non-aerosol based parameterizations, it should be noted that DeMott et al had very few IN data points < 0.1 lit-1, and those were at temperatures between -23 and -35 (DeMott Fig. 2). This is substantially colder than the temperatures considered in this work. In fact, DeMott Fig. 3B shows that these points are outliers if using the parameterization developed, predicting higher IN concentrations than were actually observed. At any rate, the fact that there isn't matching data for comparison should be discussed. This could be true of the other, older, parameterizations as well.*

**We added this discussion to the start of section 4:-**

## 4 IN parameterization comparisons

One of the aims of this study is to test the applicability of ice nuclei parameterizations (e.g. D10; Meyers et al., 1992; Fletcher, 1962; Cooper, 1986) for the Antarctic Peninsula region. The scheme of D10 was novel because, unlike the other schemes just mentioned, it also incorporated aerosol concentrations (as well as temperature) as a parameter that controls IN concentrations. D10 found that this allowed a much better fit to several IN concentration datasets than the older parameterizations that were based on temperature alone. However, it should also be noted that the datasets in D10 contained only a few observations of IN concentrations $<0.1\ l^{-1}$ and these were at temperatures of $-23$ to $-35\ ^{\circ}$C, which is slightly colder than the coldest temperatures sampled here. In fact for these points the parameterization predicted values that were somewhat high, which is consistent with our findings here (discussed shortly).

**The older parameterizations likely also did not sample low IN concentrations since they were mostly based on mid-latitude regions, which is the likely reason why they over-predict IN for clean regions. This point was already mentioned in section 4.**

*2) p. 297, lines 8-10: It's an interesting question how radiatively important clouds are over Antarctica, due to its already high albedo. Since the authors invoke radiative effects for why their measurements are important, more systematic detail in this section would be useful. Included should be a summary of relative importance of longwave and shortwave forcing of liquid vs. ice clouds in this region, and likely net effects.*

**We have expanded this part of the introduction to discuss this and made some other organizational changes to this section. Here is the revised text:-**

## 1 Introduction

Antarctica has a landmass equal to almost 10 % of the land area of Earth, and at 14.0 million km$^2$ is approximately twice the size of Australia. Most clouds over Antarctica occur in air masses at coastal regions that are moister than the dry continental interior. Kay et al. (2012, hereafter K12) showed that various satellite cloud climatologies (CALIPSO, MISR, ISCCP) reveal that the Southern Ocean is one of the cloudiest places on Earth with extensive cloud cover at all longitudes. From CERES-EBAF satellite measurements K12 also showed that the clouds throughout much of this ocean produce a large negative annual mean Shortwave Cloud Forcing (SWCF; values as low -95 Wm$^{-2}$) for the top of the atmosphere, which is larger in magnitude than the (positive) Longwave Cloud Forcing (LWCF; $\sim$ 25-35 Wm$^{-2}$). Thus, these clouds produce an overall cooling effect on Earth.

However, as well as being dependent on the cloud optical depth, SWCF is also dependent on the Solar Zenith Angle ($\theta_0$, the angle between the Sun and a line perpendicular to the surface) and surface albedo. Increases in these latter two parameters cause SWCF to become less negative. Thus, the magnitude of SWCF tends to reduce towards Antarctica over the open ocean since $\theta_0$ tends to increase, as demonstrated in K12 for the TOA radiation balance and in Pavolonis and Key (2003) (hereafter P03) for the surface. Over the ice covered landmass of the Antarctic continent, where both the surface albedo and $\theta_0$ tend to be high, SWCF is even less negative (P03) and reaches zero when there is no available sunlight in the winter months. P03 also demonstrated the diurnal variation of SWCF due to changes in $\theta_0$ throughout the day.

Relative to SWCF P03 suggested that LWCF varies only slightly with latitude and season in the Southern Ocean and Antarctic region. Thus, when the magnitude of the

SWCF is small, clouds act to warm the surface relative to clear skies. The results in P03 suggest that this is the case throughout almost the entire year at latitudes south of 75 ° S.

The region of interest for this study is the Antarctic Peninsula (hereafter AP), which is a ∼1500 km long finger of land consisting of a high mountain ridge with tops over 2 km high. It is the northernmost part of Antarctica with its tip extending to ∼63 ° S. The AP contains extensive ice shelf regions on its east side (the Larsen Ice Shelves). Between February and March, 2002 the Larsen B ice shelf experienced a dramatic disintegration when an area of 3200 $km^2$ was lost (Scambos, 2004). Crevasse propagation due to the weight of accumulated melt water is currently thought to have been the major factor in the 2002 break up, as well as in the break up of other ice shelves around the AP (Scambos et al., 2000, 2004; van den Broeke, 2005).

From over 2 years of surface radiation measurements Munneke et al. (2012) showed that most surface melting on the Larsen C ice shelf occurs in the daytime in the summer season, during cloud-free conditions and that the largest component to the melt energy was net downwelling SW radiation (see also King et al., 2008). Examination of the transition between a melting and non-melting period suggested that, despite a high surface albedo, increased cloud cover likely acted to reduce the net downwelling (SW+LW) surface radiation when considering the times of day at which melting occurred. For this reason, clouds over the Larsen and other ice shelves, as well as over sea-ice, are likely to be additionally important.

Surface processes in Antarctica may also be important in a global sense. Lubin et al. (1998) presented modelling evidence that changes to the local heating budget of Antarctica to changes in cloud properties might have global consequences through the altering of the latitudinal temperature gradient of the planet.

Given the very cold temperatures at Antarctica latitudes, ice phase processes will be important for many clouds there. A supercooled liquid cloud is likely to be more optically thick than a fully glaciated ice cloud, in part because ice particles will grow at the expense of water droplets due to the Bergeron-Findeisen process. This also leads

to increased precipitation from the cloud, depleting the overall water mass and reducing its lifetime with consequent radiative effects. Shupe and Intrieri (2004) showed that Arctic clouds containing only ice generally produced a much lower magnitude of both SWCF and LWCF than liquid containing clouds, which is consistent with them having a lower optical depth.

Thus, understanding what affects the properties of clouds in the Antarctic region (e.g. phase, optical depth, etc) is important as they are likely to have ramifications on both the local (i.e. surface) radiation balance and that of the planet. Despite this, Antarctic atmospheric processes remain poorly sampled, particularly in terms of clouds, due to its remote location and inhospitable environment. Manned surface stations provide the bulk of observations, but these are sparse, particularly in the continental interior. Some stations are equipped with ground based radar and lidar for the long term observation of cloud, e.g. Bromwich et al. (2012). However, in-situ cloud microphysical observations of Antarctic clouds have only been made rarely, for very brief periods, and with limited instrumentation. In particular, studies of ice formation in Antarctic clouds have been very limited and this will be the focus of the present work.

### 1.1 Ice in Antarctic Clouds

In the AP region only one in-situ cloud ice study of note has been published to date. Lachlan-Cope et al. (2001) describes the ground based sampling of an orographic cloud over the Avery Plateau using hand held formvar slide replicas. Ice crystals were photographed and counted under a microscope in order to calculate ice concentrations. Very large concentrations ($\sim$120 $l^{-1}$) were estimated with very few droplets observed. At the cloud temperature sampled, ($-17.5\,^{\circ}$C), this was significantly higher than predicted using the Fletcher (1962) ice nuclei (IN) parameterization. It was suggested that blowing snow from the surface that subsequently evaporated may have acted as a source of IN upwind of the measurements. This is consistent with the suggestions made in Hara et al. (2011) and Ardon-Dryer et al. (2011), that aerosol emissions from ocean and surface ice by wind driven suspension processes would result in enhance-

ment of aerosol concentrations in these size ranges making interpretation of surface sampled IN problematic.

Understanding of the relationship between ice and IN concentrations remains uncertain since it is often difficult to discriminate between observed cloud ice particle number concentrations activated through primary heterogeneous ice nucleation and those formed by secondary processes, without recourse to fast imaging spectrometers (e.g. Crosier et al., 2011). Furthermore, measurements of IN are difficult and only recently has there been a resurgence due to the development of new instruments (DeMott et al., 2011). Many in-situ cloud observations have suggested inconsistencies between measured IN concentrations and in-cloud ice concentrations (e.g. Fridlind et al., 2007; Cooper, 1986) with the suggestion sometimes being made that (as yet) uncharacterized processes might be operating. However, the difficulties mentioned above, and the lack of laboratory evidence, make this difficult to substantiate.

One aim of the present study is to examine how representative different heterogeneous ice nuclei parameterizations e.g. those described by DeMott et al. (2010, hereafter D10), Cooper (1986), Meyers et al. (1992) and Fletcher (1962), are for predicting ice crystal number concentrations for clouds prevalent in the AP and Larsen Ice Shelf regions. These schemes are all based upon measurements outside of the Antarctic region.

It was demonstrated in D10 that IN concentrations are correlated with concentrations of large ($D>0.5\,\mu m$) aerosol particles, as well as being negatively corrlelated with temperature. Due to the lack of anthropogenic aerosol sources over the continent, the coastal regions of Antarctica, such as the AP, show significantly lower aerosol concentrations than most other maritime regions (Hogan, 1986) and on the whole can be thought of as a relatively pristine environment. Combined with the results of D10 this might indicate an expectation of low IN concentrations in this region, although it should be pointed out that the Hogan (1986) measurements were of small Aitken mode aerosols rather than the large sized aerosols thought to control IN. However, evidence for the influence of long range transport of anthropogenic aerosols (fossil fuel burning)

from South America on the AP sector (to as far east as 2.5° E) was provided by Barbante et al. (1998). Other influences have also been identified by various researchers including Fiebig et al. (2009) and Hara et al. (2010), biomass burning; and McConnell et al. (2007), dust transport due to desertification. The latter study showed a doubling of alumino-silicate concentrations over the 20th century at an AP site and suggested a link to increased levels of desertification in South America.

Thus, Antarctic IN concentrations may show some anthropogenic influence and efforts to estimate them are likely to be complicated by this. Unfortunately, there have been very few observations of IN concentrations made in Antarctica and they have generally been with only surface based instrumentation. Kumai (1976) used electron diffraction analysis of residual central nuclei following the sublimation of 93 individual snowflakes collected at the South Pole to interpret IN type and possible sources. Interpretation of the results could have been confounded by the fact that many of the snowflakes contained particulate matter other than just the central nucleus due to efficient aerosol scavenging by the snowflakes. It was concluded, however, that Antarctic IN populations were likely dominated by clay particles arising from long range transport.

Bigg (1990) reviewed six different IN datasets from high southern latitudes (>60 ° S) collected between 1961 and 1988. Samples were collected in different regions, e.g. from ships close to the Antarctic Peninsula as well as surface sites. However, the measurement techniques used varied across the datasets. Reported mean IN concentrations ranged from $2 \times 10^{-4}$ to $0.2\,l^{-1}$ (at $T = -15\,°C$) with a suggestion that IN concentrations had decreased over the period.

In a more recent study, Ardon-Dryer et al. (2011) processed aerosol filters sampled at the South Pole (12 in total, 3 collected from a balloon and 9 from a laboratory rooftop). Solution droplets from the samples were tested in a freezing chamber to determine their activation temperatures; ice onset occurred at $\sim -18\,°C$. Elemental analysis subsequently verified that the composition of the aerosol was similar to that of mineral dusts collected from the Patagonian deserts in South America. Estimated IN concen-

trations ranged from 0.1 to 53 $l^{-1}$with a mean of 1 $l^{-1}$ at $T = -23\,^{\circ}$C. However, as with many similar near-ground studies, the concentrations were observed to correlate with wind speed, suggesting the filter samples were influenced by a local surface source subject to suspension processes.

In such a pristine environment as Antarctica it is possible that biogenic IN could play a relatively more important role, particularly on a seasonal basis. Alpert et al. (2011) and Knopf et al. (2011) showed that the presence of certain marine phytoplankton caused droplets to freeze at temperatures warmer than homogeneous freezing temperatures and it has been suggested that there are some bacteria that can nucleate ice at temperatures as warm as $-2\,^{\circ}$C (see Moehler et al., 2007; Hoose et al., 2010). However, concentrations of biological IN in worldwide snowfall have been found to be lowest in Antarctica compared to elsewhere (Christner et al., 2008) and also Junge and Swanson (2008) found that bacteria common in sea ice were not particularly efficient at nucleating ice at relevant temperatures.

Given these previous measurements and the general marine character of Antarctic aerosols, along with only the occasional influx of aerosol associated with non-ice covered land areas, IN concentrations in the AP region of this study would be generally expected to be fairly low, particularly as IN are generally thought to be associated with clay mineral and dust particles (e.g. Kumai, 1976; DeMott et al., 2003).

### 1.2 Airborne cloud measurements in Antarctica

Section 1.2 is as before.

*3) Most of the rest of the paper could be shortened and tightened up overall. There seems to be many meteorological details without discussing their significance, and statement of cloud locations, altitudes and LW and ice concentrations for each case, which might be better specified in the tables. Perhaps things could be reorganized into ice only (heterogeneous nucleation vs. Hallett Mossop), mixed-phase and liquid only cases for contrast and comparison. At any rate, the detailed discussion of all the cases seemed somewhat repetitive and the authors should consider if things could be condensed, without losing important points.*

**We have moved the synoptic description to an appendix to reduce the number of details in the main part of the paper. We feel that these synoptic descriptions have some merit as they place the microphysical measurements into a meteorological context. Similarly, the details of LWC, etc. can be found in the tables, but we feel that some description in the text is warranted in order to link them to specific parts of the cloud (e.g. whether in the lowermost layer of a set of 3 layers, etc.) and to provide some important details (e.g. whether there were high level clouds observed nearby from satellite, what the droplet spectra was like for Hallett Mossop considerations, etc), which may affect the microphysical interpretation. We have simplified some of the discussion related to the other flights.**

**As for dividing the results into ice-only, etc., we already have a table highlighting LWC only regions and Table 1 shows that there were very few ice only clouds. Hence we do not think that it is appropriate to re-organize the text in this way.**

## Specific:

*1) p. (17)299, line 11: What is the detection limit for soot by this satellite (in terms of concentration)?*

**Upon further investigation of this issue, we decided to remove this statement since most oceanic regions were shown to have no detectable influence of soot/absorbing aerosol with this instrument, suggesting the sensitivity is not enough to say anything meaningful about the Antarctic region. Please see the new introduction above for the new text.**

*2) p. 299, line 13: Was this "aerosol" the ice nucleating aerosol particles, or just general aerosol particles?*

**Hogan (1986) measured only small Aitken mode aerosol. A sentence detailing this has been added to this paragraph :-**

> It was demonstrated in D10 that, as well with temperature, IN concentrations are correlated with concentrations of large (D>0.5 µm) aerosol particles. Due to the lack of anthropogenic aerosol sources over the continent, the coastal regions of Antarctica, such as the AP, show significantly lower aerosol concentrations than most other maritime regions (Hogan, 1986) and on the whole can be thought of as a relatively pristine environment. Combined with the results of D10 this might indicate an expectation of low IN concentrations in this region, although it should be pointed out that the Hogan (1986) measurements were of small Aitken mode aerosols rather than the large sized aerosols thought to control IN. However, evidence for the influence of long range

**Please see the new introduction above to see how this paragraph fits into the revised text.**

*3) p.302, lines 16-17: For which probe(s) were shattered particles removed in the software, the CAS, CIP or both?*

**Shattering was removed only from the CIP measurements using this software. This has been noted in the revised text. An attempt to reduce CAS shattering was made through the removal of the flow straightener shroud as described in this section.**

*4) p.303, line 3-4: "very high concentrations" of what? Presume it's ice, but it should be specified.*

**Yes, we meant ice concentrations – this has been modified.**

*5) p.304, lines 18-25: After reading twice, I think I understand what the authors are getting at, but this paragraph is confusing. Please rewrite more clearly.*

**We apologize for the confusion and have tried to make these paragraphs more clear:-**

> The CIP-25 probe is probably unable to distinguish droplets from ice crystals for particle sizes <~112.5 µm due to its 25 µm resolution (e.g. see Avramov et al., 2011). Therefore, ice concentrations are only counted for crystals that are larger than this size, which means that newly nucleated ice particles will be undetected in this study. However, in a mixed phase cloud ice particles grow fairly rapidly so that they soon

become detectable.

Pruppacher and Klett (1997, hereafter P07) give estimates of ice growth rates through diffusion that suggests a growth time of 50–300 s to reach the detection size, depending on habit. P07 and Mitchell and Heymsfield (2005) give estimates of the fall speeds of different types of ice particle as a function of diameter and show that particles of diameter 112.5 µm have a fall speed of below ~30 $cm\,s^{-1}$. Thus, newly nucleated ice crystals will fall a maximum distance of around 90 m during growth to the detectable size, during which a temperature difference of <~1 degree would be experienced. Therefore, ice is theoretically detectable in conditions very close to those of its nucleation location.

For condensation freezing, most ice nucleation likely occurs nearer to the top of liquid layers due to the decrease of temperature with height and so it is these regions where the problem might be expected to be the worst. However, since in this study measurements were made at various depths relative to cloud top (including below cloud base), ice concentrations representative of that nucleated in the uppermost regions of cloud were likely sampled lower down in the clouds.

Finally, an important point to note is that all data in this manuscript (ice, aerosol and liquid water concentrations) have been scaled to STP values, in keeping with D10 and other studies.

*6) p.305: Based on the figures, I assume the 2D images were examined to confirm that the very low concentrations of large particles were, in fact, ice crystals and not ultra-giant aerosol particles, but this should be specified.*

**The images were manually inspected to confirm the presence of ice crystal shapes. Since we only consider ice particles larger than D=112.5 microns, it is very unlikely that ultra-giant aerosol would be detectable by the CIP instrument.**

*7) p. 307-308 discussion: Was there any effort made to sample throughout the depth of the cloud so cloud depth and nucleation regions near cloud top might be observed? Knowing the location of the samples relative to cloud top and cloud base would be useful, but it seems this was only available sometimes.*

**Unfortunately, it was not always possible to sample throughout the depth of the clouds in question, particularly for the orographic cloud. However, it is likely (as discussed above) that sampling below the cloud, or in the lower regions of clouds would provide some representation of ice that had nucleated near cloud top and precipitated downwards (except for aggregation losses, etc.) and therefore some idea of cloud top ice concentrations.**

*8) p.308, lines 8-9: "significantly lower" than what?*

**Lower than during the first mountain overpass – this has been added to the text.**

*9) p.309, line 12: What was the temperature of the low layer where the HM process was observed? I know it's discussed later, but it should be here for consistency with the rest of the discussion.*

**Done.**

*10) p.311, line 26-28: This doesn't seem to be stated correctly. How can a "lack of IN" "create the primary ice particles"?*

**We have changed this to:-**

> The most plausible remaining explanation for the lack of observed ice during the profiles then becomes a lack of primary ice particles due to a deficiency of IN. Ice particles are needed to initiate the riming process, which is fundamental to the HM mechanism. At HM zone temperatures, typical IN concentrations are predicted to be

*11) p.312, line 14: I would delete the phrase "but also demonstrate that the process is complicated". Ice nucleation does have many aspects, but it doesn't really seem that complicated in this case, which you have documented well.*

**Based on this and the comments from Reviewer #2 we have replaced this with, "but also demonstrates the somewhat chaotic and inhomogeneous nature of this process.". Since the process depends on a few different factors we wanted to get across the difficulties in representing this in GCMs.**

*12) p.312, bottom: I find it odd that a cloud with no liquid water and ice conc of 0.04 lit-1 (40 m-3) would be optically dense enough to be detected by MODIS, particularly if the cloud was over an ice surface (unclear if this was the case). Am I wrong about that, or perhaps this is a case where most of the ice is smaller than the 112 _m detection limit used for the CIP? Or the MODIS data is from a different time period? Please explain or discuss.*

**Examination of a further image at 18:15 (the original one was at 14:15 UTC and the flight period in question was at 16:45) showed some liquid cloud at temperatures around 5-7K cooler than those where the ice was sampled by the aircraft. This suggests a shifting pattern of (lee wave produced) cloud bands making it difficult to be conclusive, but with the most likely conclusion being that we did sample ice precipitating from liquid cloud above. The text has been amended and shortened to read:**

> at $T = -14.2\,^{\circ}$C. No liquid water was detected in this cloud (RH w.r.t. liquid was ~70–80 %). It is possible that there was a liquid water containing cloud above and that the ice was precipitation from this, which would be consistent with the relatively large size of the ice (mode size of 350 µm, but with an upper size limit of ~1000 µm). This is supported by MODIS cloud top temperature and cloud phase information from 14:15 and 18:15 UTC (1.5 hours before and after the period in question, respectively; not shown), which revealed the presence of bands of mixed phase cloud caused by lee waves forced by the northwesterly flow over Adelaide Island and the AP. This cloud was at temperatures similar to those sampled by the aircraft according to the earlier satellite image, but around 5-7 K cooler according to the later image.

*13) p.316, line 25: Which channels were used for the CAS aerosol totals? The DeMott parameterization has an upper size limit. Did the smaller particles dominate the concentrations here?– in which case the upper cutoff wouldn't be important, but worth mentioning.*

**An upper size limit of 2.0 microns was imposed to be consistent with the upper limit of 1.6 microns in the DeMott study. This generally made little impact outside of cloudy regions. This has been noted in the text.**

*14) p.317, line 14: Upon investigation of the tables, it appears that the 0.1-0.4 cm-3 was the range of the actual aerosol concentration observations, but this should be stated. Using a range of concentrations to assess effects of measurement uncertainty is a good approach.*

**This has been noted in the text.**

*15) Table 1, mean temp column seems to have an error: -413.5*

**Fixed – should have been -13.5.**

## New references


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